$CePt_2Si_2$: A Kondo lattice compound with no magnetic ordering down to 0.06 K

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(Received 25 July 1996)

We have performed zero-field and longitudinal field muon spin experiments on single crystals of the Kondo lattice compound $CePt_2Si_2$ down to 0.06 K. The crystals have been characterized by magnetization and specific heat measurements. We do not detect any electronic magnetic signal. Our result rules out a magnetic phase transition as the origin of the non-Fermi-liquid behavior of the specific heat and electrical resistivity observed at low temperature. [S0163-1829(97)02206-6]

The Kondo lattice compound $CePt_2Si_2$ crystallizes in the $CaBe_2Ge_2$-type tetragonal structure (space group $P4/nmm$).[1] Its Kondo temperature as determined by specific heat is $T_K \simeq 70$ K.[2] At low temperature, its specific heat, electrical resistivity, and magnetic susceptibility display non-Fermi-liquid behavior.[2,3] Whereas the thermal behavior of the specific heat C and electrical resistivity ρ follow the predictions for a Fermi-liquid system in the range 4–10 K, at low temperature they strongly deviate from the expected behavior. The Sommerfeld ratio C/T increases steeply as the temperature T decreases and concomitantly ρ has an approximate linear temperature dependence.[2] It has been argued that a collapse of the magnetic susceptibility in the c plane occurs below 4 K, although this conclusion has been reached after applying a large correction to the measured susceptibility.[3] This correction was supposed to take into account an impurity contribution to the susceptibility. The low temperature dependence of the Sommerfeld ratio, resistivity, and susceptibility may reflect a low temperature magnetic phase transition. A more exciting possibility is that $CePt_2Si_2$ is an example of a non-Fermi-liquid system.[4–6]

If $CePt_2Si_2$ has a magnetic phase transition at low temperature, this phase is certainly characterized by small magnetic moments since otherwise it would have been already detected. The most efficient method to unravel such a phase transition is to perform muon spin relaxation (μSR) experiments.[7] Here we report such experiments which show that $CePt_2Si_2$ does not display any signature of static electronic magnetism. This is in contrast to most strongly correlated electron compounds for which μSR experiments have detected static magnetism.[7]

$CePt_2Si_2$ bulk materials have been prepared by direct combination of the high quality elements (Ce : 4N, Pt : 4N, and Si : 6N5). The starting elements (for a total weight of nearly 10 g) were melted in a water-cooled copper crucible heated with a high frequency generator under a purified argon atmosphere. To improve homogeneity, the bulks were turned over and remelted several times. The weight loss is negligible using this way to prepare silicides. The material was then introduced in a triarc furnace under inert gas, equipped with a Czochralski puller.[8] Two single crystals (the c axis was either along or perpendicular to the pulling axis) were grown from the same bulk starting composition using the same seed. The single crystals were checked by conventional x-ray powder diffraction and their single-crystalline state has been confirmed using the back scattering x-ray Laue technique. They have been annealed in a resistive ultrahigh vacuum furnace during 8 days at 950 °C under 3.5×10^{-10} torr. The two crystals were then cut by spark erosion. For the μSR measurements, the slices of each crystal were carefully glued to a 5N silver plate (40×40 mm^2) in such a way as to produce sample disks of ~ 25 mm diameter and ~ 0.4 mm thickness. Therefore the μSR measurements were carried out on two samples which differ by the orientation of the crystal axes relative to the sample plane: the c axis is either perpendicular or parallel to the plane. The crystals have been characterized by magnetization and specific heat measurements. All the measurements were performed on annealed crystals, except for some magnetization measurements done on as-grown crystals.

The magnetization measurements were performed with a SQUID magnetometer. In Fig. 1 we present the magnetic field dependence of the magnetization recorded at low temperature. The field was applied either along the a or c axis. As found previously, the magnetization is very weak with a large anisotropy in favor of the basal plane.[3] This anisotropy

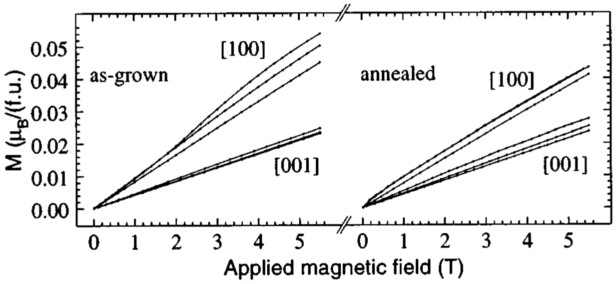

FIG. 1. Magnetization curves of an as-grown and an annealed $CePt_2Si_2$ crystal at three temperatures (2, 4.5, and 10 K). At a given field, the lower the temperature is, the larger the magnetization is except for some experimental conditions for which the magnetization is practically the same at two temperatures. The field is applied either along the [001] or [100] direction. The lines are only guides for eyes. The signature of the metamagnetic transition which is perceptible for the as-grown crystal disappears when annealed.

is explained by the crystal electric field acting on the Ce ions.[9] The signature of a metamagnetic transition which is perceptible for the as-grown crystal at $\approx$ 2.5 T when the field is applied along the a axis has disappeared in the annealed crystal. Therefore the reported metamagnetic transition[3] is not an intrinsic physical property of $CePt_2Si_2$. We have performed additional magnetic susceptibility measurements down to 2 K. The results are presented in Fig. 2. The fact that annealing increases the low temperature planar and axial susceptibilities seems to point out that the relatively large susceptibilities at low temperature is at least partly an intrinsic effect and not entirely due to impurities as suggested previously.[3] To settle the problem of the origin of the anomalous low temperature behavior of the susceptibilities, nuclear magnetic resonance (NMR) experiments on ^{195}Pt and ^{29}Si have to be performed.

In Fig. 3 we present the specific heat data. The temperature behavior is similar to that observed previously.[2] As shown by the full line, above 4 K the total specific heat follows the law $C=\gamma T+\beta T^3$ with the Sommerfeld ratio γ = 70 mJ $K^{-2}mol^{-1}$. Below 4 K, C/T departs strongly from the linearity, reaching a value of $\approx$ 0.118 J K^{-2} mol^{-1} at low temperature. The increase in C/T cannot be described by a $-\ln(T)$ law as found for $CeCu_{5.9}Au_{0.1}$.[6] Referring to the work of Ayache *et al.*,[2] we note that the electronic part in C/T is constant in the same temperature

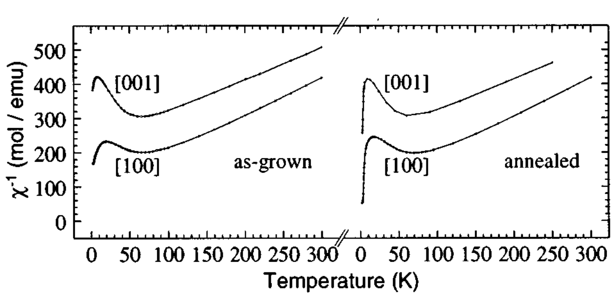

FIG. 2. Temperature dependence of the inverse of the dc magnetic susceptibility measured on as-grown and annealed $CePt_2Si_2$ crystals. The field is applied either along the [001] or [100] direction. At low temperature the planar and axial susceptibilities are larger for the annealed crystals.

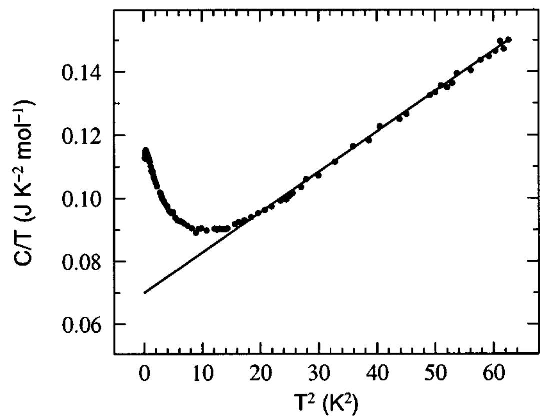

FIG. 3. Total specific heat divided by the temperature, C/T, vs T^2 measured for a crystal of $CePt_2Si_2$. These measurements clearly show that a crossover to a new electronic ground state occurs below 4 K.

range as the T^2 law is verified in the resistivity. Therefore, above 4 K, we observe a typical Fermi-liquid behavior. Below 4 K, the specific heat and resistivity[2] do not have that expected behavior. A possible reason could be the presence of a magnetic phase transition at low temperature. Below we report μSR measurements done to look for such a transition.

The μSR measurements were performed at the MuSR spectrometer[10] of the ISIS surface muon beam facility located at the Rutherford Appleton Laboratory (RAL, UK). The spectra were recorded with a ^{3}He-^{4}He dilution refrigerator for temperatures below 4.2 K and with a helium cryostat for temperatures up to 45 K. Some spectra were recorded below 4.2 K with the helium cryostat.

In the μSR technique polarized muons are implanted into a sample where their spin evolves in the local magnetic field until they decay.[11] The decay positron is emitted preferentially along the final muon spin direction; by collecting several million positrons, we can reconstruct the muon spin depolarization function $P_Z(t)$. The Z axis refers to the muon beam polarization axis which, in our case, is as well the direction of the detected positrons.[11] $P_Z(t)$ has been deduced from the raw data using the method described in Ref. 12. We have carried out measurements in zero-field and with an external applied field of 10 mT.

In Fig. 4 we present a typical zero field spectrum. The spectra are all well analyzed by the sum of two functions:

$$aP_Z(t)=a_{KT}P_{KT}(t)+a_{bg}\exp(-\lambda_{bg}t), \quad (1)$$

where $P_{KT}(t)$ is the Kubo-Toyabe function which describes the damping due to the sample and the second term accounts for the muons stopped in the sample holder, cryostat walls and windows. Because the spectra are not strongly depolarized, it is not possible to measure a_{bg}. Taking account of the size of our samples, we estimate a_{bg} = 0.080. Because this value is estimated and not measured, the absolute value of the damping rate deduced from the data suffers some uncertainty ($\sim$ 15 %). For the same reason we cannot compare the damping rate measured for the two samples in details. Note that the uncertainty on a_{bg} does not influence the temperature dependence of the damping rate. Measurements in zero field with only the silver plate and no sample showed that a good

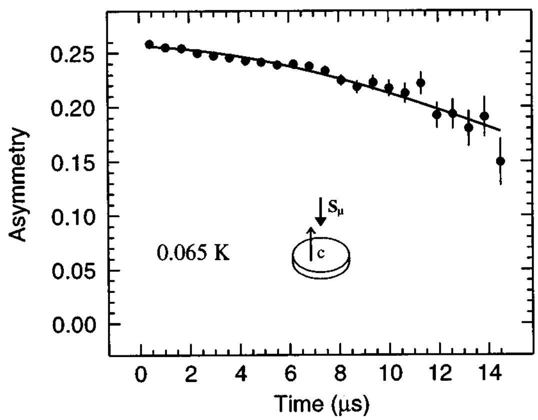

FIG. 4. A typical μSR zero-field spectrum measured on $CePt_2Si_2$ with the c axis parallel to the initial muon beam polarization, $\mathbf{S}_\mu$. The full line is a fit with the Kubo-Toyabe function.

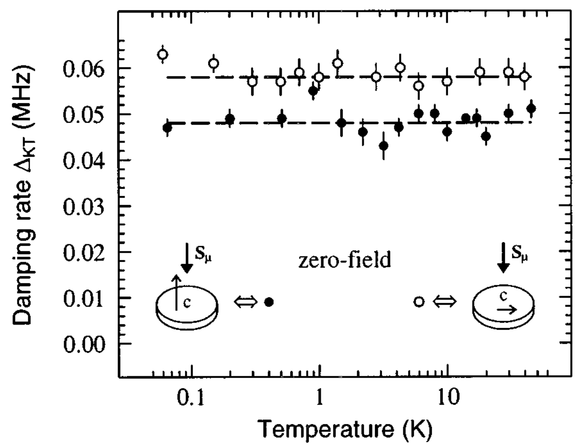

FIG. 5. Temperature dependence of the damping rate Δ_{KT} measured on two $CePt_2Si_2$ samples which differ by the orientation of $\mathbf{S}_\mu$ relative to the crystal axes: $\mathbf{S}_\mu$ is either parallel or perpendicular to $\mathbf{c}$. The dashed straight lines indicate the average Δ_{KT} values for the two orientations.

estimate for λ_{bg} is 0.012 MHz for the dilution refrigerator and virtually 0 for the helium cryostat. The $CePt_2Si_2$ zero-field spectra were therefore fitted with a_{bg} and λ_{bg} fixed to the previous values and a_{KT} as a free parameter. a_{KT} is then found to be constant over the temperature range investigated: $a_{KT} \simeq 0.175$. Since the damping due to the sample is very small, $P_{KT}(t)$ is well approximated by a parabolic function:[11]

$$P_{KT}(t)=1-\Delta_{KT}^2 t^2, \tag{2}$$

where $\Delta_{KT}=\gamma_\mu\sqrt{\langle B^2\rangle}$ describes the width of the distribution of local fields. γ_μ is the muon gyromagnetic ratio ($\gamma_\mu = 851.6$ Mrad s^{-1} T^{-1}) and $\langle B^2\rangle$ the second moment of the field distribution at the muon site. The parabolic character of the spectra is clearly seen in Fig. 4. The fact that the depolarization due to the samples is well described by the Kubo-Toyabe function is a strong indication that the spins of the muons are depolarized by a static field distribution, with a characteristic fluctuation time longer than $\sim 5\times10^{-6}$ s. This interpretation is confirmed by additional measurements performed at high and low temperature with a magnetic field of B_{ext}=10 mT applied along the Z axis: since $\gamma_\mu\Delta_{KT} \ll B_{ext}$, the spectra are not depolarized.

In Fig. 5 we present $\Delta_{KT}(T)$ for the two orientations of the crystal axes relative to the Z axis. Δ_{KT} is temperature independent for the two samples. From this observation we deduce that a possible change in the internal magnetic field induced by electronic magnetism, if it exists, has to be smaller than approximately 3 μT at the muon site over the whole temperature range.

The absence of thermal variation of $\Delta_{KT}(T)$ clearly shows that the muon spins are not depolarized by static magnetic moments of electronic origin. The observed depolarization is induced by the nuclear magnetic moments carried by the ^{195}Pt and ^{29}Si nuclei of spin 1/2 and abundance 33.7 and 4.7 %, respectively. Given a muon localization site, Δ_{KT} due to nuclear moments can be reliably computed (the lattice and electronic electric field gradients do not have any effect on 1/2 nuclear spins) but the muon localization site in $CePt_2Si_2$ is unknown. There is no obvious symmetrical interstitial site in the $CaBe_2Ge_2$ structure type where the muon could sit. In Ref. 13 a list of possible sites is given for the $ThCr_2Si_2$ structure and these sites can be transposed to the $CaBe_2Ge_2$ structure. This set of 11 sites is spread over the unit cell, and therefore we can have at least an order of magnitude for Δ_{KT}. Using the lattice parameter a = 4.25 Å and c = 9.80 Å (Ref.1) we find values ranging from 0.027 MHz to 0.14 MHz. For most of the sites the value computed for $S_\mu \perp c$ is slightly larger than the one computed for $S_\mu \| c$. The experimental value of Δ_{KT} and its orientation dependence is then consistent with the computed one.

The nonobservation of a magnetic signal of electronic origin means either that the Ce atoms carry static electronic magnetic moments smaller than $\sim 2\times10^{-4}\mu_B$ (or the conduction bands are very weakly polarized) (Ref. 14) or these moments are larger but cannot be detected by μSR. This is possible either if the magnetic moments order but for symmetry reasons the magnetic field at the muon site cancels, or if their characteristic fluctuation time τ_c is very short, i.e., smaller than $\sim 10^{-7}$ s (this time limit may be 1 order of magnitude larger or lower depending on the coupling between the muon and the moments). Because we have no information on the muon localization site, we cannot discuss reliably the possibility of cancelation of the field at the muon site for symmetry reasons. In relation to our present work, we note that while NMR and μSR do not detect any magnetic moments in UPt_3, neutron diffraction techniques do see Bragg peaks related to magnetic moments.[15] Therefore it would be of interest to look for magnetic Bragg peaks by neutron diffraction measurements in $CePt_2Si_2$. Apart from the immediate interest for $CePt_2Si_2$, it could help understanding the origin of the magnetic Bragg peaks of UPt_3. In the rest of the discussion we will take the conservative point of view that $CePt_2Si_2$ does not present magnetic ordering.

It has been pointed out that the paramagnetic ground state of compounds with strong electronic correlations is highly unstable.[7] In fact only $CeCu_6$ does not seem to present some kind of static ordering (magnetic or superconducting) at low temperature.[16] With this work we add a second compound in the list of Ce intermetallics with a nonmagnetic ground state at low temperature. We notice that the Kondo temperature in $CeRu_2Si_2$ is three times smaller than in $CePt_2Si_2$, and that the former compound presents a μSR magnetic signature starting at $\sim$ 2 K,[7] while the latter does not display this signature even at 0.06 K. In this respect, we mention here the discovery of a magnetic phase transition at $\sim$ 40 K in $CeRu_2$,[14] an intermetallic with a Kondo temperature of $\sim$ 1000 K.[17]

In conclusion, the temperature dependence in $CePt_2Si_2$ of the specific heat and electrical resistivity observed below 4 K seems to indicate that this compound is not a Fermi-liquid system. In this paper we have argued that our μSR experiment indicates that $CePt_2Si_2$ does not have a magnetic phase transition occurring above 0.06 K. To establish definitively the absence of this transition, neutron diffraction experiments should be performed since, as found for UPt_3,[15] such a transition not detected by muon relaxation measurements could be seen by neutron experiments. The exciting possibility that $CePt_2Si_2$ is an example of a non-Fermi-liquid metal calls for additional measurements to characterize its physical properties. We have in mind nuclear magnetic resonance and high field specific heat and resistivity measurements.

The researchers from the Netherlands acknowledge support from the Dutch Scientific Organization (NWO). The μSR measurements were partly supported by the Commission of the European Community through the Large Installations Plan.

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