

## Anisotropy energy measurements on single-crystal $\text{Tb}_{0.15}\text{Ho}_{0.85}\text{Fe}_2$

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The magnetocrystalline anisotropy energy constants  $K_1$  and  $K_2$ , have been examined for a  $\text{Tb}_{0.15}\text{Ho}_{0.85}\text{Fe}_2$  single crystal at various temperatures between 33 and 325 K using torque-magnetometry techniques. The data show that  $K_1$  and  $K_2$  are very sensitive to temperature changes in this range, and have room-temperature values of  $-2.2 \times 10^5$  and  $1.1 \times 10^6$  ergs/cm<sup>3</sup>, respectively. A comparison of the room-temperature values of  $K_1$  with the values reported in the literature for the binary compounds  $\text{TbFe}_2$  and  $\text{HoFe}_2$  shows that  $K_1$  for the pseudobinary is smaller than the values reported for the binary compounds by factors of 265 and 87, respectively. The low room-temperature anisotropy constants observed for the pseudobinary compound are also consistent with the prediction of the single-ion model of anisotropy and the behavior suggested by the Mössbauer-effect studies of Atzmony *et al.* Because of their small anisotropy constants and large magnetostriction, the pseudobinary compounds may be potentially useful in magnetostrictive-device applications. It was further found that easy direction of magnetization undergoes several reorientations as the temperature is increased from 33 to 325 K. A comparison of our data with the data obtained from Mössbauer-effect studies on powder samples, shows the two results to be in rather good agreement at low temperatures. At high temperatures the results differ in that the Mössbauer-effect studies suggest the presence of a triple point.

### I. INTRODUCTION

The recent discovery that certain rare-earth-iron,  $R\text{Fe}_2$ , intermetallic compounds possess unusually large room-temperature saturation magnetostriction values<sup>1</sup> has stimulated considerable interest in these compounds, particularly the  $\text{TbFe}_2$  compound which showed the largest room-temperature saturation magnetostriction. Because of their large magnetostriction constants these materials may be potentially useful in practical magnetostrictive devices. However, the large magnetic fields required to take advantage of these large strains presents a serious problem. These large fields are believed to be primarily related to the large magnetocrystalline anisotropy energy associated with these materials; however, other sources of anisotropy such as the strain and shape anisotropy may also contribute. For a practical device, it is desirable to have a material which has a low anisotropy energy while still retaining the large magnetostriction.

One group of compounds that show promise in fulfilling this requirement are the pseudobinary compounds of  $\text{Tb}_{1-x}\text{Ho}_x\text{Fe}_2$ . The qualitative behavior of the magnetocrystalline anisotropy energy for this group has been characterized, to some extent, by the Mössbauer-effect studies of Atzmony *et al.*<sup>2</sup> By examining the temperature dependence of the Mössbauer spectra for various powder samples, these authors found what was apparently a "triple point" for  $x = 0.85$ , i. e., a point where the [100], [110], and [111] are equally easy directions of magnetization. Phenomenologically, the magnetocrystalline anisotropy energy for a cubic crystal

is given by

$$E_{\text{M}} = K_0 + K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_1^2\alpha_3^2) + K_2\alpha_1^2\alpha_2^2\alpha_3^2 + \dots, \quad (1)$$

where  $K_0$  is a constant,  $K_1$  and  $K_2$  are the first and second-order magnetocrystalline anisotropy energy constants, respectively, and the  $\alpha_i$ 's are the direction cosines of the magnetization with respect to the cube axes. If all of the anisotropy energy is described by the two lowest-order anisotropy constants  $K_1$  and  $K_2$ , then such a triple point implies that  $K_1 = K_2 = 0$ .

Although the magnetocrystalline anisotropy energy constants can be deduced indirectly from the Mössbauer-effect studies by use of the single-ion model of anisotropy,<sup>2</sup> the most direct method of determination of the anisotropy constants is by use of the torque method. However, this method requires single-crystal samples, which prior to this investigation were unavailable. In this paper we report the first direct determination of the magnetocrystalline anisotropy energy for a  $\text{Tb}_{0.15}\text{Ho}_{0.85}\text{Fe}_2$  single-crystal sphere. The anisotropy energy is examined at several temperatures between 33 and 325 K, and the results are compared with room-temperature magnetocrystalline anisotropy energy constants predicted for  $\text{TbFe}_2$  and  $\text{HoFe}_2$ ,<sup>3</sup> and with the spin-reorientation results obtained from the Mössbauer-effect studies of Atzmony *et al.*<sup>2</sup>

### II. EXPERIMENTAL

The single crystal used in this investigation was prepared by arc melting the appropriate amounts of terbium, holmium, and iron in a purified argon

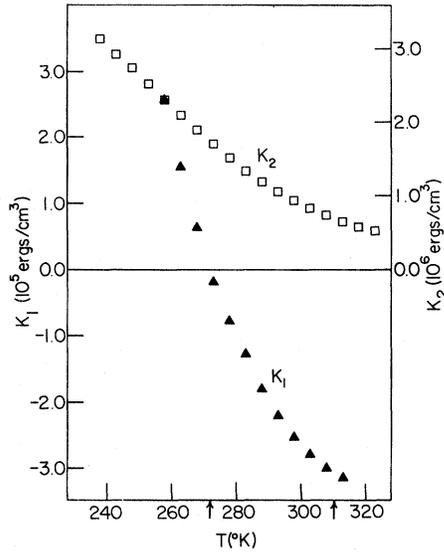


FIG. 1. Temperature dependence of  $K_1$  and  $K_2$  in the temperature range between 238 and 325 K. The arrows at 272 and 310 K indicate the spin-reorientation temperatures for spin transitions from the [100] to the [110] and from the [110] to the [111], respectively.

atmosphere. The button obtained was turned and remelted several times to ensure homogeneity. Following the arc melting an attempt was made to grow a single crystal using the Bridgeman method. Unfortunately, x-ray Laue patterns proved that the resulting sample was not single, but contained large crystallites on the order of several millimeters in size. Several  $2 \times 2$  mm cubes were cut from the crystallites and ground into spheres of about 0.085 cm in diameter.

The anisotropy constants  $K_1$  and  $K_2$  [Eq. (1)] were determined by use of the torque method. This method is based on the fact that the torque associated with rotating the magnetization away from the easy direction of magnetization is given by  $L_{hkl} = -\partial E_{hkl}/\partial \theta$ . If the plane of rotation is chosen properly, e.g., (100) or (111),  $K_1$  and  $K_2$  can be obtained directly from the torque curves providing (i) there are no other sources of anisotropy present such as strain or shape anisotropy which result in lower-order torque contributions, and (ii) the measuring field is large enough to saturate the magnetization in all directions, so that the magnetization and field directions always coincide. When other sources of anisotropy are present and/or when the plane of magnetization does not correspond to the (100) or (111), a Fourier series analysis is required to determine the various anisotropy constants. The torque measurements were made using a recently constructed torque magnetometer which is capable of detecting torques as low as  $10^{-3}$  dyn cm. The torque curves were obtained in fields up to 17.5 kG which were more than adequate to sat-

urate the magnetization in the temperature ranges considered.

### III. RESULTS AND DISCUSSION

The temperature dependence of  $K_1$  and  $K_2$  in the range between 260 and 325 K is shown in Fig. 1. These results show that  $K_1$  is positive at 260 K, passes through zero at 272 K, and becomes negative for temperatures greater than 272 K.  $K_2$ , on the other hand, is larger than  $|K_1|$  and is positive throughout this temperature range. The room-temperature values of  $K_1$  and  $K_2$  are  $-2.2 \times 10^5$  ergs/cm<sup>3</sup> and  $1.10 \times 10^6$  ergs/cm<sup>3</sup>, respectively. A comparison of the room-temperature values of  $K_1$  for this composition with the values reported by Clark *et al.*<sup>3</sup> for TbFe<sub>2</sub> and HoFe<sub>2</sub> are shown in Table I. This comparison shows that for the pseudobinary  $K_1$  is considerably less than the values for TbFe<sub>2</sub> and HoFe<sub>2</sub>, which is consistent with the behavior suggested by the Mössbauer-effect studies and the single-ion model. Also shown in Table I are the room-temperature saturation magnetostriction values,  $\lambda_s$ , and the anisotropy field,  $H_K = 2K_1/M$ , which is the field required to take advantage of the large strains. A comparison of these values shows that  $H_K$  for the pseudobinary is considerably less than the value for TbFe<sub>2</sub> by a factor of 525; however, the magnetostriction is also somewhat lower. Although there is some sacrifice in the magnetostriction and the anisotropy is not as low as desired, these results are nevertheless encouraging because they suggest that by further tailoring it is conceivable that a compound could be produced with suitable properties for magnetostrictive device applications.

Several spin reorientations or changes in the easy direction of magnetization were observed for this composition as the temperature was varied between 33 and 325 K. The conditions or relationships that  $K_1$  and  $K_2$  must satisfy when spin reorientations occur between the principal crystallographic directions are shown in Table II. These relationships can be derived directly from the anisotropy energy equations for the principal crystallographic directions,

$$\begin{aligned} E_{100} &= K_0, \\ E_{110} &= K_0 + \frac{1}{4}K_1, \\ E_{111} &= K_0 + \frac{1}{3}K_1 + \frac{1}{27}K_2. \end{aligned} \quad (2)$$

For example, when the easy direction of magnetization changes from the [110] to the [100], the [110] and [100] are equally easy directions of magnetization; therefore, the anisotropy energies associated with these directions are equal, i.e.,  $E_{110} = E_{100}$  and the first condition is obtained. The second condition  $-K_1 = \frac{1}{3}K_2$  is obtained in a similar manner by equating  $E_{110}$  to  $E_{111}$ . The third re-

TABLE I. Values of  $K_1$ ,  $K_2$ ,  $2K/M$ , and  $\lambda_s$  for  $\text{TbFe}_2$ ,  $\text{HoFe}_2$ , and  $\text{Tb}_{0.15}\text{Ho}_{0.85}\text{Fe}_2$ .

Composition	$K_1$ ( $10^7$ ergs/cm $^3$ )	$K_2$ ( $10^7$ ergs/cm $^3$ )	$2K_1/M$ (kOe)	$10^{-6}\lambda_s$
$\text{TbFe}_2$	-5.8	...	175	1720
$\text{HoFe}_2$	1.9	...	65	80
$\text{Tb}_{0.15}\text{Ho}_{0.85}\text{Fe}_2$	-0.022	0.11	0.33	325

relationship is the triple-point condition and represents the case when all three crystallographic directions are equally easy. This relationship is obtained by simultaneously equating all three anisotropy energy equations, i. e.,  $E_{100} = E_{110} = E_{111}$ . On the basis of the relationships tabulated in Table II, the spin-reorientation temperatures were determined. The first spin reorientation occurred at 42 K as shown in Fig. 2. At this temperature the first condition in Table II is satisfied, i. e.,  $K_1 = 0$  and the easy direction of magnetization changes from the [110] to the [100].

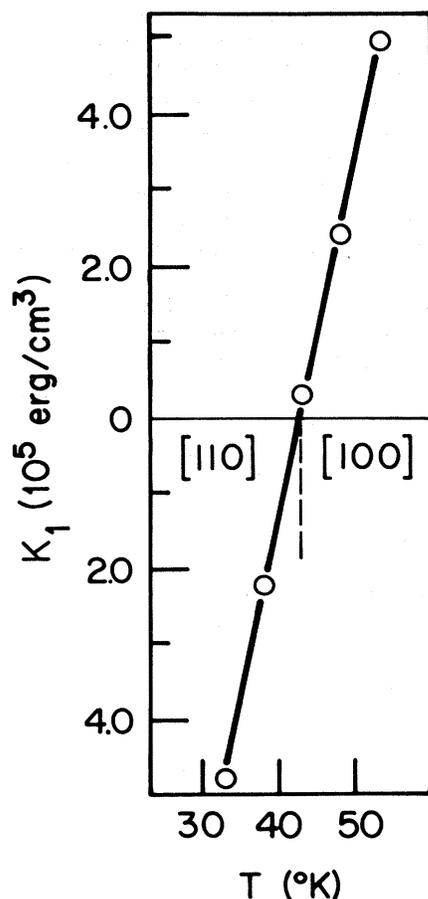


FIG. 2. Temperature dependence of  $K_1$  over the temperature range between 33 and 53 K; broken line indicates spin-reorientation temperature.

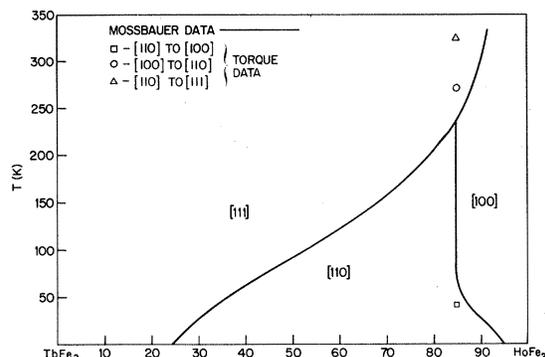


FIG. 3. Spin-reorientation diagram for  $\text{Tb}_{1-x}\text{Ho}_x\text{Fe}_2$ ; solid curve represents data of Atzmomy *et al.* (Ref. 3); open symbols represent torque results.

The higher spin-reorientation temperatures are indicated in Fig. 1 by arrows on the temperature axis. Between 42 and 272 K the [100] is the easy direction of magnetization. This conclusion is partly based on results obtained from magnetization measurements (to be discussed later). At 272 K,  $K_1 = 0$  and the easy direction changes to the [110]; at 310 K, the second condition in Table II,  $-K_1 = \frac{1}{9}4K_2$ , is satisfied and the easy direction of magnetization changes from the [110] to the [111]. Above 310 K the [111] is the easy direction of magnetization.

The spin-reorientation data are summarized and compared with the Mössbauer-effect studies of Atzmomy *et al.*<sup>2</sup> in Fig. 3. The solid curve represents the composition and temperature dependence of the spin-reorientation boundaries obtained from Mössbauer-effect studies. The symbols in Fig. 3 represent the spin-reorientation data obtained from the torque measurements. The comparison shows that the two results are in rather good agreement in the neighborhood of 42 K; however, above 42 K the results differ. The Mössbauer data show a triple point at 255 K, and a direct transition to the [111] at temperatures above 255 K. Our results show that the transition takes place in two steps: first the easy direction of magnetization changes from [100] to the [110]; then to the [111], with the [110] as an intermediate spin direction. No triple point was observed.

TABLE II. Conditions that  $K_1$  and  $K_2$  must satisfy when spin reorientation occurs between the principal crystallographic directions.

Spin reorientations	Conditions
[110] to [100]	$K_1 = 0$
[110] to [111]	$-K_1 = \frac{1}{9}4K_2$
[110] to [111] and [100]	$K_1 = K_2 = 0$

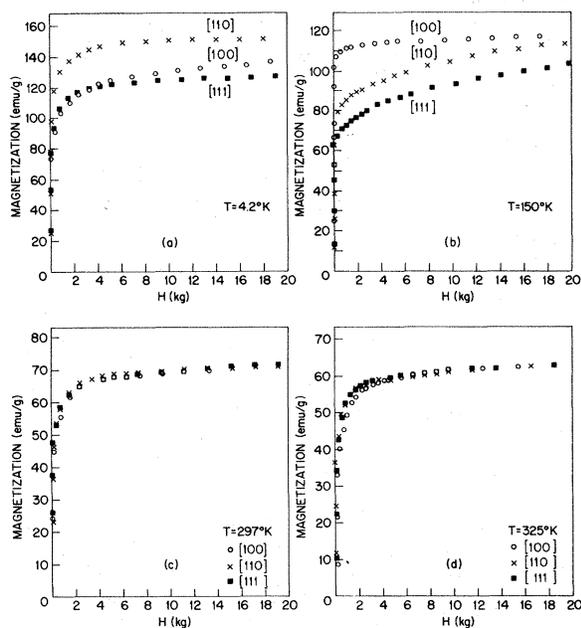


FIG. 4. (a) Magnetization plotted as a function of field at 4.2 K. (b) Magnetization plotted as a function of field at 150 K. (c) Magnetization plotted as a function of field at 297 K. (d) Magnetization plotted as a function of field at 325 K.

Since the ease of magnetization along a particular direction depends on the anisotropy energy, magnetization measurements were also made along the principal crystallographic directions at various temperatures between 4.2 and 325 K. The curves obtained after correcting for the demagnetizing factor are plotted in Figs. 4(a)–4(d). The 4.2 K curves show that the easy, intermediate, and hard directions of magnetization are the [110], [100], and [111], respectively. At 150 K, the curves are similar to those obtained at 4.2 K except the easy

and intermediate direction are interchanged. The magnetization curves obtained at the higher temperatures, 297 K, show very little difference between the principal crystallographic directions. Examination of the 325 K curve shows the [110] and [111] as equally easy directions of magnetization and the [100] as a hard direction of magnetization.

The spin directions indicated by the magnetization measurements near room temperature, like those of the Mössbauer-effect studies, differed from the spin directions obtained from the torque measurements. At low temperatures, however, all three results are in agreement. The disagreement between these results at the higher temperatures is attributed to the insensitivity of the magnetization and Mössbauer-effect studies to spin reorientations related to the magnetocrystalline anisotropy energy when sources such as strain and shape anisotropy are important. Ordinarily, one would not expect to observe a lower-order uniaxial anisotropy in a cubic crystal. However, such an anisotropy energy was detected by the presence of a large  $\sin 2\theta$  component in the Fourier-analyzed torque curve spectrum. The room-temperature value of the coefficient of the  $\sin 2\theta$  component,  $K_u$ , was found to be  $1.5 \times 10^5$  ergs/cm<sup>3</sup>, which is approximately equal to  $K_1$  and about an order of magnitude less than  $K_2$ . The origin of such a large uniaxial contribution is at present unknown, but it is believed to be related to a uniaxial strain developed during crystal growth. The different spin reorientations obtained by Mössbauer-effect studies and magnetization and torque measurements at high temperatures demonstrate the fact that the only reliable method for determining the spin reorientations due to the magnetocrystalline anisotropy energy when other sources of anisotropy are present is the torque method.

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<sup>3</sup>A. E. Clark, H. S. Belson, and N. Tamagawa, AIP Conf. Proc. **10**, 749 (1973).