Assembling and Manipulating Two-Dimensional Colloidal Crystals with Movable Nanomagnets

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Westudy crystallization of paramagnetic beads in a magnetic field gradient generated by one-dimensional nanomagnets. The pressure in such a system depends on both the magnetic forces and the hydrodynamic flow, and we estimate the flow threshold for disassembling the crystal near the magnetic potential barrier. A number of different defects have been observed which fluctuate in shape or propagate along the crystal, and it is found that the defect density increases away from the nanomagnet. We also study the melting of the crystal/fluid system after removal of the nanomagnet and demonstrate that the bond-oriental order parameter decreases with time. The nanomagnet can be moved in a controlled manner by a weak external magnetic field, and at sufficiently large driving velocities we observe self-healing crack formation due to deformation by a roughening of the lattice as well as gap formation. Finally, when confined between two oscillating nanomagnets, the colloidal crystal is shown to break up and form dipolar chains above a certain oscillation frequency.

1. Introduction

Colloid science is of considerable importance in industry as well as for the understanding of basic physical phenomena. The fact that colloids can be visualized directly using a microscope has made them ideal for model studies of structure and crystallization processes. In the past decade the photonic crystals have boosted the interest in three-dimensional colloidal crystals growth. However, also twodimensional crystals at the air—water or water—solid interface have been used as model systems for understanding physical properties and pattern formation of micrometer-sized colloidal systems. Two-dimensional self-assembly of colloidal crystals is revealed in thin liquid films or evaporating drops and these methods have been fruitful for well-controlled growth of colloidal crystals. In general, colloidal systems can be manipulated by electric and magnetic fields which induce dipolar interactions between the particles, thereby creating novel phases as well as model systems for crystal growth and disassembly. This kind of system may function as a model system for studying the influence of electric or magnetic fields on growth of thin organic films. It is well-known that crystallization in confined polymeric and colloidal systems can be enhanced by flow. Moreover, recently it has been demonstrated that localized electrostatic fields can be used to generate colloidal patterns.

In this work we demonstrate the growth of two-dimensional colloidal crystals by using a strong magnetic field gradient. That is, we take advantage of the fact that paramagnetic beads feel a force in magnetic field gradients and investigate the influence of hydrodynamic drag and confinement on the crystallization process. Moreover, we investigate how the crystal melts when the domain wall is totally removed, as well as its response to an oscillating magnetic domain wall.

2. Experimental Methods

The magnetic potential well was created experimentally using a bismuth-substituted ferrite garnet film. The garnet film has very large (1–10 mm) in-plane magnetized domains separated by domain walls, and these domain walls (of effective width approximately w = 50 nm) act as nanomagnets creating a potential well that attracts paramagnetic beads. A domain wall is defined as a continuous rotation of the magnetization vector from one direction to another. Here we use mostly Bloch walls, where the magnetization vector rotates from the positive
3. Magnetic Force on Single Magnetic Beads

The strong magnetic field gradient generated by the domain wall is very important here, since it ensures the attraction of the beads to the domain wall. The magnetic field from the domain wall acting on a paramagnetic sphere resting on the magnetic film a distance \( r = xe_x + ae_y \) from the domain wall can be approximated by

\[
H_{DW} = \frac{M_s w r}{2\pi} \tag{1}
\]

The force from the domain wall on a single magnetic bead is then found by noting that the dipole can be associated with an energy \( E = -\mu_0 m \cdot H_{DW} \), where \( \mu_0 \) is the permeability of water, and the magnetic moment is given by \( m = (4\pi/3)a^3 \lambda M_{DW} \). The magnetic field required to saturate the magnetic moment of the beads is of the order 100 kA/m. We operate here with fields that are orders of magnitude smaller than this, so that the linear relationship given above is a good approximation. Then the force is found to be (assuming \( z = a \))

\[
F_x = -\frac{Ax}{(x^2 + a^2)^{3/2}} \tag{2}
\]

\[
A = \frac{2\mu_0}{3\pi} a^3 (M_{DW})^2
\]

It should be emphasized that \( H_{DW} \) is much smaller than the field required to saturate the magnetic moment of the beads, and therefore the given expression is a good approximation. Expanding eq 2 to first order (when \( x \) is small) gives \( F_x = -kx \), where \( k = Aa^4 \).

We have measured \( k \) for a single sphere in the potential well through kinetic measurements (i.e., adsorption of a single bead to the domain wall) and also with direct sampling of the probability vs position \( x \) converted to an energy landscape assuming that the probability follows a Boltzmann distribution. A bead located close to the domain wall (i.e., \( x \) is small) has a probability density \( p(x) \) of being at a position \( x \) given by

\[
p(x) = p(0)e^{-E(x)/k_BT} \tag{3}
\]

where \( k_B \) is Boltzmann’s constant and \( T \) the temperature (\( T = 300 \text{ K} \)). Equation 3 is easily transformed into

\[
\frac{E(x) - E(0)}{k_BT} = \ln \left( \frac{p(0)}{p(x)} \right) \tag{4}
\]

Using the expression for the magnetic energy, we find that a bead in the well experiences a harmonic potential energy given by

\[
\frac{E(x) - E(0)}{k_BT} = \frac{1}{2} k \frac{x^2}{k_BT^2} \tag{5}
\]

To obtain the probability distribution, we performed about
4000 measurements of the $x$-coordinate of single beads\(^{27}\) and plotted it in a histogram (see Figure 4a). It can be seen that the distribution is approximately Gaussian, which is due to the fact that thermal excitations are responsible for the small excursions from $x = 0$. The probability distribution can then be converted into an energy landscape using eq 4, and the result is plotted in Figure 4b. Fitting eq 5 to Figure 4b gives $k = 10^{-7}$ N/m, which corresponds to an effective domain wall width of $w$.

\(^{27}\) The displacement accuracy is here about 60 nm, and we have an optical signal-to-noise ratio of about 100. Note that the displacement accuracy (change in $x$ between pictures) differs from the accuracy in absolute position, which is determined by the diffraction limit (about 0.5 $\mu$m).
beads move with velocities in the bulk (estimated by tracking beads in the bulk), the liquid flow can be as high as \( v_L \) near the interface due to the increased drag coefficient measured to be \( v_L \). Here \( v_L \) has been plotted into a histogram, whereas in (b) this probability distribution has been converted into an energy landscape. The energy can to a reasonable approximation be fitted to the parabola of eq 5.

\( \approx 20 \text{ nm} \). We emphasize that several different domain walls have been used in the current study, and measurements similar to the one above (as well as kinetic measurements) suggest that their effective widths vary between 20 and 100 nm. The reason for this variation is that important properties (e.g., magnetic uniaxial anisotropy) depend on the local environment (e.g., stress distribution), thereby creating domain walls of different width. The typical width is about \( w = 50 \text{ nm} \), which is the value we will use in our theoretical estimates.

4. Colloidal Crystallization and Melting

In this section we investigate how the crystal assembles in the presence of flow and show the different types of defects observed in it. Finally, we characterize its melting after the domain wall has been removed.

4.1. Crystal Assembly in Liquid Flow. When enough beads are in the vicinity of the domain wall, they assemble into a crystal structure due to the attraction to the domain wall. It should be emphasized that even though the domain wall may give rise to a symmetric magnetic field distribution, the liquid flow of velocity \( v_L \) ensures that the two-dimensional colloidal crystal only forms at one side of it. Although the liquid flow can be as high as \( v_L = 50 \mu \text{m/s} \) in the bulk (estimated by tracking beads in the bulk), the beads move with velocities \( v_B \) between 0.1 and 10 \( \mu \text{m/s} \) near the interface due to the increased drag coefficient there. The hydrodynamic drag is then given by \( \eta f_{\text{av}} v \), where \( v = v_L - v_B \) is the relative velocity between the liquid and the beads. Here \( \eta = 10^{-3} \text{ N s/m}^2 \) is the viscosity of water, and the hydrodynamic drag coefficient has previously been measured to be \( f = 30 \) through diffusion measurements using the Stokes–Einstein relationship. For a crystal at rest the liquid velocity is the only relevant quantity, and the hydrodynamic drag is given by \( \gamma f_{\text{av}} v \).

In addition to drag forces, the beads are also exposed to the magnetic field from the nanomagnet. A simple estimate of the maximum dipolar interactions between two spheres in a magnetic field \( H_{DW} = M_w w(2a)^3 \) gives \( E_d = \mu_0 M_w^2 / [2\pi (2a)^3] = 3k_B T \). The field acting on the magnetic beads far away from the domain wall is orders of magnitude smaller than \( H_{DW} = M_w w(2a)^3 \), and the actual dipolar energy is therefore comparable to the thermal energy also after summation over the whole lattice. Since the beads are coated with a carboxylic acid group, we estimate the electrostatic interaction between two beads in close contact to be about 50 \( k_B T \), which suggests that such interactions keep the crystal from being close packed.

However, for the moment we neglect the repulsive electrostatic interactions, so that the force on a layer of beads at rest a distance \( x \) from the nanomagnet can be given by

\[
F_x = \eta f_{\text{av}} v_L - \frac{A x}{(x^2 + a^2)^2}
\]

Here we have assumed that the hydrodynamic drag is in the positive \( x \)-direction. Moreover, we assumed that at every point in the crystal a constant drag force is acting on the beads. We will argue here that in the presence of an external flow this constant force assumption can explain our results at least qualitatively. First, we observe that the first few layers are always well ordered due to the strong magnetic forces here. Second, if the liquid flow is removed (\( v_L = 0 \)), one observes that while the rest of crystal becomes soft (and the number of defects increases) the first few layers are still well ordered. Figure 2b shows the force (eq 6) as a function of \( x \) when \( \eta = 10^{-3} \text{ N s/m}^2 \), \( f = 30 \), \( v_L = 5 \mu \text{m/s} \), \( M_s = 10^5 \text{ A/m} \), \( w = 50 \text{ nm} \), and \( \gamma = 0.17 \). Figure 2b suggests that the pressure is rather high near the domain wall but quickly decays so that about 5–10 layers away from the domain wall it is mainly governed by hydrodynamic drag. We emphasize that only the average pressure is discussed here, and fluctuations have not been considered so far.

If the drag force from the moving liquid is larger than the maximum attractive magnetic force, the beads will be pushed over to the other side of the nanomagnet and finally released. The maximum magnetic force is \( (0.6\gamma v_L / 3\gamma - M_w w)^2 \), which suggests that the maximum velocity is \( v_L^{\text{max}} = 0.6\gamma v_L / (3\gamma va/M_w w)^2 \), thus giving \( v_L^{\text{max}} = 16 \mu \text{m/s} \). This is not too far from the experimental value, found to be between 1 and 10 \( \mu \text{m/s} \), depending on the local environment and geometry of the boundary. In the rest of this study we will only be concerned with crystals which are exposed to negligible flow (i.e., \( v_L \approx 0 \)), to probe the influence of the magnetic field on the crystal in more detail.

4.2. Defects. As can be seen from Figures 2 and 3, several different defects exist in the colloidal crystals, and we will discuss them in the following.

The most common defects in colloidal crystals have 6-fold symmetry. These are basically lacking only one bead and therefore cost minimal amounts of energy. In addition, we also found 5-fold defects, in particular far away from the nanomagnets and in regions were mismatched lattices met (see, e.g., Figure 3). The 5- and 6-fold defects are seen to exhibit shape fluctuations due to the Brownian motion of the surrounding beads. Two examples of the possible shapes for a 6-fold defect are shown in panels a and b of Figure 5, which shows two successive pictures taken with an interval of 2 s. In (a) the bead on the left moves a little bit toward the vacancy, whereas in

![Figure 4. Probability distribution (a) and energy (b) for a magnetic bead in the potential well of a domain wall.](image-url)
In addition to shape fluctuations, the 6-fold defect may also move to another location. However, diffusion cannot support this process in a rigid crystal lattice where the diffusion coefficient is much smaller than that for a free particle. On the other hand, we observed that the hexagonal defect could propagate by a transformation into a 5-fold defect with two connecting dislocation lines when exposed to flow. Note that some beads are located on top of the lattice, appearing as black points.

(b) the two beads above and below the defect squeeze together. Both processes are associated with energies of approximately $k_B T$.

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anisotropic. Figure 8a shows the evolution of the average width of the colloidal system with time (note that the width is fluctuating slightly in the y-direction), which suggests a linear relationship for the time span studied here. The disassembly of the crystal is accompanied by excitations, where vacancies are created and some of the beads hop on top of the others or plow through the lattice in the x-direction thereby creating elliptical vacancies; see Figure 7b). The reason for this is that some more energetic particles push extra hard on their surrounding, thus creating vacancies upon leaving. After 3–4 s the number of vacancies is so large that the colloidal structure appears rather homogeneous. To obtain a quantitative measure of the melting of the crystal, we use the 6-fold bond-oriental order parameter.\(^2\) For a site \(k\) whose nearest neighbors labeled by \(j\) are arrayed by angles \(ı_{kj}\) from a certain reference direction, this order parameter is given by

\[
\psi_{6k} = \langle \exp(i6ı_{kj}) \rangle
\]

where the angle brackets denote an average over nearest neighbors and also over all the beads in the small area considered here (about 25 beads). Due to the finite size of this crystal, we selected a small area of it in order to describe melting using this order parameter. Its magnitude is equal to unity when we have a perfect hexagonal lattice. Figure 8b displays the evolution of the magnitude \(|\psi_{6k}|\) of the order parameter. It is clearly seen that it decreases with time, which can just as well be spotted in Figure 7. We notice that the order parameter decreases most strongly directly after release, and the melting therefore happens within few seconds. It should also be pointed out that after removal of the domain wall periodicity is also lost. However, we will not investigate this here.

Figure 7. Time evolution of a system initially in the crystalline phase (a) over a period of 20 s after the 1D nanomagnet has been removed. Here there is no flow. In image b, which was taken 2 s after removal, one observes that several holes have developed. Images c and d are taken 10 and 20 s after removal, respectively. The black scale bar is 65 \(\mu\)m.

5. Oscillating Colloidal Crystal

A particular advantage of using magnetic domain walls to assemble colloidal crystals is that they can be modulated by an external magnetic field. By applying a weak magnetic field slightly larger than the domain wall coercivity (100 A/m) in the y-direction, we may displace the domain wall in the x-direction. Reversing the sign of the field also reverses the motion. The required magnetic field is weak and does not induce significant magnetic moments in the paramagnetic beads. The only relevant field is therefore due to the domain wall.

Using a TTI waveform generator (TGA1242, 40 MHz) and a small electromagnet, we imposed a weak, oscillating in-plane magnetic field on the domain wall, thereby causing the average relative position of the layers in the crystal to oscillate as \(u_0(t) = u_{0N}(x) \sin(\omega t)\), where \(u_{0N}(x)\) is the amplitude of the Nth layer, \(x\) is the distance from the domain wall, and \(\omega = 2\pi f\) is the angular frequency. The corresponding instantaneous velocity is \(v_0(t) = v_{0N}(x) \cos(\omega t)\), where \(v_{0N}(x) = \omega u_{0N}(x)\). Figure 9a shows the oscillation of different layers of the crystal when \(u_0 = 0.4a\) and \(f = 0.3\) Hz. The second layer (\(N = 2\), open circles) oscillates fully in phase with the external magnetic field, whereas the fifth layer (\(N = 5\), open squares) is slightly out of phase. We also note that this layer does not show pure sinusoidal behavior. Moreover, in the ninth layer (\(N = 9\), open triangles) the forced excitations are of the same magnitude as the thermal fluctuations of the lattice, and it is therefore difficult to distinguish the oscillations from noise.

An important observation from Figure 9a is that the amplitude of the oscillations decreases as one moves away from the domain wall. A clearer picture of this is shown...
in Figure 9b, where $u_N/a$ is displayed as function of the layer number $N$ for two different frequencies, $f = 0.3 \text{ Hz}$ (open squares) and $f = 1 \text{ Hz}$ (open circles). In the experiments we also observed thermal fluctuations of the particles in the crystal lattice, and the position $x_d$ where the magnetic energy equals the thermal energy is given by

$$x_d = a\left(\frac{\mu_0\gamma a^3(M_s w)^2}{\pi 3k_B T} - 1\right)^{1/2} \quad (8)$$

which we here find to be $19 \mu m$. The fact that the observable oscillations extend to about $25 \mu m$ we may attribute to either the experimental inaccuracy or the result of collective effects. One may naively expect from our single particle model that the amplitude of each layer decays as $u_{N0} \propto x^{-3}$. However, experimentally we observe that the decay is nearly linear beyond the two first layers, which suggests a collective behavior where the particles interact electrostatically. We also found that the decay is very similar for different frequencies (and velocities), which may indicate that hydrodynamic interactions are not essential as long as the collective motion is periodic. Then, electrostatic interactions between the electrically charged magnetic beads are expected to be most important in a nearly close-packed lattice. The characteristic time $\tau$ required to displace a single isolated bead by $\delta/2$ about a position $x_0$ is given by $\tau \approx \eta f_0 a x_0^3/\Lambda$. As an example, assume that $\delta = a$ and $x_0 = 1.8a$ ($N = 2$), which gives $\tau = 0.3 s$. This should be contrasted to the case $\delta = 0.5a$ and $x_0 = 7a$ ($N = 5$), which gives $\tau = 9s$, and it is more difficult for this layer to follow oscillations of frequency $0.3 \text{ Hz}$, in agreement with our observations. Therefore, only by considering the single bead dynamics one may infer that the oscillations of crystal layers far away from the domain wall may not follow those close to it given that the displacement $\delta$ is sufficiently large.

In the case where the hydrodynamic drag is sufficiently larger than the magnetic force on the $N$th layer, the beads can no longer follow, and the crystal will break in its vicinity; see Figure 10. In addition, a video in AVI format available as Supporting Information shows cracking of a colloidal crystal in real time.28 Experimentally we found that the position of the crack depends on both the frequency and amplitude of the oscillations. As an example, we consider in Figure 10 the case $u_{N0} = 0.7a$ and $f = 0.3 \text{ Hz}$. In Figure 10a the crystal is maximally compressed and nearly hexagonally close-packed. Figure 10b shows the same crystal 1 s later, and it is seen that the lattice planes are no longer as well ordered as that in Figure 10a. In Figure 10c the crystal is maximally stretched, and in addition to the disordering in Figure 10b, we observe that the lattice planes are separated by gaps on several locations. In Figure 10d, the lattice is again maximally compressed, and the hexagonal structure has been restored. It should be emphasized that our crystal is different from brittle solids which exhibit fast crack propagation due to breaking of atomic bonds.29 On the other hand, we do observe both localized coarsening of the lattice as well as gap formation between the $N$th and $(N-1)$th layer, and crack formation in our system therefore has some similarities with that of a brittle solid. We will here characterize the crack formation by a roughness...
parameter $\Delta_N$ and a measure of the gap $\Delta u_N$. The root-
mean-square (rms) deviation from the average position 
$u_N(t)$ of the $N$th lattice, $\Delta u_N(t) = (\langle u_N(t) - u_N(t) \rangle^2)^{1/2}$, is taken 
to be the roughness parameter. The gap parameter $\Delta u_N$ 
is taken to be the difference between the average position 
of lattice $N$ and $N - 1$, $\Delta u_N(t) = u_N(t) - u_{N-1}(t)$. In Figure 
11a, $\Delta u_N$ is displayed as function of time for $N = 2$ (filled 
squares) and $N = 3$ (filled circles) when $f = 0.3 \text{ Hz}$. The 
roughness of layer $N = 2$ does not change significantly as 
the crystal boundary oscillates, see also Figure 10c, and 
the only contribution to $\Delta_2$ is Brownian motion. On the 
other hand, the ordering of the layer $N = 3$ is partially 
destroyed, and $\Delta_3 = 0.2$ when the crystal is maximally 
expanded. We also find that the roughening of the lattice 
is periodic with slightly varying magnitude, and almost 
in phase with the oscillation of the domain wall. In Figure 
11b we show the distance between the lattices, $\Delta u_2(t)$ (filled 
squares) and $\Delta u_4(t)$ (filled circles), when $f = 0.3 \text{ Hz}$. Note 
that the average distance between $N = 3$ and $N = 4$ 
oscillates between 2.4a and 1.8a, thus indicating a gap 
formation in addition to the roughening discussed above.

On the other hand, far away from the domain wall the 
distance between lattice planes appears to be uncorrelated 
with the domain wall motion. One may expect the crack 
to occur at a distance $x_c$ where the hydrodynamic drag 
force is equal to the magnetic force. We thus obtain

$$x_c \approx \left( \frac{2 \mu \gamma a^2 (M_w l)^2}{3 \pi \eta w_1 g} \right)^{1/3}$$

which gives $x_c \approx 2.4a$ for $u_{100} \approx 2 \mu \text{m/s}$ (experimental 
value from Figure 10) and the parameters given above.

We see that this estimate deviates slightly from the 
experimental data, which tell us that the roughening and 
gap formation start at $x \approx 4a$. Further measurements of 
$\Delta u_N$ show that the roughening of the lattice is rather 
localized; see Figure 11c, where the circles correspond to

an expanded crystal, whereas the squares correspond to 
maximal compression. The crack is seen to be localized to 
layers between $N = 3$ and $N = 7$. In a normal brittle solid 
one would expect cracks to be a major origin of stress 
relief, where big cracks may grow at the expense of smaller 
one. However, in our system these cracks do not

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Figure 10. Breaking of a colloidal crystal subject to a modulation. The images show a portion of a colloidal crystal self-assembled 
near a domain wall. Images a and b are separated by 1 s, images b and c by 0.5 s, and finally images c and d by 1.5 s. The scale 
bar is 7 \text{ \mu m}. See also a video in AVI format available as Supporting Information.

Figure 11. Crack formation is accompanied by a local change 
in the roughness of the lattice. In (a) the rms deviation $\Delta u_N/a$ 
for $N = 2$ (filled squares) and $N = 3$ (filled circles) is shown as 
function of time when the domain wall oscillates at a frequency 
f = 0.3 \text{ Hz}. In (b) the gap $\Delta u_N/a$ is shown as function of time 
for $N = 2$ (filled squares) and $N = 4$ (filled circles). In (c) the 
roughness is displayed for two different times (separated by 
about 1.5 s) in the periodic cycle. The filled circles correspond 
to an expanded lattice, and the filled squares correspond to a 
compressed lattice. We note that the roughness of the lattice 
may appear to increase away from the domain wall due to 
decreasing magnetic attraction.
represent a similar source of stress relief since there are basically no attractive interactions between the colloids, and the attraction to the domain wall is the only force holding the crystal together. In fact, the crack increases the distance to layers beyond it, and these will always try to reduce their magnetic energy by moving toward the domain wall. The cracks may therefore be said to have a self-healing behavior (which we have also observed directly by imposing a pulse instead of sinusoidal oscillations), at least if the domain wall is brought to rest. One may also wonder why there is a roughness associated with each crack, since our simple model predicts that the crystal should be cleaved nicely upon cracking. However, it must be emphasized that each bead is not identical, and therefore the hydrodynamic coupling to the underlying surface also varies. This results in a distribution of different values for $f$, and therefore also $x$. Thus, one may say that hydrodynamic coupling to the underlying surface is not so essential for the collective behavior (although it does determine some properties of the crystal) when the lattice oscillates in a relatively periodic way but becomes important when the crystal breaks.

### 6. Dynamically Induced Transition

So far we have only discussed what happens when a single domain wall is used as an oscillating interface. It is also possible to let the beads crystallize between two magnetic domain walls as seen in Figure 12. We found that by choosing the magnetization direction in the domain walls so that the $x$-component of the magnetic field from both the walls points in the same direction, a new dynamic effect occurs. As can be seen in Figure 12a, the beads crystallize near the domain wall. Moreover, the magnetic moments of the beads are aligned in the $x$-direction, thereby creating dipolar chains. A simple estimate using the field from the domain walls suggests that the dipolar energy of the chains is of the order $k_B T$, and they are therefore rather weak; see also Figure 12a. Thus, as long as the domain walls are at rest, the attraction to the domain wall is stronger than the dipolar interactions, and most of the beads prefer to form a hexagonal crystal structure. By oscillating the domain walls $180^\circ$ out of phase with $u_0 = a$, we found that at an oscillation frequency about 0.5 Hz a number of the beads are thrown out of the crystal and start forming dipolar chains. We explain this phenomenon qualitatively using the single bead model discussed above. This model predicts that a single bead is released from the domain wall when the velocity of the domain wall is larger than that allowed by hydrodynamic drag. Once the beads are "released" from the domain wall (i.e., the magnetic force is sufficiently weak), they are free.

Figure 12. Image (a) shows the crystallization between two domain walls. Since the domains outside and inside the domain walls have opposite magnetization directions (positive or negative $y$-direction), the domain walls always move in opposite directions when we apply an oscillating magnetic field in the $y$-direction, i.e., along the domain walls. When oscillating the two domain walls out of phase at $f = 1$ Hz, we observe chain formation perpendicular to the wall, as can be seen in b. The scale bar is 35 $\mu$m.

(30) This can be obtained either by choosing the magnetization vector of the domain walls to be pointing in opposite directions (positive or negative $z$-direction) or by selecting one domain wall pointing in the $z$-direction and the other in the $x$-direction (Néel wall). Near a single domain wall (with magnetization vector in the $z$-direction) one does not observe chains. The reason one can observe dipolar chains when two domain walls are present is that the fields from the two domain walls enhance each other.
to form weak dipolar chains; see Figure 12b. The critical frequency at which the beads at a distance $x_0$ are thrown out of the crystal is estimated to be

$$f_c \approx \frac{\mu_0 \gamma^2 (M_s w)^2}{\pi \eta u_0 x_0^3}$$  \hspace{1cm} (10)$$

Assuming $u_{10} = a$ gives $f_c \approx 1$ Hz when $x_0 = 1.8a$ ($N = 2$) and $f_c \approx 0.3$ Hz when $x_0 = 2.7a$ ($N = 3$). This is in qualitative agreement with what can be observed from Figure 12b, which tells us that at 1 Hz a significant portion of all layers $N > 3$ have been reformed into chains. Note that this estimate does not take into account dipolar or other interactions hindering the motion. However, a detailed study of these effects is most easily done by computer simulations, which is outside the scope of the current study.

7. Conclusion

We have studied crystallization in the magnetic field gradient generated by one-dimensional nanomagnets. It is found that the pressure in such a system depends on both the magnetic forces as well as the hydrodynamic flow, and we attempted to describe the pressure distribution in the system. A number of different defects are observed which propagate in the crystal or fluctuate in shape. Upon removal of the nanomagnet, the crystal quickly disassembles thereby forming a liquid state. The domain walls can be driven by weak external magnetic fields that do not alter the dipolar interactions between the beads, and we have found that this can be used to oscillate the crystals or probe dynamic phenomena. We showed that a single particle model can account for most of the phenomena observed here. To obtain a quantitative understanding, a theoretical model based on many-body electrostatic and hydrodynamic interactions must be developed, a task which is outside the scope of the current experimental study.

We believe that nanomagnets of reduced dimensionality can be used to assist directed assembly of colloidal systems. The method presented here offers a large amount of flexibility since it is rather easy to move the nanomagnets in a controllable manner, thus enabling high-resolution spatial and temporal control of the system. Such colloidal systems also represent a new class of self-healing materials which are not irreversibly destroyed upon cracking. In this context the self-assembled crystals studied here may improve our fundamental understanding of driven systems in soft matter physics.

Supporting Information Available: A video in AVI format showing cracking of a colloidal crystal in real time. This material is available free of charge via the Internet at http://pubs.acs.org.

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