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High-field magnetization of single crystals of $RMn_6Ge_{6-x}Ga_x$ compounds with R = Tb, Er, Tm, Yb and $x \approx 1$

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Abstract

The magnetization has been measured of single crystals of $RMn_6Ge_{6-x}Ga_x$ (R = Tb, Er, Tm and Yb and $x \approx 1$) compounds at 4.2 K in high magnetic fields up to 65 T applied along the main crystallographic directions. At low temperature, where the magnetocrystalline anisotropy is determined by the R contribution, these compounds exhibit easy-axis anisotropy with the *c*-axis as preferred moment direction. The magnetization display a variety of field-induced phase transitions which are particularly pronounced in the compounds with Tb, Er and Tm. The Yb compound exhibits the onset of a phase transition in the field region above 65 T. The results are discussed in terms of the interplay between the antiferromagnetic intersublattice R–Mn coupling and the magnetic anisotropy.

Keywords: $RMn_6Ge_{6-x}Ga_x$; High-field magnetization; Field-induced transition; Ferrimagnet; Magnetocrystalline anisotropy

1. Introduction

The hexagonal $HfFe_6Ge_6$-type (P6/*mmm*) RMn_6Ge_6 compounds, with R = Sc, Y or one of the rare-earth elements, have recently been subject of many studies [1–5]. Due to the complicated interplay between the various intra- and intersublattice magnetic interactions and the magnetocrystalline anisotropy of both the R and the Mn sublattice, they display a wide variety of magnetic behaviour. Recently, investigations of $RMn_6Ge_{6-x}Ga_x$ solid solutions (R = Sc, Y, Gd–Lu) have shown that by partial replacement of Ge by Ga the ferrimagnetic state, with Curie temperatures (T_C) well above room temperature, is stabilized for all these R elements [6–8]. In the compounds in which the R component does not possess a magnetic moment the Mn-sublattice moment orders ferromagnetically with preferred moment direction perpendicular to the *c*-axis. If also the R component has a magnetic moment, the latter couples antiparallel to the Mn moments and imposes a strong contribution to the total magnetocrystalline anisotropy upon cooling from room temperature to cryogenic temperatures, leading to spin-reorientation transitions at intermediate temperatures (T_{sr}, see Table 1). The dominance of the R-sublattice anisotropy at low temperatures leads to preferred moment directions characterized by an easy axis for the compounds with R = Tb, Er, Tm and Yb and by an easy cone for R = Dy and Ho.

In a recent investigation [9], we have further studied the magnetic properties of single crystals of $RMn_6Ge_{6-x}Ga_x$ compounds by measurements of the magnetization at 4.2 K in high magnetic fields up to 55 T applied along the main crystallographic directions. The interplay between the antiferromagnetic intersublattice R–Mn coupling and the magnetocrystalline anisotropy was found to lead to very pronounced field-induced magnetic phase transitions in the compounds with Tb, Dy, Ho, Er and Tm. The observed magnetization processes for the compounds with Tb, Er, Tm and Yb made an extension of the measurements to higher fields highly desirable. Therefore, these compounds have been selected for

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Table 1
Unit-cell parameters, Ga contents and magnetic-transition temperatures of $RMn_6Ge_{6-x}Ga_x$ compounds with R = Tb, Er, Tm and Yb (see [9] for further information)

R	x	a (Å)	c (Å)	c/a	T_C (K)	T_{sr} (K)
Tb	0.78	5.228(2)	8.221(5)	1.5725	408	196
Er	0.91	5.212(3)	8.206(5)	1.5744	361	186
Tm	0.87	5.209(3)	8.208(5)	1.5757	352	45
Yb	0.63	5.198(3)	8.182(5)	1.5741	358	210

the present study of the evolution of the magnetization in magnetic fields up to 65 T.

2. Experimental

The $RMn_6Ge_{6-x}Ga_x$ crystals for the present study have been obtained by a flux method [9]. It produces well shaped hexagonal prisms with masses of 1–5 mg. Some of the crystals have been analyzed by X-ray diffraction using a Guinier camera (Co Kα). The Ga content has been checked at the Service Commun d'Analyse par Microsondes Electroniques de l'Université de Nancy I-Henri Poincaré using an SX50 electron probe. The results of these characterizations are collected in Table 1.

High-field magnetization studies have been carried out in the High-Field Installation at the University of Amsterdam in fields up to 40 T and in the Research Center for Materials Science at Extreme Conditions at Osaka University in pulsed fields up to 65 T with pulse duration of 7 ms. The fields were applied along the *a*-axis (the [1 0 0] direction), the *b*-axis (the [1 2 0] direction) and the *c*-axis (the [0 0 1] direction) in some compounds. The high-field measurements were performed on samples consisting of several stacked platelets, all oriented in the same way. The total mass of the samples varied between 2.6 and 13.9 mg, depending on R element and the field direction.

3. Results and discussion

The high-field-magnetization of the compound $TbMn_6Ge_{5.22}Ga_{0.78}$ obtained at 4.2 K in magnetic fields up to 65 T applied in the *a*-, *b*- and *c*-directions, is shown in Fig. 1. At 4.2 K, the compound has *c*-axis anisotropy which is dictated by the Tb anisotropy. The collinear antiparellel configuration of the Tb- and Mn-sublattice moments in zero field, with a resulting moment of 2.4 μ_B, is depicted in Fig. 1: the total Mn-sublattice moment being larger than the Tb-sublattice moment. If we assume the Tb moment to be equal to the free-ion value of 9 μ_B, we can estimate the moment of the Mn sublattice to be equal to 11.4 μ_B. If the field is applied along the *c*-direction, the antiparallel moment configuration is maintained up to about 52 T, where a large jump in the magnetization occurs to a bended-moment configuration, as indicated in Fig. 1. The occurrence of hysteresis indicates that

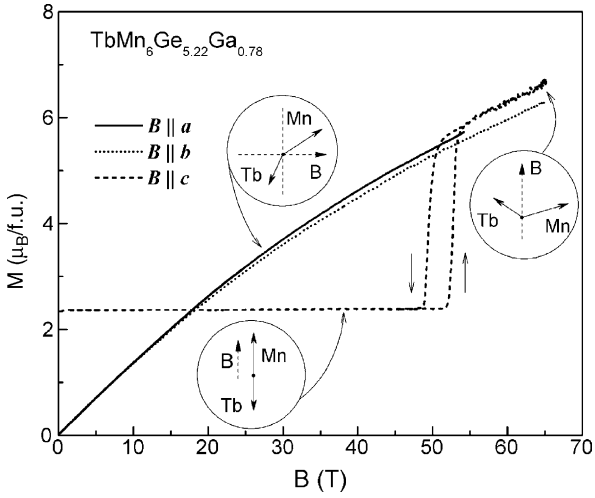

Fig. 1. Magnetization curves of $TbMn_6Ge_{5.22}Ga_{0.78}$ at 4.2 K.

it is a first-order transition. Above the transition, the magnetization further increases as both sublattice moments further bend into the direction of the applied field. When the field is applied within the basal plane, along the *a*-axis or the *b*-axis, the total moment turns away from the *c*-axis and bends towards the field direction, thereby retaining the antiparallel configuration of the sublattice moments as much as possible. The magnetizations along the *a*- and the *b*-direction are quite similar, indicating the anisotropy within the basal plane to be very small, and no magnetization jump is observed up to 65 T in the *b*-direction.

The magnetizations for $ErMn_6Ge_{5.09}Ga_{0.91}$ at 4.2 K measured by pulsed fields are presented in Fig. 2. Like in the Tb compound, also in this compound the easy magnetization direction is along the *c*-axis. The ferrimagnetic moment arrangement is the same as in the case of the Tb compound, the resulting moment being 2 μ_B. With the free-ion value of 9 μ_B for Er, we can estimate the moment of the Mn-sublattice to be 11 μ_B. At about 32 T, there is a large jump in the magnetization, from the strict antiparallel moment configuration to a

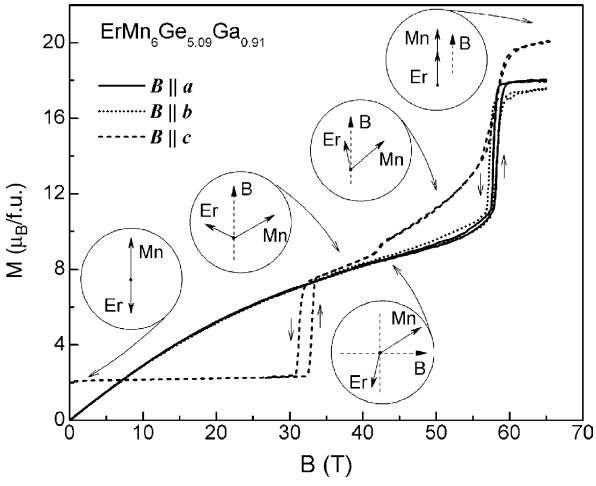

Fig. 2. Magnetization curves of $ErMn_6Ge_{5.09}Ga_{0.91}$ at 4.2 K.

bended configuration. The transition field of 32 T is appreciably lower than the field of 52 T for the Tb compound which may be due to the smaller intersublattice exchange coupling in the Er compound. At about 42 T, a second, smaller magnetization jump, presumably to the configuration indicated in Fig. 2, is observed. Above this minor jump, the Er and Mn moments gradually bend into the direction of the applied field. At 57 T, a smooth, but large, transition is observed to the forced ferromagnetic state with a saturation magnetization of 20 μ_B/f.u. is found. With the free-ion value of 9 μ_B for the Er moment, this leads to a Mn-sublattice moment of 11 μ_B/f.u., in accordance with the value derived from the saturation moment in the ferrimagnetic state. Similar to what is found for the Tb compound, the magnetizations along the *a*- and the *b*-direction are very similar, pointing to small anisotropy within the basal plane. Magnetization jumps are observed at 58 T in the *a*-axis with moment value 18 μ_B/f.u. reached and in the *b*-axis with moment value 17.5 μ_B/f.u. reached.

Fig. 3 shows the magnetizations for $TmMn_6Ge_{5.13}Ga_{0.87}$ measured in fields up to 65 T applied along the three main crystallographic directions. Like the two compounds discussed above, also this compound has the *c*-axis as easy magnetization direction. The ferrimagnetic moment arrangement is the same as for the Tb and the Er compound with a resulting moment of 4 μ_B. With the free-ion moment of 7 μ_B for Tm this leads to a Mn-sublattice moment of 11 μ_B/f.u. If the field is applied along the *c* direction, the magnetization slightly increases in the range up to about 20 T which is most likely due to a deviation of the Tm sublattice magnetization from its easy direction. At 22 T, a magnetization jump occurs from the almost antiparallel moment configuration to a bended configuration. The transition field of 22 T is much lower than the transition fields of 52 and 32 T observed for the Tb and the Er compound, respectively. This may be due to a further reduction of the intersublattice exchange coupling in the Tm compound compared to the Tb and Er compound. Above the jump, the magnetization increase is faster than that below the jump, which corresponds to a bending process that proceeds progressively with increasing field. Above 50 T, the magnetization exhibits a tendency toward saturation which is gradually accomplished above 60 T at a value of 17 μ_B/f.u. The magnetization measured with the field applied in the hard magnetization direction, along the *a*-axis, shows that there is no spontaneous magnetization below 1.5 T. Between 2 and 18 T, there is an almost rigid rotation of both sublattice moments, implying that anisotropy is weak. In this field range, the magnetization is slightly larger than that along the *c*-axis, because of a slight deviation of the Tm moment which is favored by the easy *c*-axis anisotropy of Tm. As shown in Fig. 3, the jump in magnetization around 18 T can be interpreted as a jump of the Tm moment into the easy-axis direction which is also closer to the applied-field direction at the cost of some of the antiferromagnetic intersublattice-coupling energy. The transition field is lower than that along the *c*-axis because the configuration after the transition is favored by the easy *c*-axis anisotropy of Tm. Above the magnetization jump, the magnetization increases steadily as a consequence of further bending of the two sublattice moments. Along the *b*-axis, the jump around 20 T becomes less pronounced compared to the one in 18 T along the *a*-axis. Saturation is reached via a final jump at 51 T to a value of 17 μ_B/f.u., the same value as along the *c*-axis direction. From this result we conclude that there is no appreciable anisotropy of the magnetic moment. With the free-ion moment of 7 μ_B for Tm, the saturation moment of 17 μ_B/f.u. gives a Mn-sublattice moment of 10 μ_B/f.u. On the other hand, from the observed magnetization values in the ferrimagnetic and forced-ferromagnetic state of 4 μ_B/f.u. and 17 μ_B/f.u., respectively, a Tm moment of 6.5 μ_B and a Mn-sublattice moment of 10.5 μ_B are evaluated.

The magnetizations of $YbMn_6Ge_{5.37}Ga_{0.63}$ are presented in Fig. 4. When the field is applied in the *c*-direction, which is also for this compound the easy magnetization direction, a spontaneous magnetization of about 8.8 μ_B/f.u. is found and the magnetization increases linearly to a value of 9.2 μ_B/f.u. up to 30 T. Above this field value, the magnetization starts dis-

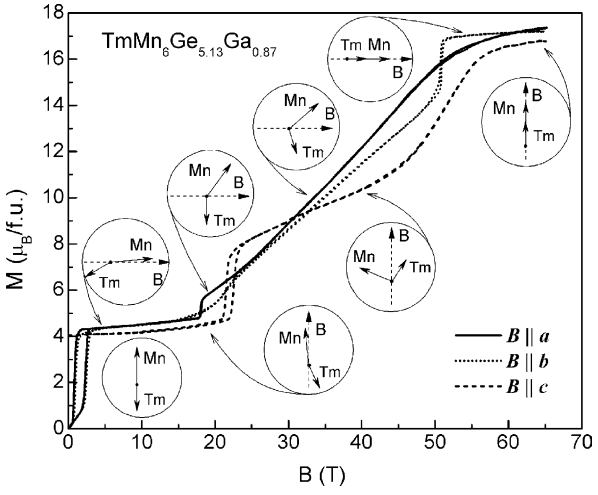

Fig. 3. Magnetization curves of $TmMn_6Ge_{5.13}Ga_{0.87}$ at 4.2 K.

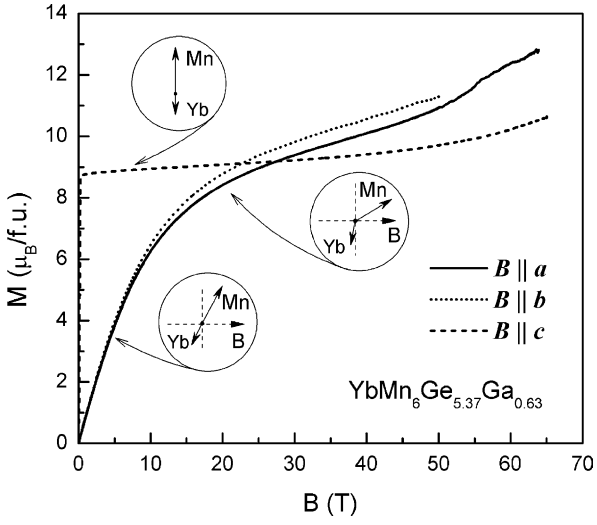

Fig. 4. Magnetization curves of $YbMn_6Ge_{5.37}Ga_{0.63}$ at 4.2 K.

playing upward curvature which becomes more pronounced with increasing field and which may be indicative for a magnetic transition above 65 T. The very high field value of this possible transition clearly does not fit into the sequence 52 T, 32 T and 22 T of the transition fields observed for the Tb, Er and Tm compound, respectively, so that we may conclude that the transition will be of another type. The magnetization along the hard direction (along the *a*-axis) shows initially almost linear behavior, corresponding to a rigid rotation of the collinearly oriented Mn- and Yb-sublattice moments. Then, a bending process in the direction of the applied field prevails and the magnetization exhibits a sluggish increase. Also the high-field part of this magnetization shows upward curvature of the magnetization, suggesting that a jump of the magnetization may occur at higher field values. The magnetization along the *b*-axis has only been measured in fields up to 50 T and does not exhibit upward curvature. Combining the free-ion value of 4 μ_B for trivalent Yb with the observed spontaneous magnetization of 8.8 μ_B/f.u., we obtain a Mn-sublattice moment of 12.8 μ_B. This value is somewhat high compared with the values estimated above for the compounds with Tb, Er and Tm that, due to experimental uncertainties, cannot be determined with great accuracy and are ranging between 10 and 12 μ_B. This, together with the unexplained high-field behavior of the magnetization, may be an indication that Yb is not perfectly trivalent in $YbMn_6Ge_{5.37}Ga_{0.63}$.

We have discussed the sequence 52 T, 32 T and 22 T of the transition fields observed for the Tb, the Er and the Tm compound, respectively, in terms of a decreasing R–Mn coupling in the same sequence. This monotonic decrease of the exchange coupling with increasing atomic number Z of the R element in a series, which is generally observed for R–T compounds, has first been reported by Beloritzky et al. [10] and has been explained in terms of an increasing 4f–5d distance and therefore decreasing 4f–5d interaction with increasing Z.

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