PHYSICAL REVIEW B VOLUME 39, NUMBER 7 1 MARCH 1989

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Coexisting static magnetic order and superconductivity in $CeCu_{2.1}Si_2$ found by muon spin relaxation

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(Received 30 November 1988)

Zero- and longitudinal-field muon-spin-relaxation measurements on a heavy-fermion system $CeCu_{2.1}Si_2$ have revealed an onset of static magnetic ordering below $T_M \sim 0.8$ K, which coexists with superconductivity below $T_c = 0.7$ K. The line shapes of the observed muon spin depolarization functions suggest an ordering in either the spin-glass or incommensurate spin-density-wave states, with a small averaged static moment of the order of $0.1\mu_B$ per formula unit at $T \rightarrow 0$.

$CeCu_2Si_2$ belongs to a group of cerium or uranium intermetallic compounds, called heavy-fermion systems,[1] which are characterized by the extremely large T-linear term $C = \gamma T$ with $\gamma \sim 1$ J/mole K^2 of the electronic specific heat C at low temperatures, which implies a very large effective mass m^*. Previously, Steglich *et al.*[2] found a superconducting transition in $CeCu_2Si_2$ at $T \sim 0.5$ K, as the first example of the superconducting ground state of a heavy-fermion system. This discovery triggered extensive study of superconductivity and its possible relation with magnetism in the highly correlated heavy-electron systems.[1,3] Although there are some susceptibility measurements[4,5] which suggest magnetic ordering of nonsuperconducting systems $Ce_{1-y}La_yCu_2Si_2$ with $y \geq 0.2$ (Ref. 5) and $CeCu_xSi_2$ with $x = 1.9$ (Ref. 4), so far it has been common to assume[1] a purely superconducting ground state without magnetic ordering for the superconducting compounds $CeCu_xSi_2$ with $2.0 \leq x \leq 2.2$ (Ref. 6). In this paper, we present the first direct evidence from zero-field muon-spin-relaxation measurements that superconductivity and static magnetic ordering coexist in $CeCu_{2.1}Si_2$. Muon spin relaxation[7] (μSR) is a very powerful tool to detecting static magnetic ordering.[8,9] Previous applications of μSR to heavy-fermion systems led to discoveries of magnetic ordering with quite small ordered moments ($0.001\mu_B$-$0.05\mu_B$) in superconducting UPt_3 (Ref. 10) and nonsuperconducting $CeAl_3$ (Ref. 11). With the present results on $CeCu_{2.1}Si_2$, we now have three heavy-fermion superconductors, $CeCu_{2.1}Si_2$, UPt_3, and URu_2Si_2 (Ref. 12), which show coexisting magnetic orderings.

Superconductivity of the stoichiometric $CeCu_{2.0}Si_2$ is known to be somewhat unstable.[5] A small amount of additional Cu helps to stabilize the superconductivity of $CeCu_xSi_2$ with $x = 2.1$–2.2, whose superconducting transition temperature T_c is around 0.7 K (Ref. 6). We therefore prepared a polycrystalline sample of $CeCu_{2.1}Si_2$ by the method described in Ref. 6. Figure 1 shows resistivity (in zero field) and susceptibility (in external field of 2.5 T) measured on a piece cut off from the sample of $CeCu_{2.1}Si_2$ used in the present μSR measurement. Reference 6 described detailed resistivity and specific-heat measurements on a few polycrystalline samples of $CeCu_xSi_2$ with $x = 1.9$–2.2 made with the same method. The present specimen has T_c at around 0.7 K, and the temperature dependence of resistivity shows the same curvature as reported in Ref. 6. The susceptibility is small (~ 0.01 emu/mole) and nearly independent of temperature below $T = 3$ K. This indicates a minimal contribution from the Curie-like term due to free impurity spins. To further characterize the present sample, we made a neutron scattering measurement of the crystal structure, and confirmed that it is a single-phase material without any minor phase within the experimental accuracy of a few volume percent.

We started zero-field μSR measurements on the present sample of $CeCu_{2.1}Si_2$ at the alternating-gradient-synchrotron muon channel of Brookhaven National Laboratory by using a ^{3}He cryostat. A rapid increase of the muon-spin-relaxation rate was found with decreasing temperature below $T \sim 0.8$ K. We then continued the measurement at the Swiss Institute for Nuclear Research (SIN), Switzerland, by using a surface muon beam and a dilution refrigerator. The facility at SIN allowed full access to a wide temperature region with high-statistics data as we report in this paper. A positive muon beam was stopped at the sample of $CeCu_{2.1}Si_2$ ($2 \times 1 \times 0.5$ cm^3) mounted on a cold finger of the dilution cryostat, and the muon-decay positrons were recorded mainly with a counter placed in the forward direction with respect to the beam direction. The counting rate $I(t)$ of this counter is given as

$$I(t) \propto \exp(-t/\tau_\mu)[1 - AG_z(t)] , \qquad (1)$$

where τ_μ is the muon lifetime 2.2 μsec, A is the initial decay asymmetry ($A \sim 0.25$ at the present condition), and the relaxation function $G_z(t)$ represents the time evolution of muon spin polarization.

Figure 2(a) shows the relaxation function $G_z(t)$ thus observed in zero field. The relaxation rate increased rapidly below $T \sim 0.8$ K with decreasing temperature. In general, the depolarization of muon spins in zero field can be due either to randomness of the static internal local field H_{int} or to the fluctuating dynamic local fields. One can distinguish between these two cases by making measurements in the longitudinal external magnetic field H_{ext} applied parallel to the initial muon spin direction (i.e., the direction of muon beam, denoted as z direction hereafter). When the internal field is static and H_{ext} is larger than H_{int}, H_{ext} can align the local field $\mathbf{H}_{\text{loc}} = \mathbf{H}_{\text{int}} + \mathbf{H}_{\text{ext}}$ to be nearly parallel to the muon spin polarization, thus keeping $G_z(t)$ finite. In contrast, the dynamic spin fluctuations are usually much faster than the corresponding Zeeman frequency $\omega = \gamma_\mu H_{\text{ext}}$ ($\gamma_\mu = 2\pi \times 13.554$ MHz/kG) of μ^+, so that there is almost no effect of H_{ext} on $G_z(t)$ for the dynamic case. We have performed such measurements in $H_{\text{ext}} = 250$ G and 1 kG at $T = 0.1$ K as shown in Fig. 2(b). The longitudinal fields suppress the depolarization and change $G_z(t)$ remarkably. This indicates that the depolarization observed in zero field is due mainly to the static random local fields of the order of 100–200 G. Similar measurements with H_{ext} confirmed that the depolarization at $T = 0.8$ K is also due to the static fields.

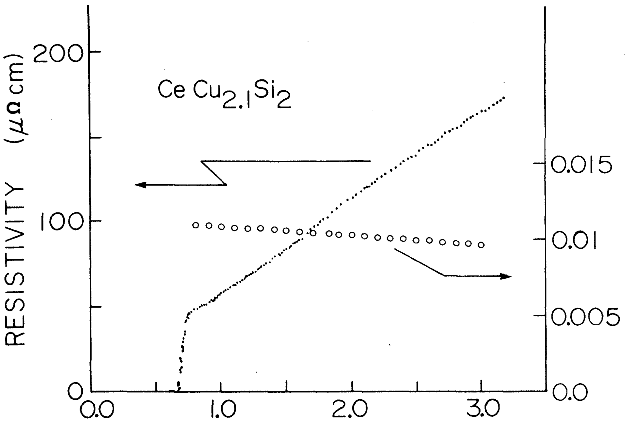

FIG. 1. Resistivity (in zero field) and susceptibility (in external field of 2.5 T) measured on a piece of $CeCu_{2.1}Si_2$ cutoff from the present specimen used in the μSR measurements. Superconducting transition occurs at $T_c = 0.7$ K.

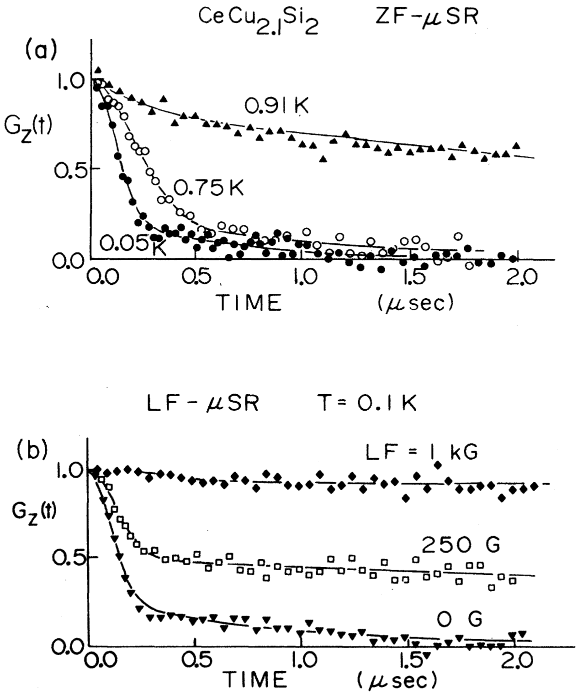

FIG. 2. (a) Muon-spin-relaxation function $G_z(t)$ in zero field observed in $CeCu_{2.1}Si_2$. Solid lines represent fits to Eq. (2). (b) Muon-spin-relaxation function in $CeCu_{2.1}Si_2$ observed at $T = 0.1$ K in longitudinal external magnetic fields LF of 0 and 250 G, and 1 kG. Solid lines are guides to the eye.

The line shapes of $G_z(t)$ in zero field shown in Fig. 2(a) resemble those observed in the dilute-alloy spin glasses *Cu*Mn or *Au*Fe (Ref. 8). The lack of coherent oscillation indicates that the magnitude of the static internal field $|H_{\rm int}|$ has a wide distribution. For a uniform $|H_{\rm int}|$, one should have seen a muon spin precession as reported in Refs. 9 and 11. $G_z(t)$ in Fig. 2(a) at $T=0.05$ K starts with a Gaussian-like shape at $t\to 0$. This is somewhat different from the case in the dilute-alloy spin glasses, where $G_z(t\to 0)$ decays with an exponential-like shape when the temperature is well below the susceptibility-cusp temperature T_g. In this paper, we do not develop a complicated theory to account for this line shape, but rather adopt a phenomenological relaxation function,

$$G_z(t)=\frac{A_1}{A}\exp(-\tfrac{1}{2}\sigma^2t^2)+\frac{A_2}{A}\exp(-\Lambda t)\ , \qquad (2)$$

to fit the observed data. The first term corresponds to the quick initial decay caused by the x and y components of the static random local fields. The second term represents the tail arising from the component of $H_{\rm int}$ parallel to the initial muon spin direction (i.e., the z direction). Reasonably good fits to the data at all different temperatures $0.05\lesssim T\lesssim 1.0$ K were obtained when we assumed $A_1/A\cong\frac{2}{3}$, $A_2/A\cong\frac{1}{3}$, with $A=0.25$. The present counter configuration without the backward counter, however, made it difficult to determine the second term of Eq. (2) accurately. Therefore, the decay rate Λ of the tail, ranging around $\Lambda\sim 1.0\ \mu{\rm sec}^{-1}$, may be subject to a large systematic error. In contrast, the first term of Eq. (2) can be determined with reasonable precision. From the fit to Eq. (2) represented by the solid lines in Fig. 2(a), we thus obtained the muon-spin-relaxation rate σ as shown in Fig. 3.

The relaxation rate σ in Fig. 3 increases rapidly with decreasing temperature below $T\sim 0.8$ K. This indicates the onset of a random but static magnetic order at the magnetic ordering temperature $T_M\sim 0.8$ K. Because of the limited accuracy of the temperature measurements and control with the cold-finger cryostat, as represented by the horizontal error bars in Fig. 3, we cannot identify whether the magnetic and superconducting orderings occur simultaneously or whether T_M and T_c are somewhat different. It is evident from Figs. 1–3, however, that the superconductivity and magnetic ordering coexist below $T\sim 0.7$ K in $\rm CeCu_{2.1}Si_2$. The present results indicate that more than 90 vol% of the specimen undergoes the magnetic ordering. The relaxation rate σ in Fig. 3 represents a measure of the width of the static internal fields as $\sigma\sim\gamma_\mu[\langle(\Delta H_{\rm int})^2\rangle]^{1/2}$. The observed value of $\sigma\sim 10\ \mu{\rm sec}^{-1}$ at $T\to 0$ corresponds to the static random local fields of the order of $\sigma/\gamma_\mu\sim 120$ G. In most magnetic materials, local fields at muon sites are due mainly to the dipolar field from the surrounding moments. Since μ^+ is a pointlike probe in real space, and since information on the muon stopping site is lacking, it is not easy to accurately estimate the spatial spin structure of the magnetically ordered state of $\rm CeCu_{2.1}Si_2$. It is, however, possible to note the following.

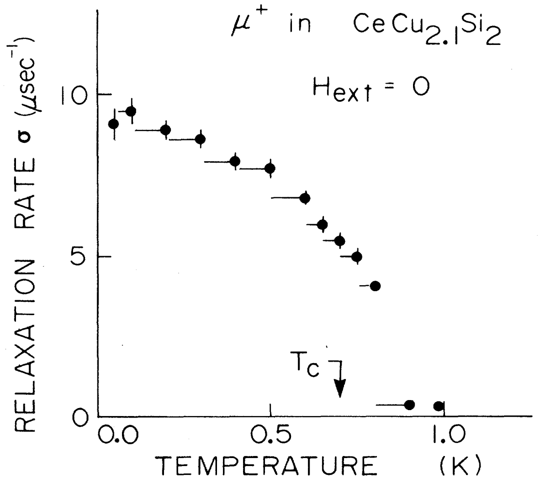

FIG. 3. Muon spin depolarization rate σ, as defined in Eq. (2), derived from the relaxation functions observed in $\rm CeCu_{2.1}Si_2$ in zero field. The onset of magnetic ordering is seen around $T_M\sim 0.8$ K.

Spin-glass ordering is one of the most likely spin structures which produce a large distribution of $H_{\rm int}$ as observed in the present experiment (see Ref. 8). This picture is consistent with the observation of a strong effect of external fields on the susceptibility measurements of $\rm CeCu_{1.9}Si_2$ (Ref. 4). If one assumes that the majority of Ce (or Cu) atoms participate in the magnetic ordering, the observed width ~ 120 G of $H_{\rm int}$ corresponds to the dipolar field from the ordered moment of the order of $0.1\mu_B$. If instead the small population of $\rm Ce^{3+}$ ions forms a spin glass with an ordered moment of $5\mu_B$, such as dilute-alloy spin glasses, then the observed value of σ corresponds to an ordering of about a few percent of the Ce atoms. The latter picture is advocated in Ref. 4 for the case of $\rm CeCu_{1.9}Si_2$. The Gaussian-like decay of $G_z(t\to 0)$ may favor the former type of spin glass, but then one faces the difficult question as to the origin of the frustration of the exchange interactions. An incommensurate spin-density-wave state, such as that observed in $\rm CePb_3$ (Ref. 13), is another possible spin structure which gives the local field at the muon site a wide distribution. Unfortunately, the present experiment alone cannot distinguish the above-mentioned three possible spin structures.

A neutron scattering experiment on $\rm CeCu_{2.1}Si_2$ is underway[14] to study spatial spin correlation. For such a small averaged moment as $\sim 0.1\mu_B$, however, the magnetic scattering intensity of neutrons is very small. Neutron measurements become even more difficult when the spin-glass ordering makes the scattering diffusive in reciprocal space. In this respect, the present experiment demonstrates the unique capability of zero-field μSR to detect magnetic orderings with small averaged moments. In the previous μSR study on the same specimen of $\rm CeCu_{2.1}Si_2$ performed in the transverse external magnetic field,[15] the depolarization of muon spins observed below $T\sim 0.8$ K was tentatively attributed to the inhomogeneous penetration of the transverse external field in the type-II superconducting state. Zero-field μSR can detect the magnetic ordering without the complication of the field penetration.

The present results indicate that a major part of the depolarization observed in the transverse field was due to the static magnetic ordering. In order to find out whether the superconductivity and magnetic ordering occur at the same temperature or at different temperatures, we are planning to perform additional μSR measurements on specimens with different Cu stoichiometry which may have different T_M (see Ref. 4).

Recently, a heavy-fermion superconductor UPt_3 ($T_c \sim 0.5$ K) was found[10,16] to order magnetically below $T \sim 5$ K with a very small averaged moment of $0.001\mu_B$–$0.02\mu_B$. URu_2Si_2 is another superconducting heavy-fermion system ($T_c \sim 1.0$ K) which orders antiferromagnetically below $T_N \sim 17$ K with an ordered moment of $0.03\mu_B$ per uranium atom.[12] UBe_{13} is so far the only remaining heavy-fermion superconductor without an identified magnetic ground state, yet there are reports[17,18] which suggest magnetic ordering in UBe_{13} doped with a small amount of Th. Together with the present results on $CeCu_{2.1}Si_2$, these features indicate that the magnetic ordering with extremely small averaged moment may be a common feature of heavy-fermion superconductors.

In summary, zero-field μSR measurements on $CeCu_{2.1}Si_2$ have shown clear evidence of static magnetic ordering below $T_M \sim 0.8$ K with a very small averaged moment of $\sim 0.1\mu_B$ in either a spin-glass or an incommensurate spin-density-wave state. This ordering coexists with superconductivity below $T_c = 0.7$ K. Further experimental and theoretical studies are clearly needed for the full understanding of the role of such magnetic orderings on the superconductivity of heavy-fermion systems.

Note added. After submitting this paper, we performed a μSR study on superconducting $CeCu_{2.2}Si_2$, and observed static magnetic order with the magnitude of the ordered moment approximately the same as the present results.

We would like to acknowledge useful discussions with P. B. Allen, K. Kakurai, M. Steiner, and E. Recknagel. We thank W. Schönig for help on the measurements at Brookhaven. This work is supported by the Division of Materials Sciences, U.S. Department of Energy under Contract No. 76-AC02-CH00016, the National Science Foundation under Grants No. DMR 8503223 and No. INT 8413978, NASA under Grant No. NAG-1-416, and by the Japanese Ministry of Education, Science, and Culture under the Grant-In-Aid for Scientific Research.

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