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1999 J. Phys.: Condens. Matter 11 5539

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Magnetocrystalline anisotropy of $Ho_2(Co_{1-x}Fe_x)_{15}Al_2$ compounds

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Received 24 November 1998, in final form 22 March 1999

Abstract. The structure and magnetic properties of $Ho_2(Co_{1-x}Fe_x)_{15}Al_2$ compounds with x = 0.0, 0.1, 0.2, 0.3, 0.4, 0.6, 0.8, 1.0 have been investigated by x-ray diffraction (XRD) and magnetization measurements. XRD patterns demonstrate that all samples are single phase with the hexagonal Th₂Ni₁₇-type structure. The substitution of Fe for Co leads to a nearly linear increase in the unit cell volume and a monotonic decrease of Curie temperature. Saturation magnetization increases with *x* and then decreases, attaining a maximum at x = 0.2. The magnetic anisotropy phase diagram of $Ho_2(Co_{1-x}Fe_x)_{15}Al_2$ compounds has been determined from the temperature dependence of magnetization and the XRD patterns of magnetically oriented powder samples. The spin-reorientation temperature of $Ho_2(Co_{1-x}Fe_x)_{15}Al_2$ compounds was found to decrease firstly with Fe content increasing for x < 0.1 and then increase for 0.1 < x < 0.4, attaining a lowest value of 381 K at x = 0.1.

1. Introduction

The binary rare-earth-iron compounds of the composition R_2Fe_{17} are of potential commercial interest as they possess good thermal stability and large Fe content, yielding the highest saturation magnetization. However, the relatively low values of the Curie temperature and the lack of a room-temperature uniaxial anisotropy have limited the practical application of these compounds as hard magnetic materials [27, 28]. While many rare-earth-cobalt compounds do not suffer from these drawbacks, they are certainly not as attractive for commercial applications from an economic standpoint. With the exception of Sm, Er, Tm, all the rare-earth compounds with the R_2Co_{17} type exhibit an easy plane anisotropy at room temperature [29, 30]. Therefore, the binary 2:17 phases do not look very promising for permanent magnets. However, gradual replacement of Co with Fe in $R_2(Co_{1-x}Fe_x)_{17}$ compounds for (R = Y, Gd, Ce, Pr, Nd, Dy and Ho) could switch the anisotropy to easy c-axis within a specific composition range [1–3, 12, 21]. Recently, it was found that the substitution of Al, Ga or Si could not only significantly increase the Curie temperature of R_2Fe_{17} , but also induce the change of easy magnetic direction (EMD) from plane to axis in $R_2Fe_{17-x}Ga_x$ (R = Y, Gd, Tb, Dy, Ho, Er, Tm) [4], $Gd_2Co_{17-x}Ga_x$ $(x \ge 1)$ [5], Gd₂Co_{17-x}Al_x $(x \ge 4)$ [6], Ho₂Co_{17-x}Al_x $(x \ge 2)$ [7] and other series. It is interesting to investigate the magnetic properties of the the (Fe, Co)-based ternary compounds. In this paper, we extended our investigation into the structure and magnetic behaviours of $Ho_2(Co_{1-x}Fe_x)_{15}Al_2$ compounds, especially the magnetocrystalline anisotropy.

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2. Experimental detail

The Ho₂(Co_{1-x}Fe_x)₁₅Al₂ compounds for $0 \le x \le 1$ samples were prepared by arc melting in an argon atmosphere of high purity. The raw materials of Ho, Fe, Co were at least 99.9% pure. The ingots were turned over and remelted at least three times to ensure homogeneity. The as-cast ingots were sealed in an evacuated quartz tube and annealed under vacuum at 1313 K for 156 h, then quenched into water. X-ray diffraction (XRD) using Cu K α radiation in a Rigaku Rint 1400 was used to determine crystal structure and lattice parameters. The saturation moment M_s at 1.5 K was deduced from the data collected on free powder samples using an extracting sample magnetometer in a field up to 6.5 T. The Curie temperature T_C and spin-reorientation temperature T_{sr} were determined from the temperature dependence of magnetization measured with a Faraday balance ($T \ge 300$ K) and the extracting sample magnetometer (1.5 K $\le T \le 300$ K) in a field of 0.1 T. The aligned samples for the magnetocrystalline anisotropy studies were prepared by mixing the powder with epoxy resin and then aligning in a magnetic field of 1 T. The XRD procedure was also used to determine the EMD.

3. Results and discussion

3.1. Structural properties

XRD patterns indicate that all samples with the compositions of Ho₂(Co_{1-x}Fe_x)₁₅Al₂ are single phase with the hexagonal Th₂Ni₁₇-type structure. Figure 1 shows the typical XRD patterns of Ho₂(Co_{1-x}Fe_x)₁₅Al₂ compounds with x = 0.2, 0.4 and 0.6. The lattice parameters *a*, *c* and the unit cell volume *v* of the samples obtained from x-ray data are listed in table 1 and are plotted in figure 2. It can be found that the substitution of Fe for Co leads to a monotonic increase in unit cell volumes. A similar increase of the unit cell was also observed in other R₂(Co_{1-x}Fe_x)_{17-y}Al_y series [8, 10].

Compounds	<i>a</i> (nm)	<i>c</i> (nm)	v (10 ³ nm ³)	
Ho ₂ Co ₁₅ Al ₂	83.9	81.5	496.83	
Ho ₂ (Co _{0.9} Fe _{0.1}) ₁₅ Al ₂	84.0	82.0	501.08	
Ho ₂ (Co _{0.8} Fe _{0.2}) ₁₅ Al ₂	84.1	82.3	504.11	
Ho ₂ (Co _{0.7} Fe _{0.3}) ₁₅ Al ₂	84.2	82.5	506.53	
Ho ₂ (Co _{0.6} Fe _{0.4}) ₁₅ Al ₂	84.4	82.6	509.56	
Ho ₂ (Co _{0.4} Fe _{0.6}) ₁₅ Al ₂	84.8	83.0	516.89	
Ho ₂ (Co _{0.2} Fe _{0.8}) ₁₅ Al ₂	85.0	83.3	521.21	
Ho2Fe15Al2	85.3	83.2	524.27	

Table 1. The lattice parameters *a*, *c* and unit cell volume *v* of $Ho_2(Co_{1-x}Fe_x)_{15}Al_2$ compounds.

3.2. Magnetization and Curie temperature

Figure 3 shows the saturation magnetization versus *x* curve of Ho₂(Co_{1-x}Fe_x)₁₅Al₂ compounds measured at 1.5 K. The saturation moment μ_s was obtained from fitting the data of M(H)versus *H* by using the law of approach to saturation and listed in table 2. It is found that the saturation moment at 1.5 K increases initially and then decreases with increasing Fe content, attaining a maximum value of 10.75 μ_B fu⁻¹ at x = 0.8. The compositional dependence of the saturation moment μ_s in Ho₂(Co_{1-x}Fe_x)₁₅Al₂ compounds behaves in a manner closely



Figure 1. X-ray diffraction patterns at room temperature of un-oriented $Ho_2(Co_{1-x}Fe_x)_{15}Al_2$ powder samples with x = 0.2, 0.4, 0.6.

related to that of the Co–Fe binary alloys [13] and other $R_2(Co_{1-x}Fe_x)_{17}$ compounds [9, 10]. The substitution of a small amount of Al or Ga only slightly decreases the value of saturation moment [11, 12].

Table 2. Saturation moment at 1.5 K (\pm 1%), Fe magnetic moment, Curie temperature T_C (\pm 5 K), spin reorientation temperature T_{sr} (\pm 5 K), Fe magnetic moment and easy magnetization type (EMT) at room temperature of Ho₂(Co_{1-x}Fe_x)₁₅Al₂ compounds.

Compounds	M_s (emu g ⁻¹)	$\mu_s \ (\mu_B \ \mathrm{fu}^{-1})$	T_C (K)	T_{sr} (K)	$\mu_{Fe} \left(\mu_B \right)$	EMT
Ho ₂ Co ₁₅ Al ₂	3.27	0.74	940	455	_	Plane
Ho ₂ (Co _{0.9} Fe _{0.1}) ₁₅ Al ₂	_	_	930	381	_	Plane
Ho ₂ (Co _{0.8} Fe _{0.2}) ₁₅ Al ₂	17.4	3.93	919	406	2.45	Plane
Ho ₂ (Co _{0.7} Fe _{0.3}) ₁₅ Al ₂	_	_	908	511	_	Plane
Ho ₂ (Co _{0.6} Fe _{0.4}) ₁₅ Al ₂	31.71	7.10	874	_	2.44	Plane
Ho ₂ (Co _{0.4} Fe _{0.6}) ₁₅ Al ₂	45.12	10.02	773	_	2.41	Plane
Ho ₂ (Co _{0.2} Fe _{0.8}) ₁₅ Al ₂	48.76	10.75	635	_	2.22	Plane
$Ho_2Fe_{15}Al_2$	36.57	8.00	411	—	1.87	Plane

For heavy rare earth element Ho, the total rare earth moment is coupled antiparallel to the 3d moment [31]. As Al is nonmagnetic, the saturation moment μ_s of Ho₂(Co_{1-x}Fe_x)₁₅Al₂ compounds can be described as:

$$\mu_s = 15\mu_{Fe-Co} - 2\mu_{Ho}.$$
 (1)



Figure 2. The lattice parameters *a*, *c* and unit cell volume *v* of Ho₂(Co_{1-x}Fe_x)₁₅Al₂ compounds as a function of concentration *x*.

Here μ_{Fe-Co} is the average moment per Fe and Co atom and μ_{Ho} is the average moment per Ho atom. Considering the rare earth is strongly local, one can assume $\mu_{Ho} = 10 \mu_B$ and it is unchanged with composition. Therefore, the average moment of Fe and Co atoms μ_{Fe-Co} can be obtained via equation (1). Furthermore, the result of polarized neutron diffraction indicated that in a crystal Fe–Co alloy the magnetic moment on the cobalt atom is shown to be broadly unaffected by alloying, while the iron moment increases with the decrease of Fe content [13]. The relation of μ_{Fe-Co} and Fe content *x* can be described as:

$$\mu_{Fe-Co} = \mu_{Co}(1-x) + \mu_{Fe}x.$$
(2)

As $\mu_{Ho} = 10 \ \mu_B$, the value of μ_{Co} in Ho₂Co₁₅Al₂ is 1.40 μ_B . Assuming that μ_{Co} does not change with Fe content and that the value of μ_{Co} in Ho₂(Co_{1-x}Fe_x)₁₅Al₂ is 1.40 μ_B , the μ_{Fe} of Ho₂(Co_{1-x}Fe_x)₁₅Al₂ compounds can be attained via equation (2) and listed in table 2. The average magnetic moment of an Fe atom decreases from 2.45 μ_B for x = 0.2 to 1.87 μ_B for x = 1.0. This is likely attributable to band-filling effects. Low Co content can cause a decrease in hole numbers of the Fe positive band above the Fermi energy level and an increase in the total magnetic moment of Fe. It seems that the majority spin d-band saturates at about the same relative Co–Fe ratio; but that the minority band filling, which determines the over-all moment, is determined by the position of minimal density of states and exchange splitting. These are clearly structure sensitive.

The Fe-concentration dependence of Curie temperature T_C is shown in figure 4. The

diagram

for



Figure 3. The magnetization m_s as a function of **Figure 4.** Magnetic phase concentration *x* at 1.5 K. Ho₂(Co_{1-x}Fe_x)₁₅Al₂ compounds.

substitution of Fe for Co leads to a monotonic decrease of Curie temperature. Generally speaking, the value of the Curie temperature in these materials is primarily dependent on the magnitude of the exchange coupling between transition metal (TM) atoms J_{TT} , which is clearly evidenced in studies of the magnetic properties of rare earth–transition metal compounds. The rare earth–transition metal (RE–TM) coupling J_{RT} and the RE–RE coupling J_{RR} are substantially smaller than J_{TT} in these compounds and J_{RT} is virtually independent of the Al contents [14]. The high-field magnetism measurement of Ho₂Co₁₇, Ho₂Co₁₄Fe₃ and Ho₂Fe₁₇ single crystal [15] shows that the Co–Co interaction is almost as twice the Fe–Fe one and J_{RT} is almost unchanged. Therefore, the Fe substitution for Co in Ho₂(Co_{1-x}Fe_x)₁₅Al₂ results in the decrease of Curie temperature.

3.3. Magnetocrystalline anisotropy and magnetic phase transition

The temperature dependence of the magnetization for $Ho_2(Co_{1-x}Fe_x)_{15}Al_2$ compounds is plotted in figure 5. It is seen that a peak or an anomaly is visible below the Curie temperature T_C for x < 0.4. Considering the temperature-induced competition between the (Fe, Co) and Ho sublattice anisotropy, this magnetic transition below T_C is a spin reorientation. The corresponding temperature is called T_{sr} . These data from the temperature dependence of the magnetization together with the XRD patterns of magnetically oriented powder samples at room temperature were used to construct the magnetic phase diagram (see figure 4). It can be seen that as the temperature increases, the $Ho_2(Co_{1-x}Fe_x)_{15}Al_2$ compounds show a change of the spin reorientation from plane to axis for x < 0.4, and the T_{sr} has a minimum 381 K at about x = 0.1. The T_{sr} diagram showing a U shape is consistent with the predictions of $Ho_2(Co_{1-x}Fe_x)_{15}Al_2$ by Callen [16].

The spin-reorientation temperature dependence on *x* can be understood in a qualitative way as follows. It is well known that the overall magnetocrystalline anisotropy Ho₂(Co_{1-*x*}Fe_{*x*})₁₅Al₂ compounds consists of two contributions $K_{1,(Fe,Co)}$ and $K_{1,Ho}$ originating from the R and (Fe,Co) sublattices respectively:

$$K_1 = K_{1,(Fe,Co)} + K_{1,Ho} = K_{1,(Fe,Co)} - 3/2\alpha_J \langle r \rangle^2 \langle O_{20} \rangle A_{20}$$
(3)

where α_J is the second order Stevens factors and $\langle r \rangle^2$ the expected ion values of the Hartree– Fock 4f radius. A_{20} is the second order crystal electric field (CEF) coefficient, which reflects the spherical charge distribution outside the 4f shell. The anisotropy of the rare-earth sublattice arises from the coupling between ion orbit magnetic moment and crystal electric field, and in lowest order approximation it can be given by the last term of equation (3). For the Ho³⁺ ion, $\alpha_J < 0$, a negative A_{20} makes $K_{1,Ho}$ negative and thus the Ho sublattice exhibits basal plane anisotropy [32]. The anisotropy of rare-earth elements is influenced strongly by the crystal electric field coefficients, which depends on the atomic configuration, the lattice and positional parameters, the atomic charges and their screening, crystal structure and composition.

The previous results of the single crystal indicate the following fact; the $K_{1,Ho}$ value of Ho sublattice in Ho₂Co₁₄Fe₃ is almost half those of the Ho₂Fe₁₇ and Ho₂Co₁₇ although the sign of the anisotropy constants seems unchanged [15]. In Y_2Co_{17} and Y_2Fe_{17} compounds, both Fe and Co sublattices exhibit the easy plane anisotropy. The anisotropy constants are K_1 , $Co = -70 \text{ J kg}^{-1}$ and K_1 , Fe = -320 J kg^{-1} [33], respectively. The Co sublattice is less anisotropic than the Fe one. So one may expect that Fe substitution for Co should increase the plane anisotropy of the TM sublattice. However, in $Y_2(Co_{1-x}Fe_x)_{17}$ compounds for $0 \le x \le 1$ [9] the negative contribution is diminished below x = 0.04, while for compositions between x = 0.04 and something less than x = 0.55, $K_{1,(Fe,Co)}$ is positive and the anisotropy of the (Fe,Co) sublattice changes from planar to axial. For x > 0.55, $K_{1,(Fe,Co)}$ is again negative. Considering the fact that the contributions of Fe and Co to anisotropy in the same crystalline sites are different: this may due to the preferential substitution of Fe. Inomata [20] has proposed that the 18f site of Co in Y_2Co_{17} makes a positive contributions to the anisotropy, while the 18h, 9d and 6c sites make negative contributions (basal plane). In particular, the 6c site was the largest negative contribution, while in Y₂Fe₁₇ the contribution of the 6c dumbbell site to the anisotropy ($K_{6c,Fe} = 6.48 \times 10^5 \text{ J m}^{-3}$) is of different sign and at least one order of magnitude larger than that in Y₂Co₁₇ ($K_{6c,Co} = -0.42 \times 10^5$ J m⁻³) [22]. So when small amounts of Fe occupy the dumbbell 6c sites, the negative contribution will be diminished. Previous studies show that Fe substitution is more or less preferential. In $Y_2(Co_{1-x}Fe_x)_{17}$ compounds, the substitution of Fe takes the order of 6c dumbbell, 18h and finally 18f site [18]. In Nd₂(Co_xFe_{1-x})₁₇ compounds, the neutron diffraction result [19] also shows that 6c transition metal sites are preferentially occupied by Fe ions. So in Ho₂(Co_{1-x}Fe_x)₁₅Al₂ compounds the anisotropy changes from planar to axial at small contents of Fe may be due to the preferential occupation in the dumbbell 6c sites. As the Fe content increases further and occupies the 18f site as well as the 6c site, the positive contribution will be diminished and the easy magnetization direction (EMD) changes to the plane.

In addition, the absolute value of negative $K_{1,(Fe,Co)}$ may be decreased by the substitution of the non-magnetic Al atoms at the 6c site. The neutron diffraction measurements and nuclear magnetic resonance (NMR) results indicate a preferential occupation of Al for the 6c site [20, 24] in $R_2Co_{17-x}Al_x$ compounds. As mentioned before, the 6c site has the largest negative contribution for the plane anisotropy of Co sublattice. Therefore, the substitution of the nonmagnetic Al atoms at the 6c site decreases the absolute value of negative $K_{1,(Fe,Ca)}$ and induces the easy-axis anisotropy of the (Fe, Co) sublattice. In $Y_2Co_{17-x}Al_x$ compounds [20, 23], the substitution of Al for Co leads to a sign reversal of the magnetocrystalline Co-sublattice anisotropy from easy-plane anisotropy for low Al concentration to easy-axis anisotropy for higher Al concentration. In the series of $Ho_2Co_{17-x}Al_x$ compounds, the spin reorientation occurs when x = 2 [7], as is in agreement with our result. On the other hand, the band structure calculations demonstrated that the second crystal field coefficient A_{20} is determined predominantly by the rare-earth 5d and 6p valence electron charge asphericity [25, 26]. It is strongly influenced by the content of non-magnetic atoms, because of the hybridization of the rare-earth 5d and 6p valence electrons with the valence electrons of its neighbouring atoms. Thus, when Al atoms preferentially substitute the neighbouring sites of a rare-earth



Figure 5. Temperature dependence of the magnetization of $Ho_2(Co_{1-x}Fe_x)_{15}Al_2$ compounds.

site, it is possible that substantial changes occur in the magnitude and sign of valence electron asphericity of this rare-earth atom.

The anisotropy switch of $Ho_2(Co_{1-x}Fe_x)_{15}Al_2$ compound temperature results from the competition between the (Fe, Co) and Ho sublattice anisotropy. According to the l(l + 1)/2power law on the temperature dependence of magnetocrystalline anisotropy [17], the anisotropy constant $K_{1,H_0}(T)(l=2) = K_{1,H_0}(0)m(T)^{l(l+1)/2}$, where l is the order of the anisotropy constant and m(T) is the magnetization of a sublattice. The reduced magnetization m(T) = $M(T)/M_0$ is given by the reduced hyperbolic Bessel function $\hat{I}_{5/2}(m(T))$, which for l = 2goes as m^3 for $T \ll T_C$, where $m \cong 1$, and recedes to m^2 for $T \cong T_C$, where $m \cong 0$. For the rare earth, since the RT exchange is small, M drops off rapidly with increasing temperature, and $K_{1,Ho}(T) = K_{1,Ho}(0)\hat{I}^{5/2}(m(T))$ drops off even more rapidly. At low x content, where T_C is large, the rare-earth sublattice of the $Ho_2(Co_{1-x}Fe_x)_{15}Al_2$ compounds is largely aligned at room temperature, and its anisotropy dominates. So at room temperature the total anisotropy of Ho₂(Co_{1-x}Fe_x)₁₅Al₂ is plane, only at higher temperature can the axis anisotropy of the Fe–Co sublattice overcome the plane anisotropy of the R sublattice. For higher Fe content, T_C is lower; the rare-earth sublattice magnetization, M(x), and hence the rare-earth anisotropy $K_{1,Ho}(m(x))$ vary with Fe content, rapidly dropping in magnitude with increasing substitution of iron for cobalt. The axis anisotropy is enhanced; therefore, the reorientation temperature T_{sr} decreases first. As Fe content increases from about 0.1 to 0.4, the axis anisotropy of the

5546 Jing-yun Wang et al

(Fe, Co) sublattice is weakened by Fe substitution; the reorientation temperature T_{sr} sharply increases. Finally, at content above 0.4, both the rare-earth sublattice and (Fe, Co) sublattice have plane anisotropy.

Acknowledgments

The authors wish to express their gratitude to T S Ning and M Hu for their assistance in works of magnetic measurement and experiment. This work was supported by the National Natural Sciences Foundation of China.

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