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Ultrasonic measurements of the off-center mode in a clathrate compound $La_3Pd_{20}Ge_6$

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The ternary compound $La_3Pd_{20}Ge_6$ has a cagelike structure. La ion in a cage vibrates between off-center positions. We have investigated two types of off-center motions 'rattling' and 'tunneling' in $La_3Pd_{20}Ge_6$ by ultrasonic measurement. The rattling is a thermally activated motion over the potential hill and the tunneling is a quantum mechanical motion through the potential hill. The elastic constant C_{44} of $La_3Pd_{20}Ge_6$ shows a Debye-type dispersion around 20 K. Similar ultrasonic dispersions was found in the C_{44} of $Ce_3Pd_{20}Ge_6$ [1] and in the $(C_{11}-C_{12})/2$ of a filled skutterudite $PrOs_4Sb_{12}$ [2]. Ultrasonic dispersion of $La_3Pd_{20}Ge_6$ is caused by a Γ_5 rattling motion of La ion between off-center positions along the threefold [111] axis in 4a-site cage. The relaxation time shows an activation-type temperature dependence $\tau = \tau_0 \exp(E/k_B T)$ with an attempt time $\tau_0 = 2.0 \times 10^{-12}$ sec and an activation energy $E = 197$ K. Besides, C_{44} shows a remarkable softening with $C_{44} = C_{44}^0(T - T_C^0)/(T - \Theta)$ below 3 K. The parameters are obteined to be $T_C^0 = -337.970$ mK, $\Theta = -338.044$ mK and $C_{44}^0 = 3.33085 \times 10^{10}$ J/m^3. This softening is probably due to the off-center tunneling motion with Γ_5 symmetry of La ion at 4a-site cage. The fact that a coupling constant of inter-cage interaction has a negative value Θ reveals that antiferro-type interaction exists between the tunneling state of 4a-site cage. The off-center tunneling mode is a new type of quantum degrees of freedom, which may bring about quantum phenomena of phase transition breaking the symmetry of ground state or unconventional superconductivity.

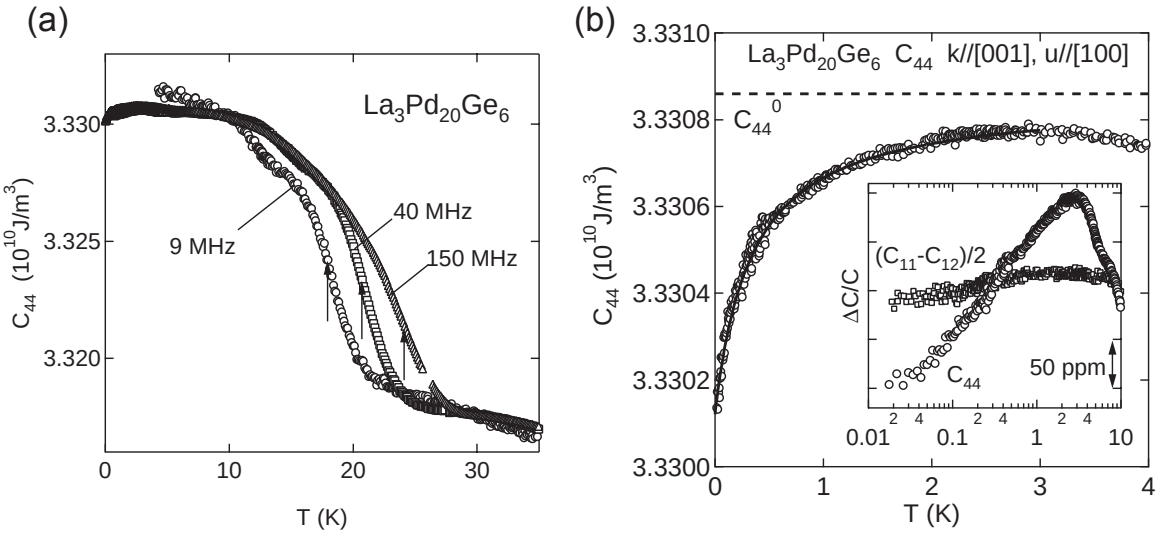

Figure 1: (a) Ultrasonic dispersion in $La_3Pd_{20}Ge_6$. (b) Softening of C_{44} at low temperature.

[1] Y. Nemoto, T. Yamaguchi, T. Horino, M. Akatsu, T. Yanagisawa, T. Goto, O. Suzuki, A. Dönni, and T. Komatsubara, Phys. Rev. B **68** (2003) 184109.
[2] T. Goto, Y. Nemoto, K. Sakai, T. Yamaguchi, M. Akatsu, T. Yanagisawa, H. Hazama, K. Onuki, H. Sugawara, and H. Sato, to be submitted Phys. Rev. Lett.