Control of lattice spacing in a triangular lattice of feeble magnetic particles formed by induced magnetic dipole interactions

Noriyuki Hirota\textsuperscript{1}, Tsutomu Ando\textsuperscript{2}, Ryo Tanaka\textsuperscript{2}, Hitoshi Wada\textsuperscript{2} and Yoshio Sakka\textsuperscript{1}

\textsuperscript{1}Nano Ceramics Center, National Institute for Materials Science, Tsukuba, Japan
\textsuperscript{2}Department of Advanced Materials Science, The University of Tokyo, Kashiwa, Japan

E-mail: hirota.noriyuki@nims.go.jp

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Abstract

We studied methods of controlling the spacing between particles in the triangular lattice formed by feeble magnetic particles through induced magnetic dipole interaction. Formation of a triangular lattice is described by the balance between the magnetic force and the interaction of induced magnetic dipoles. The intensity of the magnetic force is proportional to the volume of particles $V$ and the difference in the magnetic susceptibilities between the particles and the surrounding medium $\Delta \chi$. On the other hand, the intensity of the induced magnetic dipole interaction depends on the square of $V$ and $\Delta \chi$. Therefore, altering the magnetic susceptibility difference by changing the susceptibility of the surrounding medium, volume of the particles, and intensity and spatial distribution of the applied magnetic field effectively controls the distance between the particles. In this study, these three methods were evaluated through experiment and molecular dynamics simulations. The distance between the particles, i.e. the lattice constant of the triangular lattice, was varied from 1.7 to 4.0 in units of the particle diameter. Formation of self-organized triangular lattice through the induced magnetic dipole interaction is based on magnetism, a physical property that all materials have. Therefore, this phenomenon is applicable to any materials of any size. Consequently, structure formation through induced magnetic dipole interaction is a potential way of fabricating materials with ordered structures.

Keywords: feeble magnetic particles, triangular lattice, magnetic dipole interaction

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Recently, cryocooler-operated superconducting magnets have become widely available which can generate magnetic fields above 10 T. This allowed the use of high magnetic fields in large-scale room-temperature spaces. In response to this trend, it has become popular to study the effects of magnetic field on feeble magnetic materials, i.e. dia- or paramagnetic materials. These advances have gradually provided a better understanding of the effects of high magnetic fields on feeble magnetic materials [1–3]. One of the interesting actions of magnetic fields is mechanical effect, which acts without any direct contact and reaches deep inside the material. Through this effect, magnetic fields are expected to control materials processing.

Materials processing by magnetic fields has been eagerly explored, in particular orientation of composite materials, which relies on their magnetic anisotropy [4–9], or the flow control of materials during the crystal growth [10]. In electrochemistry, magnetic field effects are sometimes observed through the materials flow in fluids [11,12]. Those effects are mainly based on magnetic forces, which can be
expressed as interactions between feeble magnetic substances and the magnetic field.

On the other hand, interactions among feeble magnetic materials through magnetic dipoles, induced by the applied magnetic field, have not been investigated much, perhaps due to the small magnetic susceptibilities of feeble magnetic materials. Formation of ordered structures has been reported arising from the interaction among feeble magnetic materials in high magnetic fields [13–15]. In such cases, a quasi-two-dimensional system is prepared, consisting of feeble magnetic particles, made of glass, copper, or gold and dispersed in a certain solution. It is then introduced into the magnetic field generated by a superconducting magnet. When the magnetic field is applied parallel to the plane of a two-dimensional sample, a chainlike structure is formed owing to the attractive interactions among particles that arise from the magnetic dipoles. On the other hand, when the magnetic field is applied perpendicularly to the sample plane, a triangular lattice structure is formed (see figure 1). Figure 1 shows an example of a self-organized triangular lattice of 1 mm gold particles, which are dispersed in a 40 wt% MnCl2 aqueous solution.

Formation of such a triangular lattice is described by the balance between the magnetic force and the induced magnetic dipole interaction. The magnetic force is exerted on the particles owing to the distribution of the applied magnetic field in the sample plane and is expressed by equation (1). In this system, the magnetic force acts as a central force in the sample plane. The induced magnetic dipole interaction is expressed by equation (2) and acts as a repulsive force in the sample plane.

\[
F = \frac{\Delta \chi}{\mu_0} VB \cdot \nabla B, \tag{1}
\]

\[
F = \frac{3\mu_0 m_1 m_2}{4\pi r^3}, \quad m_i = \frac{\Delta \chi}{\mu_0} V B. \tag{2}
\]

Here, \(B\) is the magnetic flux density, \(\Delta \chi\) is the difference in the bulk magnetic susceptibilities between feeble magnetic particles and the surrounding medium, \(\mu_0\) is the permeability of vacuum, \(V\) is the particle volume, \(r\) is the distance between the particles and \(m_1\) and \(m_2\) are their magnetic moments.

Atoms or molecules of the same kind spontaneously form crystals. As a well-known example of self-organization on a larger scale, micrometer-sized particles form colloidal crystals thorough electrostatic interaction. In this example, however, particles must have surface charges and be dispersed in a certain fluid. Moreover, their size and density should be relatively small. Therefore, this type of self-organization is applicable only to certain materials. In the case of self-organization through induced magnetic dipole interaction in magnetic field, particles must be large enough to overcome the thermal energy because the involved magnetic forces depend on the particle volume. In case of feeble magnetic materials, which have very small magnetic susceptibilities, the particle size should be larger than a few micrometers. As seen in figure 1, this method can form regular structures from the particles as large as 1 mm. This method is based on magnetism, a physical property that all materials have,

and therefore is applicable to any material of any size. Consequently, structuring through induced magnetic dipole interaction is expected to be a potential way of fabricating ordered materials.

In figure 1, the spherical particles are not closely packed and formed triangular lattice with some spacing. This spacing between particles seems to be affected by the balance between the magnetic force and the induced magnetic dipole interaction. Therefore, control is possible over the lattice constant of a produced triangular lattice, which is very difficult to achieve by other methods.

In this paper, we evaluated the methods of controlling the distance between particles in the triangular lattice formed through induced magnetic dipole interaction.

2. Experimental procedure and simulation

The magnetic force exerted on particles by applied magnetic field is proportional to the volume \(V\) and the difference in the magnetic susceptibilities between the particles and surrounding medium \(\Delta \chi\), as expressed by equation (1). On the other hand, the intensity of the induced magnetic dipole interaction depends on the square of \(V\) and \(\Delta \chi\) (equation (2)). Therefore, the following three mechanisms should control the distance between particles: (a) altering the difference in magnetic susceptibility by changing the susceptibility of the surrounding medium, (b) changing the volume (diameter) of the particle, and (c) controlling the intensity and spatial distribution of the applied magnetic field. In this study, we evaluated these three methods through experiments and molecular dynamics simulations.

In the experiments, the magnetic field was provided by a superconducting magnet of conduction cooling type, model JMTD-10C13E-NC, manufactured by JASTEC, Ltd. This magnet has a room-temperature bore of 100 mm in diameter. It can be rotated as a whole and fixed at any angle. In our experiments, we used this magnet in the vertical position.
Along the bore axis (the $z$-axis in figure 2(a)), the magnetic field is maximum ($13 \, T$) at the center of the coil ($z = 0$), and gradually decreases toward edges of the bore (figure 2(b)). Figure 2(c) shows the magnetic field distributions in a certain $z$-plane (the magnetic field distribution in the $r$ direction). In the range $z = 149.3$–$182.0 \, mm$ used in the experiment, the magnetic field is minimum at $r = 0$, the center of the plane, and gradually increases toward the bore edge. The variation of the magnetic field in a certain $z$-plane strongly depends on $z$: it is maximum at $z = 0$, and is much smaller at $z = 182 \, mm$.

Six kinds of diamagnetic particles were used in the experiment, as summarized in table 1. Each particle was observed in an optical microscope, and the particles were selected having diameter within 2\% of a certain value. Seven particles were used in each experiment. To enhance the interaction between magnetic dipoles with the magneto-Archimedes effect [16], MnCl$_2$ aqueous solution was used as a surrounding medium. The concentration of the solution was 40 wt\% in experiments (b) and (c). To change the magnetic susceptibility of the surrounding medium in experiment (a), the concentration was changed from 10 to 40 wt\%. The densities and volume magnetic susceptibilities of each solution are listed in table 2.

The diamagnetic particles and the solution were encapsulated in a vessel to avoid the disturbance from the Moses effect and the surface tension. The vessel was introduced into the magnet bore and fixed at the prescribed place. Then, the arrangement of each particle was observed using a CCD camera. The obtained images of the particle arrangement were computer processed, the coordinate of the center of each particle, and thus the distance between the particles, was determined. In the triangular lattice formed by seven particles, there are 12 different combinations of particle pairs. This means that we can obtain 12 data points of the particle distance from one set of experiments. For each experimental condition, we chose two different sets of seven particles and carried out independent experiments using
Table 2. Properties of MnCl₂ aqueous solutions.

| Concentration (wt%) | Density (g/cm³) | Magnetic susceptibility (×10⁻⁵) |
|---------------------|----------------|-----------------------------|
| 40.01               | 1.428          | +81.04                      |
| 34.99               | 1.360          | +67.41                      |
| 29.99               | 1.297          | +54.92                      |
| 25.00               | 1.238          | +43.48                      |
| 20.01               | 1.182          | +33.01                      |
| 15.00               | 1.130          | +23.42                      |
| 10.00               | 1.082          | +14.62                      |

Table 3. Conditions of the magnetic field and magnetic force.

| Applied magnetic field at z = 0, r = 0 (T) | Position of the sample at z = 0 (mm) | Magnetic field intensity at z = 0, r = 0 (T) | Magnetic force acting on the particle at z = 0, r = 3 × 10⁻¹¹ (N) |
|------------------------------------------|--------------------------------------|------------------------------------------|--------------------------------------------------|
| 13.0                                    | 182.0                                | 6.73                                     | 1.25                                             |
| 11.5                                    | 181.5                                | 5.98                                     | 1.25                                             |
| 10.0                                    | 180.5                                | 5.25                                     | 1.25                                             |
| 8.50                                    | 179.5                                | 4.51                                     | 1.25                                             |
| 7.00                                    | 177.5                                | 3.79                                     | 1.25                                             |
| 5.50                                    | 174.3                                | 3.07                                     | 1.25                                             |
| 4.00                                    | 167.7                                | 2.37                                     | 1.25                                             |
| 2.50                                    | 149.3                                | 1.72                                     | 1.25                                             |

these two sets of particles. Therefore, the experimental data described below were averaged over 24 data points.

In the experiment based on concept (a), the concentration of the MnCl₂ aqueous solution was changed from 10 to 40 wt% in 5 wt% increments. Other conditions were fixed as follows: glass particle size 0.6 mm, magnetic field 13 T at the center of the magnet (z = 0, r = 0), and the sample position z = 182 mm.

For the experiment based on concept (b), all the particles listed in table 1 were used. To obtain several monodispersed particles with diameters ranging from 0.15 to 1.0 mm, we used particles of different materials: gold (1.0 mm, Kojyundo Chemical Co.), glass (0.8 and 0.4 mm, AZ ONE Co., and 0.6 mm, Iuchi Co.), and bronze (0.5 and 0.15 mm, Mitsui Mining and Smelting Co., Ltd.). The magnetic field was 13 T at the center of the magnet (z = 0, r = 0), the sample position was z = 182 mm, and the concentration of the MnCl₂ solution was 40 wt%.

In the experiment based on concept (c), 0.6 mm glass particles and a 40 wt% MnCl₂ solution were used. The magnetic field at t to the sample can be controlled by changing the electric current applied to the superconducting magnet. This however alters not only the magnetic field, but also the magnetic field distribution in the r direction and therefore the magnetic force exerted on particles, in accordance with the change in the induced magnetic dipole interaction. To change the applied magnetic field while fixing the magnetic force acting on the particles in the r direction, the sample position in the z-direction should be modified too. From the preliminary experimental result, it was determined that all seven 0.6 mm glass particles fall within a circle of a 3 mm radius when they form a triangular lattice. Therefore, we chose sample position z to keep constant the magnetic force acting on the glass particle at r = 3. The applied magnetic field at z = 0, the position of the sample z, the magnetic field intensity at z, r = 0, and the magnetic force acting on the particle at z, r = 3, are listed in table 3.

Numerical simulations were based on the molecular dynamics method as described in [17,18]. Only the magnetic force and the induced magnetic dipole interaction were considered. We introduce the following assumptions: (a) the inertial force of an aqueous solution is ignored because the Reynolds number of the particle is very small; (b) the change in the magnetic field in particles is ignored, and its value is regarded as the magnetic field at the particle center because the particles are very small; (c) particles are sufficiently diluted in the medium, and, as a result, the hydrodynamic interactions among particles are negligible.

The initial positions of particles were determined randomly within the circle with a radius 20 times the particle diameter. Considering the magnetic field distribution in the sample plane, the equation of motion for particle i was set up by considering their interaction with all the other particles. Then, the motion of particles was determined for the next time unit taken as 10⁻⁴ s. This procedure was repeated until the position of the particles reached the steady state. In the simulation, parameters such as the size and magnetic susceptibility of a particle, applied magnetic field and its spatial distribution, and the volume magnetic susceptibility of the surrounding medium were taken the same as in the experiment.

3. Results and discussion

Figure 3 shows images of triangular lattice structures observed in the experiments based on concept (a). With increasing concentration of the surrounding MnCl₂ solution the distance between particles gradually increased. These results are plotted in figure 4. The vertical axis of the figure represents the distance between the particle centers normalized to the particle diameter, and the horizontal axis is the difference in the volume magnetic susceptibilities between the particle and solution. The averaged values of the experimental results are indicated by the squares, and the error bars represent the standard deviation. By changing the concentration of the solution, the difference in the volume magnetic susceptibilities between the glass particles and the solution varied from 1.62 × 10⁻⁴ to 8.26 × 10⁻⁴ in SI units. In response to this, the distance between particle centers changed from 1.74 to 2.38 of the particle diameter. The circles in the figure represent the distance between particle centers obtained from simulations assuming the same conditions as in the experiments. There are slight differences in the absolute values; however, the degree and tendency of the variation are essentially consistent.

Figure 5 shows the results of the experiment and simulation when the particle diameter was changed (concept (b)). As described above, to change the particle diameter
Figure 3. Triangular lattice structures of 0.6 mm glass particles observed in the experiment based on concept (a). The concentration of the MnCl$_2$ aqueous solution was (a) 10 wt%, (b) 15 wt%, (c) 20 wt%, (d) 25 wt%, (e) 30 wt%, (f) 35 wt% and (g) 40 wt%.

Figure 4. Results of the experiments and simulation when the concentration of the surrounding solution was changed (concept (a)).

Figure 5. Results of the experiments and simulation when the particle diameter was changed (concept (b)).

Figure 6. Results of the experiments and simulation when the applied magnetic field on the sample was changed while keeping constant the magnetic force acting on the glass particle at $r = 3$ (concept (c)).

From 0.15 to 1.0 mm, particles made from different materials were used. Owing to this difference in materials, not only the particle diameter but also the difference in the susceptibilities between the particle and the solution was changed. Therefore, in figure 5 the change in the normalized distance between particle centers is plotted against the product of the volume of the particle and the absolute value of the difference in the volume magnetic susceptibilities between the particle and the solution, $|\Delta \chi| V$. The squares represent the experimental results, the error bars show the standard deviation, and the circles correspond to the simulation. Under the used experimental condition, the $|\Delta \chi| V$ value varied from $1.45 \times 10^{-6}$ to $4.42 \times 10^{-4}$ [m$^3$]. Corresponding to this change in $|\Delta \chi| V$, it was confirmed that the normalized distance between particle centers changed from 2.02 to 4.04 in the experiment. There are slight differences in the absolute values; however, the degree and tendency of the variation are essentially consistent.

Figure 6 shows the results of the experiments and the simulation when the applied magnetic field on the sample was changed while keeping constant the magnetic force acting on the glass particle at $r = 3$ (concept (c)). The horizontal axis represents the magnetic field intensity at the sample position ($z_s, r = 0$). The vertical axis of the figure represents the distance between particle centers normalized by the particle diameter as in the two previous figures. The squares represent the experimental results, the error bars show the standard deviation, and the circles correspond to the simulation. It was confirmed from these experiments that the normalized distance between particle centers varied from 1.74 to 2.38 by changing the intensity of the applied magnetic field from 1.721 to 6.729 T.
Here again, the results are slightly larger in simulation than in the experiment; however, the degree of the variation is consistent. In the experiments, the friction force acted on the particles owing to the existence of the vessel wall. This, however, was not considered in the simulation. If friction force affects the formation of triangular lattice then the distance between particles should depend on the direction of the particle movement. However, the experimental results were always the same irrespective of the initial particle position, such as the center or the wall side of the sample vessel and the direction of the movement. Therefore, the friction force does not seem to be the cause of the difference between the experiments and the simulation. On the other hand, it was assumed in the simulation that the magnetic moment of the particle exists in the center of the particle. Because an actual particle has finite volume, the spatial distribution of the magnetic moment in the particle should be considered. Therefore, this assumption may be a potential cause of the difference between the experiments and the simulation.

We are investigating this reason and will report the results elsewhere.

It was confirmed in our experiments and supported by the simulation that the distance between particles, i.e. the lattice constant of the triangular lattice, can be altered from 1.7 to 4.0 times the particle diameter by controlling the difference in the magnetic susceptibilities between the particle and surrounding medium, size of the particles, and applied magnetic field.

4. Conclusions

In this study, methods were investigated of controlling the lattice constant of the triangular lattice formed by feeble magnetic particles through induced magnetic dipole interaction. The lattice constant can be controlled by configuring the conditions of feeble magnetic particles, such as the combination of particles and the surrounding medium, size of the particles and applied magnetic field.

The structuring of feeble magnetic particles through induced magnetic dipole interaction is based on magnetism, a property all materials have. Therefore, this phenomenon applies to all materials. On the basis of the knowledge obtained here, the lattice constant of the triangular lattice can be continuously and arbitrarily controlled. This is an advantage of using magnetic fields that cannot be observed for other methods, such as the formation of colloidal crystal through electrostatic interaction. If self-organized lattice formation using magnetic fields becomes possible, even for feeble magnetic particles of micrometer size, then photonic crystals can be created that can respond to light of different wavelengths, for example. The observed here phenomenon suggests new applications of magnetic fields to various areas, such as materials processing.

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