Ru NMR and NQR Studies in $CeRu_2$ *

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We have carried out Ru NMR and NQR measurements in superconductor $CeRu_2$ having T_c = 6.1 K in the temperature range of 1.3 - 30 K. From the ^{99}Ru NMR in 10.2 T above H_{c2}, T_1T = const was found in the measured-T range, and the value of T_1TK^2 is almost the same as that in the uncorrelated Korringa relation, suggesting that there does not exist the strong spin fluctuation of Ru d-spins. The property of the superconducting state was investigated by using Ru NQR. We observed the Hebel-Slichter peak just below T_c and the exponential decrease in $1/T_1$, indicating that this superconductivity has an ordinary s-wave character. The effect of Al substitution for Ru was also studied by NMR and NQR.

Key words: Superconductor $CeRu_2$, NMR, NQR, T_1, Knight shift.

Introduction

Much interest has been paid to $CeRu_2$ due to the coexistence of superconductivity and magnetic ordering when rare-earth impurities are doped into the Ce site [1, 2]. In addition, recently much attention has been focused on a sharp transition from reversible to irreversible behavior in the superconducting mixed state near H_{c2} in $CeRu_2$ [3, 4]. This behavior may be described by the Fulde-Furrell-Larkin-Ovchinnikov (FFLO) state [5, 6, 7]. Moreover, the possibility of multiple superconducting phases as in UPt_3 was pointed out in $CeRu_2$ from ac susceptibility measurements [8]. Therefore, it is a crucial problem to identify the superconducting nature in $CeRu_2$ in order to understand these phenomena.

From specific heat measurements, Seneri et al. concluded that an axial superconducting state developed in $CeRu_2$, suggesting p-wave pairing [9]. On the contrary Huxley et al. [4] reported that the superconducting state can be interpreted by the strong coupling BCS model. The experimental results have not been settled yet. It is well known that the spin-lattice relaxation rate ($1/T_1$) gives decisive information about the superconducting gap, e. g. from the T-dependence of $1/T_1$, one can estimate the magnitude of the superconducting gap.

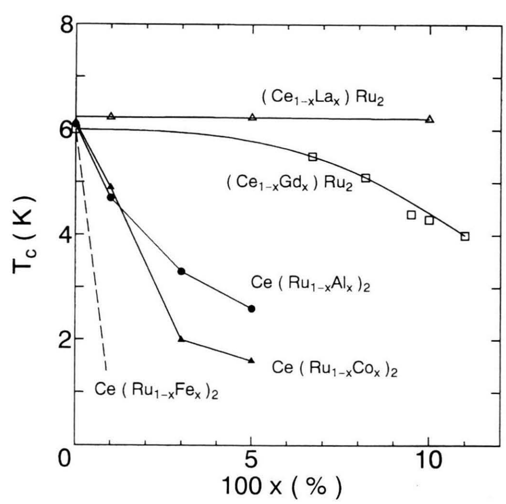

Fig. 1. Variation of T_c in $CeRu_2$ doped with various impurities.

Figure 1 shows the concentration dependence of T_c in $CeRu_2$ doped with various impurities. If the impurities are doped into the Ce site, T_c is not affected at low concentrations; on the other hand, if the impurities are doped in the Ru site, T_c decreases sharply. Therefore it is considered that the superconductivity

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occurs mainly in the Ru site. In order to investigate the electronic state of the Ru site and the superconducting property, we have carried out Ru NMR and NQR studies in $CeRu_2$.

In general, the Ru nucleus is difficult to detect since it has a low natural abandance of the isotopes for NMR, having a small gyromagnetic ratio, γ_n. Recently Burgstaller et al. [10] reported that they succeeded in detecting Ru NMR signal in hcp Ru metal. Their success gave us an impulse to try to detect Ru NMR signals in $CeRu_2$.

1. Experimental

The sample was prepared by arc melting the high-purity elements in a pure Ar atmosphere, followed by long-time anneals. T_c of this sample was determined by the ac susceptibility measurement: $T_c = 6.1$ K. For NMR measurement, the sample was crushed into powder with smaller than 35 μm diameter. The Ru NMR spectra were obtained by sweeping the external field using a superconducting magnet ($H = 12\,T$ at 4.2 K).

2. Results and Discussion

2.1. ^{99}Ru NMR

The NMR spectrum of ^{99}Ru at 20.1 MHz is shown in Figure 2. Due to the large quadrupole interaction and the random distribution of the direction of the electric quadrupole principle axis with respect to the external field, we obtained the typical powder pattern of an NMR spectrum with spin $I = 5/2$. From the splitting between two first-order satellites we deduced the quadrupole frequency (ν_Q) to be 2.22 MHz. The Knight shift of Ru is dominated by the isotropic component $K_{iso} = +0.70\%$, being T-independent, and the anisotropic component $K_{aniso} \approx +0.12\%$.

In the normal state, the nuclear relaxation rate follows the Korringa relation, $T_1 T = \text{const}$ (~ 4 (sec · K)). The value of $T_1 T K^2$ is almost the same as in the uncorrelated Korringa relation, suggesting that there does not exist a strong spin fluctuation of Ru d-spins.

2.2. ^{101}Ru NQR

By using the nuclear quadrupole moment Q in [10], we estimate ν_Q of ^{101}Ru to be 12.8 MHz. Actually, we succeeded in observing the ^{101}Ru NQR signals at 13.2 MHz for $1/2 \leftrightarrow 3/2$ transitions and 26.4 MHz for $3/2 \leftrightarrow 5/2$ transitions, as shown in Figure 3. Figure 4 shows the T-dependence of $1/T_1$, where T_1 was measured at 13.2 MHz and was determined by the single component. A small hump just below T_c and an exponential decrease in $1/T_1$ were observed. These are characteristic features of an s-wave superconductor, but we can not exclude the possibility of the BW state in p-wave superconductor because we have not

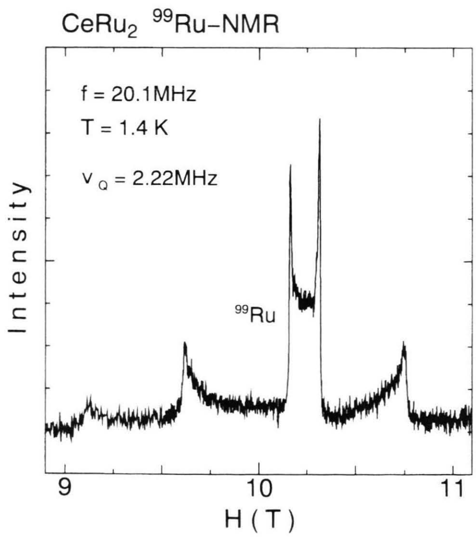

Fig. 2. ^{99}Ru NMR spectrum in $CeRu_2$ recorded at 1.4 K.

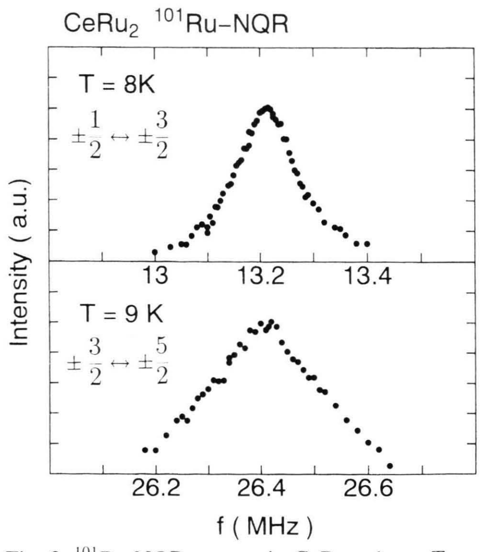

Fig. 3. ^{101}Ru NQR spectra in $CeRu_2$ above T_c.

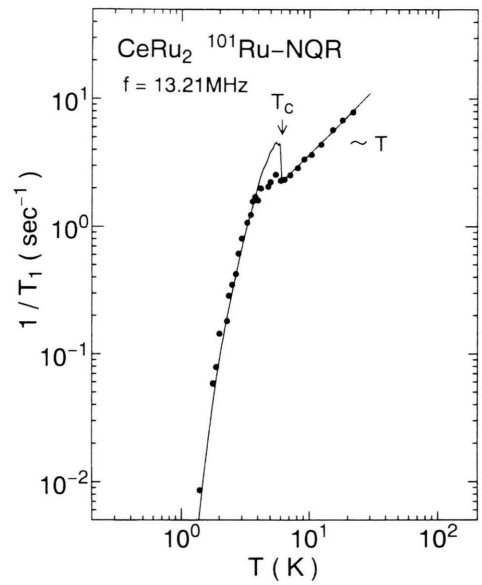

Fig. 4. T-dependence of $^{101}(1/T_1)$ in $CeRu_2$. The solid curve is the calculation with $2\Delta/k_BT_c$ = 3.8 and $\delta/\Delta(0)$ = 1 / 15, (see text).

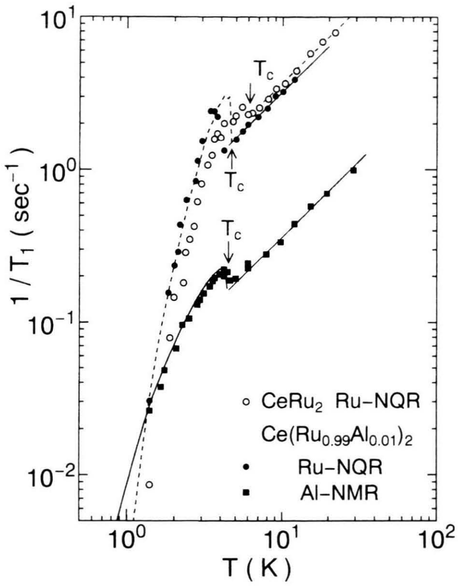

Fig. 5. T-dependence of $^{101}(1/T_1)$ and $^{27}(1/T_1)$ in $Ce(Ru_{0.99}Al_{0.01})_2$. The dotted and solid curves are the calculations with $2\Delta/k_BT_c$ = 3.8 and 2, respectively. $^{101}(1/T_1)$ in $CeRu_2$ is also presented.

yet measured the spin susceptibility in the superconducting state. The behavior of $1/T_1$ was fitted by the calculation done by Hebel and Slichter [11], where the parameters of the superconducting gap and the energy broadening in the gap edge are $2\Delta/k_BT_c \sim 3.8$, and $\delta/\Delta(0) \sim 1/15$. The value of the gap is slightly larger than that in the BCS theory ($2\Delta/k_BT_c = 3.54$). As seen in Fig. 4, $1/T_1$ is largely suppressed at the coherent peak compared with the above calculation. This may be due to the the electron-phonon damping by thermally excited phonons and/or the anisotropy of the energy gap, but the latter is considered to be more effective from the NMR study in Al-doped $CeRu_2$ as discussed later on.

In conclusion, it is evident that $CeRu_2$ is an ordinary s-wave superconductor, with a slightly larger energy gap $2\Delta = 3.8k_BT_c$. Quite recently, Matsuda [12] reported ^{101}Ru NQR measurement in $CeRu_2$ and also concluded that this superconductor is s-wave with $2\Delta = 4.0k_BT_c$. Our experimental result is in good agreement with their result.

2.3. *Al-doped $CeRu_2$*

In spite of non-magnetic impurity, Al-doping strongly reduces T_c as shown in Figure 1. 1% Al-doped $CeRu_2$ ($Ce(Ru_{0.99}Al_{0.01})_2$) was studied by Ru NQR (host site) and Al NMR (impurity site), in order to obtain information of site-selective electronic state. In the host site, the full width at half maximum of the Ru NQR signal in 1%-Al-doping is twice as broad as in non-doping. $^{101}(1/T_1)$ of the Ru site in $Ce(Ru_{0.99}Al_{0.01})_2$ is shown in Figure 5. The value of $1/T_1T$ in $Ce(Ru_{0.99}Al_{0.01})_2$ is somewhat smaller than in non-doping in the normal state. In general, $1/T_1T$ is proportional to the square of the density of states at the Fermi surface ($N(E_F)$), therefore $N(E_F)$ decreases by 1% Al-doping. We assume that the decrease of T_c is due to the reduction of $N(E_F)$, and estimate T_c in 1% Al-doping by using the well-known BCS formula $T_c = 1.13\ \Theta\ \exp(-1/N(E_F)V)$. T_c in 1% Al-doping is evaluated to be about 4.6 K, in good agreement with the experimental value 4.7 K, where we use the Debye-temperature $\Theta \sim 175$K [4]. Just below T_c, the enhancement of $1/T_1$ in 1% Al-doping is distinct compared with that in non-doping. This may be mainly due to the reduction of the anisotropy-induced broadening of the BCS peak in the density of states. The exponential decreasing rate below 0.7 T_c is almost the same as in nondoping, showing that the magnitude of the superconducting gap does not change by 1% Al-doping. These experimental results are considered to be well understood by the theory of the "dirty" superconductor proposed by Anderson [13], that is, the introduction of scattering centers mixes Bloch states with wave vectors of the pure Fermi surface, and the anisotropy in the resultant eigenstate is reduced in the alloy.

We could detect a doped 1% Al NMR signal under the external field of 9 (kOe). The value of $^{27}(1/T_1T)$ in the normal state is 0.035 $(\sec \cdot K)^{-1}$, which is more than one order of magnitude smaller than in Al-metal, suggesting that $N(E_F)$ is small at the Al site. Below T_c, the superconducting behavior in $1/T_1$ was observed even in the doped Al site as shown in Figure 5. The exponential decreasing rate in $^{27}(1/T_1)$ is smaller than in $^{101}(1/T_1)$, suggesting that the superconducting gap at the doped site is smaller than at the host site. The magnitude of the superconducting gap at the doped site is estimated to be $2\Delta/k_BT_c \sim 2$, which is half the value of the gap at the host site.

3. Conclusion

We could detect ^{99}Ru-NMR and ^{101}Ru-NQR signals in superconducting $CeRu_2$ with $T_c = 6.1$K. From the NQR measurement it was revealed that $CeRu_2$ is an ordinary *s*-wave superconductor, having the coherence peak in $1/T_1$ just below T_c and the superconducting gap, $2\Delta/k_BT_c \sim 3.8$. In 1% Al-doped $CeRu_2$, the Al-doping makes the coherence peak distinct but does not change the magnitude of the superconducting gap. The behavior of $1/T_1$ in the Al-doping is well understood in the frame of the "dirty" superconductor. The Al-doping reduces the density of states at the host site, resulting in a decrease of T_c. The superconducting behavior was observed in the doped Al site, however the magnitude of the gap decreases to half the value of the superconducting gap at the host site.

NMR studies in the irreversible region near H_{c2} are now in progress.

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