Contributions of individual Fe sites to magnetocrystalline anisotropy of $Y_{2}Fe_{17-x}Ga_{x}$ compounds

Zhao-hua Cheng a), Bao-gen Shen, and Fang-wei Wang
State Key Laboratory of Magnetism, Institute of Physics & Center of Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100080, People’s Republic of China

H. Kronmüller
Max-Planck-Institut für Metallforschung, Heisenbergstrasse 1, 70569 Stuttgart, Germany

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At present, individual Fe sites anisotropies of $Y_{2}Fe_{17}$ compound cannot be directly measured by ordinary magnetization measurement. Alternatively, they are derived from the relationship between occupancy factors of Fe atoms at different sites and anisotropy constants of $Y_{2}Fe_{17-x}Ga_{x}$ compounds. It is found that only Fe atoms at 9d site have a significantly positive contribution to the uniaxial anisotropy, while those at 6c and 18h sites have strongly negative contributions; the contribution of Fe atoms at 18f site is slightly negative and less important. The planar anisotropy is rapidly weakened and finally a uniaxial anisotropy is induced by the Ga substitution. These results can be explained by the preferential occupancies of Ga atoms at 18h site for low Ga concentration as well as 6c and 18f sites for high Ga concentration. © 1999 American Institute of Physics.

In the search for permanent magnets whose hard magnetic properties are superior to those of Nd$_2$Fe$_{14}$B, the anisotropy of R$_3$Fe$_{17}$ compounds and their carbides and nitrides has been extensively investigated. Most of the early work in this field was focused on the effect of interstitial atoms, N or C, on the R-sublattice anisotropy, the investigations of Fe-sublattice anisotropy are relatively few. Recently, it was found that the easy magnetization direction (EMD) of R$_2$Fe$_{17-x}$M$_x$ (M=Ga or Al) compounds can be modified by the introduction of M atoms. The Fe sublattice in the 2:17-type structure exhibits a uniaxial anisotropy when the Ga concentration is very high ($x \approx 7$). However, it is only possible to measure the overall Fe-sublattice magnetocrystalline anisotropy by ordinary magnetization measurement, the individual Fe sites anisotropy in R$_3$Fe$_{17}$ compounds could not be directly obtained up to now.

A previous study of Y$_2$(Co$_{1-x}$Fe$_x$)$_{17}$ compounds indicated that both Fe and Co atoms at 9d site have a predominantly planar anisotropy, Fe atoms at 6c, 18f, and 18h sites have slightly uniaxial anisotropy, while the anisotropy constants of Co atoms at 6c and 18h are more than one order of magnitude smaller than those at 9d and 18f sites. Unfortunately, there are some difficulties in comparing these conclusions with recently experimental results of Y$_2$Fe$_{17-x}$M$_x$ (M=Ga or Al) anisotropy. According to the site-occupancy studies in R$_2$Fe$_{17-x}$M$_x$ (R=Nd, Tb, Ho, Er, and Y, M=Ga or Al) systems by x-ray diffraction and neutron diffraction, M atoms occupy preferentially 18h site for low M concentration and 6c and 18f sites for high M concentration, and do not occupy 9d site at all. Thus, the introduction of Ga is expected to enhance the planar anisotropy based on the results in Ref. 5, rather than to weaken it and finally to induce a uniaxial anisotropy. The same problem can also be found in explaining the relationship between the atomic site occupancy and anisotropy of R$_2$Co$_{17-x}$Al$_x$ (R=Gd or Nd) compounds. These difficulties are probably related to the following reasons: (1) Both magnetic Fe and Co atoms share same sites of Y$_2$(Co$_{1-x}$Fe$_x$)$_{17}$ compounds, the exchange coupling between Fe and Co atoms makes the situation very complex. (2) Some uncertainties will be caused by using too many parameters $K_1$ of Co and Fe atoms at different sites for calculation. Therefore, it is necessary to investigate the contributions of these non-equivalent Fe sites to magnetocrystalline anisotropy in details. In this letter, we present the anisotropy of Y$_2$Fe$_{17-x}$Ga$_x$ to investigate the local anisotropy constants of these four nonequivalent sites. The reason for selecting this system is twofold. First, Ga atoms can substitute Fe up to a very high concentration without a structural change, except for the lattice expansion. Second, Ga and Y atoms do not posses magnetic moments, while Ga atoms can change the anisotropy of Y$_2$Fe$_{17-x}$Ga$_x$ drastically. Partial substitution of Fe by non-magnetic Ga can make the situation simpler and obtain more reliable results. Therefore, one can investigate the individual Fe sites anisotropy on the basis of a relatively simple model.

In order to determine anisotropy constants precisely, the best way is to measure magnetization curves of a single-crystal sample along different crystallographic axes. However, it is always difficult, time consuming, and even impossible to obtain a single-crystal sample. For this reason, an alternative way is generally employed to measure the magnetization curves of magnetically aligned fine powders. The sample preparation and magnetic alignment of Y$_2$Fe$_{17-x}$Ga$_x$ compounds were described in Ref. 3. The positive anisotropy constant $K_1$ of Y$_2$Fe$_{17}$Ga$_7$ was derived from the Sucksmith–Thompson method. However, since the c axes of individual crystallites of Y$_2$Fe$_{17-x}$Ga$_x$ compound ($0 \leq x \leq 6$) with planar anisotropy are randomly distributed in a plane normal to the aligned field direction, rather than are fixed in one direc-

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a) Corresponding author.
Electronic mail: cheng@vaxph.mpi-stuttgart.mpg.de
The anisotropy constant $K_1$ is negligible compared to $K_1$ in $Y_2Fe_{17}$ compound, the total energy density of single-domain particles can be therefore written as

$$E(\theta) = K_1 \sin^2 \theta - \mu_0 H M_S \cos (\theta - \theta_B),$$

(1)

where $\theta$ and $\theta_B$ are the angles of spontaneous magnetization $M_S$, and applied field $H$ deviating from $c$ axis, respectively [Fig. 1(a)].

By minimizing Eq. (1) with respect to $\theta$ one can obtain

$$\gamma \sin \delta = \sin 2(\theta_B + \delta),$$

(2)

where $\gamma = H/H_A$ is the reduced applied field with the anisotropy field given by $H_A = -2K_1/\mu_0 M_S$. $\delta = (\theta - \theta_B)$ is the angle between the spontaneous magnetization $M_S$ and the applied field $H$, $\delta = 0$. It follows that the reduced magnetization of one single-domain grain $i$, $\sigma_i = M_i/M_S = \cos \delta_i$. The change of $\delta_i$ with $\gamma$ and $\theta_B$ can be obtained by numerical calculation from Eq. (2). The reduced magnetization $\sigma$ for a given value of $\gamma$ is the sum of the individual crystallite’s magnetization and follows the equation:

$$\sigma(\gamma) = \sum_i \sigma_i = \int_0^{\pi/2} \cos \delta(\theta_B, \gamma) d\theta_B.$$

(3)

The anisotropy field, $H_A$, and hence the anisotropy constant, $K_1$, are derived from the best fit of a corresponding plot of the experimentally reduced magnetization $M(H)/M_S$ along the hard magnetization direction versus the reduced field $H/H_A$ to the calculated curve from Eqs. (2) and (3) (Fig. 2). In order to minimize the contribution of domain wall displacement, the experimental data at low magnetic fields are not taken into account. The anisotropy constant ($K_1 = -43.9$ Kf.u. or $-2.4$ MJ/m$^3$) of $Y_2Fe_{17}$ at 4.2 K obtained by this method is in good agreement with that of a single crystal ($K_1 = -2.3 \pm 0.1$ MJ/m$^3$).

Figure 3 illustrates the anisotropy constants $K_1$ as a function of temperature $T$ and $[M_S(T)/M_S(0)]^5$. The linear relationship between $K_1(T)$ and $[M_S(T)/M_S(0)]^5$ suggests that the single ion anisotropy mechanism plays an important role, as predicated in the localized moment systems. This highly localized system may originate from the $3d$ electrons orbit partial quenching in $R-T$ intermetallics.

The overall anisotropy can be decomposed into the sum of individual sites anisotropy. The compositional dependence of anisotropy constants of $Y_2Fe_{17-x}Ga_x$ compounds is shown in Fig. 4. The nonlinear change of $K_1$ with $x$ implies that the contributions of these four sites to the overall anisotropy are different. In order to take these contributions into account, the following expression is used for the Ga-containing compounds:

$$K_1(x) = \sum_i n_i f_i(x) K_{1,Fe}^i,$$

(4)

where $n_i$ and $f_i(x)$ are the number of the equivalent $3d$ atoms and the Fe occupancy factor at the $i$ site in a unit cell, and $K_{1,Fe}^i$ is the individual site anisotropy constant, which is governed by the local crystal-field parameters of site $i$ and is approximately assumed to be independent of $x$.

In order to minimize the uncertainties of site-occupancy factors, we have compared three independent results, $7,8,15$.
one of them was a high-resolution neutron diffraction study of $Y_2Fe_{17-x}Ga_x$ ($x = 5$ and 7). The site-occupancy factors obtained by x-ray diffraction, as shown in the inset of Fig. 4, are consistent with those of neutron diffraction. By considering the uncertainties of site-occupancy factors determined by x-ray diffraction, one can obtain the values of $K_{1Fe}^s = 9 \pm 1$, $20 \pm 3$, $-3 \pm 1$, and $-11 \pm 1$ K/Fe for $6c$, $9d$, $18f$, and $18h$ sites, respectively.

It can be seen that Fe atoms at $9d$ site show a significantly uniaxial anisotropy, the contribution of Fe atoms at $18f$ site to overall uniaxial anisotropy is negative and less important. The competition between magnetocrystalline anisotropy of Fe atoms at these four sites results in an overall planar anisotropy of $Y_2Fe_{17}$ compound. The overall planar anisotropy will be weakened when the Fe atoms at $18h$ and $6c$ sites are removed by the nonmagnetic Ga atoms. Since the Fe atoms at $9d$ site are not substituted by Ga atoms, it is possible to induce a uniaxial anisotropy by a further Ga substitution. Indeed, a uniaxial anisotropy is observed in $Y_2Fe_{17-x}Ga_x$ compounds with $x \geq 7$.

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