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MUON SPIN RELAXATION IN $CeCu_2Si_2$ AND MUON KNIGHT SHIFT IN VARIOUS HEAVY-FERMION SYSTEMS

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Positive muon spin precession has been observed in various heavy-fermion systems in the transverse external magnetic field. In the superconductor $CeCu_{2.1}Si_2$, the relaxation rate of muon spins increases rapidly with decreasing temperature below T_c. This is interpreted as the results of the inhomogeneous fields due to the imperfect penetration of the external field into the type-II superconducting state. The magnetic-field penetration depth λ is derived from the observed muon spin relaxation rate. λ is about 1200 Å at $T \sim 0.5T_c$, and the temperature dependence of λ is consistent with the relation expected for a BCS superconductor. We have also measured the muon Knight shift K_μ in the normal (or paramagnetic) state of various heavy-fermion systems. K_μ is large and negative (about $-1000 \sim -3000$ ppm at T = 10 K) for $CeCu_2Si_2$, UPt_3 and $CeAl_3$, while more complicated signals are measured in $CePb_3$ and CeB_6. The negative muon Knight shift in the non-magnetic heavy-fermion systems is discussed in terms of the Kondo-coupling between the conduction- and f-electrons.

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1. Introduction

Some intermetallic compounds of Cerium or Uranium, called heavy-fermion systems, have been the subject of recent extensive investigations /1/. The heavy-fermion systems are characterized by the extremely large coefficient $\gamma \sim 1$ Joule/mole K^2 for the linear term C/T of the specific heat C at low temperatures, as well as by the large magnetic susceptibility which becomes increasingly Pauli-like with decreasing temperatures. These features suggest the extremely enhanced electronic density of states at the Fermi level (i.e., a resonance with very narrow energy width of $1 \sim 10$ meV) due to quasi particles with the effective mass of about $100 \sim 1000$ times larger than the bare electron mass.

There are several different groups of materials among the heavy-fermion systems. $CeCu_2Si_2$, UBe_{13} and UPt_3 become superconducting below $T_c = 0.5 \sim 1.0$ K. Intense experimental and theoretical studies are now underway to clarify the mechanism of superconductivity of these materials, which might be different from the conventional BCS model. $CePb_3$ and CeB_6 order antiferromagnetically at low temperatures $1 \sim 3$ K, while $CeCu_6$ and $CeAl_3$ do not order magnetically nor become superconducting to the lowest temperature of current experiments.

In this paper, we first report muon spin relaxation measurement in the heavy-fermion superconductor $CeCu_2Si_2$ above and below $T_c \sim 0.7$K. The relaxation rate observed in the transverse external magnetic field increased rapidly with decreasing temperature below T_c. This is interpreted as the result of the imperfect penetration of the external field into the superconducting state, and the penetration depth λ is obtained from the observed relaxation rate.

We also report positive muon Knight shift study on various heavy-fermion systems in the normal (or paramagnetic) state. The Knight shift K_μ of the muon spin reflects the electron spin density at the muon site, which is usually some interstitial site in the crystal lattice. We compare the results for the different groups of heavy-fermion systems, and discuss the possible relation to the Kondo coupling between conduction- and f-electrons.

2. Muon Spin Relaxation in $CeCu_2Si_2$ below T_c

At the AGS muon channel of Brookhaven National Laboratory /2/, we have carried out the muon spin precession measurement on $CeCu_{2.1}Si_2$ above and below the superconducting transition temperature T_c. Our initial motivation was to study the spin susceptibility below T_c via the muon Knight shift K_μ. Unfortunately, we had difficulty in measuring K_μ accurately below T_c due to the low beam intensity, low external field, high background and fast relaxation.

Instead, however, we found that the relaxation rate σ, which represents the damping of the precession signal as $\exp(-0.5\sigma^2t^2)$, increases rapidly with decreasing temperature below T_c. Figure 1 shows the temperature dependence of σ measured in the transverse external field of 1 kG with a poly-crystal sample of $CeCu_{2.1}Si_2$ placed in a ^3He cryostat. The sample was made to be slightly off-stoichiometric in order to stabilize the superconductivity with $T_c \sim 0.7$ K.

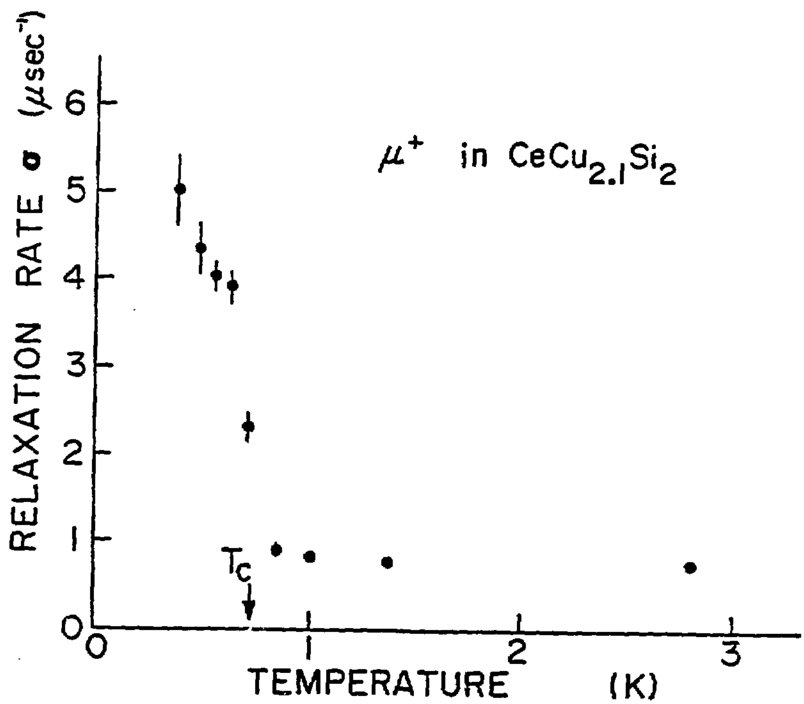

Figure 1. Muon spin relaxation rate σ in $CeCu_{2.1}Si_2$ ($T_c \sim 0.7$ K) observed in the transverse external magnetic field of 1 kG. σ represents the damping of the muon spin precession envelope as $\exp(-0.5\sigma^2t^2)$.

The rapid increase of σ below T_c can be interpreted as the result of the imperfect penetration of the external field into the specimen. Such an effect was previously studied by μSR on some BCS superconductors, e.g., Nb /3/. $CeCu_2Si_2$ is a type-II superconductor where the external field penetrates as the vortices. Each vortex carries the magnetic flux quanta $\phi \approx 2 \times 10^{-7}$ G cm^2. For the simplicity, we make the following calculation assuming the square lattice of the vortices instead of the triangular Abrikosov structure.

When we apply H_{ext} = 1 kG, there are 5×10^9 vortices per cm^2. The distance d of the adjacent vortices then becomes about 1400 Å. The mean square average of the inhomogeneous field $< (\Delta H)^2 >^{1/2}$ is given by /4/

$$< (\Delta H)^2 >^{1/2} = (H_{ext}/\sqrt{4\pi})\cdot(d/\lambda)\cdot\{1+(2\pi\lambda/d)^2\}^{-1/2} \simeq (\phi/\lambda^2)\cdot(1/\sqrt{16\pi^3})$$

where λ is the magnetic-field penetration depth. The relaxation rate σ in Fig. 1 corresponds to $\gamma_\mu<\Delta H>$ where $\gamma_\mu = 2\pi \times 13.5$ MHz/kG. Then one can directly calculate λ from the muon spin relaxation rate.

Figure 2 shows the penetration depth λ thus derived from the observed relaxation rate σ. We would like to note the following points: (1) The absolute values of λ are considerably smaller than the values obtained for UBe_{13} by a squid magnetization method /5/. (2) The temperature dependence of λ is consistent with the relation $\lambda(T) = \lambda(0)\cdot\{1-(T/T_c)^4\}^{-1/2}$ expected for a non-local BCS superconductor with nearly isotropic energy gap. The broken line of Fig. 3 shows this relation for $\lambda(0)$ = 1190 Å and T_c = 0.8 K, which agrees reasonably with the experimental results.

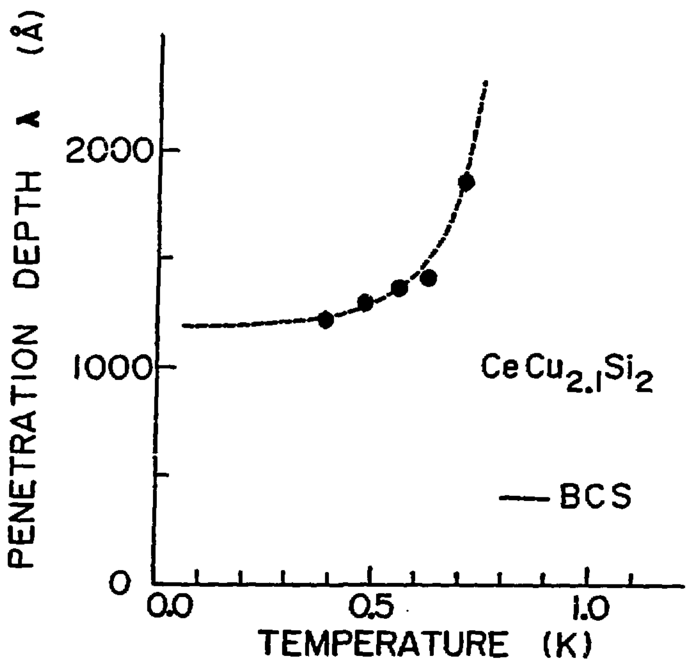

Figure 2. The magnetic-field penetration depth λ in the superconducting state of $CeCu_{2.1}Si_2$ derived from the muon spin relaxation rate shown in Fig. 1. The square lattice of the flux vortices is assumed in the calculation instead of the triangular Abrikosov structure. The broken line represents the relation expected for a BCS superconductor $\lambda(T) = \lambda(0)\cdot\{1-(T/T_c)^4\}^{-1/2}$ with $\lambda(0)$ = 1190 Å and T_c = 0.8 K.

This is the first direct observation of the penetration depth in $CeCu_2Si_2$. The present results can be explained by the conventional BCS model, but this does not necessarily preclude other models of the superconductivity. The comparison with other models and the detailed aspect of the present work will be published elsewhere.

3. Muon Knight Shift in the Normal State of Heavy-Fermion Systems

We have also measured the muon Knight shift in the normal (or paramagnetic) state of $CeCu_2Si_2$, UPt_3, $CeAl_3$, $CePb_3$ and CeB_6. This measurement was performed mainly at TRIUMF with the external magnetic field $H_{ext} \sim 3.5$ kG, while partly ($CeCu_2Si_2$) at AGS/BNL with $H_{ext} \sim 1$ kG. The sample of $CeCu_2Si_2$ was in the form of powder, CeB_6 as a single crystal, while the other three specimens were polycrystals.

Figure 3 shows an example of the raw shift (before the correction for the demagnetization, Lorentz, and dipolar fields) measured in $CeAl_3$. Although the resistivity of this material shows a maximum around $T = 20 \sim 30$ K, the frequency shift keeps the linear relation with the uniform susceptibility χ between $T = 6 \sim 130$ K.

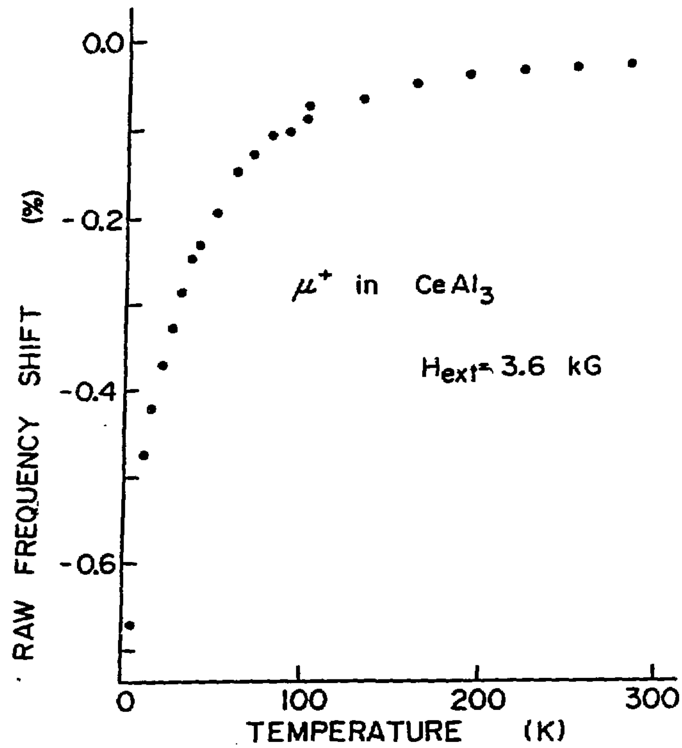

Figure 3. Positive muon Knight shift in the non-magnetic heavy-fermion system $CeAl_3$ observed in the transverse external magnetic field of ~ 3.5 kG. The raw frequency shift is plotted without corrections for the demagnetization, Lorentz, and dipolar fields. The shift is proportional to the uniform susceptibility between $T = 6 \sim 130$ K.

The amplitude and sign of the muon Knight shift in various heavy-fermion materials at low temperatures are summarized in Table 1. The Knight shift is approximately equal to the measured raw frequency shift in the case of the powder sample of $CeCu_2Si_2$. For $CeAl_3$ and UPt_3, corrections are made for the demagnetization and Lorentz fields. These corrections are subject to the systematic error of ∿ 20% due to the irregular shape of the specimens. Furthermore, we can not make correction for the atomic dipolar field within the Lorentz cavity, because we do not know the muon site in each material. Nevertheless, as seen in Table 1, the overall correction is considerably smaller than the measured raw shift, and one can obtain a rough estimate for the sign and amplitude of the Knight shift.

In the superconductors $CeCu_2Si_2$ and UPt_3, the dominant signal shows large negative Knight shift ∿ -1000 ∿ -2000 ppm. This is consistent with the case for the muon Knight shift in UBe_{13} measured at Los Alamos /6/. We also observed the negative shift in the non-magnetic $CeAl_3$. In contrast, a few different signals with positive and negative shifts were observed in $CePb_3$, and small and large positive raw shifts were observed in CeB_6. The overall tendency for the six different materials indicates the simple negative shift for the superconducting and non-magnetic heavy-fermion systems, whereas somewhat more complicated signals for $CePb_3$ and CeB_6 which have the antiferromagnetic ground states.

We would like to notice here that the nagative muon Knight shift might be related to the Kondo coupling between conduction- and f-electrons. The resistivity of heavy-fermion systems often shows an increase with decreasing temperature around T = 30 ∿ 300 K, resembling to the Kondo effect in single-impurity systems such as dilute CuFe. The effective magnetic moment, calculated from the uniform susceptibility, decreases at low temperatures in heavy-fermion systems. These phenomena are related to the spin-dependent scattering of the conduction electrons by the unpaired f-electrons via the so-called Kondo coupling.

Table 1. Positive muon Knight shift in various heavy-fermion systems.

Material		Temp.	Raw Shift (ppm)	$\frac{4}{3}\pi\chi$ (ppm)	Real Shift (ppm)
$CeCu_2Si_2$	Super.	10	- 1800	550	- 1800
UPt_3	Super.	10	- 2300	990	- 950
$CeAl_3$	Non-mag.	10	- 4700	1400	- 2900
$CePb_3$	Antiferro.	10	+ 9800 + 4100 - 1700 - 4200		
CeB_6	Antiferro.	45	+ 300 + 6500		
c.f. for comparison					
UBe_{13} (Los Alamos; Ref. /2/)	Super	1 - 10	- 2500		
Pd (ref. /7/)		300	- 124	261	- 252

These features for the resistivity and effective moment are expected when the conduction- and f-electrons are coupled with the effectively negative exchange interaction. The negative interaction would polarize the spins of conduction electrons opposite to the direction of the external field when the f-electrons are polarized along the external field.

Although we do not know the exact location of the muon site, nor the possible perturbation on the electronic system caused by the positive charge of μ^+, it is likely that muons are probing the spin density of conduction electrons at the site distant from the Cerium or Uranium atoms. In this context, the negative Knight shift is qualitatively consistent with the negative polarization of conduction electrons expected for the Kondo coupling. It would also be interesting to speculate that the complicated signals from the magnetic heavy-fermion systems $CePb_3$ and CeB_6 might be related to the oscillatory RKKY interaction which helps the magnetic ordering.

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