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NUCLEATION FIELD AND ENERGY PRODUCT OF ALIGNED TWO-PHASE MAGNETS - PROGRESS TOWARDS THE '1 MJ/m^3' MAGNET

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Abstract **- Exchange hardning of nanostructured two-phase systems composed of an aligned hard phase and a soft phase with high magnetization is investigated using a micromagnetic approach which accounts for intereactions between the soft regions. For $Sm_2Fe_{17}N_3$(2.5nm)/$Fe_{65}Co_{35}$(9nm) multilayers an energy product as high as 1 MJ/m^3 (120 MGOe) is predicted, with a rare-earth content of only 5wt%. Giant energy products may also be achieved in suitable cellular and disordered structures.**

INTRODUCTION

Due to the quadratic dependence of the theoretical energy product $(BH)_{max}$ on the saturation magnetization M_o, magnetic phases such as bcc iron with $\mu_o M_o = 2.15$ T should be excellent permanent magnetic materials. In fact, magnetocrystalline anisotropy and coercivity of bcc iron are very low so energy products of iron magnets are only of order 1 kJ/m^3, compared to the theoretical value $\mu_o M_o^2/4 = 920$ kJ/m^3. In the past it was necessary to ressort to cumbersome horseshoe shapes to avoid spontaneous demagnetization into a multidomain state by the magnet's own magnetostatic field.

Modern high-performance magnets such as $Nd_2Fe_{14}B$ [1] and $Sm_2Fe_{17}N_3$ [2] overcome this problem by exchange coupling iron atoms to rare-earth atoms in sites with strong uniaxial anisotropy. The penalty, however, is a reduced magnetization due to the rare-earth and nonmagnetic elements. Additionally, there is nothing to be gained from a coercivity much greater than M_o: the energy product cannot exceed $\mu_o M_o^2/4$, and the strong uniaxial is partly wasted. Nevertheless, it has been possible to use $Nd_2Fe_{14}B$ which has $\mu_o M_o = 1.61$ T and $\mu_o M_o^2/4 = 516$ kJ/m^3 to achieve energy products as high as 405 kJ/m^3 in laboratory-scale magnets [1].

The outlook for discovering new ternary phases with significantly higher magnetization than those available at present is poor. Interstitial modification with small atoms such as nitrogen or carbon is effective for enhancing Curie temperature and anisotropy, but the magnetization remains practically unchanged [2]. A new approach is needed if further progress is to be possible.

A possible way is to use nanocrystalline two-phase materials consisting of exchange-coupled hard-magnetic and soft-magnetic phases where the hard-magnetic phase assures the necessary coercivity. Recently enhancement of the comparatively low remanence $M_r = M_o/2$ has been achieved in the isotropic nanocrystalline composites $Nd_2Fe_{14}B/Fe_3B$ and $Sm_2Fe_{17}N_3$/Fe produced by melt-spinning [3] and mechanical alloying [4], respectively. In these systems the energy product is improved by exchange hardning [5-7] but does not reach the level attained in oriented rare-earth magnets. To obtain energy products greater than those in aligned rare-earth intermetallics, *aligned* two-phase magnets have to be used.

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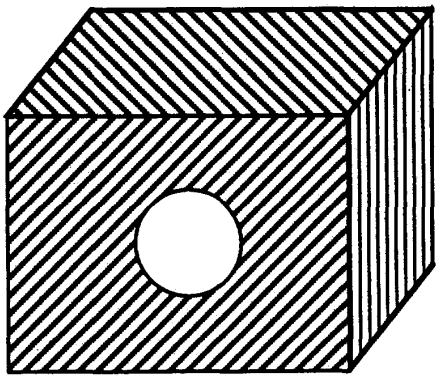

Fig.1. Spherical soft inclusion (bcc iron) in an aligned hard-magnetic matrix ($Sm_2Fe_{17}N_3$).

Here we determine nucleation fields in three-dimensional aligned two-phase magnets and calculate the maximum energy product in suitable nanostructured composites.

MODEL AND RESULTS

Micromagnetic background

Assuming short-range exchange interaction and uniaxial anisotropy, the magnetic free energy can be written as (cf. eg. [8,9])

$$F = \int \left[A(r) \left(\frac{\nabla M_i}{M_o(r)}\right)^2 - K_1(r) \left(\frac{M_i n_i}{M_o(r)}\right)^2 - \mu_o M_i H_i \right] dr$$

$$- \frac{1}{2} \sum_{ij=1}^{3} \int K_{ij}(r-r') M_i(r) M_j(r') \, dr dr' \qquad (1)$$

where $A(r)$ is the exchange stiffness, $M_i(r)$ with $|M_i(r)| = M_o$ denotes the local magnetization, and $K_1(r)$ is the first anisotropy constant. The spacially constant unit vector n_i of the easy-axis direction and the external field H_i are assumed to be parallel (aligned magnet), and the nonlocal kernel $K_{ij}(r-r')$, describes the magnetostatic dipole interaction.

If we start from the perfectly aligned state where $M_i(r) = M_o(r) n_i$, a sufficiently-high external nucleation field $H_i = -$

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$H_N n_i$ is necessary to destabilize the aligned state and to induce magnetic reversal (nucleation). Nucleation is a necessary but not sufficient condition for magnetic reversal, since there is a possibility that the reversed nucleus will not propagate.

In sufficiently large *homogeneous* ellipsoids nucleation is realized by incoherent magnetostatic modes, eg. curling in the case of a sphere. The corresponding nucleation field obeys $H_N \geq 2\,K_1/\mu_o M_o - DM_o$, where D is the macroscopic demagnetization factor of the ellipsoid [10,11]. This means that the so-called intrinsic coercivity $H_c = H_c^{ex} - DM_o$ can not be smaller than the 'much-too-high' value $2K_1/\mu_o M_o$, which is known as Brown's paradox. In fact, real systems always show a certain inhomogenity which gives, at least principially, the solution of Brown's paradox [11].

Coercivity and energy product

In the following we neglect the possible improvement of the coercivity due to pinning effects and restrict ourselves to the determination of H_N. Substituting the identity

$$M_i(r) = M_o(r)\left(m_x(r)\,e_{ix} + m_y(r)\,e_{iy} + \sqrt{1-m_x^2-m_y^2}\,e_{iz}\right) \quad (2)$$

into Eq. (1) and expanding the free energy density with respect to the small transverse components $m_i = \{m_x, m_y\} \ll 1$ yields a quadratic form whose lowest eigenvalue corresponds to the nucleation field. Two cases can be distinguished:

(i) The coercivity of modern rare-earth magnets is small compared to their large anisotropy field H_A, which cannot be explained by magnetostatic fields. Neglecting magnetostatic contributions ($M_o \ll H_A - H_c$) and spatial variations of A and M_o, the micromagnetic vector equation obtained from Eq. 1 reads

$$-A\,\nabla^2 m_i + K_1(r)\,m_i = \frac{1}{2}\mu_o M_o H_N\,m_i \quad (3)$$

The two components m_i are decoupled so Eq. (3) corresponds to Schrödinger's equation for a particle moving in a three-dimensional potential $K_1(r)$. This allows us to apply ideas familiar from quantum mechanics to discuss micromagnetics; in particular, the nucleation field H_N corresponds to the quantum-mechanical ground-state energy, and the small transverse magnetization or nucleation mode has its analog in the the wave function. It turns out that the nucleation field calculated from Eq. (3) is extremely low if the material contains soft regions (cf. Fig. 1) whose size is much larger than the Bloch-wall width δ_B of the hard matrix [9,11-14]. Fig. 2 shows the calculated nucleation field for the spherical inclusion Fig. 1 [14].

(ii) With increasing number of inhomogenities the interaction between different soft inclusions becomes important. This interaction has its quantum- mechanical analog in the formation of bonding and antibonding states and tends to reduce the nucleation field. The determination of H_N now becomes difficult, but perturbation theory can be used for macroscopically homogeneous magnets if the soft inclusions are very small (plateau region in Fig. 2). In this case the exchange coupling between hard and soft regions is very

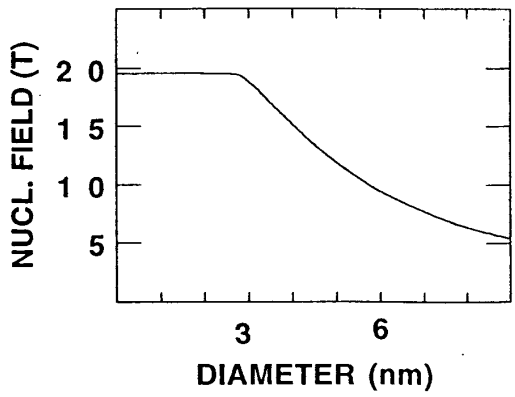

Fig.2. Nucleation field as function of 2R, the diameter of the inclusion in Fig. 1. The assumed values are : $\mu_o M_s = 2.15$ T, $\mu_o M_h = 1.55$ T, $A_s/A_h = 1.5$, $K_{Fe} = 0$, and $K_1 = 12$ MJ/m^3.

strong which leads to a more or less homogeneous magnetization state. In lowest order perturbation theory [14], the unperturbed nucleation mode, i.e. the unperturbed wave function within the quantum-mechanical analogy, can be used to calculate the nucleation field.

$$H_N = \frac{2\langle K_1\rangle}{\mu_o\langle M_o\rangle} - D\,\langle M_o\rangle \quad (4)$$

with $\langle M_o\rangle = f_h M_h + f_s M_s$. Note that this result does not depend on the shape of the inclusions.

If the saturation magnetization M_s of the soft phase is higher than the saturation magnetization M_h of the hard phase, and so long as the coercivity remains sufficiently high, the energy product of the two-phase system will be greater than that of the hard phase. The theoretical limit is given by

$$(BH)_{max} = \frac{1}{4}\,\mu_o\langle M_o\rangle^2\left(1 - \frac{\mu_o(M_s-M_h)M_s}{2K_1}\right) \quad (5)$$

where K_1 is the anisotropy constant of the hard phase. Due to the large K_1, the second term in the bracket is small so the energy product approaches the ultimate value $\mu_o\langle M_o\rangle^2/4$. The corresponding volume fraction of the hard phase is $f_h = \mu_o\langle M_o\rangle^2/4K_1$. If we consider the $Sm_2Fe_{17}N_3$/Fe system and take values $\mu_o M_s = 2.15$ T, $\mu_o M_h = 1.55$ T, and $K_1 = 12$ MJ/m^3 we obtain a theoretical energy product of 880 kJ/m^3 (110 MGOe) for a volume fraction of only 8% of the hard phase. A further increase of the energy product is possible, if iron is replaced by $Fe_{65}Co_{35}$ with $\mu_o M_s = 2.35$ T: $(BH)_{max}$ = 1090 kJ/m^3 (137 MGOe). It is remarkable that these magnets consist almost entirely of 3d metals, with only about 2% samarium.

CONCLUSIONS

Eq. (5) shows that the energy product in aligned two-phase magnets is much larger than that of the corresponding

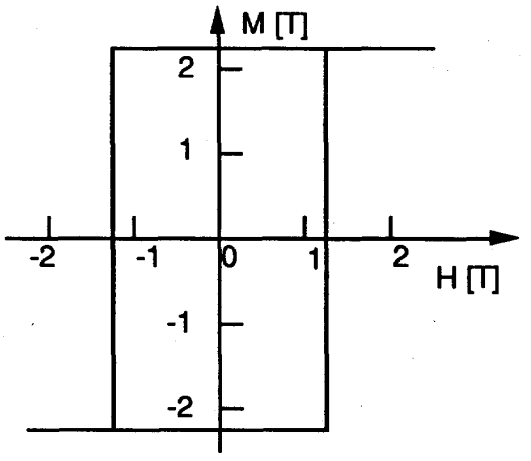

Fig. 3. Schematic hysteresis loop of a hypothetical 'MJ' magnet.

rare-earth intermetallics if the hard regions act as a skeleton to stiffen the magnetization direction of the soft regions. The practical problem however is to realize a structure where the soft regions are sufficiently small to avoid nucleation at small fields, and to have the hard regions crystallographically aligned. One conceivable solution is a disordered two-phase magnet (Fig. 4) with a common c-axis throughout the hard regions, but it is difficult to see how this might be achieved in practice.

A more realistic possibility is a multilayered structure of alternating soft and hard magnetic layers (Fig. 5). Using a slightly different theoretical approach (cf. [14,15]), an energy product of about 1000 kJ/m^3 is obtained for a multilayer composed of alternating 2.5 nm $Sm_2Fe_{17}N_3$ and 9.0 nm $Fe_{65}Co_{35}$ layers. (This structure contains 5 wt.% samarium).

Eq. (1) is based on classical micromagnetic considerations; the sizes of the hard and soft regions must be large compared to atomic dimensions (larger than about 1 nm). To compensate this size effect, and to ensure reproduceable magnetic properties, the magnet must be stabilized by slightly increasing the volume fraction of the

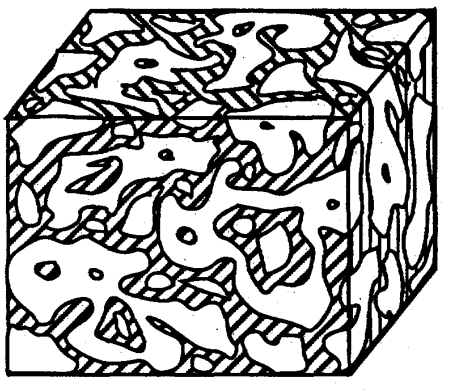

Fig.4. An oriented disordered two-phase magnet with common c axis. The size of the soft and hard regions is smaller than the Bloch wall thickness δ_B of the hard phase.

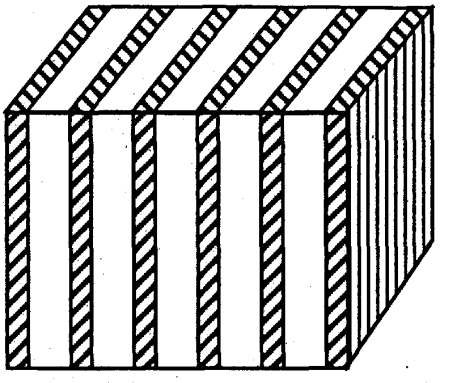

Fig.5. A multilayer structure composed of alternating hard and soft magnetic regions. Note that the exchange coupling does not depend on the c axis direction (in-plane or perpendicular).

hard phase. Nevertheless, f_h = 30 % still yields an energy product of 800 kJ/m^3 (100 MGOe) in the $Sm_2Fe_{17}N_3$/Fe system.

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