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Configurational spin reorientation phase transition in magnetic nanowire arrays

Elena V. Tartakovskaya [a], Martha Pardavi-Horvath [b,*], Manuel Vázquez [c]

[a] Institute of Magnetism NAS of Ukraine, Kiev, Ukraine
[b] SEAS ECE, The George Washington University, Washington DC, USA
[c] Instituto de Ciencia de Materiales de Madrid, CSIC, Madrid, Spain

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ABSTRACT

Classical microscopic spin reorientation phase transitions (RPT) are the result of competing magnetocrystalline anisotropies. RPTs can also be observed in discrete macroscopic systems induced by competing shape anisotropies and magnetostatic coupling. Such a configurational RPT was recently observed in series of self-organized hexagonal arrays of 2.5 μm long, 25–60 nm diameter circular permalloy nanowires grown in anodic alumina matrix. This RPT is a crossover transition from a one-dimensional easy axis "wire" behavior of weakly interacting uniaxial nanowires to a two-dimensional behavior of strongly coupled "wire film" having an easy plane anisotropy. It is shown that RPT takes place due to the competition between the intrinsic dipolar forces in individual wires and the external dipolar field of interacting nanowires in the array. The crossover occurs at a volume ratio of 0.38 for 65 nm periodicity. The experimental results are in agreement with the semi-analytical calculations of the dipolar interaction fields for these arrays of circular ferromagnetic nanowires, and are interpreted in terms of the Landau phase transition theory. The conditions for the crossover and the order of the phase transition are established. Based on the contribution to the magnetic energy from the flower state at the ends of the wires, it is concluded that the observed transition is of the first order.

1. Introduction

In a classical spin reorientation phase transition (RPT) the magnetization vector rotates continuously with respect to the crystallographic axes as a consequence of symmetry changes in magnetocrystalline anisotropy terms upon changes in temperature, crystal structure, or magnetic field. RPT is defined as the change of the stability of the magnetic structure of the spin system when the dominant anisotropy term in the free energy changes sign at a critical value of a parameter. At the RPT the next anisotropic term dominates the system, and determines the order of the phase transition [1]. Spin reorientation phase transitions can also be observed in discrete, nanostructured systems of single domain particles, induced by competing shape anisotropies of the particles and interactions, due to magnetostatic coupling [2].

Particularly, such a configurational RPT was observed in series of $25 \leq 2R \leq 60$-nm-diameter hexagonal array of L=2.5 μm long uniaxial permalloy nanowires, having a periodicity of D=65 nm [3]. The RPT was demonstrated from studies about angular dependence of the ferromagnetic resonance (FMR) spectra that reflects the symmetry of the magnetic system. A crossover RPT from an one-dimensional easy axis "wire" behavior of the saturated and weakly interacting uniaxial nanowires to a two-dimensional behavior of strongly coupled "wire film", having an easy plane anisotropy, was thus described. At the crossover, the difference in the resonance fields, measured parallel and perpendicular to the wires, changes sign, as shown in Table 1. The crossover occurs at a volume ratio of 0.38 for the array with 65 nm periodicity (i.e., for sample SU03 with $2R$=45 nm).

Similar RPTs, in other small magnetic particle systems driven by thickness, shape change, or microstructural effects were discussed in [4–6]. Actually, the problem of interacting wires, considered here, is quite similar to the one of superparamagnetic nanosphere arrays, described earlier [7,8]. Nevertheless, as it has been pointed out previously [9], the main difference is that the dipolar-like interaction between the wires is considerably stronger than in the weakly magnetic nanosphere arrays, and it can lead to the change of the sign of the expression for the *system's* shape anisotropy, i.e., to the RPT.

In the present work, we make use of the measurements previously reported on nanowire arrays [3]. Arrays of permalloy nanowires were grown on anodic alumina template exhibiting hexagonal symmetry arrangement (see Fig. 1). Further details of preparation and characterization can be found elsewhere [10]. Characteristics of nanowire arrays are collected in Table 1.

* Corresponding author. Tel.: +1 202 994 8591; fax: +1 202 994 0187.
E-mail address: mpardavi@gwu.edu (M. Pardavi-Horvath).

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Table 1
Comparison of experimental and theoretical results for demagnetizing factors for hexagonal permalloy nanowire arrays.

	SU01	SU02	SU03
$2R$	25	35	45
D	65	65	65
$H_{r\parallel}-H_{r\perp}$	1926	1393	−150
$I_{xx}^{th}(single)$	0.990	0.994	0.992
$I_{xx}^{th}(int)$	0.87	0.75	0.60
$I_{xx}^{exp}(int)$	0.81	0.72	0.61

Wire diameter: $2R$, period of the array: D (dimensions in nm). $H_{r\parallel}-H_{r\perp}$ is the FMR anisotropy field, $I_{xx}^{th}(single)$ and $I_{xx}^{th}(int)$ are the calculated normalized demagnetizing factors of an individual wire and for the interacting wire array, $I_{xx}^{exp}(int)$: experimental data from Ref. [3] (SU is the sample ID).

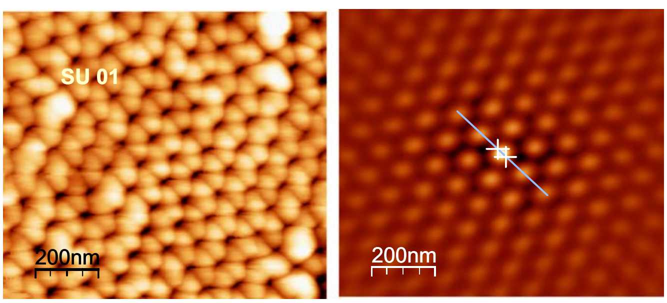

Fig. 1. AFM and self-correlation image of the self-organized permalloy nanowire array in an alumina matrix (sample SU01, see Table 1). Wire diameter: d=2R=25 nm, periodicity: D=65 nm.

The goal of the present work is to describe this configurational magnetic phase transition of a discrete system based on the classical spin reorientation model and on the Landau theory of magnetic phase transitions [1].

2. The model: RPT as a result of interwire interactions

The magnetic energy of the system of the 2D array of magnetic nanowires, W, can be written as a series expansion $W=W_1+W_2\ldots$. Here the lowest order term $W_1=(M_0^2/2)\Delta I\cos^2\theta_0$ depends on the uniform distribution of the magnetization in the individual wires, M_0 is a saturation magnetization, $\Delta I=I_{xx}-I_{zz}$ is the shape anisotropy of the system, I_{ii} are the system (array) demagnetizing factors, and θ_0 is the angle between the magnetization and the wire axis, chosen as the axis z. The second term W_2, connected to the non-uniform magnetization state at the ends of the wires, will be discussed later. The experimentally observed RPT is defined by the changes in the demagnetizing factors of the wire array, which, in turn, depend on the dipolar interaction between wires.

The calculation of demagnetizing factors for interacting wires was performed by micromagnetic methods in [11], considering the wires as long ellipsoids and using results for interacting dipoles. As it was shown in Refs. [2,12], this approximation is insufficient for the close-packed nanoparticle systems. In our case more specific considerations, taking into account the particle shape more rigorously, are necessary. In particular, in the present samples the nanowire length L is much larger, than its radius, R. In similar cases L is usually considered as infinite [13]. This simplification is unsuitable for the present task of the RPT investigation. As a result, in the calculations presented here, every wire is considered as a long, but finite length cylinder. The significant assumption that we make here is that average magnetic moments in all wires in an array are canted by the same angle θ_0, and, so, all wires in the array are in identical magnetic ground state. As the original FMR measurements were made in relatively high magnetic fields at the FRM resonance, the saturated quasi-uniform magnetization distribution is justified.

The method of obtaining the system demagnetizing factors originates from the calculations of the magnetometric demagnetizing factor for an individual rectangular prism [14]. It was first applied to *arrays* of rectangular stripes and dots in [15]. To apply it to the case of cylindrical wires we start from the known phenomenological expression, taking into account the two-dimensional order of the array

$$I_{ii}(t)=\frac{1}{Vm}\int_{-L_z}^{L_z}\int_{-L_z}^{L_z}dzdz'\sum_{n,n'}\int_{S_n}\int_{S'_{n''}}d\boldsymbol{\rho}d\boldsymbol{\rho}'\frac{d^2}{didi'}\frac{1}{|\mathbf{r}-\mathbf{r}'|}.$$

The center of the coordinate system is in the center of the gravity of a selected wire, the sum is taken over all the wire cross-sections S_n (in the film plane xy), $V_m=N2L_z\pi R^2$ is the magnetic volume, N is the number of nanowires in the sample. We use here the real-space calculations of the magnetometric demagnetizing factor as the most appropriate for RPT analysis.

The phase transition occurs when $\Delta I=0$, i.e., $I_{xx}=I_{zz}=I^{crit}=4\pi/3$ (in normalized units $I^{crit}/2\pi=0.667$). For $I_{xx}/2\pi>0.667$, the energy minimum is reached when the magnetization is directed out of plane ($\theta_0=0$), while for $I_{xx}/2\pi<0.667$ the in-plane orientation of the magnetization is preferred ($\theta_0=\pi/2$). Both the experimental and the theoretical results (Table 1 and Fig. 2) show a considerable change of the system demagnetizing factor due to the dipolar interaction between wires. In Fig. 2, the results are presented for three radii of nanowires R, as given in Table 1. As all the results depend only on two geometrical parameters of wire arrays (they can be chosen as ratios R/D and R/L), the wire length is chosen L=2500 nm throughout the calculation, as it is in the actual experiments. Note, that the calculated difference in the demagnetizing factors $I_{xx}^{th}(single)$ of individual wires in different arrays (which arise due to the difference of wire radii) is almost negligible, however the measured and calculated demagnetizing factors $I_{xx}^{th}(int)$ of interacting wires, show a marked decrease from the $I_{xx}\approx 1$. As the critical value of the array's demagnetizing factor is $\frac{2}{3}$ at the critical radius R_c=20 nm, the RPT is expected to take place in between samples SU02 and SU03, because of interwire interaction. As shown in Table 1, the RPT is experimentally observed close to the array SU03 [3]. According to the

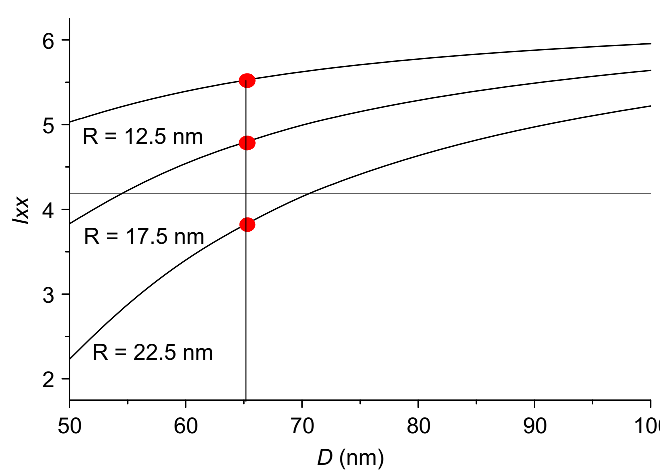

Fig. 2. The effective demagnetizing factor I_{xx} of a two-dimensional hexagonal long wire array, depending on the periodicity D of the array, for 3 different wire radii, R (see Table 1). The horizontal line indicates the critical value of I_{xx}=4π/3 when the RPT phase transition of the magnetic moments from the out-of-plane to the in-plane direction takes place. Three (red) dots on the vertical line indicate the calculated I_{xx} for the experimentally measured samples (see also Table 1). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)