Magnetization of a Dy$\textsubscript{2}$Fe$_{14}$Si$_3$ single crystal in high magnetic fields

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Abstract. The magnetization of a Dy$\textsubscript{2}$Fe$_{14}$Si$_3$ single crystal was measured at 4.2 K in pulsed fields up to 52 T along the principal axes. The compound is a ferrimagnet with $T_C = 500$ K, has a spontaneous magnetic moment of $9 \mu_B$ (at 4.2 K) and exhibits a very large magnetic anisotropy, $<100>$ being the easy axis. In fields applied along the $<100>$ and $<120>$ axes, field-induced phase transitions are observed at 33 T and at 39 T, respectively. The $c$-axis magnetization curve crosses the easy-axis curve at 23 T. At higher fields, for all directions, the magnetization continues to increase due to further bending of the sublattice moments.

1. Introduction

The collinear ferrimagnet Dy$\textsubscript{2}$Fe$_{17}$ is a representative of the R$\textsubscript{2}$Fe$_{17}$ series of rare-earth (R = Ce-Lu) - iron intermetallics. As found from measurements of the single crystals, the spontaneous magnetic moment $M_s = 16.8 \mu_B$ (at 4.2 K) is located in the basal plane of the hexagonal crystal structure of the Th$_2$Ni$_{17}$ type [1-3]. A study of polycrystalline R$_2$Fe$_{14}$Si$_3$ compounds [4], which are solid solutions of Si in R$_2$Fe$_{17}$, has revealed a strong modification of both the intra- and inter-sublattice interactions compared to R$_2$Fe$_{17}$. The effect is rather complicated, because the Fe-sublattice moment is weakened by the dilution with the non-magnetic Si, whereas the Fe-Fe exchange interaction is unexpectedly enhanced ($T_C$ is 370 K for Dy$_2$Fe$_{17}$ and 500 K for Dy$_2$Fe$_{14}$Si$_3$). It is therefore not a priori clear, whether dilution of the Fe sublattice will shift the first-order transitions in the magnetization, predicted at 54 T for $H||a$-axis and at 70 T for $H||b$-axis in Dy$_2$Fe$_{17}$ [5], to yet higher fields (possibly out of experimental reach) or whether a reduction of the transition fields will take place.

Due to the strong magnetic anisotropy of rare-earth intermetallics, single crystals are strongly desirable for quantitative studies of their magnetism. In this paper, we present the results of a high-field magnetization study of a Dy$_2$Fe$_{14}$Si$_3$ single crystal.
2. Experimental
A single crystal of Dy$_x$Fe$_{14}$Si$_3$ of 20 mm length and 4 mm diameter has been grown by a modified Czochralski method in a tri-arc furnace from a stoichiometric mixture of the pure elements (99.9% Dy, 99.98% Fe and 99.999% Si). The lattice parameters determined by X-ray powder diffraction $a = 840.8$ pm, $c = 826.8$ pm are in good agreement with literature [4]. The magnetization was measured along the $a$-axis ($<100>$), the $b$-axis ($<120>$) and the $c$-axis ($<001>$) by the extraction method in static fields up to 5 T by using a SQUID magnetometer (Quantum Design). The high-field magnetization has been measured in the Center for Quantum Science and Technology under Extreme Conditions (KYOKUGEN) at Osaka University, in pulsed fields up to 52 T with pulse duration of 20 ms. The magnetization curves presented in this paper have been corrected for the demagnetizing field.

3. Results and discussion
The magnetization curves along the principal axes show that Dy$_x$Fe$_{14}$Si$_3$ is a highly anisotropic ferrimagnet with easy $a$-axis. The ferrimagnetism is manifested in Fig. 1 by the value of the spontaneous magnetization $M_s$ that increases with increasing temperature. There is pronounced anisotropy within the basal plane, which vanishes above 160 K where the $a$- and $b$-axis magnetization curves coincide. The anisotropy between the basal plane and the $c$-axis persists at much higher temperatures; the anisotropy field still exceeding 5 T at 300 K.

![Figure 1. Magnetization curves of a Dy$_x$Fe$_{14}$Si$_3$ single crystal measured along the principal axes at different temperatures.](image)

At low temperatures, the basal-plane magnetization curves of Dy$_x$Fe$_{14}$Si$_3$ exhibit a strong characteristic hysteresis with a very low initial susceptibility, abrupt saturation in a narrow field range interval, rectangular hysteresis loops and a drastic exponential decrease of the coercive field with increasing temperature. All these features agree well with the model of high intrinsic coercivity due to narrow domain walls, applicable to highly anisotropic magnets at low temperatures. However, the easy-plane...
type of magnetic anisotropy is usually not strong enough to provide a domain-wall width of the order of several interatomic distances needed for the effective domain-wall pinning in the framework of this model. Such hysteresis is not observed in Dy$_2$Fe$_{17}$ without substituted Si whereas the magnetization and the anisotropy of Dy$_2$Fe$_{17}$ and Dy$_2$Fe$_{17}$Si$_3$ differ only quantitatively. The same situation occurs in Tb$_2$Fe$_{17}$Si$_x$ compounds: strong hysteresis appears only in the compounds with Si [6]. In our opinion, the effective domain-wall pinning in R$_2$Fe$_{17}$Si$_x$ compounds with multiaxial (e.g., easy-plane) magnetic anisotropy originates from concentration inhomogeneities on a microscopic level, which are always present in quasibinary systems. The hysteretic properties of Dy$_2$Fe$_{14}$Si$_3$ will be discussed in detail elsewhere.

Figure 3 shows the high-field magnetization curves at 4.2 K. The magnetic isotherm measured with the field applied along the $a$-axis, the easy magnetization direction in the easy plane, exhibits a spontaneous magnetization $M_s$ of about 9 $\mu_B$/f.u. Because we consider the magnetization at low temperatures, we may assume that the magnetic moment of the Dy sublattice $M_{Dy}$ is equal to the free-ion value of 20 $\mu_B$/f.u. The ferrimagnetic arrangement of the Dy and Fe moments implies that the moment of the Fe sublattice $M_{Fe}$ equals about 29 $\mu_B$/f.u. In the field range from 30 to 33 T, the $a$-axis magnetization exhibits a strongly hysteretic transition. A similar, but much less hysteretic, transition is found at about 39 T if the field is applied in the $b$-axis direction, the hard magnetization direction in the easy plane The magnetization measured along the $c$-axis, the hard magnetization direction, crosses the easy $a$- and $b$-axis curves around 22-23 T and continues to increase above this field value because the magnetization process consists not only of rotation of the total magnetic moment toward the field direction but also of field-induced non-collinearity of the ferrimagnetically coupled sublattices.

![Figure 2](image.png)

**Figure 2.** Magnetization curves of a Dy$_2$Fe$_{14}$Si$_3$ single crystal measured at 4.2 K along the principal axes in pulsed magnetic fields (lines). The symbols represent the static-field results.

The magnetization behavior observed for Dy$_2$Fe$_{14}$Si$_3$ has been observed for the first time for Ho$_2$Co$_{17}$ [7] and has later been shown to occur in many other ferrimagnetic R$_2$Fe$_{17}$ and R$_2$Co$_{17}$ compounds with easy-plane type of anisotropy. The high-field magnetic transitions, that take place in these compounds if the field is applied within the basal-plane directions, have been established to be based on the
competition between the Zeeman energy (strength of the applied field) and the strength of the 4f-3d exchange interaction.

If the general interpretation is applied to the $a$-axis magnetization of Dy$_2$Fe$_{14}$Si$_3$, it means that the low-field collinear magnetic structure, in which the Dy moments lie along the $a$ axis in the basal plane, antiparallel with the Fe moments, is broken around 30 to 33 T and a transition takes place to a moment configuration in which the Dy moments jump to another easy direction and in which magnetostatic energy is gained at the expense of exchange energy. For the field applied along the $b$-axis, this transition takes place at a field strength of about 39 T.

On the basis of simple model calculations, for the situation with the field applied along the $a$-axis, in total three transitions are expected until the parallel, forced ferromagnetic, arrangement of the two sublattice magnetizations is reached. When the field is applied along the $b$-axis, two transitions are expected. Besides predictions of high-field transitions [5], analysis of the magnetization curves of compounds in which such transitions have been observed has led to knowledge of the crystal-field coefficients for these compounds and has provided values for the strength of the 4f-3d interaction [8].

Comparison of the present experimental results on Dy$_2$Fe$_{14}$Si$_3$ and predicted [5] results on Dy$_2$Fe$_{17}$ learns that, for Dy$_2$Fe$_{14}$Si$_3$, the transition fields are distinctly lower. For the $a$-axis magnetization, 30-33 T for Dy$_2$Fe$_{14}$Si$_3$ and 54 T for Dy$_2$Fe$_{17}$. For the $b$-axis, 39 T and 70 T. In order to make a detailed comparison of the magnetization behavior of the two compounds, mean-field calculations of the magnetic isotherms are presently being carried out.

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