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Study of the Mn–Mn exchange interactions in single crystals of RMn_6Sn_6 compounds with R = Sc, Y and Lu

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Abstract

In order to investigate the behaviour of the magnetic anisotropy and the Mn–Mn exchange interaction in RMn_6Sn_6 (R = rare earth) compounds, magnetic studies have been carried out on single crystals of RMn_6Sn_6 compounds in which R is one of the non-magnetic elements Sc, Y and Lu. The temperature dependence of the magnetization of these compounds has been measured with the field applied parallel and perpendicular to the crystallographic *c*-direction. Magnetic isotherms of the three compounds were measured at 4.2 K in magnetic fields up to 50 T, applied parallel and perpendicular to the *c*-direction, sufficiently high to reach the forced ferromagnetic state and to detect multiple field-induced transitions. For $LuMn_6Sn_6$, magnetic isotherms were measured also at a higher temperature of 150 K. From our results we may conclude that the magnetic anisotropy as well as the antiferromagnetic coupling strength increases in the sequence of $ScMn_6Sn_6$, YMn_6Sn_6 and $LuMn_6Sn_6$.

Keywords: $ScMn_6Sn_6$; YMn_6Sn_6; $LuMn_6Sn_6$; High-field magnetization; Helimagnetic

1. Introduction

The $HfFe_6Ge_6$-type RMn_6Sn_6 compounds with R = Sc, Y and Lu order antiferromagnetically with Néel temperatures at 384, 333 and 353 K, respectively [1]. Because in these compounds the R atoms are non-magnetic, the magnetic properties are mainly determined by the Mn atoms. Neutron-diffraction experiments have shown that, in the whole investigated 2–300 K temperature range for $ScMn_6Sn_6$ and YMn_6Sn_6, and at lower temperatures also for $LuMn_6Sn_6$, the magnetic structure is a flat spiral consisting of ferromagnetic (0 0 1) “Mn–Sn–Sn–Mn” sheets (with the moment directions at 2 K within the layers) stacked along the *c*-axis in the sequence Mn–R–Mn–Sn–Sn–Mn–R–Mn [2]. $LuMn_6Sn_6$ displays this type of order only below $T_t \approx 200$ K [3]. Above this temperature, a collinear antiferromagnetic arrangement prevails [3]. Measurements on single-crystalline $LuMn_6Sn_6$ lead to $T_N \approx 400$ K and $T_t \approx 250$ K and, below the latter temperature, a field-induced transition around 4 T was found in the isotherms measured along the *a*-direction [4]. However, the magnetization does not show any sign of saturation, even in the highest applied field of 9 T which is quite different from the magnetization of free powder of YMn_6Sn_6 for saturation (11 μ_B/f.u.) reached around 11 T [5,6]. This, together with the occurrence of the larger temperature range of antiferromagnetic order for $LuMn_6Sn_6$, suggests larger antiferromagnetic interactions in this compound than in $ScMn_6Sn_6$ and YMn_6Sn_6.

Since the interatomic distances in $LuMn_6Sn_6$ are intermediate between those in the Sc and Y compounds, the results described above suggest that there is no simple relation between the interatomic spacings and the Mn–Mn interlayer interactions. Therefore, it is interesting to further study the relative magnetic properties of the Mn sublattice in the three compounds by high-field magnetization measurements on single crystals.

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2. Experimental

The thermal variation of the magnetization of the compounds has been measured in a MANICS magneto-susceptometer in a field of 0.1 T at temperatures between 5 and 450 K. The transition temperatures (T_C and T_{sr}) have been taken at the maximum of the first derivate dM/dT. The high-field magnetization has been measured at KYOKUGEN, Osaka University in pulsed fields up to 50 T with pulse duration of 8 ms. The high-field measurements were performed on the compounds by the standard induction method. A single crystal was mounted on a quartz stage and packed in a thin-walled cylindrical tube made of Teflon, and then sustained in liquid-He bath.

3. Sample preparation and characterization

RMn_6Sn_6 single crystals with R = Sc, Y and Lu have been synthesized in a flux method similar to the method previously reported by Clatterbuck and Gschneidner [4]. A mixture of the RMn_6Sn_6 compound, obtained by induction melting, and a large amount of pure Sn metal, with an overall atomic ratio RMn_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 with Cu Kα radiation with high-purity Si as calibration standard (a = 5.43082 Å).

Analysis of the Guinier patterns show that the structure of the investigated RMn_6Sn_6 compounds is isotypic with the $HfFe_6Ge_6$ structure. The lattice parameters, the unit-cell volumes and the c/a ratios are presented in Table 1.

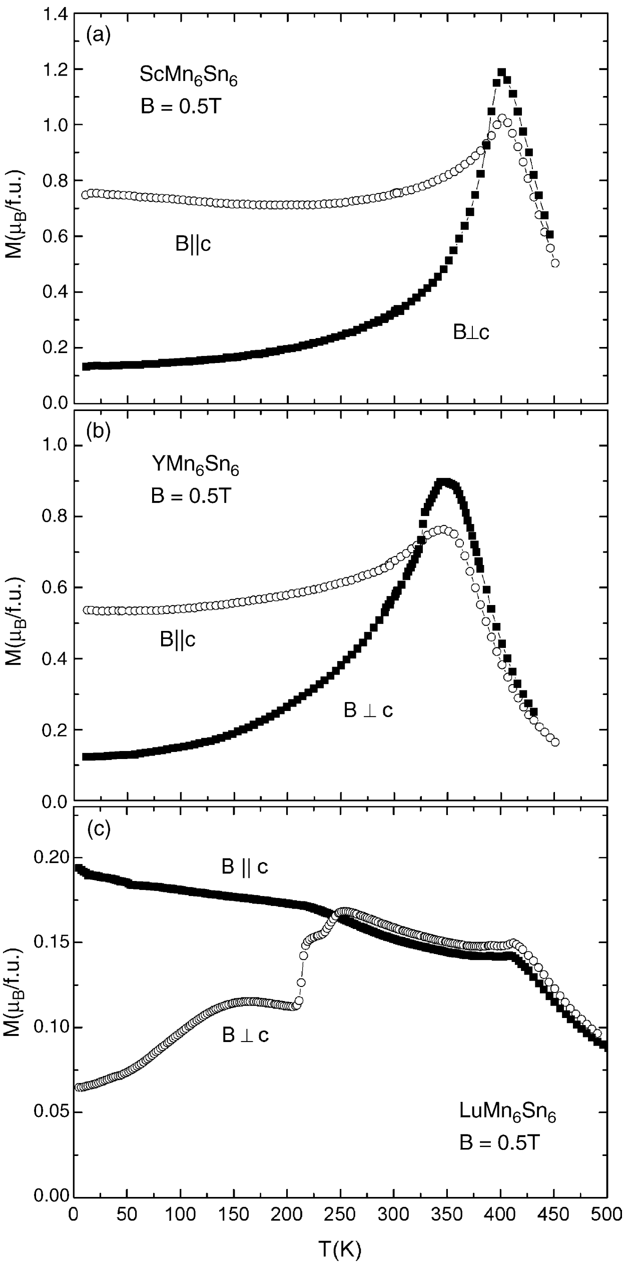

Fig. 1. Thermal variation of the magnetization of RMn_6Sn_6 compounds with R = Sc (a), Y (b) and Lu (c) in a field of 0.5 T, applied parallel and perpendicular to the c-direction. No correction has been applied for the demagnetizing field.

Table 1
Cell parameters of RMn_6Sn_6 single crystals with R = Sc, Y and Lu

	a (Å)	c (Å)	V (Å^3)	c/a
Sc	5.4692(20)	8.9741(45)	232.47(20)	1.6408
Y	5.5332(26)	9.0081(59)	238.84(37)	1.6280
Lu	5.5079(17)	8.9860(43)	236.08(26)	1.6315

4. Results and discussion

The temperature dependence of the magnetization is shown for $ScMn_6Sn_6$, YMn_6Sn_6 and $LuMn_6Sn_6$ in Fig. 1a–c, respectively. These results largely confirm those already obtained previously on powdered samples. All three compounds show a Néel-type of transition above room temperature. The curves for the compound $ScMn_6Sn_6$ do not exhibit any features indicating a transformation below this temperature. In contrast, $LuMn_6Sn_6$ shows two closely spaced transitions at about 220 and 250 K. This is close to the temperature where neutron diffraction has shown that $LuMn_6Sn_6$ abandons its spiral structure and becomes a collinear antiferromagnet up to the Néel temperature [3]. The curve recorded with the field perpendicular to the c-axis for the compound

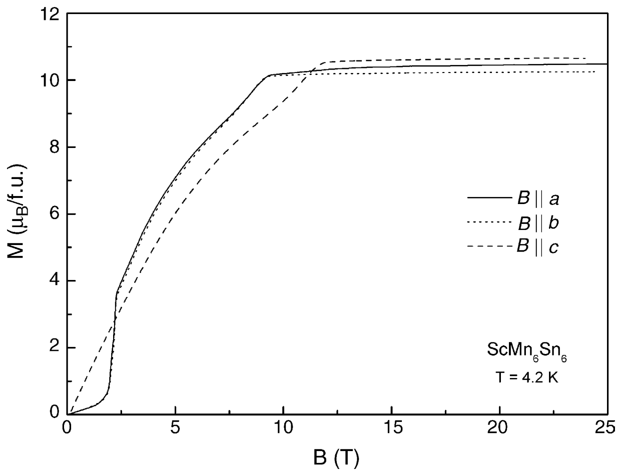

Fig. 2. Magnetization of $ScMn_6Sn_6$, measured at 4.2 K along and perpendicular to the *c*-direction.

YMn_6Sn_6 displays a sudden decrease below 328 K. This transition also appeared in previous powder measurements and seemed to be field dependent [6]. According to the behaviour of the compound $LuMn_6Sn_6$, this transition should be attributed to the same transition from collinear antiferromagnetic to helimagnetic structure.

The magnetic isotherms measured at 4.2 K in the three main crystallographic directions are shown for $ScMn_6Sn_6$, YMn_6Sn_6 and $LuMn_6Sn_6$ in Figs. 2–4, respectively. Bearing in mind that in all three cases the moment directions are perpendicular to the *c*-axis, the relatively strong increase of the magnetization in a field applied parallel to the *c*-direction can be viewed as a bending towards each other of the antiferromagnetic moment components. This bending appears to be a gradual process, leading eventually to a forced ferromagnetic arrangement of the moments at about 12, 14 and 35 T for $ScMn_6Sn_6$, YMn_6Sn_6 and $LuMn_6Sn_6$, respectively. For a simple antiferromagnet with the easy axis perpendicular to the *c*-axis, one would have expected a similar behaviour. However, in this case the isotherm would be linear and its slope would reflect the intersublattice exchange interaction. For the presently materials, the situation is more complicated because different Mn–Mn competing interlayer exchange interactions should be taken into account [2]. Nevertheless, from the fact that the forced ferromagnetic alignment is reached in fields that strongly increase when going from $ScMn_6Sn_6$, to $LuMn_6Sn_6$ we can conclude that the strength of the main antiferromagnetic coupling increases in the same sequence. It is worth noting that these relative strengths do not well fit the evolution of the inter-atomic spacing which increases following the sequence Sc, Lu, Y (Table 1). Hence, it may be concluded that the magnetic interactions are not simply related to the Mn–Mn distances. Moreover, the fact that the critical field is largely higher in the Lu compound, suggests a non-negligible effect of the filled 4f shell by modifying the rare earth transition metal d–d hybridization [7].

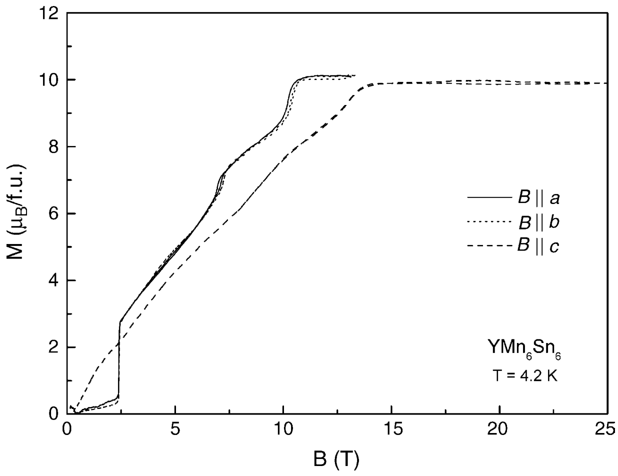

Fig. 3. Magnetization of YMn_6Sn_6, measured at 4.2 K along and perpendicular to the *c*-direction.

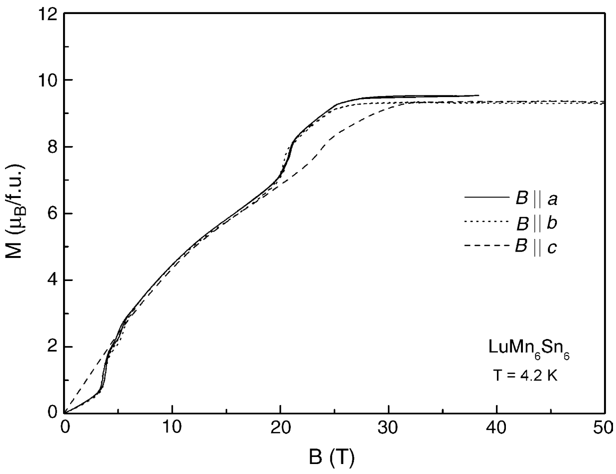

Fig. 4. Magnetization of $LuMn_6Sn_6$, measured at 4.2 K along and perpendicular to the *c*-direction.

Inspecting again the results shown for $ScMn_6Sn_6$ in Fig. 2, one finds that the general shape of the isotherms measured with the field applied perpendicular to the *c*-direction is reminiscent of a standard spin-flop process. Such spin-flop transitions are generally found in materials in which the magnetocrystalline anisotropy is relatively low. The only difference is the slightly non-linear behaviour of the $B \parallel a$ and $B \parallel b$ isotherms above the spin-flop transition and the slightly non-linear behaviour of the $B \parallel c$ isotherm. As already indicated above, these features may find their origin in the presence of more than one antiferromagnetic-coupling constant. Possibly, this is also the reason why the main spin-flop transition in YMn_6Sn_6 (and $LuMn_6Sn_6$) is followed by several other field-induced transitions. In terms of a simple spin-flop transition, the critical field associated with this transition is a measure of the anisotropy ($H_c \propto \sqrt{K}$). By comparing the results in Figs. 2–4, we find that B_c equals 1.7, 2.4 and 3.5 T for $ScMn_6Sn_6$, YMn_6Sn_6 and $LuMn_6Sn_6$, respectively. These results show that the anisotropy substantially increases when going from $ScMn_6Sn_6$ to $LuMn_6Sn_6$.

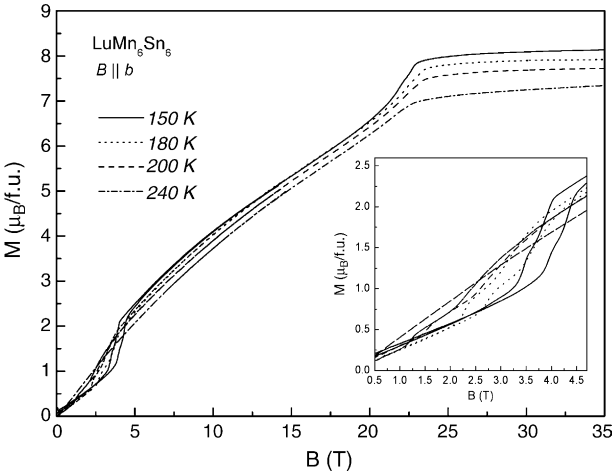

Fig. 5. Magnetization of $LuMn_6Sn_6$, measured at 150, 180, 200 and 240 K perpendicular to the c-direction.

For $LuMn_6Sn_6$, we have also measured the magnetic isotherms for $B \,||\, b$ also at higher temperatures, between 150 and 240 K, in the temperature region of the spin reorientation from the low-temperature flat-spiral structure of ferromagnetic sheets to the collinear antiferromagnetic structure. In Fig. 5, it can be seen that, at 150 K, the value of B_c is still about the same as at 4.2 K and that B_c decreases with further increasing temperature. At 240 K, above the spin-reorientation temperature, the spin-flop behaviour has vanished. The strong increase of the magnetization just before reaching the forced-ferromagnetic state has disappeared at 240 K and therefore seems to be particularly associated with the magnetization process of the spiral structure.

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