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Low temperature properties of the ternary compounds $CePt_2B$ and $CePt_3B$

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Abstract

Ground state properties of ternary $CePt_2B$ and $CePt_3B$ have been characterized from studies of various bulk properties. Both compounds order antiferromagnetically below 2.1 and 7.5 K, respectively, and the latter exhibits an additional phase transition at $T_1 = 4.5$ K. The spin wave dispersion relation of these systems is gapped, with $\Delta \approx 1$ and ≈ 7 K, for $CePt_2B$ and $CePt_3B$, respectively. Kondo-type interaction and crystal field splitting are present, reducing the absolute values of the ordered moments. The characteristic temperatures are $T_K = 3.5$–5 K for the former and $T_K \approx 6$–7 K for the latter.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

A recent exploration of phases in RE–Pt–B (RE = rare earth) and particularly Ce–Pt–B revealed the new ternary compounds $CePt_2B$ [1] and $CePt_3B$ [2]; both of them crystallize in their own structure type. While the former is hexagonal with space group $P6_222$ and lattice parameters $a = 5.4898$ Å and $c = 7.8860$ Å, the latter is tetragonal with space group $P4mm$ and lattice parameters $a = 4.004\,33$ Å and $c = 5.075\,47$ Å. A peculiarity of $CePt_3B$ is the absence of a centre of inversion. The paramagnetic state of the latter was already characterized by L_{III} absorption edge measurements, revealing a $4f^1$ electronic configuration for the Ce ion, in agreement with an effective magnetic moment $\mu_{eff} = 2.58\ \mu_B$, deduced from the temperature dependent magnetic susceptibility [1]. A study of low temperature properties by Süllow *et al* [3] evidenced long range magnetic order at $T_N = 7.5$ K, followed by a reorientation of the spin structure at $T_1 = 4.5$ K. That study, however, was lacking in that the crystal structure of

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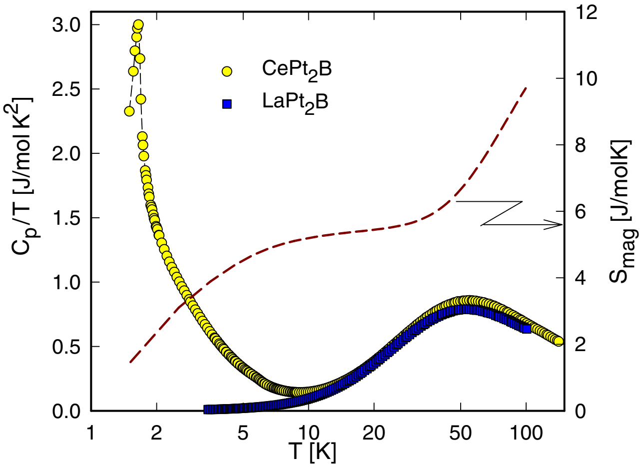

Figure 1. The temperature dependent specific heat C_p of $CePt_2B$ and of isomorphous $LaPt_2B$ plotted as C_p/T versus T. The dashed curve represents the magnetic entropy (right scale).

$CePt_3B$ was not accounted for in a proper manner. Low temperature properties of $CePt_2B$ are still largely unexplored.

Special interest in the above novel phases is drawn from the fact that isomorphous ternary $CePt_3Si$ exhibits heavy fermion superconductivity ($T_c = 0.75$ K) and, simultaneously, shows long range magnetic order ($T_N \approx 2.2$ K) [4]. The two phenomena coexist, and do not segregate in real space. The absence of a centre of inversion has severe consequences for the order parameter. In fact, NMR studies performed on this compound [5] show that the superconducting order parameter neither follows the features well known for BCS superconductors nor those of already explored unconventional heavy fermion superconducting systems.

The aim of the present work is a thorough investigation of low temperature properties of both $CePt_2B$ and $CePt_3B$ in order to work out ground state properties as well as to explore the possibility of a superconducting transition.

Polycrystalline samples were synthesized by arc melting and subsequently heat treated. A number of standard techniques served for obtaining the temperature, magnetic field and pressure dependent resistivity, specific heat, magnetization and susceptibility. For details see, e.g., [6].

2. Results and discussion

2.1. $CePt_2B$

Heat capacity measurements are sensitive tools with respect to phase transitions and renormalization of charge carrier masses, and provide information concerning a possible Fermi liquid or non-Fermi liquid behaviour. We show therefore in figure 1 the temperature dependent specific heat C_p of $CePt_2B$ and of the non-magnetic counterpart $LaPt_2B$ on a logarithmic temperature scale. Most prominent is the λ-like anomaly around 2 K, indicating a magnetic phase transition of $CePt_2B$ into a, presumably, antiferromagnetically ordered ground state below $T_N \approx 2.1$ K. Closely above the phase transition, the electronic contribution to the specific heat appears to be significantly enhanced either due to temporal or spatial fluctuations

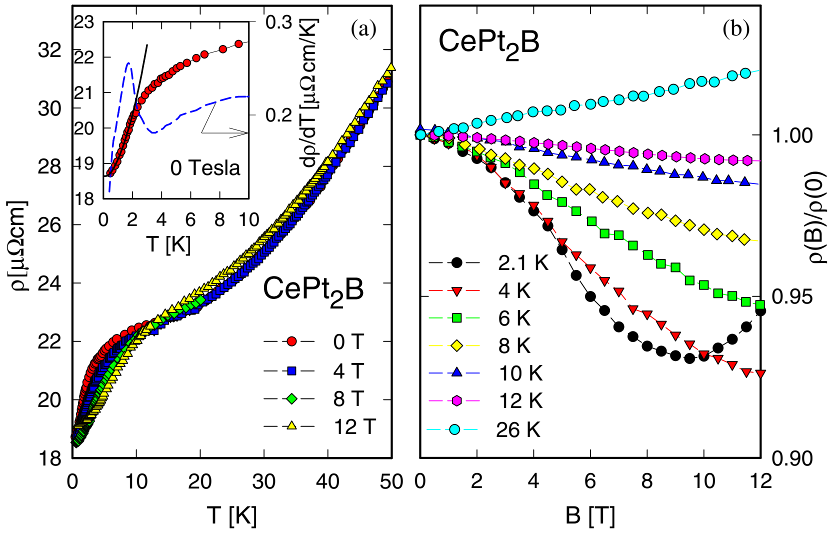

Figure 2. (a) The temperature dependent resistivity ρ of CePt$_2$B for various values of applied external fields. The inset shows low temperature details of the zero-field measurement. The dashed curve shows $\mathrm{d}\rho/\mathrm{d}T$ and the solid curve is a least squares fit according to equation (1). (b) The magnetoresistance $\rho(B)/\rho(0)$ of CePt$_2$B for various temperatures.

of the order parameter above T_N or due to the mass renormalization owing to the presence of Kondo-type interactions. Such processes are responsible for spreading entropy from below the phase transition to an extended temperature region above T_N. In fact, the magnetic entropy (dashed curve, right axis in figure 1) shows that right at $T = T_\mathrm{N}$ only about half of the entropy expected for ordering within a ground state doublet is released, while $S_\mathrm{mag} = R\ln 2$ is reached only at temperatures around 25 K. The further smooth increase of entropy with temperature indicates that thermal population of the first and the second excited levels of the CEF above the ground state doublet may happen within a temperature range from 100 to 200 K. While in a mean field-type of magnetic phase transition the jump of the specific heat at $T = T_\mathrm{ord}$ is about 12.5 J mol^{-1} K^{-1} in the case of a doublet ground state, Kondo-type interaction causes a substantial reduction of this jump height as a result of an increasing value of the ratio $T_\mathrm{K}/T_\mathrm{RKKY}$. Here, $k_\mathrm{B}T_\mathrm{K}$ is the Kondo interaction strength and $k_\mathrm{B}T_\mathrm{RKKY}$ measures the intersite interaction, responsible for establishing long range magnetic order. Beyond a critical value $|T_\mathrm{K}/T_\mathrm{RKKY}|_\mathrm{c}$ long range magnetic order vanishes; hence the jump of C_p at $T = T_\mathrm{ord}$ becomes completely suppressed. Taking into account calculations performed in terms of the mean field model for long range magnetic order [7] and the model of Schotte and Schotte [8] for the Kondo effect allows us to estimate the characteristic temperature $T_\mathrm{K} \approx 5$ K from the experimentally observed jump of C_p at $T = T_\mathrm{N}$. The magnitude of T_K naturally explains the transfer of magnetic entropy to the observed high temperature region. Moreover, in terms of a Bethe ansatz calculation of the entropy release of an effective 1/2 Kondo system, stating that at $T/T_\mathrm{K} = 1$ the entropy value amounts to about 3.75 J mol^{-1} K^{-1} [9], S_mag of CePt$_2$B would correspond to $T_\mathrm{K} \approx 3.5$ K, in fair agreement with the above sketched procedure.

The temperature and field dependent resistivity ρ for the low temperature region is shown for CePt$_2$B in figure 2(a) and the magnetoresistance for various temperatures is summarized in figure 2(b). $\rho(T)$ characterizes the metallic behaviour of CePt$_2$B and pronounced indications of phase transitions are absent in the present temperature and field range. However, the temperature derivative of the resistivity $\mathrm{d}\rho/\mathrm{d}T$ clearly signals the onset of long range magnetic order at $T_N \approx 2.1$ K (compare the inset of 2(a)). The weakness of the resistivity change at $T = T_N$ may be a hint as to small magnetic moments involved in ordering in this ternary compound and thus may corroborate the presence of the Kondo effect. The application of a magnetic field causes only slight changes of the electrical resistivity. Above about 15 K, $\rho(T)$ increases with increasing fields, while below that temperature the opposite occurs, reflecting a quenching of magnetic fluctuations. Only at temperatures below a few kelvins does the dependence become more complicated due to the presence of long range magnetic order. In order to qualitatively account for the ordered region of CePt$_2$B, a model developed in [10] is applied, yielding an analytic expression concerning the temperature dependent electrical resistivity:

$$\rho = \rho_0 + A\Delta^{3/2}T^{1/2}\exp(-\Delta/T)\left[1 + \frac{2}{3}\left(\frac{T}{\Delta}\right) + \frac{2}{15}\left(\frac{T}{\Delta}\right)^2\right]. \tag{1}$$

This expression is based on scattering of conduction electrons on antiferromagnetic magnons with a dispersion relation given by $\omega = \sqrt{\Delta^2 + D^2k^2}$, where Δ is the spin wave gap and D is the spin wave velocity; $A \propto 1/D^3 \propto 1/\Gamma^3$ and Γ is an effective magnetic coupling between Ce ions. Applying equation (1) to the experimental data yields a gap $\Delta/k_B \approx 1$ K. Equation (1) is valid only for $k_BT < \Delta$, and for temperatures well below the ordering temperature. Thus, the fit has been performed for $T < 0.5T_N$, but it nicely describes all data below T_N. This fact was already noticed in [10].

The magnetoresistance $\rho(B)/\rho(0)$ is plotted in figure 2(b) as a function of the externally applied magnetic field. $\rho(B)/\rho(0)$ shows a crossover from values below one to values above one as the temperature grows. While the former results from a quenching of magnetic fluctuations by the external field, the latter appears to be a consequence of the classical magnetoresistance, which seems to exceed the magnetic field induced change of magnetic interaction processes. The qualitative form of $\rho(B)/\rho(0)$ versus B coincides well with predictions of the Kondo model. The measurement at $T = 2$ K, however, shows distinct deviations from the smooth variations of $\rho(B)/\rho(0)$ at higher temperatures. This refers to the proximity of long range magnetic order and possibly reflects a field induced reorientation of the magnetic structure.

2.2. *CePt$_3$B*

The specific heat derived for CePt$_3$B is plotted in figure 3 as C_p/T versus T, together with that for non-magnetic isostructural LaPt$_3$B. Like in the previous case, CePt$_3$B orders magnetically at $T_N = 7.6$ K; additionally, however, a second phase transition is observable at $T_1 \approx 4.5$ K, in agreement with a previous study of Süllow *et al* [3]. Isothermal magnetization measurements (not shown here) reveal a metamagnetic phase transition at 3.5 T, corroborating antiferromagnetic order. However, data taken below T_1 indicate a very small ferromagnetic contribution possibly due to some canted magnetic structure. The contribution of antiferromagnetic spin waves to the specific heat follows from the previous model [10] as

$$C_{\text{mag}} = \delta\Delta^{7/2}T^{1/2}\exp(-\Delta/T)\left[1 + \frac{39}{20}\left(\frac{T}{\Delta}\right) + \frac{51}{32}\left(\frac{T}{\Delta}\right)^2\right], \tag{2}$$

with $\delta \propto 1/D^3$. A least squares fit of equation (2) to the data well below T_1 yields a gap in the antiferromagnetic spin wave dispersion $\Delta \approx 7$ K together with the Sommerfeld constant $\gamma = 57$ mJ mol^{-1} K^{-2}. The γ value derived is characteristic for many magnetically ordered Kondo lattices, such as prototypic CeAl$_2$.

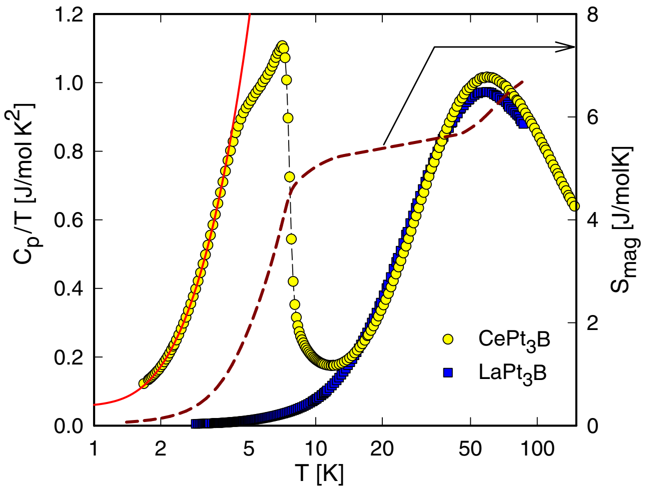

Figure 3. The temperature dependent specific heat C_p of $CePt_3B$ and of isomorphous $LaPt_3B$ plotted as C_p/T versus T. The solid curve is a least squares fit according to equation (2). The dashed curve represents the magnetic entropy (right scale).

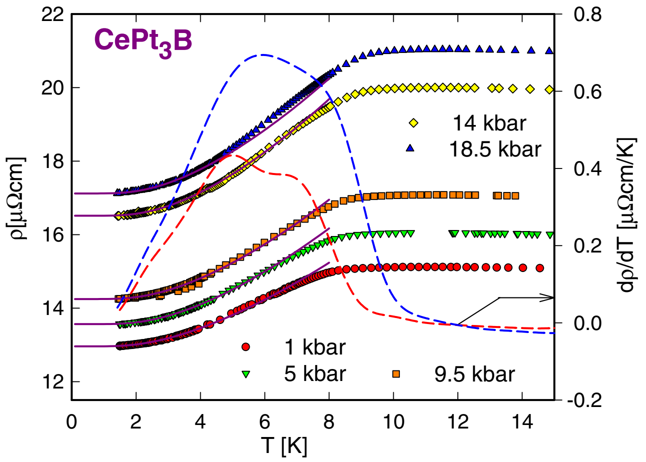

Figure 4. (a) The temperature dependent resistivity ρ of $CePt_3B$ for various values of applied hydrostatic pressure. The dashed curve shows $d\rho/dT$ and the solid curve is a least squares fit according to equation (1).

A comparison of $C_p(T)$ of $CePt_3B$ with $LaPt_3B$ allows determination of the magnetic entropy (dashed curve, figure 3). $S_{mag}(T = T_N)$ amounts to about 4.5 J mol^{-1} K^{-1}, which is about 20% below the expected $R \ln 2$. In context with the reduced jump of $C_p(T = T_N)$, Kondo-type interaction is considered responsible for that observation, with a characteristic temperature $T_K \approx 6$ K. This value coincides rather well with absolute entropy data [9], yielding $T_K \approx 7$ K. $R \ln 2$ is recovered only well above T_N and the plateau-like behaviour of S_{mag} between about 20 and 50 K indicates that the first and second excited levels of the CEF splitting of the ground state multiplet lie well above the ground state.

Low temperature details of the electrical resistivity ρ of $CePt_3B$ are shown in figure 4 for various values of applied pressure. There is an overall shift of the resistivity towards larger absolute values. The temperature derivative $\mathrm{d}\rho/\mathrm{d}T$ (shown for clarity only for the lowest and the highest value of pressure) exhibits for $p = 1$ kbar a double-peak structure, in accordance with the observations made from specific heat measurements. This feature, however, becomes washed out as the pressure grows; nevertheless, the two phase transitions seem to coexist for the pressure range shown. While T_N increases by about 1 K as a response to the applied pressure, the overall change of T_1 is slightly lower. The solid curves are least squares fits according to equation (1). For $p = 1$ kbar, the gap in the antiferromagnetic spin wave excitation is $\Delta \approx 6.8$ K, in good agreement with the figures derived from specific heat data. The increase of pressure, however, does not cause a substantial change of the gap value, at least for the pressures available in the present study.

In summary, investigations concerning $\mathrm{CePt_2B}$ and $\mathrm{CePt_3B}$ revealed long range magnetic order below 2.1 and 7.5 K, respectively. Ordering takes place with reduced magnetic moments due to the Kondo effect and the ground state multiplet is lifted by crystal electric field splitting. Superconductivity is not observed in either case, at least down to 0.4 K. Since for $\mathrm{CePt_3B}$, $\mathrm{d}T_\mathrm{N}/\mathrm{d}p > 0$, the system is placed in Doniach's phase diagram well below the maximum, relating to a dominance of the RKKY interaction, and substantial values of pressure are required to shift the system towards its quantum critical point.

Acknowledgments

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