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High-field-magnetization study of an $ErMn_6Sn_6$ single crystal

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Abstract

In previous investigations, it was shown that the compound $ErMn_6Sn_6$, that crystallizes in the $HfFe_6Ge_6$-type of structure, orders in a helimagnetic state at $T_N = 352$ K, with the moments rotating within the [0 0 1] plane. In the helimagnetic state, metamagnetic behavior is observed, the critical field increasing when the temperature decreases from T_N to 220 K and then decreasing with further decrease of the temperature from 200 to 75 K. At this latter temperature, the compound orders ferrimagnetically with the moments in the [0 0 1] plane. Very peculiarly, the in-plane magnetocrystalline anisotropy was found to decrease with temperature decreasing from 125 to 4.2 K. The magnetization of $ErMn_6Sn_6$ single crystals has been measured in magnetic fields up to 51 T. The measurements were performed at 4.2 K in the ferrimagnetic state and at 125 K in the helimagnetic state with the magnetic field applied along the three principal crystallographic directions. The measured magnetic isotherms are discussed in terms of the competition between the Er-Mn exchange interaction and the magnetocrystalline anisotropies of the Er and the Mn magnetic sublattice.

Keywords: $ErMn_6Sn_6$; High-field magnetization; Field-induced transition

1. Introduction

The compound $ErMn_6Sn_6$ crystallizes in the $HfFe_6Ge_6$-type of structure and has a Néel temperature T_N 352 K and a Curie temperature T_c 75 K [1]. In the antiferromagnetic state, metamagnetic behavior is observed. Neutron-diffraction studies and ^{166}Er Mössbauer spectroscopy have shown a high-temperature helimagnetic structure with propagation vector [0 0 q_z], the moments rotating in the [0 0 1] plane and a low-temperature ferrimagnetic structure with the moments perpendicular to the *c*-axis [2]. The metamagnetic process has been extensively studied by means of free-powder high-field (40 T) magnetization measurements in the temperature range 77–400 K [3]. An increase of the critical field has been observed from the Néel temperature down to about 220 K and then a decrease from 200 K to the Curie temperature. These results have been attributed to an exchange-related metamagnetic transition. Other free-powder high-field (38 T) magnetization measurements have been undertaken at 4.2 K [4]. At this temperature, a field-induced transition with hysteretic behavior from the ferrimagnetic to the canted-moment phase was observed around 21 T and from this measurement an estimate of the Er-Mn exchange-coupling constant $J_{ErMn}/k = -10.3$ K has been obtained. A magnetization study performed on a single-crystal in applied fields up to 9 T has revealed some additional interesting features [5]. The magnetic isotherms in the ferrimagnetic state confirm the easy-plane anisotropy but exhibit an anisotropy field that decreases with decreasing temperature. This is quite different from the behavior of ferrimagnetic $TmMn_6Sn_{6-x}Ga_x$ ($x \leq 0.51$) compounds, that show an increasing easy-plane anisotropy upon cooling [6], and indicates the importance of higher-order crystal-field terms.

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2. Experimental

$ErMn_6Sn_6$ single crystals have been synthesized in a flux method similar to the method previously reported by Clatterbuck and Gschneidner [5]. A mixture of $ErMn_6Sn_6$, obtained by induction melting, and a large amount of pure Sn metal, with an overall atomic ratio $ErMn_6Sn_{30}$, is compacted into pellets and put into a silica tube. A quartz–wool stopper is introduced into the silica tube, which is sealed under argon (267 mbar). The tube is placed in a furnace and quickly heated to 1273 K (at 50 K/h) where it remains during 24 h. The furnace is then slowly cooled down to 1223 K (6 K/h), heated again up to 1263 K at the same rate and finally slowly cooled down to 873 K during 65 h. The tube is quickly removed from the furnace, inverted and centrifuged manually using a David's sling device. After this treatment, the single crystals remain on the quartz–wool stopper and the Sn flux lies at the bottom of the tube. This method produces relatively large hexagonal prisms with masses up to 100 mg. Some of them were ground and analyzed by X-ray diffraction by means of a Guinier camera witk Cu Kα radiation with high-purity Si as calibration (a = 5.43082 Å).

The thermal variation of the magnetization $ErMn_6Sn_6$ has been measured in a MANICS magneto-susceptometer in a field of 0.1 T in the temperature range from 5 to 450 K. The transition temperatures (Tc and the spin-reorientation temperature T_{sr}) have been taken at the maximum of the first derivative dM/dT. The high-field magnetization has been measured at KYOKUGEN in Osaka University by using a pulsed magnet up to 51 T. The high-field measurements were performed on a sample consisting of stacked platelets, all oriented in the same direction. The total masses of the samples varied between 4.14 and 7.14 mg. The fields were applied along the a-axis (the [1 0 0] direction), the b-axis (perpendicular to both the a-axis and the c-axis, also indicated as the [1 2 0] direction) and the c-axis (the [0 0 1] direction).

3. Results and discussion

Analysis of the Guinier patterns of $ErMn_6Sn_6$ shows that its structure is isotypic with the $HfFe_6Ge_6$ structure. The lattice parameters are a = 5.516(2) Å, c = 8.999(5) Å. The corresponding unit-cell volume is V = 237.1(3) Å^3. The c/a ratio equals 1.631.

The temperature dependence of the magnetization of $ErMn_6Sn_6$ in a field of 0.1 T applied parallel and perpendicular to the c-axis is shown in Fig. 1. These results largely confirm the results obtained on powder samples, the values T_c = 62 K and T_N = 360 K being very close to those obtained previously. The magnetic isotherms of $ErMn_6Sn_6$, measured at 4.2 and 125 K along the [1 0 0], [1 2 0] and [0 0 1] direction, are shown in Fig. 2. The open symbols in the low-field region of the figure represent results of SQUID measure-

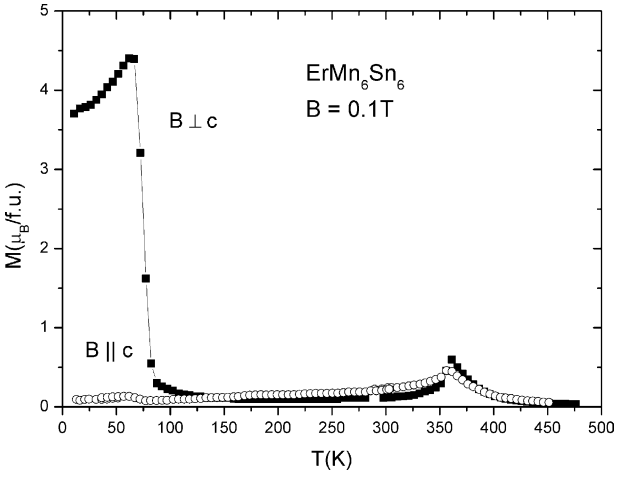

Fig. 1. Thermal variation of the magnetization of $ErMn_6Sn_6$ in a field of 0.1 T, applied parallel and perpendicular to the [0 0 1] direction. No correction has been applied for the demagnetizing field.

ments used for calibration. We will first discuss the 125 K data in Fig. 2. A comparison of the 125 K isotherms in Fig. 2a and b shows that there is hardly any anisotropy within the basal plane. The strong increase of the magnetization in the low-field part is attributed to the attainment of the ferrimagnetic configuration. This configuration is reached in substantially higher fields when the field is applied along the c-direction. The reason for this is the fact that the Er and Mn sublattice both have to abandon their easy moment direction. The steady increase of the magnetization in the higher-field parts of the 125 K isotherms points to a gradual bending of the two sublattice moments towards each other, a bending that is still incomplete at the highest field applied.

The plateau in the low-field part of the isotherm measured at 4.2 K corresponds to the ferrimagnetic configuration with $M = M_{Mn} - M_{Er}$, as in the 125 K isotherms. The magnetization of the ferrimagnetic state is lower at 4.2 K than at 125 K. The reason of this is the fact that the Er-sublattice moment increases strongly when going from 125 K to 4.2 K, while the Mn-sublattice moment does not change much. One may also notice in Fig. 2 that the value of the magnetization in the plateau region of the isotherms measured at 4.2 K is the same for all three directions of the applied field. This suggests that the magnetic anisotropy is fairly low at this temperature. In fact, when comparing the low-field part of the isotherm measured at 4.2 K for the hard direction with that measured at 125 K for the same direction (Fig. 2c), one finds that the anisotropy has substantially decreased with decreasing temperature. Such an unusual behavior is a clear signature of a crystal-field-induced anisotropy consisting of competing contributions of various orders that have a different temperature dependence. We also observe some minor differences in the magnetization behavior within the [0 0 1] plane (Fig. 2a and b). It is worth noting that the magnetization quickly and well saturates around 4 μ_B/f.u. when the

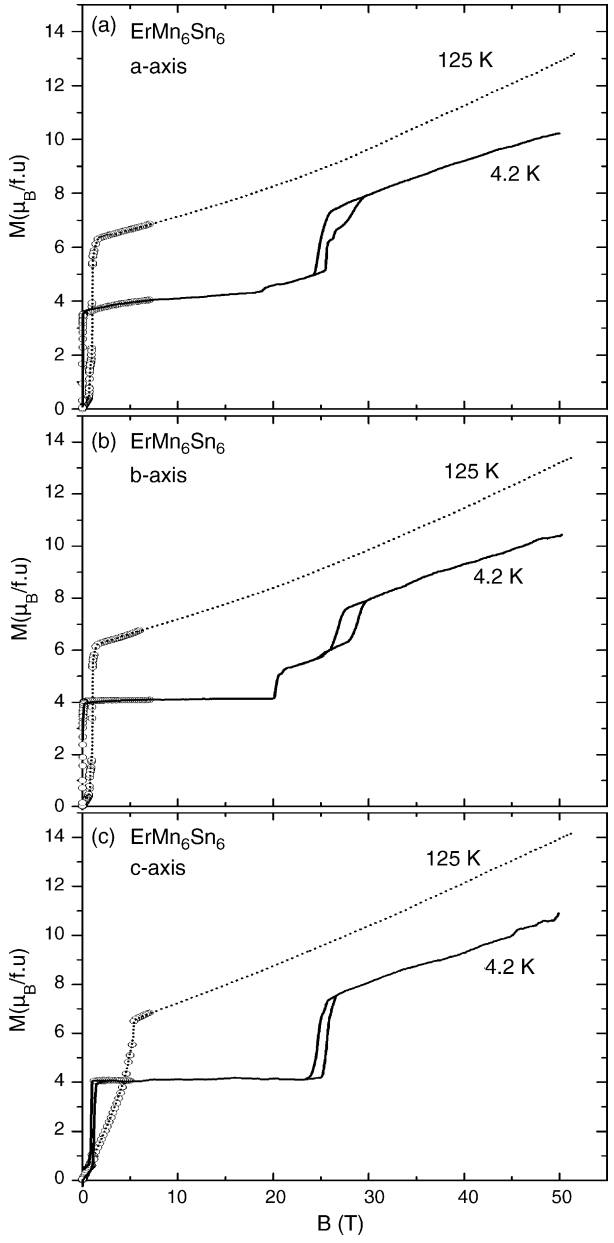

Fig. 2. Magnetization of $ErMn_6Sn_6$, measured at 4.2 K and 125 K along (a) [1 0 0]; (b) [1 2 0]; and (c) [0 0 1] direction. The symbols represent SQUID measurements for calibration.

field is applied along the *b*-direction. In contrast, when the field is applied along the *a* direction, the starting value is close to 3.5 μ_B/f.u and increases slowly in increasing fields. This suggests that the easy direction is parallel to the *b*-direction. Therefore, in zero field we should measure parallel to the *a*-axis $4 \times \cos 30^\circ = 3.46 \mu_B$/f.u. that is close to the observation. Above 20 T, all isotherms measured at 4.2 K show one (Fig. 2c) or two (Fig. 2a and b) field-induced transitions. At these field strengths, the applied field is strong enough to overcome the intersublattice molecular field and a breaking of the ferrimagnetic moment arrangement is initiated. Such transitions have been previously observed in free-powder measurements [4]. These results offer the possibility to obtain a rough value of the Er-Mn intersublattice coupling constants, as has been explained already in more detail in Ref. [4]. The occurrence of different field-induced transitions along the *a*-axis and the *b*-axis could be attributed to the symmetry of the easy *b*-axis anisotropy in the hexagonal basal plane, which causes different moment configurations during bending process.

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References

[1] G. Venturini, B. Chafik El Idrissi, B. Malaman, J. Magn. Magn. Mater. 94 (1991) 35.
[2] B. Malaman, G. Venturini, R. Welter, J.P. Sanchez, P. Vulliet, E. Ressouche, J. Magn. Magn. Mater. 202 (1999) 519.
[3] J.F. Hu, F.M. Yang, Y.H. Wang, J.L. Wang, F.R. de Boer, Phys. Stat. Sol. B 214 (1999) 135.
[4] J.H.V.J. Brabers, G.F. Zhou, J.H.P. Colpa, K.H.J. Buschow, F.R. de Boer, Phys. B 202 (1994) 1.
[5] D.M. Clatterbuck, K.A. Gschneidner Jr., J. Magn. Magn. Mater. 207 (1999) 78.
[6] F. Canepa, M. Napoletano, C. Lefevre, G. Venturini, J. Magn. Magn. Mater., submitted for publication.