

# Spin-glass permanent magnets

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## Abstract

The ground state of random-anisotropy permanent magnets, such as isotropic nanostructured  $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ , is investigated. The degeneracy of the ground-state of strongly disordered random-anisotropy magnets leads to finite-range ferromagnetic ground-state correlations and ferromagnetic configurations whose energies are comparable to the ground-state energy. This explains why the coercivity of typical rare-earth containing random-anisotropy spin glasses exceeds that of industrial ceramic magnets by more than one order of magnitude. By comparison, random-field spin glasses are unsuitable for permanent magnetic applications.

**Keywords:** Random anisotropy; Spin glasses; Permanent magnetism; Coercivity

Disordered magnets such as  $\text{CuMn}$ ,  $\text{GdAl}$  and  $\text{FeO}(\text{OH}) \cdot n\text{H}_2\text{O}$  are a subject of major scientific interest and have given rise to countless experimental and theoretical studies [1–4]. From the atomic point of view, there are different types of disorder. Spin glasses in a narrower sense are characterized by random exchange [3,4], as opposed to random-anisotropy magnets [3,5–14] and random-field spin glasses [4,9,15].

As shown by Imry and Ma [15], arbitrarily weak random magnetic fields destroy the ferromagnetic ground-state order in less than four dimensions. In the strong-disorder limit the magnetization points in field direction everywhere, and the range of ferromagnetic order equals the range of field correlations. Strong-disorder random-anisotropy magnets are subject to the same mechanism [4,6,9,11–14], but here the ground state is  $2^N$ -fold degenerate, where  $N$  is the number of magnetic moments. As we will see, this difference is responsible for the good permanent magnetic properties of random-anisotropy nanostructures involving intermetallics such as  $\text{Nd}_2\text{Fe}_{14}\text{B}$ ,  $\text{Sm}_2\text{Fe}_{17}\text{N}_3$ , and  $\text{Sm}(\text{Fe}_{11}\text{Ti})$ . For instance, in ball-milled  $\text{Sm}_2\text{Fe}_{17}\text{N}_3$  coercivities as high as  $\mu_0 H_c = 4.36$  T have been measured [16]. By comparison, the coercivity achievable in industrial  $\text{BaFe}_{12}\text{O}_{19}$ -type ceramic magnets is of order 0.3 T.

The effect of disorder is most pronounced in one-di-

mensional magnets, so we will restrict ourselves to the strong-anisotropy (strong-pinning) limit of the Hamiltonian

$$\mathbb{H} = -J \sum_i s_i \cdot s_{i+1} - K \sum_i (\mathbf{n}_i \cdot s_i)^2 - \sum_i \mathbf{h}_i \cdot s_i. \quad (1)$$

Here  $J > 0$  assures ferromagnetic coupling and  $K$  is the first-order anisotropy constant. The external magnetic field is written as  $\mathbf{h}_i = h(\mathbf{r}_i)$ , and the spin variable reads  $s_i = \cos \theta_i \mathbf{e}_z + \sin \theta_i \mathbf{e}_x$  and  $s_i = \cos \theta_i \mathbf{e}_z + \sin \theta_i \cos \phi_i \mathbf{e}_x + \sin \theta_i \sin \phi_i \mathbf{e}_y$ , in the classical planar and Heisenberg models, respectively. The disorder is assumed to consist in the randomness of the unit vector  $\mathbf{n}_i$  of the local easy-axis direction, so that  $\langle \mathbf{n} \rangle = 0$ . Defining random anisotropy as  $K(\mathbf{r}_i) \neq \langle K \rangle$  but leaving  $\mathbf{n}_i = \mathbf{e}_z$  throughout the crystal yields reduced nucleation fields and micromagnetic localization effects [14,17], which lie beyond the scope of this study.

The ground state is easily constructed by fixing one spin  $s_i$  in the direction of  $\mathbf{n}_i$  and subsequently adding neighbouring spins  $s_k = \pm \mathbf{n}_k$ . The right sign follows from the fact that the scalar product between neighbouring spins must be positive to minimize the exchange energy. The ground-state energy of the uniaxial Heisenberg model is, in lowest order,

$$E_0 = -JN/2 - KN. \quad (2)$$

Fig. 1 shows the level splitting of a uniaxial random-anisotropy Heisenberg magnet subject to a small exchange. We note that the energy of the ferromagnetic state is higher by only  $\delta E/N = J/4$  than the ground state; the anisotropy constant  $K$  does not appear in this expression. By comparison, the ground state of strong-disorder random-field magnets is non-degenerate, and  $\delta E/N = h/2$

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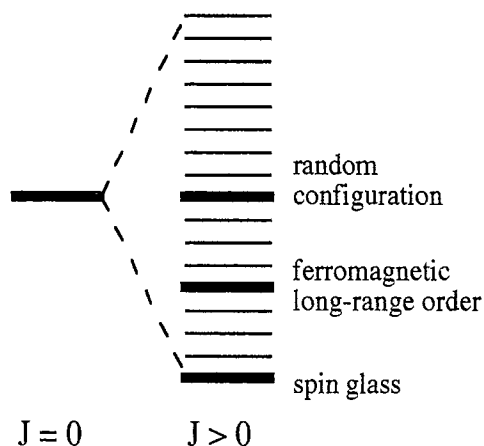


Fig. 1. Energy level splitting on small exchange coupling.

increases linearly with the strength of disorder. In other words, spin alignment in random-field magnets has to compete directly against the dominating magnetic field.

In terms of Coey's spin-disorder classification (Fig. 2), ferromagnetic long-range order corresponds to an ideally asperomagnetic configuration, whereas random configurations in the sense of Fig. 1 are speromagnetic at any length scale. The ground state turns out to be locally asperomagnetic but globally speromagnetic. The range  $R_c$  of ferromagnetic correlations, defined by  $\langle s(r)s(r') \rangle = \exp(-|r - r'|/R_c)$ , turns out to be

$$R_c = a / \ln(1 / \langle n_z \rangle_{\max}). \quad (3)$$

Here  $a = |\mathbf{r}_{i+1} - \mathbf{r}_i|$  and  $\langle n_z \rangle_{\max} = \int_{\max} n_z P(\mathbf{n}) d\mathbf{n}$  is the maximum spin projection compatible with the strong-disorder limit. For a uniaxial Heisenberg magnet  $\langle n_z \rangle_{\max} = 1/2$ , so that  $R_c = 1.443a$ . In the case of planar ferromagnets with  $n$ -fold in-plane anisotropy the correlation length is given by  $\langle n_z \rangle_{\max} = n \sin(\pi/n) / \pi$ . This leads to  $R_c/a = 2.21$  for a two-fold,  $R_c/a = 9.52$  for a four-fold, and  $R_c/a = 21.68$  for a six-fold anisotropy axis. The large- $n$  limit  $R_c/a = 6n^2 / \pi^2$  indicates asymptotic long-range order. By comparison, random-field magnets exhibit  $\langle n_z \rangle_{\max} = 0$  and  $R_c = 0$ .

Spin-glass permanent magnets such as  $\text{Sm}_2\text{Fe}_{17}\text{N}_3$  are characterized by nano-scale disorder, and it is necessary to rescale the exchange parameter  $J$  [12,13]. This is done by

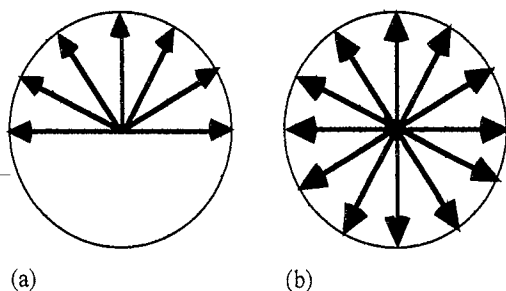


Fig. 2. Spin disorder: (a) asperomagnet and (b) speromagnet [3].

putting  $J \approx Aa^3/R_0^2$ , where  $A \approx 10^{-11}$  J/m is the exchange stiffness of the material and  $R_0$  the grain size. Using  $K = 9$  MJ/m<sup>3</sup> [18] and  $R_0 = 10$  nm we convince ourselves that the ratio  $A/KR_0^2 \approx 0.01$  is indeed much smaller than one for typical isotropic permanent magnets. This strong-pinning character explains the stability of the ferromagnetic state, regardless of whether the ground state is ferromagnetic or not. Illustratively, the nanocrystallites are decoupled and the order of magnitude of their coercivity is not much smaller than the anisotropy field  $\mu_0 H_0 = 2K/M_s$ . In the opposite weak-pinning limit the ferromagnetic exchange interaction dominates [3,7]. This enhances the size of the relevant micromagnetic units, which now feel the averaged anisotropy  $K\langle n \rangle = 0$  rather than the local anisotropy  $Kn$ .

In conclusion, we have shown that permanent magnetism does not necessarily presuppose a ferromagnetic ground state so long as there is ferromagnetic short-range order. By comparison, domain formation on magnetostatic interaction destroys ferromagnetic long-range order, predicted for instance for the two-dimensional Ising model [19], on a macroscopic length scale.

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