Magnetic and transport behavior of single-crystalline Dy_2PdSi_3

Subham Majumdar,[1] H. Bitterlich,[2] G. Behr,[2] W. Löser,[2] P. L. Paulose,[1] and E.V. Sampathkumaran[1,*]
[1]*Tata Institute of Fundamental Research, Homi Bhabha Road, Colaba, Mumbai 400005, India*
[2]*Institut für Festkörper- and Werkstofforschung Dresden, Postfach 270016, D-01171 Dresden, Germany*
(Received 11 December 2000; published 15 June 2001)

The results of magnetization, magnetic susceptibility (χ), electrical measurements, and magnetoresistance measurements as a function of temperature (2–300 K) in the single-crystalline Dy_2PdSi_3 are reported for two different directions ([0001] and $[10\bar{1}0]$) of the crystal. The results reveal that there is an onset of magnetic ordering below 8 K. The high-temperature paramagnetic Curie temperature is positive for both directions, though antiferromagnetic coupling dominates in the basal plane; the magnetic coupling along the *c* axis is of a ferromagnetic type. The electrical resistance data for both the directions indicate the formation of magnetic Brillouin-zone boundary gaps. A finding of importance is that the broad peak in the temperature dependent χ around 50 K noted for the analogous Tb compound is not observed in the present investigations, though there is a weak difference between the zero-field-cooled and field-cooled χ versus *T* curves below 40 K (the implications of which are not clear at present). The results establish that this compound exhibits giant magnetoresistance behavior at low temperatures. We also compare the observed anisotropic behavior with those in Tb and Gd compounds.

DOI: 10.1103/PhysRevB.64.012418 PACS number(s): 75.50.−y, 75.20.En, 75.70.Pa, 72.15.−v

The compounds of the type R_2TSi_3 (R= rare earth, T =transition metals), crystallizing in a AlB_2-derived hexagonal structure, have been found to exhibit many anomalies in their properties.[1–14] We have also carried out extensive studies on the single crystals of Pd containing alloys for R=Ce, Gd, and Tb (Refs. 6–8) considering the importance of some observations on the polycrystalline samples to the fields of the Kondo effect, heavy fermions, the giant magnetoresistance (GMR) effect, the magnetocaloric effect, etc. We have observed a number of interesting anisotropic anomalies, even for R=Gd (which is an *S*-state ion). This prompted us to extend the studies on the single crystals to other members of this series as well in order to get a more complete picture of the anisotropic magnetic behavior of this class of compounds. Here, we present the results of magnetization (M), magnetic susceptibility (χ), and electrical and magnetoresistance studies for single-crystalline Dy_2PdSi_3. The results confirm the magnetic behavior observed for the polycrystalline sample reported earlier,[5] in particular establishing that this compound is a low-temperature GMR system. In addition, the data reveal that the magnetic properties are qualitatively more isotropic in the paramagnetic state as compared to those in the Tb compound.[8] The magnetic couplings inferred on the basis of the present data are consistent with the neutron-diffraction results on the polycrystalline samples.[13]

The single-crystalline Dy_2PdSi_3 was grown by the floating-zone method by rf inductive heating from stoichiometric polycrystalline feed rods. This material exhibits congruent melting behavior.[15] Accordingly,[16] the growth rate was 5 to 10 mm/h. The orientation of the single crystals was determined by the x-ray Laue backscattering method. Two oriented rods with respective rod axes parallel to the crystallographic directions [0001] and $[10\bar{1}0]$ were cut from this crystal with a diamond-coated wire saw. Isothermal magnetization at selected temperatures and magnetic susceptibility studies as a function of temperature (T) (2–300 K) were carried out at two different magnetic fields, 100 Oe and 2 kOe (with H parallel to the rod axis) employing a commercial vibrating sample magnetometer (VSM) (Oxford Instruments). The electrical resistivity (ρ) measurements were performed by a conventional four-probe method employing a conducting silver paint for making electrical contacts of the leads with the samples; the magnetoresistance (MR) was also measured as a function of the external field up to 70 kOe in the longitudinal mode ($I \| H$) at selected temperatures below 40 K.

The results of temperature-dependent χ behavior are shown in Figs. 1 and 2. While the data at H=2 kOe were collected for the zero-field-cooled (ZFC) condition of the sample, the data at 100 Oe were obtained both for the ZFC and field-cooled (FC) states. There are small steplike anomalies in the data around 60–80 K, always appearing with our magnetometer; a weak change in the slope in the plot of inverse χ versus T around 270 K has also been noted with our VSM instrument, but not confirmed by measurements with the superconducting quantum interference device (SQUID) magnetometer; therefore, these are instrumental artifacts. It is apparent from these figures that there is a distinct feature at 8 K, appearing as a kink or as a peak, indicating the onset of long-range magnetic order at this temperature. There are additional upturns in χ (Fig. 1) below 3 K (though weak for $H \| [10\bar{1}0]$) which could just be due to reorientations of magnetic moments. The existence of such transitions have been inferred from the data on polycrystals as well.[5] There is a small difference between ZFC and FC χ data below 8 K, unlike the Tb case, but similar to that in the Gd compound.[6,8,11] The observed behavior rules out spin-glass freezing in this compound, a phenomenon found in isostructural U_2PdSi_3.[17] While the observation of a well-defined peak in the plot of χ versus T around 8 K for $[10\bar{1}0]$ (Fig. 2) is typical of antiferromagnetic ordering, the absence of such a peak (but the tendency for saturation below the same tem-

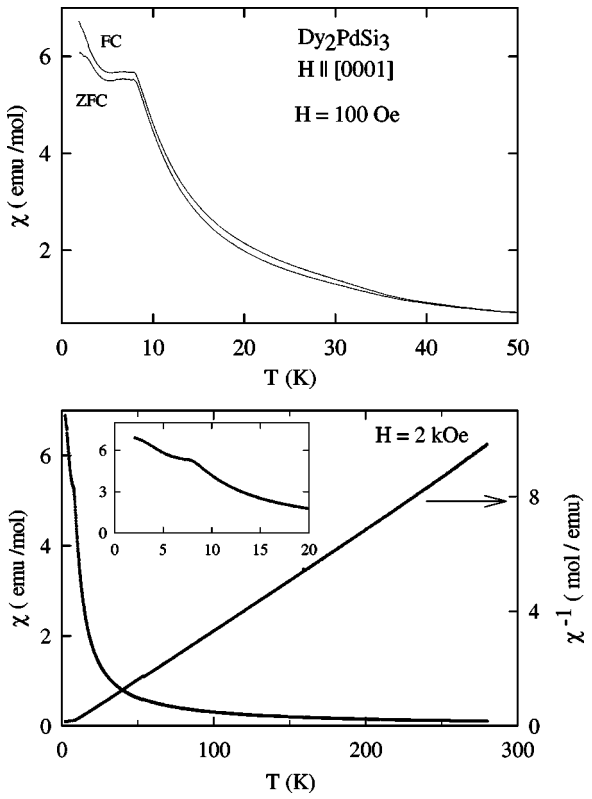

FIG. 1. Magnetic susceptibility (χ) as a function of temperature measured with $H=100$ Oe and 2 kOe for single-crystalline Dy_2PdSi_3 for the orientation H$\|$[0001] of the crystal. While the data with 2 kOe are recorded for the zero-field-cooled (ZFC) condition of the specimen, the low-field data were obtained also for the field-cooled (FC) state. The inverse χ for the 2 kOe data is also shown. The inset shows the low-temperature data in an expanded form.

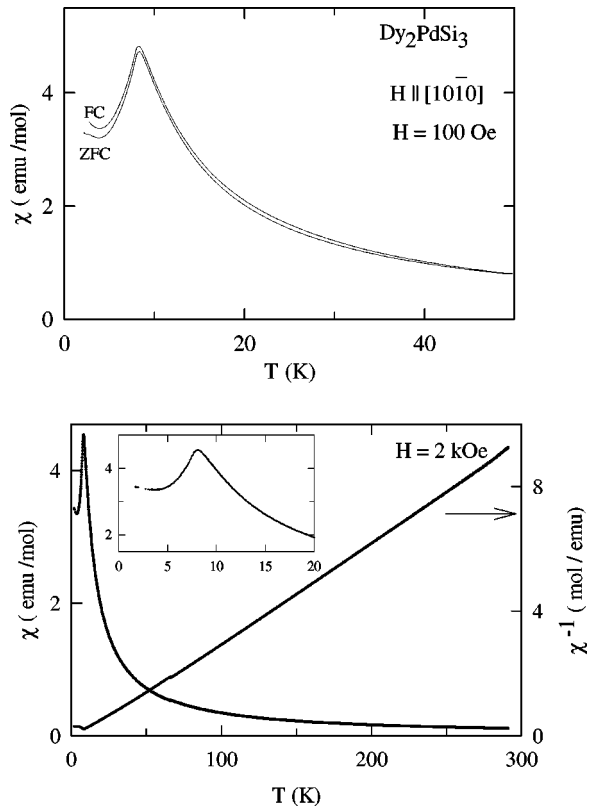

FIG. 2. Magnetic susceptibility (χ) as a function of temperature measured with $H=100$ Oe and 2 kOe for single-crystalline Dy_2PdSi_3 for the orientation H$\|$[10$\bar{1}$0] of the crystal. While the data with 2 kOe are recorded for the zero-field-cooled (ZFC) condition of the specimen, the low-field data were obtained also for the field-cooled (FC) state. The inverse χ for the 2 kOe data is also shown. The inset shows the low temperature data in an expanded form.

perature for the data at 100 Oe), combined with the M behavior (see below) for [0001] seems to suggest ferromagneticlike coupling at this temperature along this direction. With respect to the paramagnetic behavior, the plot of inverse χ versus T is linear above 100 K for both the rods with the value of the effective moment (10.5 and 11 μ_B/Dy ion for [0001] and [10$\bar{1}$0], respectively) very close to that expected for trivalent Dy ions; there is a weak continuous change in the slope of this plot with decreasing temperature below 100 K, presumably due to crystal-field effects. The sign and the magnitude of the paramagnetic Curie temperature (θ_p) above 100 K are the same (11 K) for the Dy compound for both the directions similar to that of Gd compound[6]—however, they are different from that in Tb compound.[8] The positive sign of θ_p indicates dominance of ferromagnetic correlations in the paramagnetic state for both the directions at low temperatures, though interestingly the net magnetic structure below 8 K in the basal plane appears to be antiferromagneticlike. The θ_p at low temperatures, that is for the crystal-field-split ground state, approaches 0 K, thereby signaling the existence of a competition between antiferro- and ferromagnetic interactions as the T is lowered, though it does not result in spin-glass freezing. Finally, it is important to note that the broad peak appearing in the range 40–50 K for the single-crystalline Tb compound[8] for [0001] is not observed in the present measurements; however, there appears to be a weak difference between the ZFC and FC curves below about 40 K (see Figs. 1 and 2); this observation is puzzling.

The isothermal behavior (Figs. 3 and 4), in comparison with the trends in Gd and Tb compounds, is also quite revealing. While dramatic anisotropy in M was noted in the latter compounds as elaborated in Refs. 6 and 8, qualitatively speaking, there are no such major differences in the plots of M versus H for these two directions in the Dy case. However, a careful look at the data below 10 kOe at 2 and 5 K brings out a subtle difference. At 2 K, there is an initial steep rise (below about 5 kOe) for both the directions. However, for [10$\bar{1}$0] there is a sluggish variation in the range 5–30 kOe, followed by a tendency for saturation at higher fields, in contrast to the continuous sluggish variation for [0001] in the field range 10–70 kOe. At 5 K, for [10$\bar{1}$0], there is an upward deviation in the field range 5–10 kOe from the low-field linear variation, as if there is a weak-field-induced spin-flip transition, a finding in support of antiferromagnetic ordering proposed above for this direction; this feature is absent for the [0001] direction for the 5 K data in

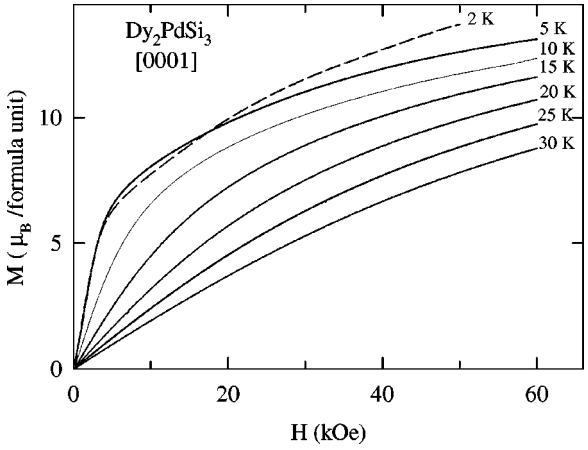

FIG. 3. Isothermal magnetization behavior for the single-crystalline Dy_2PdSi_3 for the crystal orientation $H\|[0001]$.

which case M increases steeply with H below 5 kOe followed by a sluggish variation at higher fields; this behavior is indicative of ferromagneticlike coupling for this direction at this temperature. It may be recalled[6,8] that, in both Gd and Tb compounds, antiferromagnetic coupling dominates along the [0001] direction, in contrast to that in the Dy compound. At other temperatures, otherwise, M increases steadily and monotonically with H. Finally, the fact that M versus H plots differ slightly in shape at 2 and 5 K imply that there are slight changes in the magnetic structure as the T is lowered in the magnetically ordered state. As pointed out earlier, this may be supported by the existence of upturns below 3 K in the plots of χ versus T (Figs. 1 and 2).

In Fig. 5 we show the T dependence of ρ. As expected, one observes a positive temperature coefficient in the slope around 40 K, which may be related to the anomalous difference in ZFC and FC χ curves below 40 K (Fig. 1). Apart from this, there is an upturn at the onset of magnetic ordering as the T is lowered through the magnetic transition temperature, attributable to the formation of magnetic Brillioun-zone boundary gaps. In this respect, while this property is comparable to that of the Gd compound,[6] it is distinctly different

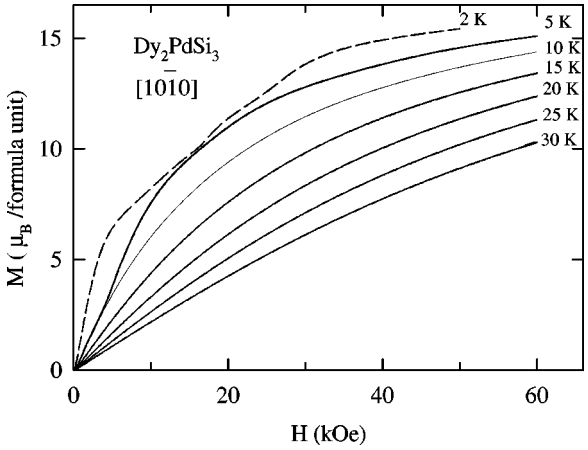

FIG. 4. Isothermal magnetization behavior for the single-crystalline Dy_2PdSi_3 for the crystal orientation $H\|[10\bar{1}0]$.

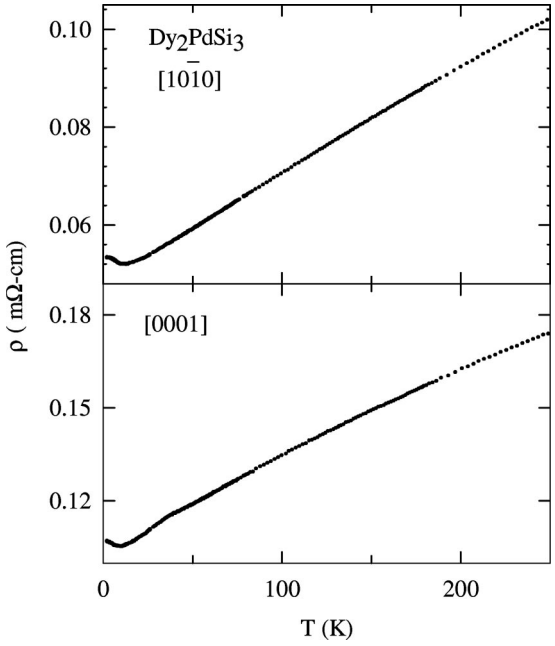

FIG. 5. Temperature-dependent electrical resistivity behavior for the single-crystalline Dy_2PdSi_3 for two different orientations of the sample, $I\|[0001]$ and $I\|[10\bar{1}0]$.

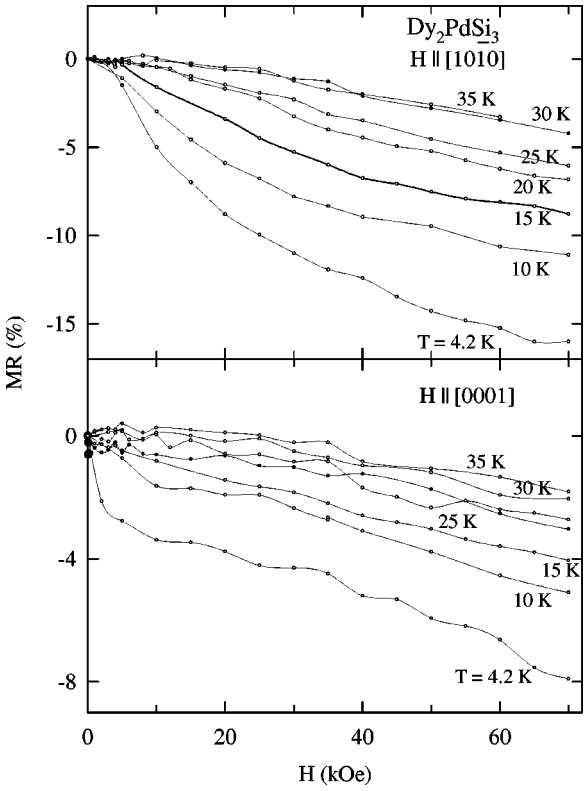

FIG. 6. Magnetoresistance (MR) as a function of an externally applied magnetic field at selected temperatures below 40 K in the longitudinal mode for two different directions of the crystals. The error bar in the values of MR is $\pm 0.5\%$. The lines drawn through the data points serve as guides to the eyes.

from that of the Tb compound.[8] It is particularly notable that this feature appears even for the [0001] direction, along which ferromagnetic coupling dominates, which implies that the magnetic structure along the *c* axis is of an incommensurate type. The application of a magnetic field should tend to diminish this gap effect, which should result in large magnetoresistance values. According to this expectation, MR, defined as $[\rho(H)\text{-}\rho(0)]/\rho(0)$, is found to be of a negative sign and large in magnitude in the magnetically ordered state, attaining as much as -17% for $H=60$ kOe for $[10\bar{1}0]$. (See Fig. 6.) Though the values for [0001] are relatively less, these are still larger than what one observes in simple metals. It is to be noted that the negative MR with a large magnitude persists even well above magnetic transition temperature, say, for instance at 40 K for $H=60$ kOe, consistent with the findings on polycrystals.[5] Some differences among the shapes of MR versus *H* plots below 10 K for these two directions have been noted, which is not unexpected considering differences in the magnetic couplings discussed above.

To conclude, the magnetic and transport behavior for the single-crystalline form of the compound Dy_2PdSi_3 have been reported. The results, while confirming the onset of magnetic ordering at 8 K and a large low-temperature magnetoresistance response reported for polycrystals, reveal that there is a dominant ferromagnetic coupling along the *c* axis whereas antiferromagnetic coupling prevails along the basal plane in contrast to that observed in the Tb compound. We have also brought out similarities and differences between the anisotropic properties among Gd, Tb, and Dy compounds. We would like to stress, in particular, that the broad peak appearing in the range 40–50 K for the single-crystalline Tb compound[8] for [0001] is not observed for the Dy case in the present studies; however, ZFC and FC χ curves differ for the Dy case below 40 K; the implications of which are not clear at present.

This work in Germany was supported by the Deutsche Forschungsgemeinschaft within the SFB463. One of us (E.V.S.) would like to thank Professor C. Laubschat for an invitation to Dresden.

*Email address: sampath@tifr.res.in

[1] P.A. Kotsanidis, J.K. Yakinthos, and E. Gamari-seale, J. Magn. Magn. Mater. **87**, 199 (1990); R.A. Gordon, C.J. Warren, M.G. Alexander, F.J. DiSalvo, and R. Pöttgen, J. Alloys Compd. **248**, 24 (1997) and references therein.

[2] R. Mallik and E.V. Sampathkumaran, J. Magn. Magn. Mater. **164**, L13 (1996).

[3] R. Mallik, E.V. Sampathkumaran, M. Strecker, and G. Wortmann, Europhys. Lett. **41**, 315 (1998).

[4] R. Mallik, E.V. Sampathkumaran, M. Strecker, G. Wortmann, P.L. Paulose, and Y. Ueda, J. Magn. Magn. Mater. **185**, L135 (1998).

[5] R. Mallik, E.V. Sampathkumaran, and P.L. Paulose, Solid State Commun. **106**, 169 (1998).

[6] S.R. Saha, H. Sugawara, T.D. Matsuda, H. Sato, R. Mallik, and E.V. Sampathkumaran, Phys. Rev. B **60**, 12 162 (1999).

[7] S.R. Saha, H. Sugawara, T.D. Matsuda, Y. Aoki, H. Sato, and E.V. Sampathkumaran, Phys. Rev. B **62**, 425 (2000).

[8] Subham Majumdar, E.V. Sampathkumarancc, P.L. Paulose, H. Bitterlich, W. Löser, and Gunter Behr, Phys. Rev. B **62**, 14 207 (2000).

[9] Subham Majumdar, R. Mallik, E.V. Sampathkumaran, P.L. Paulose, and K.V. Gopalakrishnan, Phys. Rev. B **59**, 4244 (1999).

[10] E.V. Sampathkumaran, I. Das, R. Rawat, and Subham Majumdar, Appl. Phys. Lett. **77**, 418 (2000).

[11] Subham Majumdar, E.V. Sampathkumaran, D. Eckert, A. Handstein, K.-H. Mueller, S.R. Saha, H. Sugawara, and H. Sato, J. Phys.: Condens. Matter **11**, L329 (1999).

[12] R. Mallik, E.V. Sampathkumaran, P.L. Paulose, H. Sugawara, and H. Sato, Pramana, J. Phys. **51**, 505 (1998).

[13] A. Szytula, M. Hofmann, B. Penc, M. Slaski, S. Majumdar, E.V. Sampathkumaran, and A. Zygmunt, J. Magn. Magn. Mater. **202**, 365 (1999).

[14] Subham Majumdar and E.V. Sampathkumaran, Phys. Rev. B **61**, 43 (2000) and references therein.

[15] G. Behr, W. Löser, G. Graw, H. Bitterlich, J. Fink, and L. Schultz, Cryst. Res. Technol. **35**, 461 (2000).

[16] G. Graw, H. Bitterlich, W. Löser, G. Behr, J. Fink, and L. Schultz, J. Alloys Compd. **308**, 193 (2000).

[17] D.X. Li, Y. Shiokawa, Y. Homma, A. Uesawa, A. Dönni, T. Suzuki, Y. Haga, E. Yamamoto, T. Honma, and Y. Onuki, Phys. Rev. B **57**, 7434 (1998).