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Fe substitution effects on the magnetic properties of the RCo$_{4-x}$Fe$_x$Al compounds (R: Ho and Y)

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Abstract

Fe substitution for Co leads to the increase of the Curie temperatures of both HoCo$_{4-x}$Fe$_x$Al and YCo$_{4-x}$Fe$_x$Al and of the spin-reorientation temperature of HoCo$_{4-x}$Fe$_x$Al. In contrary, the anisotropy field of YCo$_{4-x}$Fe$_x$Al decreases quickly with increasing Fe content. The results are explained in terms of the individual site anisotropy model.

RCO$_{5-x}$Al$_x$ ($x \leq 2$) compounds with CaCu$_5$ structure have recently been studied. RCo$_2$Al has been stressed in order to compare the magnetic properties with those of RCo$_2$B $^{[1,2]}$. Al substitution for Co strongly diminishes the Curie temperature, the Co atomic moment and the magnetocrystalline anisotropy (MCA) of both R and 3d sublattices. To further study the magnetic properties of the 3d sublattice of RCo$_2$Al, in this paper the effects of Fe substitution on the magnetic properties of RCo$_{4-x}$Fe$_x$Al (R = Y and Ho) have been studied.

The ingots of RCo$_{4-x}$Fe$_x$Al ($x = 0.0-4.0$) were prepared by arc melting the constituent elements of high purity (at least 99.9%) and then annealed at 900°C for about 50-70 h. The magnetically aligned samples were prepared by solidifying the mixture of sample powders and epoxy resin in a magnetic field of 1.1 T at room temperature. X-ray diffraction and magnetic measurements were carried out on the bulk, powdered and aligned samples. It turns out that the samples with $x \leq 1.8$ for YCo$_{4-x}$Fe$_x$Al and $x \leq 1.5$ for HoCo$_{4-x}$Fe$_x$Al are generally single phase with the CaCu$_5$ structure. The YCo$_2$Fe$_2$Al and YCO$_{1.8}$Fe$_{2.2}$Al samples contain a small amount of a second phase, while the samples with higher Fe content are found to be multiphase and are not considered in the following part of this paper. Pulsed magnetic fields up to 3.5 T have been used to measure the aligned samples at 77 and 300 K. Values for the anisotropy field $H_a$ of the samples with uniaxial anisotropy were approximately estimated by extrapolating the magnetization curves measured along the easy and hard magnetization directions to their intersection.

The composition dependences of the obtained magnetic parameters of HoCo$_{4-x}$Fe$_x$Al and of the Curie temperature of YCo$_{4-x}$Fe$_x$Al are shown in Fig. 1 and in the inset of Fig. 1, respectively. It can be seen that with increasing Fe content, the Curie temperature $T_C$ increases for both systems and tends to reach a constant value at about $x = 1.4$. This effect of Fe substitution on the Curie temperature of RCo$_2$Al is in good accordance with those obtained for the Co–rare-earth intermetallics $\text{RCO}_3$ and RCo$_2$B $^{[4]}$. In contrast, the ratio $T_K/T_C$ ($T_K$ is the compensation temperature) decreases when the Fe content increases. This result can be explained by taking into account the larger magnetic moment of Fe compared to that of Co, which leads to an increase of the 3d-sublattice moment $^{[4]}$.

In a low magnetic field (0.1 T), an anomalous temperature dependence of the magnetization can be observed for the HoCo$_{4-x}$Fe$_x$Al samples with $x < 1.5$, which is considered to be associated with the spin-reorientation transition as also confirmed by ac susceptibility measurements and by the temperature dependence of the magnetization of aligned samples (not shown here). In addition, an enhancement of the 002 and 001 peaks can be observed from the X-ray diffraction patterns of the aligned samples, which confirms the uniaxial anisotropy along the c-axis of the samples with $x < 1$ at room temperature. Thus, the observed spin-reorientation transition is from planar to uniaxial anisotropy with increasing temperature. It is found that the spin-reorientation transition temperature increases with increasing Fe content (see Fig. 1).

In Fig. 2, the composition dependence of $H_a$ for YCo$_{4-x}$Fe$_x$Al at 77 and 300 K is presented. The data for YCo$_4$Al were quoted from Ref. $^{[2]}$. It can be seen that with increasing Fe content, $H_a$ decreases quickly and tends to change its sign at about $x = 2.0$. In combination with X-ray diffraction patterns of the aligned samples (not

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shown here) it can be suggested that in the temperature range of 77–300 K, the samples with \( x < 2.0 \) show an uniaxial anisotropy whereas the sample with \( x = 2.2 \) has a planar one. A conical anisotropy at room temperature is also suggested for the sample with \( x = 2.0 \). It is evident that Fe substitution leads to a strong decrease in the positive MCA of YCo\(_4\)Al. Also, if we consider the observed spin-reorientation transition in HoCo\(_4\)Al as a result of the competition between the positive MCA of Co sublattice and the negative one of Ho sublattice, the observed increase of the spin-reorientation temperature with increasing Fe content can be regarded to result from the decrease of the positive MCA of the 3d sublattice. This effect of Fe substitution can be explained on the basis of the individual site anisotropy (ISA) model [4,10], in which the overall MCA of 3d sublattice is considered to be the sum of the independent contributions coming from the individual 3d sites. In rare-earth–transition-metal intermetallics with CaCu\(_4\) structure, there are two crystallographic sites for the transition metal, namely 2c and 3g. As evidenced from nuclear magnetic resonance (NMR) measurements on RCo\(_x\), the Co ions at the 2c site give a large positive contribution to the overall anisotropy whereas the Co ions at the 3g site give a small negative one [5,6]. In addition, in substituted \( R_xCo_{1-x}Fe_x \) compounds the preferential occupation at some crystallographic sites of Fe ions has been observed by means of neutron diffraction and Mössbauer effect studies [7–9]. Fe and Co ions usually give contributions of opposite signs at the same crystallographic site [4,10]. The decrease of the positive MCA of the 3d sublattice with increasing Fe content obtained above, therefore, suggests that Fe ions preferably occupy the 2c site. At this site, Fe ions have a negative MCA, which competes with the positive one of the Co ions. It should be noticed that Fe ions in RCo\(_{1-x}Fe_xB\) (CeCo\(_4\)B structure) also prefer the 2c site [7,8] whereas for RCo\(_{1-x}Fe_x\), in contrast, Fe ions prefer the 3g site [9]. Recently Konno et al. [3] have reported that the distribution of Al in RCo\(_{1-x}Al_x\) is not necessarily random. The Al atoms, furthermore, can prefer the 3g site to the 2c site [3]. Thus the Al substitution for Co in RCo\(_4\)Al is suggested to strongly modify the environment of the crystallographic sites of RCo\(_4\) compounds, which leads to a large influence on the occupation of Fe ions in RCo\(_{4-x}Fe_x\)Al.

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