

Plenary Talks

Life at Ultra low Interfacial Tension: The Interface of Demixed Colloid-Polymer Mixtures and the Isotropic Nematic Interface in Suspensions of Colloidal Platelets

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Abstract

Suspensions of colloidal particles display rich phase behaviour. Depending on shape and interactions, colloidal gas, colloidal liquid, colloid crystal and colloidal liquid crystal phases are observed. Due to the colloidal length scale the interfacial tension is much lower than in atomic and molecular systems (down to nN/m instead of mN/m). This ultra low interfacial tension has pronounced effects on the kinetics of phase separation and the wetting profile near a wall. In this talk I will present experimental results for the interface of demixed colloid-polymer mixtures and the isotropic-nematic interface in suspensions of colloidal platelets, showing the effect of ultralow interfacial tension.

Defect structure in thin shells of liquid crystal

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We use microfluidic techniques to create thin shells of liquid crystal. A water droplet is coated by a thin layer of liquid crystal and the resultant structure is immersed in water. We examine the defect structure in these geometries and show that unexpected configurations are produced as the liquid crystal shell is thinned.

Morphology and Dynamics of Microscopic Bubbles in Liquid Crystals

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ABSTRACT:

Micro- and nanometer scale gas bubbles suspended in liquids show intriguing behaviors due to their large surface/volume ratio, the variability of surface tension, the high internal pressure, and the concomitant rapid absorption. The liquid crystallinity, when used as the host liquid, adds another dimension to the bubble system, on which the molecular orientational constraint at the surface plays a positive role, introducing a significant inter-bubble interactions. We prepared microscopic air bubbles suspended in liquid crystals by using a micro-capillary injection technique under a constant flow. The most unique feature of the air bubble compared to the conventional solid and liquid micro-inclusions is the variability of the size by external pressure. By way of changing the hydrostatic pressure, the size of the bubble can be systematically controlled from hundreds of microns down to sub-microns associated with the absorption of air into the liquid crystal host. We observed orientational configuration transitions, for example, between the hyperbolic hedge hog for large bubbles and the Saturn ring for smaller bubbles upon a pressure jump, which results in a continuous reduction in the bubble diameter.

Invited Talks

Diluted colloids of nano-ferroelectric particles in liquid crystals

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Colloids in which the solute is liquid crystalline (LC) are known to possess an extremely rich set of behaviours [1]. The anchoring between the LC and microcolloidal particles ($>1\mu\text{m}$) can produce long-range orientational distortions around the particles. This results in strong inter-particle interactions – sometimes repulsive and sometimes attractive – in the mesophase. The interactions can give rise to well-ordered structures of particles in the liquid crystal matrix (both lattices and chains). However, in most cases a prerequisite for interesting LC colloidal behaviour has been a high concentration dispersion, typically with particle volume fraction $c_{part}>30\%$. In such systems aggregated particles produce director distortions extending over macroscopic scales. These suspensions scatter light strongly, and possess unique structural, mechanical, electro- and magneto-optical properties. Recently it has been shown that even at low concentrations ($c_{part} < 1\%$), LC colloids differ strongly from the pure host material [2-5]. These colloids consist of submicron ferroelectric particles suspended in the LC host. In these systems, unlike in classic LC colloids, the suspension-matrix interaction is insufficient to disturb the LC orientation.

We have investigated the physical properties of low concentration ferroelectric nematic colloids, using calorimetry, optical methods, infrared spectroscopy and capacitance studies. The resulting colloids normally remain homogeneous, but the nematic orientational coupling is significantly amplified. In particular cases, the nematic orientation coupling increases by 10% for particle concentrations of 0.2%. A manifestation of the increased orientational order is that the clearing temperature of a nematic colloid increases up to 40 Celsius degrees compared to the pure LC host. A theoretical model is proposed in which the ferroelectric particles induce local dipoles whose effective interaction is proportional to the square of the orientational order parameter.

Effective control of the order parameter of LC by ferroelectric particles allows enhance the electro-optical performance of many liquid crystal materials by not-chemical synthetic method.

The lecture is based on the results obtained by collaboration between Institute of Physics, Ukraine (Reznikov, Buchnev), Kent State University, USA (Li, West), Hoseo University, Korea (Cheon) Kyiv Shevchenko University (Reshetnyak) and University of Southampton, UK (Sluckin)

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SIMULATION OF INHOMOGENEOUS LIQUID CRYSTALS

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This talk will describe recent results in the computer simulation of liquid crystals which are spatially inhomogeneous. This may be due to the presence of planar or curved walls, suspended macroparticles, or the interface between two fluid phases. Molecular simulations are used to bridge the gap between a realistic atomic description of the system, which includes information about molecular shape and interactions, and a continuum-level description such as the Oseen-Frank, Landau-de Gennes, or Onsager density functional theory. The talk will highlight comparisons of simulations with theories of this type, and outline future prospects.

Colloids on free-standing smectic films

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Trapping colloids at a liquid interface or a surface is a reasonably simple way to create a two-dimensional colloidal system; the interaction between the particles mediated by the continuous phase depends on its structure. We discuss theoretically the different phenomena involved in adsorption of micron-size spherical colloids on micron-thick free-standing smectic films, arguing that the film structure is dictated by wetting through two phenomena: particle indentation and meniscus formation. Unlike in simple liquids, the meniscus is characterized by a sharp edge and the attraction between two adsorbed particles induced by fusion of menisci has an abrupt cutoff. This makes the system rather unique and very attractive as an experimental testbed for a variety of novel 2D colloidal structures recently predicted by simulations and theory.

2D Dispersions - liquid inclusions in smectic free standing films

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We summarize experimental and theoretical work on the preparation, stability, shape and interactions of liquid droplets in free standing smectic films. These films represent an ideal two-dimensional geometry for the study of the dynamics and structural self-organization of colloidal inclusions. Interactions of the dispersed droplets with islands (thickness gradients) on the film and with each other lead to ordering and different levels of spontaneous structure formation.

The interactions can be grouped in at least two fundamental types. Capillary forces drive the droplet motion in inhomogeneously thick films, and they may create attractive potentials around individual droplets. Additionally, elastic forces mediated by c-director distortions in tilted smectic C films can lead to complex interactions.

They depend upon the anchoring conditions of the c-director at the droplet border as well as on the topology of the c-director field. These forces may be made easily comprehensible e.g. by electrostatic analogies.

Elastic interactions of Coulomb, dipolar and quadrupolar types, may be defined for example. We describe methods for the calculation of director fields and interaction potentials based on analytical (conformal mapping) and numerical (finite element methods) techniques.

Elasticity- and anchoring-mediated interactions of particles and topological defects at the surface and in the bulk of liquid crystals.

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This presentation is an overview of recent studies of colloidal interactions of particles and topological defects in the bulk and at the surface of liquid crystals. Examples include the following. (1) Periodically ordered structures at the free surface of a liquid crystal. When placed at the free surface of a nematic liquid crystal, colloidal particles arrange in periodic lattices or chains, depending on the type of director distortions around the particles. We explain the interactions by the direct elastic mechanism (through director distortions of the nematic matrix) and also by the elasticity-mediated capillary forces that remain substantial even for micron-sized particles, i.e. at the scales much smaller than the typical length scales of buoyancy-capillary interactions. (2). Anchoring-mediated repulsive/attractive interaction of a dislocation and a liquid crystal surface. We demonstrate that a dislocation line can be either attracted or repelled from the surface of a lamellar liquid crystal depending on the balance of the anchoring and elastic parameters. (3). Elasticity-mediated colloidal interactions are measured for particles entirely submerged into the nematic bulk. The data for normally anchored particles agree well with the behavior predicted by the existing theoretical models while those for tangentially anchored droplets do not.

The work supported by NSF DMR and CRDF grants.

Wetting of micrometer-sized ellipsoids.

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Planar contact lines are very special cases of partial wetting on solids, in practice only met with spheres. We investigate, both experimentally and theoretically, the shapes of contact lines on the surfaces of micrometer-sized polystyrene ellipsoids, of varying aspect ratio (k), at the water-air interface. Combining interferometry and optical trapping, we directly observe the predicted quadrupolar symmetry of the interface deformations around such particles. Based on the numerical resolution of the partial wetting problem on ellipsoids, we deduce the shapes of the corresponding contact lines and the values of the contact angles, $\theta_{\text{c}}(k)$. Rather unexpectedly, θ_{c} is found to decrease for increasing k .

Wetting Phenomena in Liquid Crystal Colloids

Holger Stark

A surface in contact with a nematic liquid crystal at temperatures above the isotropic-nematic phase transition can induce a nematic wetting layer under appropriate conditions.

In this talk, I review wetting phenomena we have recently investigated in connection with colloids immersed in a nematic liquid crystal based on the Landau-Ginzburg-de Gennes free energy. I first summarize the wetting phase diagram for a planar geometry [1], stressing the occurrence of partial and complete wetting, prewetting and a critical line [1]. Then I explain that around spherical particles, prewetting is suppressed for a sufficiently small particle radius, i.e., for large enough curvature of the surface [2]. Thirdly, I show that the wetting layer around a colloid can be non-uniform and therefore may be used to fabricate tetra-coordinated colloids [3] which is interesting for the creation of photonic materials with a large band gap. Fourthly, I discuss the formation of a capillary bridge of condensed nematic phase between two colloids and demonstrate that it gives rise to a relatively "long-ranged" and strong colloidal interaction [4], as demonstrated by the experiments of Kočevar and Muševič [5]. Finally, the modification of the Stokes drag of a single sphere due to nematic wetting is explained on the basis of hydrodynamic equations for the alignment tensor [6].

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Exploring the phase behavior of rod dispersions using external fields

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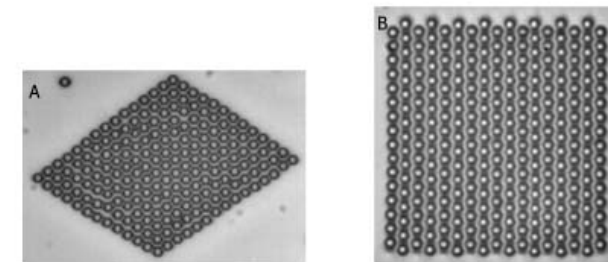
A prerequisite to study the kinetics of a phase separation process is the possibility to quench the system of interest from a stable state into a meta- or unstable state in a well defined way. Since dispersions of rods have two order parameters, concentration and orientation, both can be used to induce a quench. In this lecture I will discuss the phase separation kinetics for mixtures of rod-like viruses (fd) and polymer (dextran). A quench in orientation is performed from a flow-induced fully nematic state into the region where the nematic and the isotropic phase coexist by cessation of the shear flow. A quench in concentration is performed by applying pressure, thus compressing the solvent and increasing the concentration. With these techniques we are able to observe nucleation-and-growth and spinodal decomposition of the rods and determine the nematic-isotropic as well as isotropic-nematic spinodal points. We interpret the spinodal decomposition on the basis of very recent theory, where it is shown that fluctuations in concentration depend on fluctuations in the orientation of the rods.

Two-dimensional Nematic Colloidal Crystals Self-assembled by Topological Defects

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The ability to generate regular spatial arrangements of particles is an important technological and fundamental aspect of colloidal science. We show that colloidal particles confined to a few micrometer thick layer of a nematic liquid crystal form two-dimensional crystal structures that are bound by topological defects. Two basic crystalline structures were observed, depending on the ordering of the liquid crystal around the colloid. Colloids inducing quadrupolar order crystallize into weakly bound two-dimensional ordered structure, where the particle interaction is mediated by sharing of localized topological defects (Fig.1A). Colloids inducing dipolar order are strongly bound into antiferroelectric-like two dimensional crystallites of dipolar colloidal chains (Fig.1B). Self assembly by topological defects could be applied to other systems with similar symmetry.



Figzre 1. Quadrupolar (A) and dipolar (B) nematic colloidal crystals, formed by directed assembly using laser tweezers.

Simulations of colloidal lattices confined to thin nematic layers

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In this lecture our recent simulations of defects and colloidal structures in spatially confined nematic liquid crystals will be described. Theoretical predictions based on phenomenological approaches will be contrasted with the latest experimental studies. Colloidal couplings in nematic medium are effectively many-body interactions that are particularly sensitive to the confinement and external fields. Their complexity leads to numerous colloidal structures not present in simple liquids where van der Waals and electrostatic interactions are dominant. Here we focus on one- and two-dimensional multi-colloid structures. Number of stable and metastable colloidal arrays and their dependence on the severeness of confinement and external fields will be shown. In these structures colloids are coupled by either an array of localized disclinations or by an entangled network of delocalized disclination lines.

Specific heat spectroscopy in liquid crystal phase transitions with disorder

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Theoretical studies have shown that quenched disorder (QD) in LC destroys both orientational and positional long range order, as with other self organizing systems, the normal smectic and nematic phases being possibly replaced by corresponding glassy phases [1]. Glassy behavior has in fact been reported in both such phases [2].

In this work we report on the effect of disorder in several cases concerning LC phase transitions in the presence of disorder, namely LC with nanoparticle induced QD, confined in Millipore Membrane (MM), or in inverse micelles over the Isotropic to Nematic (IN) phase transitions, and also over the Smectic-A – Hexatic B (A-B) phase transition.

We have employed a photopyroelectric based technique which simultaneously enables polarization microscopy observations and light scattering measurements as well as high resolution calorimetry involving both specific heat (C) and thermal conductivity determination which can be performed also in restricted areas of the samples (0.01 mm^2) [3].

We show that the specific heat presents a double peak structure in the two phase coexistence region of the IN transition, as previously reported [4]. We analyze the out of equilibrium properties of the nematic material nucleated at both peaks in terms of hysteretic behavior, frequency dependence and ageing effect. We show that the nematic material nucleated at larger temperature, where the Random Dilution regime of coupling with disorder should apply, shows properties strongly reminiscent of pure material, while the material nucleated at lower temperature, where the Random Field regime should apply, shows features typical of glassy behavior. [5], [6]. The effect of progressive reduction of the MM pore size on the C behavior over the IN transition is also discussed.

Finally, we show that enthalpy is released when the temperature is lowered over the A-B transition of pure 65OBC LC, possibly related to defect annealing, such defects being responsible for the anomalous behavior of the specific heat and thermal conductivity in such cooling measurements [7]. When heating the sample over the B-A transition the specific heat and thermal conductivity behaviour restore their normal behavior and the enthalpy is strongly reduced [8].

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Defects and textures in thin films of liquid crystalline polymers

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Surfaces have a more profound effect on liquid crystal polymers because of significant entropic effects not present in ordinary low molar weight liquid crystals. In very thin films (we studied films in the range of 20-100nm) the competition of tension on the free surface, ordering effects and the substrate interaction leads to a new undulation instability. Apart from being interesting in itself, this undulation also provides a unique contrast mechanism for TEM, which allows a very high resolution for looking at topological defects and, in particular, completely resolve their cores. We shall discuss these findings, comparing them with models of nematic disclinations and their interaction.

ORAL CONTRIBUTIONS

ANOMALOUS ORIENTATION OF CHARGED ROD-LIKE COLLOIDAL PARTICLES IN EXTERNAL ELECTRIC FIELDS.

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Aqueous solutions of polyelectrolytes display intriguing and complex behavior that is partially understood only in the independent particle limit. It is universally accepted that rod-like particles in dilute aqueous suspension line up parallel to a low frequency electric field. Here we studied electrically induced orientation of charged rod-like particles in the presence of smaller, spherical particles by measuring the electrically induced optical birefringence. We find that rod-like particles, when immersed in a sea of smaller particles having similar charge, orient themselves perpendicular to the field. Such behaviour, obtained with a large variety of diluted bidisperse mixtures of rod-like and spherical particles, is beyond the scope of current models. The dependence of the anomalous orientation on frequency and on particle charge, size and concentration indicates the presence of an asymmetric crowding of spherical particles around each rod.

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A Computer Simulation Study of the Formation of Liquid Crystal Nanodroplets from a Homogeneous Solution

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Silvia Orlandi, and Claudio Zannoni

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Dispersions of minute nematic liquid crystal (LC) droplets in a suitable polymeric [1-6] matrix are of considerable importance as composite materials that act as light modulators or more generally as photonic materials [7]. The LC refractive index is anisotropic and thus can be varied by changing the alignment inside the droplet with an applied field. Moreover the LC can be chosen so that in field on/off states its refractive index contrasts with that of the isotropic matrix. This offers the possibility of realising a number of electro-optical devices [2]. While the classical case is that of polymer dispersed LC films (PDLC), a more recent application employs suitable interfering laser beams, coupled with photo-polymerisation, to create nanosize droplets (with average diameter of the order of 200 nm) corresponding to tunable diffraction gratings that can be used in holographic applications (H-PDLC). We present here the results we have obtained from classical molecular dynamics computer simulation experiments on the formation of liquid crystalline droplets of nanometric size by starting from an isotropic and uniform binary solution of an anisotropic solute in a liquid phase of spherical solvent particles. The model system we have chosen is a mixture of N=5000 Lennard-Jones (solvent) and elongated Gay-Berne [8,9] (solute) particles at several concentrations. To study the effect of sample size we have also simulated at selected temperature and concentrations a larger sample of N=40000 particles.

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Phase behaviour of nail-shaped hard particle fluids

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Hard body models have proven to be very useful for understanding the role of the entropy in the formation of liquid-crystalline phases. In surfactant systems the entropy may play crucial role in the stability of the lamellar phases, because the shape of the surfactant particles is strongly non-spherical. To study the effect of entropy we examine the phase behaviour of a system of hard body surfactants by means of Second Virial Density Functional Theory. We model the surfactant molecule as a rod-plate diblock hard body (hard rod with a hard plate attached to one end of the rod). In the perfectly align case we study the stability of the mono and bilayer smectic A phase with respect to other types of ordered phases such as the columnar and the smectic C. Using the well-known Zwanzig approximation the stability of the isotropic-nematic phase transition is examined with varying the geometrical parameters of the rod and plate segments. It has been found that the bilayer smectic C phase can be stable even if the shape of the particle is not oblique. Experimental realization of the model system of hard diblock copolymers using fd viruses is in progress.

Phase Diagrams of Binary Mixtures of Thin and Thick Hard Spherocylinders

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ABSTRACT

The hard spherocylinder is a basic liquid crystal model, especially relevant to the field of colloidal liquid crystals. It is known to exhibit both nematic and smectic A phases. Using a simple, re-summed second-virial theory, a number of phase diagrams of hard spherocylinder binary mixtures have been calculated. Smectic A phases, besides nematic and isotropic ones, have been included in these calculations. Emphasis is here given to those bidisperse mixtures which are formed by thin and thick hard spherocylinders, because of possible future comparisons with real experimental systems.

Ionic contribution to the electric current in an electrolytic cell
submitted to an external voltage.

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The ionic contribution to the electrical current in an electrolytic cell submitted to an external voltage linearly increasing with the time is evaluated. The investigation is performed in the limits of small and large electric fields, in which the density of ions depends on the actual electric field in the sample. In the analysis, it is assumed that the ionic separation induced by an external field can be described by a surface density of charge. It is shown that the ions are responsible for a peak in the current, $I(t)$, where t is the time, following with a delay the application of the external voltage.

In the case of low electric field, the chemical equilibrium relevant to the dissociation of impurities in ions is field independent. In this framework, it is shown that from the experimental data it is possible to determine the mobility of the ions and the density of ions in the sample, by analyzing the maximum of the current versus the time and the limiting value of the current for large t . In the case in which the applied field is so large such that the chemical equilibrium depends on it, the analysis has been performed by taking into account the effect predicted by Onsager, relevant to the ion formation due to the presence of the field. In this case, it is shown that in the limit of large t , the limiting value of the current depends on the Onsager coefficient. Hence, an investigation of the kind presented here can allow the determination of this coefficient.

Self assembled colloidal structures in nematics confined to a capillary

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Colloidal particles in a nematic liquid crystal confined to a supramicrometer capillary form various structures that depend particularly on the size and surface properties of the colloids. Here we present theoretical and experimental results for self assembly of micron and nano-sized colloids. Micron sized colloids form chains, which surprisingly branch in the direction perpendicular to the capillary axis. By means of numerical simulations we show that the branched colloidal configuration is within one-constant approximation metastable with the respect to the straight chain configuration. Smaller nano-sized colloids however form different structures, since they either buckle together into larger droplets or form chains with various conformations. As an example, a stable colloidal conformation of two coupled straight chains with corresponding defects is presented in Fig. 1 for different values of the scalar order parameter S .

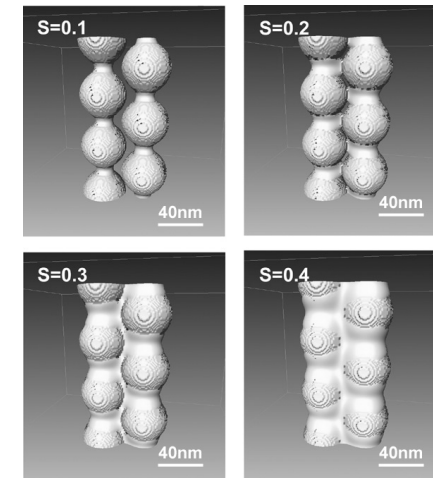


FIG. 1. Isosurfaces of different scalar order parameter for a two coupled straight chain configuration within a supramicrometer capillary are presented. Note that colloidal surfaces impose planar anchoring and thus boojum defects form along the colloidal chains.

Optical trapping of colloids in liquid crystals

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Laser trapping of particles in three-dimensions can occur as a result of the refraction of strongly focused light through micron-sized particles. The use of this effect to produce laser tweezers is extremely common in fields such as biology, but it is only relatively recently that the technique has been applied to liquid crystals. One of the exciting possibilities is to trap and manipulate the micron sized particles in an aligned liquid crystal medium is used to provide a measure of local shear viscosity coefficients and a unique test of theory at low Erickson number in liquid crystals.

Isotropic-Nematic Phase Transition in Liquid Crystalline Elastomers: A non-Boltzmann Monte Carlo Study

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Liquid crystal elastomers, consisting of cross-linked networks of polymers with mesogenic units, exhibit novel properties due to the coupling (with strength, say, γ) between the orientational order parameter S and strain parameter λ . Thermally induced phase transition from a disordered to an (orientationally) ordered state of the liquid crystalline constituents leads to a corresponding abrupt change in the strain, making these materials important for applications. Recent work to elucidate the nature of this transition by phenomenological modeling [1] and through a Monte Carlo simulation employing an appropriate lattice-based Hamiltonian [2] has led to the conclusion that the experimental observation of the smooth cross-over from disordered to ordered state could be due to, for a given γ ($\neq 1$), the heterogeneity in the sample, modeled as arising either from random-bond disorder, or from random-field disorder. Also, the computed values of S and λ are linearly related in the (limited) range of reduced temperature employed [0.8 to 0.9].

We investigate this model with the following conditions and generalizations: 1. the sample is assumed to be homogeneous with no random bond- or field-disorders; 2. the coupling strength γ is varied in steps (between 0 and 1); 3. entropic sampling is carried out covering sufficient energy ranges so as to facilitate projection of canonical ensembles over a wide temperature range; and 4. Wang-Landau algorithm is generalized [3] to make energy-wise unbiased random walks in the multidimensional space including the strain and molecular orientations. We report some new results: 1. the coupling between S and λ is indeed highly non-linear (at $\gamma = 1$) over the temperature range studied [0.2 to 1.4], and linearity is restored only at lower γ values; 2. the transition becomes relatively smoother with the decrease of coupling constant γ , even without heterogeneity; 3. different thermodynamic properties computed (like S , λ , average energy, specific heat, orientational and strain susceptibilities, and Binder's cumulant) show indications of rugged free-energy landscapes pointing to possible frustration; and 4. these observations are demonstrated with free-energy profiles computed as a function of temperature. It may be noted that another microscopic lattice-based model Hamiltonian has been subsequently proposed to account for the experimental observations [4].

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Computer experiments on the non-linear viscoelasticity of filled elastomers

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I will discuss the results of computer simulations of model polymer networks containing stiff and roughly spherical colloidal particles, using the coarse-grained "dissipative particle dynamics" model (DPD). The aim of the simulations is to elucidate the basic mechanisms at the origin of the non-linearities in the mechanical behaviour of "filled rubbers". By performing non-equilibrium molecular dynamics simulations, it is possible to probe directly the viscoelastic behavior of the composites under oscillatory shear deformations of variable amplitude and frequencies, in close analogy with the experimental investigations.

Elliptic inclusions in smectic -C films

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We present a theoretical investigation on the elliptical shapes of soft colloids in freely standing smectic C films, that have been reported in recent experiment [1].

The colloids favour parallel alignment of the liquid crystal molecules at their surfaces and, for sufficiently strong anchoring, will generate a pair of defects at the poles of the colloidal particles. In turn, the elastic free energy of the liquid crystal will affect the shape of the colloids. In this presentation we will focus on elliptical soft colloids and determine how their equilibrium shapes depend on the elastic constants of the liquid crystal, the anchoring strength, the surface tension and the size of the colloids.

A shape diagram is obtained analytically, by minimizing the Frank elastic free energy, in the limit of small eccentricities. The analytical results are verified, and generalized to arbitrary eccentricities, by numerical minimization of an appropriate Landau free energy. The latter is required for an adequate description of the topological defects when the liquid crystal correlation length is comparable to the size of the colloidal particles.

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The particle interaction in liquid crystals: beyond the permanent multipole expansion

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The director distortions in the nematic liquid crystal around particles provide the dominant mechanism of particle interaction. The existing models describe this interaction by means of permanent multipole moments that are attributed to each particle. In this work an exact solution for interaction between particles with weak surface anchoring has been obtained. Analysis of this solution reveals the existence of additional terms in the particle interaction.

Anchoring of liquid crystals on patterned self-assembled monolayers – a simulation study

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This study relates to the anchoring of liquid-crystals (LCs) on substrates coated with self assembled monolayers (SAMs). Specifically, we are investigating, by Monte-Carlo simulation, the effect of substrate chemical patterning on LC anchoring. For this, the hard gaussian overlap model is used to simulate the LC molecules and we explore their behaviour when confined between various patterned substrates.

The aim is to relate these observations to the behaviour of real LCs on patterned SAM systems being studied by our experimental collaborators in Leeds.

We present data obtained from a range of differently patterned systems, concentrating on the effects of

- sharp and diffuse pattern boundaries
- the effects induced by 1-d patterns (i.e. stripes)
- the effects induced by various 2-d patterns (squares, rectangles, circles, ...)

From these, we show that very high resolution patterning offers a viable route to controlling properties such as anchoring tilt angles and azimuthal anchoring strength.

A molecular model for the chiral nematic phase of dispersions of M13 virus

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M13 is a chiral semiflexible polyelectrolyte, with large aspect ratio. At sufficiently high concentration, its water suspensions exhibit a chiral nematic (cholesteric) phase.¹ In this liquid crystal phase virions are preferentially aligned along an axis, which rotates in space forming a helix. We have measured handedness and pitch of the cholesteric helix as a function of temperature and ionic strength.² Several sources of chirality are simultaneously present in the virus, with different characteristic length-scales: these range from the asymmetric carbons of aminoacids, to the alpha-helix structure of the capsid proteins, up to the helical organization of proteins on the virus surface and the possible chiral fluctuations of the virus shape. We propose an explanation for the microscopic origin of the cholesteric organization, which is ascribed to the helical arrangement of coat proteins. The phase organization would result from the competition between contributions of opposite handedness, deriving from steric interactions and screened electrostatic repulsions between virus particles. This hypothesis is supported by calculations based on the classical Density Functional Theory to link elastic properties and intermolecular potentials. A coarse-grained model, derived from the X-ray structure, is adopted for geometry and charge distribution of the virus.

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Electron Spin Resonance studies of order and dynamics in complex liquid crystal systems

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Electron Spin Resonance (ESR) technique has proven to be a powerful mean for investigating liquid crystals and model membranes from a molecular point of view [1]. Liquid crystals containing a suspension of silica particles of nanometric size (aerosil) present fascinating fundamental and applicative aspects. From the fundamental point of view, the interest in these “filled nematic” systems stems from the intricacies of the effect of a random perturbation on ordered phases. These complex systems also present technological interest in view of scattering displays exhibiting the so-called “memory effect” [2].

In this study we report on a comparison between the random disorder effects of an hydrophobic (R812) or an hydrophilic (A380) aerosil on the order and dynamics of the 3b-DOXYL-5a-cholestane spin probe in the liquid crystal 4-n-pentyl-40-cyanobiphenyl (5CB) using the ESR technique. Increasing the aerosil concentration up to 10 wt% the probe order parameter is depressed with a larger effect for the hydrophilic aerosil. The dynamics in the isotropic phase is of Arrhenius type and essentially concentration and aerosil-type independent. Deviations from this behavior are observed in the nematic even at 1 wt% of hydrophilic aerosil and are well represented by a Vogel-Fulcher-Tammann type law, suggesting that a glass-like state can be induced by a very small amount of random disorder.

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POSTERS

Binding energies in nematic liquid crystal colloids

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Colloidal particles confined to a few micrometer thick layer of a nematic liquid crystal form one- and two-dimensional structures that are bound by topological defects. Two basic crystalline structures were observed, depending on the ordering of liquid crystal around the colloid. Colloids inducing quadrupolar order crystallize into weakly bound ordered chains. Colloids inducing dipolar order are strongly bound into antiferroelectric-like crystallites of dipolar colloidal chains. We present the binding energies of the formed structures and explain the binding mechanism.

Modulation of the order and the N-I transition of 5CB liquid crystal doped with azo-derivatives. An ESR study.

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We report on an ESR spin-probe study [1] of the effects produced by both the *trans* and the *cis* forms of different p-azobenzene derivatives [2,3] dissolved in 5CB on the order and dynamics of a nitroxide probe. In particular, we studied the changes in the temperature dependence of the order and the shift of the liquid crystal N-I phase transition temperature (ΔT_{NI}) [4], as reported by the probe molecule, and their relationship with the composition of the mixture.

Eight closely related non-mesogenic p-azobenzene derivatives with molecular formula 4-R-phenylazobenzene (R = H, F, Br, CH₃, CF₃, OCH₃, On-Bu, Ot-Bu) were used at a mole fraction of 0.01 and 0.07.

Even at the lowest mole fraction studied, the presence of the azo-derivative caused, in all the cases, a depression of the T_{NI} . The *cis* isomers reduced the order of the 5CB nematic phase and induced a larger T_{NI} shift with respect to the *trans* isomers. At higher mole fraction, we observed an analogous behavior with enhanced effects.

Surprisingly enough, even at the highest concentration the dynamics appears to be essentially independent of the nature, the configuration and the concentration of the different solutes and very similar to that observed in the pure 5CB.

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Solvent effects in the *trans* → *cis* isomerization mechanism of azobenzene.

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Azobenzene and its derivatives undergo a major structural change under radiation with visible light, transforming from the linear *trans*-isomer to the bent *cis*-isomer. This variation, coupled with the high stability of the *cis* form and the reversibility of the isomerization, can be exploited in the design of materials with photoswitchable physical properties and nanoscale devices with very important relevance in FULCE project aims.

The mechanism of isomerization is being studied both theoretically and experimentally and it has been shown to be dependent on the excitation wavelength and solvent physical properties; it involves a $n\pi$ absorption or a $\pi\pi^*$ and occurs in the picosecond time scale. In the most common experimental conditions it is generally accepted that a three-state mechanism takes place, involving a near UV excitation from the fundamental state S_0 to the second singlet excited state S_2 , a fluorescence process or non-radiative decay to the first singlet state S_1 , and finally a non-radiative decay by conical intersection to the S_0 state. The process involves essentially two molecular degrees of freedom and relative movements, both requiring a reduction of the order of the $N=N$ bond: the torsional angle $Ph-N=N-Ph$ (rotation) and the $Ph-N=N$ angles (inversion). The relative contributions of these movement to the isomerization is still controversial.

To clarify this point and to understand the role of the solvent in the isomerization process, we have modelled the azobenzene molecule with a molecular mechanics force field which contains a quantum-mechanical potential energy surface [4] for the relevant bending and torsional degrees of freedom in the S_0 , S_1 and S_2 electronic states. We have then studied by Molecular Dynamics simulations of an azobenzene molecule in four different solvents (toluene, anisole, hexane and methyl-pentyl ether), the microscopic molecular movements that occur when the electronic state changes, and the effect of solvent polarity in favouring either the rotational or the inversion mechanism.

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Developing generalised Onsager models with Maier-Saupe interactions from experimental saturation pressure and density data

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The understanding of phase transitions in liquid crystals (LC) is becoming very important for the opto-electronic technology and for their importance in cosmetic and in pharmaceutical formulations¹.

To understand and represent the fluid-phase equilibria in model mesogenic systems, an algebraic equation of state for use in engineering applications is developed to describe the isotropic-nematic phase transition in systems represented by hard sphereocylinders (HSCs). The free energy is expressed using the trial function approach of Onsager² (OTF) and a Parson³ decoupling approximation to include the higher body term. The Maier Saupe^{3,4} theory is implemented to extend the treatment to non-spherical molecules with attractive interactions.

These Onsager models with Maier-Saupe interactions are developed from experimental data such as saturation pressure and density data. Few or no data for liquid crystals are available in the literature. An apparatus is constructed to measure the saturation pressures lower than 0.5 Pascal at temperatures between 20 and 150°C. This apparatus is based on a modified Ramsay and Young^{5,6} boiling point method which employ a glass fibre shirt extending from a thermometer into a pool of liquid placed in a boiler. Densities are determined in the temperature range 0-150°C by a pycnometric method.

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Magneto-optical tweezers measurement of interparticle potential and drag coefficient in nematic colloids

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When a particle is immersed in a liquid crystal, it causes a distortion in the surrounding director configuration depending on the size and shape of the particle as well as on the strength and type of anchoring at the particle surface. Spheres, which induce tangential anchoring at the surface, induce quadrupolar symmetry of the director field and the forces between two identical beads are either repulsive or attractive, depending on the orientation of the quadrupoles.

We used magneto-optic tweezers for measurements of liquid crystal mediated forces between microscopic beads in thin nematic samples. The main advantage of this method over optical tweezers is the low intensity of the external field used in the experiment. The magnetic field (below 10 mT) is too low to reorient the director, whereas the electric field of the trapping laser beam alters the nematic director configuration around a trapped bead [1]. We measured the force-separation dependence of the repulsive force as well as the velocity with which the particles are pushed apart by the liquid crystal [2]. The ratio yields the effective drag coefficient, which we find independent of bead separation for separations as small as 1.1-times the bead diameter.

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Novel Crystal Phases in Anisotropic Colloids

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We study crystal packings of hard ellipsoids of revolution. We have observed, in Monte Carlo simulations, spontaneous transitions from hexagonal close-packing (HCP) to a fundamentally different crystal packing (provisionally called "ellipsoid close-packing," ECP), which may be of the kind constructed by Donev et al. [1].

The unit cell consists of two ellipsoids with different orientations, producing layers of alternating orientations in the solid. This arrangement entails 14 nearest neighbors for each ellipsoid, rather than 12 (as in HCP). Our equation of state data show that at a given pressure, ECP has a higher density than HCP.

We also estimate phase boundaries. For a wide range of aspect ratios we conclude that ECP is a much more stable solid than HCP, long assumed to be the ground state of hard ellipsoids.

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Anchoring transition of nematic liquid crystals in patterned cells. Effect of confinement and electric field

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We generalize the study of the geometrically-controlled twist transition in nematic cells [Patricio et al., Phys. Rev. Lett., 2002, 88, 245502] by taking into account the effect of electric field.

Standard minimization methods are not applicable to this problem as the functional has no minima in the presence of the electric field. We perform artificial relaxation timesteps using finite element methods together with adaptive meshes to compute direct solutions of the Euler-Lagrange equations satisfied by the extrema of the free energy functional.

Ordering and demixing of a binary mixture of rod-like molecules in the presence of external field

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Effect of external field (electric/magnetic) is studied on the phase behavior of the binary mixture of the very long thick- and thin rod-like particles. Both the thick and the thin particles possess positive but different susceptibility anisotropy ($\Delta\alpha$). The difference in the extent of interaction between the external field and the two species is varied by means of a coupling constant ($f = \Delta\alpha_{\text{thick}} / \Delta\alpha_{\text{thin}}$). Isotropic-nematic phase transition and demixing phase transitions taking place both in the isotropic and in the nematic phases are examined as a function of field strength on the level of second virial theory of Onsager in the range of $0 < f < 1$. It is found that the isotropic phase becomes weakly nematic (paranematic) and the field aligns both components in the direction of the field even if the field does not have direct interaction with both components. Analytical expressions are derived for the external field induced order parameters and birefringence. The increasing field destabilizes both types of demixing transitions (isotropic-isotropic and nematic-nematic) and the isotropic-nematic phase transition. Moreover it induces closed loop immiscibility, upper and lower critical points for low values of coupling constant. It is interesting that the while the phase boundaries of the isotropic-isotropic demixing and the isotropic-nematic transition is very sensitive to the value of the coupling constant at relatively low pressures, it is practically independent from the coupling constant at high pressures.

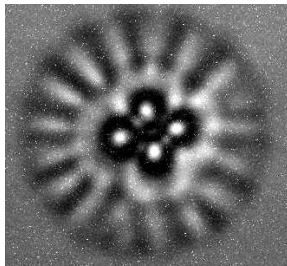
Colloids on free-standing smectic films

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We study the structure of a free-standing smectic A film around a micron-size polystyrene colloid adsorbed onto the film. We find that a colloid or a cluster of colloids is surrounded by an optically distinct and radially decorated meniscus ending with a sharp edge. The observed strong and finite-range attraction between the adsorbed colloids is driven by the fusion of menisci. We interpret the structure of the smectic meniscus in terms of a model dominated by the surface free energy and we argue that the characteristic appearance of the meniscus is due to layer undulations [1].



A group of 4 polystyrene spheres on a free-standing film.

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Wetting transition of a liquid crystal at a wedge shaped substrate

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We investigate the wetting behavior of a nematic at a wedge shaped substrate favoring homeotropic anchoring. The substrate induces a distortion of the nematic director, and an effective long-range interaction with the nematic-isotropic interface. We have used the Landau-de Gennes energy to numerically obtain the wetting phase diagram, which now depends on the wedge angle