

Ultrasonic measurements of off-center oscillations in clathrate crystals $R_3Pd_{20}Ge_6$

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The ternary rare-earth compounds $R_3Pd_{20}Ge_6$ with the cubic C_6Cr_{23} structure with space group symmetry $Fm\bar{3}m$ have cages containing rare-earth ion. Guest atom in a cage vibrates between off-center positions. We have investigated two types of off-center oscillations 'rattling' and 'tunneling' in $La_3Pd_{20}Ge_6$ by ultrasonic measurement. The rattling means a thermally activated motion over the potential hill and the tunneling means a quantum mechanical motion through the potential hill. The transverse elastic constant C_{44} of $R_3Pd_{20}Ge_6$ ($R = La, Ce$ [1] and Pr) shows a Debye-type dispersion around $10 \sim 30$ K. Similar ultrasonic dispersions was found in the $(C_{11} - C_{12})/2$ of a filled skutterudite $PrOs_4Sb_{12}$ [2]. Ultrasonic dispersion of $R_3Pd_{20}Ge_6$ is due to a Γ_5 rattling of guest atom in 4a-site cage between off-center positions along the threefold $[111]$ axis. The relaxation time shows an activation-type temperature dependence $\tau = \tau_0 \exp(E/k_B T)$. Furthermore, C_{44} of $La_3Pd_{20}Ge_6$ being absent from 4f-electron shows a remarkable softening proportional to $1/T$ below 3 K down to 20 mK. This softening is probably caused by the off-center tunneling with Γ_5 symmetry of La ion at 4a-site cage. The off-center tunneling is a new type of quantum degrees of freedom, which may bring about exotic low-temperature properties.

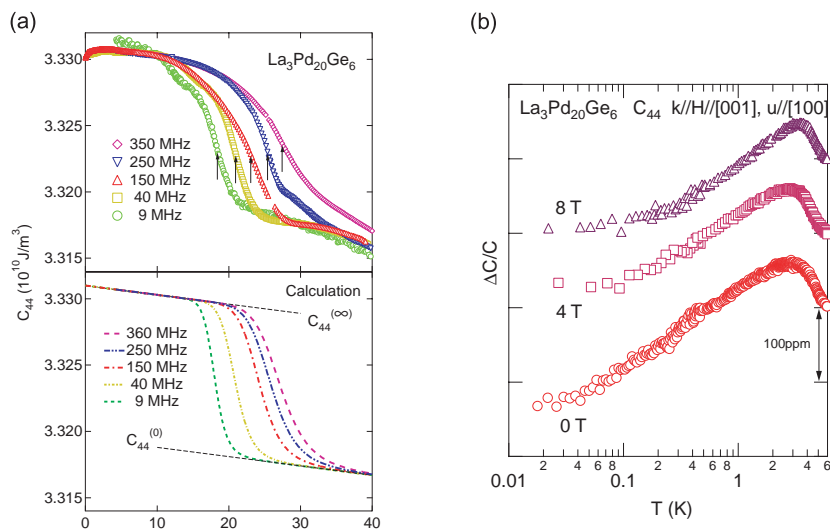


Figure 1: (a)Ultrasonic dispersion of C_{44} in $La_3Pd_{20}Ge_6$. (b)Elastic softening of C_{44} on $La_3Pd_{20}Ge_6$ in magnetic fields.

[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, Phys. Rev. B **69** (2004) 180511.