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Rattling and tunneling in clathrate compounds

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Recently we have found rattling and tunnelling motions of an off-center guest atom in oversized cage of clathrate crystals.[1-4] The elastic constant C_{44} and attenuation coefficient α_{44} of clathrate crystals $R_3Pd_{20}Ge_6$ with R= La, Ce, Pr, Nd exhibit a Debye-type dispersion around 20 K obeying the relaxation time $\tau = \tau_0 \exp(E/k_BT)$ with an attempt time τ_0 and an activation energy E. This dispersion indicates the rattling of the off-center rare-earth ion over a potential hill with a height of the activation energy E. It is remarkable that the activation energy of $E = 70$ K for $Ce_3Pd_{20}Ge_6$is smaller than $E = 197$ K of $La_3Pd_{20}Ge_6$. Furthermore a relatively slow attempt time $\tau_0 = 3.1 \times 10^{-11}$ s of $Ce_3Pd_{20}Ge_6$ is contrast to $\tau_0 = 2.0 \times 10^{-12}$ s of $La_3Pd_{20}Ge_6$. The present experiments promise that the conduction electrons being mixed strongly to $4f^1$ state of the cerium compound in particular reduces the activation energy and makes slower the attempt time. This fact indicates that the rattling is assisted by the interaction to the conduction electrons, which mixes to the 4f state. It has also been reported the transverse $(C_{11} - C_{12})/2$ mode that in a heavy Fermion superconductor $PrOs_4Sb_{12}$ exhibits the ultrasonic dispersion due to the rattling of the off- center Pr ion in a Sb-icosahedron.

At low temperatures below 3 K down to 20 mK, the C_{44} of $La_3Pd_{20}Ge_6$ shows a softening of $C_{44} = C^0_{44}(T - T^0_C)/(T - \Theta)$ with $T^0_C = -337.970$ mK and $\Theta = -338.044$ mK. Furthermore the specific heat coefficient C/T of $La_3Pd_{20}Ge_6$ increases considerably with decreasing temperature below 3K down to 100 mK. These facts indicate that the tunnelling through the potential hill in keeping O_h site symmetry in the cage becomes relevant at low temperatures, where the thermally activated rattling over the hill dies out completely. The tunnelling and rattling of the off-center rare-earth ion in the cage accompany the charge fluctuation with T_{2g} symmetry in $R_3Pd_{20}Ge_6$ and the E_g symmetry in $PrOs_4Sb_{12}$. The interaction of the off-center tunnelling to the conduction electrons in $PrOs_4Sb_{12}$ is relevant in particular for the formation of the heavy Fermion state and its superconductivity.

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