Magnetically soft phase in magnetization reversal processes of nanocomposite Sm$_2$Fe$_{15}$Ga$_2$C$_x$/$\alpha$-Fe permanent magnetic materials

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The magnetic properties and magnetization reversal processes of nanocomposite Sm$_2$Fe$_{15}$Ga$_2$C$_x$/$\alpha$-Fe permanent magnet materials have been investigated. Remanence increases and intrinsic coercivity decreases drastically with increasing magnetically soft phase $\alpha$-Fe content. About 5 vol. % excess of $\alpha$-Fe results in a significant increase in maximum energy product from 5.6 MG Oe for single-phase nanocrystalline alloys to 8.2 MG Oe for two-phase nanocomposites. A further increase in $\alpha$-Fe content leads to a decrease of the maximum energy product owing to the rapid decrease of coercivity and the deterioration of the squareness of the hysteresis loop. The nucleation field for reversing the magnetically hard phase decreases monotonically with increasing $\alpha$-Fe content. Remanence enhancement, a high degree of reversibility, and high ratios of remanence coercivity to intrinsic coercivity are observed in two-phase nanocomposites.

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Both experimental studies and numerical calculations reveal that a significant enhancement of remanence and maximum energy product can be achieved by synthesizing nanocomposites composed of magnetically hard and soft phases with strong exchange coupling.1–6 Nanocomposite permanent magnet materials can be made into inexpensively bonded magnets due to their low rare-earth concentration. The effects of grain size and the amount of magnetically soft phase on the magnetic properties of Nd-Fe-B nanocomposites have been extensively investigated.2,4,7 Some work has been performed to investigate the microstructural and magnetic properties of exchange-coupled Sm$_2$Fe$_{15}$Ga$_2$C$_x$/$\alpha$-Fe nanocomposites fabricated by melt-spinning and mechanical alloying.8–10 However, the role of magnetically soft phase $\alpha$-Fe in these nanocomposites is not yet well understood. In this work, we investigated the effect of the magnetically soft phase $\alpha$-Fe on the magnetic properties and magnetization reversal process of nanocomposite Sm$_2$Fe$_{15}$Ga$_2$C$_x$/$\alpha$-Fe permanent magnet materials.

Alloys with nominal compositions of Sm$_2$Fe$_{15+y}$Ga$_2$C$_y$ ($y = 0.0, 1.5, 3.0, 4.5, 6.0$) were prepared by arc-melting. Iron and carbon were first melted together in an induction furnace to form an Fe-C prealloy with lower melting temperature. Then Sm, Ga, Fe, and Fe-C prealloy were remelted together in a highly-pure-argon atmosphere. The elements used were at least 99.9% pure. An excess of 5% Sm was added to compensate the evaporation loss during melting. The ingot alloys were remelted at least six times to ensure good homogeneity. As-quenched ribbons with a width of about 2.4 mm and a thickness of about 36 $\mu$m were prepared by melt-spinning in a highly-pure-helium atmosphere at the substrate velocity $v_s = 17.5$ m/s. The phase composition was analyzed by means of x-ray diffraction (XRD) with Cu K$\alpha$ radiation. XRD patterns are collected at room temperature at angles ranging from 2 $\theta = 25^\circ - 85^\circ$ with a step of 0.01°. XRD patterns indicate that the as-quenched ribbon consists of a magnetically hard phase Sm$_2$Fe$_{15}$Ga$_2$C$_x$ with Th$_2$Zn$_{17}$-type structure and $\alpha$-Fe [Fig. 1(a)]. No change in the lattice constants of the 2:17 phase is found within experimental error, implying that the chemical composition of the 2:17 phase is uniform in these nanocomposites. The chemical analysis by the burning method shows that the carbon content corresponds to $x = 2.19 \pm 0.06$ in the Sm$_2$Fe$_{15}$Ga$_2$C$_x$ phase. From the values of saturation magnetization and thermomagnetic curves, the estimated volume fractions of magnetically soft phase, $f_x = 0, 5, 13, 19, \text{ and } 33$ vol. % correspond to the nominal composition of Sm$_2$Fe$_{15+y}$Ga$_2$C$_y$ with

![FIG. 1. XRD pattern of the nanocomposites with 13 vol. % Fe (a) and the grain size distribution (b). Inset of (b) is the TEM micrograph.](image-url)
respectively. On the basis of Eq. [52x459]-Fe content. The magnetization measurement was performed using a vibrating sample magnetometer (VSM) with a 70-kOe superconducting magnet.

Figure 2 illustrates the room-temperature hysteresis loops of as-quenched ribbons with different amounts of $\alpha$-Fe. The demagnetization curves show a nearly single hard magnetic phase behavior for samples with lower $\alpha$-Fe content. Separate switching of the magnetically hard and soft phases is evident with increasing $\alpha$-Fe content. High ratios of remanence to saturation magnetization, 0.63–0.70, are obtained in these exchange-coupled nanocomposites. As $\alpha$-Fe content increases, the remanence increases, but at the expense of the decrease of both intrinsic coercivity and remanence coercivity [Fig. 3(b)]. Here, we define the intrinsic coercivity as $M(H_c)=0$ from major loops in Fig. 2 and the remanence coercivity as $M_r(H_c)=0$ from dc demagnetization remanence (DCD) curves in Fig. 6. The more detailed discussion regarding remanence coercivity and intrinsic coercivity will be presented in following section. According to the Stoner-Wohlfarth model, the ideal ratio of remanence to saturation magnetization for randomly oriented crystallites or particles is well known to be 0.5 and 0.832 for a uniaxial magnetocrystalline anisotropy and $\alpha$-Fe with $K_1>$0.11 Therefore, the saturation magnetization and remanence in the isotropic crystallites of magnetically hard phase with uniaxial anisotropy $\text{Sm}_2\text{Fe}_{15}\text{Ga}_2\text{C}_2$ and $\alpha$-Fe without exchange coupling can be expected as

$$M_s=M_{s,\alpha}(1-f_\alpha)+M_{s,\alpha}\delta f_s,$$

$$M_r=0.5M_{s,\alpha}(1-f_\alpha)+0.832M_{s,\alpha}\delta f_s,$$

where $M_{s,\alpha}=9.8$ kG obtained from single-phase nanocrystalline $\text{Sm}_2\text{Fe}_{15}\text{Ga}_2\text{C}_2$ and $M_{s,\alpha}=21.6$ kG are the saturation magnetization of the magnetically hard phase and bulk $\alpha$-Fe, respectively. On the basis of Eq. (2), the expected values of remanence can be calculated and illustrated in Fig. 3(a) as dashed lines. By comparing the experimental and expected values, the values of remanence are about 10–20% higher than those of the randomly oriented exchange-decoupled grains with the same composition. A significant enhancement of remanence was observed in both exchange-coupled single-phase and nanocomposite phases. Owing to its relatively high remanence and sufficiently high coercivity, the maximum energy product of the nanocomposite materials with 5 vol. % Fe increases about 46% compared with that of the single-phase nanocrystalline alloys, from 5.6 to 8.2 MG Oe. In contrast with the two-phase $\text{Nd}_2\text{Fe}_{14}\text{Bi}$/$\alpha$-Fe nanocomposites, which still exhibit a very high maximum energy product up to a content of 30 vol. % $\alpha$-Fe, our results indicate that the maximum energy product decreases with further increasing $\alpha$-Fe content. This difference is associated with the different mean grain sizes between $\text{Sm}_2\text{Fe}_{15}\text{Ga}_2\text{C}_2$/$\alpha$-Fe and $\text{Nd}_2\text{Fe}_{14}\text{Bi}$/$\alpha$-Fe nanocomposites. Transmission electron micrograph (TEM) shows that the grain sizes of $\alpha$-Fe are distributed mainly in the range 20–40 nm [Fig. 1(b)]. The large grain sizes of $\alpha$-Fe, which are much larger than the domain wall width of $\text{Sm}_2\text{Fe}_{15}\text{Ga}_2\text{C}_2$ ($\delta_d$=4 nm), cannot guarantee that the entire volume of $\alpha$-Fe is exchange hardening by the $\text{Sm}_2\text{Fe}_{15}\text{Ga}_2\text{C}_2$ phase because the exchange coupling requires grain sizes of the order of $2\delta_d=8$ nm. A further increase in $\alpha$-Fe content results in a decrease of coercivity and deterioration of the squareness of hysteresis loop, and consequently a decrease of maximum energy product. Since the exchange coupling becomes more dominant and will be more effective in preventing the magnetization reversal of $\alpha$-Fe with decreasing mean grain size to $2\delta_d=8$ nm, high values of the coercivity and maximum energy product, as predicted by computational simulations, can be kept up to very high $\alpha$-Fe content for nanocomposites with smaller mean grain sizes. The large grain size can be reduced to a value closer to optimum by either increasing the substrate velocity $v_s$ or adding a small amount of Zr, Cu,
etc., which are immiscible in Fe and can suppress the grain growth of α-Fe. Significant improvements of magnetic properties have been obtained by the addition of Zr.\textsuperscript{12} It poses significant preparation challenges by conventional techniques, including melt-spinning, mechanical milling, and sputtering, to control both the hard and soft phases at the nanometer scale, to ensure efficient exchange coupling. The perfect microstructure can be achieved by using nanoparticle self-assembly technique. An optimum exchange coupling, and therefore an optimum energy product, can be obtained by independently tuning the size and composition of the individual building blocks. Exchange-coupled isotropic FePt-Fe\textsubscript{3}Pt nanocomposites with an energy product of 20.1 MG Oe, which exceeds the theoretical limit of 13 MG Oe for nonexchange-coupled isotropic FePt by over 50%, was reported.\textsuperscript{13}

For example, Fig. 4 shows the room-temperature DCD curves of the nanocomposites with 13 vol. % Fe. The two-phase Sm\textsubscript{2}Fe\textsubscript{13}Ga\textsubscript{2}C\textsubscript{1}/α-Fe nanocomposites are characterized by steep recoil curves and a large component of recoverable magnetization, i.e., “exchange-spring” behavior. The exchange-spring behavior is related to the reversible rotation of the magnetically soft component for fields that are not large enough to reverse the magnetically hard component. In order to determine the nucleation field \( H_n \) for irreversible magnetization reversal of the magnetically hard phase, it is useful to investigate the DC demagnetization curves in terms of the irreversible portion \( \Delta M_{\text{irr}} = (M_r - M_d(H))/2M_r \) and the reversible portion \( \Delta M_{\text{rev}} = (M_d(H) - M(H))/M_r \) of the total magnetization change as a function of the reversed field (Fig. 5), where \( M_d(H) \) and \( M_r \) are the dc field demagnetization remanence and remanence after saturation, respectively, and \( M(H) \) is the magnetization in the major demagnetization curve. The \( \Delta M_{\text{irr}} \) curves have been explained by a one-dimensional purely inhomogeneous magnetization rotation model.\textsuperscript{6} No magnetization reversal occurs when the reverse field \( H \) increases from 0 to the nucleation field \( H_n \). When \( H \) increases further to a certain value \( H_s = H_n/\cos \theta_1 \), all regions with \( 0 < \theta < \theta_1 \) have reversed, where \( \theta \) is the angle between reverse magnetic field and the easy magnetization direction of individual single-domain grain. The total irreversible change in magnetization for isotropic grains is \( 2M_r \int_0^{\theta_1} \cos \theta d\theta = 2M_r \sin \theta_1 \). Thus, the nucleation fields of 9.0, 6.5, and 4.7 kOe have been obtained by the best fit of a corresponding plot of \( \sin \theta \) vs \( H_n/\cos \theta \) to the experimental \( \Delta M_{\text{irr}}(H) \) curves for the samples with an excess of 5, 13, and 19 vol. % Fe, respectively. The \( \Delta M_{\text{rev}} \) curve shows a peak at the nucleation field. The reversible susceptibility is found to increase with increasing α-Fe content. The nucleation field of aligned nanocomposites can be described as \( H_n = 2[f_S K_S + (1 - f_S) K_A]/\mu_0 [f_S M_{S,Fe} + (1 - f_S) M_{S,A}] \), where \( K_S \) and \( K_A \) are the anisotropy constants of magnetically hard and soft phases, respectively.\textsuperscript{5} The misalignment and exchange coupling of the isotropic grains will decrease the nucleation field. As α-Fe content increases, the nucleation fields are expected to decrease monotonically. Owing to the large grain size of α-Fe and its broad distribution, some portions of irreversible magnetization (\( \Delta M_{\text{irr}} > 0 \)) occur at the nucleation field. As shown in Fig. 5, the portions are found to increase with increasing α-Fe content. It was found that the nucleation field for the lower α-Fe content is nearly equal to the intrinsic coercivity, while the difference between the nucleation field and the intrinsic coercivity increases with increasing α-Fe content. This is because the intrinsic coercivity in nanocomposites includes both reversible and irreversible changes. Figure 6 illustrates the demagnetization remanence \( M_d \) as a function of the applied reversed field. The remanence coercivity \( H_r \), which is defined by \( M_d(H_r) = 0 \), depends only on the irreversible part of the demagnetization process. The ratios of remanence co-

![FIG. 4. Room-temperature dc demagnetization remanence curves (DCD) for the nanocomposites with 13 vol. % Fe.](image)

![FIG. 5. The variation of irreversible portion \( \Delta M_{\text{irr}} \) and reversible portion \( \Delta M_{\text{rev}} \) as a function of the reversed field.](image)

![FIG. 6. Demagnetization remanence \( M_d \) as a function of the applied reversed field.](image)
ercivity to the intrinsic coercivity are found to increase from 1.14 to 1.88 with increasing α-Fe content [Fig. 3(b)]. A similar result was also observed in Nd-Fe-B nanocomposites.14 For randomly oriented uniaxial particles without exchange-coupling, the $H_r/H_c$ ratio of 1.09 was calculated by Wohlfarth.15 In the case of nanocomposites, the excess of magnetically soft phase results in a decrease of intrinsic coercivity, while the “exchange-spring” behavior, i.e., the high degree of reversibility, gives rise to an increase in remanence coercivity. Thus, an increased ratio of remanence coercivity to intrinsic coercivity can be expected in two-phase nanocomposites.

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