Video microscopy and micromechanics studies of one- and two-dimensional colloidal crystals

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Abstract. Colloidal suspension of monodispersed charged polystyrene microspheres provides an excellent experimental system for a study of condensed matter physics. Here we report a series of experiments using video microscopy to study the structure and dynamics of colloidal matter. In the first set of experiments, we give a clear demonstration of how thermal fluctuations destroy positional order in one dimension. The second set of experiments is related to the micromechanics of a colloidal solid.
1. Introduction

Monodispersed, charged colloids of submicron scales provide a convenient model system for addressing a number of important problems in condensed matter physics [1]. In this system, individual colloidal particles are large enough such that their real-time Brownian motion can be directly tracked by digital video microscopy [2], yet their dynamics are fast enough to exhibit quasi-equilibrium behaviour of a many-body system in laboratory timescale. Indeed, submicron, charged colloids have been used successfully in two-dimensional (2D) melting studies [3] and to study defect dynamics in 2D colloidal crystals [4, 5].

Colloidal crystals are ordered arrays of submicron particles suspended in a solvent [1]. The most widely studied colloidal crystals consist of monodispersed-polystyrene spheres of sizes 0.1–1 µm, suspended in deionized aqueous solutions. The surface of the particles acquires a negative charge from dissociation of sulphate groups with a typical density of the order of one electron charge per nm². Counterions in the solution form a cloud around each particle, screening the long-range Columb interaction into a short-ranged one within a Debye length, $\lambda_D$, which determines the range of inter-particle interactions. At conditions of low ionic strength (of order $10^{-6}$ M dissolved electrolyte) $\lambda_D$ is of order several hundred nm and the particles can form crystals with lattice spacing $a$ of about 1 µm. Increasing the dilution of the particles or the ionic concentration in the suspension causes the colloidal crystals to melt into a liquid-like disordered phase.

A variety of experiments have been concentrated on the behaviour of 2D colloidal crystals. Considerable amount of research has so far been conducted on the melting of these 2D crystals [3]. The availability of new tools, such as digital video microscopy, optical tweezers and...
microfabrication, opens up new directions for studying colloidal matter. These new experimental capabilities can be used to study a variety of new problems in condensed matter physics.

In section 2 we give a detailed account of the various experimental methods we used. In our experiments, a 1D or 2D colloidal crystal is formed by confining a monolayer of particles between two glass substrates. The negative charge on the glass surfaces suppresses the vertical motion of the particles. In particular, we present a highly reliable procedure for fabricating sample cells with substrate gap in the few micron scale. Our technique gives good uniformity across several millimetres and the materials we use for cell assembly are compatible with colloidal suspension purity requirements. We also describe the methods for preparing the colloidal crystals by cleaning the colloidal suspensions, the optical tweezers and the video microscopy setups.

In section 3 we report results on a model system of 1D colloidal crystals, or more precisely lattices, since true crystalline order is lost, as vividly shown here.

In section 4 we review our recent experiment [4] in which optical tweezers were used for the first time as a micromechanical tool for manipulation of colloidal crystals. Optical tweezers [6] have allowed so far precise measurements of the small forces between colloidal particles [7], as well as assembly of mesoscopic structures [8]. We show for the first time that optical tweezers can be used to create isolated point defects in a 2D colloidal crystal and we study for the first time the Brownian dynamics of these defects in real space and time.

In section 5, we show that colloidal crystals can be used to study the rheological properties of crystalline solids. The motion of dislocations in the colloidal crystal was known to be responsible for the plastic flow of these crystals. Early studies have concentrated on measuring the macroscopic stress–strain relation, and a microscopic observation of dislocation dynamics has not been seriously attempted in colloidal crystals. Here we describe a technique to directly image defects in a colloidal crystal. We use it to study the collective dynamics of dislocations during the visco-plastic deformation of a stressed 2D colloidal crystal.

2. Experimental

2.1. Setup

A method to create a 2D colloidal crystal is to confine a monolayer of particles between two glass or silica substrates [3]. The interface between silica and water has a high density of negative charge (density of OH\(^-\) groups \(\sim\)1 nm\(^{-2}\) [9]), which repels the latex particles, suppressing their vertical motions. Typically, the substrate separation has to be of order 1–2 \(\mu\)m for particles with diameter 0.3–1 \(\mu\)m. The substrates have to be smooth at the scale of the colloidal particles, i.e. have no scratches or dirt larger than \(\sim\)0.1 \(\mu\)m. Furthermore, the separation of the substrates has to be uniform across large areas, in order to avoid gradients in the chemical potential (i.e. the density) of the particles.

The experimental sample cell we used for 2D colloid experiments is shown in figure 1. We assembled it by bonding together a fused silica disk of diameter 1.27 cm and a fused silica cover slip. For flowing the suspension into the cell, we drilled two holes in the sides of the disk and machined a round circulation channel on one of its sides. Before any further processing, the surfaces of both substrates were cleaned by sonicking in successive baths of trichloroethylene, acetone and isopropanol, and subsequently a mixture of H\(_2\)SO\(_4\)–H\(_2\)O\(_2\) at 200 \(^\circ\)C and finally rinsing with deionized-water and blow drying. This procedure has been quite effective in cleaning the silica surfaces from most organic and inorganic contaminants.
Figure 1. Experimental sample-cell for studying 2D colloidal crystals (not to scale).

The separation between the two substrates was controlled by using a patterned thin polymer film, which served both as a spacer and as an adhesive. For the polymer spacer we used benzocyclobutene (BCB) (Dow Chemical, CYCLOTENE 3022), which is often found in the assembly of liquid crystal displays [10]. BCB is particularly attractive in assembling colloid sample cells as it adheres well to glass and is stable in most chemicals. It contains a small amount of ionic species (ppm) and has very small moisture absorption. We prepared the polymer film on the coverslip by spin coating. We diluted the stock BCB solution to about 30% volume in mesitylene solvent to obtain films of thickness of about 2 µm. After spin coating, we fully cured the films at $\approx 270^\circ$C for about 1 h. The cured films can be patterned using reactive-ion etching (RIE). After defining the BCB spacer, we bonded the two substrates by applying pressure while heating at 300 $^\circ$C. Finally, we sealed the rim of the cell and glued small glass tubes in the holes using Norland 88 high-purity optical adhesive. The whole assembly of the cell was carried out in a class-100 cleanroom since any micron sized particles on the surfaces would lead to failure. The thickness variation in the gap between the substrates can be less than 100 nm across the whole area of several mm$^2$ in our samples.

2.2. Sample preparation

Preparation of crystals of charged colloidal microspheres puts stringent requirements on the cleanliness of the suspension. Typically, for crystallization of 300–600 nm diameter microspheres at volume fractions of order 1% (i.e. lattice spacing of about 1 µm), screening length of the order of several hundred nm is required. Thus the ionic strength has to be of order $10^{-6}$ M. Furthermore, in order to reduce the number of uncontrolled parameters and to perform a clean experiment, the suspension should be free of possible organic contaminants, such as dissolved polymer, surfactants used to increase its stability, antibiotics, etc. Traditionally [11], preparation of charge-stabilized colloidal crystals involves initial dialysis of the suspension for cleaning organic impurities (usually directly after the microspheres are synthesized). Alternatively, the
Figure 2. Cross-flow diafiltration setup for washing latex suspension. The suspension is contained in a closed Teflon® reservoir. A peristaltic pump is used to recirculate the suspension. Deionized water buffer is contained in a 500 ml squeeze bottle and is transferred to the reservoir by a vacuum connection, continuously replacing the permeate. Medical grade silicone tubing was used all around the circuit.

Suspension can be centrifuged and the supernatant can be replaced by fresh buffer (deionized water). The drawback of this method is the formation of particle aggregates, since the suspension becomes very concentrated. Subsequently, the cleaned suspension is left to sit in a container with mixed-bed ion-exchange resin, until the ionic strength decreases to low levels and crystals begin to form. Both these techniques are very slow (days to weeks), since they rely on diffusion. Moreover, the presence of ion-exchange resin fragments in the experimental cell is not preferred always.

Instead of dialysis or centrifugation, we used cross-flow diafiltration [12] for cleaning organic impurities. This technique involves, flowing the suspension through a hollow fibre that contains pores of small size. The particles flow tangentially to the walls of the fibre, since they are bigger than the pores, but small organic molecules can permeate the fibre and get removed from the suspension. The retained suspension is continuously washed with deionized water and recirculated through the fibre cartridge (see figure 2). Washing the suspension with five times its initial volume removes about 99.3% of solute. Before washing a suspension of 360 or 640 nm diameter spheres, we diluted the stock of 10% volume suspension to about 0.2–1%. We used a MidGee™ cartridge, with polysulphone hollow fibres having pore sizes of 0.1 or 0.2 µm (obtained from A/G Technology Corporation, Needham, MA). For a recirculation rate of 10 ml min$^{-1}$, the permeate flow was about 1 ml min$^{-1}$, so that a 20 ml initial stock suspension, diluted to 100 ml, took about 2–3 h to wash five times its volume and concentrate back to 20 ml (100 times faster than dialysis).

Although cross-flow filtration cleans ionic impurities as well, the final ionic strength that our setup could reach was not low enough to induce crystallization. For further reduction in ionic

1 The website of Bangs Laboratories (http://www.bangslabs.com/) contains a variety of protocols for working with microsphere suspensions.
Figure 3. Continuous deionization setup for preparing a colloidal crystal. The suspension is contained in a glass reservoir under inert (Ar) gas. A peristaltic pump is used to recirculate the suspension through a glass column (Kontes), containing mixed-bed ion-exchange resin beads (BioRad, AG 501 X8-(D)). A conductivity meter (VWR, model 1054, flow-through cell) is used to monitor the ionic strength conditions. Teflon® (and were not possible, medical-grade silicone) tubing is used all around the circuit. The total volume in the system is about 10 ml.

strength, we used the continuous deionization setup (figure 3) described by Palberg et al [13]. The suspension is pumped peristaltically through a Teflon® tubing system connecting an ion-exchange column, a conductivity meter, the sample-cell and a reservoir for adding suspension or buffer. This scheme is very efficient and conditions of complete deionization (background stray-ion concentration of order $10^{-7}$ M) are achieved a few hours after inserting the initial suspension. Leakage of ions in the system is very small (worst estimate is a few $10^{-7}$ M h$^{-1}$). We prepared colloidal crystals in this setup that are stable for up to 2 days.

2.3. Optical tweezers

We used optical tweezers [14] to perform micromechanical experiments on the 2D colloidal crystals. Our setup is shown in figure 4. A beam from an Ar$^+$ laser is brought to a diffraction-limited spot using a high numerical aperture objective lens. Typical trapping force is in the pN range. We control the position of the trap in the sample plane by making it pivot around the back aperture of the objective using a telescope and a two-axis galvanometer scanner. The time response of the scanner is 0.5 ms for small steps (see figure 4 for details).

2.4. Video microscopy and particle tracking

Conventional optical microscopy, in transmitted light bright field mode, can resolve colloidal particles as small as 100 nm. Standard commercial video cameras, operating at 30 fps, provide adequate sensitivity and resolution for recording the dynamics of colloidal particles. The video images are digitized and transferred to a computer using a frame-grabber board, where they can be
Figure 4. Optical tweezers setup. We used the 514 nm line of an Ar$^+$ ion Coherent Innova 90 laser. The beam is expanded to a diameter of about 3 mm so that it overfills the back aperture of the objective lens. M2 is a two-axis galvanometer scanner (Cambridge Technology, model 6860). Lenses L3 and L4 comprise a telescope that creates two conjugate planes, on the back of the objective lens and on M2. Tilting the beam with M2 makes it pivot around the back aperture of the objective, thus moving the trap in the specimen plane.

further processed. In our experiments, we used a Zeiss Axiovert 135 inverted microscope with a Plan Neofluar 100× oil immersion objective (NA = 1.3). A Sony SSC-M370 monochrome CCD video camera was used for imaging and the data were recorded on a videotape using a Sony SVO-9500MD recorder. The camera captures 30 interlaced frames per second, 640 × 480 pixels, with dynamic range of 8 bit. The total system magnification is 83 nm pixel$^{-1}$. The video data were transferred to a personal computer using a National Instruments PCI-1408 image acquisition board and software written in LabVIEW.

The images of the colloidal particles were processed using the particle-tracking algorithms of Crocker and Grier [2], implemented in IDL (Interactive Data Language, Research Systems Inc.). If the imaging parameters are optimized, these routines can give the in-plane particle trajectories with a resolution of 1/20 pixels or 4 nm. In order to estimate the out-of-plane motion of the particles, we calibrated the radius of gyration of the particles as a function of their vertical position. This was done by mounting a coverslip with immobilized 360 nm diameter particles on a Burleigh TSE-150 (Burleigh Instruments) piezoelectric stage and taking images while performing 60 nm vertical steps. We find a linear increase in the apparent size of the particles as they go out of focus with slope 0.0058 pixels nm$^{-1}$.

2.5. Imaging of defects

We discovered a method for directly imaging defects (e.g. dislocations or grain boundaries) in an optically thin colloidal crystal. The scheme we followed is shown in figure 5. The colloidal crystal sample is illuminated using a high numerical aperture condenser lens (Zeiss, NA = 1.3). The condenser lens blocks the central part of the light and thus illuminates the sample with a highly divergent cone of light. The objective lens (Zeiss Achrostigmat, 20×, NA = 0.30) is outside of the cone and it can collect only light that is deflected from the sample in a solid angle.
Figure 5. Microscopy scheme for imaging defects in a 2D colloidal crystal with lattice spacing $a \approx 1 \mu m$. Size of the field of view is $240 \times 200 \mu m^2$. The elongated dark spots are the core regions of edge dislocations. Edge dislocations can be seen to align in grain boundaries. The different brightness between crystallites is due to the varying sensitivity of the CCD camera for different colours.

$\delta \Omega$. The colloidal crystal acts as a diffraction grating, since it imposes a periodic variation in the refractive index. Light that satisfies the Bragg condition for scattering in $\delta \Omega$ is collected by the objective and forms an image. Typically, the apparent colour of colloidal crystals of 360 nm diameter spheres changes from red→green→orange→purple, as the lattice spacing $a$ changes from about 1–0.5 $\mu m$. Also, crystals with the same $a$ but different relative orientations have slightly different colours. We observed that highly strained, defective regions in the crystal, such as dislocation core regions or large-angle grain boundaries, appear dark and give high contrast in images obtained with this technique. This is due to the fact that light is scattered differently...
from these regions than from the rest of the crystal. Dislocations in particular appear as elongated blobs with size about 5 µm in the direction of their Burgers vector. In section 5 below, we use this method to study the motion of dislocations during plastic flow.

3. 1D model system: loss of long-range order in 1D colloidal crystals

A 1D chain of interacting particles is probably the simplest ‘condensed matter’, for which a complete theoretical description is available for short-ranged interactions. For a 1D elastic chain, it was recognized long ago first by Landau and Peierls [15] that the long-range translational order cannot survive. The density distribution of a 1D lattice appears like a liquid even though a snapshot of the system may appear like a lattice (solid). Here we report the first direct observation of the Landau–Peierls prediction in a 1D colloidal chain.

To create a 1D chain from colloidal particles of like charges interacting with a repulsive screened Coulomb potential, these particles have to be confined within a quasi-1D space (a 1D potential well). One approach is to use a scanning laser tweezer [16] in which the laser trap moves much faster than the diffusion of the Brownian particles, creating an effective 1D potential. A simpler approach would be to take advantage of the fact that the surface of the substrate (fused-silica or quartz) is negatively charged in deionized water. We found that by creating a linear trough in one of the substrates, a 1D potential well for polystyrene colloidal particles can be created.

In the present experiment, linear troughs were created by scratching the surface of a quartz substrate using a diamond drill bit manually. Thus many troughs were created with various depths. The data presented below is for a long straight trough (>120 µm). The particles are monodispersed polystyrene-sulfate microspheres with a diameter of 632 nm (Duke Scientific).

The inset of figure 6 shows a still-frame image of a 1D colloidal chain trapped by a linear potential well created by the presence of a trough on the quartz substrate. The trapping is due to the fact that the free energy of a charged colloidal particle is lowered by the presence of a trough, since the Coulomb repulsion on the particle from the substrate is reduced. It is clear that the particles arrange themselves into a lattice along the channel of the trough. The particles are evenly spaced, indicating a smooth 1D potential well along the channel. We should also point out that one end of the trough is near the upper left corner of the image.

From the dynamics of the colloidal particles trapped in a 1D potential well, it is also clear that the trough is smooth along the channel. We could not perform an AFM measurement of the depth and the surface topology of the same trough once the sample cell is dis-assembled. However, the more important parameter is the potential well depth that can be calibrated by measuring the particle-density profile perpendicular to the channel. For the trough, we studied, the potential well depth of the channel is $1.7k_B T$ ($T = 300$ K).

We found that the 1D colloidal chain behaves like a jiggling elastic string, except that occasionally a particle on the chain can pop-out. Conversely, there are similar number of particles jumping into the chain also. One way to reveal the equilibrium behaviour of the chain is to measure the pair correlation function, as shown in figure 6. Here $g(x)$ is measured by averaging over 10 s of the video data.

Another way to reveal the equilibrium behaviour is to directly measure the spatial distribution of the time-averaged particle density, as shown in figure 7. The well-resolved particle positions in the density map near the left end of the chain is a real effect, caused by having a rigid end of the trough which reduced particle diffusion locally. The equipartition theorem allows us to
Figure 6. The pair correlation function of a 1D colloidal chain. Inset: a still-frame image from the video data. Scale: lattice spacing $a = 2 \mu$m.

Figure 7. The time-averaged particle density profile. The scale is the same as that in the inset of the above figure.

relate the average squared displacement of a particle on a chain to the effective spring constant,

$$\langle x^2 \rangle = \frac{n}{2} k_B T,$$

where $x$ is the displacement of a particle from its equilibrium position along the channel, $n$ the position of the particle away from the anchored end, $k = 2(eZ^*)^2 e^{-n/a}/\kappa^{-1}a^2$ the spring constant of the chain, $\kappa^{-1}$ the Debye length and $eZ^*$ is the effective surface charge on a particle.

Taking the values from our previous work [4], $Z^* \sim 4600$, $\kappa^{-1} = 390$ nm, and using the measured value for $a = 1.9 \mu$m, the expected mean squared displacement $\sqrt{\langle x^2 \rangle}$ is 100 times smaller than what is measured above (see figure 6, the density peak is lost at $n = 5$). We wish
to point out that in a previous study of elastic properties of similar systems of charged colloids, Weiss et al [17] also noticed the discrepancy between the measured elastic moduli of a crystal of charged colloids and those expected from a screen-Coulomb interactions. In the above case, the discrepancy was attributed to the flaw in the conventional Derjaguin–Verwey–Landau–Overbeek (DLVO) theory on the interaction between charged colloidal particles. It is entirely possible that we may have overestimated the spring constant of our 1D colloidal crystal above using the DLVO interactions. Since our system is more confined than those of Weiss et al, it is possible that the screening by the Debye layers on the top and bottom substrates may be important. Such additional screening should lead to a further reduction in the inter-particle interactions, thereby enhancing the fluctuation effects. The occasional pop-in and -out of particles into the chain could also enhance the fluctuation effects. This effect should be small since such events are rare.

4. 2D model system: unusual point defects in 2D colloidal crystals

In this section we review our recent study [4] of point defects in 2D colloidal crystals. Figures 9–11 and related text are reprinted with permission from [4], copyright (2001) by the American Physical Society. Point defects are interesting objects in 2D solids. Unlike unpaired edge dislocations, a finite amount of energy is required to create a point defect. Thus any 2D solid system at finite temperature is expected to have some thermally excited defects. A high concentration of point defects may lead to interesting new collective effects, such as forming an extremely soft supersolid phase.

Various authors have considered the energetics of point defects in a variety of 2D systems. Fisher et al [18] considered various defects in a 2D crystal of electrons (Wigner crystal), with interaction potential \( V(r) = -\epsilon_0/r \). Using numerical calculations they estimated the formation energy of the following point defect configurations: sixfold symmetric vacancy (V\(_6\)), divacancy (D) and centred and edge interstitial (CI and EI respectively), which they found to be a fraction of the crystal’s binding energy, \( \epsilon_0 \). More detailed numerical work on the 2D Wigner crystal [19] showed however that most of the configurations considered in [18] are not stable (minima of the energy), but rather correspond to saddle points. In particular, the sixfold vacancy was found to be unstable to a twofold vacancy configuration (V\(_2\)) and the EI relaxes to the CI. The same behaviour was discovered for point defects in the flux line lattice of type-II superconductors [20], in which case the inter-particle interaction is \( V(r) = \epsilon_0 K_0(r/\lambda) \). \( K_0(x) \) is the modified Bessel function and exhibits the asymptotic behaviour \( \sqrt{(\pi/2x)}e^{-x} \) for large \( x \). The characteristic decay length \( \lambda \) is the superconducting penetration depth. Further work [21], covering the range \( a/\lambda = 0.25–100 \) (\( a \) is the lattice spacing), extending also to the case of superconducting thin films [22], verified the results of [20]. However more metastable (local energy minima) vacancy configurations were discovered. In particular, the lattice was observed to relax to a threefold symmetric vacancy configuration (V\(_3\)) and a second metastable twofold symmetric configuration (V\(_2'\)) appeared at \( \lambda/a = 0.25 \).

The most complete numerical investigation has been published in [23]. Two types of interaction potentials were considered: (i) \( V(r) = \epsilon_0 K_0(r/\lambda) \), in the range \( a/\lambda = 0–7 \) and (ii) \( V(r) = 1/r^p \), for \( p = 1–12 \). It was found that the V\(_2'\) configuration (V\(_{2a}\) in [23]), appears at the longer-range interactions (\( \kappa a = 0–2 \) and \( p = 0–1 \)) whereas the V\(_6\) configuration becomes stable (with all the other vacancy configurations becoming unstable towards it) for the shorter-range interactions (\( \kappa a > 5.8 \) and \( p > 5 \)). The results for the energetics of point defects obtained in [23]
Table 1. Defect energies for $V(r) = \epsilon_0 K_0(\kappa r)$, in units of the crystal binding energy $\epsilon_0$, from [23]. Entries such as ‘$V_{2a}$’, ‘SV’ etc indicate instability towards a lower-energy defect.

| $\kappa a$ | SV  | $V_{2a}$ | $V_3$       | $V_{2b}$ | $V_6$       |
|----------|-----|---------|-------------|---------|-------------|
| 0        | $V_{2a}$ | 0.107018876 | 0.108206944 | 0.109320135 | $V_3$       |
| 1        | 0.096728537 | 0.096661116 | 0.097578530 | 0.099169907 | $V_3$       |
| 3        | 0.046095915 | SV        | 0.046131759 | 0.047174061 | $V_3$       |
| 5        | 0.0133112  | SV        | 0.0133146   | 0.0136217   | $V_3$       |
| 7        | $V_6$    | $V_6$    | $V_6$       | $V_6$    | 0.002771295 |

are reproduced in table 1. Typically, the defect energy, $\epsilon_D$, varies from $O(0.1\epsilon_0)$ for the longer range to $O(0.01\epsilon_0)$ for the shorter-range potential. The difference in energy between the various configurations, $\delta\epsilon_D$, is a small fraction of the defect energy, from $O(10^{-2})\epsilon_0$ to $O(10^{-4})\epsilon_0$ for longer and shorter potential range respectively.

Point defects in crystals with a screened-Coulomb inter-particle interaction, which would directly apply to our two-dimensional colloidal crystals, have not yet been studied. However, the qualitative behaviour discovered with numerical calculations for the vortex lattice should be applicable to our system as well. Also, there are no available numerical calculations for the energy barriers between the different configurations, which is particularly important for the dynamics of the defects. A reasonable assumption is that they are of the same order of magnitude as the energy differences $\delta\epsilon_D$, i.e. a small fraction of the elastic energy $\epsilon_0$. Thus, at not too low temperatures, one would expect the defects to be considerably mobile.

4.1. Creation of point defects in 2D colloidal crystals using optical tweezers

A 2D colloidal crystal is a very convenient system for studying point defects. Using video microscopy, such defects can be studied in real space and time, providing new insight into their microscopic dynamics. In this experiment, $a \approx 1\%$ vol. aqueous suspension of 0.360 $\mu$m diameter negatively charged polystyrene-sulfate micro spheres (Duke Scientific No 5036, polydispersity $\approx 1\%$) was completely deionized by ion exchange, reaching a conductivity $\sigma = 2.5 \mu$S cm$^{-1}$. As described in [4], this corresponds to an effective particle charge of $Z^* \approx 1650$ and screening length of $k^{-1} \approx 390$ nm. A monolayer of particles was confined between two fused silica substrates separated by $\approx 2\mu$m, resulting in a 2D colloidal crystal with a lattice constant $a \approx 1.1 \mu$m. Under these conditions, the 2D crystal is deep in the solid phase, with elastic (binding) energy $\epsilon_0 = \frac{1}{2}ka^2 = 346k_BT$ (estimated by measuring the mean square displacement of a particle from the centre of the ‘cage’ created by its six neighbours). Thus there are no thermally generated defects, due to their large formation energy. During the experiment, the 2D region extended over several hundred lattice constants, with typical crystallite size of about 200$a$. The experiments were performed in a central, perfect crystal region of size $40 \times 40$ lattice constants. Before the experiment, any impurities, such as aggregates of particles, were dragged away from this region using the optical tweezers.

We found that it is possible to create isolated point defects in an otherwise perfect 2D crystal by trapping a particle with optical tweezers and dragging it from its lattice site. The optical tweezers’ setup is described in detail in section 2.3. To overcome the restoring force from
the elasticity of the colloidal crystal and to be able to drag the particle faster than the relaxation time of the lattice, ≈50 mW of laser power was needed to form the optical trap.

The defect creation process is shown in figure 8. A trapped particle is dragged away from its lattice site at about 50 µm s⁻¹, becoming an interstitial and leaving a vacant site behind. The trapped particle is moved from its original site all the way out of the field of view (or the boundary
of the crystallite). During this process, the disturbance does not propagate through the crystal and the lattice heals behind the trace of the dragged particle. Finally the perfect lattice is restored, with the exception of a single isolated vacancy.

The same procedure can occasionally lead to divacancies. We are not able to observe the creation of the defect in detail because the entire process is completed within a few frames and the reflection of the laser beam obscures imaging around the tracked particle. The evidence available suggests that dragging a particle away from its lattice point and parallel to a crystal axis can push an entire row of particles. The tweezed particle is moved to a new lattice site leaving a first vacancy behind. The row buckles several \( \alpha \) away, creating an interstitial. Continuing to drag the particle further creates a second vacant site, resulting in a divacancy.

Single, isolated mono- and divacancies were observed in real time at 100\( \times \) magnification and recorded on videotape. The defects were tracked for 20–40 s, which is the time they take to diffuse away from the 50 \( \times \) 40\( \alpha^2 \) field of view. The positions of the particles in every frame were tracked and then linked to time-trajectories. We used a triangulation algorithm to identify the mis-coordinated particles in the defect core.

4.2. Equilibrium configurations of point defects

The real-space and real-time imaging capability here facilitates detailed observation of the structure of the defect core and the relaxed lattice around it, allowing for an experimental test of the theoretical models of 2D point defects. In our system, thermal energy causes the point defects to switch between different possible configurations, as they hop from one lattice site to the next. Typical snapshots of the system should reveal the various metastable states, since the system is expected to spend most of the time around a local energy minimum. In the case of strong interactions (\( \kappa \alpha < 5, \kappa \alpha \approx 3 \) for the 2D colloidal crystal), our observations were consistent with the numerical predictions for vortex lattices. For the monovacancy we observed that the lattice spontaneously deforms to the lower symmetry configurations \( V_3 \) and \( V_{2a,b} \). Also, we observed that the split vacancy \( SV \) is also stable for our system. For divacancies, we typically observed the twofold crushed configuration \( D_2 \) considered in [18] for Wigner crystals. Also, we discovered three more configurations (figure 9) not previously considered: a threefold symmetric \( D_3 \) and two split configurations \( SD_{a,b} \), which seem to separate two \( D_2 \) or \( D_3 \) configurations at neighbouring sites.

The configuration of the defect can be characterized by the arrangement of these broken bonds and miscoordinated particles. We performed a geometrical triangulation of the positions of the particles, identifying nearest neighbouring pairs and measuring the coordination number \( n_i \) of every particle. Our observations indicate that the following topological constraints are satisfied:

1. the miscoordinated particles are never observed isolated but rather appear as distinguishable pairs of triplets, etc. of nearest neighbours,
2. the mean coordination number of every pair, triplet, etc is always equal to six,
3. we can define vectors \( \tilde{n}_{ij} \) starting from a particle \( i \) with \( n_i < 6 \) and terminating at a nearest neighbour \( j \) with \( n_j > 6 \). The number of vectors originating from \( i \) is \( 6 - n_i \) and the number of vectors terminating at \( j \) is \( n_j - 6 \). For every configuration, \( \sum_{(ij)} \tilde{n}_{ij} \approx 0 \).

At finite temperatures, thermal fluctuations cause the system to vibrate around every local energy minimum \( C \), exploring the volume of phase space \( \Omega_C \) in which the energy is \( O(k_B T) \)
Figure 9. Configurations of point defects in the 2D colloidal crystal. Configurations of monovacancy: (A) split (SV), (B) threefold symmetric (V₃), (C, D) twofold crushed (V₂a, V₂b). Nomenclature adopted from [24]. Top-left insets in every figure show configuration of the vectors \( \vec{n}_{ij} \) defined in the text. Configurations of divacancy: (E, F) split (SDₐₙ, b), (G) twofold crushed (D₂), (H) threefold symmetric (D₃).

above the minimum energy \( E_C \), and occasionally getting enough energy (from the heat bath) to jump to a nearby local minimum. The contribution to the entropy of every configuration is roughly \( S_C = k_B \ln(\Omega_C) \), and the different configurations occur with relative probabilities \( P_C \propto \exp(-F_C/k_BT) \), where \( F_C = E_C - TSC \) is the free energy of a configuration. As long as the system remains around a local energy minimum, the distortions of the lattice are elastic.
and the topological arrangement of the particles does not change. Therefore, every energy minimum has not only a characteristic symmetry but also a certain topological configuration of the defect core. Using the topology rather than the symmetry as a criterion for identifying the different configurations \( C \), we measure their relative occurrence probabilities \( P_C \). The free energy differences, estimated from the measured \( P_C \)s, are found to be of order \( 0.1-1k_BT \) (see table 2).

In 2D crystals, a point defect can be viewed as bound dislocation pairs, or topological defects. Thus it should be possible to directly measure the dislocation–antidislocation pair potential by studying the temporal behaviour of these defects, using Boltzmann statistics. Focusing on the case in which the point defects appear as a dislocation pair, figure 10 and assuming an interaction \( V(\vec{r}) \) between the two dislocations, we expect: \( P(\vec{r}) \propto \exp(-V(\vec{r})/k_BT) \). In addition, particle conservation dictates that dislocations can only glide parallel to their Burgers vector, so \( \vec{r} = \vec{r}_c + \vec{r}_g \), with \( \vec{r}_c \approx 3\vec{a}/2\vec{a} \) for di(mono)-vacancy and \( \vec{r}_g \parallel \vec{b} \). We measured \( P(r) \), \( r = |\vec{r}| \), for the mono- and divacancies, using a few thousand snapshots of the system. The separation \( r \) of the two dislocations is identified as the separation between the two fivefold coordinated particles where the extra rows of particles terminate. Our results (figure 11) show a rather rapid decrease in \( P(r) \) as \( r \) goes beyond a couple of lattice constants, together with a modulation, more clearly seen in the case of the divacancy. Since \( V(r) \propto -\log P(r) \), the interaction of the two dislocations turns out to increase with \( r \), with an average slope \( 2.9 \pm 0.4k_BT/a \) and \( 2.0 \pm 0.2k_BT/a \) for the case of mono- and divacancy respectively. The modulation is due to a Peierls energy barrier [25] that comes from the discreteness of the lattice. To the best of our knowledge, this is the first time that the Peierls barrier of edge dislocations is observed directly in a real physical system.

A final point is that the core region of dislocations has a typical size of a few \( a \). Therefore, our measurements of \( V(r) \) are in a regime where the cores of the two dislocations overlap and linear elasticity theory is not applicable.

5. Dislocations and 2D Colloidal Plasticity

Dislocations are important defects in solids. Crystalline materials often contain a finite concentration of dislocations, incorporated into the crystal either during its growth or under deformation conditions. These defects dominate the plastic behaviour of the materials. Colloidal crystals are also useful for studying plastic deformation physics of solids.

In all solids, applying an external stress forces the dislocations to move and rearrange, resulting in plastic deformations. For applied stress higher than the yield stress, the crystal...
Figure 10. Point defects as dislocation multipoles: (A) well-defined divacancy configuration. A Burgers circuit (solid line) fails to close if it crosses the core of the defect. A Burgers circuit that surrounds the defect core without crossing it (dashed line) closes as expected. The arrows indicate the Burgers vectors of the ‘embryonic’ dislocation dipole. (B) Configuration of the same divacancy about 2 s later. The divacancy is not well-defined any more and the system resembles a pair of dislocations with opposite Burgers vectors, whose gliding lines (dotted lines) are separated by $\approx 3\bar{a}$. When those two dislocations come close to each other, the original divacancy is recovered.

behaves as a liquid and undergoes viscoplastic flow. In this regime, for a constant external stress $\sigma$, the strain rate $\dot{\gamma}$ is constant and given by Orowan’s relation [26]

$$\dot{\gamma} = \rho_m b v,$$

(1)
Figure 11. Pair interaction between dislocations. Di- (top) and monovacancies (bottom). $P(r)$ was determined from a few thousand snap-shots of the system by measuring the distribution of the separation of the two fivefold coordinated particles found. $V(r)/k_B T$ was estimated from $-\log P(r)$. The solid curves are spline fits and the straight lines are linear fits to the particular regions of data.

where $\rho_m$ is the density of mobile dislocations, $b$ their Burgers vector and $v$ their mean velocity. Obviously, such macroscopic, mean-field type of relations neglect temporal and spatial fluctuations in the density and the velocity of the dislocations. The strong interactions between a large number of cooperatively moving dislocations give rise to rather complex and heterogeneous slip processes. During deformation, complex patterns of dislocation-rich ‘walls’ separating dislocation-depleted ‘cells’ are known to form [27]. Groups of moving dislocations form slip bands. The moving dislocations can pile-up against stable structures, such as walls and boundaries, which can subsequently break apart and give large bursts of activity. Clearly,
mean-field approaches that regard viscoplastic deformation as a smooth laminar flow do not give the correct description of the observed phenomena.

The statistics of dislocation motion in single-ice crystals during viscoplastic deformation was studied by Weiss and Grasso [28] using acoustic emission (AE) measurements. Acoustic waves are generated from the sudden local strain changes during dislocation motion. The experimental results indicate an increase in the AE background with increasing strain rate, originating from the non-cooperative motion of several low-velocity dislocations. Except from this continuous background noise, the AE signal contains individual, intermittent strong bursts of activity, corresponding to coherent motion of fast dislocations, presumably triggered by collapse of the various metastable structures (walls and cells). The distribution $P(E)$ of the energy $E$ associated with these intermittent bursts exhibits a power-law behaviour spanning several decades, namely $P(E) \sim E^{\tau_E}$, with $\tau_E = 1.60 \pm 0.05$.

The discrete dislocation dynamics simulations performed by Miguel et al [29] confirmed that dislocations in stressed crystals move in a scale-free intermittent fashion. A 2D cross-section of the crystal is considered with $N \sim 650$, randomly placed edge dislocations gliding parallel to their respective Burgers vectors $\vec{b}$. $\vec{b}$ is constrained to be parallel to the $x$ axis, an approximation justified by the strong plastic anisotropy of ice crystals which leads to a single-slip system. The shear stress created by a dislocation at $\vec{r} = (x, y) = (0, 0)$ is given by [25]

$$\sigma_s(\vec{r}) = b\mu \frac{x(x^2 - y^2)}{2\pi(1 - \nu)(x^2 + y^2)^2}, \quad (2)$$

where $\mu$ is the shear modulus and $\nu$ the Poisson’s ratio. Interactions between dislocations originate from this long-range stress. The authors consider the motion of these $N$ interacting dislocations under an external stress $\sigma_e$, by solving numerically the over-damped equations of motion (along the $x$ direction)

$$\eta \frac{dx_i}{dt} = b_i \left( \sum_{j \neq i} \sigma_s(\vec{r}_j - \vec{r}_i) - \sigma_e \right), \quad (3)$$

where $\eta$ is an effective friction and $b_i$ the Burgers vector of dislocation $i$. Phenomenological reactions describe dislocation annihilation and multiplication. Two dislocations with opposite $\vec{b}$ are annihilated if they get at distance closer than $2b$. An opposite-sign dislocation pair is introduced if the local stress exceeds a certain threshold.

The simulations show that most dislocations are arranged in slowly moving metastable structures. A small fraction of the dislocations however move intermittently with much higher velocities (higher than the induced external stress $v_\sigma = b\sigma_e/\eta$). The velocity distribution $P(v_i)$ follows a power-law behaviour $P(v_i) \sim v_i^\nu$ with $\nu = 2.5$. When a single dislocation performs a sudden move with velocity $v$, an acoustic pulse is emitted whose amplitude is proportional to $v$. During the cooperative motion of a number of dislocations, the acoustic energy $E$ emitted is proportional to the square of the collective velocity $V = \sum |v_i|$, i.e. $E = V^2$. The distribution of the calculated acoustic energy $P(E) \sim E^{\tau_E}$ scales with an exponent $\tau_E = 1.8 \pm 0.2$ in agreement with the experiments on crystals of ice.

The widespread intermittency and the scaling behaviour observed suggest the existence of an underlying non-equilibrium phase transition (‘yielding transition’). The authors point out a
similarity with driven elastic manifolds in the presence of disorder, e.g. fluid flow in porous media, vortices in superconductors, etc. In these systems there is a depinning transition at a critical driving force separating the stationary and moving phases. The intermittency originates from spatial heterogeneity due to the random pinning, and scaling is observed close to the critical point. In the case of viscoplastic deformation, the random pinning potential is not applied externally but is self-generated, originating from the various metastable dislocation structures. However, the exact nature of the non-equilibrium statistical mechanics of viscoplastic deformation and, furthermore, the existence of a yielding transition are still open questions.

5.1. Rheological properties of colloidal crystals

The rheological properties of colloidal crystals have received considerable attention since the early days of the field. Under a small applied stress, the colloidal crystal yields and flows as a non-Newtonian fluid, with a shear rate-dependent viscosity. Mitaku et al [30] measured the yield stress \( \sigma_y \) for colloidal crystals as a function of particle volume fraction and electrolyte concentration and found that in general \( \sigma_y \sim O(10^{-3} \mu) \), where \( \mu \) is the rigidity modulus of the crystal. The small \( \sigma_y/\mu \) ratio suggests a dislocation mechanism for the plastic flow of a colloidal crystal [31]. Weitz et al [32] and Lindsay and Chaikin [33] questioned the existence of a finite yield stress; they observed a power law creep, \( \dot{\gamma} = \sigma^\alpha \), with \( \alpha = 3–7 \). Theoretically, the motion of screw dislocations in colloidal crystals was considered by Jorand et al [34, 35] in an attempt to explain the shear rate-dependent macroscopic viscosity of the colloidal crystal. To our knowledge there is no theoretical study of motion of edge dislocations in a colloidal crystal, which would directly apply to the 2D colloidal crystal.

5.2. Observation of dislocations in a 2D colloidal crystal

Using the microscopy scheme described in subsection 2.5, it is possible to directly image defects such as grain boundaries and dislocations in an optically thin colloidal crystal. We studied the collective dynamics of dislocations in a 2D colloidal crystal during viscoplastic deformation. The 2D crystal was prepared from a suspension of 0.3 \( \mu \)m diameter spheres in a cell with a gap \( \sim 2 \mu \)m (see section 2 for experimental details). The design of our experimental cells prohibits direct application of a mechanical stress on the 2D crystal. However, stress can be applied on the 2D colloidal crystal indirectly, e.g. through an imposed chemical potential gradient. Specifically, we discover that one can prepare a highly compressed colloidal crystal by forcing particles in the thin-gap region, using electrophoretic flow. Typically, this was achieved by applying 10–20 V across the sample for a few minutes. During the experiment we obtained a dense two-layer crystal in the middle of the thin region, and a less dense monolayer 2D crystal towards the edge of the thin region (see figure 12). The particles slowly (at \( v \sim 1 \mu \)m \( s^{-1} \)) drift from the high chemical potential region (dense thin crystal) to the lower chemical potential region (3D bulk crystal). Equilibrium is reached after several hours. The drifting velocity is varying across the sample, which corresponds to a continuous deformation of the 2D crystal. The mechanism of this deformation is the motion of dislocations in the sample, as is easily verified by microscopic observation. The stress causing this viscoplastic deformation is roughly equal to the osmotic pressure difference imposed at the boundaries of the 2D region, \( \sigma \approx \delta \Pi \).

Using the result of Alexander et al [36]:

\[
\Pi = k_B T \rho_C(a),
\]

\[ (4) \]
Figure 12. Top: scheme for applying small stress on a 2D colloidal crystal using osmotic pressure difference. The top white region corresponds to the 3D bulk colloidal crystals. The middle region is a monolayer 2D triangular colloidal crystal, with lattice spacing $a \sim 1 \mu m$. The bottom dark region is a two-layer crystal with regions of either square or hexagonal symmetry, with $a \sim 0.9 \mu m$. The thin gap region extends over almost a centimeter and the 2D and two-layer regions extend over several millimeter. The arrows indicate the drift velocity of the compressed 2D crystal, towards the bulk 3D region. The dashed lines outline the various crystallites, whose size is several hundred $\mu m$. Bottom: cartoon of the osmotic pressure profile across the sample.

where $\rho_C(a)$ is the counter-ion density at the edge of the Wigner–Seitz cell, we obtained $\sigma \approx \delta \Pi \sim O(0.1) \text{ dyn cm}^{-2}$. The value of $\sigma$ is a few times the yield stress, $\sigma_y \sim 10^{-3} \mu$, since $\mu \sim 700k_B \mu m^{-2} \approx 30 \text{ dyn cm}^{-2}$.

Data were collected for about 1 h, starting a few minutes after the initial compression. Since the equilibration of the chemical potential in the sample is a much slower process, we can safely consider the applied stress to be constant and roughly equal to $\delta \Pi$ during our observations. The dynamics of dislocations during viscoplastic deformation of the 2D crystal were observed at $20 \times$ magnification (see figure 13), video recorded and processed using the same particle-tracking procedures as described in section 2. The dislocations appear in the images as dark ellipses, with primary axis parallel to the Burgers vector and about 11 pixels long, corresponding to a core.
Figure 13. Top: observation of dislocations in a 2D colloidal crystal. Size of window is roughly $260 \times 200 \mu m^2$. The blue circles indicate the core regions of dislocations. The dislocations are arranged in cellular structures, outlined by the dotted blue lines. Bottom: blowup of a $25 \times 25 \mu m^2$ square region, at $100\times$ magnification. The crystal lattice spacing is $a \approx 1 \mu m$.

size of $r_C \approx 5 \mu m$. Typically, the trajectories of roughly 50 dislocations in the field of view were followed for 30 s.

To estimate the velocity of the dislocations, the trajectory has to be smoothed so that noise is reduced. We used boxcar (‘moving’) averaging with a window size 15 frames (0.5 s). After smoothing, the first and last 20 points of the trajectory were discarded. Figure 14 shows typical results. Dislocations that remained stationary with respect to the crystal were used to measure the drift velocity. The average drift velocity was $\vec{v}_{\text{drift}} = (v_x, v_y) = (1, -0.4) \mu m \text{s}^{-1}$. $\vec{v}_{\text{drift}}$ was used to convert the absolute, ‘camera’ coordinates to coordinates relative to the crystal frame:

$$(x_{\text{real}}, y_{\text{real}}) = (x_{\text{abs}}, y_{\text{abs}}) - (v_x, v_y)t.$$  

(5)

An example of dislocation trajectories (in the crystal frame) is shown in figure 15. We estimate the strain rate $\dot{\gamma}$ from the increase with time of the relative separation $\delta u/\delta R$ of two stationary

New Journal of Physics 7 (2005) 33 (http://www.njp.org/)
dislocations at an average distance $\vec{R}$ from each other. Typically during the measurements, $\dot{\gamma} \sim 10^{-3} \text{s}^{-1}$.

5.3. Results

The phenomenon describing the experiments on stressed ice crystals and the dislocation dynamics simulations seems to apply to the viscoplastic deformation of 2D colloidal crystals. Most of the dislocations are arranged in slowly moving cellular structures. Occasionally, we observe a few dislocations threading through the crystal at high speeds. They are emitted from highly strained regions (e.g. grain boundaries) or they enter the 2D region at the interface with the bulk 3D crystal.

Figure 14. Top: $y$ coordinate of a dislocation relative to the crystal. Inset shows the trajectory of the dislocation. Bottom: $v_y$ of the dislocation, calculated as the derivative of the boxcar averaged $y$. Averaging window is 0.5 s.
Figure 15. Trajectories of dislocations in the frame of the drifting 2D colloidal crystal. The axis labels are in \(\mu\)m. Total duration is 30 s. Arrows at top left indicate crystallographic directions. Since only dislocations with Burgers vectors parallel to \(a\) and \(b\) are observed to glide, the applied stress direction (blue dashed line) must be roughly perpendicular to the third direction \(c\).

Figure 16. Statistics of dislocation velocities. Different symbols correspond to the data collected at different regions of the 2D crystal.

They can also originate from a collapse of parts of the cellular structure. These fast dislocations typically pile-up against walls, or can be re-absorbed in regions of high strain. Following the guidelines of [29], we analysed the statistics of the velocities of the dislocations. The distribution \(P(v)\) was measured at four different places in the 2D region of the sample, separated by a few hundred \(\mu\)m from each other. The results are shown in figure 16. \(P(v)\) is quite broad, exhibiting the scaling behaviour \(P(v) \sim v^\nu\) with \(\nu \approx 2.9\), in agreement with the value \(\nu = 2.5\) found in the simulations.
Figure 17. Number of fast-moving dislocations. Typically, $N$ is a few percent of the total number of dislocations in the field of view.

Elastic distortions in colloidal crystals cannot propagate as waves since its normal modes are overdamped. Thus during the motion of dislocations, no acoustic waves are emitted and all the energy is dissipated in the hosting bath. We can still define a function $E = V^2$ as in the simulations. However, $E$ should be thought of just as a parameter that characterizes the magnitude of a rearrangement event, rather than acoustic energy. The collective dislocation velocity, $V = \sum' |v_i|$, is the sum of the magnitudes of the velocities of the fast-moving dislocations, i.e. dislocations moving faster than if moving only under the influence of the external stress. The external stress induced velocity is $v_\sigma = \sigma_e b / \eta$, where $\eta$ is the effective viscocity for dislocation motion, equal to a few times the viscosity of water [35]. For $\sigma_e \sim 10^{-2}$ dyn cm$^{-2}$, $b = 1 \mu$m and $\eta \sim 10^{-2}$ poise, $v_\sigma \approx 1 \mu$m s$^{-1}$, which is the threshold value we considered for defining fast dislocations. Typically, there were about five fast dislocations in the observation region (figure 17). The correct formula is $E = (\sum' |v_i|^2)^2$ (data not shown) resembles the acoustic emission signal from the stress ice crystal experiments, exhibiting intermittent and pronounced bursts. Each burst corresponds to collective dislocation rearrangements. The statistics of these bursts show a power-law decay, $P(E) \sim E^{-1.6}$, consistent with the results in the experiments and the simulations. Unfortunately, the maximum $E$ we can measure is limited by the small observation window, giving only 1 decade of scaling. Finally, to estimate the contribution of every fast dislocation to the $E$ signal, we determined the relation between $V$ and $N$. Since the dislocations move coherently, one expects $V \propto N$, with the proportionality constant roughly equal to the average velocity $v_m$ of fast dislocations. We obtained $v_m = 3.46 \mu$m s$^{-1}$. For an average $N$ of about five mobile dislocations in the field of view and using Orowan’s relation, we obtain $\dot{\gamma} \approx 3.5 \times 10^{-4}$ s$^{-1}$, consistent with the measured values ($O \sim 10^{-3}$ s$^{-1}$).

6. Concluding remarks

We have successfully used submicron monodispersed, charged polystyrene microspheres for studying a number of basic problems in condensed matter physics. For 1D colloidal lattice, we
showed that, aside from a quantitative issue which is likely specific to charged colloids, the qualitative feature of the experimental result confirms the Landau–Peierls prediction and the equilibrium behaviour of a 1D colloidal crystal (lattice) is indeed similar to a liquid, the density distribution is homogeneous. Anchoring the ends can induce some local order, but cannot restore long-range order.

We showed that one can introduce point defects in colloidal crystals, literally by hand, through manipulation of individual particles with optical tweezers. Using digital video microscopy, we observed that the stable configurations of the defects have reduced symmetry compared to the triangular lattice and identified distinct topological features in the arrangement of the system in every configuration. This analysis demonstrates that topologically the point defects correspond to dislocation multipoles with zero total Burgers vector. The individual dislocations are bound by an attractive interaction. Divacancies in particular are observed to dissociate into two dislocations that can separate several lattice spacings before recombination. The latter observation is particularly intriguing since it suggests that if a giant vacancy (more particles are missing) is created, it probably will disappear into the 2D solid in the form of dislocations, namely causing the whole 2D solid to melt.

A variety of further experiments could follow the work presented here. First would be to study if one can mechanically melt a 2D colloidal crystal using the optical tweezers to create giant or many vacancies. Another straightforward extension of this work is to vary the ionic strength or the inter-particle spacing, and study the properties of the defects as a function of the interaction strength and range. Performing a similar experiment in a 2D colloidal crystal of larger particles, the dynamics of the system could be slowed down appreciably (equivalently one could use a CCD camera with higher frame-rate). Utilizing magnetic particles, one could study point defects in a 2D crystal where the interaction is dipolar, \( V(r) \sim 1/r^3 \). Finally, a challenging experiment would be one that could utilize multiple optical traps (e.g. using holographic optical tweezer arrays) to create defect complexes, e.g. vacancy-interstitial pairs. Of particular interest would be the verification of numerical prediction for the defect–defect interactions and the microscopic dynamics of defect reactions and annihilation.

We have shown that it is possible to directly observe the dynamics of dislocations during plastic deformation of a 2D colloidal crystal. Using an osmotic pressure gradient, one can exert stress of order \( 10^{-1} \) dyn cm\(^{-2}\). The dislocations move intermittently, giving rise to sudden bursts of activity. The statistics of these rapid rearrangements show scaling behaviour, in agreement with experiments in ice crystals and numerical simulations. A more careful control of the externally applied stress is required in order to answer important questions regarding a possible ‘yielding’ transition and its exact nature. Furthermore, refinements of the techniques presented here can be used to address a variety of problems in plasticity (e.g. work hardening), using the colloidal crystals as a model system.

We also used charged colloids to study problems related to random pinning. This work will be described elsewhere.

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