Pressure effects on the Kondo-lattice compound Ce$_2$Pd$_3$Si$_5$

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The influence of external pressure on the electrical-resistivity behavior of Ce$_2$Pd$_3$Si$_5$, recently reported to order antiferromagnetically below ($T_N$) $8.2$ K, has been investigated in the temperature interval $2.5$–$300$ K employing a cubic anvil pressure cell up to $8$ GPa. We find that $T_N$ remains nearly constant below $4$ GPa, beyond which there is a sharp decrease, which establishes that this compound lies at the peak of Doniach’s magnetic phase diagram. Apparently this compound behaves above $2.3$ K like a nonmagnetic Kondo lattice at $8$ GPa with a distinct feature attributable to Kondo coherence effects.

I. INTRODUCTION

The investigation of ternary rare-earth compounds containing Ce remains unabated due to the constant discovery of exotic phenomena. In particular, the study of the consequences of external pressure on the physical characteristics turned out to be one of the important directions. The reason is that one is able to tune the relative strengths of the Kondo effect and magnetic ordering with pressure, as the former increases exponentially with the $4f$ exchange-coupling strength ($J$), whereas the latter increases quadratically with $J$. These functional dependencies give rise to a peak in the plot (Doniach’s magnetic phase diagram) of magnetic ordering temperature versus $J$. Thus, for instance, the compound CePd$_2$Si$_2$, known to order antiferromagnetically below ($T_N$) $10$ K under ambient pressure conditions, has been shown to exhibit a decrease of $T_N$ under pressure eventually attaining quantum critical point (QCP) at about $2.8$ GPa, thereby establishing that this compound lies at the right-hand side of the peak in Doniach’s phase diagram. In this regard, it is worthwhile to note that the compound Ce$_2$Pd$_3$Si$_5$ crystallizing in the orthorhombic $U_2$Co$_3$Si$_5$ structure is characterized by crystallographic similarities with CePd$_2$Si$_2$ in the sense that the former contains building blocks of Ce-Pd-Si slabs related to the CePd$_2$Si$_2$ unit cell sandwiched by Pd-Si layers (Fig. 1). Therefore, it is tempting to perform high-pressure experiments on Ce$_2$Pd$_3$Si$_5$, which has been reported to exhibit antiferromagnetic ordering at $8.2$ K. With this primary motivation, we carried out electrical resistivity ($\rho$) measurements in the temperature range $2.5$–$300$ K on Ce$_2$Pd$_3$Si$_5$ under pressure employing cubic anvil pressure cell ($\leq$8 GPa), the results of which are reported in this article.

II. EXPERIMENT

The polycrystalline samples employed in the present study were prepared by arc melting stoichiometric amounts of high-purity constituent elements on a water-cooled Cu hearth under argon atmosphere in an arc furnace. The molten ingots were annealed for two weeks at $800$ °C in an evacuated quartz tube. The quality of the annealed sample was examined by x-ray diffraction and metallography. The $\rho$ measurements up to $8$ GPa in the temperature $T$ interval $2.5$–$300$ K were carried out employing a cubic anvil pressure cell and the specimen employed was of the dimension $0.5\times0.6 \times 0.8$ mm$^3$. Nearly hydrostatic pressure was produced in the teflon cell filled with a fluid pressure-transmitting medium of 1:1 mixture of Fluorinert FC70 and FC77. Four-probe method was employed for the measurements and the electrical contacts with the specimen were made with $20\mu m$ φ gold wire using a conducting silver paste.

III. RESULTS

In Fig. 2 we have shown the row $\rho$ data as a function of $T$ under ambient pressure conditions. We have measured $\rho$ for the La compound as well to enable us to obtain the $4f$ contribution ($\rho_{mag}$). It is clear from Fig. 3 that $\rho_{mag}(T)=\rho_{Ce}(T)-\rho_{La}(T)$ exhibits a double-peaked structure arising from an interplay between the Kondo and crystal-field effects with the ratio of the high- and low-temperature slopes in the plot of $\rho_{mag}(T)$ in the ranges $9$–$20$ K and $200$–$300$ K, respectively, being close to the value $0.029$ expected for the doublet ground state. There is a sharp fall in $\rho$

FIG. 1. Crystal structure of Ce$_2$Pd$_3$Si$_5$ viewed along $a$ axis.
at 8.2 K as $T$ is lowered, which has been established to arise from the onset of antiferromagnetic ordering.$^{4,5}$

As the pressure is increased, we note that the overall features are qualitatively the same up to 6 GPa. However, the absolute value of $r$ increases with increasing pressure. This behavior is not expected for a Ce-based Kondo system. Actually, we measured the $r$ values while decreasing the pressure as well and we find that the trend is qualitatively the same. The exact origin of this trend is not clear to us at present. The noteworthy point is that $T$ at which the drop due to the onset of magnetic ordering occurs ~at 8.2 K! remains essentially unaltered over a wide pressure range, that is, up to about 4 GPa; however, for about 6 GPa pressure, this feature is observed at a somewhere lower temperature. In order to compare $T_N$ value at various pressures, we have defined $T_N$ as the temperature at which the two straight lines drawn through the data points just above and below this feature intersect, as shown in the inset of Fig. 2 for 6 GPa data. The value of $T_N$ thus obtained are shown in Fig. 4. A similar trend in $T_N$ is observed even if one looks at the temperature at which the $d\rho/dT$ peaks. The curve shown in Fig. 4 clearly establishes that this compound is placed at the peak of Doniach’s magnetic phase diagram, but definitely not at the right side of the peak. Interestingly, further increase of pressure to 8 GPa results in a peak in the $r_{\text{mag}}(T)$ at a slightly higher temperature, that is, at about 20 K. We believe that such an upward shift of this peak cannot arise from magnetic ordering judged by the pressure dependence of $T_N$ at lower pressures. Therefore, we attribute this 20 K peak at 8 GPa to the onset of coherent scattering among Kondo centers. We do not see any other feature below 6 K attributable to onset of magnetic ordering. We therefore expect that, at this pressure, the compound is nonmagnetic, at least above 2.5 K. It would be worthwhile to perform magnetization studies under pressure to confirm this. In any case, the trend in $T_N$, shown in Fig. 4, reveals that the pressure required (6–8 GPa) to bring the present compound to QCP is much larger than that for CePd$_2$Si$_2$ (2.8 GPa).$^3$ Finally, we would like to mention that the ratio of the resistivity slopes (discussed above) under pressure still falls in the range expected for the doublet ground state (that is, around 0.1).

\section*{IV. CONCLUSION}

To conclude, features attributable to a transformation from antiferromagnetism to Kondo lattice behavior could be observed in the pressure dependent $\rho$ data of Ce$_2$Pd$_3$Si$_5$. The present results establish that the compound Ce$_2$Pd$_3$Si$_5$ lies at the peak of Doniach’s magnetic phase diagram. It is worthwhile to extend the measurements to millikelvin range to look for superconductivity at QCP as in the case of CePd$_2$Si$_2$. $^3$

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