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LETTER TO THE EDITOR

Magnetic structure and the crystal field excitation in heavy-fermion antiferromagnetic superconductor CePt$_3$Si

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Abstract

Neutron scattering experiments have been carried out on the heavy fermion antiferromagnetic (AFM) superconductor CePt$_3$Si with $T_\mathrm{N} = 2.2$ K and $T_\mathrm{SC} = 0.75$ K. We observed clear AFM Bragg reflections with $\boldsymbol{Q_0} = (001/2)$ below and above T_SC, indicating microscopic coexistence of AFM order and heavy fermion superconductivity. The AFM structure, of two interleaved ferromagnetic sublattices of local Ce 4f moments, has inversion symmetry under simultaneous space–time reversal. However, hybridization with Pt and Si breaks this degeneracy and a combination of these two competing effects may be relevant to an understanding of the simultaneous occurrence of superconductivity and AFM order. The observed magnetic moment 0.16(1) μ_B/Ce is strongly reduced from the Curie–Weiss effective moment 2.54 μ_B/Ce. Clear crystal field excitations at 1 and 24 meV were observed. The magnetic susceptibility can be well explained in a level scheme assuming the Γ_7 ground state, Γ_6 and Γ_7 first and second excited states, respectively.

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

Heavy fermion superconductivity as observed in 4f compounds of Ce [1–6] and Pr [7, 8], as well as in the 5f U [9, 10] and Pu [11] electron systems, continues to be a field of intense interest in the study of strongly correlated electron systems. Very recently, Bauer *et al* [12] reported that CePt$_3$Si, with a non-centrosymmetric structure (space group $P4mm$) as shown

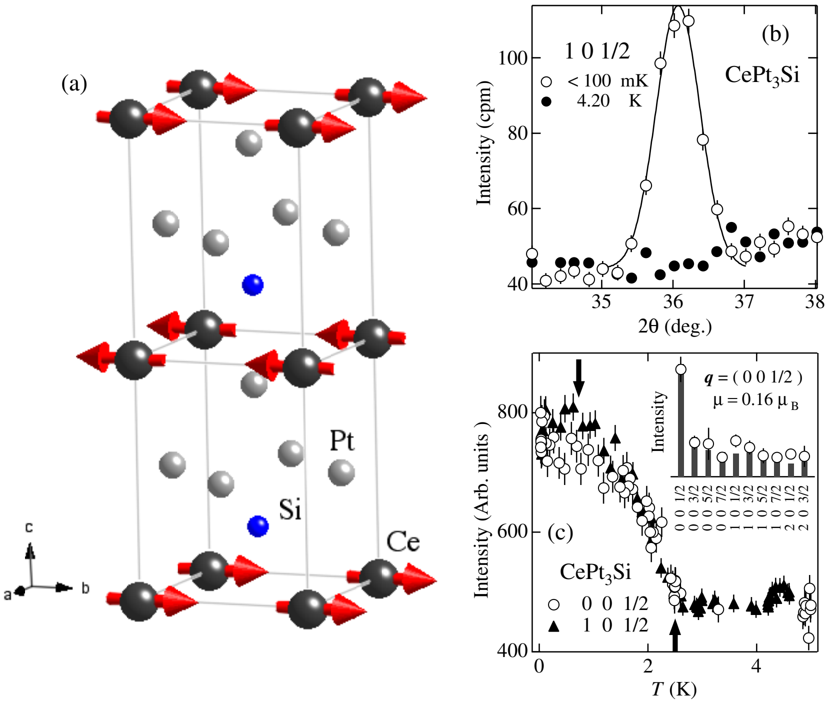

Figure 1. (a) Crystal and magnetic structure of $CePt_3Si$. The arrows on the Ce atom indicate the magnetic moment lying, with unspecified orientation, in the basal c plane. (b) The (101/2) AFM Bragg reflection observed below 0.1 K (open circles) and the background measured at 4.2 K (solid circles). (c) The intensity of (001/2) and (101/2) magnetic reflection as a function of temperature, shown by open circles and solid triangles, respectively. Up and down pointing arrows indicate T_N and T_{SC}, respectively. The inset is the observed integrated intensity (open circles) compared with model calculation (solid bars), assuming the magnetic multi-domain structure in (a).

in figure 1, exhibits antiferromagnetic (AFM) order at $T_N = 2.2$ K and enters into a heavy fermion superconducting state at $T_{SC} \approx 0.75$ K. The large Sommerfeld constant in normal and superconducting states, $\gamma_n = 0.39$ J K^{-2} mol^{-1} and $\gamma_s = 0.18$ J K^{-2} mol^{-1}, respectively, and the T^2 coefficient in resistivity $A = 2.35\ \mu\Omega$ cm K^{-2}, indicate a Fermi liquid state with significant renormalization due to electron correlation. The large $\mathrm{d}H_{c2}/\mathrm{d}T \approx -8.5$ T K^{-1} and $H_{c2} \approx 5$ T suggest that Cooper pairs form out of the heavy quasi-particle state. Bauer *et al* argued that there is a conflicting situation; a lack of spatial inversion centre in the chemical unit cell favours spin singlet pairing, while the large H_{c2}, which exceeds the estimated Pauli–Clogston limiting field, might be a signature for spin triplet pairing. It was suggested that a mixed spin singlet and triplet pairing state might be the answer to the apparent paradox. In this letter, we present the results of our neutron scattering study in order to clarify the magnetic structure, the nature of f electronic levels and crystalline electric field (CEF) parameters.

Polycrystalline samples were synthesized by arc-melting, whereas single crystals were obtained by the pulling method and mineralization. The AFM and superconducting transitions were confirmed by resistivity, specific heat and susceptibility measurements. Neutron scattering experiments were carried out on triple-axis spectrometers TAS-1, TAS-2 and LTAS installed at the JRR-3 reactor of the Japan Atomic Energy Research Institute, JAERI. Elastic scattering was measured with neutrons of energy $E = 14.7$ meV, monochromatized and analysed with pyrolytic graphite crystals, while inelastic scattering was measured at fixed final

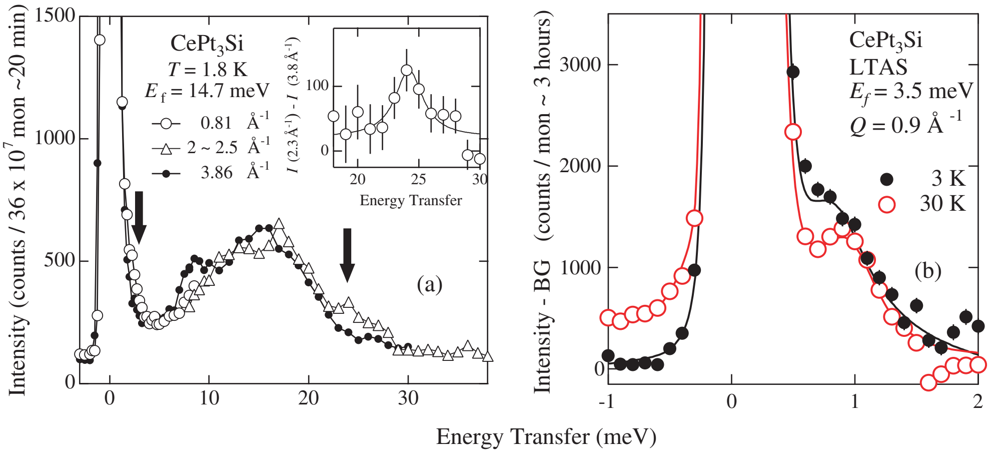

Figure 2. (a) Neutron inelastic scattering spectra of polycrystalline $\mathrm{CePt_3Si}$. The inset is the spectrum obtained by subtracting the high-$\boldsymbol{Q}$ data from the low-$\boldsymbol{Q}$ data. (b) High resolution neutron inelastic scattering spectra of polycrystalline $\mathrm{CePt_3Si}$. The solid and open circles represent data measured at 3 and 30 K, respectively.

energies of $E_\mathrm{f} = 3.5$, 14.7 and 30.5 meV. The magnetic susceptibility was measured using a commercial SQUID magnetometer.

We observed clear (001/2) and (101/2) superlattice peaks from the polycrystalline sample below T_N at $T = 1.8$ K, giving an AFM vector $\boldsymbol{Q_0} = (001/2)$. The magnetic structure was further studied with a single crystal sample. Representative data at (101/2) are shown in figure 1(b) where a clear AFM peak measured at $T < 0.1$ K disappears at $T = 4.2$ K. Figure 1(c) shows the temperature dependence of the (001/2) and (101/2) intensities with normalizing the intensity and the background to the same level. The AFM intensity is almost constant below T_SC, as marked by the down pointing arrow in figure 1(c), indicating that the AFM order coexists with the heavy fermion superconducting state.

The integrated intensities of the AFM peaks in the $(h0l)$ scattering plane are plotted in the inset of figure 1 (c). We found that the intensity can be well explained with a model calculation assuming ferromagnetic sheets of Ce moments stacked antiferromagnetically along the c-axis as shown in figure 1. The magnetic moment of $0.16 \pm 0.01\ \mu_\mathrm{B}$/Ce is strongly suppressed from the effective moment of $\mathrm{Ce^{3+}}$ ion 2.54 μ_B/Ce. This reduction is consistent with a presence of Kondo-like interaction [12]. The direction of the magnetic moment lying in the c-plane could not be determined because of the domain structure.

Figure 2 gives the inelastic scattering spectra of $\mathrm{CePt_3Si}$ measured with $E_\mathrm{f} = 14.7$ meV. The major inelastic feature, which is slightly enhanced at the larger momentum transfer, may arise from the phonon density of states. Of interest here are the two peaks at $\Delta E \approx 1$ and 24 meV in the low-$\boldsymbol{Q}$ data. On subtracting the high-$\boldsymbol{Q}$ from the low-$\boldsymbol{Q}$ data a clear peak at 24 meV appears, the existence of which was confirmed by scan at $E_\mathrm{f} = 30.5$ meV. The low energy excitation can be fitted satisfactorily assuming the inelastic response at $\Delta E \approx 1.3$ meV and a quasi-elastic response normalized with a Bose factor and convoluted with resolution function. The intensity of low energy excitation around $\Delta E \approx 1$ meV, falls with increasing $\boldsymbol{Q}$ and sample temperature. The enhancement of the peaks at low-$\boldsymbol{Q}$, together with their observation in a polycrystalline sample, suggests that they arise from van-Hove-like singularities in the magnetic excitation spectrum. Specifically we explore the consequence of attributing them to the crystalline electric field (CEF) excitations of the lowest Ce multiplet.

Table 1. CEF parameters, energy levels and wavefunctions for CePt$_3$Si.

CEF parameters

B_2^0 (meV)	B_4^0 (meV)	B_4^4 (meV)	λ (emu/mol)$^{-1}$	χ_0 (emu mol^{-1})
−0.103	−0.0679	0.304	−37	$\chi_0^{[100]} = 1.5 \times 10^{-4}$ $\chi_0^{[001]} = 0.5 \times 10^{-4}$

Energy levels and wavefunctions

E (meV)	$\lvert -5/2\rangle$	$\lvert -3/2\rangle$	$\lvert -1/2\rangle$	$\lvert +1/2\rangle$	$\lvert +3/2\rangle$	$\lvert +5/2\rangle$
24.0	−0.3661	0	0	0	−0.9306	0
24.0	0	−0.9306	0	0	0	−0.3661
1.0	0	0	1	0	0	0
1.0	0	0	0	1	0	0
0	0.9306	0	0	0	−0.3661	0
0	0	−0.3661	0	0	0	0.9306

The excitation energies $\Delta E \approx 1$ and 24 meV are consistent with a step-like behaviour observed in the electrical resistivity around 10 and 100 K [12]. The low energy excitation spectra of polycrystalline CePt$_3$Si at $Q = 0.9$ Å^{-1} for $T = 3$ and 30 K with $E_\mathrm{f} = 3.5$ meV are shown in figure 2(b). Note that the background has been subtracted. A clear inelastic peak at $\Delta E \approx 1$ meV at $T = 30$ K ($\approx 10 \times T_\mathrm{N}$) is found to coexist with a quasi-elastic response, where the quasi-elastic response increases in intensity with decreasing temperature. The broad nature around $\Delta E \approx -1$ meV is the anti-Stokes signal of the inelastic response. The solid curves indicates the model calculation, where the intensity of the CEF peak was assumed to be proportional to the population of the ground state, while the energy and intrinsic width were essentially fixed about 1 and 0.4 meV. The agreement of the model calculation is quite good. The deviation of the spectra for $\Delta E \approx 2$ meV is due to a slight shift of the background level. A low-energy CEF excitation $\approx$1 meV is consistent with the broad specific heat anomaly around $T = 2$ K [12], which integrates to an entropy change of $R \ln 2$ at $\approx$25 K.

We have made an analysis of the $\chi(T)$ data using a CEF model. The CEF Hamiltonian for Ce^{3+} ($J = 5/2$) with C$_{4\mathrm{v}}$ point symmetry can be expressed as

$$\mathcal{H}_\mathrm{CEF} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^4 O_4^4, \tag{1}$$

where the c-axis is the quantization axis, O_j^i are the Stevens operators, and B_j^i the CEF parameters. Due to the CEF, the six-fold degenerate 4f-levels are split into three set of doublets, a ground state with first and second excited levels, 1 and 24 meV, respectively, as observed in the neutron inelastic scattering spectra. In the analysis of the magnetic susceptibility, we assumed an isotropic exchange interaction λ for simplicity, and a temperature-independent Pauli paramagnetic susceptibility χ_0 for each axis. The magnetic susceptibility is thus expressed as

$$\chi = \chi_0 + \frac{\chi_\mathrm{CEF}}{1 - \lambda \chi_\mathrm{CEF}}. \tag{2}$$

Solid lines in figure 3 are the best fit curves calculated with the CEF parameters listed in table 1. The deviation of the fitting curves from the experimental data below 50 K may be due to a substantial Kondo screening at low temperatures [12]. The ground state is Γ_7, and the first and second excited states are Γ_6 and Γ_7, respectively. The lower two doublets originate from a Γ_8 quartet in the cubic point symmetry.

The magnetic structure of the CePt$_3$Si may be relevant in connection with discussions of the pairing symmetry in this compound. Bauer and co-workers pointed out the possibility

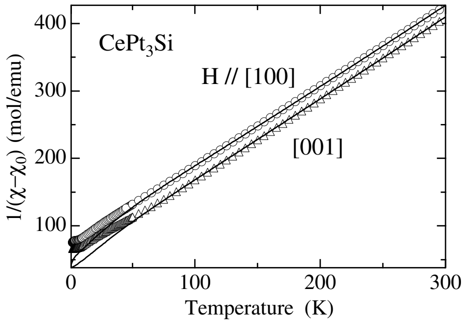

Figure 3. Temperature dependence of magnetic susceptibility of CePt$_3$Si. The solid lines show the calculated susceptibility with the CEF Hamiltonian.

that the pairing symmetry could be a mixed spin singlet and triplet state. The magnetic propagation vector $\boldsymbol{Q_0} = (001/2)$ implies two interpenetrating, Néel magnetic sublattices which are ferromagnetic in the c-plane. Within a localized model of the Ce 4f moment, on simultaneous space–time inversion a given magnetic sublattice is centrosymmetric and a pairing of spin-triplet character may be favourable. However, hybridization with Pt and Si breaks this degeneracy and a combination of these two competing effects may be relevant to an understanding of the microscopic coexistence of the superconductivity and AFM order.

A reduced, but sizeable, magnetic moment 0.16(1) μ_B/Ce, which coexists with heavy fermion superconductivity, is a surprising result in Ce-based compounds. For example, CeCu$_2$Si$_2$ is believed to show some kind of dynamical AFM order with tiny moment of the order of $10^{-2}\mu_B$/Ce [13]. No neutron data are available for CePd$_2$Si$_2$, CeIn$_3$, or the CeTIn$_5$ series in superconducting phases under pressure. On comparison with the magnetic and superconducting T–p (p; pressure) phase diagram of CeRh$_2$Si$_2$ [2, 14], one may speculate that a moment up to ~0.5 μ_B/Ce could coexist with superconductivity. However, the character of the 4f electrons under pressure is not understood.

The observation of crystal field excitations in a heavy fermion superconductor is unusual. It raises a naive question, do the 4f states have an itinerant or localized character? In the simplest case, no clear crystal field excitation should be observable in a Kondo-lattice system in which f levels are strongly hybridized with conduction states, and possess an itinerant character expressed as a renormalized heavy Fermi liquid phase. In this vein, no crystal field excitation has been reported in CeCu$_6$ [15] or CeRu$_2$Si$_2$ [16], instead a quasi-elastic response, the so-called Kondo resonance peak is superposed on the AFM fluctuations as the relevant low energy excitation. It has been reported that a broad CEF excitation can be observed in the heavy fermion superconductor CeCu$_2$Si$_2$ [20, 21]; again its ordered state is unclear yet.

An example of a heavy fermion superconductor which shows clear CEF excitations is PrOs$_4$Sb$_{12}$ [8]. The Fermi surfaces of PrOs$_4$Sb$_{12}$ are understood in terms of a localized 4f band structure [17]. Very recently a remarkable low energy excitation was observed [18] at $\boldsymbol{q} = (100)$ which corresponds to the propagation of the field induced antiferroquadrupole order [19]. It is now of intense interest to establish the low lying modes relevant to the heavy quasi-particles and superconductivity as achieved in UPd$_2$Al$_3$ [22, 23].

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