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Off-center Rattling and Tunneling in Heavy-fermion Superconductor $PrOs_4Sb_{12}$

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We have performed ultrasonic experiments in order to investigate rattling, which is thermally activated off-center motion of a guest atom in a cage, as can been seen in a filled skutterudite $PrOs_4Sb_{12}$ [1] and other clathrate compounds $R_3Pd_{20}Ge_6$ (R=La, Ce, Pr, Nd) [2,3]. Remarkable frequency dependences of the elastic constants, so-called ultrasonic dispersion, appear around 10-50 K in the range of frequencies of ultrasonic sound waves 10-400 MHz. With decreasing temperature, thermally activated rattling over an anharmonic potential hill diminishes rapidly. At low temperatures, quantum tunneling though the potential hill is reasonably expected. Actually, elastic softening proportional to the reciprocal temperature $1/T$ was observed in the elastic constants C_{44} and $(C_{11}-C_{12})/2$ in $La_3Pd_{20}Ge_6$. The C_{44} shows dominant softening below 3 K, which indicates that the ground state of the quantum tunneling is a Γ_5 triplet [3]. We note that the charge fluctuation due to tunneling remains even at 20 mK in $La_3Pd_{20}Ge_6$.

In $PrOs_4Sb_{12}$, the C_{11}, $(C_{11}-C_{12})/2$ and C_{44} show softening with decreasing temperature below 20 K well recognized by a conventional quadrupole susceptibility associated with a pseudo quartet $\Gamma_1-\Gamma_4^{(2)}$ [4,5]. In addition, the softening of $(C_{11}-C_{12})/2$ continues below 3 K which is never reproduced by a singlet Γ_1 ground and an excited $\Gamma_4^{(2)}$ triplet located at 8 K. Consequently, we proposed that the doubly degenerated tunneling state with Γ_{23} symmetry becomes apparent at low temperatures in $PrOs_4Sb_{12}$. Further notable feature is that the softening of $(C_{11}-C_{12})/2$ stops at the superconducting transition point $T_C = 1.85$ K. These results suggest quadrupolar symmetry breaks in the heavy-fermion superconducting state in $PrOs_4Sb_{12}$.

Recent extensive study on the rattling and tunneling, which is easily couples to conduction electrons, may lead to the opening of basic issues such as charge fluctuation mediated Kondo effect, heavy fermion and superconductivity.

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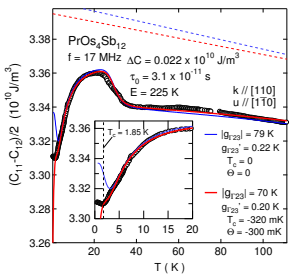

Figure 1: Temperature dependence of the elastic constant of $(C_{11}-C_{12})/2$ in $PrOs_4Sb_{12}$.