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Magnetization curves for two-dimensional rectangular lattices of permalloy nanoparticles: experimental investigation and numerical simulation

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Abstract

This work is concerned with the behaviour of a regular 2D rectangular lattice of Ni_3Fe nanoparticles, determined by the dipole interaction between them. The samples under study, prepared by electron-beam lithography, consisted of about 10^5 particles approximately 50 nm in size. The magnetization curves were studied by Hall magnetometry for different external magnetic field orientations at 4.2 K and 77 K. The results indicate a collective behaviour of the system. The magnetization curves depend on the external magnetic field direction and temperature; the system exhibits multistability. A model system of interacting 3D magnetic dipoles forming a rectangular lattice was numerically simulated by solving a system of stochastic Landau–Lifshitz equations. The multistability of the system and steps in the magnetization curves were obtained. It is shown analytically that the shape of the magnetization curves depends on the character of the interaction in the system.

1. Introduction

The interest in systems of nanosize magnetic particles arises from their potential as ultrahigh-density magnetic recording media. It is assumed that each particle carries one bit of information and the maximum density of data recording can be made as high as 10^{10} bit cm^{-2}. There are two basic limitations on the geometrical dimensions of such systems. First, the particle size must not to be too small, since the lifetime of a particle with an imparted magnetic moment is $\sim\exp(KV/kT)$, where K is the energy density of the magnetic anisotropy, V is the particle volume, T is the temperature and k is the Boltzmann constant. The effect of the thermoinduced rotation of the magnetic moment of a particle was termed superparamagnetism. Second, the energy of the particle interaction in a recording medium must be much smaller than the energy of the magnetic anisotropy. Otherwise, the particles show a collective behaviour. One fundamental type of interaction between single-domain particles, which can lead to a collective behaviour, is the dipole interaction. The energy of the two-particle interaction has the form

$$E = \frac{1}{2}\sum D_{ik}(\boldsymbol{x}-\boldsymbol{y})M_i(\boldsymbol{x})M_k(\boldsymbol{y}) \qquad D_{ik}(\boldsymbol{x}) = \frac{\delta_{ik}}{x^3} - \frac{3x_i x_k}{x^5} \tag{1}$$

where $M_i(\boldsymbol{x})$ is ith component of the magnetic moment of a particle at point $\boldsymbol{x}$. It follows from (1) that, to reduce the interaction energy, one has to make the interparticle distance longer.

By contrast, if there is a strong interaction in a system of supermagnetic particles, a situation is possible where the system becomes ordered. Such a state of the system has been named supermagnetic [1]. Owing to the anisotropic nature of the dipole interaction (1), the type of long-range order existing in the ground state strongly depends on the lattice parameters [2]. For example, two-dimensional lattices with a rhombic unit cell may have a ground state with a ferromagnetic or antiferromagnetic type of ordering, depending on a rhombicity angle [3]. This means that, by varying the 2D lattice symmetry, particle size and interparticle distance, one can, in principle, create magnetic structures with prescribed properties. The critical temperature of disordering

$$T_c \simeq M_0^2 V^2 / R^3 \tag{2}$$

depends on the characteristic value of the dipole interaction between particles and equals about 100 K for typical values of magnetization $M_0 \simeq 1000$ G, $V \simeq 10^{-18}$ cm^3 and interparticle distance $R \simeq 10^{-5}$ cm.

In this work, some results from investigations into the collective behaviour of such systems are reported. We focused on experimental and theoretical investigations of the multistability of 2D lattices with rectangular unit cells.

2. Experiment

Two-dimensional lattices of nanosize magnetic particles were formed by electron-beam lithography from permalloy films (Ni_3Fe) laser deposited on a substrate. The patterns were produced using C_{60} fullerene films as negative electron resists and Ti films as transmitting layers. In this way it is possible to form 2D lattices consisting of cylinder-shaped particles with diameters of 15 to 100 nm and height equal to the thickness of the original (Ni_3Fe) film (figure 1). The magnetic properties of the sample were studied using a differential scheme comprising two semiconductor (InSb) Hall sensors with common potential contacts

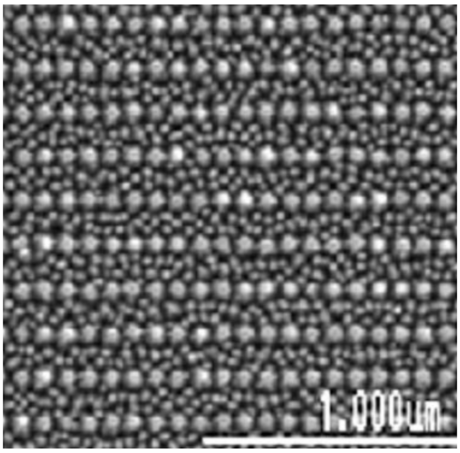

Figure 1. A SEM image of a part of a sample. The particles form a lattice with a rectangular unit cell, 90 × 180 nm. The particles, 40 nm in diameter and 45 nm high, are visible against the background of the 10 nm roughness of the sublayer.

and independent current contacts [4]. The dimensions of the Hall cross of the sensors was 50×100 μm, the thickness 10 μm. The system under study was produced in the working zone of a sensor. The method developed can be used to measure the magnetic field component perpendicular to the sensor plane over a wide temperature range.

Figures 2, 3 and 4 show the Hall signals for a sample magnetized at different orientations of the external magnetic field and $T = 4.2$ K. The external field direction is characterized by two angles, θ and ϕ, where θ is the angle measured from the normal to the sample and ϕ is the azimuthal angle measured from the axis directed parallel to the particle chains oriented along the short side of the elementary rectangular cell. The dependence of the magnetization curves on the field orientation relative to the lattice axes cannot be interpreted in terms of individual particle properties and is a manifestation of the collective behaviour of the particles. Note that the magnetization in the geometries of figures 3 and 4 is a remanent magnetization. In addition, fundamental changes in the magnetization curve of a rectangular-unit-cell sample occur when the sample temperature is raised to 77 K (figure 4). This indicates that thermal fluctuations play a significant role in the system, since the T_c of bulk permalloy is 885 K $\gg$ 77 K.

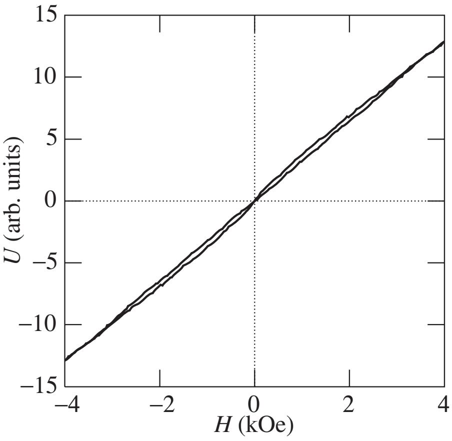

Figure 2. The Hall signal in a magnetic field with $\theta = 45°$, $\phi = 0°$.

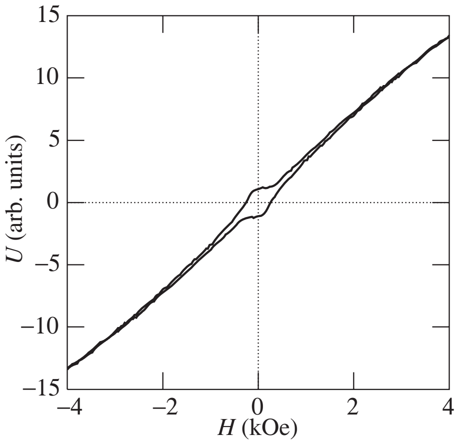

Figure 3. The Hall signal in a magnetic field with $\theta = 45°$, $\phi = 90°$.

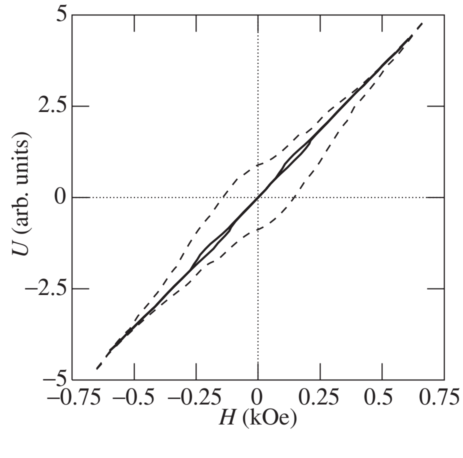

Figure 4. Changes in the Hall signal with temperature: 4.2 K (broken curves) and 77 K (full curves). H is directed at $\theta = 0°$.

Of no less interest is the Hall signal obtained with magnetization directed along the shorter side of the elementary rectangle (which corresponds to the easy-axis direction) (figure 5). It is significant that the magnetic susceptibility depends on the direction in which the magnetic field is changed. The special feature of the magnetic hysteresis obtained is as follows. With increasing external magnetic field, the magnetization follows the lower curve, and with decreasing field, the upper one, irrespective of the initial conditions. If the sign of the derivative of the external field is changed to the opposite one, the system passes from one magnetization curve to the other. So, the system is multistable, i.e. it has a number of metastable states, since different magnetization values may correspond to the same external fields. The following sections of this article are devoted to a theoretical investigation of the magnetization process of a rectangular dipole lattice in a magnetic field directed along the short side of the elementary rectangle.

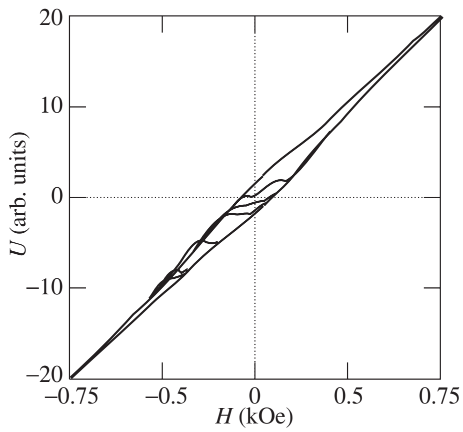

Figure 5. The Hall signal versus the external magnetic field directed along $\theta = 90°$, $\phi = 0°$.

3. Numerical simulations

To better understand the magnetization process, we provide a numerical simulation of a model system in the form of a bound 2D rectangular lattice of magnetic dipoles. The mechanisms of the reversal of infinite dipole lattices at $T = 0$ were analysed in detail in [5, 6] and it was shown that two modes are possible, depending on the lattice parameters: either by coherent rotation or by antiferromagnetic fanning. In this case the effective antiferromagnetic interaction between the neighbouring chains is weak (the integral dipole interaction between chains decays exponentially) and the states with arbitrary orientations of magnetization in the neighbouring chains are good multistable states. In a bound dipole system, the magnetostatic interaction between dipole chains is due to the magnetic charges on their edges [7]. Owing to the fact that the interchain interaction is slowly decaying ($\sim 1/r^3$, $r \gg L$; r is the distance between the chains and L is their length) and there are thermal fluctuations ($T \neq 0$), other reversal modes are possible [8]. To examine these mechanisms, a system of the classical dipoles (up to 1000 dipoles) on a rectangular lattice was numerically simulated by solving stochastic Landau–Lifshitz equations with relaxation. These numerical methods are well evaluated and widely used (see, e.g., [9]). The main feature of the reversal process is that it occurs via successive reversals of separate dipole chains. It is the sequence of the multistable states of the system that gives rise to steps in the magnetization curve (figure 6). After a reversal of the next chain occurs

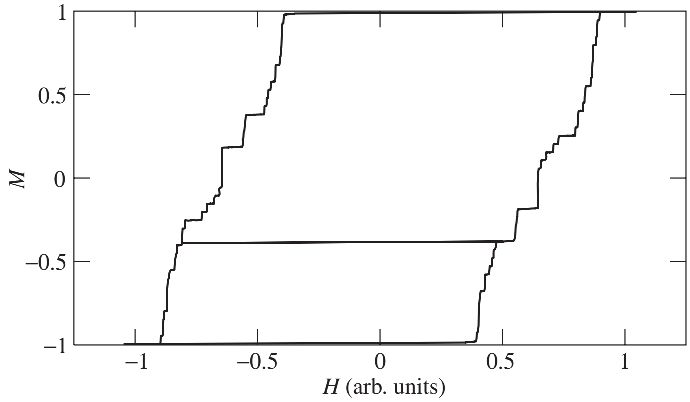

Figure 6. Magnetization versus external field directed along the dipole chains for 32×6 dipoles. The horizontal line shows one of the possible transitions between the magnetization curves. $T = 0.1M^2/a^3$ (T is the temperature, M the magnetic moment of the dipole, a the short side of the 1:2 elementary rectangle).

with increasing external field, the effective field affecting other dipole chains decreases owing to the effective antiferromagnetic character of the dipole interaction, and further increase of the field is needed to continue the reversal process. The number of steps exactly corresponds to the number of dipole chains in the system. The difference in step width is a consequence of the long-range character of the interchain interaction.

Another interesting feature is the following. If during the magnetization process we stop changing the external field and start to change it in the opposite direction, the magnetization itself does not change while we pass through the entire hysteresis loop. The number of such possible transitions corresponds to the number of steps on the magnetization curve. While being magnetized, the system goes from one metastable state to another. The width of the transitions from the left-hand magnetization curve to the right-hand one corresponds to the stability region of a definite metastable state. To make this state unstable, we must change the external field to one that is beyond these values.

The most interesting form of the magnetization curve was observed for a system of finite-length dipole chains with a periodic boundary condition in the direction perpendicular to the chains (figure 7). In this case the edge of the hysteresis loop has a self-similar character (thinner steps lie between the thicker ones). A similar behaviour, known for a system of chains of absorbed particles with mutual repulsion (i.e. antiferromagnetic interaction) between them [10], is named the ‘devil’s staircase’. To gain insight into the behaviour of the magnetic system in our case, the following simple model was solved for the system in question: a system was considered of 2^N (with $N \to \infty$) dipole chains with an antiferromagnetic interaction between them, represented in general form as

$$E = JM(\boldsymbol{r}_1)M(\boldsymbol{r}_2)/|\boldsymbol{r}_1 - \boldsymbol{r}_2|^p.$$

For the dipole interaction, $J = 1$, $p = 3$, M is the total magnetic moment of a chain. The magnetization reversal of an isolated dipole chain depends on its coercivity, considered to be same for the different chains. Reversal in the field aligned with the easy axis was examined analytically. The magnetization reversal has its onset at the external field $H_1 = H_a - 2JM\zeta(p)/a^p$ and terminates at $H_2 = H_a + 2JM\zeta(p)/a^p$, where H_a is the coercive field of an isolated dipole chain and $2\zeta(p)$ ($\zeta(p) = \sum_{n=1}^{\infty} 1/n^p$) is the field induced at the dipole chain by all other dipole chains of the system in the fully magnetized state and

a is the interchain distance. The magnetization curves have self-similar character, like that numerically calculated and shown in figure 7. In this case, the step width depends on the magnetization value:

$$\Delta H_k = \frac{2^p - 2}{2^{kp-2}} \zeta(p) \frac{JM}{a^p}$$

where $k = 1, 2, 3, \ldots$. The number of steps with this width is $N_k = 2^{k-1}$ and the corresponding magnetization values are $M = m/2^{k-1} - 1$, $m = 1, 3, 5, \ldots, 2^k - 1$. The sum of all step widths is exactly the same as the whole inclination width of the magnetization curve, equal to $4(JM/a^p)\zeta(p)$. In fact,

$$\Delta H = \sum_{k=1}^{\infty} N_k \, \Delta H_k = 2(2^p - 2) \frac{2(2^p - 2)JM}{a^p} \zeta(p) \sum_{k=1}^{\infty} \frac{1}{2^{k(p-1)}} = 4 \frac{JM}{a^p} \zeta(p).$$

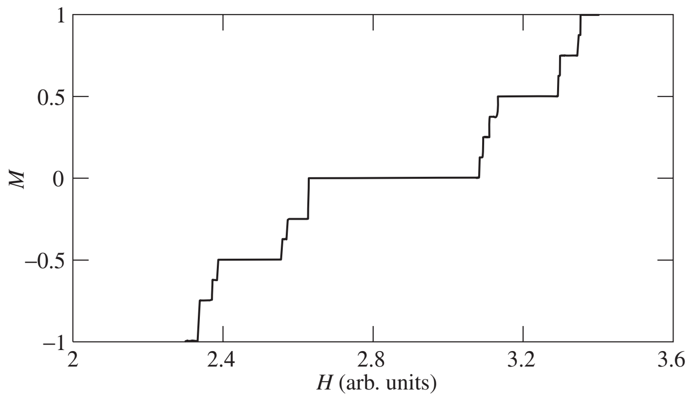

Figure 7. The edge of the hysteresis loop for a system of 16 dipole chains with 5 dipoles in each chain. The elementary rectangle is 1:2. $T = 0.05M^2/a^3$.

4. Discussion

So, we have obtained some experimental results concerning the collective behaviour of the system. Some of them (effective anisotropy of the system due to dipole interaction) are quite understandable. Another result, namely the existence of a remanent magnetization in the magnetization perpendicular to the easy axis, is unexpected. Nevertheless, this effect was observed for a number of samples. Such a behaviour is likely to be associated with the formation of nonuniform states in the system of interest. This hypothesis requires further experimental and theoretical verification. As we do not quite understand the nature of this effect, its changing with the temperature (figure 4) cannot be taken as a sign of the supermagnetic transition in the system, but it indicates a role of the fluctuations in the system. The question of whether or not this system undergoes a transition to a superparamagnetic state can be solved by investigating the system further.

Numerical simulations fail to give good correspondence with the experiment, but they are of interest in their own right. It was shown that the interaction in the system of magnetic dipoles on a rectangular lattice must lead to the appearance of steps in the magnetization curve. The steps in the ideal system are self-similar. These steps cannot be detected in experiment as yet, owing to their small width. Nevertheless, the multistability of the system (transitions from the left-hand side of the hysteresis loop to the right-hand one) were observed both experimentally and in numerical simulations. This leads us to maintain that, in the experimental samples, the magnetization process in the easy direction occurs by successive magnetization reversals of the particle chains.

Acknowledgments

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