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SOCRATIC LECTURES

PART III:PHYSICS

14TH INTERNATIONAL SYMPOSIUM, LJUBLJANA, APRIL 17, 2026

EDITED BY

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Program of the Symposium Socratic Lectures, April 17, 2026, 10:00 – 17:00 (Ljubljana time)

10.00 Welcome to participants: Veronika Kralj-Iglič

10.10 - 10.45 Plenary lecture: Sebestova Janouskova Olga: Extracellular Vesicles: nanosize natural particles with BIG biological impact

11.00 Scientific sections

Section 1: Medicine (Chairs Amon Mojca, Janša Jošt)

- 11.00 - 11.30** Knežević Ivan: From Bedside to Bench: Translating Clinical Needs into Next-Generation Regenerative Biomaterials
- 11.30 - 11.45** Jelenc Matija: Preoperative Planning of Aortic Valve Repair in Cardiac Surgery
- 11.45 - 12.00** Janša Jošt, Amon Mojca, Hawlina Simob: Beyond Tumour Biology: The Role of Body Composition in Risk Stratification of NMIBC Surgery
- 12.00 - 12.15** Trotošek Blaž, Serša Gregor, Đokić Mihajlo: Electrochemotherapy - From Lab to Clinics
- 12.15 - 12.30** Kšela Juš, Racman Mark: Environmental Determinants of Cardiovascular Disease: Integrating Air Pollution, Metal Exposure, and Early Vascular Pathology
- 12.30 - 12.45** Amon Mojca: Exercise Physiology and Ergonomics – Bridging Physiological Extremes to Pathology
- 12.45 - 13.00** Kreča Žak, Strajnar Jan, Poredoš Jana, Nikolov Žan, Mali Katra, Boc Anja: Upper limb mobility after breast cancer surgery: pathoanatomical changes and šhysiotherapy approaches

Section 2: Biomedicine (Chair Vauhnik Renata, Manoso-Hernando Daniel)

- 11.00 Keynote lecture:** Manoso-Hernando Daniel, Mateo-Quel Jesús, Villaverde-Rúa Mario, Fernández-Llamazares-Mateos Gabriel: Comparison of shoulder muscle activation and strength between sides during the Closed Kinetic Chain Upper Extremity Stability Test and its modified version in patients with rotator cuff-related shoulder pain
- 11.30 - 11.45** Jelen Matic, Weber Daša, Kejžar Nataša, Vauhnik Renata: Correlation of Knee Joint Stabilization Force With Anterior and Mediolateral Rotational Laxity Using the Dyneelax Arthrometer
- 11.45 - 12.00** Breznik Katarina, Potočnik Maja Marija, Vauhnik Renata, Frangež Maja: Electrical potential of the deep abdominal muscles during different exhalation techniques
- 12.00 - 12.15** Fijavž Staš, Jerebic Mark, Milaković Rina, Olip Nejc, Paladin Maša, Žager Marciuš Valerija, Gošnak Dahmane Raja: The role of radiation therapists in addressing the side effects of radiation exposure
- 12.15 - 12.30** Strnad Špela, Jevsnik Podlesnik Mojca, Jamnikar-Ciglenecki Urska: Short Food Supply Chains in Slovenia: Main Challenges and Future Growth
- 12.30 - 12.45** Ančimer Špela, Antončič Jerca, Hrovatin Ajda, Piliš Klemen Aleš, Vauhnik Renata: Medial discoid meniscus treated with platelet-rich plasma: A case report
- 12.45 - 13.00** Vovk Lara, Vauhnik Renata: Physiotherapy approach in linear scleroderma of the lower limb: A case report

Section 3: Extracellular particles (Chair Pocsfalvi Gabriella, Turiak Lilla)

- 11.00 - 11.25** Keynote lecture: De la Cuesta Fernando: Identity Markers for Plant EVs
- 11.25 - 11.50** Keynote lecture: Ambrosone Alfredo: Exploring Plant Extracellular Vesicles for Innovative bio-based therapies
- 11.50 - 12.00** Chang Yu-Hsin: μ Pulse TFF optimization for plant-derived extracellular vesicle isolation
- 12.00 - 12.10** Patil Pradnya, Pratiwi Feby, Tokarchuk Kateryna, Oh JaeWon, Kim Kwang Pyo, Vainio Seppo: Comparative Proteomic Analysis of Strawberry-Derived Nanovesicles and Total Protein Extracts from Biodynamic and Conventional Strawberries
- 12.10 - 12.20** Moubarak Manea: Plant tissue culture
- 12.20 - 12.30** Bautista Hernandez Olivia del Carmen: From Cells to Cytokines: A Microfluidic Approach to Study Anti Inflammatory Effects of Plant-Derived Nanovesicles

- 12:30 - 12:40 Kralj-Iglič Veronika, Romolo Anna, Istileulova Yelena, Iglič Aleš: Coacervation: a mechanism of extracellular particle formation
- 12:40 - 12:50 Murto Henri: EV lab on a farm and new possibilities of regenerative farming for circular economy
- 12:50 Discussion

Section 4: Biophysics (Chairs Iglič Aleš, Beke Somfai Tamas)

- 11:00 - 11:30 **Keynote lecture:** Beke Somfai Tamas: Supramolecular antibiotics killing Gram-negative bacteria by mechanical membrane damage
- 11:30 - 11:45 Mercaldo Beatrice, Anna Romolo, Matej Hočevár, Matic Kisovec, Apolonija Bedina Zavec, Boštjan Korenjak, Aleš Iglič, Antonio DiLoria, Veronika Kralj-Iglič: Measuring density and viscosity of samples as an improvement of interferometric light microscopy
- 11:45 - 12:00 Arko Matevž, Matej Hočevár, Boštjan Korenjak, Iglič Aleš, Kralj-Iglič Veronika: Extracellular particles from equine and bovine milk
- 12:00 - 12:15 Korenjak Boštjan, Romolo Anna, Tratenšek Armando, Nemec Svete Alenka, Drobne David, Aleš Iglič, Vladimira Erjavec, Veronika Kralj-Iglič: Comparison of extracellular vesicle characteristics of human and dog
- 12:15 - 12:30 Mesarec Luka, Kralj-Iglič Veronika, Kralj-Samo, Iglič Aleš: Effect of the concentration of curved rod-like molecules on the equilibrium shapes of two-dimensional shells
- 12:30 - 12:45 Drab Mitja: Modeling active anisotropic membrane inclusions
- 12:45 - 13:00 Klemenc Ana: Applications of surface-enhanced Raman spectroscopy in biomedicine

Section 5: Materials (Chairs Kralj-Iglič Veronika, Juan Alfredo)

- 11:00 - 11:30 **Keynote lecture:** Patrignani Mauro, Weber Jennifer, Bechthold Pablo, González Estela, Sainz Díaz Ignacio, Sandoval Mario, Figueroa Carlos, Jasen Paula, Juan Alfredo: Exploring the Effect of Oxygen on SiC/Fe Interfacial Adhesion Using Experiments and Quantum Mechanics simulations
- 11:30 - 11:45 Bar Laure, Lavrič Marta, Caf Maja, Kralj Slavko, Kumar Sadhu Raj, Škarabot Miha, Losada-Pérez Patricia, Iglič Aleš, Cordoyiannis George: Lipid-phase-modulated interactions of gold nanoparticles with supported vesicular membranes
- 11:45 - 12:00 Cepec Eva, Godič Torkar Karmen, Šunta Urška, Griessler Bulc Tjaša, Manjarres Castro June, Istenič Darja: The Microbial Comeback: Challenges in Achieving Long-Term Axenic Microalgal Cultures
- 12:00 - 12:15 Bosković Neda: Microplastics in aquatic environment: insight from research in Montenegro
- 12:15 - 12:30 Karas Tjaša: Applications of Machine Learning in Brillouin Spectroscopy
- 12:30 - 12:45 Žarković Lara, Zupan Iza, Hankić Larisa, Žager Marcuš Valerija, Gošnak Dahmane Raja: Patient Safety in Radiotherapy: Human Factors and Error Prevention Systems
- 12:45 - 13:00 Bensa Maja, Jevšnik Podlesnik Mojca, Vovk Irena: Consumers' food safety concerns in a changing food landscape

Section 6: Complexity (Chairs Jelen Andreja and Švajger Blaž)

- 11:00 - 11:30 **Keynote lecture:** Sojecka Agata Angelika, Drozd-Rzoska Aleksandra: New Insights into Global and Local Human Population Changes
- 11:30 - 11:45 Jelen Andreja: Formation of complex patterns in Ferromagnetic High-Entropy Alloys which leads to magnetic softness
- 11:45 - 12:00 Župec Matjaž: Power-law and Tsallis distribution in football goal scoring
- 12:00 - 12:15 Sgerm Jaka, Kodrin Marcel: Visualising order parameter spaces
- 12:15 - 12:30 Dovnik Damjan: Quantum Hall Effect
- 12:30 - 12:45 Fras Rene: Electrocaloric effect
- 12:45 - 13:00 Michelini Sara, Bele Marjan, Tatti Francesco, Drobne Damjana, Mandič Mulec Ines, Šimunovič Katarina: Influence of in vitro model complexity on Campylobacter jejuni-host interactions

Section 7: Soft matter (Chair Kralj Samo, Drozd-Rzoska Aleksandra)

- 11:00 - 11:30 **Keynote lecture:** Rzoska Sylwester J, Drozd-Rzoska Aleksandra: The New Philosopher's Stone: 'hard' & 'soft' matter under extreme pressures
- 11:30 - 11:45 Melani Potrč: Dynamic Light Scattering as a Tool for Characterization of DNA-quadruplexes
- 11:45 - 12:00 Javornik Tim, Kravos Iztok: Kibble Zurek Mechanism in Confined Geometries

- 12:00 - 12:15 Zid Maha: Topological charge conservation in planar confinements
12:15 - 12:30 Hölbl Arbresha: Weakly disordered soft matter
12:30 - 12:45 Ribaš Anja: Chaotic Metasurfaces
12:45 - 13:00 Švajger Blaž: Percolation in electrocaloric applications

Section 8: Artificial Intelligence in the Classroom – Partner or Challenge? and STEAM as a Catalyst for Educational Innovation (Chairs Gomes Nelson and Borghesan Mariapia)

- 11:00 - 11:15 Borghesan Mariapia: STEAM as a Catalyst for Educational Innovation
11:15 - 11:25 Brogna Daniela: STEAM Hackathon: Sky patterns on the ceramic road between IZNIK and SALERNO
11:25 - 11:45 Simac Anita: When Geometry Becomes Art: Bridging Art and Geometry in the Classroom
11:45 - 12:00 Hustiuc Nicoletta: Stem activities in eTwinning projects
12:00 - 12:10 Gomes Nelson: Artificial Intelligence in the Classroom – Partner or Challenge?
12:10 - 12:30 Holanda Leandro: Designing Culturally Sustaining Learning Environments: The Role of AI, STEAM, and Project-Based Learning
12:30 - 12:50 Videnovik Maja: From Challenge to Opportunity: How AI and Gamification Enhance Motivation and Engagement in the Classroom
12:50 - 13:00 Gomes Nelson: The STEAM StartUp and how we can participate in a collective library of interactive STEAM resources

Section 9: STEAM in Action – How Arts Enrich STEM Learning and Well-Being (Chair Istileulova Yelena)

- 11:00 - 11:05 Istileulova Yelena: How Arts Enrich STEM Learning and Well-Being
11:05 - 11:30 Makrides Gregoris: The STEAME-Academy Evolution: Latest Results
11:30 - 11:45 Kos Miha: From STEM to THE MAJESTIC: Covering Illustrations, Sonnets and Diabetes
11:45 - 12:00 Jeran Marko: Poetry from the Laboratory: Red Cabbage as a Sustainable pH Indicator and Poetry as a Teaching Tool
12:00 - 12:15 Cizelj Boris: Silver Economy
12:15 - 12:30 Adnyana Wayan Kun: Rethinking Ordinary Objects in Contemporary Painting Creation
12:30 - 12:45 Pozdorovkina Irina, Rezvukhina Lidiia, Bliznyuk Elena, Zuykova Anna: Creative Wellbeing Through Plein Air Painting: A Balance between Academic Tradition and Intuition
12:45 - 13:00 Istileulova Yelena: STEAM in Action: From Atomic Structure to Well Being through Music and Digital Interpretation of Lucretius
13:00 **Plenary lecture:** Papanikos Gregory: From TES (Technē, Epistēmē, Sophia) to STEAM: Bridging Ancient and Modern Conceptions of Knowledge

Editorial

With the participation of world top scientists from the fields and integration of science in international artistic production, Socratic lectures strive to present scientific and artistic excellence to the students, involve them in the creation of science and art and push forward development of curricula within university education.

14th Socratic Lectures symposium took place online on April 17, 2026.

It featured two plenary lectures, 9 scientific sessions, a poster session and the student session, with about 240 active participants.

With over 40 submissions received, the Proceedings of the 14th Socratic Lectures is organized into four distinct parts: Part I focuses on Breast cancer research, followed by Medicine in Part II, Physics in Part III, and Interdisciplinary sciences in Part IV. Some contributions of Physics (Part III) were contributed by the students.

This vision is driven by a unique primary mission: to invite students to actively engage in authentic research early in their academic journeys. While student participation is well-established at the doctoral level or through prestigious accolades like the Prešeren Awards, our symposium introduces a paradigm shift. We open these doors to the very youngest scholars—those who have just entered the university matrix, including those who may not yet stand out as top-tier students among their peers.

However, this radical inclusion cannot happen through institutional inertia; it requires the intense, personal engagement of professors who volunteer their time and mentorship. To bring these young minds into direct contact with cutting-edge global science completely free of charge, we have leaned heavily on digital conferencing technologies. Without the online framework, a symposium of this global magnitude would be financially impossible to sustain for un-funded student researchers.

In the Socratic circle, the individual is no longer the centre of attention, and there are no prizes for mere "compliance" or "cleverness." Instead, we step into a space where we share our ignorance, our collective helplessness, and our humility when confronting complex, unresolved problems. A student, stepping into the scientific world requires a profound psychological transition. Learning to stand before the unknown without fear, and discovering the joy of tackling these trials as a team, is the first real step toward becoming a scientist. Unlike commercial journals or conventional conferences driven by institutional metrics, Socratic lectures operate on the spirit found within Plato's Dialogues—the timeless realization that knowledge is not a commodity to be hoarded for self-promotion, but a shared horizon. Students consciously choose to align their manuscripts with strict editorial templates and technical standards, they transcend selfish interests, grades, and conquest for personal promotion. They become a part of the whole. This mental shift does not require years of advanced study; it takes only a few seconds of internal clarity and intent. While some grasp this instantly and others struggle, it remains the ultimate destination of our educational mission. It is with this ethos that we welcome you to these proceedings, not merely as readers of data, but as active participants in a community built on mutual care, resilience, and the relentless pursuit of truth.

We are counting on the Physics to provide theoretical descriptions of material phenomena, and also include the issues of the living beings. We are very happy to present here 11 original research contributions considering theoretical and experimental results. Since the beginning in 2008, Physics has been the keystone of the Socratic lectures and is remaining as such - also in the present publication.

Part III: The front cover features 3 Standard Stoppages (1913–1914) by the avant-garde artist Marcel Duchamp (1887–1968). Created by dropping three one-meter-long pieces of thread from a height of one meter and permanently fixing their random, gravity-warped curves onto canvas, the artwork includes the three custom wooden rulers seen here, crafted to match those organic lines. Though conceived primarily as a witty, conceptual critique of absolute measurement and classical Euclidean straightness, the elegant, parallel geometries of the resulting templates hold an undeniable visual rhythm. In this volume of the Socratic Lectures dedicated to Physics, the image stands as an artistic nod to the tools, models, and constructs we utilize to map the natural curves and boundaries of physical phenomena.

We conclude with a big thanks to all the participants who in the spirit of Socrates donated their contributions and to all those who made the events possible. Kindly invited to the next Socratic lectures.

Veronika Kralj-Iglič, Anna Romolo and Yelena Istileulova

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Research

Human Population Growth in Anthropocene: Active Soft Matter Perspective.

Drozd-Rzoska Aleksandra^{1,*}, Sojecka Agata Angelika^{2,*}

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Abstract:

Despite a long record of studies, the effective and fundamentally validated modelling of global population changes remains a challenge. One can recall puzzling forecasts up to 2100, and the significance of looking back, even to the onset of the Anthropocene. This report presents the recently developed Human Intelligent (HI) Active Soft Matter (ASM) concept as the fundamental reference. It is validated by describing the dynamics using universalistic scaling equations, developed in Soft Matter Physics, with a well-defined fundamental reference. They are Super-Malthusian equations and a critical-like scaling. Significantly, the analytic population dataset, covering the period from the Anthropocene onset to 2025, is examined to determine the newly proposed analytic per capita relative growth rate (RGR), which is then used to ultimately validate the scaling equations discussed via RGR-based, linearized, distortion-sensitive analysis. Notably, the discussion focuses on basic patterns in the dynamics of Soft Matter systems and also indicates some parallels to the nuclear chain reaction pattern. The report also discusses emerging links between significant ecological and historical events and characteristic changes detected in global population evolution.

Keywords: Active Soft Matter; Global Human Population; Dynamics; Scaling; Critical behavior; Stretched exponential; Compressed exponential; Carrying Capacity

1. Introduction

Today, there are 8.3 billion people on Earth, with a daily increase of ~150,000 infants. Determining changes in the global population through 2100 is crucial for global planning and management across many areas, including politics, food security, critical raw materials, and ecology (WPR, 2026; Taagapera & Nemčok, 2024; Sojecka, Drozd-Rzoska & Rzoska, 2024; Sojecka, Drozd-Rzoska & Rzoska, 2025). This is predominantly determined by the universal metric of Global Carrying Capacity (Sojecka, Drozd-Rzoska & Rzoska, 2025). However, reliable predictions require a sound understanding of the past. The importance of such past experiences was perhaps first emphasized in the Holy Bible: the phrase from *Ecclesiastes I, 10* "nihil novum sub sole", (there is nothing new under the sun), then repeatedly invoked in various forms by thinkers.

For the authors particularly notable is Cicero (106 BC to 43 BC, Ancient Roman Republic times), who specified (Everitt, 2003): *'Historia vero testis temporum, lux veritatis, vita memoriae, magistra vitae, nuntia vetustatis, qua voce alia nisi oratoris immortalitati commendatur?'* (eng.: *By what other voice than that of the orator is history, the true witness of times, the light of truth, the life of memory, the mistress of life, the heraldress of antiquity, committed to immortality?*).

Therefore, let us use this guideline to attempt to estimate future changes in the global population and the associated changes in global carrying capacity. Let us look back on past experience, dating back to the origins of the human population. However, the aim of this study is to provide insight grounded in the principles of the Scientific Method (Holman, 2023; Harper, 2012), in which empirical data and the clear validation of the models used to describe them are paramount. In this paper, we present an analysis of both the global population and the global Carrying Capacity.

The authors of the given report implement this essential rule by using the dynamics of (Human Intelligent, HI) Active Soft Matter concept as a reference, for discussing global population changes. This provides an explicitly universalistic, in-depth, and validated fundamental basis. This report summarizes the authors' research in these directions and presents new interpretations and results, particularly regarding Global Carrying Capacity, supplemented by new concepts and reasoning.

However, before we begin, it is necessary to indicate and justify the starting point of the history of the global human population discussed in this report.

A decade ago, pre-human footprints were discovered in western Crete (Greece). They show hominin-like features and date back to the late Miocene. ~5.7 million years ago (Gierliński et al., 2017). After long evolutionary development and population growth, Human ancestors in Africa and Eurasia were pushed to the brink of extinction around 900,000 years ago (Grine & Fleagle, 2009; Hu et al., 2023). About 98.7% of human ancestors were lost. This was supported by genetic tests of the fossil record from Africa and Eurasia, which date to between 950,000 and 650,000 years ago. This extremely small population, likely only 1,200–1,500 individuals, is the ancestor of all subsequent human populations (Hu et al., 2023). Not only did they survive successive long periods of global glaciation, interrupted by shorter interglacial warmings, but they also reached locations on almost all continents — Antarctica being the obvious exception. Remarkably, this progress was achieved during the last glaciation, which was perhaps the most severe in terms of climate. This can only be attributed to an increasingly evolved ability to adapt and utilize all available environmental resources, often scarce, at a given level of development. The undoubted advantage of the genus 'Homo' was adaptive intelligence, supported by social interactions.

The first anatomically 'modern' Homo Sapiens appeared in Africa around 300,000 years ago. The oldest known remains were discovered at Jebel Irhoud in Morocco. This occurred during one of the previous Ice Ages, which lasted over 50,000 years. After a brief interglacial period, however, the next Ice Ages followed. They culminated in a particularly long and severe last Ice Age that peaked only 20,000 years ago. However, as early as 19,000 years ago, a period of rapid warming known as the 'Great Thaw' began (Grine & Fleagle, 2009; Ehlers & Gibbard, 2007).

This interglacial period continues to this day and was only briefly interrupted by the Younger Dryas episode, which peaked 11,700 years ago. This also marked the beginning of the Holocene, which continues to this day. This period is associated with the rapid development of Homo sapiens and the beginning of the Anthropocene era, which is linked to 10,000 BCE. The end of the Ice Age and the subsequent Global Warming sped up so

rapidly that, by 16,000 years ago, the ice sheets in Europe had retreated to the Alps, Scotland, and the region stretching from Scandinavia to the northern coast of Poland and the Baltic states. In America, meanwhile, the ice sheet had already begun to retreat northwards along the western coast of North America, leaving a wide, clear corridor. However, the ice sheet was still relatively close to the ocean floor, meaning its level was around 100 meters lower than it is today (Ehlers & Gibbard, 2007).

This meant there were wide land passages from Asia to the Americas via the Bering Strait. The continental nature of Great Britain and Ireland was such that they were almost connected to Scandinavia by the 'Grand Doggerland', which extended beyond the North Sea. The La Manche Channel (English Channel) was also land at that time, of course (Stutz, 2026). In a warming climate, these ice-free areas provided additional opportunities for the growth and development of the Homo sapiens population.

This work analyses global population changes from the beginning of the Anthropocene, when Europe was free of glaciers and North America was experiencing a very rapid glacial retreat. A Brave New World opened up, offering an increasingly favorable climate for humans and unlimited resources within the needs and exploration capabilities of contemporary human populations. The human population had already undergone unique multidimensional evolution during the Ice Ages. It included cognitive abilities and the development and implementation of technological innovations that were passed down and further refined in subsequent generations. Perhaps most important were increasingly sophisticated social and cultural relationships.

2. Standard approaches to the global population dynamics problem

First, we would like to revisit the general overview of global population change in the Anthropocene, as depicted in the standard picture provided by the United Nations Population Fund (UNESA, 2026; UNFPA, 2026), and shown in **Figure 1**. In particular, they highlight the Neolithic period in the prehistoric era, when the population was at its terminal stage. They also indicate the Roman Empire epoch in late Antiquity.

It is worth noting the unique, complex, dynamic system features of the global population:

- (i) Global Population is a 'closed', Planetary System
- (ii) External impacts are minimal: the last great planetary extinction due to the Chicxulub Asteroid impact, near the Yucatan Peninsula, occurred 66 million years ago
- (iii) It is governed and driven only by internal features and dynamics
- (iv) There is no external population's impact (assuming UFOs, Aliens, etc, do not exist)

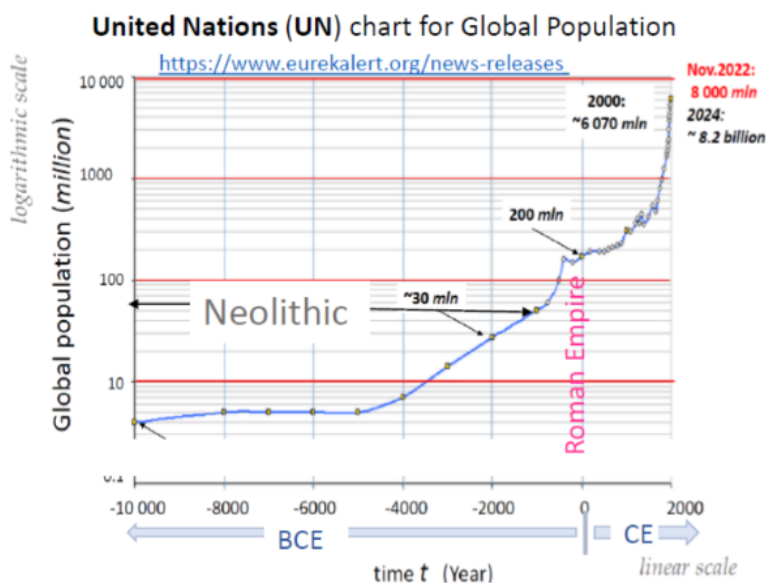


Figure 1.: The picture standard of the United Nations, with the authors' supplementation to indicate issues significant to the given report (UNESA, 2026; UNFPA, 2026).

Regarding human population growth modelling, there are two standard cognitives:

1st Path, probably dominant, aims to define the Global Population via impacts of geographical regions, social groups, education, multiple social interactions - especially the role of women, migration, education, birth/death ratio, age structure, economic development... Such a multitude of data is analyzed statistically via models developed in management, econometrics, bio-evolution, or socio-economic sciences. (Ribeiro, 2017; Kendall et al., 2011; Lima & Berryman, 2011; Cecconi et al., 2012; Kac & Lutz, 2017; Vollset et al., 2018; Dias et al. 2018; Herrington, 2020; Bystroff, 2021; van Witteloostuijn et al., 2022; McFarlane, 2023; Raftery & Ševčíková, 2023; Hill et al., 2011; Shumway & Stoffer, 2017; Dai J, Chen, 2019).

2nd Path, is related to creating a convincing model scaling equation, which subsequently is used for portraying the population data in a selected time period. (Malthus, 1798; Stokstad, 2005; Katul, 2013; Verhulst, 1847; Yumei, et al., 2007; Bacaër, 2008; Zhan, 2008; Kapitza, 2010; Lima & Berryman, 2011; Iannelli & Pugliese, 2014; Siegel, 2015; Gonzalo & Alfonsaca, 2016; Flies et al., 2018; Keilman, 2020; Gu et al., 2021; Lehman, 2021; Narayan et al., 2022; Taagapera & Nemčok, 2023; Lima, 2024; Espinosa & Koh, 2024; Lam, 2024; Taagapera & Nemčok, 2024).

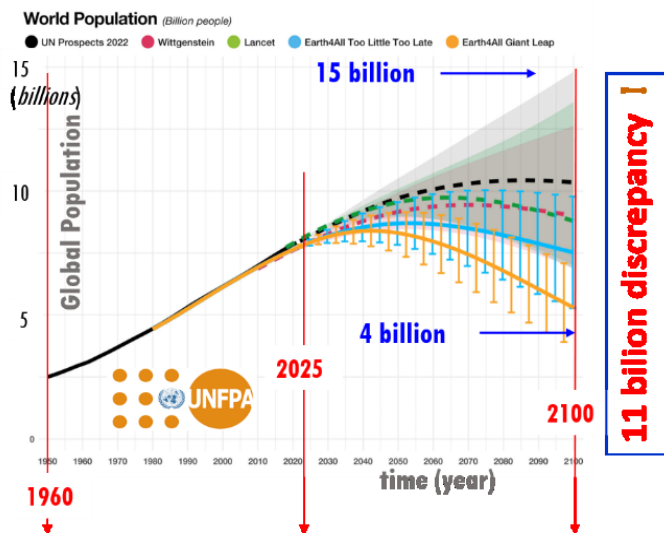


Figure 2. The United Nations Population Fund (UNFPA) standard, with the authors' supplementation, presents forecasts of global population changes until 2100 using **1st Path** modelling. The plot is based on data from 1950 to 2022, and, of course, reproduces the changes well. Then, discrepancies appear, reaching as much as 11 billion in 2100 (UNFPA, 2026).

Figure 2 illustrates the essence of the **1st Path** - related results, via the official UNFPA chart (UNFPA, 2026). Notable is the scatter in forecasts, reaching almost 11 billion (!) for 2100. This puzzling result can be explained by the following issues inherently associated with this cognitive path:

- (i) A multitude of 'empirical' data that have to be collected from many scattered sources. The types of these parameter series chosen often depend on the conceptual preferences of the authors of a given analysis.
- (ii) The meaning and interplay of the above multitude of data series require 'weighting factors', which are necessarily burdened by the arbitrariness of 'experts' opinions. Furthermore, these factors must, or at least should, be considered in the context of the time in which they are introduced, and in light of the often qualitatively different social and cultural conditions across different parts of the globe.
- (iii) The multidimensional, essentially iterative, analyses of global population change are carried out by implementing concepts from various fields of natural and cognitive sciences, with the impact of the model's underlying assumptions.
- (iv) The above points lead to the challenge of reliably determining the uncertainty of the results.
- (v) The required series of precise data are available only after 1950 (UNFPA, 2026; UNESA, 2026).

The **2nd Path** in global population dynamics modelling dates back to the pioneering work of Thomas Malthus. He was inspired by the great legacy of Isaac Newton, who demonstrated in practice the extraordinary power of the Scientific Method and the belief that there are universal laws of nature connecting various, seemingly separate phenomena, which can be represented by universal scaling relations. Notably, to highlight model interpretations, Newton developed and implemented differential calculus. Thomas Malthus, following heuristic considerations, proposed the following model equation to describe population changes (Malthus, 1798; Stokstad, 2005; Katul, 2013):

$$P(t) = P_0 \exp(rt) \Rightarrow \ln P(t) = \ln P_0 + rt \Rightarrow \frac{dP(t)}{dt} = rP(t), \quad (1)$$

where time t refers to the onset time t_0 , coupled to the prefactor P_0 , and $r = \text{const}$ is the Malthus rate coefficient.

The middle part of Eq. (1) defines the Malthus model validation via the linear pattern in the plot $\ln P(t)$ vs. time t . Malthus (1798) supplemented by the heuristic assumption for a much weaker growth of necessary resources, originally food, approximated via a linear dependence (Malthus, 1798):

$$F(t) = a + bt. \quad (2)$$

Probably, this relation resulted from Malthus's current observations that increasing food resources require a proportional rise in the size of cultivated fields. Malthus concluded (Malthus, 1798): *'The population increases in geometrical ratio and the subsistence rises only linearly, which finally leads to times of vice and misery'*. Finally, this is the Malthusian Trap (Catastrophe) (Malthus, 1798; Stokstad, 2005; Katul, 2013), when the quantity of food becomes increasingly insufficient to support the growing population. Malthus advised population self-constraints or an additional rise in subsistence (food) to escape the catastrophic Trap (Malthus, 1798). Unfortunately, the primitive escape concept, such as robbery or conquest of other countries, has often been implemented by rulers or politicians to 'solve the problem'.

In the mid-19th century, Verhulst proposed the next model scaling equation, which inherently includes the impact of resources available in the system (Verhulst, 1847; Pearl & Reed, 1920; Pearl, 1927):

$$\frac{dP(t)}{dt} = rP - sP^2 = P(r - sP) \Rightarrow \frac{dP(t)}{dt} = rP \left(1 - \frac{P}{r/s}\right) = rP \left(1 - \frac{P}{K}\right) \quad (3)$$

$$P(t) = \frac{P_0 \exp(rt)}{1 + (P_0/K)(1 - \exp(rt))} \Rightarrow (s \rightarrow 0, K \rightarrow \infty) \Rightarrow P(t) = P_0 \exp(rt), \quad (4)$$

where r means the Malthus growth rate, s is the resources (originally food) factor, the carrying capacity $K = r/s$, and $C = P_0/(K - P_0)$.

The left-hand part of Eq.(3) is for the original Verhulst notation (Verhulst, 1847), and the right one is for the reformulation by Pearl & Reed (Pearl & Reed, 1920; Pearl, 1927), who introduced the Carrying Capacity factor (K), now one of the most significant concepts in discussions on sustainable development and ecological issues. Following Eq. (3), it indicates the maximal population allowed for sustainable coexistence with the environment in a given system, i.e., $K = P_{\text{max}}^{\text{sustain}}$. The right-hand part of Eq. (3) shows the terminal approximation leading to the basic Malthus dependence.

Notably, when taking into account the Taylor expansion, Eq. (3) can be extended to the following form

$$\frac{dP(t)}{dt} = rP \left(1 - 1 + \frac{P}{K}\right) \approx -rP(t) \ln\left(\frac{P}{K}\right), \quad (5)$$

which correlates with the so-called Gompertz equation (Yumei et al. 2007), also used in population studies.

Worth noting are some problems that can be noted for the **2nd Path** implementations carried out so far.

In general, the basic Malthus, Verhulst, or Gompertz (Yumei et al. 2007) equations do not accurately reproduce global population changes. However, they are significant tools for

describing different population dynamics in biology, medicine, and food technology. When considering the results of such analyses, it is worth noting some general issues. In physics, where nonlinear fitting with scaling functions is an extremely common, even routine, way of data analysis, it is widely assumed that optimal and reliable results require 10x as many experimental points as the number of fitted parameters in a given model equation.

- (i) The Malthus relation contains only 2 free parameters that can be determined by linear regression for a straight line, which, by definition, gives reliable and optimal values, along with errors
- (ii) Verhulst and Gompertz relations for $P(t)$ changes contain 3 free parameters, and here nonlinear fittings are typically
- (iii) Numerous 'extension' models, for which the Verhulst and Gompertz equations are the limiting reference, introduce 4, 5, or more parameters.
- (iv) Especially for the latter two cases, the quality of fit should not be assessed by 'satisfactorily' portraying empirical data or by providing the associated 'favorable' parameters describing the quality of fit (R^2 , χ^2 , ...). The actual errors in the fitted parameters will be extremely unfavorable, even exceeding 50% of their values. This is due to the very flat minima of the individual fits for these parameters – meaning that even a significant change in their values will not affect the 'visual' quality of the fit.

Notably, recently Lehman et al. (2021) considered global population growth using an extended Verhulst model that takes into account three successive bio-ecological levels: (a) interactions with predators, (b) interactions with prey, and (c) intraspecific interactions. Global population changes at each level are governed by level-dependent ecological coefficients (r_i ; s_i), $i = 1; 2; 3$. These were implemented for the following extended Verhulst-type equations (Lehman et al., 2021):

$$G_P = \frac{1}{P(t)} \frac{\Delta P(t)}{\Delta t} = r_i \pm s_i P(t) \quad \Rightarrow \quad P(t) = \sum \frac{1}{(P_0 \mp (s_i/r_i) e^{-r_i t} \pm (s_i/r_i))} \quad (6)$$

where coefficients $r_i, s_i = \text{const}$, for subsequent time domains differently subjected to time-varying bio-/eco- factors. G_P denotes the Relative Growth Rate (RGR), in a commonly used discrete form,

For the empirical validation of the multiparameter Verhulst approach Lehman et al. showed the linear behavior of $G_P(P_i)$ vs. $P_i(t_i)$, in two domains, namely from 10 000 BC to ~1962 and next from 1869 to 2019, using 98 population data coveting this Anthropocene period. The mentioned crossover was associated with $P(t_{\text{cross}}) \approx 3.14 \pm 0.2$ billion (Lehman et al., 2021). However, the enormous time span of 12 millennia, combined with the colossal population change, results in a very 'squeezed' presentation at terminals. The latter can bias the RGR validation tests, as shown very recently in ref. (Sojecka & Drozd-Rzoska, 2025).

In 1960, von Foerster et al. (1960) suggested a possible preference for a qualitatively different scaling function than previously discussed. It was later referred to as 'Hyperbolic' or 'Doomsday' scaling. Global population data, covering a period of 1500 years from 400 CE to 1958, were scaled by the following function (von Foerster et al., 1960):

$$P(t) = \frac{A}{(D-t)^M} \approx \frac{A}{D-t} \quad \Rightarrow \quad \log_{10} P(t) = \log_{10} C - M \log(D-t) \quad (7)$$

where $C = (1.79 \pm 0.14) \times 10^{11}$ (years), the exponent $M = 0.99 \pm 0.01$, $D = 2026.87 \pm 5.5$ (years) which was linked to the fate 'Doomsday' on Friday, 13th Nov., 2026 in the title of (von Foerster et al., 2026).

The right-hand part of Eq. (7) defines the log-log scale analysis, with an adjustable 'Doomsday' parameter D , until reaching the linear pattern used in (von Foerster et al., 1960). This result is recalled in **Figure 3**.

Nonetheless, von Foerster et al., (1960) met extensive criticism soon after the publication. However, the objections were rather general and speculative, primarily concerning the 'strange' Doomsday singularity predicted for late 2026, noted as 'artificial' and without any real meaning. The next ones were related to no underlying model (Shinbrot, 1961; Roberstson et al, 1961; Hutton, et al, 1961; Howland, 1961; Serrina, 1975; von Hoerner, 1975;

Smith, 1977; Podum E, Whither, 1980; Umpleby, 1987; Deevay, 1988; Frasere-Smith, 1989; Kremer, 1990; Dochy, 1995; Hough, 1996; Varfolomeyev et al, 2001; Umpleby, 2005; MacIntyre, 2005; Dolgonosov & Naidenov, 2006). Till the mid-1970s, as more historical estimates of the global population have become available, evidence showing different values for the exponent in the 'Doomsday' Eq. (2), $0.7 \leq M \leq 1$. Moreover, it was noted that parameters obtained in scaling via Eq. (7) depended on the selected time domain (Kremer, 1990; Dolgonosov & Naidenov, 2006).

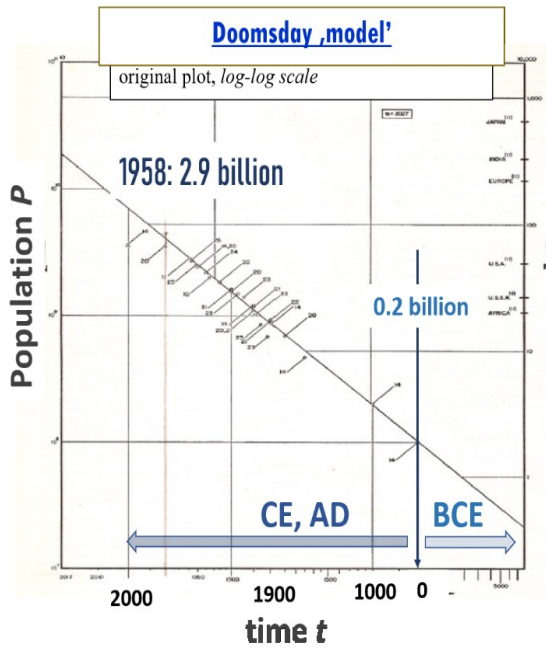


Figure 3. The original plot by von Foerster et al. showing global population changes from over 1500 years, to 1958 via the log – log scale analysis validating the ‘hyperbolic’ scaling, as shown in Eq. (7) (von Foerster et al., 1960). The plot includes supplements by the authors of the given report to facilitate reading.

The analysis by (von Foerster et al., 1960) used the $\log_{10}P(t)$ vs $\log(D - t)$ plot, with adjustable D value until obtaining the linear behaviour. However, the huge distance between the terminal of the tested empirical data and the singularity means that even relatively large changes in the value of D are poorly detectable. This problem was explicitly demonstrated in studies of the pseudospinodal behavior in near-critical liquids (Chrapeć et al, 1987).

When discussing the above scaling concept, worth recalling is the model picture by Taagapera and Nemčok (Taagapera & Nemčok, 2023; Taagapera & Nemčok, 2024), who focused on a scaling equation that correlates with the ‘Doomsday’ scaling in a ‘remote, tail’ domain. It led to the ‘tamed quasi-hyperbolic function’, or ‘T-function’ (Taagepera, 1977; Taagapera & Nemčok, 2023; Taagapera & Nemčok, 2024):

$$P(t) = \frac{A}{[\ln(B+E)]^M} \quad , \quad \text{where } E = \exp[(D - t)/\tau] \quad . \quad (8)$$

They showed a fair portrayal for two subsequent periods (1) from well below 10 000 BCE to 400 CE and (2) from 400 CE to 2022. Both domains terminated with a plateau, namely during Roman Empire times and in the last decades, up to 2022, which might suggest universalistic population terminals and the split into two leading (civilizational?) periods in the Anthropocene. A worth stressing result of refs. (Taagapera & Nemčok, 2023; Taagapera & Nemčok, 2024) is the presentation of ~ 600 collected global population data, showing significant discrepancies in estimates, which increase when shifting back in time. However, it is notable that Eq. (8) contains 5 nonlinearly fitted parameters, which makes ultimate validation difficult.

3. Global Population: Physics & Active Soft Matter Perspective

To date, studies of global population change have paid (very) limited attention to the patterns offered by the dynamics of complex systems in physics, despite their explicit, fundamental interpretative basis, which is often empirically tested and validated. Only recently, the authors of this report followed this **3rd Path** for global population growth cognition (Sojecka Drozd-Rzoska & Rzoska, 2025; Sojecka & Drozd-Rzoska, 2025; Sojecka & Drozd-Rzoska, 2024; Sojecka & Drozd-Rzoska (Proc. Soc. Lect.), 2025; Sojecka & Drozd-Rzoska, 2023; Sojecka & Drozd-Rzoska, 2026). These issues are presented in detail in the next section, alongside new and previously unpublished analyses. Dynamical processes involving exponential changes, i.e., resembling the Malthus equation, are relatively common in physics. An example is the dynamics of a nuclear chain reaction, which will be discussed in reference to the basic 'standard' population scaling patterns in the next section. More complex dynamics emerging in the global population are discussed within the framework of complex Soft Matter dynamics in subsequent sections.

3.1 Malthus scaling, Verhulst scaling, and the nuclear chain reaction parallel

In this section, we would like to draw your attention to an almost direct parallel with a process that is well known, at least by name: the nuclear chain reaction. However, neither Malthus's nor Verhulst's equations have been invoked there, so far. The nuclear chain reaction is related to a cascading increase in neutron emission, namely (Cameron, 2011):

$$\frac{dN(t)}{dt} = \left(\frac{k_{\text{eff}} - 1}{l} \right) N(t) \quad \Rightarrow \quad \frac{dP(t)}{dt} = rP(t) \quad (9)$$

where the left side is for the neutron population and the right side evokes Malthus's relation for the populations of humans, animals, bacteria, or yeast. The effective neutron multiplication factor is defined on the left side of the relation below (Cameron, 2011):

$$k_{\text{eff}} = \frac{\text{rate of neutrons production}}{\text{rate of neutrons loss}} \quad \Rightarrow \quad k_{\text{eff}}^{\text{popul.}} = \frac{\text{birth rate}}{\text{death rate}} \quad (10)$$

The right-hand side is for the possible human/species population changes.

In a nuclear chain reaction, the prompt-neutron lifetime refers to the time it takes for a neutron to undergo fission. For human populations, the parameter l can also be associated with the life-time, most often too prompt, unfortunately. Following the above, one obtains for the meaning of the Malthus growth rate:

$$r = \frac{k_{\text{eff}}^{\text{popul.}} - 1}{l} \quad \Rightarrow \quad \tau = \frac{1}{r} = \frac{l}{k_{\text{eff}}^{\text{popul.}} - 1} \quad \Rightarrow \quad P(t) = P_0 \exp\left(\frac{t}{\tau}\right) \quad (11)$$

where the right-hand part shows the formulation of the Malthus relation using the relaxation time τ , as proposed in ref. (Sojecka & Drozd-Rzoska, 2024) Such notation, commonly used in physics, enables a simple estimation of the time required to 50% change in population: $t_{50\%} = \tau \times \ln 2$.

For the nuclear chain reaction, three main domains for the effective neutron multiplication factor, which can be directly implemented in the Malthus model population analysis (Cameron, 2011):

- (i) $k_{\text{eff}}, k_{\text{eff}}^{\text{popul.}} < 1$ ('subcriticality'): the population is decreasing exponentially
- (ii) $k_{\text{eff}}, k_{\text{eff}}^{\text{popul.}} = 1$ ('criticality'): the population is maintaining a constant value
- (iii) $k_{\text{eff}}, k_{\text{eff}}^{\text{popul.}} > 1$ ('supercriticality'): the population is increasing exponentially

In a practical nuclear system, like a fission reactor, k_{eff} oscillate from slightly less than 1 to slightly more than 1. When permanently $k_{\text{eff}} > 1$ or even $k_{\text{eff}} \gg 1$, there is a danger of a nuclear explosion, with gigantic destructive force: 'Atomic Bomb' explosion.

The latter could be a fundamental justification for the concept of the 'Demographic Bomb', often used in global population studies, in connection with possible values $k_{\text{eff}}^{\text{popul.}} \gg 1$. For nuclear power plant operation, it is essential to maintain a constant, near-critical electron emission rate without actually reaching a critical or supercritical k_{eff} value, which could lead to an uncontrolled nuclear reaction. This is achieved by immersing the fuel rods in a suitable medium, such as heavy water (D₂O), which slows down neutrons and limits the nuclear chain reaction to a predetermined level. This process is described by the neutron population equation (NPE) (Cameron, 2011):

$$\frac{dN(t)}{dt} = \left(\frac{k_{\text{eff}} - 1}{l} \right) N(t) + S(t) = k_{\text{Neu}} N(t) + S(t) , \quad (12)$$

where $S(t)$ describes the influence of the medium in which the fuel rods are immersed. until reaching a state where $dN(t)/dt \rightarrow 0$, when the appropriate neutron population stops growing, but their stable 'effectively subcritical' emission ensures constant production of thermal and further electrical energy.

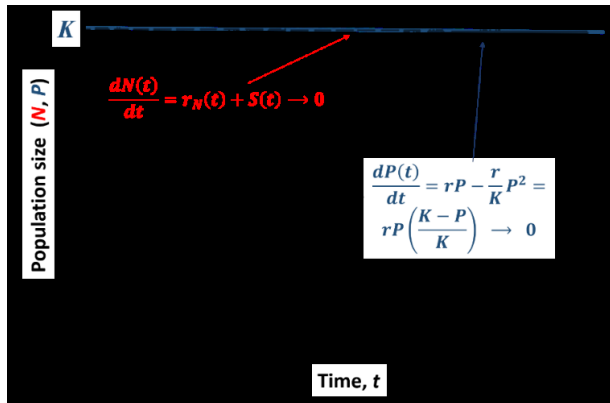


Figure 4. Bimodal pattern, consisting of the rising and stationary parts, with recalling the neutron population equation for: (1) power plants (in red) and (2) the Verhulst equation for human or any other bio-population (in blue). The latter includes the Carrying Capacity factor (K), also indicated in the plot. Arrows indicate the final, stationary population value.

Both in the NPE Eq. (12) and in the Verhulst Eq. (3), a supplementary term describing 'environment' features significant for the given population is included. Generally, $S(t)$ factor is specified with respect to the nuclear reactor implementation. On the other hand, the Verhulst scaling equation, associated with human or bio-populations, assumes a simple pattern for such a term: $S(t) = (r/K)P^2$, showing explicitly the dependence from the population $P(t)$ size, growth rate and maximal, final and sustainable population. For the Verhulst Eq. (3), the bimodal pattern shown in **Figure 4** is a standard for bacterial populations in a closed container, where the essential feature of the environment is that the food resource is constantly replenished to a constant level despite its consumption by the growing population. For a nuclear reactor, the meaning of 'resource', in the given case the moderated medium, is different, namely limiting quantitative changes in the 'electron population' to a critical level (the A-case above), essential for reaching the second 'stationary' state in **Figure 4**. This may indicate the Verhulst model's usefulness in nuclear reactor modelling. The authors believe that the direct implementation of the Carrying Capacity (K - factor) concept as a universal metric for environmental capacity relative to the needs/requirements of a given population may be particularly valuable here.

3.2 Human Active Intelligent Soft Matter Insight into Global Population Dynamics

In 1991, Pierre G. de Gennes presented the Nobel Prize Lecture entitled (de Gennes & Badoz, 1996): 'Soft Matter'. It introduced the novel category of systems, linking: liquid crystals, polymers, blends, gels, foams, supercooled glass-formers, colloids, critical liquids ... and bio-systems. Soft Matter complex systems exhibit a few common features:

- (i) dominance of mesoscale species leading to 'universal' scaling patterns of 'isomorphic' physical properties
- (ii) ability to spontaneously self-assemble
- (iii) extreme sensitivity to Exo- & Endogenic impacts, linked to weak interactions between mesoscale species

Jones (2002), in his classic monograph indicated four conceptual and cognitive 'pillars' that are essential for interpretation in any Soft Matter framework: (i) Liquid Crystals Physics, (ii) Glass Transition Physics, (iii) Polymers Physics, and (iv) Critical Phenomena Physics. In subsequent years, the Soft Matter category entered other branches of Science. One can recall Quantum Soft Matter (Thedford, 2023), Cosmological Soft Matter (Saridakis, 2021), Topological Soft Matter (Serra et al, 2020), and, perhaps most important and rapidly developing, Active Soft Matter (ASM) (Menzel, 2015; Das et al, 2020; Janzen, et al, 2026) for

complex systems composed of interacting elements that can individually consume energy and then collectively generate motion or mechanical stress.

ASM links Soft Matter physics, biology, and medicine (Tang, 2022; Gonzalez-Rodriguez, et al., 2012) to understand the unique patterns that emerge in systems such as viral assemblies, bacterial colonies (Sawada, 2017), tissue formation (Gonzalez-Rodriguez, et al., 2012) gene expression (Semerano, et al., 2025). It also explains collective motion observed in bird flocks (Cavagna, et al., 2015), schools of fish (Huang et al., 2024), bee swarms (Shpurov & Froese, 2022), and anthills (Hu, et al., 2016). Very recently, a new subcategory of Intelligent Soft Matter (ISM), that links Active Soft Matter Physics, Biology, & Cognitive Sciences, has emerged (Baulin, et al., 2025; Wrugt, 2026). This report focuses on the proposal for a new category, Human Active Intelligent Soft Matter (HAI SM). This report explores this concept to gain new insight into global population dynamics since the onset of the Anthropocene. Notable are the model features of HAI SM:

- (i) An extraordinary range of adaptation to a 'system' despite environmental changes, food resources, threats, etc...
- (ii) Creation of 'innovations', their memorization, improvement, and transfer to the next generations, supporting adaptation to the system, and even shaping it
- (iii) The ability to 'hierarchical assembling' targeting on better and more efficient acquisition of food and raw materials, but also on activities related to other 'hierarchically assembled' species, focused on defensive purposes and/or aggressive super-predator activities
- (iv) Multidimensional interactions – 'biological', 'ecological' with the environment, but also social. Significant is the rapid development of language, or the development of seemingly 'impractical' actions, indirectly strengthening communities (rituals, religion, art, etc.)

However, before proceeding to the implementation discussion, the authors would like to first draw attention to a specific drawback and limitation in the empirical validation of model scaling relations – the scatter of global population data, illustrated in refs. (Taagapera & Nemčok, 2023; Taagapera & Nemčok, 2024). It means their non-analytic nature, which excludes all methods related to numerical differentiation of empirical data, being the essential path for 'subtle', ultimate, validation tests.

The authors of this paper implemented the following protocol to address this essential challenge (Sojecka & Drozd-Rzoska, 2024; Sojecka & Drozd-Rzoska, 2025):

1. The first step focuses on the empirical data itself, which: (i) are collected from different sources, which gives them a large scatter – a big problem so far, (ii) the data are then subjected to numerical filtering (using the Savitzky-Golay principle) – until an 'analytical' set is obtained, which can be directly differentiated
2. The next focus on scaling equations with no more than 3 adjustable parameters, & used in testing dynamics of Soft Matter systems (and not for Global Population studies, so far)
3. Instead of direct fitting, the adequacy of a given scaling relation for describing a given set of 'empirical' data is tested, using the distortion-sensitive & derivative-based analysis
4. The analysis shows time domains of given relation validity and determines optimal values of parameters, and subsequently inserted into the scaling relations for portraying $P(t)$ data.

Figure 5 illustrates changes in the global population, from the Anthropocene onset (10,000 BC) to 2025 (i.e., over 12 millennia), using the dataset prepared following the above protocol. For the first nine millennia, **Figure 5** indicates a simple Malthusian pattern split into two periods corresponding to the Early and Late Neolithic epochs. Notable is the coincidence of the crossover (~5,000 BC) with the early post-final flooding of Doggerland remains. It was the landmass that connected Britain with Scandinavia, yet around 8,000 BC. Nowadays, this region is known as the North Sea, with an average depth of 80 - 90 meters (Stutz, 2026).

From around 1000 BC onwards, the dynamics of the global population, as shown in **Figure 5**, appear to be non-linear and non-Malthusian. However, two specific periods emerge, marked by red horizontal lines, during which the global population barely changed: the

Roman Empire period in late antiquity and the period from late Medieval to th Renaissance, which coincides with the times of Black Death pandemic impact.

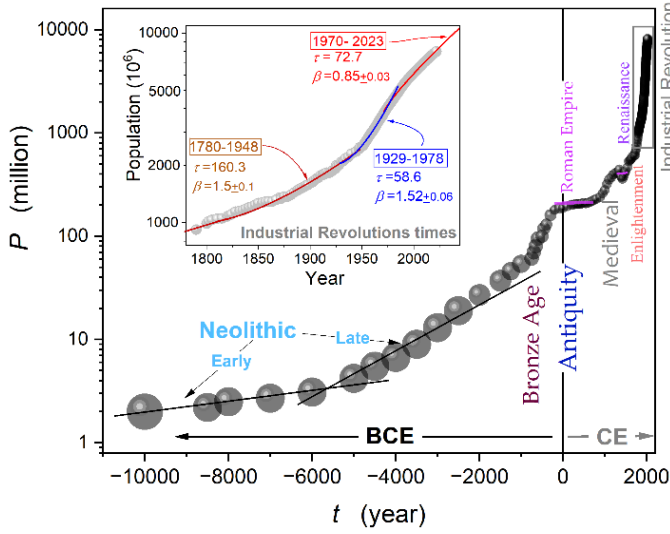


Figure 5. Global population change (200 population data points) obtained through numerical filtering of data from refs. (Sojecka AA, Drozd-Rzoska, 2024; Sojecka AA, Drozd-Rzoska, 2024; Drozd-Rzoska & Sojecka, 2026) in the semi-log scale, for which the ‘simple’ description of Malthus Eq. (1) manifests itself as a domain of linear changes. The inset shows behavior related to the Industrial Revolution period since 1800, parameterized via the empowered exponential relation, the Super Malthus Eq. (14), described above. Some significant periods and dates are indicated.

4. Super Malthusian scaling of Global Population changes

The development of an analytical dataset for the global population changes, by the authors of the given report, led to the proposal of the Analytic relative growth rate (RGR) factor (Sojecka & Drozd-Rzoska, 2025):

$$\begin{aligned} \left[G_P = \frac{1}{P(t)} \frac{\Delta P(t)}{\Delta t} \right]_{\text{discrete}} &\Rightarrow G_P(P, T) = \left[\frac{1}{P(t)} \frac{dP(t)}{dt} \right]_{\text{analytic}} \Rightarrow \\ \Rightarrow G_P(P, T) &= \frac{1}{P(t)} \frac{dP(t)}{dt} = \frac{d \ln P(t)}{dt} \end{aligned} \quad (13)$$

The analytic RGR, describing relative population changes $dP(t)/dt$ along the timeline, has been implemented as a universal testing tool for detecting subtle features of population changes, hidden in the standard $P(t)$ plot view.

Figure 5 shows that, for much of the Anthropocene, global population dynamics is non-Malthusian, i.e., they exceed the simple exponential (‘geometric’) growth defined in Eq. (1). One of the leading methods for scaling this type of empirical data in physics is the empowered exponential function. The authors of this paper implemented such an approach in ref. (Sojecka AA, Drozd-Rzoska, 2024), naming the new scaling function as Super Malthusian (SM):

$$P(t) = P_0 \exp \left(\frac{t}{\tau} \right)^\beta \Rightarrow G_P = \frac{d \ln P(t)}{dt} = \frac{\beta}{\tau} t^{\beta-1} = h(t) \Rightarrow \log_{10} G_P = A + Bx, \quad (14)$$

where $x = \log_{10} t$, $A, B = \text{const}$ and $A = \beta / \tau^\beta$, $B = \beta - 1$, and $\tau = 1/r$.

The left-hand side of Eq. (14) defines the SM dependence, and the right-hand side presents the proposed RGR-based linearized distortion-sensitive analysis, to test their validity. Namely, for the plot $\log_{10} G_P$ vs. $\log_{10} t$ linear behavior indicates domains where SM scaling is allowed. Next, linear regression can yield optimal estimates of the relevant parameters with well-defined errors.

The fundamental Soft Matter Physics interpretations of the empowered exponential (Super-Malthus) Eq. (14) (left side) can be concluded as follows:

- (i) For the Kohlrausch-Williams-Watts (KWW) model, the exponent $\beta = 1$ is for systems governed by a single relaxation time & $\beta \neq 1$ is for a distribution of relaxation times (Rzoska & Mazur, 2007).
- (ii) The most common dynamic associated with $\beta < 1$ is named the stretched exponential, and the case $\beta > 1$ is named the compressed exponential, and it has been only recently evidenced in some physical/material systems (Rzoska et al., 1994; Rzoska et al., 1997; Rzoska & Drozd-Rzoska, 2011; Drozd-Rzoska et al., 2023; Drozd-Rzoska, 2019; Martinez-Garcia, 2013; Wu, 2018; Trachenko, 2021).
- (iii) The dynamics governed by for $\beta < 1$ suggest energy loss and $\beta > 1$ energy gain in a system (Rzoska & Mazur, 2007).
- (iv) One can also recall the nucleation theory by Avrami, which yields $\beta = D + 1$, where D is for dominant (local) assemblings dimensionality (often fractal) (Rzoska & Mazur, 2007).

The latter, for the human population, can be linked to the emergence of villages, cities, states... The function $h(t)$ in Eq. (14) recalls the Weibull hazard function (Lai, et al, 2006). The Weibull distribution is used extensively in reliability applications to model failure times in complex systems. It is significant for testing failure rates or asset volatility in complex systems with random parameters across various fields, including survival analysis and failure rate modelling. It includes engineering applications for predicting component failure, weather forecasting, and challenging issues in medicine. The hazard function $h(t)$ is telling how likely something is to fail given that it has survived so far. Namely: (i) $\beta < 1$ suggests even likely to fall fast, (ii) $\beta = 1$ – failure rate is fairly constant, and (iii) $\beta > 1$ indicates increasing failure rate as time goes by. (Lai, et al., 2006).

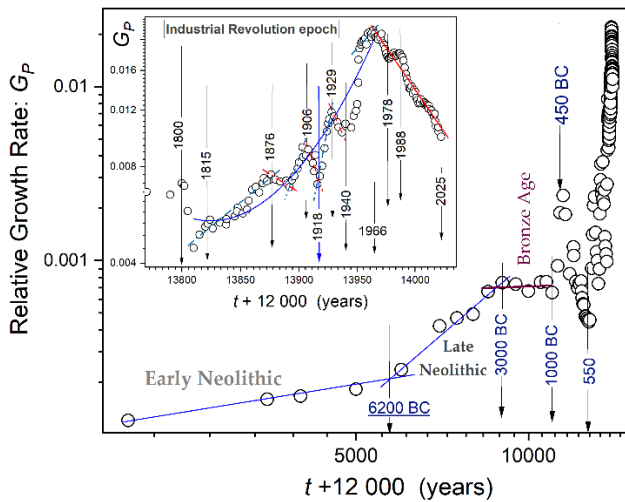


Figure 6. The Relative Growth Rate changes, from the Anthropocene dawn (12 000 BC) till nowadays, in the log-log scale as defined in Eq. (14). The onset present the behaviour focused on industrial Revolutions times, Continuous curves/lines indicate dominant trends and dashed one are for local trends revealed due to the distortions sensitive analysis. The blue and light-blue colours are for the exponent $\beta > 1$ in the Super-Malthus Eq. (14, left part) and the red is for $\beta < 1$. Horizontal lines reflect the basic Malthus pattern related to Eq. (1) or Eq. (14) with the exponent $\beta = 1$.

Figure 6 presents the evolution of the RGR factor on a log-log scale, following the data transformation defined by the right-hand side of Eq. (14), with the slopes of lines directly determining the exponent β . The ‘horizontal’ behavior is for the basic Malthus pattern (Eq. (1)). It appears in Bronze Age times, for instance. The plot reveals that for Neolithic times, the linear, Malthusian pattern in the direct presentation of $P(t)$ changes has only an ‘apparent’ meaning. The distortions-sensitive, RGR-related analysis in **Figure 6** shows that it is better described by the SM Eq. (14), within the slightly compressed pattern ($\beta > 1$). Notable is the shift of the crossover between Early and Late Neolithic times to $t \sim 6200$ BC, which surprisingly well correlates with the sinking’ time of the last Doggerland land remnants.

Notably, Bronze Age-related Malthusian behavior fairly coincides with its historical borders (3,000–1,000 BC). This period began with the rise of ancient Sumer in Mesopotamia

and Egypt along the Nile, and ended with the sudden collapse of grand Bronze Age civilizations around 1200–1100 BC. (Glass, 2021).

The times of the Roman Empire (~100 BC – 500 CE) are marked by a pronounced collapse of $G_P(t)$ changes, associated with a remarkable stabilization of the global population $P(t)$ at the level of 190-200 millions for over ~600 years (Garnsey, 2014). This is a unique situation in global population history. A similar disruption, for almost 200 years from ~1350 to ~1540, can be linked to the impact of Black Death pandemic in Asia and Europe (Wickham, 2016; Douglas, 2022).

In searching for RGR collapse during Roman Empire times, the effects of prolonged major wars, the spread of subsequent pandemics through developed communication networks, and significant urbanization can be considered. One can even consider the health effects of lead water pipes. Recently, Sojecka & Drozd (2024) have suggested yet another possible reason for this extraordinary global population collapse: the extraordinary scale of exploitation of "slave work-force" — on a scale we would today describe as "industrial." An example of this deadly exploitation of slaves can be Rio Tinto silver mines in present-day Spain. There was a constant supply of thousands of slaves, who survived for only a few months (Garnsey, 2014). By today's standards, this would be considered organized mass genocide. There were many such places in the Empire, also associated with impressive buildings, which still remains impressive. Notably, at its peak, up to 1/3 of the global population lived in the Roman Empire.

This line of reasoning also raises the question of whether the decline of the Roman Empire was driven by an economic crisis resulting from a shrinking 'barbarian slave workforce' and fewer opportunities to wage victorious wars on its borders.

The inset in **Figure 6** shows a log-log plot of the RGR factor during the ongoing Industrial Revolution era. The global population fluctuations indicated by the arrows in **Figure 6** can be easily correlated with significant historical events, as noted in **Table 1**.

Table 1. Historical events correlated with global population distortions detected in the inset in **Figure 6**.

Year	Related historical facts	Year	Related historical facts
1800	Napoleon Bonaparte terminated the post-Revolution disorder	1940	In years (1939-1941), World War II (WWII) spread across the globe
1815	The Vienna Congress established a 'new order' for the next century: the 'concert of powers'	1950	The end of post-WWII recovery in Europe, and the onset of a new path of development
1876	The terminal of post-Civil War recovery in the USA - the beginning of the global imperial path; the rise of the German Empire as a new economic and political power	1966	The years 1966–1970 were marked by great social upheaval, leading to the transformations that would transform society in the following decades. Also, the 'main wave' of decolonization
1906	Political unrest leading to a modest democratization of the post-feudal Empires, particularly disruptive for Russia	1978	The growing economic crises that led to the presidency and reforms of Ronald Reagan in the USA and Margaret Thatcher in the UK
1918	The end 1 st World War	1988	The communist system collapsed: free elections in Poland (1989), reunification of Germany (1990)
1929	The onset of the Great Economic Crisis: Black Friday		

Drozd-Rzoska & Sojecka (2024) also proposed yet another Super-Malthus equation (named type 2), with the time-dependent relaxation time or alternatively the growth rate:

$$P(t) = P_0 \exp(t/\tau(t)) \quad \Rightarrow \quad \tau(t) = t \ln(P_0/P(t)), \quad (15)$$

where the time-dependent relaxation time can be linked to the time-dependent growth rate as follows: $\tau(t) = 1/r(t)$.

The basic problem in applying the above equation for portraying empirical data is the unknown a priori form $\tau(t)$ evolution. However, this problem can be overcome by determining the changes in the relaxation time via the dependence indicated on the right-hand side of Eq. (15).

Figure 7 shows such a result for times after the collapse of the Roman Empire. The analysis reveals two surprising results. Firstly, a dominant basic Malthusian scaling pattern emerges (see Eq. (1)) from the High Middle Ages (~1100) to the onset of the Enlightenment

or the Industrial Revolution (~1700) appears, but with a significant disturbance during Black Death impact times.

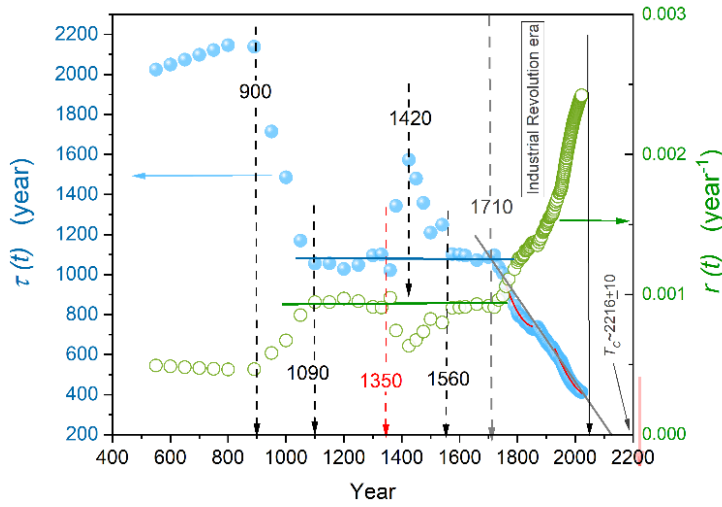


Figure 7. Temporal changes of the relaxation time and the growth rate, $\tau(t) = 1/r(t)$, from the collapse of the Roman Empire to the present day. The dashed vertical arrows indicate the onset of characteristic changes in the plot, with the red arrow corresponding to the onset of the Black Death pandemic. Horizontal lines show the behaviour associated with the dominant trend $\tau(t) = \tau = \text{const}$, related to the basic Malthus behaviour (Eq. (1)), indicated via green and blue horizontal lines. The sloped, (grey) line indicates the dominant linear trend $\tau(t) = a - bt$ in Industrial Revolutions era. Notably, there are two distortions, portrayed in red via the second-order polynomial: (1) for years 1970 -2025, and (2) a notably stronger one in years 1780-1860.

In Industrial Revolution time the exceptional linear pattern emerges: $\tau(t) = a - bt$. In the last few decades. This pattern is disturbed, in a way suggesting 'slowing down in the last few decades. It is indicated by the fitted 'red curve' in **Figure 7**. However, such behavior– in fact, even stronger – also occurred from ~1780 to ~1860, and subsequently the dominant linear trend returned.

Considering the mentioned linear trend as the dominant pattern, one can substitute the related dependence to Eq. (15) (Drozd-Rzoska & Sojecka, 2024):

$$P(t) = P_0 \exp\left(\frac{t}{\tau(t)}\right) = P_0 \exp\left(\frac{t}{a-bt}\right) = P_0 \exp\left(\frac{t/b}{a/b-t}\right) = P_0 \exp\left(\frac{b' \times t}{t_c - t}\right), \quad (16)$$

where $b' = 1/b \approx 1.62$, $t_c = a/b = 2216 \pm 10$ (year).

Notably, the above relation resembles the constrained critical behavior within the so-called Griffiths model (Drozd-Rzoska & Sojecka, 2024; Amaral & de Oliveira, 2021).

5. Global population – Critical phenomena approach

The second type of scaling relation, which is particularly characteristic, can be associated with the pioneering work of von Foerster et al. (1960), which suggested a hyperbolic scaling relation that was extremely simple yet surprisingly effective. This relation is also known as the 'Doomsday' equation, which predicted an infinite population by late 2026. This was considered a paradoxical result for a random and meaningless date, as seen in the sixties and seventies. Notwithstanding, von Foerster et al. (1960) result inspired further research, although the idea of an explicit (simple) 'hyperbolic' scaling was rejected. Below, we demonstrate that despite these objections, it is the real dominant trend, but only during the Industrial Revolution era. One can consider von Foerster scaling within the framework of Critical Phenomena Physics (Stanley, 1989).

$$P(t) = \frac{C}{|t_c - t|^M} \Rightarrow \ln P(t) = \ln C - M \ln(D - t), \quad (17)$$

where the critical year t_c can coincide with von Foerster et al. (1960) 'Doomsday' D . Generally, on approaching the critical singularity the exceptional near-critical (super-critical) state emerges. It is governed by rising multi-element fluctuations, causing the

system to become extremely sensitive to any perturbation. The right-hand part of Eq. (17) recalls the quasi-nonlinear analysis, which employs the log-log plot with an adjustable parameter D , until linear behavior emerges. In fact, it was used by von Foerster et al. (1960). Unfortunately, this type of analysis, as well as the explicitly nonlinear fit, leads to a large uncertainty in the value of the singularity D , also influencing the exponent M . This is due to the significant distance between the singularity and the empirical data terminal. In ref. (von Foerster et al., 1960), they are $D = 2026$ and $t_{\text{terminal}} = 1958$. This also constitutes an unsolved challenge for the analysis of empirical data in physics. One can recall the pseudospinodal problem (Chrapeć, 1987), or dynamics in the high-temperature region of glass-forming systems (Mallamace et al., 2010; Drozd-Rzoska & Rzoska, Starzonek, 2023), for example.

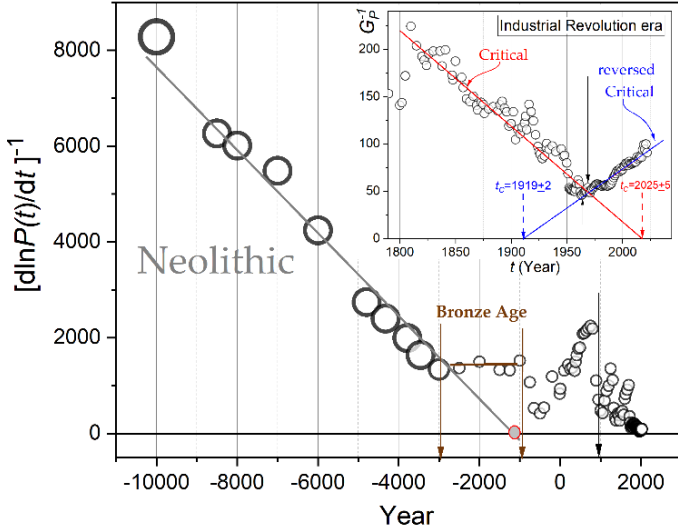


Figure 8. Temporal changes of the RGR factor in Anthropocene, focused on the distortions-sensitive test of the critical-like Eq. (18), with population data transformation defined by Eq. (19). The sloped linear behavior indicates domains where the critical-like Eq. (18) offers a fair scaling. The horizontal line indicates the basic Malthus scaling domain. The inset shows the focused behavior for the Industrial Revolution era. Dashed arrows and the red-border circle indicate singularities t_c for Eq. (18) scaling relation.

The authors of this paper proposed a solution to this problem, namely, recalling Eq. (17) (Drozd-Rzoska & Sojecka, 2026):

$$\ln P(t) = \ln C - M \ln |t_c - t| \Rightarrow \frac{d \ln P(t)}{dt} = \frac{-M}{D - t} \Rightarrow G_P^{-1}(t) = \left[\frac{d \ln P(t)}{dt} \right]^{-1} \Rightarrow$$

$$\Rightarrow G_P^{-1}(t) = -M^{-1} t_c - M^{-1} t = d - mt. \quad (18)$$

The implementation of $P(t)$ data transformation defined by Eq. (19) is shown in **Figure 8**. The most striking behavior appears for the Industrial Revolution era, shown in the inset. It reveals the following exceptional sequence (Drozd-Rzoska & Sojecka, 2026):

$$\sim 1800 \rightarrow (D_1 - t)^{M=-1} \Rightarrow \sim 1968 \Rightarrow [(t - D_2)^{M=-1}]^{-1} = (t - D_2)^{M=1.1}, \quad (19)$$

where $D_1 \sim 2025$, and $D_2 \sim 1919$, for two detected critical ($t_c = D$) singularities. It means the following pattern (Drozd-Rzoska & Sojecka, 2026):

$$\sim 1800 \rightarrow \text{critical behavior} \Rightarrow \sim 1968(\pm 4) \Rightarrow \text{reversed critical behavior} \quad (20)$$

The first 'critical' domain is associated with the singularity at $D \sim 2025$ and the exponent $M = 1$, which fairly correlates with von Foerster et al. (1960) 'Hyperbolic, Doomsday' pattern, although they overestimated it for a 1500-year period, from 400 CE (von Foerster et al., 1960). The **inset in Figure 8** validates such scaling, but for a shorter period. After the crossover at ~ 1968 , a 'tunnelling' to a 'tail' of a reversed critical behavior hidden in the past

(~1919) occurs. Notably, the ‘reversed’ critical pattern causes the population to continue rising, but at a decreasing growth rate.

The main part of the plot shows that the critical-like scaling (Eq. (18)) offer also a fair description for 7 000 years of the Neolithic era., with singularity at $D = t_c \approx 1200BC$, related to $G_P^{-1}(t_c) = 0$. This is a surprising and precise correlation with the collapse and rapid integration of the great Bronze Age civilisations and empires in the Near East and Mediterranean regions. At the same time, the decline of the classical era of ancient Egyptian civilization began after the death of Pharaoh Ramesses II and his state-exhausting wars. The Bronze Era collapse is linked to climate change, geostorms, and, above all, the invasions of the so-called Sea Peoples (Glass, 2021). But perhaps the inhabitants of the Bronze Age empires (c. 3000–1000 BC) also felt enslaved by the increasing demands and burdens of imperial authorities, and took advantage of their weakening to retreat to the wider world beyond, where such oppressive power did not exist? Or perhaps they joined the Sea Peoples warriors? A similar explanation has been put forward two Millennia later, for the rapid decline of the Maya civilization on the Yucatán Peninsula in present-day Mexico and Guatemala (Turner & Sabloff, 2012). However, a surprising systemic determinism emerges, for inevitable change in the pattern for the global population

In this context, it is worth taking a critical look at the peculiarities revealed in **the inset of Figure 8**. As for $D_1 \approx 1919$, it is a fair coincidence with the end of World War I, which wiped out the great empires of Russia, Austria-Hungary, and Germany—dominant in Europe since the Congress of Vienna (1815)—from the map of Europe. The post-feudal order lost its significance. It was also during this period that the United States unequivocally demonstrated its global power (Lowe, 2015). When von Foerster et al.’s paper (1960) was published in 1960, it was not the usual Doomsday singularity, at $D_1 \approx 2026$. At the time, it seemed like an odd, random date without any particular significance. Now, however, we recognize that it has civilizational significance, as a New World Order is emerging, amid current world-impacted Wars.

6. Discussion: Avoided Criticality and the Carrying Capacity

The inset in **Figure 6**, shows that from 1800 till ~1966/8 the dominant trend can be approximated by a next empirical exponential equation (Drozd-Rzoska & Sojecka, 2026):

$$P(t) = P_0 \exp\left(\left(\frac{t}{\tau}\right)^{\beta(t)}\right), \quad (20)$$

with the exponent changing from $\beta \sim 1$ to $\beta \sim 4$, which can be named as Hyper-Malthusian due to unique $\beta(t)$ changes.

The mentioned trend is ‘disturbed in the post-WWII period’, from ~1950 to ~1968, when the exponent $\beta \approx 1.5$ (Sojecka AA, Drozd-Rzoska, 2024). It can be associated with the post-WWII qualitative reset in the economy, social circumstances, and culture. The discussed period is associated with the increasingly compressed exponential relaxation (CER) dynamics. Such behavior, related to the exponent $\beta > 1$, has only recently been discovered in physical systems (Wu et al., 2018; Trachenko & Zaccane, 2021). It indicates the dominance of processes significantly ‘faster’ than the standard exponential case ($\beta = 1$, and Eq. (1)), also suggesting the presence of internal stresses that can lower energy barriers between different channels of processes developing in the given system, and facilitating rapid rearrangement - even in an avalanche-like manner.

From ~1968 till today, dynamics is dominated by the stretched exponential relaxation (SER) related to the exponent $\beta < 1$, as shown in the inset in **Figure 6**. SER-related dynamics can be associated with a large number of possible process channels, each with a low probability of being open. A shift between states can promote energy dissipation. This phenomenon is often observed in disordered physical systems, but the broad distribution of relaxation times associated with multichannel processes can also be linked to hierarchical constraints or to spatially heterogeneous systems that are locally ordered. The inset in **Figure 6** reveals a general crossover CER ($\beta > 1$) $\Rightarrow$ SER ($\beta < 1$), near the year 1968, for the global population ~3.5 billion.

Notably, such exceptional dynamics recall the Gardner transition pattern, which can generally be described as a crossover between two types of systems, ‘mechanically’ stable and ‘marginally’ stable. Near this transition, responses to endogenic and exogenic impacts are strongly nonlinear and are often associated with intermittent plastic events, while the

system remains globally solid. Solidity breaks down at larger deformations, leading to 'complete' reorganization (Drozd-Rzoska & Sojecka, 2026; Berthier et al., 2019; Li et al., 2021). The authors argue that this fundamental pattern strongly correlates with permanent developments in the early Industrial Revolution and with the reorganization that followed WWII. For states /domains associated with SER ($\beta < 1$) dynamics, and local networking dominate, but when shifting away from a hypothetical singularity (the case of the global population), all of these weaken. Notably, the Gardner transition can be associated with avoiding criticality (Berthier et al., 2019; Li et al., 2021), the pattern evidenced above: Figure (8) and Eq. (20).

When discussing the results of this report, it is worth bearing in mind the issue of Carrying Capacity (Sojecka, Drozd-Rzoska & Rzoska, 2024). Notably, the enormous amount of reports on this topic, over the past decade, has made it central to discussions on sustainable development and ecology (del Monte-Luna, 2004; Wang, 2022; Bradshaw, 2026). While these papers provide quantitative estimates of this key parameter, the authors of the given find them significantly unsatisfactory, particularly with respect to fundamental justification. Such estimations require the selection of significant resources under consideration and their relationships, as well as multidimensional interpretative models, often biased by initial assumptions.

The authors of this paper prefer Cohen's concept (1995), which emphasizes the direct relationship between Carrying Capacity and population changes, to be more significant. In fact, it suggests focusing rather on the time trend for Carrying Capacity changes. Building on this, Cohen proposed the 'Condorcet' relation (Cohen, 1995):

$$\frac{dK(t)}{dt} = c(t) \frac{dP(t)}{dt} = \frac{L}{P(t)} \frac{dP(t)}{dt} , \quad (21)$$

where $L = \text{const.}$

The authors of this report developed this dependence by introducing the analytic RGR factor (Sojecka, Drozd-Rzoska, Rzoska, 2025):

$$\Rightarrow \quad \frac{dK(t)}{dt} = L \frac{1}{P(t)} \frac{dP(t)}{dt} = L \frac{d \ln P(t)}{dt} = L G_P . \quad (22)$$

Following this, the RGR-related characterizations can provide insight into the evolution of Carrying Capacity. **Figures 7 & 8** are of particular interest, as they show the dominant trends in global population and Carrying Capacity across subsequent epochs, described by either the basic Malthusian dynamics (Equation (1)) or the critical growth trajectory (Equation (19)).

It should be noted that essentially the reference to the Verhulst–Pearl equation (3) describes population change using Malthus's model of infinite carrying capacity (C–C). Does this suggest a world of unlimited opportunities for a growing population in 'Malthus-relate' times ? Malthusian's dynamics is related to $G_P = \text{const.}$ In such a situation, subsequent generations of a growing population can individually have the same share of the system's important resources (Carrying Capacity) as previous generations, as indicated by Eq. (22). This conclusion can be supported by carpenters' wages in England, which remained almost unchanged from the 13th century until the early 19th century. England can be considered a model country, as it has had unchanging borders, stable governments, and no 'invasions' by hostile neighbors over such a long period. For six centuries, the share of Carrying Capacity available to individuals remained almost unchanged. During the Industrial Revolution, which began in the 18th century, Carrying Capacity increased rapidly. According to Eq. (22) and **Figure 8**, the typical worker's share of this increase has remained unchanged since the Middle Ages.

Generally, during periods of 'critical' population growth, resources and Carrying Capacity, which are crucially related to creativity, work, innovative progress, and the development of social ties, ultimately lead to an increase in individual 'well-being', i.e., the proportion strong rise of personally available part in Carrying Capacity. **Figure 8** suggests that this situation occurred during the Neolithic era and the first 7,000 years of the Anthropocene. It was an era of increasing opportunity and possibility across generations. This was not repeated until the Industrial Revolution, until 1968.

Of course, after this year, the personal share of global success increases, but on a relative scale, it seems to decrease from generation to generation.

In this work and in the refs. (Sojecka, Drozd-Rzoska, Rzoska, 2025; Sojecka & Drozd-Rzoska, 2025; Sojecka & Drozd-Rzoska, 2024; Drozd-Rzoska & Sojecka, 2026), a new method of preparing population data using numerical filtering supported by Savitzky-Gilay protocol (Luo, et al., 2005) was implemented and demonstrated. This yielded an analytical dataset of 206 points describing population changes over more than 12,000 years. Importantly, this approach was based on an initial data collection from multiple sources, thereby avoiding potential bias that could arise from relying on a single source. Next, a distortion-sensitive test feedback analysis was used to combine empirical data and test model scaling relationships. We emphasize these issues because they fulfill the fundamental requirement of the scientific method: reliable, conclusive empirical validation.

The authors argue that the difficulty of achieving reliable empirical validation is a key factor in the long-term cognitive impasse surrounding the modelling of global population change. This is exemplified by the vast and utterly inconclusive range of predictions for the year 2100, as depicted in **Figure 2**. A second issue is the attempt to test model scaling relationships over long time periods — up to thousands of years and even the entire Anthropocene — without first demonstrating that such a description is fundamentally justified over these timescales. We emphasize this fact because the present work and the refs. (Sojecka, Drozd-Rzoska, Rzoska, 2025; Sojecka & Drozd-Rzoska, 2025; Sojecka & Drozd-Rzoska, 2024; Drozd-Rzoska & Sojecka, 2026) explicitly demonstrate that no single long-term scaling equation for global population changes in Anthropocene can exist.

The authors of the given report note that the aforementioned problems of empirical validation are endemic to many analyses reported to date. In this context, and with reference to the results presented in this work, the authors would like to address two very recent trend-setting works (Zaccone & Trachenko, 2026; Yakovenko, 2026).

Representative can be two recent works which, in addition to offering interesting conceptual results, also demonstrate the aforementioned problem (Zaccone & Trachenko, 2026):

$$\frac{dP(t)}{dt} = b \times P(t) - dP(t) = (b - d)P(t) = r(t)P(t) = \frac{\exp[KP(t)]}{\tau}P(t), \quad (23)$$

where b and d are the birth rate per element per unit time and the death rate per element per unit time, respectively, and $r = b - d$ is known as the ‘productivity of the individual element’. Further, it was assumed that $r(t) = \exp(KP(t))/\tau$, $\tau = \text{const}$, and $K = \text{const}$.

The above means that τ is here an empirical constant, not the relaxation time discussed above. The authors of ref. (Zaccone & Trachenko) consider the above for the coefficient $K > 0$ as reflecting the stretched exponential function (SEF) dynamics and $K < 0$ as the compressed exponential. Further, recalling the empirical evidence, the discussion of the Zaccone-Trachenko relation (2026), proposed to describe relaxation in glass-forming materials following perturbative stress (mechanical, electrical, ...), was adopted due to its formal similarity to Eq. (23). It led the author of ref. (Zaccone A, Trachenko, 2026). Later, it was noted that for $K < 0$ and developing the exponent in Eq. (23) in a Taylor series, the Verhulst equation is recovered. For $K > 0$ the authors indicated that Eq. (23) has the Ricatti differential equation form (Zaccone A, Trachenko, 2026): one of the solutions is von Foerster et al. (1960) type ‘hyperbolic’ equation, although conditions of such solutions in the frames of the population characterizations are not satisfactorily explained. Finally, to take into account the Carrying Capacity, the ‘universal’ carrying capacity impact was taken into account (Zaccone A, Trachenko, 2026):

$$\frac{dP(t)}{dt} = \frac{\exp[KP(t)]}{\tau}P(t) \left[1 + \frac{P(t)}{\kappa} \right], \quad (24)$$

where κ is the Carrying Capacity.

The problem constitutes the multiparameter nature of the final relation describing changes in $P(t)$, which also uses an arbitrarily chosen reference constant value for Carrying Capacity, κ , ultimately leading to the prediction of a maximum population in 2064 (Zaccone A, Trachenko, 2026).

It should be recalled that $1/\kappa$ means the maximum sustainable population size given the population's essential resources. Therefore, the assumed value of Carrying Capacity κ

essentially defines when the stationary phase or population maximum occurs in the Verhulst-type equation, to which the relation introduced in ref. (Zaccone A, Trachenko, 2026) can be encountered. Then, assuming the value of κ , one assumes the eventual appearance and value of the global population a priori.

Despite the suggestion of the universality of the description from the beginning of the Anthropocene to the present, no explicit validation against 'empirical' data has been carried out in ref. (Yakovenko, 2026).

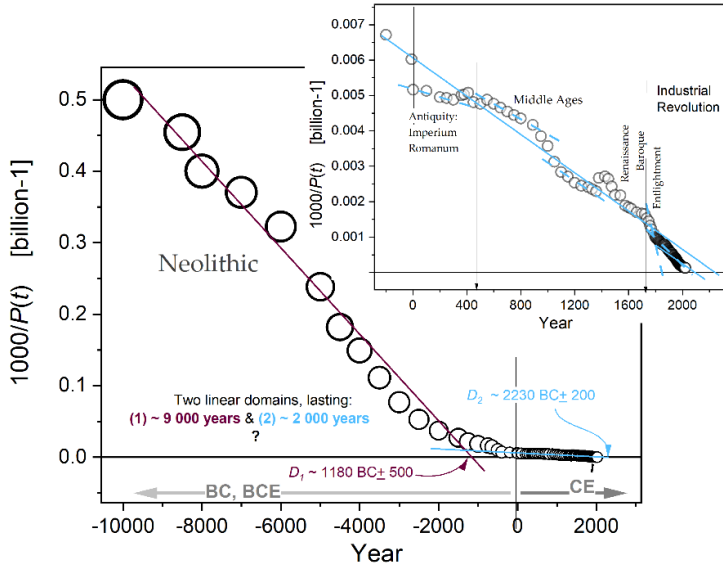


Figure 9. The normalized reciprocal of global population in Anthropocene focused on the direct apparent validation of von Forster et al. (1960). Hyperbolic scaling, i.e., via Eq. (7) with the exponent $M = 1$, indicated by linear patterns. The inset shows the focused behavior for the last two millennia. The dashed lines indicate a set of 'apparent linear domains' matched with von Foerster et al. scaling.

The next issue that should be treated with caution is the reference scale used to analyze the results. The importance of this factor was already indicated in the discussion of 'Doomsday' scaling by von Foerster et al. (1960). Another option for analysis to justify preferences is the 'reciprocal scale' analysis:

$$P(t) = \frac{A}{D-t} \quad \Rightarrow \quad \frac{1}{P(t)} = (A^{-1}D) - A^{-1}t. \quad (25)$$

Validations based on the representation of changes on the scale of $1/P(t)$ vs. t were used, for example, in (Yakovenko, 2026).

The authors of the given report present such an analysis in **Figure 9**. The central part of the plot reveals two domains described by the above relation. The first period, spanning more than 8,000 years from 10,000 BC to around 1500 BC, aligns remarkably well with the findings of the distortion-sensitive analysis depicted in Figure 8. This includes a 'critical anomaly around 1180 BC', which coincides precisely with the collapse of Bronze Age civilizations. Another 'hyperbolic' anomaly appears from antiquity to the present, with an apparent singularity at ~2230. However, as the **insight in Figure 9** shows, such an anomaly does not exist. This is the result of 'scale compression' in analyses based on Eq. (25). Such 'compression' is particularly strong in cases involving gigantic, multi-decadal changes in values — the feature exhibited by global population changes.

8. Conclusions:

This report presents a new approach to modelling global population dynamics, implementing the category of Active Soft Matter as the fundamental reference. More specifically, it implements the subcategory proposed by the authors (Drozd-Rzoska & Sojecka): Human Active Intelligent Soft Matter. Global population changes were then tested using universal model equations applicable to complex Soft Matter systems. Of particular significance are the super-Malthusian equation and critical-like scaling (Sojecka & Drozd-Rzoska, 2024). The Soft Matter provides a well-justified, fundamental basis for

interpreting the parameters in fitted scaling equations, thereby shedding new light on global population dynamics. It offers qualitative supplementation to the standard demographic approaches (see the 1st and 2nd cognitive **Paths** above). It is worth noting that the different Soft Matter based scaling relations enable us to identify and highlight the distinct process features that govern global population changes.

The introduction of distortion-sensitive analysis using the Analytic Relative Growth Rate (Sojecka & Drozd-Rzoska, 2025), is particularly important for the ultimate validation of the model descriptions discussed.

The analysis presented in this paper clearly demonstrates the non-monotonic nature of global population change in the Anthropocene. This makes it fundamentally inappropriate to describe global population changes using a single model equation in arbitrarily selected longer periods. First, it is necessary to verify the adequacy of a given description within a selected time period or epoch, as demonstrated by the methodology presented in this report. A notable outcome of this study is the strong correlation between changes in global population characteristics and specific historical events, reflecting the internal dynamics of extremely complex Human Active Intelligent Soft Matter. This also allows us to identify those events that may have had a global impact.

In conclusion, in light of the 'forecasts till 2100 challenge', this report does not identify validated hallmarks of a future decline in the global population. Instead, the growth, albeit at a slower rate, can be expected. Extrapolating the prevailing trend of recent decades suggests that the global population could reach 14 billion by 2100.

Conflicts of Interest: The authors declare no conflict of interest.

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Research

The New Philosopher's Stone: 'Hard' and 'Soft' Matter under Pressure

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Abstract:

The mythical Philosopher's Stone was expected to transform ordinary metals and even stones into gold. It symbolized the immense life-changing value for a 'disposer.' For high and extreme pressures implementations, the power to transform 'ordinary materials' is incomparably greater. The new materials with exotic features can even be a pivotal factor for a qualitative, civilizational transformation.

First, the report presents basic high-pressure facilities necessary to carry out studies across an extreme pressure and temperature range. Further, it is emphasized that for a qualitative change in properties, the pressure-related force exerted must be proportional to the strength of local bonds within a body. For standard 'Hard Matter' systems, this means pressures even close to those near Earth's core (365 GPa). The disappearance of exotic states after decompression is also a grand problem. Special attention was paid to metallic hydrogen due to its groundbreaking importance for new-generation space travel concepts.

Soft Matter systems are dominated by mesoscale collective processes, which lower 'extreme pressures' even well below ~1GPa, allowing for studies in large-volume pressure chambers. Examples presented are related to high-pressure food preservation (HPP), huge densification of solid glasses by annealing just below the glass transition, and emerging possibilities to address the great challenges of searching systems exhibiting stable negative electrical capacitance and stable negative electrical resistance.

Keywords: High pressure; Extreme pressure, Hard and soft matter; Metallic hydrogen; Space travels; High pressure processing of food; Densification of solid glasses; Negative electric capacitance

1. Introduction

Alchemy refers to Medieval philosophical and spiritual ‘cognitive activities’ that combined, according to present standards, chemistry, metallurgy, medicine, astronomy, and physics. For centuries, the *Philosopher’s Stone* was the most sought-after object in alchemy, until it was finally recognized that this goal is entirely impossible and not even worth pursuing (Ede & Cormack, 2022). The *Philosopher’s Stone* was believed to transform various materials, even a stone, into gold, leading to great wealth. Alchemists also believed that it was essential to prepare an elixir of life, which would enable rejuvenation and even immortality (Ede & Cormack, 2022; Raleigh, 2010). Isaac Newton, perhaps the greatest scientist of all time and one of the ‘fathers’ of Modern Science, who essentially supported the great cognitive success of the Scientific Method (Holman, 2023; Harper, 2012), also succumbed to the charm of the alchemical line of ‘research’. In 2016, a 17th-century manuscript handwritten by Sir Isaac Newton was purchased by the Chemical Heritage Foundation (now the Science History Institute, USA) after decades spent locked away in a private collection. Its incredible contents have finally been made public. It includes descriptions of experiments performed by Newton, plus writings from another well-known chemist of the time, George Starkey, outlining a recipe for ‘Philosophic Mercury’ (Sopnick Mercury), which was thought to be the key ingredient in the creation of the *Philosopher’s Stone* (Newman, 2018).

Today, we are surrounded by innovative materials whose properties and functional benefits would have been considered magical in earlier epochs. However, these materials resulted from a long process of ‘research and development’ grounded in the Scientific Method. Notably, during the 18th century, the impact of Newton’s colossal cognitive success and the researchers more or less associated with him was so profound that the Scientific Method became an instinctive cognitive paradigm (Holman, 2023; Harper, 2012). An interesting example of the ‘*Philosopher’s Stone*’ search is the case of the alchemist Johann Friedrich Boettger, who was hired by the Prince & Elector of Saxony to find it, produce a lot of gold, and bring great wealth to the Prince (Syndram & Weinhold, 2009). The Prince invested heavily in the ‘*Alchemist Laboratory*’ for Boettger. However, success remained elusive, and the alchemist’s situation became extremely dangerous given his Superior Prince habits. However, thanks to a ‘side’ collaboration with the mineralogist Ehrenfried Walter von Tschirnhaus, Boettger developed a technology to produce porcelain from clay-based materials, very common in the Lusatia region of Saxony. Finally, in 1710, the first porcelain factory in Europe began operating in Meissen, Saxony. Magnificent vessels, animals, and human figurines became extraordinary export commodities, far more valuable than gold. The thousand-year monopoly of China and Japan was broken by the grand innovation. The Prince had become so wealthy that, thanks to massive bribery, he was elected King of Poland in a free election, as Augustus II the Strong. Unfortunately, the great wealth acquired proved a big misfortune for Saxony and Poland, as the king’s pride drove him into senseless and ruinous wars (Syndram & Weinhold, 2009; Czok, 2006).

A canonical *Philosopher’s Stone* should transform qualitatively standard, inexpensive, and common materials into something exceptionally valuable in a single, short process. Such a ‘*Philosopher’s Stone Method*’ exists today. Some common materials can acquire extraordinary properties under high or extreme pressures – so extraordinary that they can even revolutionize our civilization.

2. Extreme Pressures Terminals

At Earth’s surface, we live under a pressure of $P \sim 0.1 \text{ MPa}$ ($\sim 1 \text{ atm}$) exerted by the weight of the atmosphere, a gaseous layer almost 200 km high. However, at the boundary of the troposphere and stratosphere (12 km), $P \sim 0.02 \text{ MPa}$ (0.22 atm), with a temperature of $T \sim -56^\circ\text{C}$. For a helium-filled balloon with pilots, the maximum altitude is $\sim 34 \text{ km}$. This is the upper stratosphere with a pressure of $P \sim 0.055 \text{ MPa}$ (0.0055 atm) and a temperature of -26.6°C .

NASA considers the altitude of 80 km to be the boundary of Space, awarding an Astronaut Badge for exceeding it. It is related to the pressure $P \sim 1 \text{ Pa}$ or 10^{-5} atm .

At the Earth's surface, the highest pressure is at the bottom of the Mariana Trench (Challenger depth): $P \sim 110 \text{ MPa}$ ($1,100 \text{ atm}$). Higher pressures occur deeper in the Earth. At a depth of 10 km, the pressure is from 2.8 GPa to 3.5 GPa ($1 \text{ GPa} = 1000 \text{ MPa}$) is noted. At 1000 km, it is from 30 GPa to 40 GPa . At Earth's core, $\sim 6370 \text{ km}$ depth: $P \sim 364 \text{ GPa}$, $T \sim 5500^\circ \text{C}$.

In the Solar System, higher pressures exist inside the large outer planets (Jupiter, Saturn, Uranus) (Marshak & Rauber, 2017; Pritchett, 2025). This pressure distribution in nature is presented graphically in Figure 1.

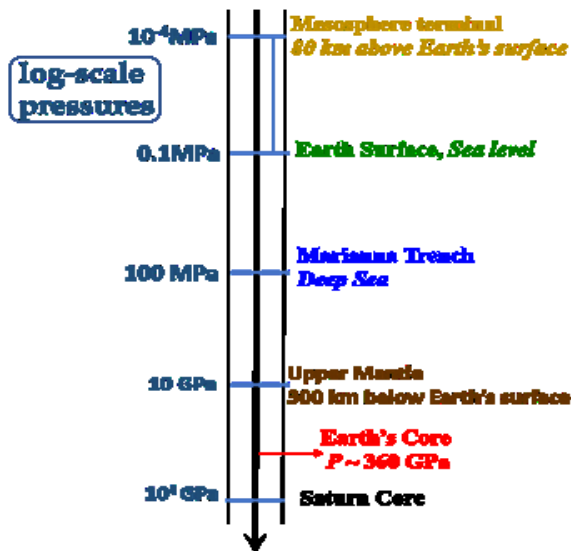


Figure 1. Illustration of the pressure scale for the Earth and the Solar System, with the support of the logarithmic scale.

Легко видеть, что во всяком равновесном состоянии давление тела должно быть положительным. Действительно, при $P > 0$ имеем $(\partial S / \partial V)_E > 0$, и энтропия могла бы увеличиться лишь при расширении тела, чему, однако, мешают окружающие его тела. Напротив, при $P < 0$ было бы $(\partial S / \partial V)_E < 0$, и тело стремилось бы самопроизвольно сжиматься, поскольку это сопровождалось бы возрастанием энтропии.

Имеется, однако, существенное различие между требованиями положительности температуры и положительности давления. Тела с отрицательной температурой были бы совершенно неустойчивы и вообще не могут существовать в природе. Состояния же (неравновесные) с отрицательным давлением могут осуществляться в природе, обладая ограниченной устойчивостью. Дело в том, что самопроизвольное сжатие тела связано с его «отрывом» от стенок сосуда или с образованием полостей внутри него, т. е. с образованием новой поверхности; это обстоятельство и приводит к возможности осуществления отрицательных давлений в так называемых метастабильных состояниях¹⁾.

It's easy to see that in any equilibrium state, pressure should be positive. Indeed, for $P > 0$, we have $(dS/dV)_E > 0$, so entropy can only increase with the expansion of the body, which is prevented by surrounding bodies. Conversely, for $P < 0$, it would be $(dS/dV)_E < 0$, and the body would spontaneously tend to compress if this led to an increase in entropy.

However, there is a qualitative difference between the requirements of positive temperature and positive pressure. A sequence with negative temperature is obviously unstable and cannot exist at all in nature. (Non-equilibrium) states with negative pressure can exist in nature, with limited stability. The point is that the spontaneous compression of a body, associated with its separation from the container walls, is linked to the formation of new surfaces within it; this opens the possibility of negative pressure in so-called metastable states.

Figure 2. A piece of the text on negative pressures from the first, Russian, edition of the monograph by Lev D. Landau, *Statisticheskaya Fizika*, p. 60 (Landau, 1937). The top part shows the original text, and below is the English translation

However, very high pressures are not the only extreme limit. It might seem that the second limit is $P = 0$, or rather the $P \sim 10^{-32} \text{ Pa}$ expected in intergalactic regions (Schilling, 2014). However, pressure is defined with respect to the gaseous state. Solids and liquids can be

isotropically stretched and pass from the isotropic compressing domain ($P > 0$) to the isotropic stretching domain ($P < 0$) without any hallmark (Imre et al., 2002).

The process continues until reaching the absolute stability limit, the spinodal, where stretching exceeds the forces determined by intermolecular, interionic, or interatomic interactions, and the body 'breaks'. It is visualized as cavitation throughout the whole volume of the body, which returns to the equilibrium state under atmospheric pressure. This phenomenon was first noticed by Boyle in the 17th century, who explained the anomalous failure of mercury to settle in early tubular barometers. In the mid-19th century, Berthelot developed the experimental method, which still remains a basic method for generating negative pressures (Imre et al., 2002).

It is worth highlighting the role of one of the greatest physicists of the 20th century, Lev D. Landau, who underscored the phenomenon's fundamental significance in his still-canonical monograph *Statistical Physics* (Landau, 1937). **Figure 2** presents the original citation commenting on this issue.

This report focuses on the extraordinary material properties that arise from the application of extreme positive pressures. However, explaining and understanding certain phenomena also requires awareness of negative-pressure states.

3. Comments on high/extreme pressures experimental techniques

Before we discuss the extraordinary impact of compression on material properties, it is important to highlight the potential and limitations of high-pressure experimental techniques.

First, the exceptional significance of Percy W. Bridgeman's research should be emphasized. He began high-pressure studies in 1905 at the age of 23 and continued along this path until the end of the 1950s. Bridgeman's contribution is not only the discovery of many high-pressure-related properties but also the development of high-pressure equipment. In 1946, Percy W. Bridgeman was awarded the Nobel Prize 'for the invention of an apparatus to produce extremely high pressures, and for the discoveries he made therewith in the field of high pressure physics' (Bridgeman, 1946; Bridgeman, 1949).

Figure 3 shows a photo of the original Percy W. Bridgeman high-pressure apparatus. Instead of being scrapped, it is on display at the Institute of High Pressure Physics 'Unipress' of the Polish Academy of Sciences (IHPP PAS) in Warsaw, Poland.

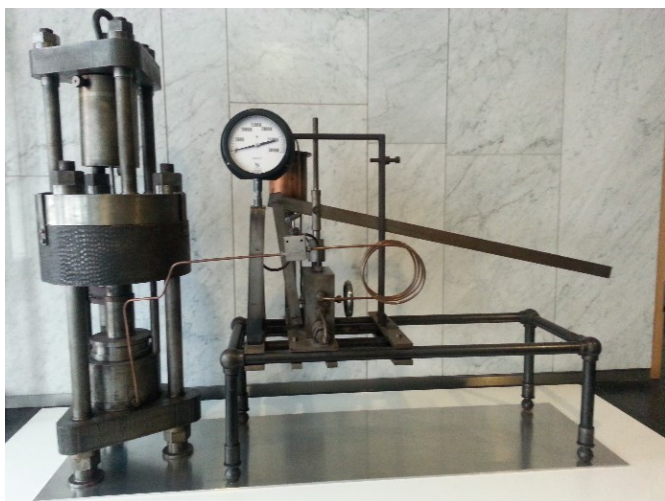


Figure 3. Original Percy W. Bridgman, Nobel Prize 1946, high-pressure apparatus/processor. For the last 5 decades, it has been on display in the Hall of the Institute of High Pressure Physics 'Unipress' PAS, Warsaw, Poland.

This Magnificent Monument of Great Science is situated in a place of honor in the entrance Hall. It came to Poland thanks to Prof. Sylwester Porowski's research fellowship at Harvard University (1967-1969), founder and long-time director of the current IHPP PAS.

High-pressure experimental studies have their own unique specificity. The most important thing is to follow a precise protocol to ensure the researcher's safety and minimize the risk of damage.

High-pressure experimental studies, particularly those involving *in situ* monitoring of physical properties under pressure, require personnel with appropriate training, practical experience, and technical aptitude. This type of research often involves targeted modules for monitoring properties, most often developed only within a given laboratory and not available commercially. They are connected via appropriate feedthroughs to external measuring equipment. Reliable isolation between the pressure-transmitting medium and the material being tested (the 'sample') poses a fundamental challenge. This is crucial for high-pressure studies of liquids and Soft Matter systems, where even tiny impurities can distort the results.



Figure 4. High-Pressure (up to 0.9 GPa, $-30\text{ }^{\circ}\text{C} < T < 150\text{ }^{\circ}\text{C}$, $V = 20\text{ mL}$) setup for *in-situ* Broadband Dielectric Spectroscopy (BDS), DTA and density tests. A special liquid serving as a pressure-transmitting medium is required. The high-pressure chamber is the silver element on the left. In the center, a high-pressure pump and the control system are visible. On the right, a large-volume thermostat with external circulation is visible. The facility is available in IHPP PAS 'Unipress', X-Press-Matter Lab, Warsaw, Poland (<https://young4softmatter.pl/>)



Figure 5. High-Pressure (up to $P = 2.4\text{ GPa}$, $-20\text{ }^{\circ}\text{C} < T < 150\text{ }^{\circ}\text{C}$, $V = 10\text{ mL}$) set up for *in-situ* Broadband Dielectric Spectroscopy (BDS) tests. Special liquid as a medium transmitting pressure. The facility available in IHPP PAS 'Unipress', X-Press-Matter Lab, Warsaw, Poland (<https://young4softmatter.pl/>)

Notably, there has been recent technological progress in the field. It is related to the reliability of high-pressure apparatuses, including new generations of seals and user-friendly design solutions. This also includes previously unavailable programming and computer control of temperature and pressure profiles. The earliest standard approach relied solely on compression (one-way changes). Now, it can be supplemented by tests involving decompression under precisely reduced and controlled pressure.

When planning a high-pressure laboratory, it is important to recognize that there is no single universal high-pressure processing system for arbitrary temperature and pressure ranges. In the temperature range from $-30\text{ }^{\circ}\text{C}$ to $+150\text{--}200\text{ }^{\circ}\text{C}$, a liquid (Plexol is the standard) is used as the pressure-transmitting medium. Modern high-pressure pumps enable precise changes during compression and decompression, as well as computer control. For pressures up to $\sim 1\text{ GPa}$, a significant volume of the high-pressure chamber is

available. It allows for the location of advanced measurement modules. Such a system is shown in **Figure 4**.

For higher pressures, a good solution is to place the measurement module with the sample inside a Teflon cylinder, without any fluid intermediary in pressure transmission. This is a manostatic concept, in which the piston acting on the Teflon cylinder exerts pressure. It causes its (reversible) deformation, yielding the pressure acting on the sample. There is no liquid or gaseous intermediary medium. Such a system, operating in the authors' X-PressMatter IHPP PAS laboratory, is shown in **Figure 5**. It acts up to $P = 2.4$ GPa, probably the highest pressure achieved using this concept.

In practice, this type of high-pressure system operates effectively for pressures above 300 – 400 MPa, and for monitoring material properties from atmospheric pressure to the above-mentioned value of 2.4 GPa, it is necessary to use coherently the processors shown in **Figures 4 & 5**.



Figure 6. High-Pressure (up to 2.4 GPa), and High-Temperature processor (> 1200 °C), with the pressure chamber volume $V = 1$ L, internal heater and gas (Nitrogen, Argon, Helium) as the medium transferring pressure. Available in IHPP PAS 'Unipress', Warsaw, Poland. The co-author of this report is saying 'Hello' in the picture.

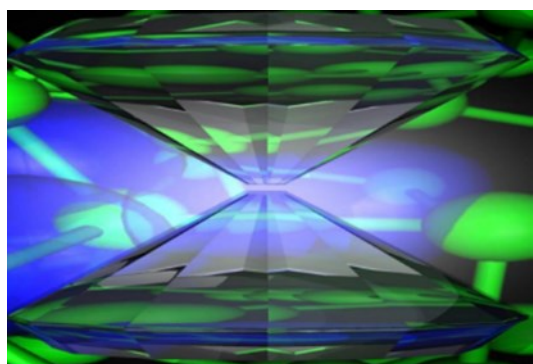


Figure 7. Diamond Anvil Cell Concept Illustration.

For high-pressure tests involving temperatures below -30 °C or above 200 °C, the pressure-transmitting medium must be a gas. It can still be associated with large pressures from a dozen or so mL to 1-2 L. **Figure 6** shows a facility operating up to the paramount pressure of ~ 2.5 GPa and temperatures even above 1200 °C. We emphasize this aspect because in high-pressure tests at temperatures above 200 °C, the primary method of operation under such conditions becomes a heating element inside the high-pressure chamber.

For research at extreme pressures ranging from $\sim 4\text{GPa}$ to as much as $300\text{--}500\text{GPa}$, i.e., pressures even greater than those at the Earth's core, the so-called anvil cell is practically the only high-pressure technique. The canonical version involves two suitably prepared small diamonds pressed together in an 'anvil,' in the simplest version controlled by a screw. The Diamond Anvil Cell is conceptually illustrated in **Figure 7**. Worth stressing is a tiny volume of compressed materials, often even lesser than $50\mu\text{L}$. The detection of physical properties is possible using techniques involving electromagnetic radiation, ranging from X-ray to infrared, in which the influence of the transparent diamond distortions can be eliminated. For pressures up to $\sim 10\text{GPa}$, the anvil cell can be made from a profiled superhard sintered material. In such a case, the compressed sample volume can reach $\sim 1\text{mL}$. It allows, for example, limited studies of dielectric properties. The photo of such a processor, available in IHPP PAS, is presented in **Figure 8**.



Figure 8. Anvil cell facility, made of superhard sintered carbide, working volume $V \sim 1\text{ mL}$, extreme pressures up to $\sim 10\text{ GPa}$. Available in IHPP PAS.



Figure 9. Processors for High Pressure Processing/Preservation (HPP) of foods, pharmaceuticals, or cosmeceuticals. Left: the lab-scale processor for pressures up to $P = 600\text{ MPa}$, $-15\text{ }^{\circ}\text{C} < T < 90\text{ }^{\circ}\text{C}$, $V = 1.1\text{ L}$. Right: the pilot (semi-industrial) scale HPP processor for pressures up to $P = 600\text{ MPa}$, and the pressure chamber volume $V = 50\text{ L}$. The facility is available in IHPP PAS 'Unipress', X-Press-Matter Lab, Warsaw, Poland (<https://young4softmatter.pl/>).

When discussing the world of high-pressure laboratory technical implementations, existing semi/industrial applications cannot be overlooked. An example that has already reached enormous market success is the High Pressure Processing/Preservation (HPP) technology for foods, which has recently been implemented also for pharmaceuticals and cosmeceuticals. This is so-called cold pasteurization, performed at near-room temperature, in which the product is placed in a high-pressure chamber and subjected to pressure in the range of $300\text{--}600\text{ MPa}$ for 3 to 15 minutes. This compression causes the lethal rupture of the cell membranes of parasitic microorganisms and fungi. Their number is significantly

reduced without protein denaturation, meaning that the product after decompression is microbiologically safe while retaining the flavor and nutritional value of the fresh product. Currently, gigantic high-pressure processors (HPP) with volumes exceeding 500L are in use. **Figure 9** shows photos of such processors on the laboratory scale (chamber volume $V \sim 1\text{L}$) and the pilot, semi-industrial scale with pressure chamber $V = 50\text{ L}$ (Sojecka et al., 2024).

4. Extreme pressures impact: 'Hard Matter'

The well-known application of high pressures to change material properties is the graphite-to-diamond conversion. It requires pressures of 5 – 6 GPa and temperatures $T \sim 1300\text{ }^{\circ}\text{C}$. Under such conditions, the relatively weak forces associated with graphite's planar bonds cannot resist the external pressure, causing it to collapse and re-organize into the dense, tetrahedral crystal structure of diamond (Hall, 1961). Furthermore, this new "diamond" state is preserved after decompressing to ambient conditions. A super-soft material (graphite) is transformed into an extraordinary, super-hard, and transparent diamond (Hall, 1961). However, this is an exceptional case, as achieving new states of matter with extraordinary properties typically requires much higher pressures, and these states are not preserved upon decompression.

In standard solid crystalline or semi-crystalline materials, the pressure-related force required to transform the state must be comparable to the strong interatomic, interionic, or intermolecular interactions, which are usually huge. This also applies to materials processed from the gas or liquid phase, as compression will first cause a transition through a sequence of phases until reaching a solid crystalline one. Finally, it can mean pressures comparable to those in Earth's core, often paired with very low or very high temperatures. For applications, another very significant challenge is obtaining materials with unusual properties under normal conditions and in large quantities. Indeed, almost routinely, after decompressing, unusual properties disappear, and the system returns to its initial state. The basic example can be the 'Holy Grail' of high-pressure research, namely the Search for Metallic Hydrogen.

A decade ago, the experimental discovery of metallic hydrogen in a diamond anvil cell was announced, for $P = 450\text{ GPa}$ and $T = 5\text{K} - 10\text{K}$ (Dias & Silvera, 2017; Silvera & Dias, 2021). However, this result has remained fiercely debated, and metallic hydrogen still seems to await its ultimate discovery (Castelvecchi, 2017; Wang et al., 2024; Zhang et al., 2026). The extreme-pressure experiment will only prove the existence of metallic hydrogen, but the results could provide fundamental inspiration for theoretical models, perhaps bringing us closer to producing such a material under normal conditions.



Figure 10. Nuclear Electric Propulsion (NEP) system for Space Ships, following National Aeronautics and Space Administration (NASA), (Atkinson, 2025). In the report, the link to metallic hydrogen 'fuel', given its unique properties, is considered.

It is predicted that metallic hydrogen can be 'the most powerful rocket fuel yet to exist'. The operating metric is the 'specific pulse, I_{sp} ', of the rocket engine. For metallic hydrogen, it is estimated to $I_{sp} \sim 1700\text{ s}$, while for the most efficient engine nowadays, based on liquid hydrogen, $I_{sp} \sim 460\text{ s}$ (Silvera & Cole, 2010).

Let's let our imaginations run wild and consider the possibilities of a Nuclear Thermal Propulsion (NTP) system (NASA, 2021), already tested in practice, but linked to metallic hydrogen unique features. NTP is conceptually similar to a chemical propulsion system, but the combustion chamber is replaced by a nuclear reactor to heat the propellant. For such an 'engine' $I_{sp} > 800$ s (NASA, 2021). A further development is Nuclear Electric Propulsion (NEP), in which the nuclear reactor also generates massive amounts of electrical power that then can drive electric/ion thrusters. Here, $I_{sp} > 3000$ s is estimated (NASA, 2021). Combining a 'condensed' energy storage system based on metallic hydrogen with the capabilities of NEP and NTP systems, a propulsion system able to launch giant payloads into orbit, even with a single-stage rocket, can be expected. Moreover, it would be a completely 'ecological' mode of transport, avoiding the dramatic atmospheric pollution associated with current rocket technology. The visualization of the NEP spaceship is shown in **Figure 10** (Atkinson, 2025).

Nowadays, the Earth-Mars journey requires ~270 days, during the optimal 'window-time'. Recently, a new optimal trajectory that shortens the travel time to 153 days has been proposed (De Oliveira, 2026). For the combination of Metallic Hydrogen (MH) and NEP technologies, one can expect a travel time reduction to ~30 days when starting from Lunar orbit. Notably, 30 days were also needed for the *Santa Maria* caravelle, guided by *Christopher Columbus*, to reach America (In fact: Hispaniola, Haiti) from the Canary Islands in 1492 (Irving, 2014).

Given the exceptional possibilities for 'fuel' obtained at high compression, one can expect that using the MH-NEP propulsion system, the shortest Earth-Jupiter travel can be only ~3 months. Since hydrogen is the fuel, it could perhaps be replenished at the destination, for example, using water from the Martian ice caps.

But large quantities of metallic, superconducting hydrogen can mean not only extremely efficient and 'condensed' fuel. It can also enable energy-efficient generation of strong magnetic fields. In such a field, ionized hydrogen, after supplementary electric charge separation, can move in a circle, accelerating with each revolution, like in a cyclotron. This could pose a challenge for a spaceship propulsion system using Metallic Hydrogen—NEP + cyclotron-like support. Such a solution could increase the I_{sp} factor even to tens of thousands. Furthermore, creating a magnetic field around the ship can provide a supplementary 'shield' to protect the crew.

Following this path, the current Earth Civilization could become a Solar Civilization in a relatively short time, perhaps even a single generation. As in Columbus's time, at the turn of the 15th/16th century, a truly global civilization can suddenly emerge.

We present these 'mildly science-fiction' comments to demonstrate how high pressures focused on new materials can 'change everything', like the *Philosopher's Stone*.

The key missing element for this goal is the challenge of metallic hydrogen, a task that requires intensified, focused high-pressure studies. But perhaps a more feasible goal will be solids based on multifold-coordinated hypermolecules, such as hypermethane or hyperethane (Seeyangnok et al., 2025).

The question also arises whether high-pressure conditions can shift the phase transition boundary of the superconducting state to near-room temperatures (Hemley & Ashcroft, 1998; Eremets et al., 2024; Xu et al., 2026). This would be the ultimate application breakthrough for such materials. This goal seems still to be distant, although, for example, for lanthanum superhydride (LaH_{10}) $T_c = 250$ K for a pressure of $P \sim 190$ GPa has been reported (Sun, 2021). However, studies of the specific impacts of compression on the internal material spacing and topology of the structure can serve as a fundamental reference for model analyses and indicate directions for further research.

The modeling and interpretative significance of experimental high-pressure studies can be illustrated by the case of Invar. In 1920, Charles Édouard Guillaume received the Nobel Prize "in recognition of the service he has rendered to precision measurements in physics through his discovery of anomalies in nickel steel alloys" (Guillaume, n.d.). In particular, this was for the discovery of Invar, an alloy with virtually zero thermal expansion over a very wide temperature range, which was crucial at the time for constructing precision measuring instruments at the Observatoire de Paris, where he worked. Today, the significance of this discovery is even greater. But only in 2009, thanks to high-pressure studies, was the ultimate explanation for the phenomenon obtained. Specifically, it was shown that electrons in Invar alloys adopt a distinct energy configuration. This energy state lies at the

boundary between two types of magnetic behavior and is highly sensitive to the alloy's precise elemental composition. If one shifts the Invar chemical composition by only a couple of percent, this specific energy configuration will disappear. High-pressure tests made such a shift, essential for explaining the phenomenon's origin, possible. (Guillaume, n.d.; Winterrose et al., 2009)

In 2007, Eremets et al. synthesized polymeric nitrogen at pressures $P > 110$ GPa and $T \sim 2000$ K, using a laser-heated diamond anvil cell (Eremets et al., 2007). It was noted that a large amount of energy would be released during the transformation from polymeric nitrogen to its molecular form, much more than that of the most powerful energetic materials currently available. It would be a non-polluting process, since only molecular nitrogen would appear. New 'ecological' 'extreme fuel' or 'extreme explosive'. (Eremets et al., 2007; Ma et al., 2009) appeared.

Another example of a potentially groundbreaking material emerging from extreme pressure is the exceptionally conductive solid superionic ammonia alpha-phase, discovered at pressures $P > 50 - 60$ GPa and $T > 700$ K (Ninet et al., 2012). This material has potentially groundbreaking implications for future generations of batteries and fuel cells.

These are just examples of groundbreaking materials obtained under high pressure, so groundbreaking that they could even transform our global civilization. However, a fundamental problem arises: the extreme pressures close to the Earth's core are required, and unique states disappear upon decompression to atmospheric pressure. The case of the diamond is exceptional because graphite exhibits an 'extraordinary softness' resulting from its specific crystal structure, which means that conversion to ultra-hard diamond requires relatively low pressures.

Unique properties emerging from compression can also be preserved in chemical reactions, in which compression forces large changes in inter-element distances and the associated characteristics of local interactions. For example, for CsI, at atmospheric pressure, the average volume occupied by an atom is ~ 4.8 nm, while at pressure of ~ 150 GPa it is only 2 nm (Hemley & Ashcroft, 1998).

Another example of a significant property that changes under extreme pressures and can be preserved after decompression is the hardness of some materials. For diamond, the hardness can rise from ~ 60 GPa to as much as ~ 150 GPa (McMillan, 2016), for instance. Yet another example is polymerization driven exclusively by high pressure, yielding super-pure polymeric materials, which are significant for advanced applications in biotechnology and medicine. Here, a sufficient pressure is $2 - 7$ GPa, at relatively low temperatures of $100 - 220$ °C (Evlyukhin et al., 2016). Even lower pressures are required to start the process in polypropylene glycol (Andersson & Andersson, 1998).

But the last topic is essentially an example for the next section, 'Soft Matter under Pressure'. For the vast majority of materials for which exotic, even civilization-breaking, properties are expected, the required pressures are really huge. This is often combined with extremely high or low temperatures. Such extreme pressures are only possible to be created in Diamond Anvil Cells, with tiny volumes samples of $50 - 100$ μ L. After decompression, they generally return to standard 'normal' properties. This raises another major challenge: obtaining them under normal conditions and in the large quantities necessary for applications.

These limitations, particularly the extreme pressure values, stem from the fact that in a given process, the forces generated by pressure must be comparable to the extremely strong bonding forces – interatomic, interionic. This is the case of standard materials, which can be named 'Hard Matter'. A common representative is a stone, which we would like to squeeze, as shown in **Figure 11**.



Figure 11. Squeezed standard Hard Matter and the less obvious soft matter bodies. For Soft Matter, pressures of ~ 1 GPa or lower can yield impacts similar to ~ 100 GPa or more in Hard Matter. For Soft Matter, 'exotic' properties induced by pressure are often preserved upon decompression. Pressures in the range of 10 GPa, 100 GPa, or more can be created only in tiny volumes (Hard Matter case). Pressures of $P < 1$ GPa can be created even in industrial-scale volumes, up to $V > 500$ L (Soft Matter case).

However, there are materials, such as a sponge, where even a light squeeze with a hand qualitatively changes the properties, even the shape. This is an example of Soft Matter, a category of materials associated with weak, or even very weak, bonds between constituent elements. It is illustrated in **Figure 11** and discussed in the next section.

5. High or Medium pressures impact: 'Soft Matter'

In 1991, Pierre G. de Gennes presented the Nobel Prize lecture entitled: 'Soft Matter', introducing the novel category, linking liquid crystals, polymers, blends, gels, foams, supercooled glass-formers, colloids, critical liquids ...and bio-systems. (de Gennes, 1991; de Gennes & Badoz, 1996). These systems/materials are illustrated in **Figure 12**.

Soft Matter complex systems exhibit some common features (de Gennes & Badoz, 1996):

- Dominance of mesoscale elements, leading to 'universal' scaling patterns of 'isomorphic' physical properties
- Ability to spontaneously self-assemble
- Extreme sensitivity to Exo- & Endogenic Impacts, linked to weak interactions between mesoscale species
- Often, the appearance of exceptional mesophases.

Soft Matter is based on macromolecules or multimolecular assemblies that are naturally associated via qualitatively weaker bonds than those in standard Hard Matter systems. Even relatively weak pressures can significantly influence their properties, as illustrated in **Figure 11**.

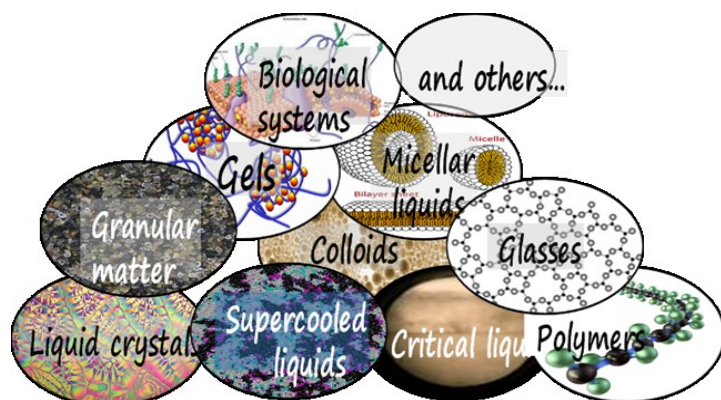


Figure 12. Basic representatives of soft matter category systems.

Below, some unique features of Soft Matter under pressure are briefly presented. In 1912, Bridgeman reported the impact of high pressure on egg white and demonstrated its coagulation at $P \sim 600$ MPa (Bridgeman, 1912). This marked the beginning of High Pressure Processing/Preservation (HPP) technology, which was implemented commercially only in the early 1990s, when it became possible to build industrial-scale

processors with large-volume pressure chambers (Sojecka et al., 2024). Industrial-scale processing is recognized for the high-pressure chamber with volumes $V > 100$ L. Standard industrial processing involves applying a high-pressure pulse in the range 350 – 600 MPa, for the period 3 to 15 minutes. Currently, HPP-related pro-health food and beverages constitute a huge global market. Food processed in this way retain the flavor and nutritional value of fresh products, while achieving a five-decade reduction in microorganism counts. This is a level of thermal pasteurization, relating to heating the product up to 83 – 86 °C. For HPP, the pasteurization is possible at room temperature (Sojecka et al., 2024). So it is also recalled as ‘cold pasteurization’ or ‘bridgemenization’. The impact of high pressure primarily involves the disruption of the cell membrane, which for particularly parasitic bacteria occurs at pressures of 400 – 500 MPa. This is also related to the destruction of intracellular organelles. **Figure 13** shows such changes in a yeast cell after 10 minutes of compression at $P = 400$ MPa.

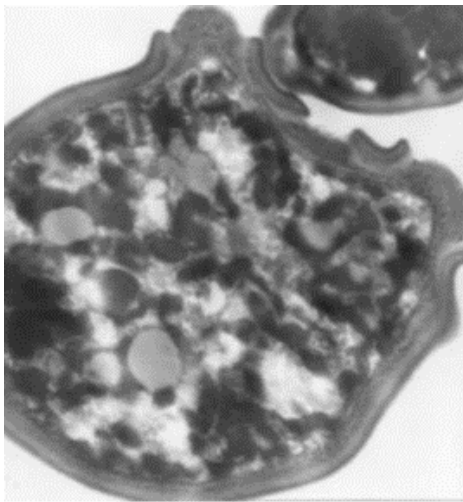


Figure 13. SEM image of a yeast cell after compressing at $P = 400$ MPa, for 10 min. Note the breaking of the cell wall (membrane) and the destruction of organelles inside. Photo: X-PressMatter Lab. IHPP PAS.

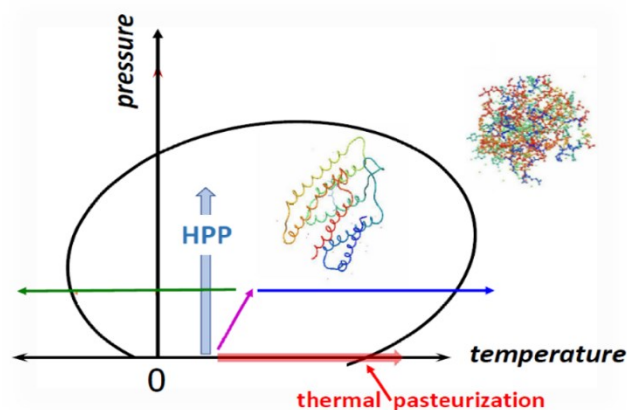


Figure 14. Denaturation curve in the pressure-temperature range. Schematic structures of the native and denatured proteins are also shown. The horizontal red ‘wide’ arrow indicates standard thermal pasteurization, which finally occurs at $T = 83 - 86$ °C. The vertical ‘light blue’ arrow indicates the direction of the standard HPP pasteurization under near-room temperatures. Thin violet, green, and blue arrows indicate the innovative high-pressure ‘cold’ sterilization solution proposed in ref. (Sojecka et al., 2024).

The unique feature of HPP technology is primarily its high-pressure lethal impact, which destroys cell membranes. This is ‘designed’ to occur at the lowest possible pressure, appropriately adjusted to the given type of microorganisms in the food product matrix. At the same time, this pressure is (most often) too low to significantly affect the food product’s texture, and it is below the limit defined by the denaturation curve shown in **Figure 14** (Sojecka et al., 2024). All of these led to the preservation of the fresh product’s nutritional

and flavor qualities after decompression because the denaturation limit, which would reduce these essential product features for consumers, is not exceeded (Sojecka et al., 2024; Sojecka et al., 2025).

Model foods can be considered as a case of particularly Complex Soft Matter. However, HPP studies related to food referred to Soft Matter are still rare. This can be explained by the fact that the vast majority of research in this field is conducted by food technologists and biologists.

A class of soft materials of enormous application importance includes rod-like liquid-crystalline (LC) compounds, for which the canonical representative is pentyl cyanobiphenyl (5CB). This is also a model Soft Matter system with a weakly discontinuous melting/freezing phase transition between the isotropic liquid and nematic LC phase. Notably, the transition is related to the freezing of a single element of symmetry: orientational, uniaxial ordering. This means that it is also a system with enormous fundamental significance for modeling, both in *Liquid Crystals Physics* and *Soft Matter Physics*.

Notable, that generally, a continuous Isotropic Liquid to Nematic phase transition for rod-like molecules is fundamentally ruled out, as evidenced by Onsager and de Gennes (de Gennes, 1974). **Figure 15**, however, shows that minimal endogenous frustration from a tiny addition of nanoparticles and adjusted compression enables the impossible to be possible, i.e., a continuous isotropic-nematic mesophasic transition (Łoś et al., 2023).

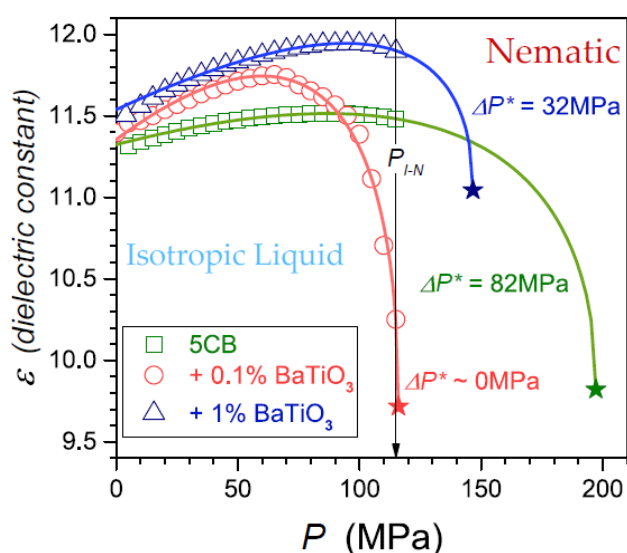


Figure 15. Pretransitional, critical-type, changes of dielectric constant for isothermal ($T = 60\text{ }^{\circ}\text{C}$) compressing on rod-like liquid crystalline pentyl cyanobiphenyl (5CB) with addition of paraelectric BaTiO_3 nanoparticles. The parameter ΔP^* is the metric of the phase transition discontinuity, i.e., the distance between the isotropic liquid terminal and the extrapolated continuous phase transition, indicated by stars. Note that when adding $x = 0.1\%$ nanoparticles, the formally ‘forbidden’ continuous phase transition appears. It is validated by $\Delta P^* \approx 0$ (Łoś et al., 2023).

Negative electric capacitance is a counterintuitive phenomenon in which increasing the applied voltage decreases the stored charge. When combining a negative-capacitance material with a traditional capacitor, an exceptional device for voltage amplification can emerge, potentially revolutionizing the energy design of Super-Efficient Transistors and Ultra-High-Density Supercapacitors. The grand challenge is to obtain a stable negative capacitance. It is suggested to appear in some ferroelectric-based devices, but only as a ‘virtual, transient event’ during state switching (Hoffmann et al., 2021).

Negative electric resistance is an electronic property where an increase in voltage across a device causes a decrease in electric current, inverting the standard rule of Ohm's law. It can be essential for generating, amplifying, and switching signals, but obtaining it remains a grand challenge (Bormashenko, 2025).

Figure 16 shows the spectra of the real and imaginary parts of the dielectric permittivity for the PolyVinylidene Fluoride (PVDF) polymer + Barium Strontium Titanate (BST) microparticle composite. For some frequencies, the real component (related to electric

capacitance) and the imaginary component (related to electric resistance) become negative (Starzonek, Rzoska, et al., 2019).

The authors of this report can suggest a preliminary qualitative explanation of this exceptional phenomenon. Namely, both PVDF and BST are ferroelectric materials with a strong tendency toward piezoelectricity, i.e., the creation of free charges at the border induced by a mechanical stress. In the given composite, large microscale BST particles (i.e., composed of one or more ferrod domains) are surrounded by a ferroelectric polymeric PVDF matrix. Relatively strong isotropic stress from external compression may cause the appearance of local free electric charges at the PVDF - BST micro-boundaries within the matrix, yielding 'additional free charges', and creating the phenomenon reported in **Figure 16** and ref. (Starzonek, Rzoska, et al., 2019). This is preliminary evidence, which is currently being further investigated in the X-PressMatter Lab IHPP PAS by the authors.

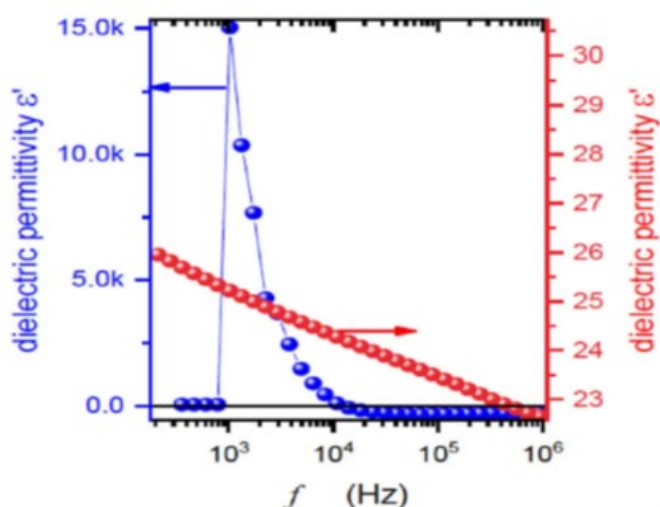


Figure 16. Real and imaginary parts of dielectric permittivity frequency spectrum (room temperature) for PVDF (Polyvinylidene fluoride) polymer +BST (Barium Strontium Titanate) microparticles composite. Note the appearance of Negative Electric Capacitance and Negative Resistance at some frequencies. Room temperature, and $P \approx 500\text{MPa}$ test (Starzonek, Rzoska, et al., 2019).

Solid amorphous glass is essentially considered within the 'Hard Matter' category discussed in the previous section. The compression by 1-3 GPa at room temperature of even $T = 100 - 300\text{ }^{\circ}\text{C}$ can change the material's density only by 1 – 2%, and it is immediately 'lost' upon decompressing. However, as shown in (Smedskjaer, ..., Rzoska, Boćkowski, et al., 2015) and (Januchta, ..., Rzoska, Boćkowski, et al., 2017), high-pressure annealing ($P < 2\text{GPa}$) in the solid amorphous phase just below the glass temperature for ~30 min can cause permanent densification of up to 18%. Such an experiment was conducted on materials designed for new-generation displays. It is important for applications, because a super-hard surface was obtained. The experiment was carried out under $P = 2\text{ GPa}$ for $T = 700 - 800\text{ }^{\circ}\text{C}$, in the solid state just below the glass temperature, using the apparatus shown in **Figure 6**.

This unusually high-pressure, high-temperature annealing process was also used in research on amorphous 'glassy' materials for future generations of batteries and fuel cells. Many glass-forming materials, when cooled, 'pass' the melting/freezing temperature, i.e., the highly discontinuous liquid-to-solid crystal phase transition without any hallmark, entering a stable, though non-equilibrium, ultraviscous, supercooled liquid state, ending with a transformation to a solid amorphous glass state.

In such materials, the standard method for determining the melting temperature is to measure it during heating from the solid glass state, when crystallization and a discontinuous phase transition to the solid crystal phase occur in the ultraviscous state, above the glass temperature.

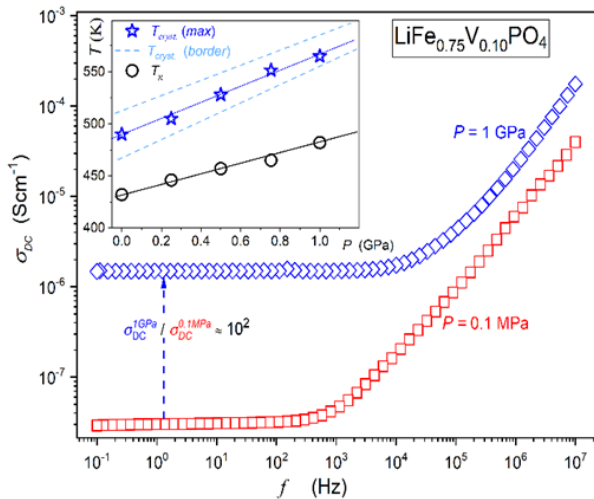


Figure 17. The real part of the electric conductivity spectrum for the native $\text{LiFe}_{0.75}\text{V}_{0.10}\text{PO}_4$ solid glass and for the nanocomposite with no grains appearance due to nanocrystallization under pressure and subsequent annealing just above the glass temperature. The inset presents the pressure dependence of the glass temperature (open circles) and the crystallization zone (stars). The horizontal pattern in the central part of the plot is related to DC electric conductivity. (Baranowski, Rzoska, et al., 2019).

This concept was implemented in materials of potential importance for the next generation of electrodes for batteries and fuel cells, such as $\text{LiFe}_{0.75}\text{V}_{0.10}\text{PO}_4$ (Baranowski, Rzoska, et al., 2019) or NaFePO_4 (Szpakiewicz-Szatan, Rzoska, et al., 2024). This operation was performed under pressure $P = 1 \text{ GPa}$, first by heating above the glass temperature $T_g = 450 - 500 \text{ }^\circ\text{C}$, until the crystallization region was reached. After a short stay in this region, approximately 1 minute, a rapid cooling to the solid glass – just below the glass temperature T_g was arranged. Next, after $\sim 30 \text{ min}$ annealing under pressure $P = 1 \text{ GPa}$, densification took place. Decompressing to normal conditions leads to the formation of a stable composite: solid glass + nanocrystalline grains, isotropically dispersed in a glass matrix. The size and quantity of nanograins can be precisely controlled by the appropriate design of P-T treatment paths.

Figure 17 shows the spectrum of electric conductivity of a native amorphous solid glass and the nanocomposite obtained due to the above treatment in the P-T space. Notably, there is over 2-decades increase in electric conductivity, reaching values that exceed the application-significant limit. This is due to high-pressure impact and ‘densification’ of crystalline nanograins. These factors facilitate the effective hopping mechanism, responsible for high electrical conductivity.

6. Conclusions

This report presents the extraordinary potential of high-pressure studies, across moderate-to-extreme ranges, to transform ‘ordinary’ materials into ‘exotic material counterparts’ with extraordinary properties.

The mythical *Philosopher’s Stone* was expected to transform ordinary metals like lead or iron, and even stones, into gold, which symbolized the immense value of a life-changing factor for a ‘disposer.’ For high & extreme pressure implementations, the power to transform ‘ordinary materials’ through compression is incomparably greater. The new materials with exotic features can even be a pivotal factor for a qualitative, civilizational transformation.

Perhaps only the ‘fight for fire’ of pre-humans, in prehistory, possessed a similar causative power.

In such a context, Soft Matter systems under medium/high pressures can be particularly important. First, the pressure transformation occurs for pressures ‘only’ around 1 GPa or lower. This is currently in the ‘industrial’ scale of pressures, which can be implemented in enormous pressure chambers with volumes of hundreds of Liters. This also means that the grand barrier to scaling up from small experimental samples to large-volume production

does not exist. Moreover, unusual and extremely beneficial properties for applications are preserved after decompressing.

In fact, such industrial applications have already been implemented for some systems. This is the case of HPP processors, already processed on a giant scale: see **Figures 9**.

Further research and application prospects for 'Soft Matter under Pressure' are still enormous, as briefly sketched above. This overview omits major fundamental challenges, where high-pressure research is crucial. For example, high-pressure studies are essential to explaining the grand glass-transition cognitive mystery, which physicists have been grappling with for decades (Drozd-Rzoska et al., 2007; Drozd-Rzoska et al.; 2023; Drozd-Rzoska, 2019a; Drozd-Rzoska, 2019b).

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Research

Formation of Complex Patterns in Refractory High-Entropy Alloys

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Abstract:

High-entropy alloys (HEAs) composed of multiple principal elements exhibit a remarkable diversity of microstructures, especially when refractory metals and magnetic *3d* transition elements are combined. Despite extensive experimental work, the fundamental mechanisms governing the formation of such complex patterns remain insufficiently understood. In this study, we investigate microstructural evolution in tin-containing refractory HEAs and propose a minimal theoretical model capable of reproducing the observed configurational richness. Experimentally, scanning electron microscopy reveals drastic microstructural changes induced by Sn and by the addition of Fe, Ni, Cu, or Nb. Theoretically, of our interest are different patterns that appear in those high entropy alloys. They consist of multiple constituents and by varying their relative concentration rich diversity of quantitatively and qualitatively different configurations could emerge. Theoretical explanation of these patterns generic mechanism is still lacking. In this contribution we present a minimal model that illustrates possible mechanisms yielding such configurational diversity. For this purpose, we use 2D Ising lattice model consisting of classical spins in an external transversal field Ω at temperature zero. In this approach we mimic key properties of high-entropy patterns by interaction matrix J . Our simple model exhibits symmetry breaking structural transitions on varying Ω . The emergent spin configurations are seeded by J eigenvectors. Therefore, different structures of J yield different patterns. Furthermore, even for a fixed system configuration one could nucleate diverse patterns on varying histories of samples where mode-coupling interactions play significant roles.

Keywords: High-entropy alloys; Refractory metals; Microstructure; Pattern formation; Ising model; SEM-BSE

1. Introduction

High-entropy alloys (HEAs) represent a paradigm shift in alloy design, where multiple principal elements are combined in near-equiatomic ratios to stabilize simple solid-solution phases through high configurational entropy. Since their introduction (Yeh et al., 2004), HEAs have attracted significant interest due to their exceptional mechanical, thermal, and chemical properties.

Refractory HEAs, containing elements such as Hf, Ti, Zr, Nb, Ta, or W, are particularly promising for high-temperature applications because of their high melting points and excellent mechanical stability (Tietz & Wilson, 1965). In this work, we focus on Hf-Ti-Zr-based alloys modified with Sn and 3d transition metals (Fe, Ni, Cu, Nb). Sn is commonly used in conventional alloys and strongly influences phase stability and segregation behavior. The addition of magnetic 3d elements introduces further complexity, enabling potential applications in electronic and magnetic devices.

Previous studies (Gačnik et al., 2021) have shown that Sn-containing refractory HEAs exhibit superconductivity and complex multiphase microstructures. However, the mechanisms behind the emergence of these patterns remain unclear. Here, we combine experimental characterization with a minimal theoretical model to elucidate the origin of microstructural diversity.

2. Material and Methods

2.1. Alloy Preparation

All alloys were synthesized by arc melting under an Ar atmosphere. First, a ternary HfTiZr alloy (HTZ) was prepared as a reference. Sn was then added to form the quaternary HfTiZrSn alloy (HTZS). Finally, Fe, Ni, Cu, or Nb were added individually to obtain five-component alloys:

- HfTiZr (HTZ)
- HfTiZrSn (HTZS)
- HfTiZrSnFe (HTZS-Fe)
- HfTiZrSnNi (HTZS-Ni)
- HfTiZrSnCu (HTZS-Cu)
- HfTiZrSnNb (HTZS-Nb)

Each ingot was re-melted several times to ensure homogeneity.

2.2. SEM-BSE Imaging

Microstructures, exhibiting complex patterns, and chemical composition was determined by Scanning Electron Microscopes (SEM) JEOL JSM-7600F Schottky FE SEM equipped with INCA Oxford 350 EDS SDD, ThermoFisher Quanta 650 ESEM equipped with EDS Oxford Instruments AZtec Live, Ultim Max SDD 40 mm², and Karl Zeiss Supra 35 VP SEM with EDS analysis. The samples' surfaces were prepared by the FIB Helios NanoLab NL650 FEI dual-beam system.

The surface of HEAs was imaged by Backscattered Electrons (BSE), technique available in all described SEMs. Backscattered electron (BSE) imaging was used to visualize compositional contrast: brighter regions correspond to heavier elements (Hf, Sn), while darker regions indicate lighter elements (Ti, Fe, Ni, Cu).

2.3. EDS Analysis

Chemical compositions of individual phases were determined by SEM-EDS (Energy Dispersive Spectroscopy). Some very small or very dark inclusions could not be reliably quantified due to detector limitations.

2.4. Theoretical Modelling

We present a simple toy model to illustrate conditions for which a rich diversity of competing configurations could appear in systems of our interest. Our focus is to identify inherent properties and frustrations needed enabling rich diversity of diverse patterns observed in our magnetic systems.

For this purpose, we use classical Ising spin lattice-type model of N spins in a transverse field (Jelen et al., 2024). The local spin orientation at the i -th site is determined by the unit vector $\hat{\mathbf{s}}_i$. We express the system in the Cartesian system (x, y, z) , which is defined by the unit vector triad $(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y, \hat{\mathbf{e}}_z)$. Mutual interactions of spins are described by interaction matrix J . The matrix component J_{ij} describes the interaction between the z -axis components of i -th and j -th spin placed at locations $\vec{\mathbf{r}}_i$ and $\vec{\mathbf{r}}_j$, respectively. For sake of simplicity use the XY-type model, where the orientation of spins is confined to the Cartesian (x, z) plane. We parametrize spin orientations in terms of angles θ_i :

$$\hat{\mathbf{s}}_i = \cos\theta_i \hat{\mathbf{e}}_x + \sin\theta_i \hat{\mathbf{e}}_z. \quad (1)$$

To increase complexity of different states we introduce presence of a homogeneous external field which favors homogeneous alignment along the x -axis. Hence, introduce a simple competition within a system and analyses under what conditions one could obtain diversity of competing configurations. The resulting dimensionless interaction energy of N -interacting spins is expressed as (Jelen et al., 2024).

$$W = -\frac{1}{2} \sum_i \sum_j J_{ij} s_i^z s_j^z - \Omega \sum_i s_i^x. \quad (2)$$

The sums run over all the spin sites of the system. Direct inter-spin interactions could impose parallel ($J_{ij} > 0$) or antiparallel ($J_{ij} < 0$) mutual alignment of interacting spins. Different probability distributions of J_{ij} values in our model mimics different constituents and spatial inhomogeneities of magnetic systems studied. The transverse field Ω introduces anisotropy into the system. To increase diversity of potential configurations we set $\vec{\Omega} = \Omega \hat{\mathbf{e}}_x$. This setting imposes frustrations: i.e., J_{ij} imposes alignment among the z -axis and Ω in the orthogonal direction. In the limit $\Omega \rightarrow \infty$ and finite values of J_{ij} this ordering field tends to align the spins homogeneously along $\hat{\mathbf{e}}_x$. In our approximate treatment we neglect thermal fluctuations.

3. Results

In the following we present typical experimentally measured patterns in our samples. Afterwards we discuss possible pattern generating mechanisms.

3.1. Microstructures

In **Figure 1** we present backscattered-electron SEM images of all six alloys in the HfTiZrSnM (M=Fe, Ni, Cu, Nb) system. The images highlight how adding Sn and different transition metals (Cu, Fe, Ni, Nb) drastically changes the microstructure. Figure 1 shows six SEM backscattered-electron (BSE) images, each corresponding to one alloy in the HfTiZrSnM system (M = Fe, Ni, Cu, Nb). BSE contrast correlates with average atomic number: brighter are heavier elements (Hf, Sn), darker are lighter elements (Ti, Fe, Ni, Cu). The figure visually demonstrates how adding Sn and different transition metals drastically changes the microstructure. Baseline alloy HfTiZr (HTZ) is presented in **Figure 1a**). It contains microstructure of μm -sized grains with dark edges. Grain interior is brighter (Hf-rich) and edges are darker (Ti-rich). It has no inclusions or secondary phases. This alloy is the only single-phase system in the series. The Ti segregation to grain boundaries is driven by lattice-strain minimization. Master Sn-containing alloy HfTiZrSn (HTZS) is shown in **Figure 1b**). It exhibits large, several $10 \mu\text{m}$ faceted grains with straight edges, which are slightly darker than interiors. Matrix is light grey and contains many small inclusions,

some very dark that can not be determined by SEM-EDS. Sn addition produces four-phase microstructure with $\text{Hf}_{20}\text{Ti}_{12}\text{Zr}_{32}\text{Sn}_{37}$ grains, rewritten as $(\text{Hf}, \text{Ti}, \text{Zr})_5\text{Sn}_3$ and $\text{Hf}_{19}\text{Ti}_{17}\text{Zr}_{32}\text{Sn}_{32}$ grain edges, appearing darker than the grain interior. This is the first appearance of the characteristic $(\text{Hf}, \text{Ti}, \text{Zr})_5\text{Sn}_3$ grains that dominate all Sn-containing alloys. **Figure 1c)** shows the master Sn-containing alloy with added Fe. Adding iron produces very large faceted grains (10-100 μm). Matrix is darker and contains many small inclusions, mostly light gray, some very dark. The darker the inclusion appear, the lighter elements it contains. Next alloy contains Ni and is presented in **Figure 1d)**. It is similar to HTZ and HTZ-Fe regarding large light-grey grains. Matrix contains many bright inclusions and some very dark inclusions. This alloy has four distinct Ni-containing phases giving strong contrast variations. **Figure 1e)** shows HfTiZrSnCu alloy with distinctive “bean-shaped” grains, unlike all other alloys. Grains contain many freckles (small contrast variations). Matrix shows variable grey levels, indicating strong compositional fluctuations. In addition, it shows some small black inclusions. HfTiZrSnNb alloy, presented in **Figure 1f)** has much simpler microstructure than Fe, Ni or Cu alloys. The large light $(\text{Hf}, \text{Ti}, \text{Zr})_5\text{Sn}_3$ grains are embedded in a HfTiNb -rich darker-grey matrix.

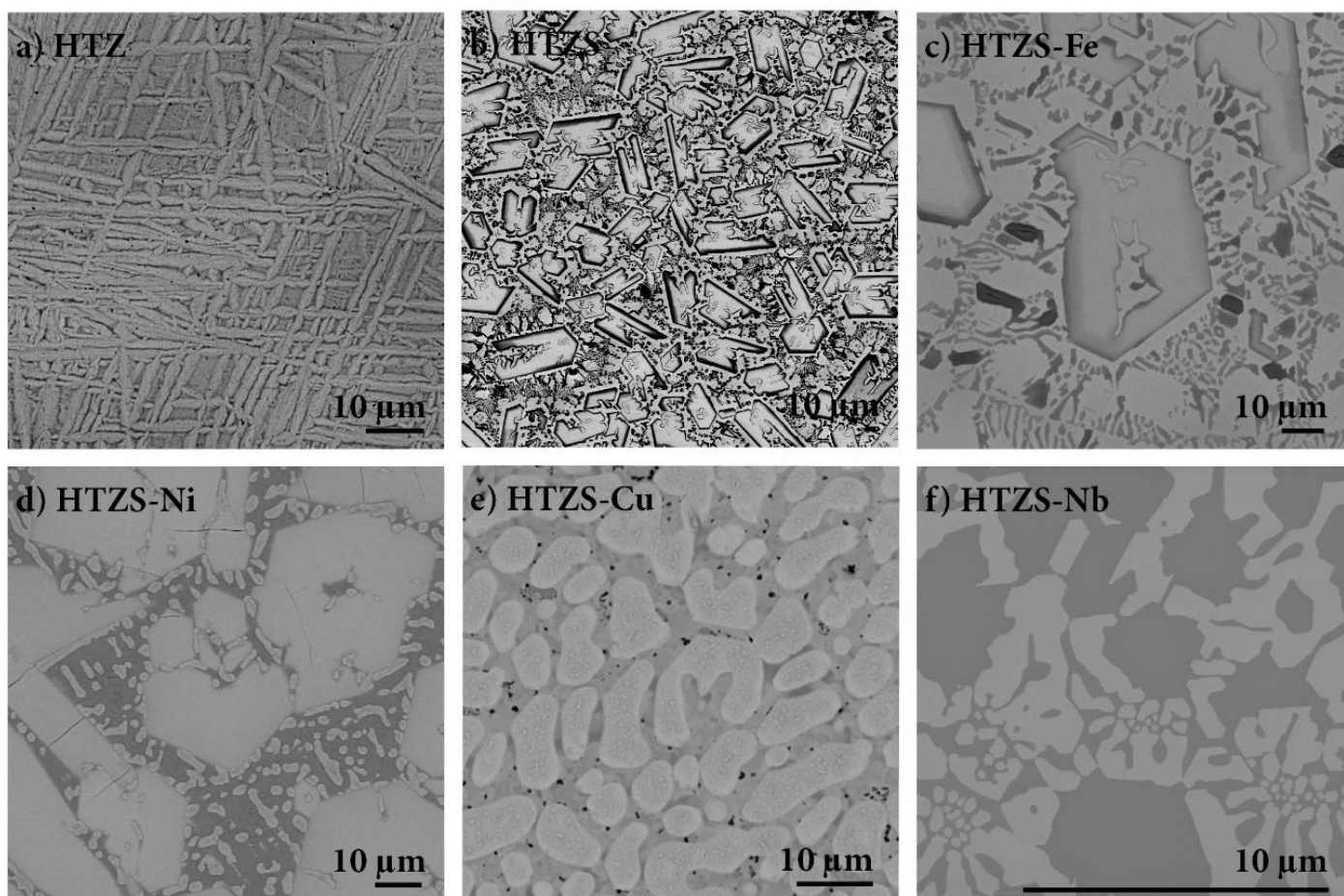


Figure 1. SEM BSE images of **a)** HfTiZr master alloy (HTZ), **b)** HfTiZrSn (HTZS), **c)** HfTiZrSn-Fe (HTZS-Fe), **d)** HfTiZrSnNi (HTZS-Ni), **e)** HfTiZrSnCu (HTZS-Cu), **f)** HfTiZrSn-Nb (HTZS-Nb).

In addition to imaging, chemical composition of individual phases (different grey shades on the BSE images) was determined by Energy Dispersive Spectroscopy (EDS) in SEM. The results are presented in **Table 1**.

Table 1. SEM-EDS chemical composition of investigated alloys. The column “Appearance of the phases” describes morphologic appearance of the phase in the SEM-determined microstructure.

Alloy	Appearance of the phases	Composition
HfTiZr	grains interior	Hf38Ti34Zr28
	grains' dark edges	Hf32Ti41Zr27
HfTiZrSn	large grains interior	Hf20Ti12Zr32Sn37
	large grains edges	Hf19Ti17Zr32Sn32
	matrix	Hf30Ti36Zr20Sn14
	dark inclusions	Can not be determined by SEM EDS
HfTiZrSnFe	large grains	Hf19Ti10Zr33Sn36Fe2
	small bright inclusions	similar to large grains, less Sn, more Ti
	matrix	Hf25Ti25Zr4Sn2Fe44
	dark inclusions	Can not be determined by SEM EDS
HfTiZrSnNi	large grains	Hf16Ti11Zr32Sn35Ni6
	small bright inclusions	Hf30Ti17Zr3Ni50
	matrix	Hf22Ti38Zr6Sn2Ni32
	dark inclusions	Can not be determined by SEM EDS
HfTiZrSnCu	grains (beans), freckles on beans	Hf18Ti11Zr29Sn32Cu10
	matrix-dark	Hf20Ti25Zr10Sn3Cu42
	matrix-bright	Hf25Ti33Zr7Sn2Cu33
	dark inclusions	~Hf18Ti48Zr7Sn3Cu24
HfTiZrSnNb	grains	Hf19Ti11Zr30Sn36Nb4
	matrix	Hf25Ti24Zr13Sn14Nb24

3.2. Theoretical Analysis

We introduce scalars $u_i = \sin\theta_i$, where $u_i \rightarrow 0$ in the limit $\Omega \rightarrow \infty$. Thus, it holds

$$\hat{s}_i = \sqrt{1 - u_i^2} \hat{e}_x + u_i \hat{e}_z. \quad (3)$$

Our interest is to show how diverse patterns of spin configurations could emerge from known statistical properties of J_{ij} on decreasing Ω where we originate from initially spatially homogeneous patten aligned along $\hat{e}_x$. The latter conditions is realized in the limit $\Omega \rightarrow \infty$ (i.e., $\Omega \gg J_{ij}$). For this aim we expand W in terms of small values of u_i up to the 4th power:

$$W \sim -\frac{1}{2} \sum_i \sum_j J_{ij} u_i u_j - \Omega \sum_i \left(1 - \frac{u_i^2}{2} - \frac{u_i^4}{8}\right) = W_h + \frac{\Omega}{8} \sum_i u_i^4. \quad (4)$$

Here W_h stands for harmonic contribution. Minimization of W_h yields the equilibrium equations $\sum_j J_{ij} u_j = \Omega u_i$. We express them in the matrix notation as

$$\underline{J} \hat{\mathbf{u}} = \Omega \hat{\mathbf{u}}, \quad (5)$$

$\underline{J} = \{J_{ij}\}$ stands for the interaction matrix. N -dimensional vectors $\hat{\mathbf{u}} = (u_1, u_2, u_3, \dots, u_N)$ describe configuration of spins that are slightly misaligned from the $\hat{e}_x$ direction. Solution of Equation (5) determines eigenvector (modes) $\hat{\mathbf{u}}^{(m)}$ of $\underline{J}$:

$$\underline{J} \hat{\mathbf{u}}^{(m)} = \Omega_m \hat{\mathbf{u}}^{(m)} \quad (6)$$

where Ω_m stands for eigenvalues. The eigenvectors are normalized and orthogonal, i.e.,

$$\sum_i u_i^{(m)} u_i^{(n)} = \delta_{mn}, \quad (7)$$

and δ_{mn} stands for the Kronecker symbol.

For relatively large values of Ω , where the harmonic approximation works well, one express $\hat{\mathbf{u}}$ as the superposition of eigenmodes

$$\hat{\mathbf{u}} = \sum_m A_m \hat{\mathbf{u}}^{(m)}, \quad (8)$$

where A_m are amplitudes of modes. Considering the expansion Eq.(8) and orto-normal condition Eq. (7), one obtains

$$W = -N\Omega + \frac{1}{2} \sum_m A_m^2 (\Omega - \Omega_m) + \frac{1}{4} \sum_m \sum_n \sum_k \sum_l \sigma_{mnkl} A_m A_n A_k A_l, \quad (9)$$

$$\sigma_{mnkl} = \frac{\Omega}{2} \sum_i u_i^{(m)} u_i^{(n)} u_i^{(k)} u_i^{(l)}. \quad (10)$$

In the following, we study configurational evolution on decreasing Ω . We first neglect the interaction among competing modes. Afterward, we consider cases where modes are coupled. In the approximation of decoupled modes it follows

$$W \sim -N\Omega + \sum_m \left(\frac{1}{2} A_m^2 (\Omega - \Omega_m) + \frac{1}{4} A_m^4 \sigma_m \right), \quad (11)$$

where we introduced

$$\sigma_m = \frac{\Omega}{2} F_4^{(m)}, \quad (12)$$

$$F_n^{(m)} = \sum_i u_i^{(m)n}, \quad (13)$$

where $n \in \{1, 2, 4\}$. Note that $F_2^{(m)} = 1$ because the/ modes are normalized. Minimization of W yields

$$A_m = \sqrt{(\Omega_m - \Omega)/\sigma_m} \quad (14)$$

for $\Omega_m < \Omega$ and $A_m = 0$ for $\Omega_m \geq \Omega$. Thus, the modes open via 2nd order phase transition on decreasing Ω below the corresponding eigenvalue Ω_m . The total response of the system is given by Eq.(8).

However, in general, the modes could be coupled where the coupling terms appear as fourth-order terms in Eq.(9). For illustration purposes, we focus to cases when only two modes are coupled. In this case

$$W = -N\Omega + \frac{1}{2} \sum_m A_m^2 (\Omega - \Omega_m) + \frac{1}{4} \sum_m (\sigma_m A_m^4 + \sum_{l \neq m} (4\mu_{ml} A_m A_l^3 + 3\gamma_{ml} A_m^2 A_l^2)), \quad (15)$$

$$\text{where } \mu_{ml} = \frac{\Omega}{2} \sum_i u_i^{(m)} u_i^{(l)3}, \quad \gamma_{ml} = \frac{\Omega}{2} \sum_i u_i^{(m)2} u_i^{(l)2}.$$

Note that contributions weighted by μ_{ml} are sensitive to relative orientation (i.e., the sign of amplitudes) of modes. An equilibrium (meta)stable configuration, in which two (i.e., m -th and l -th) modes are opened, is determined by equations

$$\frac{\partial W}{\partial A_m} = \frac{\partial W}{\partial A_l} = 0, \quad \frac{\partial^2 W}{\partial A_m^2} = \frac{\partial^2 W}{\partial A_l^2} > 0, \quad \frac{\partial^2 W}{\partial A_m^2} \frac{\partial^2 W}{\partial A_l^2} - \left(\frac{\partial^2 W}{\partial A_m \partial A_l} \right)^2 > 0. \quad (16)$$

The condition, where such a configuration becomes unstable, is determined by

$$\left(\frac{\partial^2 W}{\partial A_m \partial A_l} \right)^2 - \frac{\partial^2 W}{\partial A_m^2} \frac{\partial^2 W}{\partial A_l^2} = 0. \quad (17)$$

4. Discussion

The experimental results demonstrate that small compositional changes—particularly the addition of Sn and magnetic 3d elements—lead to qualitatively different microstructures. One obtains diversity of different patterns via relatively small changes in relative concentration of systems ingredients.

Furthermore, we present a simple toy model which pinpoints possible pattern generating mechanisms and key quantitative parameters from which one could predict emergent qualitative structure observed macroscopically. For this purpose, we assumed that key properties of different compositions could be mimicked by the interaction matrix $\underline{J}$. This matrix consists of direct interaction J_{ij} between i -th and j -th sample constituent. In our rough modelling we assumed Ising model-like inter-spin interactions. We assume that essential properties of different compositions and relative concentrations of ingredients are well fingerprinted by $\underline{J}$. For example, for a reference Sample A let us assume that all the spins are the same (i.e., we have a homogeneous sample consisting of a single element) placed on a simple square lattice. If they would exhibit short range ferromagnetic interaction, where $J_{ij} = J > 0$ has a constant positive value among 1st neighbours and is zero otherwise, $\underline{J}$ would possess in 3D six lines parallel to the diagonal where $J_{ij} = J$. As a contrast state we chose spin glass systems (Sample B) [5] where distribution of J_{ij} values is essentially random and $\langle J_{ij} \rangle = 0$ ($\langle \dots \rangle$ stands for the ensemble average). In these case random values of J_{ij} appear along the 6 parallel lines of $\underline{J}$. Furthermore, one needs in the model a parameter which could drive a structural phase transition. For this purpose we introduced in our model an external ordering field $\vec{\Omega}$, where $\underline{J}$ and $\vec{\Omega}$ have competing ordering tendencies. This allowed us to study structural transformation at zero temperature, which simplifies required mathematics of the problem. Key results are as follows. We focused on the regime on decreasing Ω where the spatially homogenous structure of classical spins initially aligned along $\vec{\Omega}$ becomes influenced by $\underline{J}$. Our analysis reveals that the formed pattern can be expressed as a superposition of modes which represent $\underline{J}$ eigenvectors. Furthermore, on decreasing Ω the 1st mode opened corresponds to the largest eigenvalue $\Omega_{\max}$ of $\underline{J}$. For instance, such a mode would be “extended” (i.e., nonlocal) and “localised” (spatial variations appear locally) (Sherington and Kirkpatrick, 1975) in Sample A and Sample B, respectively. In the case of extended modes due to the mode coupling terms (see Eq.(15)) the growth of lower modes could be suppressed on further decreasing Ω . Therefore, there would be only one mode contribution (corresponding to $\Omega_{\max}$) in overall system response, see Eq.(8). However, if $\underline{J}$ exhibits “localised” states mode blocking is essentially less significant. Consequently, the system might exhibit a pattern where several modes are superimposed. Note that coupling between modes increases on increasing the spatial overlap of the modes. In case that overlapping is important then one could by appropriate initial perturbation excite a particular $\underline{J}$ mode preferred by system, which could then persist if it is protected by suppressing mode overlapping interactions. Our analysis reveals that one could make quantitative predictions about possible states by constructing appropriate interaction matrix $\underline{J}$ of the system. Furthermore, to have a variety of different states one needs some inherent anisotropy in the system.

5. Conclusions

We considered diversity of patterns in high-entropy metallic alloys. Our microscopic analysis reveals that one could realise a rich pallet of different configurations by varying relative concentration of key constituents. We constructed a simple toy model to identify possible reasons for such diversity. We supposed that patterns are dominantly influenced by magnetic interactions among constituents which we mimicked by the interaction matrix $\underline{J}$. We believe that different ingredients in the sample are fingerprinted in rather complex

structure of $\underline{J}$ and consequently rich complexity of possible emergent patterns. For this purpose, one needs frustration within the system and competition among diverse configurations. Note that our model represents a first step towards theoretical understanding of observed pattern complexity in high-entropy metallic alloys. Namely, in the minimal model we neglected thermal fluctuations and enforced phase transition by a tuneable external field. Our future goal is to develop a more realistic model with tuning parameters reflecting real samples.

Conflicts of Interest: The authors declare no conflict of interest.

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Research

Topology Enters Physics

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Abstract:

We illustrate Gauss-Bonnet and Poincare-Hopf (GB-PH) theorems in closed two-dimensional geometrical manifolds exhibiting in-plane orientational order which should be spatially homogeneous in equilibrium. Such geometrical shapes inevitable exhibit spatial dependent Gaussian curvature. The latter is a source of local frustrations that could stabilise topological defects (TDs) in the structure. Their presence is in general avoided because they introduce free energy penalties into the system. GB-PH theorem describes total winding number of TDs and indirectly their number in such systems. Namely, due to topological origin the behaviour of TDs obeys conservation laws which are determined by topological invariants. We present applicability of the GB-PH theorem in nematic liquid crystals, where such phenomena could be relatively easily tested experimentally. Furthermore, in terms of these theorems one could also describe quantum Hall effect, via which topology first entered physics.

Keywords: Topology; Gauss-Bonnet theorem; Poincare-Hopf theorem; Topological defects.

1. Introduction

Topology is a branch of mathematics, and it focuses on properties that are conserved under continuous transformations. Not long ago the Nobel prize winner Gamow pointed out that topology is one of rare examples whose topics are not yet exploited in explaining physical phenomena. Pioneering topological entrance in the realm of physics is the discovery of the integer Quantum Hall Effect (QHE) in 1980 (Tong, 2016). It was observed that Hall resistance R_H in two-dimensional (2D) electron systems are quantized - determined by an integer number n . A typical experimental set up is depicted in **Figure 1**.

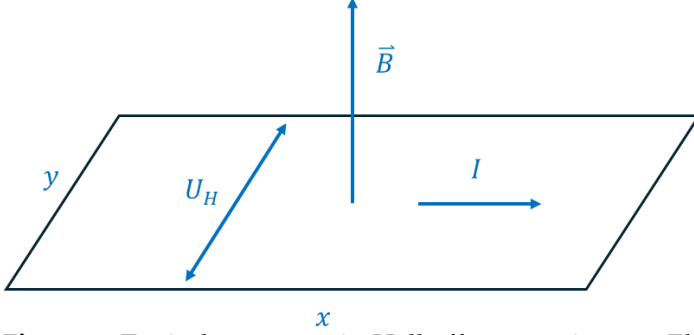


Figure 1. Typical geometry in Hall-effect experiments. Electron gas is confined to a 2D (xy) ribbon and the external magnetic field $\mathbf{B}$ is applied perpendicular to the ribbon larger surface. I denotes electric current and U_H denotes electric voltage.

The resulting QHE behaviour on increasing the magnetic field illustrated in **Figure 2**. The Hall resistance can be expressed as $R_H = h/(ne_0^2)$, where h is the Planck constant and e_0 is the elementary electric charge. Note that values of R_H are independent of material properties and consequently universal. The observed behaviour is robust, independent of microscopic imperfection because it is determined nonlocally, by the whole system. Such topological behaviours are of interest for various applications in physics. For example, it could be exploited for fault-tolerant quantum computing, where information is stored topologically (Han, 2023). This discovery opened the doors via which topology entered physics. From then on, it plays progressively increasing important role in physics. Clear evidence for this is the 2016 Nobel prize in physics (Gibney & Castelvechi, 2016) awarded to David J. Thouless, F. Duncan M. Haldane, and J. Michael Kosterlitz for their theoretical discoveries of topological phenomena in topological phases of matter. It might be that topology explains all observed discrete phenomena in nature (Wilczek, 2012).

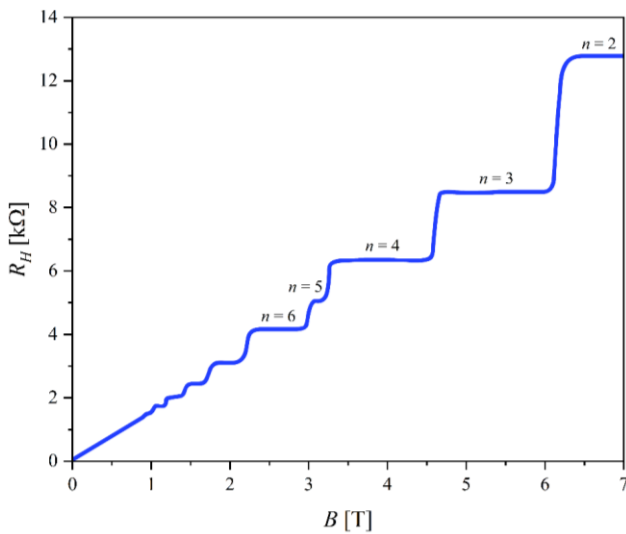


Figure 2. The Hall resistance of a low-temperature 2D electron gas enclosed within a ribbon-shaped conductive wire as the function of magnetic field strength. One observes a robust stair-like quantized behaviour. The corresponding Hall conductance (the reciprocal of Hall resistance) is given by $\sigma = ne_0^2/h$, where the integer n characterizes each plateau.

In this contribution we present the Gauss-Bonnet (GB) and Poincare-Hopf (PH) topological theorem (do Carmo, 1976; Kamien, 2002) which play the central role in various physical phenomena. They could be derived using the universal parallel transport mechanism (Levi-Civita, 1916; Persico, 1921). For illustration let us consider 2D manifolds which exhibit in-plane order described by a vector field as depicted in **Figure 3**. The field is parallel transported along a curve whenever it is rigidly conveyed in a frame that displays the least motion capable of accompanying changes of the local unit normal. Let us consider closed paths in such manifolds and parallel transport the vector field along these paths. **Figure 3a** illustrates a loop on a flat substrate. One sees that in this case the initial orientation of the field and the final one, after completing the path $A \rightarrow B \rightarrow C \rightarrow D \rightarrow A$, are identical. However, if the enclosed surface exhibits a nonzero Gaussian curvature K_g (Kamien, 2002), this introduces a frustration in the system. Gaussian curvature for a regular surface patch is defined as $K_g = \kappa_1 \kappa_2$, where $\{\kappa_1 = 1/R_1, \kappa_2 = 1/R_2\}$ stand for the principal curvatures of the patch. Here R_1 and R_2 are the corresponding curvature radii (e.g., for a flat surface it holds $K_g = 0$, and $K_g = 1/R^2$ for a spherical surface of radius R). **Figure 3b** illustrates that in such cases the initial and final orientation are different because $K_g = 1/R^2$ of the enclosed ($A \rightarrow B \rightarrow C \rightarrow A$) surface patch is finite.

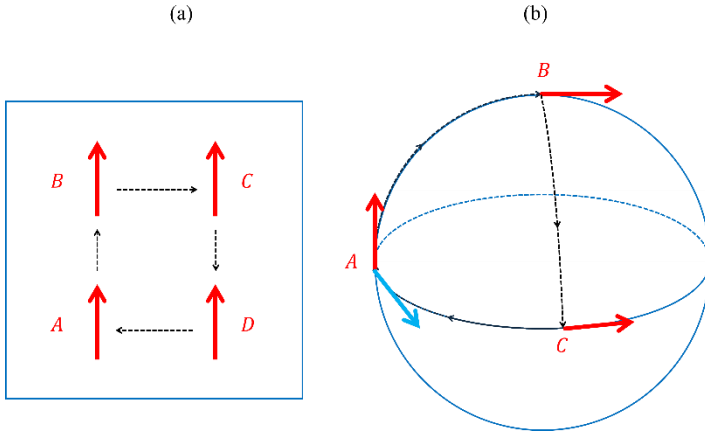


Figure 3. Parallel transport on 2D manifolds along the closed paths marked with dashed lines. a) Parallel transport $A \rightarrow B \rightarrow C \rightarrow D \rightarrow A$ of a vector field on a flat manifold. The initial and final vector field orientation on traversing a closed path is identical. b) Parallel transport $A \rightarrow B \rightarrow C \rightarrow A$ on a sphere. The initial (red arrow) and final (blue arrow) orientation of the field are different because the enclosed surface exhibits nonzero Gaussian curvature.

In the following we present the GB-PH theorems on simple 2D geometrical manifolds exhibiting polar or axial orientation order. We chose cases where phenomena could be relatively easily visualised and understood. In conclusions we describe how these theorems could be generalised and their relationship with QHE.

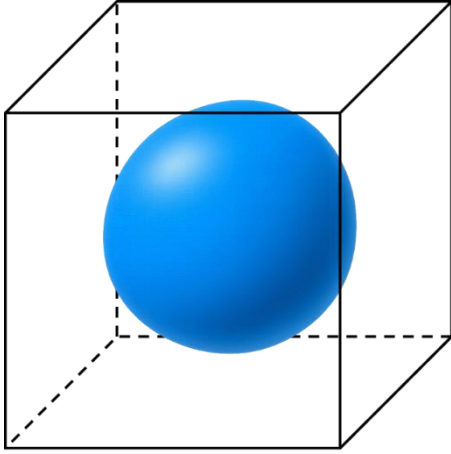
2. Results

In this section we present GB-PH theorems on 2D closed curved manifold possessing either vector or axial orientational order. We henceforth refer to such systems as *shells*. For the ease of understanding let us suppose that on flat substrates such order is spatially homogeneous in equilibrium. Such vector ordering field could represent thin closed ferromagnetic films. On the other hand, axial order is realised in thin nematic liquid crystal films (Rosso et al., 2012). A relatively simple derivation of GB-PH theorems for polar and axial fields in 2D manifolds is presented in Ref. 12.

The Gauss-Bonnet theorem (do Carmo, 1976) claims

$$\frac{1}{2\pi} \oint K_g dA = \chi_E = 2(1 - g). \quad (1)$$

Here, dA stands for an infinitesimally small surface area and the integration is carried out over the closed shell. The quantity χ_E is the Euler characteristic of the closed shape and g (genus) counts the number of holes within the system. For example, bodies exhibiting spherical or toroidal topology are characterised by $\{g=0, \chi_E = 2\}$ and $\{g=1, \chi_E = 0\}$, respectively. **Figure 4** illustrates how one could calculate χ_E of a shape by surrounding it with planar regular surfaces (Yang, 2012).



$$\chi_e = F - E + V$$

Figure 4. The Euler characteristic χ_E of a body. It is determined by the number F of surfaces ("faces"), number V of points ("vertices") and number E of lines ("edges") connecting the edges of a polyhedron within which the body is embedded. In the case shown $F=6$, $E=12$ and $V=8$ and $\chi_E = 2$.

Furthermore, the Poincare-Hopf theorem (do Carmo, 1976) claims

$$\chi_E = m_{\text{tot}}, \quad (2)$$

where m_{tot} stands for the total winding number of point topological defects (TDs) (Kurik & Lavrentovich, 1988) within the shell. TDs refer to the points where the order is not uniquely defined. There are characterised by the winding number m , which equals number of total rotations of the field on encircling the defect center once counterclockwise. Examples of TDs bearing $m = 1$ and $m = -1$ are shown in **Figure 5a** and **Figure 5b**. In 2D systems the winding number is equal to the 2D topological charge and is a conserved quantity. TDs bearing positive and negative values of m are commonly dubbed *defects* and *antidefects*, respectively. Namely, a pair $\{\text{defect}, \text{antidefect}\}$ could mutually annihilate into a defectless state, characterised by $m = 0$. **Figure 5c** illustrates a pair $\{\text{defect}, \text{antidefect}\}$ prior to annihilation. Namely, TDs are energetically expensive. Consequently, in general systems tend to be defectless, i.e., $m_{\text{tot}} = 0$. On a flat surface the pair shown in **Figure 5c** experiences an attractive force which would drive the system into a defectless, spatially homogeneous state. Note that due to energy reasons systems tend to form TDs bearing minimal winding numbers m_{min} (Kurik & Lavrentovich, 1988; Rosso et al., 2012). In case of polar and axial order it holds $|m_{\text{min}}| = 1$ and $|m_{\text{min}}| = 1/2$, respectively.

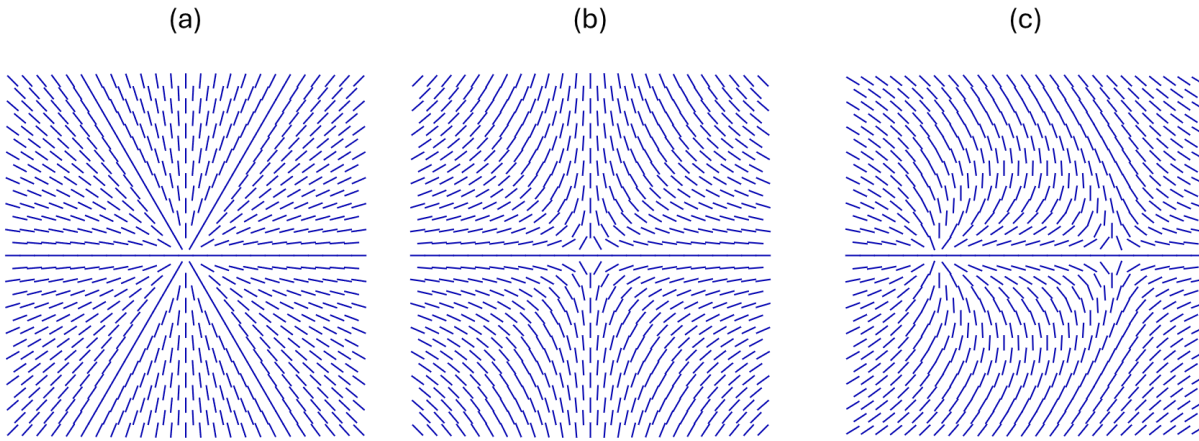


Figure 5. Topological point defects on a 2D flat surface. Single TDs characterised by (a) $m=1$ and (b) $m=-1$. (c) A pair $\{defect, antidefect\} = \{m=1, m=-1\}$ which is topologically equivalent to a defectless pattern.

However, closed shells exhibit in general surface patches that impose frustrations on ordering. This is clearly illustrated in **Figure 6**. In this case a spherical *shell* of radius R hosts orientational order. But due to the finite Gaussian curvature $K_g = 1/R^2$ the field could not be everywhere mutually parallel (such a case would be realised on a flat surface). Consequently, two $m=1$ TDs are formed at the poles of the structure, yielding $m_{tot} = 2$. Note that $K_g = 1/R^2$ yields $1/(2\pi) \oint K_g dA = 2$, fulfilling the GB-PH theorems (Eq.(1) and Eq.(2)).

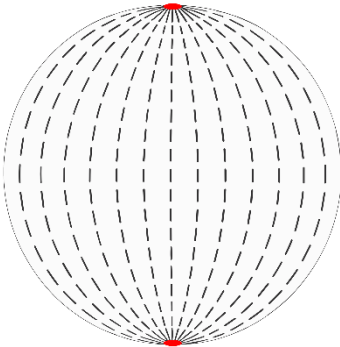


Figure 6. Topological point defects on a curved 2D spherical surface. The orientational pattern shown possesses two $m = 1$ point defects.

Experimentally, validity of these theorems could be relatively easily verified in nematic liquid crystals (NLCs) (Gharbi et al., 2013; Lopez-Leon et al., 2011). Namely, TDs in NLCs could be relatively easily created, manipulated and identified by polarising microscopy. Consequently, most of such studied are carried out in LCs. Some typical examples of resulting configurations are shown in **Figure 7**. These defect patterns were calculated numerically for a given *shell* geometry (Jesenek et al., 2015). One sees that in this case there are four $m=1/2$ on a spherical shell. Namely, the PH theorem requires $m_{tot} = 2$ and due to energy reason, the total winding number is distributed to 4 TDs bearing $m_{min} = 1/2$. Furthermore, in dumbbell-like shape one observes 8 TDs bearing $|m|=1/2$. There are two *antidefects* bearing $m = -1/2$, thus $m_{tot} = 2$ as requested for spherical topology. Note that in this case two additional pairs $\{defect, antidefect\} = \{1/2, -1/2\}$ are formed because of energy reasons. Namely, regions exhibiting $K_g > 0$ and $K_g < 0$ attract TDs bearing $m > 0$, and $m < 0$ (Jesenek et al., 2015) respectively.

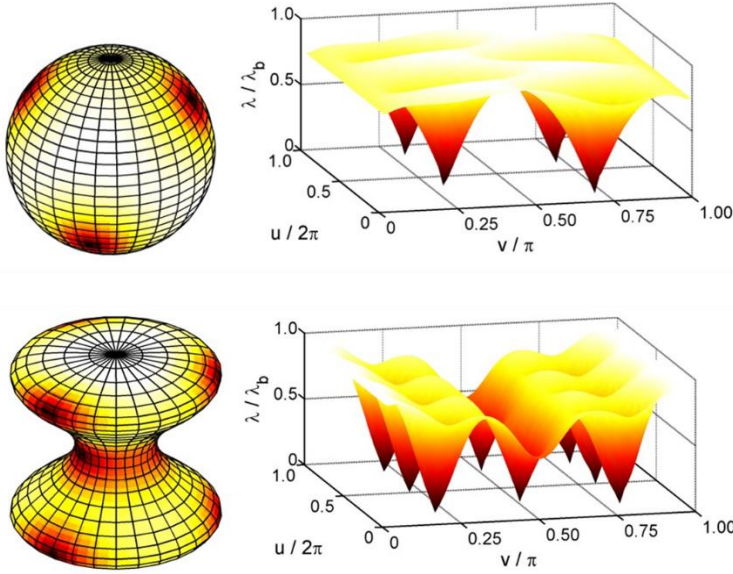


Figure 7. Topological point defects on a spherical (top panel) and dumbbell-like (bottom panel) shell. Left column: 3D view of surfaces hosting TDs. Right column: 2D view within the (u, v) plane, where u and v describe the azimuthal and zenithal angle describing the 2D shapes.

3. Conclusions

In the previous section we presented GB-PH theorems on geometric objects. We illustrated how these conservation laws are fingerprinted in topological invariants that remain unchanged under continuous transformations. However, similar behaviours could be observed also in other systems where 2D geometric shapes are described by some effective 2D physical field. For example, let us consider similar invariant physical properties for electrons in crystals. Their states are described by the Bloch wave function (Avron et al., 2003), $\psi = u_k e^{i\vec{k}\cdot\vec{r}}$. Here $u_k(\vec{r})$ exhibits the crystals translational symmetry and $\vec{k}$ describes the electron momentum $\vec{p} = \hbar\vec{k}$ with which electron hops from one atom to another and $\hbar = h/(2\pi)$. The associated de Broglie wavelength $\lambda = 2\pi/k$ cannot be smaller than the distance between neighbouring crystal cells. Consequently, the space of different momenta is restricted to the 1st Brillouin zone. Its shape reflects the arrangement of crystal atoms and plays the role of geometric space. Note that Brillouin zone exhibits periodic boundary conditions. To express topological theorem described in the previous section one needs to find an analogue of geometrical curvature. For this purpose one introduces Berry connection which is determined by u_k and related Berry curvature K_B , where details are given in (Avron et al., 2003). The resulting analogous topological conservation theorem reads

$$\frac{1}{2\pi} \oint K_B dA = n, \quad (3)$$

where $dA = d^2\vec{k}$ is the “surface” element of the 1st Brillouin zone and n is an integer, dubbed the Chern number. Furthermore, Avron et al. describe how similar approach is describing electrons in typical Hall geometrical setup show in **Figure 2** (Avron et al., 2003). This case illustrates how geometric topological theorems are present in a wide variety of physical phenomena where the relevant “shape geometry” could be described by some abstract physical field.

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Research

Chaotic Metasurfaces

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Abstract:

Metasurfaces are thin, artificially structured arrays of meta-atoms that allow manipulation of optical fields on a subwavelength scale. In this work, we show the emergence of chaotic-like behavior in a simplified model of a metasurface with spatiotemporal dynamics. The system is modeled on three levels. First, the incident light is described using a random walk model to introduce temporal phase fluctuations. Second, the metasurface is treated as an anisotropic structure that introduces spatially varying amplitude and phase shifts. Finally, the resulting field is propagated using Fresnel diffraction to obtain the output intensity. We demonstrate that this simple model exhibits signatures of chaotic-like dynamics, including the formation of attractors and characteristic relationships between frequency components and the power spectral density. These results highlight how complex behavior can emerge from simple optical systems.

Keywords: Metasurfaces; Random walk; Chaos

1. Introduction

1.1. Metasurfaces

Metasurfaces are artificially engineered two-dimensional structures composed of arrays of meta-atoms, which are subwavelength scatterers. They enable local control of electromagnetic waves, enabling manipulation of phase, amplitude, and polarization on subwavelength scales. This makes it possible to shape optical wavefronts in ways not achievable with conventional optics (Hu et al., 2021).

1.2. Classification of Metasurfaces

Metasurfaces can be classified based on their response and time dependence (**Figure 1**) (Xu et al., 2021). Passive metasurfaces have a fixed response determined by their geometry and do not require external energy, while dynamic metasurfaces can change their response in time but require external control. We can also distinguish whether the incident light is time-dependent or time-independent. In this work, we focus on a passive metasurface with time-dependent input, leading to spatiotemporal dynamics in the output.

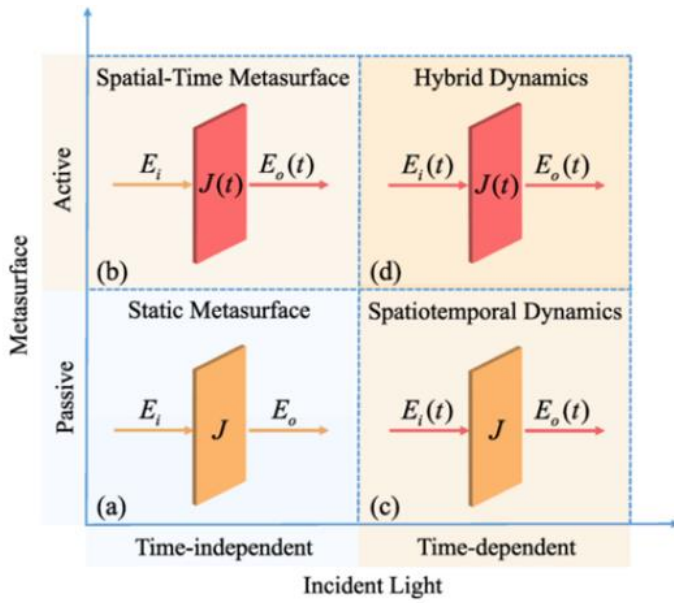


Figure 1. Classification of Metasurfaces (Xu et al., 2021). We can classify metasurfaces by whether they are passive or active in design (y-axis) and whether the incident light is time-dependent or independent (x-axis). The bottom row displays the passive metasurfaces, whose response is determined by geometry and cannot be changed. The top row shows the active metasurfaces whose configuration can be changed dynamically, but it needs an external source of energy. In the first column, the incident light is time-independent, and in the second column, the incident electromagnetic waves are time-dependent. The main focus of this work is the spatiotemporal dynamic metasurfaces that are passive, but the incident light is time-dependent.

2. Modeling

2.1. Incident light

We model the incident light $\mathbf{E}_{\text{in}}$ as having a time-dependent polarization angle $\theta(t)$:

$$\mathbf{E}_{\text{in}}(t) = (\cos \theta, \sin \theta), \quad (1)$$

which evolves as a random walk:

$$\theta(t + dt) = \theta(t) + \sqrt{2Ddt}\xi(t), \quad (2)$$

where D is a diffusion coefficient, dt time step, and ξ Gaussian white noise (Romanczuk et al., 2012). At each time step t , the new angle at $t + dt$ is given by its previous value plus a random increment, controlled by a diffusion coefficient D . If D is large, the signal becomes noisier; if it is small, the evolution is smoother. The variance of the fluctuations grows proportionally to $2D dt$. In **Figure 2**, we see how the polarization angle evolves over

time, and in **Figure 3**, how this determines the electric field components ($E_x = \cos \theta(t)$) and ($E_y = \sin \theta(t)$), and their evolution over time. Since the input is stochastic, each realization is different.

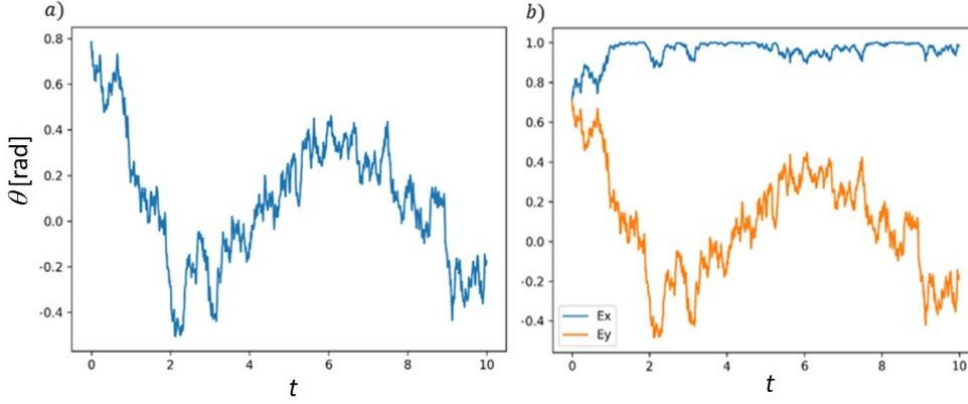


Figure 2. a): Evolution of the angle of the polarization ($\theta(t)$) with time step t . b) Evolution of electric field components (E_x blue line and E_y orange line)

2.2. Anisotropic metasurface

We model the metasurface as an optical element that transforms incoming light as follows:

$$\mathbf{E}_{\text{out}} = \mathbf{J}(\mathbf{r})\mathbf{E}_{\text{in}}, \quad (3)$$

where we use a position-dependent Jones matrix $\mathbf{J}$ (Rubin et al., 2025),

$$\mathbf{J} = \begin{pmatrix} J_{11}(x_1, y_1) & J_{12}(x_1, y_1) \\ J_{21}(x_1, y_1) & J_{22}(x_1, y_1) \end{pmatrix}, \quad (4)$$

with $\mathbf{r} = (x_1, y_1)$.

Each meta-atom introduces a local transformation that modifies the amplitude and phase of the electric field (**Figure 3**). The diagonal elements affect each component of polarization independently, while the off-diagonal elements introduce coupling between components, leading to cross-polarization. This polarization mixing is essential for generating complex behavior in the system. Therefore, each meta-atom introduces a different phase shift, resulting in a spatially varying wavefront, and this spatial disorder plays an important role in creating complex interference patterns.

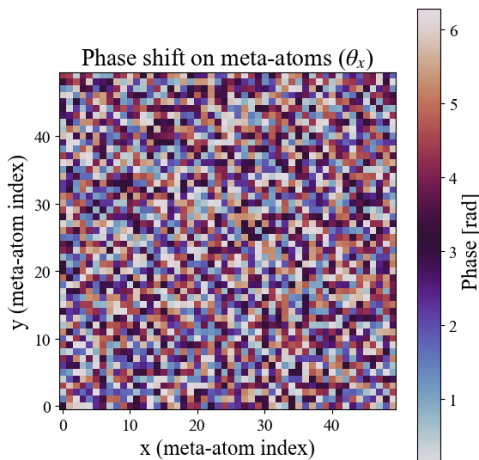


Figure 3. Phase shift on meta-atoms for x - component of angle θ (θ_x). x and y are the positions on the modelled metasurface. The colour represents the phase shift on each pixel, which corresponds to a meta-atom. Phase changes from 0 to 2π randomly for each meta-atom.

2.4. Intensity calculations

We analyze the light intensity I , which is proportional to the squared magnitude of the electric field $\mathbf{E}$:

$$I \propto |\mathbf{E}|^2. \quad (5)$$

First, the intensity is calculated directly at the metasurface. The field $\mathbf{E}_f$ at distance z from the metasurface is then evaluated using the Fresnel diffraction integral (Born, 1999):

$$\mathbf{E}_f(x, y) = \frac{e^{ikz}}{i\lambda z} \iint \mathbf{E}(x', y') e^{\frac{ik}{2z}((x-x')^2 + (y-y')^2)} dx' dy', \quad (6)$$

where λ is the wavelength of the incident light, (x', y') represent coordinates at the metasurface plane, and (x, y) coordinates represent position at a distance z from the metasurface. Since the observation screen is located in the near field, we use Fresnel diffraction, which describes the propagating in this regime. Light from different points on the metasurface acquires different phases during propagation and interference between them.

3. Results

3.1. Intensity directly after the metasurface

First we calculate the intensity directly at the metasurface. Using a diagonal Jones matrix (Eq. (4)), where off-diagonal elements are set to zero, there is no polarization mixing (**Figure 4a**), and the light intensity remains uniform. When off-diagonal terms are included (Eq. (4)) (**Figure 4b**), polarization components mix, leading to spatially varying intensity patterns. This shows that polarization coupling introduces spatial complexity.

3.2. Intensity distance z after propagation

Next, we calculate the light intensity after propagation. In both cases, interference leads to spatial variations. Even when the light intensity is uniform at the metasurface (**Figure 4a**), propagation introduces interference (**Figure 5a**). When polarization mixing is included, the patterns become even more complex (**Figure 5b**). Each pixel corresponds to a meta-atom, and we can see how the pattern continuously changes.

3.3. Temporal dynamics

We analyze the temporal behavior of the output intensity. Firstly, we plot the intensity at one time step versus the next, giving a phase-space representation of the dynamics (**Figure 6a**). We observe strong correlations between consecutive steps, resulting in an attractor-like structure. In the **Figure 6b**, we show the power spectral density. This is obtained by taking the Fourier transform of the intensity signal and computing the squared magnitude, which gives the power at each frequency. In log-log scale, the spectrum shows an approximately linear decay, indicating broadband frequency content and complex temporal behavior.

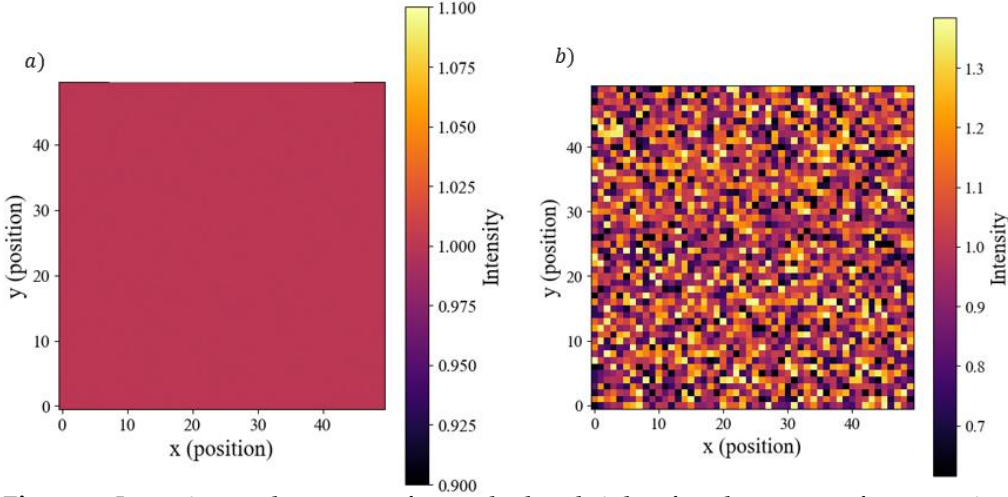


Figure 4. Intensity on the metasurface, calculated right after the metasurface. x -axis and y -axis represent the position at the metasurface. Each pixel is a modelled meta-atom on the metasurface. The intensity is represented with the color bar. a) The intensity of the each metaatom right after the metasurface, where the phase shift on the metasurface is modelled with diagonal Jones Matrix. b) Phase shift is modelled with full Jones matrix, allowing coupling between the components.

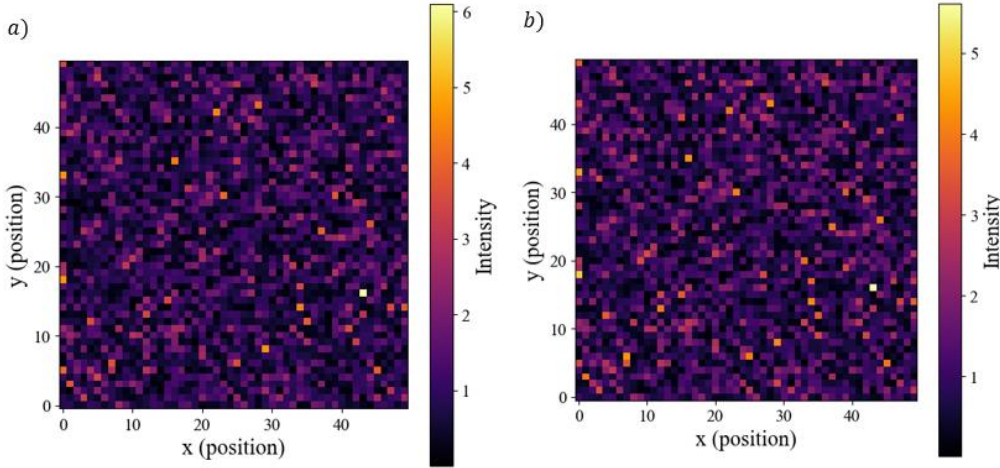


Figure 5. Intensity on the metasurface, calculated at distance z after the metasurface with Fr diffraction. x - and y -axis represent the position the metasurface. Each pixels is a modelled metaatom on the metasurfaces. The intensity is represented with the color bar. a) The intensity of the each metaatom at distance z after the metasurface, where the phase shift on the metasurface is modelled with diagonal Jones Matrix. b) Phase shift is modelled with full Jones matrix, allowing coupling between the components.

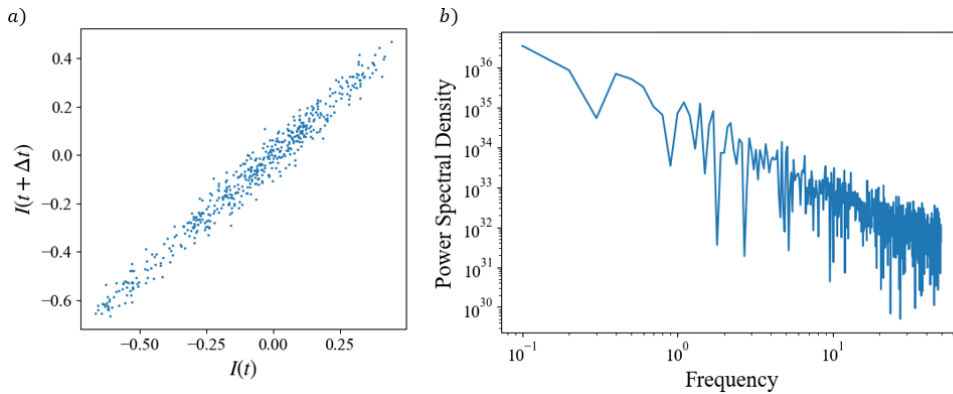


Figure 6. a) Intensity plotted at time step t on the x -axis vs the next time step $(t + \Delta t)$ on the y -axis, resulting in a chaotic like structure. b) Power spectral density (PSD) of the Intensity plotted at each frequency (x -axis), calculated by firstly taking the Fourier transform of the Intensity at time t , and calculating power at each frequency.

3.4. Poincare spheres

Polarization can be represented using the Poincare sphere (Figure 7). The polarization state is described using the Stokes parameters and corresponds to a point on the sphere. Linear polarization lies on the equator, circular polarization at the poles, and elliptical polarization elsewhere (ScienceDirect Topics, 2026). The input remains mostly linear (Figure 7a), while the output explores a broader region, indicating more complex polarization states (Figure 7b).

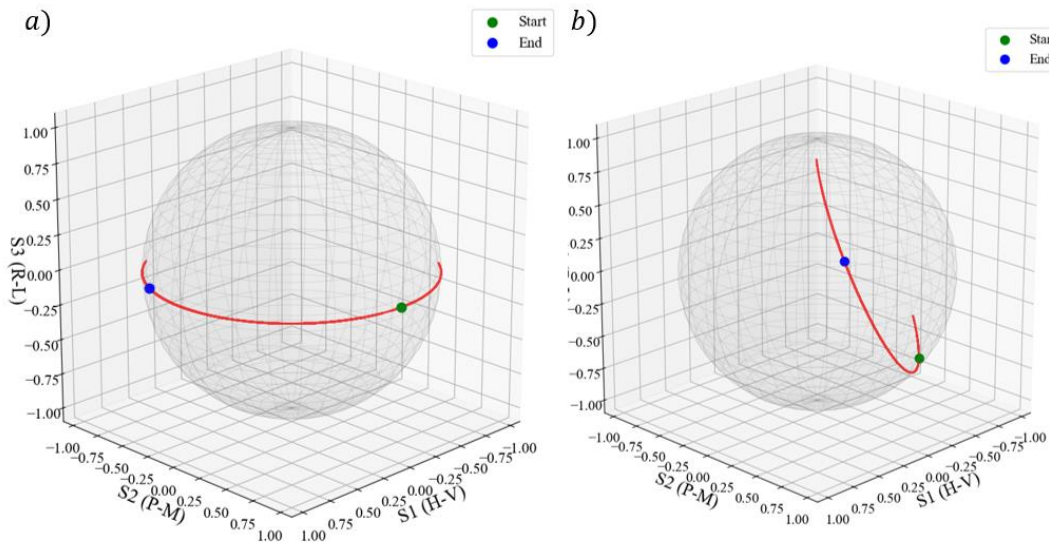


Figure 7. Poincare sphere representing polarization of the a) input field and the b) output field. On the x -axis is $S1$ Stokes parameter, on the y -axis is $S2$ Stokes parameter and on the z -axis is $S3$ Stokes parameter. Input field is linearly polarized and output field is elliptically polarized.

4. Conclusions

In this work, we have shown that a simple model combining stochastic input, spatial phase modulation, and propagation can produce rich and complex behavior. The interplay between polarization mixing and interference leads to structured, dynamic output patterns. This demonstrates how metasurfaces can act as simple platforms for generating complex, chaotic-like optical dynamics.

Conflicts of Interest: The authors declare no conflict of interest.

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Research

Percolation in electrocaloric applications

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Abstract:

We study percolation of 3D anisotropic nanoparticles immersed in nematic liquid crystal liquid (NLC). By adjusting the parameters of the simulation, we simulate ordering effects of an external electric field on NLC. We numerically calculate how shapes of particles and the degree of thermal fluctuations impact the percolation threshold. By replacing thermal diodes in an electrocaloric transistor with diodes with nanoparticles close to the threshold we create a self-regulating system which could be exploited in future electrocaloric devices.

Keywords: Liquid crystals; Phase transitions; Percolation; Electrocaloric effect

1. Introduction

Percolation theory is a theory that describes the behavior of a network when nodes or links are added (Broadbent & Hammersley, 1957). Nodes and links can be replaced with particles and their connection. This is a geometric type of phase transition, since at a critical number of particles the small, disconnected clusters of particles connect into large, connected spanning clusters. When such clusters connect two sides of a system, we say that the system percolates thus inducing a phase change.

The ElectroCaloric Effect (ECE) is a phenomenon where one triggers reversible temperature changes in a system by adiabatically applying or removing an electric field (Klemenčič et al., 2019). The effect exploits the interplay between thermal (S_T) and orientational (S_O) contributions to the total entropy $S = S_T + S_O$. In an adiabatic change increased (decreased) alignment of electric dipoles fingerprints in reduced (increased) S_O and consequently in decreased (increased) S_T . The latter change is fingerprinted in the temperature change ΔT within the electrocaloric material. An electrocaloric effect transistor is an emerging solid-state thermal management device that utilizes ECE instead of conventional cooling liquids or gases. It controls thermal flux via electrical signals.

In this contribution we show how percolation could be exploited in a representative ECE device.

2. Material and Methods

2.1. Percolation

We illustrate percolation on a simple 2D lattice consisting of squares which are either occupied or empty. In the initial state all the squares are empty. Then we occupy each of the squares of the system with the probability $p \in [0,1]$ (**Figure 1**) and we perform ensemble averaging. On gradually increasing p starting from $p = 0$ the system becomes percolated at the critical concentration p_c , dubbed percolation threshold. At p_c long-range connectivity first appear and the resulting structure exhibits fractal pattern (Mischenko et al., 2006).

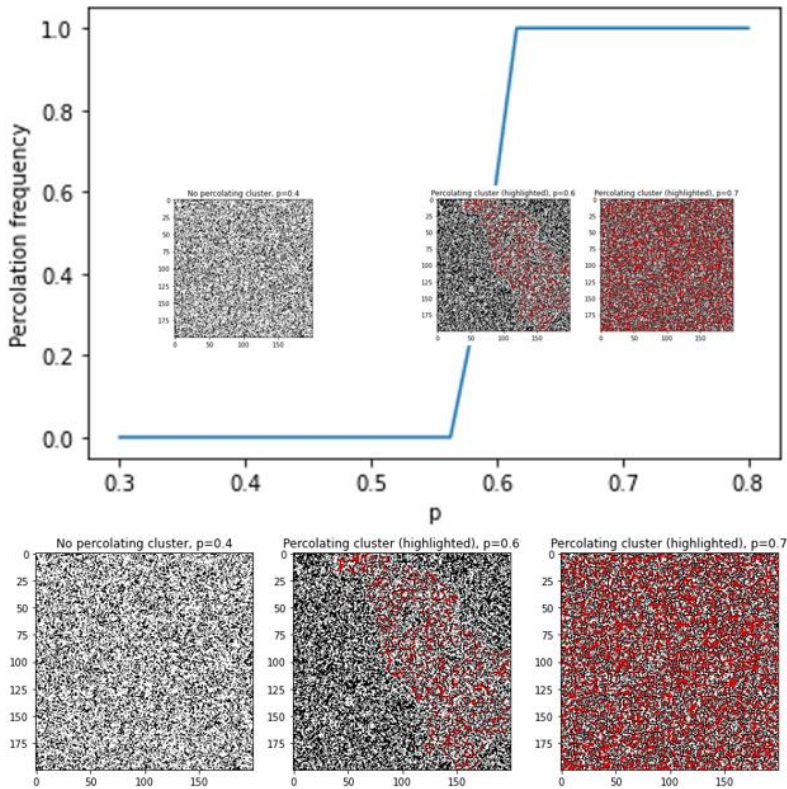


Figure 1. Percolation of 2D square lattice size 200x200. Top: Percolation frequency versus the relative number of active occupied sites (0-no occupied, 1-all occupied). Bottom: Three distributions of active sites with red coloured sites marking a cluster that successfully percolates from the bottom to the top of the lattice. Values of p are: left: $p = 0.4$, middle: $p = 0.6$, right: $p = 0.7$.

In our simulation, we consider 3D system of square objects characterised by lengths (a,b,c). We allow them to randomly fluctuate about one axis up to the maximal angle θ_m which we vary in simulation. We study at which concentration of these squares becomes percolated as a function of geometric parameters (a,b,c) and θ_m . To calculate a percolation threshold for given conditions we average results over 100 simulations.

2.2. Electrocaloric effect

The Electrocaloric effect is a phenomenon where applying or removing an electric field (E) causes a reversible temperature change under adiabatic conditions (**Figure 2**). This means that the entropy change (ΔS) is zero, therefore any change in entropy because of changing temperature (ΔS_T) must have an equal and opposite change in entropy of order (ΔS_O) (**Figure 3**):

$$\Delta S = \Delta S_T + \Delta S_O = 0. \quad (1)$$

A typical ECE device and its different stages within one cycle (in it the external electric field is switched on and off) is shown in **Figure 4**. Pay attention to thermal diodes, which control temperature flows to the regions A and B which are in contact with the ECE material. When these diodes are activated, they enable the heat transfer from ECE to the surroundings.

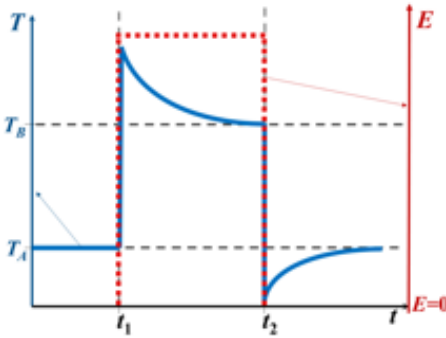


Figure 2. Electrocaloric effect. At t_1 the electric field is activated and the temperature increases from T_A to over T_B . At t_2 the electric field is deactivated and the temperature decreases from T_B to under T_A .

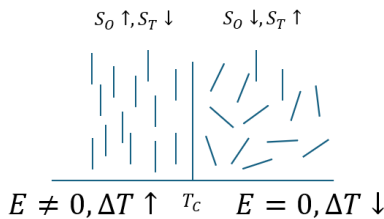


Figure 3. Changing order with electric field (E) and the changes in entropy (S_T , S_O) and temperature (ΔT).

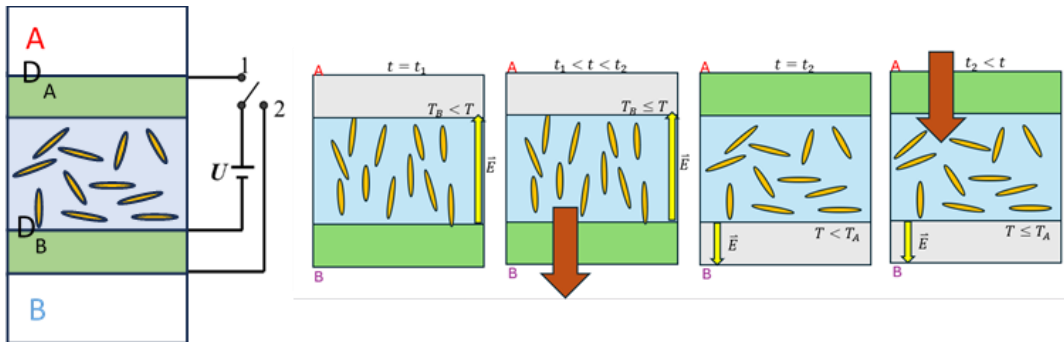


Figure 4. Electrocaloric transistor and phases of it working. A, B are outer materials, D_A , D_B are thermal diodes. The red arrow represents heat transfer, the yellow arrow represents the direction of the electric field. Times corresponds to those in **Figure 2**.

3. Results

Of our interest is to identify geometry of square objects which would exhibit large enough change in percolation threshold p_c on varying θ_m . Here $p = V_n/V_{\text{tot}}$, V_n stands for the total volume of nanoparticles (NPs) and V_{tot} is the total volume of the system. We assume that NPs are immersed in essentially spatially homogeneous nematic liquid crystal (NLC) phase. The degree of its NLC orientational order $\langle P_2^{(NLC)} \rangle$ impacts the degree of orientational order $P_2^{(NP)}(\hat{e}_i, \hat{n})$ of anisotropic NPs (van der Schoot et al., 2008). Here P_2 stands for the second order Legendre polynomial, $\langle P_2^{(NLC)} \rangle \in [-0.5, 1]$ measures the degree of NLC order, $\langle \dots \rangle$ stands for ensemble average, the unit vector $\hat{n}$ determines the average orientation of global NLC order, the unit vector $\hat{e}_i$ describes the long axis orientation of i-th nanoparticle and the overbar $\overline{(\)}$ indicates the spatial average. $\langle P_2^{(NLC)} \rangle = 1$ fingerprint saturated NLC order where NLC molecules are rigidly aligned along $\hat{n}$. In the case of $\langle P_2^{(NLC)} \rangle = 0$ NLC order is melted into ordinary liquid (the so called isotropic) phase. In our simple model we mimic the impact of $\langle P_2^{(NLC)} \rangle$ on $P_2^{(NP)}$ by θ_m . Therefore, larger values of θ_m fingerprint lower order of $\langle P_2^{(NLC)} \rangle$ and consequently lower degree of NPs global degree of order $P_2^{(NP)}$. One can tune degree of NLC order by an external electric field $\vec{E}$ (Kleman & Lavrentovich, 2003). Therefore, in our simulation different values of θ_m correspond to larger values of E .

In our simulations we systematically explored the impact of NPs geometry and θ_m on the percolation threshold p_c . We identified three geometries, described in Table 1, that exhibit large enough change of p_c on varying θ_m to be useful in practical applications. The simulation results are shown in Figure 5. Geometries of NPs are shown in the left panel of the figure and the corresponding characteristic lengths are given in Table 1. One sees that for "shape 2" the percolation threshold changes for 10% on varying θ_m from zero to 80 degrees. This relatively strong variation in p_c could be exploited in self-tunable thermal diodes in a typical ECE setup shown in Figure 4.

Table 1. Parameters of inserted objects.

Side	a	b	c	c/b	a/c
Object					
1	0.06	0.02	0.1	5	0.6
2	0.02	0.02	0.2	10	0.1
3	0.1	0.02	0.15	7.5	2/3 = 0.6667

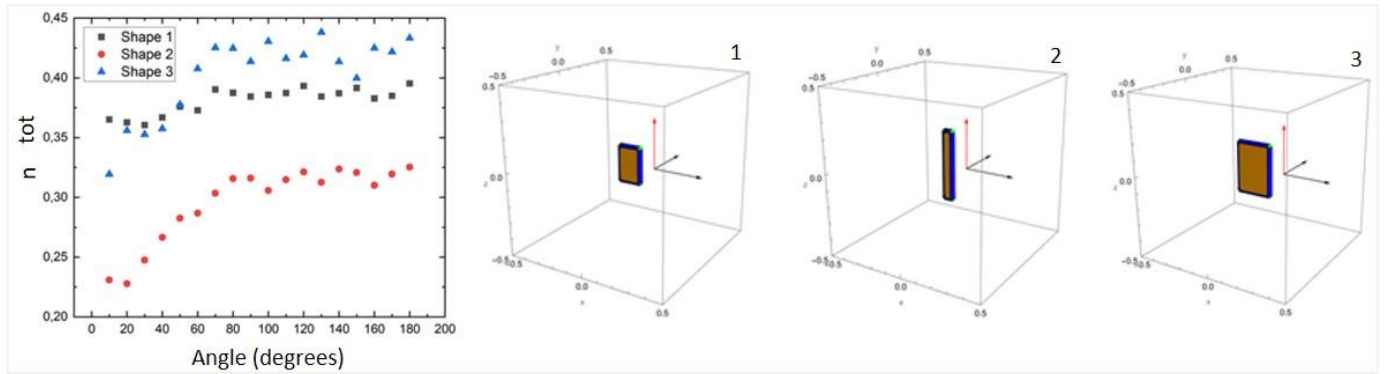


Figure 5. Objects and a graph of their percolation thresholds under different ordering strengths. Left: from top to bottom are shapes 1, 2, and 3 with the red arrow pointing in the direction of the z-axis. Right: percolation thresholds (volume of particles/total volume), depending on the permitted angle of deviation.

4. Discussion

The above studied percolation behaviour could be implemented as a self-regulating thermal diode in an ECE device, where a typical set-up is schematically presented in Fig.4. Namely, the thermal diodes in **Figure 4** consist of thermally conduction NPs in NLC matrix. If these NPs are percolated in a desired direction, then large enough heat flow could be established through the diode. This flow could be controlled via an external electric field $\vec{E}$ applied to diodes where a possible set-up is depicted in **Figure 4**. For this reason, the system should be designed as follows. In the absence of $\vec{E}$ fluctuations of NPs long axes should be large enough so that the NPs are percolated. In our simulations this would correspond to relatively large value of θ_m and $p > p_c$ (see **Figure 5**). If the field is applied, then it should switch the system in the nonopercolated regime. Therefore, in our simulations θ_m should be reduced to reach the condition $p < p_c$. Furthermore, one could design a self-regulatory diode which could be tuned to realise a desired temperature in a thermal reservoir. Therefore, if a desired temperature is crossed the diode should switch its thermal conducting state. For this purpose, a desired temperature should reflect in θ_m corresponding to $p = p_c$.

5. Conclusions

In this contribution we briefly described percolation theory and the electrocaloric effect. We described how we simulated percolation in a 3D system and presented the results of our simulation finding that the longer and thinner the object is in the direction of percolation the lower the percolation threshold and greater the effect of order in lowering the percolation threshold. We discussed how we can use the results of our simulations of percolation in diodes to control electrocaloric transistors and offered some possible uses as self regulating diodes. Further study in applying percolation theory on not only electrocaloric effect but also other phenomena to study critical values and various thresholds may lead to new discoveries or applications.

Conflicts of Interest: The authors declare no conflict of interest.

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Research

Power Law and q-Exponential Distribution in Football Goalscoring

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Abstract:

In the present study, we investigate the power law distribution as a model for heavy-tailed statistical distributions. We look at the main parameters of the power law model, as well as statistical methods used to test its applicability. The method is applied to goalscoring data from the top five European leagues from 1977/78 season to 2024/25 season on a season-by-season basis. The p -value is determined, and the results are plotted as a time series. We also consider the Tsallis entropy formalism, and focus on q-exponential distribution, which is a result of maximisation of Tsallis entropy. Particular attention is given to the parameter q . It is fitted to data of top scorers and players with most goals-and-assists (GA) of multiple national leagues across multiple seasons. The obtained values of q are found to be consistent with previously reported results. We also apply both distributions to lists of top scorers from multiple clubs. Finally, we discuss possible connections between the obtained statistical behaviour and tactical or structural aspects of modern football.

Keywords: Power law; Tsallis entropy; Q-exponential distribution; Football; Heavy tail distribution

1. Introduction

As the most popular sport in the world, football has long been the subject of research by many scientists, including physicists. Numerous studies have investigated statistical and tactical aspects of the sport. In (Greenthrough et al., 2002), distributions of goals scored by home teams, away teams, and both teams were analysed. In (Velasco Stock et al., 2022) an algorithm was adapted to predict the results of football matches. More recently, (Chacoma & Billoni, 2025) attempted to characterise playing styles using data-driven methods. In this article, we focus on the statistical properties of goalscoring. Specifically, we analyse goalscoring distributions among players and fit two candidate models: the power law distribution and the q-exponential distribution. Both distributions are particularly suitable for systems with heavy-tailed statistical behaviour, where extreme events occur more frequently than predicted by standard exponential models.

1.1. Power law

A power law is often used to describe systems with large variability spanning several orders of magnitude. We should first look into the cases where the power law would be applicable. For example, human height is concentrated around a characteristic value, with deviations larger than a factor of two being practically non-existent. In such cases, a power law description is unnecessary. In contrast, quantities such as city populations span several orders of magnitude, making power law models considerably more appropriate. In Section 2, we introduce the key parameters of the power law distribution and discuss statistical methods for testing its applicability. Power laws have previously been observed in football-related data; for instance, (Da Silva et al., 2013) showed that the relationship between team rank and points follows power law behaviour. We apply the power law model to the distribution of goals scored by players in the top five European leagues between the 1977/78 and 2024/25, as well as to all-time scoring records of multiple clubs.

1.2. q-exponential distribution

An alternative approach to modelling the observed distributions is provided by the q-exponential distribution, which generalises the standard exponential distribution. Just as the exponential distribution can be obtained, the q-exponential distribution is derived by maximising the Tsallis entropy, a generalisation of the standard Boltzmann-Gibbs entropy. The formalism was first proposed by Constantino Tsallis in 1988. We apply the q-exponential distribution to goalscoring distributions in multiple national leagues, as well as to all-time scoring records of multiple clubs.

2. Material and Methods

2.1. Power law

Let x be a quantity whose distribution we are interested in. Then we can describe a continuous probability distribution $p(x)$ such that:

$$p(x)dx = \Pr(x \leq X < x + dx) = Cx^{-\alpha}dx, \quad (1)$$

where X is the observed value, C is the normalization constant and α is the scaling exponent. However, since we are working with numbers of goals, we will need a discrete distribution. Here we write:

$$p(x) = \Pr(x = X) = Cx^{-\alpha}. \quad (2)$$

We must also note that this density diverges as $x \rightarrow 0$ (Clauset et al., 2009), so our distribution cannot hold for all non-negative values of x . We will define the lower bound for the power law behaviour by $x_{\min}$. One more thing, it is sometimes more useful to define a complementary cumulative distribution function (CCDF), defined as $P(x) = \Pr(X \geq x)$.

We will now look into estimating the values of α and $x_{\min}$. We first estimate α using the maximum likelihood estimator (MLE). The MLE for the continuous case is (Clauset et al., 2009):

$$\hat{\alpha} = 1 + n \left[\sum_{i=1}^n \ln \frac{x_i}{x_{\min}} \right]^{-1}. \quad (3)$$

For the discrete case, there is no closed-form expression. There is, however, an approximation, which works similarly to Eq. (3) and can be written as (Clauset et al., 2009):

$$\hat{\alpha} \approx 1 + n \left[\sum_{i=1}^n \ln \frac{x_i}{x_{\min} - \frac{1}{2}} \right]^{-1}. \quad (4)$$

This expression is found to be accurate to 1 % when $x \geq 6$.

We also need an accurate value for $x_{\min}$. If it is set too low, the fit includes data that do not follow a power law. If it is set too high, we lose good data. According to the method presented in (Clauset et al., 2007), we compare the distance between our observed data and the best-fit power law model. We use the Kolmogorov–Smirnov (KS) statistics, which measures the maximum distance between the two distributions:

$$D = \max_{x \geq x_{\min}} |S(x) - P(x)|. \quad (5)$$

Here $S(x)$ is the empirical distribution and $P(x)$ is the best-fit model power law. The optimal value for $x_{\min}$ is the one that minimises D .

Now we can calculate both of the desired parameters, but we still need to know whether the power law hypothesis is applicable to a given dataset. For confirmation, many studies rely on visual inspection, but this can be tricky, as many models can be visually similar to the power law. A better approach would be to use a goodness of fit test. The idea is to measure the difference between the data and the best-fit model, using the KS statistics. We also measured the difference between the synthetic data sets drawn from the same model. Then we defined the p -value as the fraction of the difference between the synthetic data sets that were larger than the difference between the data and the best-fit model. If the p -value were large, i.e. close to 1, then the power law hypothesis would be considered plausible. If the p -value were small, then our data would very unlikely follow the power law. We adopted a cut-off value of 0.1, the same as (Clauset et al., 2009). Some authors took the cut-off value of 0.05, but it is argued in (Clauset et al., 2009) that this would be too lenient, as it would let through some datasets that are very unlikely to follow the power law. We also noted that the p -value defined above did not agree with the results of some experimental studies. Therefore we wished to test our hypothesis stating that a small p -value means that the null hypothesis is very unlikely to be correct. Therefore, small p -values are desired. Here, the p -value quantifies the plausibility of the power law hypothesis itself, so we desire larger values.

2.2. q -exponential distribution

The second model we consider is the q -exponential distribution, obtained by maximising the Tsallis entropy, which was proposed by Constantino Tsallis in 1988 (Tsallis, 1988):

$$S_q = k \frac{1 - \sum_{i=1}^W p_i^q}{q-1}, \quad (6)$$

where k is the Boltzmann constant, W is the total number of possible microscopic configurations, p_i are the associated probabilities and q is a real number. In the limit of $q \rightarrow 1$, this expression reduces to the Boltzmann–Gibbs entropy.

We are interested in the q -exponential distribution, written in the form (Malacarne & Mendes, 2000):

$$N(x) = N_0 (1 - (1 - q)\lambda x)^{\frac{1}{1-q}}, \quad (7)$$

where $N(x)$ is the number of players with x goals, and N_0 , λ and q are real parameters. In the limit of $q \rightarrow 1$, the q -exponential reduces to the standard exponential function. But since the parameters N_0 and λ are not directly known, we replace them with the ones we can

determine from our data. We can evaluate the equation (7) at $x = 1$ (Malacarne & Mendes, 2000):

$$N(1) = N_0(1 - (1 - q)\lambda)^{\frac{1}{1-q}}, \quad (8)$$

which allows us to express N_0 . We can also calculate the average number of goals scored by all players with at least one goal scored. We denote this quantity by m (Malacarne & Mendes, 2000):

$$m = \frac{\int_0^\infty xN(x)dx}{\int_0^\infty N(x)dx} = \frac{1}{\lambda(3-2q)}, \quad (9)$$

which can be used to express λ . Now we can rewrite equation (7) as:

$$N(x) = N(1) \left(\frac{1 - \frac{1-q}{(3-2q)m}}{1 - \frac{1-q}{(3-2q)m}x} \right)^{\frac{1}{q-1}}. \quad (10)$$

The only remaining free parameter is q , which is determined by fitting the model to the data. The key difference between the standard exponential distribution and the q -exponential distribution is that the latter decays more slowly for large values of x . Consequently, the q -exponential distribution assigns a higher probability to extreme events and is therefore better suited for describing heavy-tailed distributions, such as the distribution of goals scored. Larger values of q correspond to heavier tails and therefore to a higher probability of extreme events.

3. Results

We now apply both distributions to the collected data. The data were collected from publicly available databases provided by Transfermarkt (2025) and MNZ Maribor (2025).

3.1 Power law

We fitted the power law model to the distribution of the number of players that scored x goals in the top five European national leagues for every season from 1977/78 to 2024/25. Only the goals scored in national leagues and national and international cups were considered. The analysis was performed using the Python package *Powerlaw* (Alstott et al., 2014), which made our work easier. We analysed the p -values for individual seasons and their evolution over time. **Figure 1** shows the development of p -values. Out of 48 seasons, 24 produced distributions were consistent with a power law, while for the remaining 24 the hypothesis was rejected. More recent seasons appear to follow a power law distribution more frequently than the earlier seasons. Since 2004, only 6 seasons were found to deviate from the power law. Additionally, the most recent 7 seasons all follow the power law.

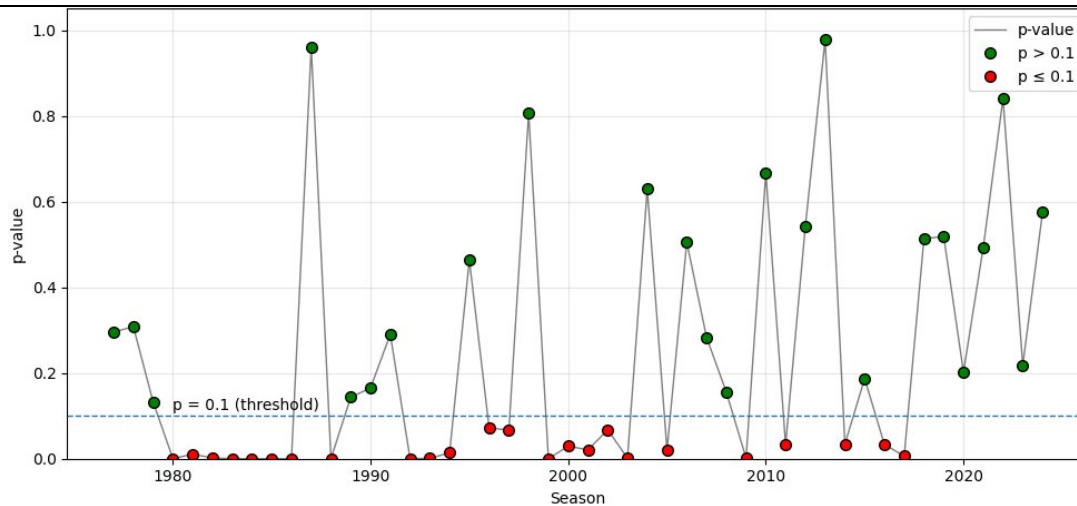


Figure 1. p -values through seasons from 1977/78 to 2024/25.

3.2 q -exponential distribution

We fitted the distribution of the number of players that scored x amount of goals in a season across multiple seasons of the English Premier League, Spanish La Liga, Slovenian Prva Liga, and the First MNZ Maribor League (the fourth tier of Slovenian football, also known as Golgeter Premium Liga) with expression (10). Only the goals scored in the leagues themselves were considered. As discussed previously, we paid special attention to parameter q . Previous studies found that $q = 1.33$ provides a good fit for multiple national leagues (Malacarne & Mendes, 2000). We tested this across multiple seasons of the aforementioned leagues (**Figure 2**). For all analysed leagues, the fitted values of q (also referred to as the Tsallis parameter) was close to 1.33, consistent with previous findings. The mean values were: 1.34 for the Premier League, 1.32 for LaLiga, 1.34 for Prva Liga and 1.35 for MNZ Maribor.

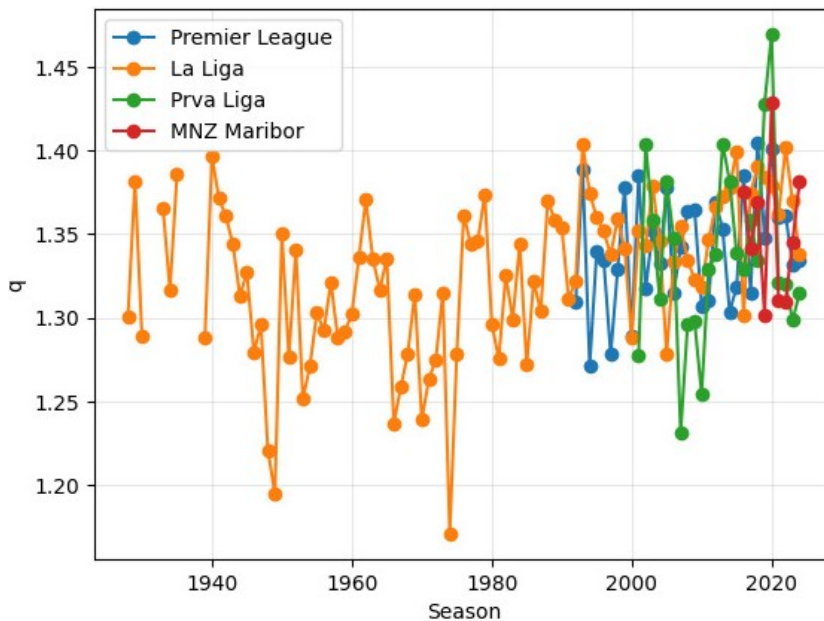


Figure 2. The values of q through seasons for Premier League, LaLiga, Prva Liga and MNZ Maribor. The two outliers from LaLiga are due to the small sample size of players during those times. The gap in LaLiga around 1940 is due to the civil war.

We also applied Eq. (10) to the distribution of goals and assists accumulated by players within a single season (further referred to as GA), as GA has become an increasingly important performance metric in modern football analysis. We then compared the fitted q -values obtained for goals alone with those obtained for GA. We also replaced MNZ Maribor with the Italian Serie A, since assist statistics are unavailable for the former league. The results are shown in **Figure 3**. At first sight, the values of q for GA are a bit smaller than for goals. The averages this time are 1.31 for the Premier League, LaLiga, and Prva Liga and 1.33 for Serie A.

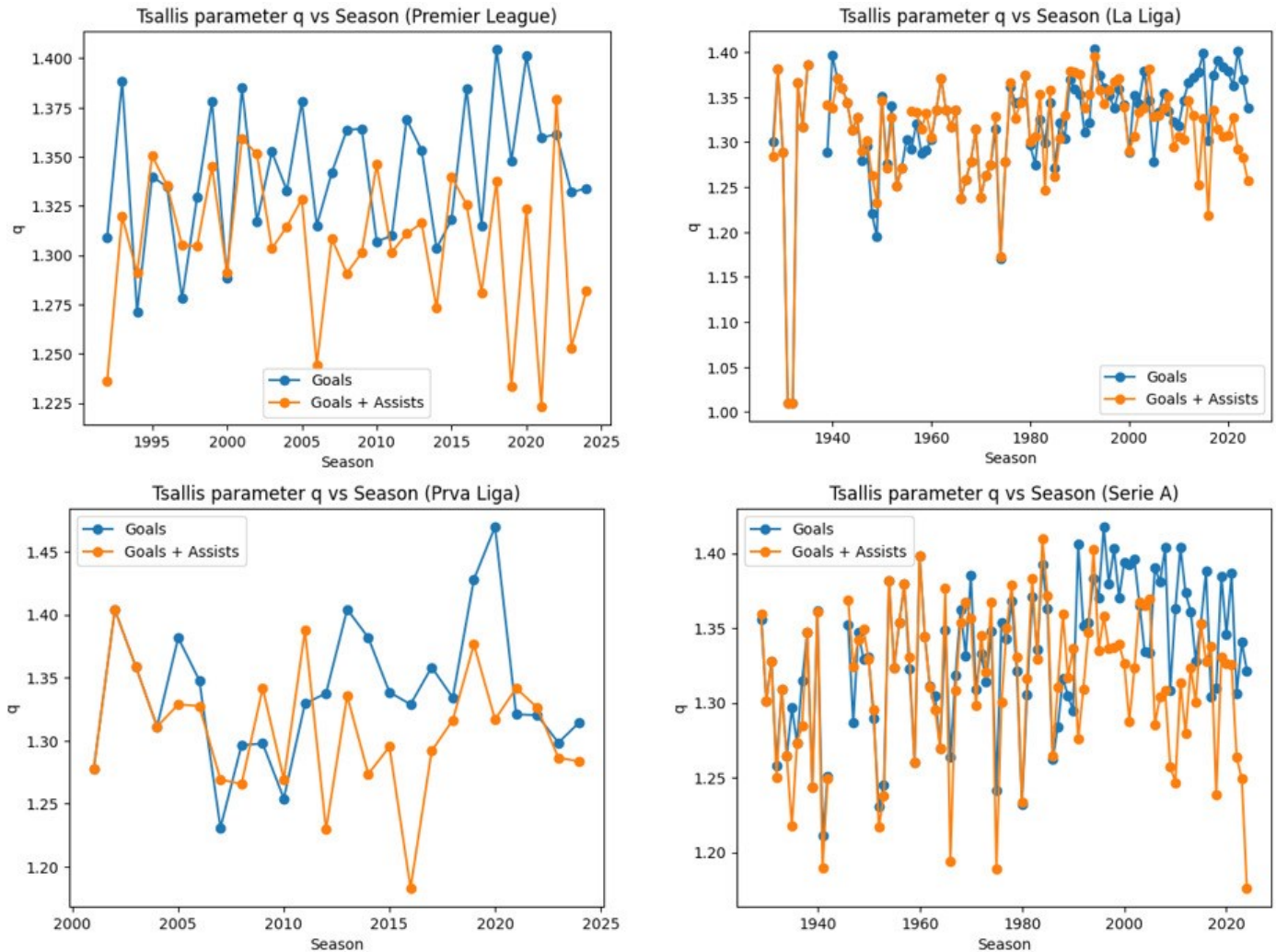


Figure 3. The values of q through seasons for Premier League, LaLiga, Prva Liga and Serie A. The two outliers from LaLiga are due to the small sample size of players during those times. The gaps in LaLiga and Serie A around 1940 are due to the civil war and the Second world war, respectively.

3.3 Scoring lists from clubs

We also fitted both models to the distributions of all-time goals scored for multiple clubs. The results are summarised in **Table 1**. The p -values indicate that only a small number of clubs can be adequately described by a power law distribution. The fitted q -values are generally larger than those obtained for seasonal goalscoring distributions.

Table 1. The values of p and q , obtained from all-time scoring lists of multiple clubs.

Club	p	q
Real Madrid	0.03	1.468
FC Barcelona	0.00	1.459
Atletico Madrid	0.04	1.449
Liverpool	0.13	1.451
Manchester United	0.01	1.449
Arsenal	0.00	1.451
Chelsea	0.09	1.446
Manchester City	0.00	1.437
Inter Milan	0.33	1.430
AC Milan	0.03	1.432
Juventus	0.02	1.437
Bayern München	0.07	1.457
Borussia Dortmund	0.04	1.437
PSG	0.28	1.437
Ajax Amsterdam	0.00	1.442
NK Maribor	0.00	1.41
NK Olimpija	0.00	1.42
NK Celje	0.00	1.37
Stoke City	0.22	1.418
Birmingham City	0.00	1.418
Exeter City	0.00	1.42
Peterborough	0.00	1.437
Al Nassr	0.56	1.37
Los Angeles Galaxy	0.00	1.435

4. Discussion

We now discuss the implications of our results. If we look back to **Figure 1**, we can see that in years after 2000, there have been considerably more seasons in which the distribution of goals scored by players can be described by a power law distribution. One possible explanation is the increasing tactical optimisation of modern football, where attacking patterns and player roles have become more structured and statistically efficient. Such optimisation may contribute to a stronger concentration of goals among a smaller subset of highly productive players, leading to distributions that more closely resemble a power law. Another possible contributing factor is the introduction of the video assistant referee (VAR) system. Interestingly, all seasons following the introduction of VAR in 2018 were found to be consistent with power law behaviour.

Turning to the q -exponential distribution, we examine the differences between the fitted values of q for goalscoring distributions (q_g) and distributions of GA (q_{ga}) (**Figure 3**). We can see that there are periods when q_g is noticeably larger than q_{ga} . One possible interpretation is that the difference between q_g and q_{ga} may be related to the overall competitive strength of a league. For Premier League, the difference became big in the late 2010s, during periods in which English clubs were particularly successful in European competitions, as well as in the late 2000s, when its clubs were also performing well in European competitions. Similar trends appear in the other analysed leagues. LaLiga has been considered as arguably the best league in the world since the late 2000s, when the difference between

both values of q appeared. For Serie A the difference appeared in the 1990s and early 2000s, during the period in which Italian clubs achieved some of their strongest European results, and for Prva Liga, the gap appeared in 2010s, during the league's most successful period in European qualification competitions.

The fitted values of q for individual clubs (Table 1) suggest that the most successful clubs tend to exhibit larger values of q . Real Madrid, FC Barcelona, and several other elite clubs have values around 1.45 or 1.46, while some less successful clubs have values around 1.42 or even lower. In terms of the q -exponential model, larger values of q correspond to heavier tails, indicating a broader distribution of high-scoring players.

5. Conclusions

We have presented the theoretical background for the power law and q -exponential distribution in the model for football goalscoring. The distributions were fitted to the number of goals scored by players across multiple seasons and leagues, and the evolution of the fitted parameters through time was analysed. For the top five European leagues, the power law model was found to be applicable in approximately half of the analysed seasons. Furthermore, power law behaviour appeared more frequently in recent years, which may reflect increasing tactical and statistical optimisation in modern football. The q -exponential distribution was applied to both goalscoring and goals-and-assists (GA) distributions across several national leagues. The obtained values of the Tsallis parameter q remained close to previously reported values, while the observed differences between the parameters for goals and GA may be related to the competitive strength and overall quality of a league. Both models were applied to all-time club scoring records. Only a small number of clubs produced a distribution consistent with the power law, while the fitted q -values were generally larger for more successful clubs, indicating heavy-tailed scoring distributions.

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Research

Assessment of Extracellular Particles from *Chlorellae sorokiniana* Culture and Suspension of Washed Erythrocytes Concentrated by Ultrafiltration

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Abstract:

Extracellular particles (EPs) are sub-micron sized particles found in biological samples containing cells. While the International Society for Extracellular Vesicles (ISEV) recommends assessment with different orthogonal methods, the technologically advanced golden standard methods for harvesting and assessment are sought. Here we report of harvesting and assessment of EPs from two types of samples: suspension of aged, washed erythrocytes and *Chlorellae sorokiniana* culture. EPs were harvested by reverse ultrafiltration and assessed by Scanning Electron Microscopy (SEM), Interferometric Electron Microscopy (ILM) and UV-vis spectroscopy. We assessed pellets and supernatants of the erythrocyte and microalgae samples (PE, SE, PA, SA, respectively). Average number density of 40 x diluted EPs in PE was $(1.16 \pm 0.09) \pm 10^9/\text{mL}$ and number density of non-diluted EPs in PA was $(0.44 \pm 0.02) \pm 10^9/\text{mL}$. Average hydrodynamic diameter in SE was (174 ± 72) nm, and in PA it was (252 ± 7) nm. The supernatants did not present notable amount of EPs. Absorption at 260 nm/280 nm in 4 x diluted SE was $(3.68 \pm 0.04)/(2.64 \pm 0.04)$ arbitrary units (a.u.), in 40 x diluted PE it was $(22 \pm 0.2)/(25 \pm 0.0)$ a.u., in undiluted PA it was $(0.31 \pm 0.11)/(0.29 \pm 0.13)$ a.u. and in undiluted SA it was $(0.03 \pm 0.06)/(0.04 \pm 0.01)$ a.u.. Algae samples contained singular lumps of self-assembly but were poor with proteins and nucleic acids. We conclude that the processing essentially determines the identity of the sample which implies that in order to reach repeatability, the methods should be minimally aggressive to the material, should be made on the same sample and should for this purpose use a minute amount of the sample to enable assessment with other methods. The methods used in this work composed such experimental pipeline that enabled assessment of the samples of in vitro aged erythrocyte EPs and EPs from microalgae culture within hours.

Keywords: Extracellular vesicles; Microalgae; Nanoalgaosomes; Erythrocyte microvesicles; Coacervates; Self-assembly

1. Introduction

EPs are a subject of increasing interest as they are considered mediators of intercellular communication and therefore vectors of vital processes in living organisms (Yanez Mo et al., 2015; Adamo et al., 2021; Welsh et al., 2024; Suresh et al., 2025; Shank et al., 2026; Liang-supree et al., 2026). There is great interest in using EPs in various scientific fields, and medical and industrial applications. The transition of extracellular particles (EPs) from laboratory discoveries to clinical and industrial applications is heavily bottlenecked by current isolation and characterization technologies (Welsh et al., 2024). Current harvesting methods struggle to produce a considerable volume of the sample, and separate EPs with required properties from similar-sized particles. Common contaminants include lipoproteins, protein aggregates, and cellular debris, which can skew downstream analysis. Furthermore, some conventional harvesting techniques suffer from significant EPs' loss. This low recovery rate is particularly problematic for scarce or precious clinical samples like patient plasma or rare cell cultures. Protocols lack uniform automation, leading to high batch-to-batch variability. Results often vary drastically based on the operator, equipment settings, and specific sample handling conditions. High-force isolation techniques, such as standard ultracentrifugation, exert intense shear stress on the sample. This stress can deform, rupture, or induce artificial aggregation of the delicate lipid membranes of EPs. Many laboratory methods require expensive instrumentation and long processing times, and such labor-intensive workflows are entirely unsuited for high-throughput clinical diagnostics or large-scale industrial manufacturing. Therefore, the quest for a "golden standard" method is taking place.

Ultrafiltration (UF) is a membrane-based separation technique widely used to concentrate EPs. It relies on semi-permeable membranes with specific molecular weight cut-offs (MWCO) or pore sizes to retain EPs while allowing smaller molecules and liquid to pass through. Traditional spin filters force the liquid downward through the membrane, smashing the EPs directly into the filter pores. The EV-Spinner operates using a reverse ultrafiltration design, where the liquid is driven through the membrane in the opposite direction of the centrifugal force to reduce membrane clogging (fouling) and severe particle trapping. For microalgae samples, which are naturally prone to clustering and high debris, such design prevents the filter from locking up mid-spin. The polyethersulfone (PES) membrane is chemically treated to be low protein-binding.

In this work we report of using reverse ultrafiltration with EV-Spinner on samples from suspension of aged, washed erythrocytes and the culture of microalgae *Chlorellae sorokiniana*.

2. Material and Methods

2.1. Culturing of microalgae

The microalgae *Chlorella sorokiniana* (strain CCAP 211/8K) were a kind gift from Jelena Danilović Luković from University of Belgrade. They were sourced from the Culture Collection of Algae and Protozoa, UK. They were grown in deionized distilled water with addition of Bold Basal Medium (PhytoTech Labs, Lenexa, Kansas City, USA). The composition of the BBM was: Boric Acid (11.42 mg/L), Manganese Chloride×4H₂O (1.44 mg/L), Calcium Chloride Anhydrous (18.87 mg/L), Potassium Hydroxide (31.00 mg/L), Cobalt Nitrate×6H₂O (0.49 mg/L), Potassium Phosphate Dibasic (75.00 mg/L), Cupric Sulfate×5H₂O (1.57 mg/L), Potassium Phosphate Monobasic (175.00 mg/L), EDTA Disodium Salt (63.61 mg/L), Sodium Chloride (25.00 mg/L), Ferrous Sulfate×7H₂O (4.98 mg/L), Sodium Molybdate (1.19 mg/L), Magnesium Sulfate Anhydrous (36.63 mg/L), Sodium Nitrate (250.00 mg/L), Zinc Sulfate×7H₂O (8.82 mg/L). Microalgae grew in a 50 mL falcon tube in a shelf of the laboratory having sunlight and conditioned air at 18 °C. We observed no bacteria under optical microscope in the culture.

2.2. Blood sampling

Blood was donated by an author (a female with no record of disease). Collection was established in the morning after fasting for a minimum of 12 h overnight. A G21 needle (Microlance, Becton Dickinson, Franklin Lakes, NJ, USA) and 2.7 mL evacuated tube with

trisodium citrate (BD Vacutainers, 367714A, Becton Dickinson, Franklin Lakes, NJ, USA) were used.

2.3. Preparation of erythrocyte suspension

To sediment the erythrocytes, blood was centrifuged for 10 min at $300 \times g$ and 18°C (centrifuge Centric 400/R, Domel, Železniki, Slovenia with swinging rotor). The supernatant (plasma) was haemolytic (**Figure 1**). We removed the plasma from above the buffy coat. The erythrocytes were resuspended in PBS by turning the eprouvette upside-down and centrifuged for 10 min at $300 \times g$ and 18°C (centrifuge Centric 400/R, Domel, Železniki, Slovenia, with swinging rotor). The supernatant was removed and replaced with PBS. Erythrocytes were resuspended by turning the eprouvette upside-down and centrifuged for 30 min at $500 \times g$ and 18°C (centrifuge Centric 400/R, Domel, Železniki, Slovenia). The procedure was repeated to yield transparent supernatant above the erythrocytes. The washed erythrocytes suspended in PBS were stored at 4°C for 15 days.



Figure 1. Blood sample after centrifugation for 10 min at $300 \times g$ and 18°C showing haemolytic plasma.

2.4. Ultrafiltration of samples

Extracellular particles (EPs) were harvested from 1 mL samples of either suspension of aged washed human erythrocytes or *Chlorella sorokiniana* culture using reverse-flow ultrafiltration. Briefly, individual 1 mL aliquots were loaded directly into EV-Spinner ultrafiltration concentrators (HansaBioMed, Tallinn, Estonia; 100 kDa molecular weight cut-off [MWCO]). The loaded filter units were placed in a centrifuge Centric 400R (Domel d.o.o.) with swinging rotor. The samples were centrifuged at 18°C and $2500 \times g$ for 15 minutes to drive the liquid and low-molecular-weight elements through the low-protein-binding polyethersulfone (PES) membrane. This step effectively concentrated the retained EPs into a liquid boundary concentrate layer within the retentate chamber, yielding the final pellets (designated PE for the microalgae culture and PA for the erythrocyte suspension). The volume of the retrieved PE was cca 100 μL and of the PA it was cca 50 μL . The corresponding ultrafiltration supernatants (SE for microalgae and SA for erythrocytes) were collected from the filtrate compartment. Both the harvested pellets and collected supernatants were assessed immediately after harvesting, with Interferometric Light Microscopy (ILM), and UV-vis spectroscopy.

2.5. Electron microscopy

Scanning Electron Microscopy (SEM). A drop of 5 μL of sample was loaded onto aluminum stands covered with thin layer of polymers based on cyanoacrylates (Jampani et al., 2024) and let to dry in air. The sample was examined with a Thermo Fisher Scientific Apreo 2 microscope with EDS/EBSD detector, low vacuum mode.

2.6. Assessment of density and viscosity

We measured the density of the supernatants. There was not enough volume of the pellets (required 1mL) available for the measurement with the above described instrument, so we used the supernatant density as an estimate also for pellets. The density was then used as an input to assess viscosity, and viscosity was used as an input to assess hydrodynamic diameter of nanoparticles by ILM. Density measurement was performed using an oscillating U-tube Density Meter (DMA 1002, Anton Paar, Graz, Austria). 1 mL sample was placed in the measuring cell, which was subjected to acoustic oscillations. Dynamic viscosity was measured using a falling ball micro viscometer (Lovis 2000 M, Anton Paar, Graz, Austria with Lovis 1.59 Short Capillary). Temperature and measurement angle were set at 20°C and 45° respectively.

2.7. Interferometric Light Microscopy (ILM)

EP samples were measured with ILM using Videodrop (Myriade, Paris, France) (Boccara et al., 2016; Romolo et al., 2022; Korenjak et al., 2024). Samples were diluted to enable optimal performance of the instrument (in the range of $(0.5 \text{ to } 5) \times 10^9/\text{mL}$). 7 μL of sample was placed between cover glasses and illuminated by a 2 W blue LED light. Threshold value was 4.2. The average concentration (ILMc) and D_h of EPs were determined for each sample. The position of a particle was captured in the recorded video (movie). We set a limit of 20 movies or 300 detected EPs to assess the EPs in the sample. We used the software QVIR 2.6.0 (Myriade, Paris, France) to process the images (movies). D_h was determined by the Einstein-Stokes equation using the measured data of viscosity and density obtained as described above.

2.8. UV - vis spectroscopy

Three microliters droplets of samples were placed on the stand of a Nanodrop UV spectrometer (ThermoFischer Scientific, Waltham, MA, USA) and assessed for ultraviolet-visible spectrum (UV-Vis) absorbance. The wavelengths 260 nm and 280 nm were considered in all samples, while the wavelength 430 nm was considered in erythrocyte samples, and wavelengths 460 nm and 690 nm were considered in microalgae samples. Samples were mixed before measurement.

2.9. Statistical Analysis

All data were analysed using descriptive statistics. All measurements were performed in triplicate and are presented as an average and standard deviation.

3. Results

Pellets of ultrafiltration (PE and PA) indicated the presence of EPs in samples while supernatants of ultrafiltration (SE and SA) were practically devoid of EPs (**Table 1**); in supernatants, the number densities of EPs were under the operational detection limits of the ILM (**Table 1**).

Table 1. Number density of EPs n , hydrodynamic diameter D_h and absorbances (A) at respective UV and visual light wavelengths for pellets and supernatants of the ultrafiltration. PE: ultrafiltration pellet from in vitro aged washed erythrocyte suspension, SE: ultrafiltration supernatant from in vitro aged washed erythrocyte suspension, PA: ultrafiltration pellet from microalgae culture, SA: ultrafiltration supernatant from microalgae culture. The results are given by the averages of three measurements $\pm$ standard deviations.

Variable	Dilution	$n \times 10^9/\text{mL}$	D_h (nm)	A260	A280	A430	A460	A690
PE	40	1.16 ± 0.09	168 ± 6	22.3 ± 0.23	24.8 ± 0.0	73.6 ± 0.70	/	/
SE	4	0.01 ± 0.00	174 ± 72	3.68 ± 0.04	2.64 ± 0.04	6.63 ± 0.06	/	/
PA	1	0.44 ± 0.02	252 ± 7	0.31 ± 0.11	0.29 ± 0.13	/	0.04 ± 0.04	0.04 ± 0.04
SA	1	0	0	0.03 ± 0.06	0.04 ± 0.01	/	0	0

Imaging the pellet PE, **Figure 2A** shows extensive dendritic crystalline structures, which are classic signatures of dried buffer salts (like PBS) or crystallized proteins (such as hemoglobin). Panel **B** highlights distinct, spherical nano-sized particles (erythrocyte EPs) embedded within dense matrix. Imaging the supernatant SE, **Figure 2C** reveals scattered, cuboidal/geometric crystal lattices, typical of clean salt precipitation from a dried buffer solution. Panel **D** appears largely smooth and featureless, displaying an empty background devoid of the characteristic globular clusters seen in the pellet. Panels **E**, **F** (Fixed/Stained SEM Control) and **G** (cryogenic transmission electron microscope image) (Božič et al., 2021; Jozelj et al., 2022) are shown for comparison. A notable amount of EPs survived the treatment by 80 °C. The protein and nucleic acid content in the erythrocyte samples was considerable, in particular the clearly expressed peak pertaining to haemoglobin. The values of the absorbance at 430 nm in the ultrafiltration pellet were the highest detected among the samples. The pellet from microalgae PA presented a detectable amount of EPs but did not define a peak at the chlorophyll absorbance wavelengths 460 nm and 690 nm.

C (Panel E). The cryo-transmission electron micrograph (**Figure 2G**) reveals intact, well-defined, oval-to-elongated vesicular membranes filled with dense internal contents, confirming the presence of true biogenic lipid-bilayer extracellular vesicles.

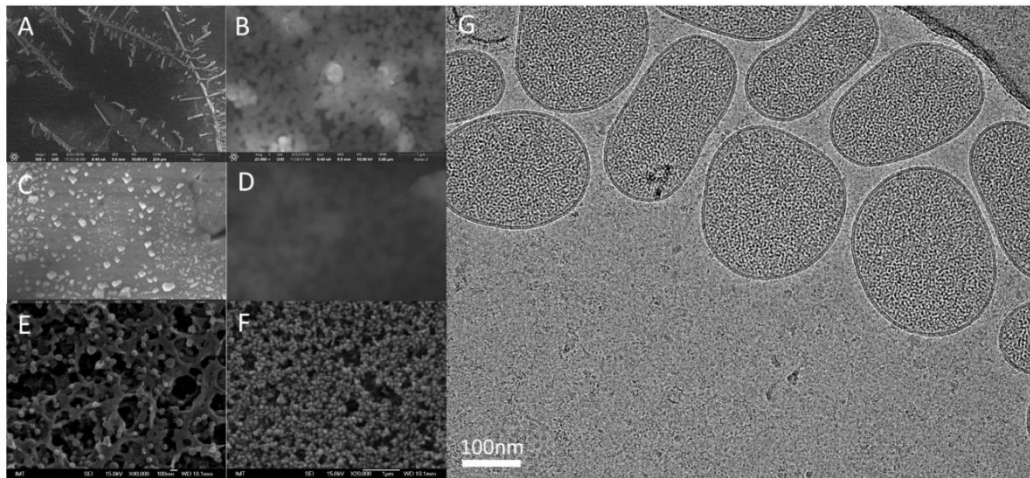


Figure 2. Scanning electron microscopy (SEM) and cryogenic transmission electron microscopy (cryo-TEM) images of erythrocyte-derived fractions. (A, B) SEM micrographs of the raw ultrafiltration pellet containing erythrocyte extracellular particles (EPs) embedded in crystalline matrix. (C, D) SEM micrographs of the corresponding raw ultrafiltration supernatant, displaying clean buffer salt precipitation and an absence of vesicular clusters. (E) SEM micrograph of fixed, stained, and heavy-metal-sputtered erythrocyte EPs following thermal treatment at 80 °C. (F) SEM micrograph of control fixed, stained, and heavy-metal-sputtered erythrocyte EPs. (G) Cryo-TEM micrograph demonstrating intact, well-defined biogenic erythrocyte extracellular vesicles (EVs). Panels E–G are reproduced from Božič et al., 2021 (available under a Creative Commons Attribution 4.0 International License [CC BY 4.0]).

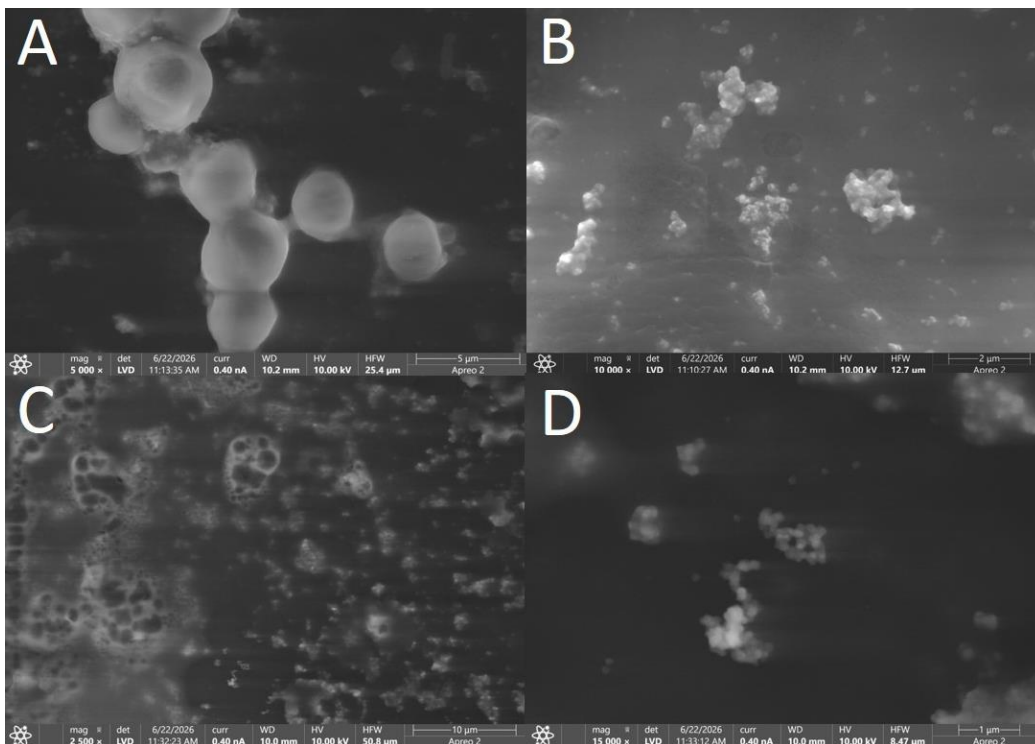


Figure 3. Scanning electron microscopy (SEM) micrographs of raw ultrafiltration pellet fractions (PA) from a *Chlorellae sorokiniana* microalgae culture. (A) Low-magnification overview showing distinct, spherical microalgae cells tightly associated with clusters of sub-micron globular extracellular particles (EPs). (B–D) Intermediate and high-magnification views of alternate fields revealing a repeating, porous extracellular polymeric substance network and self-assembled organic film embedded with clustered, nano-sized structures.

Imaging the pellet PA, **Figure 3A** shows distinct, spherical microalgae cells (~3–5 μm in diameter) tightly associated with clusters of much smaller, globular extracellular particles (EPs). **Figures 3B–D** reveal a repeating, porous network structure resembling a dried liquid film or a polymer surface with embedded clustered nano-sized globular particles. Microalgae cultures often contain complex extracellular polymeric substances, sticky polysaccharides, and rigid cell walls that can build up globular nano and microstructures.

The protein and nucleic acid content in the erythrocyte samples was considerable, in particular the clearly expressed peak pertaining to haemoglobin. The values of the absorbance at 430 nm in the ultrafiltration pellet were the highest detected among the samples. The pellet from microalgae PA presented a detectable amount of EPs but did not define a peak at the chlorophyll absorbance wavelengths 460 nm and 690 nm.

4. Discussion

4.1. Thermodynamic dispersity and dilution-induced colloidal aggregation

Morphological analysis via scanning electron microscopy (SEM) revealed distinct lumped nanoparticles within the *Chlorella sorokiniana* microalgae fractions. In highly dilute solutions, dispersed individual particles operate in a state of thermodynamic instability due to their high free energy. To minimize the free energy, adjacent particles drift into a coordination sphere, co-localizing into cohesive clusters or "particle swarms" (**Figures 3B–D**). This phenomenon is accelerated during sample preparation. As a droplet desorbs and dries upon the SEM stub covered with nanoscale polymer films (Jampani et al., 2024), the receding capillary meniscus acts as a physical net. The fluid front drives individual scattered particles and pre-existing particulate swarms into closer proximity, ultimately locking them into highly visible, clumped networks. Consequently, the micro-structures observed within the microalgae supernatants (SA) are best characterized as dilution-induced colloidal aggregates or flocculated particle swarms.

4.2. Biopolymer condensation paradox: microalgae mucilage vs. erythrocyte hemoproteins

The self-assembly behavior in microalgae samples is driven by the unique biochemistry of phycological secretomes. Unicellular green algae are notorious for secreting vast quantities of extracellular polymeric substances, commonly referred to as mucilage. This complex extracellular matrix consists primarily of high-molecular-weight heteropolysaccharides, glycoproteins, and proteoglycans. Within the raw culture media, the mucilage exists in a highly dynamic equilibrium between fully dissolved single polymer chains and larger assembled gel networks. During pressure-driven reverse ultrafiltration, unassembled polysaccharide chains or low-molecular-weight glycoprotein subunits align linearly under convective flow, easily slipping through the 3–10 nm pore channels of a 100 kDa membrane. Upon entering the filtrate chamber, the localized shear forces from fluid processing combined with subsequent concentration-driven cross-linking trigger these amphiphilic biopolymers to spontaneously condense into globular nano-aggregates. Importantly, pseudo-EP structures lack an authentic phospholipid bilayer. They represent highly dynamic, transient gel matrices capable of shifting morphology under localized environmental conditions. Such behavior highlights a critical, source-specific artifact unique to microalgal harvesting workflows that must be carefully distinguished from biogenic, bilayer-enclosed extracellular vesicles (EVs). In contrast, washed erythrocyte suspensions do not secrete sticky, highly cross-linking plant polysaccharides. The background of mammalian red blood cell streams is instead dominated by structurally stable proteins, specifically hemoglobin. If individual 64 kDa hemoglobin tetramers pass through the ultrafiltration barrier, they remain stable monodisperse units in solution or form standard geometric crystalline precipitates upon drying (**Figures 2A and 2C**) rather than self-assembling into biomimetic nano-spheres.

4.3. Historical nomenclature and the biophysical context of ultrafiltration

The isolation and characterization of submicron extracellular structures—historically labeled as microparticles, exosomes, paramural bodies, or exocellular fragments, and modernly consolidated under the umbrella of extracellular vesicles (EVs) and extracellular particles (EPs)—has long presented a biophysical challenge. Foundational researchers in the 1970s and 1980s recognized that there are different methods to resolve membranous nanoparticles from complex macro-solute matrices. Early pioneers successfully integrated ultrafiltration to concentrate and purify submicron vesicles (Blatt et al., 1970; Porter, 1972;

Cotton et al., 1975; Tutunijan, 1982; Cheryan, 1986). Also, mammalian body fluids were considered as sources of EPs (Lutz et al., 1977; Panakov et al., 1977; Stagmayer et al., 1982; Greenwalt et al., 1984; Wiggins et al., 1986; Johnstone et al. 1987). Johnstone et al. coined the term "exosome" in describing how maturing sheep reticulocytes clear unwanted proteins (like the transferrin receptor) via the fusion of multivesicular bodies with the plasma membrane (Johnstone et al., 1987).

When extracting extracellular fractions from unicellular green algae or plants, early investigators were forced to confront thick, carbohydrate-rich matrices and dense wall components. To isolate EPs without inducing mechanical cell lysis, researchers implemented sequential filtration and pressure-driven membrane concentration systems to successfully harvest exocellular particles (Halperin et al., 1967; McLean et al., 1972; Marchant et al., 1986; Atkinson et al., 1972; Grov et al., 1981, Vreeland et al., 1984). It was identified that the harvesting of the particles was severely hampered by the co-purification by the mucilage (Atkinson et al., 1972).

Our current pipeline directly addresses these historical limits by utilizing reverse ultrafiltration to harvest EPs from both washed erythrocytes and *Chlorellae sorokiniana* cultures. In membrane separation physics, dead-end filtration configurations suffer from rapid concentration polarization and gel-layer cake formation (Blatt et al., 1970; Porter, 1972), wherein accumulated macro-solutes blind the membrane pores and trap smaller vesicles. By implementing a reverse ultrafiltration strategy, we successfully mitigated this cake formation. This processing efficiency is directly reflected in our characterization data. The high concentration of EPs captured in the erythrocyte pellet (PE: $(1.16 \pm 0.09) \times 10^9$ /mL) at $40 \times$ dilution) versus the microalgae pellet (PA: $(0.44 \pm 0.02) \times 10^9$ /mL), non-diluted) demonstrates that reverse ultrafiltration functions effectively across radically different biological matrices.

Crucially, our supernatants (SE and SA) presented no notable amounts of EPs, confirming that the molecular cutoff of the filter membrane efficiently retained the target particles without letting them slip through or shear into smaller fragments—a common issue when using early, non-optimized membrane configurations.

4.3. Spectroscopic fingerprints and the dilution-identity law

Our UV-vis spectroscopy data reveals a striking contrast between the two sources that can be explained through historical biochemical frameworks. The 40-fold diluted erythrocyte pellet (PE) showed high absorption at 260/280 nm ($(22 \pm 0.2 \text{ a.u.}) / (25 \pm 0.0 \text{ a.u.})$)(**Table 1**), pointing to a dense corona of proteins and hemoglobin remnants typical of aged, ATP-depleted erythrocytes. Conversely, the *Chlorellae sorokiniana* pellet (PA) was remarkably poor in proteins and nucleic acids ($(0.31 \pm 0.11) \text{ a.u.} / (0.29 \pm 0.13 \text{ a.u.})$)(**Table 1**). Low protein/nucleic acid profile in algae agrees with previous results that *Chlorella sorokiniana* cell-wall-associated fractions are primarily composed of structural lipids and non-proteinaceous polymers, rather than soluble cytoplasmic proteins (Grov and Taylor, 1981). The singular lumps of self-assembly seen in our algae samples likely represent the structural arrangement of these exocellular matrices as they interact with trace ambient polysaccharides, a behavior predicted by Vreeland and Laetsch (1984). Structurally identical, micro-sized globular lumps are present in both the ultrafiltration pellet (PA; **Figures 3A and 3B**) and the ultrafiltration supernatant (SA; **Figures 3C and 3D**). The primary differentiation between the two fractions is quantitative, with a lower density of these micro-structures observed in the supernatant. Rather than indicating a failure of the ultrafiltration membrane retention, this phenomenon points to a continuous, post-filtration spontaneous thermodynamic self-assembly of mucilage and sticky polysaccharides. Free macromolecules passing through the membrane pores into the supernatant fluid continuously aggregate to reach an operational energy minimum, organizing into macro-molecular clusters in situ after the filtration process is complete.

Because of this highly dynamic behavior, we have explicitly reported the number densities of the EPs at their exact measured dilutions (**Table 1**). Due to the fragile colloidal nature of EPs, subjecting these samples to additional or un-optimized dilution steps alters their thermodynamic equilibrium. Such shifts are likely to drive individual particles into entirely different structural states, shifting the identity, composition, size, and shape of the observed clusters. Therefore, to ensure reproducible characterization, processing pipelines must consume minute sample volumes and avoid aggressive post-isolation modifications.

The high concentration of EPs captured in the erythrocyte pellet ($1.16 \pm 0.09 \times 10^9$ /mL) versus the microalgae pellet ($0.44 \pm 0.02 \times 10^9$ /mL) (**Table 1**) demonstrates that reverse ultrafiltration functions effectively across radically different biological matrices. Crucially, our supernatants (SE and SA) presented no notable amounts of EPs, confirming that the molecular cutoff of our membranes efficiently retained the target particles without letting them slip through or shear into smaller fragments—a common issue when using early, non-optimized membrane configurations (Cheryan, 1986).

The post-filtration assembly clarifies the discrepancy observed with our ILM measurements, which recorded no particles for the SA fraction (**Table 1**). Because these self-assembled organic lumps rapidly exceed the sub-micron scale, they transcend the operational detection ceiling of standard ILM tracking (approximately 800 nm). Crucially, however, the optical background of the SA was notably clearer than that of the PA (**Figure 3**).

5. Conclusions

By implementing a reverse ultrafiltration processing pipeline paired with non-destructive, minute-volume orthogonal characterization methods (SEM, ILM, and UV-vis spectroscopy), we established a rapid experimental workflow capable of assessing EPs in samples within a matter of hours. The qualitative and quantitative results from the included analytical methods demonstrated excellent agreement across both biological matrices. The mammalian erythrocyte pellet (PE) was highly enriched with both EPs and co-isolated proteins and nucleic acids, exhibiting distinct globular morphologies of EPs consistent with membrane-enclosed vesicles. Conversely, the microalgae pellet (PA) was comparatively poor in native EPs, proteins, and nucleic acids. In both post-filtration supernatants (SE and SA), sub-micron EPs were thoroughly depleted, falling beneath the operational detection limits of the ILM tracking system. However, while the erythrocyte supernatant (SE) retained a high background of stable soluble hemoproteins and nucleic acids, the microalgae supernatant (SA) was largely devoid of macromolecular signatures. Morphologically, the micro-lumps observed in the algal supernatant (SA) lacked the distinct smooth geometry characteristic of membrane-enclosed biogenic vesicles. Instead, they represent dynamic, dilution-induced colloidal matrices that undergo spontaneous thermodynamic self-assembly from cell wall polysaccharides and mucilage subunits after passing through the filtration membrane. Ultimately, this work demonstrates that reverse ultrafiltration provides a highly efficient, gentle, but low-volume mechanism to concentrate and harvest native EPs from radically different cellular sources.

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Conflicts of Interest: The authors declare no conflict of interest.

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Research

Measuring Density and Viscosity of Samples as an Improvement of Determination of Hydrodynamic Diameter of Extracellular Particles by Interferometric Light Microscopy

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Abstract:

We present technological advancement of the method for assessment of the hydrodynamic diameter D_h of nanoparticles suspended in liquid, by interferometric light microscopy (ILM). In ILM, D_h is determined by Einstein-Stokes equation which includes dynamic viscosity η of the sample as an input. Hitherto, η was estimated by the respective value for water at given temperature. It was of our interest to see how the results obtained by direct measurement of η differ from this estimate. We measured the density of the sample by oscillating U-tube Density Meter and used this value as an input to measure η by a rolling and falling ball Micro viscometer. We determined D_h by interferometric light microscope. An overestimation of D_h by using the “water” estimation was found to range from 1% to 7%. Furthermore, a significant difference was observed between D_h obtained by the respective data ($t = 25.985$; $p < 0.001$). The percentage difference between the two methods of determining D_h showed a moderate and statistically significant negative correlation with plasma density (Spearman coefficient $\rho = -0.365$, $p < 0.001$) and a very strong and significant negative correlation with dynamic viscosity ($\rho = -0.968$, $p < 0.001$). The inherently negative Spearman coefficients demonstrate that the overestimation error introduced by the measurement using water dynamic viscosity increases significantly with increase of the density and dynamic viscosity of the sample. The correction method based on direct measurement of η offers a more accurate, scientifically sound measurement of D_h of extracellular particles derived from different biological matrices (e.g., plants, micro-organism cultures, bodily fluids, etc.).

Keywords: Extracellular vesicles; Interferometric Light Microscopy; Nanoparticles; Density; Viscosity; Biophysical Properties

1. Introduction

Nanomedicine and innovative biotechnologies have recently introduced as focus of research the development of nanoparticle-based systems for diagnostic and therapeutic purposes. Among these, particular attention has been paid to extracellular particles (EPs), including extracellular vesicles (EVs) (Welsh et al., 2024).

Research is focusing on the isolation and use of these particles as biomarkers and carriers derived from biological fluids (blood, plasma), but also on the study, within the field of plant biology, of so-called plant-derived extracellular particles (PEVs) (Wiklander et al., 2019; Welsh et al., 2024). These structures, in particular, are of great interest due to their biocompatibility, stability and lack of immunogenicity (Cui et al., 2024).

Both for plant and animal-derived particles, a careful assessment of their physical characteristic is the key to studying them properly. This is because there is a strong correlation between physical properties of nanoparticles and their bio-functional properties (bioavailability, pharmacokinetics, etc.) (Welsh et al., 2024). The hydrodynamic diameter (D_h) of nanoparticles, in particular, is a variable on which the ability to cross biological barriers, load-carrying capacity, bioavailability and pharmacokinetics strictly depend. Among the techniques developed to study the physical characteristics of these structures, research has focused on the development of innovative technologies such as Interferometric Light Microscopy (ILM). This is an optical technology within the field of single-particle tracking (SPT), which aims to overcome the limitations of other mass analysis techniques, such as Dynamic Light Scattering (DLS) (Filipe et al., 2010; Boccara et al., 2016; Berry et al., 2024; Korenjak et al., 2024).

D_h is calculated in ILM system following the Stokes-Einstein equation. The equation requires the viscosity of medium and the temperature value. To accurately translate Brownian motion into a metric dimension, it is essential to have viscosity of the medium examined. In experimental practice, it is generally assumed that the viscosity of the matrix—since most cases involve dilute solutions—is comparable to that of water. However, when working with complex biological matrices, such as plasma or plant extracts—even when diluted—it must be considered that these are complex colloidal suspensions rich in proteins and lipid fractions that increase their viscosity (Windberger et al., 2003). Failure to apply a specific viscosity correction result in an incorrect evaluation, and the instrument, interpreting the kinetic slowdown as an effect due to mass rather than medium friction, generates a systematic overestimation of the particles D_h . For this reason, the aim of this study is to verify, using correct measurements of canine plasma density and dynamic viscosity (dynVis), whether the dimensional determination of nanoparticles using ILM is more accurate.

2. Material and Methods

2.1. Samples Management

A total of 79 canine plasma samples were used in the study. Samples were randomly collected from dogs of various ages, sexes, breeds, and health statuses at the Veterinary Teaching Hospital of the Department of Veterinary Medicine and Animal Production, University of Naples “Federico II.” 3 ml of blood sample were drawn from the jugular vein and collected in EDTA-containing tubes. The samples were subsequently centrifuged at 1500 g for 15 minutes at 4°C to obtain the plasma, which was transferred to Eppendorf tubes, stored at -80°C, and sent to the Laboratory of Biophysics at the Faculty of Electrical Engineering, University of Ljubljana, Slovenia, for further analysis. All the analyses were performed for diluted 10x plasma sample, and all the following results are considered for diluted 10x sample.

2.2. Assessment of density and viscosity

Density evaluation was performed using an oscillating U-tube Density Meter (DMA 1002, Anton Par, Graz, Austria). The sample was placed in the measuring cell, and the tube was mechanically oscillated. The oscillation frequency changes according to the internal mass, so this variation allows the density of the sample to be calculated with very high precision. The minimum sample quantity required to perform the analysis is 1 ml. Measurements were performed setting the temperature at 20°C. For the same samples, dynamic viscosity

was measured, using a rolling and falling ball Micro viscometer (Lovis 2000 M, Anton Par, Graz, Austria). A small metal ball rolls and falls inside an inclined glass or quartz measuring capillary tube with a known internal diameter filled with the sample. Gravity causes the ball to move, but the viscosity of the liquid slows it down, and the instrument measures the rolling and falling time of the ball. Capillary Lovis 1.59 Short was used, temperature and measurement angle have been set at 20°C and 45° respectively, and density was entered for each sample.

2.3. Interferometric Light Microscopy (ILM)

For evaluating EPs, samples were processed with ILM using Videodrop (Myriade, Paris, France). All samples were 10 times diluted with saline solution. Measurements were performed at room temperature, recorded at the time of measurement and entered manually for each sample (20-22°C) to achieve the optimal EPs concentration and dimension for detection between 5×10^8 and 5×10^9 particles/ml. A total of 7 μ l of sample was placed between cover glasses and illuminated by a 2 W blue LED light, using a threshold value of 4.2. The average concentration (ILMc) and D_h of EPs were determined for each sample. Particle number density was estimated as the ratio between the number of particles detected and the scanned volume. The D_h was determined by tracking the position of the particle captured in the recorded video. A limit of 20 movies or 300 detected particles was set for each sample. Image and movie processing was performed using the associated software QVIR 2.6.0 (Myriade, Paris, France). The D_h was determined by setting both the dynVis of water, generated automatically by the system (Water viscosity: 0.954 mPas), and the actual dynVis of the sample, which had been measured previously. For statistical purposes, the measurement based on the dynVis of water is defined as "estimated D_h " (eD_h), while the measurement based on the actual dynamic viscosity of the sample is defined as "corrected D_h " (cD_h), and the absolute difference (in nm) and the percentage difference between the two measurements were calculated.

2.4. Statistical Analysis

All data were analysed using descriptive statistics. All measurements were performed in triplicate and are presented as median and interquartile range. Shapiro-Wilk test was used to perform the normality of data. Paired samples T-test was used to highlight differences between eD_h and cD_h , considered statistically significant for a p-value < 0.05. The correlation between variables was performed using Spearman's rank correlation coefficient (ρ). A significant correlation was observed for $p < 0.05$ and classified according to the ρ value as follows: i) $\rho < \pm 0.1$ no correlation; ii) ρ from ± 0.1 to ± 0.3 weak; iii) ρ from ± 0.3 to ± 0.5 moderate; iv) ρ from ± 0.5 to ± 0.7 strong; v) $\rho > \pm 0.7$ very strong. Graphical analysis with Bland-Altman plots were used to show the agreement between variables (Bland & Altman, 1986).

3. Results

3.1 Data distribution and comparisons between methods

The data followed normal distribution, except for the values relating to plasma density and the percentage difference between the methods ($p = 0.001$). A summary of values for eD_h , cD_h , absolute difference in nm, percentage difference, density and dynamic viscosity is shown in **Table 1**. When examining the values obtained from measurements using the method with measured viscosity, an overestimation of particle size by using the data for water was found to range from 1% to 7%. Furthermore, a significant difference was observed between measurements based on the dynamic viscosity of water and those based on specific dynamic viscosity of the samples ($t = 25.985$; $p < 0.001$).

Table 1. Average, Standard Deviation, Minimum, and Maximum Value for estimated and corrected hydrodynamic diameter, absolute and percentage differences between the two measures, and density and dynamic viscosity of samples.

Variable	Average	Standard Deviation	Minimum value	Maximum Value
Estimated D_h	145.06 nm	± 19.40	107.33 nm	194.00 nm
Corrected D_h	139.15 nm	± 18.55	103.74 nm	189.05 nm
Absolute Difference ($cD_h - eD_h$)	-5.90 nm	± 2.02	-11.40 nm	-1.04 nm
Percentage Difference ($cD_h - eD_h$)	-4.13%	± 1.30	-7.00%	-1.00%
Density	1.00693g/cm ³	± 0.00053	1.00499g/cm ³	1.00859g/cm ³
Dynamic Viscosity	0.994 mPas	± 0.012	0.960 mPas	1.026 mPas

D_h : hydrodynamic diameter; nm: nanometres; mPas: millipascal second

3.2 Correlation between measurement differences and the biophysical properties of the sample

To verify if the differences in the observed measurements were due to biological variability in plasma, Spearman's correlation (ρ) was applied. The percentage difference between the two methods of determining particle size showed a moderate and statistically significant negative correlation with plasma density ($\rho = -0.365$, $p < 0.001$) and a very strong and significant negative correlation with dynVis ($\rho = -0.968$, $p < 0.001$). Since percentage difference values are inherently negative, the negative Spearman coefficients demonstrate that the overestimation error introduced by the measurement using water dynamic viscosity increases significantly with increase of the specific density and dynamic viscosity of the sample. Furthermore, the Bland-Altman plot revealed a constant systematic error in the overestimation of the hydrodynamic diameter using the water-dynVis-based method, as can be seen in **Figure 1**. The 95% confidence intervals were wide, ranging from -1.95 nm to -9.85 nm, indicating considerable inter-individual variability in the degree of error.

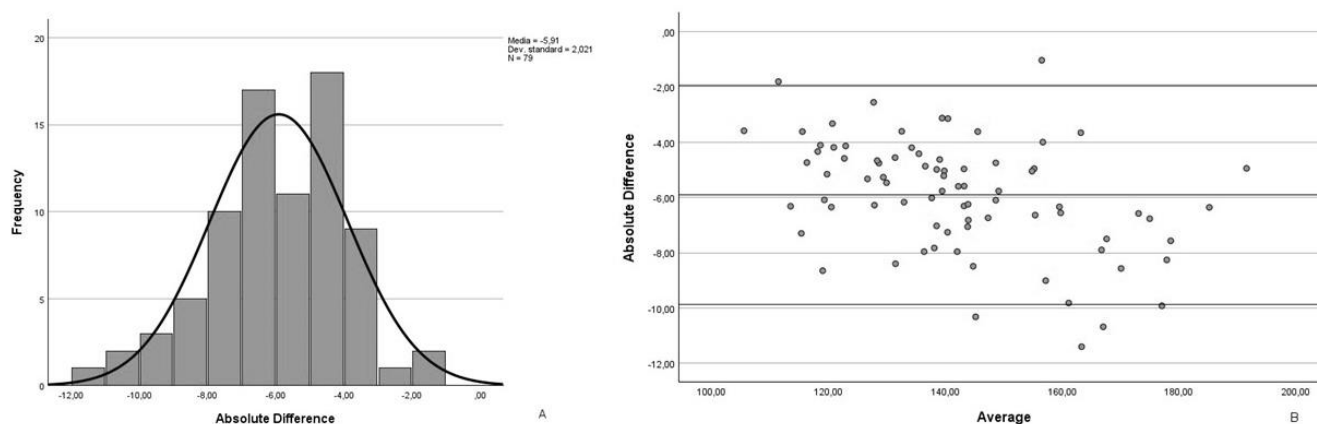


Figure 1. Analysis of the discrepancy between the hydrodynamic diameter measurement using water dynamic viscosity and the corrected measurement using the sample specific dynamic viscosity. (A) The histogram shown in the figure displays the distribution of absolute differences (expressed in nanometres) with a superimposed normal curve; the distribution appears symmetric and centred around a mean bias of -5.91 nm (SD = 2.021). (B) The Bland-Altman plot highlights the agreement between the two methods, showing the presence of a systematic error of overestimation in the measurement based on water viscosity and an error trend proportional to the increase in average particle size.

4. Discussion

The aim of this study was to assess the impact of the biophysical properties of canine plasma (density and viscosity) on the determination of the hydrodynamic diameter of nanoparticles using ILM, by comparing the approach based on the theoretical viscosity of water with a correction protocol which takes into account the actual viscosity of the individual sample. The results clearly show that the uncritical assumption of water viscosity introduces a significant systematic error, leading to a constant overestimation of the actual particle size. This phenomenon is not an artefact, but is governed by precise

physical laws and directly reflects the intrinsic biological variability of the plasma matrix of the samples examined (Boccara et al., 2016). The explanation for this systematic bias, quantified by the Bland-Altman plot as an average error of -5.91 nm, can be found in the theoretical foundations of Brownian motion as tracked individually by the ILM. The instrument calculates the root-mean-square displacement of individual particles based on the variation in their interference pattern over time, and then calculates their hydrodynamic diameter using the Stokes–Einstein equation (Young & Kukura, 2019). In the equation, the hydrodynamic diameter is inversely proportional to the viscosity of the medium. As plasma has a density and viscosity that are inherently higher than those of water—due to the complex suspension of macromolecular proteins such as albumin, globulin and fibrinogen, as well as lipid fractions—the free motion of the particles tracked by ILM is slowed down (Kwaan, 2010). Consequently, if the instrument’s algorithm processes the video tracking on the assumption of water viscosity, it interprets this reduced particle movement as the result of a larger size, leading to an overestimation (Filipe et al., 2010). The strong negative correlation observed between the percentage difference and plasma viscosity provides direct evidence of this mechanism. The negative correlation coefficient demonstrates that, as plasma viscosity and density increase, the magnitude of the error introduced by the method that takes water viscosity into account increases linearly. This finding has crucial biological significance: plasma composition is not constant, but varies drastically depending on age, nutritional status, and the presence of inflammatory, infectious or metabolic conditions (Geyer et al., 2016). Consequently, the wide range of agreement limits shown in the Bland-Altman plot demonstrates that it is impossible to apply a fixed or universal mathematical correction factor to all samples, making it essential to measure viscosity individually in order to obtain the actual particle sizes. Furthermore, an interesting finding is the evidence of a proportional error within the Bland-Altman plot. The distribution of the data points shows a negative slope, indicating that the overestimation caused by the method using water viscosity is not only present but becomes progressively more pronounced as the difference in the average size of the analysed nanoparticles increases.

Although a linear variation in diameter of between 1% and 7% may appear modest at first analysis, its true biological impact becomes clear when the three-dimensional geometry of the particles is considered. As these particles can be approximated as spherical structures, their volume (V) is related to the radius (r) by a cubic relationship ($V = 4/3\pi r^3$). An estimate of the total particle volume that differs by 7% from the actual value is a significant error, because when the cubic effect is taken into account, the percentage error rises to 22.5% (Gardiner et al., 2013).

The inclusion of the room temperature, recorded manually for each sample, is a key factor in the stability of the method. Temperature influences the Stokes-Einstein equation both directly and indirectly through the modulation of viscosity itself, which decreases as temperature rises. Actively tracking the thermal microfluctuations in the laboratory allows the pure effect of the biological matrix on the interferometric measurement to be isolated, eliminating environmental biases and conferring high reproducibility on the proposed method.

It is important to emphasise that the value of performing accurate measurements is not limited exclusively to animal-derived biological matrices, but also applies to the study of plant-derived materials. In recent years, the isolation of natural nanoparticles derived from plant matrices – in particular PEVs or those derived from fruit – has become central to nanomedicine, nutraceuticals and drug delivery, thanks to their biocompatibility and lack of toxicity (Cui et al., 2024). The plant extraction fluids, cell juices or apoplastic fluids from which these nanoparticles are isolated are characterised by a high degree of biochemical complexity. The massive presence of soluble sugars, organic acids, polyphenols and structural polymers such as pectins drastically alters the rheological properties of the medium, resulting in density and viscosity values significantly higher than those of water. The proposed approach therefore constitutes a transferable, cross-disciplinary methodological standard, indispensable for the accurate characterisation of nanoparticles across all fields of application.

5. Conclusions

Data collected demonstrate that the conventional approach based on water viscosity is not biometrically suitable for ILM analysis in canine plasma. The correction method tested offers a more accurate, scientifically sound and clinically necessary solution for standardising the characterisation of nanoparticles derived from different biological matrices.

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Research

Derivation of Bending Energy of Thin, Mildly Curved, Laterally Isotropic Elastic Shell

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Abstract:

Minimization of the energy of the system is a principle behind comprehension of stable membraneous structures, making the expression for the membrane energy a central problem in description of different systems. Here we present a derivation of the bending energy of a thin, mildly curved, laterally isotropic elastic shell from the basic assumptions regarding the deformation and stress. Using the equations of the differential geometry, the bending energy is expressed in terms of the invariants of the curvature tensor: trace and determinant. It is shown which assumptions are necessary to arrive to the result which is widely used in particular in membrane biophysics for description of the shapes of membrane-enclosed microscopic structures.

Keywords: Bending energy; Curvature; Differential geometry; Thin shells; Elastic modulus; Membrane

1 Free energy volume density of a deformed body

When impacted by external forces, the bodies deform. Deformation can be described in a way to give the change of the radius vector from the origin of the coordinate system to a chosen point on the body before deformation $\mathbf{r} = (x_1, x_2, x_3)$ and after the deformation $\mathbf{r}' = (x'_1, x'_2, x'_3)$,

$$\mathbf{u} = \mathbf{r}' - \mathbf{r}, \quad (1)$$

where $\mathbf{u}$ stands for the deformation vector. Written in components:

$$u_i = x'_i - x_i, \quad i = 1, 2, 3. \quad (2)$$

The distances between two nearby points before and after the deformation dl and dl' , respectively, are mutually connected,

$$dl'^2 - dl^2 + \sum_{i,k} 2\epsilon_{ik} dx_i dx_k, \quad (3)$$

where ϵ_{ik} stands for the deformation tensor $\underline{\epsilon}$. For small deformations,

$$\epsilon_{ik} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i} \right). \quad (4)$$

It can be seen that $\underline{\epsilon}$ is a symmetrical tensor.

An external force $\mathbf{F}$ is acting on a chosen infinitesimal volume of an isotropic body,

$$\mathbf{F} = \int \mathbf{f} dV, \quad (5)$$

where $\mathbf{f}$ is the force density and dV is the volume element. Since the forces, which are causing deformation, are of a short range, the force $\mathbf{F}$ is acting on the chosen volume at the surface. Therefore integration over the volume can be replaced by the integration over the surface. Following the vector analysis, this can be done if we express the vector $\mathbf{f}$ as a divergence of a tensor of the 2.nd range,

$$f_i = \sum_{i,k} \frac{\partial \sigma_{ik}}{\partial x_k}, \quad (6)$$

where σ_{ik} is a component of the stress tensor. The stress tensor $\underline{\sigma}$ is symmetric,

$$\sigma_{ik} = \sigma_{ki}. \quad (7)$$

The change of the free energy volume density of the chosen volume of material due to bending is

$$dw_b = -s dT + \sum_{i,k} \sigma_{ik} d\epsilon_{ik}, \quad (8)$$

where s is the volume density of the entropy, and T is temperature. In the equilibrium at constant temperature, $dT = 0$,

$$\sigma_{ik} = \left(\frac{\partial w_b}{\partial \epsilon_{ik}} \right)_T. \quad (9)$$

We express the free energy volume density w_b in terms of the deformation tensor. We consider small deformations so that we can expand w_b in terms of elements of the deformation tensor. If

the membrane is not deformed at a constant temperature $\epsilon_{ik} = 0$, there is no stress $\sigma_{ik} = 0$. It follows from Eq.(9), that the expansion should not include linear terms $\epsilon_{i,k}$. We consider that like w_b , the expansion terms should be scalars. From the deformation tensor we can compose two independent scalars of the second order: $(\sum_i \epsilon_{ii})^2$ and $\sum_{i,k} \epsilon_{ik}^2$. Volume density of the free energy is

$$w_b = \frac{\lambda}{2} \left(\sum_i \epsilon_{ii} \right)^2 + \mu \sum_{i,k} \epsilon_{ik}^2, \quad (10)$$

where λ and μ are the Lamée coefficients. We express the deformation as a sum of squeezing and shear deformation,

$$\epsilon_{ik} = \left(\epsilon_{ik} - \frac{1}{3} \delta_{ik} \sum_l \epsilon_{ll} \right) + \frac{1}{3} \delta_{ik} \sum_l \epsilon_{ll}, \quad (11)$$

where $\delta_{ik} = 1$ when $i = k$ and 0 when $i \neq k$. Inserting Eq.(11) into Eq.(10) yields

$$w_b = \frac{K}{2} \left(\sum_i \epsilon_{ii} \right)^2 + \mu \sum_{i,k} \left(\epsilon_{ik} - \frac{1}{3} \delta_{ik} \sum_l \epsilon_{ll} \right)^2 \quad (12)$$

where $1/K$ is called compressibility

$$K = \lambda + \frac{2}{3} \mu. \quad (13)$$

The coefficient μ is called the shear modulus. In order for f_b to reach a minimum at $\epsilon_{ik} = 0$, the coefficients K and μ should both be positive. The coefficients are connected with the Young modulus E and Poisson coefficient ν (Landau and Lifshitz, 1986, Kralj-Iglič, 1993),

$$E = \frac{8K\mu}{(3K + \mu)}, \quad (14)$$

$$\nu = \frac{(3K - 2\mu)}{2(3K + \mu)}. \quad (15)$$

Since K and μ are always positive, also E is positive, while ν can have different values between

$$-1 \leq \nu \leq \frac{1}{2}. \quad (16)$$

2 Free energy volume density of a thin, mildly curved shell

We consider that the shell is very thin and that the displacement of the membrane from the plane xy around a chosen point on the shell is described by a function $\zeta(x, y)$,

$$u_z = \zeta(x, y). \quad (17)$$

We neglect external forces comparing to the stresses, so the stresses are only tangential,

$$\sigma_{xz} = \sigma_{zx} = 2\mu\epsilon_{zx} = 0, \quad (18)$$

$$\sigma_{xz} = \sigma_{zx} = 2\mu\epsilon_{zx} = 0, \quad (19)$$

$$\sigma_{zz} = -\frac{\lambda}{2} \left(\epsilon_{xx} + \epsilon_{yy} + \left(1 + \frac{2\mu}{\lambda}\right) \epsilon_{zz} \right) = 0. \quad (20)$$

If we insert Eq.(4) into Eqs.(18)-(20), we get

$$\frac{\partial u_x}{\partial z} = -\frac{\partial \zeta(x, y)}{\partial x}, \quad (21)$$

$$\frac{\partial u_y}{\partial z} = -\frac{\partial \zeta(x, y)}{\partial y}, \quad (22)$$

$$\epsilon_{zz} = -\frac{1}{(1 + 2\mu/\lambda)}(\epsilon_{xx} + \epsilon_{yy}). \quad (23)$$

We integrate Eqs.(24) and (25) from 0 to z and consider that at $z = 0$ it should be fulfilled

$$u_x = z \frac{\zeta(x, y)}{\partial x}, \quad (24)$$

$$u_y = -\frac{\partial \zeta(x, y)}{\partial y}. \quad (25)$$

With derivation of expressions (24) and (25) we yield

$$\epsilon_{xx} = z \frac{\partial^2 \zeta(x, y)}{\partial x^2}, \quad (26)$$

$$\epsilon_{yy} = z \frac{\partial^2 \zeta(x, y)}{\partial y^2}, \quad (27)$$

$$\epsilon_{xy} = z \frac{\partial^2 \zeta(x, y)}{\partial x \partial y}. \quad (28)$$

It follows from Eqs.(18) and (19) that

$$\epsilon_{xz} = \epsilon_{yz} = 0, \quad (29)$$

and from Eq.(23) that

$$\epsilon_{zz} = -\frac{\lambda z}{(\lambda + 2\mu)} \left(\frac{\partial^2 \zeta(x, y)}{\partial x^2} + \frac{\partial^2 \zeta(x, y)}{\partial y^2} \right). \quad (30)$$

The free energy volume density is obtained by inserting Eqs.(26) - (30) into Eq.(10). After some rearrangement, we get

$$f_b = z^2 \left(\frac{2\mu(\lambda + \mu)}{(\lambda + 2\mu)} \left(\frac{\partial^2 \zeta(x, y)}{\partial x^2} + \frac{\partial^2 \zeta(x, y)}{\partial y^2} \right)^2 + 2\mu \left(\left(\frac{\partial^2 \zeta(x, y)}{\partial x \partial y} \right)^2 - \frac{\partial^2 \zeta(x, y)}{\partial x^2} \frac{\partial^2 \zeta(x, y)}{\partial y^2} \right) \right). \quad (31)$$

Bending energy of the shell W_b is

$$W_b = \int \int \int f_b \, dx \, dy \, dz. \quad (32)$$

Integration is performed over the surface and the thickness of the shell.

3 Curvature of the surface

The surface is described by the radius vector $\mathbf{R} = (x, y, \zeta(x, y))$. The equations of differential geometry yield the expressions for the trace and the determinant of the diagonalized curvature tensor $\underline{C}$, the trace being $\text{Tr } \underline{C} = C_1 + C_2$ and $\text{Det } \underline{C} = C_1 C_2$,

$$C_1 + C_2 = \frac{(EN + GL - 2FM)}{(EG - F^2)}, \quad (33)$$

$$C_1 C_2 = \frac{(LN - M^2)}{(EG - F^2)}, \quad (34)$$

where C_1 and C_2 are the principal curvatures.

$$E = 1 + \left(\frac{\partial \zeta(x, y)}{\partial x} \right)^2, \quad (35)$$

$$F = \frac{\partial \zeta(x, y)}{\partial x} \frac{\partial \zeta(x, y)}{\partial y}, \quad (36)$$

$$G = 1 + \left(\frac{\partial \zeta(x, y)}{\partial y} \right)^2, \quad (37)$$

$$L = \frac{\frac{\partial^2 \zeta(x, y)}{\partial x^2}}{\sqrt{\left(1 + \frac{\partial^2 \zeta(x, y)}{\partial x^2} + \frac{\partial^2 \zeta(x, y)}{\partial y^2} \right)}}, \quad (38)$$

$$M = \frac{\frac{\partial \zeta(x, y)}{\partial x} \frac{\partial \zeta(x, y)}{\partial y}}{\sqrt{\left(1 + \frac{\partial^2 \zeta(x, y)}{\partial x^2} + \frac{\partial^2 \zeta(x, y)}{\partial y^2} \right)}}, \quad (39)$$

$$N = \frac{\frac{\partial^2 \zeta(x, y)}{\partial y^2}}{\sqrt{\left(1 + \frac{\partial^2 \zeta(x, y)}{\partial x^2} + \frac{\partial^2 \zeta(x, y)}{\partial y^2} \right)}}. \quad (40)$$

For small $\partial \zeta(x, y)/\partial x$,

$$EG - F^2 \simeq 1, \quad (41)$$

$$C_1 + C_2 \simeq \frac{\partial^2 \zeta(x, y)}{\partial x^2} + \frac{\partial^2 \zeta(x, y)}{\partial y^2} \quad (42)$$

and

$$C_1 C_2 = \frac{\partial^2 \zeta(x, y)}{\partial x^2} \frac{\partial^2 \zeta(x, y)}{\partial y^2} - \left(\frac{\partial^2 \zeta(x, y)}{\partial x \partial y} \right)^2. \quad (43)$$

The surface area element is

$$dA = \sqrt{(EG - F^2)}, \quad (44)$$

and the volume element is

$$dV = dx \, dy \, dz. \quad (45)$$

4 Bending energy of thin mildly curved membrane

Comparing the expression (31) with expressions (42) and (43), we recognize (Struik, 1961; Do Carmo, 2016),

$$W_b = \frac{1}{2}k_c \iint (C_1 + C_2)^2 dA + \frac{1}{2}k_G \iint C_1 C_2 dA, \quad (46)$$

where the elastic constants k_c and k_G are

$$k_c = \frac{2\mu(\lambda + \mu)h^3}{6(\lambda + 2\mu)}, \quad (47)$$

$$k_G = -\frac{1}{3}\mu h^3, \quad (48)$$

and h is the thickness of the membrane. If we express the elastic constants with the Young modulus E and the Poisson coefficient σ , we get

$$k_c = \frac{Eh^3}{12(1 - \sigma^2)}, \quad (49)$$

$$k_G = -\frac{Eh^3}{6(1 + \sigma)}. \quad (50)$$

Since the Young modulus is always positive and the Poisson coefficient cannot be smaller than -1 and larger than 1/2, it follows from Eqs.(16), (49), and (50) that the bending constant k_c is positive and the Gaussian constant k_G is negative.

5 Conclusions

Starting from deformation of the material due to forces acting on a surface, and taking into account the equations of differential geometry, we derived the bending energy of a thin, mildly curved elastic shell.

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Research

Equivalent Characterisations of Singularity in Curvature-Coupled Membrane Functionals

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Abstract:

We study a curvature-based functional describing the coupling between isotropic bending and curvature anisotropy in membrane shapes. The functional is reformulated in invariant geometric form, enabling a structural analysis from three complementary viewpoints: the quadratic curvature kernel, the linearised shape operator, and the Euler–Lagrange equations. In each formulation, a common threshold is identified at which the highest-order curvature contribution degenerates. This provides an equivalent characterisation of the resulting singular behaviour, linking it to a loss of ellipticity of the associated operator and to a vanishing of curvature stiffness in the deviatoric mode. The coincidence of these conditions across all three perspectives demonstrates that the singularity is intrinsic to the model and independent of the chosen representation. These results clarify the structural origin of degeneracy in curvature-coupled membrane theories.



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Keywords: Curvature-based functionals; Membrane bending; Curvature anisotropy; Loss of ellipticity; Singular behaviour; Variational formulation

1 Introduction

The equilibrium shapes of fluid membranes are commonly described within the framework of curvature elasticity, where the bending energy is expressed in terms of the principal curvatures of the surface. In the classical Helfrich model (Helfrich, 1973; Deuling and Helfrich, 1976), the energy density depends quadratically on the mean curvature, leading to a variational problem governed by a fourth-order geometric operator. This approach successfully captures a wide range of membrane morphologies observed in lipid vesicles and related systems (Helfrich, 1973, Deuling and Helfrich, 1976, Seifert, 1997).

In many situations of physical and biological interest, however, the membrane exhibits anisotropic behaviour. This may arise from orientational ordering of lipid molecules, the presence of embedded proteins, or other structural inhomogeneities. Such effects introduce a coupling not only to the mean curvature but also to the curvature anisotropy, measured by the difference of the principal curvatures. This motivates the inclusion of additional terms in the energy functional that depend on the curvature deviator.

A specific model incorporating such effects was proposed in (Kralj-Iglič et al., 2006), based on a microscopic description of quadrupolar ordering of phospholipid molecules. In this formulation, the curvature energy density is given by

$$f = \frac{1}{4}(c_1 + c_2)^2 - \kappa \ln \cosh(2\vartheta d), \quad (1)$$

where c_1 and c_2 denote the principal curvatures of the surface normalized by the curvature of the sphere with the same area A , $c_1 = C_1 R_s$, $c_2 = C_2 R_s$, $A = 4\pi R_s^2$. The normalized mean curvature h and the normalized curvature deviator d are defined by

$$h = \frac{c_1 + c_2}{2}, \quad d = \frac{c_1 - c_2}{2}. \quad (2)$$

The parameter κ characterises the strength of the anisotropic interaction, while ϑ controls the sensitivity of this interaction to curvature anisotropy. The first term corresponds to the classical isotropic bending energy, whereas the second term introduces a nonlinear dependence on the curvature deviator.

This model predicts a rich variety of equilibrium shapes and, in particular, exhibits the formation of narrow neck regions in axisymmetric configurations. Numerical continuation results reported in (Kralj-Iglič et al., 2006) reveal the presence of parameter intervals in which the governing equations become singular and solutions cannot be continued. While this behaviour is clearly visible at the level of the axisymmetric shape equations, its structural origin is not immediately apparent.

In particular, it is not clear whether the observed singularity is a consequence of the chosen parametrisation, a limitation of the numerical method, or an intrinsic feature of the underlying variational formulation. Addressing this question requires an analysis that does not rely on a specific coordinate representation, but instead examines the structure of the geometric functional itself.

The aim of the present work is to provide such a structural interpretation. Rather than focusing on individual solutions, we analyse the curvature functional and identify the mechanism responsible for the loss of regularity. Our approach is based on three complementary viewpoints: the quadratic curvature kernel, which captures the highest-order contribution to the energy; the linearised shape operator, whose principal symbol determines ellipticity; and the full Euler–Lagrange equations, which describe the nonlinear equilibrium conditions.

We show that all three perspectives lead to the same threshold condition. At this threshold, the contribution associated with curvature anisotropy cancels the isotropic bending stiffness

in the deviatoric mode. As a result, the quadratic form loses coercivity, the principal symbol becomes non-positive, and the highest-order term in the Euler–Lagrange equations vanishes. This identifies the singular behaviour observed in (Kralj-Iglić et al., 2006) as a manifestation of a loss of ellipticity of the underlying geometric operator.

The present analysis thus provides a unified interpretation of the transition observed in the model, connecting its energetic, differential, and nonlinear aspects. In this way, the neck singularity can be understood as a structural consequence of the loss of curvature stiffness in the anisotropic mode.

2 Invariant form of the functional

Let C_1, C_2 denote the principal curvatures,

$$H = \frac{C_1 + C_2}{2}, \quad D = \frac{C_1 - C_2}{2}. \quad (3)$$

The functional derived in (Kralj-Iglić et al., 2006) can be written as

$$F(S) = \int (w_i(2H)^2 - w_a\Phi(D)) \, dA, \quad (4)$$

where

$$\Phi(D) = \ln \cosh(\alpha D), \quad (5)$$

w_i, w_a and α are constants, and S is the arclength of the contour.

The first (isotropic) term in Eq.(4) penalises mean curvature, while the second (deviatoric) term favours curvature anisotropy.

3 Connection to the dimensionless formulation

Using the dimensionless quantities,

$$f = w_b + f_d, \quad w_b = \frac{1}{4}(c_1 + c_2)^2, \quad f_d = -\kappa \ln \cosh(2\vartheta d), \quad (6)$$

with

$$h = \frac{c_1 + c_2}{2}, \quad d = \frac{c_1 - c_2}{2}. \quad (7)$$

The correspondence is

$$w_i = \frac{1}{4}, \quad w_a = \kappa, \quad \alpha = 2\vartheta. \quad (8)$$

4 Quadratic curvature kernel

To analyse the structural well-posedness of the functional, we isolate the highest-order quadratic dependence on curvature. This corresponds to extracting the leading contribution of the energy density in terms of the principal curvatures.

4.1 Expansion of the deviatoric term

The deviatoric contribution is given by

$$\Phi(D) = \ln \cosh(\alpha D). \quad (9)$$

For small D , we expand:

$$\ln \cosh(\alpha D) = \frac{\alpha^2}{2} D^2 + O(D^4). \quad (10)$$

Substituting into (4), the functional becomes, to quadratic order,

$$Q = \int \left(w_i (2H)^2 - w_a \frac{\alpha^2}{2} D^2 \right) dA. \quad (11)$$

This expression captures the highest-order curvature dependence and therefore determines the principal part of the associated Euler–Lagrange operator.

4.2 Representation in principal curvatures

We now rewrite the quadratic kernel in terms of the principal curvatures C_1, C_2 ,

$$H = \frac{C_1 + C_2}{2}, \quad D = \frac{C_1 - C_2}{2}. \quad (12)$$

Then

$$(2H)^2 = (C_1 + C_2)^2, \quad D^2 = \frac{(C_1 - C_2)^2}{4}. \quad (13)$$

Substituting into (11), we obtain

$$Q = \int \left[w_i (C_1 + C_2)^2 - \frac{w_a \alpha^2}{8} (C_1 - C_2)^2 \right] dA. \quad (14)$$

Expanding both terms,

$$(C_1 + C_2)^2 = C_1^2 + 2C_1 C_2 + C_2^2, \quad (15)$$

$$(C_1 - C_2)^2 = C_1^2 - 2C_1 C_2 + C_2^2. \quad (16)$$

Thus

$$Q = \int \left[\left(w_i - \frac{w_a \alpha^2}{8} \right) (C_1^2 + C_2^2) + \left(2w_i + \frac{w_a \alpha^2}{4} \right) C_1 C_2 \right] dA. \quad (17)$$

4.3 Mode decomposition

It is more instructive to diagonalise the quadratic form in curvature modes. We consider two fundamental modes: Mean curvature mode ($C_1 = C_2$) and deviatoric mode ($C_1 = -C_2$).

4.3.1 Mean curvature mode ($C_1 = C_2 = C$)

$$H = C, \quad D = 0, \quad (18)$$

$$Q \sim 4w_i C^2. \quad (19)$$

Thus the corresponding curvature stiffness, meaning the coefficient multiplying the quadratic curvature contribution in this mode, is denoted by λ_+ :

$$\lambda_+ = w_i. \quad (20)$$

4.3.2 Deviatoric mode ($C_1 = -C_2 = C$)

$$H = 0, \quad D = C, \quad (21)$$

$$Q \sim -\frac{w_a \alpha^2}{2} C^2. \quad (22)$$

Combining both contributions yields eigenvalues

$$\lambda_{\pm} = w_i \pm \frac{w_a \alpha^2}{4}. \quad (23)$$

Here λ_+ and λ_- are the eigenvalues of the quadratic curvature form after diagonalisation into the mean and deviatoric curvature modes. They represent the effective curvature stiffnesses associated with these two independent modes.

4.4 Coercivity condition

The quadratic form is positive definite if and only if both eigenvalues, namely the mean-mode eigenvalue λ_+ and the deviatoric-mode eigenvalue λ_- , are positive:

$$\lambda_+ > 0, \quad \lambda_- > 0. \quad (24)$$

This gives

$$-4w_i < w_a \alpha^2 < 4w_i. \quad (25)$$

Within this interval, the quadratic curvature kernel is coercive, and the highest-order contribution to the energy is strictly positive.

4.5 Interpretation of the loss of deviatoric curvature stiffness

The isotropic term always contributes positive stiffness through the mean curvature mode. In contrast, the deviatoric term acts as a *negative stiffness* in the anisotropic mode. As the parameter $w_a \alpha^2$ increases, it progressively cancels the isotropic bending stiffness in the deviatoric direction. At the threshold

$$w_a \alpha^2 = 4w_i, \quad (26)$$

the deviatoric stiffness vanishes:

$$\lambda_- = 0. \quad (27)$$

Beyond this point, the quadratic form becomes indefinite, and the curvature functional is no longer coercive in the deviatoric mode.

4.6 Connection to the dimensionless formulation

We now repeat the analysis in the dimensionless variables used in (Kralj-Iglič et al., 2006). The density reads

$$f = \frac{1}{4}(c_1 + c_2)^2 - \kappa \ln \cosh(2\vartheta d). \quad (28)$$

For small d ,

$$\ln \cosh(2\vartheta d) = 2\vartheta^2 d^2 + O(d^4). \quad (29)$$

Thus

$$f_{\text{quad}} = \frac{1}{4}(c_1 + c_2)^2 - 2\kappa\vartheta^2 d^2. \quad (30)$$

In terms of

$$h = \frac{c_1 + c_2}{2}, \quad d = \frac{c_1 - c_2}{2}, \quad (31)$$

this becomes

$$f_{\text{quad}} = 2h^2 - 2\kappa\vartheta^2 d^2. \quad (32)$$

The eigenvalues are therefore

$$\lambda_h = 1, \quad \lambda_d = 1 - 2\kappa\vartheta^2. \quad (33)$$

Positivity requires

$$2\kappa\vartheta^2 < 1. \quad (34)$$

4.7 Conclusion (kernel viewpoint)

Both invariant and dimensionless formulations lead to the same structural condition. The quadratic curvature kernel loses positivity when the deviatoric stiffness vanishes. This threshold will reappear in the analysis of the linearised operator and the Euler–Lagrange equations.

5 Coercivity and ellipticity

Positivity of the quadratic curvature kernel implies positivity of the principal part of the linearised operator. In particular, the fourth-order geometric operator is elliptic when both eigenvalues are positive (Evans, 2010; Gazzola, 2010). Thus ellipticity is equivalent to

$$\lambda_+ > 0, \quad \lambda_- > 0. \quad (35)$$

This establishes the structural threshold for well-posedness at the level of the quadratic kernel.

6 Interpretation of neck singularity

In the original axisymmetric formulation, singular behaviour appears in the curvature equations when a coefficient vanishes. From the present viewpoint, this corresponds to the loss of stiffness in the deviatoric mode, i.e. when

$$\lambda_- \rightarrow 0. \quad (36)$$

At this point, the quadratic kernel ceases to be positive definite and the operator loses ellipticity. Thus the neck singularity reflects a structural degeneration of the variational operator.

7 Linearised operator and principal symbol

We now analyse the same structural condition at the level of the linearised shape operator. While the quadratic curvature kernel determines the energetic coercivity, the linearised operator governs the differential structure of the Euler–Lagrange equations. In particular, ellipticity is determined by the principal symbol of this operator.

7.1 Normal variation of the surface

Let Σ be a smooth reference surface with unit normal n . Consider a normal variation

$$x \mapsto x + \eta n, \quad (37)$$

where η is a scalar perturbation field. To leading order, the variation of the mean curvature is given by

$$\delta H = -\Delta_{\Sigma} \eta + \text{lower-order terms}, \quad (38)$$

where Δ_{Σ} is the Laplace–Beltrami operator on the surface. The deviatoric curvature

$$D = \frac{C_1 - C_2}{2} \quad (39)$$

also depends linearly on second derivatives of η . To leading order, one may write

$$\delta D = \mathcal{D}_{\Sigma} \eta + \text{lower-order terms}, \quad (40)$$

where $\mathcal{D}_{\Sigma}$ is a second-order tangential operator. Its precise form depends on the local geometry, but its principal part is of the same differential order as Δ_{Σ} .

7.2 Second variation of the functional

We now substitute these variations into the quadratic part of the energy. From Section 4, the quadratic density is

$$f_{\text{quad}} = 2H^2 - 2\kappa\vartheta^2 D^2. \quad (41)$$

The second variation therefore contains terms of the form

$$\delta^2 F \sim \int_{\Sigma} [2(\delta H)^2 - 2\kappa\vartheta^2 (\delta D)^2] dA. \quad (42)$$

Substituting (38)–(40), we obtain, at leading order,

$$\delta^2 F \sim \int_{\Sigma} [2(\Delta_{\Sigma} \eta)^2 - 2\kappa\vartheta^2 (\mathcal{D}_{\Sigma} \eta)^2] dA. \quad (43)$$

Thus both contributions involve second derivatives of η , and the resulting operator is of fourth order [6].

7.3 Principal symbol

To determine ellipticity, we analyse the operator in local coordinates or Fourier space. Let k denote the tangential wavevector. Then:

$$\Delta_{\Sigma}\eta \longrightarrow -|k|^2\hat{\eta}, \quad (44)$$

$$\mathcal{D}_{\Sigma}\eta \longrightarrow \sigma_D(k)\hat{\eta}, \quad (45)$$

where $\sigma_D(k)$ is homogeneous of degree two in k . Consequently,

$$(\Delta_{\Sigma}\eta)^2 \sim |k|^4|\hat{\eta}|^2, \quad (46)$$

$$(\mathcal{D}_{\Sigma}\eta)^2 \sim \gamma(\theta)|k|^4|\hat{\eta}|^2, \quad (47)$$

where $\gamma(\theta)$ captures the angular dependence of the deviatoric mode in tangent space. The principal symbol of the linearised operator is therefore

$$\sigma(k) = (2 - 2\kappa\vartheta^2\gamma(\theta)) |k|^4. \quad (48)$$

7.4 Ellipticity condition

Ellipticity requires that the principal symbol be strictly positive for all nonzero k (Evans, 2010):

$$\sigma(k) > 0 \quad \text{for all } k \neq 0. \quad (49)$$

Since $\gamma(\theta)$ is bounded and achieves its maximum in the pure deviatoric direction, the worst-case coefficient is

$$2(1 - 2\kappa\vartheta^2). \quad (50)$$

Thus ellipticity holds if and only if

$$2\kappa\vartheta^2 < 1. \quad (51)$$

7.5 Interpretation of the loss of ellipticity

The operator consists of two competing fourth-order contributions:

- the isotropic bending term, proportional to $(\Delta_{\Sigma}\eta)^2$, which provides positive stiffness,
- the deviatoric term, proportional to $(\mathcal{D}_{\Sigma}\eta)^2$, which acts as a negative stiffness in the anisotropic mode.

As the parameter $\kappa\vartheta^2$ increases, the deviatoric contribution progressively cancels the isotropic bending stiffness. At the critical threshold

$$2\kappa\vartheta^2 = 1, \quad (52)$$

the principal symbol vanishes in the deviatoric direction. At this point, the operator is no longer strictly elliptic.

7.6 Conclusion (operator viewpoint)

The linearised operator loses ellipticity precisely at the same threshold identified in the quadratic curvature kernel. This confirms that the loss of coercivity at the energy level corresponds to a loss of ellipticity at the level of the governing differential operator.

8 Euler–Lagrange structure and degeneracy

We now analyse the same structural condition at the level of the full Euler–Lagrange equations. While the quadratic kernel determines energetic coercivity and the linearised operator determines ellipticity, the Euler–Lagrange system describes the fully nonlinear governing equations of equilibrium shapes.

8.1 Structure of the axisymmetric equations

In the original formulation of [4], the surface is represented by an axisymmetric meridian curve. The Euler–Lagrange equations obtained from the functional involve curvature-dependent coefficients multiplying the highest-order derivatives. In particular, the shape equations contain a factor of the form

$$B = 1 - \frac{2\kappa\vartheta^2}{\cosh^2(2\vartheta d)}, \quad (53)$$

where d is the deviatoric curvature. This factor multiplies the highest-order curvature terms in the differential equation governing the meridian profile. This degeneracy is consistent with the behaviour observed in earlier numerical studies of the same class of functionals [4]. In particular, a finite parameter interval exists in which the Euler–Lagrange system ceases to admit regular numerical solutions. Within this interval, the associated Lagrange multipliers tend to zero, indicating a loss of constraint enforcement at the level of the variational formulation, while the total energy remains smooth and does not exhibit any discontinuity. This separation between the regularity of the solution and the behaviour of the energy reflects the vanishing of the highest-order curvature contribution, rather than any energetic instability.

8.2 Degeneracy condition

The function $\cosh^2(x)$ satisfies

$$\cosh^2(x) \geq 1 \quad \text{for all } x. \quad (54)$$

Therefore,

$$\frac{1}{\cosh^2(2\vartheta d)} \leq 1, \quad (55)$$

and the factor B satisfies

$$B \geq 1 - 2\kappa\vartheta^2. \quad (56)$$

The first occurrence of degeneracy is therefore at

$$2\kappa\vartheta^2 = 1, \quad (57)$$

which corresponds to $d = 0$. At this point,

$$B = 0, \quad (58)$$

and the coefficient of the highest-order derivative vanishes.

8.3 Interpretation of the singularity

When the factor B vanishes, the differential equation loses its highest-order term. As a result, the system ceases to be of fourth order at that point, and the standard theory of elliptic equations no longer applies (Evans, 2010). This leads to the appearance of singular behaviour in the curvature equations, observed in the form of narrow necks in the membrane shape. From this perspective, the singularity is not merely a geometric feature of the surface, but reflects a degeneration of the differential operator governing the equilibrium.

8.4 Relation to curvature stiffness

The factor B can be interpreted as an effective stiffness coefficient associated with the deviatoric curvature mode. As the parameter $\kappa\vartheta^2$ increases, this coefficient decreases. At the threshold

$$2\kappa\vartheta^2 = 1, \tag{59}$$

the effective stiffness vanishes, and beyond this point it becomes negative in certain regimes. This behaviour mirrors the loss of positivity of the quadratic curvature kernel discussed in Section 4, and the loss of ellipticity of the linearised operator in Section 7. Representative numerical solutions of the same functional are shown in **Figure. 1**.

8.5 Conclusion (Euler–Lagrange viewpoint)

The degeneracy of the Euler–Lagrange equations occurs at the same threshold identified in the quadratic kernel and linearised operator analyses. This shows that the singularity observed in the axisymmetric formulation corresponds to the loss of ellipticity of the underlying geometric operator.

9 Unified interpretation and conclusion

We have analysed the curvature functional from three complementary viewpoints:

- the quadratic curvature kernel (energy level),
- the linearised operator (principal symbol),
- the Euler–Lagrange equations (nonlinear level).

Each of these approaches captures the behaviour of the system at a different level: energetic, differential, and fully nonlinear. Despite this difference, all three lead to the same structural condition. At the level of the quadratic kernel, the deviatoric eigenvalue vanishes and the energy loses coercivity. At the level of the linearised operator, the principal symbol becomes non-positive and ellipticity is lost. At the level of the Euler–Lagrange equations, the coefficient of the highest-order derivative vanishes, and the system becomes singular. These three phenomena therefore represent different manifestations of the same underlying transition. The neck singularity observed in the membrane shapes can thus be interpreted as a consequence of the loss of curvature stiffness in the deviatoric mode. This provides a consistent structural explanation that connects the variational formulation, the differential operator, and the nonlinear governing equations. The present framework clarifies the mathematical nature of the model and provides a basis for further extensions, including the study of stabilising mechanisms and nonlinear saturation effects.

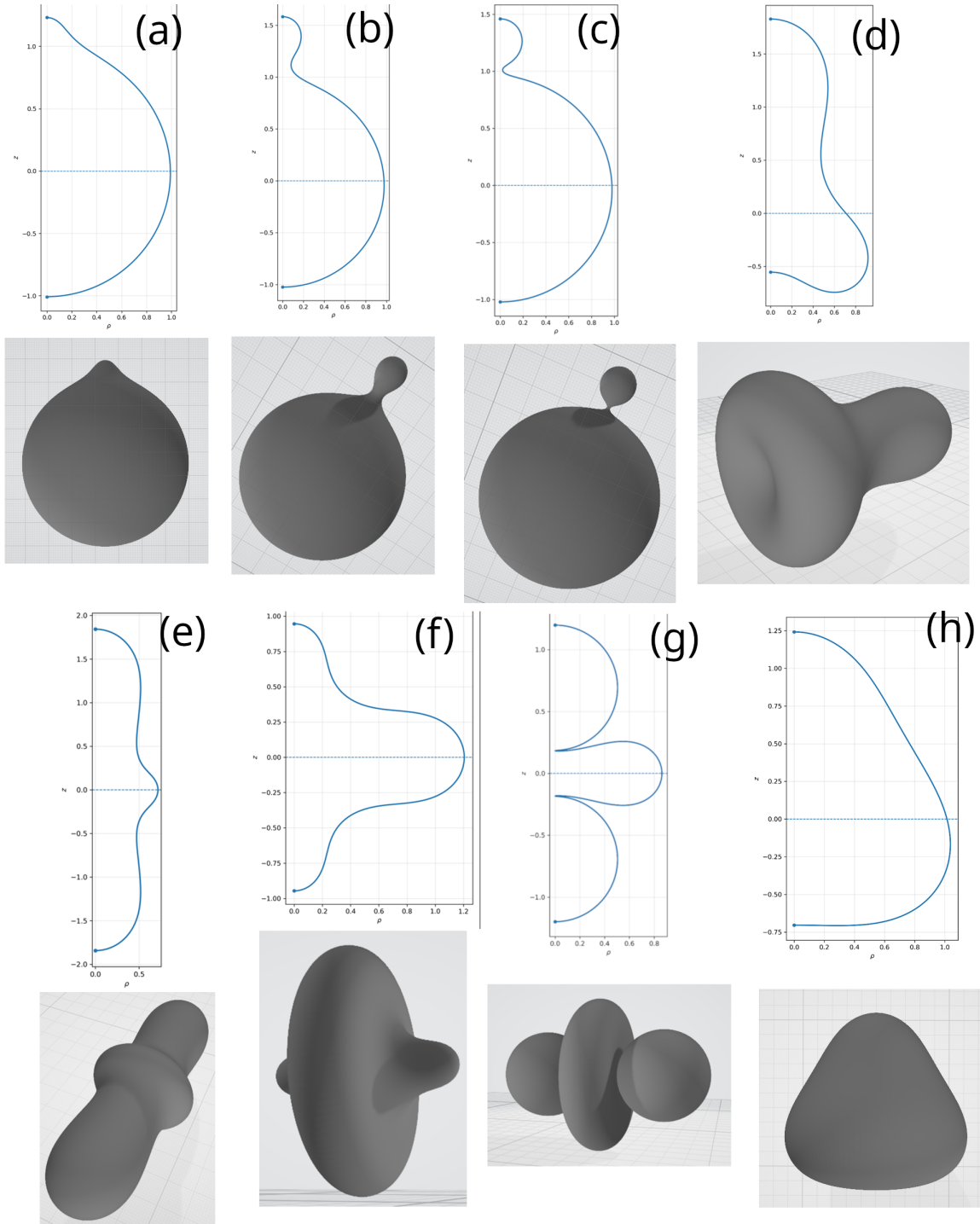


Figure 1: Representative axisymmetric equilibrium shapes obtained numerically from the analysed functional using a predictor-corrector scheme. Panels (a)–(h) show generating profiles $\rho(z)$ together with the corresponding reconstructed three-dimensional surfaces. The examples illustrate the diversity of stationary solutions obtained for different prescribed values of volume V and surface area A . The parameters of the entire shape given in panels (a) $v = 0.9920$, $\langle h \rangle = 1.0101$, (b) $v = 0.9500$, $\langle h \rangle = 1.1324$, (c) $v = 0.9500$, $\langle h \rangle = 1.1664$, (d) $v = 0.7325$, $\langle h \rangle = 1.1473$, (e) $v = 0.6950$, $\langle h \rangle = 1.2380$, (f) $v = 0.6950$, $\langle h \rangle = 1.1120$, (g) $v = 0.4950$, $\langle h \rangle = 1.7379$, (h) $v = 0.9500$, $\langle h \rangle = 1.0165$. Shape (b) data coordinates available at (Židánek et al., 2025). All shapes data on plots and 3D objects data available at (Židánek et al., 2026).

The singularity identified in the present analysis should not be interpreted merely as a numerical artefact or a consequence of parametrisation. Rather, it reflects a change in the structure of the governing equations. At the critical threshold, the coefficient of the highest-order curvature term vanishes, and the associated operator is no longer strictly elliptic. In this regime, the standard elliptic framework no longer applies, and the behaviour of solutions is governed by a degenerate structure. This suggests that the observed singular behaviour is linked to a loss of curvature stiffness in the deviatoric mode, although a detailed analysis of the resulting solution properties lies beyond the scope of the present work.

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