6 Nanostructured Exchange-Coupled Magnets

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6.1 Introduction

In recent years, exchange-coupled magnetic nanostructures have attracted much attention in the areas of permanent magnetism, magnetic recording, sensors, soft magnetism and spin-electronics. The reason is that suitable nanostructuring may improve the performance of an artificial material beyond that of naturally occurring substances, realizing what is known as the materials-by-design concept. This chapter focuses on permanent magnets, whose ability to store magnetostatic energy is described by the energy product. In the 20th century, energy product doubled every twelve years, and the present-day record-holder \( \text{Nd}_2 \text{Fe}_{14} \text{B} \) has energy products in excess of 451 kJ/m\(^3\). However, the outlook for discovering new ternary phases with magnetizations significantly higher than \( \text{Nd}_2 \text{Fe}_{14} \text{B} \) has been poor, and new approaches are necessary if the energy product is ever to double again. For example, as predicted in Skomski and Coey (1993), adding a soft material with a high polarization, such as \( \text{Fe}_{65} \text{Co}_{35} \) (\( M_s = 2.43 \) T), to an oriented hard magnet improves the energy product if the grain size of the soft regions is sufficiently small. Compared to the present-day theoretical limit of 516 kJ/m\(^3\) for single-phase \( \text{Nd}_2 \text{Fe}_{14} \text{B} \), the energy product in suitably nanostructured \( \text{Sm}_2 \text{Fe}_{17} \text{N}_3/\text{Fe}_{65} \text{Co}_{35} \) composites was predicted to be as high as 1090 kJ/m\(^3\) (Skomski and Coey, 1993). Related structures have been investigated by a number of authors (Al-Omari and Sellmyer, 1995; Liu et al., 1998a; Sawitchi et al., 2000; Bowden et al., 2000; Skomski et al., 1999; Hadjipanayis, 1999; Fullerton et al. 1999).

The development of exchange-coupled magnets has several starting points. From a technological point of view, Coehoorn et al. (1988) first exploited the remanence enhancement in isotropic Nd-Fe-B magnets; this research has its scientific root in earlier random-field (Imry and Ma, 1975) and random-anisotropy theories (Alben et al., 1978; Chudnovsky et al., 1986). A second starting point is the investigation of magnetic multilayers (Nieber and Kronmüller, 1989; Kneller and Hawig, 1991; Skomski and Coey, 1993), which is now widely associated with Kneller’s concept of exchange-spring magnetism. Third, attempts to predict the nucleation-field coercivities \( H_c = \)
Nanostructured Exchange-Coupled Magnets

$H_N(H_N$ is the nucleation field) for three-dimensional two-phase nanostructures have given rise to a quantitative analysis of the permanent-magnet performance of oriented two-phase nanostructures (Skomski, 1992; Skomski and Coey, 1993). A nanostructure where this principle has been realized in practice is FePt: the energy product of the composite exceeds that of hard-magnetic FePt, in spite of the soft magnetic character of the magnetic phase with a composition close to Fe$_3$Pt. In fact, the energy product of the composite, 420 kJ/m$^3$, approaches that of record-holding Nd$_2$Fe$_{14}$B and is superior to that of FePt (Liu et al., 1998a).

As the best available permanent magnets, rare-earth-transition-metal intermetallics have a lower saturation magnetization $M_s$ than many soft magnetic materials. Furthermore, these materials are chemically very reactive and also expensive due to a substantial rare-earth content. Therefore, exchange-coupled composite permanent magnets are preferred, consisting of both magnetically soft and hard phases, in which the former provides a high saturation polarization and the latter contributes to a high coercive field. Kneller and Hawig (1991) theoretically predicted a high maximum magnetic energy product and an unusually high remanence ratio or reduced remanence $M_r/M_s$, due to exchange coupling between nano-grains of the soft and hard magnetic phases. In addition they predicted a unique magnetic behavior characterized by a reversible demagnetization curve; that is, a maximum recoil permeability as distinguished from the conventional single-phase permanent magnets, where the demagnetization curves reflect essentially the distribution of the irreversible switching fields. It is for this unique magnetic behavior of the nanocomposites, in a sense resembling a mechanical spring, that such magnets have been termed exchange-spring magnets by Kneller and Hawig.

In nanomagnetism it is important to distinguish between isotropic and aligned magnets. Isotropic structures are comparatively easy to produce, but the remanent magnetization $M_r$ of randomly oriented grains with uniaxial anisotropy is only half the saturation magnetization $M_s$. Since energy product scales as $M_s^2$, this amounts to an energy-product reduction by a factor of 4. Intergranular exchange in isotropic magnets, realized by grain-size reduction, improves the remanence by favoring parallel spin alignment in neighboring grains (Coehoorn et al., 1988; Ding et al., 1993a; Muller et al., 1991). This important feature is known as remanence enhancement. On the other hand, intergranular exchange reduces the coercivity by averaging the anisotropy of grains with different crystallite orientations. This random-anisotropy effect (Imry and Ma, 1975; Callen et al., 1977; Alben et al., 1978; Chudnovsky et al., 1986) may overcompensate the benefit of the remanence enhancement, and structures with very small grain sizes are actually used as soft magnets. Typical examples are nanocrystalline Nd$_2$Fe$_{14}$B/Fe$_3$B-Fe and Sm$_2$Fe$_{17}$Ni$_x$/Fe composites produced by melt-spinning (Coehoorn et al., 1988) and mechanical alloying (Ding et al., 1993a), respectively. In oriented or aligned