Vol 11 No 2, February 2002
1009-1963/2002/11(02)/0174-04

Chinese Physics

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Structural and magnetic properties of $Dy_2Co_{17-x}Mn_x$ compounds*

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(Received 10 September 2001; revised manuscript received 18 October 2001)

Using x-ray diffraction and magnetic measurements, we have studied the structural and magnetic properties of $Dy_2Co_{17-x}Mn_x$ (x=0∼5) compounds with a rhombohedral Th_2Zn_{17}-type structure. With an increasing Mn concentration x, the unit-cell volume V was found to increase linearly. The Curie temperature T_C decreases linearly, and the saturation magnetization M_s at 5K first increases slightly for x <1, then decreases rapidly for x >1 with a further increase of Mn concentration x. In compounds for x=1∼3, a spin reorientation was found. A magnetic diagram of the compounds is given.

Keywords: 2:17-type compounds, crystal structure, magnetism
PACC: 7530, 7590

1. Introduction

In recent years, R_2Fe_{17} rare-earth ferrum intermetallic compounds and their carbides and nitrides with excellent intrinsic magnetic properties have attracted much attention as new possible permanent magnets. Investigations indicate that the partial substitution of Ga, Al or Si for the transition metal not only significantly improves the Curie temperature and the uniaxial magnetocrystalline anisotropy field of R_2Fe_{17} compounds,[1−7] but also changes the compounds from the easy-plane type to the uniaxial type. Recently, much attention has been paid to 2:17 Co-based compounds and partial substitution of the 3d transition metal elements for Co. It is found that the substitution of the transition metal elements can enhance the magnetocrystalline anisotropy field of the 2:17-type compounds. A previous study has indicated that the Mn atom has a complex magnetic behaviour in Mn-based compounds and alloys. The magnetic moment arrangement of Mn and Co atoms is dramatically related to the difference between atoms in separation and structural configuration. In the search for permanent-magnet materials with excellent intrinsic magnetic properties, it is very important to study the structural and intrinsic magnetic properties of the 2:17-type compounds of the partial substitution of Mn[8] for Co atom. In this paper, we report on a study of the structural and magnetic properties of $Dy_2Co_{17-x}Mn_x$ compounds (x=0∼5).

2. Experimental details

The $Dy_2Co_{17-x}Mn_x$ alloys with x=0∼5 were prepared by arc melting in a high-purity argon atmosphere. The elements used were of at least 99.9% purity. An excess of Dy (5%) and Mn (10%) was added to compensate for loss during the arc melting. The ingots were re-melted at least three times to ensure homogeneity. Subsequently the ingots were sealed in an evacuated quartz tube and annealed under vacuum at 1050°C for 150h, then quenched in water. The crystallographic structure of the ingots was measured using x-ray diffraction (XRD) with CuKα radiation. In order to study the magnetocrystalline anisotropy, orientated samples were prepared by mixing powders with epoxy resin first, and then aligned in a magnetic field of 1T at room temperature. The magnetic properties were measured by a magnetic balance and a superconducting quantum interface device (SQUID).

3. Result and discussion

The XRD patterns indicated that all samples with the composition $Dy_2Co_{17-x}Mn_x$ (x=0∼5) are sin-

*Project supported by the National Natural Science Foundation of China (Grant No. 50071041).

gle phase compounds with a rhombohedral Th_2Zn_{17}-type structure[9] (space group $R\bar{3}m$). The lattice parameters a, c and the unit-cell volume V are listed in Table 1. The unit-cell volume V of the compounds is found to increase linearly with increasing Mn concentration x, as shown in Fig.1. The XRD patterns of the normal-aligned and rotating-aligned samples at room temperature are shown in Fig.2. For $Dy_2Co_{17-x}Mn_x$(x=0~5) compounds, the enhancement of ($hk0$) diffraction peaks and the disappearance of (00l) diffraction peaks for the normal-aligned samples, and the enhancement of (00l) diffraction peaks and the disappearance of ($hk0$) diffraction peaks for rotating-aligned samples, show that the $Dy_2Co_{17-x}Mn_x$ (x=0~1) compounds have their easy magnetization directions (EMDs) in the basal plane at room temperature.

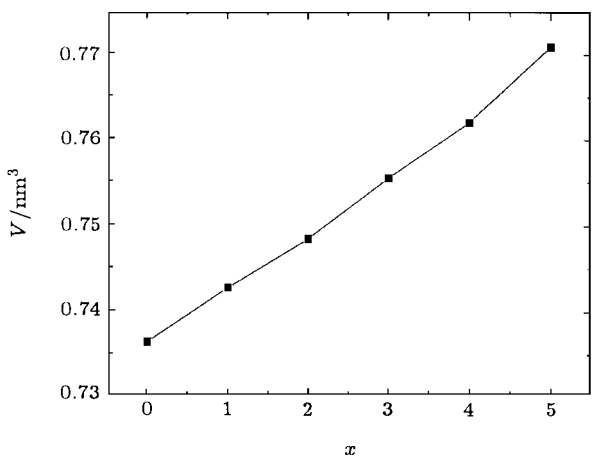


Fig.1. Unit-cell volume V of $Dy_2Co_{17-x}Mn_x$ compounds as a function of Mn concentration x.

The thermal magnetization curves of the $Dy_2Co_{17-x}Mn_x$(x=1~5) compounds are plotted in Fig.3. For x=1~3, the observed anomaly in the magnetization–temperature (M–T) curves is related to the spin reorientation. The temperature at which the peak appears in the M–T curves is called the spin reorientation temperature, T_{sr}. The anisotropy of the compounds changes from the easy-plane type below T_{sr} to the uniaxial type (c-axis) above T_{sr}.

It is well known that the overall magnetocrystalline anisotropy of the $R_2Co_{17-x}Mn_x$ compounds is determined from the sum of the 3d-sublattice and rare-earth R-sublattice anisotropies. The rare-earth sublattice anisotropy is determined by the product $\alpha_J A_{20}$, which is positive for Dy, thus leading to basal plane anisotropy ($K_{Dy} < 0$). The 3d sublattice exhibits a uniaxial anisotropy ($K_{3d} > 0$). The anisotropy constant K_{Dy} of the rare-earth sublattice becomes dominant at low temperatures. The competition between K_{3d} and K_{Dy} results in an overall planar anisotropy of the $Dy_2Co_{17-x}Mn_x$ (x=0~5) compounds. With increasing temperature, the decreasing rate of K_{Dy} is larger than that of K_{3d}, which becomes dominant at a certain temperature. The easy magnetization direction of the $Dy_2Co_{17-x}Mn_x$ compounds changes from basal plane to c-axis.

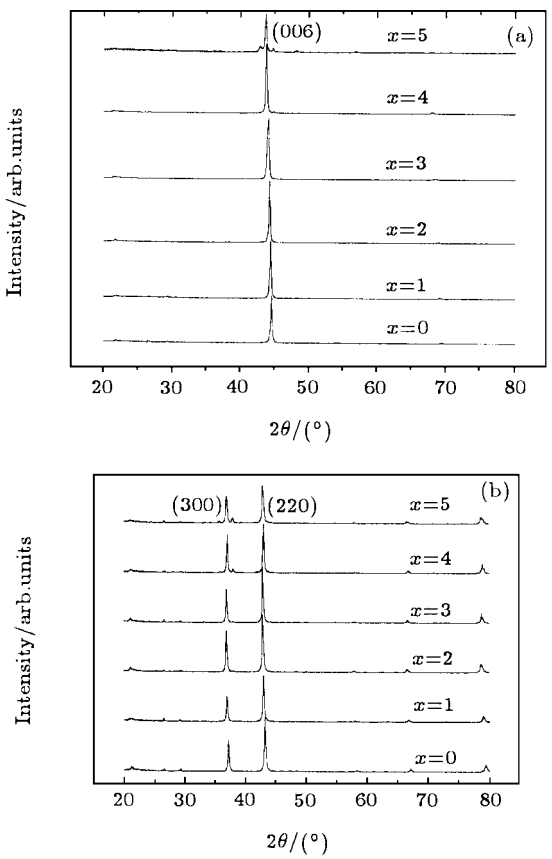


Fig.2. XRD diffraction patterns of the normal (a) and rotating (b) aligned samples of $Dy_2Co_{17-x}Mn_x$.

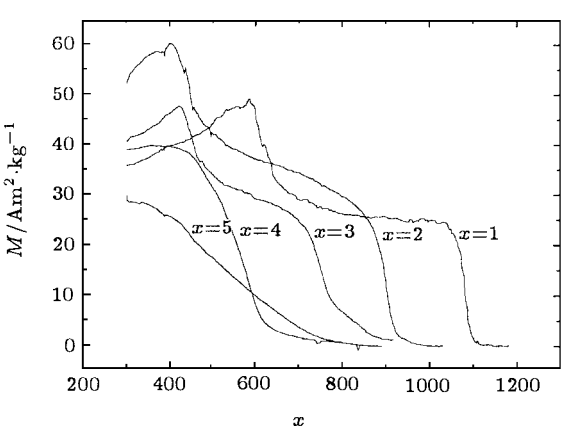


Fig.3. Temperature dependence of the magnetization of $Dy_2Co_{17-x}Mn_x$(x=1~5) compounds.

The magnetization curves of $Dy_2Co_{17-x}Mn_x$ compounds at 300K have been measured by SQUID. Table 1 lists the saturation magnetization M_s, obtained by plotting M–H^{-1} curves and extrapolating H^{-1} to zero. Figure 4 shows M_s as a function of Mn concentration x. It can be seen that M_s increases slightly first, reaches a maximum around x=1, then decreases rapidly with a further increase of Mn concentration. This is due to the fact that Mn atoms have a complex magnetic behaviour. The magnetic moments of Co and Mn atoms are arranged parallel for low Mn concentration (x <1), which results in the increase of the saturation magnetic moment. With increasing Mn concentration (x >1), the magnetic moments of Co and Mn atoms are arranged anti-parallel, even resulting in the disappearance of the Mn atomic magnetic moment at certain sites, and leading to a rapid decrease of the saturation magnetic moment.

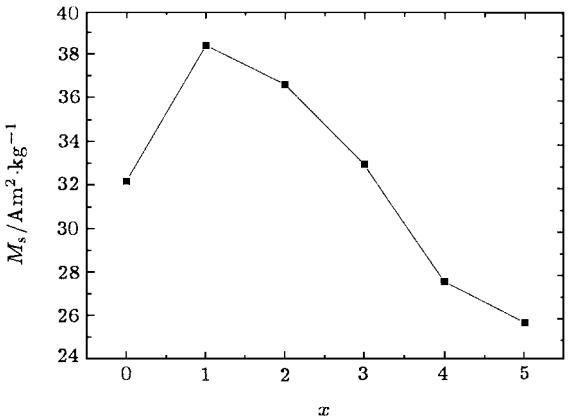


Fig.4. Saturation magnetization M_s of $Dy_2Co_{17-x}Mn_x$ compounds as a function of Mn concentration x (T=300K).

Table 1. The lattice parameters a, c and unit-cell volume V, saturation magnetization M_s at 300K, Curie temperature T_C, spin reorientation temperature T_{sr} of the $Dy_2Co_{17-x}Mn_x$ compounds.

Compounds	a/nm	c/nm	V/nm^3	M_s/Am^2·kg^{-1}	T_C/K	T_{sr}/K
Dy_2Co_{17}	0.8355	1.2179	0.7363	32.19	1215	
$Dy_2Co_{16}Mn$	0.8378	1.2217	0.7427	38.44	1090	625
$Dy_2Co_{15}Mn_2$	0.8399	1.2251	0.7484	36.66	925	435
$Dy_2Co_{14}Mn_3$	0.8415	1.2319	0.7554	33.05	785	465
$Dy_2Co_{13}Mn_4$	0.8405	1.2454	0.7619	27.69	550	
$Dy_2Co_{12}Mn_5$	0.8446	1.2479	0.7709	25.80	470	

Figure 5 shows the magnetic phase diagram of the $Dy_2Co_{17-x}Mn_x$ (x=0∼5) compounds. Table 1 also lists the Curie temperature T_C, obtained by plotting M^2–T curves and extrapolating M^2 to zero, and the spin reorientation temperature T_{sr}. The Curie temperature decreases linearly with increasing Mn concentration x. In rare-earth cobalt intermetallic compounds, the Curie temperature is mainly determined by T–T and R–T exchange interactions. The increase of the Co–Co interatomic distance, caused by the increase of the unit-cell volume V, leads to a decrease of the Co–Co exchange interaction. The Co–Co exchange interaction is partially substituted by the Co–Mn and Mn-Mn exchange interactions, therefore leading to a decrease of the T–T exchange interaction, while the magnetic moments of Gd and Mn atoms are arranged anti-parallel, resulting in the decrease of the R–T exchange interaction. The decrease of the T–T and R–T exchange interactions result in a rapid decrease of the Curie temperature with increasing Mn concentration x. T_{sr} first decreases with increasing Mn concentration x and then slightly increase. The decrease of T_{sr} is attributed to the enhancement of the Co sublattice uniaxial anisotropy with increasing Mn concentration x. With a further increase of Mn concentration, Mn atoms preferentially take the sites contributing to uniaxial anisotropy, which leads to a decrease of Co sublattice uniaxial anisotropy. Thus, the decrease of the Co sublattice uniaxial anisotropy results in the increase of T_{sr}.

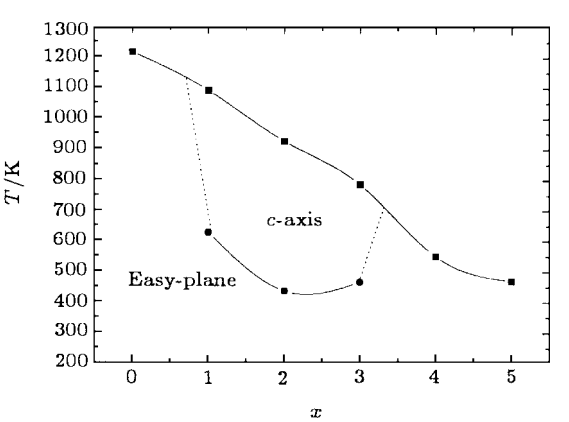


Fig.5. Magnetic phase diagram of $Dy_2Co_{17-x}Mn_x$ compounds.

References

[1] Shen B G, Cheng Z H, Liang B, Guo H Q and Zhang J X 1995 *Appl. Phys. Lett.* **67** 1621

[2] Liang B, Shen B G, Wang F W, Zhao T Y, Cheng Z H, Zhang S Y, Gong H Y and Zhan W S 1997 *J. Appl. Phys.* **82** 3452

[3] Cheng Z H, Shen B G, Zhang J X, Liang B, Guo H Q and Kronmuller H 1997 *Appl. Phys. Lett.* **70** 3467

[4] Zhang L G *et al* 1997 *Acta. Phys. Sin.* **11** 2241 (in Chinese)

[5] Zhang S Y *et al* 1998 *Acta Phys. Sin.* (*Overeas Edition*) **7** 518

[6] Zhang H W *et al* 1999 *Acta. Phys. Sin.* **48** 163 (in Chinese)

[7] Gao S X *et al* 1999 *Acta. Phys. Sin.* **48** 1751 (in Chinese)

[8] Stayanarayana M V, Fujii H and Wallace W E 1984 *J. Magn. Magn. Mater.* **40** 241

[9] Strnat K J 1988 *Handbook of Magnetic Materials* vol 4, ed K H J Buschow, p 131