Large positive magnetoresistance at low temperatures in a ferromagnetic natural multilayer, $LaMn_2Ge_2$

R. Mallik, E. V. Sampathkumaran,[a] and P. L. Paulose
Tata Institute of Fundamental Research, Homi Bhabha Road, Mumbai-400005, India

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The results of magnetoresistance measurements on a naturally occurring multilayer $LaMn_2Ge_2$, which is ferromagnetic below 326 K, are reported. The magnitude of magnetoresistance is found to be positive below 70 K gradually increasing to an unusually large value (nearly 100%) at 4.2 K in the presence of a field of 70 kOe as the temperature is lowered, similar to the recent observations by Verbanck, Temst, Mae, Schad, Van Bael, Moshchalkov, and Bruynseraede [Appl. Phys. Lett. **70**, 1477 (1997)] in Cr/Ag/Cr trilayers. The positive sign of magnetoresistance for a ferromagnet is unexpected and possible explanations are offered. *© 1997 American Institute of Physics.* [S0003-6951(97)02142-6]

It is well-known that the magnetic multilayers, for instance Fe–Cr,[1] exhibit a large magnetoresistance of the order −100% at 4.2 K in the presence of a magnetic field (H) of 60 kOe and the investigation of such multilayers is an active topic research. With the observation of giant magnetoresistance in La based manganates,[2] the investigations of magnetoresistance behavior of materials in general have been receiving special attention. Recently, we have intensified our efforts in this direction on a number of rare-earth (R) containing intermetallics (besides on Ce alloys) and several valuable results were obtained.[3] In particular, the compounds of the type $RMn_2(Si,Ge)_2$, crystallizing in the $ThCr_2Si_2$-type tetragonal structure,[4] 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, with several magnetic anomalies. Therefore, the layered nature of this structure (stacked in the sequence R–Si–Mn–Si–R) and its derivatives provides a novel route to model artificial multilayer physics. A number of magnetic and magnetoresistance anomalies have been reported recently in $SmMn_2Ge_2$ (Ref. 5). As a continuation of our efforts in probing this class of natural multilayered compounds, we report here the results of magnetoresistance measurements in $LaMn_2Ge_2$. We notice a large positive magnetoresistance similar to that observed recently in Cr/Ag/Cr trilayers.[6] What is puzzling is that the sign is positive though this compound is believed to be a bulk ferromagnet.

The magnetism of this compound has attracted considerable attention in the recent post. It had been believed that, below 326 K, the Mn sheets undergo paramagnetic to ferromagnetic ordering with the easy axis of magnetization along the c axis, with the interplanar interaction also being ferromagnetic (simple collinear ferromagnetism).[4] Recent (doped) ^{57}Fe Mossbauer spectroscopic studies[7] and careful neutron diffraction[8] provided new and unexpected results: in the ferromagnetically ordered state the Mn moments have a conical arrangement (instead of a collinear arrangement) with a ferromagnetic coupling of the ferromagnetic component along the c axis; the in-plane component however seems to persist well above T_C giving rise to antiferromagnetic structure till about 420 K above which the sample becomes paramagnetic. Thus, the transition at 326 K is currently believed to originate from antiferro to ferromagnetic transition (with decreasing temperature).

The polycrystalline sample $LaMn_2Ge_2$ was prepared by melting together constituent elements in an arc furnace. Slight excess of Mn was added to compensate for weight loss while melting. The ingot was homogenized in an evacuated sealed quartz tube at 800 °C for one week. X-ray diffraction pattern (Cu K_α) confirms single phase nature of the alloy. The magnetoresistance, $\Delta\rho/\rho[=\{\rho(H)-\rho(0)\}/\rho(0)]$, data were obtained as a function of temperature in the range 4.2–300 K in the presence of an external magnetic field (H) of 50 kOe and also as a function of H at selected temperatures with the direction of constant current (50 mA) being the same as that of H. The area of cross section of the specimen used is of the order of 7 square mm with the separation between the voltage leads being about 2 mm. A conducting silver epoxy was used to make the electrical contacts of the leads with the sample. Considering that there are microcracks in the sample, which is intrinsic to this class of alloys, the error in the absolute values of resistivity (ρ) is estimated to be of the order of 20% by repeated measurements.

The electrical resistivity as a function of temperature (4.2–60 K) in zero field as well as in the presence of a field of 50 kOe are shown in Fig. 1(a). Since the values of $\Delta\rho/\rho$ above 60 K are negligible, we do not show the data above this temperature. The absolute value of resistivity (ρ) is of the order of 300 $\mu\Omega$ cm at 70 K and it varies by a factor of about 10 as the temperature is lowered to 4.2 K in zero field. The application of a field of 50 kOe enhances the value of ρ noticeably (the corresponding change in the voltage across the leads is well above the resolution of the nanovoltmeter used) and the magnitude of $\Delta\rho/\rho$ increases gradually with decreasing temperature reaching as high a value as 80% at 4.2 K [Fig. 1(b)]. The values of $\Delta\rho/\rho$ were also obtained as a function of H at 4.2, 12, 30, and 60 K (Fig. 2). The values at 60 K, though small, are positive. For other temperatures, $\Delta\rho/\rho$ increases nearly linearly with increasing field, attaining a value of about 80% at 12 K and 100% at 4.2 K in a field of

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[a] Electronic mail: SAMPATH@tifrvax.tifr.res.in

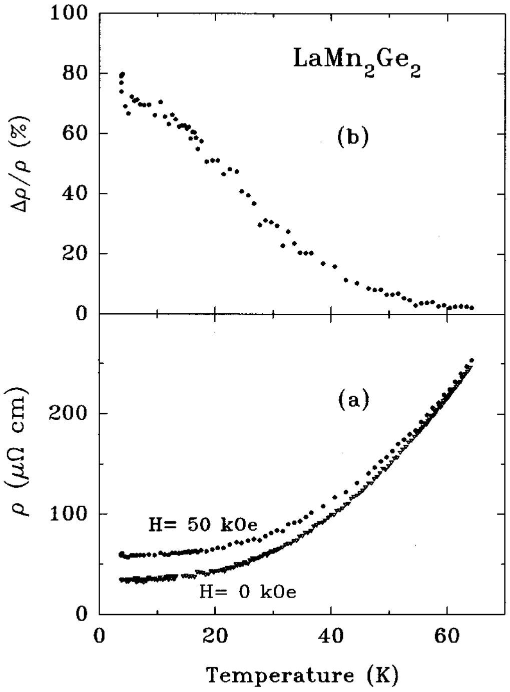

FIG. 1. (a) The electrical resistivity (ρ) of LaMn$_2$Ge$_2$ as a function of temperature (4.2–60 K) in the presence and in the absence of a magnetic field of 50 kOe. (b) The magnetoresistance ($\Delta\rho/\rho$) obtained from the data shown in (a) is plotted.

70 kOe; it is a well-known fact that in nonmagnetic metals and alloys, $\Delta\rho/\rho$ varies quadratically with H and therefore the linear field dependence in the alloy under study must in some way be related to magnetism. The monotonic increase with H implies that the magnitude will still be larger for further high fields. The above data brings out two puzzling findings: (i) the magnitude of $\Delta\rho/\rho$ at high fields as temperature tends to zero is very large, typical of artificial multilayer systems; it may be recalled that in nonmagnetic metals, the corresponding values are known to be less than few percent. (ii) The sign of $\Delta\rho/\rho$ is positive whereas for a ferromagnetically ordered bulk polycrystalline material one usually expects negative sign. The origin of positive sign is not clear at present. Similar anomalies have been noted in a few other ferromagnetic materials recently by us, viz., GdNi (Ref. 3) and SmMn$_2$Ge$_2$ (Ref. 5). We offer below various explanations. As mentioned in the introduction, recent neutron diffraction results suggest that the magnetic moment alignment is not strictly collinear, resulting in an antiferromagnetic component of the Mn moments in the basal plane. Generally, in polycrystalline metallic antiferromagnets, the $\Delta\rho/\rho$ is positive in case there is no formation of superzone boundary gaps at the Néel temperature. In view of this, it is not clear whether the positive value of $\Delta\rho/\rho$ arises from the antiferromagnetic component; however, the observed magnitude is generally uncharacteristic of antiferromagnetic metals. Alternatively, the application of H presumably induces more spin fluctuations (in contrast to the usual influence of H to suppress spin fluctuations) in an otherwise more stable conical arrangement, as a result of which ρ in the presence of a field is larger. The positive sign may also arise if the presence of a field induces some pseudogaps in some portions of the Fermi surface. It is also possible that the scattering of the conduction electrons by the nonmagnetic layers La/Ge becomes dominant at low temperatures similar to the Ag interface effect observed in Cr/Ag/Cr trilayers.[6] These factors may also contribute to the low temperature positive $\Delta\rho/\rho$ in SmMn$_2$Ge$_2$.[5] Whatever may be the origin, the $\Delta\rho/\rho$ of LaMn$_2$Ge$_2$ presents an unusual and interesting situation. It is worthwhile to probe the role of anisotropy, stoichiometry and, in particular, disorder on these anomalies. Such studies become relevant in view of the recent report of the role of residual resistance on the magnitude of $\Delta\rho/\rho$ in multilayers.[6]

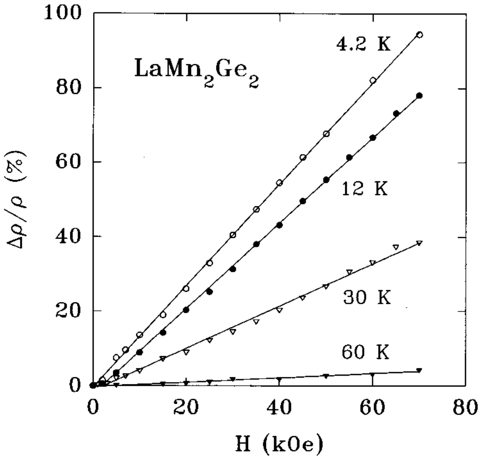

FIG. 2. The magnetoresistance ($\Delta\rho/\rho$) as a function of magnetic field at various temperatures for LaMn$_2$Ge$_2$. The lines through the data points serve as a guide to the eye.

Finally, we reiterate[5] that this class of Mn alloys, particularly those in which rare earths also order magnetically at lower temperatures, serve as model systems for artificial multilayers. Many of the alloys of the type RMn$_2$Si$_2$ are antiferromagnetic around 300 K (Ref. 4) and it would be interesting to look for magnetoresistance behavior of these alloys. Detailed investigation of all these alloys, particularly in thin films, will be a novel route to pursue the field of magnetism in multilayers.

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