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Magnetic properties of $LnMn_xT_{12-x}$ alloys in high magnetic fields

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Abstract

Earlier, preliminary magnetic data of $LnMn_xT_{12-x}$ alloys with Ln = Ce–Nd, Sm and Y, and T = Fe, Co, Ni have been determined in magnetic fields up to 140 kOe in the temperature range 1.7–300 K. In the present paper, the measurements for selected compounds from the family mentioned above have been extended to magnetic fields up to 350 kOe at 4.2 K. The measurements up to 140 kOe were performed in permanent field whereas, above this field in pulsed field with a pulse duration of 10 ms. The measurements were carried out on bulk and loose-powder samples. All investigated materials exhibit ferro(ferri) magnetic properties but without clear saturation and with tiny hysteresis. The results are discussed in terms of the magnetic coupling between three magnetic sublattice systems and the Mn–Mn separation.

Keywords: Magnetic properties; Rare-earth intermetallics

1. Introduction

The $ThMn_{12}$-type f-electron ternaries containing rare earths and 3d metals are subject of intensive research because of their potential applications as permanent magnets (for a review, see Ref. [1]). They are mostly ternaries obtained with the p-electron elements (Si, Al), early 3d (Ti, V, Cr, Mn), 4d (Zr, Nb, Mo) or 5d (Hf, Ta, W, Re) metals as stabilizing components. However, the compounds with Mn are not very popular and they are reported in combination with Fe as a transition element and in principle for the heavier rare earths and Y [2–6]. Up to now, only one piece of information has appeared concerning $LnMn_4Co_8$ compounds in which, besides heavier Ln elements, Pr and Y were used [7].

All these materials crystallize in the tetragonal $ThMn_{12}$-type of structure (I4/mmm-space group) in which the 2(a) site is occupied by rare-earth atoms and the 3d atoms are located at the 8(f), 8(i) and 8(j) positions.

Single-phase YMn_xCo_{12-x} compounds are reported for $0 \leqslant x \leqslant 6.7$, and for $x = 4$ no long-range magnetic order was found above 77 K [2].

$LnMn_xCo_{12-x}$ compounds with Ln = Gd, Tb, Dy and Ho exist in single-phase form for $x \geqslant 4$. It

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has been shown that the ordering temperature is almost independent of Ln and has a maximum at about $x = 7$ [3] or 6 [4]. In Er compounds [6], ferromagnetic ordering of the Er moments is observed at low temperature and the maximum T_C value is found for $x = 4$ and the maximum T_N, corresponding to magnetic ordering of the 3d sublattices, for $x = 6$ [4]. Abad et al. [5] claim, in contrast to Ref. [4], that in $LnMn_xFe_{12-x}$ compounds with Ln = Tb or Y, there are positive interactions in the Fe and negative ones in the Mn sublattice. In $HoMn_{4.8}Fe_{7.2}$, that is ferrimagnetic below 204 K [6], two magnetic sublattices have opposite moment directions, but the authors do not specify the composition of the two sublattices. Wang et al. [7] report that, among the $LnMn_4Co_8$ (Ln = Y, Pr, Gd, Tb, Dy) compounds, the compounds with Y, Pr and Gd are ferromagnetic, and suggest that Pr does contribute to the magnetism, whereas the compounds with Tb and Dy are ferrimagnetic with magnetic moments on all three (Ln, Co and Mn) sublattices.

In the present paper, we report the extension of the preliminary results published previously [8,9] to magnetization measurements performed in pulsed magnetic fields up to 350 kOe on powder and bulk samples of selected materials for possible observation of the contributions of the individual sublattices and the magnetic anisotropy.

2. Experimental

The $LnMn_xT_{12-x}$ alloys have been prepared as it was described previously [8]. The magnetic properties have been investigated in principle in the temperature range 1.7–300 K using a SQUID-magnetometer technique in a magnetic field of 5 kOe. The samples with magnetic-ordering temperature above the room temperature have been investigated at higher temperature. The magnetization has been measured on bulk polycrystalline samples in steady fields, at 1.7 K up to 50 kOe and at 4.2 K up to 140 kOe, and on bulk and loose powder samples in pulsed fields up to 350 kOe with a pulse duration of 10 ms.

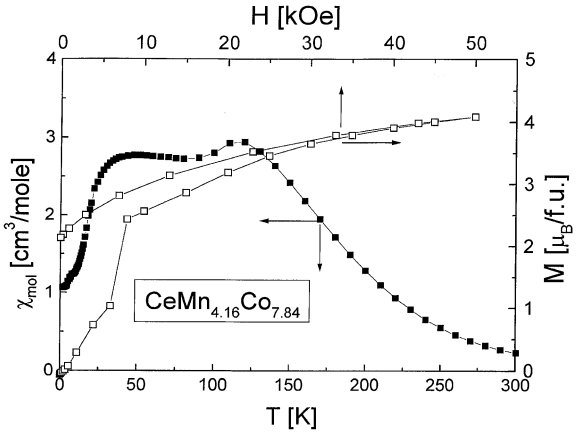

Fig. 1. Magnetic susceptibility of $CeMn_{4.16}Co_{7.84}$ measured at 5 kOe versus temperature (left-hand side and bottom) and magnetization versus magnetic field at 1.7 K (right-hand side and top).

3. Results and discussion

All the investigated samples are in principle single phase. They form a variety of stoichiometric types, as it was also observed previously [2–5]. The absence of La compounds, reported previously [3,6], seems to indicate that the La atom is too large to be accommodated in the $ThMn_{12}$-type of crystal lattice. The results are presented in Figs. 1–6. The susceptibility, χ_{mol}, of $CeMn_{4.16}Co_{7.84}$ measured in 50 kOe shows a strong increase up to about 25 K, reaches a first maximum around 40 K and a second maximum at about 110 K, followed by smooth decrease (Fig. 1). M versus H exhibits ferromagnetic character with small hysteresis and a "saturation" magnetic moment $p_s = 4.08\mu_B$/f.u. (Fig. 1). The temperature dependence of the susceptibility of $PrMn_4Co_8$ [7] is very similar, but the susceptibility of $CeMn_{4.96}Fe_{7.04}$ exhibits $T_C \sim 230$ K and the saturation moment equals $16.81\mu_B$/f.u. [9]. Magnetization measurements on $CeMn_{4.16}Co_{7.84}$ carried out at 4.2 K up to 350 kOe in pulsed field do not show saturation, and, in 350 kOe, the magnetization of the loose-powder sample is 41% higher then of bulk material, suggesting considerable anisotropy of the alloy.

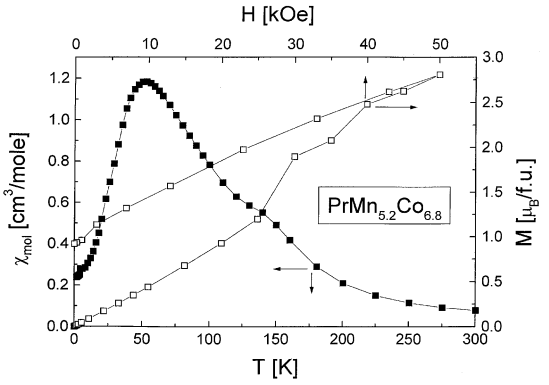

Fig. 2. Magnetic susceptibility of $PrMn_{5.2}Co_{6.8}$ measured at 50 kOe versus temperature (left-hand side and bottom) and magnetization versus magnetic field at 1.7 K (right-hand side and top).

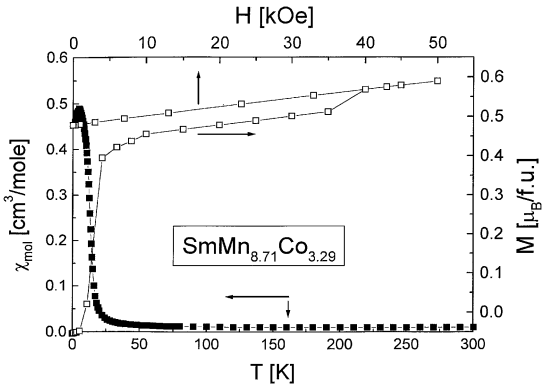

Fig. 4. Magnetic susceptibility of $SmMn_{8.71}Co_{3.29}$measured at 50 kOe versus temperature (left-hand side and bottom) and magnetization versus magnetic field at 1.7 K (right-hand side and top).

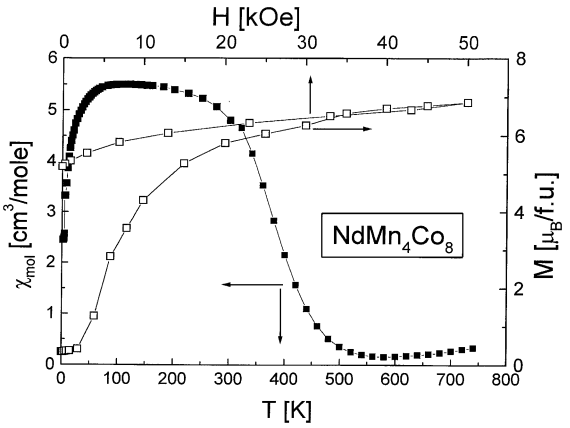

Fig. 3. Magnetic susceptibility of $NdMn_4Co_8$ measured at 50 kOe versus temperature (left-hand side and bottom) and magnetization versus magnetic field at 1.7 K (right-hand side and top).

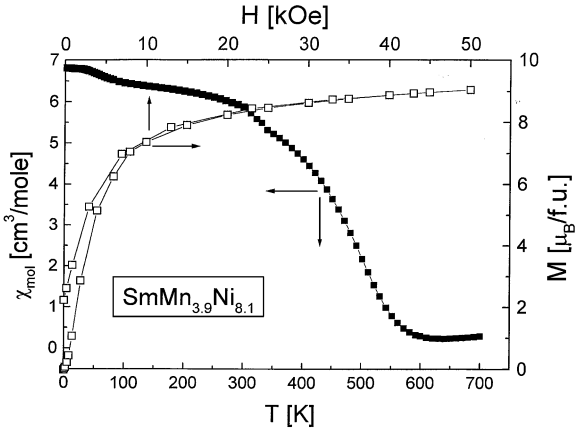

Fig. 5. Magnetic susceptibility of $SmMn_{3.9}Ni_{8.1}$ measured at 50 kOe versus temperature (left-hand side and bottom) and magnetization versus magnetic field at 1.7 K (right-hand side and top).

The susceptibility of $PrMn_{5.2}Co_{6.8}$ shows a maximum at 53 K (Fig. 2) and the magnetization shows hysteresis below about 40 kOe. The magnetization of a bulk sample increases almost linearly above 100 kOe with p_s (140 kOe) = $4\mu_B$/f.u. [9], and p_s (350 kOe) = $6\mu_B$/f.u. (Fig. 6). Unfortunately, the magnetization of the powder sample could not be measured in pulsed fields above 100 kOe, but at this field value the magnetization of powder sample is about two times higher than the bulk magnetization (Fig. 6).

The susceptibility of $NdMn_4Co_8$ is very similar to that of $CeMn_{4.16}Co_{7.84}$ and $PrMn_4Co_8$ [7], but the Nd compound has a higher Curie temperature of about 400 K (Fig. 3). The decrease of χ_{mol} at low temperature can result from domain effects. At 4.2 K, there is tiny hysteresis below 80 kOe [9] and the saturation moment reaches about $3.5\mu_B$/f.u. in 140 kOe. There is practically no difference between the magnetization of the bulk and the powder (Fig. 6) which points to small magnetic anisotropy.

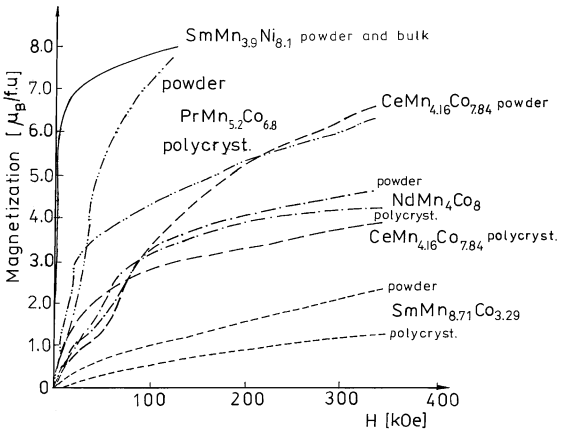

Fig. 6. Magnetization of $LnMn_xT_{12-x}$ alloys, measured in bulk and loose-powder form, versus pulsed magnetic field up to 350 kOe at 4.2 K.

$SmMn_{8.71}Co_{3.29}$ is apparently ferromagnetic below 18 K (Fig. 4) with a low saturation moment $p_s = 0.58\mu_B$/f.u. at 1.7 K and in 50 kOe with hysteresis below 40 kOe and with relatively large remanence. As can be seen in Fig. 6, the magnetization difference between the bulk and the loose-powder sample is 47% in 350 kOe, but the value of magnetization of powder sample is also rather low. All these results suggest that magnetic structure is not simply ferromagnetic. It is also possible that the ferromagnetic behavior is due to an impurity phase. This idea is supported by the observation that compounds with high Mn concentration are usually paramagnetic (see e.g. Ref. [9]).

$SmMn_{3.9}Ni_{8.1}$ is ferromagnetic below about 550–600 K with a saturation moment $p_s = 9.1\mu_B$/f.u. at 1.7 K and in 50 kOe (Fig. 5). The bulk and the loose-powder sample of this compound could only be investigated in steady fields. Up to 140 kOe, there is a high magnetization, almost the same for bulk and powder sample (Fig. 6) and no hysteresis (Fig. 5).

It is clear that an interpretation of these complex magnetic properties of $LnMn_xT_{12-x}$ alloys is very difficult. The most important experiment that should be done is the determination of the distribution of the transition elements over the individual crystallographic positions. A neutron diffraction (ND) investigation should elucidate this problem as well as provide information on magnetic structure. Available ND data (see e.g. Ref. [5]) suggest the following competing interaction in $TbMn_xFe_{12-x}$: positive for Fe (F) and negative for Mn (AF). The Tb sublattice is ordered ferromagnetically. The following sequence of the strengths of the exchange interactions J_{ij}, has been proposed [5]:

$$|J_{TT}| > |J_{LnT}| > |J_{LnLn}| > |J_{LnMn}| \sim |J_{MnMn}|.$$

This sequence may only be valid for the $TbMn_xFe_{12-x}$ system but it can also be a convenient starting point for the discussion of the results reported above. The magnetization dependence on magnetic field for f–d materials has got an explanation in terms of two-sublattice models (see e.g. Refs. [10,11], but in $LnMn_xT_{12-x}$ compounds a description in terms of three (or more) magnetic sublattices may be necessary. This depends on the question whether the Mn sublattice(s) are magnetically ordered. It seems that the existence and a type of magnetic ordering depends critically on the Mn–Mn separation. Unfortunately, this question cannot be answered properly without ND studies. The apparent difference between the results obtained on loose-powder and bulk samples suggests strong anisotropy in all materials except $NdMn_4Co_8$ and $SmMn_{3.9}Ni_8$. At present, the contribution of Mn to the anisotropy is still obscure.

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