Strong renormalization of Ba vibrations in thermoelectric type IX clathrate Ba24Ge100

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Résumé

We report a combined experimental and theoretical study of the lattice dynamics of the type IX clathrate Ba24Ge100 by inelastic neutron scattering (INS) experiments and Density Functional Theory (DFT) calculations. We observe low-energy optical modes at about 2-3 meV due to the motion of the heavy Ba atoms along the high symmetry axis and the largest dimension of the open Ge@20 cages present in the compound. Even though the phonon participation ratio indicates that these low-energy modes are localized, their Q dependence shows that the dynamics of the Ba guests are correlated. We observe a strong change in the spectral weight of these modes when the compound undergoes a temperature-induced structural transformation in the temperature range 190-230 K. In the high-temperature phase the low-energy optical modes show high intensities in the INS data and frequencies rather insensitive to temperature changes up to about 550 K. In the low-energy structural modification the mode intensities are strongly depleted and apparently shifted to higher energies a behavior in line with an off-centering of the Ba atoms at low temperatures. Our DFT calculations successfully approximate the essential features in the dynamics of the high-temperature Ba24Ge100 structure.

Mots-Clés: Clathrate, phonons, neutrons, DFT

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