

Raman scattering of filled skutterudite – CEF excitation and filling rate –

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In this meeting, we pay attention to the crystal electric field (CEF) excitation for $\text{PrRu}_4\text{P}_{12}$ and the filling effect of Nd ion for $\text{Nd}_x\text{Fe}_4\text{Sb}_{12}$ observed in Raman spectra

1. CEF excitation for $\text{PrRu}_4\text{P}_{12}$

In the Raman spectra of $\text{PrRu}_4\text{P}_{12}$, CEF excitations are observed at every temperature we measured. We have assigned the CEF peaks observed below 250 cm^{-1} by applying magnetic field, and determined the energy level scheme. In the polarization dependence of the CEF spectra, we have observed a spectral asymmetry originated from the Th symmetry, that is, the difference of T_g spectra between (x,y) and (y,x) polarization conditions, where (α,β) denotes polarization directions of incident(α) and scattered(β) light. Figure 1 shows excitation energy dependence of T_g Raman spectra (x,y) and (y,x) . Broad peaks assigned as the CEF excitations below 200 cm^{-1} show a significant spectral difference between (x,y) and (y,x) . In the perturbation theory for Raman scattering, the phenomenon is expected to be observed at a resonance scattering. In fact, the resonance scattering is observed at around $\lambda \simeq 530\text{nm}$ ($\simeq 2.3\text{eV}$), as shown in the Fig. 1.

2. filling effect of Nd ion $\text{Nd}_x\text{Fe}_4\text{Sb}_{12}$

Figure 2 shows the polarization dependence of $\text{Nd}_x\text{Fe}_4\text{Sb}_{12}$ Raman spectra for $x=0.744$ and 0.901 . As shown in the Fig. 2, with decreasing the filling rate x , the phonon energies decrease about 2 cm^{-1} , and a decrease in $4\sim 5\text{ cm}^{-1}$ is especially observed for A_{g1} and E_{g1} . The energy decrease for most phonons suggests the increase of the lattice parameter. The large decrease of A_{g1} and E_{g1} suggests that these modes are sensitive for the rare earth behavior, because A_{g1} and E_{g1} phonons correspond to the stretching motion of pnictogens with the rare earth ion.

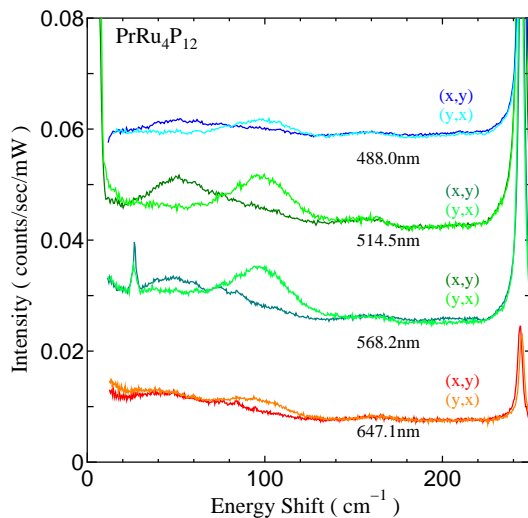


Figure 1: Excitation energy dependence of T_g spectra (x,y) and (y,x) for $\text{PrRu}_4\text{P}_{12}$ measured at $T=300\text{K}$.

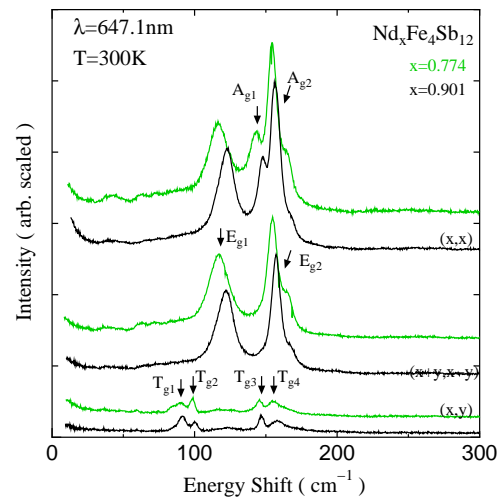


Figure 2: Polarization dependence of $\text{Nd}_x\text{Fe}_4\text{Sb}_{12}$ for $x=0.774$ and 0.901 measured at $T=300\text{K}$.