Coexistence of magnetism and superconductivity in the heavy-fermion superconductor CePt₃Si

A. Amato,[1,*] E. Bauer,[2] and C. Baines[1]

[1]*Laboratory for Muon-Spin Spectroscopy, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland*
[2]*Institut für Festkörperphysik, Technische Universität Wien, A-1040 Wien, Austria*
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The interplay between magnetism and superconductivity in the heavy-fermion superconductor CePt₃Si has been investigated using the zero-field muon-spin relaxation (μSR) technique. The μSR data indicate that the whole muon ensemble senses spontaneous internal fields in the magnetic phase, demonstrating that magnetism occurs in the whole sample volume. This points to a microscopic coexistence between magnetism and heavy-fermion superconductivity.

I. INTRODUCTION

In recent years, strongly correlated electron systems have played a leading role in solid state physics. The importance of this research field is illustrated by the discovery of atypical phases in metals, intermetallics, and oxides at low temperatures. One of the most relevant examples is the discovery of unconventional superconductivity in heavy-fermion systems.

Unconventional superconductivity seems to result from the nature of the mechanism providing the attractive force necessary for the Cooper-pair formation. In conventional superconductors, the electrons are paired in a spin-singlet zero-angular-momentum state ($L=0$), which results from the fact that their binding is described in terms of the emission and absorption of phonons. This leads to the formation of an isotropic superconducting gap in the electronic excitations over the whole Fermi surface. On the other hand, heavy-fermion superconductivity is observed to show a close interplay with magnetic fluctuations. This seems to indicate that the attractive effective interaction between the strongly renormalized heavy quasiparticles in the superconducting heavy-fermion systems is not provided by the electron-phonon interaction as in ordinary superconductors, but rather is mediated by electronic spin fluctuations. This nonconventional (i.e., non-BCS) mechanism is believed to lead to an unconventional configuration of the heavy-fermion superconducting state, which may involve anisotropic, nonzero-angular-momentum states ($L \neq 0$, see Ref. 1 for a review, and references therein).

An additional feature in a number of systems is the observation of an apparent coexistence of heavy-fermion superconductivity and static magnetism. However, at ambient pressure, such a coexistence was up to recently solely confirmed on uranium-based heavy-fermion systems, and was discarded on cerium-based systems. Such conclusions were deduced from microscopic studies, in particular from the sensitive μSR technique.[2] In this frame, the heavy-fermion superconductor CeCu₂Si₂ is exemplary as it exhibits a competition between both ground states, i.e., magnetism and superconductivity do not coexist, but appear as two different, mutually exclusive ground states of the same subset of electrons. Such competition was first discovered by μSR[3,4] and only recently confirmed by neutron studies.[5]

Recently another Ce-based heavy-fermion system, namely CePt₃Si, was found[6] showing antiferromagnetism and superconductivity ($T_N = 2.2$ K and $T_c = 0.75$ K) at ambient pressure. This material crystallizes in a tetragonal structure (space group $P4mm$) lacking a center of inversion symmetry. This feature is presently attracting much interest since unconventional superconductivity with spin-triplet was to date thought to require such inversion symmetry to obtain the necessary degenerated electron states.[7,8] In this paper, we present μSR studies aiming to test—at the microscopic level—the coexistence between magnetic and superconducting state.

II. EXPERIMENT

CePt₃Si was prepared by high frequency melting and subsequently heat treated at 880 °C for one week. Phase purity was evidenced by x-ray diffraction. The μSR experiments were carried out at the Swiss Muon Source of the Paul Scherrer Institute (Villigen, Switzerland). Measurements were performed on the GPS and LTF instruments of the πM3 beamline, using a He-flow cryostat (base temperature 1.7 K) and a ^{3}He—^{4}He dilution refrigerator (base temperature 0.03 K), respectively. In order to avoid a depolarizing background μSR signal, the sample was glued onto a high-purity silver holder. Measurements on both instruments were performed on the same sample and showed a very good agreement in the overlapping temperature range. Measurements were performed in zero applied field (ZF), with an external-field compensation of the order of ± 20 mOe.

III. RESULTS AND DISCUSSION

ZF μSR is a local probe measurement of the magnetic field at the muon stopping site(s) in the sample. If the implanted polarized muons are subject to magnetic interactions, their polarization becomes time dependent, $\mathbf{P}_\mu(t)$. By measuring the asymmetric distribution of positrons emitted when the muons decay as a function of time, the time evolution of $P_\mu(t)$ can be deduced. The function $P_\mu(t)$ is defined as the projection of $\mathbf{P}_\mu(t)$ along the direction of the initial polarization: $P_\mu(t) = \mathbf{P}_\mu(t) \cdot \mathbf{P}_\mu(0) / P_\mu(0) = G(t) P_\mu(0)$. Hence, the depolarization function $G(t)$ reflects the normalized muon-spin
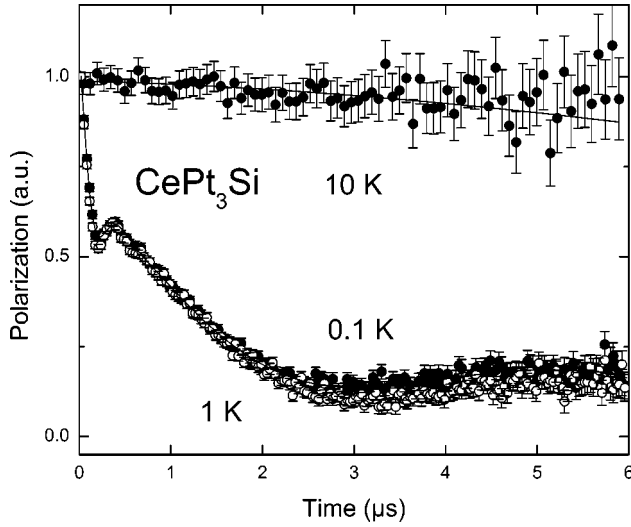
FIG. 1. Example of ZF μSR signals obtained in polycrystalline CePt$_3$Si in the paramagnetic phase (10 K), the magnetic phase (1 K), and below the superconducting transition (0.1 K). The lines represent fits obtained using Eqs. (1) and (2). Note that for clarity, the fit for the data obtained at 1 K is omitted.

FIG. 2. Temperature dependence of the spontaneous μSR frequencies ν_1 and ν_2 obtained by fitting Eq. (2) to the μSR data. The measurements were performed in a polycrystalline CePt$_3$Si sample.

autocorrelation function $G(t) = \langle \mathbf{S}(t) \cdot \mathbf{S}(0) \rangle / S(0)^2$, which depends on the average value, distribution, and time evolution of the internal fields, and therefore contains all the physics of the magnetic interactions of the muon inside the sample.[9]

Above T_N, the time evolution of the muon polarization is best described by the well-known Kubo-Toyabe function[10]

$$G^{\mathrm{para}}(t) = \frac{1}{3} + \frac{2}{3}(1 - \Delta^2 t^2)\exp\left(-\frac{\Delta^2 t^2}{2}\right), \qquad (1)$$

where Δ^2/γ_μ^2 represents the second moment of the local field distribution at the muon site (γ_μ is the gyromagnetic ratio of the muon). Such a depolarization function is characteristic of a paramagnetic state where the muon depolarization is solely due to the dipolar fields of the nuclear moments (^{29}Si and ^{195}Pt). In the paramagnetic state, the electronic magnetic moments are often not observable by μSR due to their fast fluctuation rates. Alternatively, the nuclear magnetic moments appear static within the μSR time window and create a Gaussian field distribution at the muon stopping site, leading to the Kobo-Toyabe depolarization function reported in Eq. (1). Note that this function posses an initial Gaussian character $[\simeq \exp(-\Delta^2 t^2)$ for $t \ll \Delta^{-1}]$ as observed in the data reported in Fig. 1. Fitting Eq. (1) to the data provides a depolarization rate $\Delta = 0.06$ MHz corresponding to field distribution width of ~ 0.7 G at the muon site, in line with theoretical values computed for several possible stopping sites.

Below T_N, clear spontaneous oscillations are detected in the μSR signal indicating the occurrence of static finite magnetic fields sensed by the muons and arising from static electronic magnetic moments. In the antiferromagnetic state, the μSR signal is best described by the sum of two components, i.e.:
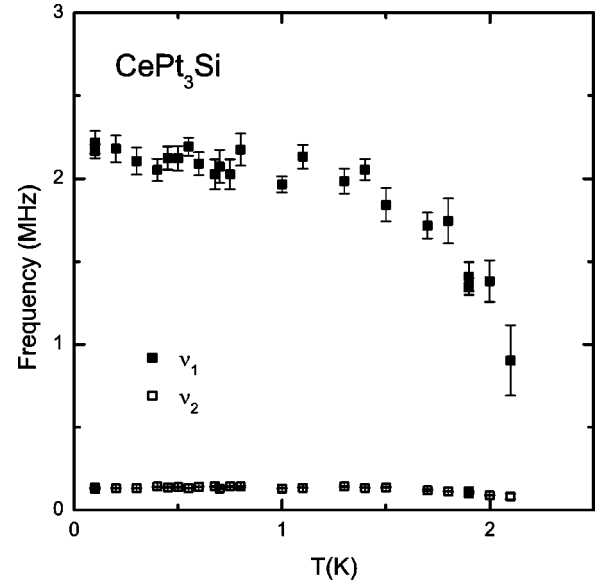
$$\begin{aligned}
G^{\mathrm{AF}}(t) = A_1 &\left[\frac{1}{3}\exp(-\lambda_1 t) + \frac{2}{3}\exp(-\lambda_1' t)\cos(2\pi\nu_1 t + \phi_1) \right] \\
+ A_2 &\left[\frac{1}{3}\exp(-\lambda_2 t) + \frac{2}{3}\exp(-\lambda_2' t)\cos(2\pi\nu_2 t + \phi_2) \right].
\end{aligned}$$
$$(2)$$

These components indicate the presence of two magnetically inequivalent muon stopping sites sensing internal fields $|\mathbf{B}_\mu^i| = 2\pi\nu_i/\gamma_\mu$. As expected for a polycrystalline sample, the "1/3 term" of each component represents the fraction of the muons possessing an initial polarization along the same direction of the internal field. Therefore, the depolarization rates related to these fractions (λ_i) reflect solely the internal spins dynamics, whereas the depolarization rates λ_i' are ascribed by both dynamical and static effects. The temperature evolution of the spontaneous frequencies ν_i are reported in Fig. 2. The values of the frequencies at $T \rightarrow 0$ correspond to internal field values of ~ 160 and 10 G, respectively.

Very recently, neutron scattering experiments determined the magnetic structure of CePt$_3$Si.[11] Magnetic Bragg reflections observed at wave vector values $\mathbf{Q} = (0, 0, 1/2)$ and $(1, 0, 1/2)$, indicate that magnetic moments align ferromagnetically in the basal plane and stack antiferromagnetically along the c axis with a strongly reduced value of 0.16 μ_B. By considering this magnetic structure, the values of the spontaneous μ^+ frequencies provide information for a tentative determination of the muon stopping sites in the tetragonal structure. By assuming the magnetic moment direction pointing along the a or b axis, the most probable muon sites are located at two different 1(b) Wyckoff-positions, i.e., $(1/2, 1/2, 0.65)$ for the low-frequency component and $(1/2, 1/2, 0.82)$ for the high-frequency component. These sites are respectively located in the center of the Pt plane formed by the Pt(1) ions and between the planes formed by
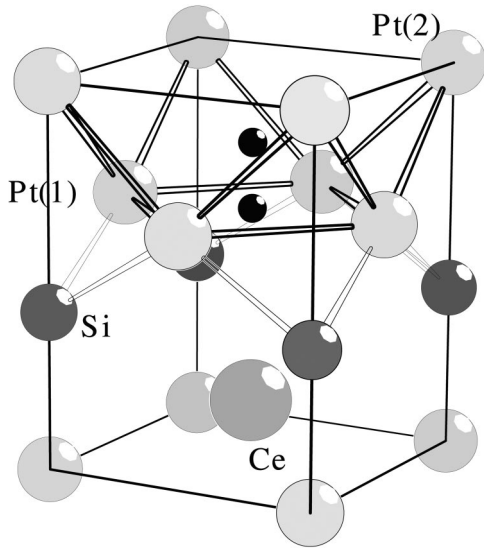
FIG. 3. Crystal structure of CePt$_3$Si. The smallest spheres represent the muon stopping sites discussed in the text.

Pt(1) and Pt(2) ions (see Fig. 3 and notation from Ref. 6). In addition, for these sites, the calculated field distributions due to nuclear dipole moments are found in reasonable agreement with the observed depolarization rate Δ observed in the paramagnetic regime [see Eq. (1)]. Note also that both sites have the same multiplicity, which is in line with the observation that $A_1 \simeq A_2$ as shown in Fig. 4.

The first relevant observation is that $A_1 + A_2 = 1$ for all temperatures below T_N. This means that the whole muon ensemble is sensing the magnetic state, which in turn unambiguously demonstrates that the *whole* sample volume is involved in the magnetic phase below T_N. Together with thermodynamical studies, demonstrating that superconductivity has a bulk character, the present observation indicates a microscopic coexistence between magnetism and superconduc-
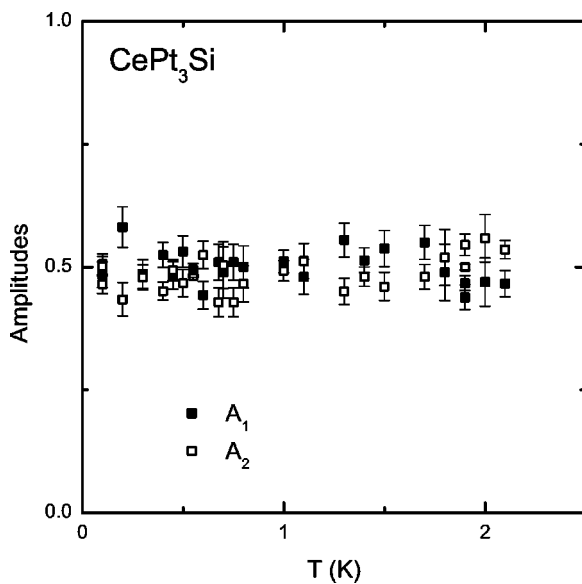

FIG. 4. Temperature dependence of the amplitudes A_1 and A_2 of the spontaneous μSR frequencies in CePt$_3$Si [see Eq. (2)].
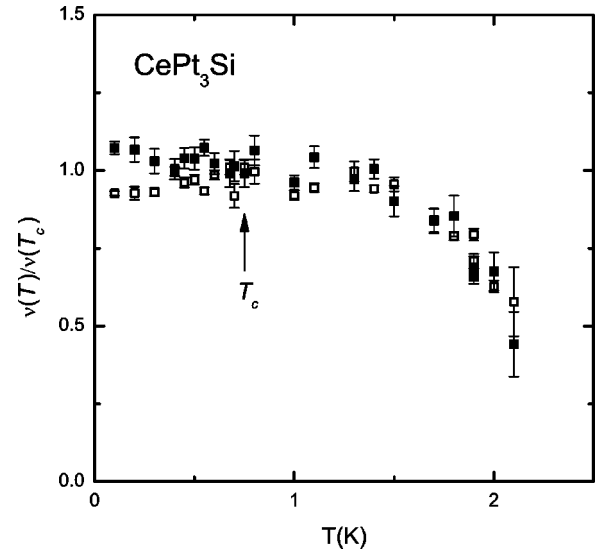

FIG. 5. Temperature dependence of the spontaneous μSR frequencies normalized to their values at $T_c = 0.75$ K. Note the very slight change below T_c. The symbols correspond to those of Fig. 2.

tivity. A similar conclusion was very recently drawn from NMR studies performed at different frequencies.[12] Note that the conclusion obtained by μSR is independent of the exact knowledge of the muon stopping sites. The behavior observed here in CePt$_3$Si is opposite to the one reported for CeCu$_2$Si$_2$ (see above), where the magnetic state is expelled from the sample upon cooling below T_c. The observed coexistence in CePt$_3$Si is reminiscent of the situation observed in U-based heavy-fermion systems, as UPd$_2$Al$_3$ (Ref. 13) or UNi$_2$Al$_3$ (Ref. 14), where a model of two independent electronic subsets, localized and itinerant (responsible for magnetism and superconductivity, respectively), was proposed in view of similar microscopic studies[13] and thermodynamics measurements.[15]

Upon cooling the system into the superconducting state, the μSR data suggest a slight change of the absolute spontaneous internal fields at the muon sites. As shown on Fig. 5, one observes, for $T < T_c$, a slight reduction and increase of the low and high frequency signals, respectively. Note that such changes are at the limit of the measurement accuracy. In any case, two possible explanations could be invoked for these changes. The first one would be to consider a coupling between the superconducting and magnetic order parameters, reminiscent to the situation observed in UPt$_3$.[16] Alternatively, the frequency changes could have a more simple origin, as the muon is sensing interstitial fields and therefore only indirectly probes the strength of the magnetic order parameter. Hence, in addition to the dipolar interaction, the static $4f$ magnetic moments will change the spin polarization of the conduction electrons at the muon site,[2] which results in an increased hyperfine field acting on the muon. Such a contribution is a function of the density of normal electron states and will therefore be affected upon cooling the sample into the superconducting state. Below T_c, one expects a decrease in absolute value due to the opening of the superconducting gap. Depending on the muon stopping site and due to the oscillatory character of the Ruderman-Kittel-Kasuya-Yosida

(RKKY) interaction between the static $4f$ moments and the conduction electrons, a decrease of the hyperfine field contribution can actually lead to either an increase or a decrease of the total internal fields at the muon site, as possibly observed in the present μSR data.

IV. CONCLUSION

Our zero-field μSR data have demonstrated the bulk character of the antiferromagnetic state in the heavy-fermion superconductor CePt$_3$Si, suggesting therefore a microscopic coexistence between magnetism and superconductivity. In addition, a slight change of the μSR response upon cooling the sample below T_c can be ascribed to a coupling of the superconducting and magnetic order parameters and/or to the decrease of the hyperfine contact contribution acting on the muon.

*Electronic address: alex.amato@psi.ch

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