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Ultrasonic Investigation of Off-Center Oscillator in Clathrate Compounds $R_3Pd_{20}Si_6$ and $R_3Pd_{20}Ge_6$

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Clathrate compounds $R_3Pd_{20}Si_6$ and $R_3Pd_{20}Ge_6$ (R=rare-earths) crystallize in a cubic $Cr_{23}C_6$-type structure with O_5^h space group symmetry. The rare-earths are located on the two crystallograhically non-equivalent 4a- and 8c-sites. Using grown single crystals, we have carried out ultrasonic measurements to investigate rattling and tunneling of a rare-earth ion in a cage consisting of Pd and Si or Ge. The transverse elastic constant C_{44} of $R_3Pd_{20}Ge_6$ (R=La, Ce, Pr, Nd) shows frequency dependence together with considerable ultrasonic attenuation around 10-30 K, while no frequency dependence has been observed in $(C_{11}-C_{12})/2$. The ultrasonic dispersion in C_{44} is well described by a Debye-type formula with a relaxation time $\tau = \tau_0 exp(E/k_BT)$. This is caused by thermally activated rattling of the rare-earth ion over a potential barrier in the cage [1,2]. Furthermore, the C_{44} of $La_3Pd_{20}Ge_6$ without 4f electron shows softening below 3 K down to 20 mK in proportion to 1/T suggesting a triply degenerated tunneling state of the rare-earth ion through the potential barrier at low temperatures [2]. In the present study, we have measured elastic constants to investigate the rattling and tunneling in the $R_3Pd_{20}Si_6$ system. A monotonous increase of all elastic constants of $La_3Pd_{20}Si_6$ in decreasing temperature has been found. Fig. 1 shows the temperture dependence of elastic constants in $Ce_3Pd_{20}Si_6$. C_{11}, $(C_{11}-C_{12})/2$ and C_{44} exhibit softening with decreasing temperature below 60 K, which is originated in quadrupoablar effect of 4f-electron.The absence of the ultrasonic dispersion in Si clathrates is in contrast to the case of compound $R_3Pd_{20}Ge_6$. As shown in the inset of Fig. 1, $(C_{11}-C_{12})/2$ shows a minimum around 0.15K indicates appearance of a Kondo signlet state. The result of $Ce_3Pd_{20}Si_6$ suggests strongly correlated phenomena of heavy fermion state due to a Γ_8 ground state.

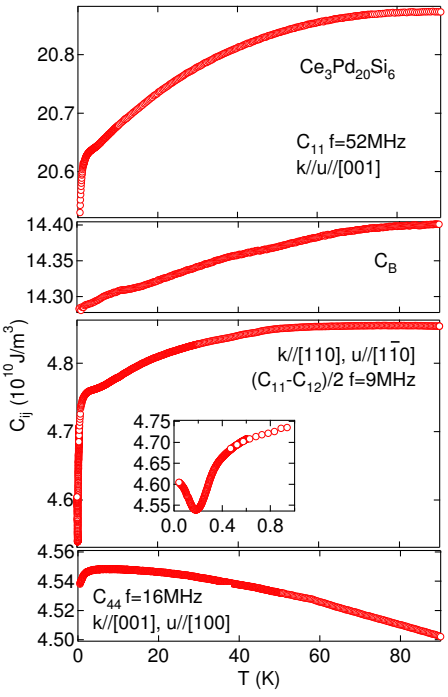

Figure 1: The temperature dependence of the elastic constant in $Ce_3Pd_{20}Si_6$

[1] Y. Nemoto et al., Phys. Rev. B **68** (2003) 184109.
[2] T. Goto et al., Phys. Rev. B **70** (2004) 184126.