

## Magnetic anisotropy, first-order-like metamagnetic transitions, and large negative magnetoresistance in single-crystal $\text{Gd}_2\text{PdSi}_3$

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Electrical resistivity ( $\rho$ ), magnetoresistance (MR), magnetization, thermopower, and Hall-effect measurements on the single-crystal  $\text{Gd}_2\text{PdSi}_3$ , crystallizing in an  $\text{AlB}_2$ -derived hexagonal structure, are reported. The well-defined minimum in  $\rho$  at a temperature above Néel temperature ( $T_N=21$  K) and large negative MR below  $\sim 3T_N$ , reported earlier for the polycrystals, are reproducible even in single crystals. Such features are generally uncharacteristic of Gd alloys. In addition, we also found interesting features in other data, e.g., two-step first-order-like metamagnetic transitions for the magnetic field along  $[0001]$  direction. The alloy exhibits anisotropy in all these properties, though Gd is an  $S$ -state ion. [S0163-1829(99)00341-0]

The observation of large negative magnetoresistance (MR) above respective magnetic transition temperatures in some polycrystalline Gd (also Tb, Dy) alloys is of considerable interest.<sup>1-5</sup> Among the Gd alloys, we have studied the transport properties of the compound  $\text{Gd}_2\text{PdSi}_3$ , crystallizing in an  $\text{AlB}_2$ -type structure, which has been found to show unusual nature. While this compound orders antiferromagnetically at ( $T_N=$ ) 21 K, there is unexpectedly a distinct minimum in the temperature-dependent electrical resistivity ( $\rho$ ) at about 45 K. This minimum disappears by the application of a magnetic field ( $H$ ), thereby resulting in large MR in the vicinity of  $T_N$ .<sup>3</sup> These properties are also characteristic of Ce/U-based Kondo lattices, but uncharacteristic of Gd systems, considering that the Gd-4*f* orbital is so deeply localized that it cannot exhibit the Kondo effect. Though magnetic-polaronic effect (even in metallic environments) has been proposed in Refs. 1-5 as one of possible mechanisms behind this large MR, its origin is not clear yet. It, however, appears that short-range correlation as a magnetic precursor effect may be the primary ingredient<sup>5</sup> for the origin of the resistivity minimum above  $T_N$  and negative MR. The importance of such findings is obvious from similar recent reports from other groups<sup>6-10</sup> and among these the observation of  $\rho$  minimum and resultant colossal magnetoresistance (CMR) in a pyrochlore-based oxide,  $\text{Ti}_2\text{Mn}_2\text{O}_7$ ,<sup>7,8</sup> has attracted recent attention. In view of the importance of the observations on polycrystals of this Gd compound, we considered it important to confirm the findings on the single crystals. With this primary motivation, we have investigated  $\rho$ , MR, thermopower ( $S$ ), Hall effect, and magnetization behavior on single crystals of  $\text{Gd}_2\text{PdSi}_3$ , and these results are presented in this article.

Single crystals of  $\text{Gd}_2\text{PdSi}_3$  have been prepared by the Czochralsky pulling method using a tetra-arc furnace in an argon atmosphere. The single-crystalline nature has been confirmed using backscattering x-ray technique. The  $\rho$ , MR, and Hall-effect (employing a magnetic field of 15 kOe) measurements have been performed by a conventional dc four-

probe method down to 1.2 K; the MR and Hall-effect measurements have also been performed as a function of  $H$  at 4.2 K. The magnetic measurements have been carried out with a Quantum Design superconducting quantum interference device magnetometer. The thermopower data were taken by the differential method using Au-Fe (0.07%)-chromel thermocouples.

Figure 1(a) shows the temperature dependence (1.2–300 K) of  $\rho$  for the sample with the current  $j//[10\bar{1}0]$  and  $j//[0001]$  in zero field. In Fig. 1(b), the low-temperature data, normalized to the 300 K value, in the absence of a

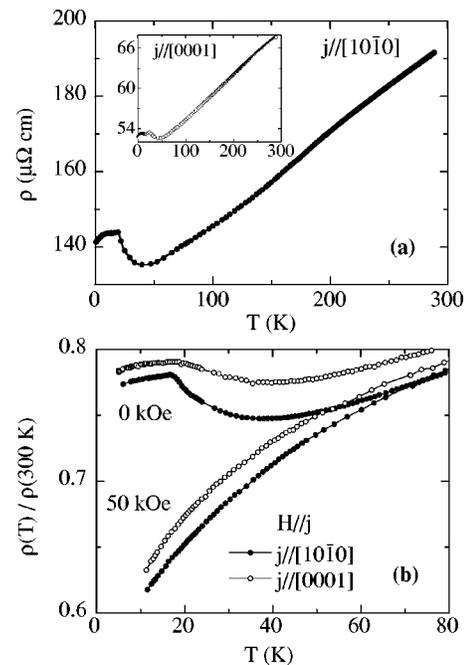


FIG. 1. (a) The electrical resistivity ( $\rho$ ) of single-crystalline  $\text{Gd}_2\text{PdSi}_3$  as a function of temperature (1.2–300 K) for  $j//[10\bar{1}0]$  and  $j//[0001]$  (inset). (b) The low-temperature  $\rho$  data in the absence of a magnetic field and in the presence of a field of 50 kOe are shown in an expanded form after normalizing to 300-K values.

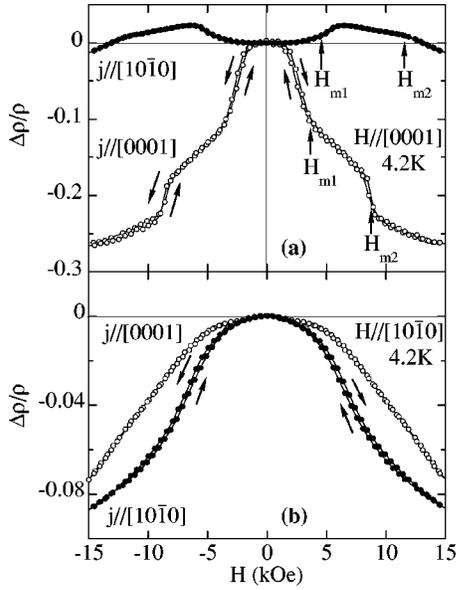


FIG. 2. The magnetic-field dependence of magnetoresistance for  $\text{Gd}_2\text{PdSi}_3$  in the transverse and longitudinal geometries, as labeled in the figure. The arrows indicate the directions of the field sweep.

magnetic field as well as in the presence of 50 kOe (in the longitudinal geometry,  $H//j$ ) are shown. In zero field,  $\rho$  gradually decreases with decreasing temperature as in ordinary metals, however, only down to about 45 K below which there is an upturn. There is a kink at about 21 K for both directions, marking the onset of magnetic ordering.<sup>3</sup> The  $\rho$ , however, does not drop sharply at  $T_N$  expected due to the loss of spin-disorder contribution, but exhibits a tendency to flatten or a fall slowly with decreasing temperature. Presumably, the magnetic structure could be a complex one, resulting in the formation of superzone-zone boundary gaps in some portions of the Fermi surface. The application of a magnetic field, say  $H=50$  kOe, in the geometry discussed above, however, depresses the  $\rho$  minimum restoring metallic behavior in the entire temperature range of investigation. This naturally means that there is a large negative magnetoresistance,  $\text{MR} = \Delta\rho/\rho = [\rho(H) - \rho(0)]/\rho(0)$ , at low temperatures, the magnitude of which increases with decreasing temperature. A large negative value close to  $-30\%$  could be seen for moderate fields (15 kOe) at 4.2 K [Fig. 2(a)], an indication of giant magnetoresistance. Thus all these features observed in polycrystals are reproducible in single crystals as well. It is obvious [Fig. 1(a)] that the absolute values of  $\rho$  are relatively higher for  $j//[10\bar{1}0]$  than that for  $j//[0001]$ . It is to be noted that, though these values still fall in the metallic range, the temperature dependence is rather weak, for instance,  $\rho(4.2\text{ K})/\rho(300\text{ K})$  is not less than 0.75 (in zero field), in sharp contrast to a value of about 0.2 even for polycrystalline  $\text{Lu}_2\text{PdSi}_3$ .<sup>11</sup> It is not clear whether this fact is associated with some kind of disorder effect on  $\rho$  in magnetic sample compared to that in nonmagnetic  $\text{Lu}_2\text{PdSi}_3$  or with an intrinsic mechanism responsible for the  $\rho$  minimum.

We have also measured MR as a function of  $H$  at 4.2 K with  $H$  varying from  $-15$ – $15$  kOe, both in the longitudinal and transverse geometries for  $H//[0001]$  and  $H//[10\bar{1}0]$ . For  $H//[0001]$ , the transverse MR ( $j//[10\bar{1}0]$ ) is positive with a small magnitude, while the longitudinal MR

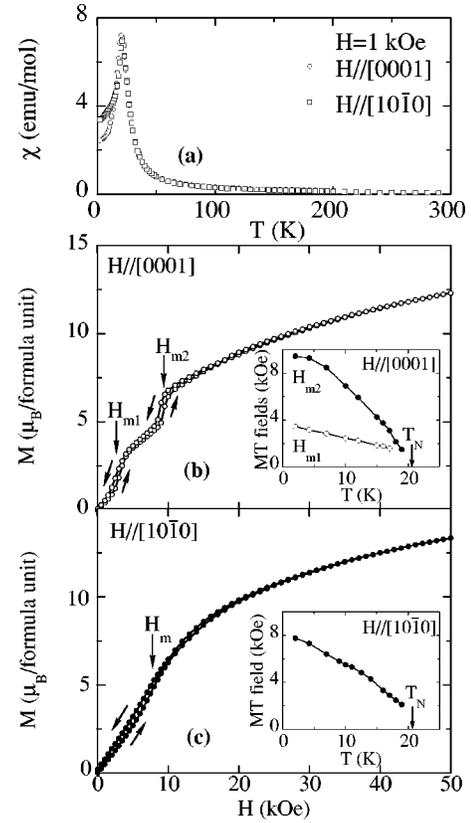


FIG. 3. (a) The magnetic susceptibility versus temperature (2–300 K) for single-crystals  $\text{Gd}_2\text{PdSi}_3$ . The isothermal magnetization behavior at 2 K for  $H//[0001]$  and  $H//[10\bar{1}0]$  are shown respectively in (b) and (c); the insets show the metamagnetic transition (MT) fields obtained as described in the text as a function of temperature.

( $j//[0001]$ ) is negative [see Fig. 2(a)]. The contribution from the anisotropic MR due to spin-orbit coupling is negligible for the Gd ion. The cyclotron contribution to the resistivity is also small. Possible contribution resulting from the reduction of magnetic scattering due to the metamagnetic transitions should be the same for both geometries, since the field directions are the same. Therefore the anisotropy in MR reflects the anisotropy of the Fermi surface for the two current directions. We believe that the conductivity parallel to  $[0001]$  is favored by the disappearance of the magnetic superzone gaps in some portions of the Fermi surface, resulting in a decrease of  $\rho$  in the longitudinal MR geometry for  $H//[0001]$ . However, it is interesting to note that, for  $H//[10\bar{1}0]$ , one sees negative MR for both geometries ( $j//[10\bar{1}0]$  and  $j//[0001]$ ). A noteworthy finding is that, for  $H//[0001]$ , there are sharp changes in MR when measured as a function of  $H$  as indicated by arrows in Fig. 2(a), but occurring at different fields for the two geometries due to the difference in the demagnetizing fields. There is a small hysteresis at the region around the sharp changes and we see similar behavior even in the magnetization data (see below); such sharp variations are absent for  $H//[10\bar{1}0]$ . All these results bring out anisotropic nature of MR.

Figure 3(a) shows the temperature dependence of magnetic susceptibility ( $\chi$ ), measured in the presence of a field of 1 kOe for both  $H//[0001]$  and  $H//[10\bar{1}0]$ . There is a well-

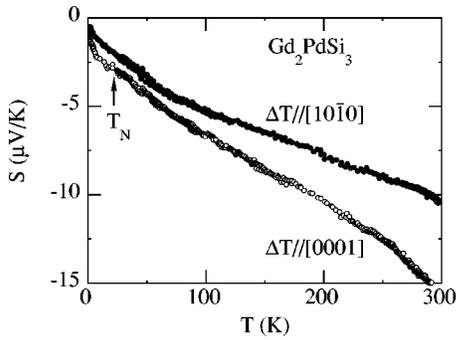


FIG. 4. The thermopower as a function of temperature for two different directions of thermal gradient on  $\text{Gd}_2\text{PdSi}_3$  single crystals.

defined peak in  $\chi$  at 21 K confirming the antiferromagnetic nature of the magnetic transition; below 21 K, however, there is only a small difference in the values for these two geometries. The paramagnetic Curie-temperature turns out to be the same for both geometries, with the same magnitude as that of  $T_N$ , however, with a positive sign suggesting the existence of strong ferromagnetic correlations. There is no difference between field-cooled (FC) and zero-field-cooled (ZFC)  $\chi$  values below 21 K, unlike the situation in polycrystals.<sup>3</sup> This suggests that such a difference in FC and ZFC data in polycrystals is not intrinsic to this material. This fact supports our earlier conclusion<sup>3</sup> that this alloy is not a spin glass, unlike  $\text{U}_2\text{PdSi}_3$ .<sup>12</sup>

The isothermal magnetization ( $M$ ) behavior at 2 K is shown in Fig. 3(b), both for increasing and decreasing fields. For the field along [0001], there are two steplike metamagnetic transitions, one around 3 kOe and the other around 9 kOe. Apparently, there is a small hysteresis around these transitions, indicating first-order nature of the transitions. The inset of Fig. 3(b) shows the metamagnetic transition fields  $H_{m1}$  and  $H_{m2}$  (the magnetic fields corresponding to the highest  $dM/dH$  at the low-field and high-field transitions, respectively) versus temperature; both  $H_{m1}$  and  $H_{m2}$  decrease with increasing temperature.  $M$  vs  $H$  for  $H//[10\bar{1}0]$  also shows a faint metamagnetic anomaly [Fig. 3(c)], however, with  $M$  varying relatively smoothly with  $H$ , unlike the situation for  $H//[0001]$ ; the inset shows the characteristic magnetic field,  $H_m$  (estimated in the same way as  $H_{m1}$  and  $H_{m2}$ ). The results establish the existence of anisotropy in the isothermal magnetization. The observation of metamagnetic transitions are consistent with the anomalies in MR, discussed above.

Figure 4 shows the temperature dependence of thermopower. The absolute value is large at 300 K as in the case of  $\text{Lu}_2\text{PdSi}_3$ .<sup>3</sup> Therefore the large  $S$  might arise from  $4d$  band of Pd as in the case of  $3d$  band of Co in  $\text{YCo}_2$ .<sup>13</sup> There is no anomaly, however, at  $T_N$ .  $S$  decreases with decreasing temperature and the features are qualitatively the same as those observed in the nonmagnetic  $\text{Lu}_2\text{PdSi}_3$ .<sup>3</sup> Though the overall  $S$  behavior mimics the one in polycrystals,<sup>3</sup> there is a distinct anisotropy in the values when measured along different directions, that is, the value of  $S$  depends on whether the temperature gradient,  $\Delta T$ , is parallel to  $[10\bar{1}0]$  or  $[0001]$ . The absence of any peaklike behavior expected for Kondo system<sup>14</sup> indicates the absence of Kondo effect in  $\text{Gd}_2\text{PdSi}_3$ .

The temperature dependence of Hall coefficient ( $R_H$ ),

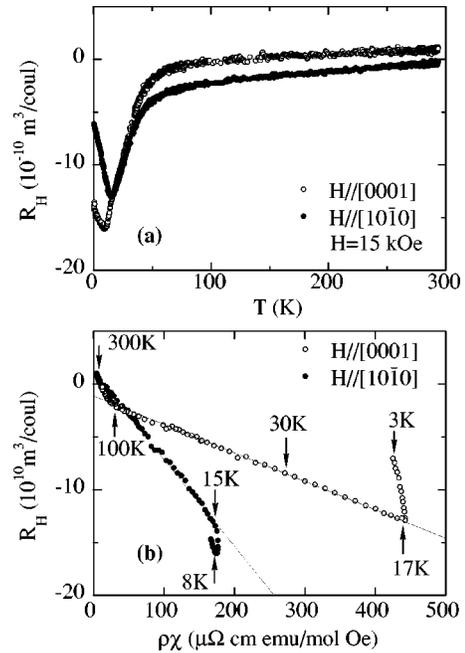


FIG. 5. The temperature dependence of Hall coefficient  $R_H$  for  $\text{Gd}_2\text{PdSi}_3$  single crystals with two different orientations is plotted in (a). In (b) the values of  $R_H$  are plotted as a function of the electrical resistivity times magnetic susceptibility, with temperature as an intrinsic parameter. The temperature region in which the  $R_H$  varies linearly is shown by drawing continuous lines through the data points. For  $H//[0001]$ , it is obvious that there is a deviation from this line above 100 K, besides the one near  $T_N$ .

shown in Fig. 5(a), also reflects anisotropic nature of this material. The  $R_H$  shows large temperature dependence, in contrast to the temperature independent behavior in  $\text{Lu}_2\text{PdSi}_3$ ,<sup>3</sup> with a negative peak for both geometries,  $H//[0001]$  and  $H//[10\bar{1}0]$ , in the vicinity of  $T_N$ , however, at slightly different temperatures (the reason for which is not clear). Clearly there is a dominant  $4f$  contribution in the Gd case. The Hall effect of magnetic metals like those of Gd is generally a sum of two terms—an ordinary Hall effect ( $R_0$ )

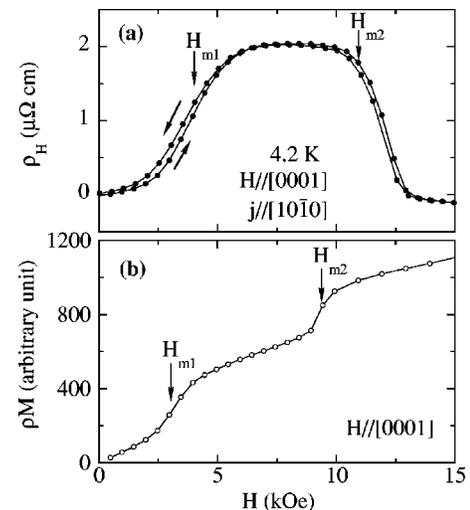


FIG. 6. (a) The field dependence of Hall resistivity in  $\text{Gd}_2\text{PdSi}_3$  for  $H//[0001]$ . The arrows indicate the directions of the field sweep. (b) Field dependence of  $\rho M$  for  $H//[0001]$ .

due to Lorentz force and an anomalous part arising from magnetic scattering (skew scattering). Thus in the paramagnetic state,  $R_H = R_0 + A\rho\chi$ , where  $A$  is a constant. Using this relation,  $R_0$  is estimated by plotting  $R_H$  versus  $\rho\chi$  [Fig. 5(b)]. From Fig. 5(b), it is obvious that the plot is linear for  $H//[10\bar{1}0]$  in the paramagnetic state with a value of  $R_0 = 0.92 \times 10^{-10} \text{ m}^3/\text{C}$ . However, for  $H//[0001]$ , there is a distinct change in the magnitude as well as in the sign around 110 K as if there is a change in the sign of the carrier ( $1.6 \times 10^{-10} \text{ m}^3/\text{C}$  and  $-1.3 \times 10^{-10} \text{ m}^3/\text{C}$  for  $T > 110 \text{ K}$  and  $T < 110 \text{ K}$ , respectively). Below 17 K, the data however deviate from the high-temperature linear variation as the state is no longer paramagnetic.

Figure 6(a) shows the field dependence of Hall resistivity ( $\rho_H$ ) for  $H//[0001]$ .  $\rho_H$  shows distinct anomaly across the two metamagnetic transition fields and traces the hysteresis. Again considering the concept of anomalous Hall effect discussed in the above paragraph, the field dependence of the corresponding  $\rho M$  is plotted in Fig. 6(b). Theoretically, when anomalous Hall effect is dominant,  $\rho_H$  should vary linearly with  $\rho M$ . In other words,  $\rho_H$  and  $\rho M$  should vary in the same way with the corresponding applied fields. But in the present case, the field dependence of  $\rho_H$  completely differs from that of  $\rho M$ , particularly around  $H_{m2}$ . This fact strongly

indicates the modification of the Fermi surface across the metamagnetic anomaly.

Summarizing, we have explored anisotropy in the transport and magnetic properties on the single-crystal  $\text{Gd}_2\text{PdSi}_3$ . Possibly the anisotropic exchange interaction due to crystal-line anisotropy and the anisotropy in the Fermi surface are responsible for the observed anisotropy. Interestingly, there are magnetic-field induced first-order-like magnetic transitions in the magnetically ordered state, resulting in large MR and consequent Fermi-surface modification. Recently, first-order transition has been reported in another Gd alloy,  $\text{Gd}_5(\text{Si,Ge})_4$ ,<sup>15</sup> which has been found to be a simultaneous crystallographic and magnetic transition, and in view of this it is of interest to explore whether there is any structural transition with the application of  $H$  in our case as well. In short, the single-crystal  $\text{Gd}_2\text{PdSi}_3$  exhibits interesting features. Above all, there is a well-defined  $\rho$  minimum above  $T_N$ , the origin of which is still not completely clear; the negative MR persists till about  $3T_N$  even in single crystals with the magnitude gradually increasing with decreasing temperature towards  $T_N$ . The results overall establish that this compound is an interesting magnetic material.

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