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Controlling magnetic ordering in coupled nanomagnet arrays

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Abstract. We have fabricated using high-resolution electron beam lithography circular magnetic particles (nanomagnets) of diameter 60 nm and thickness 7 nm out of the common magnetic alloy supermalloy. The nanomagnets were arranged on rectangular lattices of different periods. A high-sensitivity magneto-optical method was used to measure the magnetic properties of each lattice. We show experimentally how the magnetic properties of a lattice of nanomagnets can be profoundly changed by the magnetostatic interactions between nanomagnets within the lattice. We find that simply reducing the lattice spacing in one direction from 180 nm down to 80 nm (leaving a gap of only 20 nm between edges) causes the lattice to change from a magnetically disordered state to an ordered state. The change in state is accompanied by a peak in the magnetic susceptibility. We show that this is analogous to the paramagnetic–ferromagnetic phase transition which occurs in conventional magnetic materials, although low-dimensionality and kinetic effects must also be considered.

Contents

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1. Introduction

One of the most exciting recent developments in magnetism has been the use of nanometre fabrication techniques to form nanometre-scale magnets [1]. These so-called nanomagnets possess very different magnetic properties from their parent bulk material and may provide advanced replacements for hard disk media [2], non-volatile computer memory chips [3] and an environment in which to implement quantum computing [4]. A nanomagnet is analogous to a single giant atom of a new magnetic element; new artificial magnetic materials can be constructed from a lattice of these artificial atoms. In this paper we demonstrate how the magnetic properties of a lattice of nanomagnets can be profoundly changed not only by the properties of each constituent nanomagnet but also by the magnetostatic interactions *between* nanomagnets within the lattice. We find that simply reducing the lattice spacing in one direction causes a fundamental change in behaviour by causing the lattice to change from a magnetically disordered state to an ordered state.

2. Experimental

The samples were made by high-resolution electron beam lithography. Two layers of polymethylmethacrylate (PMMA), one of molecular weight 495 000 and one of weight 950 000, were spun onto a single-crystal (100) silicon substrate. Arrays of circular discs were then exposed onto the sample in a JEOL 4000EX SEM/TEM. A number of different arrays were made on a single substrate (and therefore processed together to enable comparison), each comprising approximately 5000 identical nanomagnets of diameter 60 nm arranged on a two-dimensional rectangular lattice. The y-direction lattice period was kept constant at 180 nm (i.e. three times the diameter of the nanomagnets), whereas the x-direction lattice period varied in different arrays from 180 nm down to as small as 80 nm (i.e. leaving only 20 nm between neighbouring edges). The sample was developed for 30 s in a $1:3$ solution of methyl iso-butyl ketone (MIBK)/iso-propyl alcohol (IPA). A 7 nm thick layer of $Ni_{80}Fe_{14}Mo_5$ ('supermalloy') was then deposited at a rate of 0.08 nm s^{-1} by electron beam evaporation in an ultra-high vacuum chamber with a base pressure of 4×10^{-9} mbar. An unpatterned substrate was also present in the chamber to allow structural and magnetic characterization of the unpatterned magnetic film. Ultrasonic-assisted lift-off in acetone was used to remove the magnetic film from the unexposed parts of the patterned sample. The lower PMMA layer, having a smaller molecular weight, had a lower exposure threshold, causing slightly larger discs to be developed in it compared to the upper layer. The resulting undercutting assisted the lift-off process.

Transmission electron microscopy (TEM) and cross sectional TEM showed the deposited supermalloy to have a random polycrystalline microstructure with grains of size ~ 10 nm and a surface roughness of less than 0.5 nm. Scanning electron microscopy (SEM) was used to check the size and shape of the nanomagnets. Figure 1 shows some of the SEM images. From these we are able to set an upper bound of 2% on the random ellipticity of the nominally circular structures, independent of the x-direction lattice period. Only arrays with an x-direction periodicity of 80 nm showed any occasional failure of the lithography, leading to two neighbouring nanomagnets coalescing (these were removed from the statistical analysis of ellipticity).

In addition to this structural characterization, we have also performed magnetic characterization. Magneto-optical magnetometry was used to measure the coercivity (~ 1 Oe)

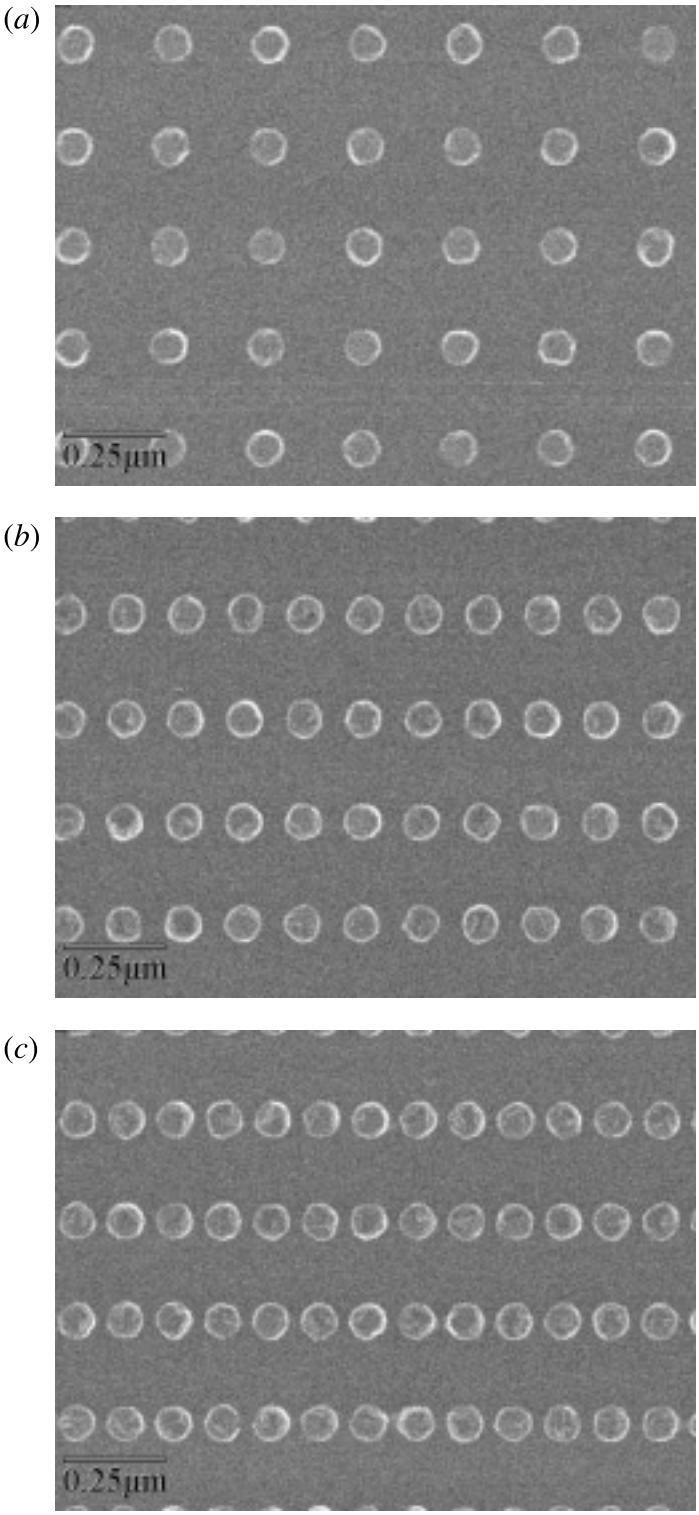

Figure 1. Scanning electron micrographs of some of the nanomagnet lattices. Each nanomagnet is of 60 nm diameter and has a y-direction lattice parameter of 180 nm. The x-direction lattice parameter is (*a*) 180 nm, (*b*) 110 nm and (*c*) 90 nm.

and the anisotropy (4 ± 1 Oe, uniaxial in-plane) of the unpatterned film. A B–H looper was used to check the thickness and saturation magnetization (800 ± 60 emu cm^{-3}) of the unpatterned film. Temperature-dependent measurements showed the unpatterned films still to be ferromagnetic at 300 °C, which is not inconsistent with the expected Curie temperature of 400 °C and hence an exchange stiffness of $\sim 10^{-6}$ erg cm^{-1}. Surface oxidation of the unprotected magnetic film in air was found to be limited to the top 1 nm of supermalloy.

We have determined the magnetic properties of these different lattices by measuring their hysteresis loops (M–H loops) using a high-sensitivity magneto-optical method [5]. The shape of a magnetic hysteresis loop is a sensitive probe both of the internal magnetic structure of the nanomagnets and of any interactions between them. An optical microscope allows the silicon surface to be viewed while a focused laser spot (size ~ 5 μm, the approximate size of each array of nanomagnets) is moved into position on top of one of the lattices. Polarization analysis of the reflected laser beam allows us to probe the component of lattice magnetization lying in the optical plane of incidence, via the longitudinal magneto-optical Kerr effect [6]. This magnetization is then recorded while an alternating magnetic field is applied in the plane of the lattices. All measurements were performed at a stabilized room temperature.

Figure 2 shows hysteresis loops obtained from the different lattices for the cases of the field applied along the lattice x- and y-directions. One sees that when the nanomagnets are widely separated (e.g. figure 2(*a*)) the hysteresis loops have a characteristic 'S' shape and are fully closed (i.e. zero area inside the loop). As the x-axis spacing is reduced, however, the loops show a significant change in their central region, the x-axis loop opening up, while the y-axis loop becomes more sheared (e.g. figures 2(*c*) and (*d*)).

We explain these changes as being due to magnetostatic interactions between nanomagnets [7]. Each nanomagnet can be represented to a good approximation as a point magnetic dipole located at the nanomagnet centre. The magnetic field emanating from such a magnetic dipole falls off with the cube of the distance from it. The largest lattice period ($X = 180$ nm, see figures 1(*a*) and 2(*a*)) causes the nanomagnets to be spaced by three times their own diameter, which is a sufficiently large distance for magnetostatic interactions between nanomagnets to be relatively weak. The measured average property of the lattice is thus *approximately* the same as the individual property of an isolated nanomagnet [5]. It has already been shown that the shape of a nanomagnet imposes a magnetic anisotropy of related symmetry order [8]. In our case, the circular symmetry means that all in-plane magnetization directions are equally favourable and so even though each nanomagnet is small enough to avoid domain formation [9], large thermal fluctuations of the giant spin vector occur, leading to a time-averaged magnetization of zero. This phenomenon is usually called superparamagnetism [10] and in well-spaced spherical particles is described by the statistical mechanical Langevin function [11]. The prismatic shape of our nanomagnets and the small interaction field which is present even in the $X = 180$ nm arrays mean, however, that a numerical calculation using a Monte Carlo algorithm is a more accurate way of predicting the behaviour. We have performed such a calculation, the details of which will be published elsewhere, in order to test this interpretation and have fitted the result to the $X = 180$ nm hysteresis loops of figure 1 to obtain a value for the ratio $m/k_{\mathrm{B}}T$ and hence m, the magnitude of the magnetic dipole moment which is undergoing thermal fluctuations (k_{B} is the Boltzmann constant and T is temperature). The value of m thus obtained was $m = (1.5 \pm 0.2) \times 10^{-14}$ emu, which compares favourably with the expected value of 1.6×10^{-14} emu found from $M_{\mathrm{s}}V$, the total moment carried by a single nanomagnet. Here M_{s} is the saturation magnetization of supermalloy (measured by B–H looping of our unpatterned films

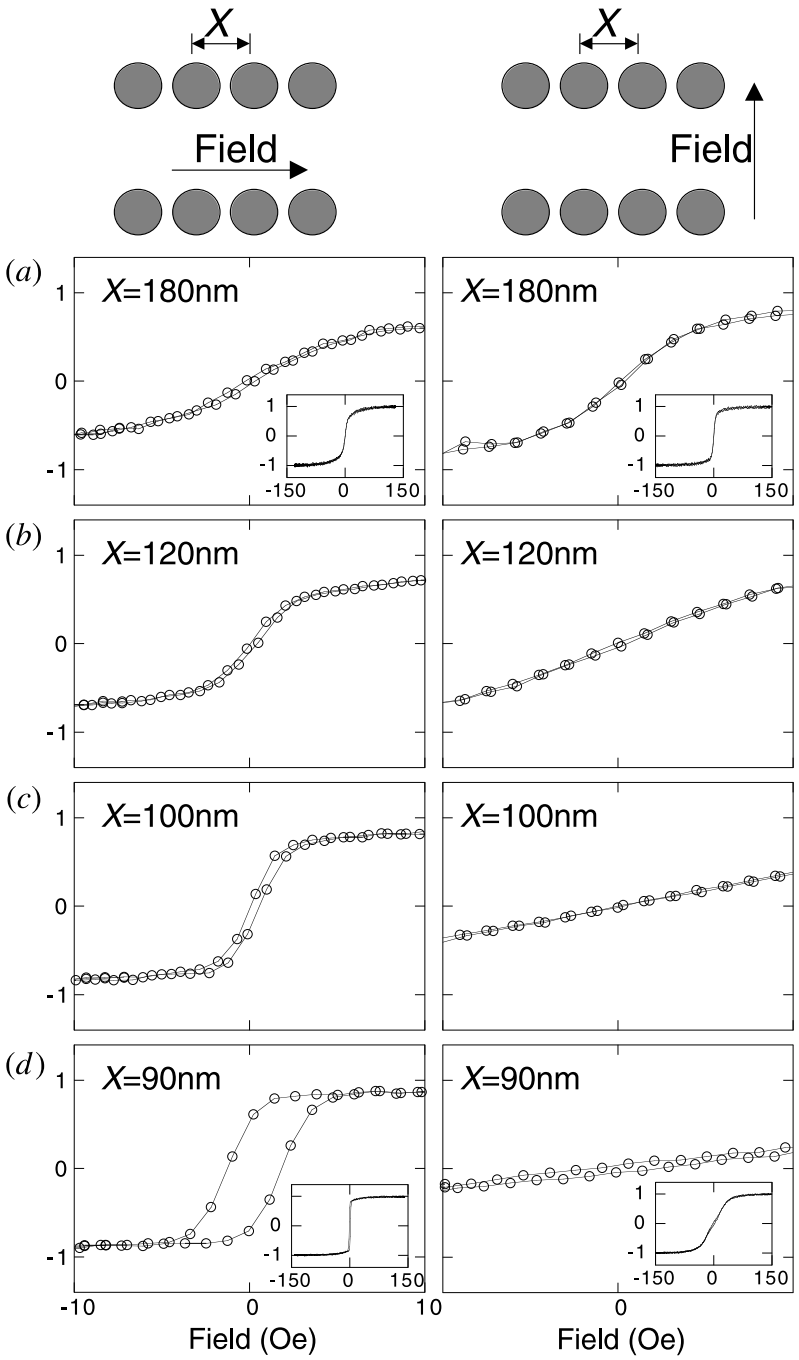

Figure 2. Hysteresis loops measured for different lattice spacings and applied field directions. All loops were measured within the field range ± 150 Oe: the large panels show high-magnification views around zero field; insets show the full measured loop. The vertical axis of all loops is magnetization normalized by the saturation value. Panels (*a*) and (*b*) show the lattice to be magnetically disordered. Panels (*d*) show the lattice to be magnetically ordered. Panels (*c*) are near to the transition point between an ordered and disordered lattice.

as 800 emu cm^{-3}) and V is the volume of a nanomagnet. This confirms that each nanomagnet is indeed behaving as an isolated giant fluctuating spin and is hence in the superparamagnetic state.

A lattice of nanomagnets each in the superparamagnetic state is directly analogous to a sample of a conventional *paramagnetic* material such as oxygen where, although each constituent atom carries a net spin, the lack of sufficient coupling between atoms allows thermal fluctuations to reduce the average moment to zero.

As the separation between nanomagnets is now reduced, as shown in figures 1(*b*) and (*c*), the magnetostatic coupling between nanomagnets (especially, but not exclusively, between nearest neighbours) becomes stronger to the point at which it can overcome the thermal fluctuations. When this occurs, the spins essentially remain parallel and locked together in the x-direction even under zero applied field, leading to increased remanence (magnetization under zero field) in the loops of figures 2(*c*) and (*d*). This is directly analogous to the onset of *ferromagnetism* in an atomic lattice, and is particularly interesting because spin ordering has in our experiments been achieved through *magnetostatic* interactions. Although historically Weiss [12] was able to explain many features of ferromagnetism in conventional materials by assuming an interaction field (called the molecular field), he was also aware that magnetostatics alone could never be powerful enough to overcome thermal fluctuations in single atoms. Our lattice *is* able to achieve ordering through magnetostatic interactions because of the giant magnetic moment ($\sim 10^6$ Bohr magnetons, cf ~ 1 Bohr magneton for a magnetic atom) carried by each nanomagnet. By simply changing the spacing between nanomagnets we can thus control whether the lattice adopts a magnetically ordered or disordered state.

A paramagnetic–ferromagnetic phase transition in conventional magnetic materials is of second order and so is accompanied by a peak in the isothermal susceptibility at the point of transition between phases (called the critical point). The susceptibility χ_T is defined (in c.g.s. units) as $\chi_T = 4\pi\, \mathrm{d}M/\mathrm{d}H|_{H=0}$ where M is the spontaneous magnetization under an applied field H and hence can be found from the gradient of the central portion of the hysteresis loops. In order to see whether a similar signature exists at the onset of ordering of our nanomagnet lattices, we have measured χ_T as a function of the x-direction lattice period as that period is varied through the transition point. Figure 3 presents the results for the cases of the field applied in the x- and y-directions. We have also plotted the remanence of the loops on the same axes, as this signals which state the lattice is in (zero x-direction remanence is disordered, finite x-direction remanence is ordered). Indeed, in figure 3(*a*) one sees a peak in the susceptibility at $X = 100$ nm coinciding with the onset of remanence and hence the ordered state.

In contrast, the signature is not observable in the y-direction of the lattice (figure 3(*b*) shows no peak in the susceptibility and no rise in remanence) because this is the 'hard' magnetization direction and so displays remanence in neither the disordered nor the ordered phase. Nevertheless, the effect of magnetostatic interactions is still very much in evidence with the susceptibility falling with decreasing separation as the nanomagnets become increasingly coupled. The fact that the x- and y-direction susceptibilities do not become exactly equal in the limit of $X = Y = 180$ nm is due to the presence of weak uniaxial anisotropy in the supermalloy.

3. Discussion

The ratio $J_\mathrm{c}N/k_\mathrm{B}T$, where J_c is the interaction energy between a pair of neighbours at the critical point and N is the coordination number, is of prime importance when considering order–disorder phase transitions. In a simple picture one can think of each element (be it a nanomagnet or an

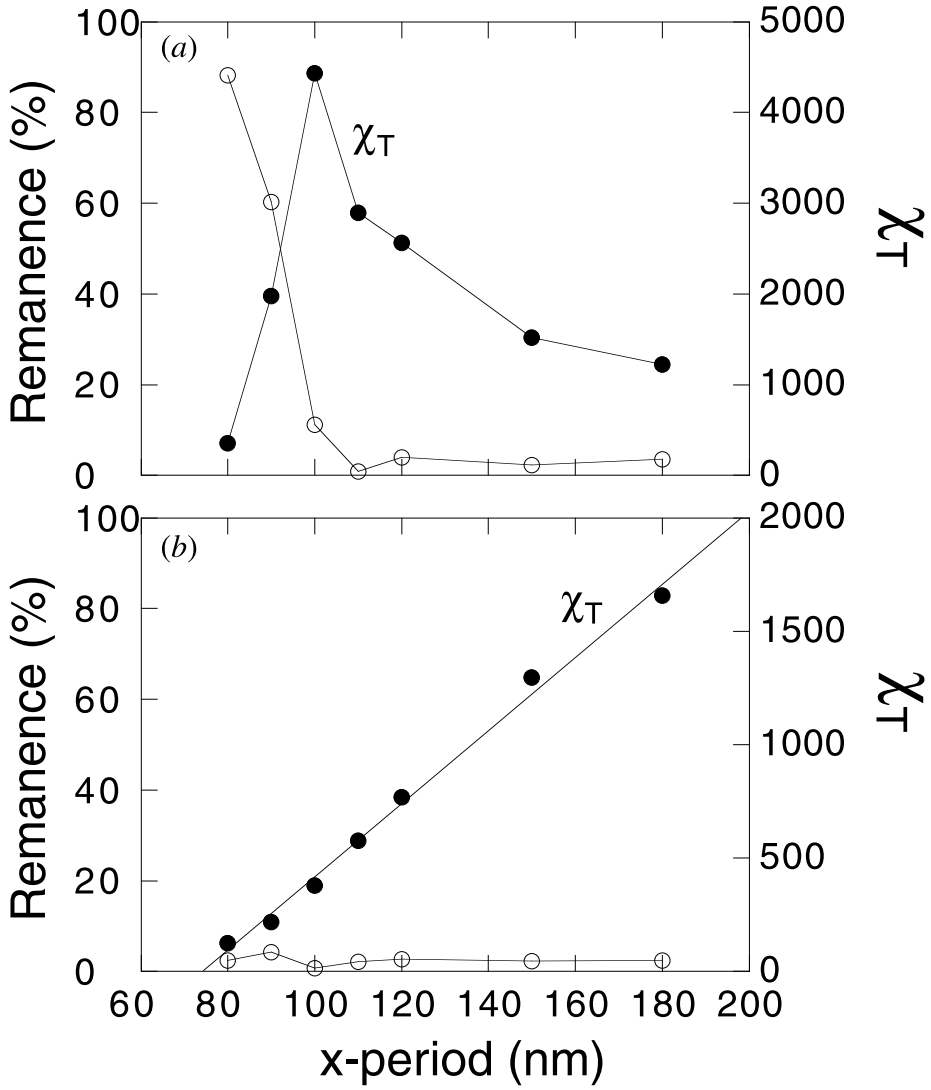

Figure 3. Remanence (open circles) and susceptibility (full circles) measured as a function of lattice spacing for the field applied along (*a*) the lattice x-direction and (*b*) the lattice y-direction. The peak in susceptibility coinciding with a rise in remanence in (*a*) marks the transition from lattice disorder to order and is analogous to the peak which occurs in the paramagnetic–ferromagnetic phase transition of conventional materials. Error bars are approximately equal to the size of the points.

atom) as possessing $k_{\mathrm{B}}T/2$ thermal energy and JN interaction energy. These two terms work in opposition, the thermal energy encouraging disorder and the interaction energy encouraging order and so the value of the ratio at the critical point is a measure of how willing the system is to order: in the simplest picture one could anticipate a value of the order of unity. The magnetostatic coupling energy between two moments m separated by a distance X is m^2/X^3 and N is 2 (for small X the system approximates to a one-dimensional (1D) chain of spins) which at the critical point of our system (i.e. $X = 100$ nm) gives $J_{\mathrm{c}}N/k_{\mathrm{B}}T = 12$. This is a high value, meaning

that a high interaction energy is required to effect ordering. It is, however, a general feature of theoretical 1D systems that they do not order easily. Neither the 1D Ising model nor the 1D XY model [13], the two closest theoretical relatives to our system, order at all theoretically at finite temperature. This is generally because there is always a finite probability of nucleating a defect where the direction of one spin in an ordered chain abruptly reverses, which leads to a pair of spin waves which can freely propagate along the entire chain thus destroying all knowledge of the magnetization direction. In an infinite chain there will always be at least one of these defects which is enough to destroy its entire order. In a *finite* chain, however, it is also necessary to consider *kinetics*: there will be an average waiting time before the defect nucleates. If this time is long compared to the experimental measurement time (~ 0.1 ms in our case) then full ordering will be observed. A simple application of the Boltzmann probability factor [14] gives the waiting time as

$$\tau = \frac{\ln 2}{2N\nu \exp(-\Delta E/k_{\mathrm{B}}T)}$$

where N is the number of nucleation sites, ν is the attempt frequency (conventionally taken as 10^{10} s^{-1}) and ΔE is the magnetostatic energy of a defect. If the defect nucleation begins at the end of the chain then $N = 2$ and ΔE can be estimated to be $4m^2/X^3$. The critical X-direction period for ordering is thus $X = [4m^2/k_{\mathrm{B}}T \ln(4\nu\tau/\ln 2)]^{1/3}$ which evaluates to 112 nm. This is in good qualitative agreement with experiment ($\sim$100 nm) given the simplicity of the model. A more precise model would also take into account dimensionality cross-over effects due to the small but finite interactions between chains and the precise shape (and hence energy) of the nucleated defect.

4. Conclusion

We have used electron beam lithography to make circular nanomagnets arranged on rectangular lattices of differing x-direction spacing. A high-sensitivity magneto-optical method was used to measure their magnetic properties. When the nanomagnets are spaced on a relatively large pitch (180 nm in both x- and y-directions) magnetostatic coupling between nanomagnets is sufficiently weak that thermal fluctuations are able to destroy long-range order in the array. Such arrays are found to possess zero remanence, and, if one maps nanomagnets onto atoms, can be considered to be analogous to paramagnetic materials. As the x-direction lattice period is reduced, we show experimentally that magnetostatic interactions become stronger until the point is reached where they can overcome thermal fluctuations in the system and long-range order is established. Such arrays possess a high remanence and can be considered to be analogous to ferromagnetic materials. We identify experimentally a characteristic peak in the isothermal susceptibility at the transition point between the disordered and ordered states, as would be seen in the analogous paramagnetic–ferromagnetic phase transition. A simple kinetic model shows that all of these effects can be understood if one considers the effect of the lattice spacing, and hence magnetostatic coupling energy, on the defect nucleation time. When this is less than the experimental observation time (weak coupling), a magnetically disordered lattice is observed. When it is greater than the experimental observation time (strong coupling), a magnetically ordered lattice is observed.

Acknowledgments

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