Neutron scattering study of phonon dynamics on type-1 clathrate cage compounds (Ba,Sr)8Ga16Ge30

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Large vibration of an atom in an oversized atomic cage, so called rattling, has attracted a great interest since it can be the origin of some exotic physical properties. For example, the rattling can be responsible for an extremely low thermal conductivity. Electronic properties could also be affected via electron-phonon coupling. To clarify the relation between the rattling motion and exotic physical properties in materials having oversize atomic cages, it is very important to understand the nature of the rattling.

(Ba,Sr)8Ga16Ge30 is one of compounds that has large Ga and Ge atomic cages filled with Ba or Sr guest atoms. In the compounds, the guest atoms are considered to behave as rattlers in the oversized atomic cages. To clarify the vibrational motion of filling atoms, we have conducted neutron scattering measurements on (Ba,Sr)8Ga16Ge30 compounds using the 3-axis spectrometers, GPTAS, TOPAN, HQR and AKANE, at JRR-3M reactor of Japan Atomic Energy Agency in Tokai. Sample vomules of Ba8Ga16Ge30 and Sr8Ga16Ge30 single crystals used for the measurements were about 1cc and 2cc, respectively.

Fig. 1 shows the phonon dispersion of Ba8Ga16Ge30 with propagation vector of [100]. The optical phonon mode observed at E = 4.5 meV corresponds to a guest mode, in which Ba atoms vibrate largely. The guest mode shows anti-crossing behavior with acoustic phonon mode around q=(0.5,0,0). The low phonon energy indicates that Ba atoms bound loosely with surrounding atoms consistent with the previous expectation. On the other hand, the anti-crossing behavior indicates that the guest mode mixes strongly with the acoustic mode. Although the energy dependence is weak, the guest mode is still dispersive suggesting the existence of Ba-Ba interaction. In Sr8Ga16Ge30, on the other hand, the energy dependence of the guest mode is weaker and energy gap between the guest and the acoustic mode is smaller than in Ba8Ga16Ge30. It seems that the guest mode in Sr8Ga16Ge30 becomes more Einstein-like mode than in Ba8Ga16Ge30. In Sr8Ga16Ge30, the guest atoms tend to split from the center position of atomic cages, whereas in Ba8Ga16Ge30 they are located on the center. The results show that the guest mode is more Einstein-like in a sample where the atomic splitting is larger.

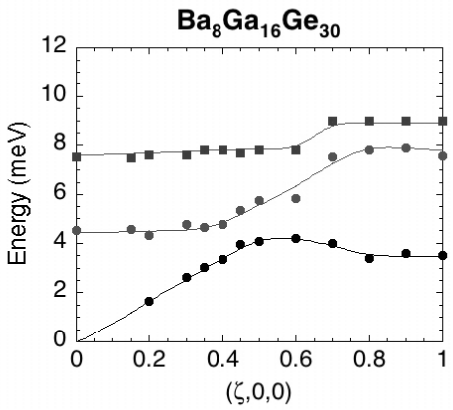

Fig. 1. Phonon dispersion curves of transverse phonon modes with propagation vector [100] in Ba8Ga16Ge30.