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Reentrant melting of two-dimensional colloidal systems

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Abstract. When a two-dimensional colloidal suspension of highly charged particles is subjected to a periodic one-dimensional (1D) light field, a liquid–solid transition can be induced. This phase transition is well known as light-induced freezing. However, upon further increase of the intensity, the crystal is found to remelt (laser-induced melting) to a 1D liquid. This reentrance behaviour is in good agreement with predictions based on Monte Carlo simulations. We suggest explaining this intriguing phenomenon in terms of particle fluctuations which tend to stabilize the crystalline phase.

1. Introduction

Order can be established in disordered fluidlike systems in different ways. Probably the most convenient method is to lower the temperature and thus to enhance one particular density mode which leads to translational and orientational order. Alternatively, however, order can be also induced by periodic external potentials which reduce thermal fluctuations and thus lead to long-ranged density modes. Barrat and Xu predicted by means of density functional theory calculations that the freezing transition of a two dimensional (2D) system of hard spheres is slightly decreased when the substrate is not flat but exhibits a periodic one-dimensional (1D) structure [1]. A similar effect has been experimentally observed by Ackerson and coworkers for the case of a 2D colloidal system of highly charged colloidal spheres in the presence of a standing laser field [2–4]. Since dielectric polystyrene (PS) particles are polarized by the electric field of a laser beam, the interaction of these dipole moments with an interference pattern leads to a periodic external potential for these particles. When the light intensity was increased the colloidal liquid was observed to crystallize. This transition which has been also verified by means of simulations is known as laser-induced freezing (LIF) [3].

When this phenomenon was theoretically investigated by Chakrabarti *et al* in addition to LIF [5] a novel reentrant laser-induced melting (LIM) transition from the crystal to a modulated liquid phase was predicted when the light intensity is increased even further [6]. This was the starting point of our experimental investigations of the phase behaviour of a 2D colloidal system under the influence of a modulated laser field.

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2. Experiment

The samples used in our experiments consisted of aqueous suspensions of charged PS spheres 3 μm in diameter whose pair interaction potential is well known to be described by the repulsive DLVO potential [7, 8]

$$\Phi(r) = \frac{(Z^*e)^2}{4\pi\epsilon} \left(\frac{\exp(\kappa R)}{1 + \kappa R} \right)^2 \frac{\exp(-\kappa r)}{r}. \quad (1)$$

Here Z^*e is the renormalized charge of the particles which has been roughly determined as $Z^* \approx 20\,000$ [9], ϵ the dielectric constant of water and κ the inverse Debye screening length and r the distance between the particle centres. In order to control the Debye screening length, the cell, which was composed of two silica flats and contained the colloidal suspension, was connected to a circuit consisting of a peristaltically driven pump, a vessel containing ion exchange particles and an electrical conductivity probe [10]. This allowed us to perform measurements at very low salt concentrations corresponding to about $0.5 \mu\text{S cm}^{-1}$.

To produce an external periodic potential we used an optical setup as schematically shown in figure 1 [11]. The beam of a linearly polarized argon ion laser (TM₀₀ mode, $\lambda = 514 \text{ nm}$, $I_{\text{max}} = 2.6 \text{ W}$) is split into two parallel beams of equal intensity by means of two beam splitters (BS1 and BS2) and two mirrors (M1, M2). The separation s of the two rays behind BS2 can be adjusted by the position of M2 which is mounted on a motor controlled translational stage. After passing the lens L, the two beams are directed by means of a mirror (M3) onto the sample where they overlap and create an interference pattern with line spacing $d = \lambda/2 \sin(\Theta/2)$ where Θ is given by the crossing angle between the beams which can be adjusted by variation of s . This leads to a lateral external potential for the colloidal particles which can be written as [4]

$$V(x) = -V_0 \cos(2\pi x/d). \quad (2)$$

Here $V_0 = (3n_w P \sigma_0^3 (n^2 - 1)/(n^2 + 2) c r_0^2) (j_1(\pi \sigma_0/d) d/2\pi \sigma_0)$ with P being the laser power, c the velocity of light in vacuum, n the ratio of the refraction indices of polystyrene n_P and water n_w , σ_0 the colloidal particle diameter, j_1 the first order spherical Bessel function, r_0 the waist radius of the Gaussian laser beam and d the line spacing of the interference pattern, respectively. Since the two optical rays are directed from above onto the horizontally adjusted sample, in addition to the potential of equation (2) the particles experience a vertical optical force F_{sc} due to light scattering which pushes them towards the bottom silica plate of our sample cell [12]. This vertical force which under our experimental conditions is in the range of pN largely reduces vertical fluctuations of the particles and thus leads to an almost ideal 2D system. In order to keep F_{sc} constant independent of the potential depth V_0 , the latter being varied in our experiments, a $\lambda/2$ -plate was inserted in one of the arms of the interferometer shown in figure 1. Accordingly, the polarization of one beam could be rotated with respect to the other one, thus leading to a variation in V_0 while F_{sc} remained unchanged. The sample cell is additionally illuminated with white light (not shown in figure 1) and imaged with a microscope objective onto a CCD camera chip. The intense Ar⁺-laser light is blocked with an optical filter. From the pictures, which were stored on video tape, a particle identification algorithm identified the particle centres. From this we obtained the 2D single particle density $\rho(x, y)$ and the pair correlation function $g(x, y)$. To obtain sufficient statistics $\rho(x, y)$ and $g(x, y)$ were averaged over 200 pictures, spanning time intervals of 3 s each.

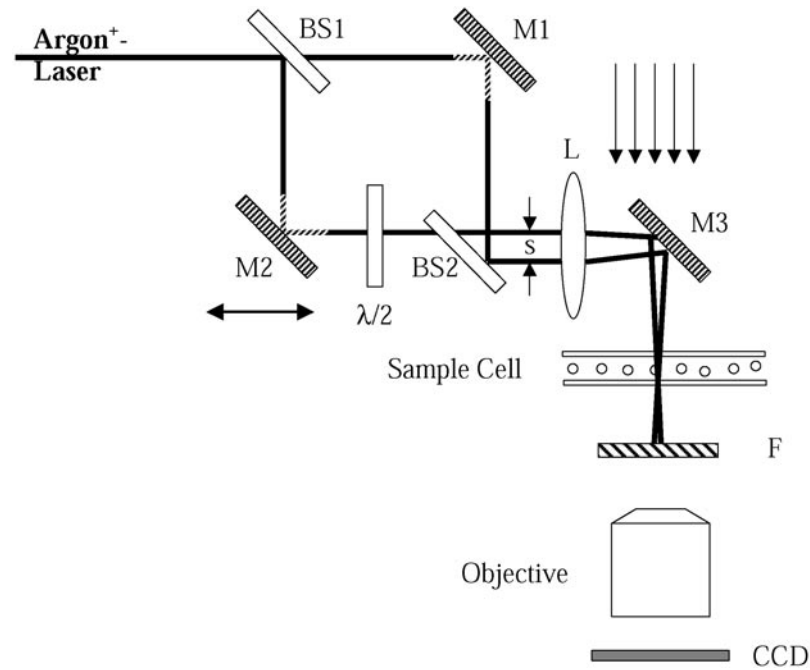


Figure 1. Schematic view of the optical setup. M1, M2, M3 are mirrors, BS1, BS2 beamsplitters, L a positive lens and F an optical high pass filter. The position of M2 can be adjusted by a translational stage and determines the fringe spacing d of the optical interference pattern. The sample is illuminated with white light from above to image the particles onto a CCD camera.

3. Results and discussion

The particle density was kept constant during our experiments and was chosen to be somewhat smaller than that required for spontaneous crystallization. Accordingly, in the absence of an interference pattern, the system always exhibited a liquid structure as can be seen from the homogeneous $\rho(x, y)$ and isotropic $g(x, y)$ in figures 2(a) and (b). From the distance between the ring centres in figure 2(b) we obtained a mean distance of particle centres $a = 9.3 \mu\text{m}$. In the following we applied an interference pattern with period $d \approx a\sqrt{3}/2$ which is commensurate with a triangular lattice at our particle density. As can be seen in figures 2(c) and (d) which correspond to $V_0 = 0.6 \text{ kT}$, the light field causes the particles to be aligned parallel to the interference lines which are in the vertical direction. Although a short-ranged hexagonal order can be observed in figure 2(d), no long-ranged correlation effects occur. This is also supported by an analysis of the pair correlation function along the interference lines which shows an exponential decay as expected in the absence of short-range order. Accordingly, in the following we will refer to this phase as 1D modulated liquid [4]. Increasing the potential to 2.1 kT, one observes that now $\rho(x, y)$ and $g(x, y)$ are modulated both in the x and y direction which demonstrates that at this intensity the system responds by forming a 2D crystal (see figures 2(e), (f)). According to Loudiyi and Ackerson this transition can be qualitatively explained when the 1D liquid in figures 2(c), (d) is viewed as being comprised of small, randomly oriented, crystallites which become oriented in the external light field [4]. Despite the occurrence of long-range order which is also supported by the algebraic decay of $g(y)$ it can be clearly seen that the particles are not perfectly localized at their equilibrium positions,

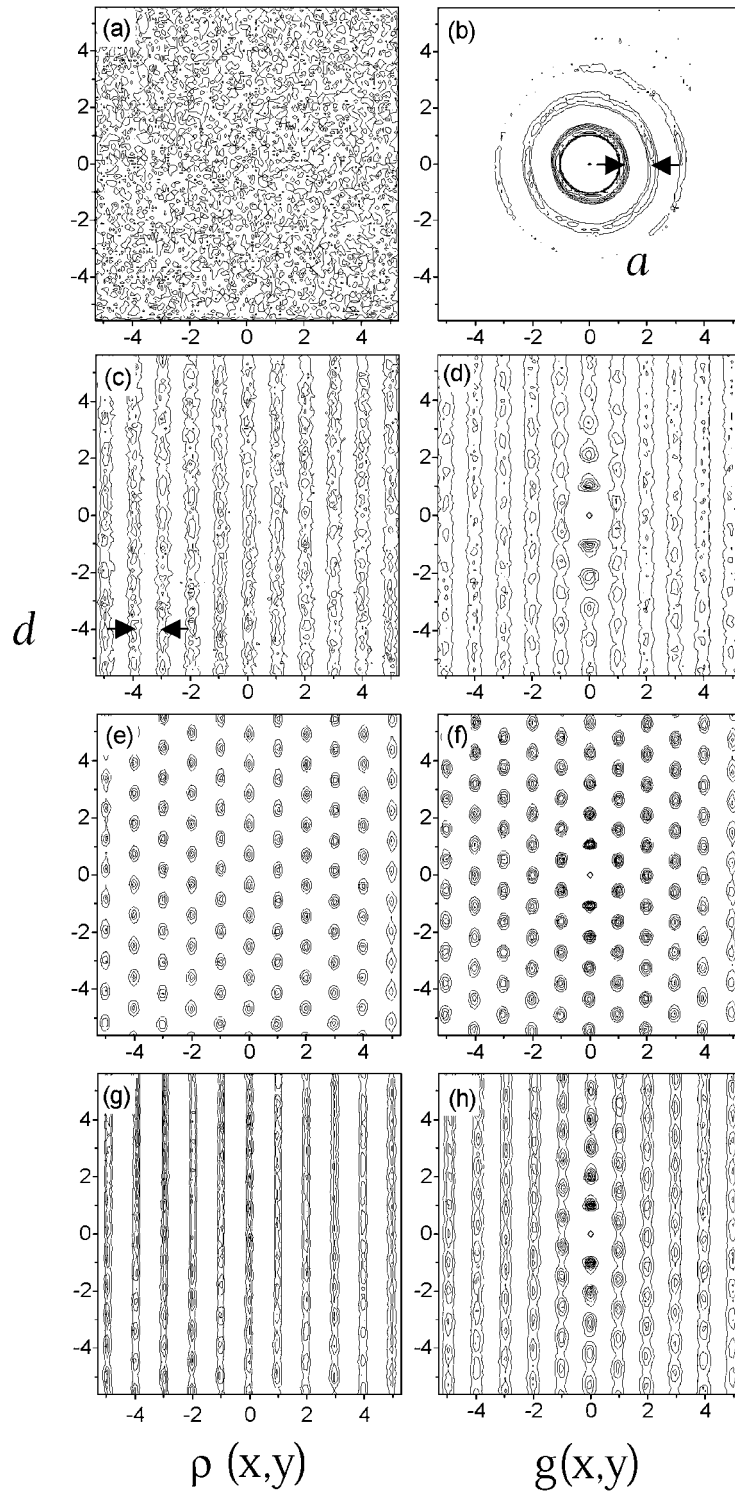


Figure 2. Contour plots of the averaged values of $\rho(x, y)$ and $g(x, y)$ for $V_0 = 0$ (a), (b), 0.6 kT (c), (d), 2.1 kT (e), (f) and 6.3 kT (g), (h), respectively. The horizontal and vertical axes are x and y , respectively. All length scales are in units of the potential period $d = 8.3 \mu\text{m}$.

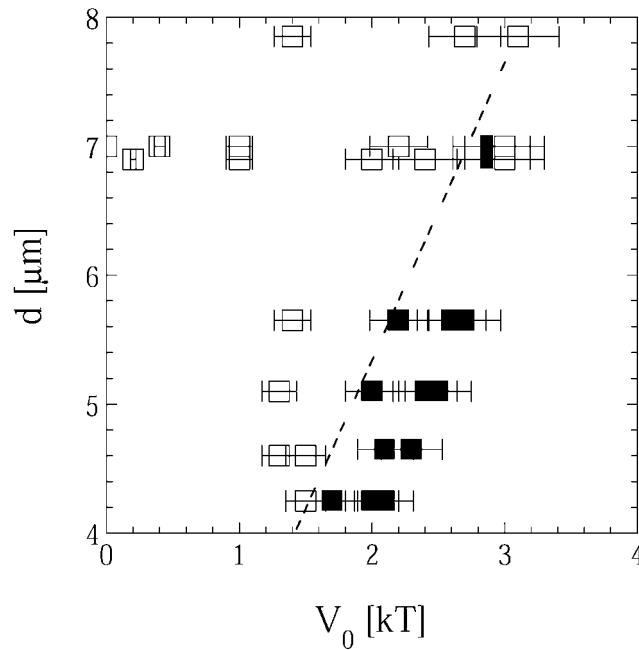


Figure 3. Light-induced freezing transition as a function of the potential depth V_0 and the spacing of the modulated light field d . The open and closed symbols denote the liquid and the crystalline phase, respectively. The dotted line is a guide to the eye. The salt concentration in the experiment differs slightly from that in figure 2.

but perform relatively large fluctuations. In figure 2(e) the particle–particle distances vary by up to 30% with respect to their mean value. The horizontal fluctuations of the particles, in combination with the strongly repulsive particle–particle interaction, cause the registration of adjacent lines. In that sense, the fluctuations stabilize the crystalline structure. This can be also seen from figure 3 where the line spacing d was varied between 8 and 4 μm , respectively. With decreasing d the onset of LIF is shifted to smaller intensities because the fluctuations can mediate the coupling between adjacent lines more easily at smaller values of d .

When the intensity is increased even further as shown in figures 2(g), (h) the signatures of crystalline order disappear and $\rho(x, y)$ and $g(x, y)$ become again very similar to the modulated liquid phase discussed above. In order to understand this at first glance surprising phenomenon one has to recall the role of fluctuations in the 2D crystalline phase. As can be seen the lateral fluctuations are reduced by increasing V_0 . Accordingly, the stabilization of adjacent rows is now reduced and the crystal remelts into a 1D liquid. This behaviour is in good agreement with the theoretically predicted LIM [6].

It has been suggested that the LIM transition is not restricted to crystals formed by LIF but may also appear in systems where the particle concentration is slightly above that for spontaneous crystallization [6]. This, however, is strongly questioned by very recent calculations by Frey *et al* who investigated the LIM taking a different approach [13]. Using the concepts which have been successfully applied in the context of dislocation-mediated melting [14, 15], they described the system by considering the continuum elastic free energy. Their results also support the existence of LIM of a light-induced crystal as observed in our experiments. However, when the density of particles is above the critical value for spontaneous crystallization LIM should not occur.

4. Summary

In summary, we have studied the phase transition of a 2D colloidal system subjected to a 1D spatially modulated light potential. We have demonstrated that, as the light potential is gradually increased, a 2D liquid first crystallizes in predominantly hexagonal order and then melts again to a modulated liquid. This reentrant melting is in agreement with theoretical predictions.

Acknowledgments

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