Enzymes are Active Matter

Steve Granick and Tsvi Tlusty

IBS Center for Soft and Living Matter, South Korea

sgranick@gmail.com

tsvitlusty@gmail.com

The traditional view that enzyme kinetics is only a matter of catalyzing chemical reactions is challenged by recent experiments and theory showing that catalysis enhances enzyme mobility. This is significant to programming spatio-temporal patterns of molecular response to chemical stimulus. This talk will report that the enhanced diffusivity of enzymes is a "run-and-tumble" process analogous to that performed by swimming microorganisms, executed in this situation by molecules that lack the decision-making machinery of microorganisms. One consequence is that enzymes migrate in the direction of lesser reactant concentration when they turn over substrate; they display "anti-chemotaxis." This run-and-tumble process offers the possible biological function to homogenize product concentration, which could be significant in situations when the reactant concentration varies from spot to spot. Attempts will be made to place these and our related recent findings in the context of larger puzzles in the active matter intellectual community.

Cooperativity and frustration in the dynamics of flagella, cilia, and related systems

Nariya Uchida*

Department of Physics, Tohoku University

Flagella and cilia play vital roles in locomotion of aqueous microorganisms and material transport in the body of higher organisms. The motion a single flagellum or cilium in viscous fluids and their hydrodynamic properties are now understood quantitatively. On the other hand, collective motion of flagella and cilia still remains as an open problem, partly due to difficulty in measuring and controlling the motion of individual filaments. After introducing some recent results on single filament dynamics [1,2], we will show a couple of examples where collaborated experimental and theoretical efforts are made toward quantitative understanding of the cooperative dynamics of flagella and cilia.

In the first part, we consider non-equilibrium phase transition in bacterial carpets. Bacterial carpets are grafted monolayers of bacteria generating active surface flow. Onset of collective flow has been observed by tuning the flagellar activity, and mean-field-like critical behavior has been confirmed [3] in agreement with previous theoretical predictions [4]. We will also discuss the effect of impurities on the transition behavior.

In the second part, we address synchronized collective dynamics of cilia. We have emulated their periodic beating motion by optically driven colloidal particles orbiting circular paths. By tuning the flexibility of the orbit and the amplitude of the modulated driving force, we find that the both factors control the patterns of synchronization in a competitive manner [5].

The long-ranged hydrodynamic interaction also introduces inherent frustration in the collective behavior. In order to illustrate their effects, we will address the dynamics of non-locally coupled oscillators where spatio-temporal heterogeneity emerges from uniformly interacting elements.

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* uchida@cmpt.phys.tohoku.ac.jp

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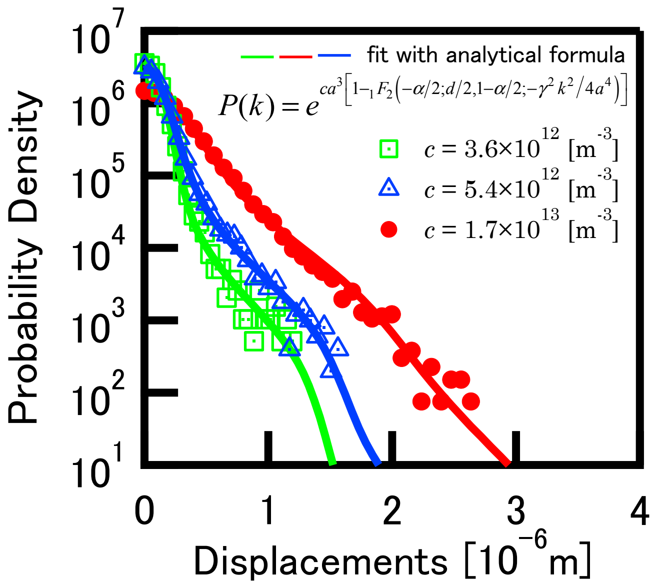
Non-Gaussian limit fluctuations in active swimmer suspensions

Daisuke Mizuno
Kyushu University, Fukuoka 812-8581, Japan

Hydrodynamic fluctuations in suspensions of swimming microorganisms (Chlamydomonas and E-coli) exhibit heavily-tailed distribution which is not Gauss nor Levy (Figure), for which both the classical and extended central limiting theory do not apply. In this study, the physical limit distribution, instead of mathematical ones, was derived in an analytical form by summing the general power-law interactions from field sources (here, swimming microorganisms) randomly distributed in general spatial dimensions. The origin of the non-Gaussianity is not just the power-law decay of hydrodynamic fields (simple mathematical sum of which should lead to Gauss or Levy distribution) but the summing procedure of the fields, which we refer to as the physical limit operation [1].

The non-Gaussian shape of the hydrodynamic fluctuations in active swimmer suspensions obeys the analytic theory concomitantly with independently determined parameters such as the strength of force generations and the concentration of Chlamydomonas. Time evolution of the distributions collapsed to a single master curve, except for their extreme tails, for which our theory presents a qualitative explanation. Investigations thereof and the complete agreement with theoretical predictions revealed broad applicability of the formula to dispersions of active sources of fluctuations [2].



Hydrodynamic fluctuations in active swimmer suspension

Fluctuations show up differently in different spatial dimensions since dimensionality affects the spatial correlations of fields (exponents of their power-law decays) and the population of field sources as a function of system size. Swimmers that resist to sedimentation or ordinary force-dipolar swimmers confined in 2D generate $1/r$-decaying fields that are long-ranged in the sense that there is no thermodynamic limit. As system size grows, the variance of fluctuations also increase as long as Stokes' approximation is valid. On the other hand, fluctuation distribution rapidly converges to Gauss. If time permitted, I will discuss the implication of our study for the non-Gauss fluctuations in Glassy systems.

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Active Soft matter Physics: From colloid to vibrobots

Harmut Löwen*
Institut für Theoretische Physik II: Weiche Materie
Heinrich-Heine-Universität Düsseldorf Universitätsstrasse 1

Ordinary materials are "passive" in the sense that their constituents are typically made by inert particles which are subjected to thermal fluctuations, internal interactions and external fields but do not move on their own. Living systems, like schools of fish, swarms of birds, pedestrians and swimming microbes are called "active matter" since they are composed of self-propelled constituents. Active matter is intrinsically in nonequilibrium and exhibits a plethora of novel phenomena as revealed by a recent combined effort of statistical theory, computer simulation and real-space experiments. After an introduction into the physics of active matter focussing on biological and artificial microswimmers as key examples of active soft matter [1], a number of single-particle and collective phenomena in active matter will be adressed ranging from active colloidal clusters [2] to inertial delay effects of granular vibrobots [3].

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* Corresponding author: hlowen@hhu.dec

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Orientational order and topological defects in biological active matter

Masaki Sano
Department of Physics, Graduate School of Science, University of Tokyo,
7-3-1 Hongo, Tokyo 133-0033, Japan

Active systems can exhibit greater variety of ordered states than conventional equilibrium systems. They consist of a sufficiently high density of particles like filamentous protein, bacteria, cells, vibrated granular rods, fish and birds that are either self-propelled or actively driven. Broken time reversal symmetry at the particle level in these systems is distinct from conventional physical systems. Those systems exhibit intriguing behaviors such as solitary moving band, moving droplet, and long range orientational order. Thanks to recent development of active matter research, some of complex behaviors in dense population of biological cells are now within reach of theoretical and experimental physics. In this talk, I will give two examples with different symmetries. Recently, we reported intriguing collective behaviors in tissue of neural stem cells (NSC). Elongated NSC created long range nematic order with topological defects. We found that topological defects control collective behavior such as accumulation or dilution of cell density[1]. These behaviors can be described by a simple continuum theory of active matter. Equations governing the dynamics can be deduced from data of traction force microscopy and individual cell motions.

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Nonequilibrium Self-Organization Phenomena in Droplet Spinodal Decomposition

Che-Min Chou*[,a], Wei-Tsung Chuang[a], and Po-Da Hong[b]
[a]*National Synchrotron Radiation Research Center, Hsinchu, 30076, Taiwan*
[b]*Department of Materials Science and Engineering, National Taiwan University of Science and Technology, Taipei, 10607, Taiwan*

In the spinodal decomposition (SD), the simulations and experiments have shown that the percolation can occur only for the volume fraction of the minority phase $\Phi \geq 0.36$, and the remainder would undergo the so-called "droplet-SD" [1,2]. Recently, Shimizu and Tanaka [1] reported that the Marangoni force is the main reason for the collision and coalescence of the droplets. However, another easily observed phenomenon is the collision-induced collision via flow [2,3], where the droplet coalescence leads to an outward flow field which would significantly accelerate the next coalescence. It would be important to clarify whether each droplet feels the active or the passive hydrodynamic effects. At this point, we need the exact structural information of the droplet-SD, which is implicitly contained in the small-angle-scattering (SAS) experiments.

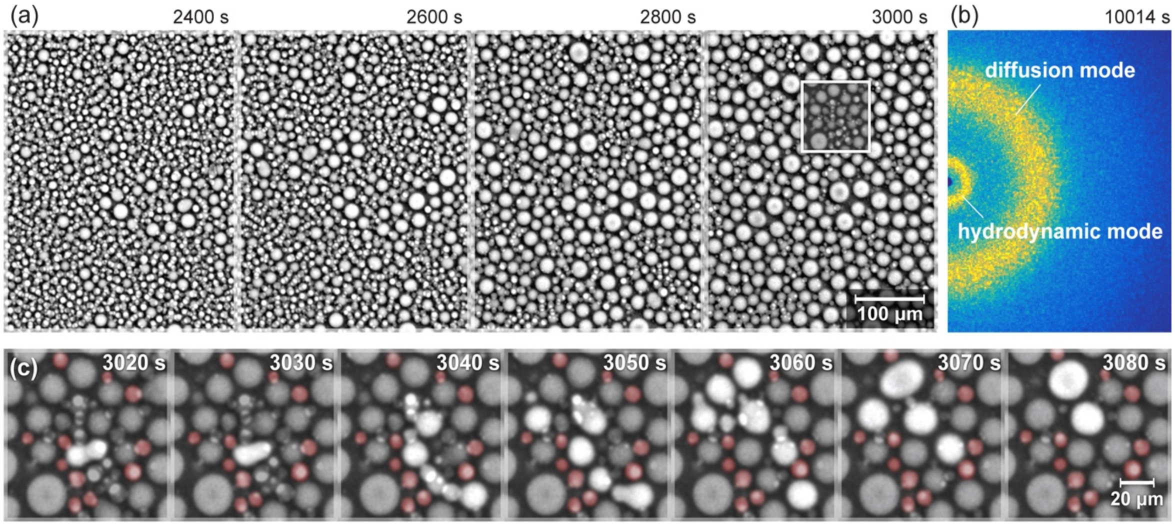

Figure 1 (a) Morphologies evolution of the droplet spinodal decomposition for the $\phi = 0.54$ polymer mixture at 363K. (b) Double-ring pattern of polarized scattering. (c) A series of chain collisions (highlight) and the unperturbed small droplets (pseudo-red color).

By solving the inverse SAS problems, we yield quantitatively accurate descriptions of the droplet-SD structure, especially in the droplet-size distribution, the well-defined short-range order, and the stress-optical phenomenon. Since the "attractive" hydrodynamic effect between the droplets is not so obvious in the early coarsening, the Brownian-coagulation mechanism is still a candidate. Further, at later times, the most striking feature is the outward flow field induced by the shape relaxation of coalescing droplets which looks like a "ripple" propagating and triggering a series of chain collisions. It is interesting that the phenomenon seems to be a nonequilibrium self-organization process and can occur only if the size of the coalescing droplets is greater than a threshold value.

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*Corresponding author: chou.cm@nsrrc.org.tw

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Multi-scale Design of Hydrogels with Reversible Sacrificial Bonds
– From Toughness to Adhesion to Composites –

Jian Ping Gong
Faculty of Advanced Life Science,
Global Station for Soft Matter GI-CoRE, Hokkaido University
Sapporo 001-0021, Japan
gong@sci.hokudai.ac.jp

Invention of the tough double network hydrogels (DN gels), consisting of interpenetrated rigid/brittle network and soft/stretchable network, shows that the effective energy dissipation by the breaking of the covalent bond of the brittle network prevents catastrophic crack propagation upon deformation, and thus, gives the extraordinarily high toughness of the material[1]. Such sacrificial bond effect has been successfully applied to develop tough double network hydrogels of diverse chemistry and also to double and triple network elastomer materials. Thus, sacrificial bond concept is proved to be a general approach for developing tough soft materials.

As the internal rupture of DN gels is due to the irreversible breaking of the covalent bonds of the brittle network, the conventional DN gels deteriorate gradually after repeated deformation. To address this problem, many recent works have replaced the covalent bonds with non-covalent bonds to allow the sacrificial bonds to be reformed.

In this talk, novel hydrogels with reversible sacrificial bonds developed in author’s group are reviewed and their excellent mechanical behaviors such as high toughness, self-healing [2,3], adhesion to biological tissues[4], and fast underwater adhesion are demonstrated[5]. Furthermore, this principle is extended to develop tough composites using tough hydrogels as energy dissipative soft matrix[6].

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Fate of accelerating slip on self-affine rough interfaces

So W. Ozawa, Takahiro Hatano*, and Nobuki Kame
Earthquake Research Institute, The University of Tokyo

We consider friction dynamics on a rough interface that has fractal height profile. First we review the universal feature of unstable accelerating slip on a flat interface [1] and the underlying friction law [2]. We then discuss how the interface roughness affects this universal feature. The roughness profile is characterized by three parameters: the RMS amplitude, the minimum cutoff wavelength of self-affine structure, and the Hurst exponent. We show that their effects on slip dynamics are described by a single parameter. This is interpreted as an effective stress that inhibits slip. If this effective stress becomes comparable to the driving stress, the slip is stabilized and does not accelerate. We discus the relevance of this result to earthquake faults, which are in general self-affine [3].

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* Corresponding author: hatano@eri.u-tokyo.ac.jp

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Density fluctuations and solute-induced phase separation in a fluid mixture composed of a binary solvent and a nonionic hydrophobic solute

Ryuichi Okamoto[1]* and Akira Onuki[2]

[1]*Research Institute for Interdisciplinary Science, Okayama University*

[2]*Department of Physics, Kyoto University*

We study density fluctuations and phase separations in fluid mixtures in which a small amount of hydrophobic solute can significantly alter the phase behaviour. First, assuming small solute density and a binary solvent, we discuss thermodynamic stability and density fluctuations in a one-phase state, and present a new expression for the second osmotic virial coefficient B_2. When the solute density exceeds the spinodal value $n_{\rm spi} \simeq -1/2B_2$, the fluid undergoes solute-induced instability. Our calculation based on the Mansoori-Carnahan-Starling-Leland model reveals that $n_{\rm spi}$ exhibits a minima at an intermediate solvent composition, owing to the coupling between the solvent composition fluctuation and the solute density. We then discuss gas-liquid and liquid-liquid phase transitions induced by a small amount of a solute. In the latter we calculate the binodal and spinodal curves in the triangular phase diagram, and discuss nucleation.

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* Corresponding author: `okamoto-ryuichi@okayama-u.ac.jp`

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Drag Coefficient of a Circular Liquid Domain in a Near-Critical Binary Fluid Membrane

Youhei Fujitani*
School of Fundamental Science & Technology,
Keio University, Yokohama 223-8522, Japan

Hisasi Tani
Department of Mechanical Engineering, Texas A & M University, TX 77843-3123, USA

We calculate the drag coefficient of a circular liquid domain, which is placed in a flat fluid membrane composed of a binary fluid mixture lying in the homogeneous phase near the demixing critical point. Assuming a sufficiently small correlation length $\xi_{\rm c}$, we regard the domain dynamics as independent of the critical fluctuation and use the Gaussian free-energy functional for the mixture. Because of the near-criticality, the preferential attraction between the domain component and one of the mixture components generates a significant composition gradient outside the domain, which can affect the drag coefficient. Writing $\eta_{\rm i}$ and $\eta_{\rm o}$ for the membrane viscosities inside and outside the domain, we first consider the case of $\eta_{\rm i} = \eta_{\rm o}$. The drag coefficient is expanded with respect to a dimensionless strength of the preferential attraction, λ. It is numerically shown that the magnitude of the expansion coefficient markedly decreases as the order of λ increases and that the first-order term, *i.e.*, the term linear to λ, usually gives a good approximation for practical material constants. The effect of the preferential attraction is shown to be able to become significantly large in practice. We secondly consider cases of $\eta_{\rm i} \neq \eta_{\rm o}$ by utilizing the calculation procedure devised in Ref. 1. The first-order term of the expansion series with respect to λ decreases to approach zero as $\eta_{\rm i}$ increases to infinity. This agrees with previous numerical results showing that the hydrodynamics makes the effect of the preferential attraction negligibly small for a rigid disk [2]. Details are published in Ref. 3, where some errors in Ref. 4 are corrected and and the results of Ref. 5 regarding the transport coefficients in the near-critical fluid membrane are improved and extended.

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* Corresponding author: `youhei@appi.keio.ac.jp`

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Interfacial tensions near the critical endpoints and the tricritical point of three-phase equilibria: Mean-field density-functional model

Kenichiro Koga and Joseph O. Indekeu
Okayama University, Japan.
koga@okayama-u.ac.jp

We investigate a mean-field density-functional model of the structures and tensions of the interfaces between fluid phases near the tricritical point of three-phase equilibrium. The main objective is to examine a critical exponent associated with the vanishing interfacial tension on approach to a critical endpoint. Specifically, we consider three fluid phases α, β, and γ in equilibrium, with the interfacial tensions $\sigma_{\alpha\beta}$, $\sigma_{\beta\gamma}$, $\sigma_{\alpha\gamma}$. (The $\alpha\gamma$ interface may be wet by β or three interfaces may meet at a line of common contact.) A class of density-functional models describes the variation of those tensions as the three-phase region is traversed from the $\alpha\beta$ critical endpoint, where $\sigma_{\alpha\beta} = 0$, to the $\beta\gamma$ critical endpoint, where $\sigma_{\beta\gamma} = 0$ at any one temperature, and how one of the tensions vanishes on approach to the critical endpoint:

$$\sigma_{\beta\gamma} \sim \text{const}\,.\,(\sigma_{\alpha,\beta\gamma} - \sigma_{\alpha\beta})^{\mu},$$

where $\sigma_{\alpha,\beta\gamma}$ is the tension between the noncritical α phase and the critical $\beta\gamma$ phase. An analytically soluble one-density model gives $\mu = 3/2$ while $\mu \sim 1.3$ by experiment [1]. A recent study of two different two-density models, supposedly more realistic than the one-density model, found different values of μ, 3/2 and 3 [2]. The present study accounts for the inconsistency in the earlier models. Furthermore, we conjecture an "exact" expression of the interfacial tensions in the class of density-functional models, from which one may explore the phase diagram near the tricritical point [3].

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Water in a Capacitor: Structure, Fluctuation, and Response

Kyohei Takae[1]* and Akira Onuki[2]
[1]*Institute of Industrial Science, University of Tokyo*
[2]*Department of Physics, Kyoto University*

The dielectric response of condensed matter is often studied under the assumption that the external electric field is a controllable parameter. However, it is needed to solve the Poisson equation under an appropriate boundary condition to obtain the macroscopic electric field, because it depends on the geometry and the polarization of the system. We construct a method of molecular simulation for calculating the electrostatic interactions among charged particles between parallel metallic plates [1]. By applying this method, we study structure and dynamics of water in a capacitor [2,3]. Image interaction arising from the dielectric discontinuity at the interface induces polarization near the surface, which decreases the bulk electric field. Homogeneous correlation of polarization fluctuation emerges in this geometry (see Fig.1). However, the dynamics of each molecule is governed by the local electric field, which is considerably larger than the macroscopic electric field. The electrostatic interactions between the nearest and second-nearest molecules are dominant contribution to the local electric field, and therefore the responses of water molecules to external voltage applied on the electrodes becomes highly cooperative.

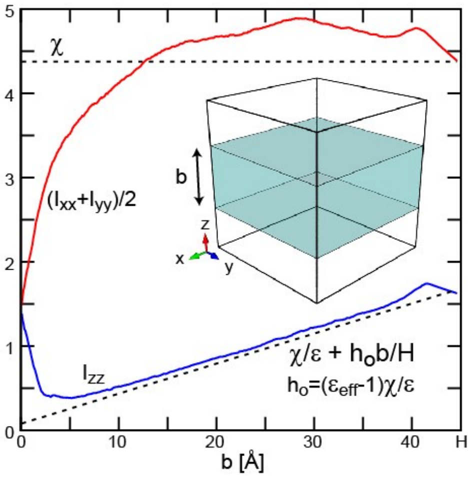

FIG. 1: Space integral of the polarization correlation function in shaded region is displayed. Homogeneous contribution is observed for perpendicular (zz) component.

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* Corresponding author: takae@iis.u-tokyo.ac.jp

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Collective dynamics of externally driven colloids along a rectilinear path

T. Taniguchi,* K. Teshigawara, J. Molina, and R. Yamamoto
Department of Chemical Engineering, Kyoto University, Kyoto 615-8510, Japan

N. Oyama
Mathematics for Advanced Materials. OIL, AIST.
Tohoku University, Sendai 980-8577, Japan

The collective dynamics of externally driven N_p-colloidal systems (N_p=1,2,3,4) in a confined viscous fluid (See Fig.1) have been investigated using three-dimensional direct numerical simulations with fully resolved hydrodynamics. The dynamical modes of collective particle motion are studied by changing the particle Reynolds number as determined by the strength of the external driving force and the confining wall distance. For a system with $N_\mathrm{p} = 3$, we found that at a critical Reynolds number, a dynamical mode transition occurs from the doublet-singlet mode to the triplet mode, which has not been reported experimentally. For a system with $N_\mathrm{p} = 4$, we found similar dynamical mode transitions from the doublet-singlet-singlet mode to the triplet-singlet mode and further to the quartet mode[1].

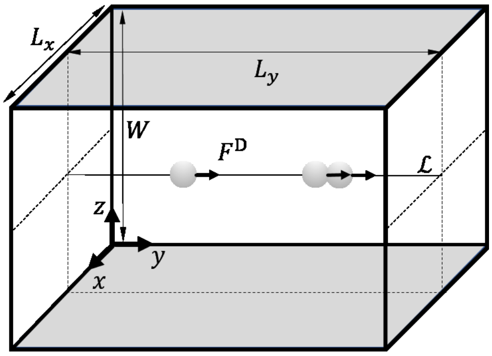

FIG. 1: Schematic picture of the system. The particles are driven along the rectilinear line $\mathcal{L}$ located at the center line zx-plane. The system size is $L_x \times L_y \times W$ ($L_x = L_y = 64D/3$, $2D \leq W \leq 64D/3$, where D is the diameter of a particle)

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[*] Corresponding author: `taniguch@cheme.kyoto-u.ac.jp`

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Jones-Ray effect can be explained by charged impurities

Yuki Uematsu*
Department of Chemistry, Kyushu University

Surface charges of air-water interface are one of the central problems in physical chemistry. Although electrophoretic measurements of bubbles in pure water imply negative surface charges at neutral pH, i.e. OH^- adsorption, most of other experimental and simulation studies suggest H_3O^+ adsorption and OH^- depletion. In my talk, I focus on the surface tension of air-electrolyte interfaces and Jones-Ray effect which is a minimum of the surface tension at millimolar electrolyte concentration. The theoretical model including ion-specific affinity to the surface is constructed and agrees with the experimental data for high concentration. Since at low concentration the minimum does not appear, we consider impurity effect in the solutions. Then, we theoretically demonstrate that when the impurity is charged and surface-active (like SDS), only the nanomolar concentration is enough to induce the minimum observed in experiments [1]. We also performed experiments and show that intentionally added ionic surfactants at micromolar concentration cause the minimum, which perfectly agrees with the model [2]. These results suggest that the Jones-Ray effect was caused by unexpected contamination of the solution with charged impurities, which does not contradict with H_3O^+ adsorption, OH^- depletion, and negative zeta potential assuming the impurity is negatively charged [3].

This work has been done in collaboration with D. J. Bonthuis, R. R. Netz, K. Chida, and H. Matsubara.

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* Corresponding author: `uematsu@chem.kyushu-univ.jp`

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Theory of linear response in glasses: Shear modulus and dielectric permittivity

Akira Onuki[a], Takeshi Kawasaki[b], and Kyohei Takae[c]

[a] Kyoto University, [b] Nagoya University, [c] University of Tokyo.

We construct a linear response theory[1] of applying shear strains from boundary walls with the Hamiltonian $\mathcal{H}-\gamma_{\rm ex}(t)\mathcal{A}$. Here, $\gamma_{\rm ex}(t)$ is the applied mean strain and $\mathcal{A}$ is its conjugate variable determined by the particles near the walls. Our theory is applicable to any solids and fluids. For glasses, we assume quasi-equilibrium around a fixed inherent state. Then, we obtain linear-response expressions for any variables using the inherent average $\langle\cdots\rangle_{\rm ih}$ despite the interior inhomogeneity. Let us consider the response function $\boldsymbol{\chi}_i(t) = \langle \boldsymbol{u}_i(t)\mathcal{A}(0)\rangle_{\rm ih}/k_BT$ for the displacement $\boldsymbol{u}_i$ of particle i. In Fig.(a), we show the response $\boldsymbol{\chi}_i(0)$ at steady strain, where the affine and nonaffine parts coexist. In (b,c) we show the response $\boldsymbol{\chi}_i(0)-\boldsymbol{\chi}_i(t)$ after a stepwise strain, where ω_1 is the lowest vibrational frequency. Here, shock-like transverse sounds propagate after the wall motion. For oscillating strain, we also find resonant sound amplification when the frequency ω approaches that of the first transverse sound mode.

We also consider ferroelectric glasses[2], where the critical fluctuations near the transition freeze due to impurities at low T. The dielectric permittivity $\epsilon'(\omega) - i\epsilon''(\omega)$ exhibits the characteristic features of relaxors. In (d), we show an example of ferroelectric domains.

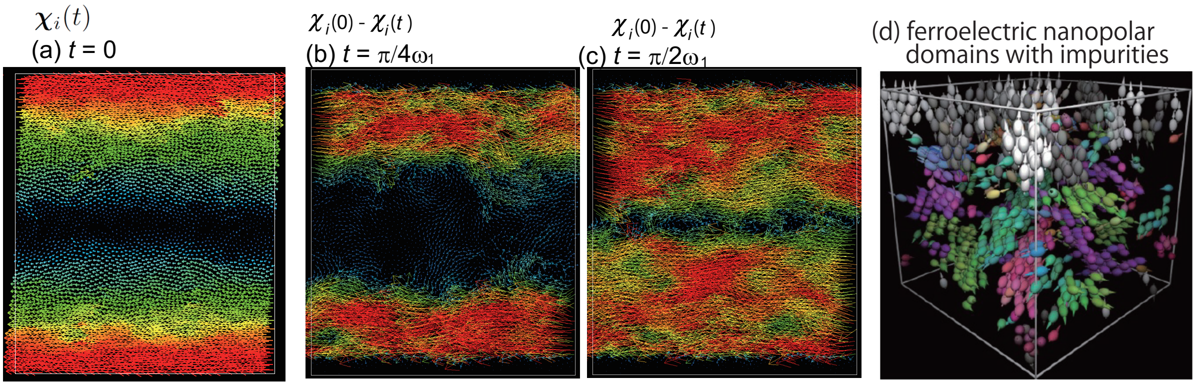

FIG. 1: (a) Static rsponse to applied shear strain. (b,c) Displacements after a stepwise shear motion of the walls. (d) Ferroelectric nanopolar regions in a glassy state.

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Rigid-body Rotations of Chiral and Achiral Liquid Crystalline Droplets Driven by Linear Fluxes

Yuka Tabe,[1,2] **, Shinji Bono**[1]**, Shin-ya Sugisawa,**[1,2] **and Chuhei Oshima**[1,2]

[1] **Department of Pure and Applied Physics, Waseda University, Tokyo, Japan**
[2] **Kagami Memorial Research Institute for Materials Science and Technology, Waseda Univ.**

Linear flows often drive unidirectional and coherent molecular rotations in chiral liquid crystals (LCs). In this report, we show two examples of rigid-body rotations of LC samples composed of chiral and achiral compounds, respectively. The first one is a heat-driven rigid-body rotation of cholesteric droplets coexisting with isotropic media. When a heat flux is given to a cholesteric droplet, the director is known to exhibit unidirectional rotation, but it has been questioned whether the heat flow could also drive the rigid-body rotation of the whole cholesteric droplet. In order to answer the question, we manipulated aggregates of cholesteric cylinders characterized by double-twist structure and observed their motion under heat flux. Since the aggregates of the cylinders don't possess the cylindrical symmetry, it is easy to distinguish the rigid-body rotation from the director rotation under optical microscopy. The experimental result showed that the aggregates rotated as a rigid body, which was observed even under non-polarized light. The angular velocity of the aggregates increased with the heat flux and decreased with the number of the cylinders, which suggests that the angular momentum of the director rotation is converted into that of the rigid-body rotation.

Another example is an untypical rotation of non-chiral LC films driven by normally incident electron beam. We used a 1:1 mixture of achiral LC compounds taking Cry-E phase, which is characterized by a layered structure and an in-plane herringbone lattice. When a free-standing film of the Cry-E sample was irradiated by a low-energy electron beam, a clear diffraction pattern was obtained successfully, corresponded to the herringbone lattice in the film plane. Then, we found that the diffraction pattern started a unidirectional and continuous rotation with keeping the shape, indicating the rigid-body rotation of the film itself. We tested more than 100 films composed of the same mixture and found both clockwise and counter clockwise rotations appeared at the equal ratio. Although the mechanism has not been revealed yet, the induced chirality originated from the alternative molecular arrangement possibly determines the rotational direction.

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Helical Inversions of Chiral Liquid Crystalline molecules

Akihiko Matsuyama*
Kyushu Institute of Technology, Fukuoka, Japan

A mean field theory is introduced to describe a helical inversion in nematic-cholesteric mixtures and binary cholesteric mixtures.[1] Taking into account a chiral coupling between unlike molecules,[2] the helical pitch is derived as a function of orientational order parameters and concentration. We find the conditions of helical inversions for the mixtures and derive the concentration of the helical inversions, depending on the chiral interaction. The numerical results are in agreement with the experimental results.[3] We also examine phase separations on the temperature-concentration plane, including the helical inversions. A large variety of phase separations, such as the two-phase coexistence between a right-handed rich and a left-handed rich phase, etc., are predicted.

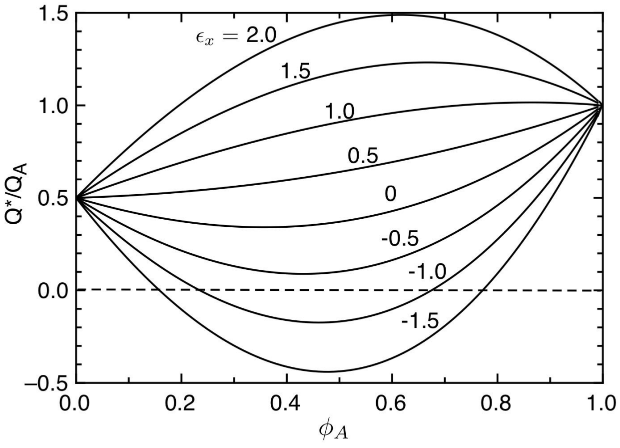

FIG. 1: Pitch wavenumber Q^*/Q_A plotted against the concentration ϕ_A for various values of $\epsilon_x (= \lambda_x/Q_A)$ at $\tau = 0.92$ in the binary cholesteric mixture with a right handed helix of $Q_A = 0.01$ and $Q_B = 0.005$. For larger values of the ϵ_x, the wavenumber has a maximum as a function of ϕ_A.

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* Corresponding author: `matuyama@bio.kyutech.ac.jp`

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Exotic mesoscale structures in a thin film of a chiral liquid crystal

Jun-ichi Fukuda*

Department of Physics, Kyushu University

Liquid crystal mesophases without rotational and/or translational symmetry have fascinated physicists because they provide intriguing examples of self-organized mesoscopic structures. A chiral liquid crystal exhibits spontaneously twisted orientational order that is understood by the presence of a pseudo-scalar term in the free energy in terms of the orientational order parameter. By numerical calculations based on a Landau-de Gennes theory describing the orientational order by a second-rank tensor Q_{ij}, we show [1] that a thin film of a chiral liquid crystal can exhibit various exotic mesoscale structures that are not seen in bulk liquid crystals. These structures are understood as a result of frustrations between the bulk ordering and the surface anchoring, and involve topological line defects of orientational order (disclination lines), and also Skyrmions, vortex-like topological entities that have been shown to emerge in various condensed matter systems. We also show[2] that some of such structures, including a hexagonal lattice of Skyrmions, are indeed observed experimentally by optical measures, and experimental results are perfectly accounted for theoretically by solving Maxwell equations for light waves. Liquid crystals offer an interesting platform for the investigation of various self-organized structures involving topological defects, and a Landau theory provides a powerful tool for the study of such structures of a liquid crystal.

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* Corresponding author: `fukuda.jun-ichi@phys.kyushu-u.ac.jp`

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Ferroelectric-like Order along the Director in Fluid Liquid Crystals

H. Nishikawa[1], K. Shiroshita[2], H. Higuchi[1], Y. Okumura[1],
Y. Haseba[3], S. Yamamoto[3], K. Sago[3], and H. Kikuchi[1*]

[1]Institute for Materials Chemistry and Engineering (IMCE), Kyushu University
[2]Interdisciplinary Graduate School of Engineering Sciences, Kyushu University
[3]JNC Petrochemical Corporation

The dioxane type liquid crystal (DIO) is found to show a surprisingly large positive Δε that is several hundred times larger than those of conventional liquid crystals. X-ray diffraction observed that there was no long-range periodic structure like a smectic layer in LC phases of DIO. The exceptionally large dielectric anisotropy larger than or equal to 10000 of DIO was observed at a LC region, suggesting that highly polar ordering could be spontaneously generated. The DIO also exhibited a parallelogram-shaped *P*-*E* hysteresis loop which is observed generally in ferroelectric materials. Furthermore, considerably large second harmonic generation is detected along the director. These results indicate clearly that a unidirectional, ferroelectric-like parallel polar arrangement of the molecules is generated along the director in the liquid crystal phase of DIO which is highly fluid.

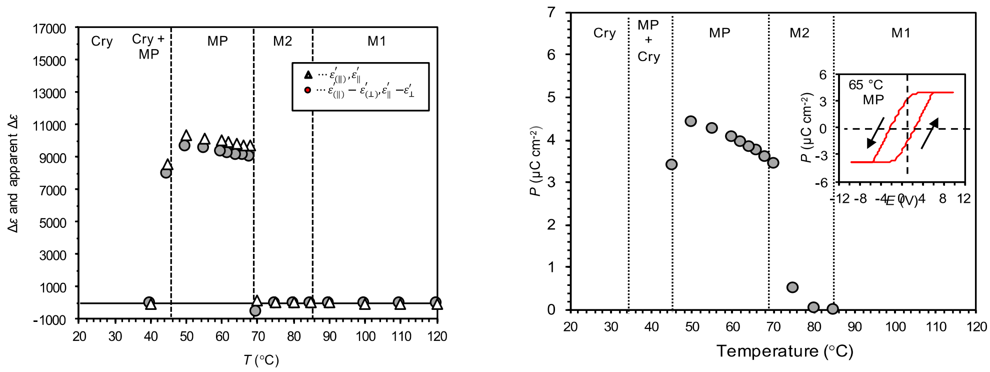

FIG.1 : Temperature dependences of dielectric permittivity along the director and dielectric anisotropy (left) and switching polarization of DIO (right).

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* Corresponding author: kikuchi@cm.kyushu-u.ac.jp

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The ideal glass: More than just vibrations

Walter Kob*

Laboratoire Charles Coulomb, University of Montpellier

Recent theoretical work suggests that glass-forming systems will make an equilibrium ideal glass transition if a certain number of particles are frozen in a permanent manner. We use this approach to probe the properties of such an ideal glass and find that contrary to naïve expectation, the motion of the fluid particles is not purely vibrational. These non-trivial excitations have a consequence for the entropy and lead to a non-trivial relaxation dynamics of certain observables.

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* Corresponding author: walter.kob@umontpellier.fr

Skating and skiing: the surface of ice and snow

Daniel Bonn

For ice skating, we have investigated the friction coefficient of steel on ice over a large temperature range, revealing a very high friction at low temperatures (-100'C) and a steep decrease in the friction coefficient with increasing temperature. Very low friction is only found over the limited temperature range typical for ice skating. The strong decrease in the friction coefficient with increasing temperature exhibits Arrhenius behavior with an activation energy Ea ~ 11.5 kJ/mol. Remarkably, molecular dynamics simulations of the ice-air interface reveal a very similar activation energy for the mobility of surface molecules. Weakly hydrogen-bonded surface molecules diffuse over the surface in a rolling motion, their number and mobility increasing with increasing temperature. This correlation between macroscopic friction and microscopic molecular mobility indicates that slippery ice arises from the high mobility of its surface molecules.

For skiing, we have investigated the mechanism of ageing of snow. This is also important since the evaporation (sublimation) of ice and snow has a major impact on global climate, since the amount of ice and snow determines Earth’s albedo. Yet, due to their complex geometry with several sharp regions which are singular for the evaporation, the precise evaporation dynamics of snow and ice crystals remains challenging to predict. We studied the sublimation of snowflakes and pointy ice drops and show that, surpsrisingly, the evaporation rates of water and ice drops are similar; they are both limited by the diffusive transport of the vapor. This allows us to predict ice and snowflake evaporation quantitatively by solving the diffusive free-boundary problem, which correctly predicts the rapid self-similar rounding of sharp edges and points: this is the reason why skiing in freshly fallen snow is different.

Transient stagnant formation in a thermal convection

Rei Kurita* and Kazuya U. Kobayashi
Department of Physics, Tokyo Metropolitan University

Convection driven by the density difference between horizontal fluid layers when they are heated from below is called Rayleigh-Bénard convection (RBC). RBC is chiefly governed by the non-dimensional Rayleigh number, defined as $Ra = \frac{g\alpha\Delta T H^3}{\nu\kappa}$, where g, α, ΔT, H, ν and κ are acceleration due to gravity, the thermal expansion coefficient, the temperature difference across the fluid, sample height, kinematic viscosity and thermal diffusivity, respectively.

Recently, we have studied thermal convection near the critical Rayleigh number in binary solutions. We observe that a domain without flow is transiently formed in an upwelling near the upper surface and the mode of heat transfer in the fluid changes repeatedly between convection and conduction over time [1, 2]. Furthermore, we also observed transient stagnant domain (TSD) in thermally driven convective flow. It is important to understand the origin of the transient stagnant domain formation since it induces a large deformation of convection patterns. Here, we find that stagnant domain formation is ubiquitous in two component mixtures. In addition, we find that difference in viscosity between the two components is crucial for transient stagnant domain formation, more so than the concentration gradient induced by the temperature gradient [3].

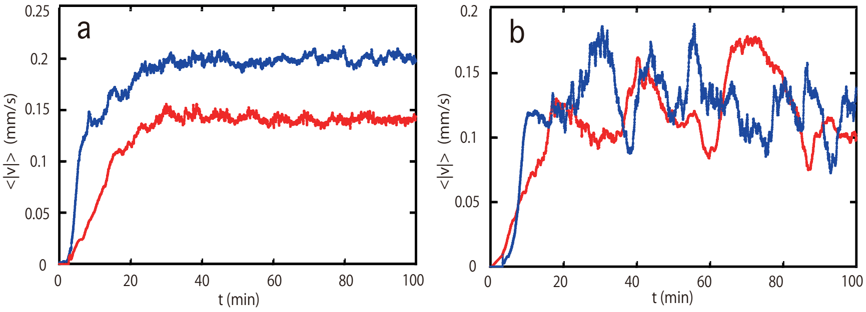

FIG. 1: Time evolution of the average velocity near the top surface in glycerol and silicone oil. (a) shows results for pure glycerol (blue line) and silicone oil 50cs (red line). (b) shows results for 60 % glycerol (blue line) and 1cs/100cs silicone oil mixtures (red line).

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Fluctuation of Diffusion Coefficient in Coarse-Grained Models of Entangled Polymers

Takashi Uneyama*
Graduate School of Engineering, Nagoya University

Polymer melts and solutions with relatively large molecular weights exhibit characteristic relaxation behavior due to so-called the entanglement effect. The entanglement effect originates from the uncrossability between chain-like molecules, and the motion of the chains are strongly constrained. To reproduce the entanglement effect in coarse-grained simulations, various phenomenological models such as the tube model have been proposed.

Recently, the diffusion behavior of the center of mass of an entangled polymer chain has been studied for several different coarse-grained models. The diffusion coefficient for the center of mass was shown to be not constant but fluctuate in time[1]. The center of mass position $\boldsymbol{r}_{\mathrm{CM}}$ obeys the following Langevin equation with the fluctuating diffusivity:

$$\frac{d\boldsymbol{r}_{\mathrm{CM}}(t)}{dt} = \sqrt{2}\boldsymbol{D}^{1/2}(t) \cdot \boldsymbol{w}(t). \tag{1}$$

Here $\boldsymbol{w}(t)$ is a Gaussian white noise which has a unit variance ($\langle \boldsymbol{w}(t) \rangle = 0$ and $\langle \boldsymbol{w}(t)\boldsymbol{w}(t') \rangle = \mathbf{1}\delta(t - t')$, with $\langle \dots \rangle$ representing the statistical average) and $\boldsymbol{D}^{1/2}(t)$ is the matrix square root (Cholesky decomposition) of a time-dependent and fluctuating diffusion coefficient tensor. $\boldsymbol{D}(t)$ obeys another stochastic process and thus it fluctuates in time. In this work, we study the diffusion behavior of the centers of mass of entangled polymers by the simple reptation model and simulations with a responsive particle dynamics (RaPiD) type model[2].

In the reptation model, the diffusion coefficient of the center of mass reflects the conformation of the polymer chain (the end-to-end vector), and thus it fluctuates. The simulation results also show large fluctuations for the diffusion coefficient. In the RaPiD type model, a polymer chain is modelled as a single particle without any internal degrees of freedom, and thus this fluctuation cannot be related to the chain conformation. The fluctuation of the diffusion coefficient in the RaPiD type model originates from the entanglements between different chains. Threfore, both the conformation of a chain and the entanglements between chains contribute to the fluctuating diffusion coefficient.

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* Corresponding author: `uneyaam@mp.pse.nagoya-u.ac.jp`

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A non-equilibrium phase transition in particle trajectories near the jamming transition

Takeshi Kawasaki* and Kunimasa Miyazaki
Department of Physics, Nagoya University

Kentaro Nagasawa
Department of Physics, University of Tokyo

The reversible-irreversible (RI) transition of particle trajectories in colloidal suspensions under cyclic shear deformation is an archetypal non-equilibrium phase transition and attracts much attentions. In the low density limit, the RI transition is predicted to belong to a universality class of the absorbing state transitions [1], whereas at the high densities well above the jamming transition density, $\varphi_{\rm J}$, it has more to do with the yielding transition of the amorphous solids [2]. The link to bridge the gap between the low and high densities is missing and the relation of the RI transitions with mechanical and flowing behaviors of colloidal suspensions are largely unexplored. In this study, we investigate the RI transitions over a wide range of densities above and below $\varphi_{\rm J}$ by using oscillatory sheared molecular dynamics simulation. It is revealed that the nature of the RI transitions dramatically change across $\varphi_{\rm J}$. When the density is above $\varphi_{\rm J}$, the discontinuous RI transition and concomitant yield transition are observed. Below $\varphi_{\rm J}$, however, the nature of the RI transition becomes surprisingly rich. We find the three distinct phases; (i) continuous RI transition at small amplitudes and low densities followed by (ii) reentrance to the reversible phase at larger strain amplitudes, and (iii) semi-discontinuous RI transitions in the vicinity of $\varphi_{\rm J}$. Here, we have confirmed that these results are quantitatively agreed with those obtained from the simulations in the athermal quasi-static (AQS) limit. We show that these transition behaviors are strongly correlated to the number of the contacts, characterized in AQS limit. This implies that these distinct transitions strongly correlated with hidden geometrical properties of particle configurations. We also show that the RI transition in the regime (iii) is even related with another non-equilibrium transition called the shear jamming.

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* Corresponding author: `kawasaki@r.phys.nagoya-u.ac.jp`

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Glassy cytoplasm driven by non-thermal forces

Kenji Nishizawa*, Daisuke Mizuno
Dept. of Phys., Kyushu Univ., Fukuoka, Japan.

Cell interiors are crowded with various soft objects which is furthermore driven out of equilibrium with metabolic activity. The effect of "crowding" and "activity" on the cell mechanics presents intriguing challenge which we here investigates by performing microrheology experiments in cell extracts and in living cells.

For that purpose, we prepared several models of cytoplasm which lacks cytoskeletons (E. coli, Xenopus eggs, and HeLa cells). We found that viscosity of cell extracts showed super-exponential increase with the protein concentrations. Furthermore, viscoelastic properties of cytoplasm in the living cultured cells (HeLa cells) were measured by optical-trap-based microrheology under 3-dimentional feedback of a piezo-mechanical sample stage [1]. Volume fraction of intracellular macromolecules was changed in the range of (0.2 - 0.6 g/ml) by adjusting the osmotic pressure. We found that the viscosity in living cells showed purely exponential increase with respect to the macromolecule concentration.

The Angell plot of our data clarified that the cell extract and living cytoplasm is a fragile and a strong glass former, respectively (Fig. 1). Glasses are fragile when their components cannot individually relax local frustrations in their arrangements. In living cytoplasm (strong glass former), non-thermal forces are generated by e.g. motor proteins as we observed the violation of fluctuation-dissipation theorem (Fig. 2a). This non-thermal forces can activate the local structural relaxations. We speculate that such active mixing can erase the origin of cytoplasmic fragility (dynamic heterogeneity), and thus convert fragile cytoplasm to strong glass formers [2]. From the comparing fluctuations of a probe measured by Passive MR (Fig. 2b) and a response of the probe measured by Active MR (Fig. 2c), we discuss to quantify the non-thermal forces in living cells.

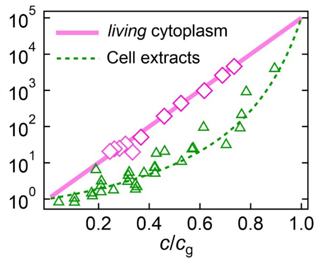

FIG 1: The Angell plot.

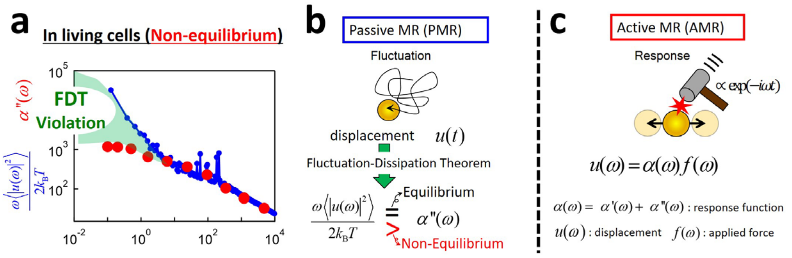

FIG 2: (a) FDT violation in a living cell. (b) Passive microrheology. (c) Active microrheology.

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Liquid crystallinity and rheology of non-aqueous colloids of clay nanosheets

Nobuyoshi Miyamoto* and Ryo Iwashita
Department of Life, Environment, and Materials Chemistry, Fukuoka Institute of Technology,3-30-1,
Wajiro-higashi, Higashi-ku, Fukuoka 811-0295, Japan.

Anisotropic colloids of rod- or plate-like particles have been interested in condensed matter physics field because of their unique properties such as liquid crystallinity (LC) and unusual rheological properties. Especially, inorganic nanosheets that are obtained by exfoliation of layered materials are highlighted as ultimately anisotropic nanoparticle[1-2]. Especially, nanosheets of layered clay minerals are important materials for many industrial applications such as paint, cosmetics, and plastics.[3] In those applications, dispersion in non-aqueous solvents are required; however, it is generally difficult to prepare good dispersion of fully-exfoliated nanosheets in non-aqueous solvents and, therefore, the property of those colloids have hardly been investigated. The non-aqueous colloidal systems are also curious form the fundamental point of view because there have been few detailed studies for those system. Here we demonstrate successful preparation of the colloidal sols of a fluorinated layered clay mineral, fluorohectorite, dispersed in *N*,*N*-dimethylformamide (DMF)/water mixture. We investigated liquid crystallinity, structure of the colloidal sols, and rheological property.

Regardless of the solvent composition, textures and colors due to birefringent LC phases were observed by polarized microscopy above the same critical concentration, indicating that the nanosheets are well-dispersed in the mixed solvents. In the LC colloids, swollen lamellar-like structures with the basal spacing of up to ca. 100 nm were identified by small-angle X-ray scattering. With the increase of DMF content, the basal spacing increased significantly, indicating that the DMF facilitated the repulsive interaction between the nanosheets. The dependence of viscosity of the colloid on DMF content showed an unexpected trend. It increased with the increase of DMF content below ca. 50 wt% of DMF but it decreased afterwards.

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* Corresponding author: miyamoto@fit.ac.jp

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Structures and Slow Dynamics of a Simple Active Matter Model

Kunimasa Miyazaki

Department of Physics, Nagoya University, Nagoya, 464-8602, Japan

Active matters are ideal bench models to assess the effects of nonequilibrium fluctuations at microscopic levels on the collective dynamics and structures at macroscopic levels. In this talk, we present two numerical results on one of simplest active matter models called the Active Ornstein-Uhlenbeck model (AOUP). In the first part, we discuss about slow dynamics near the glass transition point at relatively high activity parameters. Dynamic heterogeneities and slow dynamics are shown to be strongly correlated with collective dynamics. In the second part, we generalize the AOUP model to seamlessly bridge the nonequilibrium system with the equilibrium but non-Markovian dynamical systems. We discover an anomalous structural correlation, called hyperuniformity, for a set of parameter space.

Causes and consequences of long-range stress correlations in glasses

Anaël Lemaître*
Université Paris-Est, Laboratoire Navier (UMR 8205), CNRS, ENPC, IFSTTAR,
2 allée Képler, F-77420 Marne-la-Valle, France

It can be rigorously demonstrated that liquid inherent states carry long-range anisotropic stress correlations that are fixed by certain scaling properties of local stress fluctuations. These stress correlations were also found semi-empirically to cause long-range correlations of local elasticity moduli, a feature believed to be related to a recently observed excess of acoustic scattering as compared with the Rayleigh law. We will review recent results on these related topic and discuss open questions.