Lattice thermal conductivity on argyrodite compounds Ag_8TS_6 (*T*= Si, Ge and Sn): Experimental and Theoretical approach

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Thermoelectric materials offer a promising solution for energy conservation and environmental protection by converting significant heat waste into consumable electrical energy and vice versa. Among them, argyrodite-type materials have lately sparked a lot of research interest due to their extraordinary thermoelectric properties.[1–3] In this work, we have studied Ag_8TS_6 (*T*= Si, Ge, Sn) argyrodite family by combining chemical-bonding analysis based on the Local Orbital Basis Suite Towards Electronic-Structure Reconstruction (LOBSTER)[4] and lattice vibrational properties simulation through foundation machine-learning interatomic potentials (MLIP)[5] and Density functional theory (DFT) to elucidate their structural and thermal transport properties.[6] The machine-learning potentials accurately reproduce experimentally measured lattice thermal conductivities over the entire temperature range while reducing computational cost compared to conventional approaches. Additionally, we propose a two-channel lattice-dynamics model that incorporates phonon lifetimes derived from Grüneisen parameters and compare its performance with the MLIP.[5] Both models successfully reproduce experimentally observed lattice thermal conductivities, highlighting their potential for high-throughput screening of thermoelectric materials. Additionally, we confirm predictive power of the chemical bonding situation, highlighting additional data-driven screening opportunities for materials with extreme thermal conductivity. Our results also confirm that the pronounced bond heterogeneity,[7] together with a large Grüneisen parameter and low sound velocities arising from the weakly bonded Ag^+ ions, are responsible for the low lattice thermal conductivity of Ag_8SnS_6, Ag_8GeS_6, and Ag_8SiS_6.

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