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First-principles Study of Guest Impurities of Na and Ba on Lattice Thermal Conductivity of Type-I Si Clathrate YI XIA, University of California, Los Angeles, FEI ZHOU, Lawrence Livermore National Laboratory, VIDVUDS OZOLINS, University of California, Los Angeles — The type-I clathrate compounds, known as good thermoelectric materials, exhibit phonon-glass electron-crystal properties with their open cages filled with guest atoms. We present a first-principles study of the intrinsic impact of these rattlers on the lattice thermal conductivity in type-I Si clathrate compounds. We observe both coherent and incoherent coupling between guest and framework acoustic modes which could be ascribed to the difference in atomic radius, confirming the "avoided crossing" behavior. Our calculated lattice thermal conductivities for Si_{46} (37.64 W/(m K)), Na_8Si_{46} (2.75 W/(m K)) and Ba_8Si_{46} (1.37 W/(m K)) are in good agreement with experimental measurements and simulations at room temperature. Significant reductions in both phonon lifetime and group velocity due to guest atoms are responsible for the reduction in lattice thermal conductivity. The energy widths of acoustic modes of both empty and filled silicon clathrates are beyond the resolution limit of 0.2 meV of inelastic x-ray scattering and thus cannot be used to exclude the Umklapp scatterings as main cause of reduction in lattice thermal conductivity. In addition, the Umklapp scatterings of acoustic modes are directly mediated by the guest modes.

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