Elastic properties of Re-Ru_4Sb_{12} (Re; La, Pr)

T. Kumagai [a], Y. Nakanishi [a,1], H. Sugawara [b], H. Sato [b], and M. Yoshizawa [a]

[a] *Department of Materials Science and Engineering, Iwate University, Morioka 020-8551, Japan*

[b] *Department of Physics, Tokyo Metropolitan University, Hachioji 192-0397, Japan*

Abstract

We have studied elastic properties of the filled skutterudite compounds Re-Ru_4Sb_{12} (Re=La and Pr) by means of ultrasonic measurement. They both undergo a superconducting transition at temperature of T_c=3.2 K for La Ru_4Sb_{12} and Tc=1.0 K for Pr Ru_4Sb_{12}. No distinct anomaly was observed at T_c in both compounds. In Pr Ru_4Sb_{12}, the determination of $4f$ ground state was desired whether to be Γ_1 or Γ_3. The obtained result exhibit no softening towards low temperature in both $(C_{11}$-$C_{12})/2$ and C_{44}, indicating the $4f$ ground state of Pr ion to be Γ_1 singlet.

Key words: ultrasonic measurement; crystalline electric field effect; Re-Ru_4Sb_{12} ; singlet ground state

The Re- Ru_4Sb_{12} compounds (Re=rare earth) with the filled skutterudite crystal structure have attracted much attention since the discovery of non-Fermi liquid behavior in $CeRu_4Sb_{12}$ at low temperatures[1-3]. In these system, the band structure and $4f$ ground state affect pretty sensitively on their physical properties. It is because the hybridization between conduction band and $4f$ state is effectively much large due to its unique crystal structure. In order to elucidate the physical interpretation of observed unusual phenomena, systematic investigations such as substitution by La impurity, replacing the kind of rare earth element are inevitably necessary. From this point of view, $LaRu_4Sb_{12}$ and Pr Ru_4Sb_{12} were selected as the first compounds for investigation. In Re- Ru_4Sb_{12} groups $LaRu_4Sb_{12}$ and Pr Ru_4Sb_{12} undergo a superconducting transition at temperature of T_c=3.2 K and T_c=1.0 K, respectively[3]. In Pr Ru_4Sb_{12}, in particular, the determination of $4f$ ground state of Pr ion split by crystalline electric field (CEF) effect has not been clarified. However, it was selected whether to be Γ_1 singlet orΓ_3 non Kramers doublet by the magnetic susceptibility measurement[3]. Ultrasonic measurement is powerful method to determine the $4f$ ground state of rare-earth ion. An elastic softening in $(C_{11}$-$C_{12})/2$and/or C_{11} towards low temperature is expected in the case of Γ_3 non Kramers doublet, on the other hand no softening in all elastic constants is expected in the case of Γ_1 singlet ground state[4]. Thus, ultrasonic measurement was eager. In this paper, we report the elastic property of both $LaRu_4Sb_{12}$ and $PrRu_4Sb_{12}$. Especially in Pr Ru_4Sb_{12}, the $4f$ ground state of Pr ion and the possible level scheme is discussed by comparing previous $4f$-level scheme determined by other experimental measurement.

The single crystals of $LaRu_4Sb_{12}$ and Pr Ru_4Sb_{12} were grown by the Sb-flux method. The samples with rectangular shape of 2.2 2.3 2.8 mm^3 for La Ru_4Sb_{12}, and of 2.2 2.3 2.8 mm^3 for Pr Ru_4Sb_{12} were prepared for the ultrasonic measurement. In order to observe the ultrasonic pulse echoes with an exponential decay, we polished carefully the surfaces of sample to be parallel. The $LiNbO_3$ transducers for the generation and detection of the sound waves with frequencies 5'10 MHz were bonded on the surfaces of the sample by an elastic polymer Thiokol. The sound-wave velocity v was detected by an ultrasonic apparatus based on the phase-comparison method. In the estimation of the elastic constant $C = \rho v^2$, we used the mass density $\rho = 8.338$ g/cm^3 and 8.367 g/cm^3 from the lattice constant a = 9.277 (Å) and 9.270 (Å) of

[1] Corresponding author. E-mail: yoshiki@iwate-u.ac.jp

the present sample of La Ru_4Sb_{12} and Pr Ru_4Sb_{12}, respectively. Figure1 shows the temperature dependence of elastic constants C_{11}, $(C_{11}$-$C_{12})/2$ and C_{44} for $LaRu_4Sb_{12}$. All measured elastic constants increase monotonically with decreasing temperatures. No distinct anomaly was observed at $T_c = 3.2$ K in all measures elastic constants. Figure 2 shows the temperature dependence of elastic constants C_{11}, $(C_{11}$-$C_{12})/2$ and C_{44} for $PrRu_4Sb_{12}$. All measured elastic constants increase monotonically with decreasing temperatures. The absolute values of each elastic constants and calculated bulk modulus C_B=$(C_{11}$+$C_{12})/3$ and Poisson ratio ν=$C_{12}/(C_{11}$+$C_{12})$ from C_{11} and $(C_{11}$-$C_{12})/2$ for $LaRu_4Sb_{12}$ and $PrRu_4Sb_{12}$ at 77 K@are listed in Table I

Table 1
The absolute values of each elastic constants (GPa) and estimated bulk modulus C_B (GPa), Poisson ratio ν of $PrFe_4P_{12}$, $NdFe_4P_{12}$ and $CeRu_4Sb_{12}$ at 77 K.

	a (Å)	C_{11} (GPa)	$(C_{11}-C_{12})/2$ (GPa)	C_{44} (GPa)	C_B (GPa)	ν
$LaRu_4P_{12}$	9.2774	167.6	34.5	15.2	121.6	0.37
$PrRu_4P_{12}$	9.2701	127.5	30.2	14.2	80.5	0.33

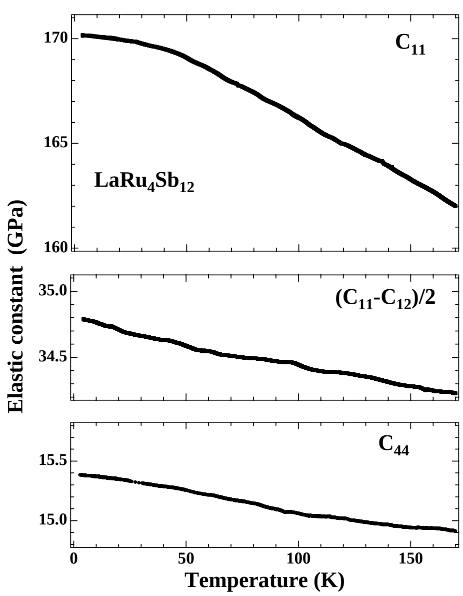

Fig. 1. Temperature dependence of elastic constants C_{11}, $(C_{11}$-$C_{12})/2$ and C_{44} for $LaRu_4Sb_{12}$. Inset shows the low-temperature behavior of them on an expanded scale. Arrows indicate a superconducting transition temperature.

The obtained results of Pr Ru_4Sb_{12} provide us the following suggestions. The experimental fact; no softening towards low temperatures was observed, indicates that the ground state of $4f$-multiplet is Γ_1 singlet, not Γ_3 non-Kramers doublet. If it is latter, an

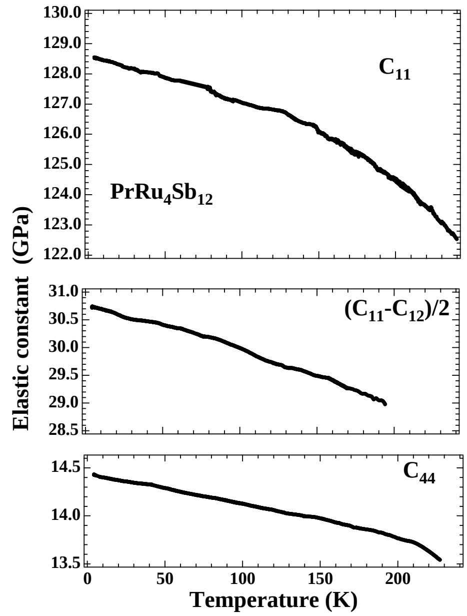

Fig. 2. Temperature dependence of elastic constants C_{11}, $(C_{11}$-$C_{12})/2$ and C_{44} for $PrRu_4Sb_{12}$.

elastic softening is expected in $(C_{11}$-$C_{12})/2$. Present result strongly supports Γ_1 singlet as ground state in Pr Ru_4Sb_{12} which was proposed by Takeda *et al* [3]. However, no information gives us concerning the high-lying $4f$ state in present study. The superconducting transition hardly influences the elastic constants in La Ru_4Sb_{12}. This result was also found in La Fe_4P_{12}.[5] This might be a common behavior in these material groups.

As a summary, we reported the elastic properties of the filled skutterudite compounds $LaRu_4Sb_{12}$ and $PrRu_4Sb_{12}$ by means of ultrasonic measurement. From present results we obtain the following conclusions. For $LaRu_4Sb_{12}$, all measured elastic constants increase monotonically with decreasing temperatures. No distinct anomaly was observed at T_c in $LaRu_4Sb_{12}$. For $PrRu_4Sb_{12}$, all measured elastic constants increase monotonically with decreasing temperatures. No elastic softening towards low temperatures was observed, indicating that the ground state of $4f$-multiplet is Γ_1 singlet, not Γ_3 non-Kramers doublet. The lower temperature condition is needed to investigate the elastic properties around T_c =1.0 K in $PrRu_4Sb_{12}$.

This study was financially supported in part by the Grant-in-Aid for Scientific Research from the Ministry of Education, Sport, Science and Culture of Japan.

References

[1] N. Takeda *et al.*; Physica B **259-261** (1999) 92.

[2] N. Takeda *et al.*; Physica B **281-282** (2000) 388.

[3] N. Takeda *et al.*; J. Phys. Soc. Jpn. **69** (2000) 868.

[4] Y. Nakanishi *et al.*; J. Phys. Soc. Jpn. (suppl.) **69** (2001) 868.

[5] Y. Nakanishi *et al.*; (unpublished)