Observation of enhanced magnetic transition temperature in Nd_2PdGe_3 and superconductivity in Y_2PdGe_3

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The results of electrical resistance (1.4–300 K), magnetization (2–300 K) and heat-capacity (2–50 K) measurements in Nd_2PdGe_3 and Y_2PdGe_3, found to crystallize in an AlB_2-derived hexagonal structure, are reported. The results establish that there is an onset of magnetic ordering (of a ferromagnetic type) for the Nd compound at a temperature close to 6 K and this temperature is considerably enhanced compared to the de Gennes scaled value. The Y compound is found to be superconducting below 3 K and this compound turns out to be a superconductor among the ternary members derived from the AlB_2 structure. The identification of an AlB_2-derived superconductor not containing B may contribute to the overall understanding of the physics of recently discovered high-temperature superconductor, MgB_2.

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During the last few years, we have been actively involved in synthesizing new compounds of the type R_2XY_3, with crystal structures derived from that of AlB_2.[1] We noticed several interesting magnetic anomalies,[2–12] in particular relevant to the fields of Kondo lattices and heavy fermions,[2,3] non-Fermi liquids,[4] colossal magnetoresistance,[2,3,5] magnetic refrigeration,[6] and possibly low-dimensional magnetism.[7,8] Some of the compounds of this type have also been reported to belong to the class of ''stoichiometric compounds exhibiting spin-glass ordering,'' possibly due to intrinsic crystallographic disorder.[13,14] It is thus evident that, considering a variety of interesting magnetic and crystallographic[1] features exhibited by these compounds, this class of ternary compounds will attract as much attention as the $BaAl_4$-derived tetragonal structures; it is, however, to be noted that none of the present family of compounds has been reported to be superconducting. It is thus of interest to synthesize new compounds with this stoichiometry and to probe their magnetic and possible superconducting characteristics. Here, we report the formation of the compounds Nd_2PdGe_3 and Y_2PdGe_3 in an AlB_2-derived structure. It may be remarked that the analogous Gd alloy has been reported earlier and found to order antiferromagnetically below 10 K;[11] our attempts to synthesize other members of this series with R beyond Gd have not been successful; among the light rare-earth compounds, La, Ce, and Pr appear to form in an orthorhombic structure, the properties of which will be investigated after proper structural characterization. While the present results establish that the Nd compound exhibits magnetic ordering of a ferromagnetic type below 6 K, the Y compound is identified to be a superconductor among this family of ternary compounds (that is, with 2:1:3 stoichiometry) with a superconducting transition temperature (T_c) of 3 K.

The samples were prepared by arc melting stoichiometric amounts of constituent elements in an inert atmosphere. The ingots were homogenized in evacuated, sealed quartz tubes at 850°C. X-ray diffraction patterns were obtained employing Cu K_α radiation and the patterns (Fig. 1) confirm that the compounds crystallize in an AlB_2-derived hexagonal structure. For the Y compound, there is an additional weak line (marked by asterisk in Fig. 1), which we believe is due to a parasitic phase with the stoichiometry 1:2:2. We do not find any superstructure line in the x-ray-diffraction patterns within the detection limit and hence we believe that there is no doubling of unit-cell parameters unlike in some other isostructural compounds.[1,12] The electrical resistivity (ρ) measurements (T=1.4–300 K) were performed by a conventional four-probe method employing a silver paint for making electrical contacts. For the Nd sample, the magnetic susceptibility (χ) measurements (2–300 K) were performed employing a commercial superconducting quantum interfer-

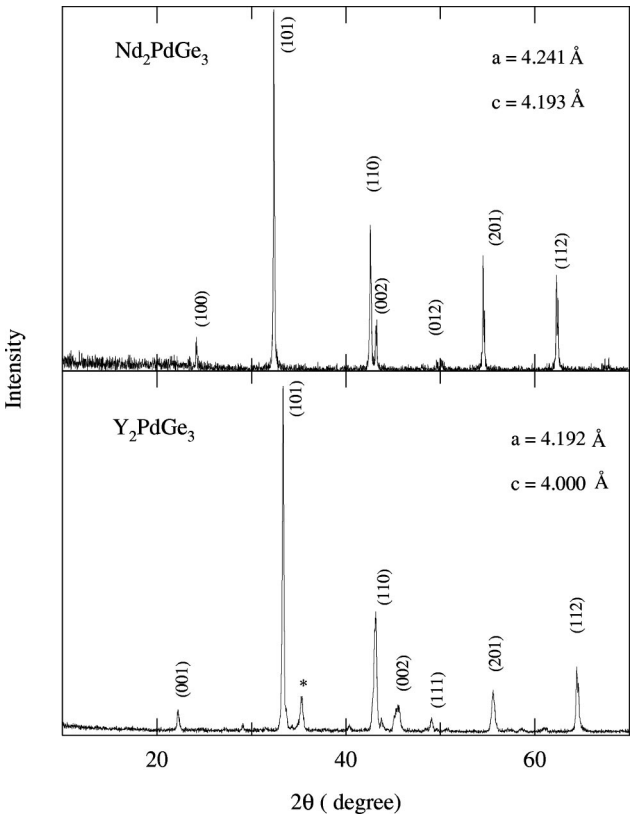

FIG. 1. X-ray-diffraction pattern (Cu K_α) for Nd_2PdGe_3 and Y_2PdGe_3. The unindexed line is marked by an asterisk for Y_2PdSi_3.

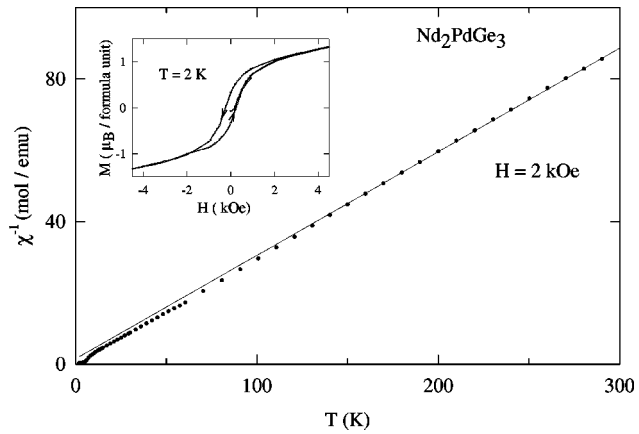

FIG. 2. Temperature-dependent magnetic susceptibility behavior for Nd_2PdGe_3 measured in the presence of $H=2$ kOe for the zero-field-cooled (ZFC) state of the specimen, plotted in the form of inverse χ vs T; the inset shows hysteresis loop at 2 K.

ence device magnetometer (Quantum Design) in the presence of a magnetic-field (H) of 2 kOe; in addition, the low-temperature data for the field-cooled (FC) and zero-field-cooled (ZFC) states of the specimen were taken in an H of 100 Oe; the magnetic hysteresis loop was obtained at 2 K; the heat-capacity (C) data (2–60 K) were obtained by a semiadiabatic heat-pulse method; the magnetoresistance ($MR=[\rho(H)-\rho(0)]/\rho(0)$) behavior in the longitudinal mode was also tracked at selected temperatures in the vicinity of magnetic transition temperatures, by the measurement of ρ as a function of H. For the Y sample, FC and ZFC χ behavior ($H=25$ Oe) below 10 K, the ρ (1.4–300 K) and the C data (2.5–30 K) were obtained.

The results of magnetization, C, ρ, and MR measurements are shown in Figs. 2–4 for Nd_2PdGe_3. The plot of inverse χ versus T is found to be linear above 100 K and the effective moment (μ_{eff}) obtained from the linear region is found to be close to 3.7 μ_{eff}/Nd ion, which is consistent with the trivalent nature of Nd ions. The value of the paramagnetic Curie temperature (Θ_p) obtained from this linear region is found to be -5.4 K. There is a marginal deviation from linearity below 100 K, which may arise from crystal-field effects; if one extends the linear fit to a wider temperature range (say, in the range 10–300 K) ignoring this marginal deviation, the value of Θ_p comes closer to that of low temperature (<20 K) Θ_p(=zero Kelvin). A careful look at the low temperature data reveals that the compound may be classified as a ferromagnet. For instance, the isothermal M behavior at 2 K is hysteretic (see Fig. 2, inset), but with a small coercive field (close to 200 Oe). However, considering that the sign of Θ_p tends to be negative, we infer that there is a competition from antiferromagnetic correlations at least in the paramagnetic state. The existence of magnetic ordering at 6 K is also confirmed by a well-defined upturn (just below 6 K) in C and a drop in ρ due to loss of spin-disorder contribution (Fig. 3). There are also qualitative changes in the plots of MR vs H (Fig. 4) as the temperature is varied; while the sign of MR is negative throughout the wide range of H below 30 K varying quadratically with H in the para-

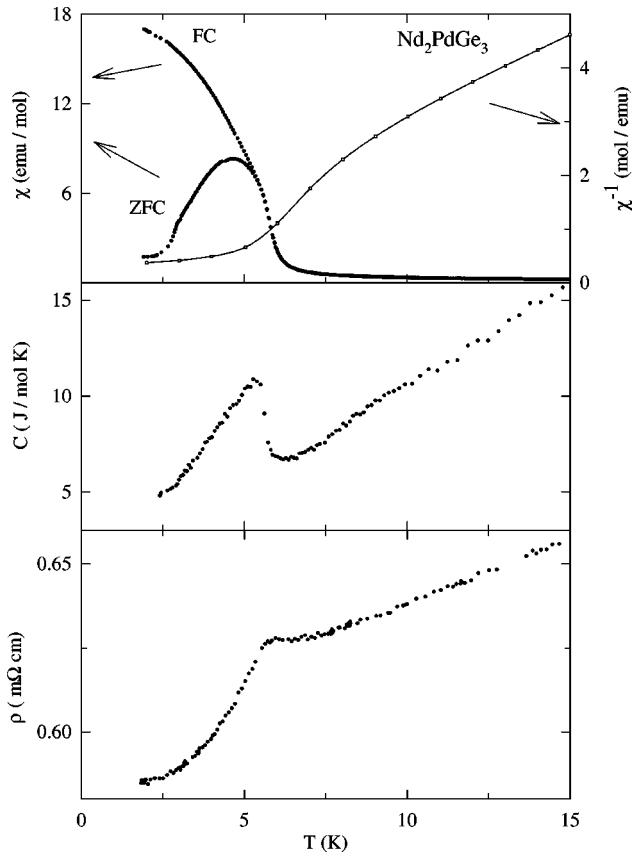

FIG. 3. The low-temperature (below 15 K) inverse χ ($H=2$ kOe), heat-capacity (C) and electrical resistivity (ρ) behavior for Nd_2PdGe_3 to highlight the features due to magnetic ordering, along with the χ data measured in the presence of $H=100$ Oe for the field-cooled (FC) and ZFC states of the specimen. The lines through the data points for the inverse χ data serve as a guide to the eyes.

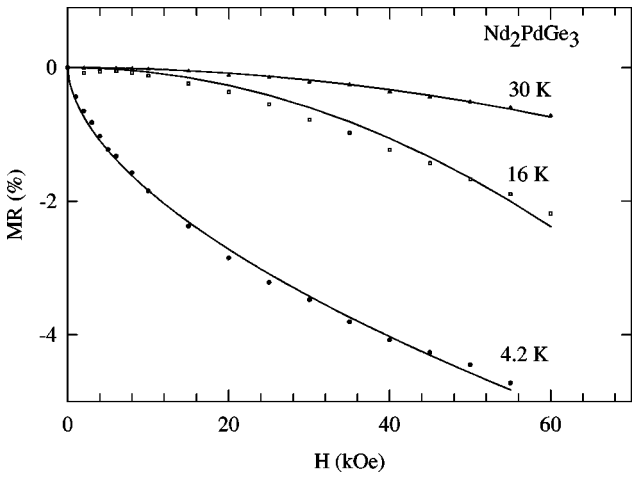

FIG. 4. Magnetoresistance, defined as $MR=[\rho(H)-\rho(0)]/\rho(0)$, at selected temperatures for Nd_2PdGe_3. The continuous lines represent a fit to the power-law dependence and the exponent is 2 for 16 and 30 K and 0.56 for 4.2 K data, within the limits of experimental error ($\pm 0.1\%$).

magnetic state (for instance, at 16 and 30 K) expected for dominance of paramagnetic fluctuations, there is a sharp decrease for initial applications of H at 4.2 K (with MR varying with a reduced power-law behavior, e.g., $H^{0.56}$ dependence), a behavior commonly observed in ferromagnets. Finally, it may also be remarked that the FC and ZFC χ data deviate at the magnetic onset temperature; such a behavior is not necessarily a characteristic of spin glasses and has been known even in systems with long-range magnetic ordering.[15]

Now, turning to the properties of Y_2PdGe_3, there is a normal metallic behavior of ρ with the variation of T; however, at 3 K, there is a sharp drop of ρ (Fig. 5) to zero (within the accuracy of nanovoltmeter employed to measure the voltage drop across the leads) below 3 K, as if this compound is superconducting below ($T_c =$) 3 K. To support this finding, the χ has also been measured at low fields (25 Oe) and we find that there is an onset of strong diamagnetism below 3 K and the divergence of ZFC and FC χ is typical of that expected for type-II superconductors. A comparison of the magnitude of the value of ZFC-χ at 2 K with that of other known standard superconductors like Pb establishes bulk nature of superconductivity. The plot of M versus H (see Fig. 5, inset) at 1.7 K is typical of that of type-II superconductors with a value of lower critical field of about 400 Oe. In order to render further support to the bulk nature of superconductivity, C was also measured down to 2.5 K and the upturn observed in the data obtained below 3 K (see Fig. 5, bottom inset) is sufficient to establish bulk superconductivity; the broadened signal around the transition could possibly be due to inhomogeneities. The values of the Debye temperature and the electronic term (γ) inferred from the data in the range 5–15 K turn to be about 130 K and 2.5 mJ/mol K^2, respectively. If one employs this value of γ, the value of $\Delta C/\gamma T_c$ turns to be close to 3, which is far away from the weak-coupling value of 1.35.

To conclude, we synthesized two compounds, Nd_2PdGe_3 and Y_2PdGe_3, and studied their physical characteristics. While the Nd compound undergoes long-range magnetic ordering below 6 K, the magnitude of the ordering temperature is far beyond that expected on the de Gennes scaling; that is, considering that the Néel temperature of Gd_2PdGe_3 is 10 K,[11] the corresponding value for the Nd compound should have been close to 1 K. In this sense, this Nd compound exhibits anomalous magnetism, presumably due to strong anisotropic $4f$ hybridization.[16] Finally, the compound

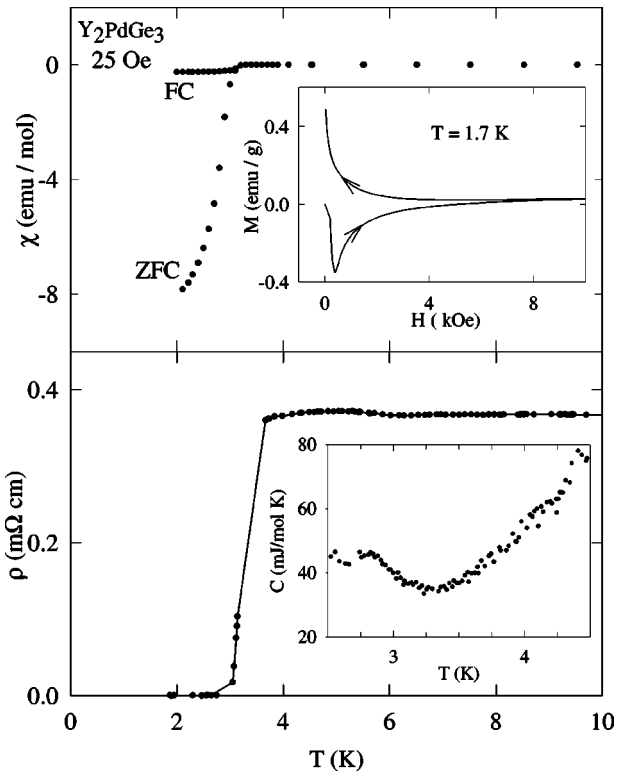

FIG. 5. Electrical resistivity and magnetic susceptibility ($H = 25$ Oe) behavior at low temperatures for Y_2PdGe_3 to highlight the features due to superconducting transition. The top inset shows the field dependence of M at 1.7 K for increasing and then decreasing field for the ZFC state of the specimen and the bottom inset shows the plot of heat capacity vs temperature to highlight the feature around superconducting transition. The line through the ρ data serves as a guide to the eyes.

Y_2PdGe_3 has been identified to be the lone superconductor to date in this family of ternary rare-earth compounds.

Note added. Finally, with respect to the recent excitements on MgB_2, it is worthwhile to note that superconductivity is scarce even among binary AlB_2-type rare-earth intermetallic compounds. There are also currently some proposals that B plays a crucial role to bring about superconductivity in this compound. In light of these, the observation of superconductivity in a AlB_2-type compound without B, viz. Y_2PdGe_3, gains importance. See Ref. 17 for details.

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