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Raman scattering study of RB_6

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Raman scattering spectra of RB₆, with R=Ca, La, Ce, Pr, Sm, Gd, Dy and Yb, have been systematically investigated. From the observed spectra, we have obtained the clear valence- or carrier-dependence and the importance of electron-phonon interaction for the trivalent crystals. The observed Raman scattering spectra can be classified into the following three different excitations; Low-energy excitations below 200cm^{-1} , Raman-active three phonons (T_{2g}, E_g, A_{1g}) at $700 \sim 1300 \text{cm}^{-1}$, and higher-energy mode at $\sim 1400 \text{cm}^{-1}$. From the energy comparison with the phonon dispersion curves, the low-energy excitations and the higher-energy mode are assigned as the second order Raman spectra of the acoustic branches and T_{2g} mode, respectively, at Brillouin boundary.

For the divalent crystals (CaB₆, YbB₆), the doublet line shape of the T_{2g} and E_g phonons suggests the anisotropic distribution of a few carrier on B₆. For the trivalent crystals, we summarize the important results obtained by the present experiments.

1. Low-energy excitations

The intensity decreases with decreasing temperature and this dependence well correlates with the reported thermal movement of R ions. The energy decreases with the increase of the cage size surrounded by the B_6 octahedra. Thus, the low-energy excitations are the vibrations related to the R ion movement.

2. T_{2g} phonon

The energy of the trivalent crystals is smaller by $\sim 100 \text{cm}^{-1}$ than that of the divalent one. The energy-decrease from the divalent-crystal energy corresponds to the valence of R ion. Furthermore, the asymmetric line shape of T_{2g} for the trivalent crystal is the experimental evidence of the electron-phonon interaction.

3. Higher-energy mode Its intensity is greater by 20 times than that of the divalent one. To explain this remarkable difference, some kind of coupling between electron and T_{2g} branch is necessary.

Finally, we summarize the remaining problems related to the present RB_6 study.

- The determination of electron-phonon interaction by the first principle calculation, especially for the T_{2q} phonon.
- Measurements of the crystal field excitation in the quadrupole order for CeB_6 .

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