Modelling and Measurement of Remanence Enhancement in Two Phase Nanocrystalline Materials

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In this study the exchange interaction between a hard magnetic phase and a soft magnetic phase has been numerically modelled using a two dimensional array of uniaxially anisotropic micromagnetic moments. Hysteresis curves, calculated by minimising the total energy of the array, show that the coercivity and the enhancement of magnetization depend on the dimension of the soft magnetic phase. The results of the numerical study are shown to be consistent with measurements in a nanocrystalline two phase α-Fe/$Sm_2Fe_{17}N_{2.6}$ alloy.

1. INTRODUCTION

In the absence of intercrystalline exchange coupling the remanent magnetization, M_r, of a crystallographically isotropic array of grains should equal one half of the spontaneous magnetization, M_s, for materials exhibiting uniaxial magnetocrystalline anisotropy. Recent studies [1-6] have reported signficantly higher values of M_r in nanocrystalline two phase mixtures of magnetically hard and soft phases associated with exchange coupling across the interfaces between the phases. Coehoorn et al. [1] reported remanence values of 0.7-0.8 M_s in melt spun ribbons of NdFeB containing approximately 85% Fe_3B. In their investigations of nanocrystalline two phase mixtures prepared by mechanical alloying Ding et al. [3-6] reported M_r values of up to 0.8 M_s in α-Fe/$Sm_2Fe_{17}N_{2.6}$ and Sm_2Co_{17}/$SmCo_5$ alloys. Maximum energy products of up to 26 MGOe were reported in isotropic two phase α-Fe/$Sm_2Fe_{17}N_{2.6}$ alloys containing approximately 40 vol% α-Fe [3].

These studies and others [7] have shown that remanence enhancement in exchange coupled nanocrystalline materials is a potentially important mechanism for achieving high values of $(BH)_{max}$ in isotropic magnets since elimination of the need for crystallographic alignment should substantially reduce manufacturing costs. Recent developments in micromagnetism and numerical modelling [8-9] provide a basis for modelling the effect of crystal size and other microstructural features on the magnetic behaviour of exchange coupled nanocrystalline grains. In this paper we report initial results of a numerical study of the effect of crystal size on magnetization behaviour using a two dimensional crystal model.

2. MODELLING PROCEDURE

A two dimensional array of micromagnetic moments is used to represent a nanostructure magnetic material on an atomic scale. Associated with each moment of magnitude $m_s[i,j]$ and orientation $\theta[i,j]$ is a coefficient of magnetocrystalline anisotropy (assumed to be uniaxial) $k[i,j]$ and an easy direction of magnetization with respect to the the applied field $\theta_0[i,j]$. The number of neighbouring elements with common values of m_s, k and θ_0 represents the size of a phase. The energy at each site in the array, $E[i,j]$, can be expressed as the sum of magnetostatic, demagnetizing, magnetocrystalline and exchange energies. In this study the component due to demagnetizing fields, which is relatively small in nanoscale systems [10], has not been included. Thus for a given applied field, H,

$$E[i,j] = \mu_o m_s[i,j] H \cos(\theta)$$
$$+ k[i,j]\sin^2(\theta - \theta_o)$$
$$+ \frac{J_{ex}}{2d^3}\left\{\begin{array}{l}\cos(\theta[i-1,j]-\theta[i,j]) \\ +\cos(\theta[i,j]-\theta[i+1,j]) \\ +\cos(\theta[i,j-1]-\theta[i,j]) \\ +\cos(\theta[i,j]-\theta[i,j+1])\end{array}\right.$$

where J_{ex} is the exchange constant between any pair of nearest neighbour moments and d^3 is the unit volume. The equilibrium configuration of the system of moments for a given applied field is established by using an iterative solution technique to find the minimum total energy, $E_{tot} = \Sigma E[i,j]$.

3. RESULTS AND DISCUSSION

The numerical modelling results presented in this paper are for a two phase material comprising a uniform distribution of hard magnetic grains in a soft magnetic matrix, as shown in Figure 1. The easy direction of magnetization of the hard magnetic phase is aligned with the field, $\theta_0 = 0$, and for the matrix phase $\theta_0 = \pi/4$. Values of the saturation magnetization and first order anisotropy constant of $Nd_2Fe_{14}B$ and α-Fe have been used for k and m_s of the hard and soft magnetic phase respectively. The lattice dimension is assumed to be d = 3Å.
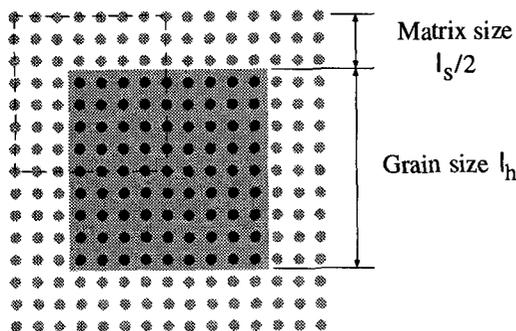
Figure 1.   Schematic diagram of the model.
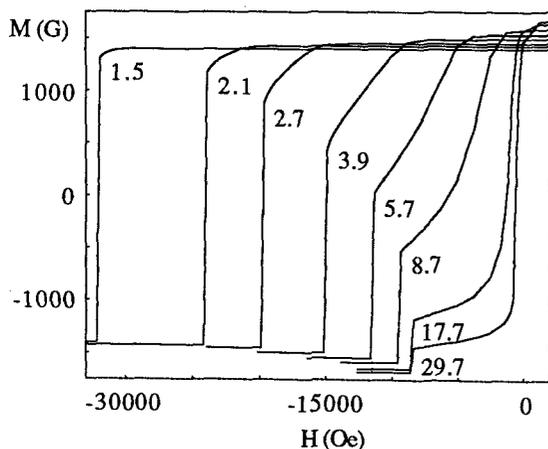
Figure 2.   Demagnetization curves for a constant size of grains of the hard magnetic phase ℓ_h = 10 nm and a range of values for the minimum dimension in the soft magnetic matrix ℓ_s from 1.5 nm to 29.7 nm.

In Figure 2 the effect of the minimum dimension of the soft magnetic phase on the calculated demagnetization curves for a constant size of the hard magnetic phase is shown. The demagnetization process is reversible until a critical field, the coercivity, H_c, at which point all the moments flip irreversibly. The dependence of coercivity on the size of the soft magnetic phase for a constant size of the hard magnetic phase is shown in Figure 3. The largest coercivity is achieved in the system of smallest dimension. For this size all the moments of the soft phase remain approximately aligned with the hard magnetic phase in fields many times greater than the anisotropy field of the soft magnetic phase, $H_{k(soft)}/H_{k(hard)}$ = 0.006. As the dimension of the matrix increases, so does the extent of rotation of soft magnetic moments further from the hard magnetic phase. Coercivity decreases because of the resulting nonuniform magnetization at the interface between the phases where the magnetic moments of the hard phase are disturbed from their easy direction of magnetization. For sufficiently large dimensions of the matrix there is no significant increase in the size of the interfacial region in which the magnetization is nonuniform such that coercivity remains approximately constant.
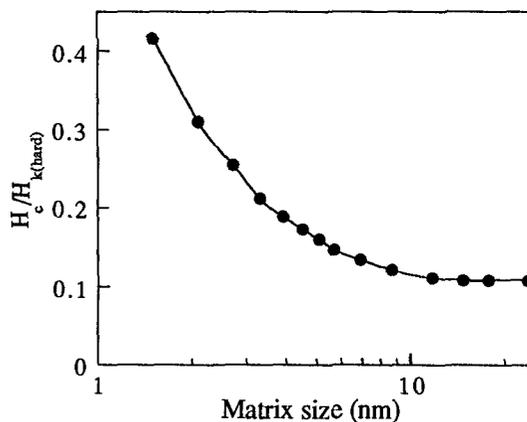
Figure 3.   Coercive field, H_c, normalised by the anisotropy field of the hard phase, $H_{k(hard)}$ = $2k/\mu_0 m_s$, as a function of the minimum dimension of the soft magnetic phase.

Evolution of magnetization reversal in the the soft magnetic phase is illustrated in Figure 4. At zero field, represented by Figure 4a,
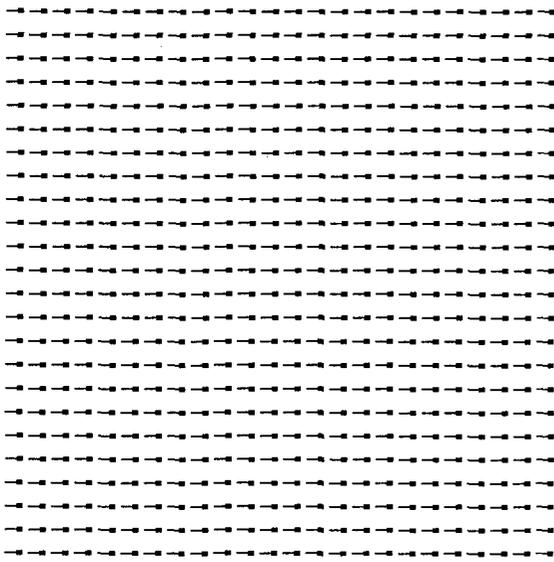
a) $H/H_{k(hard)} = 0$; $M/M_{sat} = 1.0$
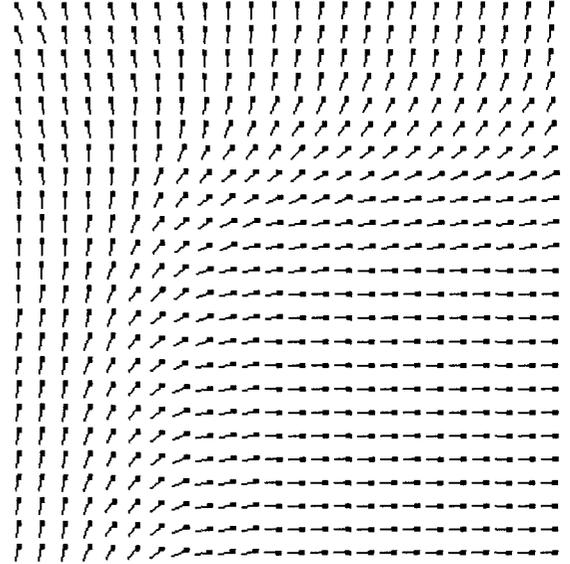
b) $H/H_{k(hard)} = -0.16$; $M/M_{sat} = 0.6$
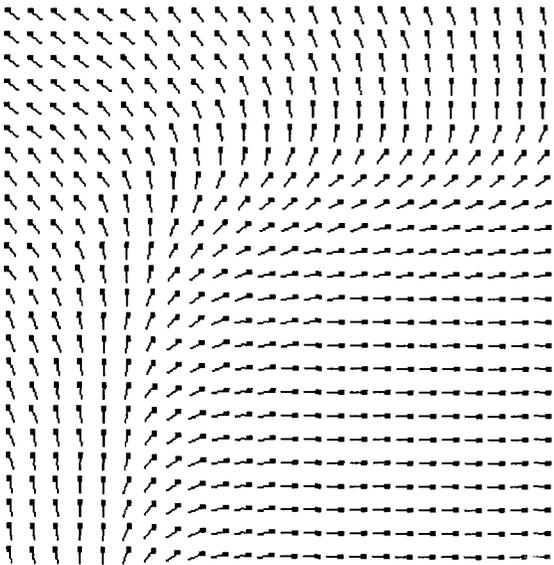
c) $H_c/H_{k(hard)} = -0.19$; $M/M_{sat} = 0.3$
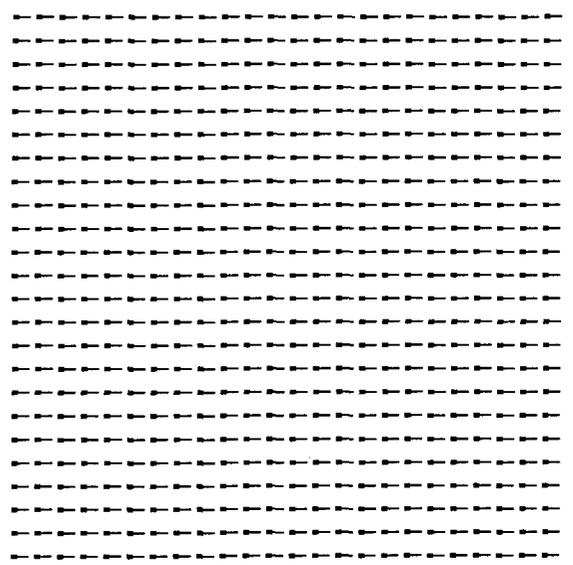
d) $H < H_c$; $M/M_{sat} = -1.0$

Figure 4.   Distribution of the micromagnetic moments at a) remanence, b) a field between remanence and coercivity, c) coercivity and d) saturation for a material with grain size $\ell_h = 10$ nm and minimum matrix dimension $\ell_s = 4$ nm. The corresponding fraction of the hard magnetic phase is 0.5. (Note that only one quarter of the array is displayed.)

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the exchange coupling with the hard magnetic phase causes all the soft magnetic moments, which would otherwise be orientated at $\pi/4$, to lie along the easy direction of magnetization of the hard magnetic phase. Figure 4b corresponds to a field at which the change in magnetization is midway between that at remanence and coercivity. The change in the magnetization that has occurred can be seen to be due largely to rotation in the soft magnetic phase. In the hard magnetic phase, only the moments at the interface have rotated out of their easy direction of magnetization. Figure 4c is a configuration just prior to coercivity. Thereafter the moments flip and the system is saturated in the reverse direction, as depicted in Figure 4d.

independently such that there is a large change in magnetization in a small field due to rotation in the soft phase. In contrast to the results of the numerical model in Figure 2, the approach to saturation for these systems with a large soft magnetic phase is gradual because there is a distribution of easy axes of magnetization.

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Figure 5. Demagnetization curves measured in isotropic Sm_9Fe_{91} - nitride (approximately 30 vol.% Fe) with a constant size of the hard magnetic phase 10-20 nm and a range of sizes of the soft magnetic phase.

Experimental measurements of the effect of crystallite size on the magnetization curves in two phase $Sm_2Fe_{17}N_x/\alpha$-Fe samples are shown in Figure 5. The measurements show the type of dependence on the size of the soft magnetic phase that is predicted by the modelling work. The largest coercivity is exhibited by the material with the smallest soft magnetic phase. In this material the magnetization is enhanced even in relatively large fields because the soft phase remains approximately aligned with the hard phase until the system becomes unstable and the magnetization changes irreversibly. As the size of the soft phase increases, the hard and soft magnetic phases tend to behave