On phase behaviour and dynamical signatures of charged platelet suspensions

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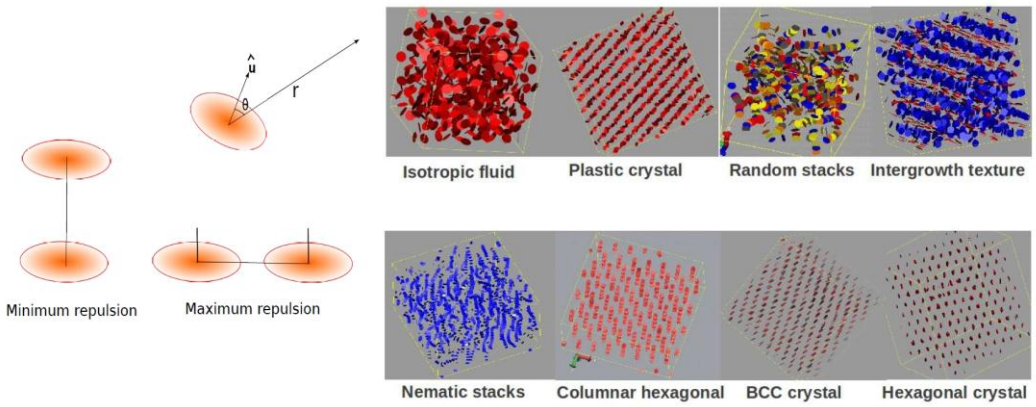
Charged platelet suspensions, such as swelling clays, disc-like mineral crystallites or exfoliated nanosheets are ubiquitous in nature. Their phase behaviours are nevertheless still poorly understood: while some clay suspensions form arrested states at low densities, others exhibit an equilibrium isotropic-nematic transition at moderate densities. In the absence of electrostatic interactions, hard platelets undergo an isotropic-nematic transition as a result of the competition between orientational and positional entropy as predicted by the pioneering work of Onsager [1]. The main question that arises is: how electrostatic interactions influence the isotropic-nematic transition and the organisation of charged platelets?

To address this key issue, we investigate the competition between anisotropic excluded-volume and repulsive electrostatic interactions in suspensions of charged colloidal discs, by means of Monte-Carlo simulations and characterization of the dynamics of the structures. We use a first principle derived orientation-dependent effective pair potential [2] to investigate the phase behavior. The angular dependence of effective pair potential has a peculiar form that makes an asymmetry between two states of parallel disks in co-planar and stacked configurations (see figure, left part).



Exploring the phase behaviour as a function of platelets density and ionic strength, we find a rich a rich phase diagram (see figure, right part) that consists of various crystalline and liquidcrystalline structures. We show that the original intrinsic anisotropy of the electrostatic potential between charged platelets not only rationalizes generic features of the complex phase diagram of charged colloidal platelets such as Gibbsite and Beidellite clays, but also predicts the existence of novel structure of *intergrowth texture* that is composed of disks arranged in alternating nematic (red colour) and antinematic (blue colour) layers. Furthermore, studying the dynamics as a function of density, we provide a strong evidence of slowing-down of dynamics in the orientationally disordered states of charged platelets. This points to the potential formation of arrested states in some regions of phase diagram.

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