$CeRu_2$: A magnetic superconductor with extremely small magnetic moments

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Muon-spin-relaxation experiments and ac low-field magnetization measurements have been carried out on the superconductor $CeRu_2$. The relaxation rate of the muon-spin polarization in zero field exhibits a small but significant increase below $T_M \simeq 40$ K, which is suppressed by applying a longitudinal field. This result taken together with magnetization measurements provides definite evidence for the occurrence of static electronic magnetism involving extremely small magnetic moments. Our work shows that $CeRu_2$ is a member of a restricted family of superconducting compounds that order magnetically with extremely small magnetic moments at a temperature much higher than that at which they become superconducting.
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The interplay between magnetism and superconductivity engulfs two of the richest areas of solid state physics. The coexistence of the two phenomena was first extensively studied in the Chevrel phases[1] where in certain pure compounds magnetic order appears below the superconducting transition temperature, T_c. For these materials the magnetically ordered ions are only weakly coupled to the conduction electrons. A different situation pertains for some of the heavy fermion superconductors such as $U_{1-x}Th_xBe_{13}$, UPt_3, and URu_2Si_2. The small size of the ordered magnetic moments in these compounds relative to their Néel temperatures attests to the more complex many-body origins of their magnetism.[2–4] It is remarkable that the latter two compounds have magnetic ordering temperatures roughly an order of magnitude higher than T_c and that the magnetic order persists into the superconducting state. In $CeCu_2Si_2$, the most studied Ce-based heavy fermion superconductor, the magnetism is relatively strong and in competition with superconductivity rather than coexisting with it.[5]

Our finding is that the cubic Laves phase superconductor $CeRu_2$ condenses into a static magnetic state at a temperature $T_M \simeq 40$ K which persists into the superconducting state below $T_c = 6.1$ K. The evidence comes from both muon-spin-relaxation (μSR) measurements and ac susceptibility measurements on a single crystal. Our work supports the interpretation that anomalies seen in recently presented high field measurements are due to the occurrence of static magnetism at T_M.[6]

In the superconducting state of $CeRu_2$ an abrupt transition from irreversible magnetic behavior near the upper critical field to almost perfectly reversible behavior at lower fields occurs.[7–11] The robustness and well-defined nature of this transition has led to the contention that it might be due to some underlying transition within the superconducting state, rather than due to a continuous evolution of flux pinning effects alone. In a recent neutron study[12] the correlation length of the flux line lattice was measured. When interpreted within a theory of weak collective pinning, a pin spacing of the order of the superconducting coherence length was deduced. So far no evidence as to the physical origin of the pinning mechanism has been forthcoming. In this light, the existence of magnetic order raises the possibility that the pinning is magnetic in origin and not necessarily related to crystalline defects.

Early studies[13–15] concerning the coexistence of magnetism and superconductivity related to $CeRu_2$ considered compounds where the Ce had been partially substituted by a third ionique species. It was found that the replacement of Ce with significant quantities of other lanthanide metals can give rise to short-range ferromagnetic correlations. On substituting higher concentrations of these elements the superconductivity is eventually destroyed and replaced by long-range ferromagnetic order. These results should not be confused with the data presented in this article, where we examine only the pure unsubstituted compound. In the pure compound the transition is indeed quite subtle and explains why it was not picked up in previous dc magnetization studies.[16,17] As in a previous investigation[18] we do not resolve any anomaly in the resistivity near T_M.

The μSR sample was a disk of $\sim$25 mm diameter and $\sim$0.5 mm thickness, comprising of a mosaic of slices glued on a 5N silver plate (40×40 mm^2). These slices were cut from a large grain polycrystalline ingot of $CeRu_2$. The single crystal used in the susceptibility study was grown by the Czochralski method and had a mass of 1.7 g. No second phases were detectable in similarly prepared crystals in both electron microprobe and high resolution electron microscope

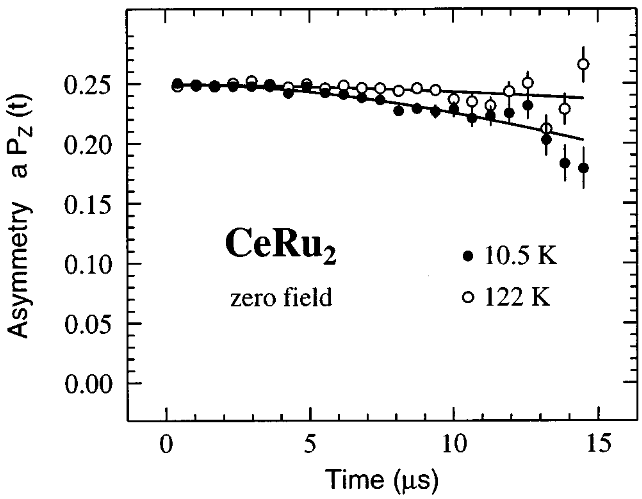

FIG. 1. Typical zero field spectra recorded on $CeRu_2$ at 10.5 and 122 K. The lines are fits to the sum of a parabolic depolarization function and a constant term. The relaxation rate is clearly stronger at low temperature.

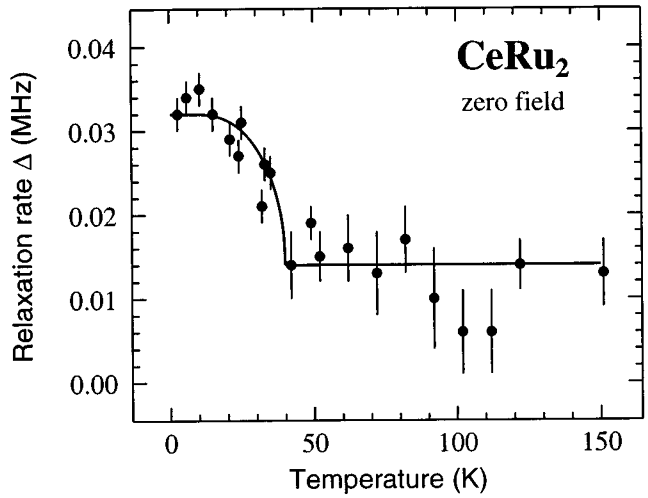

FIG. 2. Temperature dependence of the Gaussian muon-spin-relaxation rate, Δ, in $CeRu_2$ at zero field. The line is the Brillouin function prediction for a spin $S = 1/2$ and $T_M = 40$ K. This result provides evidence for the occurrence of static electronic magnetism at $T_M \simeq 40$ K.

studies. The residual resistivities of similarly prepared crystals are of the order 10 $\mu\Omega$ cm.

The μSR measurements were performed with the MuSR spectrometer[19] at the ISIS surface muon beam facility (Rutherford Appleton Laboratory, U.K.). The spectra were recorded with a closed cycle refrigerator for temperatures between 21 and 151 K and with a helium (''Orange'') cryostat for low temperatures down to 2.8 K. Some cross checked spectra were recorded at temperatures up to 49 K with the helium cryostat. The ac susceptibility was measured by the usual inductive technique with a driving field of 3.5 mT at 35 Hz. The crystal was oriented with a low symmetry direction parallel to the ac field for geometric convenience since any magnetic anisotropy is expected to be insignificant ($CeRu_2$ is cubic).

The basic physical quantity measured in our μSR experiment is the muon-spin depolarization function $P_Z(t)$ which is simply related to the distribution of fields experienced at the muon stopping site.[20] The measurements correspond to a longitudinal geometry, in which the muon beam polarization is parallel to the incident beam (Z axis) and the positron detectors.[21] We have carried out measurements in zero field and with an external applied field of 1 mT (parallel to Z). The residual magnetic field on the sample during the zero field measurements was $\lesssim 1$ μT.

In Fig. 1 we present typical zero field spectra. All the spectra are well analyzed by the function

$$aP_Z(t) = a_s P_s(t) + a_{bg} . \tag{1}$$

$P_s(t)$ describes the relaxation due to the sample and the second term in Eq. 1 accounts for the muons stopped in the sample holder, cryostat walls and windows. By definition $P_Z(0) = P_s(0) = 1$. Measurements at zero field with only the silver plate and no sample showed that the second component does not relax. The data are well described by $P_s(t) = 1 - \Delta^2 t^2$. The parabolic character of the spectra is clearly seen in Fig. 1. A transverse field measurement in the superconducting phase allowed us to determine a_{bg} : a_{bg} $= 0.051$ (2). This $a_{\rm bg}$ value was used as a fixed parameter in the fit. a_s is then found to be constant over the temperature range investigated : $a_s = 0.198$ (1).

In Fig. 2 we display Δ versus the temperature. While at high temperatures Δ is roughly temperature independent with a value of ~ 0.014 MHz, it increases sharply below $T_M \sim 40$ K to a value of $\sim$ 0.032 MHz at low temperature. Superconductivity does not seem to influence the relaxation rate. That $P_s(t)$ is quadratic in time is a strong indication that the muons are stationary and their spin depolarized by either a static field distribution or a very small coherent field at the muon site.[20] This interpretation is confirmed by the measurements at 1 mT (for an example, see Fig. 3), which show that the depolarization of the muon spin is supressed at both low and high temperature.

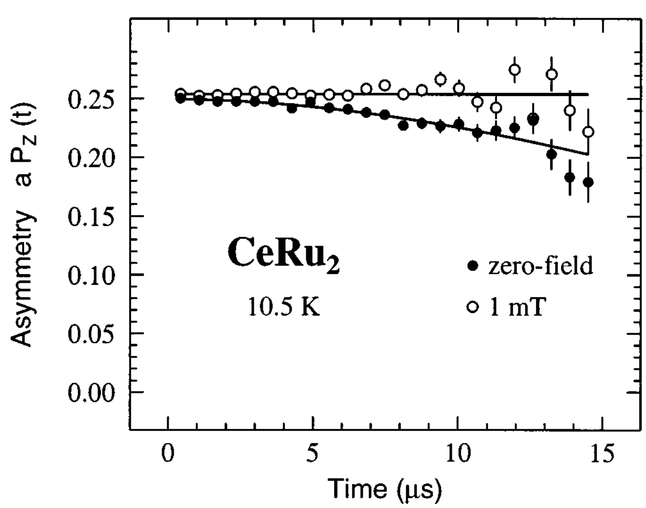

FIG. 3. Comparison between zero field and longitudinal field spectra recorded on $CeRu_2$ at 10.5 K. The fact that the depolarization is suppressed by an applied longitudinal magnetic field is an additional proof that the field distribution at the muon site is static.

As a first step to interpret the data of Fig. 1, given a stationary muon, we calculate the relaxation at high temperatures induced by the nuclear magnetic moments (uniquely carried by the ^{99}Ru and ^{101}Ru nuclei of abundance 12.8% and 17%, respectively). Such a depolarization mechanism would indeed give a parabolic form for $P_s(t)$ for which Δ is then identified with the Kubo-Toyabe relaxation rate due to the nuclear moments, $\Delta_{\mathrm{KT},n}$.[20] There are three possible interstitial muon stopping sites in the cubic Laves phase structure, denoted 2-2, 3-1, and 4-0 where the first (second) digit denotes the number of nearest-neighbor ruthenium (cerium) atoms.[22] For the lattice parameter $a=7.538$ Å,[23] and neglecting the electric field gradient (EFG) acting on the Ru atoms due to the muon and the lattice environment (the Ru atoms are not in a site of cubic symmetry) we find $\Delta_{\mathrm{KT},n}=$ 0.042, 0.056, and 0.070 MHz for the three sites, respectively. None of these values can explain the measured small damping rate. The difficulty encountered to explain the measured $\Delta_{\mathrm{KT},n}$ at high temperatures is not new. The 2-2 site which has been deduced for the isostructural compound $CeAl_2$ from transverse field measurements[22] does not explain its zero field spectra:[24] the observed $\Delta_{\mathrm{KT},n}$ is again much smaller than given by the simple calculation. These difficulties are probably all related to the neglect of the EFG in the calculation. An alternative possibility is that the muons stop at atomic voids. In this case we cannot compute $\Delta_{\mathrm{KT},n}$ reliably because the position of the atoms are then drastically changed relative to the unperturbed lattice.

Having ruled out the possibility of a mobile muon, the increase of the relaxation rate below T_M must result from the appearance of a very small coherent magnetic field or a broadening of the field distribution which can be either of nuclear or electronic origin. A nuclear origin for the broadening can be eliminated since it would require an unreasonable change of the crystal lattice that has not been detected:[25] a lattice contraction of $\sim$25% is needed to explain the fractional change in Δ with temperature. Therefore the additional relaxation rate detected at low temperatures must be due to magnetic moments of electronic origin. This interpretation is strongly reinforced by the low field magnetization measurements presented below and the high field data of Nakama *et al.*[6] which are consistent with a magnetic transition at T_M.

Because of the extremely small value of the relaxation rate, the parabolic shape of $P_s(t)$ is a limiting form of either the Kubo-Toyabe function[20] or of an extremely low frequency oscillating signal. The Kubo-Toyabe depolarization function corresponds to a Gaussian field distribution of width Δ_e (in frequency units, $\Delta_e^2 = \Delta^2 - \Delta_{\mathrm{KT},n}^2$) at the muon site which characterizes a spatially disordered or incommensurate magnetic state, whereas a low frequency oscillating signal is the signature of a coherent magnetic structure with an appreciable correlation length and small magnetic moments. The μSR data cannot distinguish between these possibilities. Under the assumption that the muon spin is depolarized by a field distribution and senses only the dipolar fields from the electronic magnetic moments localized on the Ce atoms, we estimate the Ce magnetic moment: $\mu_{\mathrm{Ce}}\gtrsim 10^{-4}\mu_B$. Assuming equal moments on both Ce and Ru sites, as suggested by a recent polarized neutron study,[26] we find about $10^{-4}\mu_B$. If we suppose that the increase in damping is in fact due to the

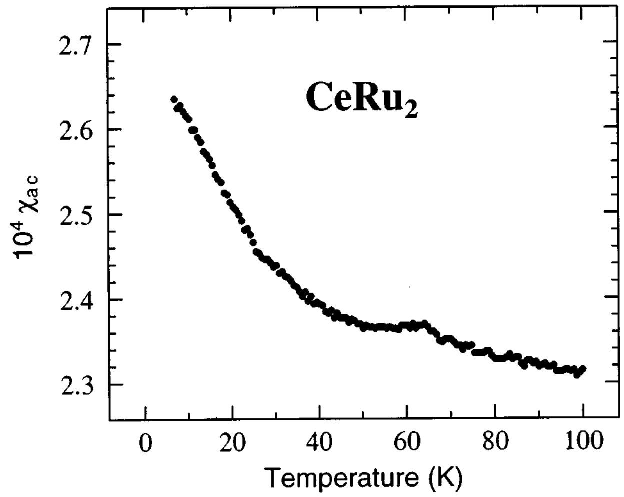

FIG. 4. Temperature dependence of the real part of the ac susceptibility of a crystal of $CeRu_2$. In SI units, the susceptibility is dimensionless. The measurements were made for increasing temperature after initially cooling the sample to just above T_c. We observe a plateau followed by an increase at $\sim T_M=40$ K, the ordering temperature deduced in the μSR experiment.

appearance of a coherent magnetic field at the muon site, this field would be 0.05 mT. This corresponds to a μ_{Ce} of the same range as previously estimated. These are the smallest values of electronic moments ever detected. They have however been derived using a simple localized magnetic model. In view of their extremely small value, a bandlike model is probably more appropriate.

We have analyzed our spectra supposing that the small detected moment is uniformly distributed in the sample. Another possibility that might be considered is that the depolarization is caused by only a small volume fraction of the sample. From the magnitude of the depolarization at 14.5 μs we can conclude that at least 15 volume % of the sample is responsible for the depolarization. A magnetic moment greater than $\approx 10^{-3}\mu_B$ would be inconsistent with the observed quadratic shape of the depolarization. We note that such a large fraction of any second phase was not detected in our sample.

The ac susceptibility data displayed in Fig. 4 shows a plateau starting at $\sim$60 K followed by a strong increase below T_M. We do not have a definite explanation for the occurrence of the plateau, but the accumulated evidence for a weak magnetic signal in $CeRu_2$ at T_M, from our zero field μSR and low field susceptibility measurements as well as the high field results from Nakama *et al.*,[6] points definitively to the occurrence of a magnetic transition at T_M. In order to better characterize the magnetic state we have also carried out some measurements in low field with commercial dc superconducting quantum interference device magnetometers, in particular to test for the possible occurrence of magnetic hysterisis. Within our experimental uncertainties we fail to find any such effects.

Small static moments, but still larger by an order of magnitude, have been observed for $U_{1-x}Th_xBe_{13}$ (Ref. 2) and $CeRu_2Si_2$.[27] It is only in the former compound that the parabolic character of the μSR depolarization function at small times has been established. While in UPt_3, magnetic Bragg peaks are seen by neutron and x-ray scattering, most other experimental techniques including μSR fail to detect a signal of magnetic origin.[28] URu_2Si_2 exhibits a magnetic phase transition with a relatively long correlation length and is characterized by a small uranium magnetic moment.[29] The functional form of $P_s(t)$ confirms that URu_2Si_2 is a relatively well-ordered magnet.[30] The other three widely studied heavy fermion superconductors, UNi_2Al_3, UPd_2Al_3, and $CeCu_2Si_2$, all exhibit relatively large ordered moments.[31,32,5] and therefore may not belong to the same class of compounds as $CeRu_2$.

In summary, $CeRu_2$ appears to be an ordered magnetic superconductor[33] characterized by a small magnetic moment. This invites comparison to similar characteristics in the U-based materials $U_{1-x}Th_xBe_{13}$, UPt_3, and URu_2Si_2. Relative to the latter three compounds, it exhibits even smaller magnetic moments. The shape of the μSR depolarization function is quadratic in time. While this result does not identify the precise nature of the order, we note that magnetic moments located on the Ru ions would lie on a three-dimensional lattice of corner-sharing tetrahedra: this situation is known to give rise to frustration.[34] This frustration might lead to a glasslike state and would nicely explain the μSR results. Whatever the nature of the magnetic order, it is likely to influence the pinning of the vortex lattice and presents an important ingredient that needs to be considered to understand the unusual transition from reversible to irreversible behavior in the superconducting state.

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