Low-Energy Phonon Anomaly and Electron-Lattice Interaction in the Pr-Filled Skutterudites

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The rare-earth filled skutterudites RT₄X₁₂ (R = rare earth, T = transition metal, X = pnictogen) have been studied in terms of the anomalously anharmonic lattice properties, as well as the various strongly correlated electron phenomena involving ⁴f electrons. The filled ions are located within the icosahedral X cage, and vibrate with large amplitude detected as the large effect of the Debye-Waller factors. Ultrasonic measurements of PrOs₄Sb₁₂ revealed dispersion of elastic constants indicative of anomalous Pr-ion motion within the Sb cage, and vibrated with large amplitude detected as the large effect of the Debye-Waller factors. Recent inelastic x-ray scattering experiment of Sm-based compounds (S. Tsutsui et al.: Physica B 383 (2006) 142, JMMM in press) and inelastic neutron scattering experiment of CeRu₄Sb₁₂ (C. H. Lee et al.: JPSJ 75 (2006) 123602) revealed lower-frequency flat phonon branches corresponding to the motion of filled atoms.

We have investigated the low-energy phonon spectra of PrOs₄Sb₁₂ and PrRu₄Sb₁₂ using the triple-axis spectrometers TOPAN (6G) and HER (C1-1).

The left figure shows phonon spectra of PrRu₄Sb₁₂ measured at the reciprocal lattice point Q = (0 3 3). It shows distinct softening of excitation energy by 17% with decreasing temperature from 300 down to 12 K. By the measurements in the Brillouin zone around Q = (0 3 3), this mode was confirmed to be less dispersive like an optical mode. We measured phonon spectra also at Q = (6 ζ ζ), as shown in the right figure. The spectrum measured at ζ = −0.4 is composed of the peak “R” around 4 meV which exhibits softening behavior similar to the less-dispersive optical mode at Q = (0 3 3) and that around 6 meV ascribed to the transverse acoustic (TA) one which has no softening behavior. The temperature dependence of the integrated intensity of the 4 meV peak is explained by the phonon state dominated by the Pr-ion vibration, so that the low-energy optical mode is given by the large Pr-ion motion “rattling” inside the Sb cage. The spectrum at the zone center Q = (6 0 0) is unexpectedly quasielastic, whose tail extends beyond 4 meV. Such overdamped response can be attributed to a relaxation phonon due to additional interaction. We will compare the experimental data with the phenomenological phonon susceptibility based on the mode coupling.
Fig. 1. The upper figure shows energy spectra at $\mathbf{Q} = (0 \ 3 \ 3)$ of PrRu$_4$Sb$_{12}$ as a function of temperature, and the lower one shows those at $\mathbf{Q} = (6 \ z \ z)$ measured at 300 K.