

# Exchange Coupling and Energy Product in Random Two-Phase Aligned Magnets

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**Abstract** - Size effects in two-phase Boolean magnets composed of an aligned hard phase and a soft phase with high magnetization are investigated. In 'overcritical' magnets, where the size of the soft regions is larger than the Bloch-wall width  $\delta_h$  of the hard phase, the energy product is reduced compared to the corresponding 'undercritical' magnets with small soft regions, where theoretical energy products as high as  $1 \text{ MJ/m}^3$  have been predicted. Nevertheless, the theoretical energy product only gradually drops with increasing size of the soft regions, and it may still exceed the theoretical energy product  $516 \text{ kJ/m}^3$  of  $\text{Nd}_2\text{Fe}_{14}\text{B}$ .

## I. INTRODUCTION

The energy product  $(BH)_{\max}$ , which gives the maximum magnetostatic energy available from a magnet, is a key figure of merit for permanent magnetic materials. The energy product increases with the coercivity but never exceeds its theoretical value  $\mu_0 M_0^2/4$ . The theoretical energy product of bcc iron is  $\mu_0 M_0^2/4 = 920 \text{ kJ/m}^3$ , but in fact pure iron exhibits nearly no coercivity so the large theoretical energy product is far from being realized in practice. In modern permanent magnetic alloys such as  $\text{Nd}_2\text{Fe}_{14}\text{B}$  [1] and  $\text{Sm}_2\text{Fe}_{17}\text{N}_3$  [2], the saturation magnetization does not reach the high value  $\mu_0 M_0 = 2.15 \text{ T}$  of iron but the high uniaxial anisotropy makes them useful permanent magnets. At present, energy products as high as  $405 \text{ kJ/m}^3$  are obtained in laboratory-scale  $\text{Nd}_2\text{Fe}_{14}\text{B}$  magnets [1].

The outlook for discovering new ternary phases with significantly higher magnetization than those available at present is poor [3], but as it has been shown recently [4], exchange coupling in nanostructured aligned two phase magnets consisting of alloys available at present yields theoretical energy products exceeding that of  $\text{Nd}_2\text{Fe}_{14}\text{B}$  ('giant energy product'). The nucleation field, which determines the energy-product in nucleation-controlled magnets, is calculated from the magnetic free energy integral [5] - [8]

$$F = \int \left[ A(\mathbf{r}) \left( \frac{\nabla M_i}{M_0(\mathbf{r})} \right)^2 - K_1(\mathbf{r}) \left( \frac{M_i n_i}{M_0(\mathbf{r})} \right)^2 - \mu_0 M_i H_i \right] d\mathbf{r} \\ - \frac{1}{2} \sum_{i,j=1}^3 \int \mathbb{K}_{ij}(\mathbf{r}-\mathbf{r}') M_i(\mathbf{r}) M_j(\mathbf{r}') d\mathbf{r} d\mathbf{r}' \quad (1)$$

where  $A(\mathbf{r})$  is the exchange stiffness, the  $M_i(\mathbf{r})$  with  $|M(\mathbf{r})| = M_0$  denote the components of the local magnetization, and  $K_1(\mathbf{r})$  the first anisotropy constant.  $H_i = e_{zi} H_z$  denotes the external magnetic field,  $n_i$  is the unit vector of the easy-axis direction, and the nonlocal kernel  $\mathbb{K}_{ij}(\mathbf{r}-\mathbf{r}')$  describes the magnetostatic dipole interaction.

The basic assumptions made in [4] are:

(i) The size of the soft regions is smaller than or comparable to the domain-wall width of the hard phase. This leads to an exchange coupling between hard and soft regions which is comparable to the well-known remanence enhancement in isotropic two-phase magnets (cf. [4], [9]).

(ii) The hard regions are aligned ( $n_i = e_{zi}$ ) so the exchange coupling leads to an effective increase in remanence magnetization and energy product.

(iii) The first order anisotropy constant  $K_h$  of the hard phase is much larger than magnetostatic energy density of the soft phase.

The upper limit of the theoretical energy product is [4]

$$(BH)_{\max} = \frac{1}{4} \mu_0 \langle M_0 \rangle^2 \left( 1 - \frac{\mu_0 (M_s - M_h) M_s}{2K_h} \right) \quad (2)$$

where  $M_s$  and  $M_h$  denote the magnetizations of the soft and hard phases, respectively [4], and  $\langle M_0 \rangle = f_s M_s + f_h M_h$  the average magnetization of the magnet. For instance, Eq. (2) predicts an energy product as high as  $1090 \text{ kJ/m}^3$  for the system  $\text{Sm}_2\text{Fe}_{17}/\text{Fe}_{65}\text{Co}_{35}$ , with a volume fraction of the hard phase  $f_h$  of only 11 vol. %.

However, the production of magnets structured on a scale smaller than the Bloch-wall width  $\delta_h$  of the hard phase is a difficult problem, and in practice it might be suitable to resort to magnets where the soft regions are larger than  $\delta_h$ . It is well known that there is no closed general solution for the corresponding micromagnetic problem [5] - [8], and only for a few configurations, such as homogeneous ellipsoids, the nucleation problem can be solved analytically [7]. Here we investigate the effect of exchange-stiffness and anisotropy inhomogeneities, and random magnetic stray fields to predict the behaviour of magnets structured on a overcritical scale.

## II. RESULTS AND DISCUSSION

### Random stray fields

From the theoretical point of view, the magnetostatic interaction is the most difficult in (1) to deal with. This is due to the non-local kernel

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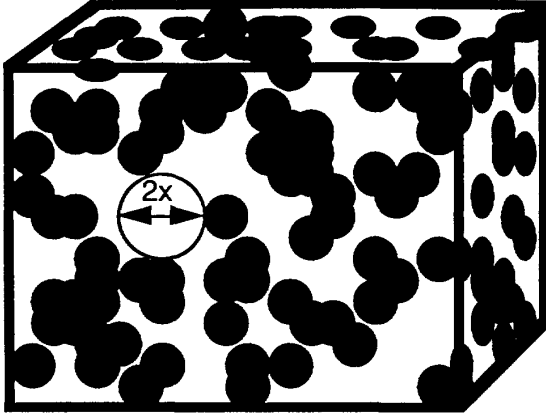


Fig. 1. Boolean model describing the structure of disordered two-phase magnets. The position of the spheres (hard regions) is random, but many geometrical properties, such as the typical distance  $x$  to the next sphere, are given analytically [15].

$$\mathbb{K}_{ij}(\mathbf{r}) = \frac{\mu_0}{4\pi} \frac{3 r_i r_j - \delta_{ij} r^2}{r^5} \quad (3)$$

which describes the long-range dipole interaction between different magnetic regions. A simple but nevertheless non-trivial approximation is to replace  $H_z$  in (1) by  $H_z - D\langle M_0 \rangle$ , where  $D$  is the macroscopic demagnetization factor of the ellipsoid.

In disordered aligned two-phase magnets (cf. Fig. 1) there are random stray fields which are incompatible with the assumption of a homogeneous demagnetizing field. These stray-field inhomogeneities are harmful to coercivity if they cover a volume larger than about  $\delta_h^3$ . Smaller stray-field inhomogeneities are ineffective: exchange coupling prevents regions smaller than  $\delta_h$  from being switched even if they are subject to a strong demagnetizing field.

The local stray fields are given by

$$\mu_0 H_i^L(\mathbf{r}) = \int \sum_{j=1}^3 \mathbb{K}_{ij}(\mathbf{r}-\mathbf{r}') M_j(\mathbf{r}') d\mathbf{r}' \quad (4)$$

where the integration includes all points inside a sufficiently large sphere. Note that the angular integration  $\sin\theta d\theta$  yields zero for all contributions spherical around  $\mathbf{r}$ . In particular,  $H_i^L(\mathbf{r}) = \langle H_i^L(\mathbf{r}) \rangle = 0$  for  $M_i(\mathbf{r}) = \langle M_i(\mathbf{r}) \rangle$ , where  $\langle \dots \rangle$  denotes the volume average.

Situation changes if the variation of  $H_i^L(\mathbf{r})$  is considered. To calculate  $\langle [H_i^L(\mathbf{r})]^2 \rangle = \langle \delta H_{i||} \rangle^2 + 2 \langle \delta H_{i\perp} \rangle^2$ , we have to know the correlation function  $\langle M_o(\mathbf{r}) M_o(\mathbf{r}') \rangle$  which will vary from sample to sample. A reasonable, since largely uncorrelated, ansatz is

$$\langle M_o(\mathbf{r}) M_o(\mathbf{r}') \rangle = \langle M_o \rangle^2 + V_o \delta(\mathbf{r}-\mathbf{r}') [\langle M_o^2 \rangle - \langle M_o \rangle^2] \quad (5)$$

describing inhomogeneities whose size (correlation length) is of order  $V_o^{1/3}$ . Calculation yields

$$\langle \delta H_{i||} \rangle^2 = \frac{4}{15} [\langle M_o^2 \rangle - \langle M_o \rangle^2] \quad (6)$$

and

$$\langle \delta H_{i\perp} \rangle^2 = \frac{1}{5} [\langle M_o^2 \rangle - \langle M_o \rangle^2] \quad (7)$$

For the  $\text{Sm}_2\text{Fe}_{17}/\text{Fe}_{65}\text{Co}_{35}$  system we predict  $\mu_0 \delta H_{i||} = 0.07$  T. This means that random stray fields in typical aligned two-phase magnets hardly exceed a few tenth of a Tesla, and a small compensating increase of the volume fraction  $f_h$  of the hard phase will be sufficient to re-establish the necessary coercivity.

#### Exchange stiffness and Inhomogeneous Anisotropy

We now neglect random stray fields in (1) and expand the free energy with respect to the small transverse components  $m_x = M_x/M_o \ll 1$  and  $m_y = M_y/M_o \ll 1$ :

$$F = F_o + \int [A(\mathbf{r}) (\nabla m_i)^2 + K_1(\mathbf{r}) m_i^2 - \frac{1}{2} \mu_0 m_i^2 M_o H_z] d\mathbf{r} \quad (8)$$

Next we have to calculate the free-energy minimum and to look whether the equilibrium is stable or not. To minimize  $F = \int \eta d\mathbf{r}$  we use the formula

$$\frac{\delta F}{\delta m_i} = -\nabla \left( \frac{\partial \eta}{\partial \nabla m_i} \right) + \frac{\partial \eta}{\partial m_i} = 0 \quad (9)$$

where  $\delta F/\delta m_i(\mathbf{r})$  denotes the functional derivative (see e.g. [10]) and obtain the eigenvalue problem

$$-\nabla [A(\mathbf{r}) \nabla m_i] + K_1(\mathbf{r}) m_i = \frac{1}{2} \mu_0 M_o H_N m_i \quad (10)$$

with the nucleation field  $H_N$ . Note that the two components  $m_i$  are decoupled in demagnetizing-field approximation.

The  $\nabla A(\mathbf{r})$ -contribution in (10) is due the fact that the magnetization in magnets with large exchange stiffness tends to be more homogeneous than in those with a lower exchange stiffness. However,  $A$  is of order  $10^{-11}$  J/m in both phases so the  $\nabla A(\mathbf{r})$ -term, though included in [4], does not change the qualitative behaviour of the system. In particular, it does not affect the energy product in overcritical two-phase magnets.

The  $K_1(\mathbf{r})$  term in (10) describes the effect of a soft phase, where  $K_1(\mathbf{r}) = K_s \approx 0$ , on the nucleation field  $H_N = -H_z$ . To calculate the nucleation field, we use the similarity between Eq. (10) with  $\nabla A(\mathbf{r}) = 0$  and Schrödinger's equation for a particle moving in a three-dimensional potential  $U(\mathbf{r}) = K_1(\mathbf{r})$  (cf. [4]). Up to second order perturbation theory

$$H_N = \frac{2\langle K_1(\mathbf{r}) \rangle}{\mu_0 M_o} - \frac{2}{\mu_0 M_o A V} \sum_{\mathbf{k}} (k^2)^{-2} \int_V e^{i\mathbf{k}\mathbf{r}} \langle K_1(\mathbf{r}) K_1(0) \rangle d\mathbf{r} \quad (11)$$

where the  $\mathbf{k}$  summation includes all plane waves  $\exp(i\mathbf{k}\mathbf{r})$ . It can be shown that this assumption is reasonable for macros-

copically large magnets, where modes such as curling are curling are negligible (see e.g. [3] and [5] - [8]).

The linear term in (11) corresponds to the introduction of a volume-averaged anisotropy constant  $K_{\text{eff}}(\mathbf{r}) = \langle K_1(\mathbf{r}) \rangle$  and reproduces the energy product in (2). As can be shown by using the quantum-mechanical analogy, the quadratic term in (11) always reduces  $H_N$ , so this equation may serve as a simple illustration of Brown's paradox. As opposed to the first-order correcture, which does not depend explicitly on the geometrical form of the soft and hard regions, the second order contribution involves the correlation function  $\langle K_1(\mathbf{r})K_1(0) \rangle$ . This correlation function strongly depends on the real structure of the magnet, and it is not possible any longer to obtain closed expressions such as (2).

Equation (11) gives the first two terms of a perturbation series. The small parameter of this expansion is  $s^2/\delta_h^2$ , where  $s$  is the correlation length of the inhomogeneities, i.e. roughly speaking the size of the hard and soft regions. For small  $s$  a 'plateau' behaviour [4] is obtained, whereas the series (11) breaks down for large correlation lengths (for other approaches see e.g. [11] - [14]).

Compared to random stray fields and exchange-stiffness inhomogeneities, large soft regions are much more harmful to coercivity and energy product. To discuss this effect we use a Boolean model [15] where overlapping randomly distributed spheres with radius  $R$  serve to model the hard regions Fig. 1. For this model the probability  $p(x > L)$  that the distance  $x$  between a typical soft-phase point and the closest hard-phase point is larger than  $L$  [15]

$$p(x > L) = (1 - f_h) (1 + L/R)^3 - 1 \quad (12)$$

To remain in the plateau region we have to choose  $L = \delta_h/2 = 1.5$  nm. On the other hand, the size of the hard regions cannot be smaller than about 1 nm [4], so we obtain with the typical value  $f_h = 11$  % the probability  $p(x > 1.5 \text{ nm}) = 0.06$  %. This means that less than one out of 1000 soft-phase atoms are in a too-large region, which makes the production of macroscopically large magnets with totally random distribution of the hard phase unrealistic. However, a small increase of the fraction of the hard phase strongly improves the prospects for making a reliable magnet: taking  $f_h = 20$  % instead of  $f_h = 10$  % reduces the energy product of the  $\text{Sm}_2\text{Fe}_{17}/\text{Fe}$  system by only 6 %, while  $p(x > 1.5 \text{ nm}) = 7.8 \cdot 10^{-5}$  %. A similar effect is achieved by leaving the plateau region, which must be compensated by increasing  $f_h$ , and structuring the magnet on a somewhat larger scale. Note that these difficulties do not occur if the hard regions are more regularly dispersed.

### III. CONCLUSIONS

Disordered two-phase magnets with a common  $c$  axis throughout the hard regions structured on an 'overcritical' scale are subject to random stray fields which reduce the coercivity by a few tenth of a Tesla. More harmful to coercivity are fluctuations of the size of the soft regions, which occur in particular in totally random structures. Using a Boolean model we have calculated nucleation probabilities which show that increasing the amount of the hard phase strongly improves the skeleton performance of the hard phase

in 'overcritical' magnets. This stabilization is paid by a slight decrease in saturation magnetization, so the ultimate energy product  $1090 \text{ kJ/m}^3$  can be approached but not reaches in 'overcritical' magnets.

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