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First Principles Study of the Vibrational and Thermal Properties of the Type-II Clathrates $A_xGa_xSn_{136-x}$(x = 8,16,24; A = Rb,Cs)
DONG XUE, CHARLES MYLES, Texas Tech University — We have performed first-principles calculations of the vibrational and thermal properties of the semiconductor clathrates $Rb_xGa_xSn_{136-x}$ and $Cs_xGa_xSn_{136-x}$ for x = 8, 16, and 24. Our calculations used the VASP code to obtain the equilibrium geometries and the PHONOPY code to obtain the harmonic phonon modes. For x = 24, the phonon dispersion relations predict an upshift of the low-lying optical modes ($<30cm^{-1}$) in the presence of the light guest ("rattler") Rb. We also find large isotropic atomic displacement parameters (U_{iso}) when the Rb occupies the large cages (Sn_{28}). The modes associated with these guests should contribute strongly to lowering the lattice thermal conductivity (k_L). This is reinforced by our evaluation of the guest-associated effective potential energy curves E(x). Our calculated effective harmonic spring constants K for these guests show that a simple harmonic oscillator model is in good agreement with the first principles lattice dynamical calculations. The similarity between $\omega_{os} = (K/M)^{1/2}$ and our computed guest phonon frequencies implies that anharmonic contributions to the guest vibrational modes are not significant. Our calculations of the vibrational contribution to the specific heat and our estimation of $k_L$are also presented and discussed.

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