Arranging Diamagnetic Particles in a Modulated Magnetic Field Originating in Microelectromechanical Systems Compatible with an Integrated Circuit upon Halbach Array Magnet

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ABSTRACT: In this study, we attempted to expand the applicability of the mechanism for arranging diamagnetic particles in a modulated magnetic field. A Halbach array magnet was prototyped as a portable device for generating a high magnetic field. Despite the magnet being palm-size with dimensions of $50 \times 50 \times 20$ mm, the magnetic field is $1.31$ T at $1$ mm from the surface. Additionally, an Si substrate on which an Fe thin film is formed and patterned to be compatible with the integrated circuit (IC)—utilizing the microelectromechanical systems process technology—is prototyped as a tool to generate a modulated magnetic field. Regarding the deposition condition of the Fe thin film, holes with diameters of $30 \mu m$ are arranged in an array at intervals of $60 \mu m$, and the thickness is approximately $0.5 \mu m$. Finally, a particle magnetic-adsorption experiment was conducted using the prototypes. The diamagnetic particles ($\text{diameter: } 25 \mu m$) dispersed in the paramagnetic surrounding medium were observed to be arranged in the hole portions. This result indicates that the microparticles are absorbed in their arbitrary positions by the modulated magnetic field. In the end, we succeeded in achieving the portability and implementation on IC for the particle arrangement magnetic mechanism.

1. INTRODUCTION

Particle assembly is the regular accumulation of particles to create materials and devices. It is considered to be a promising seed technology owing to its numerous applications, including biosensors,1,2 catalysts,3 magnetic recording media,4 and optical interconnections.5 It is expected that many materials and devices will be developed based on this seed technology, leading to the creation of new functions and technologies. For these purposes, the capability to arrange precisely the particles in arbitrary positions is important. Furthermore, the ability to operate in a noncontact and nondestructive manner is necessary for viable applications to biology and medicine—for which further development of the technique is desired.

One technique proposed for arranging the particles involves using optical tweezers, a technology for trapping and manipulating microscopic objects utilizing the radiation force of a beam.6 A precise arrangement could be produced because this method handles particles one-by-one. Another method of arranging a plurality of particles was developed;7 however, it was not suitable for industrial applications because the throughput was too low, and many particles could not be arranged. Moreover, a dielectrophoresis method (DEP)8 has been proposed along with another method utilizing a modulated magnetic field9 as techniques for arranging many particles without contact; however, a precise particle arrangement could not be produced by either. The DEP—which involves manipulation of particles using the interaction of the polarization effect in a nonuniform electric field—has been advanced to the application of immunoassay10 as it is consistent with integrated circuit (IC) technology; although, to get to that stage, it was necessary to fabricate an electrode wiring pattern by lithography technology.

On the other hand, the applications of the magnetic method have not progressed significantly in terms of convenience; however, attempts are being made to assemble diamagnetic particles with high accuracy using a solution of paramagnetic holmium salt11 and the patterning of cells.12 In the past, an electromagnetic device was able to generate a magnetic field of $1 \text{ T as a high magnetic field environment;}$ however, this device could not be easily carried because it was large and heavy. Additionally, in the case of commercially available permanent magnets, the magnetic field was too weak to control the diamagnetic particles easily, unless the magnetic susceptibility difference between a substance and the surrounding medium was increased.11 Another inconvenience was that a tool in which two types of thin films having different magnetic susceptibilities were alternately stacked was used for generating a modulated magnetic field in a magnetic field; therefore, it was not practical to implement the tool on another device post-process. There were also limitations with that tool in adjusting the design and size of the modulated magnetic field.
To overcome these issues, in this study, a Halbach array magnet (HAM) was prototyped as a portable device for generating a high magnetic field. Likewise, a substrate on which an Fe thin film was formed and patterned—utilizing the microelectromechanical systems (MEMS) process technology compatible with an IC—was prototyped as a tool to generate a modulated magnetic field, assumed to be implemented on the IC. Furthermore, whether the diamagnetic particles could be arranged magnetically was verified utilizing these prototypes.

The purpose of this study is to expand the applicability of the mechanism for arranging diamagnetic particles in a modulated magnetic field while achieving portability and implementation on ICs.

2. MATERIALS AND METHODS

2.1. Creation of the Portable High Magnetic Field Environment. It is necessary to reduce the size of the equipment that generates a magnetic field identical to the magnetic field generated by the electromagnet to improve the convenience of the magnetic method. A HAM could offer a solution as a portable device that generates a high magnetic field. The Halbach array is a structure considered to have the best magnetic efficiency in a magnetic circuit composed of only permanent magnets; further, this array could maximize the magnetic field in a specific direction. In recent years, the utilization of the HAM has been steadily increasing in applications, such as maglev trains, electric motors, NMR, and energy harvesters. Furthermore, the HAM is also beginning to be utilized in the manipulation of diamagnetic particles.

Figure 1 shows the schematic of the three-dimensional HAM design used in this study. The dimensions of the HAM were 50 × 50 × 19 mm. The HAM was composed of rectangular parallelepiped subpermanent magnets arranged in a rectangular parallelepiped formation. The dimensions of the central magnet were 7.5 × 7.5 × 19 mm, and the dimensions of the perimeter magnets were 21.25 × 21.25 × 19 and 21.25 × 7.5 × 19 mm. The magnets were composed of neodymium (N-48H). The magnetization direction (indicated by the arrow in Figure 1) of the central magnet was the vertical upward direction. The magnetization directions of the perimeter magnets were the same as those in the central magnet. Hence, in the central magnet, the magnetic fluxes of the perimeter magnets were gathered, and the density of the emitted magnetic flux was amplified. The ferritic stainless steel SUS430 was employed as a fixing plate with dimensions of 50 × 50 × 1 mm. Figure 2 shows the assembly method. Then, a prototype of a single block neodymium (N-48H) magnet (50 × 50 × 19 mm) was employed and the ferritic stainless steel SUS430 (50 × 50 × 1 mm) was bonded to the bottom of the magnet—similar to the prototyped HAM. The magnets were obtained from a specialized manufacturer of magnetic-applied products (Shimonishi Seisakusho, Co., Ltd., Japan). Furthermore, the magnetic field distribution over the surface of the prototyped HAM and the neodymium magnet were measured using a special specification high precision magnetic analysis system apparatus (Magnet Force, Co., Ltd., MMA-3D, Japan) along with incorporating a gauss meter owned by the manufacture shown in Figure 3. The magnetic field distribution over the surface of the prototyped HAM was calculated using an electromagnetic simulation software (ELF, ELF/MAGIC, Japan).

Figure 1. Schematic of the HAM-devised design. (A) Top surface view. (B) Cross-sectional view of the broken line b in panel (A).

Figure 2. Assembly method of HAM.

Figure 3. High precision magnetic analysis system apparatus with special specifications of the magnetic field distribution. Photograph courtesy of Shimonishi Seisakusho, Co., Ltd. Copyright 2020.

2.2. Development of Tool for Modulating the Magnetic Field Implemented on an IC. It was necessary that the implementation of the tool modulating the magnetic field be easy; moreover, the ability to control the design and size of the modulated magnetic field was required to improve the convenience of the magnetic method. MEMS process technology could offer a solution as a novel fabrication method for the tool. The technology refers to microelectromechanical devices and their creation technology; it is essential for diverse products in a wide range of fields such as automotive airbag sensors, inkjet printer heads, accelerometers in cell phones, and blood pressure sensors. In this case, the patterned Fe thin-film substrate as the tool was fabricated by the MEMS process.
technology, which is compatible with the IC. The dimensions of the Fe thin film were 5 mm × 5 mm × 0.5 μm. The design was arranged in holes of 64 rows × 64 columns. The distance between the holes is 60 μm. The diameter categories of the hole were selected to be 10, 30, and 50 μm. The schematic cross-sectional view of the substrate is shown in Figure 4. In the substrate, a polyimide film was used as the protective film to isolate the Fe thin film from the solution.

![Figure 4. Schematic cross-sectional view of the Fe and polyimide films on the Si. Reprinted with permission from ref 22. Copyright 2017 Institute of Electrical and Electronics Engineers.](image)

The procedure for forming and patterning the Fe thin film and forming the polyimide film on the Si substrate is as follows:

1) Create a photo mask of the Fe pattern design.
2) Deposit SiO₂ (thickness: 1 μm) on the Si substrate (thickness: 0.5 mm).
3) Sputter Fe (thickness: 0.5 μm) with Ti (thickness: 0.05 μm) as the base.
4) Perform photolithography using the prepared photo mask.
5) Etch (ion milling) the Fe/Ti film.
6) Remove the resist.
7) Coat with polyimide (thickness: 3 μm).

After the procedure, the substrate surface was observed with an optical microscope (Olympus, TH4-100 with FX380, Japan) to confirm the film formation and the patterning state of the prototyped substrate. Furthermore, the cross-sectional view of the substrate was observed with a field emission scanning electron microscope (JEOL, JSM-7100F, Japan) after the cross section of the substrate was prepared by a cross-section polisher (JEOL, IB-19520CCP, Japan).

**2.3. Particle Magnetic-Adsorption Experiment.** An experiment was conducted to investigate the influence of the magnetic field distribution—created by the combination of the prototype HAM and the prototype Fe thin-film pattern substrate—on the behavior of particles. The force acting on a substance when under a magnetic field is expressed by eq 1 with the following variables: $\chi_1$ and $\chi_2$ are the magnetic susceptibilities of the microparticle and the surrounding medium, respectively; $V$ is the volume of the substance; $B$ is the magnetic field; $dB/\text{dy}$ is the magnetic field gradient; and $\mu_0$ is the vacuum permeability. When the magnetic susceptibility of the microparticles is lower than that of the surrounding medium in which the microparticles are dispersed, the magnetic force acts on the microparticles in the direction of the low magnetic field, and then, the microparticles move in that direction.

$$ F = \frac{(\chi_1 - \chi_2)V}{\mu_0} B \frac{dB}{dy} \quad (1) $$

As an experimental environment, the substrate was terminated on a plastic plate (thickness: 1 mm) and placed on the center top surface of the prototype portable HAM. The experiment was performed in a manner in which the surrounding medium of hexacyanoferrate(III) (concentration: 40 mmol/L; density: 1.013 g/cm³), where polystyrene microparticles (diameter: 25 μm; density: 1.05−1.19 g/cm³) were dispersed at 1 wt %, was poured on the substrate. The magnetic susceptibility of polystyrene is smaller than that of hexacyanoferrate(III) because polystyrene is a diamagnetic material ($\chi_1 < 0$; $\chi = -8.21 \times 10^{-6}$) and hexacyanoferrate(III) is a paramagnetic material ($\chi_2 > 0$; $\chi_{\text{mol}} = \pm 2290 \times 10^{-6}$ emu/mol). The perimeter of the Fe thin-film area was framed so that the solution did not spread. As a control, we performed the same experiment without a magnet. Then, the behavior of the microparticles was observed using the optical microscope, identical to the step mentioned earlier.

The magnetic field distribution in the space at certain distances (3, 18, and 33 μm) from the Fe patterned thin film—under the same conditions as the particle adsorption experiment—was calculated using the same simulation software mentioned earlier.

**3. RESULTS AND DISCUSSION**

**3.1. Evaluation of Magnetic Efficiency for the Prototyped HAM.** The prototyped HAM is shown in Figure 5. The dimensions were 50 × 50 × 20 mm, and the weight was 376 g. Figure 6 shows the result of the measurement of the magnetic field distribution in a horizontal line over the central region of the prototyped HAM at the five levels of distances (from 1 to 5 mm per 1 mm) from the magnet surface (line A in Figure 5). As the distance from the magnet decreased, the magnetic field increased. Although falling was observed in the magnetic field at the edge of the center magnet, the magnetic field distribution was confirmed to be virtually homogeneous.
Figure 7 shows the magnetic field and the magnetic force field over the center of the prototyped HAM and the neodymium magnet at the same distance in Figure 6. It also shows the simulated result of the magnetic field distribution over the center of the prototyped HAM. The magnetic field gradient was calculated from the magnetic field. In the case of the neodymium magnet, the magnetic field was within the range of 0.32–0.38 T and only marginally attenuated. Thus, the magnetic force field was extremely weak. On the other hand, in the case of HAM, the magnetic field was within the range of 0.75–1.31 T and attenuated, as shown in Figure 6. Moreover, it was significant that the magnetic field gradient was large, and the magnetic force field was within the range of 89–201 T²/m. Therefore, it was noted that the prototype HAM has a high magnetic field and a high magnetic force field, as compared to the neodymium magnet. Likewise, it was confirmed that the prototype HAM could reproduce the magnetic field (1 T) obtained by the electromagnet under a 3 mm distance from the surface. Furthermore, the measured result was slightly lower than the simulated result of the prototype HAM; however, this difference was negligible. Therefore, a desirable magnet was obtained.

3.2. Evaluation of the Film Condition as to the Prototyped Substrate. The deposition condition on the Si substrate was evaluated to confirm whether the Fe film was formed and etched in accordance with the pattern design. The results from observing the surface of the prototype substrate with an optical microscope are shown in Figure 8. It was observed under the condition in which 10, 30, and 50 μm holes were arranged in an array at intervals of 60 μm on each substrate. The results confirmed that the design in the Fe thin film was successfully made.

Figure 9 shows a field emission scanning electron microscopy (FE-SEM) image of the cross-sectional view of the prototype substrate with a 30 μm hole diameter. The thickness of the Fe thin film was approximately 0.5 μm, as intended. It was also discovered that the Fe film was completely covered with the polyimide film. From these results, it was confirmed that the film formation and patterning of the Fe thin film was achieved as desired.

3.3. Analysis of Microparticle Behavior under the Created Magnetic Field. The state of the microparticles dispersed in the solution under the magnetic field—created by the combination of the prototype portable HAM and the prototype Fe thin-film pattern substrate—was observed and analyzed. Optical microscope images of the state over the substrate surface with the solution and dispersed microparticles are shown in Figure 10. In the case without a magnet and a hole with a 10 μm diameter, the microparticles were randomly distributed and aggregated. On the other hand, in the case with
30 and 50 μm diameters, some microparticles were arranged in the hole portions.

The simulation results are shown in Figure 11. The magnetic field increased at the upper part of the Fe portion due to the attraction of magnetic fluxes to the Fe portion and decreased at the upper part of the hole portion. In the vicinity of the Fe thin film, the space having the modulated magnetic field was formed like in Figure 11A. In addition, as the distance from the Fe thin film increased, the magnetic field became more uniform. Furthermore, at the boundary between the hole portion and the Fe thin film, the maximum value and the minimum value of the magnetic field were adjacent.

In the case with 30 (Figure 11C) and 50 μm (Figure 11D) hole diameters in Figure 11, the space having the modulated magnetic field formed even at a distance of 18 μm from the Fe film. These results indicate that a modulated magnetic field was generated on the surface of the Fe thin film in the magnetic field; the microparticles were absorbed into the hole portion by the modulated magnetic field and by the difference in the magnetic susceptibility between the microparticles and the surrounding medium. Incidentally, because the prototype Halbach array magnet has a large magnetic force, it is possible that the magneto-Archimedes effect may have influenced particle behavior. Magnetic levitation for diamagnetic subjects due to the magneto-Archimedes effect has been previously reported.26−28 It was assumed that the apparent weight of the particles was reduced, and the influence of the modulated magnetic field was relatively increased. Therefore, it is believed that they contributed to the magnetic absorption of the microparticles in their arbitrary positions. In addition, the number of microparticles with the center of the particles inside the hole was investigated for each hole of diameters (30 and 50 μm). Figure 12 shows the number of holes for each number of microparticles located in the holes. It can be seen that a significantly higher proportion (more than double) of one microparticle is located at each hole portion in the hole size of 30 μm compared to 50 μm. That is, the hole size of 30 μm rather than 50 μm was deemed appropriate for arranging one microparticle per hole portion. The reason was that a plurality of microparticles were arranged within a hole because the portion of the smallest magnetic field was near the hole edge when the hole diameter was much larger than the microparticle...
magnetic field by increasing the thickness of the Fe thin film. The second is increasing the external magnetic field by changing the design of the HAM. The third was increasing the magnetic susceptibility difference between a substance and the surrounding medium.11 In addition, utilizing magnetophoresis,2,23 which is the controlled manipulation of microparticles in space and time through the insertion of the flow path mechanism, it is possible to reduce the size variation of microparticles, disperse the microparticles with low concentrations in the surrounding medium, and remove excess microparticles distributed in any area other than the holes. It is expected that potential application fields will be expanded by increasing the arrangement accuracy.

4. CONCLUSIONS

In this study, we attempted to expand the applicability of the mechanism for arranging diamagnetic particles in a modulated magnetic field.

One of the measures taken was that a HAM was prototyped as a portable device for generating a high magnetic field. The magnetic efficiency using the prototyped HAM was then evaluated. It was confirmed that the prototype HAM has a high magnetic field and generates a high magnetic force field, even though it is approximately palm-sized at 50 × 50 × 20 mm. The magnetic field was 1.31 T, and the magnetic force field was approximately 201 T²/m, at a distance of 1 mm from the surface. The magnetic field distribution in the horizontal line over the central region was almost homogeneous. Thus, it was confirmed that the prototype HAM could reproduce the magnetic field of the electromagnet and that the desirable magnet was successfully developed.

The other success was that a substrate on which an Fe thin film was formed and patterned utilizing the MEMS process technology, compatible with the IC, was prototyped as a tool to generate a modulated magnetic field assumed to be implemented on the IC. According to the evaluation of the deposition condition as to the prototyped substrate, holes having 10, 30, and 50 μm diameters were arranged in an array at intervals of 60 μm, and the thickness was approximately 0.5 μm, similar to that of the Fe thin film. The Fe thin film was completely covered with the polyimide film. Therefore, it was confirmed that the film was manufactured as desired.

Finally, whether the diamagnetic particles that were dispersed in the paramagnetic surrounding medium could be arranged magnetically was verified utilizing the prototypes. It was observed that the microparticles were arranged in the hole portions. According to the simulation result, the magnetic field increased at the upper part of the Fe portion and decreased at the upper part of the hole portion. These results indicated that a modulated magnetic field was generated on the surface of the Fe thin film in the magnetic field and that the microparticles were absorbed into the arbitrary positions by the modulated magnetic field and by the difference in the magnetic susceptibility between the microparticles and the surrounding medium. As a consequence, we succeeded in creating the mechanism for arranging the diamagnetic particles in the modulated magnetic field while achieving portability and implementation on an IC.

Incidentally, it is necessary to improve the arrangement accuracy indicating the proportion of the state in which one particle is arranged into one hole. The first measure for improvement was to increase the amplitude of the modulated magnetic field by increasing the thickness of the Fe thin film.

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