Novel superconductivity and magnetism in $CePt_3Si$ *)

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$CePt_3Si$ exhibits antiferromagnetic order at $T_N \approx 2.2$ K and superconductivity (SC) at $T_c \approx 0.75$ K. Large values of $H'_{c2} \approx -8.5$ T/K and $H_{c2}(0) \approx 5$ T indicate Cooper pairs formed out of heavy quasiparticles. The mass enhancement originates from Kondo interaction with a characteristic temperature $T_K \approx 8$ K. NMR and μSR measurements evidence coexistence of SC and long range magnetic order on a microscopic scale. Moreover, $CePt_3Si$ is the first heavy fermion SC without an inversion symmetry. This gives rise to a novel type of the NMR relaxation rate $1/T_1$ never reported before for other heavy fermion superconductors. Studies of Si/Ge substitution allow us to establish a phase diagram.

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Quantum critical fluctuations in solids can lead to strong renormalization of normal metallic properties as well as to novel exotic phases. One of the most exciting observations is the occurrence of SC with various unconventional features. A recently discovered example in this respect is tetragonal $CePt_3Si$ [1], the first heavy fermion SC without a centre of inversion. This implies non-degenerate electrons, except along some high-symmetry lines in the Brillouin zone [2]. Lifting of degeneracy, however, weakens or even suppresses SC. To obtain SC with spin-triplet pairing inversion symmetry was assumed to be prerequisite for having the necessary degenerate electron states [3]. Thus, it became a widespread view that a material lacking an inversion center would be an unlikely candidate for spin-triplet pairing

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[4]. Non-centroymmetric $CePt_3Si$, however, exhibits an extremely large value of the upper critical field ($H_{c2}(0) \approx 5$ T). This figure is inconsistent with spin-singlet Cooper-pairs, hinting at some novel features of the SC order parameter.

The aim of this paper is to highlight a number of extraordinary features found for $CePt_3Si$ and to locate the system in the standard generic phase diagram of heavy fermion compounds.

Normal state and superconducting properties of $CePt_3Si$

Fig. 1 (a) shows the magnetic contribution to the specific heat of $CePt_3Si$ displayed as $C_{mag} \sim \Delta C_p/T$ vs. T. This plot exhibits three distinct features: i) the SC transition of $CePt_3Si$ at $T_c = 0.75$ K (see below), ii) a magnetic transition at $T_N \approx 2.2$ K and iii) an almost logarithmic tail of C_{mag}/T above T_N, stretching roughly up to 10 K. Well above 10 K, Schottky contributions dominate in the specific heat. The integrated entropy up to 20 K is nearly $R \ln 2$ and reveals about 8.7 J/mol-K around 100 K. The total entropy deduced up to 100 K is slightly less than $R \ln 4$. These results clearly indicate that the ground state of Ce^{3+} ions is a doublet with the first excited level above 100 K. The logarithmic temperature

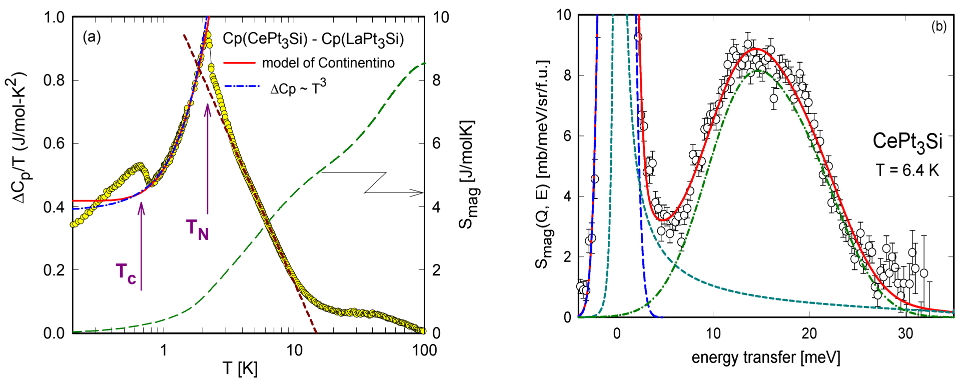

Fig. 1. (a): Temperature dependent magnetic contribution to the specific heat, ΔC_p of $CePt_3Si$ plotted as $\Delta C_p/T$ on a logarithmic temperature scale. The long-dashed line represents the magnetic entropy (right axis). The short-dashed line is a guide to the eyes and roughly indicates the non-Fermi liquid behaviour. The solid line is a fit (see text) and the dashed-dotted line is a fit according to $\Delta C_p \sim T^3$. (b): Magnetic scattering obtained at 6.4 K with the incident energy of 35 meV. The dashed line is for the elastic component while the short-dashed line represents the quasi-elastic component. The dashed-dotted line is for the sum of two Lorentzian components centered at 13 and 20 meV with FWHM=10.0 meV.

dependence observed just above T_N may be considered as hint of non-Fermi liquid behaviour. It is thus a unique finding that non-Fermi liquid behaviour, magnetic ordering and eventually a SC transition consecutively occurs in the same sample upon lowering temperature.

The magnetically ordered regime reveals antiferromagnetism, evidenced from both a model of Continentino et al. [5], as well as from a model with simple antiferromagnetic spin waves [solid and dashed-dotted lines, Fig. 1(a)]. These fits allow estimation of the Sommerfeld coefficient γ, resulting in 0.41 and 0.39 J/molK^2. Neutron elastic scattering, in fact, confirms antiferromagnetic ordering with $\mathbf{k} = (0, 0, 1/2)$ [6].

For better understanding of the pronounced paramagnetic features, evident from the heat capacity data, neutron inelastic scattering experiments were carried out at HET, ISIS (UK). Results concerning the magnetic scattering are shown in Fig. 1 (b), which can be explained in terms of a crystal field Hamiltonian with tetragonal symmetry and parameters B_2^0=-0.4972 meV, B_4^0=0.0418 meV, and B_4^4=0.2314 meV. At low energy excitations a weak feature around 1.4 meV occurs. The dispersion of that intensity at $T = 5$ K, particularly around $Q = 0.8$ Å^{-1} is a signature for the development of short-ranged magnetic correlations and explains the anomalous behaviour of the specific heat above T_N.

To study the upper critical field of CePt$_3$Si, specific heat measurements at low temperatures and at various external magnetic fields are shown in Fig. 2(a). External magnetic fields reduce T_c, giving rise to a rather large initial slope of

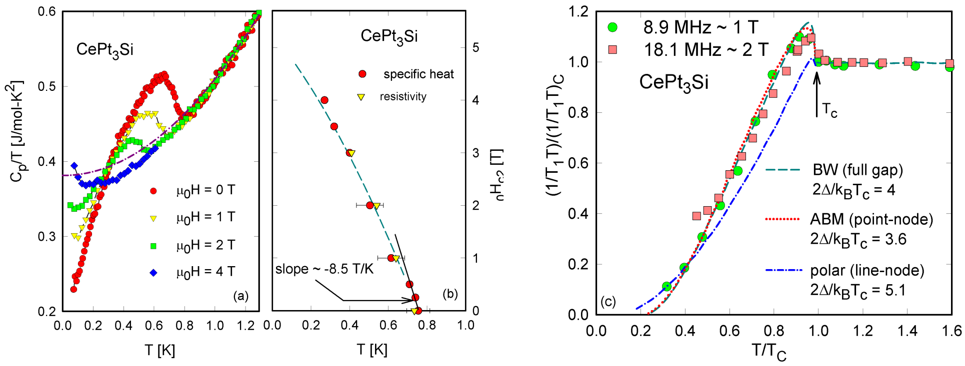

Fig. 2. (a): (b): Temperature dependent specific heat C_p/T of CePt$_3$Si for various values of applied fields; the dashed line is a T^3 extrapolation of $C_p(T)$ at 0 T. (b): Temperature dependence of the upper critical field H_{c2}. The solid straight line yields $H'_{c2} \approx -8.5$T/K; the dashed line is a guide to the eyes. (c): A plot of $(1/T_1T)/(1/T_1T)_{\mathrm{C}}$ vs T/T_c at 8.9 MHz ($H \sim 1$ T) and 18.1 MHz ($H \sim 2$ T). The solid line corresponds to the Balian-Werthamer model (BW isotropic triplet SC state, $2\Delta/k_BT_c = 3.9$); the dotted line is a fit according to a point node model with $2\Delta/k_BT_c = 3.6$; the dashed-dotted line represents a fit to a line-node gap with $2\Delta/k_BT_c = 5.1$.

$dH_{c2}/dT \equiv H'_{c2} \approx -8.5$ T/K, in good agreement with the conclusion drawn from electrical resistivity [see Fig. 2(a)]. An extrapolation of $T_c(H)$ towards zero yields $H_{c2}(0) \approx 5$ T, well above the Pauli - Clogston limiting field [1]. The upturn of C_p/T

at lowest temperatures growing with increasing magnetic fields is most likely due to the nuclear contribution of ^{195}Pt.

The jump in the specific heat $\Delta C_p/T|_{T_c} \approx 0.1$J/molK^2, leads to $\Delta C_p/\gamma_n T_c \approx 0.25$, which is much smaller than expected from the BCS theory, $\Delta C_p/\gamma T_c \approx 1.43$. Even using the electronic specific heat coefficient in the SC state, $\gamma_s \approx 0.18(1)$ J/molK^2, we obtained $\Delta C_p/\gamma_s T_c \approx 0.55$ that is still below the BCS value.

Two scenarios may well explain such a substantial reduction of $\Delta C_p/\gamma T_c$ with respect to the BCS value; i) spin triplet SC like Sr_2RuO_4 exhibits a similarily reduced magnitude of $\Delta C_p/\gamma T_c$ [7] and ii) not all electrons condense into Cooper pairs; thus only a fraction of the carriers mediates the super-current.

Microscopic evidence for the latter conclusion can be found from zero-field μSR spectroscopy data obtained in the magnetic phase below and above T_c in the magnetic phase (not shown here). High statistic runs indicate that the whole sample orders magnetically. This points to a novel state for SC Ce-based heavy-fermion systems at ambient pressure, for which, to date, magnetism was found to be either absent [8] or strongly competing against SC [9]. The observed coexistence is reminiscent of the situation observed in UPd_2Al_3 [10], where a model of two independent electron subsets, localized or itinerant, was proposed in view of similar microscopic data [11].

Another microscopic information about the SC state can be obtained from the temperature dependent ^{195}Pt nuclear spin-relaxation rate $1/T_1$ [12], plotted in Fig. 2(b). The relaxation behaviour $1/T_1T$ of $CePt_3Si$ is characterized by a kind of Hebel-Slichter anomaly [13] indicating coherence effects as in the conventional BCS case. The peak height, however, is significantly smaller than that observed for conventional SC and, additionally, shows no field dependence at the 8.9 MHz ($H \sim 1$ T) and 18.1 MHz ($H \sim 2$ T) run. $(1/T_1T)$ at $H \sim 2$ T seems to saturate at low temperature, which can be attributed to the presence of vortex cores where the normal-state region is introduced. $1/T_1T$ at 8.9 MHz, however, continues to decrease down to $T = 0.2$ K, the lowest temperature accessed. Neither an exponential law, a point node model, nor a T^3 behaviour (line nodes) are sufficient to account for the data in the entire SC temperature range. Thus, we conclude that $CePt_3Si$ is the first HF SC that exhibits a peak in $1/T_1T$ just below T_c and, moreover, does not follow the T^3 law reported for most of the unconventional HF SC (see e.g., Ref. [14] and Refs. therein).

In almost all previous studies on either conventional or unconventional SC, the crystals possess an inversion center, which allows separate consideration of the even (spin-singlet) and odd (spin-triplet) components of the SC order parameter [2]. In $CePt_3Si$, however, a center of symmetry is absent. Therefore, the novel relaxation behaviour found below T_c hints at a possibly new class of a SC state being realized in noncentrosymmetric $CePt_3Si$.

Gor'kov and Rashba [16] demonstrated that in the absence of inversion symmetry the order parameter becomes a mixture of spin-singlet and spin-triplet components. A novel idea with respect to the order parameter of systems without inversion symmetry was put forward very recently by Saxena and Monthoux [17]. In their model for the case of broken inversion symmetry, the spins rotate clockand anticlockwise in the momentum space around the surface.

Evolution of magnetism and superconductivity in $CePt_3(Si, Ge)$

The primary response of a Si/Ge substitution is an enlargement of the unit cell volume, which should result in a decrease of hybridsation and a strengthening of the magnetic state of Ce.

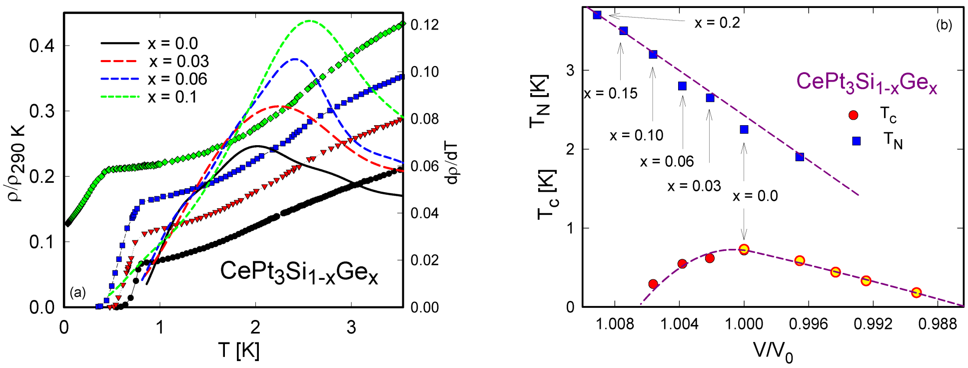

Fig. 3. (a): Low temperature electrical resistivity ρ for various concentrations x of $CePt_3Si_{1-x}Ge_x$ (symbols), and temperature derivative $d\rho/dT$ (lines, right axis). (b): Phase diagram of $CePt_3(Si_{1-x}Ge_x)$.

The electrical resistivity ρ of $CePt_3(Si_{1-x}Ge_x)$ is shown for several concentrations x in Fig. 3 (a) for temperatures below about 4 K. $\rho(T)$ increases due to increasing Si/Ge substitutional disorder which also drives a suppression of superconductivity. A magnetic instability becomes obvious from a $d\rho/dT$ plot (lines, Fig. 3, right axis). The respective transition temperatures increase continuously with growing Ge content, in good agreement with specific heat data [18].

The phase diagram shown in Fig. 3(b) summarizes characteristic temperatures deduced for $CePt_3(Si_{1-x}Ge_x)$, as well as data derived from resistivity studies on $CePt_3Si$ under hydrostatic pressure [19]. To compare chemical substitution with hydrostatic pressure, Murnaghan's equation of state is adopted, assuming a bulk modulus $B_0 = 1000$ kbar. 15 kbar of pressure corresponds then with a reduction of the unit cell volume of about 1 %. The phase diagram resembles very well those characteristics associated with a quantum phase transition. The increase of the unit cell volume by the Si/Ge substitution is responsible for a lowering of hybridisation. This has two consequences: i) the Kondo interaction decreases and ii) magnetic interaction strengthens, causing the observed increase of the magnetic transition temperature. Concomitantly, the SC transition temperature is reduced and finally, SC vanishes beyond $x > 0.1$. Increasing hydrostatic pressure is responsible for a decrease of both T_N and a decrease of T_c. Hence, CePt$_3$Si is situated at the maximum position of the "superconducting dome". Whether or not this SC dome is constrained within the magnetic phase is still unknown. Depending on the particular choice of B_0, i.e. smaller or larger than $B_0 = 1000$ kbar the volume below $V/V_0 = 1$ becomes strechted or narrower. In any case, T_c^{max} is well below the magnetic phase line, being a signature that Cooper pairing may be mediated by magnetic fluctuations rather than by the standard phonon mechanism.

We summarize that non-centrosymmetric CePt$_3$Si is a heavy fermion SC with $T_c = 0.75$ K that orders magnetically at $T_N = 2.2$ K. The NMR relaxation rate $1/T_1$ shows unexpected features which were found before neither in conventional nor in heavy fermion SC, indicative of very unusual shapes of the SC order parameter. The Si/Ge substitution simply drives a volume expansion, thus magnetism is stabilized and SC finally ceases to exist.

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