Magnetoresistance anomalies and multiple magnetic transitions in $SmMn_2Ge_2$

E. V. Sampathkumaran,* P. L. Paulose, and R. Mallik
Tata Institute of Fundamental Research, Homi Bhabha Road, Colaba, Mumbai 400005, India
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The results of magnetoresistance ($\Delta\rho/\rho$), heat capacity (C), and ac susceptibility (χ) measurements are reported for a polycrystalline specimen of $SmMn_2Ge_2$. In addition to sharp variations of the order of 3–5 % at 110 and 142 K, there are anomalies in the sign and the magnitudes of $\Delta\rho/\rho$. A finding of considerable interest is that there is a gradual increase of $\Delta\rho/\rho$ from a small negative value at 35 K to *a very large value* of about 24% at 4.2 K in the presence of a magnetic field of 80 kOe, though apparently there is no corresponding anomaly in the C data. This finding signals the existence of a phase transition below 30 K attributable to magnetic ordering from Sm sublattice, thereby making the origin of the onset of Mn ordering around 110 K a puzzle. [S0163-1829(96)50530-8]

The layered compounds RMn$_2$Ge$_2$ (R=rare earth), known to crystallize in the $ThCr_2Si_2$-type tetragonal crystal structure, are of considerable interest[1–4] as the nature of magnetic ordering of the Mn sublattice depends sensitively on the planar Mn-Mn distance (d). Thus for R alloys in which the value of d exceeds 2.87 Å , ferromagnetism is observed and for $d<2.87$ Å antiferromagnetism prevails.[3–14] Among these alloys, $SmMn_2Ge_2$ presents an interesting situation as $d\cong 2.87$ Å; until recently, three magnetic transitions have been believed to exist:[3–11] below 345 K ($t1$), para to ferro; 160 K ($t2$), ferro to antiferro; 80–105 K ($t3$), antiferro to ferromagnetic. It is to be noted that there are some discrepancies in the reported values of these transition temperatures. The series of magnetic transitions in this compound cannot be reconciled with the systematics observed in the d dependence of magnetic behavior in this series, the reason being that the thermal contraction of the lattice upon the lowering of temperature should have resulted in only one transition, viz., ferromagnetism to antiferromagnetism. In order to explain this, it was generally assumed in the literature that magnetic ordering of the rare-earth sublattice in the close vicinity of $t3$ triggers a transition from the Mn sublattice. In contrast to this assumption, (doped) ^{57}Fe Mössbauer spectroscopic investigations[14] indicate the existence of an additional magnetic transition at 30 K ($t4$), presumably arising from Sm. [It may be added that these authors also find a transition at 385 K ($t5$) which is proposed to arise from the onset of antiferromagnetism from Mn with decreasing temperature.] There is, however, a need to verify the existence of this 30 K transition, as this conclusion is based on a study involving doping of Fe as an impurity. Clearly the magnetic phase diagram[6,7] of this compound needs to be modified if the existence of these new transitions is shown beyond doubt. The present investigation of magnetoresistance ($\Delta\rho/\rho$), heat capacity (C), and ac susceptibility (χ) studies on $SmMn_2Ge_2$ was initiated with the main motivation of probing the transition at $t4$. The results, besides rendering evidence for the existence of a magnetic transition at 30 K, bring out interesting aspects of this compound.

This material is of importance[10] considering that it shows promise as a model system for understanding artificial multilayers. Some of the findings reported in the recent literature are summarized below. The pressure dependencies of these transition temperatures, in terms of sign and magnitude, are found to be widely different.[6,7] At 104 K, a moderate field of 10 kOe is sufficient to bring about a metamagnetic transition and the corresponding magnitude of the magnetoresistance is as much as 4%.[7] There are noticeable changes in the lattice constants at $t2$ and $t3$. ^{149}Sm and ^{55}Mn NMR frequencies and linewidths also exhibit anomalies as a function of temperature and pressure[12,13] particularly below 100 K.

The polycrystalline sample of $SmMn_2Ge_2$ was prepared by arc melting and excess Mn (about 5%) was taken to compensate for the loss while melting. The ingot was homogenized at 800 °C in an evacuated, sealed quartz tube. An x-ray diffraction pattern confirms the single phase nature of the alloy. Electrical resistivity (ρ) measurements in the absence of a magnetic field (H) and in the presence of H =80 kOe were performed in the temperature interval 4.2–300 K by a conventional four-probe method employing a silver paint to make electrical contacts; in addition, ρ was also measured at 4.2 K as a function of H up to 80 kOe in the longitudinal mode (with the direction of the current being the same as that of H). The ac magnetic susceptibility (χ) (107 Hz, 0.08 Oe, 4.2–200 K) measurements were also performed to probe the magnetic transitions. Heat-capacity (C) behavior was also obtained (3–60 K) by a semiadiabatic heat-pulse method employing a home-built calorimeter.[15] In order to allow comparison of the transition temperatures, all these measurements were performed on the same specimen.

The results of ρ and χ measurements are shown in the temperature interval 4.2–200 K in Fig. 1. With decreasing temperature, ρ increases by about 5% at 110 K following antiferro to ferromagnetic transition, though ac χ starts increasing distinctly only below 90 K with a peak at 75 K. Since the measurements were performed on the same specimen, some physical significance should be attached to the apparent difference in this transition temperature. It may be remarked that the temperature dependence of reported dc χ (Ref. 14) is quite similar to that of our ac χ and hence the apparent discrepancy of ac χ with ρ cannot be attributed to frequency dependence of transition temperature. We believe

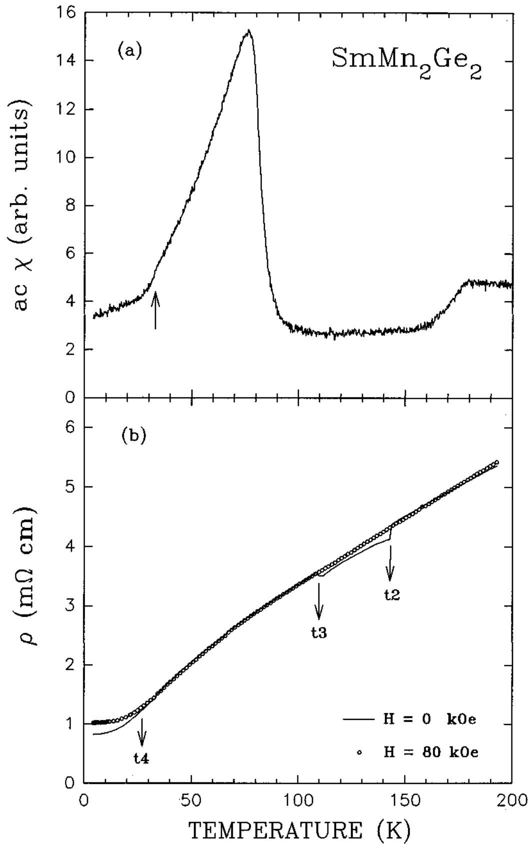

FIG. 1. (a) ac magnetic susceptibility and (b) electrical resistivity (ρ) in the absence (continuous line) and in the presence of a magnetic field (circle) (H) as a function of temperature (4.2–200 K) for polycrystalline $SmMn_2Ge_2$. The shoulder in the ac χ data around 35 K is indicated by a vertical arrow in (a).

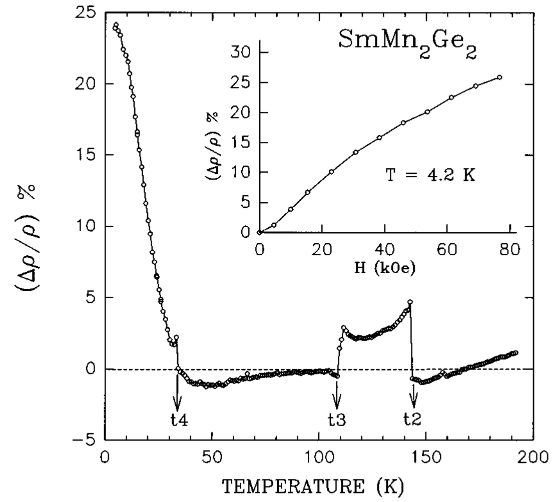

FIG. 2. The magnetoresistance, $\Delta\rho/\rho=[\rho(H)-\rho(0)]/\rho(0)$, for an applied field of 80 kOe for $SmMn_2Ge_2$. $t2$, $t3$, and $t4$ transitions are indicated ($t1$ and $t5$ are outside the temperature range of investigation). A line is drawn through the data points to serve as a guide to the eyes. The inset shows the variation of $\Delta\rho/\rho$ as a function of H at 4.2 K.

that the first order transition at 110 K sensed by ρ results in a very small change in magnetization, but the magnetization starts building only below a lower temperature. The transition around $t2$ also presents an interesting situation. It is clear that ρ exhibits a sharp drop by about 5% at 142 K following ferro to antiferromagnetic ordering. However, ac χ starts falling continuously from 180 K with decreasing temperature. This might imply that (short range) magnetic fluctuations precede long range antiferromagnetic ordering at 142 K sensed by ρ. Thus these results emphasize that there are hitherto unrecognized aspects associated with these transitions, thereby calling for further investigations particularly by neutron scattering technique.

Figure 1 also shows the temperature dependence of ρ in the presence of a field of 80 kOe. It is clear that the sharp variations in the values of ρ seen at 110 and 142 K in the zero field data are absent for $H=80$ kOe. Thus the nature of the spin alignment (viz., ferromagnetic) remains the same over the entire temperature range of investigation with the application of a field of $H=80$ kOe and this is consistent with the metamagnetic behavior in the antiferromagnetic state known earlier.[10] The magnetoresistance, defined as $\Delta\rho/\rho=[\rho(H)-\rho(0)]/\rho(0)$, is positive above 170 K ($>1\%$ above 200 K), decreases monotonically to -1% at about 145 K, increases again to 5% after the 142 K transition, followed by about a 3% drop at the 110 K transition (Fig. 2) with decreasing temperature. The values of $\Delta\rho/\rho$ in the range 110–140 K are comparable to those reported on single crystals in the metamagnetic state.[10] The finding to be emphasized is that there is a sharp increase below about 30 K from a value of about -1.5% at 35 K to a large positive value of about 25% at 4.2 K, as if there is a change in the spin dependent scattering contribution in ρ. We therefore attribute this significant variation of $\Delta\rho/\rho$ to the onset of another magnetic transition at about 30 K, presumably arising from Sm sublattice. This corroborates the conclusions of Nowik *et al.*[14] A closer look at the dc χ (Ref. 14) and ac χ data suggests that there is in fact a change in the slope in the plot of magnetization versus temperature at about 35 K, as indicated by a vertical arrow in Fig. 1(a). We have measured heat capacity (C) to look for the features due to this transition (Fig. 3) and surprisingly we do not see any noticeable anomaly around 35 K. This observation is puzzling. It is possible that this feature is smeared out for some reasons. It may be remarked that the magnetic moment for fully degenerate trivalent Sm ion is already low ($0.72\mu_\beta$). In the presence of crystal-field effects and transferred hyperfine field from Mn sublattice, the degeneracy of the ground state as well as the magnetic moment of Sm in the ordered state should be considerably reduced. Thus the entropy associated with the transition with the ground state must be rather low. In order to understand this aspect better, it is necessary to estimate the phonon contribution to C. Such an estimate is unreliable in the presence of the Mn sublattice ordering in the temperature range 75–110 K, which is bound to extend its influence on the C data at lower temperatures; in addition, La and Y analogs may not serve as good references as the Mn sublattice in these compounds exhibit magnetic ordering around 300 K only.[3] Alternatively, the magnetism from Sm is more of itinerant type due to strong hybridization between Mn $3d$ electrons and partially extended $4f$ orbital. Whatever may be the origin of the smearing out of C anomaly around

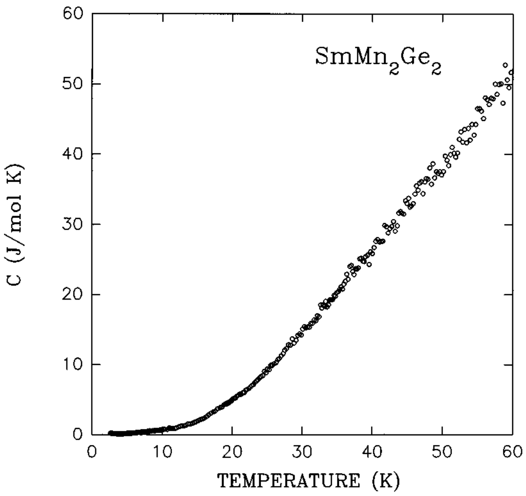

FIG. 3. Heat-capacity (C) as a function of temperature (T) for $SmMn_2Ge_2$.

30 K, it is clear that $\Delta\rho/\rho$ measurement serves as a powerful bulk method to detect magnetic transitions which are otherwise not so clearly detectable by other well known bulk methods.

We note some anomalies in the sign and magnitude of $\Delta\rho/\rho$. First, the magnitude of $\Delta\rho/\rho$ at 4.2 K is rather large and increases with H (Fig. 2, inset) attaining a very large value of about 24% at $H=80$ kOe. It is puzzling to note that the sign of $\Delta\rho/\rho$ is positive (*with the magnitude being large*), while a ferromagnetically ordered state (as proposed for Sm in $SmMn_2Ge_2$ in the literature[12,14]) generally should be characterized by a negative value of $\Delta\rho/\rho$. At this juncture, we like to recall[16] that we have reported a rather large negative value (−60%) of $\Delta\rho/\rho$ in the ferromagnetic state of $CeIr_2B_2$. Secondly, the loss of spin-dependent interlayer magnetic scattering contribution in the antiferromagnetic state[17] with the application of magnetic field in the temperature interval 110–140 K should have resulted in negative $\Delta\rho/\rho$, in contrast to the finding. We therefore believe that the magnetoresistance of $SmMn_2Ge_2$ is dominated by very subtle effects. It may be stated that partial replacement of Sm by Y inverts the sign of $\Delta\rho/\rho$ above $t3$ in this system[8] and this observation strengthens the above view. Thirdly, the values are negative in the temperature range 142 to 170 K, beyond which it is positive, thus resulting in the sign reversal at about 170 K. The negative value in this temperature range renders support to the proposal of the existence of magnetic fluctuations (which are suppressed by the application of a field) inferred from our ac χ data (see above).

In view of our observation of a transition below 30 K, we throw some light on the interpretation of the temperature dependence of ^{149}Sm and ^{55}Mn NMR frequencies[12] in this compound. Lord *et al.*[12] had difficulties in the interpretation of their data under the original assumption that Sm orders magnetically around 75 K. One of the main NMR findings is that Sm NMR could be observable only below 19 K. We propose that the nonobservation at higher temperatures, which was attributed to a rapid increase of the transverse relaxation rate with temperature, is in fact due to the disappearance of ferromagnetic ordering of Sm. This Sm NMR behavior thus renders support to our interpretation for the origin of $\Delta\rho/\rho$ anomaly at 30 K in terms of Sm magnetic ordering. As a consequence, one cannot invoke the collapse of Sm magnetism at 75 K to explain the continuous reduction of ^{55}Mn frequency above 75 K; considering that the plot of ^{55}Mn NMR frequency versus temperature (Fig. 9 in Ref. 12) resembles that of our ac χ data, we interpret the Mn NMR data in terms of continuous evolution of Mn magnetization below 100 K following the antiferro to ferromagnetic transition.

To conclude, the magnetoresistance of $SmMn_2Ge_2$ exhibits certain anomalies at low temperatures, which indicate the existence of a magnetic transition at about 30 K. This transition is attributed to Sm, consistent with the features in the reported Mössbauer and NMR data of this compound.[12,14] Therefore, the original idea that the magnetic ordering of Sm around 100 K results in the reentrance of Mn sublattice to ferromagnetic state at $t3$ may have to be revised and this makes the origin of a Mn transition at $t3$ a puzzle. Interesting anomalies in the sign and magnitude of $\Delta\rho/\rho$ are noted in the entire temperature range of investigation. There is an urgent need to confirm whether all these anomalies are related to complicated Fermi surface and/or magnetoelastic effects. A comparison of the ρ and χ data also shows interesting features around 110 K and 142 K transitions. In short, the physics of $SmMn_2Ge_2$ warrants further investigations, particularly to address immediately the questions on the exact nature of spin alignment as a function of temperature, dimensionalities of the transitions, interlayer and intralayer coupling strengths (between Sm and Mn), etc.

*Electronic address: SAMPATH@tifrvax.BITNET

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