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Magnetic properties of $Tb_2(Fe, Cr)_{17}$ single crystal

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Abstract. A $Tb_2(Fe, Cr)_{17}$ single crystal with the Th_2Ni_{17}-type structure has been prepared by the Czochralski method and investigated by means of Laue back-reflection, metallographic observation, x-ray diffraction and magnetic measurements. The magnetization curves along the easy and hard directions in the temperature range from 5 K to 293 K are presented. The spontaneous magnetization increases with increasing temperature below 300 K. A magnetohistory effect was observed below 167 K. A first-order magnetization process (FOMP) of type I was also observed below 250 K. The FOMP critical fields occur at about 5.5 T at low temperature.

1. Introduction

Since the structure and magnetic properties of R_2Fe_{17} (R = rare earth) were investigated by Strnat *et al* in 1966 [1], a fair number of studies have been performed on R_2Fe_{17} iron-based intermetallic compounds. The two drawbacks of a rather low Curie temperature and a planar anisotropy at room temperature restrict the use of binary R_2Fe_{17} compounds as permanent magnets. A significant breakthrough was the discovery of the ternary $R_2Fe_{17}C_y$interstitial compound by Gueramian *et al* [2] in 1987 and another was that of $R_2Fe_{17}N_y$ by Coey and Sun [3] in 1990. The carbides and nitrides show a dramatic modification in anisotropy and a pronounced increase in the Curie temperature after the introduction of C and N as interstitial atoms. Unfortunately, the thermal instability of the carbides and nitrides prepared by gas–solid reaction prevents their application as permanent magnets. As an effective method for surmounting the problems, $R_2Fe_{17}C_y$ and $R_2Fe_{1-x}Ga_xC_y$ compounds, which had a high carbon concentration of $y = 2.8$ as well as a high decomposition temperature up to 1100 °C, were prepared by Shen *et al* using the melt-spun technique [4]. Another efficient method is the replacement of Fe atoms by a third element. It was found that the Curie temperature can be markedly increased by Ni [5] and Co [6] substitution. In particular, a drastic change from the planar anisotropy of R_2Fe_{17} to a uniaxial anisotropy at room temperature occurred with an appropriate substitution of Co for Fe [6]. A similar enhancement of the Curie temperatures and drastic changes in anisotropy were also found in investigations in which Fe was substituted

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for with Si [7], Ga [8] and Al [9]. In order to obtain more and accurate information on the structure and magnetic properties of R_2Fe_{17}, a study on a single crystal of R_2Fe_{17} with substitution is necessary. In the present work, a $Tb_2(Fe, Cr)_{17}$ single crystal was prepared using the Czochralski method and its magnetic properties were investigated.

2. Experimental details

The alloy, with a composition of $Tb_{10.16}Fe_{83.60}Cr_{6.24}$ ($Tb_2Fe_{16.46}Cr_{1.23}$), was prepared by arc melting the starting materials Tb, Fe with a purity of 99.9% and Cr with a purity of 99.99%. A slim bar of Tb_2Fe_{17}single crystal with an $\langle 001\rangle$ orientation was used as the seed. The $Tb_2(Fe, Cr)_{17}$ crystal was grown from the original $Tb_{10.16}Fe_{83.60}Cr_{6.24}$ alloy melt using an MCGS-3CZ piece of equipment with growth rates of 15–25 mm h^{-1} and a rotation rate of 30 rpm [10]. As was checked by Laue back-reflection and metallographic observation, the $Tb_2(Fe, Cr)_{17}$ crystal was a single crystal with a Th_2Ni_{17}-type structure. The hard-magnetization direction was fixed using the rotation alignment method [11]. During the fixing procedure, a small spherical crystal sample with a diameter of 2 mm was placed in an applied rotating field. The hard-magnetization direction was fixed as the direction perpendicular to the plane of the rotating field. The easy-magnetization direction was determined as that of the field when the crystal sample was placed freely in the applied field. The Curie temperature was measured using a vibrating-sample magnetometer. At 5 K–293 K the thermomagnetic and magnetization curves along the easy and hard directions were carried out using an extracting-sample magnetometer in fields of up to 6.5 T.

3. Results and discussion

The x-ray diffraction patterns of the powder and the scanning surface perpendicular to the direction of pulling the crystal are presented in figure 1 for $Tb_2(Fe, Cr)_{17}$ single crystal. The

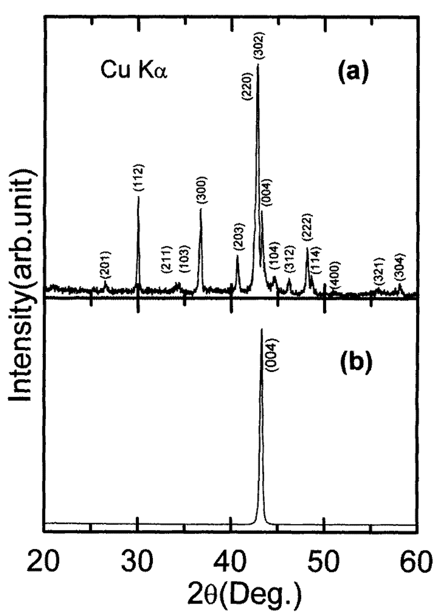

Figure 1. X-ray diffraction patterns of (a) powder and (b) the scanning surface for $Tb_2(Fe, Cr)_{17}$ single crystal, where the surface is perpendicular to the direction of the pulling crystal.

powder patterns of the single crystal in figure 1 clearly show that the crystal has a Th_2Ni_{17}-type structure. The scanning patterns imply that the single crystal was grown along the c-axis since only the line (004) is present in the scanning diffraction pattern. A low-field thermomagnetic trace of the crystal above room temperature is plotted in figure 2. The Curie temperature, as determined from figure 2, is 457 K. The Curie temperature is higher than that of Tb_2Fe_{17}, namely 408 K [12]. According to Givord and Lemaire, there are two types of exchange interaction in the Fe–Fe pairs in the 2:17 structure [13]. The exchange interaction is positive when the Fe–Fe-atom distance is larger than the critical distance of 2.45 Å, while it is negative for smaller distances. Neutron powder diffraction showed for $Y_2Fe_{15}Cr_2$ that the Cr atom preferentially occupies the 4f site [14], which has a negative interaction due to the distance being less than 2.45 Å within the Fe–Fe pairs. The increase in the Curie temperature is reasonable considering that the Cr atom, which has a weaker ferromagnetism than Fe, preferentially replaces the Fe atom at the 4f position in $Tb_2(Fe, Cr)_{17}$ single crystal.

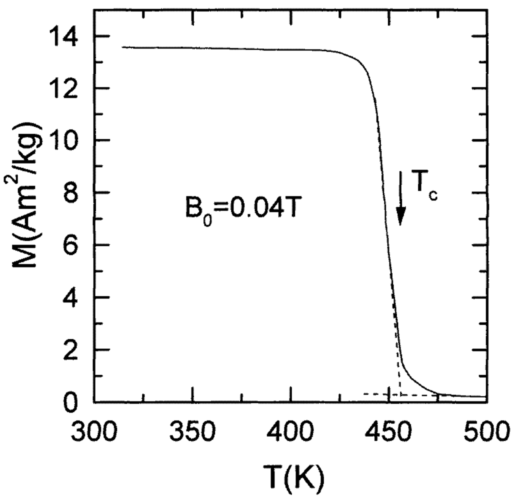

Figure 2. A thermomagnetic scan trace measured by a VSM above room temperature in a low field of 0.04 T for $Tb_2(Fe, Cr)_{17}$ single crystal.

The low-temperature field-cooling (FC) and zero-field-cooling (ZFC) thermomagnetic curves along the easy direction in a low field are shown in figure 3. It can be seen that the ZFC curve deviates from the FC curve at about 200 K and reaches the largest deviation at

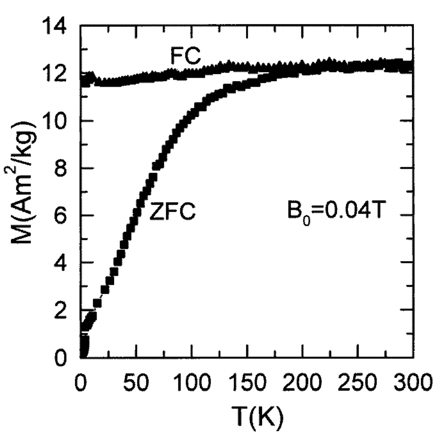

Figure 3. Field-cooling (FC) and zero-field-cooling (ZFC) thermomagnetic curves along the easy direction in a low field of 0.04 T for $Tb_2(Fe, Cr)_{17}$ single crystal.

1.5 K. The irreversible thermomagnetic behaviour is a result of magnetohistory effects, as seen in other rare-earth compounds such as $RFe_{12-x}Mo_x$ (R = Y [15], Nd [16] and Dy [17]) and $Tb_3(Fe, Cr)_{29}$ [18]. In order to determine the temperature of the magnetohistory effect precisely, ac susceptibility (χ_{ac}) measurements were carried out and the results are shown in figure 4. The temperature at which the magnetohistory effect occurred was below 167 K, as was determined from the cusp of the χ_{ac}-curve.

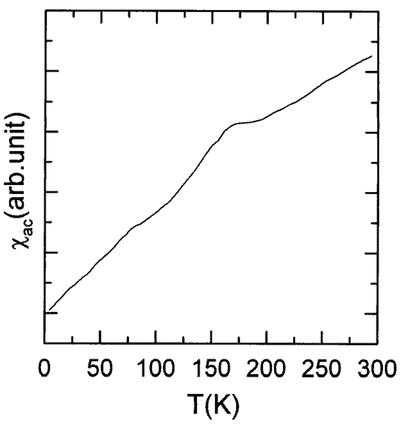

Figure 4. The temperature dependence of the ac susceptibility (χ_{ac}) at 4.2–293 K for $Tb_2(Fe, Cr)_{17}$ single crystal when the small spherical sample was placed freely in the ac field.

The hard-magnetization direction, i.e. the *c*-axis, can be accurately fixed using the rotation alignment method taking into consideration that $Tb_2(Fe, Cr)_{17}$ exhibits a planar anisotropy at room temperature. Figure 5 shows the typical magnetization curves at 5 K and 293 K measured along the easy and hard directions. Employing the law of approach to saturation to the easy-magnetization curve, the spontaneous magnetization M_s is obtained and this is plotted in figure 6. One can see that M_s increases with increasing temperature. This means that the Tb

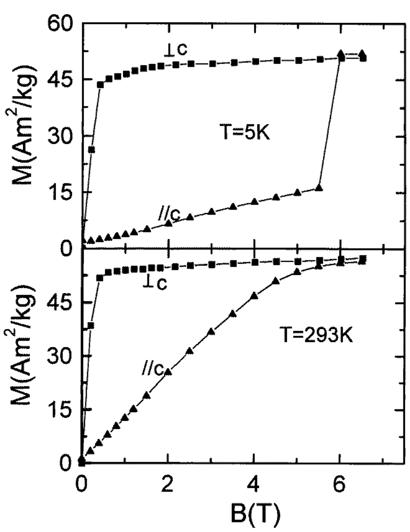

Figure 5. Magnetization curves along the easy and hard directions at 5 K and 293 K for $Tb_2(Fe, Cr)_{17}$ single crystal.

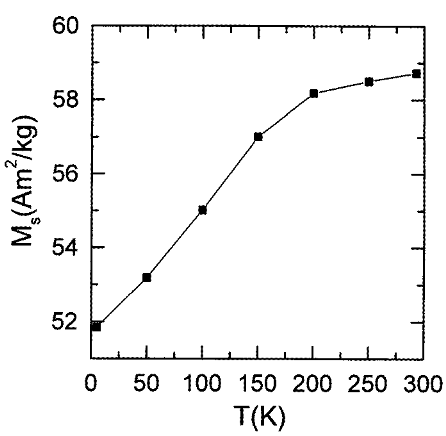

Figure 6. The temperature dependence of the spontaneous magnetization for $Tb_2(Fe, Cr)_{17}$ single crystal.

sublattice is antiferromagnetically coupled with the 3d sublattice. Spontaneous magnetization values of 51.8 A m^2 kg^{-1} (11.7 μ_B/f.u.) at 5 K and 58.7 A m^2 kg^{-1} (13.2 μ_B/f.u.) at 293 K are obtained, respectively. Taking 9 μ_B as the Tb moment, the average moment of 1.98 μ_B for Fe at 5 K is obtained. The value is somewhat lower than the values of 2.01 μ_B/Fe obtained for Y_2Fe_{17} [19] and 2.15 μ_B/Fe obtained for Tb_2Fe_{17} single crystals at 4.2 K [20] and may be connected with the antiferromagnetic coupling of Cr atoms.

We can also see from the magnetization curves at different temperatures that an obvious FOMP effect occurs at low temperature and disappears at about 250 K for the Tb_2(Fe, Cr)$_{17}$ single crystal. The critical field B_{cr} for the FOMP effect is 5.5 T at 5 K for Tb_2(Fe, Cr)$_{17}$ crystal, which is larger than that of 3.9 T for Tb_2Fe_{17} single crystal at 4.2 K [20]. The FOMP effect occurring in Tb_2(Fe, Cr)$_{17}$ crystal is considered as a FOMP of type I. The magnetization along the hard direction almost reaches saturation as a result of the FOMP. It behaves differently from Tb_2Fe_{17} single crystal, in which a type-II effect was confirmed, as the magnetization along the hard direction was at saturation until the magnetization went up a slight incline at about 10.0 T at 4.2 K. The anisotropy field B_a is determined as the field at which the magnetization along the hard direction equals the one along the easy direction (see figure 5). The temperature dependences of the anisotropy field B_a and the critical field B_{cr} of the FOMP effect are plotted in figure 7. We can see that B_a and B_{cr} are nearly equal and remain unchanged below 250 K. For the high temperatures of 250 K and 293 K, the FOMP effect is not seen. The anisotropy constants K_1 and K_2 for a system with a planar anisotropy can be calculated using the Klein equation [21]. The expression is written as

$$\frac{B}{\mu_0 M_c} = -\frac{2K_1 + 4K_2}{\mu_0 M_s^2} + \frac{4K_2}{\mu_0 M_s^4} M_c^2$$

where μ_0 is the permeability of free space, B is the applied field, M_c is the magnetization along the hard direction and M_s is the spontaneous magnetization. Figure 8 shows a Sucksmith–Thompson plot at 293 K for the Tb_2(Fe, Cr)$_{17}$ single crystal obtained using the Klein expression. At 293 K the values of $K_1 = -1.06$ MJ m^{-3} and $K_2 = 0.15$ MJ m^{-3} were obtained from figure 8 for Tb_2(Fe, Cr)$_{17}$.

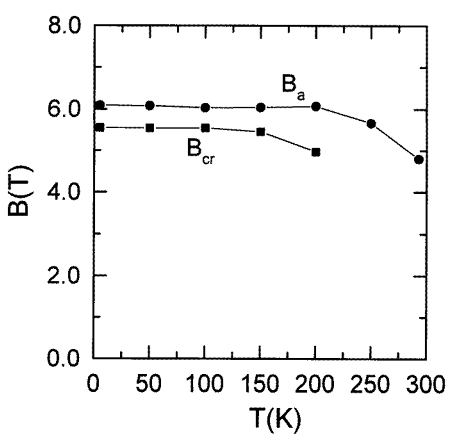

Figure 7. The temperature dependence of the magnetocrystalline anisotropy field B_a and the critical field B_{cr} for the FOMP effect for Tb_2(Fe, Cr)$_{17}$ single crystal.

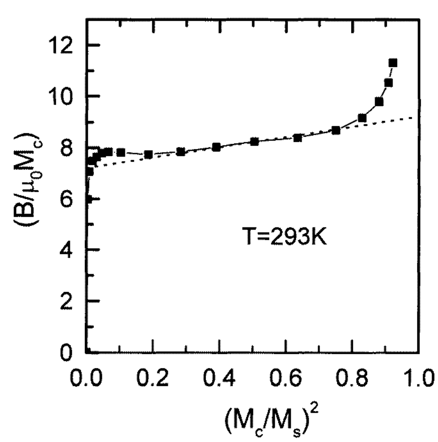

Figure 8. A Sucksmith–Thompson plot obtained using the Klein equation for Tb_2(Fe, Cr)$_{17}$ single crystal at 293 K.

In conclusion, a Tb_2(Fe, Cr)$_{17}$ single crystal was prepared by the Czochralski method. The Curie temperature is 457 K. The spontaneous magnetizations are 51.8 A m^{-2} kg^{-1}

(11.7 μ_B/f.u.) at 5 K and 58.7 A m^2 kg^{-1} (13.2 μ_B/f.u.) at 293 K. A remarkable magneto-history effect is observed below 167 K. $Tb_2(Fe, Cr)_{17}$ still exhibits a planar anisotropy in the temperature range from 5 K to 293 K, like Tb_2Fe_{17}. A FOMP of type I is found below 250 K and the critical fields are around 5.5 T for various temperatures for $Tb_2(Fe, Cr)_{17}$ single crystal.

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