Magnetic anisotropy, first-order-like metamagnetic transitions and large negative magnetoresistance in the single crystal of Gd_2PdSi_3

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Electrical resistivity (ρ), magnetoresistance (MR), magnetization, thermopower and Hall effect measurements on the single crystal Gd_2PdSi_3, crystallizing in an AlB_2-derived hexagonal structure are reported. The well-defined minimum in ρ 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 a S-state ion.

The observation of large negative magnetoresistance (MR) above respective magnetic transition temperatures in some polycrystalline Gd (also Tb, Dy) alloys is of considerable interest.[1–5] Among the Gd alloys, we have studied the transport properties of the compound, Gd_2PdSi_3, crystallizing in an 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 (ρ) 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 [Ref. 3]. These properties are also characteristic of Ce/U-based Kondo lattices, but uncharacteristic of Gd systems, considering that the Gd-4f orbital is so deeply localised that it cannot exhibit the Kondo effect. Though magnetic-polaronic effect (even in metallic environments) has been proposed in references 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[5] 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[6–10] and among these the observation of ρ minimum and resultant colossal magnetoresistance (CMR) in a pyrochlore-based oxide, $Tl_2Mn_2O_7$ [Ref. 7,8], has attracted recent attention. In view of the importance of the observations on polycystals of this Gd compound, we considered it important to confirm the findings on the single crystals. With this primary motivation, we have investigated ρ, MR, thermopower (S), Hall-effect and magnetization behavior on single crystals of Gd_2PdSi_3, and these results are presented in this article.

Single crystals of Gd_2PdSi_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 back scattering x-ray technique. The ρ, 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. The thermopower data were taken by the differential method using Au-Fe (0.07%)-chromel thermocouples.

Fig. 1a shows the temperature dependence (1.2-300 K) of ρ for the sample with the current j//[10$\bar{1}$0] and j//[0001] in zero field. In Fig. 1b, the low temperature data, normalised to the 300 K value, in the absence of a magnetic field as well as in the presence of 50 kOe (in the longitudinal geometry, H//j) are shown. In zero field, the ρ(T) gradually decreases with decreasing temperature like 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.[3] The ρ, 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 ρ minimum restoring metallic behavior in the entire temperature range of investigation. This naturally means that there is a large negative magnetoresistance, $MR = \Delta\rho/\rho = [\rho(H) - \rho(0)]/\rho(0)$, 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. 2a), an indication of giant magnetoresistance. Thus, all these features observed in polycrystals, are reproducible in single crystals as well. It is obvious (Fig. 1a) that the absolute values of ρ 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, ρ(4.2K)/ρ(300K) is not less than 0.75 (in zero field), in sharp contrast to a value of about 0.2 even for polycrystalline Lu_2PdSi_3 (Ref. 11). It is not clear whether this fact is associated with some kind of disorder effect on

ρ in magnetic sample compared to that in nonmagnetic Lu_2PdSi_3 or with an intrinsic mechanism responsible for the ρ minimum.

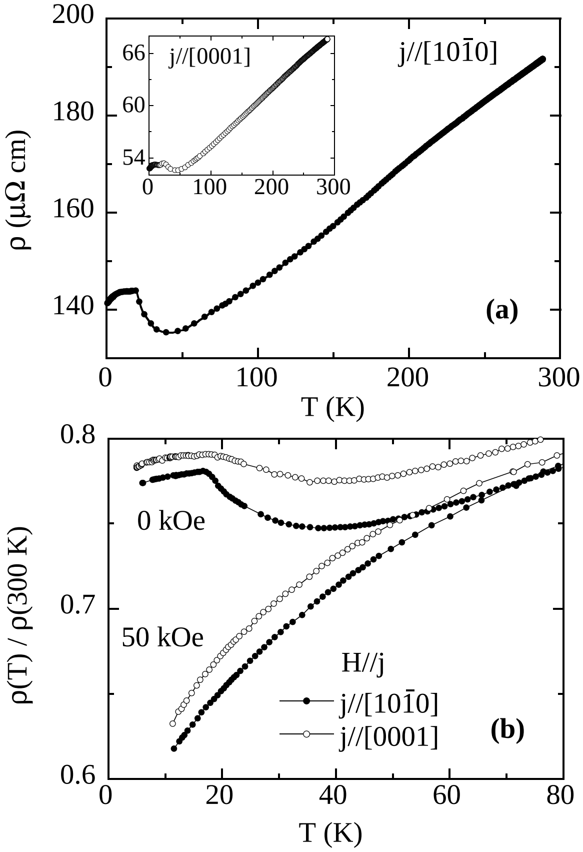

FIG. 1. (a) The electrical resistivity (ρ) of single crystalline Gd_2PdSi_3 as a function of temperature (1.2-300 K) for j//[10$\bar{1}$0] and j//[0001] (the inset). (b) The low temperature ρ data in the absence of a magnetic field and in the presence of a field of 50 kOe is shown in an expanded form after normalizing to 300 K values.

We have also measured MR as a function of H at 4.2 K with H varying from -15 kOe to 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}$0]) is positive with a small magnitude, while the longitudinal MR (j//[0001]) is negative (see Fig. 2a). The contribution from the anisotropic MR due to spin-orbit coupling is negligible for the Gd ion. The cyclotron contribution to the resistivity is also small. Possible contribution resulting from the reduction of magnetic scattering due to the metamagnetic transitions should be the same for both geometries, since the field directions are the same. Therefore, the anisotropy in MR reflects the anisotropy of the Fermi surface for the two current directions. We believe that the conductivity parallel to [0001] is favored by the disappearance of the magnetic superzone gaps in some portions of the Fermi surface, resulting in a decrease of ρ in the longitudinal MR geometry for H//[0001]. However, it is interesting to note that, for H//[10$\bar{1}$0], one sees negative MR for both geometries (j//[10$\bar{1}$0] and j//[0001]). A noteworthy finding is that, for H//[0001], there are sharp changes in MR when measured as a function of H as indicated by arrows in Fig. 2a, but occurring at different fields for the two geometries due to the difference in the demagnetizing fields. There is a small hysteresis at the region around the sharp changes and we see similar behavior even in the magnetization data (see below); such sharp variations are absent for H//[10$\bar{1}$0]. All these results bring out anisotropic nature of MR.

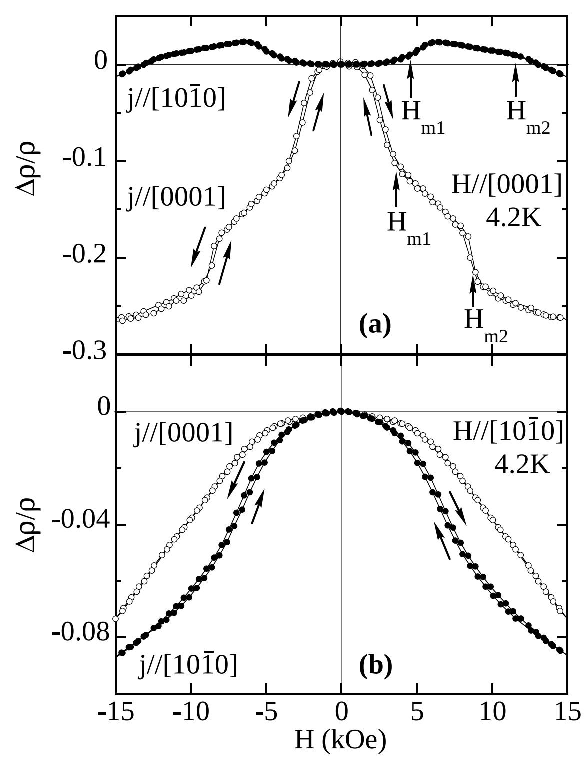

FIG. 2. The magnetic field dependence of magnetoresistance for Gd_2PdSi_3 in the transverse and longitudinal geometries, as labelled in the figure. The arrows indicate the directions of the field sweep.

Fig. 3a shows the temperature dependence of magnetic susceptibility (χ), measured in the presence of a field of 1 kOe for both H//[0001] and H//[10$\bar{1}$0]. There is a well-defined peak in χ 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 ferromagneticcorrelations. There is no difference between

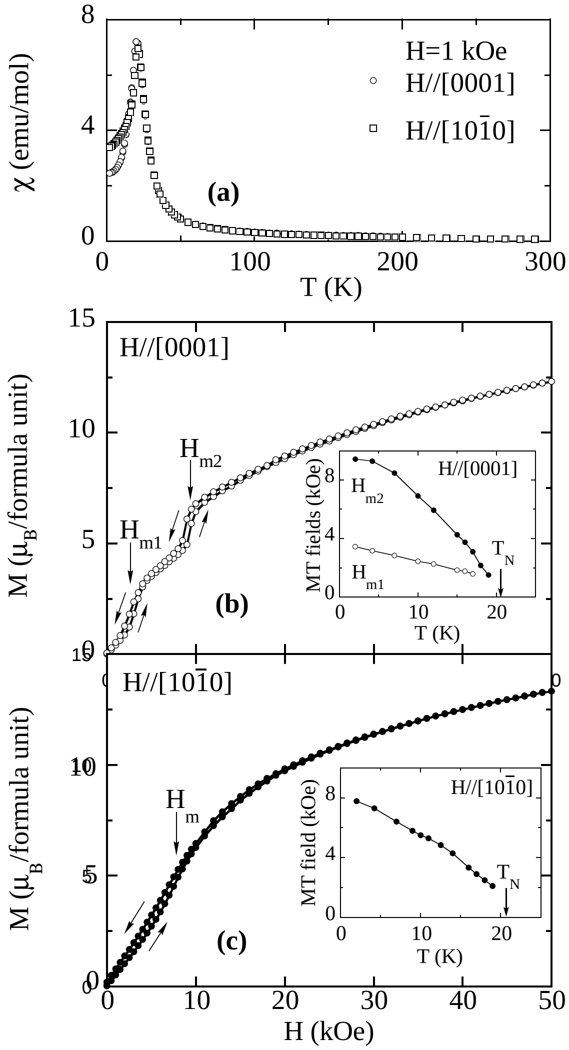

FIG. 3. (a) The magnetic susceptibility versus temperature (2-300 K) for single crystals of Gd_2PdSi_3. The isothermal magnetization behavior at 2 K for H//[0001] and H//[10$\bar{1}$0] are shown respectively in (b) and (c); the insets show the metamagnetic transition (MT) fields obtained as described in the text as a function of temperature.

field-cooled (FC) and zero-field-coooled (ZFC) χ values below 21 K, unlike the situation in polycrystals.[3] This suggests that such difference in FC and ZFC data in polycrystals is not intrinsic to this material. This fact supports our earlier conclusion[3] that this alloy is not a spin-glass, unlike U_2PdSi_3 (Ref. 12).

The isothermal magnetization (M) behavior at 2 K is shown in Fig. 3b, both for increasing and decreasing fields. For the field along [0001], there are two step-like 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 figure 3b 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 meta-magnetic anomaly (Fig. 3c), 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.

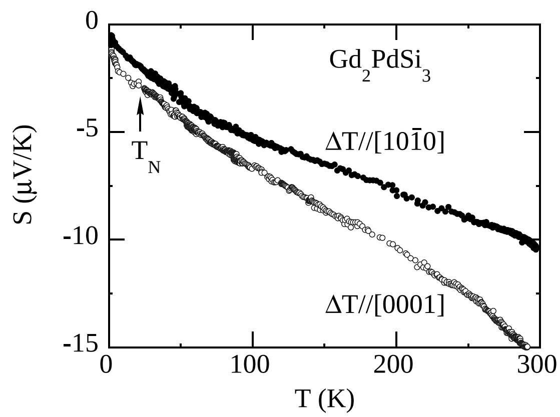

FIG. 4. The thermopower as a function of temperature for two different directions of thermal gradient on Gd_2PdSi_3 single crystals.

Fig. 4 shows the temperature dependence of thermopower. The absolute value is large at 300 K as in the case of Lu_2PdSi_3 (Ref. 3). Therefore, the large S might arise from 4d band of Pd as in the case of 3d band of Co in YCo_2.[13] 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 non-magnetic Lu_2PdSi_3.[3] Though the overall S behavior mimics the one in polycrystals,[3] 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, ΔT, is parallel to [10$\bar{1}$0] or [0001]. The absence of any peak-like behavior expected for Kondo system[14] indicates the absence of Kondo effect in Gd_2PdSi_3.

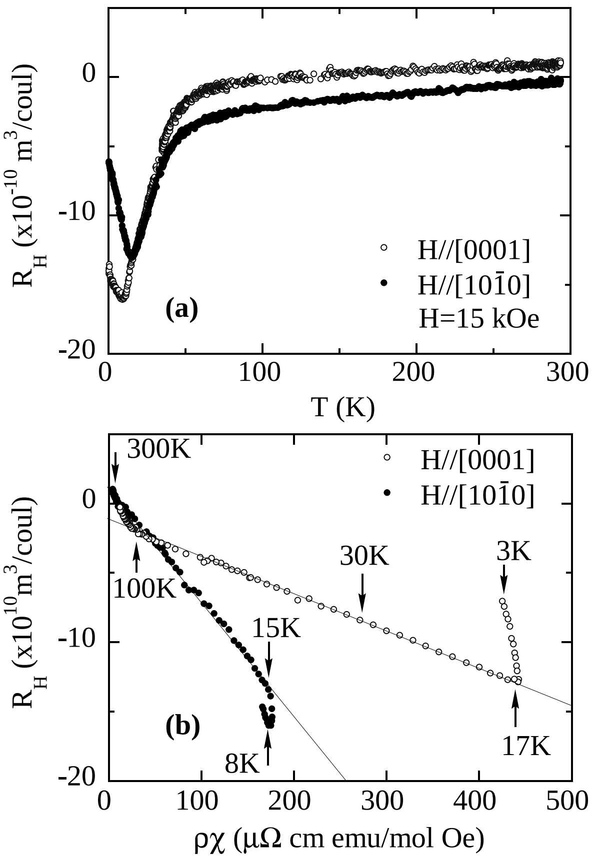

FIG. 5. The temperature dependence of Hall coefficient, R_H, for Gd_2PdSi_3 single crystals with two different orientations is plotted in (a). In (b) the values of R_H are plotted as a function of the electrical resistivity times magnetic susceptibility, with temperature as an intrinsic parameter. The 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.

The temperature dependence of Hall coefficient (R_H), shown in Fig. 5a, also reflects anisotropic nature of this material. The R_H shows large temperature dependence, in contrast to the temperature independent behavior in Lu_2PdSi_3 (Ref. 3), 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) due to Lorentz force and an anomalous part arising from magnetic scatterring (skew scatterring). 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. 5b). From Fig. 5b, it is obvious that the plot is linear for H//[10$\bar{1}$0] in the paramagnetic state with a value of $R_0 = 0.92\times10^{-10}$ m^3/coul. 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; (1.6×10^{-10} m^3/coul and -1.3×10^{-10} m^3/coul for T> 110 K and T<110 K, respectively). Below 17 K, the data however deviate from the high temperature linear variation as the state is no longer paramagnetic.

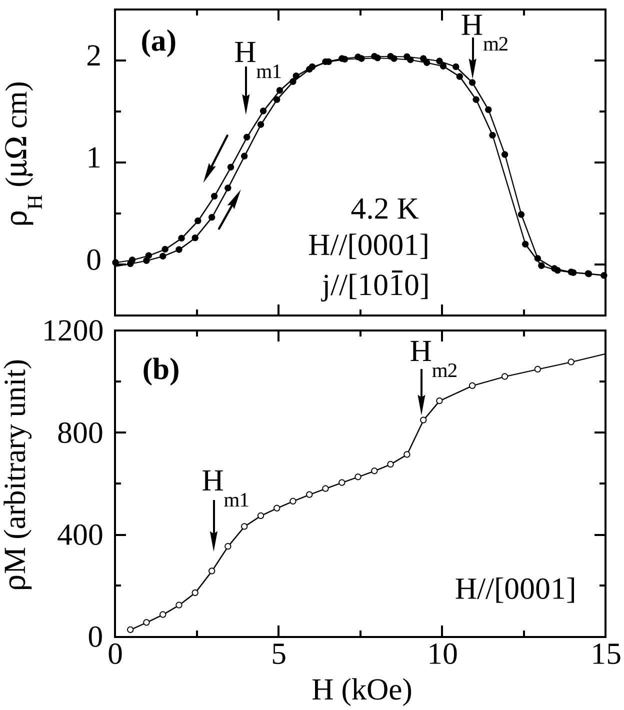

FIG. 6. (a) The field dependence of Hall resistivity in Gd_2PdSi_3 for H//[0001]. The arrows indicate the directions of the field sweep. (b) Field dependence of ρM for H//[0001].

Fig. 6a shows the field dependence of Hall resistivity (ρ_H) for H//[0001]. ρ_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 ρM is ploted in Fig. 6b. Theoretically, when anomalous Hall effect is dominant, ρ_H should vary linearly with ρM. In other words, ρ_H and ρM should vary in the same way with the corresponding applied fields. But in the present case, the field dependence of ρ_H completely differ from that of ρM, particularly around H_{m2}. This fact strongly indicates the modification of the Fermi surface across the metamagnetic anomaly.

Summarising, we have explored anisotropy in the transport and magnetic properties on the single crystal Gd_2PdSi_3. Possibly the anysotropic exchange interaction due to crystalline anisotropy and the anisotropy in the Fermi surface are responsible for the observed anysotropy. 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, $Gd_5(Si,Ge)_4$ (Ref. 15), which has been found to be a simultaneous crystallograpic 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 of Gd_2PdSi_3 exhibits interesting features. Above all, there is a well-defined ρ 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 a novel magnetic material.

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