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Magnetic properties of $\text{Y}(\text{Fe}_{0.8}\text{M}_{0.2})_{11.3}\text{Nb}_{0.7}$ compounds with $\text{M}=\text{Mn}, \text{Fe}, \text{Co}, \text{Ni}, \text{Al}, \text{and Ga}$

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Structural and magnetic properties of $\text{Y}(\text{Fe}_{0.8}\text{M}_{0.2})_{11.3}\text{Nb}_{0.7}$ compounds with $\text{M}=\text{Mn}, \text{Fe}, \text{Co}, \text{Ni}, \text{Al}$, and Ga have been investigated. The x-ray diffraction patterns of aligned samples show that, for $\text{M}=\text{Mn}, \text{Fe}, \text{Co}, \text{Ni}$, the compounds exhibit easy axis anisotropy while for $\text{M}=\text{Al}$ and Ga the compounds have planar anisotropy. The anisotropy fields of $\text{Y}(\text{Fe}_{0.8}\text{Mn}_{0.2})_{11.3}\text{Nb}_{0.7}$ increase monotonously with decreasing temperature. The influence on the Curie temperature and the magnetization due to the various substitutional atoms are discussed. © 1997 American Institute of Physics. [S0021-8979(97)20808-X]

I. INTRODUCTION

Since Coey and Sun¹ found that the introduction of nitrogen can strongly influence the intrinsic magnetic properties of R_2Fe_{17} -type compounds, the introduction of N and other elements in rare earth (R) transition-metal (T) compounds has attracted much attention. For instance, it has been found² that $\text{Sm}_3(\text{Fe}, \text{Ti})_{29}\text{N}_y$ exhibits potentially interesting permanent magnet properties. Recently Hu *et al.*³ have found that the ThMn_{12} structure can be stabilized with a small amount of Nb ($x=0.65$). A small amount of stabilization element is of interest, since in this case the magnetization is only slightly reduced, which may result in an improvement of the performance. The magnetic anisotropy in these compounds results from the R and T sublattices. However, in most cases, the anisotropy of the T sublattice is much weaker than that of the R sublattice and can be neglected. Recently, it has been found⁴ that the easy magnetization direction at room temperature can be changed from plane to c axis by the substitution of Ga or Al for Fe in the 2:17 structure. Huang and Ching⁵ have performed first principles calculations on the influence of Al substitution in 2:17 compounds on the electronic structure and magnetism.

In the present article, we have studied the influence of substitution of various elements for Fe on the Curie temperature and the magnetic anisotropy. In order to be able to investigate only the contribution of the anisotropy of the T sublattice we have studied the Y compounds, since Y is non-magnetic.

II. EXPERIMENT

Compounds with composition $\text{Y}(\text{Fe}_{0.8}\text{M}_{0.2})_{11.3}\text{Nb}_{0.7}$ with ($\text{M}=\text{Mn}, \text{Fe}, \text{Co}, \text{Ni}, \text{Al}$, and Ga) were prepared by arc-melting of 99.9% pure materials in a purified argon atmosphere. Additional amounts of Y and Mn were added to compensate the loss during the melting. X-ray diffraction patterns of powder samples that were magnetically aligned at room temperature were used to determine the easy magnetization direction of the compounds. The Curie temperature T_c was determined from σ^2-T plots by extrapolating σ^2 to zero.

The magnetic isotherms were recorded with the external field applied either parallel or perpendicular to the alignment direction of the samples using a superconducting quantum interference device (SQUID) magnetometer. The latter samples were prepared in cylindrical form by aligning powder particles at room temperature in a magnetic field of 1 T applied parallel and perpendicular to the cylinder axis and by fixing their direction with epoxy resin. Above 77 K, the anisotropy fields for the compounds with easy axis anisotropy were determined by the means of the singular point detection (SPD) technique. The anisotropy constants K_i of the compounds with easy axis anisotropy at 5 K have been obtained by fitting the magnetization curve measured along the hard direction.

III. RESULTS AND DISCUSSION

The x-ray diffraction patterns show that all the compounds crystallize in the ThMn_{12} -type of structure while for $\text{M}=\text{Al}$ and Ga , a small amount of second phase is found in form of Fe_2Nb and Nb_5Ga_3 , respectively. In Table I, it can be seen that substitution of Al and Ga for Fe leads to a clear increase of the lattice constants while the replacement of Fe

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TABLE I. Lattice constants a and c , unit cell volume V , Curie temperature T_c , saturation magnetization M_s at 5 K, anisotropy field B_a at room temperature (RT), 77 K, and 5 K, easy magnetization direction at 300 K, anisotropy constants K_1 and K_2 at 5 K of $Y(Fe_{0.8}M_{0.2})_{11.3}Nb_{0.7}$ compounds.

M	a (Å)	c (Å)	V (Å ³)	M_s ($\mu_B/f.u.$)	T_c (K)	Anisotropy at RT	$B_a(T)$			$K_i(5\text{ K})$	
							RT	77 K	5 K	(J/kg)	(J/kg)
Fe	8.53	4.80	349	18.0	507	uniaxial	1.65	2.63	3.47 ^a	47	92
Ni	8.49	4.79	345	15.7	614	uniaxial	0.81	0.95	1.56 ^a	29	30
Co	8.51	4.79	347	17.3	739	uniaxial	1.86	2.48	3.14 ^a	42	83
Mn	8.55	4.80	351	5.6	331	uniaxial	0.23	0.72	1.65 ^a	20	21
Al	8.59	4.84	357	...	367	planar	^b	^b	...	...	...
Ga	8.59	4.84	357	...	475	planar	^b	^b	...	...	...

^aObtained from fitting magnetization curves along the hard direction.

^bEasy plane anisotropy and B_a cannot be obtained by SPD.

by other elements results in a slight change of a and c . The changes of the lattice constants can be understood in terms of the different atomic radii of the various elements.

The Curie temperatures are also listed in Table I. It can be seen that substitution of Co for Fe leads to the largest increase of T_c while substitution of Mn results in the strongest decrease. In R-T compounds three exchange interactions play a role: the R-R, the T-T, and the R-T interaction. Among them the T-T interaction is the strongest and in 1:12 or 2:17 compounds T_c is mainly determined by this interaction.⁶ The Co-Co exchange interaction is the largest. Substitution of Mn for Fe may lead to a negative exchange interaction. Al and Ga are nonmagnetic elements and do not contribute to the exchange interaction. The increase of T_c due to substitution of Ni for Fe may be associated with preferential occupation of certain sites by Ni atoms.⁷

The x-ray diffraction patterns of magnetically aligned $Y(Fe_{0.8}M_{0.2})_{11.3}Nb_{0.7}$ samples are shown in Fig. 1. It can be seen that for $M=Ni$, the (002) peak in the diffraction pattern is dominant, which suggests that the easy magnetization direction is along the c axis in this compound. The cases with $M=Mn$ and Co are similar. However, for $M=Al$ and Ga , the (301) and (400) peaks are dominant, which means that the anisotropy has changed from easy axis to planar type. Apparently, substitution of Al and Ga favors planar anisotropy and weakens the easy axis anisotropy. This result is opposite to the conclusion for R_2Fe_{17} compounds, for which it has been reported that substitution of Al and Ga enhances the easy axis anisotropy.⁸

Figure 2 shows the SPD signals at 77 K as a function of applied field for $Y(Fe_{0.8}M_{0.2})_{11.3}Nb_{0.7}$ compounds with $M=Mn, Fe, Co$, and Ni . The position of the peak in the SPD signal corresponds to the anisotropy field. The values of the anisotropy fields for the various substitution elements, as derived from the SPD measurements, are listed in Table I. It can be seen that substitution of Co for Fe does not lead to an appreciable change of the anisotropy field, whereas substitution of Mn and Ni leads to a clear decrease of the anisotropy field at 77 K: from 2.63 to 0.95 T for Ni substitution and from 2.63 to 0.72 T for Mn substitution. Since Y is nonmagnetic, the contributions to the anisotropy in the $Y(Fe_{0.8}M_{0.2})_{11.3}Nb_{0.7}$ compounds are entirely due to the Fe and M atoms. In intermetallic compounds with the $ThMn_{12}$ structure, the Fe sublattice favors easy axis anisotropy and the Co sublattice, planar anisotropy. Substitution of Mn and

Ni for Fe leads to a distinct decreases in easy axis anisotropy. In the 1:12 compounds, there are three different transition-metal sites, $8i$, $8j$, and $8k$. Hu *et al.*⁹ have reported that the contribution to the magnetization and the anisotropy of these three sites is different. Therefore, it is reasonable to assume that the effects found upon substitution of Mn, Co, and Ni may be related to preferential occupation of the various sites by these atoms.

Figure 3 shows the SPD signal at various temperatures as a function of the applied magnetic field for $Y(Fe_{0.8}Mn_{0.2})_{11.3}Nb_{0.7}$. The anisotropy field B_a decreases with increasing temperature. With increasing temperature, the values of B_a decrease from 0.72 T at 77 K to 0.23 T at 300 K.

FIG. 1. X-ray diffraction patterns of aligned samples of $Y(Fe_{0.8}M_{0.2})_{11.3}Nb_{0.7}$ compounds with $M=Ni, Ga$, and Al .

FIG. 2. The SPD signal at 77 K as a function of applied magnetic field for $Y(Fe_{0.8}M_{0.2})_{11.3}Nb_{0.7}$ compounds with $M=Mn, Fe, Co,$ and Ni .

The magnetization curves at 5 K of magnetically aligned samples with easy axis anisotropy measured along the hard direction and of free powder samples are shown in Fig. 4. It is seen that the substitution of the various atoms for Fe leads to a decrease of the magnetization at 5 T. As has been shown by Huang *et al.*,¹⁰ the anisotropy constant K_i can be derived from the magnetization curves measured with the magnetic field applied perpendicular to the alignment direction, taking into account the imperfect alignment of the powder particles in the samples. The anisotropy constants K_1 and K_2 at 5 K, obtained by fitting magnetization curves of the compounds with easy axis anisotropy are also listed in Table I. The cor-

FIG. 3. The SPD signal at various temperatures as a function of the applied magnetic field for $Y(Fe_{0.8}M_{0.2})_{11.3}Nb_{0.7}$.

FIG. 4. Experimental (empty symbols) and calculated (lines) magnetization curves along the hard direction at 5 K of $Y(Fe_{0.8}M_{0.2})_{11.3}Nb_{0.7}$ compounds with $M=Mn, Fe, Co,$ and Ni , and magnetizations curves of $Y(Fe_{0.8}M_{0.2})_{11.3}Nb_{0.7}$ measured on free powder samples with $M=Mn, Fe, Co, Ni, Al,$ and Ga (filled symbols).

responding anisotropy field at 5 K can be obtained by $B_a/\mu_0 = (2K_1 + 4K_2)/M_s$. The calculated anisotropy fields are also listed in Table I. It can be seen that the substitutions of both Ni and Mn for Fe leads to a clear decrease of K_1 and K_2 .

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