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Magnetic order in the Kondo-lattice compound CePdIn

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Out of a systematic research program on equiatomic ternary compounds RETM (RE is a rare earth, T a transition metal and M a metal out of the *p* block of the periodic table) crystallizing in the ZrNiAl structure some results on CePdIn are reported. Transport and magnetic properties and specific heat were investigated as functions of temperature. An antiferromagnetic transition at 1.65 K shows up in the specific heat as a maximum and in the resistivity as a distinct change of slope. Other salient features are the occurrence of two maxima in resistivity, a huge anomalous Hall effect and a high electronic-specific-heat coefficient of 123 mJ/mole K^2, reminiscent of low T_{sf} spin-fluctuation systems. The isostructural compound LaPdIn becomes superconducting at 1.6 K.

INTRODUCTION

The ground state of Ce, Eu, and Yb in intermetallic compounds shows a variety of valencies depending on ion-ion spacing, neighboring atoms, lattice pressure, and so on. Within the REPdIn (RE is rare earth) series the Yb and Eu compounds behave anomalously. YbPdIn has been established to be mixed-valent and EuPdIn, that crystallizes in a different structure, is divalent.[1] No information is available in literature about the valence state of Ce in CePdIn. We have investigated the transport and magnetic properties and the specific heat of CePdIn in order to find out whether any anomalies associated with a possible mixed-valent state would show up in the macroscopic properties of this compound.

SAMPLE PREPARATION

Polycrystalline samples of CePdIn were prepared by arc melting high-purity starting elements in a titanium-gettered argon atmosphere. To account for evaporation losses of indium an excess of 3 wt. % indium was taken. The buttons were turned and remelted several times. Whiskers jump spontaneously out of the melt at fast cooling on a water-cooled copper hearth. From x-ray powder and Weissenberg measurements the ZrNiAl structure was established with lattice parameters $a = 7.698$ Å, $c = 4.076$ Å, which is in agreement with Ref. 2. By DTA the melting point of the compound was determined at 1366 K; the origin of an additional very small endothermic effect at 1070 K is not yet clear.

RESULTS

The magnetic susceptibility was measured between 1.6 and 300 K. The inverse susceptibility plotted versus temperature (Fig. 1) displays nonlinear behavior up to about 200 K indicating appreciable crystal-field splitting. From the approximate Curie–Weiss behavior between 200 and 300 K an upper limit for the effective moment of $2.64\mu_B$, which exceeds the free ion moment by 4%, and a paramagnetic Curie temperature of -41 K can be derived. Assuming Curie–Weiss behavior in the interval from 2 to 7 K an effective moment of $1.6\mu_B$ is found which is consistent with the expected doublet ground state. At liquid-helium temperatures the magnetization depends linearly upon the field below 1T, shows curvature at higher fields (insert in Fig. 1), but does not saturate in fields as high as 35 T.

Specific-heat results between 1.2 and 40 K are shown in Fig. 2. For comparison we also show the specific heat of LaPdIn (dashed line and insert in Fig. 2) that becomes superconducting at 1.6 K. For CePdIn at 1.65 K a peak is observed in C/T vs T. In magnetic fields, that were applied up to 5 T (Fig. 3), this peak broadens and shifts to lower temperatures. The temperature T_{max} at which the maximum of the peak is located depends quadratically upon the field: $T_{max} = 1.65 - \alpha B^2$ with $\alpha \simeq 11$ mK/T^2. Due to the lack of data below 1.2 K the entropy connected with the anomaly can only be estimated to be less than 50% of $R \ln 2$ at the peak position. C/T plotted versus T^2 is linear between 10 and 20 K. From this an electronic-specific-heat coefficient of 123 mJ/mole K^2 and a Debye temperature of 200 K are determined.

Electrical-resistivity measurements between 1.5 and 300 K on polycrystalline samples and between 0.5 and 200 K on whiskers along the *c* axis are shown in Figs. 4 and 5, respectively. Due to the presence of cracks in the polycrystalline samples and due to the very small dimensions of the whiskers (length $\simeq 1$ mm, diameter $\simeq 40\,\mu$m) the resistivity values may be wrong by about 20%. The polycrystalline samples exhibit a pronounced double-peak structure, similar to what has been found in $CeCu_2Ge_2$,[3] with a sharp maximum at 3 K and a broad maximum around 70 K. For the whiskers the lower maximum is shifted to 3.5 K and the upper maximum shows up as a broad shoulder. Residual resistivity is not reached for the polycrystals at 1.5 K nor for the whiskers at 500 mK. The resistivity of the whiskers shows a quadratic temperature dependence between 500 mK and 1.6 K with a temperature coefficient of about 6 $\mu\Omega$ cm/K^2 (insert in Fig. 5), which is in the same order of magnitude as what is found in heavy fermion systems.[4] However, one has to keep in mind that in the neighborhood of a magnetic transition not only electron-electron interaction may cause a quadratic increase in resistivity. Above 1.7 K the slope is much lower and flattens towards 3 K. The behavior right at the temperature of the specific-heat maximum was not measurable due to contact failure. A reason for this may be huge

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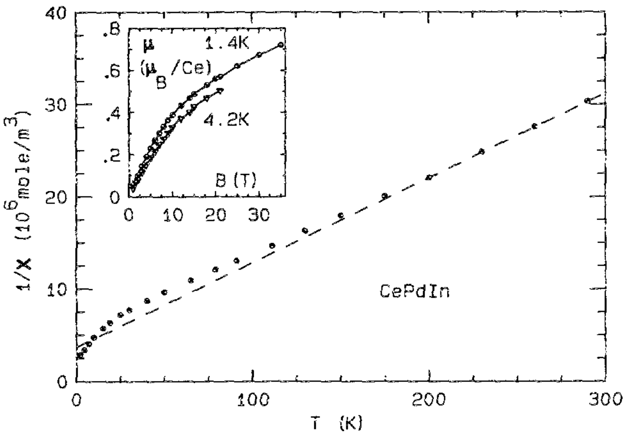

FIG. 1. Inverse dc susceptibility vs temperature for CePdIn. Insert: High-field magnetization vs magnetic field at 1.4 and 4.2 K.

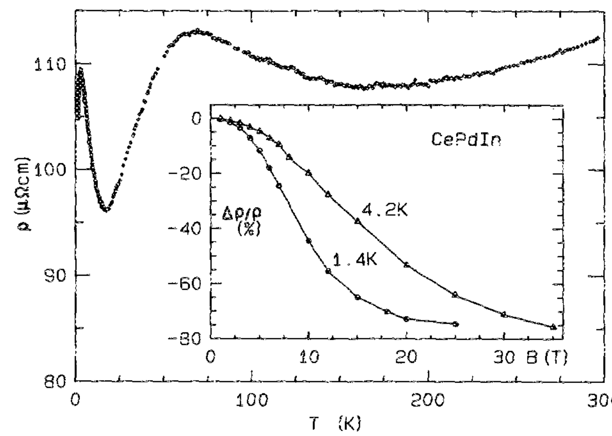

FIG. 4. Resistivity of polycrystalline CePdIn vs temperature. Insert: Relative reduction of the resistivity as function of the applied field.

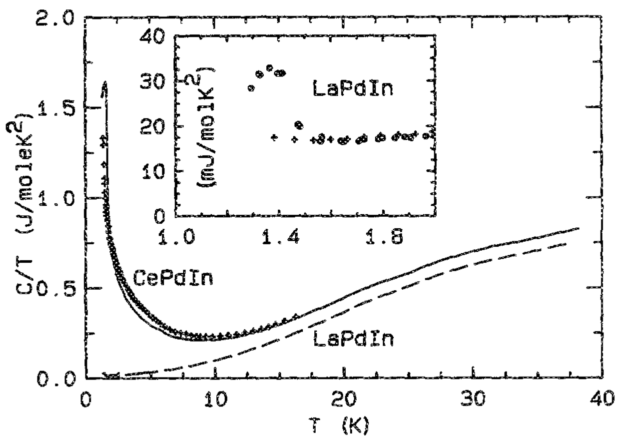

FIG. 2. Specific heat in a plot of C/T vs T of CePdIn in zero field (line) and in an applied field of 5 T (+) and of LaPdIn (dashed line). Insert: Low-temperature specific heat of LaPdIn in zero field (O) and in an applied field of 5 T (+).

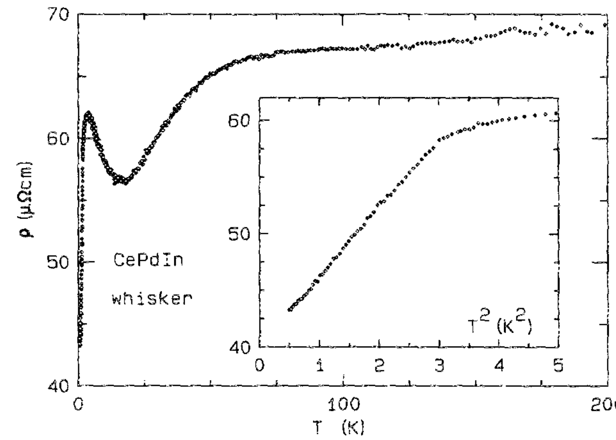

FIG. 5. Resistivity vs temperature for CePdIn whiskers measured along c axis. Insert: Low-temperature resistivity vs T^2.

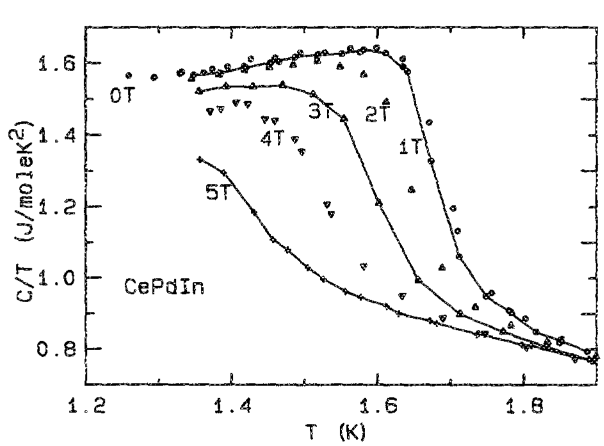

FIG. 3. Specific heat of CePdIn around the maximum in different magnetic fields.

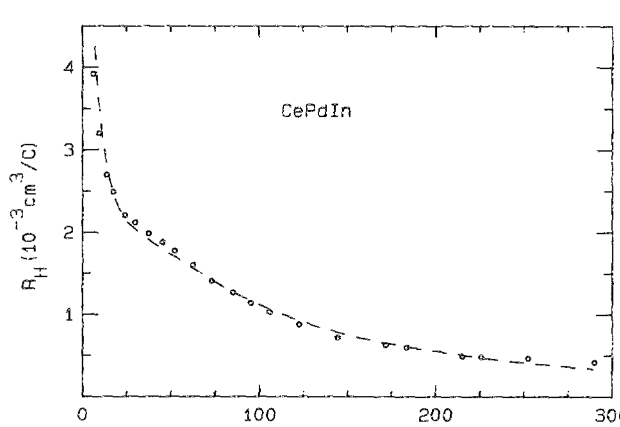

FIG. 6. Hall effect of CePdIn as a function of temperature. The dashed line represents a fit to $R_H = R_0 + K\rho\chi$ (see text).

lattice distortions causing movement of the contacts over the sample.

At the fixed temperatures of 1.4 and 4.2 K the field dependence of the resistivity was measured on a polycrystalline sample (insert in Fig. 4). The magnetoresistance is found to be negative and dependent on the current density, at a current density of 30 A/cm^2 the relative reduction of the resistivity is about half of what is found at current densities below 10 A/cm^2, where the magnetoresistance is independent of the current density within the accuracy of the measurement. The values given in Fig. 4 are taken at current densities of 7.5 A/cm^2 and 3 A/cm^2 at 4.2 and 1.4 K, respectively. Oscillations reminiscent to Shubnikov–de Haas oscillations are observed at 1.4 K and in fields above 25 T. Up to 7 T the resistivity decreases quadratically with the field, at 1.4 K saturation at about 25% of the zero-field value is reached at about 25 T and at 4.2 K above 35 T.

The Hall effect between 6 and 300 K is shown in Fig. 6. Its magnitude and temperature dependence are similar to what is found in cerium heavy-fermion systems or low-T_{sf} mixed-valent compounds.[5,7] An astonishing good fit is obtained by $R_H = R_0 + K_{\rho\chi}$ with $R_0 = -3.33\times 10^{-4}$ cm^3/C and $K = 1.81\times 10^{-3}$ mole/C $\mu\Omega$ cm (dashed line in Fig. 6). The Hall resistivity is proportional to the applied magnetic field up to 5 T over the whole temperature interval.

CONCLUSION

Macroscopic properties of CePdIn are anomalous, however, the behavior can still be conceived consistent with an integral valency of cerium. The field dependence of the maximum in the specific heat is typical for antiferromagnetic order that also shows up as a kink in the electrical resistivity. From the pronounced anomalies in the transport properties and its high electronic-specific-heat coefficient CePdIn can be classified as a low-T_{sf} spin fluctuation or Kondo-lattice compound. Magnetic-susceptibility results are consistent with a trivalent groundstate and appreciable crystal-field splitting with a low-lying doublet. To establish the influence of the anisotropic crystal structure on all macroscopic properies, further work on single crystals is planned.

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