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Magnetic study on single crystals of YMn_6Ge_6 and $LuMn_6Ge_6$

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Abstract

The magnetic properties of single crystals of the $HfFe_6Ge_6$-type compounds YMn_6Ge_6 and $LuMn_6Ge_6$ obtained in an In flux have been investigated by thermomagnetic measurements and by a high-field magnetization study. Both the compounds display Néel point at $T_N = 482$ and 527 K, respectively. The compound YMn_6Ge_6 saturates at $M_s = 12\ \mu_B$/f.u. above 33 T, whereas $LuMn_6Ge_6$ is still not saturated in the largest applied field ($M(50\ T) = 9\ \mu_B$/f.u.), thus indicating different strengths of the antiferromagnetic interactions. The magnetization curves of YMn_6Ge_6 and $LuMn_6Ge_6$ recorded with the field applied along the *c*-axis display a field-induced transition at relatively low field (around 10 and 15 T, respectively), which should be related to spin reorientation process. In larger applied fields, all magnetization loops display two field-induced transitions with pronounced hysteretic behaviour which may be related to the successive stabilization of helical and fan structures.

Keywords: YMn_6Ge_6; $LuMn_6Ge_6$; High-field magnetization; Competing magnetic interactions; Field-induced transition

1. Introduction

The $HfFe_6Ge_6$-type compounds RMn_6Ge_6 (R = Sc, Y, rare-earth element) display a wide variety of magnetic behaviour (see [1] and references therein), indicating a complicated interplay between the Mn–Mn, R–Mn and R–R interactions and the magnetocrystalline anisotropy of both sublattices. For a better understanding of the magnetic behaviour of these compounds, a more detailed knowledge is needed of the properties of the Mn sublattice alone, which can be obtained by studying the compounds involving non-magnetic R elements, $ScMn_6Ge_6$, YMn_6Ge_6 and $LuMn_6Ge_6$, which have Néel temperatures of 516, 473 and 509 K, respectively [2]. Neutron-diffraction studies have shown that YMn_6Ge_6 orders in a collinear antiferromagnetic structure characterized by a doubling of the *c*-axis and displays a double-cone structure below 80 K [3]. A collinear antiferromagnetic structure has also been observed for $LuMn_6Ge_6$ [4]. Further neutron-diffraction studies on this compound suggested a possible double-cone structure at low temperature with a rather small semi-cone angle [5]. Finally, $ScMn_6Ge_6$ displays a quite different behaviour with an antiferromagnetic structure above 255 K and a helical structure below this temperature [5]. The magnetic-structure changes in the three compounds are not reflected in the electrical-resistivity versus temperature curves [6]. Free-powder high-field magnetization studies ($B_{max} = 38$ T) have been performed on YMn_6Ge_6 and $LuMn_6Ge_6$ [7]. It was observed that YMn_6Ge_6 displays a giant hysteresis loop extending over a field range of 15 T and that the magnetization saturates above 30 T. In contrast, the magnetization curve of $LuMn_6Ge_6$ displays only the onset of a metamagnetic transition, indicating stronger antiferromagnetic interactions. In order to obtain additional information on these compounds, high-field magnetization measurements have been performed on single crystals of YMn_6Ge_6 and $LuMn_6Ge_6$.

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2. Experimental

An In flux mixture of RMn_6Ge_6 and RGe_3 has been used to obtain single crystals of RMn_6Ge_6 with R = Y and Lu. The RMn_6Ge_6 and RGe_3 alloys, obtained by induction melting, and pure Ge were compacted into pellets and put into a silica tube with a large amount of In metal with a total atomic ratio of $R_2Mn_6Ge_{10}In_{60}$. A quartz-wool stopper is introduced in the silica tube, which is sealed under argon (267 mbar). The tube is placed in a furnace and quickly heated to 1273 K (50 K/h) where it is kept for 24 h. The furnace is then slowly cooled down to 1223 K (6 K/h), heated again up at the same rate to 1263 K, and finally slowly cooled down to 873 K in 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, the In flux being found at the bottom of the tube. The remaining flux is dissolved in diluted hydrochloric acid. Most of the crystals display a thin hexagonal platelet-like shape (with mean diameter up to 1 mm), while some of them display a needle-like shape (with length up to 2.5 mm). The crystals have been characterized by means of Guinier patterns (with Co-Kα radiation) with high-purity Si as standard (a = 5.43082 Å) and by Weissenberg photographs. The In content has been checked at the Service Commun d'Analyse par Microsondes Electroniques de l'Université de Nancy I, Henri Poincaré using the SX50 electron probe. It was found that the In concentration is smaller than the detection limit ($c_{In} < 0.1\%$)

The thermomagnetization of YMn_6Ge_6 and $LuMn_6Ge_6$ in a low field of 0.8 T has been measured with a magnetosusceptometer in the temperature range 10–520 K. The magnetization was measured with the field applied parallel and perpendicular to the c-axis. The high-field magnetization has been measured in the Research Center for Materials Science at Extreme Conditions at Osaka University in pulsed fields up to 55 T with pulse duration of 20 ms, again with the field applied parallel and perpendicular to the c-axis. The high-field experiments have been carried out on samples each consisting of several stacked platelets, all oriented in the same direction. The total mass of the samples ranged from 2.5 to 11.6 mg, depending on R (Y or Lu) and the field direction.

3. Results and discussion

The results of the thermomagnetic measurements in 0.8 T, shown in Figs. 1 and 2, indicate Néel temperatures of 482 K for YMn_6Ge_6 and 527 K for $LuMn_6Ge_6$. These ordering temperatures are slightly higher than those previously reported [2]. For both compounds, the magnetization measured with the field applied parallel to the c-axis is smaller and varies somewhat stronger with temperature than the magnetization measured with the field applied perpendicular to the c-axis. These results are in good accordance with the orientation of the moments along the c-axis as deduced from neutron

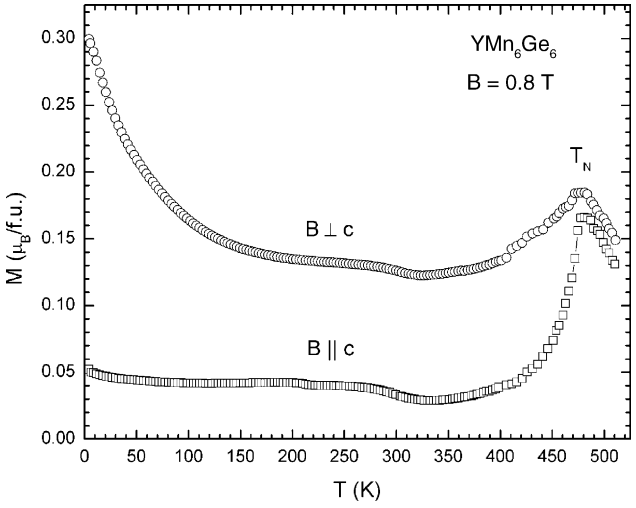

Fig. 1. Thermal variation of the magnetization of YMn_6Ge_6 in a field of 0.8 T applied parallel and perpendicular to the c-axis.

studies [3–5]. The pronounced increase of magnetization for both $B \parallel c$ and $B \perp c$ in $LuMn_6Ge_6$ at low temperatures suggests a possible parasitic paramagnetic impurity contribution. In contrast, the more sluggish increase of magnetization for $B \perp c$ in YMn_6Ge_6 may very well be intrinsic, particularly because it is absent for $B \parallel c$.

The magnetic isotherms for YMn_6Ge_6 and $LuMn_6Ge_6$ at 4.2 K are shown in Figs. 3 and 4, respectively. The isotherms of the two compounds display similar features, although the field-induced transitions in $LuMn_6Ge_6$ appear to be considerably shifted towards the high-field region and saturation is still not reached in the highest applied field of 50 T. Moreover, the hysteretic behaviour is more pronounced in YMn_6Ge_6 than in $LuMn_6Ge_6$. The general aspect of the curves well accounts for previous high-field free-powder magnetization measurements [7].

The magnetization process with the field applied perpendicular to the c-axis clearly displays two field-induced

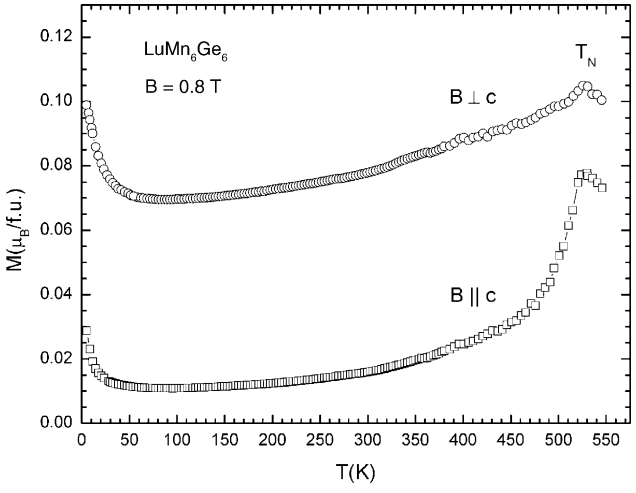

Fig. 2. Thermal variation of the magnetization of $LuMn_6Ge_6$ in a field of 0.8 T applied parallel and perpendicular to the c-axis.

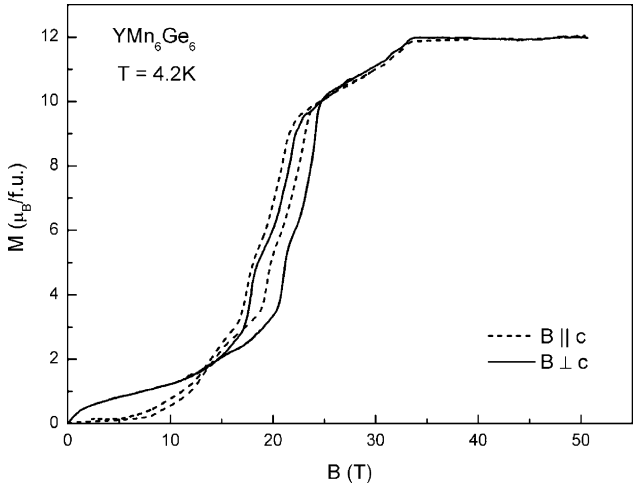

Fig. 3. Magnetization of YMn_6Ge_6 measured at 4.2 K parallel and perpendicular to the *c*-axis.

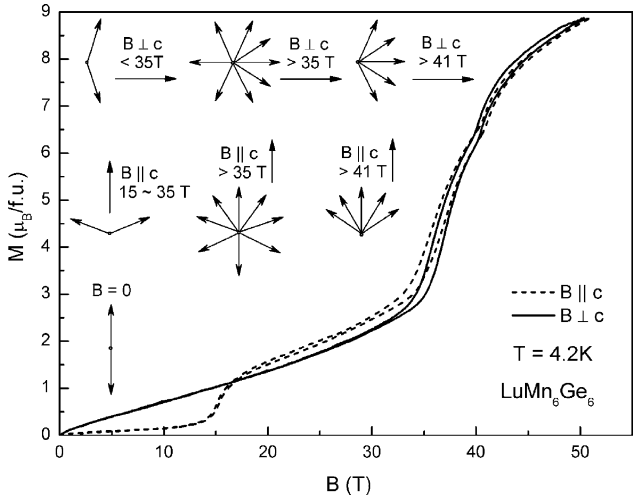

Fig. 4. Magnetization of $LuMn_6Ge_6$ measured at 4.2 K parallel and perpendicular to the *c*-axis. The possible moments configurations in $LuMn_6Ge_6$ are indicated.

transitions, around 21 and 24 T for YMn_6Ge_6 and 37 and 41 T for $LuMn_6Ge_6$. The curves with the field applied parallel to the *c*-axis display the same transitions with more or less pronounced shifts. They also show additional transition around 10 T for YMn_6Ge_6 and 15 T for $LuMn_6Ge_6$. The transitions at low field observed along the *c*-axis should be due to the spin flop transition, in which spins rotate almost 90° keeping an antiferromagnetic configuration, as indicated in Fig. 4. The other large field-induced transitions are more intricate. Within the RMn_6X_6 (R = rare-earth, X = Ge, Sn) compounds, helimagnetic structures are often observed [1]. This behaviour could be related to competing successive interlayer interactions. Possibly, the first transition at 37 T observed in the *c* plane magnetization of $LuMn_6Ge_6$ is from the canted antiferromagnetic structure towards a deformed helical structure. On the other hand, molecular-field approximation suggests a transition from a helical structure to a fan structure in the highest applied fields [8], which might explain the second jump of the magnetization at 41 T. It is possible that at higher field strengths a fan-like structure becomes stabilized, as is frequently the case. This possibility has been illustrated for $LuMn_6Ge_6$ in Fig. 4. Here, we have to bear in mind that, for $LuMn_6Ge_6$, the fan probably has not yet completely closed in the highest field applied since the magnetization is not yet saturated in Fig. 4. The fact that saturation has been reached in the case of YMn_6Ge_6 (Fig. 3) indicates that in this compound the antiferromagnetic Mn–Mn coupling is less strong than that in $LuMn_6Ge_6$.

Acknowledgments

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