Specific heat of $CePb_3$ in magnetic fields

R. Pietri and B. Andraka
Department of Physics, University of Florida, P.O. Box 118440, Gainesville, Florida 32611-8440
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The specific heat of polycrystalline $CePb_3$ was measured in magnetic fields to 14 T and temperatures down to 0.4 K. An anomaly related to an aniferromagnetic phase transition is effectively attenuated by magnetic fields and disappears for fields larger than 6 T. The electronic specific heat coefficient γ increases for small fields, has a maximum value at 6 T, and decreases for larger fields. This field dependence of γ correlates with previously studied quadratic temperature coefficient of the resistivity as a function of a magnetic field. A/γ^2 is field dependent below 6 T, but constant ($\sim 10^{-5}$ $\Omega\,\mathrm{cm\,K^2\,mol^2\,J^{-2}}$) for fields larger than 6 T within the experimental uncertainty.

Alloying experiments performed on $CePb_3$ (Ref. 1) have provided the strongest evidence yet for Kondo-impurity behavior of Ce-based heavy fermions. Both the specific heat C and magnetic susceptibility χ of $Ce_xLa_{1-x}Pb_3$ scale with the concentration of Ce at temperatures larger than 1.5 K. Moreover, the temperature dependence of the specific heat above 1.5 K can be described, with high accuracy, by the Kondo model with $T_K = 3.3$ K.[2] This latter value is also consistent with the magnetic susceptibility values at low temperatures.

For the same reason, $CePb_3$ can also be considered highly anomalous. It orders antiferromagnetically at about 1.1 K, and can be classified as a magnetic Kondo lattice. The ratio of the low temperature χ to γ (related to the Wilson ratio), where γ is the linear coefficient of the specific heat, for magnetic Kondo lattices, is much larger than that derived from the single impurity model. Magnetic interactions between Ce moments renormalize χ and γ in a very different manner. In fact, χ/γ is used as an important criterion to distinguish between magnetic and nonmagnetic heavy fermions.[3] Accordingly, $CePb_3$ should be a nonmagnetic heavy fermion system.

An unusual coexistence between magnetic and heavy-fermion–Fermi-liquid degrees of freedom below T_N is another outstanding feature of $CePb_3$. A pronounced antiferromagnetic anomaly in the specific heat (see Fig. 1) leads to only a partial reduction of the electronic density of states. There is a huge residual γ of about 1000 $\mathrm{mJ/K^2\,mol}$. The electrical resistivity at temperatures well below T_N has a familiar Fermi-liquid temperature dependence[4] $\rho = \rho_0 + AT^2$, with $A = 45$ $\mu\Omega\,\mathrm{cm\,K^{-2}}$. This latter value is among the largest ever observed in heavy fermions and comparable to the ones corresponding to $CeCu_6$ and $CeAl_3$,[5] canonical heavy fermion compounds generally regarded as nonmagnetic. The ratio A/γ^2 for $CePb_3$ is about 4×10^{-5}, thus larger than the average value for nonmagnetic heavy fermions (1×10^{-5}), but in line with an experimentally observed enhancement of this ratio for magnetic Kondo lattices. According to the results obtained by McDonough and Julian,[4] the quadratic temperature coefficient of the resistivity A increases in moderately high magnetic fields, reaches a maximum somewhere near 5 T, and decreases at larger fields. In addition, the resistivity acquires a large term linear in temperature near this critical field value. An increase of A suggests an increase of the electronic density of states and of γ. An experimental verification of this increase, a search for a scaling relation between A and γ as a function of field, and the nature of heavy fermion state of $CePb_3$ in moderately high and high magnetic fields were among the objectives of our study.

All reported measurements were performed on a polycrystalline sample obtained by arc melting the highest commercially available elements. Special care was taken to compensate for Pb losses during the melting. The sample was annealed in excess of Pb and according to the prescription published in Ref. 1. No additional phases were detected by x-ray diffraction analysis. The results of magnetic susceptibility and zero-field specific heat measurements were in agreement with other published results.

The results of specific heat measurements for several fields between zero and 14 T are shown in Fig. 1 in the form of C/T versus T. No phonon subtraction was performed since the phonon contribution is negligible below 2 K. The peak that is observed just above 1.1 K in zero field is reduced in magnitude by magnetic fields and shifted to lower temperatures. Using the temperature position of the maximum in $C/T(T_m)$, we have constructed an H-T phase diagram shown

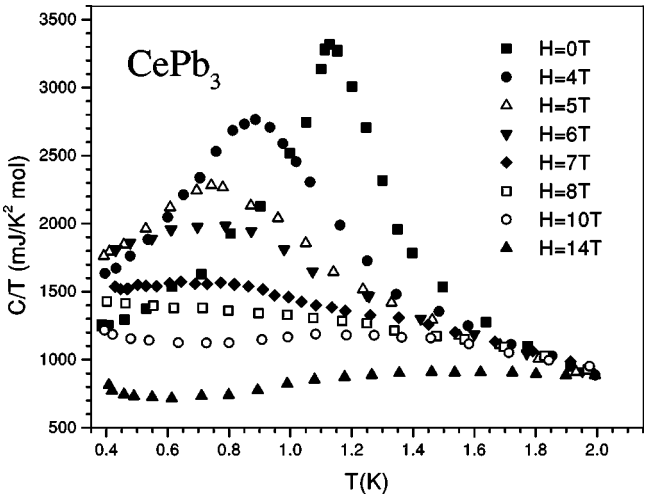

FIG. 1. C/T versus T for $H = 0$, 4, 5, 8, 10, and 14 T.

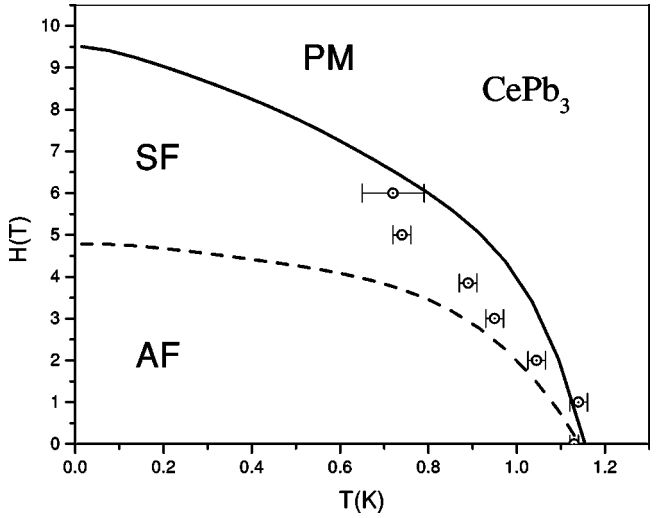

FIG. 2. H-T phase diagram for $CePb_3$. The solid and dashed lines represent magnetoresistance data from Ref. 4.

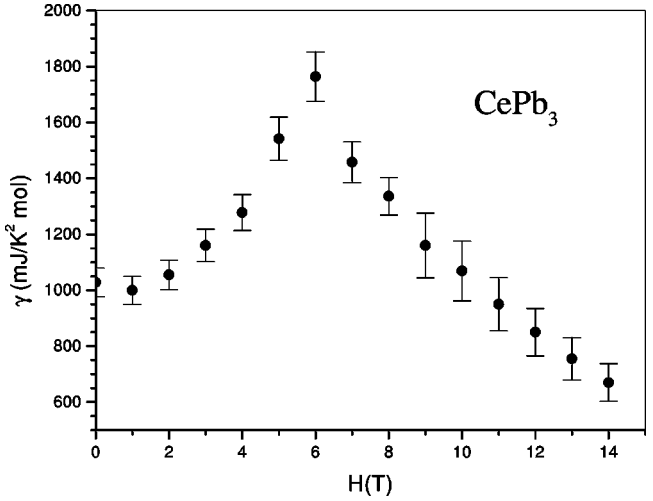

FIG. 3. γ versus H for $CePb_3$.

in Fig. 2. A similar phase diagram could be also constructed from the position of a maximum in C versus T. However, these latter maxima are much less pronounced, particularly for fields of order 5 T. For nonzero fields, there is also a difference in the temperature position where C/T and C peak. A maximum in C corresponds to a somewhat larger value than T_m and this difference grows with H. Figure 2 shows also two lines of transitions inferred from transport measurements[4,6] performed on single crystals of $CePb_3$, for magnetic fields applied along the (110) crystallographic axis. The broken lower line at lower field is believed to be due to transitions between a long range aniferromagnetic phase[7] (AF), incommensurate with the lattice, and a spin-flop (SF) phase, and the upper line due to transitions between a SF and a paramagnetic phase. However, the spin-flop assignment to the intermediate field phase has to be treated tentatively, since the nature of this phase has not yet been determined by neutron diffraction studies. Our points obtained from the specific heat on the polycrystalline sample fall in between the two lines. We have not observed double transitions for any value of the applied field. This can be due to the fact that according to the magnetorestance measurements at 20 mK,[4] the lower field transition is well defined only when the magnetic field is applied along the (110) direction or when it makes a small angle this direction. When the field is rotated away from the (100) direction, the AF-SF transition smears out and is observed at a higher field. Only one broad transition was observed in the resistivity at 20 mK when the field was applied along the (100) direction.

No distinct anomalies in C/T that could be related to the antiferromagnetic transition were detected in fields higher than 6 T, i.e., for $H=7$ and 8 T. Note that T_m for the 6 T data is still above 0.7 K and the rate of the suppression of T_m by magnetic field is relatively small between 4 and 6 T. Thus the absence of the anomaly in the data for 7 and 8 T cannot be explained by a shift of T_m below the lowest temperature of our measurements but rather by the fact that the magnitude is strongly attenuated by the fields and the maximum disappears near 6 T, i.e., before T_m reaches 0.4 K. On the other hand, there is a broad shoulder in C/T versus T for the highest fields applied, 10 and 14 T (Fig. 1). This shoulder cannot be related to the upper field transition reported in Ref. 4 since it moves to higher temperatures with an increase of H. A similar and even more pronounced shoulder can be also found in the 16.1 T data reported by Fortune *et al.*[8]

Our observation that 6 T is one of the characteristic fields of this system is also consistent with the field dependence of the electronic specific heat coefficient. γ versus H is shown in Fig. 3. For fields up to 6 T, γ values have been determined from the fits of C/T versus T^2 to straight lines at temperatures well below T_m. For fields larger than 6 T, i.e., for fields for whose C/T does not vary appreciably below 0.8 K, γ values have been taken as C/T at 0.4 K. There is a small tail in the 14 T data below 0.5 K, whose magnitude is consistent with the nuclear contribution of Pb. This contribution at 0.4 K is much smaller than the uncertainty of our data of about 10%. This way determined γ initially increases with H, reaches a maximum value of 1770 mJ/K^2 mol for $H=6$ T, and decreases for fields larger than 6 T. γ is only about 600 mJ/K^2 mol at 14 T. We have disregarded a small decrease of γ between 0 and 1 T, since this decrease is within the uncertainty of our results.

The plot of γ versus H (Fig. 3) is reminiscent of the plot of A versus H for the field parallel to the (110) direction,[4] with the following exception. The coefficient A peaks sharply at 5 T as opposed to γ, which reaches a maximum near 6 T. This discrepancy is possibly related to the fact that our results for γ represent some averages over all crystallographic directions. On the other hand, the results for magnetoresistance seem to suggest that while rotating the field away from the (110) direction decreases the magnitude of the maximum of A versus H, it also does change appreciably its field position.

The correlation between A and γ is explored in Fig. 4, which displays the ratio A/γ^2 as function of magnetic field. A has been taken for the field along the (110) direction from Ref. 4. This ratio for fields smaller than 6 T is clearly larger than the average A/γ^2 value found in heavy fermion metals.[5] This average ratio, $\sim 1\times 10^{-5}$ $\Omega\,\mathrm{cm\,K^2\,mol^2\,J^{-2}}$ (postulated also to be a universal value for nonmagnetic heavy fermion compounds) is represented by a broken line in Fig. 4. On the other hand, we have observed a rather flat A/γ^2 in the range $1-2\times 10^{-5}$ $\Omega\,\mathrm{cm\,K^2\,mol^2\,J^{-2}}$ for $H\geqslant 6$ T. This

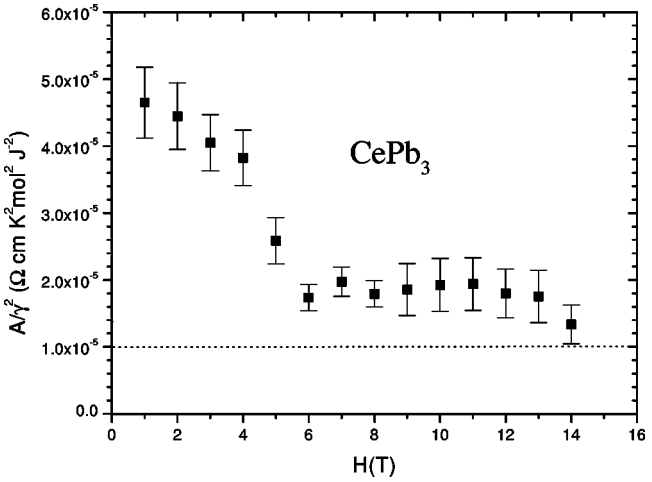

FIG. 4. A/γ^2 versus H for $CePb_3$. Values for A are from Ref. 4. The dotted line corresponds to the universal ratio postulated in Ref. 5.

latter observation seems to suggest that there is a good scaling between A and γ independent of the field in the nonmagnetic regime. A similar conclusion has been previously made for $CeCu_{5.9}Au_{0.1}$.[9] This alloy exhibits non-Fermi-liquid behavior in $H=0$, but in finite fields its A/γ^2 is constant and almost equal to the postulated universal value. Enhanced values of A/γ^2 below 6 T for $CePb_3$ are consistent with usually larger values of this ratio reported for systems in whose heavy electrons coexist with a magnetic order. For example, the above ratio for heavy fermion antiferromagnets $CeAl_2$,[10] $CeAuAl_3$,[11] $CePd_2In$,[12] and YbPdBi[13] (all with T_N of order 1 K) is 5, 10, 3, and 30×10^{-5} $\Omega\,\mathrm{cm\,K^2\,mol^2\,J^{-2}}$, respectively. We are not aware of any theoretical investigations of this enhancement. According to the study by Takimoto and Moriya[14] in the framework of spin fluctuations, A/γ^2 is essentially independent on the strength of spin fluctuations, except in the very vicinity of their critical value ($T_N=0$) where this ratio is enhanced. On the other hand, if for $H=0$ one uses a value for γ calculated from $T_K=3.3$ K (determined from the fit of the specific heat above T_N; $\gamma=1700$ $\mathrm{mJ/K^2\,mol}$) then the calculated A/γ^2 is about 1.6×10^{-5} $\Omega\,\mathrm{cm\,K^2\,mol^2\,J^{-2}}$, i.e., of the same magnitude as that for $H=6$ T. Thus the value of A coefficient, measured well below T_N, seems to be more consistent with T_K, characterizing the paramagnetic state, than the measured low temperature γ.

Finally, we have not observed non-Fermi-liquid properties for any value of applied magnetic field. Such non-Fermi-liquid properties have been reported for antiferromagnets for whose a magnetic order has been suppressed by pressure or magnetic field; e.g., for $CeCu_{4.8}Ag_{1.2}$ at 3 T.[15] Lack of non-Fermi-liquid characteristics might be explained by the fact that thermodynamic signatures of a magnetic order in $CePb_3$ disappear in a magnetic field long before T_N reaches zero. Quantum phase transition models relate such non-Fermi-liquid behavior to critical fluctuations corresponding to $T_N=0$.[16]

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