Magnetic behavior of Gd_2CuGe_3: Electrical resistance minimum above the Néel temperature

Subham Majumdar and E. V. Sampathkumaran*
Tata Institute of Fundamental Research, Homi Bhabha Road, Colaba, Mumbai-400005, India
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We report the formation of a compound, Gd_2CuGe_3, in a AlB_2-derived crystal structure and its temperature-(T) dependent magnetic susceptibility, heat-capacity, electrical-resistivity (ρ), and magnetoresistance behavior. The results suggest that this compound orders antiferromagnetically below 12 K. The most notable finding is that the T derivative of ρ is negative below about 25 K, resulting in a minimum in the plot of ρ versus T around this temperature, a feature not very common among Gd alloys.

The observation of a distinct electrical-resistivity (ρ) minimum in the plot of ρ versus temperature (T) *well beyond the Néel temperature* (T_N), mimicking the behavior of Kondo lattices, in Gd_2PdSi_3, has raised interesting questions and calls for a deeper understanding of magnetic precursor effects.[1,2] It is of interest to search for Gd alloys with similar behavior, as it appears that this finding bears relevance to current trends in the fields of the Kondo effect and colossal magnetoresistance.[1,2] In the recent past,[1,3–18] there has been considerable interest in the synthesis and investigation of new compounds crystallizing in the AlB_2-derived hexagonal crystal structures which the compound Gd_2PdSi_3 belongs to. Considering this, we wanted to extend the search for the formation of new Gd compounds with similar transport behavior within the AlB_2-derived family. Here we report the formation of Gd_2CuGe_3, in which a distinct minimum in the plot of ρ versus T is noted around 25 K, which is far above the value of T_N (=12 K).

The sample was prepared by arc melting stoichiometric amounts of constituent elements in an atmosphere of argon. The ingot was homogenized in an evacuated, sealed quartz tube at 750 °C for 7 days. An x-ray diffraction pattern, obtained by employing Cu $K\alpha$ radiation, established that the present compound essentially crystallizes in an AlB_2-derived hexagonal structure, though at present it is difficult to ascertain whether the structure is an ordered invariant of this type.[14] In the absence of any superstructure line in the x-ray diffraction pattern, the lattice parameters are obtained as $a = 4.079$ Å and $c = 4.089$ Å. There is an additional weak line around $2\theta = 35.15°$, which is attributed to the presence of a small component (about 5%) with stoichiometry 1:2:2. The ρ measurements (2–300 K) were performed by a conventional four-probe method employing silver paint for making electrical contacts. The magnetic susceptibility (χ) measurements (2–300 K) were performed employing a commercial superconducting quantum interference device in the presence of a magnetic field (H) of 100 Oe as well as 2 kOe. The heat-capacity (C) data (2–40 K) were obtained by a semiadiabatic heat-pulse method. The (longitudinal mode) magnetoresistance (MR) data were obtained in the presence of a magnetic field of 50 kOe in the temperature range 4.2–100 K and also as a function of H at $T = 4.2$ K.

The temperature dependence of ρ, χ ($H = 100$ Oe), and C below 50 K is shown in Fig. 1. The inverse χ varies linearly with T above 80 K; however, there is a marginal deviation from linearity as T is lowered [Fig. 2(a)], resulting in an increase of the magnitude of $d\chi/dT$ below 80 K but attaining a negative minimum at 17 K. The effective moment obtained from the linear region turns out to be $8\mu_B$ per Gd ion which is very close to that expected for tripositive Gd ions; the sign of the paramagnetic Curie temperature (Θ_p) is found to be negative with a magnitude of 18 K. The χ, measured at both the fields 100 Oe and 2 kOe, exhibits a peak at 11.9 K. Though the plot of the isothermal magnetization in the magnetically ordered state measured up to 50 kOe is not a linear function of H, M does not undergo saturation [Fig. 2(b)] even at high fields; however, there is a weak curvature (up to high fields) of the plot, say, at 5 K,

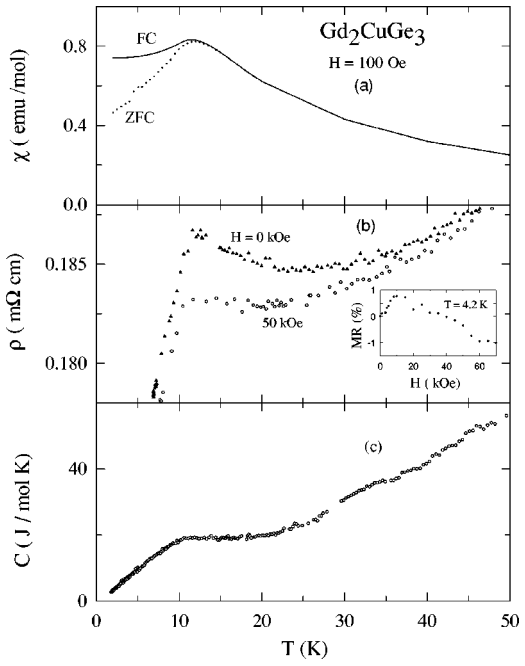

FIG. 1. (a) Magnetic susceptibility (χ) measured in the presence of 100 Oe, (b) electrical resistivity (ρ), and (c) the heat capacity (C) in the temperature range 2–50 K for Gd_2CuGe_3. FC (solid line) and ZFC (dotted line) for the χ data in (a) represent field-cooled and zero-field-cooled states of the specimen. The ρ data as a function of temperature in the presence of $H = 50$ kOe are also shown in (b). The inset in (a) shows the plot of magnetoresistance as a function of field at 4.2 K.

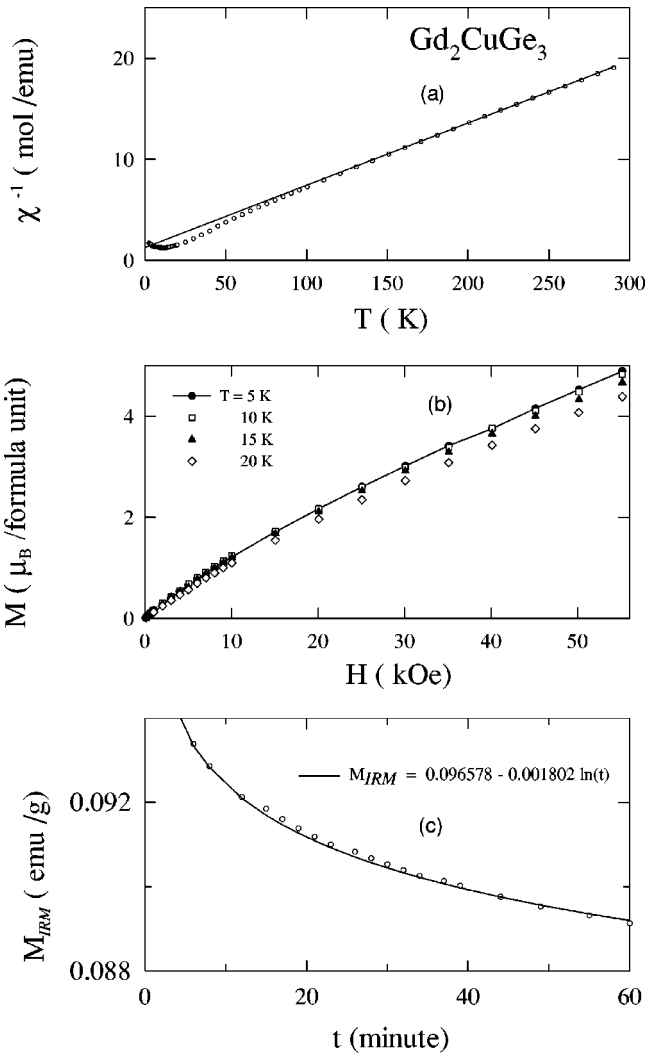

FIG. 2. (a) Inverse magnetic susceptibility as a function of temperature (2–300 K), (b) isothermal magnetization at various temperatures, and (c) isothermal remanent magnetization at 4.2 K as a function of time (t) for Gd_2CuGe_3. The solid lines in (a) and (c) represent a fit to the Curie-Weiss (80–300 K) behavior and a logarithmic function, respectively. The line through the data points at 5 K in (b) is a guide to the eyes and the data for and 5 and 10 K nearly overlap.

which might arise from the small amount of secondary phase of a ferromagnetic type. These findings suggest that Gd ions undergo distinct antiferromagnetic ordering in the close vicinity of 12 K in this compound. Other notable observations in the magnetization data are that (i) the field-cooled and zero-field-cooled χ data deviate from each other below 13 K (slightly above the peak temperature) and (ii) the sample was cooled to 4.2 K in zero field and then magnetized by keeping it in a field of 5 kOe for 5 min; the field was subsequently switched off and the isothermal remanent magnetization (M_{IRM}) value was recorded as a function of time; it is noted that M_{IRM} exhibits logarithmic decay with time as shown in Fig. 2(c). Though these properties are characteristic of spin glasses, such features are common among magnetic materials with long-range magnetic order as well.[19] As discussed below, the feature due to the onset of magnetic order in the ρ data is well defined (at 11.7 K) unlike in spin glasses and the MR is positive for initial applications of H at 4.2 K in contrast to the negative MR expected for spin glasses. Hence we attribute the magnetic transition to long-range magnetic ordering. The C data also exhibit an anomaly in the vicinity of T_N, however with a broadening of the feature extending to a rather wide temperature range (until about 25 K), as in the case of Gd_2PdSi_3 (Ref. 1), presumably due to short-range correlation effects as indicated by the deviation of inverse χ below 80 K from the high-temperature linear behavior [see Fig. 2(a)]; it is also possible that there is an amplitude modulation of the magnetic structure considering that the peak value of C (near T_N, 10 J/Gd mol K) is far below that expected (above 20 J/mol K) for equal-moment magnetic structure.[20] We are, however, not able to derive the magnetic contribution (C_m) to C, as there are difficulties in the synthesis of single-phase nonmagnetic analogs. From the shape of the plot of C versus T, one can infer that the tail of C_m above T_N might persist at least over a decade of temperature.

Now turning to the ρ data, it is found to decrease linearly in the temperature interval 100–300 K with decreasing temperature; there is no other worthwhile feature in the data in this temperature interval and hence not shown in the form of a figure above 50 K. There is a distinct drop in ρ at 11.7 K as the temperature is lowered. This drop arises from the onset of a magnetic ordering as evidenced by other measurements. The most notable finding is that the temperature derivative of ρ below 25 K is negative, thus resulting in a minimum at about 25 K in the plot of ρ versus T. The observation of such a ρ minimum, mimicking the behavior of Kondo lattices, is not very common among Gd alloys. This minimum, however, gets washed out with the application of a large magnetic field [Fig. 1(b)], resulting in negative magnetoresistance [defined as $\rho(H)-\rho(0)/\rho(0)$], as in the case of Kondo systems. Finally, we also measured the MR as a function of H at 4.2 K [see Fig. 1(b), inset] and we note that the MR is positive for $H<10$ kOe and becomes negative for higher values of H. This behavior is typical of antiferromagnets exhibiting metamagnetic transitions with the application of H (Ref. 2). It is, however, interesting to see that no such transition could be detected in the isothermal magnetization data [Fig. 2(b)] and thus there is no apparent correlation between isothermal MR and M behavior below T_N.

To conclude, we report the synthesis of a new compound Gd_2CuGe_3 crystallizing in an AlB_2-derived hexagonal structure. This compound undergoes antiferromagnetic ordering below 12 K, presumably undergoing amplitude modulation as indicated by the heat-capacity data. The most important finding is that there is a well-defined minimum at about 25 K, *far above* T_N in the ρ data, the T-dependent ρ behavior mimicking that of the magnetically ordering Kondo lattices, e.g., $CeAl_2$ and $CePd_2Si_2$ (Ref. 21). It may be recalled that the negative temperature coefficient of ρ before the onset of long-range magnetic order has been known even in Gd and Tb metals,[22] but the temperature (about 110% of the magnetic ordering temperature) at which this occurs is so close to the magnetic ordering temperature that it could still be visualized in light of traditional critical point effects (with a critical exponent of the order of 0.1). However, in the Gd alloys under discussion, the feature appears even beyond $2T_N$; therefore one has to assume an unusually large value of the critical exponent if one has to attribute it to critical point

effects. We attribute the resistivity minimum to possible s-f interaction-induced magnetic localization above T_N (see Refs. 1 and 2). In any case, the present finding firmly augments our earlier emphasis[1,2] that one needs to be cautious while attributing such a ρ minimum to the Kondo effect in magnetically ordering Ce or U systems as is generally the case in the literature. Finally, it is worthwhile to note that this ρ behavior is qualitatively similar to that seen in $Tl_2Mn_2O_7$ (Ref. 23), a compound gaining considerable attention in the field of colossal magnetoresistance due to the absence of double-exchange and Jahn-Teller effects (which is true even in the case of Gd alloys).

*Electronic address: sampath@tifr.res.in

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