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Effect of Th doping on superconductivity in $CePt_3Si$

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ABSTRACT

Alloys from the solid solution $Ce_{1-x}Th_xPt_3Si$ ($x = 0.0$, 0.02, 0.04, 0.08, 0.1, 0.2 and 1.0) were prepared by arc-melting. X-ray Rietveld powder analyses revealed that alloys in the compositional range $0 \le x \le 0.2$ crystallize with the $CePt_3B$-type with a random distribution of Ce and Th atoms in positions *1(b)* (1/2, 1/2, z) of the noncentrosymmetric space group *P4mm*. Th-doping results in a rapid suppression of the superconductivity. The alloy with $x = 0.02$ shows the onset of superconducting state at $T_c = 400$ mK, while that with $x = 0.04$ remains in normal metallic state at least down to 70 mK.

1. Introduction

In heavy fermion superconductors, Cooper pairs may have either spin singlet or spin triplet configuration and as these Cooper pairs consist of heavy particles carrying the supercurrent, the upper critical field H_{c2} may attain large values [1]. Many of the heavy fermion systems show weak magnetic order or are in the proximity of a magnetic instability where magnetic fluctuations may be responsible for Cooper pairing. While time reversal invariance is a precondition for spin singlet pairing, spin triplet pairing requests an inversion center. Therefore the discovery of heavy fermion superconductivity in $CePt_3Si$ ($CePt_3B$-type), a material without inversion symmetry [2], has spurred scientific interest in the pairing mechanism itself as well as in the physical behavior of nonmagnetic superconductors without inversion symmetry.

The present paper is concerned with the solid solution $Ce_{1-x}Th_xPt_3Si$ ($0 < x < 1$), which crystallizes with the $CePt_3B$-type, and is designed to monitor the dependency of superconducting behavior on the replacement of cerium (magnetic ground state Ce^{3+}) by thorium, a nonmagnetic element of almost identical size.

2. Experimental details

Polycrystalline alloys $Ce_{1-x}Th_xPt_3Si$ ($x = 0.0$, 0.02, 0.04, 0.08, 0.1, 0.2 and 1.0), each with a total weight of about 1 g, were prepared by repeated arc-melting appropriate amounts of master alloys $CePt_3Si$ and $ThPt_3Si$ which were arc melted from elemental constituents (Ce-ingot, 99.9% mass%, Auer Remy; Th-ingot: 99.9 mass%, Si-pieces: 99.99 mass%, Alfa Ventron). The syntheses were carried out on a water-cooled copper hearth under Ti-gettered high purity argon. Insignificant weight losses generally were below 0.4 mass%. Parts of the melted buttons were vacuum-sealed in quartz capillaries and annealed at 760 °C for up to 7 days prior to quenching in cold water. As-cast and annealed specimens were used for X-ray powder diffraction investigations.

Crystal structure identification and determination of precise lattice parameters and standard deviations were performed by least squares refinement of the room temperature X-ray powder diffraction data, collected on a Guinier-Huber Image Plate Recording System with CuKα1 radiation employing 99.9999 mass% pure Ge ($a_{Ge} = 0.5657906$ nm) or Si ($a_{Si} = 0.5431065$ nm) as internal standards. Quantitative refinements of the atom positions were done using the FULLPROF program [3].

Magnetic measurements were performed on polycrystalline samples in the temperature range 1.72–600 K and in magnetic fields up to 5 T using a Quantum Design MPMS-5 SQUID magnetometer. The electrical resistivity was measured in the temperature range 4.2–300 K employing a conventional dc four-point technique. In these measurements the electrical leads were attached to bar-shaped polycrystalline specimens by silver-epoxy paste. The electrical resistivity was measured in the temperature interval 0.07–8 K and in applied magnetic fields up to 2 T using a conventional ac four-point technique and a home-made setup installed inside a Cryogenics 3He/4He dilution refrigerator. Heat capacity measurements were carried out in the temperature range 0.35–10 K using a thermal relaxation method implemented in a Quantum Design PPMS platform.

3. Results and discussion

3.1. Crystal structure of alloys $Ce_{1-x}Th_xPt_3Si$

Guinier X-ray powder intensity profiles of the alloys from the series $Ce_{1-x}Th_xPt_3Si$ were all indexed completely on the basis of a primitive tetragonal unit cell for $0 \le x \le 0.2$. X-ray powder intensities with absence of systematic extinctions and the unit cell dimensions suggest isotypism with the tetragonal noncentrosymmetric $CePt_3B$-type (space group P4*mm*). Assuming an atomic arrangement corresponding to the $CePt_3Si$ structure a satisfactory

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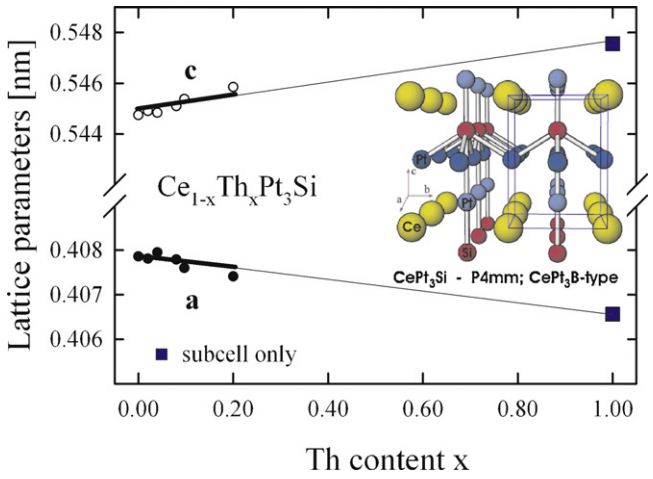

Fig. 1. Compositional dependence of the lattice parameters and the unit cell volume in the $Ce_{1-x}Th_xPt_3Si$ system.

description of the Guinier X-ray diffraction patterns was obtained. Albeit the main reflections of the X-ray spectra of $ThPt_3Si$ are consistent with the $CePt_3B$-type, large regions of inelastic scattering on the basis of some of the Bragg-reflections do not comply with a satisfactory description of the X-ray intensities recorded and indicate a more complicated structure based on a $CePt_3B$-type subcell.

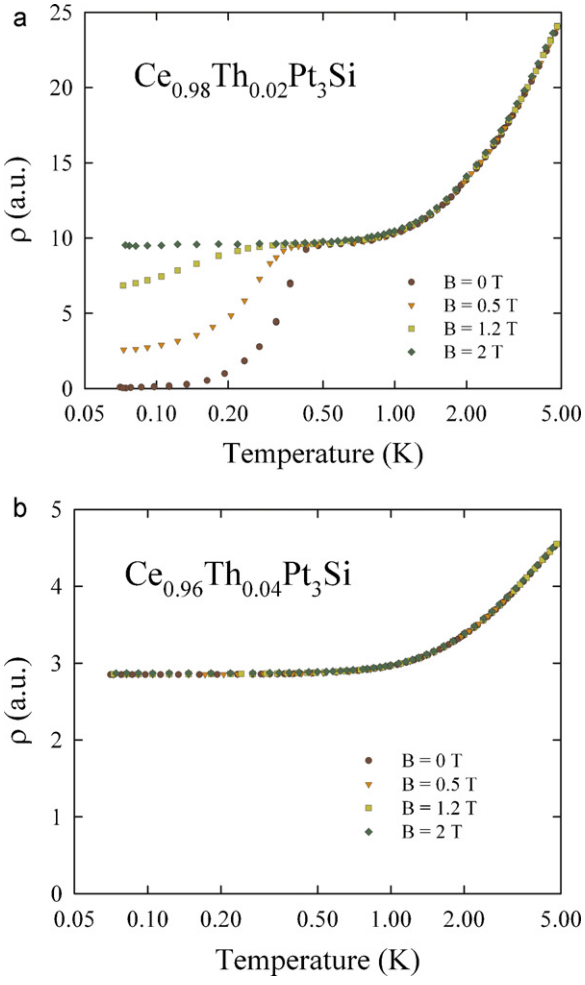

Fig. 2. Temperature variation of the electrical resistivity of (a) $Ce_{0.98}Th_{0.02}Pt_3Si$ and (b) $Ce_{0.96}Th_{0.04}Pt_3Si$ measured in a few different external magnetic fields.

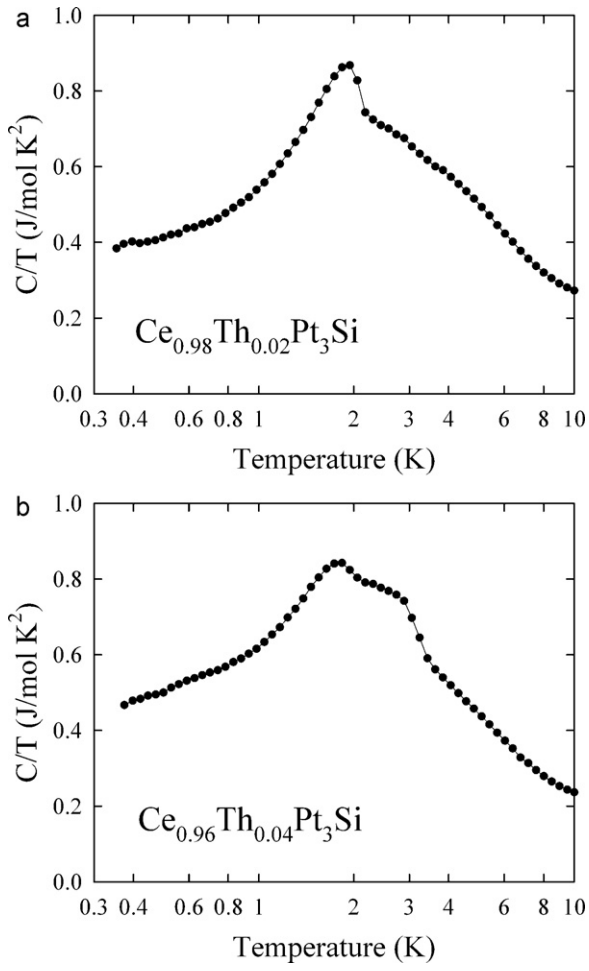

Fig. 3. Temperature variation of the specific heat over temperature ratio for (a) $Ce_{0.98}Th_{0.02}Pt_3Si$ and (b) $Ce_{0.96}Th_{0.04}Pt_3Si$.

Table 1 and Fig. 1 show the concentration-dependent lattice parameters, *a* and *c*, as a function of Ce/Th substitution, together with the unit cell volume. The monotonic decrease of both *a*, *c* parameters yields a shrinking unit cell volume upon substitution.

3.2. Physical properties of Ce-rich alloys $Ce_{1-x}Th_xPt_3Si$

Fig. 2 presents the low-temperature electrical resistivity data (in arbitrary units) of the alloys $Ce_{0.98}Th_{0.02}Pt_3Si$ and $Ce_{0.96}Th_{0.04}Pt_3Si$. Clearly, the former sample exhibits in zero magnetic field a superconducting transition at $T_c = 0.4$ K. This critical temperature is significantly reduced compared to the value of 0.75 K reported for pure $CePt_3Si$ [2]. The superconducting transition is rather broad being about 100 mK. In an applied magnetic field, T_c shifts to lower values and simultaneously the transition width notably increases. Already in a field of 0.5 T the resistivity does not drop to zero in the temperature range covered. In a field of 2 T the sample remains in a normal metallic conductivity state down to the lowest temperature reached (70 mK). On the contrary, for $Ce_{0.96}Th_{0.04}Pt_3Si$ no hint at superconductivity is observed in the temperature range accessible in this work. The specimen shows a plateau in $\rho(T)$ that should be interpreted as a residual resistivity due to scattering the conduction electrons on structural defects.

The low-temperature dependencies of the specific heat of $Ce_{0.98}Th_{0.02}Pt_3Si$ and $Ce_{0.96}Th_{0.04}Pt_3Si$ are displayed in Fig. 3. For both alloys, the dominant feature in $C/T(T)$ is a peak located at about 2 K, which manifests an antiferromagnetic ordering established for

Table 1
Crystallographic data for alloys $Ce_{1-x}Th_xPt_3Si$.

Th content x	Lattice parameters [nm]	
	a	c
0.0	0.40786(4)	0.54475(4)
0.02	0.40781(3)	0.54491(8)
0.04	0.40795(3)	0.54484(7)
0.08	0.40779(2)	0.54510(6)
0.10	0.40760(2)	0.54538(4)
0.20	0.40741(1)	0.54585(4)
1.0	0.40656(2)	0.54756(8)

the parent compound $CePt_3Si$ to set in at $T_N = 2.2$ K [2]. In turn, the superconducting phase transition, identified from the electrical resistivity data of $Ce_{0.98}Th_{0.02}Pt_3Si$ to occur at $T_c = 0.4$ K, can hardly be recognized as a tiny hump in $C/T(T)$ being of the order of the experimental error. Nevertheless, no similar anomaly is seen for the other alloy, in concert with its low-temperature normal electrical transport behavior.

As can be inferred from Fig. 3b, the specific heat of the measured sample of $Ce_{0.96}Th_{0.04}Pt_3Si$ forms an additional hump at about 3 K. This feature has probably an extrinsic nature; likely it occurs due to formation of a small amount of a Pt-rich $Ce_{1+x}Pt_{3+y}Si_{1+z}$ alloy, reported in Ref. [4] to be ferromagnetic below $T_c = 3$ K.

4. Summary

The alloys $Ce_{1-x}Th_xPt_3Si$ ($x = 0.0$, 0.02, 0.04, 0.08, 0.1, 0.2) crystallize with the $CePt_3B$-type with a random distribution of Ce and Th atoms in positions *1*(*b*) (1/2, 1/2, z = 0.1468) of the noncentrosymmetric space group *P*4*mm*. The physical properties reveal an extremely strong Cooper pair breaking effect of Th atoms substituted in a minute amount for Ce atoms in $CePt_3Si$. This finding may be considered as another indication for unconventional character of the superconductivity in the latter compound.

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