Pattern formation in a monolayer of magnetic spheres

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Pattern formation is investigated for a vertically vibrated monolayer of magnetic spheres. The spheres of diameter D encase cylindrical magnetic cores of length l. For large D/l, we find that the particles form a hexagonal-close-packed pattern in which the particles' dipole vectors assume a macroscopic circulating vortical pattern. For smaller D/l, the particles form concentric rings. The static configurational magnetic energy (which depends on D/l) appears to be a determining factor in pattern selection even though the experimental system is driven and dissipative.

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I. INTRODUCTION

Two-dimensional (2D) dipolar systems have been investigated using many different experimental and theoretical approaches [1]. Patterns have been observed experimentally in micron- to mm-sized magnetic particles suspended on the surface of a liquid [2–5], in a magnetic fluid containing nonmagnetic particles [4,6], in magnetic bubbles in garnet films [7], in mm- to cm-sized magnetically excited magnetized spheres [8], at the atomic level in micron-sized thin film magnetic materials [24–26], and recently in mm-sized vertically vibrated magnetic spheres [9]. Studies of biological systems also suggest that dipole-dipole interactions between proteins may be an important part, e.g., of microtubule formation [10] and pattern formation in a lipid monolayer [11]. Monte Carlo simulations of the thermal equilibrium of large 2D systems of hard spheres with centrally embedded point dipoles have shown that ordering in the absence of external forces depends on the dipolar strength, the temperature of the system, and the number density of the spheres [5,12–14]. At intermediate temperature, high density systems of dipolar spheres form hexagonal-close-packed (HCP) macrovortices [Fig. 1(a)], while intermediate density systems form networks of long branching chains [12]. At high temperatures, systems of dipolar spheres dissociate and become gaseous [9,12]. We report observations of pattern formation in a 2D system of vertically vibrated magnetic spheres, which depends also on the shape of the magnetic core. Our results show that spheres containing short cylindrical magnets form HCP macrovortices, while spheres containing long cylindrical magnets form concentric rings. In addition to the previous work on vertically vibrated magnetic spheres [9], several groups have investigated vertically vibrated monolayers of unmagnetized particles [15–17].

A calculation done by Belobrov *et al.* [18] showed that the zero-temperature ground state of a finite (<500 particle) HCP lattice of dipoles is a macrovortex [Fig. 1(a)], while the zero-temperature ground state of a finite square-packed lattice of dipoles is a microvortex [Fig. 1(b)]. The macrovortex state was later observed in simulations of a dense system of dipolar spheres in thermal equilibrium [12], and in an experimental system of settled magnetic spheres (a system of magnetic spheres that are initially magnetically excited with a solenoid, then allowed to settle at the bottom of a container under zero external magnetic field) [8]. A similar vortex pattern made by atoms in micron-sized thin film magnetic materials is also being investigated as an element of a future nonvolatile memory device [24–26]. However, to our knowledge, a macrovortex pattern has never been observed in an excited system of dipolar spheres. We provide direct experimental evidence of the macrovortex pattern in an excited system of dipolar spheres. We also provide direct experimental evidence of a microvortex pattern in a zero-temperature system of dipolar spheres.

II. EXPERIMENTAL SETUP

The magnetic particles used were cylindrical, ceramic, permanent magnets securely encased in a spherical hard plastic shell. We investigate particles encasing magnetic cores of

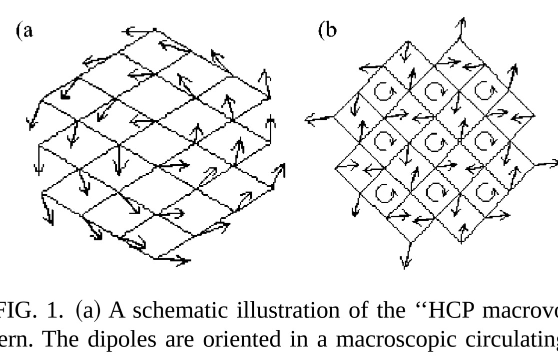

FIG. 1. (a) A schematic illustration of the ''HCP macrovortex'' pattern. The dipoles are oriented in a macroscopic circulating vortical pattern about the center. (b) A schematic illustration of the ''square-packed microvortex'' pattern. The dipole axes are tilted by about 45° with respect to the axes of the square pattern, and the dipole orientations are such that they define alternating directions of circulation (indicated by the rotational arrows in the centers of the squares) on a checker-board pattern of squares (after Ref. [18]).

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two types, which we call ''long'' and ''short'' magnets. The long magnets we used had length $l=1.42$ cm and diameter $d=0.94$ cm. The short magnets we used had $l=0.64$ cm and $d=0.95$ cm. Once the *long* magnets had been encapsulated in the spherical shells, the particles (*long* particles) were 5.30 g in mass, the maximum strength of the magnetic field on the surface of a long particle was 0.54 kG$\pm$16%, and the particle diameter D was 1.69 cm. The particles containing the *short* magnets (*short* particles) were 4.25 g in mass, the maximum strength of the magnetic field on the surface of a short particle was 0.12 kG$\pm$24%, and the particle diameter D was again 1.69 cm. All particles were constructed to have a light colored north hemisphere and a dark colored south hemisphere so that dipole orientations could be discerned. (Previous experiments on thermally or externally excited systems did not sense the dipole orientations of individual particles.)

A horizontal, cylindrical container with a diameter 17.5 times the particle diameter and a height of 1.7 particle diameters was constructed with a polyvinyl chloride bottom and a rigid transparent acrylic top (for imaging). 190 particles were placed in the container and sinusoidally vibrated vertically at 30 Hz, using an electromagnetic shaker. The maximum strength of the magnetic field from the shaker at the inner container surface was 23.1 G at zero acceleration amplitude, although at typical acceleration amplitudes the rms magnetic field strength was of order 1 G. No noticeable effect due to any external magnetic field from the shaker was observed. The large masses of our particles and the high driving accelerations prevented charging of the container wall from being a factor in our experiments. The acceleration amplitude was a parameter that could be varied and was measured, using an accelerometer. The system was imaged at a rate of 2 Hz for 500 frames with a 1024$\times$1024 pixel camera. At high accelerations ($>10g$ for the long particles or $>6g$ for the short particles, where g is the acceleration of gravity), the particles dissociate from each other and the system becomes gaseous. At low accelerations ($<6g$ for the long particles or $<3g$ for the short particles), particles do not detach from each other and the positional arrangement of the system remains roughly unchanged (although the orientations of individual particles may change). The system was usually driven at accelerations high enough to allow particles to rearrange but low enough so they could form patterns. Typical accelerations for the long particles were $4-8g$, while short particles were usually driven at about $2-5g$.

III. RESULTS

Figure 2(a) shows the ''network'' pattern of 95 short particles (area number density $\rho=0.31$) after being shaken at $4.3g$ for 250 s. In this case [Fig. 2(a)], the initial state was created by pouring the particles randomly into the container. The observed chainlike structure is similar to the pattern observed by Blair *et al.* [9] in intermediate density systems of vertically vibrated magnetic spheres, and by Weis [12] in simulations of intermediate density systems of dipolar spheres. Figure 2(b) shows the configuration of 190 short particles (area number density $\rho=0.62$) after being shaken at $4.3g$ for 250 s. The initial state of the particles was random. The final state of the particles after being driven is mostly an HCP macrovortex state similar to the ground state of a dipolar hexagonal lattice predicted by Belobrov *et al.* [18] [Fig. 1(a)] and seen in simulations by Weis [12]. We find that this HCP-based macrovortex is a stable state for the short particles. When other patterns such as square-packed microvortices [Figs. 1(b) and 2(c)] or concentric rings are set up as the initial condition, after driving, the system always becomes an HCP-based macrovortex [Fig. 2(d)]; if the system is set up as a macrovortex, it remains in a macrovortex state. Also, when several single chains of the particles are brought together in an antiferromagnetic square-packed state of 190 particles (each chain has the opposite dipolar orientation from that of the two neighboring chains), they immediately, without driving, reorient themselves into the microvortex state [Figs. 1(b) and 2(c)]. If the particles are initially set up in a ferromagnetic HCP pattern, under small ($2.4g$) agitation, the macrovortex state is assumed.

Figure 3(a) is the final concentric ring state of 190 long particles after being shaken at $7.8g$. The initial state of the particles was random. The existence of the concentric ring state is independent of the initial conditions of the system. If the system is set up in any other state, it rearranges into a concentric ring state with sufficient driving. Systems of all lower densities also prefer ring and line states at intermediate and low accelerations ($<8g$).

We can track the positions of all particles during our experiments. To gauge quantitatively how much HCP clustering occured in each experiment, we designed a program that finds the number of particles with six touching neighbors in each image. The program identifies particles as ''touching'' if the centers of the particles are within 9/8 particle diameters of each other. We found that for 190 short particles initially in a random state and then shaken at $4.3g$, the mean number of particles with six touching neighbors, N_6, averaged over 500 frames, was 43.31. For 190 initially random long particles shaken at $7.8g$, $N_6=0.14$. For these two experiments, where 190 particles began in a random state, N_6 of the short particles was more than 300 times N_6 of the long particles. Using this measure, in all experiments with similar initial conditions, the short particles were quantitatively more clustered than the long particles.

Boundary conditions do not appear to affect the qualitative features of the final states for either type of particle. We performed similar experiments in a square box of height and area equal to those of the cylindrical container, and the final states were always qualitatively similar to the final state in the cylindrical container. For example, see Fig. 3(b), which shows the state of a system of 190 long particles after shaking in the square box under similar initial conditions as for Fig. 3(a).

IV. ENERGETIC CONSIDERATIONS

To gain insight into the reasons why the long particles form different patterns than the short particles, we numerically examined a model of impenetrable spherical particles of diameter d containing two equal and opposite magnetic

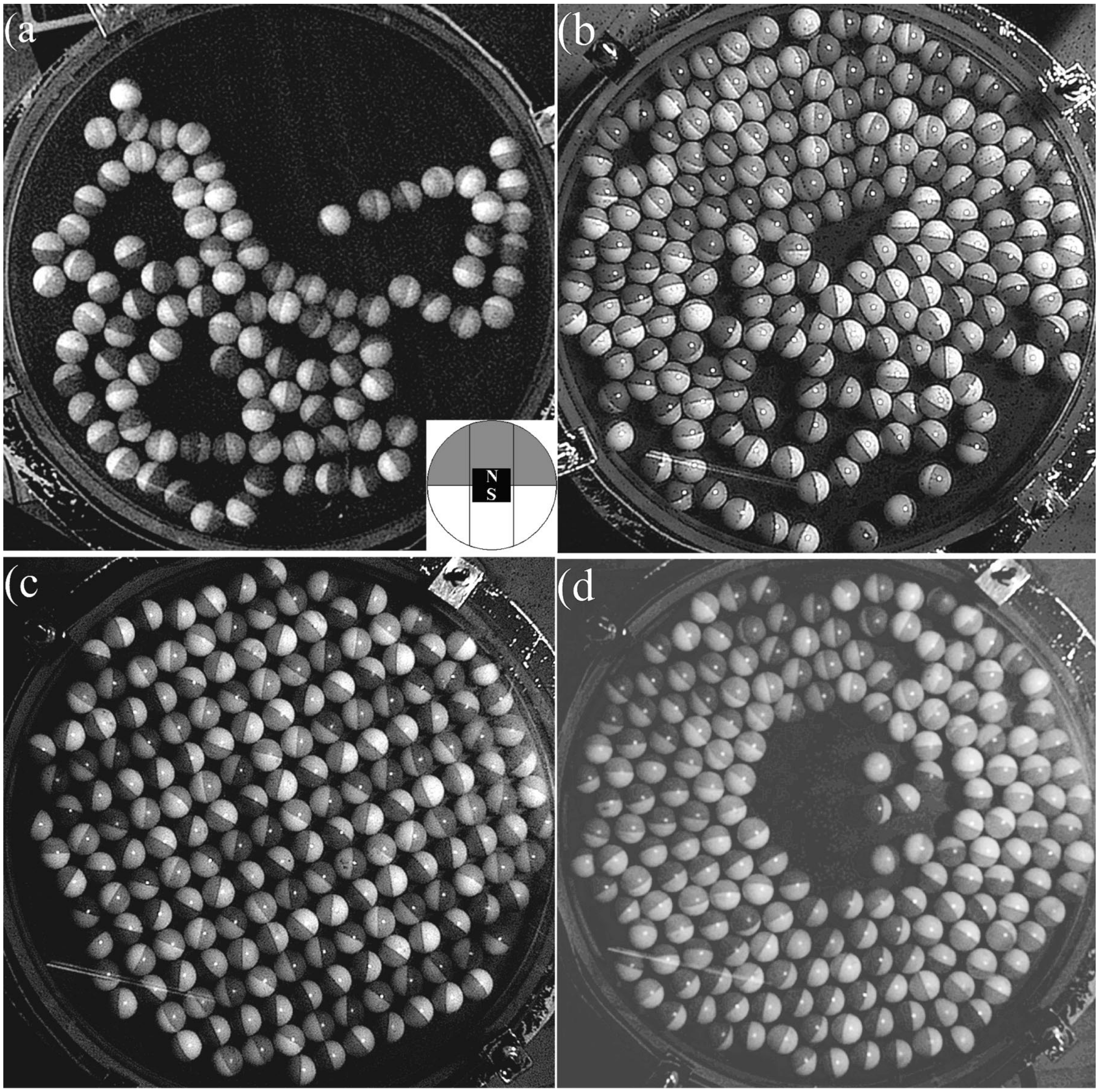

FIG. 2. (a) The network pattern of an intermediate density (95 particles) system of short particles after being shaken at $4.3g$ for 250 s. (b) The final macrovortex configuration of a dense (190 particles) system of short particles after being shaken at $4.3g$. (c) The microvortex pattern made by the square-packed setup, undriven, dense (190 particles) system of short particles. (d) A ''hollow macrovortex'' configuration of the short particles after being set up in an aligned, concentric rings pattern and then shaken at $4.3g$.

charges $\pm q$, separated by a distance s $(0 \leqslant s \leqslant d)$, with the line connecting the charges running through the center of the sphere. We vary q with s so that the dipole moment qs is constant. This model is similar to that used in simulations by Schneider *et al.* [11] to model lipid headgroups (which are also dipolar), and gives qualitatively similar results to the two-parallel-dipole model used by Camp *et al.* [19]. Our model also reduces to a central point dipole model as $s/d \rightarrow 0$.

We use the usual Coulomb energy formula

$$U_{ij} = \frac{\mu_0}{4\pi} \frac{q_i q_j}{R_{ij}} \tag{1}$$

and sum over all interparticle pairs of charges to obtain the energy of a pattern of particles (excluding the energy of pairs of charges within the same particle). When we take the limit $s/d \rightarrow 0$, our calculations are consistent with the energy calculations of rings and lines of spheres containing single point dipoles done by Clarke *et al.* [20].

In Fig. 4, we have plotted the energies of two touching particles as the angle between their axes is symmetrically bent for several values of s/d. Note that at $\Theta = 0$, the particles are in the head-to-tail configuration and at $\Theta = \pi/2$, the particles are in the antialigned side-to-side configuration. For large s/d, the head-to-tail $(\Theta = 0)$ configuration is much more favorable than the antialigned side-to-side $(\Theta = \pi/2)$ configuration (e.g., for $s/d = 0.75$, the head-to-tail energy is more than six times the antialigned side-to-side energy). For $s/d = 0.001$, the head-to-tail energy is only twice the antialigned side-to-side energy. This indicates that, as the distance between the charges increases, the relative importance

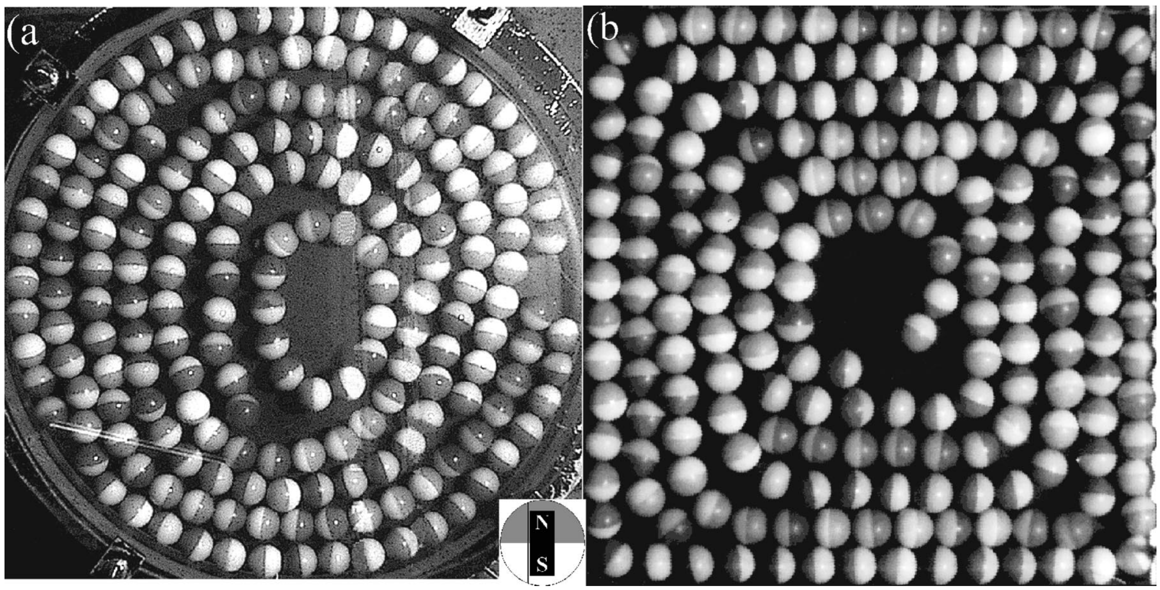

FIG. 3. (a) The final concentric ring state of a dense (190 particles) system of initially random long particles after being shaken at 7.8g for 250 s. (b) The final state of 190 long particles after being shaken in a square box at 7.8g for 250 s. The pattern is one of squared-off concentric rings, similar to the final state of the particles in the cylindrical container of equal height and area.

of the side-to-side interaction decreases. Particles with large s/d (e.g., our long particles) have a strong preference to align head-to-tail, while particles with small s/d (e.g., our short particles) do not. This is in contrast to recent models of dipolar "spherocylinders" (cylinders with hemispherical ends), which encapsulate a single centrally located point dipole [21,22]. These spherocylindrical particles increase the relative importance of side-to-side interactions. Also in Fig. 4, the slope of the curves increases as s/d increases. This indicates that chains of particles with large s/d are stiffer than those with small s/d.

Figure 5 is a plot of the energy gained by bringing together ten 10-particle chains in various patterns normalized to the energy of ten infinitely separated 10-particle chains versus s/d. At low s/d (<0.19), the lowest energy state is the HCP macrovortex (as predicted by Belobrov *et al.* [18] for $s/d=0$ particles). At $s/d>0.19$, the particles energetically prefer a ring state. This is consistent with our observations of high density systems; particles with short cylindrical magnetic cores form a macrovortex [Fig. 2(b)], while particles with long magnetic cores form rings [Fig. 3(a)]. Also, the orientations of the long particles in each ring appears to be independent from the orientations of the particles in neighboring rings. The square-packed microvortex state is more favorable than the square-packed antiferromagnetic arrangement only at very small s/d (<0.1). This is also consistent with our observations; when we attempt to set up the short particles in a square-packed antiferromagnetic pattern, they immediately, without driving, reorient into a microvortex pattern. The long particles never form a microvortex pattern when set up in a square-packed antiferromagnetic state, even when driven. The HCP ferromagnetic state is energetically unfavorable compared to separated lines at all s/d. This is in contrast to the result for an *infinite* HCP lattice, in which

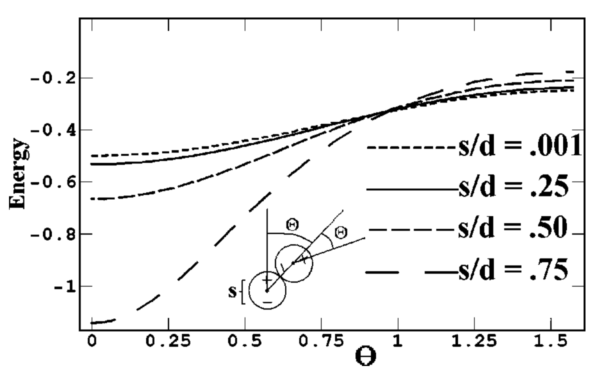

FIG. 4. The energies in units of $q^2 s^2 \mu_0 / d^3 \pi$ of two touching particles as a function of the angle Θ between their axes for several values of s/d.

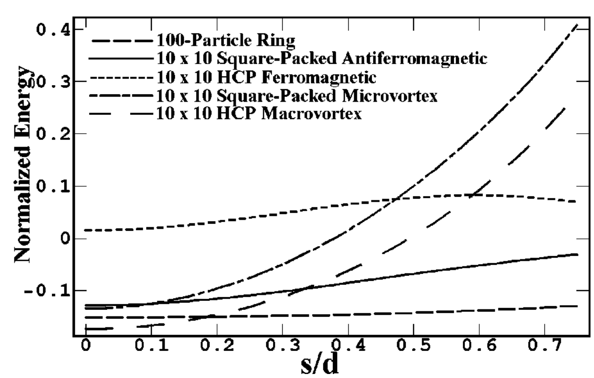

FIG. 5. The energy of ten 10-particle chains brought together in various patterns normalized to the energy of ten infinitely separated 10-particle lines vs s/d. The energy of two infinitely separated 10-particle lines is normalized to 0 for all s/d. Note that the HCP macrovortex is the lowest energy conformation only for s/d <0.19; for $s/d>0.19$, a single 100-particle ring state is of lower energy.

case the *ground state* is ferromagnetic [23]. It is also interesting to note that the ground state of an HCP lattice of magnetic dipoles can change under an external tangential magnetic field. Bologa *et al.* [8] observed a transition in an unexcited system of magnetic spheres from an HCP macrovortex state to an HCP ferromagnetic state under an increasing external tangential magnetic field.

We conclude that particles with a large s/d, such as our long particles, are more likely to form long, stiff, weakly interacting chains and rings. Particles with small s/d are more likely to aggregate into clustered patterns. Energetically, it has been shown that the ground state of a hexagonal lattice of point dipoles is a macrovortex [18]. It has also been shown that for systems of between 4 and 13 (point) dipolar particles, the global ground state is a single ring [20]. For system of 14 or more dipolar spheres, a ''double ring state'' has lower energy, and it is expected that the lowest energy configuration changes once again to triple, quadruple, and higher-order ring states as the number of particles increases. We conjecture that, as the number of particles increases, the global ground state may begin to look like a ''hollow macrovortex,'' similar to that in Fig. 2(d).

V. CONCLUSION

Our experimental and theoretical results show that the shape of the magnetic core in particles can be a determining factor in pattern formation, even in simple systems. As s/d increases, the energetically favored pattern changes qualitatively. Indeed, as $s/d \to 1$ in our model, the head-to-tail arrangement of the dipoles becomes infinitely more favorable than any other configuration. Our unique particles allow us to provide the first direct experimental evidence of the square-packed microvortex pattern in a zero-temperature system of dipolar spheres, and the first observations of an HCP macrovortex state in an excited system of dipolar spheres. Possible future work includes studies of larger systems of particles with which orientations can be discerned, and obtaining color images to facilitate the extraction of orientations by computer. Finally, we note that the observed patterns can be understood as either minimum magnetostatic energy equilibria (in the macrovortex and microvortex cases) or [in the case of low-density network patterns, Fig. 2(a)] as thermal equilibrium states.

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