

Probing $3d-4f$ Exchange Interactions by High-Field Magnetization Measurements

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In a $4f-3d$ ferrimagnet the molecular field (a net value expressing the relevant exchange interactions) can be determined from high-field magnetization measurements. If the applied field is high enough, it breaks the ferrimagnetic ground state and drives the system towards ferromagnetic order via non-collinear intermediate phases. Magnetization curves of $\text{Er}_2\text{Fe}_{17}$ single crystal were measured along the [100] crystallographic direction in magnetic fields up to 50 T. An unfixed sample, free to rotate, was measured as well. The magnetization measured along the easy magnetization direction shows jumps at 37.5 and 44 T. The first jump was used for the determination of the molecular field (66.4 T). This agrees with the data for the unfixed sample where a kink in the magnetization at 33.5 T yields a molecular field of 65.7 T. The obtained values are in a good agreement with literature data.

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1. Introduction

Intermetallic R–T compounds, where R is a rare earth element, and T is a $3d$ transition metal, are intensively investigated in last decades. The interest is justified by their applicability as a permanent magnet material as well as extremely rich phase diagram. In this class of compounds the highly anisotropic, localized $4f$ shell of the R ion is embedded in the matrix of T sublattice possessing large magnetic moment. *Light* rare earths tend to order parallel to the T sublattice, which is favorable for applications. In the case of *heavy* rare earths antiparallel, or ferrimagnetic order is observed. The latter fact is responsible for numerous field-induced phase transitions [1].

The molecular H_{mol} is a fundamental quantity, expressing the total of the exchange interactions binding R and T magnetic sublattices. Direct measurement of this quantity is hindered by interference of the crystal field (CF), also acting on the R ion. The most direct method of measuring field acting on the R ion — the inelastic neutron scattering — is unable to disentangle the H_{mol} and CF in most cases [2].

For the R–T compounds based on heavy rare earth ions one can break the ferrimagnetic order by applying magnetic field, thus overcoming R–T exchange interactions. Deducing the value of H_{mol} may be not straightforward in this case either, due to the magnetic anisotropies of

both subsystems. The anisotropy is also closely related with the crystal fields.

In this work we show that in case of easy-plane type of magnetic anisotropy one can obtain the value of molecular field from high-field magnetization experiments. The idea is illustrated by magnetization curves of $\text{Er}_2\text{Fe}_{17}$ single crystals measured on unclamped and fixed samples.

2. Experimental details

The starting ingot of $\text{Er}_2\text{Fe}_{17}$ was prepared by inductive melting of 99.9%-pure erbium and 99.998%-pure iron in an alundum crucible. It was cooled down at a rate of 10 K/min to provide favorable conditions for growing large enough single-crystalline grains. The ingot was broken up by ultrasound and large (up 200 mg) grains were picked up. The quality of single crystals was checked by the Laue backscattering measurements, optical metallography, and atomic force microscopy. The samples were spherically shaped, the stressed surface was relaxed by electro-chemical polishing. The crystals used for experiments on fixed samples were oriented by the Laue backscattering.

The magnetization measurements were performed at the pulsed-field facility of the IFW Dresden. Magnetic fields up to 47 T with a rise time of about 8 ms were used [3]. The magnetization value was obtained by integrating the voltage induced in a pick-up coil surrounding

the sample. The voltage induced by the varying magnetic field was compensated by connecting a compensation coil of equal effective area opposite to the measurement pick-up coil.

Two pulsed magnetometers were involved in the measurements. The magnetization of the fixed sample was measured by a coaxial magnetometer, where the compensation coil was surrounding the measurement pick-up coil. The sample space had a diameter of 5 mm [4].

The free-rotation measurements were performed by a purpose-made magnetometer, where measurement pick-up and an identical compensation coils were placed one-by-another. The sample space in this case was about 1 mm.

3. Free rotation

The idea of the method is the following: If magnetic anisotropy of one of the sublattices is zero and the crystal is free to rotate, the crystal lattice will follow the magnetization vector of the other, anisotropic sublattice, effectively excluding the anisotropy from consideration. The only case of isotropic R subsystem (with non-zero magnetic moment) is R = Gd. A free-rotation experiment on the GdCo₅ compound was performed earlier [5].

The anisotropy of the T-subsystem is usually non-negligible. However, in the case of easy plane anisotropy there is still a way to circumvent the anisotropy. Here we exploit the fact that in this case all the movements of the magnetization vectors take place within the basal plane. The *in plane* anisotropy of T-subsystem is usually very small and can be neglected. So we can use the free rotation method assuming that the crystal lattice will follow the magnetization of the R-subsystem. Of course, we only can succeed if applied magnetic field is high enough to break down the intersublattice interaction.

Figure 1 shows the magnetization curve measured on an Er₂Fe₁₇ single crystal. The sample was free to rotate being restricted, however, within the pick-up coil. An upturn in the magnetization is observed at 33.5 T. According to the simple phenomenological theory developed by Clark and Callen, an isotropic ferrimagnet would abandon the collinear ordering at a critical field

$$H_{cr} = \lambda |M_T - M_R|, \quad (1)$$

where λ is intersublattice exchange constant, M_T and M_R are magnetizations of T- and R-sublattices, respectively [6]. As we now get the value of H_{cr} experimentally, we immediately obtain the value of $\mu_0 H_{mol} \equiv \mu_0 \lambda M_T = 65.7$ T. Here we take the value of the measured saturation magnetization of 82 A m²/kg below the transition, and the value of free Er ion for M_R . Despite its apparent simplicity, the described method is far from being straightforward. In pulsed fields it is limited by the field sweep rate and the final reaction time of the sample. Below the transition point, for a certain sample orientation an energy minimum exists. At the transition point this minimum flattens out, and then becomes a local maximum, surrounded by two equivalent minima. It takes

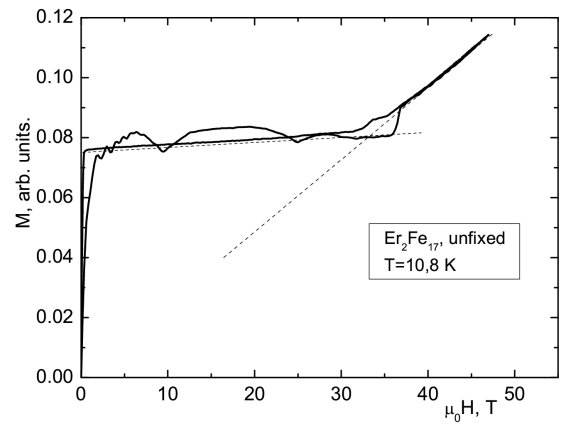


Fig. 1. Magnetization curve of the unfixed Er₂Fe₁₇ single crystal.

time for the sample to orient itself one of the minima. This process is reflected in the magnetization curve. The bigger the sample, the slower the reaction time and eventually the transition point could be missed completely.

4. Fixed sample

The presence of the magnetic anisotropy will modify the simple scenario described by Clark and Callen. Instead of linear increase the magnetization curve will show a number of jumps as the magnetization vector of the R sublattice passes energy minima. A full mathematical description of that is rather complicated and can be found elsewhere [7]. Here we limit ourselves to some essential formulae.

The key model parameter is the ratio of the sublattice magnetizations, $m = M_R/M_T$, usually, a well-known function. It uniquely determines the value of the so-called first tricritical field h_{C_1} (a dimensionless auxiliary function) through the following explicit expression:

$$h_{C_1} = \frac{1}{3} - m + \frac{2}{3} \sqrt{1 + \frac{9}{35}m} \times \cos \left(\frac{1}{3} \arccos \frac{1 + \frac{27}{70}m - \frac{81}{70}m^2}{(1 + \frac{9}{35}m)^{3/2}} \right). \quad (2)$$

Let a first-order transition be observed at a field H_1 , where the magnetization receives a relative increment $\Delta\sigma_1$. Then the molecular field is given by

$$H_{mol} = \frac{H_1}{h_1}, \quad (3)$$

where

$$h_1 = h_{C_1}(m) + \Delta\sigma_1 \left[\frac{1-m}{1-m/4} - h_{C_1}(m) \right] \times \frac{2-m+m^2/2}{m(1-m/4)}. \quad (4)$$

The magnetization curve of Er₂Fe₁₇ measured along the easy axis a , [100] is shown in Fig. 2. Two transitions are visible. The higher-field one at about 44 T has a hysteresis width of about 2 T. For our purpose

the first, low-field transition is important. It is located at $\mu_0 H_1 = 37.5$ T. The magnetization plateau below this first transition is $82 \text{ A m}^2/\text{kg}$, the relative height of the first jump is $\Delta\sigma_1 = 0.18$. For $\text{Er}_2\text{Fe}_{17}$ we have the following parameters: $m = 0.49$, $h_{C_1} = 0.531$, and $h_1 = 0.565$. For the molecular field we obtain $\mu_0 H_{\text{mol}} = \mu_0 H_1/h_1 = 66.4$ T.

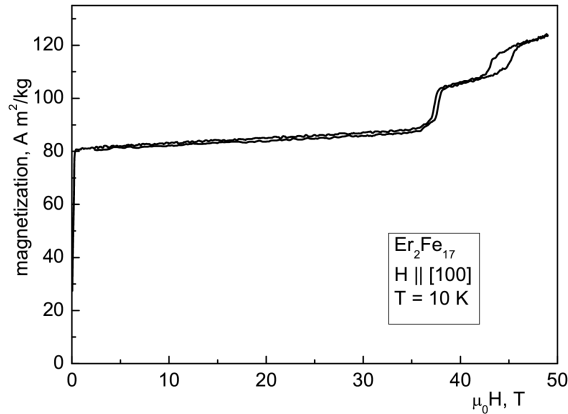


Fig. 2. Magnetization curve of the $\text{Er}_2\text{Fe}_{17}$ measured along the a [100] crystallographic direction.

The value obtained by this technique is in a good agreement with the value obtained by the free rotation method. Both values are consistent with the previously published data on $\text{Er}_2\text{Fe}_{17-x}\text{Al}_x$. The authors of [8] have obtained the value of $\mu_0 H_{\text{mol}} = 66.7$ T.

5. Conclusions

Magnetization curves of $\text{Er}_2\text{Fe}_{17}$ single crystal have been measured along the [100] crystallographic direction in magnetic fields up to 47 T. An unfixed sample, free

to rotate, was measured as well. The magnetization measured along the easy magnetization direction shows jumps at 37.5 and 44 T.

We have proven the possibility to determine the value of molecular field for easy-plane R-T compounds. The first technique is based on eliminating magnetic anisotropy by allowing a free sample rotation. The second one is a mathematical algorithm for calculating H_{mol} from the magnetization curve of the easy direction showing a first-order transition.

Both methods deliver relevant results, which are consistent with each other as well as with literature data.

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