Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.
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Supplementary Information: First-Principles Study of Strain Effect on Thermoelectric Properties of LaP and LaAs

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(Dated: August 22, 2020)

In this document, we show the computational settings and convergence tests of density functional theory (DFT) calculations for various software packages discussed in the main text. These include structural relaxation in VASP, transport and thermoelectric properties in BoltzTraP2, phonon dispersion in Phonopy, and lattice thermal conductivity in Phono3py. All results presented below are based on the generalized gradient approximation (GGA) exchange-correlation energy functional.

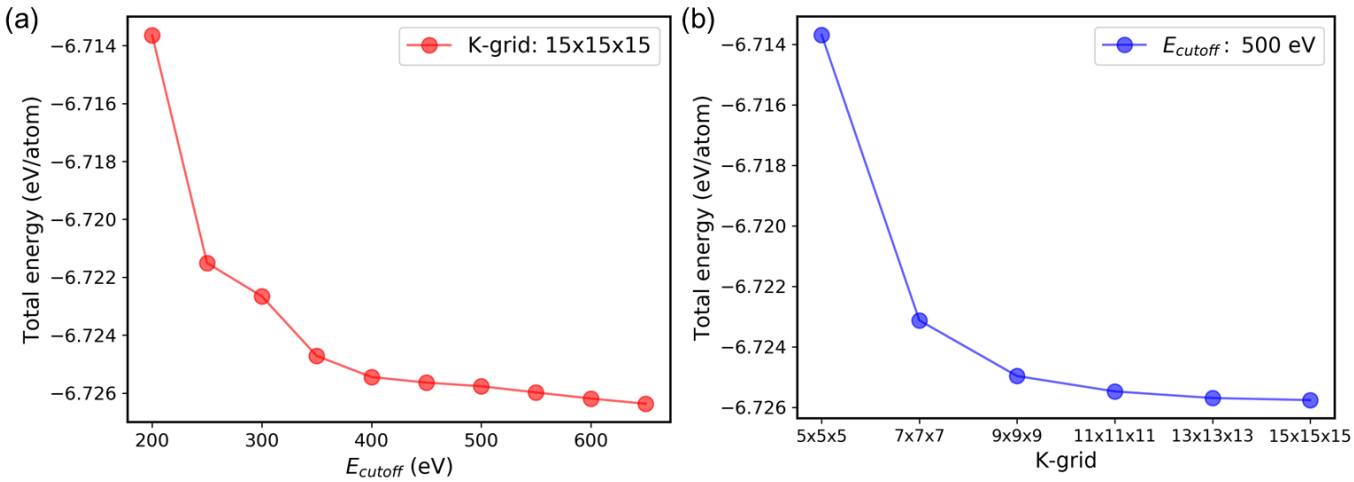

FIG. S1. Convergence tests on (a) plane-wave cutoff energy, and (b) k-point sampling, for VASP DFT total energy of LaP.

I. STRUCTURAL RELAXATION

In our calculations, we use the primitive cell of the rock-salt structure for the structural relaxation and thermoelectric properties. We first conduct convergence tests of plane-wave energy cutoff (E_{cutoff}) and k-grid for structural relaxation. The test results of DFT total energy for LaP are shown in FIG. S1, which indicates that a $E_{\text{cutoff}} = 500$ eV and a k-grid of 15×15× 15 points is sufficient to achieve an energy convergence within 1×10^{-4} eV/atom.

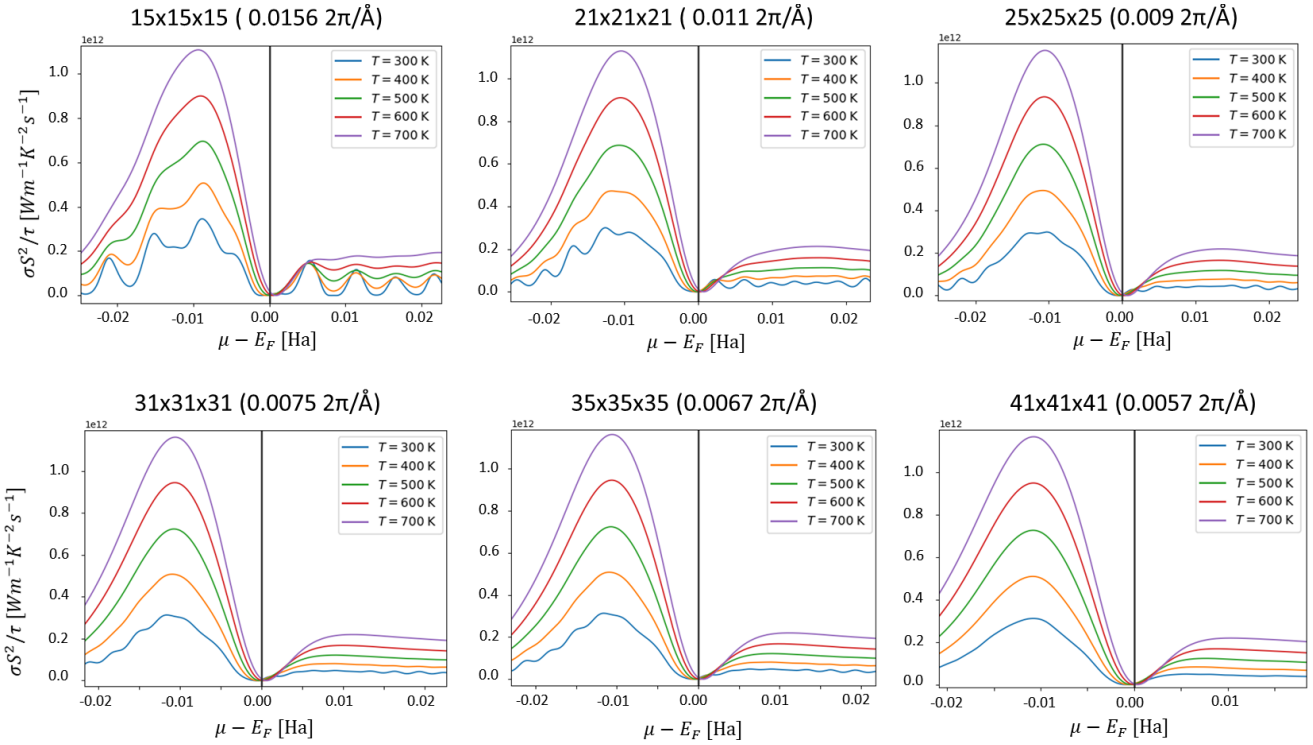

FIG. S2. Convergence tests on k-point sampling for LaP's power factor calculations by BoltzTraP2. The k-grid size ranges from 15×15 ×15 to 41×41×41 points. The result is well-converged with the 41 × 41 × 41 k-grid for temperature ≥ 300 K.

II. TRANSPORT AND THERMOELECTRIC PROPERTIES

The calculations of transport and thermoelectric properties using BoltzTraP2 require a very refined k-grid to achieve proper convergence, especially for low temperature. Here, we show the k-grid convergence test on power factor (σS^2, where σ and S are respectively the electrical conductivity and Seebeck coefficient) divided by the relaxation time τ. FIG. S2 shows systematic convergence tests for k-grid size ranging from $15\times15\times15$ to $41\times41\times41$ points. The results show that insufficient k-point sampling would cause artificial oscillating behavior in the temperature-dependent power factor. The power factor is well-converged using a 41 × 41 × 41 k-grid for temperature ≥ 300 K.

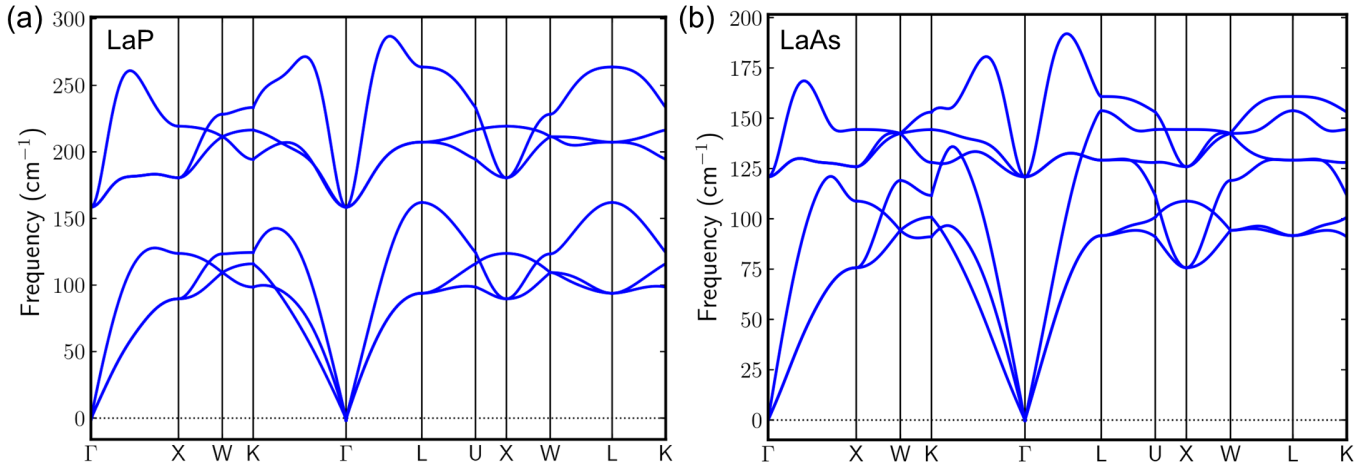

FIG. S3. Phonon dispersion spectra of (a) LaP and (b) LaAs, calculated by using a 5×5×5 supercell with the finite-displacement method in PHONOPY.

III. PHONON DISPERSION

In FIG. S3, we show the phonon dispersion spectra for LaP and LaAs. The calculations are performed using a 5×5×5 supercell with the finite-displacement method. The absence of any imaginary (negative) phonon mode indicates dynamical stabilities of these structures.

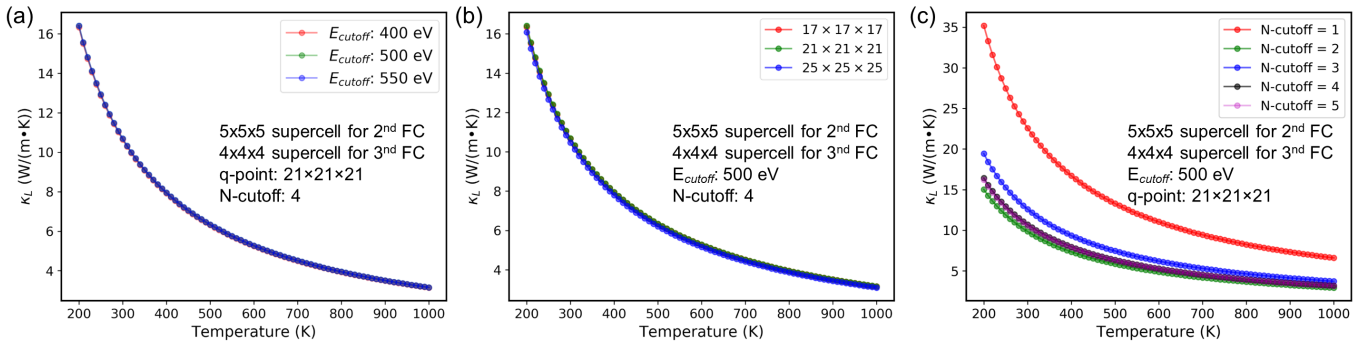

FIG. S4. Convergence tests for (a) E_{cutoff}, (b) q-point, and (c) neighbor cutoff N-cutoff, on LaP lattice thermal conductivity calculated by PHONO3PY.

IV. ENERGY CUTOFF, q-POINT, AND NEIGHBOR CUTOFF CONVERGENCE TESTS FOR LATTICE THERMAL CONDUCTIVITY

In FIG. S4, we perform convergence tests on energy cutoff E_{cutoff}, q-point, and neighbor cutoff N-cutoff, for the lattice thermal conductivity κ_L. The calculations utilize 5×5×5 and 4×4×4 supercells for the second- and third-order force constants, respectively. We find that the lattice thermal conductivity is converged using a 500 eV E_{cutoff} and a 21×21×21 q-point sampling. Besides, we find that N-cutoff = 4 (or radius cutoff ~ 6.1 $\mathring{A}$ for unstrained LaP) is sufficient for the third-order force constants to achieve converged lattice thermal conductivity.