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Editorial

Soft matter research in India

Ranjini Bandyopadhyay[1,*] **and Jürgen Horbach**[2]

[1] Raman Research Institute, CV Raman Avenue, Sadashivanagar, Bengaluru 560080, India
[2] Institut für Theoretische Physik II, Heinrich-Heine-Universität Düsseldorf, Universitätsstraße 1, 40225 Düsseldorf, Germany

E-mail: ranjini@rri.res.in and horbach@thphy.uni-duesseldorf.de

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Abstract

Research on soft matter and biological physics has grown tremendously in India over the past decades. In this editorial, we summarize the twenty-three research papers that were contributed to the special issue on Soft matter research in India. The papers in this issue highlight recent exciting advances in this rapidly expanding research area and include theoretical studies and numerical simulations of soft and biological systems, the synthesis and characterization of novel, functional soft materials and experimental investigations of their complex flow behaviours.

Keywords: soft, matter, research, India

1. Introduction to the special issue

Personal care products such as lotions, shampoos and shaving creams, foods such as jelly and mayonnaise, detergents, dyes, paints, polymers, liquid crystals in our laptop displays, and biological matter are some examples of soft materials. They are *soft* as they are easily deformed by thermal fluctuations at room temperature and are constituted by macroscopic molecular aggregates or *macromolecules* of sizes between a few nanometres to about a micrometer [1]. The predominantly entropic interactions manifest as very weak inter-constituent forces and very low mechanical moduli in comparison to atomic solids. Large responses to the application of small perturbations is a hallmark of driven soft systems. Soft materials are characterised by disorder and slow dynamics, nonlinear flows, rich nonequilibrium behaviours and long-lived metastable states [2]. This allows the use of soft materials as model systems to study several complex physical processes. In spite of their apparent diversity, soft materials are characterised by structural complexity and mechanical flexibility. This highly interdisciplinary field has seen the active involvement of researchers with backgrounds in physics, chemistry, materials science, biology, nanotechnology, and engineering [3]. This special issue showcases research on soft matter in India.

While Indian researchers have been working on liquid crystals since the 1950s [4], research on other topics in soft matter and biophysics has grown spectacularly over the past three decades. According to the Institute for Scientific Information Science Citation Index and Social Science Citation Index, published by data.worldbank.org in 2018, India ranks 3rd in the publication of scientific and technical articles in journals. According to the Web of Science database, India ranks 4th in the world in publication of research on soft matter. The contributions in this special issue include theoretical and experimental research on a broad range of topics including but not limited to the characterization of functional and adaptive soft materials, measurements of their complex flows and studies of their spontaneous self-assembly.

2. Theoretical studies of the morphologies, flow and phases of soft materials

Organic molecules with bent cores (BC) can assemble into exotic liquid crystalline mesophases and are used in the fabrication of displays and devices. Of particular interest is the B_7 phase which comprises smectic layers of tilted BC

* Author to whom any correspondence should be addressed.

molecules with in-plane polar ordering. In an interesting contribution, Madhusudana [5] proposes a novel theoretical model to explain the polarization-modulation stripes that have been observed experimentally in the B_7 phase. An earlier model had contended that the stripes owe their origin to an interplay between the molecular tilt and the divergence of the polarization vector. The model proposed in [5] argues that the stripes arise from the conformational variety of the BC molecules. The stripes are explained by considering that domains of different layer spacings, formed by BC molecules in their excited and ground states, are characterized by distinct splay energies. Minimization of the system energy results in the observed stripes. The model correctly predicts the stripe width as also the weak first order transition from the B_7 to the uniform B_2 phase observed upon lowering the temperature.

Mandal *et al* [6] use nemato-hydrodynamics to explore complex fluid flows driven by both controlled shear stresses and shear rates and predict complex dynamical features such as spatiotemporal chaos and transient shear banding. Their minimal model considers the anisotropic nature of the constituents in terms of a nematic alignment tensor and reproduces behaviours that are reminiscent of elastic turbulence in polymers and rheological chaos in micellar solutions. The authors consider spatial nonlinearities only in the gradient direction in the present work. They speculate that incorporation of nonlinearities in both gradient and vorticity directions may be essential in an accurate description of shear banding instabilities observed experimentally in, say, sheared polymer solutions.

While a true crystalline state with long ranged translational order cannot exist in two dimensions at a non-zero temperature, theories predict that quasi-long ranged orientational order can indeed be sustained before the system eventually melts into a disordered liquid phase. In their contribution, Shankaraiah *et al* [7] use Monte Carlo simulations to study a two-dimensional fluid of repulsive particles and show that the incorporation of quenched defects gives rise to local hexatic order. These hexatic neighbourhoods are seen to extend well beyond the separation distance between the pins, and are accompanied by an enhancement of the local ordering at increased particle density and in the neighbourhood of the liquid–solid transition. In another interesting and related contribution, Pal and Chakrabarti [8] show using Brownian dynamics simulations that the presence of a spatially modulated periodic potential causes a two-dimensional colloidal system, wherein the particles interact via screened repulsive interactions, to enter into a modulated phase. Their calculations of self-van Hove functions from the distribution of particle displacements are non-Gaussian, thereby indicating the presence of heterogeneous dynamics due to the pinning of particles at the minima of the modulating field. Arjun and Chaudhuri [9] use Monte Carlo simulations to study a two-dimensional system comprising a 50:50 mixture of size-bidisperse hard disks. They show that such bidispersity and the incorporation of quenched disorder disrupt the formation of ordered structures in their system. Their simulations shed light on some interesting dynamical aspects such as the response of the system to external potentials, the creation of defects and the eventual fluidization of the system. These aspects are summarized in a phase diagram. Presumably, the ideas presented in [7–9] can be tested experimentally in optical experiments involving confined colloidal systems.

A defining feature of glassy dynamics is the breakdown of the Stokes–Einstein (SE) relation between diffusivity and viscosity at low temperatures. Nandi and Bhattacharyya [10] use an extended mode coupling theory (MCT) to address such breakdown in the supercooled liquid phase. They show that at low temperatures, activated dynamics determine structural relaxation processes, while diffusion is best described using MCT-like dynamics. They show that decoupling in these two dynamics gives rise to the breakdown of the SE relation. They calculate the dynamic correlation length of cooperatively rearranging regions and report that this length grows faster than the static correlation length.

In their study of the morphologies and interactions in semi-flexible polymeric systems, Mitra and Chatterji [11] use a bead-spring model to demonstrate the emergence of transient helical structures under the combined influence of spherically symmetric long-range repulsions between the constituent monomers and the ubiquitous thermal fluctuations. While the aforementioned helix formation depends strongly on persistence length and charge density, it is insensitive to the molecular details of the monomers. The authors demonstrate that the formation of helices can be controlled by applying suitable time-dependent potentials and that the helix lifetime can be increased by tethering the polymer chain at its two ends. Incidentally, experimental studies of conformation of single polymer chains are possible using techniques such as the one discussed in reference [25] of this issue (see section 5 below).

3. Computational studies of biological matter: morphologies and interactions

The organization of chromatin and proteins into highly folded chromosomal architectures is key to replication and gene expression processes. Using a coarse-grained homopolymer model, and the assumption of an attractive potential between chromatin and binder proteins that share repulsive interactions between themselves, Kumar and Chaudhuri [12] have shown, via molecular dynamics simulations, that the chromatin folding phenomenon is a continuous transition. The authors have systematically analysed this binder mediated transition and the associated chromatin morphologies. Their analyses show the dominance of simply connected loops at the critical point, and the presence of zippering between polymeric segments mediated by crosslinkers in the folded phase.

The delivery and functionalization of nanoparticles (NPs) has important implications in biomedicine and drug delivery. In a contribution that involves the numerical simulation of the binding process, Basak *et al* [13] use coarse grained molecular dynamics simulations to study the interactions that prevail between lipid bilayer membranes and hydrophilic and hydrophobic NPs of different sizes. They show that uncharged NPs penetrate the lipid bilayers and embed themselves in the hydrophobic regions both in the gel and fluid membrane phases. They further report that cationic NPs interact with the

bilayer in both membrane phases, in stark contrast to anionic NPs which are repelled by the bilayer. Their simulations show that larger NPs disrupt the bilayer, with both anionic and cationic hybrid functionalized NPs showing comparable binding ability. Interestingly, the membrane easily recovers from disruptions when smaller hydrophobic NPs are added. The authors report that only cationic NPs can bind to the bilayer in the second scenario.

Chowdhury and Ghanti [14] have developed a stochastic kinetic model to compute the lifetimes of the transient attachments between microtubules (MTs) and binding proteins or crosslinking molecules in a mitotic spindle. Their *in-silico* analogues of *in vitro* molecular force spectroscopy experiments are useful in understanding the formation of molecular hubs or joints by MTs in such a mitotic spindle. This work is important from the viewpoint of soft mechano-chemistry and can explains complex emergent biophysical phenomena such as the ubiquitous force-induced conformational changes in biological systems, for example, the unfolding of biomolecules.

Rani *et al* [15] use atomistic simulations to study the morphologies of aggregates of methacrylate-based ternary biomimetic antimicrobial polymers bearing hydrophobic, charged cationic and polar groups in solution. Besides exploring the effects of the functional groups on the polymers on their aggregation, the authors also investigate the influence of both random and block sequences on aggregate morphologies. Their interaction calculations point to a marked reduction in the formation of clusters of hydrophobic moieties and therefore an enhancement in polymer solubilisation in the presence of polar groups during the self-assembly process. They demonstrate the important contribution that weakly attractive electrostatic interactions between charged groups have during aggregate formation. Notably, the resulting loosely packed aggregates, which can be tuned by careful selection of the functional groups, have important implications in antibacterial applications.

4. Synthesis and characterisation of functional soft materials

While theoretical studies are important in predicting useful material properties, synthesis and characterization of new functional and meta-materials have gained currency due to their diverse applications such as in catalysis, sensing, additive manufacturing etc. Owing to their enhanced stability, their sensitive response to solution pH, temperature and salt condition, and their applicability in fields such as sensing and imaging, polymers hybrid quantum dots (QDs), wherein the QDs are immobilized within the polymer matrix, are being intensely investigated. Singh *et al* [16] have synthesized a pH- and temperature-sensitive poly(N-isopropylacrilamide)-cationic QD hybrid microgel that exhibits photoluminescence. The photoluminescent behaviour is enhanced when the QDs are produced *in situ* during preparation of the hybrid. Transmission electron microscopy (TEM) is performed to ensure the successful incorporation of QDs into the hybrid microgel. The authors study the temperature-dependent swelling and the pH dependence of the photoluminescence of the hybrids using dynamic light scattering (DLS) and fluorescence measurements respectively. They analyse their data and cite previous reports to argue that maximum photoluminescence can be achieved when the two species in the hybrid bear opposite charges. In another contribution from the same research group, a facile protocol is used to synthesize a smart microgel hybrid of gold and silver NPs [17]. As in the previous contribution [16], the researchers characterize the temperature and pH response of the hybrids using DLS. They perform TEM and UV-visible spectroscopy to ensure the incorporation of the metallic NPs in their samples. By controlling the catalytic ability of these bimetallic hybrids over a broad range of pH and temperature conditions, they demonstrate the smart catalytic action of these hybrids, and speculate on their use as synthetic enzymes.

In their contribution on polyelectrolyte-bilayer complexes formed by the anionic polyelectrolyte sodium polyacrylic acid (PAA-Na), and the cationic bilayer-forming surfactant didodecyldimethylammonium bromide (DDAB), Gupta and co-workers [18] use small angle x-ray scattering experiments to study swelling and deswelling of the complex upon increasing salt concentration in the suspending medium. The authors report the presence of lamellar structures at low and high salt conditions, and an intervening swollen sponge phase at intermediate salt concentrations. They understand their observations by considering the polymer-mediated interparticle interactions and the concomitant changes in the rigidity modulus of the bilayer. In another study that attempts to decipher colloidal interactions, this time under an optothermal gradient, Sharma *et al* [19] study the large-scale two-dimensional assembly of silica colloids into a poly-crystal placed near a gold microplate that is evanescently excited by a laser beam. The authors demonstrate that the resulting optothermal assemblies are more stable for larger particle sizes and can be tuned by changing the polarization of the excitation field or the surface to volume ratio of the gold microplate. The authors speculate upon harnessing this technique for the sorting of colloids in optical fields.

5. Experimental studies of complex fluid flows

Soft materials are viscoelastic and exhibit interesting rheology (flow and deformation behaviour). As discussed earlier, their weak inter-constituent interactions and macromolecular nature result in their viscoelastic response to mechanical stresses [2]. Besides being excellent candidates for studies of complex nonlinear flows, dense suspensions, prepared by dispersing hard particles in a fluid medium, are also excellent model systems to study non-equilibrium phenomena. Using *in situ* imaging and steady-state rheological measurements in one such study, Dhar and co-workers [20] study the onset of shear jamming in dense particulate suspensions of polystyrene (PS) colloids suspended in polyethylene glycol (PEG). They show that the Krieger–Dougherty model systematically underestimates the viscosity of dense suspensions for large applied stresses and high suspension volume fractions. They show, via steady-state rheological experiments, that the flow at the solid-like shear jammed state is best predicted by the Wyart–Cates model.

They further demonstrate that the onset stress for shear jamming decreases with volume fraction. Based on their results, they hypothesize that microscopic failures at random spatial locations grow and eventually merge at higher stresses to cause bulk sample failure. These results are valid for a range of particle sizes. In a related investigation of the unjamming transition in a spontaneously ageing colloidal suspension of charged particles of the synthetic clay Laponite®, Suman *et al* [21] perform dissolution studies to probe the effect of the interface on the formation of fragile particulate networks in aqueous Laponite® suspensions. They study the ageing clay suspensions using rheological tools while varying the solution pH and also following the incorporation of the additive tetrasodium pyrophosphate (TSPP). Their results imply that the fragile Laponite suspension structures that are formed during spontaneous sample ageing are constituted by attractive assemblies of individual Laponite disks.

Bera *et al* [22] uncover signatures of a nonequilibrium transition in their study of the interfacial response to oscillatory deformations of a monolayer of the surfactant sorbitan tristearate that is confined at the air–water interface. Such a transition has been observed before only in particulate colloidal suspensions. The time-dependence of the rheological parameters, measured over several stress cycles, indicates critical-like behaviour. A power law divergence of the number of cycles required to reach the steady state is observed at a strain amplitude value that corresponds to the crossover of the elastic modulus G' and the viscous modulus G'' obtained in amplitude sweep oscillatory measurements. Using Fourier transform rheology measurements, the authors demonstrate the order parameter-like behaviour of the higher harmonics at the nonequilibrium transition point. Their energy dissipation measurements are consistent with the predictions of the continuum nonequilibrium shear transformational zone model for amorphous viscoplasticity.

While macroscopic rheological measurements such as discussed in [20–22] are of great use in materials research and processing, particle-scale studies of heterogeneous soft materials or biological systems, for example the viscoelastic intra-cellular environment, require the use of microrheological experiments. Microrheology measures the microscopic dynamics of a probe particle trapped in a medium to indirectly predict the rheological properties of the latter. In a significant advance to existing techniques, Paul *et al* [23] propose a novel active microrheological technique involving reliable measurements of the fast phase response of a probe particle to external perturbations. The probe particle is trapped using an optical tweezer in a viscoelastic fluid. The authors modulate their trapping beam with a square wave excitation. They use a lock-in algorithm to decompose the phase response in terms of a large number of sinusoidal frequencies spanning a wide range in a single shot. They validate their technique by correctly computing the viscoelastic response of a polyacrylamide-water mixture, thereby laying out the framework to probe more complex environments in their future research. Pal *et al* [24] implement a different active microrheological technique to locally probe the mechanical properties of biological systems at a very fast rate and with high spatial resolution. They directly visualize and analyse the movement of helical nanorobots/nanomotors driven by an external magnetic field in a heterogeneous environment. They show that this active microrheology technique works well in complex macromolecular environments, with successful internalization of the nanorobots (nanobots) by He–La cells. The researchers use this nanobot microrheology technique to correctly estimate the viscosity of the fluid-phase cytoplasm of the cells. These nanobots are easily manoeuverable and can be tracked in real time. These properties make them good candidates for local measurements in highly complex and crowded environments.

While [20–24] involve the measurement of viscoelastic properties using shear rheology, the rheological properties of filamentous objects such as axons of neuronal cells, synthetic and natural fibres, and polymer melts undergoing extensional flows require to be probed under extensional perturbations. In [25], Dubey *et al* demonstrate the use of a cantilever-based device that can be coupled to a microscope to perform extensional deformation studies along with simultaneous imaging. They use their device to accurately measure the extensional rheology of axons, spider and drag silks and the nonequilibrium properties of bacterial baths. Major advantages of the micro-extensional rheometer include the ability to impose a controlled tensile strain to a sample volume as small as a nanolitre, and the ability to track real-time deformations due to the application of forces ranging between millinewtons to piconewtons.

While the experiments described above involve complex instrumentation, Kumar *et al* [26] have studied colloidal transport in aqueous suspensions by exploiting a very ubiquitous phenomenon: the desiccation of droplets. The authors image drying droplets of sessile and pendant droplets of suspensions of latex particles. The morphologies of the coffee stain deposits obtained after completion of the drying process are carefully studied for various latex particle sizes and substrate wettabilities. With decrease in substrate wettability and increase in particle diameter during the drying of pendant drops, the authors report a transition from coffee ring to central dome deposition patterns. In all other experiments, the coffee ring deposit is observed. Numerical simulations suggest that the gravity-driven settling of the colloidal particles prevents the accumulation of the particles at the three-phase contact line in the pendant drop mode, giving rise to the observed central dome deposit patterns for large particle sizes and low wettability of the substrate.

In an experimental study of 2D and 3D turbulence, Paraz and Bandi [27] use laser Doppler velocimetry to quantify the temporal second-order structure functions for integer powers of turbulent fluid velocity fluctuations. While the 2D experiments are performed using gravity assisted soap films, the 3D experiments are set up with rotating jets on the floor of a water tank. The authors report that the enstrophy cascade in 2D experiments shows logarithmic convergence, while the 3D experiments display exponential convergence. This difference in convergence features is attributed to intra-scale couplings in the flows. The work of Paraz and Bandi has important implications for the theory especially of 2D turbulence as

well as the understanding of wind power fluctuations and atmospheric flows.

6. In memoriam Surajit Sengupta (1962–2021)

On March 18, 2021, one of the guest editors of this special issue, Surajit Sengupta, passed away tragically of a heart attack, soon after presenting an invited talk at the APS March meeting. Surajit's sudden and untimely death, deeply saddening to his colleagues, friends, and family, has left a void in the condensed matter physics community in India. Using statistical mechanics and various simulation techniques, his research focussed on diverse problems in soft matter, bio physics and materials physics. His last talk at the APS meeting was on the statistical mechanical theory of rigidity and yielding in crystalline solids. This theme had thrilled him in recent years. With great enthusiasm and excitement, he, along with students and collaborators, developed a novel view on the plasticity of crystals. Those who knew Surajit and had scientific collaborations with him miss him dearly. Surajit is one of the authors of reference [7] of this issue. An obituary for Surajit with personal reminiscences of close colleagues can be found in reference [28].

Data availability statement

No new data were created or analysed in this study.

ORCID iDs

Ranjini Bandyopadhyay https://orcid.org/0000-0002-4290-6404
Jürgen Horbach https://orcid.org/0000-0001-9062-9213

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