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Magnetocrystalline Anisotropy in the Cubic TbAl_2 Compound

By

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The isothermal magnetization of a TbAl_2 single crystal has been measured along the main symmetry directions in static magnetic fields up to 150 kOe and in the temperature range from 1.5 to 300 K. $\langle 111 \rangle$ is the easy direction of magnetization at all field strengths and for all temperatures below the Curie point $T_c = 105$ K. The spontaneous magnetization along $\langle 111 \rangle$ follows a Brillouin law; however, there is a strong magnetization anisotropy. By neglecting magnetostriction, the thermal variations of the anisotropy parameters of 4th and 6th order have been obtained: They are of the same type as those anticipated by Callen and Callen.

Nous avons mesuré les variations isothermes de l'aimantation d'un monocristal de TbAl_2 suivant les principales directions cristallographiques dans des champs statiques atteignant 150 kOe, entre 1,5 et 300 K. L'axe $\langle 111 \rangle$ est de facile aimantation à toutes températures inférieures à la température de Curie $T_c = 105$ K. Selon cet axe la variation thermique de l'aimantation spontanée obéit à une loi de Brillouin; il apparaît cependant une forte anisotropie de l'aimantation. En négligeant la magnétostriction, les variations thermiques des constantes d'anisotropie d'ordres 4 et 6 ont pu être obtenues: elles sont du type de celles prévues par Callen et Callen.

1. Introduction

The RAl_2 compounds where R represents a rare-earth element crystallize in the cubic MgCu_2 type structure [1]. Magnetization measurements on polycrystalline samples show that most of the RAl_2 compounds are ferromagnetically ordered [2]. Recent low-temperature measurements on single crystals of ErAl_2 [3] and TbAl_2 [4] demonstrate the importance of the crystalline electric field in these compounds. This is also reflected in the magnetic excitation spectra measured recently in TbAl_2 [5] and NdAl_2 [6]. In the present paper we study the magnetization of a single crystal of TbAl_2 . In particular, we discuss the thermal variation of the magnetocrystalline anisotropy.

2. Experimental Methods

The TbAl_2 single crystal has been prepared by the Czochralski method [7] from 99.9% pure terbium and 99.999% pure aluminium. The crystal was cut to a sphere of 3 mm in diameter and analysed by X-ray diffraction. The amount of parasitic phase is estimated to be less than 0.5%.

The measurements have been performed at the Service National des Champs Intenses at Grenoble. The magnetization has been measured from 1.5 to 300 K by the induction method in static fields up to 150 kOe provided by a water-cooled Bitter magnet [8, 9].

3. Experimental Results

Fig. 1 shows a plot of the magnetization versus the magnetic field applied in the three symmetry directions, at several temperatures below the ordering point. In all cases $\langle 111 \rangle$ and $\langle 100 \rangle$ are observed respectively as the easy and hard directions of magnetization. For example at 4.2 K, for the three directions $\langle 111 \rangle$, $\langle 110 \rangle$, and $\langle 100 \rangle$, the initial magnetization curve in low field is a straight line with a slope equal to the reciprocal demagnetization factor. Along the $\langle 111 \rangle$ axis the saturation is obtained as soon as the field reaches the demagnetizing field strength but this curve exhibits a very small superposed susceptibility. Along the $\langle 110 \rangle$ axis the increase of magnetization abruptly becomes very small at 100 kOe but the magnetization still remains inferior to that measured along $\langle 111 \rangle$. Along the $\langle 100 \rangle$ axis, such a change of slope appears only on the curves for temperatures higher than 40 K. By extrapolating the saturation curve along the $\langle 111 \rangle$ direction to zero internal field we find at 4.2 K a value of $(8.90 \pm 0.05)\mu_B/\text{Tb}^{3+}$ for the spontaneous magnetization. This value is very close to the magnetic moment of the free Tb^{3+} ion and is in good agreement with recently reported data [4, 10]. At 100 kOe the slope of the magnetization curve is $1.1 \times 10^{-6} \mu_B/\text{Oe}$.

The points of intersection of the demagnetizing field and saturation straight lines were plotted versus temperature to determine below 91 K the temperature dependence of the spontaneous magnetization M_s in the $\langle 111 \rangle$ direction. Above 91 K Belov and Goryaga [11] and Kouvel's [12] method was used. By this method the Curie temperature of (105 ± 2) K has been obtained from the isotherms $M(H)$ measured at 91, 101, 110, and 120 K. The curve of spontaneous magnetization versus temperature is compared to the B_6 Brillouin curve in Fig. 2.

From accurate measurements of the spontaneous magnetization of TbAl_2 between 1.5 and 40.4 K along the $\langle 111 \rangle$ easy magnetization axis we have deduced the thermal variation of the difference between the spontaneous magnetization at 0 K and that at any temperature T . The curve obtained versus $T^{3/2}$ presents positive concavity (Fig. 2, insert). It is linear as a function of $T^{3/2} \exp(-\Delta/kT)$ with $\Delta = 1.6$ meV.

Below T_c the values of the zero field magnetization along the $\langle 110 \rangle$ and $\langle 100 \rangle$ directions correspond to the projections of the zero field moment measured along $\langle 111 \rangle$. At 4.2 K (Fig. 1) a field of 100 kOe is needed to align the moments along $\langle 110 \rangle$. However, for $\langle 100 \rangle$ the largest available field of 150 kOe is not sufficient to obtain full alignment. Above 40 K alignment of the moments is obtained in all directions. An important feature of the magnetization behaviour of TbAl_2 is that the measured moments in the case of full alignment vary according to the symmetry direction in which they are aligned; the intrinsic moment depends on direction. This anisotropy of magnetization must be taken into account for the evaluation of the magnetocrystalline energy. In the insert of Fig. 3 the variation of the intrinsic moment is shown as a function of temperature.

Above the Curie point no anisotropy is observed in TbAl_2 . The effective moment has a value of $9.5\mu_B$ which is close to the value of $9.7\mu_B$ obtained from Hund's rule.

4. Determination of the Anisotropy Energy

Measurements of TbAl_2 lattice parameters at low temperature are in progress in order to ultimately reveal any distortion of the cubic lattice. Lacking this information we can not at this time take into account all the magnetostrictive effects.

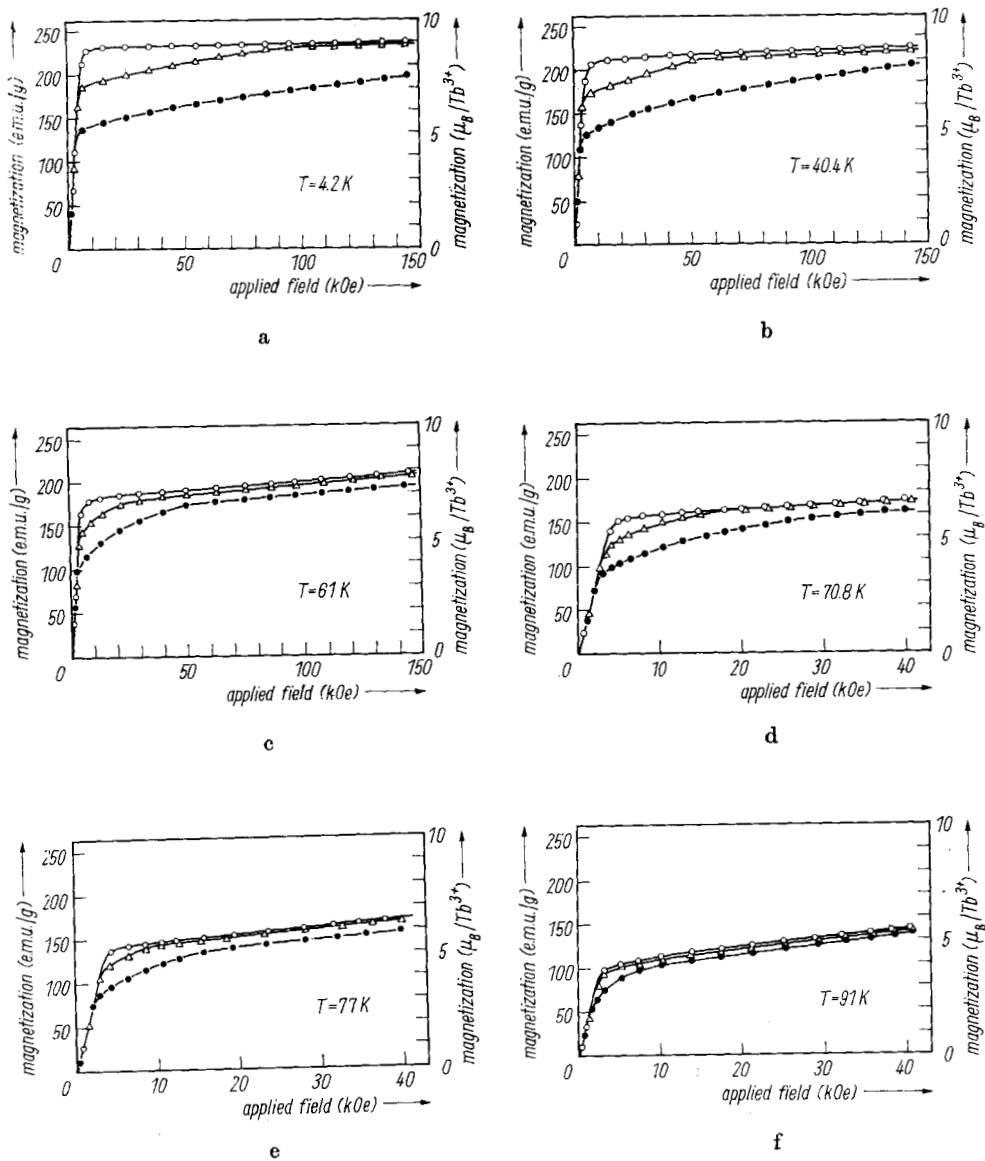


Fig. 1. Magnetization of a single crystal of $TbAl_2$ as a function of external magnetic field applied along the $\langle 111 \rangle$ (\circ), $\langle 110 \rangle$ (Δ), and $\langle 100 \rangle$ (\bullet) directions at several temperatures

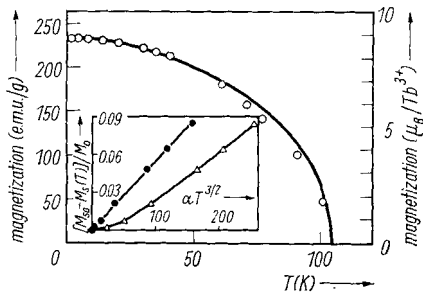


Fig. 2. Spontaneous magnetization as a function of temperature. The continuous curve represents the B_6 Brillouin law. Insert: relative difference between the spontaneous magnetization M_{s0} and $M_s(T)$ vs. $\alpha T^{3/2}$; $\alpha = 1$ (Δ), $\alpha = \exp - \Delta/kT$ (\bullet)

The energy necessary to rotate the magnetization into the direction of the applied field is obtained (Fig. 1) by evaluating the area enclosed by the isothermal magnetization curve and the line of saturation of the same isothermal curve extrapolated to internal zero field. Above 40 K the magnitude of the anisotropy of magnetization is obtained along the three directions and the method described above gives this energy directly. At 40 K and below, this magnitude was not detected on the $\langle 100 \rangle$ direction and the magnetization anisotropy was estimated by extrapolating to zero Kelvin the thermal variation of the intrinsic moment measured on the $\langle 100 \rangle$ axis (Fig. 3, insert). The energy thus obtained is mainly of magnetocrystalline origin. The contribution due to the imperfections of the crystal is eliminated by evaluating the area above the curve along the $\langle 111 \rangle$ axis. The errors on the magnetocrystalline energies in the $\langle 100 \rangle$ direction are $\pm 8\%$ at 4.2 K, $\pm 7\%$ at 20.0 K, $\pm 6\%$ at 40 K, and $\pm 2\%$ for all temperatures studied above 40.4 K. For the $\langle 110 \rangle$ direction this error is $\pm 2\%$ over the whole temperature range.

The measured magnetocrystalline energies $V_c \langle 100 \rangle - V_c \langle 111 \rangle$ and $V_c \langle 110 \rangle - V_c \langle 111 \rangle$ are plotted in Fig. 3 as a function of temperature. The former decreases slowly between 4.2 and 20 K, and very quickly between 30 and 60 K. The latter $V_c \langle 110 \rangle - V_c \langle 111 \rangle$ decreases more rapidly. Both energies vanish with increasing temperature.

As the magnetostrictive effects are not taken into account in this paper, an eventual second order term may be neglected in the magnetocrystalline energy, which is then given by the following expression [13]:

$$V_c = K_4(T) [x^4 + y^4 + z^4 - \frac{3}{5}] + K_6(T) [11(x^6 + y^6 + z^6) - 15(x^4 + y^4 + z^4) + \frac{30}{7}],$$

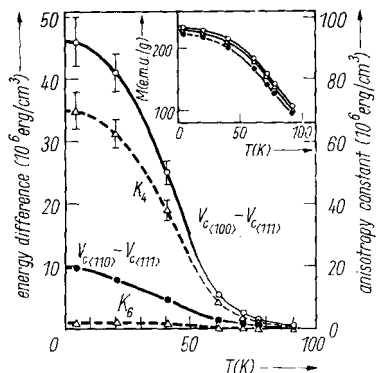


Fig. 3. Temperature dependence of the magneto-crystalline anisotropy energy difference, between the $\langle 100 \rangle$ and the $\langle 110 \rangle$ hard directions and the $\langle 111 \rangle$ easy direction (continuous curves). Thermal variations of the anisotropy parameters of 4th and 6th order (broken curves). Insert: thermal variation of the intrinsic moment measured along the main symmetry directions. Δ $\langle 110 \rangle$ axis, \circ $\langle 111 \rangle$ axis, \bullet $\langle 100 \rangle$ axis

where x , y , and z are the direction cosines and $K_4(T)$ and $K_6(T)$ the rare-earth ion anisotropy constants.

The results for $K_4(T)$ and $K_6(T)$ are given in Fig. 3. Both parameters are positive for the whole temperature range in which a sufficiently accurate determination is possible. $K_6(T)/K_4(T)$ is constant to a good approximation below 40 K but decreases abruptly around 50 K. $K_6(T)$ decreases more rapidly than $K_4(T)$ with increasing temperature. The temperature dependence of $K_4(T)$ and $K_6(T)$ is described approximately by the $[M(T)/M(0)]^{l(l+1)/2}$ law [14].

5. Conclusion

We have shown that in TbAl₂ $\langle 111 \rangle$ is the easy and $\langle 100 \rangle$ the hard direction of magnetization over the whole temperature range below $T_c = 105$ K. We have evaluated the magneto-crystalline anisotropy energy along the $\langle 100 \rangle$ and $\langle 110 \rangle$ directions and the anisotropy constants of the 4th and 6th order under the assumption that magnetostriction is not important. At low temperature $K_6(T)/K_4(T)$ is approximately equal to 3×10^{-2} and the value of $K_4(T)$ extrapolated to 0 K is 7×10^7 erg/cm³. These values lead to crystal field parameters which have the same sign and order of magnitude as the parameters used in earlier works for TbAl₂ [4, 5], CeAl₂ [15, 16], NdAl₂ [6], ErAl₂ [3], and Tm_xY_(1-x)Al₂ [17].

These parameters account for the result obtained by NMR for the easy direction of magnetization in several RAl₂ compounds [18]. We note that in RAl₂ the constants $K_4(0)$ and $K_6(0)$ can be obtained approximately by assuming that the aluminium ions have no charge and that the nearest terbium ions have a charge of 3+.

In TbAl₂, the crystalline field involves a strong anisotropy and also leads to a reduction of about 7% in the intrinsic moment when the moments are aligned along the $\langle 100 \rangle$ direction. This value is approximately that obtained from a quantum mechanical calculation using the molecular field approximation [19].

The temperature dependence of the magnetization at low temperatures is of the $T^{3/2} \exp(-\Delta/kT)$ type, showing the crystalline field effect on spin wave properties. Recent magnon dispersion measurements on TbAl₂ [5] have given $\Delta = (1.6 \pm 0.1)$ meV for the value of the minimum energy necessary for the excitation of the acoustic spin wave mode. From the zero Kelvin extrapolated values of the anisotropy constants $K_4(T)$ and $K_6(T)$, $\Delta = 1.1$ meV is calculated.

From these parameters, the 4f contribution to the superposed susceptibility on the $\langle 111 \rangle$ axis calculated in the molecular field approximation is found to be ten times too small. The increase of magnetization with increasing field along $\langle 111 \rangle$ at 4.2 K is then mainly due to conduction electron magnetization. This is supported by recent measurements on CeAl₂ where a superposed susceptibility of $2 \times 10^{-6} \mu_B/\text{Oe}$ was found. Since the light rare earths seem to have densities of states of conduction electrons roughly double that of the heavy rare earths [20] the results of TbAl₂ and CeAl₂ are consistent.

Despite the large anisotropy in TbAl₂ the observed agreement between the B₆ Brillouin curve and the experimental points is explained by the fact that the exchange field of 450 kOe is sufficient to create nearly pure $|J_z\rangle$ eigenstates. Furthermore, the introduction of the crystalline field parameters into the calculation of the spontaneous magnetization at several temperatures below T_c results in only minor corrections to the B₆ Brillouin curve.

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