Abstract. We have performed $^{27}$Al-NQR measurements in CeOs$_2$Al$_{10}$ which exhibits a novel phase transition at $T_0=29$ K. The NQR parameters determined for all the Al sites in ambient pressure were compared with those in CeRu$_2$Al$_{10}$ with $T_0=27$ K and CeFe$_2$Al$_{10}$ with no phase transition. The distinct NQR splitting just below $T_0=32.5$ K under pressure 0.66 GPa ensures an enhancement of $T_0$ and a homogeneous transition. Despite the increase of $T_0$, the nuclear spin-lattice relaxation rate $1/T_1$ is suppressed over whole range of temperature than in ambient pressure. The characteristic features of no critical slowing down at $T_0$ and of the remarkable decrease of $1/T_1T$ starting at $T > T_0$ become prominent under pressure, suggesting an approach to Kondo semiconductor in a valence fluctuation regime.

1. Introduction

CeOs$_2$Al$_{10}$ exhibits a transition to the novel phase at $T_0=29$ K. The transition temperature is extraordinary high to ascribe it to the RKKY interaction because of the long Ce-Ce distance of $\sim 5.2$ Å. In the early stage of the study, the transition is ascribed to the nonmagnetic origins; CDW[1], the stuructural transition to lower symmetric phase[2], and a spin singlet formation[3, 4]. However recent neutron scattering (NS) confirmed the existence of the long range antiferromagnetic (AF) order below $T_0$ in both CeRu$_2$Al$_{10}$[5, 6, 7] and CeOs$_2$Al$_{10}$.[7] However it is still under devate why the $T_0$ is so high and, moreover, how to map the novel phase to the Doniach criterion.[8]

In this context, an interesting feature is the scaling behaviors of the physical properties against pressure among the isomorphic compounds, CeRu$_2$Al$_{10}$ with $T_0=27$ K, CeOs$_2$Al$_{10}$ with $T_0=29$ K and CeFe$_2$Al$_{10}$ with no phase transition.[1] In applying pressure, $T_0$ increases initially and decreases abruptly at about 2 GPa for CeOs$_2$Al$_{10}$ and 4 GPa for CeRu$_2$Al$_{10}$ as like the first order transition in the pressure-temperature ($p-T$) plane.[1] The electronic state of CeOs$_2$Al$_{10}$ in ambient pressure corresponds to that of CeRu$_2$Al$_{10}$ under 2 GPa.

We focus on Al-NQR in CeOs$_2$Al$_{10}$ in the present report. A part of Al-NQR measurements have been reported previously.[9] We will report whole the NQR spectra including all the Al sites in ambient pressure. Moreover NQR measurements are newly performed under pressure. We focus on the NQR splitting just below $T_0$ and the $T$-dependence of $1/T_1$ under pressure, comparing with the previous results in ambient pressure and those in CeRu$_2$Al$_{10}$ as well.

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$^{27}$Al-NQR Study on Novel Phase Transition in CeOs$_2$Al$_{10}$

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Figure 1. The Al-NQR spectrum in CeOs$_2$Al$_{10}$ at 77 K.

Table 1. NQR parameters in CeOs$_2$Al$_{10}$.

| site  | Al(1) | Al(2) | Al(3) | Al(4) | Al(5) |
|-------|-------|-------|-------|-------|-------|
| $\nu_Q$ (MHz) | 1.80  | -     | 1.45  | 1.82  | 2.61  |
| $\eta$     | $\sim$0.56 | -     | $\sim$0 | $\sim$0 | $\sim$0 |

2. Experiments
The single crystal CeOs$_2$Al$_{10}$ was crushed into powder below 53 $\mu$m in the present measurement. The preparation of the single crystal had been reported already.\[1\] The NQR measurements were carried out using phase coherent spin echo method. The so called $\nu^2$ correction has been made for spin echo intensity at respective frequencies $\nu$. The inversion-recovery method for the nuclear magnetization was used to evaluate $1/T_1$ for Al(5)$_{HF}$ NQR peak, where the recovery function of the nuclear magnetization is written as, $p(t) = 1 - m(t)/m(\infty) = a(0.427e^{-3t/T_1} + 0.573e^{-10t/T_1})$ with the fitting parameter $a$.\[10\] The piston-cylinder cell was used in the NQR measurement under pressure. The pressure media is the mixture (1:1) of fluorinert FC-77 and FC-70. The pressure was determined by the superconducting transition temperature of Pb metal.

3. Experimental Results and Discussions
Figure 1 shows the NQR spectrum at 77 K. The five inequivalent Al sites is numbered as Al(i) (i=1 $\sim$ 5) and the two NQR lines at high and low frequencies for each site are labeled as Al(i)$_{HF}$ and Al(i)$_{LF}$, respectively, which are the same as previous report.\[9\] Even including the new NQR lines Al(3)$_{LF}$ and Al(4)$_{LF}$, the previous assignment is supported. The resonances associated with Al(2) site is out of our observation. The NQR parameters, the quadrupole frequency $\nu_Q$ and the asymmetry parameter $\eta$, are listed in Table I. The value $\eta$ is almost the same as those in CeRu$_2$Al$_{10}$\[2\] and CeFe$_2$Al$_{10}$\[12\], confirming the common local symmetry. The magnitude of $\nu_Q$ in CeOs$_2$Al$_{10}$ is largest among the isomorphic compounds. The NQR parameters in CeRu$_2$Al$_{10}$ are substantiated quantitatively by the band structure calculation.\[2\] The band calculations also for CeOs$_2$Al$_{10}$ and CeFe$_2$Al$_{10}$ are desired.

The NQR splitting near neighbor to $T_0$ for Al(4)$_{HF}$ under 0.66 GPa is shown in Fig. 2. The qualitative feature of the NQR splittings due to magnetic order in ambient pressure is the same as those in CeRu$_2$Al$_{10}$\[2\] except for the slight quantitative difference (not shown). As shown in Fig. 2, the splitting below 32 K and no splitting above 33 K indicates $T_0 \sim 32.5$ K under 0.66 GPa, which means the increase of $T_0$ from 28.6 K in ambient pressure. No residual
resonances around 3.66 MHz which is the peak position at $T > T_0$ imply the homogeneous bulk transition, excluding the possibility of the co-existence of the ordered and disordered regions. Thus the smaller entropy decrease $\Delta S (\sim 0.3R \ln 2)$[11] than in CeRu$_2$Al$_{10}$ ($\sim 0.7R \ln 2$)[1] at the transitions is not a volume fraction effect but an essential bulk feature, where $R$ is the gas constant.

Figure 3(a) shows the $T$-dependence of $1/T_1$ under ambient and 0.66 Gpa. The arrows indicate $T_0$ for respective cases. As already pointed out previously, $1/T_1$ does not exhibit even prominent hump not to mention critical divergence, and the remarkable decrease seems to start above $T_0$. This characteristic behavior becomes more prominent under pressure 0.66 GPa. No anomaly of $1/T_1$ at $T_0$ is not due to an inhomogeneity effect, but is essential one as mentioned before.

In general, $1/T_1$ is written as, $1/T_1 \propto \sum_q A(q)^2 \chi^2(q, \omega_0)/\omega_0$, where $A(q)$ is the Fourier $q$
component of the hyperfine interaction between Al nuclear spin and the electron spins relating to the relaxation mechanism, $\chi'(q, \omega)$ the imaginary part of dynamical susceptibility at the $q$ mode and NQR frequency $\omega_0$, $\Gamma_q$ the characteristic fluctuation frequency of the $q$ mode in electron system. Assuming Lorentz type $\chi(q, \omega) = \chi(q)/(1 - i\omega/\Gamma_q)$, and fast fluctuation regime, $\Gamma_q \gg \omega_0$, $1/T_1 \propto \sum_q A(q)^2 \chi(q)/\Gamma_q$.

When the temperature approaches to $T_0$ from above, $1/T_1$ diverges because the staggered susceptibility $\chi(Q)$ with the AF wave vector $Q$ diverges, and also $\Gamma_q$ slows down with growing up of the correlation length of the $Q$ mode. The divergence is eventually ceased if $A(Q)$ is zero geometrically. However these are not zero for all the Al sites in CeT$\text{Al}_10$ ($T=$Ru, Os and Fe) for the magnetic structure proposed by NS. 1/T$_1$ should exhibit critical slowing down more or less if the transition is cooperative 2nd order AF one. The AF order in CeRu$\text{Al}_10$ or CeOs$\text{Al}_10$ might not be primary to the phase transition at $T_0$ but induced one by some hidden ordering accompanying by no collective mode of $f$ electron spins.

Figure 3(b) shows the T-dependence of $1/T_1T$ together with our previous results of CeRu$_2$Al$_{10}$[2] and CeFe$_2$Al$_{10}$.[12] We have interpreted T-dependence of $1/T_1T$ in CeRu$_2$Al$_{10}$ in terms of dense Kondo scenario; localized moment of Ce at higher temperature, coherent state below $T^* \sim 60$ K and after that the novel phase transition occurs at $T_0$.[2] On the other hand, Kondo semiconductor with narrow hybridized band has been applied to CeFe$_2$Al$_{10}$.[12] The T-dependence of $T_1T$ above $T_0$ in Fig 3(b) represents that CeOs$_2$Al$_{10}$ is in the intermediate state between CeRu$_2$Al$_{10}$ and CeFe$_2$Al$_{10}$. Then the temperature $T^*$ might be assigned to higher one than room temperature in CeOs$_2$Al$_{10}$, and CeFe$_2$Al$_{10}$ as well.

The gap magnitude below $T_0$ is nearly the same among CeRu$_2$Al$_{10}$, CeOs$_2$Al$_{10}$ in ambient pressure and in 0.66GPa as easily seen in Fig. 3(b). However the T-dependence in the low temperatures $T \ll T_0$ in CeOs$_2$Al$_{10}$ is different from those both in CeRu$_2$Al$_{10}$ and CeFe$_2$Al$_{10}$. The latters follow Korringa law ($T_1T=\text{const.}$), suggesting a pseudo gap with the density of coherent state at the Fermi energy $\epsilon_F$. On the other hand, $1/T_1 \sim \text{const.}$ in CeOs$_2$Al$_{10}$ might be ascribed to the relaxation process through incoherent states in the gap, hence suggest a full gap opening at $\epsilon_F$. These difference with respect to the density of states at $\epsilon_F$ is consistent with the specific heat measurements.[1]

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