
Melting of mesoscopic lattices of magnetic fluid thin film subjected to perpendicular fields

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Abstract

Applying a magnetic field on the magnetic fluid thin film perpendicularly, leads a phase separation that is concentrated in particles separating from a dilute phase. The concentrated phase forms cylindrical columns that construct two-dimensional lattices. This kind of artificial lattice is a novel mesoscopic system and has been explored with optical microscope, CCD, and digital imaging analysis. We explore the ordering evolution of the two-dimensional extraordinary lattice by varying the applied field. The ordering of these lattices is analyzed in terms of translational and bond-orientation correlation functions to address the two-dimensional melting.

1. Introduction

The versatile structures of magnetic fluid thin film subjected to an external field have attracted much interest because of the induced optical properties and potential applications [1–9]. It is well known that the application of a magnetic field perpendicular to the magnetic fluid thin film leads to a phase separation—the separation of the concentrated portion in magnetic particles from the liquid matrix of magnetic fluid. The concentrated phase forms cylindrical columns that construct two-dimensional ordering lattices under appropriate condition. Henceforth, the remarkable magneto-optical effects in ordered structure of magnetic fluid thin films, such as birefringence...
field-dependent transmission [12,15], and magnetochromatics [16,17] have been investigated.

In this study we explore the ordering variation of the lattices formed in the magnetic fluid thin film by varying the applied perpendicular field. The two-dimensional lattices present hexagonal phase with exotic topological defects due to distortion of the structure under excitation. The ordering evolution of the mesoscopic lattices can be addressed with the two-dimensional melting theory. There has been considerable interest in two-dimensional melting both in theoretical and experimental condensed matter physics [18,19], since Kosterlitz and Thouless (KT) constructed an elegant phase transition theory with $XY$ model in two-dimensional systems [20]. They claimed that the two-dimensional $XY$ model provides an unusual type of phase transition driving by vortices. The KT transition of superfluidity in two dimensions is the unbinding of vortex pairs of dilute gas. This novel transition is topological defect-mediated melting. Subsequently, Nelson and Halperin [21,22] and Young [23] have extended the ideas of the KT mechanism to the two-dimensional melting problem in contrast to the conventional order–disorder phase transition. They proposed a new scenario with two successive continuous phase transitions with an intermediate hexatic phase instead of a single direct first-order transition. In the two-dimensional crystal, the bond orientation order (BOO) parameter is long-range order (LRO), but the translational order parameter possesses only quasi-long-range order (QLRO). At a temperature $T_m$, the melting is driven by the breakup of thermally generated bound dislocation pairs, and the two-dimensional crystal transits the hexatic phase. Above $T_m$, the increase of the density of free dislocations will result in an exponential decay of the translational order parameter. However, the orientation order persists, in the sense the BOO decays only algebraically and displays QLRO. Such transition is a continuous phase transition. Subsequently, the spontaneous breakup of free dislocations into their constituent disclinations will drive a second continuous transition to an isotropic fluid from the intermediate hexatic phase. Experimental systems for studying the novel KTHNY two-dimensional melting theory have been explored by some researchers. These include electrons on helium [24], noble gases physisorbed on substrates [25], liquid crystals [26,27], polystyrene colloids [28], magnetic bubble arrays [29], and vortex arrays in high-temperature superconductors [30].

In this study we explore the lattices of magnetic fluid thin film subjected to perpendicular fields with microscope and pick up images with charge coupled device (CCD). Digitized images of the extraordinary lattices are examined and analyzed to address the novel mesoscopic two-dimensional system. The ordering evolution of the two-dimensional lattice is investigated by varying the applied field. We mimic that the usual thermal melting transition from the low-temperature ordered phase to a high-temperature disordered phase with the high-field ordered phase to a low-field disordered phase. Making analysis of the ordering of these extraordinary lattices in terms of translational and bond-orientation correlation functions. We address the ordering evolution of magnetic fluid thin-film lattices under the varied fields with the exotic two-dimensional melting theory.

2. Experiments and analysis method

Magnetic fluids are disperse systems consisting of colloidal magnetic particles and a continuous carrier medium [1–4]. Our homogeneous magnetic fluid consists of MnFe$_2$O$_4$ magnetic particles, which are dispersed in kerosene liquid matrix. The average diameter of the magnetic monodomain particles ranges from 5 to 10 nm. The nanoparticles are coated by a surfactant layer with the co-precipitation technique. The steric repulsion of surfactant covering makes the suspensions homogeneous. Its saturated magnetization is 8.2 emu/g. The magnetic fluid sample is sealed in a rectangular glass cell of 6μm thickness. The applied perpendicular magnetic fields are generated by Helmholtz coils, which are cooled by circulating water in copper tube. The thin film sample is maintained at room temperature. Because the viewing range is limited in the microscopic observation, the magnetic field is uniform in the small region of exploration. Through dipolar
interaction, dispersed particles attract or repel one another depending on their positions. In the absence of an external magnetic field, thermal energy leads the particles in Brownian motion. Whereas, with application of an external magnetic field, the magnetic particles tend to align with the field, and partially overcomes the thermal agitation. As the strength of the field reaches some critical point, the violation of thermodynamical stability occurs and the condensation initiates. The concentrated portion in magnetic particles will aggregate into droplet form. Both the size and the number of the droplet tend to increase with the magnitude of the applied field at this stage. As we intensify the magnetic field furthermore, the larger droplet splits into parts of appropriate size, because the repulsive force among parallel domains overcomes the bundling surface tension. In an intense magnetic field, the magnetic droplet divides into similar-sized columns that have similar magnitude of magnetic moment. The size of columns, and the separation between columns vary with the magnitude of the applied terminal magnetic field, the raising rate of the field, and the concentration of the magnetic particles in the fluid, etc. [5].

Through the dipolar interaction among columns, the columnar lattices of high-quality magnetic fluid in a thin film subjected to perpendicular magnetic fields can form two-dimensional lattices. The disordered pattern appears at lower magnetic fields first, and will arrange to be ordered two-dimensional hexagonal lattices at higher fields. Patterns are visualized with an Olympus transmission microscope (BX-51). CCD images are digitized with a threshold chosen to render the pattern faithfully recorded. They display high contrast black and white images. Digital images can be used to locate the center of each magnetic column.

In order to analyze the ordering of the two-dimensional lattice of the system, we use the digital data to calculate both translational and bond-orientation correlation functions [31]. The translational correlation function, \( G(r) \), is defined as the possibility of finding another point at relative distance \( r \) apart and can be written as

\[
G(r) = \langle \rho(r_0) \rho(r') \rangle,
\]

where the ensemble average is taken over all reference points \( r_0 \) and points \( r' \) with \( |r_0 - r'| = r \). The bond-orientation correlation function \( G_6(r) \) relating bond-orientation at relative distance \( r \) apart is defined as follows:

\[
G_6(r) = \langle \phi_6(r) \phi_6^*(r') \rangle,
\]

where \( \phi_6(r) \) is the local orientation order parameter and is defined as follows:

\[
\phi_6(r_i) = \frac{1}{6} \sum_{n.n.} e^{i \theta_{ij}}
\]

where \( \theta_{ij} \) is the angle between a fixed reference axis and the bond linking particles \( i \) and \( j \). The summation is taken over all the bonds of nearest neighbors. The bond-orientation correlation function measures the extent to which sixfold orientational order persists for separations comparable to \( r \). In this study we analyze the ordering and characterize the phase of two-dimensional lattices forming with magnetic fluid subjected to perpendicular fields in terms of the translational correlation function, \( G(r) \), and the bond-orientation correlation function, \( G_6(r) \), respectively.

3. Results and discussion

We acquire images of the magnetic fluid thin films subjected to perpendicular magnetic fields. Fig. 1 displays the typical image of ordered phase at field of 500 and 170 Oe, respectively. There are about 1340 centers in the plot. The insertion figure is the fast Fourier transformation (FFT) of the image. That the insertion figure shows six distinct bright spots around the center one indicates a fairly ordered hexagonal structure. The columns of concentrated magnetic fluid form a two-dimensional hexagonal lattice is evident. The distance between columns can be worked out by directly counting the number of image pixels from the location of the columns or calculating with the formula \( d = 2\pi/k \) from the \( k \)-space distance. The average distance between columns in this case is about 3.4 \( \mu \)m. Fig. 1(c) and (d) display the Delaunay triangulation plot of the image of Fig. 1(a) and (b), respectively. The lines connecting lattice points almost are parallel to one another.
Except in the small-hatched area, almost every lattice point has six coordinates. This also demonstrates that the observed structure is a well-ordered hexagonal lattice at high-field region.

While the applied field is decreased, the defects appearing in the ordered lattice increase abruptly below 130 Oe. Fig. 2 displays the images of 130 and 120 Oe of the applied field during the change.
The hatched region of the Delaunay triangulation plot displays the appearing of defects in the lattice. Shown in the Fig. 3 is the concentration of the regular six-bond lattice points and the defective five- and seven-bond lattice points. In the ordered state, the perfect six-bond lattice points are dominant, and less than 1% lattice points are occupied by the defect. The transition occurs at 120 Oe, where there exists a steep rising of the concentration of the defective lattice points.
In order to measure the ordering, we analyze with translational and bond-orientation correlation functions. The data of correlation functions are the ensemble average of 40 pictures taken at the specific magnetic field. We inspect the ordered phase first. Apply the perpendicular magnetic field of 500 Oe on the film for 2 h, the system will establish the equilibrium ordered phase. We then lower the applied field in 2 Oe step to explore the variation of the columnar lattice structure. In each step, the field is quickly decreased and we wait 5 min for equilibrium. Afterward we pick up 40 pictures in 2 min. In the high-field region, there is no significant variation of the ordered lattices. The analysis of typical ordered lattices at 170 Oe was made with the translational correlation function, $G(r)$, and of the bond-orientation correlation function, $G_6(r)$. The diagrams of $G(r)$, and $G_6(r)$ at 170 Oe, are shown in Fig. 4(a) and (b), respectively. They display succession of spikes, and therefore the two-dimensional lattice ascertains ordered phase. It is noted that the spike height of the bond-orientation correlation function almost keep constant, while that of the translational correlation function decays slowly. This demonstrates that the bond-orientation is more likely to persist despite the appearing of defects.

Shown in Fig. 5 is the evolution of translational correlation functions $G(r)$ of 170, 130, 120, 110 and 100 Oes. Series variation is presented. As for the bond-orientation correlation function $G_6(r)$, there appears distinctive variation from 120 Oes. Shown in Fig. 6 is the evolution of bond-orientation correlation functions $G_6(r)$ of 170, 130, 120, 110 and 100 Oes. The bond-orientation correlation function shows constant value in the high-field region, where defects are rare. As we
lower down the applied magnetic field, the hexagonal lattice gets disordered. Even defects increase to a significant amount, the bond-orientation correlation is persisted, and the peak height of oscillating bond-orientation correlation function decay algebraically slows. We make the algebraic function fitting, \( \sim r^{-\eta} \), and plot the diagram for the fitting exponents \( \eta \) and \( \eta_b \) for the correlation functions, \( G(r) \) and \( G_6(r) \), respectively. The exponents \( \eta_6 \) and \( \eta_b \) versus the magnetic field is shown in Fig. 7. As the field decreases further, the applied magnetic field will not be able to hold the systems’ ordered structure. The ordered lattice melts through a quick transition. We notice that the exponent of the fitting algebraic function increases rapidly at the melting threshold. The theoretical threshold exponent of bond-orientation correlation function, \( G_6(r) \), that decays in the two-dimensional system has the value of \( \frac{1}{4} \) [20,32]. Inspecting the experimental result, the exponent goes beyond \( \frac{1}{4} \) when the applied magnetic field decreases to 120 Oe which ascertains that the two-dimensional melting occurs. We also notice that the bond-orientation correlation function presents exponentially rapid decaying behavior below 120 Oe, which means that the observed lattice loses its bond-orientation correlation.

We further examine the behavior of the correlation length. The exponential fitting of the correlation functions was made with \( \sim \exp(-r/\xi) \). Fig. 8 shows the correlation length \( \xi \) and \( \xi_6 \) of translational and bond-orientation correlation functions versus the applied fields. Long correlation length addresses the buildup of LRO from 120 Oe. It is noted that the bond-orientation correlation function displays longer correlation length. This also demonstrates that the bond-orientation is more likely to persist. At 120 Oe, both correlation
lengths steeply decrease to the order of lattice spacing. Thus, we ascertain that the two-dimensional lattice melts as the applied magnetic field decreases to 120 Oe, with the mesoscopic lattice demonstrating randomly isotropic distribution.

4. Conclusion

By applying magnetic field on the magnetic fluid thin films perpendicularly, the concentrated portion phase separated from the liquid carrier matrix. The as-formed columns will arrange to ordered lattices under appropriate conditions. The applied field can be manipulated to simulate the temperature control processes. Allowing the mesoscopic lattice to get disturbed, leads to the order–disorder transition. Since this kind of order–disorder transition displays distinguished features of an equilibrium phase transition, it will be suitable to mimic two-dimensional phase transition. The mesoscopic crystal is essentially a defected crystal with dislocations destroying the translational symmetry in two-dimension. Nevertheless, the bond-orientation order will be sustained, thus the nonconventional order will be kept in the system. We study the ordering of the extraordinary magnetic fluid thin film lattices with the bond-orientation correlation function as well as the translational correlation function. The quantitative measurements reveal the following features of the transition. In a narrow range of magnetic fields, the correlation length $\xi$ and $\xi_6$ falls from a value comparable to the size of the system to a small value of lattice spacing. In the transition from the high-field phases characterized by constant bond-orientation correlation function to a low-field disordered phase, the long-range orientation order in the lattice decreases substantially. Examining the algebraic fitting of the bond-orientation correlation function, we find the power-law exponent shows an abrupt increase and at 120 Oe passes through $\frac{1}{4}$, which is the theoretical threshold for two-dimensional melting. The melting evolution in the artificial mesoscopic lattices of magnetic fluid thin films can be addressed with the exotic two-dimensional melting phenomena in this study.

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References