



XVI International Workshop on COMPLEX SYSTEMS 13-17 March, 2023 - Andalo (Trento)

Book of abstracts

Contents

I	Programme	3
II	Talks	21
	Poster	109
Index		146

Part I Programme

Monday, 13 March

15.00

Opening

Giacomo Baldi, Roberto Menichetti, Raffaello Potestio, and Marco Zanatta

Chair: Walter Schirmacher

15.10

Absence of cooperativity near the glass transition

Matthieu Wyart

15.30

Probing the structure and relaxation in the "no-man's land" of phase-change materials Shuai Wei

15.50

Studying dynamics and crystallization in nanometric size vapor-deposited films of molecular glass-former

Karolina Adrjanowicz

16.10

Recent results on intrinsic droplet dynamics in emulsions from experiments in microgravity onboard the International Space Station

Luigi Cristofolini

16.30 - 17.00

Coffee Break

Chair: Luigi Cristofolini

17.00

Designing enhanced entropy binding in associative polymers Lorenzo Roviqatti

17.20

Programmable soft matter

Jasna Brujic

17.40

Distance-as-time in physical aging

Jeppe Dyre

18.00

Data-driven modeling of intrinsically disordered proteins: phase separation and proteomewide analysis

Giulio Tesei

18.20

Chair: Giancarlo Ruocco

18.30 **Multiple equilibria** Giorgio Parisi

19.15

End of the session

20.00

Dinner

21.00 - 23.00

POSTER SESSION

Tuesday, 14 March

Chair: Miguel Angel Ramos

8.30

Theory of non-phononic excitations in disordered systems

Walter Schirmacher

8.50

Vibrational properties of particulate physical gels

Hideyuki Mizuno

9.10

Hump universality and singularity in low-temperature heat capacity of crystalline and amorphous solids: boson peak or van Hove signature

Alexander I. Krivchikov

9.30

On the microscopic nature of two-level systems in glasses

Felix-Cosmin Mocanu

9:50

From the vibrational properties of silicate glasses to the viscosity of their parental melts: a model

Michele Cassetta

10.05

The Debye-Waller factor of amorphous glass from static speckle patterns

Charlotte F. Petersen

10.20 - 10.50

Coffee Break

Chair: Shuai Wei

10.50

Transverse dynamics in fluids: how a collective excitation sets in

Ubaldo Bafile

11.10

Cooperative dynamics in bulk water

Margarita Russina

11.30

Structural relaxation and nano-domain dynamics in highly concentrated electrolytes for zinc anode batteries

Antonio Faraone

11.50

Microscopic collective dynamics in liquid neon-deuterium mixtures: neutron scattering and quantum simulations

Daniele Colognesi

12.05

Where is the hydrodynamic limit?

Jesper Schmidt Hansen

12.20

Supercooled liquid tellurium: water's distant relative?

Peihao Sun

12.35 - 15.30

Lunch Break

Chair: Simone Capaccioli

15.30

Athermal bond-breaking pathways in glassy chalcogenide materials

Konstantinos Konstantinou

15.50

Reversing Peierls-like distortions elicits the polyamorphic transition in GeTe and GeSe Tomoki Fujita

16.10

Microscopic origin of resistance fluctuation in germanium telluride glass

Sebastian Walfort

16.25

Structural, dynamical and electronic heterogeneities in supercooled-liquid antimony from abinitio simulations

Nils Holle

16.40

The ESRF Extremely Brilliant Source for X-ray photon correlation spectroscopy at the beamline ID10

Federico Zontone

17.00 - 17.30

Coffee Break

Chair: Alessandro Paciaroni

17.30

Capillary thinning of polymer solution threads: self-similar configurations and instabilities Alexander N. Semenov

17.45

Untangling the role of tau in neurodegeneration and neurodevelopment using 3D human cortical organoids

Silvia Di Angelantonio

18.05

Self-organization in neural rosette morphogenesis

Giorgio Gosti

18.20

Exploring non-equilibrium nanoscale processes in a heated egg yolk using coherent X-rays Nimmi Das Anthuparambil

18.35

Sperm cells in micro-traps: motility sorting and swimming precision Claudio Maggi

> 18.55 End of the session

19.20

Meeting @ Malga Dosson

Wednesday, 15 March

Chair: Jasna Brujic

8.30

Intra-protein interacting collective modes in the terahertz frequency region

Valeria Conti Nibali

8.50

Probing cage relaxation in concentrated protein solutions by XPCS

Yuriy Chushkin

9.10

Diffusive dynamics of bacterial proteome as a proxy of cell death

Alessandro Paciaroni

9.30

Dynamic cluster formation, viscosity and diffusion in monoclonal antibody solutions Ilaria Mosca

9.45

Optimality in self-organized molecular sorting

Andrea Gamba

10.05

Transition path sampling with a quantum computer Danial Ghamari

10.20 - 10.50

Coffee Break

Chair: Jeppe Dyre

10.50

Do structural relaxations in supercooled liquids have the generic line shape? Simone Capaccioli

11.10

Excitation populations provide a thermodynamic order parameter for liquids

Marcus T. Cicerone

11.30

The dielectric and rheological responses of strongly asymmetric mixtures of glycerol/propanol Jan Philipp Gabriel

11.50

The Johari-Goldstein relaxation as probe of the mosaic structure of supercooled liquids Federico Caporaletti

12.05 Reaching the yield point of a glass during X-ray irradiation

Alessandro Martinelli

12.20

Amorphous-amorphous transformation induced in glasses by intense X-ray beams Erica Alfinelli

12.35 - 16.00

LUNCH BREAK

Chair: Margarita Russina

16.00

Origin of the anomalous properties of water determined using X-ray lasers Anders Nilsson

16.20

Water on the mesoscale and progress towards IXS with 0.3 meV resolution Alfred Baron

16.40

Progress and ppportunities for XPCS and XSVS at LCLS

Paul Fuoss

17.00

Unique phase transformations in glassy aqueous solutions containing ionic liquids Johannes Bachler

17.15

Instantaneous normal modes in liquids: theory and simulation

Stefano Mossa

17.30 - 18.00

Coffee Break

Chair: Matthieu Wyart

18.00

Stochastic atomic acceleration during the X-ray-induced fluidization of a silica glass Francesco Dallari

18.15

Pressure-driven disorder in glassy $As_2Se_3\colon$ structure, low-energy phonon dynamics and optical band gap

Giovanna D'Angelo

18.35

Numerical evidence of glass and pseudo-localization transitions in the mode-locked p-spin model for random lasers

Jacopo Niedda

18.50

Multimode fibers: an extremely flexible tool from fast imaging to reservoir computing Luca Leuzzi

19.10

Optical computation of the spin glass dynamics Marco Leonetti

> 19.30 End of the session

20.00

Social Dinner

Thursday, 16 March

Chair: Hideyuki Mizuno

8.30

Predicting scaling properties from a single configuration Thomas B. Schrøder

8.50

Liquid dynamics eases the equilibration of glasses

Simone Napolitano

9.10

Long-range concentration fluctuations and their effect on the thermodynamics of model supercooled liquids

Robin Cortes-Huerto

9.30

Fluctuation-dissipation relation and effective temperatures in out of equilibrium systems Federico Corberi

9.50

Experimental evidence of a crossover between cooperative relaxation and liquid growth dynamics

Marta Gonzalez-Silveira

10.05

The difference of aging near or far from equilibrium

Florian Pabst

10.20 - 10.50

Coffee Break

Chair: Lorenzo Rovigatti

10.50

Dynamics of passive and activated self assembled patchy particle architectures

Peter Bolhuis

11.10

Hydrodynamically-induced morphologies in active and driven matter

Ignacio Pagonabarraga

11.30

Towards designing of actively moving nanoparticles: theoretical estimation of optimal geometry and engine parameters

Irina A. Nyrkova

11.50 **Colloidal crystals and gels from active apolar colloids** Pietro Tierno

12:05

Quantifying the deviation from thermal equilibrium Lorenzo Costigliola

12:20 **2D crystals of squares** Peter Keim

12.35 - 16.00

LUNCH BREAK

Chair: Alexei Sokolov

16.00

Suppression of two-level systems with increasing stability in TPD glasses

Miguel Angel Ramos

16.20

Low-temperature thermal anomalies in ordered and disordered crystalline phases Josep Lluís Tamarit

16.40

Acoustic attenuation and dispersion in the vicinity of the boson peak

Marie Foret

17.00

Electric fields for tuning molecular orientation in TPD-modified glasses: experiments and simulations

Marta Rodríguez-López

17.15

Effect of secondary interactions on the dynamics of glass forming liquids: insights from the comparison of dielectric and mechanic techniques

Silvia Arrese-Igor

17.30 - 18.00

Coffee Break

Chair: Peter Bolhuis

18.00

A soft matter perspective of proteins: from phase behavior to dynamics Anastasia Ragulskaya 18.20

A computational strategy to modulate the compatibility of biomolecular interfaces: the importance in protein design

Lorenzo Di Rienzo

18.40

Bio-Inspired topological supramolecular materials: from circular catenanes to kDNA Luca Tubiana

19.00

Dissecting the role of division noise in promoting cancer phenotypic heterogeneity $\mathsf{M}\mathsf{attia}\ \mathsf{M}\mathsf{iotto}$

19.15

Brillouin light scattering microspectroscopy for biomedical applications Daniele Fioretto

19.35 End of the session

20.00

Dinner

21.00 - 23.00

POSTER SESSION

Friday, 17 March

Chair: Ignacio Pagonabarraga

8.30

Investigation of the mechanism of gellan aggregation at the molecular scale Letizia Tavagnacco

8.50

Static and dynamical properties of modulated hard-spheres fluid Michele Caraglio

9.10

Characterization of interpolyelectrolyte complexes

Matteo Chamchoum

9.25

Aggregation of fullerenes C60 in liquids: investigations and theoretical description Timur Tropin

9.45

Superparamagnetic nanoclusters in a strong applied field: understanding static and dynamic magnetic response

Andrey Kuznetsov

10.05

Self-assembly and magnetic susceptibility of colloidal magnetic nanoplatelets Margaret Rosenberg

10.20

Coffee Break

Chair: Simone Napolitano

10.50

Revealing the mechanism of high fragility in polymers

Alexei P. Sokolov

11.10

Chemical vitrification and universal scaling between cooperative motion, caging dynamics, and emergent elasticity

Silvia Corezzi

11.30

Evolution of relaxation time and dynamic heterogeneity through aging in metallic glasses Eloi Pineda

11.50

Denser glasses relax faster: a competition between rejuvenation and aging during in-situ high pressure compression at the atomic scale

Antoine Cornet

12.05

Exploring the densification effect on dynamics of metallic glass-forming liquid

Jie Shen

12.20

Mechanical memory of metallic glasses: time and temperature dependence of the anelastic relaxation

Mehran Nabahat

12.35

CONCLUSIVE REMARKS

Posters

Dismantling some common believes on the Debye relaxation of monoalcohols Silvia Arrese-Igor

Exploitation of the unique capabilities of an ultrafast nanocalorimeter at a synchrotron coherence beamline

Jacopo Baglioni

Combining X-ray diffraction and X-ray Raman spectroscopy for the investigation of irradiated shock-compressed silica glass samples

Lena Bussmann

Fast differential scanning calorimetry: advancements in data treatment to investigate organic molecular glass-formers

Simone Capaccioli

Micro-eV vibrational dynamics in amorphous SiO₂: evidences of additional modes? Federico Caporaletti

Thermoresponsivity of poly(N-isopropylacrylamide) microgels in water-trehalose solution and its relation to protein behaviour

Silvia Corezzi

D4pdf: from constant-wavelength total neutron scattering data to the pair distribution function Gabriel Cuello

Spatial organization of hydrophobic and charged residues affects protein thermal stability and binding affinity

Fausta Desantis

Reactive MD simulation of phosphate-based glasses Zohreh Fallah

Vibrational dynamics of ultra-stable and conventional glasses Irene Festi

CANVAS: a fast, accurate, and system-specific variable resolution approach for simulating biomolecules

Raffaele Fiorentini & Thomas Tarenzi

Isocompositional liquid-liquid transition at ambient pressure in dilute aqueous LiCl solutions Iohannes Giebelmann

Researching the formation of CO₂ structures in cryocondensate thin films Oleg Golikov

Electrostatic complementarity evaluation at the protein interfaces: prediction of transient protein-protein interactions

Greta Grassmann

Isochronal superposition of the structure for cumene Erik Hillo Lørup

Information-theoretical measures identify accurate low-resolution representations of proteins Margherita Mele

Dynamical and kinetic assessment of nucleic acids systems Manuel Micheloni

The shape complementarity evaluation at the interfaces of protein-protein complexes through a novel strategy based on 2D Zernike formalism Edoardo Milanetti

Magneto-mechanical response of magnetic filaments with solvophobic, super-paramagnetic colloids, in bulk

Deniz Mostarac

Investigating the action mechanism of pore-forming toxins with molecular dynamics simulations at different resolution scales

Costanza Paternoster & Thomas Tarenzi

Modelling liquid dynamics by random walks within overlapping hyperspyheres Mark Railton

Amorphizing or modifying materials by MeV ion irradiation Miguel Angel Ramos

Effect of high pressure on the atomic dynamics of a Au-based metallic glass Alberto Ronca

Spatial resolution of glass relaxation dynamics: a real-time microscopic view Marta Ruiz-Ruiz

Theory of non-phononic excitations in disordered systems Walter Schirmacher

The electric and magnetic disordered Maxwell equations as eigenvalue problem Walter Schirmacher

Thermodynamic limit in computer simulations via finite-size integral equations Mauricio Sevilla

Exploring pharmaceuticals with conventional and fast calorimetry Daniele Sonaglioni

Exploring the glassy dynamics of the Gaussian core model Vittoria Sposini

On the characteristic thermal conductivity plateau in model amorphous silica Daria Szewczyk

Pressure Scanning Volumetry: a useful alternative to Scanning Calorimetry Elpidio Tombari

Interaction of ionic surfactants with epoxy-based hydrogel investigated by SANS Timur Tropin Vibrational phenomena in glasses at low temperatures captured by field theory of disordered harmonic oscillators

Florian Vogel

Effect of recondensation on the glass transition of a cryofilm mixture with nitrogen Olga Vorobyova

Measuring the dynamics of complex systems using coherent X-ray radiation at beamline P10 Fabian Westermeier

Part II Talks

STUDYING DYNAMICS AND CRYSTALLIZATION IN NANOMETRIC SIZE VAPOR-DEPOSITED FILMS OF MOLECULAR GLASS-FORMER

K. Adrjanowicz

Institute of Physics, University of Silesia, 75 Pulku Piechoty 1a, 41-500 Chorzow, Poland.

E-mail: kadrjano@us.edu.pl

Vapor-deposited glasses got wide scientific attention as they are remarkably stable and dense materials with significant practical applications. The physical properties of ultrathin films have been widely investigated in the past decade, but the challenge lies in the fact that they possess characteristics deviating from the bulk behavior when they are confined by the thickness. Here, we present the results of experimental work on the effect of thickness and surface roughness on the α -relaxation dynamics and crystallization tendency of vapor-deposited films of low-molecular glass-former, celecoxib (CXB) probed by dielectric spectroscopy. Among that, we report differences in relaxation dynamics of vapor-deposited thin films above T_g compared to spin-coated counterparts of the same thickness and provide evidence of the presence of irreversibly adsorbed material in vapor-deposited films on a silicon substrate.

Amorphous-amorphous transformation induced in glasses by intense X-ray beams

E. Alfinelli¹, F. Caporaletti², F. Dallari³, A. Martinelli⁴, G. Monaco⁴, B. Ruta^{5,6}, M. Sprung³, M. Zanatta¹, G. Baldi¹

¹Physics Department, University of Trento, I-38123, Povo, Trento, Italy.
²Van der Waals-Zeeman Institute, Institute of Physics, University of Amsterdam, 1098XH Amsterdam, The Netherlands.
³Hasylab at DESY, Notkestr. 85, D-22607 Hamburg, Germany.

⁴Physics and Astronomy Department, University of Padova, I-35122, Padova, Italy.

⁵ESRF- The European Synchrotron, F-38043, Grenoble, France.

⁶Institut Lumière Matière, UMR5306 Université Lyon 1–CNRS, Université de Lyon, 69622, Villeurbanne Cedex,

France.

E-mail: erica.alfinelli@unitn.it

We report a recent investigation on microscopic dynamics in pure boron-oxide and a set of sodium silicate glasses by means of the X-ray Photon Correlation Technique (XPCS). Recent observations [1–3] on oxide glasses have revealed a fast structural relaxation well below the glass transition temperature with a inverse flux dependence of the measured characteristic time. The radiolysis is the responsible phenomenon for this beam induced effect in which the pumped photoelectrons by the x-rays have sufficient energy to break atomic bounds and make the nearby atoms to rearrange.

In this talk I will show how, at high delivered doses, a low energy $\epsilon \sim 8$ KeV X-rays beam induces a transformation of the initial glass into a new amorphous state which remains stable under irradiation. The radiation dose needed for the initial structural variation is sample dependent and correlates well with the number of constraints per vertex, within the framework of rigidity theory developed by Gupta [4]. The interaction with the X-ray beam propels the system to move across the energy landscape in the new amorphous phase which continues to rearrange under the beam with a stretched exponential relaxation suggesting a release of the stresses in the new configuration.

- [1] B. Ruta et al., Sci. Rep. 7, 3962 (2017).
- [2] K. Holzweber et al., Phys. Rev. B 100, 214305 (2019).
- [3] G. Pintori et al., Phys. Rev. B 105, 104207 (2022).
- [4] P.K. Gupta et al., J. Am. Chem. Soc. 76, 1088 (1993).

Exploring non-equilibrium nanoscale processes in a heated egg yolk using coherent X-rays

N. Das Anthuparambil^{1,2}, A. Girelli³, S. Timmermann², M. Kowalski², M. S. Akhundzadeh², S. Retzbach³, M. D. Senft³, M. Dargasz², D. Gutmüller³, A. Hiremath³, M. Moron⁴, Ö. Öztörk², H.-F. Poggemann³, A. Ragulskaya³, N. Begam³, A. Tosson², M. Paulus⁴, F. Westermeier¹, F. Zhang³, F. Schreiber³, M. Sprung¹, C. Gutt²

¹Deutsches Elektronen-Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany.
 ²Department Physik, Universität Siegen, 57072 Siegen, Germany.
 ³Institut für Angewandte Physik, Universität Tübingen, 72076 Tübingen, Germany.
 ⁴Fakultät Physik/DELTA, Technische Universität Dortmund, 44221 Dortmund, Germany.

E-mail: nimmi.das.anthuparambil@desy.de

Eqq yolk is widely utilized as a culinary component due to its high nutritious value and excellent emulsifying and gelling abilities [1,2]. When heated, it undergoes a solution-to- gel transition. It is known that the texture and visco-elastic properties of the final gel are a result of the high concentration and diversity of proteins and lipids found in the yolk [2]. Despite its versatile use in the food industry [1,2] and pharmaceuticals [3], a little is known about the functional contribution of its constituents to the final gel microstructure. Using low-dose X-ray photon correlation spectroscopy [4] in ultra-small angle X-ray scattering geometry, we follow the time-resolved structural and dynamical evolution of multiple non-equilibrium processes occurring in a heated hen egg yolk. Following key structural and dynamical features, we identify non-equilibrium processes such as denaturation and aggregation of proteins, protein gelation, gel aging, two-step aggregation of yolk low-density lipoproteins (LDLs), and gelation of yolk granules for wide time-temperature combinations. We find that the overall kinetics and dynamics governing protein denaturation, aggregation, and gelation follow Arrhenius-type time-temperature superposition (TTS). This implies an identical reaction pathway underlying these consecutive processes, with a temperature-dependent reaction rate. At high temperatures, TTS breaks down during gelation and temperature-independent gelation dynamics is observed. This indeed reflects the complex association of protein aggregates that results in a gel network. Furthermore, the two-step complex aggregation of LDLs contributes to the grainy microstructure of the yolk. Consolidating the evidence we create a time-temperature phase diagram that delivers a wealth of information about the physics of nanoscale processes occurring in an egg yolk during cooking. In a broader sense, our research provides an illustration of how to comprehend the fascinating non-equilibrium events in inherently complex, multi-component, thermally driven biological systems on length scales ranging from nanometers to micrometers in a time spectrum of milliseconds to hours.

- [1] M. Anton et al., J. Sci. Food Agric. 93, 2871-2880 (2013).
- [2] Y. Zhao et al., Food Chem. 355, 129569 (2021).
- [3] L. Gu et al., Food Sci. Biotechnol. 32, 121133 (2023).
- [4] F. Perakis and C. Gutt, Phys. Chem. Chem. Phys. 22, 19443-19453 (2020).

EFFECT OF SECONDARY INTERACTIONS ON THE DYNAMICS OF GLASS FORMING LIQUIDS: INSIGHTS FROM THE COMPARISON OF DIELECTRIC AND MECHANIC TECHNIQUES

S. Arrese-Igor¹, A. Alegria^{1,2}, J. Colmenero^{1,2,3}

¹Centro de Física de Materiales (MPC), Centro Mixto CSIC-UPV/EHU, Paseo Manuel de Lardizabal 5, 20018 San Sebastián, Spain.

²Departamento de Polímeros y Materiales Avanzados UPV/EHU, Apartado 1072, 20080 San Sebastián, Spain. ³Donostia International Physics Center, Paseo Manuel Lardizabal 4, 20018 San Sebastián, Spain.

E-mail: silvia.arreseigor@ehu.eus

The presence of different kind and degree of specific non-covalent interactions can potentially modify the dynamic response of liquids. Hydrogen bond interactions for example are known to produce specific effects, being plenty of works just dedicated to understand the family of monoalcohols only, not to mention the case of water. Lately, dipolar interactions too are attracting increasing interest due to singular characteristic found in the glassy dynamics of these systems (e.g. narrower lineshapes of their dielectric response). From an experimental point of view, the use of different techniques in a complementary way frequently reveals key details for knowledge, but often too the comparison is not straightforward and poses new questions.

In this work, we will overview some examples where the dynamics of glass forming systems were studied following the same procedure irrespective of their nature trying to obtain generic phenomenological relations that could later aid in answering system specific questions [1–4]. In particular, we have studied in detail the shear and dielectric relaxation characteristics exploring the relations between the different experimental quantities and representations generally used in the literature.

- [1] S. Arrese-Igor et al., Phys. Chem. Chem. Phys. 20.44, 27758-27765 (2018).
- [2] S. Arrese-Igor et al., J. Mol. Liq. 312, 113441 (2020).
- [3] S. Arrese-Igor et al., J. Mol. Liq. 318, 114215 (2020).
- [4] S. Arrese-Igor *et al.*, arXiv:2210.01947.

UNIQUE PHASE TRANSFORMATIONS IN GLASSY AQUEOUS SOLUTIONS CONTAINING IONIC LIQUIDS

J. Bachler¹, F. Caporaletti², M. Demmenie², S. Woutersen², I. Daidone³, L. Zanetti-Polzi⁴, T. Loerting¹

¹Institute of Physical Chemistry, University of Innsbruck, 6020 Innsbruck, Austria.

²Van't Hoff Institute for Molecular Sciences, University of Amsterdam, 1098XH Amsterdam, The Netherlands.

³Department of Physical and Chemical Sciences, University of L'Aquila, 67010 L'Aquila, Italy.

⁴Center S3, CNR-Institute of Nanoscience, 41125 Modena, Italy.

E-mail: Johannes.Bachler@uibk.ac.at

Many of the anomalous properties of water can be explained using a two-liquid model [1]. It is believed that water can be separated into a high-density liquid (HDL) and a low-density liquid (LDL) at sufficiently low temperatures. While both liquids are connected via a first-order liquid–liquid transition (LLT), this LLT is observed to be in a temperature region where ice crystallization is nearly instantaneous, thereby limiting its experimental observation [2].

Zhao and Angell took a different route by employing aqueous solutions containing ionic liquids instead of pure water [3]. They discovered that an aqueous mixture containing hydrazinium trifluoroacetate with a solute mole fraction $x \approx 0.16$ shows an unprecedented first-order transition around 190 K, which they believed to be the long-sought LLT. This claim was supported by Woutersen *et al.* employing IR spectroscopy experiments [4]. However, it was argued that the transformation cannot be related to a genuine LLT of water as the solutions do not exhibit water-like compression behavior [5]. More recently, MD simulations were used to show that a transition between low- and high-density states is only possible if a mixing–unmixing transition is far from being fully understood.

Here, we use a combination of different methods, IR spectroscopy, X-ray diffraction and calorimetry to shed light on the nature of the transition. We study the "Angell mix" itself and several other ionic liquid solutions, in which cations and anions are altered. These studies show a complex nature of the phase transition involving a glass transition that shifts with chemical composition.

- [1] P. Gallo et al., Eur. Phys. J. E. 44, 143 (2021).
- [2] P.H. Handle, T. Loerting and F. Sciortino, Proc. Natl. Acad. Sci. U.S.A. 114, 13336-13344 (2017).
- [3] Z. Zhao and C.A. Angell, Angew. Chem. Int. Ed. Engl. 55, 2474-2477 (2016).
- [4] S. Woutersen et al., Science 359, 1127-1131 (2018).
- [5] J. Bachler, L.-R. Fidler and T. Loerting, Phys. Rev. E 102, 060601(R) (2020).
- [6] L. Zanetti-Polzi, A. Amadei, I. Daidone, J. Chem. Phys. 155, 104502 (2021).

Transverse dynamics in fluids: how a collective excitation sets in

U. Bafile¹, D. Colognesi¹, M. Neumann², A. De Francesco^{3,4}, F. Formisano^{3,4}, A. Cunsolo⁵, W. Montfrooij⁶, E. Guarini⁷

¹Consiglio Nazionale delle Ricerche, Istituto di Fisica Applicata "Nello Carrara", via Madonna del Piano 10, I-50019 Sesto Fiorentino, Italy.

²Fakultät für Physik der Universität Wien, Kolingasse 14–16, A–1090 Wien, Austria.

³CNR-IOM & INSIDE@ILL c/o Operative Group in Grenoble (OGG) F-38042 Grenoble, France.

⁴Institut Laue Langevin (ILL), F–38042 Grenoble, France.

⁵Department of Physics, University of Wisconsin at Madison, 1150 University Avenue, Madison, WI, 53706, United States.

⁶Department of Physics and Astronomy, University of Missouri, Columbia, MO, 65211, United States.

⁷Dipartimento di Fisica e Astronomia, Università degli Studi di Firenze, via G. Sansone 1, I-50019 Sesto Fiorentino, Italy.

E-mail: u.bafile@ifac.cnr.it

The transverse-current autocorrelation function $C_T(Q, t)$, obtained by molecular dynamics simulation of a dense Lennard–Jones fluid, has been analyzed in the wavevector range between the hydrodynamic region and $Q_p/2$ (where Q_p is the position of the main peak in the static structure factor). The exponential expansion theory (EET) provides a full and accurate account of $C_T(Q, t)$, and reveals a varied dynamical behaviour. We identify and accurately locate the Qvalue at which a shear wave begins to propagate, and show how this phenomenon is described as the continuous transition of a harmonic oscillator from the overdamped to the underdamped state. Moreover, this analysis reveals the existence of a second pair of modes equivalent to another oscillator that undergoes, at higher Q values, a similarly smooth over– to underdamped transition [1].

[1] E. Guarini et al., Phys. Rev. E, (2023), in press.

Water on the mesoscale and progress towards IXS with 0.3 $\ensuremath{\mathsf{meSo}}$ resolution

A.Q.R. Baron¹, D. Ishikawa²

¹Materials Dynamics Laboratory, RIKEN SPring-8 Center, Sayo, Hyogo 679-5148, Japan.
²Precision Spectroscopy Division, CSRR, SPring-8/JASRI, Sayo, Hyogo, 679-5198, Japan.

E-mail: baron@spring8.or.jp

Higher resolution inelastic x-ray scattering (IXS) is critically needed to facilitate understanding of disordered materials, including liquids and glasses. The mesoscale region, say, momentum transfers of 0.1 to 3 nm^{-1} or correlation lengths of 60 to 2 Å is where one should, often, in principle, most clearly observe the onset of interesting physical processes, including line broadening, structural relaxation, and fast sound. Liquid water is just one example with dramatic changes in dynamical response, $S(Q, \omega)$, as the momentum transfer, Q, increases in this region: the sound velocity changes, linewidths increase, and the quasi-elastic intensity grows from a small fraction of the phonon intensity at small Q to dominate the spectra at higher Q. However, in practice, the endpoints at 0.1 and $\sim 1 \text{ nm}^{-1}$ may be observed but the intermediate region can not be probed by present methods. Here we will discuss using IXS [1-5] to enter this region. Extending IXS measurements to below 1 nm⁻¹ is very challenging, as, to be effective, both the energy and momentum transfer resolution must improve, reducing count-rates in what is already a count-rate limited technique. Thus, one is faced with the combined hurdles of needing high quality precisely controlled optics and extremely low countrates, requiring long-term stability and very low backgrounds. We are working to overcome these hurdles using the Si(15 15 15) back reflection at 29.7 keV. Resolution of 0.3 meV has been achieved, and optimization is in progress.

^[1] A.Q.R. Baron, arXiv:1504.01098.

^[2] D. Ishikawa et al., J. Synchrotron Radiat. 22, 3 (2015).

^[3] D. Ishikawa and A.Q.R. Baron, J. Synchrotron Radiat. 28, 804 (2021).

^[4] D. Ishikawa and A.Q.R. Baron, J. Phys. Soc. Japan 90, 83602 (2021).

^[5] A.Q.R. Baron and D. Ishikawa, submitted.

ACTIVATING SELF-ASSEMBLED PATCHY PARTICLE ARCHITECTURES

H. Jonas, P.G. Bolhuis

Van 't Hoff institute for Molecular Science, University of Amsterdam.

E-mail: p.g.bolhuis@uva.nl

Patchy particles have become a standard model in soft matter physics to investigate complex molecular behavior, from proteins to large colloidal systems. Synthetic colloidal particles with specific directional interactions can act as a playground to deeper understand their molecular counterparts (such as proteins and smaller molecules), but also open up avenues in the design of novel materials, and even mimic active, living matter. A particularly sensitive way of controlling the attraction between the patches is by critical Casimir interactions, which allows colloids to assemble into various superstructures, such as chains and networks. To understand and explore the behavior of these Casimir systems, we developed a quantitatively accurate potential model [1]. Using these optimized potentials in large–scale simulations we can predict the phase behavior of mixtures of patchy particles, understand the relaxation behavior of colloidal molecules, and explain the experimentally observed anomalous excess of monomers [2]. We can also take the system out of equilibrium, by including self–propelled active particles, causing ring and chains in the network to undergo breakage and rearrangement [3]. Such activated viscoelastic architectures can possibly act as model for understanding the behavior of living matter.

[1] H.J. Jonas et al., J. Chem. Phys 155, 034902 (2022).

[2] H.J. Jonas et al., J. Chem. Phys 157, 094903 (2022).

[3] H.J. Jonas et al., (2023) in preparation.

PROGRAMMABLE SOFT MATERIALS

A. McMullen¹, M. Basagoiti², Z. Zeravcic², J. Brujic¹

¹Center for Soft Matter Research, New York University. ²Gulliver Lab, ESPCI.

E-mail: jb2929@nyu.edu

The problem with self-assembling materials from a gas of particles is that the number of random combinations is so vast that achieving a unique structure requires all particles to be distinct, like in a jigsaw puzzle. This method is therefore expensive and demands the design and manufacture of building blocks of different shapes, sizes, and specific matching rules. To circumvent this problem, we borrow the idea of folding polypeptide chains into proteins from biology and we apply it to colloidal self-assembly. Our system of choice are chains of DNA-functionalized emulsion droplets, which have the advantage of freely rearranging even after binding along the folding pathways. Their diffusive mobility allows the system to avoid kinetic traps and reach a rigid state on the timescale of minutes. Theoretically enumerating all possible configurations allows us to program a hierarchical sequence of folding interactions such that a unique folded structure (i.e. foldamer) is achieved. While two droplet DNA flavors are sufficient to encode 2% of the available rigid geometries in 2D, adding an extra flavor improves folding success to ~50% in chains up to 14 droplets long. Our palette of foldamers opens material science to the hierarchical assembly of foldamers into metamaterials with exotic mechanical or optical properties.

This work was supported by the NSF grants No. NSF PHY17-48958 and No. NSF DMR-1710163.

Do structural relaxations in supercooled liquids have the generic line shape?

S. Capaccioli^{1,2}, K.L. Ngai²

¹Dipartimento di Fisica "E. Fermi", Università di Pisa, Largo Pontecorvo 3, 56127, Pisa, Italy. ²CNR-IPCF, Largo Bruno Pontecorvo 3, I-56127, Pisa, Italy.

E-mail: simone.capaccioli@unipi.it

Recent papers [1–2], relying on a collection of almost superimposing light scattering spectra of very different systems (e.g., H-bonding, van der Waals and ionic liquids), proposed a generic line shape of the structural relaxation, following $\propto \omega^{-1/2}$ at high frequencies. Based on this finding it was proposed that the frequency dispersion of the structural α -relaxation in supercooled liquids is generic, supported also by dielectric spectra of some liquids with small dipole moment. However, such evidence is not found in highly dipolar liquids, whose dielectric α -loss peak is much narrower than the generic line shape [3]. According to [1], this is due to the presence of a strong Debye-like contribution from cross-correlation between the dipole moments of different molecules to dielectric α -relaxation, masking the generic structural relaxation would have tremendous impact on theories and experiments of liquids and glass transition. In this paper we present results of crucial experimental data and simulations to demonstrate that the structural relaxation of liquids is far to be generic and that, instead, the distribution of relaxation times characterizing the relaxation provides rich information on glass-former dynamics.

[1] F. Pabst et al., J. Phys. Chem. Lett. 12, 3685 (2021).

[2] F. Pabst et al., Phys. Rev. E 102, 010606 (2020).

[3] K.L. Ngai et al., Sci. Rep. 11, 22142 (2021).

The Johari-Goldstein relaxation as probe of the mosaic structure of supercooled liquids

F. Caporaletti¹, S. Capaccioli², D. Bessas³, A. I. Chumakov³, F. Dallari⁴, A. Martinelli⁴, G. Monaco⁴

¹Laboratory of Polymer and Soft Matter Dynamics, Experimental Soft Matter and Thermal Physics (EST), Université

libre de Bruxelles (ULB), Brussels 1050, Belgium.

²Dipartimento di Fisica, Università di Pisa Pisa, Italy.

³ESRF-The European Synchrotron, Grenoble, France.

⁴Dipartimento di Fisica ed Astronomia, Università di Padova, Italy.

E-mail: federico.caporaletti@ulb.be

The dynamics of supercooled liquids is dominated by the structural relaxation, which becomes very slow on approaching the glass-transition temperature T_g , where its characteristic relaxation time becomes eventually of 100 s. At slightly elevated temperatures of $\sim 1.2T_g$, however, a second faster process, the Johari–Goldstein relaxation (β_{JG}) [1], decouples from the structural one and remains active even in the glass. The β_{JG} -process is known to be a genuine feature of the glass–transition and strongly coupled to the α -relaxation [2], though its dedicated role in the glass–transition remains under scrutiny, also for the lack of a firm microscopic theory of the glass–transition.

By employing X-ray scattering time-domain interferometry [3-6], a technique able to investigate microscopic density fluctuations in the 10ns-10us time-window, we have demonstrated that the molecules participating to the β_{JG} -relaxation are characterized, in the deep supercooled state, by two intriguing features [7]: (i) their mean-squared displacement satisfies the Lindemann criterion for structural instability and (ii) their number matches the threshold for site percolation. This suggests that the molecules participating to the β_{JG} -relaxation escape the cages formed by the nearest neighbors on a timescale faster than the α -relaxation and form a percolating cluster pervading the whole sample, in line with the basic idea of the random first theory of the glass-transition. Such result shows that the β_{JG} -relaxation can be used an effective probe for the mosaic structure of supercooled liquids, and by applying this approach to several samples we have recently found out that such percolating structure is rather insensitive to both interaction potential and temperature above T_g , and survives even in the glassy state [9].

- [1] G.P. Johari and M. Goldstein, J. Chem. Phys. 53, 2372 (1970).
- [2] K. Ngai, Relaxation and Diffusion in Complex Systems (Springer 2011).
- [3] A.Q.R. Baron et al., Phys. Rev. Lett. 79, 2823 (1997).
- [4] M. Saito et al., Phys. Rev. Lett. 109, 115705 (2012).
- [5] F. Caporaletti et al., Rev. Sci. Instrum. 88, 105114 (2017).
- [6] F. Caporaletti et al., Sci. Rep.9, 14319 (2019).
- [7] F. Caporaletti et al., Nat. Comm. 12, 1867 (2021).
- [8] J. D. Stevenson, et al., Nat. Phys. 2, 268-274 (2006).
- [9] F. Caporaletti et al.,, submitted.

STATIC AND DYNAMICAL PROPERTIES OF MODULATED HARD-SPHERES FLUID.

C. F. Petersen¹, K. Macmillan², **M. Caraglio**³, A. Ahmadirahmat³, V. Krakoviack⁴, S. U. Egelhaaf², T. Franosch³

¹Australian Institute for Bioengineering and Nanotechnology, The University of Queensland, Brisbane, Queensland 4072, Australia.

²Heinrich Heine University, Condensed Matter Physics Laboratory, Düsseldorf, Germany.

³Institut für Theoretische Physik, Universität Innsbruck, Technikerstraße 21A, A-6020, Innsbruck, Austria.

⁴Laboratoire de Chimie, UMR CNRS 5182, École Normale Supérieure de Lyon, 46 Allée d'Italie, 69364 Lyon Cedex 7, France.

E-mail: michele.caraglio@uibk.ac.at

The structure of a liquid can be manipulated with externally applied fields. This can be achieved experimentally with interfering lasers and has been implemented with colloidal particles [1,2]. This possibility also paves the way to a better understanding of confined liquids [3], which are prevalent in nature and necessary in many industrial applications. Considering a hard-sphere fluid with a modulated density profile, we investigate its static and dynamical properties by means of simulations and theory. In this system, we control three dimensionless parameters: the packing fraction, the amplitude of the potential relative to temperature, and its wavelength relative to particle diameter. Similar to a liquid confined in a slit, a non-monotonic evolution of the static structure factor peak and the pressure is observed upon variation of the potential wavelength. From a theoretical perspective, we approach the problem by using fundamental measure theory for the static and mode coupling theory for the dynamics.

- [1] C. Bechinger, M. Brunner, and P. Leiderer, Phys. Rev. Lett. 86, 930 (2001).
- [2] F. Evers et al., Phys. Rev. E 88, 022125 (2013).
- [3] S. Saw and C. Dasgupta, J. Chem. Phys. 145 054707 (2016).
From the vibrational properties of silicate glasses to the viscosity of their parental melts: a model

M. Cassetta^{1,2}, D. Di Genova³, F. Vetere⁴, M. Biesuz¹, N. Daldosso², G. Mariotto², M. Zanatta⁵

¹University of Trento, Dept. Of Industrial Engineering, Trento-Povo, Italy.

²University of Verona, Dept. Of Computer Sciences, Verona, Italy.

³Institute of Environmental Geology and Geoengineering (IGAG), Italian National Research Council (CNR), Roma,

Italy.

⁴University of Siena, Dept. of Physical Sciences, Earth and Environment, Siena, Italy. ⁵University of Trento, Dept. Of Physics, Trento-Povo, Italy.

E-mail: michele.cassetta@univr.it

The temperature and chemical dependence of the viscosity of melts is central in the numerical modeling of both volcanic dynamics and industrial glass production-design. The fast incursion of nanoscopic modifications (i.e., crystallization), affects drastically the measurements and our knowledge of viscosity results quite elusive. This increases the requirement for quantification of pure melt phase contribution to the measured viscosity of a system that probably undergoes crystallization. In this contribution, we present an alternative technique that enables careful estimation of viscosity and its temperature-dependence, starting from the vibrational dynamics (by Brillouin and Raman scattering) of the melt's parental glass. Our results show that the melt fragility is correlated to the ratio between bulk and shear moduli and the boson peak frequency position of glasses. However, a universal law addressing the why some of these quantities are related in the glassy state, is still missing. At this effort, we explored the extension of the observed trend by testing conventional binary and industrial systems, up to post-run experiments of volcanic multiphase systems. Concluding, our strategy allows the estimation of the melt viscosity as a function of temperature avoiding its direct measurement, ridding also the measurement from unwished crystallization.

CHARACTERIZATION OF INTERPOLYELECTROLYTE COMPLEXES

M. Chamchoum^{1,2}, M. Gradzielski², O. Czakkel¹, S. Prevost¹

¹Institut Laue-Langevin, 71 Avenue des Martyrs, 38000 Grenoble, France.

²Stranski-Laboratorium für Physikalische und Theoretische Chemie, Institut für Chemie, Technische Universität Berlin, Straße des 17. Juni 124, 10623 Berlin, Germany.

E-mail: chamchoum@ill.fr

Advances in polymer science allow to create evermore complex self-assembled structures, which are driven mostly by using electrostatic and hydrophobic forces, such as multicompartment interpolyelectrolyte complexes (MIPECs) [1–2]. These water-soluble colloids of 50–200 nm size combine different solubilisation properties, functionalities and variable mesoscopic structure that make them interesting e.g. in the field of drug delivery. A deep understanding of the morphology of such complexes and a predictive framework are still elusive, requiring further investigation.

In this talk, we are going to present an in-depth analysis of SANS data of oppositely charged IPECs, coupled with a computational description. The investigation has been performed at ANSTO facility (Bilby instrument [3]) and it has been focused on the capability of such complexes to form hydrophobic domains. The main parameter to be varied in our system is the hydrophobic modification of the polyacrylate. Using a coarse-grained description of the IPECs, the SANS pattern has been simulated from a collection of configurations sampled through molecular dynamics. The match between experimental data and the model confirms the latter, which suggests further insights on the dynamics' behavior and the stability of such structures.

The funding of the PhD project support "Structure and Dynamics of MIPECs of Dual Polarity" by the ILL and TU Berlin is gratefully acknowledged.

- [1] D.V. Pergushov, A.H.E. Müller & F.H. Schacher, Chem. Soc. Rev. 41, 6888 (2012).
- [2] S. Riemer et al., Polymer 70, 194-206 (2015).
- [3] A. Sokolova et al., J. Appl. Crystallogr. 52, 1-12 (2019).

Probing cage relaxation in concentrated protein solutions by XPCS

Y. Chushkin¹, A. Gulotta², F. Roosen-Runge^{2,3}, A. Pal², A. Stradner^{2,4}, P. Schurtenberger^{2,4}

¹ESRF, The European Synchrotron,71 Avenue des Martyrs, CS40220, 38043 Grenoble Cedex 9, France.

²Division of Physical Chemistry, Lund University, Naturvetarvägen 14, 22100 Lund, Sweden.

³Department of Biomedical Science and Biofilms Research Center for Biointerfaces (BRCB), Faculty of Health and Society, Malmö University, Sweden.

⁴Lund Institute of advanced Neutron and X-ray Science LINXS, Lund University, Lund, Sweden.

E-mail: chushkin@esrf.fr

X-ray Photon Correlation Spectroscopy (XPCS) is a well-established technique to study slow dynamics in disordered materials at nanometer down to angstrom length scales [1]. The method exploits the coherent fraction of the synchrotron radiation and benefits enormously from the recent upgrade of the ESRF source (EBS) [2]. The high degree of coherence opens new avenues for application of XPCS. One of the promising avenues is the study of dynamics in concentrated protein solutions at nearest neighbor distances. Diffusion of proteins on length scales of their own diameter in highly concentrated solutions is essential for understanding biological systems such as a living cell, but its experimental characterization remains a challenge. Our work addresses this problem and discusses the use of X-ray Photon Correlation Spectroscopy at a recently upgraded 4th generation sunchrotron source for this purpose. While X-ray radiation damage was generally believed to seriously threaten the application of XPCS to biological systems, we now present a dedicated experimental and analysis strategy [3] to overcome this obstacle. We report a successful test of this approach to highly concentrated solutions of the eye lens protein alpha crystallin [4], which has previously been established as a model protein exhibiting the classic behavior of hard sphere colloids under these conditions [5]. The thus obtained intrinsic relaxation times for so-called long-time cage diffusion indeed agree with macroscopic measurements of the relative zero shear viscosity [6]. Our experiments also reveal a complex dependence of the key structural and dynamic properties of the protein solutions on both the total absorbed radiation dose as well as the dose rate. We discuss possible mechanisms responsible for the observed radiation effects and their consequences for future applications of XPCS.

- [1] G. Grübel, A. Madsen, and A. Robert, in Soft Matter Characterization (Springer Netherlands, 2008) pp. 953–995.
- [2] P. Raimondi, Synchrotron Radiation News 29, 8 (2016).
- [3] Y. Chushkin, J. Synchr. Rad. 27, 1247 (2020).
- [4] Y. Chushkin et al., Phys. Rev. Lett., 129, 238001 (2002).
- [5] G. Foffi et al., Proc. Natl. Acad. Sci. U.S.A. 111, 16748 (2014).
- [6] S. Bucciarelli et al., Sci. Adv. 2, e1601432 (2016).

Excitation populations provide a thermodynamic order parameter for Liquids

M.T. Cicerone¹, J. Zahn¹, J.P. Stoppelman¹, K. Badilla-Nunez², J.G. McDaniel¹

¹Department of Chemistry and Biochemistry, Georgia Institute of Technology, 950 Atlantic Drive, Atlanta, GA 30332, USA.

²School of Chemical and Biochemical Engineering, Georgia Institute of Technology, 900 Atlantic Drive, Atlanta, GA 30332, USA.

E-mail: cicerone@gatech.edu

We have shown previously that parameters describing elastic deformation of inherent states (IS) and interbasin (IB) transitions on a potential energy landscape (PEL) can be extracted quantitatively from the picosecond timescale β_{fast} relaxation in liquids using neutron scattering [1-2] or nonlinear optical methods [3], and that these can be used to predict liquid dynamics over 14 orders of magnitude [1]. Here [4], we also obtain excitation populations from β_{fast} profiles. There is mounting evidence that local structural excitations play an important role in the dynamics of liquids and glasses. However, to our knowledge, they have not previously been measured in real liquids. Having excitation populations over a wide temperature range in several liquids, we show that their temperature-dependent population can be predicted from entropy and enthalpy of melting. We further show that the excitation population in the first solvent shell serves as an order parameter for the appearance of dynamic heterogeneity and for driving transformations between distinct mechanistic regimes of liquid relaxation, including the Arrhenius to super-Arrhenius and fragile to strong transitions. We propose a scenario that provides simple physical explanations for these previously enigmatic aspects of liquid behavior and support our hypotheses with simulations.

[1] M. Cicerone and Q. Zhong, Phys. Rev. Lett. 113, 117801 (2014).

[2] M. Cicerone and M. Tyagi, J. Chem. Phys. 146, 054502 (2017).

[3] J. Bender, M. Zhi and M. Cicerone, Soft Matter. 16, 5588 (2020).

[4] M. Cicerone et al., arXiv:2201.12593v3.

Microscopic collective dynamics in liquid neon-deuterium mixtures: neutron scattering and quantum simulations

D. Colognesi¹, U. Bafile¹, E. Guarini², and M. Neumann³

¹Consiglio Nazionale delle Ricerche, Istituto di Fisica Applicata "Nello Carrara", via Madonna del Piano 10, I-50019 Sesto Fiorentino, Italy.

²Dipartimento di Fisica e Astronomia, Università degli Studi di Firenze, via G. Sansone 1, I-50019 Sesto Fiorentino, Italu.

³Fakultät für Physik der Universität Wien, Kolingasse 14–16, A–1090 Wien, Austria.

E-mail: d.colognesi@ifac.cnr.it

In this talk a combined neutron scattering and quantum simulation study of the collective dynamics in liquid Ne–D₂ mixtures, at a temperature of T=30 K and in the wave-vector transfer range 4 nm⁻¹ < q < 51 nm⁻¹, is presented. Two D₂ concentrations have been investigated, one close to 25% molar fraction and the other close to 50% molar fraction, together with pure Ne. The dynamic structure factor for the centers of mass of the two molecular species has been extracted from the neutron scattering data and subsequently compared with that obtained from three different quantum simulation methods, such as ring polymer molecular dynamics and two slightly different versions of the Feynman–Kleinert approach. A general agreement was found, even though some discrepancies both among simulations, and between simulations and experimental data, could be observed. In order to clarify the physical meaning of the present spectroscopic results, an analysis of the longitudinal current spectral maxima has been carried out showing the peculiarities of the D₂ center–of–mass dynamics in these mixtures. A comparison with the centroid molecular dynamics results obtained for the D₂ center–of–mass self–dynamics in the same liquid mixtures is finally proposed [1].

[1] D. Colognesi et al., Phys. Rev. E 105, 054603 (2022).

NANODNA SEQUENCES IN AQUEOUS SOLUTIONS: STRUCTURAL AND THERMODYNAMIC RESULTS IN NATIVE AND DRUG-COMPLEXED SAMPLES

V. Libera^{1,2}, F. Ripanti¹, F. Bianchi¹, R. Biehl³, A. Martel⁴, B. Rossi⁵, F. D'Amico⁵, C. Masciovecchio⁵, G. Schirò⁶, C. Petrillo¹, A. Paciaroni¹, L. Comez²

¹Dipartimento di Fisica e Geologia, Università di Perugia, 06123.
 ²CNR-IOM c/o Dipartimento di Fisica e Geologia, Università di Perugia, 06123 Perugia, Italy.
 ³JCNS & ICS, Forschungszentrum Julich GmbH, Leo-Brandt Strasse, 52425 Julich, Germany.
 ⁴Institut Laue-Langevin-71 avenue des Martyrs, 38042 GRENOBLE Cedex 9, France.
 ⁵Elettra Sincrotrone Trieste, S.S. 14 Km 163.5, 34012 Trieste, Italy.
 ⁶Univ. Grenoble Alpes, CNRS, CEA, IBS, F-38000 Grenoble, France.

E-mail: comez@iom.cnr.it

Nucleic acid sequences rich in quanines can organize into noncanonical DNA G-quadruplexes (G4s) of variable size. The high degree of polymorphism together with conformational switching properties enables the structural reconfiguration of G4s and promotes their applications in sensing, analytical biochemistry, and logic gate development. In this context, the design of small molecules stabilizing the structure of G4s is a rapidly growing area for the development of novel anticancer therapeutic strategies and bottom-up nanotechnologies. Here, we adopted an integrated experimental approach to study the structural and thermodynamic properties of the G4 formed by a human telomeric 22-mer sequence (Tel22) in the presence of model drugs, upon thermal unfolding or irradiation with visible light [1–3]. Singular value decomposition analysis applied to circular dichroism and resonant Raman scattering spectra allowed us to identify a temperature region populated with intermediate conformers along the path from the native to the unfolded state, for both the quadruplex alone and the complexes. We found evidence for complex unfolded states with persistent residual stacking. Small angle scattering experiments have provided a valuable contribution to gather information at the guaternary structure level. Overall, our results suggest that, even under external stimuli, the complex is prevented to adopt a self-avoiding random-coil conformation due to the interaction with examined drugs, that possibly stabilizes a structure where the oligonucleotide is partially folded.

[1] F. Bianchi et al., Nucleic Acids Res. 46, 11927-11938 (2018).

- [2] V. Libera et al., J. Phys. Chem. Lett. 12, 8096-8102 (2021).
- [3] V Libera et al., Int. J. Mol. Sci. 23, 5123, (2022).

INTRA-PROTEIN INTERACTING COLLECTIVE MODES IN THE TERAHERTZ FREQUENCY REGION

V.C. Nibali¹, F. Sacchetti², A. Paciaroni², C. Petrillo², M. Tarek³, G. D'Angelo¹

¹Department of Mathematical and Computational Sciences, Physical Science and Earth Science, Messina University, Viale Ferdinando Stagno D'Alcontres 31, 98166, Messina, Italy.
²Department of Physics and Geology, Perugia University, Via Alessandro Pascoli, I-06123 Perugia, Italy.

²Université de Lorraine, CNRS, LPCT, F–54000 Nancy, France.

E-mail: vcontinibali@unime.it

We investigate herein the intra-protein collective dynamics of a model globular protein by analysing – in terms of an interacting phonon model – its longitudinal and transverse modes, calculated from molecular dynamics simulations. We reveal a complex low-frequency vibrational landscape which consists of two acoustic-like and two optic-like modes with a mixed symmetry character. The dispersion curves display two avoided crossing events that we relate to the sidechains and the hydrogen-bonded networks dynamics, a behaviour likely to be encountered for all globular and membrane proteins. The present insights provide a molecular-level perspective for understanding the pathways for energy redistribution in the interior of proteins.

Fluctuation-dissipation relation and effective temperatures in out of equilibrium systems

F. Corberi

¹Dipartimento di Fisica "E. R. Caianiello" and INFN, Gruppo Collegato di Salerno, via Giovanni Paolo II 132, 84084 Fisciano (SA), Italy.

E-mail: fcorberi@unisa.it

I will review the topic of the fluctuation-dissipation relation and the related concept of effective temperature is systems out of equilibrium and their relation with the structure of the equilibrium state. In particular, after discussing the longly studied problem of the fluctuation-dissipation relation in aging systems, like magnetic systems or spin-glasses after a thermal quench, I will present some new results regarding non-equilibrium stationary states of systems in contact with a multibath, namely two or more thermal reservoirs with different temperatures and timescales.

Chemical vitrification and universal scaling between cooperative motion, caging dynamics, and emergent elasticity

S. Corezzi¹, C. De Michele², S. Capaccioli³, A. Orecchini¹, L. Comez⁴

¹Dipartimento di Fisica e Geologia, Università di Perugia, I-06123 Perugia, Italy.
²Dipartimento di Fisica, Sapienza Università di Roma, I-00185 Roma, Italy.
³Dipartimento di Fisica, Università di Pisa, I-56127 Pisa, Italy.
⁴CNR-IOM, Dipartimento di Fisica e Geologia, Università di Perugia, I-06123 Perugia, Italy.

E-mail: silvia.corezzi@unipg.it

The basic ideas of cooperative motion, caging dynamics, and emergent elasticity intertwine within different theories of glass formation, intuitively suggesting relations between them: building on the idea that particles become increasingly trapped in transient cages formed by their nearest neighbours, inside which they rattle and from which escape via cooperative motions, a connection is envisaged between the average escape time from the cage, i.e. the relaxation time τ_{i} and the mean square amplitude of the rattling motion, $\langle u^{2} \rangle$. From a separate perspective, breaking the cage may be related to the capacity of the material to resist shear deformation, implying also a connection between $\langle u^2 \rangle$ and the elastic modulus G. Establishing quantitative relations, however, and assessing the extent to which they describe universal glass-forming behaviour has proved controversial [1]. The problem so far has only been addressed in glasses produced by cooling, and, notably, never assessed by using chemical vitrification, a process where glasses are formed via particle bonding [2]. Here, we use the polymerization of epoxy resins as a prototypical chemical vitrification process, and combining coarse-grained simulations with dielectric and QENS experiments we find the same nonlinear scaling relation between log τ and $1/\langle u^2 \rangle$ as in physical vitrification [3]. Moreover, Brillouin spectroscopy is used to monitor material elasticity. Surprisingly, for the first time, we find that $1/\langle u^2 \rangle$ is directly related to the short-time, not infinite-frequency, emergent shear modulus, implying that the same scaling between cooperative motion and caging dynamics applies to a well-defined measure of elasticity [4]. Overall, chemical vitrification helps uncover quantitative relations and to assess universality of glass formation.

- [1] B.A.P. Betancourt et al., Proc. Natl. Acad. Sci. U.S.A. 112, 2966 (2015)
- [2] S. Corezzi, D. Fioretto, and P. A. Rolla, Nature 420, 653 (2002)
- [3] L. Larini, A. Ottochian, C. De Michele, and D. Leporini, Nat. Phys. 4, 42 (2008)
- [4] C. De Michele, S. Capaccioli A. Orecchini, L. Comez, and S. Corezzi, Proc. Natl. Acad. Sci. U.S.A., submitted

PRESSURE INDUCED FAST ATOMIC MOTION IN METALLIC GLASSES AT EXTREME CONDITIONS

A. Cornet^{1,2}, A. Ronca^{1,2}, J. Shen^{1,2}, T. Deschamps¹, Y. Chushkin², F. Zontone¹, G. Garbarino², M. Mezouar², B. Ruta^{1,2}

¹Institut lumière matière (iLM), UMR5306 Université Lyon 1-CNRS, Université de Lyon 69622 Villeurbanne, France. ²ESRF – The European Synchrotron, 71 avenue des Martyrs, 38000 Grenoble, France.

E-mail: antoine.cornet@esrf.fr

A fascinating feature of metallic glasses is their ability to explore different configurations under mechanical deformations. This effect is usually observed through macroscopic observables, while little is known on the consequence of the deformation at atomic level. Using the new generation of synchrotrons, we probed the atomic motion and structure in a metallic glass under hydrostatic compression, from the onset of the perturbation up to a severely-compressed state. While the structure indicates reversible densification under compression, the dynamic is dramatically accelerated and exhibits a hysteresis with two regimes. At low pressures, the atomic motion is heterogeneous with avalanche-like rearrangements suggesting rejuvenation, while under further compression, aging leads to a super-diffusive dynamics triggered by internal stresses inherent to the glass. These results highlight the complexity of the atomic motion in non-ergodic systems and support a theory recently developed to describe the surprising rejuvenation and strain hardening of metallic glasses under compression.

A. Cornet et al. arXiv:2301.02551.

LONG-RANGE CONCENTRATION FLUCTUATIONS AND THEIR EFFECT ON THE THERMODYNAMICS OF MODEL SUPERCOOLED LIQUIDS

M. Sevilla, A. Banerjee, J. Rudzinski, R. Cortes-Huerto

¹Max Planck Institute for Polymer Research, Ackermannweg 10, 55128 Mainz, Germany.

E-mail: corteshu@mpip-mainz.mpg.de

The supercooled state challenges our understanding of the theory of liquids. The connection between dynamics, which varies considerably upon supercooling, and structure, which appears to remain essentially unchanged, is the subject of intense research. Model systems with reduced complexity provide a direct route to investigate this problem. Kob–Andersen mixtures with purely repulsive Weeks-Chandler-Andersen (KAWCA) interactions exhibit substantially different dynamics than their Lennard-Jones (KALJ) counterpart. By contrast, their radial distribution functions (RDF) are relatively similar.

In this talk, we present partial structure factors, Kirkwood–Buff integrals (KBI) [1] and chemical potentials [2] of KAWCA and KALJ supercooled liquids [3]. We show that minor differences in the tail of the RDF result in contrasting thermodynamic properties. Indeed, upon decreasing temperature, the structure factors of the KALJ system display anomalous behaviour in the low–k limit. KBI in the thermodynamic limit confirm this picture, identifying the anomaly with long–range concentration fluctuations. By contrast, the KAWCA system remains miscible and only reveals qualitatively similar concentration fluctuations in the crystalline state. Also, differences in isothermal compressibilities and chemical potentials show that thermodynamics is not entirely governed by the short–range repulsive part of the interaction potential, emphasising the nonperturbative role of attractive interactions.

- [1] R. Cortes-Huerto, K. Kremer, and R. Potestio, J. Phys. Chem. 145, 141103 (2016).
- [2] M. Heidari et al., J. Chem. Theor. Comput. 14, 3409 (2018).
- [3] A. Banerjee et al., Soft Matt. 18, 2373 (2022).

QUANTIFYING THE DEVIATION FROM THERMAL EQUILIBRIUM

S. Saw, L. Costigliola, J.C. Dyre

¹Glass and Time, IMFUFA, Department of Science and Environment, Roskilde University, Roskilde, Denmark.

E-mail: lorenzoc@ruc.dk

This talk will investigate the possibility of using the configurational temperature T_{conf} for quantifying how far a given out-of-equilibrium system is from thermal equilibrium. We measure this "distance" by the ratio of the systemic temperature T_s to T_{conf} , where T_s is the canonicalensemble temperature leading to the same average potential energy as that of the out-ofequilibrium system. T_{conf} is determined by the local variation of the potential-energy function, while T_s is a global quantity. The quantity T_s/T_{conf} is easy to evaluate in a computer simulation; equilibrium simulations in conjunction with a single steady-state active-matter configuration are enough to determine T_s/T_{conf} . We validate the suggestion that T_s/T_{conf} quantifies the deviation from equilibrium by computer simulations of the 3d Kob-Andersen and 2d Yukawa active-matter models with active Ornstein-Uhlenbeck and active Brownian Particle dynamics. The ratio T_s/T_{conf} , structure, and dynamics of the homogeneous phase, are all approximately constant along the motility-induced phase separation (MIPS) boundary in the phase diagram of the 2d Yukawa model. This confirms that T_s/T_{conf} measures the degree of deviation from thermal equilibrium, which is expected to be constant along the MIPS phase boundary. The proposed measure is not limited to active matter and can be used for quantifying how far any system involving a potential-energy function, e.g., a driven Hamiltonian system, is from thermal equilibrium. The results presented in this talk are published in Ref. [1].

[1] S. Saw, L. Costigliola, and J.C. Dyre, arXiv:2212.09041.

RECENT RESULTS ON INTRINSIC DROPLET DYNAMICS IN EMULSIONS FROM EXPERIMENTS IN MICROGRAVITY ONBOARD THE INTERNATIONAL SPACE STATION

L. Cristofolini¹, D. Orsi¹, V. Lorusso¹, D. Cassi¹, M. Vaccari¹, L. Liggieri², F. Ravera², E. Santini², G.

Loglio²

¹SMFI Università di Parma. ²CNR-ICMATE, Genova.

E-mail: luigi.cristofolini@unipr.it

Microgravity experiments on emulsions can unveil important information on drop-drop interactions and destabilization mechanisms in marginally stable or unstable emulsions. Emulsions are central to industrial processes in diverse fields including food, pharma, cosmetics, detergency, oil and automotive industries. Knowledge of emulsions is mostly empirical, and many processes are based on recipes making use of excess surfactants to stabilize emulsions, often with negative impact for the environment. The purpose of our research is the advancement of basic knowledge on emulsions, with positive effects in the development of more environmentally friendly processes and products. The destabilization of emulsions is inextricably linked to the droplet dynamics, their aggregation and coalescence. Under conditions of normal gravity, the study of these processes is complicated by the concomitant sedimentation or buoyancy. Experiments in microgravity, eliminating these complications, offer the unique opportunity to study droplet dynamics driven solely by intrinsic interactions, such as Brownian diffusion, and the DLVO droplet interactions. The relevant properties are obtained from Diffusing Wave Spectroscopy (DWS) [1,2], exploiting the Soft Matter Dynamics (SMD) [3] facility, developed by Airbus for ESA. The project involves academic and industrial partners from Italy, Greece, Germany, France, USA, Russia and Japan. We will discuss the dynamic transitions describing the motion of droplets, which is governed in the steady state by diffusion, but with important transient regimes characterized by faster and superdiffusive dynamics, presumably linked to Marangoni forces due to surfactant release as a consequence of drop-drop coalescence events. The interpretation of this phenomenology, the magnitude and frequency of events as a function of the emulsion composition (e.g. the volumetric ratio of immiscible liquids, the nature and quantity of surfactant) will allow us to advance knowledge relating to the destabilization mechanisms of emulsions.

[1] D.A. Weitz and J.D. Pine, *Diffusing-wave spectroscopy*, in *Dynamic Light Scattering: The Method and Some Applications*, edited by W. Brown (Clarendon Press, 1993).

- [2] V. Lorusso et al., Adv. Colloid Interface Sci. 288, 10234 (2021).
- [3] D. Orsi et al., Colloids Surf. A Physicochem. Eng. Asp. 580, 123574 (2019).
- [4] F. Salerni et al., Colloids Surf. A Physicochem. Eng. Asp. 580, 123724 (2019).
- [5] P. Born et al., Rev. Sci. Instrum. 92, 124503 (2021).

$\label{eq:pressure-Driven} \begin{array}{l} \text{Pressure-Driven Disorder in Glassy } As_2Se_3\text{: structure, low-energy} \\ \text{phonon dynamics and optical band gap} \end{array}$

G. D'Angelo¹, M. Federico¹, V. Romano², G. Carini¹

¹Department of Mathematical and Computational Sciences, Physical Science and Earth Science,Messina University, Viale Ferdinando Stagno D'Alcontres 31, 98166, Messina, Italy. ²Department of Physics, Politecnico di Milano, Piazza Leonardo da Vinci 32, 20133 Milano, Italy.

E-mail: gdangelo@unime.it

In this talk the vibrational dynamics and the structure of As_2Se_3 glasses compacted under growing GPa pressures and temperatures are presented and compared with the polycrystalline counterpart. Raman measurements evidence changes in the short and medium range order. The results of the pressure dependence of internal vibrational bands and of the boson peak are analyzed. In particular, the role of the structural interconnection of ring– and cage–like As_nSe_m nanoclusters in determining the low–frequency Raman spectra and low temperature heat capacity is discussed. Furthermore, interrelation of structure, low energy vibrational dynamics and optical band gap in the investigated samples is also addressed, in view of the possibility of tuning the band gap of chalcogenide glasses by high temperature treatment at high pressure for specific photovoltaic and optoelectronic applications.

Stochastic atomic acceleration during the X-ray-induced fluidization of a silica glass

F. Dallari^{1,2}, A. Martinelli¹, F. Caporaletti³, G. Baldi⁴, M. Sprung², G. Monaco¹

¹Dipartimento di Fisica e Astronomia "Galileo Galilei", Università degli Studi di Padova, Via F. Marzolo, 8, 35131 Padova, Italy.

²Deutsches Elektronen-Synchrotron DESY, Notkestraße 85, 22607 Hamburg, Germany.

³Van der Waals-Zeeman Institute, Institute of Physics/Van't Hoff Institute for Molecular Sciences, University of Amsterdam, Amsterdam, the Netherlands.

⁴University of Trento, Department of Physics, via Sommarive 14, 38123 Povo (TN), Italy.

E-mail: francesco.dallari@unipd.it

Upon X-ray irradiation, a number of glasses undergo a fluidization process: The atoms move from their original positions while keeping a similar distribution of interatomic distances, as for two snapshots of a liquid. In the past years, this process has been studied looking at the atomic displacements over interatomic distances. We here extend these investigations to much longer length scales in the few nanometer range. Our experiments clarify that this process is not characterized by the common atomic diffusion typical of liquids: It is rather the outcome of an atomic acceleration due to X-ray-induced local stresses random in both time and space, as in the famous example of stochastic acceleration of charged particles by interstellar fields. The X-ray-induced, nonthermal fluidization of the prototypical SiO₂ glass is investigated by X-ray photon correlation spectroscopy in the small-angle scattering range. This process is initiated by the absorption of X-rays and leads to overall atomic displacements which reach at least few nanometers at temperatures well below the glass transition. At absorbed doses of \sim 5 GGy typical of many modern X-ray-based experiments, the atomic displacements display a hyperdiffusive behavior and are distributed according to a heavy-tailed, Lévy stable distribution. This is attributed to the stochastic generation of X-ray-induced point defects which give rise to a dynamically fluctuating potential landscape, thus providing a microscopic picture of the fluidization process.

Untangling the role of tau in neurodegeneration and neurodevelopment using 3D human cortical organoids

S. Di Angelantonio^{1,2,3}, F. Cordella^{1,2}, L. Mautone^{1,2}, D. Salerno^{1,2}, E. Parente^{1,2}, L. Tondo^{1,2}

¹Center for Life Nano- and Neuro-Science, Istituto Italiano di Tecnologia, Roma, Italy.
²Department of Physiology and Pharmacology, Sapienza Università di Roma, Italy.
³D-Tails s.r.l., Roma, Italy.

E-mail: silvia.diangelantonio@uniroma1.it

Tau is a protein that plays a crucial role in the structure and function of neurons. Tauopathies, such as Alzheimer's Disease and Frontotemporal dementia, are characterized by the presence of tau protein aggregates called neurofibrillary tangles. A specific mutation in the tau protein, called the MAPT 10+16 mutation, is known to cause Frontotemporal dementia by increasing the expression of a form of tau protein called 4R tau, which is more prone to forming tangles than the normal form of tau. We investigated the impact of the IVS10+16 tau mutation on cortical development and maturation using 3D patterned human cortical organoids. Results showed that the tau mutation had a significant effect on neuronal, astrocytic and synaptic maturation, resulting in downregulation of specific synaptic and axonal genes and alteration in cytoskeleton structure. Additionally, functional analysis of the organoids revealed that the mutation impaired neuronal activity and reduced the number of active neurons. These findings suggest that inherited tau mutations have a major impact on neurodevelopment and have common features with both neurodevelopmental and neurodegenerative disorders.

A COMPUTATIONAL STRATEGY TO MODULATE THE COMPATIBILITY OF BIOMOLECULAR INTERFACES: THE IMPORTANCE IN PROTEIN DESIGN

L. Di Rienzo¹, M. Miotto¹, E. Milanetti^{2,1}, G.Ruocco^{1,2}

¹Center for Life Nano-& Neuro-Science, Istituto Italiano di Tecnologia, Viale Regina Elena 291, 00161 Rome, Italy.
²Department of Physics, Sapienza University of Rome, Piazzale Aldo Moro 5, 00185 Rome, Italy.

E-mail: lorenzo.dirienzo@iit.it

The complex network of interactions between biomolecules is responsible for the physiological regulation of almost all cellular functions. The molecular recognition between partners relies on the fine-tuning of the physicochemical properties of the amino acids involved on both sides of the interface. Indeed, the disruption of a molecular association can be caused even by a single residue mutation, often leading to a pathological modification of a biochemical pathway. Therefore, the evaluation of the effects of amino acid substitutions on binding, and the ad hoc design of molecular interfaces, is an important challenge in computational structural biology. Modeling the shape of the interaction surface properties using the orthonormal basis of the Zernike polynomials, we describe the geometry of binding sites with a compact ordered set of rotationally invariant descriptors, making the evaluation of complementarity between interacting surfaces possible. Including in the description a compact model of chemical complementarity, we present a novel strategy for computational mutagenesis and optimization of protein interfaces. Using a simulated annealing approach, we can obtain protein mutants with controlled molecular complementarities. The applications of this method to various biological systems of interest demonstrate its effectiveness in the identification of a small set of amino acid substitutions capable of modulating the stability of the corresponding molecular complexes.

DISTANCE-AS-TIME IN PHYSICAL AGING

J.C. Dyre, I.M. Douglass

Glass and Time, IMFUFA, Department of Science and Environment, Roskilde University, P.O. Box 260, DK-4000 Roskilde, Denmark.

E-mail: dyre@ruc.dk

It has been known for 50 years that the physical aging of glasses is described well by a linear thermal-history convolution integral over the "material time", but its microscopic definition remains a mystery. We propose that the material-time reflects the distance traveled by the system's particles [1]. Different possible distance measures are discussed, starting from the mean-square displacement and its inherent-state version. The viewpoint adopted, which is closely related to pioneering works of Cugliandolo and Kurchan from the 1990s, implies a "geometric reversibility" and a "unique-triangle property" characterizing the system's path in configuration space during aging. Both properties are inherited from equilibrium and here confirmed by computer simulations of an aging binary Lennard-Jones system. Our simulations demonstrate, moreover, that the material time is dominated by the slowest particles' inherent mean-square displacement, conveniently quantified by the inherent harmonic mean-square displacement. This distance measure collapses data for potential-energy aging well such that the normalized relaxation functions following temperature jumps is (almost) the same function of the material time. Finally, the standard linear material-time convolution-integral description of physical aging is derived assuming that when time is replaced by distance in the above sense, an aging system is described by the same expression as that of linear-response theory.

[1] I.M. Douglass and J.C. Dyre, Phys. Rev. E 106, 054615 (2022).

Structural relaxation and nano-domain dynamics in highly concentrated electrolytes for zinc anode batteries

J.A. Dura¹, A. Faraone¹, S. Takeuchi^{1,2}, F. Wang³

¹National Institute of Standards and Technology, Center for Neutron Research, Gaithersburg, MD 20899–6100, USA.
²Materials Science and Engineering, A. James Clark School of Engineering, University of Maryland, College Park, MD 20742, USA.

³Department of Materials Science, Fudan University, Shanghai 200438, China.

E-mail: afaraone@nist.gov

Aqueous batteries are regarded as desirable alternatives to lithium-ion batteries using nonaqueous electrolytes due to their intrinsic non-flammable nature. Metallic zinc has long been regarded as an ideal anode material for the aqueous batteries systems. Highly Concentrated Zinc Electrolytes (HCZEs) are a new type of Water in BiSalt Electrolytes (WIBSE) which might allow the practical use of zinc anode batteries, improving their cycle life and energy density [1]. HCZEs contain both a zinc salt and Li(TFSI) [TFSI being bis(trifluoromethanesulfonyl)imide], this latter at very high concentrations. Ultimately, the effectiveness of HCZEs in improving the characteristics of zinc batteries is believed to be due to the curbing of zinc hydroxide formation, through the depletion of water in the zinc ion solvation shell, the segregation of water molecules in the solvation shell of the lithium ions, the suppression of the water activity [2], and the disruption of its H-bond network. Using QuasiElastic Neutron Scattering (QENS), we investigated the structural relaxation of water and the nanodomain dynamics in HCZEs, at the molecular scale. The relation between the measured dynamics and the macroscopic transport properties, viscosity and conductivity, will be discussed.

[1] F. Wang et al., Nat. Mater. 17, 543 (2018).

[2] F. Wang et al., Adv. Energy Mater. 11, 2102016 (2021).

BRILLOUIN LIGHT SCATTERING MICROSPECTROSCOPY FOR BIOMEDICAL APPLICATIONS

D. Fioretto

Department of Physics and Geology, University of Perugia, 06123 Perugia, Italy.

E-mail: daniele.fioretto@unipg.it

Brillouin light scattering is an ancient technique for investigating condensed matter that has recently gained renewed interest as an optical elastography technique to provide a nondestructive, non-contact probe of the micromechanics of biological matter. Here we report some recent advances of Brillouin light scattering micro-spectroscopy for biomedical applications. Furthermore, we show how these studies can contribute to the understanding of long-standing problems in complex systems, such as the dynamics of hydration water and the nature of the glass transition.

Acoustic attenuation and dispersion in the vicinity of the boson peak

P.-J. Wanq¹, A. Huynh², B. Perrin², B. Rufflé³, R. Vacher³, C.-K. Sun¹, M. Foret³

¹ Department of Electrical Engeneering and Graduate Institute of Photonics and Optoelectronics, National Tawain University, Taipei 10617, Taiwan.

² Sorbonne Universités, CNRS-UPMC Univ. Paris 06, UMR 7588, Institut des Nanosciences de Paris, F-75005 Paris, France.

³ Laboratoire Charles Coulomb, Université de Montpellier, CNRS, F-34095, Montpellier, France.

E-mail: marie.foret@umontpellier.fr

Pump-probe optical methods made it possible to explore acoustic properties of glasses in the difficult but crucial frequency region reaching the THz frequency range. A universal property of glasses is that a large excess of modes exists, with a maximum density of states near 1 THz, forming the so-called boson peak. The acoustic modes are expected to be strongly affected as their frequency nears the boson peak. Here, we present an experimental study of the onset of this effect in v-SiO2 as a function of temperature. The anticipated transition, from an ω^2 to ω^4 law for the mean free path of the longitudinal acoustic modes is observed well below 1 THz, along with a negative velocity dispersion.

Reversing Peierls-like distortions elicits the polyamorphic transition in GeTe and GeSe

T. Fujita¹, Y. Chen², S. Takahashi³, H. Kasai³, M. Bernasconi⁴, Y. Kono⁵, E. Nishibori³, R. Mazzarello², S. Wei^{1,6}

¹Department of Chemistry, Aarhus University, 8000 Aarhus C, Denmark.

²Department of Physics, Sapienza University of Rome, Rome 00185, Italy.

³Department of Physics, Faculty of Pure and Applied Sciences and Tsukuba Research Center for Energy Materials Science (TREMS), Tsukuba, Ibaraki 305–8571, Japan.

⁴Dipartimento di Scienza dei Materiali, Università di Milano-Bicocca, I-20125 Milano, Italy.

⁵Geodynamics Research Center, Ehime University, Matsuyama 790–8577, Japan.

⁶iMAT Centre for Integrated Materials Research, 8000 Aarhus C, Denmark.

E-mail: tofu@chem.au.dk

While polymorphism is well known in crystalline states, the polyamorphic transition in amorphous states have not been well understood in various types of glasses. Recently, direct evidence of a liquid-liquid transition has been reported in the supercooled liquid of phasechange materials (PCMs), where temperature induces Peierls-like distortions (PLD) that play a key role in the transition [1–3]. On the other hand, influence of the other factors such as pressure on PLD is still not identified in the polyamorphic transitions. Here we investigated the structural changes of amorphous GeSe and GeTe under in-situ high pressure conditions by 100 keV X-ray diffraction experiment. The analysis of structural factors and pair distribution functions shows the clear evidence that pressure can reverse the PLD that elicits a polyamorphic transition. This is supported by our molecular dynamics simulations based on a neuron-network potential. A higher transition pressure in GeSe than that in GeTe can be rationalized in terms of PLD. The high-pressure state is characterized by vanishing PLD, a larger coherence length, and a lower compressibility. In addition, the reversed PLD implies a closing of the pseudo-bandgap, which may result in a semiconductor-to-metal transition. Our work highlights PLD as an essential feature of many amorphous octahedral systems including most PCMs, which might be controlled by pressure and chemical components for designing properties in PCM applications.

- [1] P. Zalden et al., Science 364, 1062-1067 (2019).
- [2] S. Wei, P. Lucas, and C. A. Angell, MRS Bull. 44, 691-698 (2019).
- [3] S. Wei, J. Non-Cryst. 14, 100094 (2022).

PROGRESS AND OPPORTUNITIES FOR XPCS AND XSVS AT LCLS

P.H. Fuoss¹, M. Chollet¹, H. Li¹, T. Sato¹, S. Song¹, Y. Sun¹, D. Zhu¹, M. Sutton²

¹LCLS, SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA. ²Physics Department, McGill University, Montrèal, Quebec, Canada H3A 2T8.

E-mail: fuoss@slac.stanford.edu

The coherent, femtosecond hard x-ray pulses from free electron laser x-ray sources provide unprecedented opportunities to observe the structure of disordered materials, for example liquids and glasses, down to the atomic length scale, as well as the dynamics of atomic motion in these systems down to the femtosecond time scale. This presentation will focus on recent progress at the Linac Coherent Light Source (LCLS) using x-ray photon correlation spectroscopy (XPCS) and x-ray speckle visibility spectroscopy (XSVS) to measure ultra-fast dynamics in disordered materials [1-4]. We will discuss the new instruments recently commissioned [5,6,7], the FEL multi-pulse modes that have been implemented [8], and the new detectors being developed [9]. Finally, we will highlight the experimental opportunities and challenges associated with the new capabilities enabled by the LCLS-II project including >20 keV photons from the new vertically-polarized hard x-ray undulator and the use of high repetition rate pulses from third harmonics with the LCLS-II superconducting linac.

This work is supported by the U.S. Department of Energy Office of Science under Contract No. DE-AC02-76SF00515.

- [1] M.H. Seaberg et al., Phys. Rev. Lett. 119, 067403 (2017).
- [2] W. Roseker et al., Nat. Comm. 9, 1704 (2018).
- [3] F. Perakis et al., Nat. Comm. 9, 1917 (2018).
- [4] Y. Sun et al., Phys. Rev. Lett. 127, 058001 (2021).
- [5] D. Zhu et al., Proc. SPIE, 10237,102370R (2018).
- [6] H. Li et al., Optics Letters, 45, 2086 (2020).
- [7] H. Li et al., Phys. Rev. Res. 3, 043050 (2021).
- [8] F. Decker et al., Proc. FEL2010, WEPB33, 467 (2010).
- [9] M. Sikorski et al., J. Synchrotron Rad. 23, 1171 (2016).

The dielectric and rheological responses of strongly asymmetric mixtures of glycerol/propanol

J.P. Gabriel, K. Niss, J.C. Dyre, T. Hecksher

Glass and Time, Roskilde University, 4000 Roskilde, Denmark.

E-mail: jgabriel@ruc.dk

Significant progress has been made in recent years regarding the understanding of pure hydrogen-bonded systems. Currently, monohydroxy- and polyalcohols are considered to form transient hydrogen-bonded supramolecular structures in the form of chains, rings, and networks. Consistent with this, complex dynamic behavior can be identified for network-forming glycerol [1] and chain-forming propanol [2] by combining dielectric and shear rheological spectra.

We transfer these concepts to study the combined dielectric and shear rheological spectral shape of glycerol/propanol mixtures. Although glycerol differs from propanol by having two additional hydroxy groups, the mixtures are strongly asymmetric ($\Delta T_{melting} = 291 \text{ K} - 147 \text{ K} = 143 \text{ K}$, $\Delta T_{glass} = 190 \text{ K} - 98 \text{ K} = 92 \text{ K}$). The strong asymmetry is manifested by the existence of two distinct glass transitions in calorimetric measurements at a glycerol concentration $\chi_{gly} = 0.3$. Between 0.7 and 0.5 molar glycerol concentration a change in the shear modulus G_{∞} . We interpret this as a percolation transition. We discuss our findings of extremely broad spectral shapes compared to neat liquids and assign molecular interpretations of the spectral features by performing comprehensive analyses of the three experimental techniques.

[1] J.P. Gabriel et al., Phys. Chem. Chem. Phys. 22, 11644-11651 (2020).

[2] J.P. Gabriel, F. Pabst, and T. Blochowicz, J. Phys. Chem. B 121, 8847-8853 (2017).

OPTIMALITY IN SELF-ORGANIZED MOLECULAR SORTING

M. Zamparo¹, D. Valdembri^{2,3}, G. Serini^{2,3}, I. Kolokolov^{4,5}, V. Lebedev^{4,5}, L. Dall'Asta^{6,7,9}, A. Gamba^{6,7,8}

¹Dipartimento di Fisica, Università degli Studi di Bari, 70126 Bari, Italy.

²Department of Oncology, University of Torino School of Medicine, 10060 Candiolo, Italy.

³Candiolo Cancer Institute IRCCS, Fondazione del Piemonte per l'Oncologia, 10060 Candiolo, Italy.

⁴L.D. Landau Institute for Theoretical Physics, 142432, Moscow Region, Chernogolovka, Russia.

⁵National Research University Higher School of Economics, 101000, Moscow, Russia.

⁶Department of Applied Science and Technology, Politecnico di Torino, 10129 Torino, Italy.

⁷Italian Institute for Genomic Medicine, Candiolo, 10060, Italy.

⁸Istituto Nazionale di Fisica Nucleare (INFN), Italy.

⁹Collegio Carlo Alberto, 10122, Torino, Italy.

E-mail: andrea.gamba@polito.it

One of the main mechanisms involved in the generation and maintenance of distinct intracellular compartments in eukaryotic cells is molecular sorting, where specific biomolecular cargos are concentrated and distilled into submicrometric lipid vesicles, that are later delivered to appropriate destinations. We propose a theoretical model of the process based on the coupling of phase separation and membrane bending. In the model, localized microdomains enriched in specific cargo and adaptor molecules form by phase separation on lipid membranes, grow by absorbing laterally diffusing molecules, and induce membrane bending and the nucleation of a lipid vesicle when they reach a characteristic size. Since the newly generated vesicle is constitutively enriched in the biochemical factors of the engulfed domain, this results in a natural distillation process. We study the efficiency of this process at the steady state in terms of a few phenomenological parameters: the aggregation strength and the number of molecules sorted per unit time. Theoretical analysis and numerical simulations show that the distillation process is most efficient in an intermediate, optimal range of the aggregation strength. In this optimal range, both the steady-state surface concentration of sorted molecules and their average sorting time are minimal, and obey simple scaling laws. Quantitative measures of endocytic sorting performed in primary endothelial cells are compatible with the hypothesis that such optimal conditions are realized in living cells, suggesting that optimal parameter ranges may have been evolutionary selected.

[1] M. Zamparo et al., Phys. Rev. Lett. 126, 088101 (2021).

[2] E. Floris et al., Phys. Rev. E 106, 044412 (2022).

TRANSITION PATH SAMPLING WITH QUANTUM COMPUTERS

D. Ghamari^{1,2}, P. Hauke^{1,4}, R. Covino³, P. Faccioli^{1,2}

¹Department of Physics, University of Trento, Via Sommarive 14, Trento, 38123, Italy. ²INFN-TIFPA, Via Sommarive 14, Trento, 38123, Italy. ³Frankfurt Institute for Advanced Studies, Ruth-Moufang-Straße 1, Frankfurt am Main, 60438, Germany.

⁴INO-CNR BEC Center, University of Trento, Via Sommarive 14, Trento, 38123, Italy.

E-mail: danial.ghamari@unitn.it

Complex biomolecular systems such as proteins can undergo large functional structural rearrangements. However, these transitions provide prototypical examples of rare events. Transition path sampling (TPS) is a rigorous Monte Carlo method that aims at capturing these rare and complex events by refocusing the computational resources of Molecular Dynamics (MD) onto the relative transitions. However, realistic applications of this method to complex biomolecular transitions is fundamentally limited by the difficulties in sampling previously unexplored regions of a large configuration space and by the very long autocorrelation time of the Monte Carlo Markov chain. In order to overcome both of these problems, we have devised a new TPS framework that integrates MD, machine learning and both classical and quantum computers. We report on a first successful application [1] performed using the DWave quantum computer, on a benchmark molecular system that demonstrates that our framework is able to improve on the challenges of conventional TPS. Furthermore, we show preliminary results on a realistic protein, which demonstrates the scalability of our framework to systems that cannot be investigated with plain MD nor by conventional TPS on a classical computer.

[1] D. Ghamari et al., Sci. Rep. 12, 16336 (2022).

EXPERIMENTAL EVIDENCE OF A CROSSOVER BETWEEN COOPERATIVE RELAXATION AND LIQUID GROWTH DYNAMICS

M. Gonzalez-Silveira, A. Vila-Costa, C. Rodríquez-Tinoco, M. Rodríguez-López, J. Rodríguez-Viejo

Departament de Física. Facultat de Ciències, Universitat Autònoma de Barcelona, 08193, Bellaterra, Spain. Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and BIST, Campus UAB, Bellaterra, 08193, Barcelona, Spain.

E-mail: marta.gonzalez@uab.cat

In stark contrast with the conventional understanding of the glass transition, where the transition from glass to liquid appears as a dynamic process with atoms/molecules cooperatively relaxing into the equilibrium phase, we experimentally show that the nature of the glass transition depends at a given temperature on the ratio between the relaxation time of the glass, τ_{glass} , taken as its transformation time, and the alpha relaxation time, τ_{α} . Although the relaxation of liquid-cooled glasses is not totally synchronous, due to the existence of a distribution of relaxation times, there has been no clear observation of phase separation. However, at temperatures at which $\tau_{glass}/\tau_{\alpha}$ is large, high mobility regions nucleate into the liquid phase that subsequently grow by dynamic facilitation before – or while – cooperative glass relaxation sets into play. On the contrary, at temperatures associated to smaller $\tau_{glass}/\tau_{\alpha}$ the glass transition proceeds by cooperative relaxation dynamics all-across the material. This behavior is independent of the experimental procedure or protocol to produce the glass.

Self-organization in neural rosette morphogenesis

G. Gosti^{1,2}, M. Miotto^{1,3}, M. Rosito^{1,4}, M. Paoluzzi⁵, V. de Turris¹, V. Folli^{1,6}, M. Leonetti^{1,2,6}, A. Rosa^{1,7}, G. Ruocco^{1,3}

¹Center for Life Nano & Neuro Science, Istituto Italiano di Tecnologia, Viale Regina Elena 291, 00161, Rome, Italy.
²Soft and Living Matter Laboratory, Institute of Nanotechnology, Consiglio Nazionale delle Ricerche, 00185, Rome, Italy.

³Department of Physics, Sapienza University of Rome, Piazzale Aldo Moro 5, 00185 Rome, Italy

⁴Department of Physiology and Pharmacology "V. Erspamer", Sapienza University of Rome, Piazzale Aldo Moro 5, 00185 Rome, Italu.

⁵Departament de Física de la Matèria Condensada, Universitat de Barcelona, C. Martí Franquès 1, 08028 Barcelona, Spain.

⁶D-TAILS srl, 00161, Rome, Italy.

⁷Department of Biology and Biotechnologies "Charles Darwin", Sapienza University of Rome, Piazzale Aldo Moro 5, 00185 Rome, Italy.

E-mail: giorgio.gosti@iit.it

Under certain conditions the process of differentiation of human pluripotent stem cells into neural progenitors is characterized by the formation of structures known as neural rosettes. Even if neural rosette formation is recognized as an example of self-organization, we still do not understand the fundamental mechanisms guiding the process. I show that rosette formation is the result of a series of phase transitions that can be modeled in a disordered system framework in which cells differentiate and self-organize in space. This process of self organization allows cell to form an emergent supracellular cytoarchitecture which characterizes the adult tissue. Ultimately, to understand rosette formation, we first need to fully understand the complex interplay between growth, migration, cytoarchitecture organization, and cell type evolution.

WHERE IS THE HYDRODYNAMIC LIMIT?

J.S. Hansen

"Glass and Time", IMFUFA, Dept. of Science and Environment, Roskilde University, Denmark.

E-mail: jschmidt@ruc.dk

Hydrodynamics is often referred to as a macroscopic theory [1] that breaks down on "small length scales". In this talk I will revise that claim [2] comparing detailed molecular dynamics simulation data and theory. First, it is relevant to discuss what we mean by a break down and and how this is manifested in the dynamics. This, in turn, leads to the need for a precise definition of hydrodynamics, namely, as a set of balance equations and constitutive models that can be extended and generalized giving raise to the important distinction between classical and extended hydrodynamics. The main point of the talk is that classical hydrodynamics based on simple local linear constitutive models can, in fact, predict at least the qualitative behavior of the fluid dynamics of systems like toluene and water on the nano-scale, and that the degree of validity depends on the actual dynamics we study. This is still often a surprising conclusion for the fluid dynamic community. Furthermore, by proposing generalized constitutive models [3], extended descriptions [4], and including relevant coupling phenomena [3] it will be shown that the validity can be improved considerably.

- [3] J.S. Hansen, Nanoscale Hydrodynamics of Simple Systems (Cambridge University Press, 2022).
- [4] T. Bryk et al., J.Chem.Phys. 133, 024502 (2010).

^[1] See for example, B. Lautrup, *Physics of Continuum Matter* (IoP Publishing, 2005).

^[2] This has been done by many others, of course, see e.g. C. Bardos et al., J. Stat. Phys. 63, 323 (1991).

Structural, dynamical and electronic heterogeneities in supercooled-liquid antimony from *Ab-Initio* simulations

N. Holle¹, S. Walfort¹, J. Ballmaier¹, R. Mazzarello², M. Salinga¹

¹University of Münster, Institute of Materials Physics, Wilhelm-Klemm-Str. 10, 48149 Münster, Germany. ²Sapienza Università di Roma, Department of Physics, Piazzale Aldo Moro 2, Roma, 00185, Italy.

E-mail: nils.holle@uni-muenster.de

Computer simulations of typical glass formers are often limited by relaxation times exceeding the time scale of the simulation by orders of magnitude. The fast dynamics in pure supercooled-liquid antimony, a highly fragile glass former, allow simulation studies even under deep supercooling and of the subsequent glass transition. Using *ab-initio* simulations of this very simple system, we show here how structural heterogeneities in terms of local Peierls-like distortions govern both dynamical and electronic heterogeneities in the supercooled liquid. These heterogeneities are observed at temperatures below a liquid-liquid transition from a high-temperature state without Peierls-like distortions to a distorted low-temperature state. Furthermore, relaxation in the rapidly quenched glassy state is observed on the time scale of the simulation, which facilitates *ab-initio* simulation studies of structural relaxation in a realistic material system. Our results provide a new picture of dynamics and electronic effects in the supercooled-liquid and glassy phase of a whole class of materials, commonly known as phase change materials [1]. While this insight is of direct relevance to applications in nanoelectronics and photonics, it can also advance our understanding of the connection between structure and dynamics in glass forming systems in general.

[1] M. Salinga et al., Nat. Mater. 17, 681-685 (2018).

Melting of 2D crystals of squares

P. Keim¹, L. Siedentop²

¹Max Planck Institute for Dynamics and Self-Organization, 37073 Göttingen. ²University of Konstanz, Department of Physics, 78464 Konstanz.

E-mail: Peter.Keim@ds.mpg.de

Melting in 2D is described by the celebrated Kosterlitz-Thouless-Halperin-Nelson-Young (KTHNY) theory [1–3]. The unbinding of two different types of topological defects destroys translational and orientational order at different temperatures. The intermediate phase is called hexatic and has been measured in 2D colloidal monolayers [4,5]: it is a fluid with six-folded quasi-long-ranged orientational order. The six-folded director field stems from the closed packed crystal structure in the low temperature phase, being hexagonal for isotropic particles.

Here, the melting of a quadratic, 4-folded crystal is investigated, consisting of squares of about $5 \times 5\mu m^2$. The anisotropic particles are manufactured from a photo-resist using a 3D nanoprinter (nanoscribe GT). In aqueous solution, particles sediment by gravity to a thin cover slide where they form a monolayer. The curvature of the cover slide can be adjusted from convex to concave, which allows to vary the area-density of the monolayer in the field of view. For low densities, the squares are free to diffuse and form a 2D fluid while for high densities they form a quadratic crystal. Using a four-folded bond-order correlation function, we resolve the tetratic phase with quasi long ranged orientational order but short ranged translational one.

- [1] M. Kosterlitz, D. Thouless, Jour. of Phys. C 5, L124 (1972).
- [2] B. Halperin D. Nelson, Phys. Rev. Lett. 41, 121 (1978).
- [3] P. Young, Phys. Rev. B 19, 1855 (1979).
- [4] P. Keim, G. Maret, and H.H. von Grünberg, Phys. Rev. E 75, 031402 (2007).
- [5] U. Gasser et al., Chem. Phys. Chem. 11, 963 (2010).

ATHERMAL BOND-BREAKING PATHWAYS IN GLASSY CHALCOGENIDE MATERIALS

K. Konstantinou¹, J. Akola^{1,2}, S.R. Elliott³

¹Computational Physics Laboratory, Tampere University, Tampere FI-33014, Finland.
²Department of Physics, Norwegian University of Science and Technology (NTNU), Trondheim NO-7491, Norway.
³Physical and Theoretical Chemistry Laboratory, University of Oxford, Oxford OX1 3QZ, UK.

E-mail: konstantinos.konstantinou@tuni.fi

Hybrid density-functional theory calculations are used to provide an accurate picture of the role played by carrier injection in the bond-breaking processes within native defective atomic environments in glassy Ge-Sb-Te chalcogenide materials. We demonstrate that the localization of one and/or two extra electrons and holes, that are spontaneously trapped at the intrinsic near-linear triatomic structural precursors from the axial bonds in "see-saw" 4-coordinated or 5-coordinated configurations, can result in bond stretching, and therefore potential breaking, of these long (soft) bonds in the defective octahedral-like sites, which also can lead to bond switching with a nearby atom in the vicinity of the host defect environment [1,2]. Moreover, we utilize the Berry-phase formalism to illustrate how these highly polarizable bonds in such geometries can break under the application of finite periodic electric fields, resulting in an annihilation, without thermal annealing, of mid-gap defect electronic states from the band gap of the glassy models [3]. The observations suggest that electron and hole trapping, as well as the application of a sufficiently high external electric field, induce atomic relaxations of the glassy structure which can reduce the energy barrier for athermal bond breaking within the amorphous network of the chalcogenide materials. In that way, we demonstrate a materialsengineering concept for tuning the atomic and electronic behaviour in glasses.

- [1] K. Konstantinou, S.R. Elliott, and J. Akola, J. Mater. Chem. C 10, 6744 (2022).
- [2] K. Konstantinou, and S.R. Elliott, Phys. Stat. Solidi RRL, accepted.
- [3] K. Konstantinou, F.-C. Mocanu, J. Akola, and S.R. Elliott, Acta Mater. 223, 117465 (2022).

Hump universality and singularity in low-temperature heat capacity of crystalline and amorphous solids: boson peak or van Hove signature

A.I. Krivchikov^{1,2}, A. Jeżowski¹, M.S. Barabashko²

¹Institute of Low Temperature and Structure Research, Polish Academy of Sciences, 50-422 Wrocław.
²Verkin Institute for Low Temperature Physics and Engineering of the National Academy of Sciences of Ukraine, Kharkiv 61103, Ukraine.

E-mail: krivchikov@ilt.kharkov.ua

One of the unsolved problems in the physics of disordered solids is the nature of their lowtemperature anomalies in thermal properties. A large set of experimental data has been accumulated, for example, on heat capacity and thermal conductivity, various hypotheses for their description have been proposed, and various theoretical approaches have been developed to describe the dynamics of disordered solids.[1] The report will present the results of an analysis of the low-temperature heat capacity in normalized form, C_p/T^3 of a large amount of literature data for various crystalline, amorphous materials and glass structures such as molecular, metallic, and other non-magnetic materials including polymers. It was carried out to find the universal features of the $C(T)/T^3$ hump (a function with a maximum at $[C(T)/T^3]_{max}$ and T_{max}) for all complex solids and taking into account the assumed curve, which may be due to the structural features of the complex shape of solids. Quantities $[C(T)/T^3]_{max}$ and Debye temperature, along with T_{max} , are a unique set for the characterization of a solid. Good correlations between these quantities and a universal scaling dependence [2,3,4] are found, which allows one to observe the features of the excess low-temperature heat capacity both in the case of ordered crystals and disordered solids. The new information indicates that the origin of the boson peak in glasses is closely related to a hump C_p/T^3 in a complex crystal with a dispersion law, not to the van Hove singularity of simple crystals.

[1] M.A. Ramos (Ed.) Low-temperature Thermal And Vibrational Properties Of Disordered Solids (World Scientific, 2022).

- [2] D. Szewczyk et al., Sci. Rep. 11, 1 (2021).
- [3] Y. Miyazaki et al., J. Phys. Chem. Lett. 12, 2112 (2021).
- [4] A.I. Krivchikov et al., J. Phys. Chem. Lett. 13, 5061 (2022).

SUPERPARAMAGNETIC NANOCLUSTERS IN A STRONG APPLIED FIELD: UNDERSTANDING STATIC AND DYNAMIC MAGNETIC RESPONSE

A.A. Kuznetsov¹, E.V. Novak², E.S. Pyanzina², V.S. Zverev², S.S. Kantorovich^{1,3}

¹Faculty of Physics, University of Vienna, Vienna, 1090, Austria.
²Ural Federal University, Ekaterinburg, 620000, Russian Federation.
³Research Platform "Mathematics-Magnetism-Materials", University of Vienna, Vienna, 1090, Austria.

E-mail: andrey.kuznetsov@univie.ac.at

We present a theoretical study of superparamagnetic nanocluster ensembles in the presence of a static magnetic field of arbitrary magnitude. We model nanoclusters as rigid spherical particles filled with a fixed number of interacting point-like magnetic dipoles of constant magnitude [1]. Dipoles are distributed within the cluster volume randomly and uniformly, their spatial positions are fixed. However, they fully retain rotational degrees of freedom and are subjected to a rotational Brownian motion.

The first part of the work is dealing with the equilibrium magnetization curve of concentrated ensembles. A simple phenomenological model is proposed that describes magnetostatic response at low to intermediate values of the dipolar coupling constant. However, direct comparison with Langevin dynamics simulations demonstrate a possibility of a field-induced aggregation of nanoclusters, which results in an anomalous increase of the magnetic response and a unique "S-like" shape of magnetization curves [2].

In the second part we use Langevin dynamics to investigate a stochastic magnetomechanics of an isolated nanocluster under an oscillating magnetic field.

[1] A.A. Kuznetsov, Phys. Rev. B 98, 144418 (2018).

[2] A.A. Kuznetsov et al., J. Mol. Liq. 359, 119373 (2022).

Optical computation of the spin glass dynamics

M. Leonetti^{1,2}, L. Leuzzi^{2,3}, E. Hörmann³, G. Parisi³, G. Ruocco^{1,3}

¹Center for Life Nano Science@Sapienza, Istituto Italiano di Tecnologia, 00161 Rome, Italia. ²CNR NANOTEC-Institute of Nanotechnology, Soft and Living Matter Lab, 00185 Rome, Italy. ³CDepartment of Physics, University Sapienza, 00185 Roma, Italy.

E-mail: marco.leonetti@cnr.it

Optical computing is an emerging trend and it involves an optical layer paired with a traditional digital processing to improve performances while reducing computational costs and processing time. It promises parallel processing at high bandwidth that may be eventually performed in free space, with limited power consumption. Optical approach is becoming leading in quantum transport, quantum simulation [1], and machine learning: the calculation is in this case performed at the "speed of light" [2]. Spin glasses [3] serve as prototype models, capable of providing both equilibrium and off-equilibrium nontrivial phenomenology. In particular, the dynamics in an energy landscape with many equilibrium states and the origin of (multiple) relaxation times in finite dimensional systems are open questions in statistical mechanics. Indeed, novel methods for the calculation of the equilibrium states and of the dynamics of a spin glass system are highly sought after. By exploiting last-generation optical modulation devices, in a recent paper [4] we propose an optical system to compute the dynamics of a given spin glass state. By implementing an analog optical calculation layer, we realized an optical spin glass simulation. We observed that the overall intensity on a screen placed downstream of a strongly scattering medium shone with N coherent light rays can be formally written as a spin glass Hamiltonian. We demonstrate the optical supremacy of our approach with respect to digital computing.

- [1] N.C. Harris et al., Nat. Photon. 11, 447, (2017).
- [2] N.H. Farhat et al., Appl. Opt., 24, 1469 (1985).
- [3] M. Mezard, G. Parisi, and M. Virasoro, *Spin glass theory and beyond: An Introduction to the Replica Method and its Applications* (World Scientific Publishing Company, 1987).
- [4] M. Leonetti et al., Proc. Natl. Acad. Sci. U.S.A. 118, e2015207118 (2021).

MULTIMODE FIBERS: AN EXTREMELY FLEXIBLE TOOL FROM FAST IMAGING TO RESERVOIR COMPUTING

L. Leuzzi^{1,3}, D. Ancora^{2,1}, D. Ballarini³, P. Cazzato³, M. De Giorgi³, L. Dominici³, A. Gianfrate³, M. Negri^{4,1}, F. Ricci–Tersenghi^{4,1}, D. Sanvitto³, D. Trypogeorgos³

> ¹CNR-NANOTEC, Rome. ²EMBL, Rome. ³CNR-NANOTEC, Lecce. ⁴Dept. Of Physics, Sapienza University, Rome.

E-mail: luca.leuzzi@cnr.it

In disordered photonics, a fundamental task is to characterize the transmission matrix of the optically opague material. Multi-mode optical fibers are extensively studied because they are cheap and easy-to-handle devices. Given the sensitivity of a flexible medium, the speed of transmission matrix recovery is a crucial element for real life applications. Iterative phaseretrieval methods provide the fastest reconstructing frameworks: the less the iterations, the faster the learning from training data. We present a new kind of phase retrieval, reconstructing the transmission matrix in a single iteration, using a smaller amount of experimental data to feed the learning procedure than in state-of-the-art phase protocols [1]. The novelty is simple: to exploit the speckle spatial correlations. Furthermore, mm fibers can been turned into computing tools. They are used as random projectors introducing a sufficient amount of interaction to perform non-linear classification, as a reservoir computing resource [2]. Training a single logistic regression layer on the data images projected by the fiber improves the accuracy with respect to learning it on the raw images. Consistently with the current theory of deep neural networks, we also show that the classifier lives in a flatter region of the loss landscape when trained on fiber data, suggesting that mm fibers exhibit robust generalization properties, making them promising tools also for optically-aided machine learning.

[1] D. Ancora et al., arXiv:2112.09564.

[2] D. Ancora et al., arXiv:2210.04745.
SPERM CELLS IN MICRO-TRAPS: MOTILITY SORTING AND SWIMMING PRECISION

C. Maggi¹, L. Caprini², B. Nath³, R. Di Leonardo⁴, A. Puglisi³

¹NANOTEC-CNR, Piazzale A. Moro 2, I-00185, Rome, Italy.
²Heinrich-Heine-Universität Düsseldorf, D-40225 Düsseldorf, Germany.
³ISC-CNR, Institute for Complex Systems, Piazzale A. Moro 2, Rome, Italy.
⁴Dipartimento di Fisica, Sapienza Università di Roma, Rome, Italy.

E-mail: claudio.maggi@roma1.infn.it

Sperm swimming is crucial to fertilise the egg in nature and in assisted reproductive technologies. In this talk I will show how we have implemented a passive technique for the amplification of motile bull sperm concentration [1]. This is realised by building micro-chambers capable of trapping swimming sperms yielding a significant enhancement of motile cells density in the traps. Fabricating chambers with different shape and size highlights the ingredients that are key to optimal trap design. In the second part of the talk I will show we have used an array of micro-cages to trap the sperms' cell bodies and accurately track the spatial modes of the sperms' flagella during their beating cycle [2]. By studying accurately the fluctuations of the dynamics of the flagellum we find that the maximum precision of this motion is close to the estimated precision of one single dynein molecular motor actuating the flagellar axoneme. I will discuss how the maximum precision of individual molecular motors is related to their energy dissipation by the thermodynamic uncertainty relation and how the precision level of the whole flagellum can be explained by a schematic model with strong motor-motor coupling.

[2] C. Maggi et al., arXiv:2211.07779.

^[1] B. Nath et al., Lab on Chip 2023, in press (preprint at arXiv:2208.14098).

Reaching the yield point of a glass during X-ray irradiation

A. Martinelli¹, F. Caporaletti^{2,3,4}, F. Dallari¹, M. Sprung⁵, F. Westermeier⁵, G. Baldi⁶, G. Monaco¹

¹University of Padova, Department of Physics and Astronomy "Galileo Galilei", Via F. Marzolo, 8 – 35131 Padova,

ltaly.

²Van der Waals-Zeeman Institute, Institute of Physics, University of Amsterdam, Science Park 904, 1098 XH Amsterdam, The Netherlands.

³Van 't Hoff Institute for Molecular Sciences, University of Amsterdam, Science Park 904, 1098 XH Amsterdam, The Netherlands.

⁴Present address: Laboratory of Polymer and Soft Matter Dynamics, Experimental Soft Matter and Thermal Physics (EST), Université libre de Bruxelles (ULB), Brussels 1050, Belgium.

⁵Deutsches Elektronen–Synchrotron DESY, Notkestr. 85, 22607 Hamburg, Germany.

⁶University of Trento, Department of Physics, I-38123, Trento, Italy.

E-mail: alessandro.martinelli@unipd.it

A solid loaded beyond the yield stress looses its elastic properties and becomes plastic. From a microscopic point of view, this corresponds to the condition where plastic regions become so densely packed to give rise to system-spanning structures [1]. This limit for glasses is abrupt, which makes experimental investigations challenging: Brittle glasses exhibit mechanical failure at yield and the transition is strongly dependent on the glass history and preparation protocol [2].

In this talk we present recent experimental results in an oxide glass obtained with X-ray photon correlation spectroscopy [3]. We show that X-rays create point defects which behave as plastic regions and their number is directly controlled by the absorbed dose. We follow the atomic length-scale dynamics as a function of the exchanged momentum and distinguish different regimes: for a low-density of defects, they behave as isolated plastic regions that induce atomic displacements typical of an elastic solid. As the density of defects increases, the mechanical response of the glass changes from elastic to more and more plastic, until reaching the limit where it becomes that characteristic of a flowing system, which signals the reaching of the yield point.

[1] R. Dasgupta et al., Phys. Rev. Lett. 109, 255502 (2012).

[2] M. Ozawa et al., Proc. Natl. Acad. Sci. U.S.A. 115, 6656–6661 (2018).

[3] A. Martinelli et al., submitted.

Dissecting the role of division noise in promoting cancer phenotypic heterogeneity

M. Miotto

CLN2S, Center for Life Nano & Neuro Science, Italian Institute of Technology, Rome, Italy. Department of Physics, Sapienza University of Rome, Rome, Italy.

E-mail: mattia.miotto@roma1.infn.it

Phenotypic heterogeneity in cancer cell populations arises due to extrinsic and intrinsic sources of biological noise [1]. In particular, intrinsic cell-to-cell variability is understood as the inevitable consequence of stochasticity in molecular processes, such as RNA/protein level fluctuations and partition of cellular components at the cell division. Indeed, partitioning noise, i.e. fluctuations in the cellular compounds during cellular division, is regarded as one of the major enhancers of phenotypic variability in genetically identical cell populations. Depending on their extent, such fluctuations can lead to dramatic changes in the cell state and thus to transitions between different phenotypes. This phenotypic plasticity increases the heterogeneity of the tumor and changes its ability to resist treatments. Here, we present a novel experimental method combined with a minimal model to quantity the partitioning statistics of different cellular elements [2]. Testing our method by measuring simultaneously the division of cytoplasm, membrane proteins, and mitochondria in human leukemia cells, we found that although cell cytoplasm is partitioned symmetrically, mitochondria and membrane lipids are asymmetrically partitioned between daughter cells. Finally, we show that our experimental apparatus and theoretical framework can be generalized to different cell kinds, providing a tool for understanding partitioning-driven biological processes.

[1] A De Martino, T Gueudré, and M Miotto, Phys. Rev. E 99, 012417 (2019)

[2] G Peruzzi et al., Comm. Phys., 4, 188 (2021)

VIBRATIONAL PROPERTIES OF PARTICULATE PHYSICAL GELS

H. Mizuno, M. Hachiya, A. Ikeda

Graduate School of Arts and Sciences, The University of Tokyo, Tokyo 153-8902, Japan.

E-mail: hideyuki.mizuno@phys.c.u-tokyo.ac.jp

There are several kinds of amorphous materials in our world, where "constituents", such as atoms, molecules, polymers, colloidal particles, granular particles, and so on, condense into amorphous states. For example, in glasses, atoms and/or molecules are packed tightly into dense states. On the other hand, in gels, polymer molecules or colloidal particles are connected via bonds to form sparse networks.

Thanks to past efforts, our understanding of vibrational properties of high-density amorphous materials, glasses, has much advanced. Glasses exhibit characteristic properties which are markedly different from crystalline counterparts. Vibrational excitations in crystals are described in terms of phonons, whereas glasses show non-phonon excitations in addition to phonon vibrations. The non-phonon properties are observed as excess density of states over the Debye value, referred to as boson peak (BP), and also existence of quasi-localized vibrations (QLV).

Here, in this work, we would focus on low-density amorphous materials, particulate gels (or colloidal gels). In contrast to the glasses, the gels are sparse materials. Based on the results obtained from molecular dynamics simulations, we show that the BP and QLV, which are observed universally in many glasses, are reduced or even absent in the gels. Rather, in the low-frequency regime of gels, phonon-like vibrations are developed on the fractal-like, network structures.

[1] H. Mizuno, M. Hachiya, and A. Ikeda, J. Chem. Phys. 155, 234502 (2021).

[2] H. Mizuno, M. Hachiya, and A. Ikeda, J. Chem. Phys. 156, 204505 (2022).

On the microscopic nature of two-level systems in glasses

F.-C. Mocanu¹, L. Berthier^{2,3}, S. Ciarella¹, D. Khomenko^{4,5}, D. Reichman⁴, C. Scalliet⁶, F. Zamponi¹

¹Laboratoire de Physique de l'Ecole normale supérieure, Paris, France.

²Laboratoire Charles Coulomb, Montpellier, France.

³Yusuf Hamied Department of Chemistry, University of Cambridge, United Kingdom.

⁴Department of Chemistry Columbia University, New York, United States.

⁵Dipartimento di Fisica, Sapienza Università di Roma, Italia.

⁶Department of Applied Mathematics and Theoretical Physics, University of Cambridge, United Kingdom.

E-mail: felix-cosmin.mocanu@phys.ens.fr

Tunneling two-level systems (TLS) are an elegant model for the quasi-universal thermal and mechanical properties of glasses at cryogenic temperatures [1,2]. Despite the success of the model, many puzzles remain. Recent simulations probing the potential energy landscape of simple model glass formers have shown that such defects deplete with increasing glass stability [3]. The advent of swap Monte-Carlo dynamics now enables the simulation of model metallic glasses, in equilibrium, at temperatures near the corresponding glass transition. By looking at minimum-energy paths between inherent glassy states obtained from simulations, we can begin to form a picture on the atom nature of TLS [4]. We have explored the relationship between TLS and quasi-localised modes and the interplay between quasi universality and different aspects of glass phenomenology. Overall, understanding the properties of two-level systems in glasses is important for optimizing the thermal and mechanical properties of glass-based materials, and can be useful in the development of new glass-based technologies.

- [1] P.W. Anderson, B.I. Halperin, and C.M. Varma, Phil. Mag. 25, 1 (1972).
- [2] W.A. Phillips, Rep. Prog. Phys. 50, 1657 (1987).
- [3] D. Khomenko et al., Phys. Rev. Lett. 124, 225901 (2020).
- [4] F.-C. Mocanu et al., J. Chem. Phys. 158, 014501 (2023).

Dynamic cluster formation, viscosity and diffusion in monoclonal antibody solutions

I. Mosca^{1,2}, C. Beck^{1,2}, K. Pounot³, O. Matsarskaia², C. Grapentin⁴, L. Matthews³, T. Seydel², F. Schreiber¹

¹Institut für Angewandte Physik, Universität Tübingen, Germany. ²Institut Max von Laue – Paul Langevin, Grenoble, France. ³European Synchrotron Radiation Facility, Grenoble, France. ⁴Lonza AG, Ltd., Basel, Switzerland.

E-mail: mosca@ill.fr

Antibodies are proteins playing an essential role in the immune response of mammals. Monoclonal antibodies (mAbs) are particularly relevant for therapeutic applications due to their high specificity and versatility. A current pharmaceutical challenge is to formulate highly concentrated antibody solutions to achieve a significant therapeutic effect, while minimizing their viscosity and keeping it under the subcutaneous injectability limit [1,2], thus rendering the drug administration to patients less difficult and painful.

Since the understanding of macroscopic viscosity requires an in-depth knowledge on protein diffusion, mutual interactions and dynamic cluster formation [3,4], we study the self-diffusion of five mAbs of the IgG1 subtype (produced and characterized at Lonza AG) in aqueous solution as a function of antibody type and concentration, by quasi-elastic neutron scattering (QENS). QENS allows to determine the hydrodynamic cluster size of the solutions [5] and sheds light on the mAb internal dynamics. Data have been treated using analysis frameworks we developed [6] and interpreted using colloid physics theory.

A subset of these mAbs has been also investigated using small angle neutron and X-ray scattering (SANS and SAXS) to probe mutual interactions between mAb molecules. Complementary information is provided by molecular dynamics (MD) simulations and rheology measurements. As a reference, we use polyclonal antibody (IgG from bovine serum) solutions [7], thus obtaining a comprehensive picture of mAb diffusion.

- [1] C. Berteau et al., Medical Devices (Auckl.) 8, 473, (2015).
- [2] I. Usach, et al., Adv. Ther. 36, 2986–2996 (2019).
- [3] S. von Bülow et al., Proc. Natl. Acad. Sci. U.S.A. 116, 984 (2019).
- [4] M. Heinen et al., Soft Matter 8, 1404 (2012).
- [5] M. Grimaldo et al., Quart. Rev. Biophys., 52, e7 (2019).
- [6] C. Beck et al., EPJ Web Conf. 272, 01004 (2022).
- [7] M. Grimaldo et al., J. Phys. Chem. B 118, 7203 (2014).

INSTANTANEOUS NORMAL MODES IN LIQUIDS: THEORY AND SIMULATION

S. Mossa¹, T. Bryk², G. Ruocco³, W. Schirmacher⁴

¹Univ. Grenoble Alpes, CEA, IRIG-MEM-LSim, 38054 Grenoble, France.

²Institute for Condensed Matter Physics, National Academy of Sciences of Ukraine, UA-79011 Lviv, Ukraine, ³Dipartimento di Fisica, Universitá di Roma "La Sapienza", Piazzale Aldo Moro 5, I-00185, Roma, Italy.

⁴Institut für Physik, Staudinger Weg 7, Universität Mainz, D–55099 Mainz, Germany.

E-mail: stefano.mossa@cea.com

The vibrational density of states in crystals and glasses is mirrored in liquids by the Instantaneous Normal Modes spectrum, which provides important information on liquid dynamics and transport properties. The concept of heterogeneous-elasticity theory has been successful in explaining low-frequency excitations in glasses, and in this talk we present an extension of this theory to the liquid state. Our findings, based on theory and Molecular Dynamics simulations of a model liquid, show that the spectrum of eigenvalues of the Hessian matrix has a sharp maximum near $\lambda = 0$ and decreases monotonically with $|\lambda|$. The spectral shape is found to be temperature dependent, being symmetric at high temperatures and becoming asymmetric at low temperatures close to the dynamical critical temperature. Importantly, the theory naturally reproduces a zero-energy spectral singularity with a cusp-like character that develops in the vibrational spectra upon cooling. This feature has been generally overlooked in the past, due to misleading representation of data.

Mechanical memory of metallic glasses: time and temperature dependence of the anelastic relaxation

M. Nabahat, E. Pineda

Department of Physics, Institute of Energy Technologies, Universitat Politècnica de Catalunya - BarcelonaTech, Barcelona, 08019, Spain.

E-mail: mehran.nabahat@upc.edu

In this contribution we will present our recent work on $Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5}$ allog [1,2]. This well-known glass former shows outstanding stability against crystallization or decomposition, and a rather "simple" mechanical spectrum with just an α peak and no evidence of secondary processes. This makes this material a good model to get insight into the universal features of the relaxation dynamics of metallic glasses. Here we will focus on the anelastic behavior, or delayed elasticity, in the range of temperatures between $0.85T_g$ and T_g . Deformation recovery experiments after various loading times indicate that the temporal distribution of reversible deformation modes is invariant across a wide temperature range and has a time distribution, τ_n , that can be directly related to the shape of the high-frequency wing of the α relaxation. The results also show a simple relationship between the relaxation spectrum and the corresponding distribution of activation energies, revealing the origin of transient creep and anelastic recovery behaviors and allowing to model the mechanical memory of the metallic glass, which is a critical knowledge to interpret the rate and temperature dependence during mechanical probes.

[1] N. Amini et al., Phys. Rev. Mater. 5, 055601 (2021).

[2] M. Nabahat et al., Phys. Rev. Mater. 6, 125601 (2022).

LIQUID DYNAMICS EASES THE EQUILIBRATION OF GLASSES

S. Napolitano, Z. Song, K. Wang, C. Li, F. Pabst, E. Thoms

Laboratory of Polymer and Soft Matter Dynamics, Experimental Soft Matter and Thermal Physics (EST), Université libre de Bruxelles (ULB), Brussels 1050, Belgium.

E-mail: simone.napolitano@ulb.ac.be

The rate at which a nonequilibrium system decreases its free energy is commonly ascribed to molecular relaxation processes, arising from spontaneous rearrangements at the microscopic scale. While equilibration of liquids usually requires density fluctuations at timescales quickly diverging upon cooling – known as the α -modes –, growing experimental evidence indicates the presence of different pathways exhibiting a temperature-invariant activation energy on the order of 100 kJ/mol. Based on a large series of molecular dynamics and equilibration experiments, we identified the underlying molecular process responsible for this class of Arrhenius equilibration mechanisms with a slow mode (SAP), universally present in the liquid dynamics [1]. We verified that this process is present also in bulk melts (layer thickness > 200 nm) and that the activation energy and the characteristic molecular time of the SAP are not affected by either the macromolecular or the sample size. In line with recent experiments by mechanical spectroscopy [2], our measurements indicate that this process extends for length scales much larger than those of the α -modes. The SAP, which we show is intimately connected to high temperature flow, can efficiently drive melts and glasses towards more stable, less energetic states. Our results show that measurements of liquid dynamics can be used to predict the equilibration barrier in the glassy state and the relaxation rate in proximity of free interfaces.

[1] Z. Song et al., Sci. Adv. 8, eabm7154 (2022).

[2] H. Yuan et al., Sci. Adv. 8, eabq5295 (2022).

NUMERICAL EVIDENCE OF GLASS AND PSEUDO-LOCALIZATION TRANSITIONS IN THE MODE-LOCKED P-SPIN MODEL FOR RANDOM LASERS

J. Niedda^{1,2}, G. Gradenigo^{3,4,3}, L. Leuzzi^{1,2}, G. Parisi^{1,2,5,6}

¹Dipartimento di Fisica, Università di Roma "Sapienza", Piazzale A. Moro 2, I-00185, Roma, Italy.
²NANOTEC CNR, Soft and Living Matter Lab, Roma, Piazzale A. Moro 2, I-00185, Roma, Italy.
³Gran Sasso Science Institute, Viale F. Crispi 7, 67100 L'Aquila, Italy.

⁴INFN-Laboratori Nazionali del Gran Sasso, Via G. Acitelli 22, 67100 Assergi (AQ), Italy.

⁵INFN, Sezione di Roma-1, P.le A. Moro 5, 00185, Rome, Italy. ⁶Accademia Nazionale dei Lincei, Palazzo Corsini – Via della Lungara, 10, I-00165, Roma, Italy.

E-mail: jacopo.niedda@uniroma1.it

Optical waves in active disordered media display the typical phenomenology of complex systems. The multiple scattering of light with randomly placed scatterers inside a material confines the electromagnetic field and entails the existence of well-defined cavity modes with long lifetimes competing for amplification. The presence of modes in random lasers can be revealed from the highly structured spectra measured in experiments. If one takes several spectral shots from the same piece of material, the position of the intensity peaks changes from shot to shot, meaning that there is no specific frequency which is preferred in the lasing phenomenon, but depending on the initial state, with the disorder kept fixed, the modes gaining the highest intensity change every time. In order to explain this behaviour, a statistical mechanical model derived from spin-glass theory has been developed, where the light modes are described as non-linearly interacting phasors on the so-called mode-locked diluted graph [1]. The specific mode-coupling selection rule, which naturally emerges in the study of lasing modes dynamics, impairs the analytical solution of the model out of the narrow bandwidth limit, where the interaction network is fully connected.

In this talk we present recent results from numerical simulations of the mode-locked glassy random laser. The presence of a phenomenology compatible with a glass transition when tuning the optical power above a threshold value of the system is put in evidence from the divergence of the specific heat and a non-trivial structure of the Parisi overlap distribution function [2]. By means of a refined finite-size scaling analysis of the critical region, the transition is assessed to be compatible with a mean-field universality class [3]. A pseudo-localization transition to a phase where the intensity of light is neither properly localized on a single mode nor equiparted among all the modes is revealed from the measure of the inverse participation ratio and of the spectral entropy [4]. The two transitions occur at the same temperature as different manifestations of the same underlying phenomenon, the breaking of ergodicity.

- [1] F. Antenucci et al., Phys. Rev. Lett. 114, 043901 (2015).
- [2] G. Gradenigo, F. Antenucci and L. Leuzzi, Phys. Rev. Res. 2, 023399 (2020).
- [3] J. Niedda, G. Gradenigo, L. Leuzzi and G. Parisi, arXiv:2210.04362.
- [4] J. Niedda, L. Leuzzi and G. Gradenigo, arXiv:2212.05106.

Origin of the anomalous properties of water determined using X-ray lasers

A. Nilsson

Department of Physics, Stockholm University, Sweden.

E-mail: andersn@fysik.su.se

The finding that the thermodynamic response and correlation functions appear to diverge in supercooled water towards a singular temperature estimated by power-law fits of about 228 K has led to several hypotheses about the origin of waters anomalous properties. One hypothesis to explain the apparent divergence is that there exists a liquid-liquid transition with a liquid-liquid critical point at rather high positive pressures. In this scenario the Widom line, defined as the locus of correlation length maxima in the P-T plane, emanates from the critical point as a continuation of the liquid-liquid transition line into the one-phase region and the divergence in the response functions is towards this line. The challenge is that the temperature Ts lies below the homogeneous ice nucleation temperature 232 K, a region of the phase diagram that has been denoted as "no man's land", since ice crystallization occurs on much faster time scale compared to the experimentally accessible time scale in a typical laboratory setting. Here I will present how X-rays from X-ray lasers and synchrotron radiation sources have been used to probe the liquid phase of water from hot to the deep supercooled water regime inside no-man's land. In particular I will discuss if a liquid-liquid transition, Widom line and a critical point exists in deep supercooled water causing fluctuations all the way up to ambient temperature.

Towards designing of actively moving nanoparticles: theoretical estimation of optimal geometry and engine parameters

I.A. Nyrkova and A.N.Semenov

Institut Charles Sadron, CNRS, 23 rue du Loess, BP 84047, 67034 Strasbourg, France.

E-mail: irina.a.nyrkova@gmail.com

Studying/developing of artificial micro-swimmers/nano-robots (NR) has a strong application potential ranging from water treatment (waste removal) to nano-medicine (drug delivery, microsurgery). Active swimming of micro-objects presents a special challenge: at low Reynolds numbers the hydrodynamics becomes essentially time-reversible, rendering simple back-andforth paddle flaps inefficient. On the other hand, thermal noise is an important competitor for NR' active motion, hence, depending on the engine efficiency and the observation time, there are three basic regimes of NR' behaviour: Brownian diffusion, active drift and active diffusion. Several working principles were already proposed for the artificial engines in microworld (chemical, catalytic, phoretic self-propulsion, active external field, revolving molecular motors). However, the configurational details of an NR assembly between the engines and the transported cargo have been largely left out of scope of the research. In our talk we discuss these questions paying attention to the optimal size/shape of the load, the engine-load attachment (joint length & flexibility), the number of engines per NR and their geometrical organization (with respect to each other and around the load). We determine the individual engine driving force and the engine's possible frustation (overload) point, motor efficiency, role of convection and the active/thermal diffusion ratio for various NR architectures.

The difference of aging near or far from equilibrium

F. Pabst, S. Napolitano

Université libre de Bruxelles, Polymer and Soft Matter Dynamics, 1050 Brussels, Belgium.

E-mail: florian.pabst@ulb.be

Physical aging, the evolution of a glassy material toward equilibrium, is commonly assumed to proceed on the time scale of the structural relaxation (α -modes). However, growing experimental evidence shows that glasses can age at timescales shorter than those of the α -relaxation, and that the equilibration kinetics can undergo a two-step mechanism [1]. In such cases, a final step occurring at the timescale of structural mobility follows a first step at much shorter times. The microscopic origin of the fast equilibration mechanism was very recently discovered by dielectric spectroscopy [2], and associated to the so-called slow Arrhenius process (SAP). This molecular process is slower than the structural relaxation above T_q but, because of its weaker temperature dependence, eventually relaxes faster than the α -modes in the glassy state. Here, we show that a decisive point for the contribution of the SAP to the aging process is the distance of the system from equilibrium. For small temperature steps, aging proceeds solely via the structural relaxation and can be predicted by the Tool-Narayanaswamy-Moynihan (TNM) model. For large temperature steps or at large cooling rates, the TNM model fails, and a two-step aging process is observed. A model free isoconversional analysis shows that, in the latter cases, the activation energy grows with the aging time and varies from the value of the SAP to that of the structural relaxation. We conclude that the aging at long times is driven by the α -modes while at short times it is governed by the SAP; in between these limiting region a broad crossover is observed, which explains the large dispersion of activation energy values reported in the literature.

[1] D. Cangialosi et al., Phys. Rev. Lett. 111, 095701 (2013).

[2] Z. Song et al., Sci. Adv. 8, 7154 (2022).

DIFFUSIVE DYNAMICS OF BACTERIAL PROTEOME AS A PROXY OF CELL DEATH

A. Paciaroni¹, D. Di Bari^{1,2}, S. Timr³, M. Guiral⁴, M.-T Giudici-Orticoni⁴, T. Seydel⁵, C. Beck⁵, C. Petrillo¹, P. Derreumaux⁶, S. Melchionna⁷, F. Sterpone⁶, J. Peters²

¹Dipartimento di Fisica e Geologia, Università degli Studi di Perugia. ²Laboratoire Interdisciplinaire de Physique, Université Grenoble Alpes, CNRS. ³J. Heyrovský Institute of Physical Chemistry, Czech Academy of Sciences. ⁴Laboratoire de Bioénergétique et Ingénierie des Protéines, IMM, CNRS, Aix-Marseille Université. ⁵Institut Laue-Langevin, Grenoble, France. ⁶ISC-CNR, Dipartimento di Fisica, Università Sapienza. ⁷Laboratoire de Biochimie Théorique, CNRS, Université de Paris Cité.

E-mail: alessandro.paciaroni@unipg.it

Temperature variations have a big impact on the bacterial metabolism and death, yet an exhaustive molecular picture of these processes is still missing. For instance, whether the thermal death is determined by the deterioration of the whole or a specific part of the proteome is hotly debated. In our work, by monitoring the proteome dynamics of E. coli we show that only a minor fraction of the proteome unfolds at the cell death. First, we prove that the dynamical state of the E. coli proteome is an excellent proxy for the temperature dependent bacterial metabolism and death. The proteome diffusive dynamics peaks at about the bacterial optimal growth temperature, then a dramatic dynamical slowdown is observed which starts just below the cell's death temperature. Next, we show that this slowdown is caused by the unfolding of just a small fraction of proteins which establish an entangling inter-protein network—dominated by hydrophobic interactions— across the cytoplasm. Finally, we prove that the deduced progress of the proteome unfolding and its diffusive dynamics are both key to correctly reproduce the E. coli growth rate.

Hydrodynamically-induced morphologies in active and driven matter

I. Pagonabarraga

Department of Condensed Matter Physics, University of Barcelona, Carrer Martí i Franqués 1, 08028-Barcelona, Spain.

E-mail: ipagonabarraga@ub.edu

Suspensions intrinsically out of equilibrium disturb the liquid medium in which they are embedded as a result of nonequilibrium processes, such as chemical reactions, or inhomogeneous thermal heating. These are intrinsically out of equilibrium systems, which makes them very versatile, with a natural tendency to self-assemble. Due to their small size, these out of equilibrium dynamical states generates flows that induce long range hydrodynamic interactions. These interactions have profound effects in the transport, spontaneous self-assembly and emergence of stable mesoscopic structures.

I will analyze the role that hydrodynamics play in the rectification mechanism that leads to particle motion, as well as the possibility that hydrodynamic instabilities lead to novel, dynamically-stabilized mesostructures. I will discuss the impact that these mechanisms have in the emergence of patterns and different type of morphological structures in active suspensions. I will combine theoretical simple models and computer simulations to gain insight in the role of hydrodynamics in these out of equilibrium system

The Debye-Waller factor of amorphous glass from static speckle patterns

C.F. Petersen^{1,2}, P. Harrowell²

¹School of Chemistry, University of Melbourne, Melbourne, VIC, Australia. ²School of Chemistry, University of Sydney, Sydney, NSW, Australia.

E-mail: charlotte.petersen@unimelb.edu.au

Scattering experiments that measure the static structure factor are the standard structural determination tool for crystalline materials. However, their utility is limited for amorphous materials, where the lack of periodic order means structural signatures become washed out; Only average structural information can be obtained. In addition to these measurement difficulties, we lack a complete theory to predict the rigid behaviour of glass from its amorphous microscopic structure.

Recent progress has connected the presence of localised soft normal modes to the dynamics of glass [1,2]. Building on this success, the capacity of a configuration to restrain particle motion has been formalised as a structural order parameter that characterises the solidification of a liquid into an amorphous solid on cooling [3]. However, this quantity cannot be measured in experiments. Here, we demonstrate that the Debye-Waller factor, which quantifies particle vibrations from the static structure factor, is a suitable experimentally accessible structural order parameter. Despite the weak temperature dependence of the spherically averaged static structure factor, we find that the Debye-Waller factor of a simulated glass shows a strong temperature dependence when we consider the full speckle scattering pattern.

[1] A. Widmer-Cooper and P. Harrowell, Phys. Rev. Lett. 96, 185701 (2006).

[2] R. L. Jack, A. J. Dunleavy, and C. P. Royall, Phys. Rev. Lett. 113, 095703 (2014).

[3] G. Sun and P. Harrowell, J. Chem. Phys. 157, 024501 (2022).

Evolution of relaxation time and dynamic heterogeneity through aging in metallic glasses

E. Pineda, M. Nabahat, Y. Duan

Department of Physics, Universitat Politècnica de Catalunya - BarcelonaTech.

E-mail: eloi.pineda@upc.edu

Physical aging in disordered metallic systems is usually studied by determining the evolution of viscosity and static properties like volume and enthalpy. The microscopic dynamics has been also recently studied by X-ray photon correlation spectroscopy finding intermittent behaviors, avalanche like processes, and surprisingly fast, stationary off-equilibrium dynamics. In this contribution, we present an unprecedented detailed study of the evolution of the macroscopic dynamics with aging. The initial glass state was obtained by fast guenching, with a high fictive temperature, and the evolution of the macroscopic relaxation dynamics through aging was studied isothermally in the range of temperatures from $0.85T_q$ to T_q . The relaxation time τ and the level of dynamic heterogeneity were obtained from the experimental mechanical relaxation modulus $\mathcal{M}(t, t_w)$. Contrary to microscopic observations, the macroscopic aging is found to evolve continuously, with a $\tau \propto t_w^{\mu(T)}$ law, driving the system toward increasingly flat regions of the energy landscape. The aging exponent, $\mu < 1$, tends to $\mu \sim 1$ as temperature decreases and the glass state is more distant from equilibrium. Besides, we found that aging produces a small but continuous increase of dynamic heterogeneity. In addition to present the experimental results, we will discuss if the macroscopic data can be related to the microscopic observations and how can be described by current aging models.

A SOFT MATTER PERSPECTIVE OF PROTEINS: FROM PHASE BEHAVIOR TO DYNAMICS

A. Ragulskaya

Institut für Angewandte Physik, Universität Tübingen, 72076 Tübingen, Germany.

E-mail: anastasia.ragulskaya@uni-tuebingen.de

Proteins are considered the machinery of life. They are an exciting subject of study for many branches of modern science and technology, from biology to medicine and pharmacy, but also in colloid science, chemical engineering and nanotechnology. In many cases, not only the behavior of individual proteins needs to be understood, but rather the collective phenomena, which remains a challenge, in particular quantitatively. In many cases, models derived from soft matter, colloid or polymer-based, can be successfully employed. After some introductory remarks on the soft matter perspective of proteins, we discuss concepts for controlling and understanding protein phase diagrams, including aggregation pathways and the branching between them in aqueous solution by addition of multivalent ions. This route for the tailoring of the interaction potential is exploited for controlling a) crystallization, b) gelation and amorphous aggregation, c) network formation, as well as d) smaller aggregate formation including their dynamics and kinetics.

First, we present a real-time study of protein crystallization induced by multivalent ions using small-angle scattering and optical microscopy. Based on the crystallization kinetics, we propose a multistep mechanism. In the first step, an intermediate phase is formed, followed by the nucleation of crystals within the intermediate phase. In the next step, the intermediate phase is consumed by nucleation and slow growth. Competing pathways for amorphous aggregation and arrested states are also discussed.

Second, we present complementary investigations of the dynamics of these systems using quasielastic neutron scattering, showing a remarkably universal behavior of the effective diffusion as a function of concentration and salt under suitable conditions. We also discuss the mapping on models for diffusion of colloids.

Third, we show how these concepts can be transferred to protein–interface interactions, and how adsorption behavior can be manipulated by multivalent charges. We try to connect the interface and the bulk behavior, including reentrant adsorption and anomalous interface behavior upon approaching bulk phase boundaries.

Invaluable contributions by numerous collaborators are gratefully acknowledged as is financial support by BMBF, DFG, and ANR.

- [1] F. Roosen-Runge et al., Proc. Natl. Acad. Sci. U.S.A. 108, 11815 (2011).
- [2] A. Sauter et al., J. Am. Chem. Soc. 137, 1485 (2015).
- [3] M. Grimaldo et al., Q. Rev. Biophys. 52(e7), 1 (2019).
- [4] M. R. Fries et al., Phys. Rev. Lett. 119, 228001 (2017).
- [5] A. Girelli et al., Phys. Rev. Lett. 126, 138004 (2021).
- [6] N. Begam et al., Phys. Rev. Lett. 126, 098001 (2021).
- [7] M. Reiser et al., Nat. Comm. 13, 5528 (2022).

Suppression of two-level systems with increasing stability in TPD glasses

M.A. Ramos^{1,2}, M. Moratalla¹, R.J. Jiménez-Riobóo³ M. Rodríguez-Lopez^{4,5}, C. Rodríguez-Tinoco^{4,5}, J. Rodríguez-Viejo^{4,5}

¹Laboratorio de Bajas Temperaturas, Dpto. de Física de la Materia Condensada, and Instituto Nicolás Cabrera (INC), Universidad Autónoma de Madrid, Spain.

²Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, Spain.

³Instituto de Ciencias de Materiales de Madrid (ICMM-CSIC), Campus de Cantoblanco, Madrid, Spain.

⁴Departamento de Física, Facultat de Ciències, Universitat Autònoma de Barcelona, 08193 Bellaterra, Spain.

⁵Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and BIST, Campus UAB, Bellaterra, 08193 Barcelona, Spain.

E-mail: miguel.ramos@uam.es

The preparation and study of highly-stable structural glasses, which are expected to approach the concept of ideal glass, have recently attracted much attention. In particular, the question has been raised as to whether or not the controversial low-temperature "anomalous" properties of glasses at low temperature, usually attributed to additional low-energy excitations such as two-level systems (TLS), persist in these ultrastable glasses of much lower configurational entropy. Some previous attempts to answer this question have given contradictory results. To shed light on this puzzle, a particular glass-forming substance has been studied in this work, TPD, which can be grown by physical vapor deposition (PVD) in a highly-stable state, either anisotropic or isotropic, and also as an ordinary glass and in the crustalline state. Besides performing structural, calorimetric and acoustic characterization of the different samples prepared, we have measured the specific heat in the temperature range 0.4-20 K. While the ordinary glass presented the typical glassy features (a linear term attributed to TLS, a broad maximum in C_p/T^3 , i.e. the boson peak) and the crystal exhibited the expected Debye cubic dependence at very low temperatures, a strong depletion of the TLS contribution was found in both anisotropic and isotropic ultrastable glasses. The different degree of anisotropy and layering of the grown films only seem to influence the height of the vibrational background.

Electric fields for tuning molecular orientation in TPD-modified glasses: experiments and simulations

M. Rodríguez-López¹, C. Rodríguez-Tinoco², C. Melis³, L. Colombo⁴, M. Gonzalez-Silveira⁴, J. Rodríguez-Viejo^{5,6}

¹Group of Thermal properties of Nanoscale Materials (GTNaM), Universitat Autònoma de Barcelona (UAB) & Catalan Institute of Nanoscience and Nanotechnology (ICN2), Barcelona, Spain.

²Dipartimento di Fisica, Università di Cagliari, Cittadella Universitaria, Monserrato (Ca), Italy.

E-mail: marta.rodriguez@uab.cat

Molecular organic glasses are nowadays gaining attention due to the great advantages they present when used in the electronics industry, such as flexibility, freedom of choice of the underlying layers, low cost, easily control of the thickness and composition and simple fabrication process. However, their lack of efficiency and long-term stability prevent their presence in our daily life. These inconveniences can be solved if those glasses are prepared by Physical Vapour Deposition (PVD) [1]. This technique permits to tune glasses properties by changing the preparation conditions such as the deposition temperature and rate. One of the main assets of PVD is the possibility of obtaining ultrastable glasses, which are special highly-packed glasses that exhibit exceptional thermodynamic and kinetic stability [2], thus improving the efficiency of electronic devices. Another utility of PVD is the possibility of tuning the average spatial orientation of the molecules in the glass, differing from the random distribution that a traditional isotropic glass would have, therefore enhancing the electrical, thermal and optical properties of the glassy materials [3,4]. This two important parameters are strongly linked to the substrate temperature and it is therefore desirable to decouple them in order to obtain ultrastable glasses in which the orientation of its molecules can be chosen as demanded by the final application. The goal of this study is to change the molecular orientation while maintaining the stability of the glass by applying an electric field during the deposition of TPD and TPD-modified molecules. Ultraviolet-visible spectroscopy [5] will be used for quantifying the degree of anisotropy and the possible changes inferred by the electric field. In addition, Molecular Dynamics simulations have been performed to gain access to the behaviour of each molecule in the glass as well as the possibility of applying electric fields that are not achievable in the laboratory.

- [1] S.F. Swallen et al., Science 315(5810), 353-356 (2007).
- [2] L. Kenneth and S.F. Kearns, J. Phys. Chem. B 112(16), 4934-4942 (2008).
- [3] D. Yokoyama, J. Mater. Chem. 21, 19187 (2011).
- [4] J. Ràfols-Ribé et al., Phys. Rev. Mat. 2, 035603 (2018).
- [5] Y. Sakai et al., Appl. Phys. Express, 8, 096601 (2015).

Self-Assembly and magnetic susceptibility of colloidal magnetic nanoplatelets

M. Rosenberg¹, P. Camp², S. Kantorovich¹

¹Department of Physics, University of Vienna, Vienna, 1090, Austria. ²School of Chemistry, University of Edinburgh, David Brewster Road, Edinburgh EH9 3FJ, Scotland.

E-mail: margaret.rosenberg@univie.ac.at

Adding a magnetic component to conventional Soft Matter opens up many interesting research avenues to manipulate materials, due to both intrinsic interactions and coupling to an externally applied magnetic field. In case of magnetic colloidal systems, in which the magnetic nano- or micron-sized particles are suspended in magnetopassive carriers, both the magnetic anisometry and the shape anisotropy of colloidal nanoparticles have been shown to strongly affect the macroscopic properties of the resulting fluid. This contribution focuses on a system that sits at the intersection of colloidal liquid crystal and magnetic soft matter research; a suspension of colloidal magnetic nanoplatelets with dipole moments oriented perpendicular to the platelets' plane, which have been shown to exhibit a macroscopic ferromagnetic nematic phase. Using Langevin and Brownian Dynamics simulations, we study the microstructure of the suspension to understand how the cluster formation differs from suspensions of dipolar hard spheres, and calculate the static and dynamic magnetic susceptibilities. Comparing simulation findings to known theories and available experimental results we show the limitations of a single-dipole approximation, as well as how strongly the shape anisotropy affects the self-assembly scenario.

DESIGNING ENHANCED ENTROPY BINDING IN ASSOCIATIVE POLYMERS

L. Rovigatti, F. Sciortino

Physics Department, Sapienza University of Rome.

E-mail: lorenzo.rovigatti@uniroma1.it

Associative polymers are a class of functionalised polymeric objects in which a fraction of the monomers can bind to each other, forming either intra- or inter-polymer bonds. If the chain flexibility is large enough and the associative monomers can form only single bonds, then at low density bonding takes place essentially within the same polymer, forming soft nanoobjects named single-chain nanoparticles (SCNP) [1]. At larger densities, these particles form an interconnected spanning network, and a continuous cross-over from isolated SCNPs to a network state has been observed in experiments [2,3] and simulations [4], with no hints of a first-order transition, consistent with predictions of mean-field theory [5]. In this contribution I will numerically show that a judicious design of the patterns of reactive monomers along the polymer chain can drive a fully-entropic gas-liquid phase separation in SCNP systems, achieving control over the discontinuous (first-order) change, from a phase of diluted (fullybonded) polymers to a phase in which polymers entropically bind to each other to form a (fully-bonded) polymer network. Such a sensitivity arises from a delicate balance between the distinct entropic contributions controlling the binding, which can be exploited to design new self-assembling systems, as well as to better understand phase separation phenomena in associative (bio–)macromolecular systems [6].

[1] A.J. Pomposo, Single-Chain Polymer Nanoparticles: Synthesis, Characterization, Simulations, and Applications (John Wiley & Sons, 2017).

- [2] E.D. Whitaker, C.S. Mahon, and D. A. Fulton, Angew. Chem. 125, 990 (2013).
- [3] S. Tang, M. Wang, and B.D. Olsen, J. Am. Chem. Soc. 137, 3946 (2015).
- [4] M. Formanek et al., Macromol. 54, 6613 (2021).
- [5] A.N. Semenov and M. Rubinstein, Macromol. 31, 1373 (1998).
- [6] L. Rovigatti and F. Sciortino, Phys. Rev. Lett. 129, 047801 (2022).

COOPERATIVE DYNAMICS IN BULK WATER

M. Russina¹, F. Mezei², B. Faragó³

¹Helmholtz Zentrum Berlin, Hahn-Meitner Platz 1, 14109 Berlin, Germany.
²KFKI Campus, H–1525 Budapest, Konkoly Thege u 29–33, Hungary.
³Institute Laue–Langevin – 71 avenue des Martyrs, CS 20156, 38042 GRENOBLE Cedex 9 – France.

E-mail: Margarita.Russina@helmholtz-berlin.de

Liquid water exhibits many unusual properties, setting water aside from other molecular liquids. Theoretical concepts of many anomalies involve cooperative effects, such as the formation of hydrogen-bonded networks and/or rearrangements of molecular ensembles. However, experimental investigations of such cooperative phenomena have been extremely difficult and limited to very few studies.

Neutron scattering is very well suited for investigation of the structure and molecular dynamics at nanoscale. Cooperative processes are usually reflected in the coherent neutron scattering signal, while incoherent neutron scattering is related to the self-particle motion. Up to now the investigation of cooperative processes in H_2O has been hindered by the weak coherent signal compared to the incoherent one.

We have developed a new experimental approach combining of polarization analysis and neutron spin echo spectroscopy. Using our new approach, we were able for the first time to unambiguously detect and successfully separate cooperative water dynamics from the stochastic self-particle motion in H_2O on the picosecond to nanosecond time scale and on the nanometer length scale. Our study reveals the existence of a picosecond cooperative process in water, whose nature could be related to the cooperative rearrangements between several neighboring water molecules. Investigations of the light and heavy water reveal subtle but pronounced differences in hydrogen bond formation.

THEORY OF NON-PHONONIC EXCITATIONS IN DISORDERED SYSTEMS

W. Schirmacher^{1,2}, M. Paoluzzi^{3,4}, G. Ruocco^{2,4}

¹Institut für Physik, Staudinger Weg 7, Universität Mainz, D-55099 Mainz, Germany.

²Center for Life Nano Science @Sapienza, Istituto Italiano di Tecnologia, 295 Viale Regina Elena, I-00161, Roma, Italy.

³Departament de Física de la Matèria Condensada, Universitat de Barcelona, Carrer de Martí i Franquès 1, 08028 Barcelona, Spain.

⁴Dipartimento di Fisica, Università di Roma "La Sapienza", Piazzale Aldo Moro 5, I-00185, Roma, Italy

E-mail: walter.schirmacher@uni-mainz.de

Due to the absence of long-range symmetries the vibrational excitations of glasses differ very much from those of crystals (phonons). Only at low frequencies phonon-like waves exist, because their wavelength is so large that they are not sensitive to the structural disorder. At high frequencies (in the THz range) and/or in small computer-generated samples the vibrational excitations differ very much from phonons. They resemble more the eigenstates of random matrices.

Investigating the continuum limit of the Hessian of a glass, using a novel coarse-graining procedure, we identify two different types of non-phononic vibrational excitations. A first type (Type-I.) arises from spatially fluctuating elastic constants and are responsible for vibrational anomalies, which are observed in glasses in the THz range ("boson peak") [1]. A second type (Type-II.) becomes distinct in very small samples, in which the low-frequency waves are suppressed. These Type-II excitations are caused by local stresses and exhibit non-irrotational displacement patterns (vortices). We demonstrate that the frequency scaling of the Type-II excitations are non-universal and depend on the details of the underlying interaction potentials. Recent small-sample findings of non-phononic spectra, which scale with the fourth power of the frequency are shown to be an artifact of the smooth potential cutoff used in the simulations.

[1] W. Schirmacher and G. Ruocco, *Heterogeneous Elasticity: The tale of the boson peak*, in *Low-Temperature Thermal* and Vibrational Properties of Disordered Solids edited by R. A. Ramos (World Scientific, New Yersey, USA, 2022); preprint at arxiv:2009.05970.

PREDICTING SCALING PROPERTIES FROM A SINGLE CONFIGURATION

T. B. Schrøder

Glass & Time, Roskilde University, Denmark.

E-mail: tbs@ruc.dk

Time-dependent dynamical properties of a liquid cannot be estimated directly from a single configuration without performing a simulation. Here, however, we present a method that predicts the scaling properties of both structure and dynamics from a single configuration. The method is demonstrated to work well for the Lennard–Jones fluid, and the viscous Kob–Andersen Lennard–Jones mixture, both in and out of equilibrium [1], as well as for small stiff molecules [2]. These systems have isomorphs, which are curves in the high–density phase diagram along which the dynamics, structure, and excess entropy are all invariant in reduced units. In this context, the new method is a method to identify isomorphs, which is here compared to existing methods.

Finally, we test the new method on molecular models with flexible bonds, which do *not* have isomorphs; Instead, they have curves in the phase diagram with invariant reduced-unit dynamics, and structure, but *not* invariant excess entropy. These curves, termed pseudo-isomorphs, are not described by the isomorph theory, and have until now been complicated to identify [3].

[1] T.B. Schrøder, Phys. Rev. Lett. 129, 245501 (2022).

[2] Z. Sheydaafar, J.C. Dyre, and T.B. Schrøder, arXiv:2105.13736.

[3] A.E. Olsen, J.C. Dyre, & T.B. Schrøder, J. Chem. Phys. 145, 241103 (2016).

EXPLORING THE DENSIFICATION EFFECT ON DYNAMICS OF METALLIC GLASS-FORMING LIQUID

J. Shen, A. Cornet, A. Ronca, B. Ruta

Université Claude Bernard Lyon, CNRS, Institut Lumière Matière, F-69622, Lyon, France. European Synchrotron Radiation Facility, 71 Avenue des Martyrs, CS 40220, F-38043, Grenoble, France

E-mail: jie.shen@univ-lyon1.fr

For supercooled liquids, both the cooling and densification can slow down their dynamics and ultimately trigger the glass transition according to the jamming phase diagram. Differently from the cooling, the effect of isothermal densification on glass-forming liquids dynamics is far less studied. We explored the effects of ex-situ densification performed around the glass-transition temperature (T_g) on $Zr_{46.8}Ti_{8.2}Cu_{7.5}Ni_{10}Be_{27.5}$ metallic glasses by the combinations of fast-calorimetric and X-ray phonon correlation spectroscopy (XPCS) methods. Densification in the equilibrium liquid (i.e., $T_g + 30$ K) induces an enhancement of T_g by 3 - 5K/GPa, which is derived by an enthalpy-recovery method. The kinetics of enthalpy relaxation is suppressed if the glass is densified below T_g (i.e., $T_g - 30$ K), leading instead to a sub- T_g enthalpy-recovery peak. Differently, XPCS reveals that pressure accelerates the atomic mobility of the glass by two orders of magnitude as well as makes its dynamics more localized and stress-dominant. This result highlights the difference between kinetics of enthalpy relaxation and atomic motions in this glass under *ex-situ* hydrostatic compression, where the former is slowed down and the latter is rejuvenated.

Revealing the mechanism of high fragility in polymers

A. P. Sokolov

University of Tennessee, Knoxville, TN 37996, USA. Oak Ridge National Laboratory, Oak Ridge, TN, USA.

E-mail: sokolov@utk.edu

If fragility of molecular liquids is usually limited to $m \sim 80 - 100$, many polymers exhibit fragility $m \sim 150$ or even higher. The mechanism of this unusually steep temperature dependence of structural relaxation in polymers still remains a puzzle [1]. To reveal additional hints for understanding the underlying mechanism, we analyzed molecular weight (MW) dependence of chain and segmental dynamics in several polymers. While fragility of segmental dynamics in many polymers increases sharply with MW, fragility of chain dynamics appears to be essentially independent of MW and comparable to fragility of small molecular liquids [2,3]. Our surprising discovery is that the temperature dependence of the chain relaxation follows well the Adam-Gibbs relationship, which fails in the case of segmental relaxation [2]. Moreover, many correlations that fail for segmental dynamics fragility work well for chain fragility [2]. These results suggest that the molecular level relaxation (chain relaxation) still follows the behavior usual for small molecules even in polymers. It is the segmental relaxation that has the unusually high fragility. We suggest that many polymers cannot reach an ergodic state on the time scale of structural relaxation and this leads to a steeper temperature dependence of segmental relaxation.

- [1] A.P. Sokolov et al., J. Phys.Cond.Matt. 19, 205116 (2007).
- [2] C. Dalle-Ferrier et al., J. Chem. Phys. 145, 154901 (2016).
- [3] A.L. Agapov et al., Macromolecules 51, 4874 (2018).

SUPERCOOLED LIQUID TELLURIUM: WATER'S DISTANT RELATIVE?

P. Sun¹, G. Monaco¹, P. Zalden², K. Sokolowski-Tinten³, J. Antonowicz⁴, R. Sobierajski⁵, Y. Kajihara⁶, A.Q.R. Baron⁷, P. Fuoss⁸, A. Chihpin Chuang⁹, J.–S. Park⁹, J. Almer⁹, J.B. Hastings⁸

¹Dipartimento di Fisica e Astronomia "Galileo Galilei", Università degli Studi di Padova, Via F. Marzolo, 8, 35131 Padova, Italy.

²European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany.

³Faculty of Physics and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, Lotharstrasse 1, 47048 Duisburg, Germany.

⁴Faculty of Physics, Warsaw University of Technology, Koszykowa 75, Warsaw, 00-662, Poland.

⁵Institute of Physics of the Polish Academy of Sciences, PL-02-668 Warsaw, Poland.

⁶Graduate School of Advanced Science and Engineering, Hiroshima University, Higashi-Hiroshima, Hiroshima 739-8521, Japan.

⁷Materials Dynamics Laboratory, RIKEN SPring-8 Center, 1–1–1 Kouto, Sayo, Hyogo 679–5148, Japan. ⁸SLAC National Accelerator Laboratory, 2575 Sand Hill Rd, Menlo Park, CA 94025, USA.

⁹X-ray Science Division, Advanced Photon Source, Argonne National Laboratory, Lemont, Illinois 60439, USA.

E-mail: peihao.sun@unipd.it

Traditionally, liquids are modeled as disordered systems with a given short-range order. However, lately there has been accumulating evidence which challenges this view. The most famous example is water, known to exhibit a multitude of thermodynamic anomalies earning it the name "the most anomalous liquid". Recent studies [1,2] have provided strong support for the existence of two forms of liquid water – a phenomenon generally known as "liquid polymorphism" – as the mechanism behind the anomalies. Meanwhile, thermodynamic anomalies and evidence for liquid polymorphism have been found in several other systems, including liquid tellurium (Te) [3] where experimental input is relatively lacking.

In our work [4], we performed simultaneous small- and wide-angle X-ray scattering measurements on liquid Te supercooled to more than 130 K below the melting point. We observe clear maxima in its thermodynamic response functions around 615 K, suggesting the possible existence of liquid polymorphism. The wide-angle scattering data further allows us to uncover the underlying structural evolution, revealing a development of intermediate-range order upon cooling, which we attribute to bond-orientational ordering. Comparing our results to those of water, striking similarities can be found despite the lack of hydrogen-bonding and tetrahedrality in Te. Thus, our results indicate that water-like anomalies may be a general phenomenon among liquid systems with competing bond- and density-ordering.

[1] K.H. Kim et al., Science 358, 1589 (2017).

[2] P.G. Debenedetti, F. Sciortino, and G.H. Zerze, Science 369, 289 (2020).

[3] A. Angell, Nat. Nanotechnol. 2, 396 (2007).

[4] P. Sun et al., Proc. Natl. Acad. Sci. U.S.A. 119, e2202044119 (2022).

LOW-TEMPERATURE THERMAL ANOMALIES IN ORDERED AND DISORDERED CRYSTALLINE PHASES

J.F. Gebbia¹, C. Escorihuela-Sayacedo¹, C. Cazorla¹, A. Krivchikov^{2,3}, J.L. Tamarit¹

¹Grup de Caracterizació de Materials, Departament de Fisica, EEBE, and Barcelona Research Center in Multiscale Science and Engineering, Universitat Politècnica de Catalunya, 08019 Barcelona, Catalonia.

²Institute of Low Temperature and Structure Research, Polish Academy of Sciences, 50–422 Wrocław.

³Verkin Institute for Low Temperature Physics and Engineering of the National Academy of Sciences of Ukraine,

Kharkiv 61103, Ukraine.

E-mail: josep.lluis.tamarit@upc.edu

Before the seminal works by Zeller and Pohl [1] and Anderson *et al.* [2], it was expected that at low temperatures amorphous or glassy solids (*i.e.*, aperiodic and vastly structurally degenerated although still macroscopically crystalline over extended timescales) should behave similarly to their ordered counterparts. By assuming that long wavelength excitations dominate the vibrational landscape, it follows from the Debye theory that the density of states $g(\omega)$ behaves like $g_D(\omega^2)$ and, accordingly, at low temperatures ($k_BT < h\omega$) the heat capacity like $c_D(T) = C_DT^2$, where $C_D \propto \theta_D^{-3}$ and θ_D is the Debye temperature.

However, experimental works [1,2] made soon evident that the low-temperature heat capacity and also thermal conductivity of glassy solids strongly differed from those of standard solids (thus, were called *"anomalous"*). In particular, the existence of a linear temperature dependence for the specific heat at ultra-low temperatures was evidenced for several non-metallic non-crystalline crystals along with the appearance of a "bump" in the $c_v(T)/T^3$ representation, called *boson peak*. This *boson peak* departs from the heat capacity deduced from the Debye model and manifests in a broad maximum in the reduced vibrational density of states, $g(\omega)/\omega^2$, obtained at low frequencies, as experimentally evidenced by Buchenau *et al* [3].

Our communication will present results on the low-temperature heat capacity of several materials displaying the anomalous thermal properties described above [4–7]. The degree of disorder in the selected materials can be arbitrarily reduced, all the way down to the fully ordered system, while they still display the aforementioned thermal anomalies.

- [1] R.C. Zeller and R.O. Pohl, Phys. Rev. B 4, 2029-2041 (1971).
- [2] P.W. Anderson, B.I. Halperin and C.M. Varma, Philos. Mag. 25, 1-9 (1972).
- [3] U. Buchenau, N. Nucker, and A.J. Dianoux, Phys. Rev. B 53, 2316-2319 (1984).
- [4] A.I. Krivchikov et al., J. Phys. Chem. Lett. 13, 5061-5067 (2022).
- [5] Y. Miyazaki et al., J. Phys. Chem. Lett. 12, 2112-2117 (2021).
- [6] M. Moratalla et al., Phys. Rev. B 99, 024301 (2019).
- [7] D. Szewczyk et al., Sci. Rep. 11, 18640 (2021).

INVESTIGATION OF THE MECHANISM OF GELLAN AGGREGATION AT THE MOLECULAR SCALE

L. Tavagnacco

CNR-ISC and Department of Physics, Sapienza University of Rome, Piazzale A. Moro 2, 00185, Rome, Italy.

E-mail: letizia.tavagnacco@roma1.infn.it

Among hydrocolloids, gellan is one of the most studied polysaccharides because of its ability to form mechanically stable gels with several technological bio-applications in medicine, pharmaceutical and cosmetic industries. More recently it was also demonstrated that microgels made of gellan are promising materials to be used for paper preservation to restore both ancient and modern paper artworks [1].

Understanding the mechanism of gelation of gellan and the interactions occurring at the molecular level is thus of fundamental importance to allow the adjustment of the processes of gel formation to the specific application requirements. In this talk, I will report on a molecular investigation of the mechanism of aggregation of gellan carried out using atomistic molecular dynamics simulations based on a newly developed gellan force field which is capable to detect both the coil to single helix transition at dilute conditions [2] and the formation of higher-order aggregates at high concentration. By analyzing the solution behavior of a suspension of gellan chains in aqueous solution as a function of the polymer concentration I will provide evidence of the occurrence of a two-step aggregation process [3]. Moreover, I will compare the solution behavior of gellan chains in the presence of monovalent and divalent cations, showing a different capability of solution structuring, in qualitative agreement with atomic force microscopy and rheological measurements.

[1] B. Di Napoli et al., ACS Appl. Polym. Mater 2, 2791-2801 (2020).

[2] M. Diener et al., ACS Macro Lett. 9, 115–121 (2020).

[3] L. Tavagnacco et al., arXiv:2209.08975v1.

DATA-DRIVEN MODELING OF INTRINSICALLY DISORDERED PROTEINS: PHASE SEPARATION AND PROTEOME-WIDE ANALYSIS

G. Tesei¹, A. I. Trolle¹, T. S. Schulze¹, N. Jonsson¹, K. E. Johansson¹, R. Crehuet^{1,2}, K. Lindorff-Larsen¹

¹Structural Biology and NMR Laboratory, Linderstrøm-Lang Centre for Protein Science, Department of Biology, University of Copenhagen, DK-2200 Copenhagen, Denmark.

²CSIC-Institute for Advanced Chemistry of Catalonia (IQAC), E-08034 Barcelona, Spain.

E-mail: giulio.tesei@bio.ku.dk

Many intrinsically disordered regions (IDRs) participate in the formation of biomolecular condensates through a reversible phase separation process. While condensates play important biological roles, the dysregulation of their formation and dissolution may lead to pathogenic insoluble aggregates associated with neurodegenerative diseases and cancer. To gain insight into the sequence-dependence of condensate formation, we developed CALVADOS (Coarsegraining Approach to Liquid-liquid phase separation Via an Automated Data-driven Optimization Scheme) [1,2], a residue-level model for molecular dynamics simulations of IDRs. We based our model development on an automated Bayesian procedure which trains the aminoacid-specific parameters of the model against experimental data reporting on single-chain conformational properties. The training procedure results in significantly improved predictions of the self-association and condensate formation of diverse and unrelated protein sequences. We further made use of the computational efficiency and accuracy of the CALVADOS model to simulate all the IDRs of the human proteome. Based on the confidence score assigned in AlphaFold2 structure predictions of the full-length proteins, we identified 29,998 IDRs ranging in length between 30 and 500 residues. We characterized the conformational ensemble of each sequence and related chain compaction to gene ontology annotations and clinical data.

[1] G. Tesei et al., Proc. Natl. Acad. Sci. U.S.A. 118, e2111696118 (2021).

[2] G. Tesei and K. Lindorff-Larsen, Open. Res. Europe 2, 94 (2023).

Colloidal crystals and gels from active apolar colloids

H. Massana-Cid¹, J. Codina¹, I. Pagonabarraga^{1,3}, P. Tierno^{1,2,4}

¹Departament de Física de la Matèria Condensada, Universitat de Barcelona.

²Universitat de Barcelona Institute of Complex Systems, Universitat de Barcelona.

³Centre Européen de Calcul Atomique et Moléculaire, École Polytechnique Fédérale de Lausanne, 1015 Lausanne,

Switzerland.

⁴Institut de Nanociéncia i Nanotecnologia, Universitat de Barcelona.

E-mail: ptierno@ub.edu

Artificial active particles are autonomous agents able to convert energy from the environment into net propulsion, breaking detailed balance and the action—reaction law, clear signatures of their out-of-equilibrium nature. In this talk I will start illustrating how clusters composed of passive and catalytically active apolar colloids may undergo directed motion using a lightinduced chemophoretic flow [1]. Further, I will show that routes toward the formation of two dimensional clusters and gels may be uncovered by using a few active apolar colloidal dopants that are dispersed in a population of inert passive particles. The activity and interactions between the synthetic dopants are controlled by light, and the absence of self-propulsion in the gel makes them ideal dockers for the formation of stable and strong bonds that chemophoretically glue different inert particles, creating solid, space-filling networks [2]. This contrasts with previous works based on active polar, i.e., self-propelling, units that were activated by external fields or chemical reactions.

[1] J. Codina et al., Soft Matter 18, 5371-5379 (2022).

[2] H. Massana-Cid et al., Proc. Nat. Acad. Sci. USA 115, 10618-10623 (2018).

Aggregation of fullerenes $C_{\rm 60}$ in liquids: investigations and theoretical description

T. V. Tropin¹, N. Jargalan², P. A. Selyshchev³, M. V. Avdeev⁴, V. L. Aksenov⁵

¹Frank Laboratory of Neutron Physics, Joint Institute for Nuclear Research, Dubna, Russia.
²Department Material Science, Institute of Physics and Technology, Ulaanbaatar, Mongolia.
³Department of Physics, University of Pretoria, Pretoria, South Africa.

E-mail: ttv@jinr.ru

Unlike classic carbon allotropes, graphite and diamond, fullerenes may dissolve in different liquids [1]. Their solutions often bear colloidal character, may serve as initial samples for making complex systems or are considered complex themselves. Depending on solvents polarity, a number of interesting properties and effects is observed in these systems [1,2].

In present report we review our recent theoretical and experimental investigations of fullerenes aggregation in different solutions. For low-polar liquids, the kinetics of aggregation of C_{60} in solutions in toluene, benzene and chlorobenzene were measured by the dynamic light scattering (DLS). It is proposed, that clusters start growing around fullerene oxides that are formed by photooxidation of C_{60} . For the polar liquids, on the other hand, cluster growth is supposed to be connected with formation of charge-transfer complexes between fullerene and solvent molecules. Here various experimental data is considered for C_{60}/N -methyl-pyrrolidone (NMP) solution, including DLS, UV-Vis absorption and SAXS measurements [3]. The most recent experiments cover the aggregation that follows the extraction of fullerene molecules from C_{60}/h exane solution to pure NMP. This way one obtains a system were aggregation proceeds together with an input flux of monomers to the "sealed" volume of polar solvent. The obtained results are evaluated by utilizing a set of kinetic equations for clusters formation and growth.

- [2] S. Nath, H. Pal, and A. Sapre, Chem. Phys. Lett. 327, 143 (2000).
- [3] T. Tropin et al., Ukr. J. Phys. 65, 701 (2020).

^[1] M. Avdeev, et al., J. Phys. Chem. A 84, 1273 (2010).

BIO-INSPIRED TOPOLOGICAL SUPRAMOLECULAR MATERIALS: FROM CIRCULAR CATENANES TO KDNA

L. Tubiana¹, E. Orlandini², F. Ferrari³, D. Michieletto⁴, C. Dellago⁵

¹University of Trento, Trento, Italy.
 ²University of Padova, Padova Italy.
 ³University of Szcezcin, Szcezcin, Poland.
 ⁴University of Edinburgh, Edinburgh, United Kingdom.
 ⁵University of Vienna, Vienna, Austria.

E-mail: luca.tubiana@unitn.it

Supramolecular materials built of topologically interlocked polymer rings have recently gained considerable interest in supramolecular chemistry, biology, and soft matter. Two typical examples are polycatenanes, linear chains of concatenated rings, and the kDNA, a natural occurring, two-dimensional surface of linked DNA rings. Here I present the results of several ongoing collaborations, all highlighting the role of topological interactions in shaping the physical properties of supramolecular objects and how one can exploit them to tune the behavior of such materials. We will show that a relation Twist + Writhe = constant, typical of dsDNA rings, holds for circular polycatenanes, and that a similar effect holds for 2D sheets of rings. Finally, we will report experimental results on the kDNA and show that its typical shower-cap shape in solution can be explained by the topological interaction between the rings on its border. Our results suggest that supramolecular topological objects can form a new category of highly designable structures with potential applications in supramolecular chemistry and material science.

MICROSCOPIC ORIGIN OF RESISTANCE FLUCTUATION IN GERMANIUM TELLURIDE GLASS

S. Walfort, H. Treppke, N. Holle, M. Salinga

University of Münster, Institute of Materials Physics, Wilhelm-Klemm-Strße 10, 48149 Münster, Germany.

E-mail: sebastian.walfort@uni-muenster.de

Germanium telluride (GeTe) is a fragile glass former with a large electrical property contrast between a conductive crystalline phase and highly resistive glass states. It is further possible to repeatedly switch a small, confined volume between crystalline and amorphous on nanosecond timescales, which makes GeTe an interesting material for electronic memory applications [1], [2] and as a model system for observing glassy dynamics. For instance, physical aging of GeTe glass seems to manifest in a continued evolution of the electronic properties, e.g. in a power-law-like increase of the resistivity [3]. In this experimental study, resistance serves as the observable to probe the structural dynamics of the glass. We demonstrate that, as a consequence of reducing the nanoscopic amorphous volume, individual resistance states can be resolved in time. The fluctuations between these states are measured across a broad temperature range, six orders of magnitude in time and modeled as a (hidden) Markov process. The resulting attempt frequencies of the structural dynamics range from the typical phonon timescales of terahertz, down to few kilohertz, which would point towards collective structural events involving a large number of atoms. Beyond their relevance for application, these results illustrate the feasibility of the experimental approach for probing the dynamics of a nanoscopic glass volume through a fluctuating resistance.

- [1] Z. Wang, H. Wu, and G. Burr, Nat. Rev. Mater. 5, 173–195 (2020).
- [2] D. Lencer, M. Salinga, and M. Wuttig, Adv. Mater. 23, 2030–2058 (2011).
- [3] F. Zipoli, D. Krebs, and A. Curioni, Phys. Rev. B 93, 115201 (2016).

PROBING THE STRUCTURE AND RELAXATION IN THE "NO-MAN'S LAND" OF PHASE-CHANGE MATERIALS

S. Wei

Department of Chemistry, Aarhus University, Aarhus, 8000, Denmark.

E-mail: shuai.wei@chem.au.dk

Theories suggested that a liquid-liquid critical point (LLCP) may exist at the end of a liquidliquid transition line that separates two distinct liquid phases in some substances [1]. The most discussed example is water that exhibits a range of anomalous behaviors possibly originating from a LLCP [2,3]. However, the challenge is that the liquid anomalies might lie in a metastable supercooled liquid state, often obscured by fast crystallization. This establishes a "no-man's land", in which experimental observation is difficult. Here we show a striking parallel of the thermodynamic anomalies between liquid phase-change materials (PCMs) (e.g. Ge-Sb-Te) and supercooled water [4,5]. Using X-ray free electron lasers, we can access the liquid structure of no-man's land with the femtosecond-resolution and demonstrate the evidence of a continuous liquid-liquid transition [6]. The latter is not only a high- to low-density transition, but also associated with a metal-to-semiconductor transition [7] with a drastic change in viscosity (a fragile-strong transition) [8]. While mixing water with a second component often diminishes its anomalies, the anomalies of PCMs bear a large tunability through altering the compositions, offering new opportunities for exploring a possible LLCP in a wide temperature-pressure space. At last, the liquid anomalies have nontrivial effect on crystallization, which is relevant to the functionality of PCMs in the application of non-volatile memory devices.

- [1] H. Tanaka, J. Chem. Phys. 153, 130901 (2020).
- [2] P.G. Debenedetti, F. Sciortino, and G.H. Zerze, Science, 369, 289 (2020).
- [3] K.H. Kim et al., Science 370, 978 (2020).
- [4] S. Wei, P. Lucas, and C.A. Angell, MRS Bulletin 44, 691 (2019).
- [5] S. Wei, J. Non-Cryst. Sol. X 14, 100094 (2022).
- [6] P. Zalden et al., Science 364, 1062 (2019).
- [7] S. Wei et al., Phys. Rev. Appl. 7, 034035 (2017).
- [8] S. Wei, P. Lucas, and C. A. Angell, J. Appl. Phys. 118, 034903 (2015).
Absence of cooperativity near the glass transition

M. Pica Ciamarra^{1,2}, W. Ji^{3,4}, M. Wyart⁵

¹Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, 21 Nanyang Link, 637371, Singapore.

²Consiglio Nazionale delle Ricerce, CNR-SPIN, Napoli, I-80126, Italy.

³Department of Physics of Complex Systems, Weizmann Institute of Science, Rehovot, 234 Hertzl St., Israel.

⁴John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138, USA.

⁵Institute of Physics, École Polytechnique Fédérale de Lausanne, Lausanne, CH-1015, Switzerland.

E-mail: matthieu.wyart@epfl.ch

Under cooling, a supercooled liquid undergoes a glass transition and stops flowing. Physicists do not agree on the microscopic reasons that make a glass solid. Some view this phenomenon as being collective in nature: it may be a signature of a thermodynamic phase transition, or being caused by kinetic constraints (where particles seek to solve a sort of Chinese puzzle). Others view it as simply reflecting elementary barriers for rearrangements, controlled by the elasticity of the material. Here I will focus on polydisperse numerical glasses, which are receiving a considerable attention, because they can be equilibrated as efficiently as experimental molecular liquids. I will introduce a novel algorithm to systematically extract elementary rearrangements in a broad energy range. It allows to make, for the first time, a quantitative prediction on the relaxation time, assuming that relaxation is not collective in nature. The comparison with observation is very good. I will explain why, in such a scenario, dynamical correlations emerge even if they have little effects on the dynamics. I will discuss the nature of elementary rearrangements under cooling, and how these aspects relate to mean field description of glasses.

The ESRF Extremely Brilliant Source for X-ray photon correlation spectroscopy at the beamline ID10

F. Zontone¹, Y. Chushkin¹, B. Ruta²

¹ESRF – The European Synchrotron, 71 avenue des Martyrs, 38043 Grenoble, France. ²Institut Lumière Matière (ILM), UMR5306 Université Lyon 1–CNRS, Université de Lyon 69622 Villeurbanne, France

E-mail: zontone@esrf.fr

The Extremely Brilliant Source (EBS) at the European Synchrotron Radiation facility (ESRF) is the first high-energy storage ring for synchrotron radiation hard X-rays based on the new hybrid multi-bend achromat concept, allowing a huge increase of the source brilliance [1]. As the coherent fraction of the beam is proportional to the brilliance, a large coherent flux is expected even at high energies above 20 keV with sufficient contrast, allowing experiments with coherent X-ray beams that were not possible so far. The technique benefitting the most from EBS is certainly X-ray Photon Correlation Spectroscopy (XPCS), thanks to the quadratic dependence of the accessible time scales on the brilliance gain. The ESRF beamline ID10 has pioneered and applied successfully XPCS at wide angles (WA-XPCS) to probe structural relaxations in molecular glasses and supercooled liquids [2,3,4]. We show how the coherent beam can be efficiently tailored for WA-XPCS for high signal-to-noise ratio measurements at unprecedented short times scales thanks to the EBS, the upgrade of the focusing optics and modern detectors. The new possibilities for XPCS at ID10 will be shown on an interesting "*in operando*" application on hydrogenated metallic glasses.

- [1] P. Raimondi et al., Comm. Phys., submitted.
- [2] B. Ruta et al., Phys. Rev. Lett. 109, 165701 (2012).
- [3] N. Neuber et al., Comm. Phys. 5, 1-10 (2022).
- [4] A. Cornet et al., arXiv:2301.02551.

Part III Poster

DISMANTLING SOME COMMON BELIEVES ON THE DEBYE RELAXATION OF MONOALCOHOLS

S. Arrese-Igor¹, A. Alegria^{1,2}, J. Colmenero^{1,2,3}

¹Centro de Física de Materiales (MPC), Centro Mixto CSIC-UPV/EHU, Paseo Manuel de Lardizabal 5, 20018 San Sebastián, Spain.

²Departamento de Polímeros y Materiales Avanzados UPV/EHU, Apartado 1072, 20080 San Sebastián, Spain. ³Donostia International Physics Center, Paseo Manuel Lardizabal 4, 20018 San Sebastián, Spain.

E-mail: silvia.arreseigor@ehu.eus

The "Debye" relaxation of monoalcohols has attracted the attention of many researchers for decades. In this poster we want to draw the attention to some aspects that are often obviated and/or rarely mentioned in the literature. There are some 'common believes' –as the exponential character of the mentioned slow process or a proportional relation between its strength and the timescale separation relative to the alpha relaxation– which are frequently just assumed during literature data analysis. Although the qualitative picture may not change radically, these assumptions may have important implications in detailed quantitative determination of Kirkwood correlation factor or alpha relaxation shape and characteristic times. Moreover, apart from quantitative determinations, we should not overlook such behaviour, as understanding the reasons under these deviations (when and how much) could be key for understanding unsolved aspects of the dynamics of monoalcohols.

Exploitation of the unique capabilities of an ultrafast nanocalorimeter at a synchrotron coherence beamline

J. Baglioni

Dipartimento di Fisica e Astronomia "Galileo Galilei", Università degli Studi di Padova, 35131 Padova, Italy.

E-mail: jacopo.baglioni@phd.unipd.it

The ultra-high heating/cooling rates achievable with fast nano-calorimeters allow one to study the amorphous phase of countless materials. We show that a commercial nano-calorimeter can be installed in a synchrotron beamline, enabling real-time and online calorimetric analysis of the samples investigated with X-rays. In particular, we have installed a Mettler-Toledo nano-calorimeter (Flash DSC 2) at the coherence beamline (P10) of Petra3 (DESY), and we demonstrate that XPCS measurements can still be performed on samples mounted on the calorimeter chip sensor. The sensor gives a negligible contribution in terms of scattered intensity even for low Z materials. As an example, we show that the beam induced dynamics – previously observed on amorphous LiBO₂ thin disks – can also be measured on samples melted on the chip. The glass can be guenched at a controlled cooling rate, and its either dynamic or thermodynamic properties can be studied without ever removing the sample from the beamline. Furthermore, the calorimeter is usable as a powerful and precise furnace. Temperatures up to 1000 K, with an accuracy well below 1 K, can be reached in a few seconds. As an example, we have measured the alpha relaxation of some metallic glasses controlling the temperature with the chip sensor: the calculated decorrelation times as a function of T are consistent with the results reported in literature.

Combining X-ray Diffraction and X-ray Raman Spectroscopy for the investigation of irradiated shock-compressed silica glass samples

L. Bussmann^{1,3}, T. Schlothauer^{2,3}, G. Heide^{2,3}, C. Sternemann⁴, S. Fuhrmann^{1,3}

¹TU Bergakademie Freiberg, Institute of Glass Science and Technology. ²TU Bergakademie Freiberg, Institute of Mineralogy. ³TU Bergakademie Freiberg, Freiberg High Pressure Center (FHP). ⁴Technische Universität Dortmund, Fakultät Physik/DELTA.

E-mail: lena.bussmann@igt.tu-freiberg.de

The university of Freiberg is one of the few institutions with direct access to an underground mine. The shock wave laboratory (SWL), installed in 2006, is used for material synthesis and other experiments. Silica glass is a suitable material for experimenting on impact processes, since the pressure-related properties of silica glass have been vastly investigated and are fairly well known. At the same time, many questions remain unsolved, and silica glass is still pertinent and omnipresent in several disciplines such as geosciences or material sciences. A set of silica glass powder samples have been shock-compressed in the SWL, and investigated at the P01 beamline of PETRA III at DESY, using a combination of X-ray Raman spectroscopy (XRS) and X-ray diffraction (XRD). Studying the Si L-edge via XRS allows to classify the effects of shock-compression on the electronic short-range structure, while the position of the amorphous hump in XRD patterns indicates intermediate-range correlations. Both methods combined allow to analyse the samples in terms of the resulting shock pressure achieved in the SWL. Time-resolved recording of XRD patterns furthermore revealed a structural instability aqainst X-ray irradiation leading to a "relaxation" process in the shock-compressed samples. This process is characterized by collating the complementary XRD and XRS results and comparing the dynamics of the changing network on different order scales.

Fast differential scanning calorimetry: advancements in data treatment to investigate organic molecular glass-formers

S. Capaccioli^{1,2}, Daniele Sonaglioni^{1,3}, E. Tombari⁴

¹Dipartimento di Fisica "E. Fermi", Università di Pisa, Largo Pontecorvo 3, 56127, Pisa, Italy.

²CISUP, Centro per l'Integrazione della Strumentazione dell'Università di Pisa, Lungarno Pacinotti 43, 56126, Pisa, Italu.

³Center for Materials Interfaces-Electron Crystallography, Istituto Italiano di Tecnologia, Viale Rinaldo Piaggio 34, 56025, Pontedera (Italy).

⁴Istituto per i Processi Chimico-Fisici del CNR, via G. Moruzzi 1, 56124 Pisa, Italy.

E-mail: simone.capaccioli@unipi.it

Fast differential scanning calorimetry is a commercial technique very appreciated for its capability of suppressing reorganization processes, thanks to its wide interval of scanning rates, several orders higher than that of conventional scanning calorimeters [1,2]. Nevertheless, difficult experimental aspect must be solved: the evaluation of the dynamic thermal lag under non optimal conditions, as for organic molecular materials; the application of reliable models to rescale the specific heat to that from conventional calorimetry; the use of a feasible way to reveal static thermal gradients within the sample causing smearing effects.

In a recent paper [3] we have provided the following advancements: i) a novel way to estimate the dynamical thermal lag by using the temperatures of maximum slope during cooling and heating through the glass transition; ii) a novel interpretation of the heat flow losses affected by the sample and depending on the scanning rate sign; iii) the use of the glass to liquid transition measured on heating to reveal static thermal gradients. These novel approaches have been here tested on three prototypical organic glass-forming materials, i.e. orto-terphenyl, glycerol and poly(propylene glycol).

[1] C. Schick and V. Mathot, *Fast Scanning Calorimetry of Organic Materials from Low Molecular Mass Materials to Polymers*,(Springer Nature, 2016).

- [2] G.W.H. Höhne, W.F. Hemminger, and H.-J. Flammersheim, Differential Scanning Calorimetry (Springer Berlin, 1996).
- [3] D. Sonaglioni, E. Tombari, and S. Capaccioli, Thermochim. Acta 719, 179385 (2022).

Micro-eV Vibrational Dynamics in amorphous SiO₂: evidences of additional modes?

F. Caporaletti¹, D. Bessas², A.I. Chumakov², M. Zanatta³, G. Monaco⁴, G. Baldi³

¹Laboratory of Polymer and Soft Matter Dynamics, Experimental Soft Matter and Thermal Physics (EST), Université

libre de Bruxelles (ULB), Brussels 1050, Belgium.

²ESRF-The European Synchrotron, Grenoble, France. ³Dipartimento di Fisica, Università di Trento.

⁴Dipartimento di Fisica ed Astronomia, Università di Padova, Italy.

E-mail: federico.caporaletti@ulb.be

The low-temperature vibrational properties of glasses are known to be distinctively different from those of crystals as a result of their disorder. Examples of this anomalous behavior are long-wavelength phonons being scattered from elastic disorder [1,2] or the possible presence of quasi-localized soft modes [3,4]. To experimentally probe these features it is required to access the vibrational properties of glasses at very low frequencies (< 1 meV), which is experimentally challenging.

We here report the measurement of the low-frequency tail of the vibrational density of states (DOS) of the prototypical glass of silica using a recently developed μ eV hard X-rays spectrograph [5]. This new experimental setup allowed us to investigate the vibrational properties of SiO₂ with an energy resolution of 170 μ eV at 300 and 150 K. We observed that at both temperatures the DOS does not reach the Debye level down to 300 μ eV.

Further, by exploiting the temperature dependence of the low-energy tails of the DOS, we could disentangle the non-vibrational contributions, coming from additional relaxation modes and anharmonicity, and extrapolate the purely vibrational DOS. Our data suggests that additional vibrational modes accumulate above sound waves in the low frequency tail of the DOS, if we assume that transverse and longitudinally phonons are equally attenuated, as suggested by MD simulations.

[1] W. Schirmacher, G. Ruocco, and T. Scopigno, Phys. Rev. Lett. 98, 025501 (2007).

[2] G. Baldi et al., Phys. Rev. Lett. 104, 195501 (2010).

[3] H. Mizuno et al., Proc. Natl. Acad. Sci. U.S.A. 114, E9767 (2017).

[4] C. Rainone et al., Proc. Natl. Acad. Sci. U.S.A. 117, E5228 (2020).

[5] A.I. Chumakov et al., Phys. Rev. Lett. 123, 097402 (2019).

Thermoresponsivity of poly(N-isopropylacrylamide) microgels in water-trehalose solution and its relation to protein behaviour

S. Corezzi¹, B. P. Rosi¹, L. Tavagnacco^{2,3}, L. Comez⁴, P. Sassi⁵, M. Ricci⁵, E. Buratti^{2,3}, M. Bertoldo^{6,7}, C. Petrillo¹, E. Zaccarelli^{2,3}, E. Chiessi⁸

> ¹Dipartimento di Fisica e Geologia, Università di Perugia, I-06123 Perugia, Italy. ²CNR-ISC, Sapienza Università di Roma, I-00185 Roma, Italy.

³Dipartimento di Fisica, Sapienza Università di Roma, I-00185 Roma, Italy.

⁴CNR-IOM, Dipartimento di Fisica e Geologia, Università di Perugia, I-06123 Perugia, Italy.

⁵Dipartimento di Chimica, Biologia e Biotecnologie, Università di Perugia, I-06123 Perugia, Italy.

⁶Dipartimento di Scienze Chimiche, Farmaceutiche ed Agrarie, Università di Ferrara, I-44121 Ferrara, Italy. ⁷CNR-ISOF. Area della Ricerca. I-40129 Bologna. Italu.

⁸Dipartimento di Scienze e Tecnologie Chimiche, Università di Roma "Tor Vergata", I-00133 Roma, Italy.

E-mail: silvia.corezzi@unipg.it

Additives are commonly used to tune macromolecular conformational transitions. Among additives, trehalose is an excellent bioprotectant and among responsive polymers, PNIPAM is the most studied material. Nevertheless, their interaction mechanism so far has only been hinted without direct investigation, and, crucially, never elucidated in comparison to proteins. Here [1] the effect of trehalose on the swelling behavior of PNIPAM microgels is monitored by dynamic light scattering, while Raman spectroscopy and molecular dynamics simulations are used to explore changes of solvation and dynamics across the swelling-deswelling transition at the molecular scale. We find that strongly hydrated trehalose molecules develop water-mediated interactions with PNIPAM microgels, thereby preserving polymer hydration below and above the transition while drastically inhibiting local motions of the polymer and of its hydration shell. The study, for the first time, demonstrates that slowdown of dynamics and preferential exclusion are the principal mechanisms governing trehalose effect on PNIPAM microgels, at odds with preferential adsorption of alcohols, but in full analogy with the behavior observed in trehalose-protein systems. The new insights help understand to what extent PNIPAM microgels can effectively be used as synthetic biomimetic materials, to reproduce and study, at the colloidal scale, isolated protein behavior and its sensitivity to interactions with specific cosolvents or cosolutes.

[1] B.P. Rosi et al., J. Colloid Interface Sci. 604, 705-718 (2021).

D4PDF: FROM CONSTANT-WAVELENGTH TOTAL NEUTRON SCATTERING DATA TO THE PAIR DISTRIBUTION FUNCTION

G. J. Cuello¹, J. Robledo², J. Dawidowski³

¹Institut Laue Langevin, Grenoble, France.
 ²Forschungszentrum Juelich GmbH, Juelich, Germany.
 ³National Atomic Energy Commission and CONICET, Bariloche, Argentina.

E-mail: cuello@ill.fr

In total scattering experiments the detectors register not only the elastic scattering but also the diffuse signal. The latter contains information on the local order, which could be different from the average structure explored by the Bragg scattering. The Fourier transform of this total scattering is the pair distribution function (PDF), which represents the probability of finding two atoms apart from each other at a given distance and allows to get correlation distances and coordination numbers in disordered systems (like liquids, glasses, quasi-crystals, nanoparticles, etc.). Neutrons and X-rays are the commonly used probe particles for this kind of studies, although electrons are recently used too. Data processing must be thorough to correct for many effects that other techniques (for example those based on Rietveld refinement) do not take into account.

We present here the workflow to get the PDF from total neutron scattering experiments in the constant wavelength configuration, the usual one for stationary sources, such as nuclear research reactors like the one operated by the Institute Laue Langevin (ILL). At ILL, the instrument dedicated to total scattering experiments is D4, which uses hot neutron beams. This technique is the most used to study the short range structure of glasses.

A series of routines specially designed for D4, but suitable for any two-axis neutron diffractometer, are grouped in Python modules and implemented as web-based Jupyter notebooks. In this way, the whole process can be followed step by step, thus avoiding black box software. These notebooks allow to get dead-time and efficiency corrected diffractograms, to perform the calibration of the wavelength and zero-angle correction, the correction of the sample by the attenuation and multiple scattering (using a Monte Carlo simulation), to get the differential scattering cross section in absolute units, to get the static structure factor and finally, by Fourier transformation, to get the correlation functions in the real space.

An example of a mixture of oxide glasses measured on D4 will be shown, though this software have been successfully used for hydrogenated samples and using polarised neutrons with polarisation analysis.

Spatial organization of hydrophobic and charged residues affects protein thermal stability and binding affinity

F. DeSantis

Italian Institut of Technology, 16163, Genoa, Italy.

E-mail: fausta.desantis@iit.it

What are the molecular determinants of protein-protein binding affinity and whether they are similar to those regulating fold stability are key issues in molecular biology having important implications both under a theoretical and an applicative perspective. Here, we analyze chemico-physical features on a large dataset of protein-protein complexes of known experimental binding affinity data and compare them with a set of monomeric proteins of available melting temperature data. Firstly, we probed the spatial organization of protein intra- and inter-molecular interaction energies among residues showing that strong Coulombic interactions associate with a high protein thermal stability, while strong intermolecular van der Waals energies correlate with protein-protein binding affinity. Given the role of van der Waals interface interactions in binding affinity, we focused on the molecular surfaces of the binding regions and evaluated their shape complementarity, employing a 2D Zernike polynomials expansion, thus managing to quantify the correlation between local shape complementarity and binding affinity. Moreover, considering the solvent interactions via the residue hydropathy, we found that the hydrophobicity of the binding regions dictates their shape complementary. These results pave the way to the fast and accurate prediction and design of optimal binding regions as the 2D Zernike formalism allows a rapid and superposition-free comparison between possible binding surfaces.

REACTIVE MD SIMULATION OF PHOSPHATE-BASED GLASSES

Z. Fallah¹, J.K. Christie¹

¹Department of Materials, Loughborough University, Loughborough, LE11 3TU, UK.

E-mail: z.fallah@lboro.ac.uk

Phosphate-based glasses (PBGs) have different applications based on their dissolution properties which can be tuned over several orders of magnitude via their composition targeting the desired application. The ReaxFF forcefield can describe the formation and dissociation of chemical bonds during molecular dynamics simulation allowing us to understand the dissolution mechanisms of the glass at the atomic level. In this work, we have developed for the first time, a reactive interatomic force field to describe the structure of these glasses. Firstly, we reparametrized the ReaxFF parameters of binary sodium phosphate glasses and validated them based on the structural properties of the phosphate glasses. Then we developed ReaxFF parameters of ternary PBGs which include the interaction between phosphorus and calcium atoms, as these are the compositions used in biomedical applications. Optimization of the parameters has been done against a large training set of quantum mechanical data, mostly from small boxes of the phosphate glasses (30–300 atoms). The developed ReaxFF parameters can describe structural properties of both binary and ternary glass compositions which are consistent with the experimental results.

VIBRATIONAL DYNAMICS OF ULTRA-STABLE AND CONVENTIONAL GLASSES

I. Festi¹, E. Alfinelli¹, F. Caporaletti², A.I. Chumakov³, D. Bessas³, J. Rodríguez-Viejo^{4,5}, C. Rodríguez-Tinoco^{4,5}, M. Rodríguez-López^{4,5}, M. Moratalla⁶, M.Á. Ramos⁶, G. Baldi¹

¹Department of Physics, University of Trento, I–38123, Povo, Trento, Italy.

²Laboratory of Polymer and Soft Matter Dynamics, Experimental Soft Matter and Thermal Physics (EST), Université libre de Bruxelles (ULB), Brussels 1050, Belgium.

³European Synchrotron Radiation Facility, BP 220, F-38043 Grenoble, France.

⁴Departament de Física. Facultat de Ciències, Universitat Autònoma de Barcelona, 08193 Bellaterra, Spain.

⁵Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and BIST, Campus UAB, Bellaterra, 08193

Barcelona, Spain.

⁶Departamento de Física de la Materia Condensada, Condensed Matter Physics Center (IFIMAC), and Instituto Nicolás Cabrera, Universidad Autónoma de Madrid, 28049 Madrid, Spain.

E-mail: irene.festi@unitn.it

The aim of our work is to measure the density of vibrational states (DOS) at low frequency of an ultra-stable glass and to compare it with that of the corresponding conventional glass. At low temperature, the thermodynamic properties of glasses are remarkably different from that of their crystalline counterparts. At temperatures below 1 K, the specific heat of glasses differs significantly from that of crystals and shows a linear temperature dependence: this anomaly is described by the phenomenological model of tunneling between two level states (TLS). We have some evidence that in ultra-stable glasses, prepared through physical vapor deposition, this anomaly is absent [1]. We have measured the DOS of amorphous samples of TPD ($C_{38}H_{32}N_2$) to quantify the different vibrational contributions to the specific heat. The measurements were carried out at the ID18 beamline of the ESRF synchrotron, in Grenoble. By exploiting the inelastic X-ray scattering with nuclear resonance analysis, we managed to measure the DOS down to 0.3 meV. The data analysis shows that the DOS of the ultra-stable glass is lower than that of the conventional one, as expected. Furthermore, a good agreement between the specific heat measured with calorimetry experiments and that evaluated starting from the DOS is found. The DOS allows us to conclude that a small contribution from the TLS is still present in the ultra-stable glass, although the linear term is not observed in the specific heat measurement.

[1] T. Pérez-Castañeda et al., Proc. Natl. Acad. Sci. U.S.A. 111, 11275 (2014).

CANVAS: A fast, accurate, and system-specific variable resolution approach for simulating biomolecules

R. Fiorentini, T. Tarenzi, R. Potestio

Department of Physics, University of Trento, via Sommarive 14, Trento, Italy. INFN-TIFPA, Trento Institute for Fundamental Physics and Applications, via Sommarive 14, Trento, Italy.

E-mail: raffaele.fiorentini@unitn.it

The field of multi-scale modeling and simulation has enjoyed significant success in soft matter research within the past decade also thanks to the boost impressed by the necessity to overcome the expensive cost of a single, highly detailed resolution. Several methodologies have been developed in the past few years, where different system components are simultaneously modelled at different levels of resolution [1]. In the case of biomolecules, functionally relevant parts of the system can be modelled at a high a level of detail, while the remainder of the system can be represented using less expensive models. Here, we propose a novel multiresolution scheme dubbed coarse-grained anisotropic network model for variable resolution simulations, or CANVAS, which allows one to employ and smoothly couple virtually any desired degree of coarse-graining within the same model [2]. The novelty of this method lies in the possibility of setting the level of resolution of the coarse-grained subdomain(s) in a quasi-continuous range; furthermore, the interaction network within and between resolution domains is straightforwardly parametrized on the basis of the properties of the specific system under examination. The model is validated by comparing results from all-atom and multi-scale simulations of two biomolecules, namely the enzyme adenylate kinase and the IqG4 antibody pembrolizumab.

^[1] M. Giulini et al., Front. Mol. Biosci., 8, 676976 (2021).

^[2] R. Fiorentini, T. Tarenzi, and R. Potestio, arXiv:2207.03187.

Isocompositional liquid-liquid transition at ambient pressure in dilute aqueous LiCl solutions

J. Giebelmann, J. Bachler, T. Loerting

Institute of Physical Chemistry, University of Innsbruck, Innrain 52c, A-6020 Innsbruck, Austria.

E-mail: Johannes.Giebelmann@uibk.ac.at

The two liquid model of water involving a first-order liquid-liquid transition is among the most promising ideas to explain the anomalous nature of H_2O [1]. At ambient pressure the liquid-liquid transition line cannot be accessed. Upon cooling liquid water significant changes occur near 220 K, which have been interpreted to involve the crossing of a Widom line, but this is masked by simultaneous crystallization of ice [2]. Likewise, upon heating of high-density amorphous ice (HDA) [3] cold-crystallization and rather high glass transition temperature of low-density amorphous ice (LDA) near 136 K [4] interfere, so that it is not easy to make the case for a direct observation of a high-to-low density liquid-liquid transition near 140 K. In this work we investigate the glass transitions and phase behaviour of dilute, aqueous LiCl solutions, where hyperquenching is used to avoid crystallization. It has recently been shown [5] that densified hyperguenched glassy water (d-HGW) shows two glass transitions at ambient pressure: One that corresponds to low- (LDA) and a second one pertaining to high-density amorphous ice (HDA). We show that both glass transitions are observed even after LiCl is introduced but its effect on their onset temperatures differs. While the first glass transition decreases from 136 K to 126 K, the second stays at around 118 K. Furthermore we observe a first-order transition in such solutions at 137-141 K, which clearly takes place in the deeply supercooled liquid domain. That is, by contrast to pure water, we are able to access an isocompositional transition between two liquids of different densities.

- [1] P. Gallo et al., Eur. Phys. J. E 44, 143 (2021).
- [2] P.H. Handleet al., Proc. Natl. Acad. Sci. U.S.A. 114, 13336 (2017).
- [3] Y.P. Handa et al., J. Chem. Phys., 84, 2766 (1986).
- [4] M.S. Elsaesser et al., Phys. Chem. Chem. Phys. 12, 708 (2010).
- [5] J. Bachler et al., J. Chem. Phys., 157, 64502 (2022).

Researching the formation of CO2 structures in cryocondensate thin films

D. Sokolov^{1,2}, O. Vorobyova¹, **O. Golikov**¹, D. Yerezhep¹, A. Aldiyarov¹

¹Al-Farabi Kazakh National University, Al-Farabi av., 71. Almaty, Kazakhstan. ²Almaty Technological University, Tole bi av., 100. Almaty, Kazakhstan.

E-mail: yasnyisokol@gmail.com

This paper focuses on the study of hydrate and gas hydrate compounds formed in $CO_2 + H_2O$ mixtures. The research was carried out in thin films, using the physical vapour deposition (PVD) method in a pure vacuum at a pressure of $P = 0.5 \,\mu$ Torr. The FTIR spectra obtained using the Fourier IR spectrometer FSM 2203 (INFRASPEK, Russia) suggest the emergence of sl-type hydrate and clathrate structures in the mixture under study, as indicated by the characteristic peaks at frequencies of 2275 cm⁻¹, 2280 cm⁻¹ and 2340 cm⁻¹. Typical gas hydrate peaks at frequencies of the second Fermi resonance were not observed, which is obviously linked to the high absorption intensity of water ice in the frequency range of 3580-3620 cm⁻¹. Mass spectroscopic data (obtained using the Extorr XT100 (Extorr Inc., USA) complement the FTIR spectra and clearly demonstrate the change in sublimation temperature at the selected pressure. During the experiment, the majority of the CO2 molecules sublimated at 147-150 K rather than at 93 K, the temperature at which CO₂ sublimates under a pressure of $P = 0.5 \mu$ Torr. PMT P25A (Sens-Tech, UK) was used to study the refractive indices of $H_2O + CO_2$ mixtures at different concentrations. When the concentration of water approached 25%, the formation of structures less dense than the amorphous CO_2 condensates and amorphous H_2O ice was observed. For the concentration of CO_2 (25%) – H_2O (75%), the changes in the spectra and data recorded via mass spectroscopy indicate incomplete hydration of the mixture. Some of the CO₂ molecules remain in a free state and sublimate at a temperature of about 93 K.

Electrostatic complementarity evaluation at the protein interfaces: prediction of transient protein-protein interactions

G. Grassmann^{1,2}, L. Di Rienzo², G. Gosti^{2,3}, M. Leonetti^{2,3}, G. Ruocco^{2,4}, M. Miotto², E. Milanetti^{2,4}

¹Department of Biochemical Sciences "Alessandro Rossi Fanelli", Sapienza University of Rome, P.Le A. Moro 5, 00185, Rome, Italy.

²Center for Life Nano & Neuro Science, Istituto Italiano di Tecnologia, Viale Regina Elena 291, 00161, Rome, Italy.
³Soft and Living Matter Laboratory, Institute of Nanotechnology, Consiglio Nazionale delle Ricerche, 00185, Rome,

Italy.

⁴Department of Physics, Sapienza University, Piazzale Aldo Moro 5, 00185, Rome, Italy.

E-mail: greta.grassmann@iit.it

Understanding the mechanisms driving bio-molecules interactions is fundamental for the predicition of protein binding and the characterization of complex systems such as protein assemblies. Features like the preferentially hydrophobicity of the binding interfaces and the role of van der Waals interactions are well established. However, no consensus has yet been reached on the role of electrostatic. Here, we perform a computational analysis to evaluate the role of the electrostatic match between interacting protein interfaces, showing an inversely proportional relationship between the electrostatic complementarity and the experimentally binding affinity. Moreover, we propose a novel, fast and efficient computational method, based on the 2D Zernike polynomial formalism, to compactly describe the electrostatic properties of protein regions. In accordance with electrostatic analysis based on Poisson-Boltzmann, we suggest that less stable complexes (often transient interactions) have greater electrostatic complementarity and less shape complementarity. The results of this approach help to shed light on the non-trivial relationship between the hydrophobic and electrostatic contributions in the binding interfaces, thus favoring the development of new predictive methods for binding affinity, given that the proposed descriptor is able to discriminate between transient and permanent protein complexes with an AUC of the ROC of ~ 0.8 .

ISOCHRONAL SUPERPOSITION OF THE STRUCTURE FOR CUMENE

E.H. Lørup

Glass & Time, IMFUFA, Roskilde University, Roskilde, Denmark.

E-mail: Hillo@ruc.dk

The fundamental prediction of the isomorph theory is that exist curves in the phase diagram, where along these lines structure and dynamics are invariant [1]. Previous test on Cumene have successfully tested the dynamical prediction of isomorph theory [2]. The relaxation time is invariant along isomorphs making isochrones suitable experimental identifiers for isomorphs. We present the results of one of the first experimental test of structural prediction of theories of isomorph theory. We compare the main diffraction peak along isobars, isotherms, isochors and isochrones, and find that the main diffraction peak collapse along isochrones.

[1] J.C. Dyre, J. Phys. Chem. B 118, 10007-10024 (2014).

[2] H.W. Hansen et al., J. Chem. Phys. 149, 214503 (2018).

INFORMATION-THEORETICAL MEASURES IDENTIFY ACCURATE LOW-RESOLUTION REPRESENTATIONS OF PROTEINS

M. Margherita, R. Menichetti, R. Potestio

Department of Physics, University of Trento, via Sommarive 14, Trento, Italy. INFN-TIFPA, Trento Institute for Fundamental Physics and Applications, via Sommarive 14, Trento, Italy.

E-mail: margherita.mele@unitn.it

"What is the appropriate level of detail in this case?" This question occurs in many situations, whether to tell a story, save a photo, or study a complex system. In this latter case, finding the optimal level of detail is a crucial task. In fact, the ever-growing computational power employed to perform molecular dynamics simulations of biological macromolecules produces large amounts of data, from which it is necessary to extract useful, synthetic, and intelligible information to make the crucial step from knowledge to understanding. The theoretical framework of resolution and relevance [1] and the information-theoretic measure dubbed mapping entropy [2,3] serve this scope, as these general quantities can be employed to make unsupervised processing of data, in particular to compress the available raw input in a way that maximises the emergence of useful information. In our work, we employ these quantities and the relationship between them to identify, in a quantitative manner, a simplified representation of the system that holds the most information about the generative process underlying an empirical, finite data set. When applied to the analysis of the configurational space of proteins, this approach reveals that clustering methods inducing ultrametric structure in their projected, low-dimensional spaces are more physically accurate in preserving the information content of the high-dimensional counterpart, consistent with the hypothesis that the configurational landscape of proteins has a self-similar organisation [3].

^[1] M. Marsili and Y. Roudi, Phys. Rep. 963, 1 (2022).

^[2] R. Holtzman, M. Giulini, and R. Potestio, Phys. Rev. E 106, 044101 (2022).

^[3] M. Mele, R. Covino, and R. Potestio. Soft Matter 18, 7064 (2022).

Dynamical and kinetic assessment of nucleic acids systems

M. Micheloni, G. Mattiotti, L. Petrolli

Department of Physics, University of Trento, Via Sommarive 14, I-38123 Trento, Italy. INFN-TIFPA, Trento Institute for Fundamental Physics and Applications, I-38123 Trento, Italy.

E-mail: manuel.micheloni@unitn.it

The in silico characterization of the dynamics of nucleic acids at the molecular scale by means of numerical techniques, such as Molecular Dynamics (MD), has been instrumental in depicting the fundamental mechanics underlying most cellular processes. Yet, atomistic MD simulations are still bound by a high numerical overhead, affecting the statistical significance of the sampling of the conformational space – all the more so in contexts of massive biological systems. Hence, coarse–grained (CG) models, such as oxDNA [1], have been developed that capture the global behavior of nucleic acids, while keeping an appropriate level of resolution accounting for local, sequence–specific thermodynamic properties.

Here, we show two scenarios of application of the oxDNA CG force field that relate to extremely relevant biological phenomena. Firstly, we characterize the (structural, dynamical) behavior of a viral RNA fragment [2] (2774 nucleotides) at equilibrium, upon diverse salt concentrations, as well as out-of-equilibrium, by enforcing a time-dependent bias mimicking the (electrostatic, structural) constraints of its viral capsid. On the other hand, we carry out an assessment of the kinetics of the DNA disruption by diverse double strand break motifs, whereby we observe an exponential, Arrhenius-like increase of the characteristic rupture times as function of the distance between the cuts of the DNA backbone.

[1] B.E. Snodin et al., J. Chem. Phys. 142, 06B613_1 (2015).

[2] R.F. Garmann, PhD thesis, University of California, Los Angeles (2014).

The shape complementarity evaluation at the interfaces of protein-protein complexes through a novel strategy based on 2D Zernike formalism

E. Milanetti

Department of Physics, Sapienza University of Rome, P. Le Moro, 5, 00185 Rome, Italy.

E-mail: edoardo.milanetti@uniroma1.it

Understanding the molecular mechanisms driving the interaction between proteins plays a crucial role both in the understanding of cellular activities and in the development of new biotechnologies. Despite the complete knowledge of the human genome and the set of protein structures, which can be predicted with high accuracy by novel machine learning based algorithms, to date predicting the interaction between two proteins still remains an open problem in the field of computational biology. Indeed, even if the information about the interaction is encoded in the chemical and geometric features of the interacting structures, the number of possible binding regions on a protein surface, and their relative orientations, are too large to be computationally characterized in a reasonable time. To address this issue, we developed a novel method for efficiently and effectively assessing whether and where two proteins can interact with each other to form a complex. The method can rapidly and quantitatively measure the geometrical shape complementarity between interacting proteins. It compares the molecular iso-electron density surfaces of the two proteins, expanding the surface patches in terms of 2D Zernike polynomials. The comparison between all the patches of two proteins allows the association, with low time and computational cost, each patch with each other through a contact probability score. This make the analysis of a large amounts of data possible, allowing for instance the application on all human proteins or molecular dynamics simulation data.

MAGNETO-MECHANICAL RESPONSE OF MAGNETIC FILAMENTS WITH SOLVOPHOBIC, SUPER-PARAMAGNETIC COLLOIDS, IN BULK

D. Mostarac¹, E. Novak², S. Kantorovich^{1,2}

¹University of Vienna, Faculty of Physics, 1090, Vienna, Austria. ²Ural Federal University, Ekaterinburg, Russia.

E-mail: deniz.mostarac@univie.ac.at

Incorporating magnetic nanoparticles within permanently cross-linked structures, opens the possibility for synthesis of highly magneto-responsive systems, with improved rheological, transport and optical properties compared to conventional magnetic fluids. Magnetic filaments (MFs) are polymer-like chains of magnetic, nano-sized colloids, and are an example of the aforementioned complex systems. Understanding the interplay between solvent selectivity, and dipolar interactions in conjunction with backbone architecture is crucial to be able to tune the macroscopic response of MFs and control the formation of self-assembled superstructures. In this contribution we study suspensions of MFs and show that considering nonlinear contributions to the magnetisation of super-paramagnetic colloids is essential to capture the magneto-mechanical response of MFs in bulk, consisting of magnetisable colloids with a varying degree of solvophilicity. Based on the degree of solvophilicity of magnetic colloids in MFs, we report substantial differences in the equilibrium structure of filament clusters, and the mechanical and magnetic response of filament suspensions, to static and time dependant magnetic fields. On the level of a single filament, magneto-mechanical properties are in general more dependent on the crosslinking approach than on the magnetic nature of colloids. In bulk, however, the long-range nature of dipolar interactions in quasi-infinite systems makes up for a tremendous difference and underlines the necessity of a sophisticated model of magnetic colloids that can be magnetised.

[1] D. Mostarac et al., Nanoscale 12, 13933-13947 (2020).

[2] D. Mostarac and S.S. Kantorovich, Macromolecules 55, 6462-6473 (2022).

INVESTIGATING THE ACTION MECHANISM OF PORE-FORMING TOXINS WITH MOLECULAR DYNAMICS SIMULATIONS AT DIFFERENT RESOLUTION SCALES

C. Paternoster, T. Tarenzi, R. Potestio, G. Lattanzi

Department of Physics, University of Trento, via Sommarive 14, Trento, Italy. INFN-TIFPA, Trento Institute for Fundamental Physics and Applications, via Sommarive 14, Trento, Italy.

E-mail: costanza.paternoster@unitn.it

Pore-forming toxins (PFTs) represent an ancient protein family found in all kingdoms of life. Once secreted in the extracellular space, PFTs undergo a functional rearrangement that leads inactive, soluble, and monomeric proteins to assemble as a multimeric pore in the membrane of the target cell, leading to the cell's death by leakage or apoptosys [1]. A detailed understanding of the mechanism of transition between soluble and membrane-bound forms of PFTs is fundamental for the design of antivirulence agents against PFT-producing pathogens, such as the methicillin-resistant *Staphylococcus aureus* [2]. Here, through molecular dynamics simulations at different resolution scales, ranging from atomistic to coarse-grained, we have investigated the first steps of pore formation of γ -hemolysin, a bi-component PFT constituted by four copies of the LukF and Hlg2 proteins. A comparison between the two monomers has been performed in terms of membrane-binding residues and stability of the anchoring on the bilayer surface [3]. We have then employed these results to investigate the subsequent stage of spontaneous dimerization on the membrane, shedding light on the LukF and Hlg2 interaction surface and the putative mechanism of transition between the soluble monomers and the membrane-bound dimer.

[1] M. Dal Peraro and F.G. Van Der Goot, Nat. Rev. Microbiol. 14, 77 (2016).

[2] E.S. Seilie and J. B. Wardenburg, Semin. Cell. Dev. Biol. 72, 101-116 (2017).

[3] T. Tarenzi, G. Lattanzi, and R. Potestio, BBA-Biomembranes 1864, 183970 (2022).

Modelling liquid dynamics by random walks within overlapping hyperspheres

M.F.B. Railton, J.C. Dyre, T.B. Schrøder

"Glass & Time", IMFUFA, Roskilde University.

E-mail: mfbr@ruc.dk

It has been shown [1] that the real part of the frequency-dependant fluidity conforms to the prediction of the random barrier model for nine glass-forming liquids of different chemistry. We have studied the hypersphere model, which is a simple one parameter model of a potential energy landscape, and is defined by a percolating system of equally sized n-balls centered on randomly distributed points in \mathbb{R}^{3N} . Using the mean squared displacement for the inherent dynamics of random walks, we have shown that the average behaviour in the 42-dimensional hypersphere model (corresponding to a 14 particle system) encapsulates the behaviour of the random barrier model, and thus also to that of glass-forming liquids, several decades prior to the onset of diffusion. A method to generate potential energy landscapes, similar to that of the hypersphere model, on-the-fly has been used. Combining this with the n-fold way Kinetic Monte Carlo method and several other optimization techniques, allowed simulations of the inherent dynamics of random walks at experimental time scales.

[1] S. Bierwirth, R. Böhmer, and C. Gainaru, Phys. Rev. Lett. 119, 248001 (2017).

Amorphizing or modifying materials by MeV ion irradiation

M.A. Ramos^{1,2,3}, A. Andrino^{1,2,3,4}, M. Moratalla^{1,2,3}, J.L. Sánchez-Toural⁴, A. Redondo-Cubero^{1,3,4}, G. Tabares^{1,3,4}, J.L. Pau^{3,4}, G. García¹, N. Gordillo^{1,3,4}

¹Centre for Micro Analysis of Materials (CMAM), Universidad Autó noma de Madrid, Spain.

²Laboratorio de Bajas Temperaturas, Dpto. de Física de la Materia Condensada, Universidad Autó noma de Madrid,

Spain.

³Instituto Nicolá s Cabrera (INC), Universidad Autó noma de Madrid, Spain.
⁴Departamento de Física Aplicada, Universidad Autó noma de Madrid, Spain.

E-mail: miguel.ramos@uam.es

In our highly-technological world, there is an increasing demand and use of quantum properties of materials for new devices and sensors of unprecedented precision including, but not limited to, biomedical applications, key elements in micro- and nano-electronics, or the basis for the expected bloom of quantum computation in the near future. Our research project on *Quantum materials and sensors by MeV ion implantation* is focused on exploiting the capabilities of our 5 MV ion-beam accelerator at CMAM aiming to modify several target materials with potential quantum applications.

Our first goal is to tune the properties of synthetic diamond crystals: (i) by irradiating with nitrogen (N) ions, we aim to create NV colour centres in diamond, in order to develop sensitive quantum magnetic sensors; (ii) by irradiating with boron (B) MeV ions, we could fabricate structures or micropatterns of superconducting boron-doped diamond.

Our second research line is the investigation of bismuth-antimony alloys, ranging from pure Bi to about 20% of Sb doping. In amorphous state, they are appealing superconducting materials with critical temperatures above 6 K. They are considered potential candidates for topological superconductivity, an emergent research field with promising expectations for quantum computing, among others. Although Bi–Sb alloys unfortunately tend to crystallize unless at very low temperatures, we are working out several hypotheses and strategies to overcome such difficulty.

EFFECT OF HIGH PRESSURE ON THE ATOMIC DYNAMICS OF A AU-BASED METALLIC GLASS

A. Ronca, B. Ruta

Institute of Light and Matter, UMR5306 Université Lyon 1–CNRS, Université de Lyon, F–69622 Villeurbanne, France. European Synchrotron Radiation Facility, 71 avenue des Martyrs, CS 40220, Grenoble 38043, France.

E-mail: alberto.ronca@univ-lyon1.fr

The understanding of how pressure influences the rich phenomenology of glasses attracts the scientific community not only from a theoretical point of view, but also for technological applications. This is particularly true for metallic glasses (MGs), which exhibits suppression of shear banding and inhibition of catastrophic mechanical failures under quasi-hydrostatic compression, making deformed MGs appealing for technological applications [1].

Historically, the use of pressure as an experimental variable mainly concerned the study of macroscopic quantities, owing in part to experimental convenience. Nowadays, our understanding of the microscopic structural and dynamical effect of deformation in MGs is limited to ex situ densified glasses. The recently developed 4th generation synchrotron sources, like the ESRF synchrotron in France, made in situ pressure-dependent studies experimentally achievable thanks to a 100 times higher coherent flux (up to 10¹² photon/s).

This work investigates the atomic scale in situ pressure-dependence of a Au-based MG by X-rays photon correlation spectroscopy (XPCS). From the structural point of view, pressure is found to induce a reversible elastic densification of the system, while the dynamics exhibits an hysteresis upon compression and decompression. Moreover, under hydrostatic compression the atomic motion accelerates, as indicated by a decrease in the characteristic relaxation time of the system, which suggests rejuvination. Our results support recently developed theories predicting rejuvination of MGs under compression [2–3].

- [1] J. Pan et al., Nature 578, 559-562 (2020).
- [2] A.D. Phan et al., Phys. Rev. Lett. 126, 025502 (2021).
- [3] N.K. Ngan et al., Phys. Status Solidi (RRL) 15, 2100235 (2021).

Spatial resolution of glass relaxation dynamics: a real-time microscopic view

M. Ruiz-Ruiz^{1,2}, A. Vila-Costa^{1,2}, T. Bar³, C. Rodríguez-Tinoco², M. Gonzalez-Silveira^{1,2}, J. Antonio Plaza^{1,2}, J. Alcalá⁴, J. Fraxedas², J. Rodriguez-Viejo^{1,2}

¹Departamento de Física. Facultad de Ciencias, Universitat Autònoma de Barcelona, 08193, Bellaterra, Spain.

²Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and BIST, Campus UAB, Bellaterra, 08193, Barcelona, Spain.

³Instituto de Microelectrónica de Barcelona, IMB-CNM (CSIC), Esfera UAB, Campus UAB, Cerdanyola, Barcelona, Spain.

⁴InSup, ETSEIB, Universitat Politècnica de Catalunya, 08028 Barcelona, Spain.

E-mail: marta.ruiz@uab.cat

he understanding of glassy dynamics above the devitrification temperature of a glass remains poorly understood. Here, we use real-time AFM imaging to provide a direct microscopic view of the highly heterogeneous transformation of thin film stable glasses with bulk-like dynamics following a temperature jump above the glass transition temperature. We show the existence of localized fast mobility regions separated by giant length scales and their progression inside the glass via dynamic facilitation. This approach may also be applicable to spatially study the microscopic dynamics structure of other glass forming systems with much shorter length and time scales, including liquid-cooled glasses.

The electric and magnetic disordered Maxwell equations as eigenvalue problem

W. Schirmacher^{1,2}, T. Franosh³, M. Leonetti^{1,4}, G. Ruocco^{1,5} ¹Center for Life Nano science@Sapienza,

Istituto Italiano di Tecnologia, Viale Regina Elena, 291, I-00161 Roma, Italy

²Institut für Physik, Universität Mainz, Staudinger Weg 7, D-55099 Mainz, Germany

³Institut für Theoretische Physik, Universität Innsbruck, Technikerstrasse 21A, A-6020 Innsbruck, Austria

⁴Soft and Living Matter Laboratory, Institute of Nanotechnology, Consiglio Nazionale delle Ricerche, 00185 Rome,

Italy

⁵Dipartimento di Fisica, Sapienza Università di Roma, Piazzale Aldo Moro 2, I-00185, Roma, Italy

We consider Maxwell's equations in a 3-dimensional material, in which both, the electric permittivity, as well as the magnetic permeability, fluctuate in space. Differently from all previous treatments of the *disordered* electromagnetic problem, we transform Maxwell's equations and the electric and magnetic fields in such a way that the linear operator in the resulting secular equations is manifestly Hermitian, in order to deal with a proper eigenvalue problem. As an application of our general formalism, we use an appropriate version of the Coherent–Potential approximation (CPA) to calculate the photon density of states and scattering–mean–free path. Applying standard localization theory, we find that in the presence of both electric and magnetic disorder the spectral range of Anderson localization appears to be much larger than in the case of electric (or magnetic) disorder only. Our result could explain the absence of experimental evidence of 3D Anderson localization of light (all the existing experiments has been performed with electric disorder only) and pave the way towards a successful search of this, up to now, elusive phenomenon.

CAPILLARY THINNING OF POLYMER SOLUTION THREADS: SELF-SIMILAR CONFIGURATIONS AND INSTABILITIES

A.N. Semenov¹, A. Subbotin², I.A. Nyrkova¹

¹Institut Charles Sadron, University of Strasbourg, Strasbourg, France. ²Topchiev Institute of Petrochemical Synthesis, R A S, Moscow, Russia.

E-mail: semenov@unistra.fr

Long liquid cylinders are unstable and tend to break in many droplets. However, adding a small amount of long-chain polymer can suppress the breakup: a beads-on-a-string structure of nearly uniform stable threads connecting well-separated droplets is formed instead. This phenomenon is of importance in many industrial and biological processes; it also serves as a basis of modern rheological devices (FSR, CaBER). Yet, polymer solution threads can suffer secondary instabilities which are often undesired. This talk summarizes our theoretical advances in this area. Based on recently established anti-Bernoulli law for the Oldroyd-B model we obtain a self-similar solution for the thread shape and corner flow in the transition zone [1]. Then we turn to the thread pearling instabilities. One mechanism is related to anisotropic interactions of stretched polymer fragments, another involves dynamical phase separation due to capillary-driven squeezing of solvent in polymer matrix [2]. A hierarchical structure of polydisperse annular droplets around a thin polymer fibril emerges as a result [3]. We show that such pearling instabilities in polymer solutions and gels occur when the thread radius falls below a critical value which increases as concentration or solvent quality is lowered.

^[1] A.N. Semenov and I.A.Nyrkova, Polymers 14, 4420 (2022).

^[2] A.V. Subbotin and A. N. Semenov, Macromolecules 55, 2096 (2022).

^[3] A.V. Subbotin, A. N. Semenov, J. Rheol. 67, 53 (2023).

Thermodynamic limit in computer simulations via finite-size integral equations

M. Sevilla, R. Cortes-Huerto

Max Planck Institute for Polymer Research.

E-mail: sevillaj@mpip-mainz.mpg.de

Integral equations (IE) are in the core of statistical mechanics of liquids as they connect the local structure with thermodynamic properties as compressibility, activity coefficients and excess entropy. IE are normally defined in the grand canonical ensemble and calculated in the thermodynamic limit (TL). By contrast, computer simulations are performed with finite-size systems and emulate the TL by using periodic boundary conditions (PBC). This procedure introduces several finite-size contributions whose effects must be identified and corrected in order to approximate the simulation results to the TL. In this talk, we present a generic method to compute IE from molecular dynamics simulations. In our approach, we define finite-size IE, integrating them in Fourier space to trivially introduce PBC. This strategy allows us to identify and isolate ensemble, finite-volume domains and PBC effects and accurately obtain the corresponding thermodynamic quantities in the TL or artificially for any finite size system or ratios for anisotropic boxes. To validate our method, we compute isothermal compressibilities, chemical potentials and excess entropies of simple liquids and liquid mixtures, including water and aqueous alcohol solutions, showing good agreement with results available in the literature.

Exploring pharmaceuticals with conventional and fast calorimetry

D. Sonaglioni^{1,2}, E. Tombari³, S. Capaccioli^{1,3,4}

¹Dipartimento di Fisica "E. Fermi", Università di Pisa, Largo Pontecorvo 3, 56127, Pisa, Italy.

²Center for Materials Interfaces-Electron Crystallography, Istituto Italiano di Tecnologia, Viale Rinaldo Piaggio 34, 56025. Pontedera(Italu).

³Istituto per i Processi Chimico–Fisici del CNR (CNR-IPCF), Via Moruzzi 1, 56124 Pisa, Italy.

⁴CISUP, Centro per l'Integrazione della Strumentazione dell'Università di Pisa, Lungarno Pacinotti 43, 56126, Pisa,

ltaly.

E-mail: daniele.sonaglioni@phd.unipi.it

Pharmaceuticals have always attracted attention, partly because their industrial application is often hindered by their low dissolution rate. Amorphisation has been proven to enhance their dissolution rate [1], although reverse to the crystalline state hampers their use. Additionally, in some cases it is impossible to reach the glassy state. Generally, there exists three classes of pharmaceuticals, inversely ranked according to crystallization tendency [2].

Differential scanning calorimetry (DSC) has been largely used for characterizing glassy and crystalline pharmaceuticals [3–7]. A step further is represented by fast DSC: it allows scan rate up to 10^5 K/s and to probe faster dynamics [8].

Our study focuses on the glass forming ability and stability of two poorly soluble drugs: indoprofen and bifonazole.

Indoprofen, class I, has a high crystallization tendency, being difficult to reach the glassy state through DSC [6]. Fast DSC unlocks the possibility to obtain glassy indoprofen, enabling to characterize its recrystallization, form both the melt and the glass.

Bifonazole, class II, is a good glass-formers, but little characterized [7]: DSC will be employed to study its glassy and crystalline states, unveiling the peculiarities of this compound, whereas fast DSC will allow the study of glass-forming dynamics.

Fast DSC data analysis will be done with a new method: relying on raw data only, it estimates the thermal lag and the absolute value of the specific heat without further experiments [8].

- [1] C. Bhugra and M.J. Pikal, J. Pharm. Sci. 97, 1329 (2008).
- [2] J.A. Baird, B. Van Eerdenbrugh, and L.S. Taylor, J. Pharm. Scie. 99, 3787-3806 (2010).
- [3] E. Tombari et al., J. Phys. Chem. B 112, 10806–10814 (2008).
- [4] M. Descamps and E. Dudugnon, J. Pharm. Scie. 103, 2615–2628 (2014).
- [5] Y. Gao et al., Prog. Mater. Sci. 104, 53-137 (2019).
- [6] M. Vasanthavada et al., Pharm. Res. 22, 440-448 (2005).
- [7] J.J. Moura Ramos and H. P. Diogo, J. Therm. Anal. Calorim. 145, 3077-3085 (2021).
- [8] D. Sonaglioni, E. Tombari, and S. Capaccioli, Thermochim. Acta 719, 179385 (2023).

Exploring the glassy dynamics of the Gaussian core model

V. Sposini¹, M. Camargo², C. N. Likos¹

¹Computational and Soft Matter Physics, Faculty of Physics, University of Vienna, Vienna, Austria. ²FIMEB & CICBA, Universidad Antonio Nariño-Campus Farallones, Cali, Colombia.

E-mail: vittoria.sposini@univie.ac.at

Within the realm of soft colloids a prominent role is played by the Gaussian Core Model (GCM) introduced by Stillinger in the 70's [1]. The GCM describes point particles interacting by means of a Gaussian-shaped potential and is one of the simplest models for the description of systems such as polymer or dendrimer solutions [2,3]. Whereas at low temperatures and densities the GCM is believed to be described by an effective hard-sphere mapping, at high densities a mean-field description sets in, giving rise to re-entrant melting [4] and a glass state compatible with a geometric transition [5]. The broad intermediate glass regime remains unexplored [6]. In this talk I will present results from molecular dynamics simulations exploring the slow dynamics of the GCM in this regime. In particular, I will discuss the transition from the low density hard-sphere-like glass to the high density glass, featuring novel characteristics such as anomalous diffusion after cage escape as well as intermediate scattering function with logarithmic decay. We attribute these unique features to the interplay between temperature and soft decay tail of the GCM.

- [1] F.H. Stillinger, J. Chem. Phys. 65, 3968 (1976).
- [2] A.A. Louis et al., Phys. Rev. Lett. 85, 2522 (2000).
- [3] I.O. Götze et al., J. Chem. Phys. 120, 7761 (2004).
- [4] A.Lang et al., J. Phys. Condens. Matter 12, 5087 (2000).
- [5] D.Coslovich et al., Phys. Rev. E 93, 042602 (2016).
- [6] J.-M. Bomont et al., Phys. Rev. E 105, 024607 (2022).

On the characteristic thermal conductivity plateau in model amorphous silica

D. Szewczyk^{1,2}, M.A. Ramos^{2,3}

¹Department of Low Temperature and Superconductivity, Institute of Low Temperature and Structure Research PAS, Wroclaw, Poland.

²Departamento de Física de la Materia Condensada & Instituto "Nicolás Cabrera", Universidad Autónoma de Madrid, Madrid, Spain.

³Centro de Microanálisis de Materiales (CMAM), Universidad Autónoma de Madrid, Madrid, Spain.

E-mail: d.szewczyk@intibs.pl

The temperature dependence of thermal conductivity at low temperatures in amorphous materials is often considered as an already well-known feature, although its true nature still demands understanding. Generally, the thermal conductivity of non-crystalline materials is several orders of magnitude lower than in the case of crystals. When lowering the temperature, the thermal conductivity coefficient decreases monotonically until reaching a characteristic plateau, where the value is assumed to be almost independent of the materials' chemical composition followed by a further quadratic decrease as T^2 at the lowest temperatures. Nevertheless, in the most examined model case of paradigmatic amorphous silica, the thermal conductivity values found in literature, in the plateau region, differentiate more than 50% from each other, only to mention the well-renowned works of Zeller and Pohl (1971), Cahill (1987) or Freeman and Anderson (1986). Therefore a need for detailed, low temperature thermal conductivity studies of silica has risen again. For our studies, we chose a UV-grade SiO_2 (commercially available spectrosil 3000) and performed thermal conductivity measurements both in a commercial setup (PPMS) and a specially designed for the purpose insert to the He_4 cryostat to unravel the encountered question. Additionally, the details of the newly constructed device will be presented including the methodology of operating the system.

Pressure Scanning Volumetry: a useful alternative to Scanning Calorimetry

E. Tombari¹, D. Sonaglioni², G.P. Johari³

¹Istituto per i Processi Chimico-Fisici del CNR, via G. Moruzzi 1, 56124 Pisa, Italy.
 ²Dipartimento di Fisica "E. Fermi", Università di Pisa, Largo Pontecorvo 3, 56127, Pisa, Italy.
 ³Department of Materials Science and Engineering, McMaster University, Hamilton, ON L8S 4L7, Canada.

E-mail: elpidio.tombari@ipcf.cnr.it

In the commonly used technique of scanning calorimetry, one measures the heat capacity, C_p , as a function of temperature T on heating or cooling a material at a fixed rate. A liquid also becomes glass on isothermal pressurizing and the glass is expected to become liquid on depressurizing. We describe the formalism of a technique in which the volume, V, may be measured as a liquid is isothermally pressurized at a fixed rate, dp/dt, to form glass, and as the glass is isothermally depressurized at a fixed rate until it becomes liquid. We name the technique as pressure scanning volumetry (PSV), and simulate the pressurizing and depressurizing scans of $(dV/dp)_T$ within the framework of non-exponential, non-linear structural relaxation [1, 2]. Thus we obtain the features of simulated PSV scans by varying (i) the pressurizing and depressurizing rates, (ii) the parameters β and x, and (iii) the volume of activation for characteristic relaxation time, ΔV^* . We also describe how the limiting fictive pressure, can be determined from a PSV scan, and fit the PSV formalism to the available bulk modulus against p plot of a polymer to obtain β , x and ΔV^* . In addition to being of academic interest in determining the role of pressure *vis-a-vis* that of temperature, pressure scanning volumetry would be useful for modeling polymer extrusion processes.

[1] C.T. Moynihan et al., Ann. N. Y. Acad. Sci. 279, 15-35 (1976).

[2] I.M. Hodge and A.R. Berens, Macromolecules 15, 762-770 (1982).

INTERACTION OF IONIC SURFACTANTS WITH EPOXY-BASED HYDROGEL INVESTIGATED BY SANS

T. Tropin¹, I. Krakovsky², O.I. Ivankov¹, V.I. Petrenko^{3,4}

¹Frank Laboratory of Neutron Physics, Joint Institute for Nuclear Research, Dubna, Russia.
²Department of Macromolecular Physics, Faculty of Mathematics and Physics, Charles University, Czech Republic.
³Basque Center for Materials, Applications & Nanostructures, Leioa, Spain.

⁴Ikerbasque, Basque Foundation for Science, Bilbao, Spain.

E-mail: ttv@jinr.ru

Stoichiometric amphiphilic epoxy network containing polyoxyethylene (POE) and polyoxypropylene (POP) was prepared by reaction of α , ω -diamino terminated POP and POE bis(glycidyl ether) of molar masses ca 4,000 and 526 g.mol⁻¹. A series of hydrogels was obtained by swelling of the network in D₂O and excess volume of solutions of two surfactants: myristyltrimethylammonium bromide (C₁₄TAB) and sodium dodecylsulfate (SDS), respectively, in D₂O. Composition and structure of hydrogels were investigated by gravimetry and small-angle neutron scattering (SANS). A two-phase structure with Bragg's distance in the range ca 86 – 110 Å was revealed in the hydrogels obtained by swelling in neat D₂O, C₁₄TAB solutions and subcritical SDS solutions. Details of the hydrogels structure were obtained by fitting experimental SANS profiles to two model scattering functions. Either water-poor polydisperse nanodomains or micelles dispersed in the mayor water-rich phase are considered in the models. SANS results shown that macroscopic observations are accompanied by significant change of the two-phase structure of hydrogels.
VIBRATIONAL PHENOMENA IN GLASSES AT LOW TEMPERATURES CAPTURED BY FIELD THEORY OF DISORDERED HARMONIC OSCILLATORS

F. Vogel, M. Fuchs

University of Konstanz.

E-mail: florian.vogel@uni-konstanz.de

We investigate the vibrational properties of topologically disordered materials by analytically studying coupled harmonic ocillators in the thermodynamic limit at T = 0. Exploiting field theory, we build up a self-consistent field theory by analysing the Hessian utilizing Euclidean Random matrix theory. In accordance with earlier findings [1], we take non-planar diagrams into account to correctly address multiple local scattering events. By doing so, we end up with a first principles theory that can predict the main anomalies of athermal disordered materials, i.e the Boson peak, sound softening and a transition from Rayleigh damping to a weaker dependence of the sound attenuation around the loffe-Regel limit. Additionally, we argue that Rayleigh-damping implies a localisation of the eigenvectors of the random matrices. This is explained by the hybridisation of the phonons with quasi-localised modes (QLMs), which we rationalise the vibrational properties of disordered materials on a microscopic scale.

[1] T. Grigera et al., J. Stat. Mech. P02015 (2011).

EFFECT OF RECONDENSATION ON THE GLASS TRANSITION OF A CRYOFILM MIXTURE WITH NITROGEN

O. Vorobyova^{1,2}, D.Y. Sokolov^{1,2}, M.A. Ramos^{3,4}

¹Department of Thermal Physics and Technical Physics, Al-Farabi Kazakh National University, Al-Farabi Av. 71, Almaty 050040, Kazakhstan.

²Department of Machines and devices of manufacturing processes, Almaty Technological University, Tole Bi Av. 100, Almaty 050012, Kazakhstan.

³Departamento de Física de la Materia Condensada, Instituto "Nicolás Cabrera", Universidad Autónoma de Madrid, E-28049 Madrid, Spain.

⁴Campus de Cantoblanco, Centro de Micro-Análisis de Materiales (CMAM), Universidad Autónoma de Ma-drid, E-28049 Madrid, Spain.

E-mail: olga.vorobyova842@gmail.com

Different mixtures of cosmic ice have different sublimation temperatures, which under certain temperature conditions can lead to drastic changes in their structure. For example, for a two-component sample, when the medium temperature reaches a value above the sublimation temperature of the low-temperature component, it will evaporate, resulting in the release of the second component and its subsequent recondensation. Cryovacuum recondensates are prone to the formation of glassy states with their subsequent transformation with a change in temperature or pressure. However, these amorphous and polymorphic structures can only form at low and ultralow temperatures. Changing the thermal parameters (pressure and temperature) will also affect the structural transformations of cryocondensates, so these structures can be studied using infrared spectroscopy (IR spectrometry). Simple molecules such as H₂, N₂, CO, Ar and CH₄ adsorbed in amorphous or crystalline ice are often used as an IR label to study the properties of ice and glass. Experiments of the recondensation of thin films of waternitrogen mixtures were carried out at a pressure of 5 μ Torr. The mixture was applied in a cryovacuum chamber at a temperature of 16 K, at which a thin film 3.9 μ m thick was formed. The refractive indices of thin films of a mixture of water and nitrogen were calculated by the method of physical vapor deposition (PVD) and a two-beam interferometer in the range from 15 to 180 K

Measuring the dynamics of complex systems using coherent X-ray radiation at beamline P10

F. Westermeier¹, Z. Ren¹, N.D. Anthuparambil^{1,2}, V. Kartik¹, R. Rysov¹, D. Weschke¹, H. Xu¹, M. Sprung¹

¹Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany. ²Department of Physics, University Siegen, Siegen, Germany.

E-mail: fabian.westermeier@desy.de

One tool for the investigation of complex systems is synchrotron radiation, which allows probing inter alia optically opaque samples or the bulk of a sample system. While most synchrotron studies concentrate on the structural behavior of complex systems, usually on the nanometer to some hundreds of nanometers length scale, the use of a partially coherent X-ray beam allows to additionally investigate sample dynamics by using X-ray Photon Correlation Spectroscopy (XPCS).

One of the places to perform XPCS experiments is the Coherence Applications Beamline P10 at PETRA III, DESY, which is dedicated to coherent X-ray scattering experiments. It offers several experimental setups – two of these setups are especially well suited for XPCS. On the one hand a SAXS/WAXS instrument, where the detector can be translated between 0° and 30° at a sample to detector distance of 2 m or 5 m. And on the other hand a ultra-small angle X-ray scattering (USAXS) setup, where the detector is positioned at a sample to detector distance of around 21 m. This long pathway allows to use a large fraction of the coherent flux in an unfocused X-ray beam, while providing a fairly strong speckle visibility. This setup is therefore ideally suited for radiation sensitive samples such as most complex liquids, as the flux per sample area can be considerably reduced using a comparatively large X-ray beam. This contribution aims to highlight the experimental possibilities of these two setups.

Index

Adrjanowicz, Karolina, 5, 23 Alfinelli, Erica, 11, 24 Arrese-Igor, Silvia, 14, 18, 26, 111 Bachler, Johannes, 11, 27 Bafile, Ubaldo, 7, 28 Baglioni, Jacopo, 18, 112 Baron, Alfred Q.R., 11, 29 Bolhuis, Peter G., 13, 30 Brujic, Jasna, 5, 31 Bussmann, Lena, 18, 113 Capaccioli, Simone, 10, 18, 32, 114 Caporaletti, Federico, 10, 18, 33, 115 Caraglio, Michele, 16, 34 Cassetta, Michele, 7, 35 Chamchoum, Matteo, 16, 36 Chushkin, Yuriy, 10, 37 Cicerone, Marcus T., 10, 38 Colognesi, Daniele, 8, 39 Comez, Lucia, 40 Conti Nibali, Valeria, 10, 41 Corberi, Federico, 13, 42 Corezzi, Silvia, 16, 18, 43, 116 Cornet, Antoine, 17, 44 Cortes-Huerto, Robin, 13, 45 Costigliola, Lorenzo, 14, 46 Cristofolini, Luigi, 5, 47 Cuello, Gabriel, 18, 117 D'Angelo, Giovanna, 11, 48 Dallari, Francesco, 11, 49 Das Anthuparambil, Nimmi, 9, 25

Desantis, Fausta, 18, 118 Di Angelantonio, Silvia, 9, 50 Di Rienzo, Lorenzo, 15, 51 Dyre, Jeppe C., 5, 52 Fallah, Zohreh, 18, 119 Faraone, Antonio, 7, 53 Festi, Irene, 18, 120 Fiorentini, Raffaele, 18, 121 Fioretto, Daniele, 15, 54 Foret, Marie, 14, 55 Fujita, Tomoki, 8, 56 Fuoss, Paul, 11, 57 Gabriel, Jan Philipp, 10, 58 Gamba, Andrea, 10, 59 Ghamari, Danial, 10, 60 Giebelmann, Johannes, 18, 122 Golikov, Oleg, 18, 123 Gonzalez-Silveira, Marta, 13, 61 Gosti, Giorgio, 9, 62 Grassmann, Greta, 18, 124 Hansen, Jesper Schmidt, 8, 63 Holle, Nils, 8, 64 Keim, Peter, 14, 65 Konstantinou, Konstantinos, 8, 66 Krivchikov, Alexander I., 7, 67 Kuznetsov, Andrey, 16, 68 Lørup, Erik Hillo, 18, 125 Leonetti, Marco, 12, 69 Leuzzi, Luca, 12, 70

Maggi, Claudio, 9, 71 Martinelli, Alessandro, 11, 72 Mele, Margherita, 19, 126 Micheloni, Manuel, 19, 127 Milanetti, Edoardo, 19, 128 Miotto, Mattia, 15, 73 Mizuno, Hideyuki, 7, 74 Mocanu, Felix-Cosmin, 7, 75 Mosca, Ilaria, 10, 76 Mossa, Stefano, 11, 77 Mostarac, Deniz, 19, 129 Nabahat, Mehran, 17, 78 Napolitano, Simone, 13, 79 Niedda, Jacopo, 11, 80

Nilsson, Anders, 11, 81 Nyrkova, Irina A., 13, 82

Pabst, Florian, 13, 83 Paciaroni, Alessandro, 10, 84 Pagonabarraga, Ignacio, 13, 85 Parisi, Giorgio, 6 Paternoster, Costanza, 19, 130 Petersen, Charlotte F., 7, 86 Pineda, Eloi, 16, 87

Ragulskaya, Anastasia, 14, 88 Railton, Mark F.B., 19, 131 Ramos, Miguel Angel, 14, 19, 89, 132 Rodríguez–López, Marta, 14, 90 Ronca, Alberto, 19, 133 Rosenberg, Margaret, 16, 91 Rovigatti, Lorenzo, 5, 92 Ruiz-Ruiz, Marta, 19, 134 Russina, Margarita, 7, 93 Schirmacher, Walter, 7, 19, 94, 135 Schrøder, Thomas B., 13, 95 Semenov, Alexander N., 8, 136 Sevilla, Mauricio, 19, 137 Shen, Jie, 17, 96 Sokolov, Alexei P., 16, 97 Sonaglioni, Daniele, 19, 138 Sposini, Vittoria, 19, 139 Sun, Peihao, 8, 98 Szewczyk, Daria, 19, 140

Tamarit, Josep Lluís, 14, 99 Tarenzi, Thomas, 18, 19, 121, 130 Tavagnacco, Letizia, 16, 100 Tesei, Giulio, 5, 101 Tierno, Pietro, 14, 102 Tombari, Elpidio, 19, 141 Tropin, Timur, 16, 19, 103, 142 Tubiana, Luca, 15, 104

Vogel, Florian, 20, 143 Vorobyova, Olga, 20, 144

Walfort, Sebastian, 8, 105 Wei, Shuai, 5, 106 Westermeier, Fabian, 20, 145 Wyart, Matthieu, 5, 107

Zontone, Federico, 8, 108

Scientific Committee

Giacomo Baldi, University of Trento (Italy) Simone Capaccioli, University of Pisa (Italy) Joe Feldman, Naval Research Laboratory (USA) Aldo Fontana, University of Trento (Italy) Luca Leuzzi, CNR-Nanotech (Italy) Claudio Masciovecchio, Elettra Sincrotrone Trieste (Italy) Giulio Monaco, University of Padova (Italy) Kristine Niss, Roskilde University (Denmark) Kia Naai, University of Pisa (Italy) Raffaello Potestio, University of Trento (Italy) Giancarlo Ruocco, Sapienza Università di Roma & IIT (Italy) Beatrice Ruta, CNRS Lyon (France) Domenico Truzzolillo, CNRS Montpellier (France) Luca Tubiana, University of Trento (Italy) Emanuela Zaccarelli, CNR-ISC (Italy) Marco Zanatta, University of Trento (Italy) Alessio Zippo, University of Trento (Italy)

Organizing committee

Erica Alfinelli, University of Trento (Italy) *Giacomo Baldi*, University of Trento (Italy) *Fabio Brugnara*, University of Trento (Italy) *Irene Festi*, University of Trento (Italy) *Aldo Fontana*, University of Trento (Italy) *Roberto Menichetti*, University of Trento (Italy) *Raffaello Potestio*, University of Trento (Italy) *Marco Zanatta*, University of Trento (Italy)

Organised by







Supported by





event.unitn.it/complexsystems2023