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Transport and thermodynamic properties of $CePt_3X$ intermetallics

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Abstract

A series of $CePt_3X$ intermetallics (X = Si, Al, Ge, B) has been synthesized, characterized by X-ray powder diffraction and microprobe analysis. $CePt_3Si$ and $CePt_3B$ compounds crystallize in a non-centrosymmetric $CePt_3B$-type structure with a space group $P4\,mm$, while the diffraction patterns of $CePt_3Al$ and $CePt_3Ge$ show pronounced splitting of diffraction lines indicating a disordered $CePt_3B$-type structure or rather a new crystallographic structure with a lower symmetry. The results of specific heat and electrical resistivity measurements are discussed. While the heavy fermion superconductivity coexists with the long-range antiferromagnetic ordering in $CePt_3Si$, the other compounds show a rather complex magnetic ordering at low temperatures; however, no sign of superconductivity has been observed down to 0.4 K.

Keywords: $CePt_3X$ Compounds; Magnetic ordering; Specific heat; Electrical resistivity; Metamagnetism

$CePt_3Si$ is known as a heavy fermion superconductor crystallizing in a structure without inversion center of the unit cell, in which the long-range antiferromagnetic ordering ($T_N = 2.2$ K) coexists with the heavy fermion superconductivity ($T_C = 0.7$ K) [2]. Concerning the crystallochemical trends within the periodic table, a very interesting aspect is the non-existence of the hypothetic $CePt_3$ cubic phase, whereas its analogues $CeNi_3$ and $CePd_3$ do form; only the low-symmetry variant $CePt_3B$-type (space group $P4\,mm$) has been proposed in [1] and recently confirmed by a single-crystal X-ray diffraction [2]. This structure can be derived from the cubic $CeAu_3$-type by filling the pyramidal cavities formed of the Pt_5 polyhedron by a *p*-metal atom, which causes a tetragonal distortion. These interesting features have encouraged us to synthesize two potential isostructural compounds $CePt_3X$, where X = Al or Ge, together with the already known $CePt_3B$ to discuss the influence of *p*-metal species on the crystal structure and the magnetic properties.

$CePt_3X$,X = B,Al, Si and Ge samples have been prepared by arc melting high purity constituents (3N Ce, 4N Pt, 6N Si, Ge, 5N Al) in an Ar protective atmosphere (6N) using several different procedures. First, two different amounts (approximately 2 and 3.5 g) of melt have been introduced to compare an influence of mass on the homogeneity of final product. Half of each sample (wrapped in a tantalum foil) was then sealed under vacuum in a quartz tube, annealed for 8 days at 850 °C and slowly cooled down to room temperature.

Microprobe analysis showed a perfectly homogeneous bulk for samples prepared from the smaller-mass melt, while small precipitates of non-diluted metallic phases have been observed in the other case. The resulting stoichiometry was determined as 1:3:1 within an experimental error. Powder X-ray diffraction measurements have been performed on both as cast and annealed samples using Seifert diffractometer (Cu Kα -radiation with a monochromator to reduce the high fluorescence of Ce) in a 2θ range 20–120° at a room temperature. While the diffraction pattern of the as cast $CePt_3Si$ sample showed only the one phase (yielding values of lattice parameters are in a good agreement with [2]), the patterns of $CePt_3B$, $CePt_3Al$ and $CePt_3Ge$ revealed significant splitting of diffraction peaks into several distinct lines. According to previous observations [1], we have ascribed this feature to a partial distribution of the *p*-metal into intersticial positions. Although the diffraction patterns of annealed samples have shown fair reduction of splitting of the diffraction lines, the proposed tetragonal structure type has been confirmed only

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in the case of B compound. For $CePt_3Al$ and $CePt_3Ge$ the significant splitting of diffraction lines persisted indicating a possibility of forming a new crystal structure with lower symmetry than that of tetragonal.

The electrical resistivity and the specific heat were measured in the temperature range 0.4–330 K on PPMS 9T and PPMS 14T devices (Quantum Design) using standard PPMS options.

A change of slope at 2.2 K, reflecting an AF ordering, followed by a rapid decrease of resistivity to 0 at 0.75 K has been observed on the temperature dependence of the electrical resistivity (R) of $CePt_3Si$ (not shown) in a good agreement with [2]. The $R(T)$ curve for $CePt_3Al$ exhibits a broad maximum at around 4 K, while for $CePt_3Ge$ a pronounced step-like decrease at around 3 K (indicating AF ordering) is observed followed by a change of slope at around 0.5 K, respectively (see Fig. 1). Moreover, both the $R(T)$ dependencies reflect a Kondo-like behavior reflected in a significant increase of the resistivity with decreasing temperature below 30 K. The $R(T)$ curve observed for $CePt_3B$ shows a significant decrease at around 7 K together with a slender step on the $R(T)$ curve at around 1 K (see Fig. 1). No indication of superconductivity was observed for the three compounds at temperatures down to 0.4 K.

The magnetoresistance in $CePt_3Si$ measured at 0.45 K reflects the suppression of the SC state at approximately 3 T which is in a good agreement with [2]. While $CePt_3Al$ exhibits a rather small decrease of resistivity in fields increasing up to 5 T (at 0.6 K), $CePt_3Ge$ at 0.5 K shows a pronounced negative magnetoresistance as expected for an antiferromagnet together with a step-like metamagnetic transition located at 1 T as seen Fig. 2. Two magnetoresistivity steps (at 1 and 6 T, respectively) observed for $CePt_3B$ at 0.5 K can be ascribed to a cascade of metamagnetic transitions, similarly as in the case of other non-superconducting $CePt_3X$ compounds. Unfortunately, the lack of magnetization data below 2 K prevents us to discuss the origin of the observed transitions more in detail. The microscopic aspects of this metamagnetic behavior deduced from the magnetoresistance measurements should be studied by a detailed neutron diffraction experiment.

The specific heat of as cast and annealed samples clearly reflects a dramatic heat treatment effect. While all the observed anomalies are fixed on the temperature scale, their character is very much modified, as shown in Fig. 3. The low-temperature specific heat of $CePt_3Al$ shows two anomalies, at 1.9 and 0.7 K, respectively. The former one seems to be due to the para-to antiferromagnetic transition as deduced from effects induced by magnetic fields (see Fig. 4). The latter corresponds to a possible order-to-order magnetic phase transition. While both peaks are practically equivalent, placed on a broad bump on the $C(T)$ curve of the as cast sample, a huge redistribution of magnetic entropy is clearly visible on the $C(T)$ dependence measured for the annealed sample. The $CePt_3Ge$ exhibits several discontinuities of the specific heat below 4 K, as shown in Fig. 4. The anomaly below 2 K

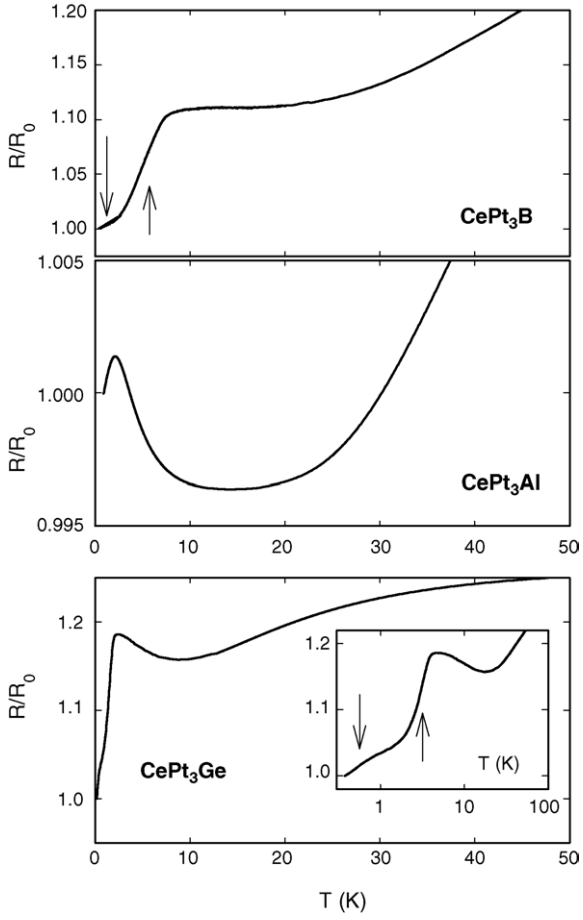

Fig. 1. Relative electrical resistivities $R(T)/R_0$ (R_0 represents the el. resistivity at 0.4 K) of $CePt_3X$, X = B, Al, Ge compounds. The inset on the last figure shows the low-temperature detail of the curve of $CePt_3Ge$, the arrows mark the magnetic phase transitions at inflection points of curves.

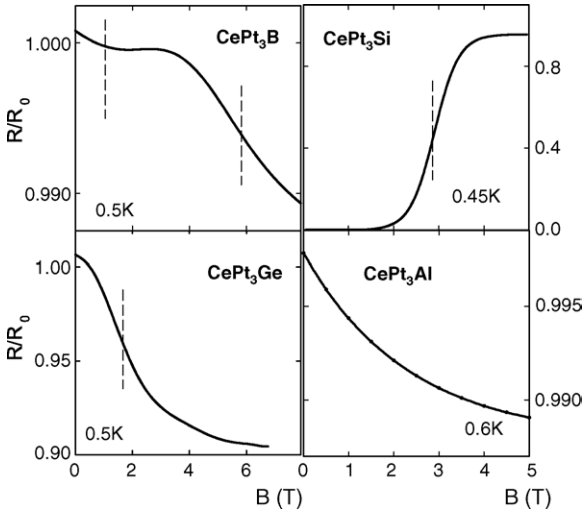

Fig. 2. Low-temperature $R(B)$ dependencies for $CePt_3X$, X = B, Al, Si and Ge compounds. The dashed lines mark the collapse of superconductivity in $CePt_3Si$ and possible metamagnetic phase transitions at inflection points of the $R(B)$ curves for other compounds, respectively.

shows a prompt of splitting for the as cast sample, which is then suppressed by annealing yielding the sharp symmetric peak similar to the corresponding one in the $CePt_3Al$ sample, while the higher lying one is surprisingly more clearly separated on the annealed sample. The low temperature $C(T)$ dependence of as cast $CePt_3B$ differs much from the specific heat behavior of the annealed sample, which is in agreement with $C(T)$ data presented in [1]. Such dramatic effect can be ascribed to a pronounced redistribution of the B atoms in the lattice, which is consistent with the annealing effect on the X-ray diffraction pattern. The anomaly at around 7 K marks the N'eel temperature whereas the feature found around 1.8 K probably reflects an order-to-order magnetic phase transition. An additional transition from the antiferromagnetic to a canted ferromagnetic state can be deduced from magnetization measurements. The sign of the transition at T_C can be recognized as a small shoulder around 5.5 K on the $C(T)$ curve. On the other hand, we still cannot exclude a possibility of presence of a non-negligible amount of impurity becoming ferromagnetic at 5.5 K and causing this feature. The clarify the origin of the observed ferromagnetic component microscopic measurements, e.g. neutron scattering experiment are strongly required. Finally, slightly enhanced values of the Sommerfeld γ coefficients were found: ~70, ~80 and ~110 mJ/molK^2 for Al, B and Ge compound, respectively.

In conclusion, we have prepared 4 compounds of $CePt_3X$ composition, with X = B, Si, Al, Ge, and studied both the as cast and annealed samples by electrical resistivity and specific heat as functions of temperature and magnetic field. Except for $CePt_3Si$, which was found to behave in agreement with [2], no sign of superconductivity at temperatures down to 0.35 K have been indicated in our data. A possible reason for

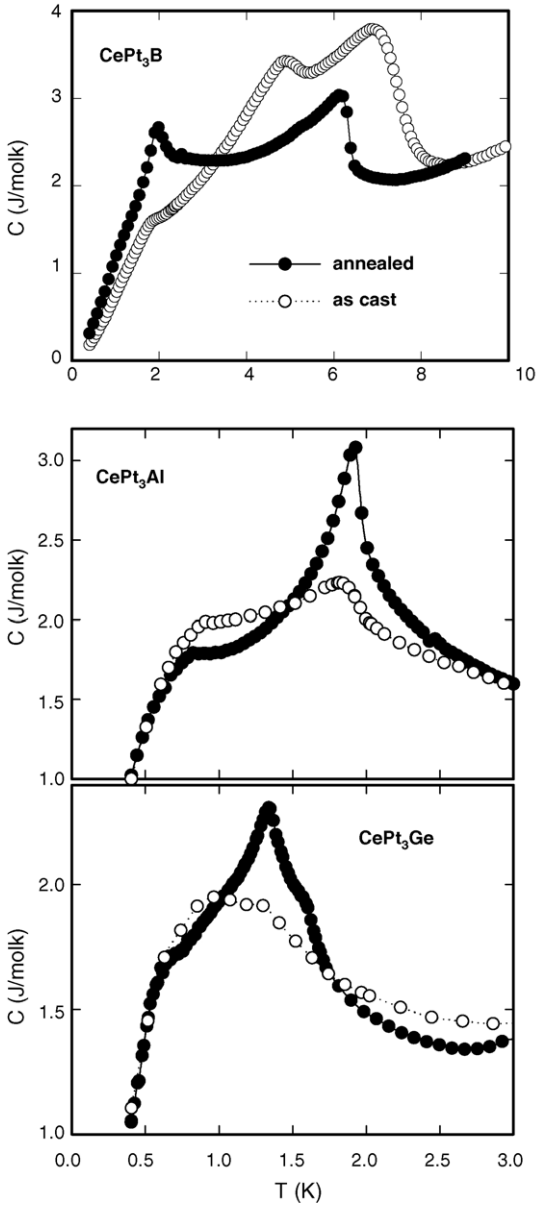

Fig. 3. The dramatic annealing effect on the specific heat of $CePt_3X$, X = B, Al, Ge compounds.

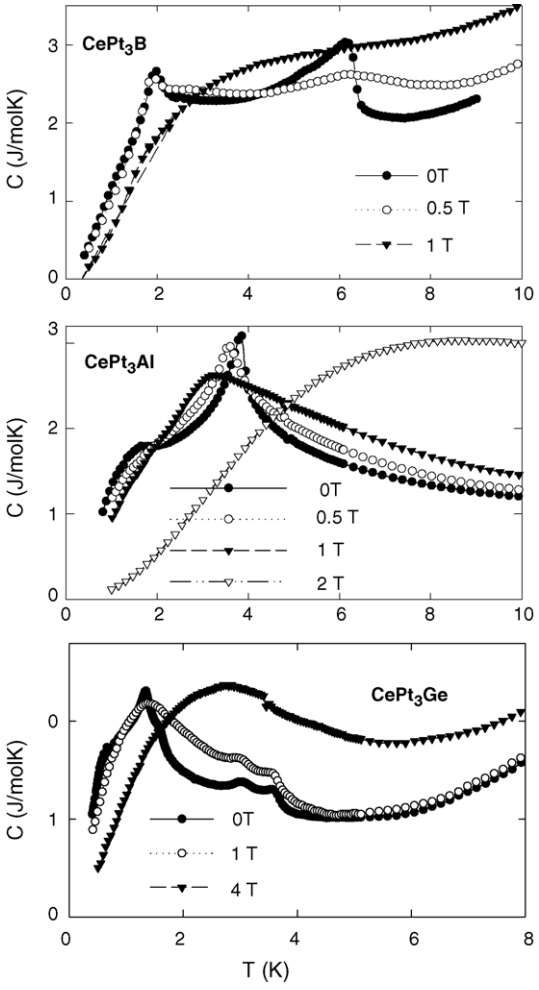

Fig. 4. C(T) of $CePt_3X$, X = B, Al, Ge compounds in several magnetic fields reflecting the AF magnetic ordering.

the lack of superconductivity may be either the pronounced structural disorder in the novel $CePt_3Al$ and $CePt_3Ge$ compounds or formation of a new structure-type. If the role of *p*-metal is only to stabilize the low-symmetry variant of the hypothetic $CePt_3$ structure, the electron–electron interactions in the CeT_3 sublattice would play a prominent role governing the cooperative phenomena. To support this assumption, we plan to synthesize structurally rigid samples of all $CePt_3X$ compounds using the ultravacuum Solid State Electrotransport technology to enable further structural and electronic properties studies. The rather complex low-temperature magnetic behavior of all studied compounds will be further investigated by already arranged neutron diffraction experiment.

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References

[1] S. Sullow, et al., Physica B 199 (1994) 644.
[2] E. Bauer, et al., Phys. Rev. Lett. 92 (2004) 027003.