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Magnetotransport properties of $Gd_2(Mo_{1-x}V_x)_2O_7$ with pyrochlore structure

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Abstract. Magnetization and magnetoresistance (MR) measurements were performed on pyrochlores $Gd_2Mo_2O_7$ and $Gd_2(Mo_{0.6}V_{0.4})_2O_7$. It was shown that both compounds are ferromagnets below $T_C = 80$ K with the same alignment of magnetic moments of 4f and 4d ions. The substitution of Mo ions by V leads to the increase of the resistivity without changes in the magnetic state. $Gd_2Mo_2O_7$ exhibits a large negative magnetoresistance ratio, especially in the low-magnetic-field regime, which increases strongly with decreasing temperature. In $Gd_2(Mo_{0.6}V_{0.4})_2O_7$ below T_C the magnetoresistance ratio changes from negative to positive with decreasing temperature. These data indicate that the mechanism of the magnetoresistance effect in the molybdates differs markedly from that for manganites studied in detail with perovskite and pyrochlore structures.

1. Introduction

The discovery of the 'colossal' magnetoresistance (MR) in the manganites with perovskite [1] and pyrochlore [2] structures has stimulated the search for new compounds exhibiting the large magnetoresistance. In this paper we are reporting the data obtained for $Gd_2(Mo_{1-x}V_x)_2O_7$ with the pyrochlore structure. It was established that $R_2Mo_2O_7$ (R = Nd, Sm, Gd) compounds exhibit metallic behaviour of resistivity in a wide range of temperatures [3, 4]. The results of magnetization studies of $Gd_2Mo_2O_7$ in strong magnetic fields are in agreement with ferromagnetic alignment of both Gd and Mo sublattices [4]. The data on the Curie temperature of $Gd_2Mo_2O_7$ are contradictory [3, 4]. The vanadates $R_2V_2O_7$ (R = Lu, Tm, Y, Er, Ho, Dy) with the pyrochlore structure are ferromagnetic semiconductors with T_C around 70–80 K [5, 6]. The magnetotransport properties of these classes of compounds have not been studied yet. In this paper we report briefly the MR behaviour of the $Gd_2(Mo_{1-x}V_x)_2O_7$ system. We find that the MR has interesting dependences on the composition, magnetic field and temperature, and for $Gd_2Mo_2O_7$ the MR can be comparable with that observed earlier for manganites.

2. Experiment

The samples $Gd_2Mo_2O_7$ and $Gd_2(Mo_{1.2}V_{0.8})O_7$ were prepared from starting materials Gd_2O_3, MoO_2 and VO_2 at pressure $P = 4$ GPa and temperature 1800 K. Powder x-ray diffraction patterns showed the single-phase nature of the samples. The lattice parameters

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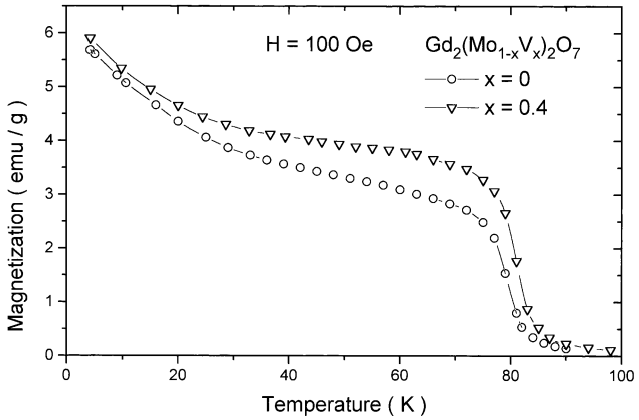

Figure 1. Magnetization–temperature curves for $Gd_2(Mo_{1-x}V_x)_2O_7$ ($x = 0$, 0.4) measured at $H = 100$ Oe.

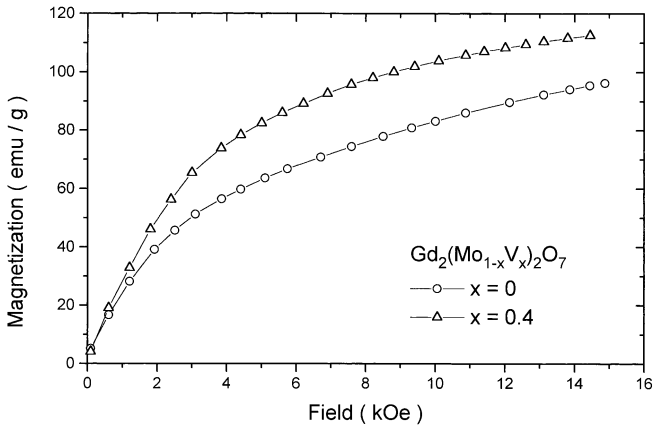

Figure 2. Magnetization as a function of magnetic field for $Gd_2(Mo_{1-x}V_x)_2O_7$ ($x = 0$, 0.4) at 4.2 K.

of $Gd_2(Mo_{1-x}V_x)_2O_7$ obtained by the analysis of the x-ray data based on the space group $Fd\bar{3}m$ are 1.0332 nm ($x = 0$) and 1.0293 nm ($x = 0.4$). The magnetization measurements were made with a vibrating sample magnetometer. The MR measurements were performed by a standard four-probe method. A superconductivity coil was used to produce steady magnetic fields up to 120 kOe and the magnetic field was applied perpendicular to the current during the MR measurements.

3. Results and discussion

Figure 1 shows the thermal dependence of the magnetization on heating after field cooling down to 4.2 K for the $x = 0$ and $x = 0.4$ samples. For both samples a rapid decrease of magnetization at around $T_C = 80$ K is observed. The low-temperature magnetization behaviour is modified apparently by the Gd-sublattice contribution. However, there is no magnetic phase transition at lower temperatures, because the magnetization increases

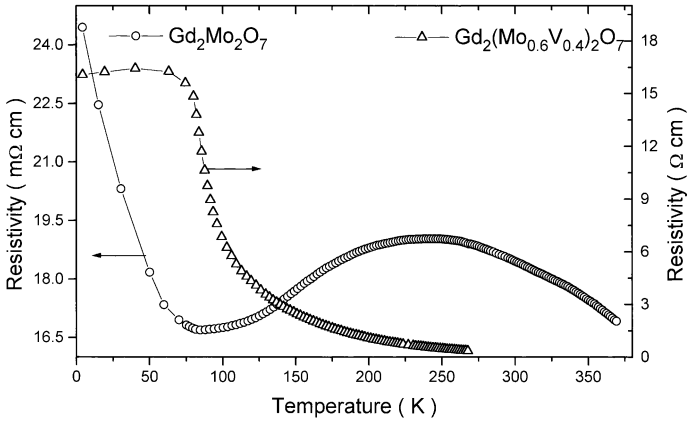

Figure 3. The thermal behaviour of resistivity for $Gd_2Mo_2O_7$ and $Gd_2(Mo_{0.6}V_{0.4})_2O_7$.

gradually with decreasing temperature due to positive f–d superexchange. The magnetization as a function of magnetic field is plotted in figure 2. The magnetic field of 15 kOe is too low to saturate the magnetization. The $Gd_2Mo_2O_7$ compound has a magnitude of magnetization 90 emu g^{-1} or approximately 11 μ_B per formula unit at magnetic field of 15 kOe. A large magnetic moment arises due to the Gd-sublattice contribution. Magnetic moments of Gd^{3+} ions are aligned parallel to magnetic moments of the 4d sublattice. The substitution of Mo ions by V leads to an increase of magnetization (figure 2). Apparently f–d superexchange is larger for the V-containing compound. Figure 3 shows the electrical resistivity behaviour as a function of temperature. The resistivity of $Gd_2Mo_2O_7$ decreases with decreasing temperature below 250 K; however at low temperatures it starts to increase on cooling in spite of the magnetic ordering. The $Gd_2(Mo_{0.6}V_{0.4})_2O_3$ is semiconducting in the whole investigated temperature range. Below T_C the resistivity weakly depends on temperature. The measurements of the Seebeck coefficient indicate an n type of charge carriers for both compounds. The results of the MR (defined as $MR = [R(H) - R(0)] \times 100\%/R(0)$) measurements are presented in figure 4. As one can clearly see from figure 4 the MR of $Gd_2Mo_2O_7$ increases gradually with decreasing temperature. The data obtained at 4.2 K resemble those at 15 K. The effect is the most pronounced in the low-field regime. The MR at 4.2 K in a field of 10 kOe reaches a magnitude of 20%. This value is comparable with the MR in the manganites $La_{1-x}Pb_xMnO_3$ in the vicinity of T_C. In contrast to manganites the MR of $Gd_2Mo_2O_7$ weakly changes with field, increasing from 20 to 120 kOe.

Quite different results have been obtained for the V-substituted sample. The MR of this sample is much less than that for $Gd_2Mo_2O_7$. At around T_C the MR is negative up to 120 kOe. However below T_C the increase of the magnetic field leads to the change of the sign of MR which becomes positive. A weak negative MR has been again observed in low magnetic fields at 4.2 K (figure 4(b)). In fields higher than 10 kOe the positive MR linearly depends on the magnetic field up to 120 kOe (figure 4(b)).

The magnetic properties of $Gd_2Mo_2O_7$ samples studied in this work differ from those observed for $Gd_2Mo_2O_7$ nominal composition in the work [4]. According to [4] the $Gd_2Mo_2O_7$ is a ferromagnet with $T_C = 56$ K in which both Gd and Mo sublattices order ferromagnetically at the same temperature T_C. The $Gd_2Mo_2O_7$ received under high-pressure conditions exhibits the ferromagnetic order below 80 K, and the Gd-sublattice starts to order at around 30 K (figure 1). The chemical composition of compounds with the pyrochlore

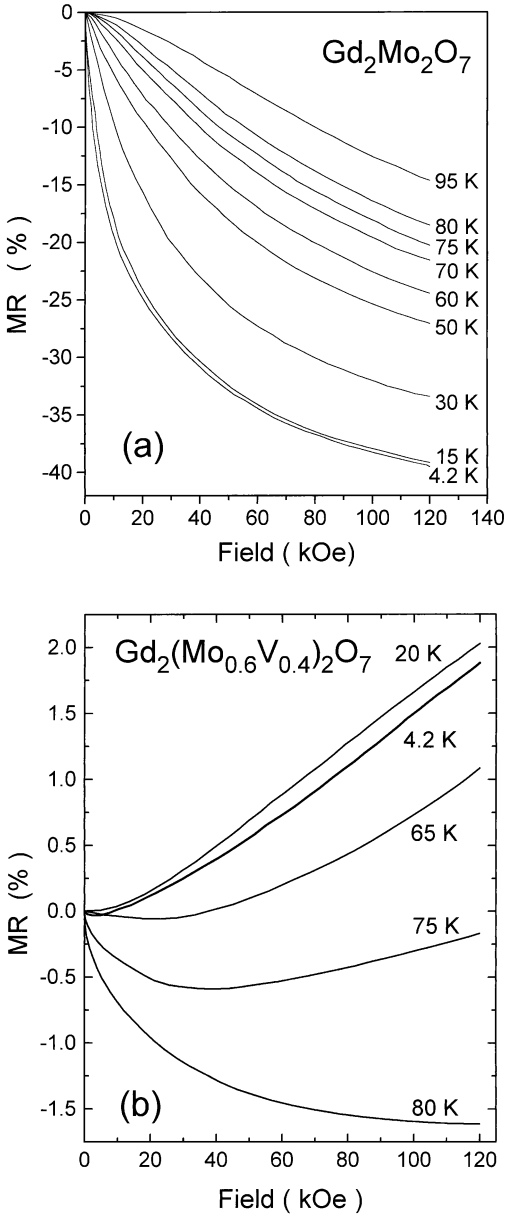

Figure 4. The magnetoresistance ratio as a function of field for $Gd_2Mo_2O_7$ (a) and $Gd_2(Mo_{0.6}V_{0.4})_2O_7$ (b) at different temperatures.

structure probably depends strongly on the conditions of preparation. Very complex cationic distribution has been revealed by an NMR method for $In_2Mn_2O_7$ pyrochlore [7]. It is worth noting that positive f–d superexchange is a common feature of the $R_2B_2O_7$ (R = rare earth ion, B = Mo, Mn, V) compounds with pyrochlore structure [3–7]. All these compounds exhibit ferromagnetic or spin glass behaviour. The topological frustration of the pyrochlore lattice favours the ferromagnetic type of ordering [2–7]. There is no doubt

that superexchange via oxygen is the dominant magnetic interaction [2–7]. In contrast to $Tl_2Mn_2O_2$ and $La_{1-x}A_xMnO_3$ manganites the $Gd_2(Mo_{1-x}V_x)_2O_7$ system does not exhibit any peak of MR at Curie temperature. The MR value increases gradually with decreasing temperature (figure 4). Such a behaviour was observed for many systems, for example in oxides with spinel structure and inhomogeneous manganites [8, 9], metallic multilayers and granular systems [10, 11]. In inhomogeneous materials the giant MR could be attributed to the spin-dependent scattering of the charge carriers on the interface. The origin of the positive MR in magnetic semiconductors is not understood at present. Weak positive MR was observed for the metallic ferromagnet $La_{0.5}Sr_{0.5}CoO_3$ where the MR effect increases with decreasing temperature [12]. In the vanadium-substituted pyrochlore the positive MR is linked closely with a ferromagnetic ordering, because above $T_C = 80$ K the MR is negative up to 120 kOe (figure 4). The preliminary measurements indicate that the positive MR increases strongly with increasing vanadium content. Unfortunately the $R_2V_2O_7$ pyrochlores exhibit very large resistivity near the temperature of magnetic ordering. So we cannot carry out MR measurements of these compounds. A detailed investigation of the MR and magnetic properties of the $R_2(Mo_{1-x}V_x)O_7$ (R = rare earth ion) system are in progress.

Acknowledgments

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