Magnetic properties of the novel $Y_3(Fe,Cr)_{29}$ single crystal

1999 J. Phys.: Condens. Matter 11 6843

(http://iopscience.iop.org/0953-8984/11/36/301)

J. Phys.: Condens. Matter **11** (1999) 6843–6849. Printed in the UK PII: S0953-8984(99)01320-X

Magnetic properties of the novel $Y_3(Fe,Cr)_{29}$ single crystal

C P Yang†‡‖, Y Z Wang‡§, G H Wu‡, X F Han‡, B P Hu§, C L Ma†, X Y Chen† and J Zhu†

† Department of Material Science and Engineering, Tsinghua University, Beijing 100084, People's Republic of China
‡ State Key Laboratory for Magnetism, Institute of Physics and Center for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100080, People's Republic of China
§ San Huan Research Laboratory, Chinese Academy of Sciences, PO Box 603, Beijing 100080, People's Republic of China

E-mail: yangcp@263.net

Received 27 January 1999, in final form 21 June 1999

Abstract. A novel $Y_3(Fe,Cr)_{29}$ single crystal with the $Nd_3(Fe,Ti)_{29}$-type structure has been successfully prepared using the Czochralski method followed by a heat treatment on a $Y_2Fe_{15}Cr_2$ single crystal with a Th_2Ni_{17}-type structure. The magnetization curves measured along the easy and hard direction in the temperature range from 1.5 to 293 K are presented. The lattice parameters of the crystal are $a = 10.645$ Å, $b = 8.455$ Å, $c = 9.678$ Å and $\beta = 97.462^\circ$, respectively. The Curie temperature is 410 K. The saturation magnetizations decrease with increasing temperatures and are in good agreement with the spin-wave relation. The average magnetic moment is 1.54 μ_B per Fe atom at 1.5 K. The crystal keeps a planar anisotropy from 1.5 to 293 K. The magnetocrystalline anisotropy constants at 1.5 K are $K_1 = -0.87 \times 10^6$ J m^{-3} and $K_2 = 0.19 \times 10^6$ J m^{-3}.

1. Introduction

In the past decade, a novel iron-based metastable intermetallic compound $R_3(Fe,M)_{29}$ and its nitride of $R_3(Fe,M)_{29}N_y$, where R = rare earth, M = Ti, V, Cr and Mo, were investigated extensively for potential application as permanent magnets [1]. It has been found that the new family of $R_3(Fe,M)_{29}$ possesses the monoclinic $Nd_3(Fe,Ti)_{29}$-type structure with the space group $P2_1/c$, and can be considered as a intermediate structure consisting of the 1:12 structure and the 2:17 structure at the ratio of 1:1 [2]. The subsequent investigations of Kalogirou *et al* [3] and Psycharis *et al* [4] suggested the $R_3(Fe,M)_{29}$ structure can be described more accurately by the space group $A2/m$ instead of $P2_1/c$. Like the iron-based intermetallic compounds of 2:17 and 1:12, a marvellous improvement in the Curie temperature and a drastic modification in anisotropy take place in the carbides and nitrides upon the introduction of C, and especially N, as the interstitial atoms in 3:29. The permanent magnetic properties of $Sm_3(Fe,Ti)_{29}N_y$ [5] and $Sm_3(Fe,Ti)_{29}C_y$ [6] developed by the ball-milling technique were reported in our previous work. However, all the investigations mentioned above were performed on the polycrystalline $R_3(Fe,M)_{29}$ samples. Compared with the systematic studies on other iron-based rare-earth compounds, such as $R_2Fe_{14}B$, R_2Fe_{17} and $R(Fe,M)_{12}$, there are some significant questions keeping us from understanding the $R_3(Fe,M)_{29}$ compound yet. For instance, the substantial value of the magnetization and anisotropy contributed from the Fe atom at different sites in the

‖ Corresponding author.

3:29 structure remains unknown both in theory and experiment. It is not confirmed by experiment yet that $R_3(Fe,M)_{29}$, as supposed by Li *et al*, possesses two unequal R sites and results in the spin reorientation in $Nd_3(Fe,Ti)_{29}$ and the first order magnetization process (FOMP) in $Sm_3(Fe,Ti)_{29}$ due to the competition between the uniaxial anisotropy and the planar anisotropy originating from the two different R sites [7]. In order to have accurate data, the investigation on a single crystal is always highly desirable. However, the metastable feature prevents us from directly growning an $R_3(Fe,M)_{29}$ single crystal. To our knowledge, there is no data on a single crystal of $R_3(Fe,M)_{29}$ except for $Y_3(Fe,V)_{29}$ [8, 9] and $Tb_3(Fe,V)_{29}$ [10] single crystals investigated by Courtois *et al*. In the present work, a $Y_2Fe_{15}Cr_2$ single crystal with a hexagonal 2:17 structure was grown by the Czochralski method, followed by a proper heat treatment. We successfully obtained a $Y_3(Fe,Cr)_{29}$ single crystal with the $Nd_3(Fe,Ti)_{29}$-type structure after a solid transition. The magnetic properties of the crystal are also present in this contribution.

2. Experiment

The alloy with composition of $Y_2Fe_{15}Cr_2$ was prepared by arc-melting the starting material Y,Fe with the purity of 99.9% and Cr of the purity of 99.99%. An excess of 5 wt% was added for Y to compensate the loss during melting. A slim Fe bar was used as the seed in the procedure of pulling the crystal. The original crystal was grown from such a Y–Fe–Cr alloy melt using MCGS-3CZ equipment with a growth rates of 15–25 mm h^{-1} and a rotation rate of 30 rpm [11]. Checked by Laue back-reflection and metallographic observation, the $Y_2Fe_{15}Cr_2$ original crystal was confirmed as a single crystal with a Th_2Ni_{17}-type structure. The $Y_3(Fe,Cr)_{29}$ single crystal with an $Nd_3(Fe,Ti)_{29}$-type structure was finally obtained via the original single with Th_2Ni_{17} structure vacuum annealed at 1000 °C for four days and followed by quenching in water. The hard magnetization direction (i.e. the [102] axis in the 3:29 structure) was fixed on a spherical sample with a diameter of 2 mm. The Curie temperature was determined from the thermomagnetic scan trace measured by a vibrating sample magnetometer (VSM) above room temperature. The magnetization curves measured along the easy and hard direction were carried out by an extracting sample magnetometer. The anisotropy field at room temperature was detected by the singular point detector (SPD) technique [12].

3. Results and discussion

Figure 1 presents the x-ray powder diffraction patterns of the Y–Fe–Cr crystal before and after the annealing procedure. It can be seen clearly that the characteristic spectrum of the 3:29 structure appears in the patterns after heat treatment, such as line (322), which was shifted slightly below 40°, and the three lines (10−4), (40−2) and (23−1), which were split apart after heat treatment. This implies that the former $Y_2Fe_{15}Cr_2$ crystal with a hexagonal structure has been changed into the $Y_3(Fe,Cr)_{29}$ crystal with a monoclinic structure after a solid transition. The lattice parameters of the $Y_3(Fe,Cr)_{29}$ crystal respectively are $a = 10.645$ Å, $b = 8.455$ Å, $c = 9.678$ Å and $\beta = 97.462°$ by indexing the x-ray powder diffraction patterns.

The Curie temperature of $Y_3(Fe,Cr)_{29}$ single crystal determined by VSM is 410 K. It is similar to $T_c = 439$ K for the $Y_3(Fe_{0.93}V_{0.07})_{29}$ single crystal [9]. Figure 2 shows the magnetization curves from 1.5 to 293 K measured along the easy and hard direction in fields up to 6.5 T for the $Y_3(Fe,Cr)_{29}$ single crystal. Employing the law of approach to saturation, the saturation magnetization M_s is obtained and summarized in figure 3. The saturation magnetization is 8.34×10^5 A m^{-1} (115.26 A m^2 kg^{-1}) at 1.5 K and 6.36×10^5 A m^{-1} (87.86 A m^2 kg^{-1}) at 293 K respectively. The average moment of 1.54 μ_B at 1.5 K is

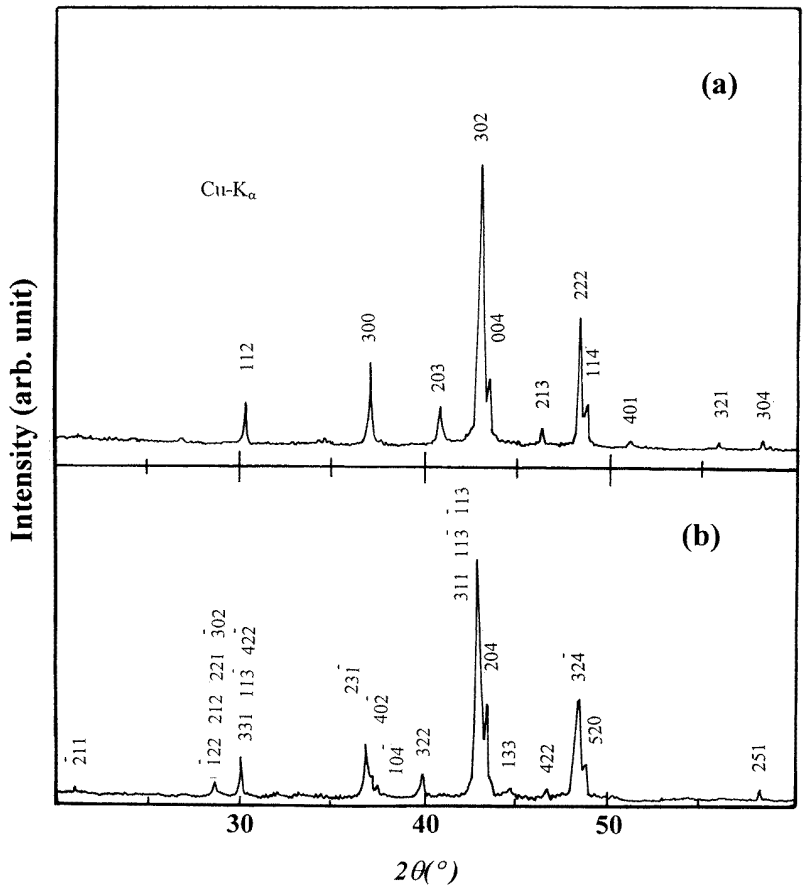

Figure 1. X-ray diffraction patterns of (a) before and (b) after the annealing procedure on a Y–Fe–Cr single crystal.

calculated per Fe atom. The value is somewhat smaller than 1.79 μ_B/Fe at 5 K for the $Y_3(Fe_{0.93}V_{0.07})_{29}$ crystal [9]. According to the spin-wave relation, the temperature dependence of the spontaneous magnetization of a ferromagnetic material is written by:

$$M_s = M_s(0)(1 - aT^{3/2}) \tag{1}$$

where $M_s(0)$ is the spontaneous magnetization at 0 K and a is a coefficient factor connected with the structure and properties of the material. The M_s calculated by the spin-wave relation is also shown in figure 3 with a solid line, together with the magnetization values calculated on the basis of the Brillouin function (shown in a dashed line). It can be seen that the experimental data are in good agreement with the spin-wave relation but somewhat different from the results of the Brillouin function, which can describe the variation of spontaneous magnetization with temperature for an Fe single phase with $J = 1/2$ very well. The coefficient a is 12.6×10^{-5} K$^{-3/2}$ for the $Y_3(Fe,Cr)_{29}$ single crystal. A similar relationship of M_s versus T was also found in Y_2Fe_{17} and Y_2Co_{17} single crystals with $a = 7.55 \times 10^{-5}$ K$^{-3/2}$ and $a = 3.42 \times 10^{-6}$ K$^{-3/2}$, respectively [13].

The magnetocrystalline anisotropy field B_a also can be obtained from figure 2 by extrapolating the hard magnetization curve across the easy magnetization curve. The values of B_a using the extrapolation method are shown in figure 4. At 1.5 and 293 K, B_a is 1.95 and

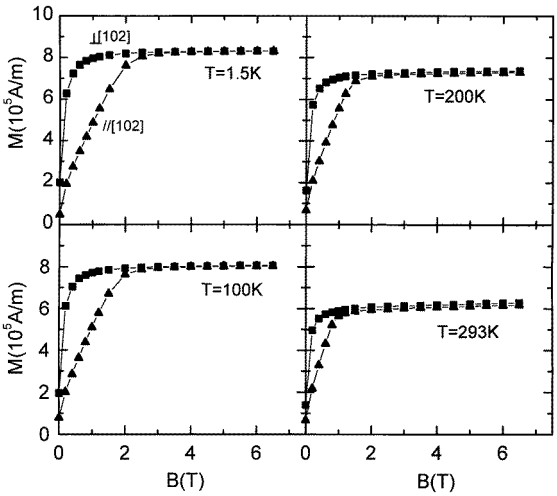

Figure 2. Magnetization curves along the easy and hard direction at several temperatures for the $Y_3(Fe,Cr)_{29}$ single crystal.

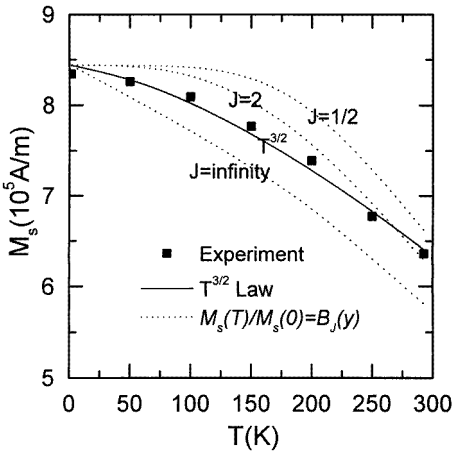

Figure 3. Temperature dependence of the saturation magnetization for the $Y_3(Fe,Cr)_{29}$ single crystal. The solid square symbol presents the experimental values of M_s obtained by the law of approach to saturation. The solid and dashed curves respectively show the calculation results using the spin-wave relation and Brillouin function.

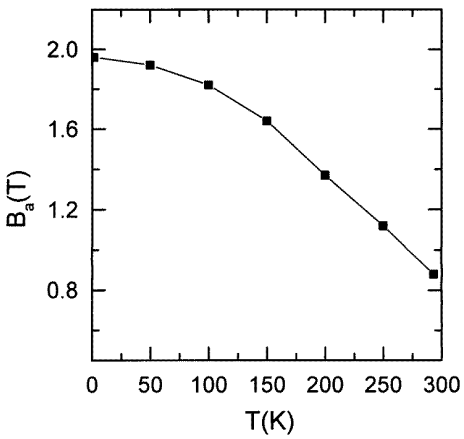

Figure 4. The variation of the magnetocrystalline anisotropy field B_a with temperature for the $Y_3(Fe,Cr)_{29}$ single crystal.

0.88 T respectively. The SPD curves measured along the hard magnetization direction at room temperature are presented in figure 5. The peak at the curve of the second-order derivative d^2M/dt^2 versus the applied field B is defined as the anisotropy field. B_a is 0.75 and 0.84 T determined from the d^2M/dt^2 curve of the magnetization and demagnetization procedure, respectively. The average value of B_a at room temperature using the SPD technique is in good agreement with that of 0.88 T using the extrapolation procedure.

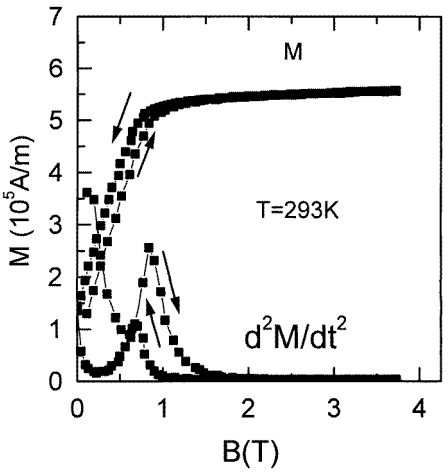

Figure 5. A room temperature SPD signal measured along the hard direction for the $Y_3(Fe,Cr)_{29}$ single crystal.

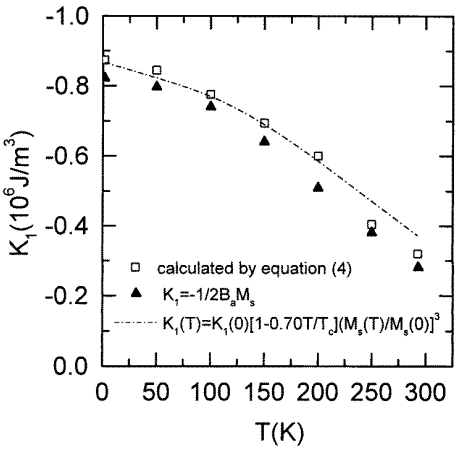

Figure 6. Temperature dependence of magnetocrystalline anisotropy constant K_1 in the temperature range from 1.5 to 293 K. The dashed line presents the results calculated by the Perkins equation. The open square and solid triangle symbols are respectively the calculation values using the Klein equation and first approximation.

To a first approximation, the magnetocrystalline anisotropy constant of $Y_3(Fe,Cr)_{29}$ single crystal can be calculated by:

$$K_1 = -\tfrac{1}{2} M_s B_a \tag{2}$$

where M_s is the saturation magnetization, B_a is the magnetocrystalline anisotropy field determined by the extrapolation method. The results of K_1 calculated by the above equation are shown in figure 6 by a solid up triangle symbol. The temperature dependence of K_1 for a planar system can be calculated by the following expression, which was proposed by Perkins and Nagel [14].

$$K_1(T) = K_1(0)\left(1 - \alpha \frac{T}{T_c}\right)\left(\frac{M_s(T)}{M_s(0)}\right)^3 \tag{3}$$

where α is a constant independent of temperature. T_c, $M_s(0)$ are the Curie temperature and the saturation magnetization at 0 K respectively. α is 0.7 for the $Y_3(Fe,Cr)_{29}$ single crystal and in the same as Y_2Fe_{17} single crystal, but smaller than that of 1.5 for the Y_2Co_{17} single crystal [14]. The results calculated by the Perkins expression are also presented with a dashed line in figure 6. One can see that the experimental data are coincident with the calculation data using the Perkins expression for the $Y_3(Fe,Cr)_{29}$ single crystal at a low temperature.

The anisotropy constant K_1 and K_2 for a planar anisotropy system can be calculated by the following expression, which was suggested by Klein *et al* [15]:

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

where μ_0 is the permeability of free space, B is the applied field, M_c is the magnetization measured along the hard direction and M_s is the saturation magnetization. As an example, a Sucksmith–Thompson-like plot at 1.5 K is presented in figure 7 for the $Y_3(Fe,Cr)_{29}$ single crystal using equation (4). The temperature dependence of K_1 and K_2 is shown in figure 8.

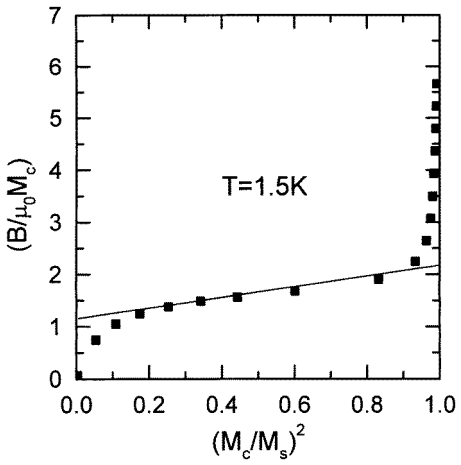

Figure 7. A Sucksmith–Thompson-like plot at 1.5 K using the Klein equation for the $Y_3(Fe,Cr)_{29}$ single crystal.

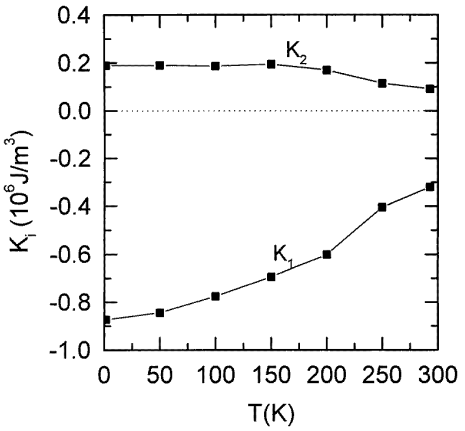

Figure 8. Temperature dependence of anisotropy K_1 and K_2 obtained by the Klein equation for the $Y_3(Fe,Cr)_{29}$ single crystal.

At 1.5 K, the anisotropy constants $K_1 = -0.87 \times 10^6$ J m^{-3} and $K_2 = 0.19 \times 10^6$ J m^{-3} are obtained from figure 7 for the $Y_3(Fe,Cr)_{29}$ single crystal. The value of K_1 at 1.5 K is much smaller than that of -2.3×10^6 J m^{-3} ($T = 4.2$ K) for the binary Y_2Fe_{17} single crystal [16].

In conclusion, a $Y_3(Fe,Cr)_{29}$ single crystal with an $Nd_3(Fe,Ti)_{29}$-type structure has been successfully prepared using the Czochralski method followed by an annealing procedure. The lattice parameters are $a = 10.645$ Å, $b = 8.455$ Å, $c = 9.678$ Å and $\beta = 97.462^\circ$ for $Y_3(Fe,Cr)_{29}$ single crystal, respectively. The Curie temperature is 410 K. The saturation magnetization decreases with increasing temperature and is in good agreement with the spin-wave relation. The average magnetic moment is 1.54 μ_B per Fe atom at 1.5 K. The crystal keeps a planar anisotropy from 1.5 to 293 K. $M_s = 8.34 \times 10^5$ A m^{-1}, $K_1 = -0.87 \times 10^6$ J m^{-3} and $K_2 = 0.19 \times 10^6$ J m^{-3} at 1.5 K are also obtained for the $Y_3(Fe,Cr)_{29}$ single crystal.

References

[1] Cadogan J M, Li H S, Margarian A, Dunlop J B, Ryan D H, Collocott S J and Davis R L 1994 *J. Appl. Phys.* **76** 6138

[2] Li H S, Cadogan J M, Davis R L, Margarian A and Dunlop J B 1994 *Solid State Commun.* **90** 487

[3] Kalogirou O, Psycharis V, Saettas L and Niarchos D N 1995 *J. Magn. Magn. Mater.* **146** 335

[4] Psycharis V, Kalogirou O, Niarchos D and Gjoka M 1995 *Proc. 3rd Int. Symp. on Physics of Magnetic Material, ISPMM'95 (Seoul, 1995)* p 752

[5] Hu B P, Liu G C, Wang Y Z, Nasunjilegal B, Zhao R W, Yang F M, Li H S and Cadogan J M 1994 *J. Phys.: Condens. Matter* **6** L197

[6] Hu B P, Liu G C, Wang Y Z, Nasunjilegal B, Tang N, Yang F M, Li H S and Cadogan J M 1994 *J. Phys.: Condens. Matter* **6** L595

[7] Li H S, Cadogan J M, Hu B P, Yang F M, Nasunjilegal B, Margarian A and Dunlop J B 1995 *J. Magn. Magn. Mater.* **140–144** 1037

[8] Courtois D, Givord D, Lambert-Andron B, Bourgeat-Lami E, Amako Y, Li H S and Cadogan J M 1998 *J. Magn. Magn. Mater.* **189** 173

[9] Courtois D, Amako Y, Givord D, Cadogan J M and Li H S 1998 *J. Magn. Magn. Mater.* **177–188** 995

[10] Courtois D, Li H S and Cadogan J M 1997 *IEEE Trans. Magn.* **33** 3844

[11] Wu G H, Zhao X G, Wang J H, Li J Y, Jia K C and Zhan W S 1995 *Appl. Phys. Lett.* **67** 2005
[12] Asti G and Rinaldi S 1974 *J. Appl. Phys.* **45** 3600
[13] Sinnema S 1988 *PhD Thesis* Natuurkundig Laboratorium der Universiteit Van Amsterdam
[14] Perkins R S and Nagel H 1975 *Physica* B **80** 143
[15] Klein H P, Menth A and Perkins R S 1975 *Physica* B **80** 153
[16] Averbuch-Pouchot M T, Chevalier R, Depórtes J, Kebe B and Lemaire R 1987 *J. Magn. Magn. Mater.* **68** 190