Magnetic behavior of single-crystal Ho_2PdSi_3

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The magnetic behavior of single-crystal Ho_2PdSi_3, crystallizing in an AlB_2-derived hexagonal structure, is investigated by magnetic susceptibility (χ) and electrical resistivity (ρ) measurements along two directions. There is no dramatic anisotropy in the high-temperature Curie-Weiss parameter or in the ρ and isothermal magnetization data, though there is a noticeable anisotropy in the magnitude of ρ between two perpendicular orientations. The degree of anisotropy is overall less prominent than in the Gd (which is an *S*-state ion) and Tb analogs. A point of emphasis is that this compound undergoes long-range magnetic ordering below 8 K as in the case of analogous Gd and Dy compounds. Considering this fact for these compounds with a well-localized *f* orbital, the spin-glass freezing noted for isomorphous U compounds in the recent literature could be attributed to the role of the *f*-ligand hybridization, rather than just Pd-Si disorder.

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The investigation of intermetallic compounds of the general formula R_2TX_3 (R=rare earth or U, T= transition metal, and X=Si or Ge), derived from AlB_2-type hexagonal structure,[1] is of great interest, considering that the properties of many of these compounds are relevant to current problems in the field of magnetism and superconductivity (see, for instance, Refs. 1–12). We have also carried out intense studies on the single-crystalline forms of the series R_2PdSi_3 for R=Ce, Gd, and Tb and found novel anisotropic behavior in the magnetic and transport data.[5–7] It was, however, noticed that such anisotropy is less evident in the case of Dy,[8] and this finding is surprising considering that even the Gd (which is an *S*-state ion) compound exhibits strongly anisotropic properties. In view of this situation, we considered it worthwhile to extend single-crystal investigations to other members of this series. Therefore, we have investigated single-crystalline Ho_2PdSi_3, the results of which are reported in this article. Another motivation to take up this investigation is that the analogous U compounds have been found[12] to undergo spin-glass freezing, whereas previous reports[1,5,8,10,11] for the above heavy rare-earth compounds reveal features due to long-range magnetic ordering. It is therefore of interest to carefully investigate single crystals of additional *f*-localized systems, in order to explore possible role of *f* hybridization to decide the spin-glass ground state in U compounds.

The single crystals of Ho_2PdSi_3 (25–50 mm in length and 7 mm in diameter) were grown by the floating zone method by rf inductive heating from stoichiometric polycrystalline feed rods. The orientation of the single crystals was determined by x-ray Laue backscattering. Pieces (2 mm $\times$2 mm$\times$10 mm) with two orientations $[10\bar{1}0]$ and [0001] were cut from this rod for measurements. Isothermal magnetization (M) measurements at 2 and 5 K and magnetic susceptibility (χ) measurements in the temperature (T) interval 1.8–300 K were performed employing a commercial superconducting quantum interference device (Quantum Design) as well as a vibrating sample magnetometer (Oxford Instruments). The electrical resistivity (ρ) measurements were performed by a conventional four-probe method employing silver paint for electrical contacts.

The T dependence of χ at low temperatures taken at 100 Oe and 2 kOe for the zero-field-cooled (ZFC) condition of the crystal is shown in Fig. 1. For H=100 Oe, we have in addition taken the data for the field-cooled (FC) state of the specimen. It is distinctly clear from this figure that, for $H\|[0001]$, there is a well-defined peak at 8 K, establishing the onset of magnetic ordering. The transition temperature is higher than that reported (6 K) for polycrystals.[13] It should be noted that there is no separation of ZFC-FC curves as in

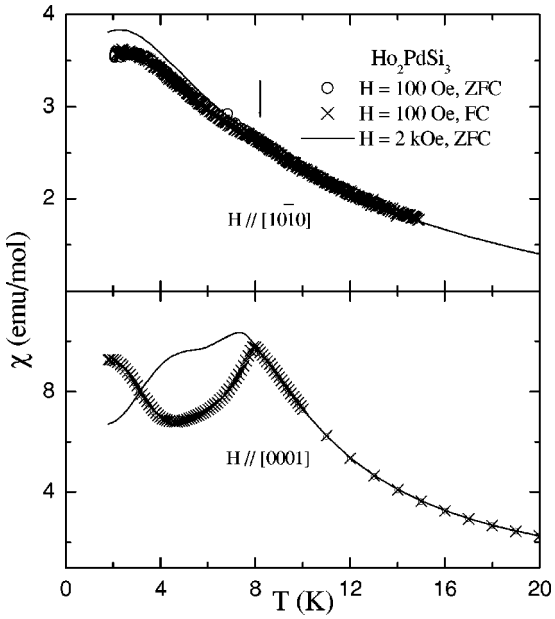

FIG. 1. The low-temperature magnetic susceptibility behavior of Ho_2PdSi_3 for two crystallographic orientations measured in magnetic fields of 100 Oe and 2 kOe. ZFC and FC represent zero-field-cooled and field-cooled state of the specimens to 1.8 K, and the curves corresponding to these two situations overlap. The vertical arrow shows the onset of magnetic ordering appearing as a shoulder in the H=100 Oe data.

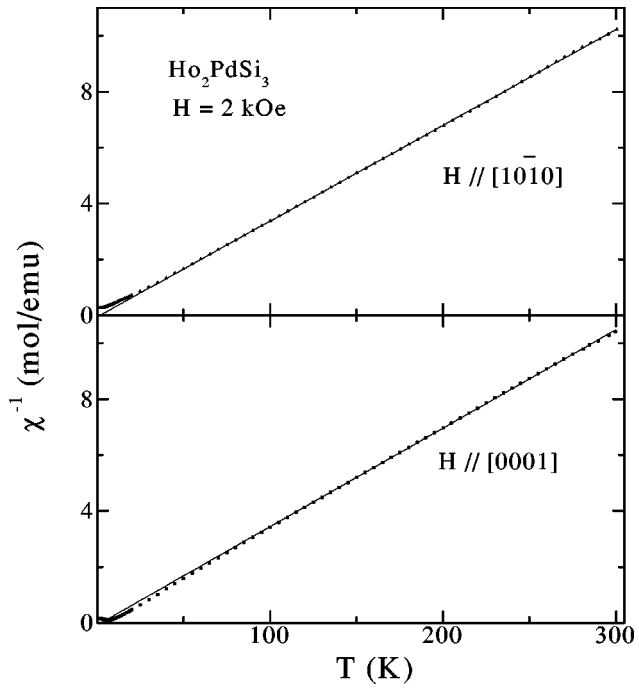

FIG. 2. Inverse susceptibility as a function of temperature for two crystallographic orientations of Ho_2PdSi_3 in a magnetic field of 2 kOe. The lines represent least-squares fitting of the data above 100 K.

the case of single crystals of Gd, Tb, and Dy alloys,[5,7,8] which is a sufficient proof that this compound undergoes long-range magnetic ordering, rather than spin-glass freezing. As T is lowered, there is an upturn in χ values below 4 K, which is indicative of possible spin reorientation effects apparently characteristic of this class of compounds.[5,7,8] If χ data are collected at $H=2$ kOe, this upturn vanishes; however, a broad shoulder appears around 4 K in the plot of χ versus T as though there is a change in the magnetic structure with decreasing T. Now turning to the data for $H\|[10\bar{1}0]$, the 8 K transition appears only as a broad shoulder in the plot of χ versus T; however, the transition below 4 K appears as a prominent feature—that is, a tendency to flatten for both fields. The χ values are noticeably lower for this direction compared to those for $H\|[0001]$. In short, χ behavior appears to be anisotropic in the magnetically ordered region.

With respect to the paramagnetic behavior (Fig. 2), the plot of inverse χ versus T is nearly linear, though there is a tendency for a small deviation below about 50 K presumably due to crystal field effects. The nature of the deviation is different for these orientations (see Fig. 2) in the sense that the experimental values at low temperatures in the paramagnetic state (say, in the range 10–30 K) are higher for $H\|[10\bar{1}0]$ with respect to the straight line extrapolated from the high-temperature (>100 K) linear region, whereas for $H\|[0001]$ the values are lower. The value of the effective moment obtained from the high-temperature linear region is very close to that expected for trivalent Ho ions (close to $10.6\mu_B$/Ho mol) and the Curie-Weiss temperature (θ_p) is practically the same for both directions (close to 2 K). Thus, the anisotropy is insignificant in the high-temperature χ data.

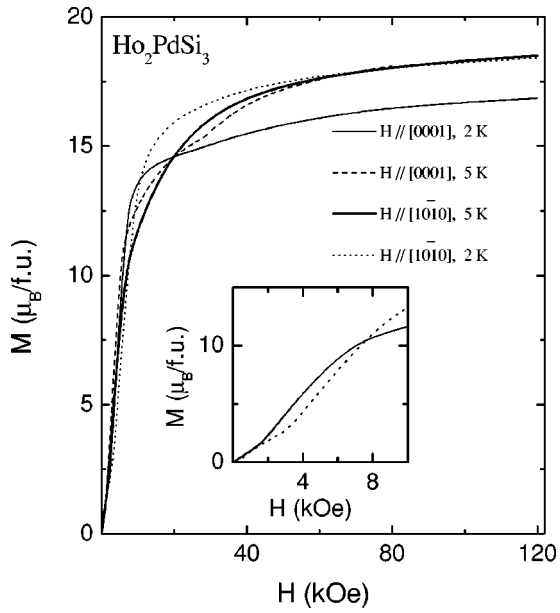

FIG. 3. Isothermal magnetization data at 2 and 5 K for two crystallographic orientations of Ho_2PdSi_3. Low-field behavior is shown in the inset to highlight the existence of a metamagneticlike transition.

It is to be remarked that the absolute value of θ_p is nearly the same as the Néel temperature (T_N) for $H\|[0001]$, as inferred from the linear region below 30 K; however the sign of θ_p is positive, implying the existence of significant ferromagnetic correlations. The low temperature θ_p, however, changes sign (to -3 K) for $H\|[10\bar{1}0]$ consistent with antiferromagnetic coupling.

The isothermal M behavior (Fig. 3) shows no hysteresis at 2 or 5 K. A close inspection of the data at 5 K suggests that

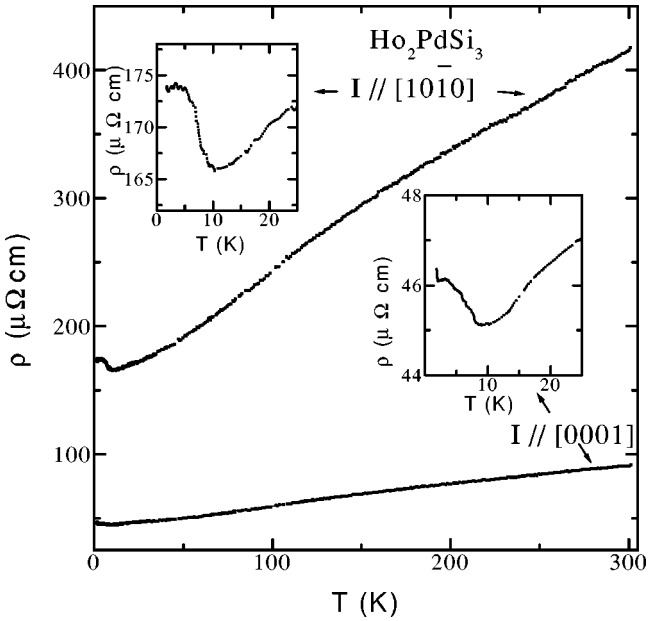

FIG. 4. Electrical resistivity as a function of temperature for Ho_2PdSi_3 for two different crystallographic directions. The low-temperature behavior is shown in expanded form as insets to highlight the existence of an upturn.

there is a sudden increase in the slope in a field of about 2 kOe, as though there is a spin reorientation, for both directions; the field at which this occurs increases to about 3 kOe at 2 K as shown in an expanded form in the inset for one direction. This finding suggests that the zero-field magnetic ordering is antiferromagnetic. The magnetic moment at high fields tends towards saturation to a constant value, which is indicative of field-induced ferromagnetism. The value of the saturation moment is somewhat lower than that expected for trivalent Ho ions ($10\mu_B$/Ho ion) and it appears that higher fields are required to attain this saturation value. It is to be noted that, for the 2 K data for $H\|[0001]$, the values at high fields are comparatively lower than those at 5 K, which may be a consequence of the change in spin orientation as T is lowered. Needless to say, isothermal M in the paramagnetic state is a linear function of H (not shown in the figure).

In Fig. 4, we show the T dependence of ρ. The low-T data have also been shown in an expanded form in the insets. Above T_N, ρ for both crystallographic directions of excitation current has a positive temperature coefficient typical of metals (as expected). Though the values for the two directions are different, the features are qualitatively the same, even below T_N. The value of ρ, instead of exhibiting a drop at T_N due to the loss of the spin-disorder contribution, shows an upturn. This observation signals the formation of magnetic Brillouin-zone boundary gaps below 8 K, thereby endorsing the previous conclusion from neutron diffraction data[3] that the magnetic structure is of a modulated type (incommensurate with the lattice).

To conclude, we have reported the magnetic behavior of single-crystalline Ho_2PdSi_3 for two orientations. Among various findings, we would like to emphasize that the overall features in isothermal M, temperature dependence of ρ, and paramagnetic χ behavior are essentially isotropic, though the shapes of the χ versus T plots in the magnetically ordered state appear to show some anisotropy and the magnitude of ρ is noticeably different for two perpendicular orientations. In this sense, the properties of this compound are similar to that of Dy_2PdSi_3.[8] In sharp contrast to this situation, the features in isothermal M and ρ near the magnetic region are strongly anisotropic not only in the Tb analog but also for the Gd (which is an S-state ion) analog.[5,7] Finally, with the observation of long-range magnetic ordering in another heavy rare-earth compound of this structure in single-crystalline form, it can be stated that the spin-glass behavior of the isostructural U compounds is determined in some fashion by f hybridization (endorsing the conclusion in Ref. 12), rather than Pd-Si disorder alone, with the assumption that the degree of possible Pd-Si site disorder is the same for U as well as for the heavy rare-earth members of this series with long-range magnetic order.

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