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Unconventional superconductivity and magnetism in $CePt_3Si_{1-x}Ge_x$

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Abstract

$CePt_3Si$ is a novel ternary compound exhibiting 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 NMR relaxation rate $1/T_1$ which is very unique and never reported before for other heavy fermion superconductors. Studies of Si/Ge substitution allow us to establish a phase diagram.

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Keywords: $CePt_3Si$; Superconductivity; Antiferromagnetism

1. Introduction

Strongly correlated electron systems have been one of the most interesting topics in condensed

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matter physics. The key importance in such research is undoubtedly the discovery of unexpected features and new phases of metals, intermetallics and oxides at low temperatures. Among the topics, quantum phase transitions and related quantum critical phenomena are of particular importance. Quantum critical fluctuations can lead to strong renormalization of normal metallic properties as well as to novel exotic phases emerging from these strongly fluctuating environments. One of the most exciting features in this context is the occurrence of superconductivity (SC).

A recently discovered example is tetragonal $CePt_3Si$ [1], the first heavy fermion SC without a centre of inversion. This implies that the electron bands are non-degenerate, except along some high-symmetry lines in the Brillouin zone [2]. Reduced degeneracy, however, weakens or even suppresses SC. Superconductivity with spin-triplet pairing should require inversion symmetry to obtain the necessary degenerate electron states [3]. Thus, it became a widespread view that a material lacking an inversion centre would be an unlikely candidate for spin-triplet pairing [4]. Nevertheless, the extreme large value of the upper critical field $H_{c2}(0) \approx 5\,\mathrm{T}$ of $CePt_3Si$ is inconsistent with spin-singlet Cooper-pairs, hinting at some novel features of the SC order parameter.

The aim of this paper is to provide a review of current research on $CePt_3Si$ and to locate the system in the standard generic phase diagram of heavy fermion compounds through both substitution and pressure experiments. The paper is organized as follows: After a discussion of normal state properties of $CePt_3Si$, the SC features of $CePt_3Si$ are examined before reporting the evolution of magnetism and SC in $CePt_3(Si, Ge)$.

2. Normal state properties

Physical properties of ternary $CePt_3Si$ are dominated by long range magnetic order below $T_N \approx 2.2\,\mathrm{K}$ and SC below $T_c = 0.75\,\mathrm{K}$. Crystal electric field (CEF) splitting and Kondo interaction substantially modify Hund's $J = \frac{5}{2}$ ground state of the Ce ion. The response of the system associated with the mutual interplay of these phenomena will be highlighted below.

Fig. 1 displays $\Delta C_p/T$ vs. T of $CePt_3Si$, where ΔC_p is defined by the difference between the $C_p(T)$ data of $CePt_3Si$ and $LaPt_3Si$, i.e. $C_{mag} \sim \Delta C_p$. This plot exhibits three distinct features: (i) the SC transition of $CePt_3Si$ at $T_c = 0.75\,\mathrm{K}$ (see below), (ii) a magnetic transition at $T_N \approx 2.2\,\mathrm{K}$ and (iii) an almost logarithmic tail of $\Delta C_p/T$ 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 the entropy of $8.7\,\mathrm{J/mol\,K}$ integrated up to 100 K is slightly less than $R \ln 4 = 11.5\,\mathrm{J/mol\,K}$. 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 dependence observed just above the magnetic transition may be considered as hint of non-Fermi liquid behaviour. Therefore, it is a unique observation that non-Fermi liquid behaviour, magnetic ordering and eventually a SC transition consecutively arises on the same sample upon lowering temperature.

In order to analyse in more detail the magnetically ordered region of the system, a model by Continentino [5] is applied with the following

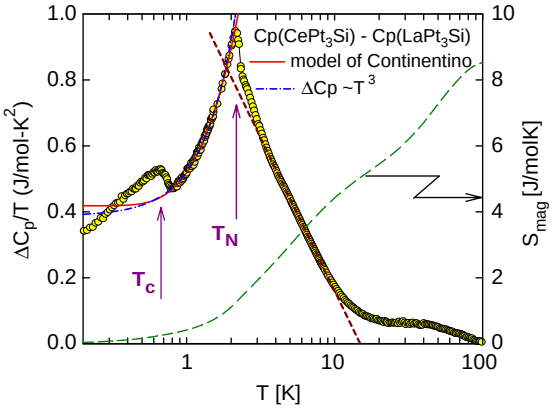

Fig. 1. Temperature dependent magnetic contribution to the specific heat, ΔC_p of $CePt_3Si$ plotted as $\Delta C_p/T$ on a logarithmic temperature scale. The long-dashed line represents the magnetic entropy (right axis). The short-dashed line is a guide to the eyes and roughly indicates the non-Fermi liquid behaviour. The solid line is a fit according to Eq. (1) and the dashed-dotted line is a fit according to $\Delta C_p \sim T^3$.

analytic expression for the specific heat well below T_{mag}:

$$C_{mag} = g\Delta_{SW}^{7/2} T^{1/2} \exp(-\Delta_{SW}/T) \times \left[1 + \frac{39}{20}\left(\frac{T}{\Delta_{SW}}\right) + \frac{51}{32}\left(\frac{T}{\Delta_{SW}}\right)^2\right]. \quad (1)$$

This expression is based on antiferromagnetic magnons with a dispersion relation given by $\omega = \sqrt{\Delta_{SW}^2 + D^2k^2}$, where Δ_{SW} is the spin-wave gap and D is the spin-wave velocity; $g \propto 1/D^3 \propto 1/\Gamma^3$, and Γ is an effective magnetic coupling between Ce ions. A least-squares fit of Eq. (1) to the data below T_N (solid line, Fig. 1) reveals $\Delta_{SW} \approx 2.7$ K, a reasonable gap value with respect to the ordering temperature. Another model calculation with simple antiferromagnetic spin waves with $C_{mag} \propto T^3$ gives reasonable agreement with the data, too. A recent neutron diffraction study on $CePt_3Si$ reveals antiferromagnetic ordering below $T_N \approx 2.2$ K with a wave vector $\mathbf{k} = (0, 0, \frac{1}{2})$, i.e. doubling of the magnetic unit cell along **c**-direction [6]. Using both models, we estimate a Sommerfeld coefficient of 0.41 and 0.39 J/mol K^2 for the former and latter models, respectively. These figures are in good agreement with an extrapolation of high-field specific heat data where SC is suppressed by applying magnetic fields.

Considering Kondo type interactions to be responsible for the significant renormalization of electrons in $CePt_3Si$ as evidenced by the Sommerfeld value γ, the magnetic entropy allows to estimate the Kondo temperature T_K. Taking the results derived in Ref. [7] yields $T_K \approx 7.2$ K. A second possible estimate T_K follows from the competition of the RKKY interaction and the Kondo effect, which leads to a significant reduction of the specific heat jump at $T = T_N$. Following the procedure developed in Ref. [8] gives $T_K \approx 9$ K, in reasonable agreement with the previous estimate.

For better understanding of the magnetic ground state and expected localized character of Ce 4f electrons, inelastic neutron scattering experiments at the HET spectrometer of ISIS, UK, were carried out. In order to accurately and reliably determine magnetic scattering from the magnetic moments of Ce, both $CePt_3Si$ and $LaPt_3Si$ were investigated in powder form under identical conditions. For phonon subtraction, we used two well-established methods [9], with almost the same results. Our main finding is that there are strong and clear features from 10 to 30 meV. As shown in Fig. 2 scattering intensity ought to be of magnetic origin as the phonon contributions have been subtracted. In order to explain the data, we used the CEF Hamiltonian for Ce^{3+} with C_{4v} point symmetry: $H_{CEF} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^4 O_4^4$. With $B_2^0 = -0.4972$ meV, $B_4^0 = 0.0418$ meV, and $B_4^4 = 0.2314$ meV, the observed magnetic scattering is reasonably well explained (solid line, Fig. 2). Keeping the CEF parameters unchanged, we could also explain the data obtained at 94 K equally well. Moreover, the two CEF excitations centred at 13 and 20 meV are also consistent with the heat capacity data as discussed above. We furthermore studied low-energy excitations using lower incident energy to find a weak feature around 1.4 meV. 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 magnetic ordering. At higher

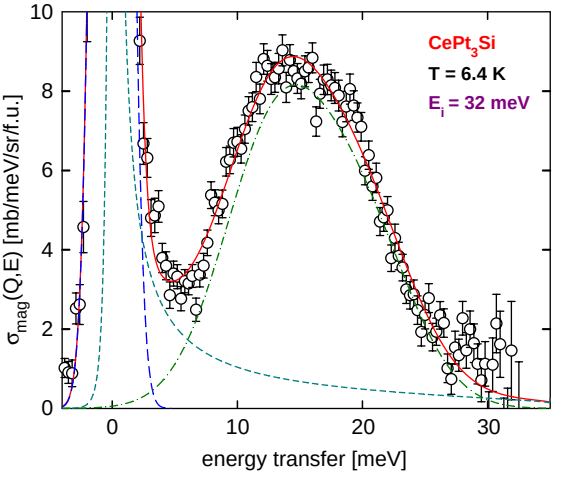

Fig. 2. Magnetic scattering obtained at 6.4 K with the incident energy of 35 meV. The dashed line is for the elastic component with FWHM = 2.4 meV while the short-dashed line represents the quasi-elastic component with FWHM = 0.8 meV. The dashed-dotted line is for the sum of two Lorentzian components centred at 13 and 20 meV with FWHM = 10.0 meV.

temperatures ($T \approx 30\,\mathrm{K}$) scattering becomes Q-independent. This feature is completely absent in non-magnetic $LaPt_3Si$. We note that a recent inelastic neutron scattering experiment [6] reported two CEF peaks at 1.0 and 24 meV. However, our data show that their second excitation is most likely to be mistaken.

3. Superconducting properties of $CePt_3Si$

Signs of bulk SC of $CePt_3Si$ below $T_c = 0.75\,\mathrm{K}$ are numerous: zero resistivity, diamagnetic signal in the susceptibility, a jump in the specific heat and NMR relaxation rate at T_c. Here, we show in Fig. 3(a) the field-dependent specific heat at low temperatures and at various external magnetic fields. Sommerfeld coefficient $\gamma_n \approx 0.39\,\mathrm{J/mol\,K^2}$ of $CePt_3Si$ at zero field can be obtained from a careful extrapolation of the normal state invoking the T^3 dependence associated with antiferromagnetic ordering. This extrapolation also satisfies the basic requirement of an entropy balance between the SC and normal state regions.

The application of magnetic fields reduces T_c, giving rise to a rather large change of $dH_{c2}/dT \equiv H'_{c2} \approx -8.5\,\mathrm{T/K}$, in good agreement with the conclusion drawn from electrical resistivity [see Fig. 3(b)]. An extrapolation of $T_c(H)$ towards zero yields $H_{c2}(0) \approx 5\,\mathrm{T}$, well above the Pauli–Clogston limiting field [1]. Furthermore, an estimation of the Sommerfeld coefficient from the high field data gives $0.36\,\mathrm{J/mol\,K^2}$, in fair agreement with the value obtained from an extrapolation of the normal state in the zero field data (see Fig. 3). The upturn of C_p/T at lowest temperatures that gets stronger with increasing magnetic fields is most likely due to the nuclear contribution of ^{195}Pt.

The jump in the specific heat $\Delta C_p/T|_{T_c} \approx 0.1\,\mathrm{J/mol\,K^2}$, 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, $\gamma_s \approx 0.18(1)\,\mathrm{J/mol\,K^2}$, we obtained $\Delta C_p/(\gamma_s T_c) \approx 0.55$ that is still below the BCS value.

We can think of two scenarios that may well explain such a substantial reduction of $\Delta C_p/\gamma T_c$ with respect to the BCS value; (i) spin triplet SC's like Sr_2RuO_4 exhibit a similarily reduced magnitude of $\Delta C_p/(\gamma T_c)$ [10] and (ii) not all electrons condense into Cooper pairs; thus, only a fraction of the carriers mediate the super-current. It implies that electrons responsible for normal state features, like antiferromagnetic order, coexist with those forming the Cooper pairs. In fact, the finite value of $\gamma_s \approx 0.18\,\mathrm{J/mol\,K^2}$ provides evidence that even at $T = 0\,\mathrm{K}$ a significant portion of the Fermi surface is still not involved in the SC condensate.

Microscopic evidence for the latter conclusion can be found from zero-field μSR spectroscopy

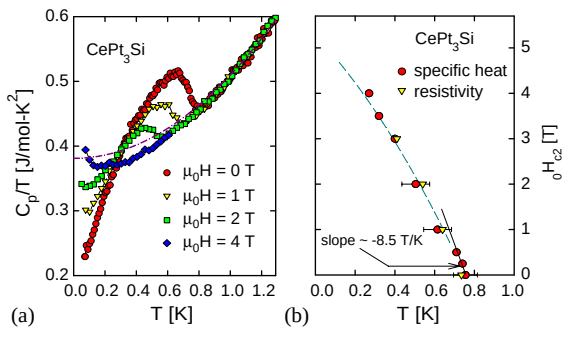

Fig. 3. (a) Temperature-dependent specific heat C_p/T of $CePt_3Si$ for various values of applied fields; the dashed line is a T^3 extrapolation of $C_p(T)$ at 0 T. (b) Temperature dependence of the upper critical field H_{c2}. The solid straight line yields $H'_{c2} \approx -8.5\,\mathrm{T/K}$; the dashed line is a guide to the eyes.

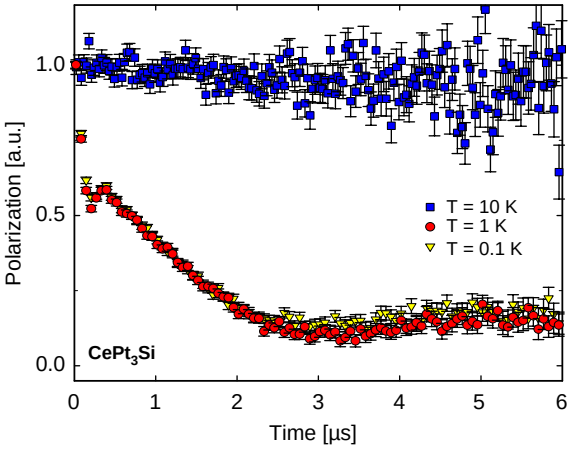

Fig. 4. Zero-field depolarization rate of $CePt_3Si$ at various temperatures.

data obtained in the magnetic phase below and above T_c in the magnetic phase. (Fig. 4). At temperatures much above T_N, the μSR signal is characteristic of a paramagnetic state with a depolarization solely arising from nuclear moments. Below T_N the μSR signal indicates that the full sample volume orders magnetically. High statistic runs performed above and below T_c did not show any change of the magnetic signal, supporting the view of a microscopic coexistence between magnetism and SC. 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 [11] or strongly competing against SC [12]. The observed coexistence is reminiscent of the situation observed in UPd_2Al_3 [13], where a model of two independent electron subsets, localized or itinerant, was proposed in view of similar microscopic data [14].

Another microscopic information about the SC state can be obtained from the temperature dependent ^{195}Pt nuclear spin-relaxation rate $1/T_1$ [15]. Results are shown as $(1/T_1T)_{SC}/(1/T_1T)_N$ vs. T/T_c plot in Fig. 5 for 8.9 and 18.1 MHz. The relaxation behaviour $1/T_1T$ of $CePt_3Si$ is characterized by a kind of the Hebel–Slichter anomaly

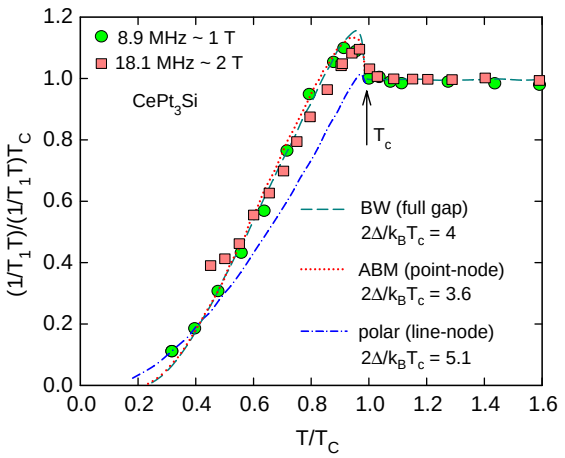

Fig. 5. A plot of $(1/T_1T)/(1/T_1T)_{T_c}$ vs. T/T_c at 8.9 MHz ($H \sim 1$ T) and 18.1 MHz ($H \sim 2$ T). The dashed line is for the Balian–Werthamer model (BW isotropic triplet SC state) with a value of $2\Delta/k_BT_c = 4$. The dotted line assumes a point-node model with $2\Delta/k_BT_c = 3.6$ and the dashed-dotted line represents a fit by a line-node gap model with $2\Delta/k_BT_c = 5.1$.

[16] indicating coherence effects as in conventional BCS SC. The peak height, however, is significantly smaller than that observed for conventional BCS 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 ($H \sim 1$ T), however, continues to decrease down to $T = 0.2$ K, the lowest measured temperature. Neither an exponential law nor a T^3 behaviour is observed for the data down to $T = 0.2$ K. Therefore, $CePt_3Si$ 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. [17] and references therein).

To account for the relaxation behaviour below T_c in non-centrosymmetric $CePt_3Si$, three models were adopted for a description of the temperature dependence of $1/T_1$ at $H \sim 1$ T. The dashed line in Fig. 5 represents a fit according to the Balian–Werthamer model (isotropic spin-triplet SC state) with a value of $2\Delta/k_BT_c = 3.9$ [18], while the dashed-dotted line is a fit using a line-node model with $2\Delta/k_BT_c = 5.1$. The dotted line refers to a point-node model with $2\Delta/k_BT_c = 3.6$. The models used, however, failed to give satisfactory description of the observed temperature dependence of $1/T_1$ over the entire temperature range. While the line-node model gives a reasonable agreement with the data at lowest temperatures, the BW model describes reasonably well the data just below T_c. The peak in $1/T_1T$ would indicate the presence of an isotropic energy gap, even though a coherence effect—inherent for the isotropic spin-singlet s-wave pairing state—is absent.

In almost all previous studies on either conventional and unconventional SC, it was assumed that the crystal has an inversion centre, which allows separate consideration of the even (spin-singlet) and odd (spin-triplet) components of the SC order parameter. In $CePt_3Si$, however, a centre 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 non-centrosymmetric $CePt_3Si$.

Gorkov and Rashba [19] demonstrated that in the absence of inversion symmetry the order parameter becomes a mixture of spin-singlet and spin-triplet components, which leads, for instance, to the Knight shift attaining a non-zero value at $T = 0$ K. A novel idea with respect to the order parameter of systems without inversion symmetry was put forward very recently by Saxena and Monthoux [20]. In their model for the case of broken inversion symmetry, the spins might rotate in the momentum space around the surface.

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

In order to follow the evolution of physical properties upon Si/Ge substitution, we prepared several alloys. Guinier X-ray powder intensity profiles of the alloys from the series $CePt_3(Si_{1-x}Ge_x)$ with $x = 0.02$, 0.03, 0.06, 0.10, 0.15 and 0.20 were all indexed completely on the basis of a primitive tetragonal unit cell, confirming isotypism with the structure type of $CePt_3Si$ [21] with a random substitution of the Si/Ge atoms in the 1a sites of space group P4/mmm. Small secondary peaks arise beyond $x \approx 0.2$ indicating the limit of the $CePt_3B$ type phase region. Fig. 6 shows the concentration-dependent lattice parameters a and c as a function of Si/Ge substitution, together with the unit cell volume. The monotonic increase of both a and c parameters yields a growing unit cell volume upon substitution. The general increase of the unit cell volumes reduces chemical pressure on the Ce atoms leading to a decrease of hybridization.

The temperature dependent electrical resistivity ρ of $CePt_3(Si_{1-x}Ge_x)$ is shown for several concentrations x in Fig. 7 for temperatures below about 4 K. The overall $\rho(T)$ values increase due to increasing substitutional disorder on the 1a site. Samples investigated are characterized by a concentration dependent onset of SC, where the Si/Ge substitution suppresses the SC roughly above $x = 0.1$. At somewhat elevated temperatures, $\rho(T)$ exhibits a pronounced curvature which can be taken as a signature for long range magnetic order, in agreement with specific heat measurements. A magnetic instability is evidenced from the resistivity data in terms of a $d\rho/dT$ plot (lines, Fig. 7, right axis). $d\rho/dT$ shows pronounced anomalies, referring to broadened magnetic phase transitions; the respective temperatures increase continuously with increasing Ge content.

The magnetic contribution to the specific heat of $CePt_3(Si_{1-x}Ge_x)$ is plotted in Fig. 8 as C_{mag}/T vs. T for several compositions up to $x = 0.2$ from 1.8 to 20 K. C_{mag} is derived as in Fig. 1 using the measured data of isostructural non-magnetic $LaPt_3Si$. A common feature in all the data sets is a mean-field-like anomaly, which is associated with magnetic order of the cerium sublattice. T_N

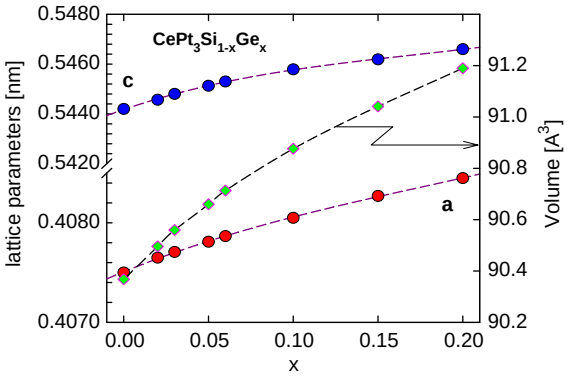

Fig. 6. Concentration dependent lattice parameters a, c and unit cell volume V of $CePt_3Si_{1-x}Ge_x$.

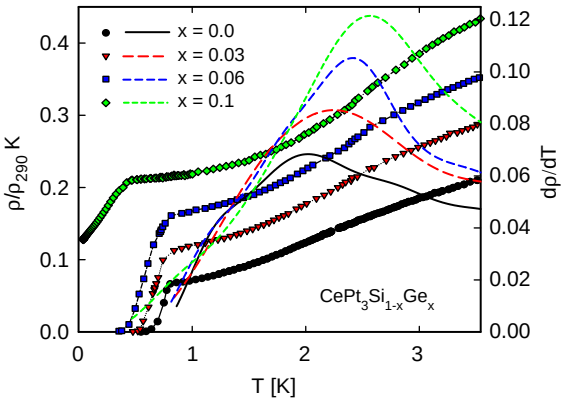

Fig. 7. Low temperature electrical resistivity ρ for various concentrations x of $CePt_3Si_{1-x}Ge_x$ (symbols), and temperature derivative $d\rho/dT$ (lines, right axis).

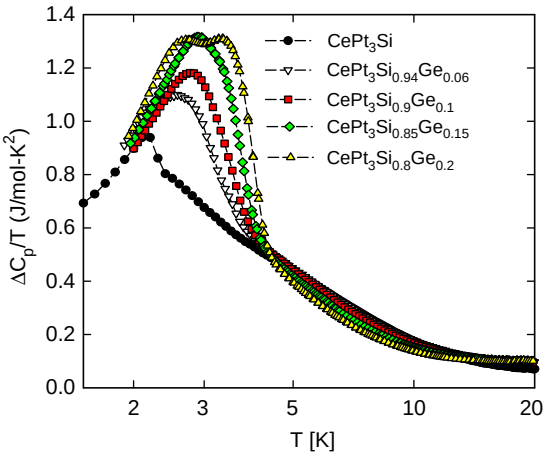

Fig. 8. Magnetic contribution to the specific heat ΔC_p plotted as C_p/T vs. T for various alloys of $CePt_3Si_{1-x}Ge_x$.

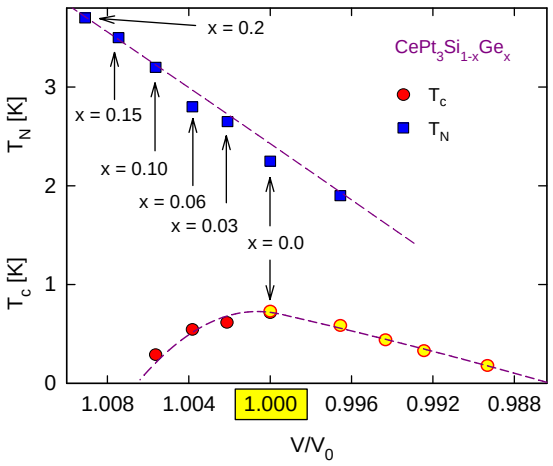

Fig. 9. Phase diagram of $CePt_3(Si_{1-x}Ge_x)$.

rises with increasing Ge content and, simultaneously, the phase transitions appear to be broadened, in agreement with the observation made from $d\rho/dT$. While for $x = 0$, the logarithmic behaviour well above T_N is quite pronounced and obvious, its temperature range becomes narrower with increasing Ge content as a consequence of increasing magnetic interactions, driving a magnetic instability at much higher temperatures. The double-peak feature for $x = 0.2$ is most likely due to a secondary phase as indicated by our XRD analysis.

The phase diagram shown in Fig. 9 summarizes characteristic temperatures deduced for $CePt_3(Si_{1-x}Ge_x)$ at ambient pressure, as well as data derived from resistivity studies on $CePt_3Si$ under hydrostatic pressure up to about 15 kbar [22]. In order to make comparison between substitution and pressure, Murnaghan's equation of state is adopted, with a bulk modulus $B_0 =$ 1000 kbar. Pressure of 15 kbar corresponds then to a reduction of the unit cell volume of about 1%. The phase diagram thus deduced resembles very well those characteristics, which determine the standard generic phase diagram associated with a quantum phase transition. The increase of the unit cell volume by the Si/Ge substitution is, in a first-order approximation, responsible for a decrease of hybridization. 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 becomes suppressed and finally, SC vanishes beyond $x > 0.1$. Increasing hydrostatic pressure is responsible for a decrease of both T_N and T_c. Ternary $CePt_3Si$ thus appears to be, by chance, 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 stretched 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 $CePt_3Si$ 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. In fact, a number of theoretical scenarios support these observations. The Si/Ge substitution simply drives a volume expansion, thus magnetism is stabilized and SC finally ceases to exist.

Acknowledgements

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