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LETTER TO THE EDITOR

Unusually large negative magnetoresistance around 200 K in the alloys $Gd_{1-x}La_xMn_2Ge_2$

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Abstract. We report here the observation of large negative magnetoresistance (MR) at 200 K (for instance, about −5.5% for the application of a magnetic field of 70 kOe) in magnetic alloys, $Gd_{1-x}La_xMn_2Ge_2$ ($x = 0.07$ and 0.08). There is a distinct sign reversal of MR, with a comparable magnitude, for $x = 0.07$ at 123 K thereby establishing the presence of temperature dependent magnetic structures. The observation of a large MR at temperatures as high as 200 K is uncommon among the bulk form of alloys.

The direction of magnetoresistance (MR) research in solids has gained importance following the observation of giant magnetoresistance (GMR) in Fe–Cr multilayers [1]. This topic of research became more prominant with the observation of GMR in La based manganates [2] around 200 K due to potential applications, and there have been constant efforts in the literature to look for materials with GMR near room temperature. It is to be noted that all such materials discovered recently are based on oxides and semiconductors [3, 4]. But no alloy in the bulk form [5] has hitherto been known to exhibit GMR at high temperatures (T); however, many rare-earth intermetallics with large low-temperature MR due to magnetic anomalies have been identified [6]. In this regard, the compounds of the type $RMn_2(Si,Ge)_2$, crystallizing in the $ThCr_2Si_2$-type tetragonal structure [7], are of special interest, as this is the only series of alloys of this structure in which the transition metal ions carry a magnetic moment (close to 3 μ_B), ordering magnetically near room temperature, and in addition exhibiting a novel low-temperature MR anomaly [8]. The nature of the magnetic structure, whether it is ferro-, ferri- or antiferromagnetic, sensitively depends on Mn–Mn separation. In some cases, one observes multiple magnetic transitions as a function of temperature, if Mn–Mn separation falls near the 'so-called' critical value of 2.85 Å; we believe [8] that such 'sensitive' systems are very good candidates to look for high-temperature GMR behaviour due to possible changes in the spin-dependent scattering effects. Thus, in $SmMn_2Ge_2$ [8, 9], MR of magnitude of about 4% has been reported in the T interval of 110–150 K, and considering that the classical contribution to MR is expected to be less than, or very close to 1% at such high temperatures, this system is considered to be a GMR system [9]. As a continuation of our efforts in this direction, we have investigated the MR behaviour of several alloys belonging to the pseudoternary series, $Gd_{1-x}La_xMn_2Ge_2$ [10], at the Gd rich end. While readers may see [10] for a detailed magnetization behaviour of these alloys, it is sufficient here to mention that $GdMn_2Ge_2$ and $LaMn_2Ge_2$ order antiferro- and ferromagnetically below about 370 and 327 K respectively,

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while the intermediate compositions at the Gd rich end exhibit multiple transitions as a function of temperature.

The samples, $Gd_{1-x}La_xMn_2Ge_2$ ($x = 0.0, 0.04, 0.06, 0.07, 0.08$ and 0.09), used in the present studies are the same as those employed in earlier investigations [10]. The electrical resistance (R) as a function of magnetic field (H) up to 70 kOe was measured by a conventional four-probe method employing a conducting silver paint for making electrical contacts of the leads with the sample at selected temperatures (4.2, 10, 30, 123 and 200 K) and these temperatures were chosen such that we probe different regions and magnetic phases in the magnetic phase diagram of this class of compounds (see figure 2 of [10]). The excitation current is of the order of 50 mA and a Keithley nanovoltmeter (sensitivity 10 nV) was employed to measure the voltage drop across the leads. Isothermal magnetization (M) measurements were also performed for comparative purposes employing a commercial (Oxford Instruments) vibrating sample magnetometer at relevant temperatures.

Though we tried to collect data at several temperatures, we concentrate here (figure 1) mainly on the data at 200 K for $x = 0.07$ and 0.08 (as well as at 123 K for $x = 0.07$), as the net

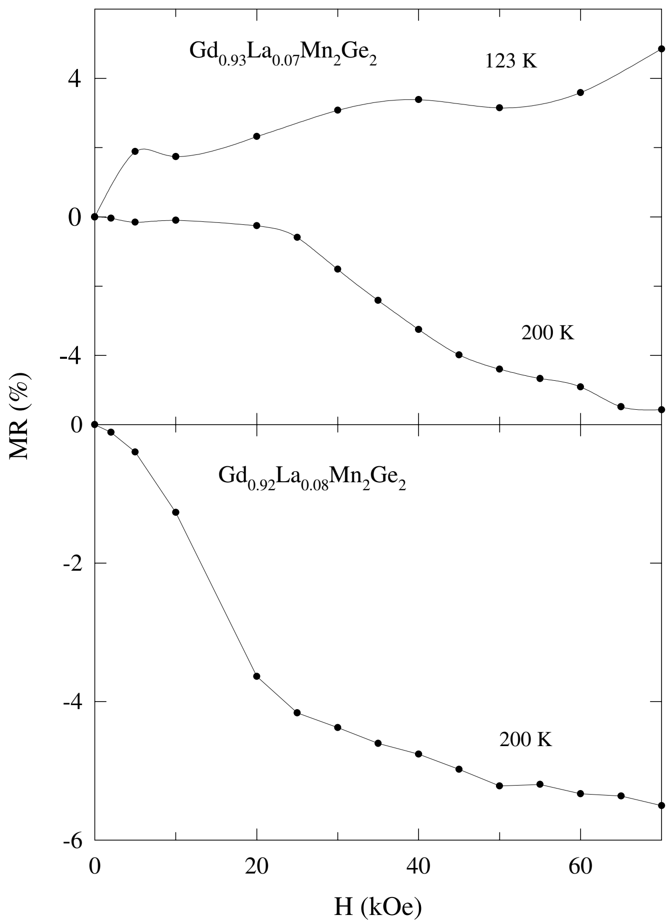

Figure 1. The magnetoresistance, $\Delta R/R = [R(H) - R(0)]/R$ for the alloys, $Gd_{1-x}La_xMn_2Ge_2$ ($x = 0.07$ and 0.08) as a function of applied magnetic field (H) at 200 K. The data for $x = 0.07$ at 123 K are also shown. The lines through the data points are guides to the eye.

R change ($\Delta R = R(H) - R(0)$) induced by the application of H is well outside the detection limit of nanovoltmeter for these cases, particularly at high fields. For instance, the zero field R values at 200 K for the specimens of these two compositions employed are close to 3 mΩ and a few percent variation of R is well outside the sensitivity range of the nanovoltmeter. However, at very low temperatures, the values of R were drastically reduced and hence we could not place much confidence on the values of ΔR (and hence are not presented here). It is to be remarked that ΔR is negligibly small for other compositions even at 200 K. The main point of emphasis therefore is that the R of the above two alloys at 200 K exhibit noticeably large response for the application of a H. Now turning to the composition $x = 0.07$, the MR, defined as MR $= [R(H) - R(0)]/R(0)$, is negligible for $H < 25$ kOe and the magnitude, with a negative sign, increases for higher fields, attaining a value of about -5% at $H = 70$ kOe. There is also a metamagnetic-like transition in the field range 20–60 kOe in the isothermal M data (figure 2), qualitatively mimicking the plot of MR versus H. Thus, it appears that the field-induced changes (towards ferromagnetism) in the magnetic structure and associated spin-dependent scattering effects are responsible for the observed effects. We also observed

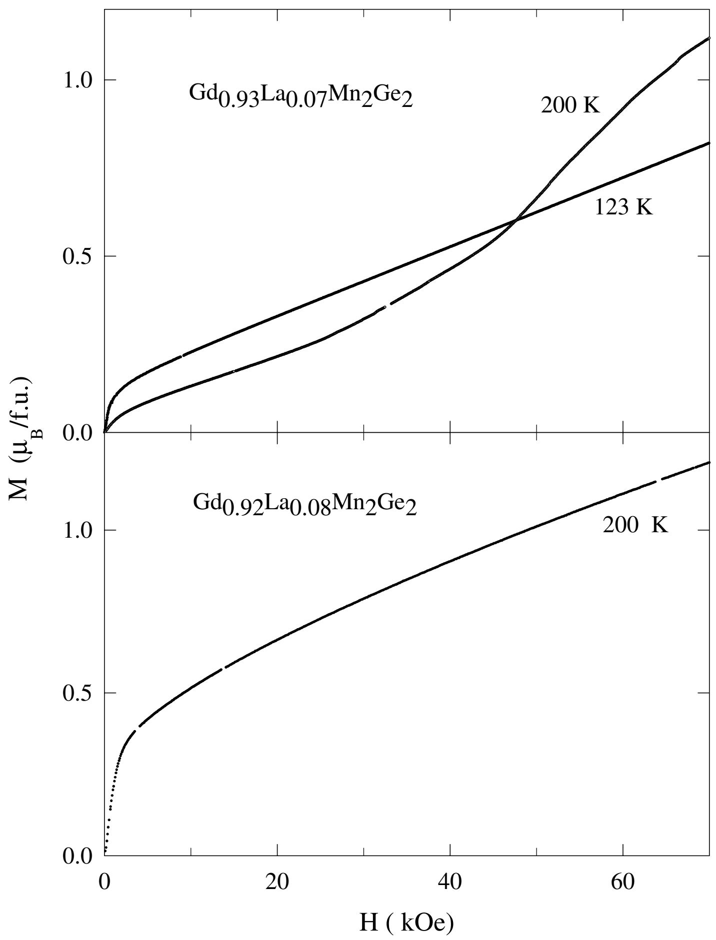

Figure 2. The isothermal magnetization for the alloys, $Gd_{1-x}La_xMn_2Ge_2$ ($x = 0.07$ and 0.08) at relevant temperatures.

large MR at 123 K as well at higher fields with a somewhat similar magnitude, however, with a positive sign. According to [10], for this composition, at 123 K, the magnetic state at zero field is located at just near the critical temperature for ferrimagnetic to antiferromagnetic transition. Therefore, an applied magnetic field apparently causes a drastic spin rearrangement at 123 K, somehow tending towards antiferromagnetism as indicated by the positive sign of MR (a characteristic feature in antiferromagnets without Brillouin-zone boundary gaps) and linear dependence of isothermal M on H without a tendency for saturation at high fields (figure 2); it is worthwhile to focus future studies on understanding the magnetic structure modified by H in view of the fact that the proposed explanation in terms of field-induced antiferromagnetism is surprising and unexpected.

Now, turning to $x = 0.08$, while the MR is negligibly small at 123 K (and hence not shown here), the magnitude starts increasing as H is applied if the measurement is performed at 200 K, thus clearly indicating that there is a change in the nature of the magnetism with increasing temperature, consistent with the conclusions of [10]; at 200 K, the value of MR reaches a large value comparable to that in $x = 0.07$. Though the shape of MR versus H plot for $x = 0.08$ at 200 K is nearly the same as that for $x = 0.07$, the field around which one sees a metamagnetic-like change is shifted to relatively lower values (5–20 kOe) for the former alloy. In sharp contrast, the isothermal M at 200 K does not show any such metamagnetic tendency for this composition in this field range; however, there is a sharp variation in M at much lower fields, but a corresponding sharp variation in MR is not observed. Thus, in this sense, in these two compositions, R responds differently with respect to the application of H, while comparing with M data. One therefore wonders why spin-dependent scattering mechanism is not transparent for the $x = 0.08$ alloy.

To conclude, we have identified some pseudoternary alloys in which the magnetoresistance is unusually large at rather high temperatures (around 200 K) [11]. In these alloys, this observation is a characteristic feature of the antiferromagnetic phase which extends to temperatures as high as 250 K. This implies that there is a strong possibility [8] to observe GMR at room temperature as well in such magnetic alloys, thereby providing scope for device applications, particularly considering that these alloys are easy to synthesize and stable in air and also the features noted are for the bulk form of materials.

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[11] After submission of our article for publication, we became aware of the following report in which the authors observed MR of about −15% in a field of 60 kOe at 145 K in $Gd_{0.925}La_{0.075}Mn_2Ge_2$
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