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Magnetic anisotropy of $R_2Co_{14}B_{1-x}C_x$ compounds (R≡Pr, Nd)

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Abstract

High field magnetization curves have been measured at 4.2 K on magnetically aligned $R_2Co_{14}B_{1-x}C_x$ (R≡Pr, Nd; $x=0.0$, 0.2 and 0.5) samples with the magnetic field applied parallel and perpendicular to the alignment direction. It is found that, for both the praseodymium and the neodymium compounds, the saturation magnetization decreases on substitution of carbon for boron. In the $Nd_2Co_{14}B_{1-x}C_x$ system, the critical field of the first-order magnetization process (FOMP) increases with increasing carbon content. No FOMP is observed in the $Pr_2Co_{14}B_{1-x}C_x$ system. The temperature dependence of the a.c. susceptibility shows that in $Nd_2Co_{14}B_{1-x}C_x$ the spin reorientation temperature decreases with increasing carbon content. A spin reorientation phenomenon is also observed in $Pr_2Co_{14}B_{0.8}C_{0.2}$ and in $Pr_2Co_{14}B_{0.5}C_{0.5}$. Also in these compounds, the spin reorientation temperature decreases with increasing carbon content.

1. Introduction

$R_2T_{14}B$-based compounds have been extensively studied in recent years [1]. In particular, substitution of other elements on the R and T sublattices has received much attention. As for as substitution for boron in $R_2Fe_{14}B$ compounds is concerned, until now only carbon has been reported as possible substitution element for boron in this type of compound [1, 2]. All published results show the Curie temperature to decrease on carbon substitution, whereas the spin reorientation temperature and the anisotropy field increase. The $R_2Fe_{14}C$ compounds were prepared and investigated for the first time by Buschow *et al.* [3]. All rare earth elements which form $R_2Fe_{14}B$ compounds

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also form $R_2Fe_{14}C$, except yttrium, lanthanum, cerium and ytterbium. Some substitutions in $R_2Fe_{14}C$ compounds, on both the R and the Fe sublattices, have been performed [4].

Until now, not much work has been done on substitution of carbon for boron in $R_2Co_{14}B$ compounds [5]. The $R_2Co_{14}B$ compounds, having the same structure as the $R_2Fe_{14}B$ compounds, have much higher Curie temperatures than the $R_2Fe_{14}B$ compounds [6–9]. The magnetic anisotropy is also different: in $R_2Fe_{14}B$, the iron sublattice exhibits easy-axis anisotropy, whereas in $R_2Co_{14}B$, the anisotropy of the cobalt sublattice is of easy-plane type. Carbon substitution for boron in $R_2Fe_{14}B$ compounds leads to a decrease in the iron moment, which may be attributed to an increased filling of the 3d band. In this respect, it is also interesting to investigate the effect of carbon substitution in $R_2Co_{14}B$ compounds. Sanchez *et al.* [5] have investigated the dependence of the Curie temperature on the cobalt content in $Dy_{15}(Fe_{1-x}Co_x)_{77}C_8$ with $0 \leqslant x \leqslant 0.5$ and found a strong increase in the Curie temperature with increasing cobalt content.

In the present work, the magnetic anisotropy of two systems, $Pr_2Co_{14}(B,C)$ and $Nd_2Co_{14}(B,C)$, has been investigated.

2. Experimental procedure

Samples were prepared by melting together appropriate amounts of pure rare earth, pure cobalt and Co–B (15.6% B) or Co–C (3.9% C) alloy in an arc furnace under a purified-argon atmosphere. The ingots were wrapped in tantalum foil and sealed into an evacuated quartz tube, filled with high purity argon. They were annealed for two weeks at 900 °C. X-ray diffraction showed the main phase to possess the $Nd_2Fe_{14}B$-type tetragonal structure and small amounts of second phase to be present. Aligned samples were prepared by mixing a fine powder of the compounds with epoxy resin and by letting it solidify in a cylindrical holder in an external field of about 1 T.

High field magnetization curves were measured at 4.2 K in the 40 T High Field Facility at the University of Amsterdam. Magnetization curves were measured by applying external fields parallel and perpendicular to the alignment direction. The spontaneous magnetization of the compounds was derived by extrapolating the parallel magnetization curve to zero field. A.c. susceptibility measurements were carried out in the temperature range from 4.2 to 290 K on polycrystalline bulk samples.

3. Results and discussion

3.1. $Nd_2Co_{14}B_{1-x}C_x$

Figure 1 shows the high field magnetization curves of $Nd_2Co_{14}B_{1-x}C_x$ compounds measured at 4.2 K. The magnetization curves have been measured with the field applied parallel and perpendicular to the alignment direction

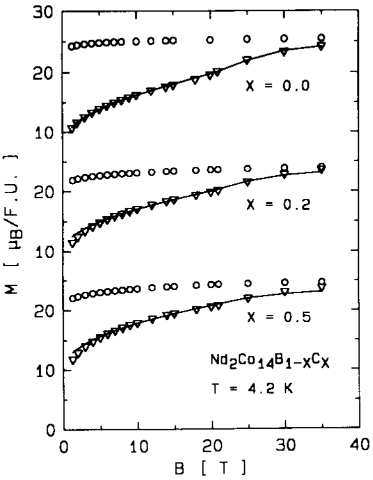


Fig. 1. High field magnetization curves of $Nd_2Co_{14}B_{1-x}C_x$ compounds for the field applied parallel (○) and perpendicular (▽) to the alignment direction.

TABLE 1

Magnetic properties of $Nd_2Co_{14}B_{1-x}C_x$ compounds at 4.2 K

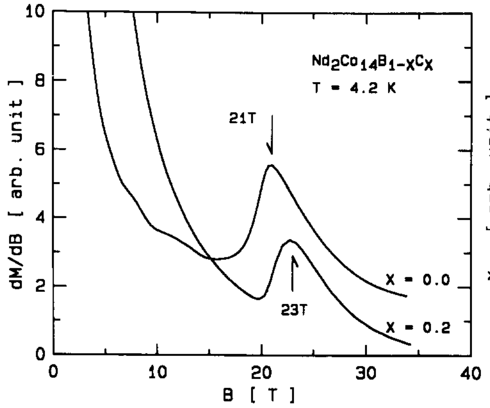
	M_s (μ_B (formula unit)$^{-1}$)	M_R (μ_B ion^{-1})	M_{Co} (μ_B ion^{-1})	B_{tr} (T)	T_{sr} (K)
$Nd_2Co_{14}B$	24.9	3.27	1.31	21	≈40
$Nd_2Co_{14}B_{0.8}C_{0.2}$	23.9	3.27	1.24	23	≈20
$Nd_2Co_{14}B_{0.5}C_{0.5}$	22.7	3.27	1.22		
$Pr_2Co_{14}B$	24.3	3.20	1.27		
$Pr_2Co_{14}B_{0.8}C_{0.2}$	24.0	3.20	1.25		105
$Pr_2Co_{14}B_{0.5}C_{0.5}$	23.2	3.20	1.21		85

of the samples. It is found that the spontaneous magnetization M_s decreases with increasing carbon content (Table 1). By assuming that the rare earth ion moment possesses the free ion value, the magnetic moment per cobalt ion can be derived from the spontaneous magnetization. The magnetic moment per cobalt ion decreases from $1.31\mu_B$ for $Nd_2Co_{14}B$ to $1.22\mu_B$ for $Nd_2Co_{14}B_{0.5}C_{0.5}$. This shows that the influence of carbon substitution for boron on the magnetic moment of the transition metal ions is qualitatively the same for $R_2Fe_{14}B$ and $R_2Co_{14}B$ compounds. This reduction in the transition metal moment can be understood in terms of an increased filling of the 3d band on carbon substitution.

In the perpendicular magnetization curves of $Nd_2Co_{14}B$ and $Nd_2Co_{14}B_{0.8}C_{0.2}$ (Fig. 1), a phenomenon is observed similar to that found earlier for aligned $Nd_2Fe_{14}B$ samples and which corresponds to a first-order magnetization process (FOMP) [10]. For $Nd_2Co_{14}B$, the FOMP field is about

21 T which is the same as the result in ref. 6. In order to show clearly the influence of carbon substitution on the critical field, the field dependence of the differential susceptibility d*M*/d*B* is shown in Fig. 2 for the perpendicular magnetization curves of $Nd_2Co_{14}B$ and $Nd_2Co_{14}B_{0.8}C_{0.2}$. One can see that the FOMP field clearly increases on carbon introduction which is similar to the effect of carbon introduction in $Nd_2Fe_{14}B$ [4].

The temperature dependence of the a.c. susceptibility of the $Nd_2Co_{14}B_{1-x}C_x$ compounds is shown in Fig. 3. Clear peaks, corresponding to spin reorientation transitions, are observed for $Nd_2Co_{14}B$ and



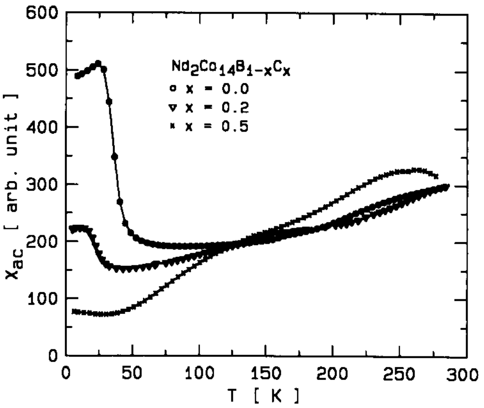
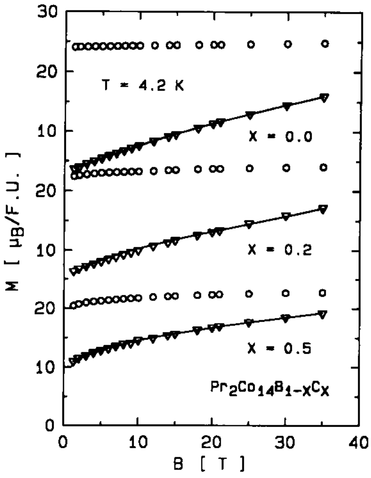


Fig. 2. Field dependence of d*M*/d*B* of the perpendicular magnetization curves of $Nd_2Co_{14}B$ and $Nd_2Co_{14}B_{0.8}C_{0.2}$.

Fig. 3. Temperature dependence of the a.c. susceptibility of $Nd_2Co_{14}B_{1-x}C_x$ compounds.



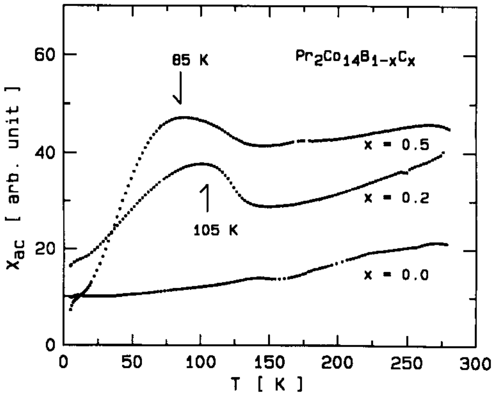


Fig. 4. High field magnetization curves of $Pr_2Co_{14}B_{1-x}C_x$ compounds for the field applied parallel (○) and perpendicular (▽) to the alignment direction.

Fig. 5. Temperature dependence of the a.c. susceptibility of $Pr_2Co_{14}B_{1-x}C_x$ compounds.

$Nd_2Co_{14}B_{0.8}C_{0.2}$. The spin reorientation temperature decreases with increasing carbon content from about 40 K for $x=0$ to near 20 K for $x=0.2$.

3.2. $Pr_2Co_{14}B_{1-x}C_x$

Figure 4 shows the magnetization curves of aligned $Pr_2Co_{14}B_{1-x}C_x$ compounds measured at 4.2 K with the field applied either parallel or perpendicular to the alignment direction. The spontaneous magnetization decreases with increasing carbon content, corresponding to a magnetic moment per cobalt ion that decreases from $1.27\mu_B$ in $Pr_2Co_{14}B$ to $1.21\mu_B$ in $Pr_2Co_{14}B_{0.5}C_{0.5}$. This can again be explained by an increased filling of the 3d band on carbon substitution. One can see that the perpendicular magnetization curves of all three compounds do not saturate even in the highest applied field of 35 T. No FOMP is observed in these compounds. From the experimental results it is difficult to decide whether substitution of carbon for boron has led to a change in the magnetic anisotropy field. From the magnetization curves for the field applied perpendicular to the alignment direction one can see that the zero field extrapolations of the magnetization are different for each carbon content. Since all samples have been aligned at room temperature, this observation indicates a change in the magnetic anisotropy between room temperature and liquid helium temperature in at least two of the compounds.

This change in magnetic anisotropy is confirmed by the a.c. susceptibility measurements shown in Fig. 5. Spin reorientation transitions occur at 105 K in $Pr_2Co_{14}B_{0.8}C_{0.2}$ and at 85 K in $Pr_2Co_{14}B_{0.5}C_{0.5}$. These results show that partial substitution of boron by carbon not only causes a decrease of the magnetization but also strongly influences the magnetocrystalline anisotropy of the $Pr_2Co_{14}B$ compound. A low temperature spin reorientation transition has never been observed before in praseodymium-based compounds of the 2–14–1 type.

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