NMR spin-lattice relaxation rate of heavy fermion superconductor UBe$_{13}$

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Abstract. $^9$Be NMR measurements have been carried out for a single crystal UBe$_{13}$ with $T_c = 0.85$ K, in order to clarify unusual properties in the normal state. For the applied field parallel to [111] direction, the quadrupole split Be(II) lines gather around the central line. $^9$Be nuclear spin-lattice relaxation rate was measured for Be(II) sites for $H \parallel [111]$ at 0.85, 7, 15 T. $1/T_1$ does not depend on applied fields above $T_{NMR} \approx 5 - 9$ K and weakly depends on temperature. $1/T_1$ at 0.85 and 7 T is proportional to $T^n$ with $n = 0.5 - 0.6$ down to $T = 2$ K, suggesting that antiferromagnetic spin fluctuations exist. On the other hand, $1/T_1$ is suppressed by applied magnetic field. The present field dependence of $1/T_1$ at low temperatures would give important information about the formation of the heavy quasiparticles in UBe$_{13}$.

1. Introduction

Since the discovery of heavy Fermion superconductivity in UBe$_{13}$ having the extremely large electronic specific heat coefficient of $\gamma_e = 1100$ mJ/(mol K$^2$),[1, 2] extensive experimental and theoretical works have been carried out in order to clarify the nature of the superconducting state of UBe$_{13}$. For instance, specific heat [1, 2], neutron diffraction [3, 4], $\mu$SR [5, 6], and NMR measurements [7, 8, 9, 10], etc., have probed the unconventional nature of both normal and superconducting states.

The superconducting transition occurs at around $T_c \approx 0.85$ K with large specific heat jump, $\Delta C/\gamma_e T_c \approx 1$ [1, 2], indicating that the heavy-quasiparticles are responsible for the superconductivity. In the superconducting state, the power law temperature dependence of the specific heat [2], the NMR spin-lattice relaxation rate [8], etc., suggest that the superconducting energy gap vanishes at points and/or lines on the Fermi surface. These features have been interpreted as evidence for an anisotropic pairing state, most likely a $p$-wave triplet state, in UBe$_{13}$.

In the normal state, the temperature dependence of the electric resistivity shows $\rho(T) \propto -\ln T$ above 40 K [1, 11, 12], associated with the Kondo scattering. On the other hand, $T^2$ of resistivity and Pauli paramagnetic susceptibility expected for the heavy Fermi liquid state are not observed down to 2 K; $\rho(T)$ is almost temperature independent in the temperature range of 40 K – 2 K, and decreases gradually with decreasing temperature below $T^* = 2$ K.
down to $T_c$. $\rho(T_c) \approx 139 \, \mu\Omega\text{cm}$ is quite large in comparison with that in the other heavy fermion superconductors [12]. Temperature dependence of magnetic susceptibility $\chi(T)$ shows logarithmic divergence down to 2 K. These anomalies have attracted much attention as a non-Fermi liquid behavior. In order to explain the non-Fermi liquid behavior in UBe$_{13}$, a two-channel Kondo effect is proposed theoretically [13].

In order to investigate the normal state in UBe$_{13}$, we report magnetic field dependence of $^9$Be NMR relaxation rate $1/T_1$ in the single crystal UBe$_{13}$ at various magnetic fields.

2. Experimental

Single crystals of UBe$_{13}$ were grown by the Al-flux method. Details of sample preparation techniques for single crystals of UBe$_{13}$ was reported elsewhere [11]. Samples were characterized to be a single phase by a Laue photograph as well as the X-ray diffraction. UBe$_{13}$ crystallizes in the cubic NaZn$_{13}$-type structure (space group (Fm3c)) with the lattice constant of $a = 10.257$ Å, where the U atoms are in the position of 8(a) site of (0, 0, 0), $\pm(1/2, 1/2, 1/2)$, and Be atoms have two crystallographically inequivalent sites. Be(I) atoms are in the position of 8(b) of $\pm(1/4, 1/4, 1/4)$, and Be(II) atoms are in the position of 96(i) sites. The superconducting phase transition was confirmed to occur at $T_c \approx 0.85$ K by means of DC electrical resistivity and surface impedance measurements [13, 14].

![Figure 1](image-url). $^9$Be NMR spectra measured at $H = 7$ T and for $H \parallel [001]$ and $H \parallel [111]$.

$^9$Be-NMR measurements were carried out by using a conventional pulsed NMR spectrometer in the temperature range of 1.5-100 K. A field angle was determined from the angular dependence of the $^9$Be-NMR peak position. The field calibration was carried out with $^{27}$Al resonance ($^{27}K \approx 0.161\%$ at 4.2 K) of a reference sample [12]. We confirmed that the linewidth of $^9$Be-NMR spectrum measured at $H = 0.85$ T and for $H \parallel [001]$ is as quite narrow as $\approx 10$ G. Furthermore no extra signals associated with impurity phases were observed [12], which guarantee the high-quality of the single crystal from a microscopic level. In order to investigate the normal state in UBe$_{13}$, we report temperature dependence of the $^9$Be NMR relaxation rate $1/T_1$ in the single crystal UBe$_{13}$ at $H = 0.85$, 7 and 15 T and for $H \parallel [111]$. 

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3. Results and Discussions

Figure 1 shows $^9$Be NMR spectra measured at $H = 7$ T and for $H \parallel [001]$ and $H \parallel [111]$. When the magnetic field is applied to $[111]$ direction, quadrupole split Be(II) lines merge into the central resonance line around 7 T because the effect of the electric field gradient (EFG) is effectively canceled out. The peak observed around 7.012 T corresponds to the signal from Be(I) atoms in 8(b) site [12, 15].

Typical examples of $^9$Be-NMR relaxation curves for the Be(II) site are shown in Fig. 2(a) and (b). Since the effect of the EFG is canceled out for $H \parallel [111]$, the relaxation curve shows a single exponential relaxation curve, which can be fitted by $(M_0 - M(t))/M(t) = \exp(-t/T_1)$ by two digits. Therefore $1/T_1$ can be determined uniquely. The solid lines are the results of the fitting. Temperature dependence of $1/T_1$ was measured at the main peak of the Be(II) line.

Figure 2. (a) $^9$Be NMR spin-lattice relaxation curve for $H = 0.85$ T and $T = 1.5$ K. (b) $^9$Be NMR spin-lattice relaxation curve for $H = 15$ T and $T = 1.6$ K. (c) Temperature dependences of $1/T_1$ for $H \parallel [111]$ and at $H = 0.85$ (open triangles), 7 (closed circles), and 15 T (open circles). Solid lines in (a) and (b) are fits to relaxation data (see text). Solid line in (c) represents $1/T_1 \propto T_{0.15}^{\infty}$ as a guide for eyes.

Figure 2(c) shows the temperature dependences of $1/T_1$ in the paramagnetic normal state measured at $H = 0.85, 7, 15$ T. Overall temperature dependences of $1/T_1$ for $H = 0.85$ T and 7 T are almost consistent with the temperature dependence of $1/T_1$ reported by MacLaughlin et al.[7]. $1/T_1$ does not depend on applied fields above 10 K and exhibits weak temperature dependence. The solid line in Fig. 2(c) is a guide for the eyes for $1/T_1 \approx T_{0.15}^{\infty}$. With decreasing temperature, $1/T_1$ deviates from the line at $T_N^{\text{MR}}$. $T_N^{\text{MR}}$ is obtained as $T_N^{\text{MR}} \approx 5$ K for $H = 0.85$ T and 7 T, whereas $\approx 9$ K for $H = 15$ T. Below $T_N^{\text{MR}}$, the Korringa relation of $1/T_1 T = const.$, which is connected to the heavy Fermi liquid state, cannot be observed, but rather it obeys $1/T_1 \propto T^n$ with $n = 0.5 - 0.6$ down to $T = 2$ K, suggesting a non Fermi liquid (NFL) state by spin fluctuations near the quantum critical point.

Below $T_N^{\text{MR}}$, $1/T_1$ at $H = 15$ T is suppressed compared to that at $H = 0.85$ T and 7 T. Such a suppression of $1/T_1$ by applying magnetic fields is analogous with the breakdown of the heavy Fermi liquid state. Then, if one assumes that the heavy-quasiparticles are formed through Kondo mechanism, the characteristic critical field, $H^*$, corresponding to the breakdown of the Kondo coherency is estimated to be $7 \, T < H^* < 15$T. This field range is comparable to
\( T^*_{NMR} \approx 5 - 9 \) K, suggesting that the Kondo mechanism works for the formation of the heavy quasiparticles in UBe\(_{13}\).

However, one can find that the NFL like behavior observed below \( T^*_{NMR} \) is robust against the applied magnetic field. These features are contrasted with the typical relaxation behavior of the heavy Fermion system, in which \( 1/T_1 \) undergoes a moderate crossover from the \( 1/T_1 = \text{const.} \) behavior at higher temperatures than a characteristic temperature, \( T^K \approx T_K \), to the \( T_1T = \text{const.} \) behavior at low temperatures. Actually, the temperature independent Pauli susceptibility, which is expected for the heavy Fermi liquid state, is not observed down to \( \approx 2 \) K [12]. No observation of the Korringa relation, i.e., the NFL like behavior, implies the competition between the Kondo effect and other effect, e.g. crystal electric field states and/or multipoles, which would disturb the formation of the Kondo singlet[16].

4. Summary

The NMR results in this study provide insight into how the magnetic response is modified by applied fields. \( 1/T_1 \) does not depend on applied fields above \( T^*_{NMR} \approx 5 - 9 \) K and weakly depends on temperature. Below \( T^*_{NMR} \), \( 1/T_1 \) starts to decrease which is reminiscent of the development of the Kondo coherency. However \( 1/T_1 \) does not follow the Korringa relation. Especially for \( H = 15 \) T, the Korringa relation cannot be observed though \( 1/T_1 \) is suppressed compared with that at low fields. The NFL like behavior implies the competition between the Kondo effect and other effect, e.g. crystal electric field states and/or multipoles, which would disturb the formation of the Kondo singlet.

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