Supporting Information (SI)

A Combined First-Principles and Machine Learning Study of Phase Stability and Thermoelectric Properties in ZrSiPt

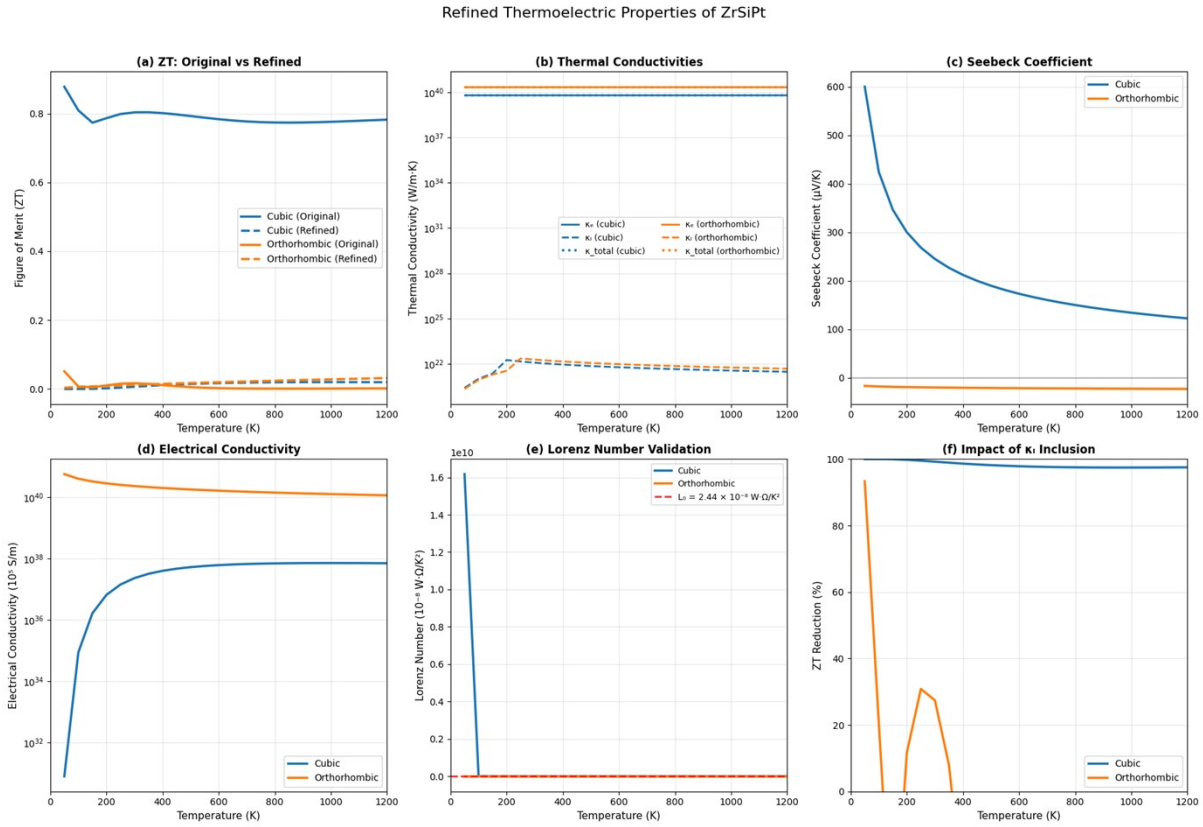

Fig. S1: Comprehensive Refined Thermoelectric Transport Properties and Validation for ZrSiPt Phases.

(a) Refined Figure of Merit (ZT): Comparison of the original (constant τ, assumed κ_L) and refined (deformation-potential τ, Debye-Callaway κ_L) ZT values for cubic and orthorhombic ZrSiPt as a function of temperature (0–1200 K). The drastic reduction for the cubic phase highlights the critical impact of its high lattice thermal conductivity.

(b) Thermal Conductivity Components: Temperature dependence of the total thermal conductivity (κ) and its constituents: electronic thermal conductivity (κ_e) and lattice thermal conductivity (κ_L) for both phases. The dominance of κ_L in the cubic phase and the ultralow κ_L in the orthorhombic phase are evident.

(c) Seebeck Coefficient (S): Temperature-dependent Seebeck coefficient for cubic (positive, p-type) and orthorhombic (negative, n-type) phases, calculated with the refined methodology.

(d) Electrical Conductivity (σ/τ): Electrical conductivity per relaxation time (σ/τ) versus temperature, derived from the deformation potential theory, showing the activated behavior of the cubic semiconductor and the semi-metallic character of the orthorhombic phase.

(e) Lorenz Number Validation: Calculated temperature-dependent Lorenz number (L) for both phases compared to the Sommerfeld constant ($L_0 = 2.44 \times 10^{-8}$ WΩK^{-2}, dashed line). The deviation, especially for the cubic phase at low T, validates the treatment beyond the constant L approximation and confirms the consistency of our electronic transport calculations [1, 2].

(f) Impact of Lattice Thermal Conductivity Inclusion: Percentage reduction in ZT upon the explicit inclusion of the calculated κL, defined as $\frac{ZT_{\text{orig}} - ZT_{\text{refined}}}{ZT_{\text{orig}}} \times 100\%$. This quantifies the dominant effect of κ_L on the cubic phase's performance.

Calculation Details for Panel (e): The Lorenz number was calculated from the transport distribution function within the Boltzmann framework as implemented in BoltzTraP2 [3]:

$$L(T) = \frac{\kappa_e(T)}{\sigma(T)T} = \frac{1}{e^2T}\left[\frac{L^{(2)}}{L^{(0)}} - \left(\frac{L^{(1)}}{L^{(0)}}\right)^2\right],$$

where $L^{(\alpha)}$ are the transport integrals:

$$L^{(\alpha)}(T,\mu) = \frac{1}{\Omega}\int \sigma(E)(E-\mu)^{\alpha}\left[-\frac{\partial f(T;\mu)}{\partial E}\right]dE.$$

Here, $\sigma(E)$ is the transport distribution, f is the Fermi-Dirac distribution, μ is the chemical potential, and Ω is the unit cell volume. This calculation does not assume a constant L and is performed self-consistently with the Seebeck and conductivity calculations [4].

References:

[1] M. Inglot et al., *Phys. Rev. B* 91, 115410 (2015).
[2] A. B. Pippard, *Proc. R. Soc. Lond. A* 305, 291 (1968).
[3] G. K. H. Madsen et al., *Comput. Phys. Commun.* 231, 140 (2018).

[4] Basak, K., Nath, S., Mondal, R. and Jana, D. (2023), Electric Field-Induced Phase Transition on HPX_6 (X = C, Si, Ge, Sn) Monolayers. Phys. Status Solidi B, 260: 2300112. https://doi.org/10.1002/pssb.202300112