Magnetic precursor effects, electrical and magnetoresistance anomalies, and heat-capacity behavior of Gd alloys

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The electrical resistivity (ρ), magnetoresistance ($\Delta\rho/\rho$), heat capacity (C), and magnetization (M) behavior in a number of Gd alloys are reported. Particular attention is paid to the ρ and $\Delta\rho/\rho$ behavior at temperatures just above respective long-range magnetic ordering temperatures (T_0). In some compounds, e.g., $GdNi_2Sn_2$ and $GdPt_2Ge_2$, there is an enhancement of ρ prior to long-range magnetic order over a wide temperature range which can be highlighted by the suppression of ρ caused by the application of a magnetic field. However, such features are absent in many other Gd compounds, e.g., $GdCu_2Ge_2$, $GdAg_2Si_2$, $GdAu_2Si_2$, $GdPd_2Ge_2$, and $GdCo_2Si_2$. Attempts to relate these features to magnetic precursor effects in C are made. We observe interesting features in the C data, like shoulders and additional transitions in the magnetically ordered state and peak values of C at T_0 varying from one compound to the other. The implications of these observations are discussed. In addition, various $\Delta\rho/\rho$ anomalies, including large positive values at low temperatures in some cases, and the sensitivity of magnetoresistance to field-induced magnetic transitions in comparison with the isothermal M data, are also brought out. [S0163-1829(98)03438-9]

I. INTRODUCTION

One of the points of debate in the field of giant magnetoresistance is the origin of negative temperature coefficient of resistivity (ρ) above the Curie temperature (T_C) and the resultant large negative magnetoresistance at T_C (Refs. 1,2). Keeping such trends in the field of magnetism in recent years in mind, we have been carefully investigating the magnetoresistance behavior of some of the Gd alloys in the vicinity of respective magnetic ordering temperatures (T_0), in order to address the question whether such features can arise from some other factor. We have indeed noted an extra contribution to ρ over a wide temperature range above T_0 in $GdPt_2Si_2$, $GdPd_2In$, $GdNi_2Si_2$ (Ref. 3), GdNi (Ref. 4), and Gd_2PdSi_3 (Ref. 5), as a result of which the magnetoresistance is negative just above T_0, attaining a large value at T_0, similar to the behavior in manganites. In fact, in one of the Gd compounds, Gd_2PdSi_3, the temperature coefficient of ρ is even negative just above Néel temperature (T_N), with a distinct minimum at a temperature far above T_N. Similar resistance anomalies have been noted above T_0 even in some Tb and Dy alloys.[6] Since critical spin fluctuations may set in as one approaches T_0, the natural tendency is to attribute these features to such spin fluctuations extending to unusually higher temperature range. In our opinion,[4–6] there exists a more subtle effect, which is yet to be understood. In the case of manganites, there is a proposal that a decrease in mobility of the carriers are primarily responsible for negative temperature coefficient of ρ above T_C and large magnetoresistance.[7,8]

The results on the Gd alloys mentioned above are also important to various developments in the field of heavy fermions and Kondo lattices, as discussed in Refs. 3–5, 9–11. Thus, the investigation of magnetic precursor effects in relatively simple magnetic systems is relevant to current trends in magnetism in general; the Gd systems are simple in the sense that Gd does not exhibit any complications due to double-exchange, crystal-field, Jahn-Teller, and Kondo effects.

We therefore consider it worthwhile to get more experimental information on Gd compounds in order to arrive at an overall picture of magnetic precursor effects. With this primary motivation, we carried out ρ, magnetoresistance ($\Delta\rho/\rho$), heat capacity (C), and magnetization (M) measurements in a number of other Gd alloys crystallizing in the same (or closely related) structure. Among the Gd alloys investigated, interestingly, many do not exhibit such resistance anomalies; in addition, we find that there is no one-to-one correspondence between the (non)observation of excess ρ and a possible enhancement of heat capacity (C) above T_0 in these Gd alloys. The compounds[12] under investigation are $GdCu_2Ge_2$ ($T_N = 12$ K), $GdAg_2Si_2$ ($T_N = 17$ K), $GdPd_2Ge_2$ ($T_N = 18$ K, Ref. 13), $GdCo_2Si_2$ ($T_N = 44$ K), $GdAu_2Si_2$ ($T_N = 12$ K), $GdNi_2Sn_2$ ($T_N = 7$ K), and $GdPt_2Ge_2$ ($T_N = 7$ K). The crystallographic and some of the magnetic properties of these compounds have been known,[12] to our knowledge with the exception of $GdPt_2Ge_2$. We have chosen this set of compounds, since all of these compounds are crystallographically related: most of these form in $ThCr_2Si_2$-type tetragonal structure, while $GdNi_2Sn_2$ and $GdPt_2Ge_2$ appear to form in a related structure, viz., $CaBe_2Ge_2$ or its monoclinic modification.[14,15] It may be added that the transition-metal ions, except Mn, are known not to carry any moment in these crystal structures; in the case of the cobalt alloy, the observed effective moment in the paramagnetic moment is marginally higher (by about $0.3\mu_B$) compared to that expected for trivalent Gd ion, presumably due to the polarization of the conduction band induced by Gd.[12]

II. EXPERIMENT

The samples were prepared by arc melting stoichiometric amounts of constituent elements in an arc furnace in an at-

mosphere of argon and annealed at 800 °C for 7 days. The samples were characterized by x-ray diffraction. The electrical resistivity measurements were performed in zero field as well as in the presence of a magnetic field (H) of 50 kOe in the temperature interval 4.2–300 K by a conventional four-probe method employing a silver paint for electrical contacts of the leads with the samples; in addition, resistivity was measured as a function of H at selected temperatures; no significance may be attached to the absolute values of ρ due to various uncertainties arising from the brittleness of these samples, voids and the spread of silver paint. The C measurements were performed by a semiadiabatic heat-pulse method in the temperature interval 2–70 K in order to look for certain correlations with the behavior in ρ; respective nonmagnetic Y or La compounds have also been measured so as to have an idea on the lattice contribution, though it is not found to be reliable at high temperatures (far above T_0). In order to get further information on the magnetic behavior, the magnetic susceptibility (χ) was also measured in a magnetic field of 2 kOe (2–300 K) employing a superconducting quantum interference device; the data are shown only in the vicinity of T_0, as the high-temperature data are featureless agreeing with the results known in the literature; the behavior of isothermal M was also obtained at selected temperatures.

III. RESULTS AND DISCUSSION

The results of ρ measurements in the absence and in the presence of a magnetic field are shown in Fig. 1(a) below 45 K for $GdPt_2Ge_2$. The C data are shown in Fig. 1(b). The χ data in the same temperature interval are shown in Fig. 1(c) to establish the value of T_N. The magnetoresistance, defined as $\Delta\rho/\rho=[\rho(H)-\rho(0)]/\rho(0)$, as a function of H at selected temperatures are shown in Fig. 1(d). From the comparison of the data in (a), (b), and (c), it is clear that this compound undergoes long-range magnetic ordering at ($T_N=7$ K), presumably of an antiferromagnetic type, considering that the Curie-Weiss temperature (θ_p) obtained from high-temperature Curie-Weiss behavior of χ is negative (-8 K) and the isothermal magnetization (M) at 4.5 K does not show indication for saturation [and, in fact, varies linearly with H, Fig. 1(c), inset]. There is an upturn in ρ below 7 K, instead of a drop, presumably due to the development of magnetic Brillouin-zone boundary gaps.[16] However, with the application of a magnetic field, say 50 kOe, this low-temperature upturn in ρ gets depressed; the point to be noted is that there is a significant depression of ρ with the application of H even above 7 K, the magnitude of which decreases with increasing temperature. Thus, there is a significant negative magnetoresistance not only below T_N, but also above it over a wide temperature range. This point can be emphasized more clearly when one measures $\Delta\rho/\rho$ as a function of H at various temperatures [Fig. 1(d)]. There is a quadratic variation with H (up to about 50 kOe) at all temperatures mentioned in the plots, attaining a large value at higher fields, and these are characteristics of spin-fluctuation systems. In order to explore whether any such magnetic precursor effects are present in the C data, we show the magnetic contribution (C_m) to C in Fig. 1(b) after subtracting the lattice contribution (derived from the C data of YPt_2Ge_2) as

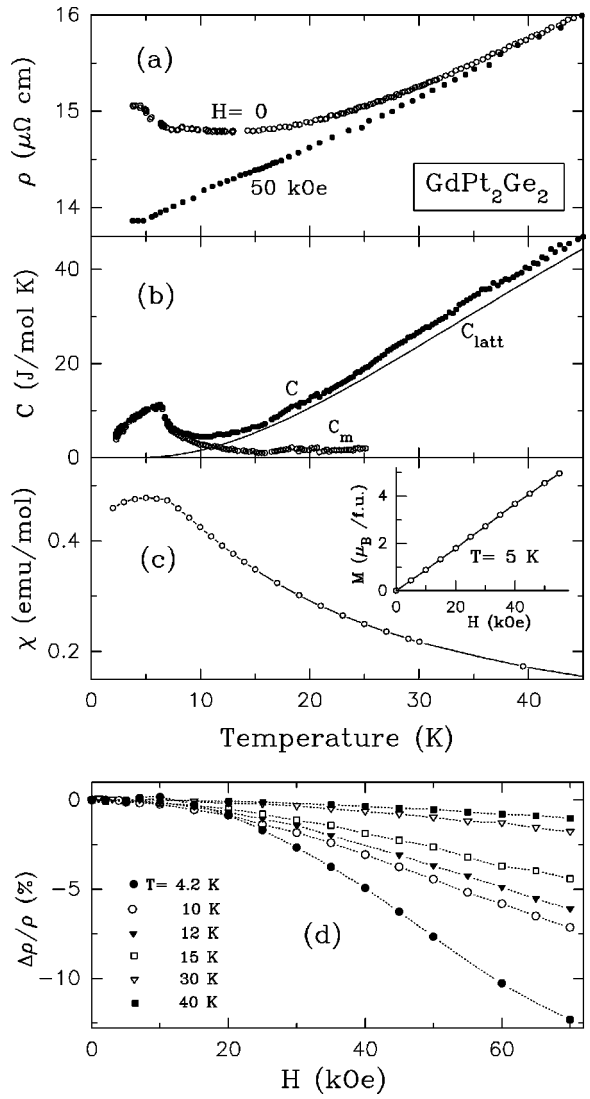

FIG. 1. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, (b) heat capacity (C), lattice contribution to C and the magnetic contribution (C_m) to C and (c) the magnetic susceptibility below 45 K as well as the isothermal magnetization (inset) at 5 K for $GdPt_2Ge_2$. The magnetoresistance, $\Delta\rho/\rho$, as a function of magnetic field (H) at various temperatures is shown in (d). The lines drawn through the data points serve as guides to the eyes.

described in Refs. 9,17. It appears that this may not be the perfect way of determination of C_m above 30 K as the derived lattice part does not coincide with the measured data for the sample, though the magnetic entropy (obtained by extrapolation of C_m to zero Kelvin) reached its highest value (R ln 8) around 40 K; there may possibly be a different degree of crystallographic disorder between Gd and Y alloys, which is responsible for this discrepancy. Clearly the feature is rounded off at the higher temperature side of T_N, resulting in a tail extending to higher temperature range and this feature is free from the error discussed above. The data basically provide evidence for the fact that the full magnetic entropy (R ln 8) is attained only in the range 30–40 K and it is exactly the same temperature range until which we see an enhancement of ρ, depressing with the application of H. In short, this compound exhibits magnetic precursor effects both in C and ρ data.

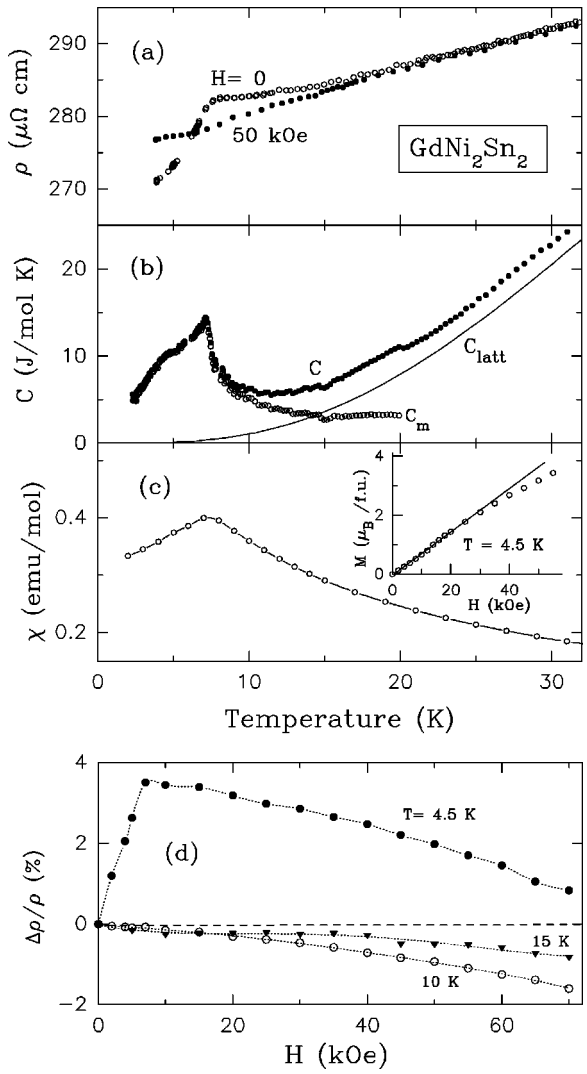

FIG. 2. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, (b) heat capacity (C), lattice contribution to C, and the derived magnetic contribution (C_m) to C and (c) the magnetic susceptibility for $GdNi_2Sn_2$ below 32 K. The magnetoresistance, $\Delta\rho/\rho$, as a function of magnetic field (H) at various temperatures is shown in (d). The lines drawn through the data points serve as guides to the eyes. The isothermal magnetization behavior at 4.5 K is plotted in the inset of (c) and the low field linear region is shown by a continuous line.

As in the case of $GdPt_2Ge_2$, the results obtained from various measurements for $GdNi_2Sn_2$ are shown in Fig. 2 below 35 K. It is clear from the features in ρ, C, and χ that this compound orders magnetically at about 7 K; from the reduced value of peak C_m (lattice contribution derived from the values of YNi_2Sn_2) (Ref. 17) and negative $\theta_p(-13\ \mathrm{K})$, we infer that the magnetic structure is of an amplitude-modulated (AM) antiferromagnetic type. The main point of emphasis is that there is an excess resistivity until about 15 K, which is highlighted by the depression of ρ with the application of H. Though there are problems similar to $GdPt_2Ge_2$ in deducing precise lattice contribution at higher temperature, we are confident that C_m data (qualitatively) exhibit a tail until about 15 K and the total magnetic entropy is released around the same temperature. The magnetoresistance appears to vary nearly quadratically with H above T_N,

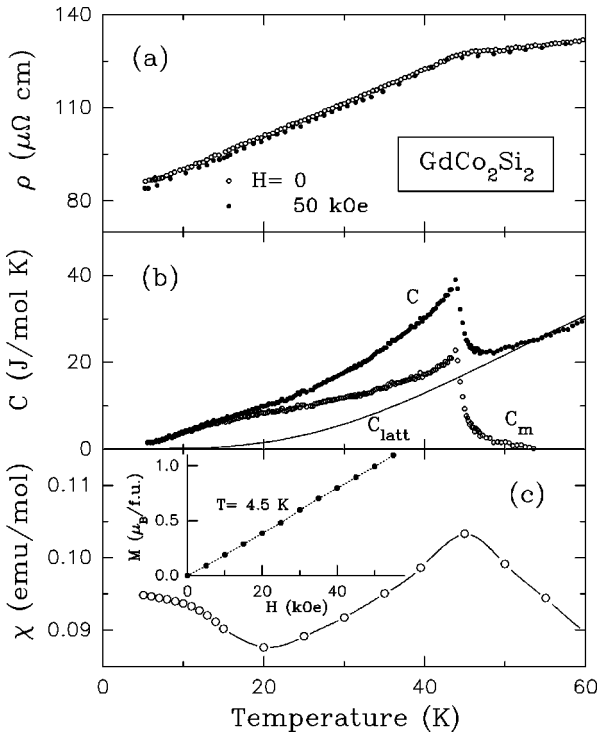

FIG. 3. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, (b) Heat capacity (C), lattice contribution to C and the magnetic contribution (C_m) to C and (c) the magnetic susceptibility below 60 K as well as the isothermal magnetization (inset) at 4.5 K for $GdCo_2Si_2$. The lines drawn through the data points serve as guides to the eyes.

say, at 10 and 15 K. Thus, ρ and C data show magnetic precursor effects for this alloy as well.

We now present the results on a series of Gd alloys in which the excess resistance (in the sense described above) is not observable above T_0. These alloys are $GdCo_2Si_2$ (Fig. 3), $GdAu_2Si_2$ (Fig. 4), and $GdPd_2Ge_2$ (Fig. 5). It is clear from Figs. 3–5 that the resistivity in the presence and in the ab-

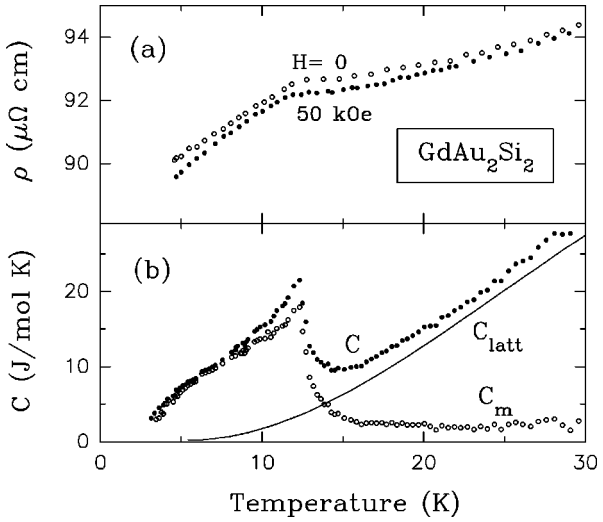

FIG. 4. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, and (b) heat capacity (C), lattice contribution to C, and the magnetic contribution (C_m) to C for $GdAu_2Si_2$ below 30 K.

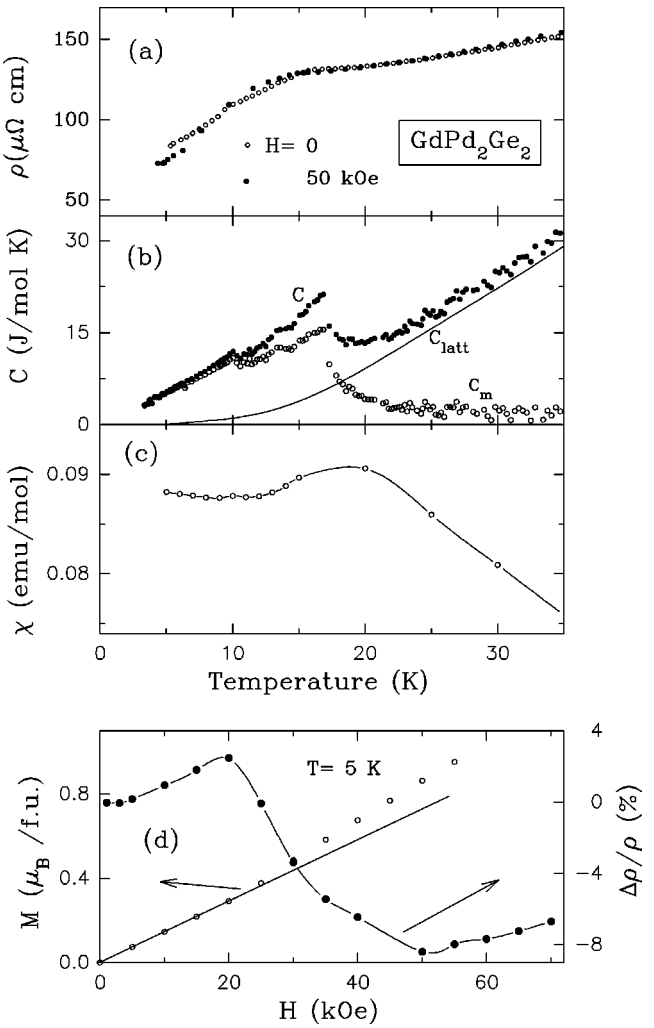

FIG. 5. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, (b) heat capacity (C), lattice contribution to C, and the magnetic contribution (C_m) to C and (c) the magnetic susceptibility below 35 K for $GdPd_2Ge_2$. The magnetoresistance, $\Delta\rho/\rho$, and isothermal magnetization as a function of magnetic field (H) at 5 K are shown in (d). The lines drawn through the data points serve as guides to the eyes in all the plots except for the M versus H plot, in which case the straight line represents the low field linear region.

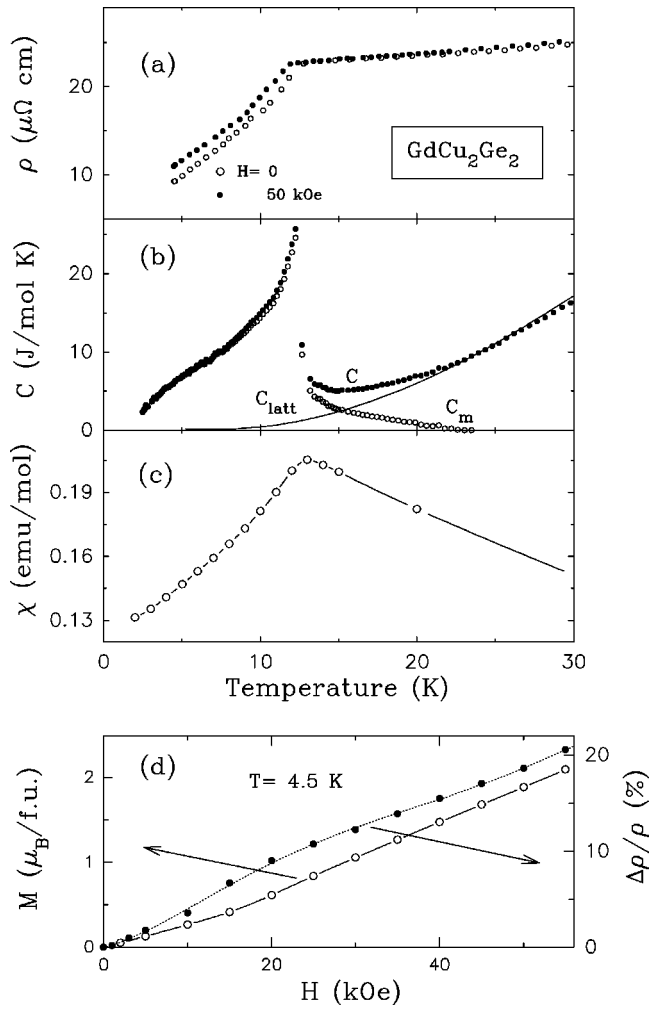

FIG. 6. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, (b) heat capacity (C), lattice contribution to C and the magnetic contribution (C_m) to C and (c) the magnetic susceptibility below 30 K for $GdCu_2Ge_2$. The magnetoresistance, $\Delta\rho/\rho$, and isothermal magnetization as a function of magnetic field (H) at 4.5 K are plotted in (d). The lines drawn through the data points serve as guides to the eyes.

sence of H are practically the same (within 0.1%) above their respective ordering temperatures, thereby establishing the absence of an additional contribution to ρ before long-range ordering sets in. It is to be noted that even in Co alloy in which excess effective moment has been reported[12] (also confirmed by us), the excess ρ is absent. In order to look for the tail in C_m above T_0, we attempted to obtain respective lattice contributions (employing the C values of YCo_2Si_2, YAu_2Si_2 and YPd_2Ge_2, respectively). We can safely state that the continuous decrease in C_m just above T_0, if exists, does not proceed beyond $1.2T_0$ [see Figs. 3(b), 4(b), and 5(b)]. Thus, it appears that the magnetic precursor effects in C, if present, are negligible, thus tracking the behavior of ''excess resistance.''

In $GdCu_2Ge_2$ and $GdAg_2Si_2$ as well, clearly there is no excess resistivity above T_N, as the application of H does not suppress the value of ρ (Figs. 6 and 7). However, in contrast to the cases discussed in the previous paragraph, it appears that there is no correlation between C and ρ behavior prior to long-range magnetic order. YCu_2Ge_2 and $LaAg_2Si_2$ have been used as references to obtain lattice contributions to C, respectively. The finding of interest is that the magnetic contribution to C appears to exhibit a prominent tail (without any doubt in $GdCu_2Ge_2$), at least until 10 K above respective T_N. This behavior is similar to that noted for $GdCu_2Si_2$ earlier.[11,17]

We have also made various other interesting findings. Bouvier *et al.*[17] and Blanco *et al.*[18,19] have elegantly shown for Gd alloys how the peak C_m value at T_0 can be used to derive information on magnetic structure. According to these reports, for an equal moment (EM) magnetic structure (simple antiferro, ferro or helimagnetic), this value for Gd should be 20.15 J/mol K and the amplitude modulation (AM) is expected to reduce this value. This guideline to infer the magnetic structure appears to be superior even to neutron diffraction in some respects, particularly for Gd considering that Gd is not very convenient for neutron-diffraction stud-

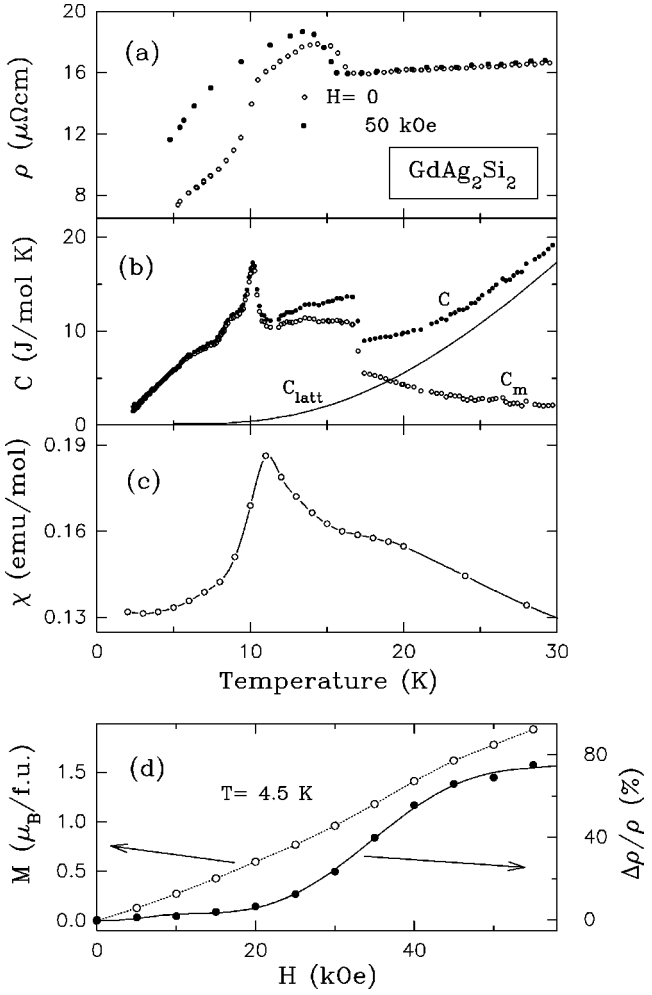

FIG. 7. (a) Electrical resistivity in zero field and in the presence of a magnetic field of 50 kOe, (b) heat capacity (C), lattice contribution to C, and the magnetic contribution (C_m) to C and (c) the magnetic susceptibility below 30 K for $GdAg_2Si_2$. The magnetoresistance, $\Delta\rho/\rho$, and isothermal magnetization as a function of magnetic field (H) at 4.5 K are plotted in (d). The lines drawn through the data points serve as guides to the eyes.

ies. On the basis of such ideas, we look at the C behavior of our alloys. The peak values of C_m for $GdPt_2Ge_2$ and $GdNi_2Sn_2$ are much smaller than that expected for EM magnetic structures and the fact that the value is reduced by at least a factor of about 1/3 shows that the magnetic structure is modulated. The situation is somewhat similar for $GdPd_2Ge_2$. However, for $GdCo_2Si_2$ and $GdAu_2Si_2$, the peak values of C_m are very close to the expected value for EM magnetic structures, thus suggesting that the (antiferromagnetic) magnetic structure is not modulated. Interestingly, for $GdCu_2Ge_2$, the corresponding value is unusually high (about 25 J/mol K), the implications of which are not clear at the moment.

In addition, Blanco *et al.*[18,19] also derive that the exact shape of the C versus T curves below T_0 strongly depends on the details of the exchange coupling; sudden jumps in C well below T_0 due to a transition from an AM- to EM-type structure or additional shoulders due to an evolution to a full antiphase structure also can appear in the C versus T plots. In addition, a hump arises around $T/T_N = 0.25$, corresponding to a Schottky-like anomaly in the ordered state involving quantum levels of $(2J+1)$-fold degenerate multiplet the energy positions of which depend on the temperature through the thermal variation of the exchange field.[18,19] Clearly, the C behavior in the ordered state can be very complex. In light of these ideas, we now look at the features in the C data of the present compounds. The most prominent behavior is seen for $GdAg_2Si_2$; there are two prominent magnetic transitions, (interestingly) a discontinuous one near 17 K and the other at 11 K [see the features in C and χ in Figs. 7(b) and 7(c)]. There are additional shoulders at about 6 and 8 K. Due to such a complex C versus T behavior, we avoid inferring magnetic structure from the peak value of C_m for this compound. At the 17 K transition in this compound, there is a sudden upward jump in C, and at the same temperature ρ shows a sudden upturn instead of a decrease (Fig. 7), possibly due to the formation of antiferromagnetic energy gaps. It would be interesting to probe whether the transition is first order in nature. There are very prominent shoulders at about 10 and 14 K for $GdPd_2Ge_2$ and at 5 K for $GdNi_2Sn_2$ well below respective T_0. There are relatively less intense, but clearly visible shoulders below T_0 for $GdCo_2Si_2$ (at about 20 K), $GdAu_2Si_2$ (at about 7 and 10 K) and $GdCu_2Ge_2$ (around 10 K and below) as well; such features are apparently rounded off for $GdPt_2Ge_2$. It is to be noted that the additional magnetic transition in $GdCo_2Si_2$ around 20 K can be inferred from the upturn in the susceptibility also [Fig. 3(c)]. Thus, the C behavior of these Gd alloys in the magnetically ordered state is quite rich in features.

We now compare the field dependence of magnetoresistance with that of isothermal M. For $GdNi_2Sn_2$ [Fig. 2(d)], $\Delta\rho/\rho$ as a function of H at 4.5 K exhibits a sharp rise for initial applications of H with a positive peak near 8 kOe. While the positive sign may be consistent with antiferromagnetism, corresponding anomaly in the isothermal magnetization at 4.5 K is not very prominent; the plot of M versus H, however, is not perfectly linear at 4.5 K, showing a weak metamagnetic tendency around 30 kOe [Fig. 2(c), inset]. It appears that the peak in the magnetoresistance is a result of significant changes in the scattering effects from a weak metamagnetism. Even in the case of $GdAg_2Si_2$, there is a weak feature in the plot of M vs H at 4.5 K in the field range 20–40 kOe due to possible metamagnetic transition [see Fig. 7(d)], which is pronounced in the magnetoresistance beyond 20 kOe. In the case of $GdPd_2Ge_2$, at 5 K, $\Delta\rho/\rho$ shows a positive value until 20 kOe, beyond which the value is negative exhibiting a nonmonotonic variation with H [Fig. 5(d)]; the plot of M versus H shows only a small deviation from linearity around this field. Thus there are very weak metamagnetic effects which have subtle effects on the scattering processes in the magnetically ordered state in these compounds. The plot of magnetoresistance versus H and that of isothermal magnetization look similar for $GdCu_2Ge_2$ [Figs. 6(d)], with a very weak metamagnetic tendency near 35 kOe, as reflected by nonlinear plots. These results suggest that the magnetoresistance technique is a powerful tool to probe metamagnetism, even the weak ones, which may not be clearly detectable by magnetization measurements. Additional points to be noted in the magnetoresistance data are (i) the value of magnetoresistance is very large (about 80%) at high fields at 5 K [see Fig. 7(d)] for $GdAg_2Si_2$; the corre-

sponding values are reasonably large for $GdCu_2Ge_2$ as well (Fig. 6). It may be added that sufficiently large positive values of magnetoresistance have been reported by us in layered compounds of this type, not only on antiferromagnets,[20] but also on isostructural paramagnetic[21] and ferromagnetic[22] compounds, which prompts us to believe that the magnetoresistance mainly originates from nonmagnetic layers similar to the interface effect reported for Cr/Ag/Cr trilayers;[23] alternatively, the role of granularity to give rise to such large values cannot be excluded; (ii) an inspection of Fig. 2(a) and Fig. 5(a) suggest that the magnetoresistance changes sign from positive to negative at a temperature well below T_0 as the temperature is raised from 4.2 K for $GdNi_2Sn_2$ and $GdPd_2Ge_2$; these may be related to the changes in the temperature-induced changes in the magnetic structures.

IV. CONCLUSIONS

To summarize, on the basis of our investigations on Gd alloys, we divide the Gd compounds into two classes: Class I, in which there is an excess contribution to ρ prior to long-range magnetic order over a wide temperature range, as a result of which the magnetoresistance is large and negative, e.g., GdNi, $GdNi_2Si_2$, $GdPt_2Si_2$, $GdPt_2Ge_2$. $GdNi_2Sn_2$, $GdPd_2In$, Gd_2PdSi_3; Class II, in which such features are absent, e.g., $GdCu_2Si_2$, $GdCu_2Ge_2$, $GdAg_2Si_2$, $GdAu_2Si_2$, $GdCo_2Si_2$, $GdPd_2Ge_2$. (At this juncture, we would like to add that we performed similar studies on compounds such as $GdCu_2$, $GdAg_2$, $GdAu_2$, $GdCoSi_3$, and $GdNiGa_3$ and we do not find any magnetic precursor effects). The present study on isostructural compounds establishes that there is no straightforward relationship between the observation of the excess ρ, on the one hand, and the crystal structure or the type of transition metal and s-p ions present in the compound, on the other. The fact that all the compounds studied in this investigation are of the layered type suggests that possible onset of magnetic correlations within a layer before long-range magnetic order sets in cannot be offered as the sole reason for excess resistivity selectively in some cases. One is tempted to attribute the observation of excess ρ to critical spin fluctuations extending to higher temperature range, as inferred from the tail in C_m above T_0. If so, the conclusions of several reports in the literature assuming the validity of the Ginzburg criterion of critical point effects restricting such effects to a narrow temperature range above T_0 are questionable. However, one does not get a consistent picture, the reason being that, in some of the class II alloys, there is a distinct tail in C_m (in which we are confident, though unambiguous determination of absolute C_m values is found to be difficult in general in most of the present compounds). It is therefore clear that there must be more physical meaning for the appearance of excess ρ in class I alloys. If the speculative idea proposed in Refs. 5 and 6, viz., ''magnetic disorder-induced localization of electrons'' before the onset of long-range order in some alloys is confirmed, one should explore various factors determining the presence or the absence of the possible ''magnetic-localization'' effects; possibly, the relative magnitudes of mean free path, localization length,[7] and short-range correlation length, in addition to the strength of polarization of the conduction band (as measured, say, by the excess effective moment), may be some of the deciding factors. The results imply that there are conceptually open questions in understanding the magnetic behavior of even relatively simple compounds like those of Gd. Finally, this paper also brings out interesting features in the heat capacity and magnetoresistance in the magnetically ordered state as well in these Gd compounds. It is worthwhile to probe carefully the magnetic phase diagram (field- and temperature-induced magnetic phases) for these compounds.

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