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HIGH-FIELD MAGNETIZATION AND ANISOTROPY CONSTANTS OF $\text{R}_2\text{Fe}_{14}\text{B}$
(R = Tb, Dy, Ho, Er, Tm) INTERMETALLIC COMPOUNDS

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Abstract. – High-field magnetization measurements on the $\text{R}_2\text{Fe}_{14}\text{B}$ compounds have been performed under the magnetic fields up to 18 T in the temperature range 4.2-275 K. The magnetic anisotropy constants were precisely determined by the Sucksmith-Thompson plot. The temperature dependence of $K_1$ varies systematically across the series.

1. Introduction

The sintered R-Fe-B permanent magnets have received considerable interest in recent years in connection with a mechanism of coercivity [1, 2]. In order to understand the microscopic magnetic hardening mechanism in the R-Fe-B magnets, it is a prerequisite to know the intrinsic magnetic properties of the $\text{R}_2\text{Fe}_{14}\text{B}$ matrix phase. It is especially desirable to determine the magnetocrystalline anisotropy constants on the $\text{R}_2\text{Fe}_{14}\text{B}$ series. Since the large magnetic anisotropy results mainly from the crystal field of the $\text{R}^{3+}$ ions [3], an experimental study of the fundamental magnetic properties of the $\text{R}_2\text{Fe}_{14}\text{B}$ compounds has been needed under high magnetic field. In the present paper, the temperature dependence of the anisotropy constants of the $\text{R}_2\text{Fe}_{14}\text{B}$ (R = heavy rare-earth element) single crystals is described.

2. Experimental procedure

The single crystals of the $\text{R}_2\text{Fe}_{14}\text{B}$ compound with R = Tb, Dy, Ho, Er and Tm were prepared by the floating zone melting technique. The sample was successively oriented to the three principal directions [100], [110] and [001] within a few degree by using back-reflection Laue photographs. High-field magnetization measurements have been performed under the magnetic fields up to 18 T at the Service National des Champs Intenses in Grenoble. Temperature variation of the magnetization curve along each principal direction has been measured in the temperature range between 4.2 and 275 K.

3. Results

From the magnetization curves of the $\text{R}_2\text{Fe}_{14}\text{B}$ compounds measured for each of the principal axes, magnetic easy direction is parallel to the [001] axis for the compounds with $R =$ Tb and Dy. It is expected that these compounds have a large uniaxial anisotropy and a small in-plane anisotropy. For the $\text{Ho}_2\text{Fe}_{14}\text{B}$ compound, the [100] and [110] components of magnetization spontaneously appear at low temperatures only, resulting from a spin reorientation phenomenon. The compounds with $R =$ Er and Tm have magnetic easy-axis along the [100] direction through the temperature range of the measurements. The hard-axis magnetization surpasses the easy-plane magnetization above about 16 T at 4.2 K.

The magnetic total free energy $E$ composed of the anisotropy energy for a uniaxial crystal and the magnetostatic energy is phenomenologically expressed by the following equation:

$$E(\theta, \phi, \psi) = K_1 \sin^2 \theta + K_2 \sin^4 \theta + K_3 \sin^4 \theta \cos 4\phi - I_s H_{\text{eff}} \cos (\psi - \theta), \quad (1)$$

where $K_1$, $K_2$ and $K_3$ are anisotropy constants, $\theta$ and $\phi$ the polar and the azimuthal angles of magnetization, $\psi$ the angle between the direction of magnetic field and the c-axis, $H_{\text{eff}}$ the effective magnetic field assumed to be parallel to the applied field, $I_s$ the saturation magnetization deduced from the magnetization curve taken along the easy direction. In order to determine these anisotropy constants, the method of Sucksmith-Thompson [4] based on the equilibrium condition for $E$ is usually applied to the magnetization curve measured along a direction perpendicular to the easy direction.

For the uniaxial compounds with $R =$ Tb and Dy, the proportional relation in $(H_{\text{eff}} / I) \text{ versus } I^2$ plot holds good excepting in the low field range even at 4.2 K. The anisotropy constants were determined by the Sucksmith-Thompson method. But for the $\text{Ho}_2\text{Fe}_{14}\text{B}$ compound, the magnetization curves taken along the [100] and [110] directions do not yield the proportional relation below about 60 K because of the spin reorientation phenomenon. In the present analysis, the anisotropy constants were estimated from the low field region of the Sucksmith-Thompson plot where it can be expressed approximately by the following equation:

$$(H_{\text{eff}} / I) \propto a I^2 + b I + c, \quad (2)$$

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where the coefficients $a$, $b$ and $c$ are a function of $K_1$, $K_2$, $K_3$ and of the canting angle [5]. For the compounds with $R = \text{Er}$ and $\text{Tm}$, the Sucksmith-Thompson plot mainly consists of two straight lines above 150 K: one of the high-field side is corresponding to a region of magnetization saturated; the other is a process reflecting magnetic anisotropy, from which the anisotropy constants can be estimated.

The temperature dependence of the anisotropy constant $K_1$ of the $R_2\text{Fe}_{14}\text{B}$ series is shown in the figure 1. The temperature dependence of $K_1$ varies systematically across the series: $K_1$ increases monotonically with decreasing temperature in the $\text{Tb}_2\text{Fe}_{14}\text{B}$ compound; $K_1$ begins to decrease at low temperatures in the $\text{Dy}_2\text{Fe}_{14}\text{B}$ and then alters the sign from positive to negative in the $\text{Ho}_2\text{Fe}_{14}\text{B}$ at the spin reorientation temperature; on the other hand, in the compounds with $R = \text{Er}$ and $\text{Tm}$, $K_1$ is always negative below about 300 K and the absolute value increases with decreasing temperature.

4. Discussion

The values of the anisotropy constants of the $R_2\text{Fe}_{14}\text{B}$ series at 4.2 K are in agreement with the literature [6, 7]. The present data are thought to have higher precision because of the measurements under high-magnetic field and along each principal axis. The value of $K_2$ of all the present compounds are positive in the temperature range of the measurements, and increases monotonically with decreasing temperature in the compounds with $R = \text{Tb}$, $\text{Dy}$, $\text{Er}$ and $\text{Tm}$; however, $K_2$ increases rapidly below about 100 K in the $\text{Ho}_2\text{Fe}_{14}\text{B}$ compound, resulting in the spin reorientation phenomenon; in fact, the canting angle has been estimated to be $23^\circ$ at 4.2 K by $\sin \theta = \sqrt{(-K_1/2K_2)}$, which is in agreement with the value obtained from the torque measurements [8]. The absolute value of $K_3$ is an order of magnitude smaller than that of $K_1$ across the series. The sign of $K_3$ of the compounds with $R = \text{Er}$ and $\text{Tm}$ is negative, which is consistent with the result of [100] easy direction.