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Coexisting static magnetic order and superconductivity in $CeCu_2Si_2$ found by nuclear quadrupole resonance

Cheng Tien
Department of Physics, National Cheng Kung University, Tainan, Taiwan, Republic of China

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^{63}Cu nuclear quadrupole resonance (NQR) has been observed in a heavy-fermion system $CeCu_2Si_2$. In the superconducting $CeCu_2Si_2$ specimens ($T_c \simeq 0.6$ K), NQR experiments have revealed an onset of a phase transition below ~ 1 K. Zero- and longitudinal-field muon-spin-relaxation measurements suggest a static magnetic ordering below ~ 0.8 K, either in the spin-glass or in incommensurate spin-density-wave states. The exotic transition observed in NQR measurements should be closely related to the static magnetic ordering in muon-spin-relaxation measurements. NQR data from a nonsuperconducting $CeCu_2Si_2$ sample are consistent with extensive disorder in Cu site occupation. Therefore, the spin-density wave may be depressed in the nonsuperconducting $CeCu_2Si_2$. The superconducting transition temperature T_c can be enhanced by the presence of spin-density waves such that, if the spin-density wave is depressed, $CeCu_2Si_2$ will lose its superconducting properties.

INTRODUCTION

$CeCu_2Si_2$ belongs to a group of lanthanide and actinide compounds,[1–4] called heavy-fermion systems, that exhibiting a large value of the low-temperature magnetic susceptibility χ and coefficient γ of the linear term in the specific heat. Enhanced electron masses $m^* \sim 100 m_e$ result from the standard analyses of χ and γ, but the ratio χ/γ retains a value appropriate to a free-electron gas. Previously, Steglich *et al.*[1] found a superconducting transition in $CeCu_2Si_2$ at $T \sim 0.5$ K as the first example of the superconducting ground state of the heavy-fermion system. Recently Uemura *et al.*[5] reported that static magnetic ordering can coexist with superconductivity in $CeCu_2Si_2$. By zero-field muon-spin-resonance measurements on $CeCu_2Si_2$, Uemura *et al.* suggested this static magnetic ordering state in either the spin-glass or incommensurate spin-density-wave states.

The interplay between superconductivity and antiferromagnetism has been investigated extensively both experimentally and theoretically for a long time.[6–8] Two heavy-fermion superconductors, UPt_3 and UBe_{13} show antiferromagnetic fluctuations in inelastic neutron scattering experiments.[6] Another heavy-fermion superconductor, URu_2Si_2, exhibits superconductivity at $T_c \cong 1.5$ K, below the spin-density-wave transition ($T_N \cong 17.5$ K).[9] In fact, a small amount of doping ultimately induces a spin-density-wave long-range order in $(U,Th)Be_{13}$,[10,11] $(U,Th)Pt_3$,[12,13] and $U(Pt,Pd)_3$.[14,15] It is suggested that the origin of the exotic superconductivity observed in heavy-fermion systems is closely associated with this magnetic phenomenon.

In this paper we present a ^{63}Cu zero-field nuclear-quadrupole-resonance (NQR) experiment in the ternary compound $CeCu_2Si_2$. Above $T_c \sim 0.6$ K the NQR measurements show a clear phase transition at around 1 K. This phase transition might be associated with the magnetic ordering observed by the muon spin relaxation.[5] The features of this phase transition are discussed from the point of view of the nuclear quadrupole resonance.

SAMPLE PREPARATION

Our $CeCu_2Si_2$ specimens were obtained from the following sources: sample No. 1, from J. Aarts and F. R. de Boer, Natuurkunding Laboratorium, University of Amsterdam, Amsterdam, the Netherlands; sample No. 2, from L. C. Gupta, Tata Institute of Fundamental Research, Bombay, India. Both samples No. 1 and No. 2 are superconducting. One nonsuperconducting $CeCu_2Si_2$ sample, sample No. 3, was acquired from Z. Fisk, Los Alamos National Laboratory, Los Alamos, New Mexico.

Sample No. 1 was prepared[16] by arc melting and annealing at 1120 °C for 100 h. X-ray spectra did not show any indication of a second phase. This sample was powdered to a grain size of less than 90 μm in order to allow radio-frequency penetration of the entire sample.

In $CeCu_2Si_2$, superconductivity is extremely sensitive to sample preparation and to the presence of defects[17] so that measurements on several specimens are necessary to separate intrinsic effects. Therefore, another polycrystalline $CeCu_2Si_2$ sample, sample No. 2, was also made by the arc-melting method, but by a different annealing procedure.[18] Sample No. 2 was synthesized by melting together stoichiometric amounts of the appropriate elements under an argon atmosphere in an arc furnace. The buttons that were obtained after repeated arc melting were homogenized at about 800 °C for a week. Subsequently, the buttons were crushed into fine power and annealed for another week. At first, sample No. 2 was ground to 90-μm powder, and a series of NQR measurements were made. Then the sample was further powdered to 45 μm to study the size dependence of the heavy-fermion superconductivity. Sample No. 3 was grown as a single crystal in a liquid indium flux[19] by slow cooling from 1400 to 500 °C at 4 °C/hr in an In:$CeSi_2$:Cu mixture (0.95:0.01:0.04 atomically) in an aluminum crucible. The crystals were leached from the In matrix with HCl. In order to test the quality of this sample, x-ray fluorescence, energy dispersive spectroscopy, and lattice parameter measurements were performed. The results of these tests indicated that the stoichiometry of this single crystal $CeCu_2Si_2$ sample is comparable (within +5%) to the correct 1:2:2 ratio.

EXPERIMENT

The transition temperature T_c of a superconducting sample can be measured by its susceptibility variations which can be obtained by determining the ac impedance of a coil containing the sample. The coil that contains the sample is connected to an oscillator. If one writes the inductance as

$$L=L_0(1+4\pi f_0\chi)\ ,$$

where L_0 is the inductance of the empty coil and f_0 is the filling factor of the coil, then a change in the susceptibility, χ, causes a proportional change ΔL in the inductance of the coil. A frequency meter provides a digital output of the oscillator frequency, f, where

$$2\pi f=1/\sqrt{LC}\ ,$$

such that Δf can be observed by a chart recorder after the digital frequency output passes through a digital-to-analog converter. In order to get more sensitivity, the frequency meter was set to read period $(1/f)$ instead of frequency. Figure 1 gives the temperature dependence of $\Delta(1/f)$ for sample No. 1 and sample No. 2. Sample No. 1 and sample No. 2 exhibited sharp superconducting transitions at 0.60±0.03 K and 0.65±0.03 K, respectively, with no precursor diamagnetism above the transition temperatures to better than 10^{-3} the signal change at T_c. Sample No. 3 showed no superconductivity down to 0.35 K, the limit of our ^{3}He evaporation cryostat.

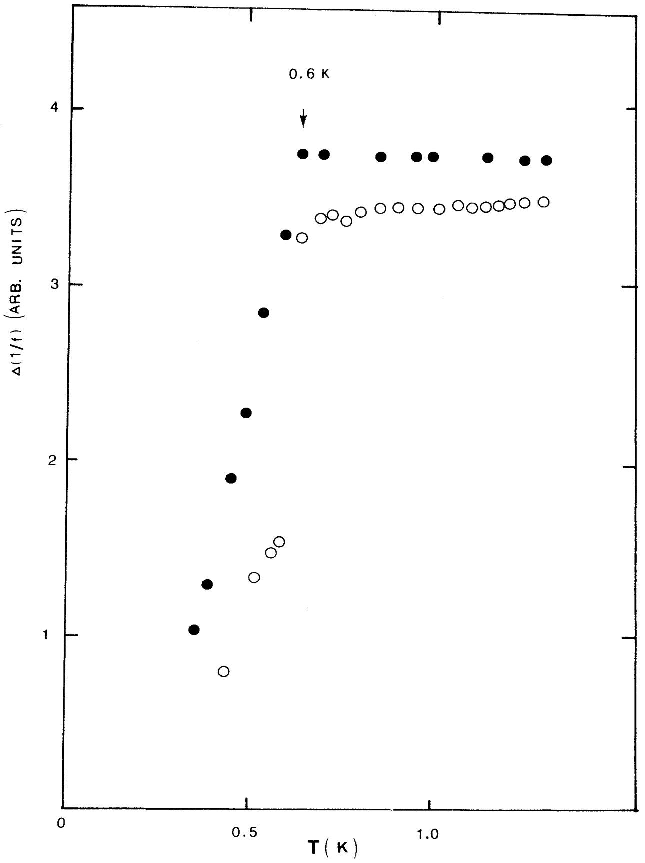

FIG. 1. Temperature dependence of the inductance change in the $CeCu_2Si_2$ sample No. 1 (filled circles) and sample No. 2 (open circles).

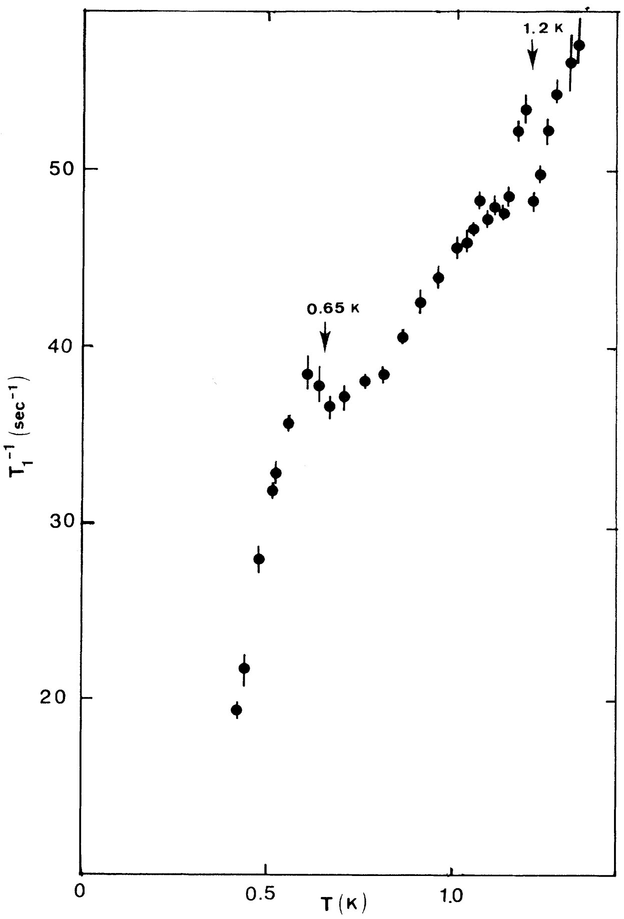

FIG. 2. Temperature dependence of the ^{63}Cu nuclear quadrupole resonance (NQR) longitudinal (spin-lattice) relaxation rate $1/T_1$ in the $CeCu_2Si_2$ sample No. 1.

Conventional pulse nuclear resonance techniques were used to measure spin-lattice relaxation times T_1, spin-echo decay times T_2, and inhomogeneous linewidths $1/T_2^*$ at the ^{63}Cu $(I=\frac{3}{2})$, NQR frequency $\omega_Q/2\pi=3.43$ MHz. The spin-lattice relaxation rate $1/T_1$ was related to the fluctuation noise spectrum $J(\omega)$ of nuclear local-field fluctuations at $\omega=\omega_Q$.[20] Figures 2 and 3 give the

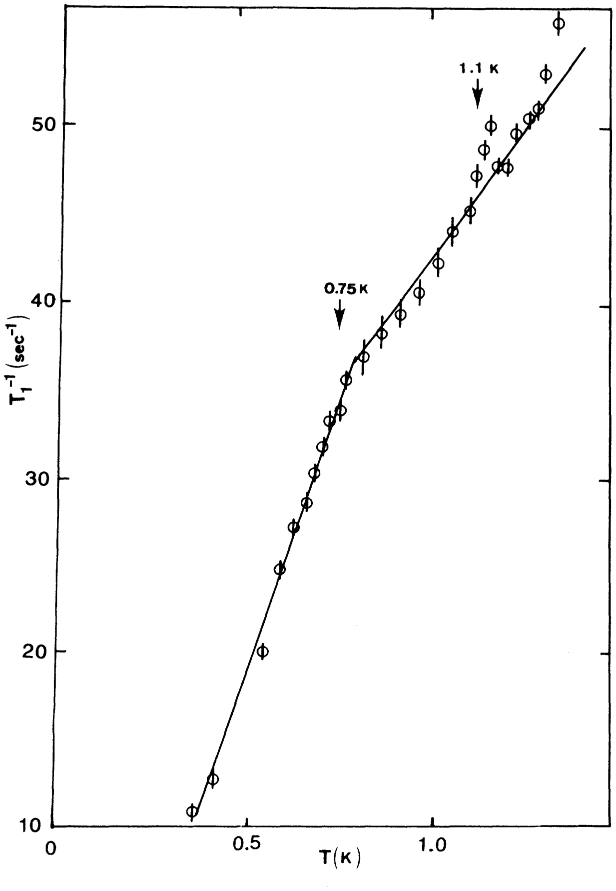

FIG. 3. Temperature dependence of the ^{63}Cu NQR longitudinal (spin-lattice) relaxation rate $1/T_1$ in the CeCu$_2$Si$_2$ sample No. 2.

temperature dependence of the ^{63}Cu spin-lattice relaxation rate $1/T_1$ of sample No. 1 and sample No. 2, respectively, at 90 μm grain size. The $1/T_1$ fall off sharply when the temperature drops below the superconducting transition temperature $T_c \simeq 0.6$ K. Besides the superconducting phase transition, we found a sharp anomaly at 1.1 K, which are well outside the error bars. The T_1 measurements between 1.3 K to 1.0 K were repeated at least three times to confirm that these anomalous peaks were unlikely to be experimental artifacts. A rapid increase of the muon-spin-relaxation rate was found with decreasing temperature below $T \sim 0.8$ K by Uemura *et al.*[5] The 1.1-K anomaly which observed in NQR measurements might correspond to the magnetic ordering that is found by muon-spin-relaxation measurements. Uemura *et al.* claimed that the depolarization of muon spins in zero field could be due either to randomness of the static internal local field H_{int} or to the fluctuating dynamic local fields. Since an external longitudinal field $H_{\text{ext}} = 250$ G suppressed the depolarization of muon spins remarkably, they suggested that the depolarization was due mainly to the static random local field.

Figure 4 gives the frequency dependence of the ^{63}Cu spin-echo signal amplitude S_{SE} at 1.2 K. The line width $\Delta\omega$ is the width at half maximum intensity of resonance line at $\omega = \omega_Q$. The dependences of ω_Q and $\Delta\omega$ on temperature are shown in Figs. 5(a) and 5(b), respectively. The NQR frequency will shift due to the change of electron surrounding of the resonant nucleus. Therefore, the independence of ω_Q on temperature indicates that the 1.1-K anomaly cannot be related to any electronic effect, such as the structural phase transition. The broadening of $\Delta\omega$ below 1.0 K might be due to the static random local field as it is suggested by Uemura *et al.*[5] The dynamics spin fluctuation usually much faster than NQR frequency $\omega_Q/2\pi = 3.43$ MHz; therefore it has no effect on

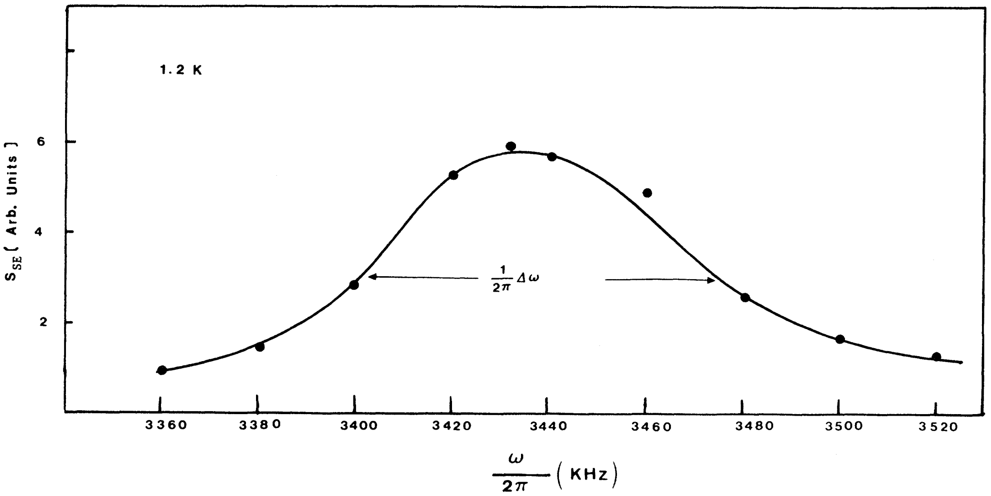

FIG. 4. Frequency dependence of the ^{63}Cu spin-echo signal amplitude S_{SE} in the CeCu$_2$Si$_2$ sample No. 1 at 1.2 K. The line width $\Delta\omega$ is the width at half maximum intensity of the resonance line at $\omega = \omega_Q$.

T_1. The independence of ω_Q also indicates that the anomaly in NQR T_1 measurements will not be made by fluctuating dynamics local field.

One of the most likely spin structures which produces a static random local field is spin-glass ordering. The host nuclear magnetic-resonance measurements in *Cu*Mn spin glasses have been investigated by MacLaughlin and Alloui.[21] They have shown that through the freezing temperature T_g there are gradual decreases in the longitudinal and transverse relaxation times T_1 and T_2, respectively, accompanied by a progressive diminution in the resonance intensity. They also found that the relaxation signal varies nonexponentially, but rather like $\exp(-\sqrt{t/T_1})$. The spin-echo recovery of $CeCu_2Si_2$ sample No. 1 is shown in Fig. 6. The form of the relaxation function of ^{63}Cu can be fitted by a single exponential function to three decades. Since we do not observe a deviation from a single exponential, we conclude that only one macroscopic phase is present. The temperature dependence of the longitudinal relaxation rate in the nonsuperconducting $CeCu_2Si_2$ sample is shown in Fig. 7. The NQR signal of the nonsuperconducting sample is much less than that of the superconducting samples. This phenomenon is according to that the absence of superconductivity can be correlated with the presence of defects, particularly in the Cu sublattice.[22] The reduced

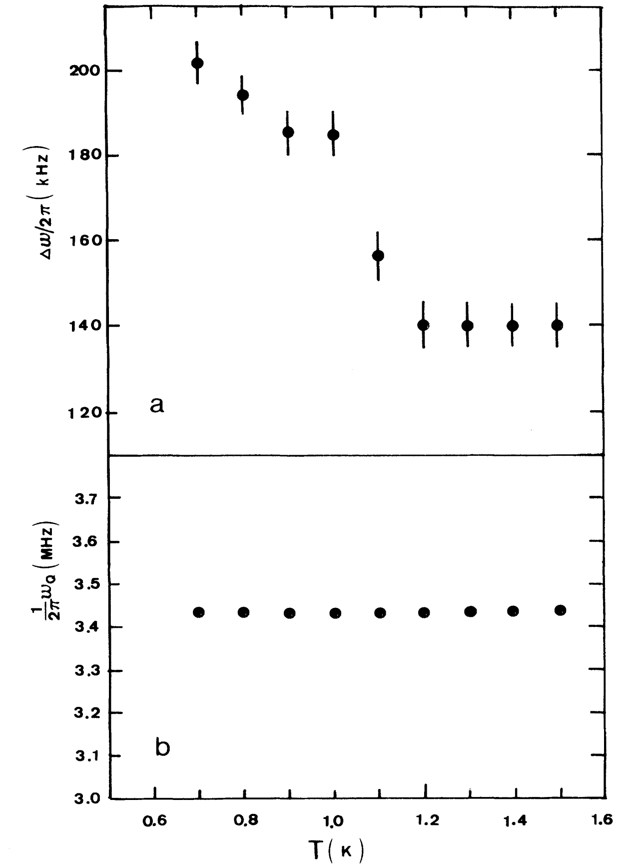

FIG. 5. (a) Temperature dependence of the ^{63}Cu NQR resonance frequency ω_Q in the $CeCu_2Si_2$ sample No. 1. (b) Temperature dependence of the ^{63}Cu NQR resonance line width $\Delta\omega$ in the $CeCu_2Si_2$ sample No. 1.

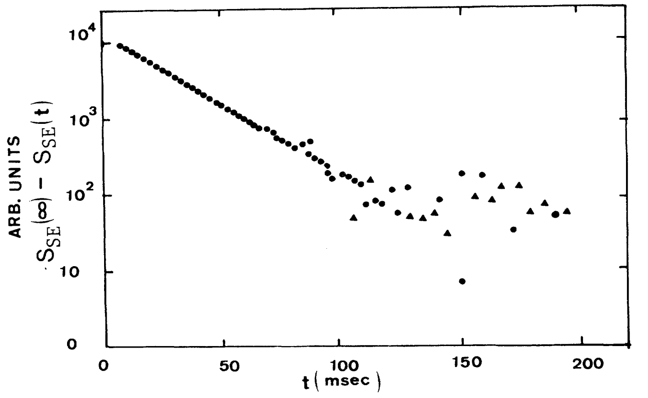

FIG. 6. ^{63}Cu magnetization recovery function $S_{SE}(\infty) - S_{SE}(t)$ vs recovery time t. The spin-echo height $S_{SE}(t)$ is directly proportional to the magnetization $M(t)$. Therefore, if the relaxation process is exponential, $S_{SE}(t) = S_{SE}[1 - \exp(-t/T_1)]$. The triangles denote negative values because of noise.

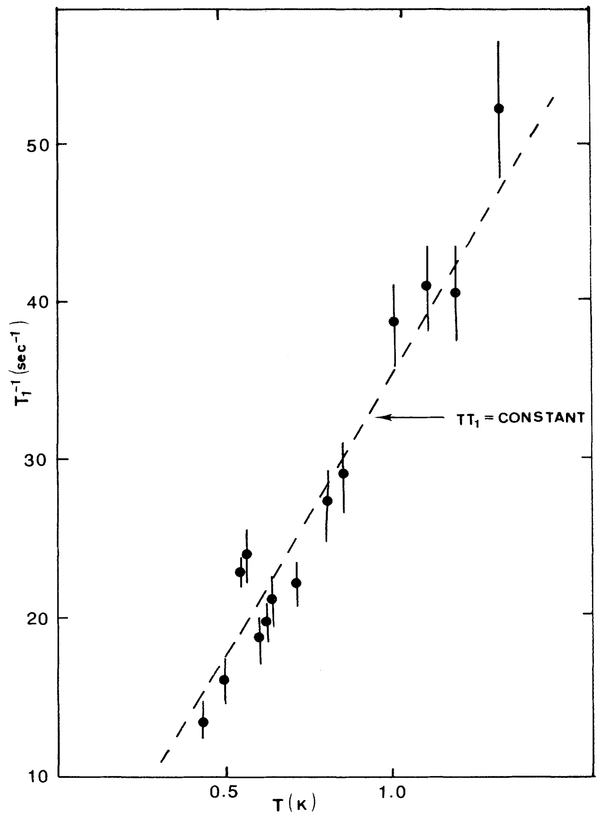

FIG. 7. Temperature dependence of the ^{63}Cu NQR longitudinal (spin-lattice) relaxation rate $1/T_1$ in the nonsuperconducting $CeCu_2Si_2$ sample (sample No. 3).

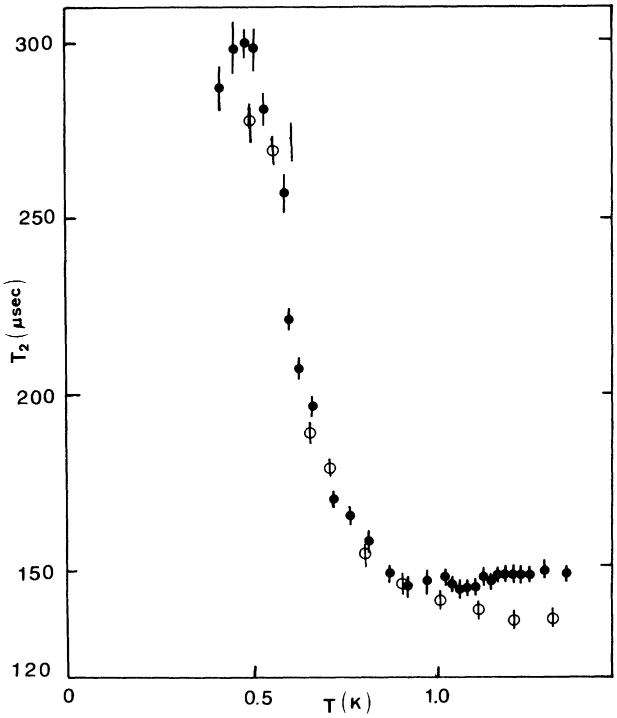

FIG. 8. Temperature dependence of the ^{63}Cu NQR transverse (spin-spin) relaxation time T_2 in the $CeCu_2Si_2$ sample No. 1 (filled circles) and sample No. 2 (open circles).

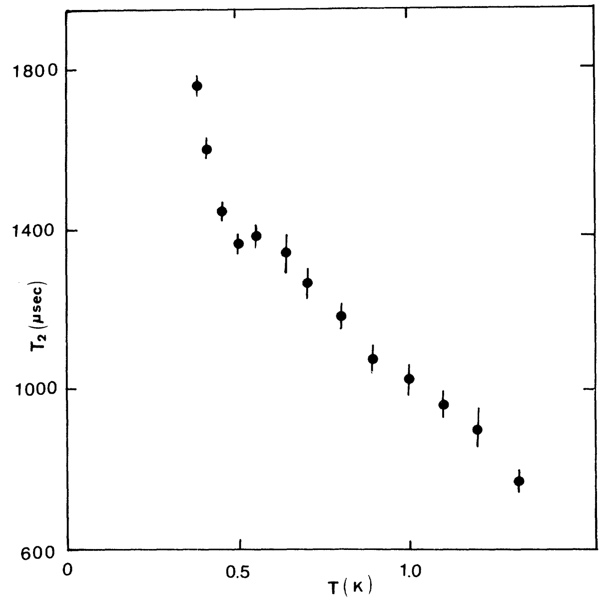

FIG. 9. Temperature dependence of the ^{63}Cu NQR transverse (spin-spin) relaxation time T_2 in the nonsuperconducting $CeCu_2Si_2$ sample (sample No. 3).

signal strength in nonsuperconducting sample means that the observed resonance is representative only of nuclei in unstrained regions of the specimen. However, the reduced NQR signal produced poor signal-to-noise ratio and resolution of the 1-K anomaly would have been difficult even if it were present.

Figure 8 gives the temperature dependences of the transverse relaxation time T_2 in sample No. 1 and sample No. 2, respectively. The most striking result is that an increase of T_2 with decreasing temperature is observed below 0.9 K, instead of the superconducting transition temperature $T_c \simeq 0.6$ K. This does not seem to be due to the onset of superconductivity at 0.6–0.65 K. The static random local fields which are produced by spin-glass ordering as well as an incommensurate spin-density-wave states[23] could "detune" neighboring nuclei and render their mutual dipolar interaction less effective in inducing mutual spin flips. The temperature dependence of T_2 in the nonsuperconducting sample is given in Fig. 9. The temperature dependence of the nonsuperconducting sample is quite different from that of superconducting $CeCu_2Si_2$. The transverse relaxation time T_2 increases linearly with decreasing temperature. No anomaly at 0.9 K was observed. The slight change of slope in T_2 near 0.5 K might be due to a small amount of sample that becomes superconducting. Besides, T_2 in the nonsuperconducting sample is much longer than that in the superconducting samples. Long T_2 might also be related to that the copper sublattice is considerably less perfect in a nonsuperconducting sample than that in a superconducting sample. The spin-echo amplitude $S_{SE}(t)$ of ^{63}Cu in the $CeCu_2Si_2$ sample No. 1 versus the $\pi/2$-π rf pulse separation time t is plotted in Fig. 10. The transverse relaxation of ^{63}Cu is quite close to being exponential.

The NMR signal intensity in a spin-glass sample will considerably lose when the temperature nears T_g.[24] The lost intensity near T_g results from an experimental difficulty that the unobserved nuclei possess very short spin-echo decay time T_2 so that the echo from those nuclei decays before the spectrometer has recovered from rf pulse overload.[24] In (U,Th)Be_{13} the spin-density-wave long-range order appears below the superconducting transition temperature and coexists with it,[6] however, the ^{9}Be NMR signal intensity is not reduced. It implies that the spin-density wave will not significantly change the NMR signal but the spin-glass state will. The product TS_{SE} of the temperature T and the ^{63}Cu NQR spin-echo amplitude S_{SE} (normalized to the sample mass) in the superconducting $CeCu_2Si_2$ sample No. 1 and the nonsuperconducting $CeCu_2Si_2$ sample No. 3 are given in Fig. 11. The spin-echo amplitude S_{SE} is proportional to the average ^{63}Cu nuclear magnetic moment M. According to the nuclear Curie law, M is in inverse proportional to T, and so does S_{SE}. Therefore, by forming the product TS_{SE}, the temperature dependence of S_{SE} is removed. As shown in Fig. 11, the significant loss of signal that is observed below the superconductive transition temperature T_c for sample No. 1 can be partially attributed to the expulsion of rf field in the superconducting state. Besides, we notice that in Fig. 11, the signal reduction in the superconducting samples is observed above the T_c determined by the superconducting diamagnetism in the ac susceptibility, $T_c(\chi_{ac})$, and above the T_c determined by spin-lattice relaxation time, $T_c(T_1)$. However, the reduc-

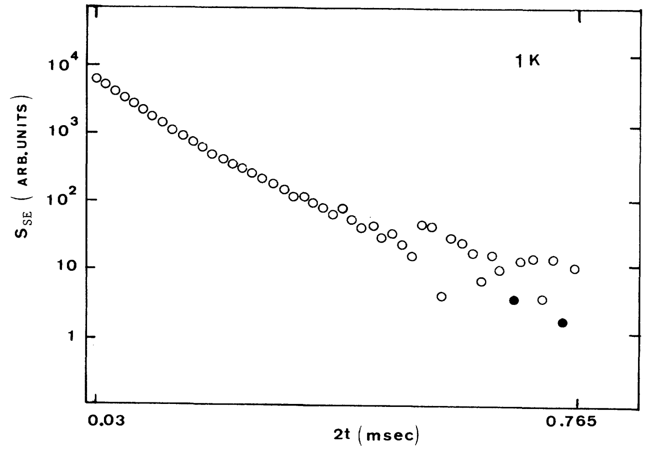

FIG. 10. The spin-echo amplitudes S_{SE} of ^{63}Cu in the $CeCu_2Si_2$ sample No. 1 vs $2t$ where t is the separation of $\pi/2-\pi$ rf pulses. The filled circles denote negative values because of noise.

tion of the NQR signal at $0.6\ K < T < 0.9\ K$ in $CeCu_2Si_2$ is so small[24] that it is difficult to be interpreted as an effect of a spin-glass-type phase transition. NQR signal amplitude measurements in the nonsuperconducting sample show no appreciable variation in TS_{SE} down to 0.35 K, which is consistent with the absence of a superconducting transition in the ac susceptibility. The significant loss of signal in the nonsuperconducting sample relative to the superconducting sample might also be due to that the copper sublattice is considerably less perfect in nonsuperconducting sample than that in superconducting sample.

The NQR signal will decay due to the field inhomogeneity as well as the spin-lattice and spin-spin interactions. The time constant which describes the NQR signal decay is denoted as T_2^* because it is so closely related to T_2. Without any field inhomogeneity, the time constant is T_2; therefore,

$$1/T_2 = 1/T_2^* + \gamma_n \Delta H_0 ,$$

where ΔH_0 is field inhomogeneity and γ_n is a constant. Usually $1/T_2^*$ is called the inhomogeneous linewidth.

The T_2^* measurements of $CeCu_2Si_2$ sample No. 1 is shown in Fig. 12. Below T_c, T_2^* decreases sharply. A dip near 0.9 K is observed, which might correspond to the onset of static magnetic ordering in the muon-spin-relaxation measurements.[5] The T_2^* measurements were repeated many times to confirm that this anomalous deep was unlikely an experimental artifact. However, this behavior is not clear in the $CeCu_2Si_2$ sample No. 2 T_2^* measurements (Fig. 13). The shortening of T_2^* near 0.9 K suggests that, in the frequency domain, the NQR signal of ^{63}Cu is broader. The static random field which is caused either by the spin-glass or by incommensurate

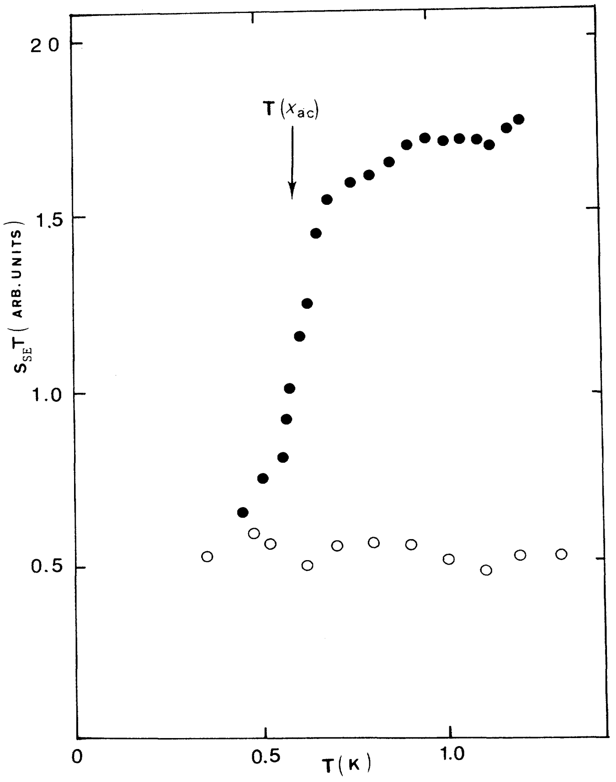

FIG. 11. Product TS_{SE} of temperature and the ^{63}Cu NQR spin-echo signal amplitude S_{SE} (normalized to sample mass) in sample No. 1 (filled circles) and sample No. 3 (open circles).

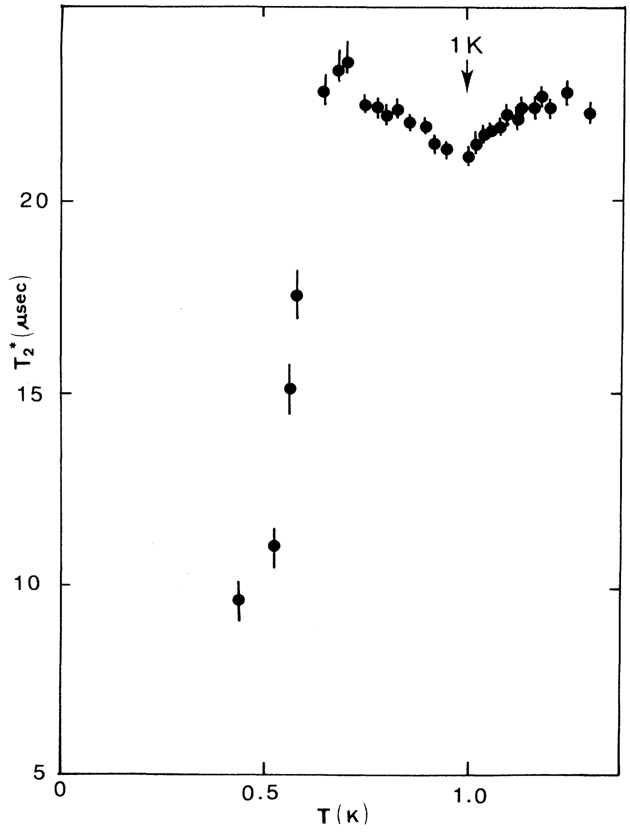

FIG. 12. Temperature dependence of the NQR signal decay time T_2^* in sample No. 1.

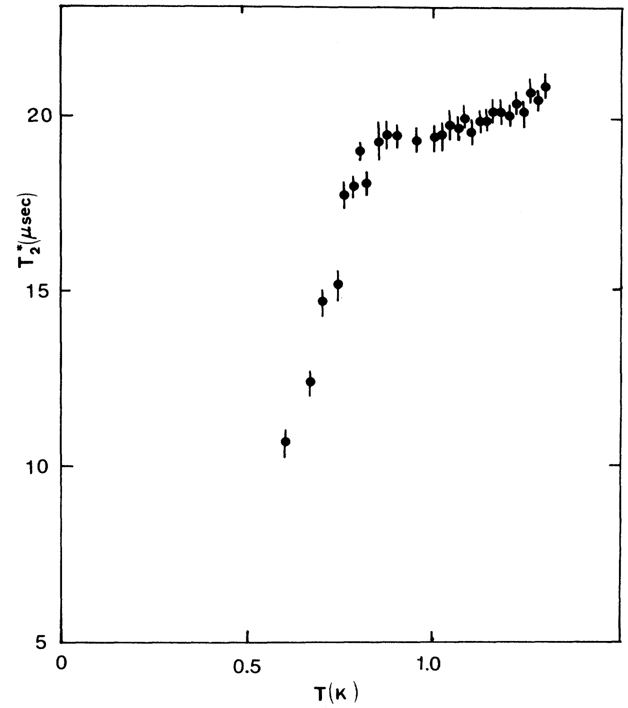

FIG. 13. Temperature dependence of the NQR signal decay time T_2^* in sample No. 2.

spin-density-wave states will cause the broadening of the ^{63}Cu NQR spectrum. The T_2^* measurements of the nonsuperconducting $CeCu_2Si_2$ sample are shown in Fig. 14. The T_2^* of the nonsuperconducting $CeCu_2Si_2$ is much shorter than that of superconducting samples. The shortening of T_2^* in nonsuperconducting $CeCu_2Si_2$ suggests that the copper sublattice in the nonsuperconducting $CeCu_2Si_2$ is considerably less ordered than that in the superconducting sample, so that the internal electric field gradient experienced by the Cu nucleus at the different position is not the same. This fact is consistent with the large reduction of the ^{63}Cu spin-echo amplitude in the nonsuperconducting $CeCu_2Si_2$ sample.

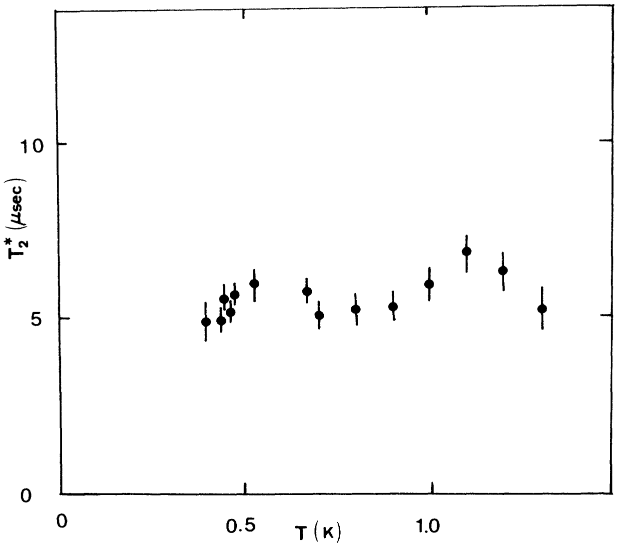

FIG. 14. Temperature dependence of the NQR signal decay time T_2^* in sample No. 3.

By measuring the electrical resistivity of $CeCu_2Si_2$, Bellarbi *et al.*[25] found a steep increasing pressure to about 2 K near 25 kbar, and found no evidence for superconductivity about 1.2 K for $p > 100$ kbar. Nevertheless, in their data, T_c first rises slowly between 0 and 15 kbar, then seems to level off up to ~25 kbar, with $T_c \sim 1$ K from 15 kbar to 25 kbar. One might expect that the anomaly of NQR measurements in $CeCu_2Si_2$ at ~1 K is due to a part of the sample under an internal pressure in the 15–25-kbar caused by the sample imperfections, such that a small amount of the sample become superconducting before the ambient pressure $T_c \sim 0.7$ K. During the experiment, samples were powdered and sieved to <90 μm, and none was heat treated after powdering. Since the internal stress in the $CeCu_2Si_2$ samples will depend on the size of the sample, the samples were further powdered to 45 μm; there is no sample size effect observed. Therefore, the anomaly of NQR measurements in $CeCu_2Si_2$ at ~1 K is not caused by internal pressure. Besides, no diamagnetism in the susceptibility measurements being observed at ~1 K also suggests that the 1-K anomaly in $CeCu_2Si_2$ is not due to a part of the sample become superconducting.

CONCLUSIONS

Nuclear quadrupole resonance experiments on $CeCu_2Si_2$ have revealed a transition at ~0.9 K. Since zero-field μSR measurements on this heavy-fermion system have shown a clear evidence of static magnetic ordering below ~0.8 K, the exotic transition observed in NQR measurements should be closely associated with the static magnetic ordering in μSR measurements. The line shapes of the observed muon-spin-depolarization function suggest an ordering in either the spin-glass or incommensurate spin-density-wave state with a very small averaged moment of ~0.1 μ_B. The NQR measurements suggest that the ordering is unlikely the spin-glass type. NQR in a nonsuperconducting specimen reveals the presence of considerable disorder in the copper sublattice such that the spin-density wave may not exist in nonsuperconducting $CeCu_2Si_2$. The superconducting transition temperature T_c can be enhanced by the presence of spin-density wave.[26] Therefore, if the spin-density wave is depressed, $CeCu_2Si_2$ will lose the superconductivity.

ACKNOWLEDGMENTS

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