Revisiting lattice thermal conductivity of CsCl: The crucial role of quartic anharmonicity

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We utilized *ab initio* density functional theory as implemented in VASP, combined with projector-augmented wave (PAW) pseudopotentials and PBEsol functionals. The structural optimization was carried out with a cutoff energy of 650 eV and convergence thresholds of 10^{-4} eV/Å for Hellmann-Feynman forces on each atom and 10^{-8} eV for the total energy. Harmonic phonon and interatomic force constants (IFCs) calculations were conducted using the finite displacement technique utilizing a 4 × 4 × 4 supercell and a 11 × 11 × 11 Monkhorst-Pack *k*-grids. Subsequently, we performed a 2000-step *ab initio* molecular dynamics (AIMD) simulation at 300 K with 2 ps step sizes, selecting 40 random incoherent structures. Our approach to extracting third- and fourth-order IFCs was based on a compressive sensing lattice dynamics (CSLD) technique [1-3] to select the most physically relevant IFCs from force-displacement data under constraints imposed by the space group symmetry and operation. Combining all IFCs, thermal transport properties were calculated using our modified ShengBTE code for CsCl.

By performing various functionals, we provide a more comprehensive comparison in lattice constant and lattice thermal conductivity (κ_L) from computation and experimental results as shown in follows TABLE. I.

Lattice constant (Å)	Functional	κ_L (W m^{-1} K^{-1})	References
4.073	PBE	1.12@250 K	**Our work**
4.039	LDA	1.14@300 K	[4]
4.225	GGA	/	[4]
3.990	GGA	/	[5]
4.085	GGA	/	[6]
3.970	LDA	1.38@300 K	[7]
4.116	Experimental	1.11@250 K	[8]
4.090	Experimental	1.01@300 K	[9]

TABLE. SI. Comparison of lattice constants, functionals, and lattice thermal conductivity (κ_L) from computational and experimental results for CsCl.

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