

Probing Anharmonic Lattice Dynamics and Thermal Transport in Layered Perovskite LiYTlO₄ Anode

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Layered perovskite lithium yttrium titanate (LiYTlO₄) has recently emerged as a promising low-potential, ultrahigh-rate anode material for lithium-ion batteries. However, its lattice dynamics and thermal transport behavior remain largely unexplored, limiting a complete assessment of its practical applicability. Here, we combine experimental thermal conductivity measurements with a systematic theoretical study based on a machine-learning neural evolution potential (NEP). Our computational framework integrates the temperature-dependent effective potential method with the Wigner thermal transport (WTT) formalism, explicitly accounting for both diagonal and off-diagonal components of the heat-flux operator. Phonon calculations at 0 K reveal dynamical instabilities arising from rotations of the TiO₆ octahedra, which are stabilized at finite temperatures through anharmonic phonon renormalization. By incorporating particlelike phonon propagation and wavelike phonon tunneling within the WTT framework, we predict an orientation-averaged room-temperature lattice thermal conductivity (κ_L) of $3.6 \text{ Wm}^{-1} \text{ K}^{-1}$, which is qualitatively consistent with the experimentally measured value of $3.2 \pm 0.08 \text{ Wm}^{-1} \text{ K}^{-1}$ for polycrystalline samples. To further elucidate the role of ionic motion at elevated temperatures, we compute κ_L using the Green-Kubo approach within equilibrium molecular dynamics simulations based on the trained NEP, yielding improved agreement with experiment relative to the WTT prediction. Our results identify intrinsically low thermal conductivity as a key limitation for the practical application of LiYTlO₄ and provide broader insight into anharmonic lattice dynamics and thermal transport in complex battery materials.

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I. INTRODUCTION

Lithium-ion batteries (LIBs) stand as the cornerstone of modern energy storage technology, enabling widespread applications, including portable electronic devices and electric vehicles [1–4]. However, the ongoing pursuit of higher energy density, power density, and fast-charging capabilities has led to significant Joule heating during high-rate cycling [5,6], posing a fundamental challenge to both the performance and safety of batteries. This internal

heat generates from multiple sources: internal resistance, entropic contributions of electrochemical reactions, and polarization losses [7,8]. If not efficiently dissipated, accumulated heat can give rise to uneven temperature distribution, accelerated material degradation, shortened cycle life, and, in severe cases, thermal runaway [5,6,9]. Hence, effective thermal management is essential to ensure the overall performance, safety, and operational reliability of battery devices.

The effectiveness of battery thermal management is fundamentally governed by the intrinsic thermal transport properties of its constituent components, particularly the active electrode materials [9]. Poor thermal transport in electrode materials can lead to localized heat accumulation, accelerating performance degradation and increasing safety risks during battery operation [10]. Despite its importance for battery performance, safety, and operational reliability, current research on electrode materials has predominantly focused on electrochemical metrics,

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such as specific capacity, rate capability, and cycling stability, whereas their fundamental thermophysical properties, especially thermal conductivity and its microscopic origins, remain largely unexplored. This knowledge gap limits a comprehensive evaluation of the practical applicability and long-term reliability of electrode materials under realistic operating conditions. Recently, submicrometer lithium yttrium titanate (LiYTiO_4) with a layered perovskite structure has emerged as a promising low-potential and ultrahigh-rate intercalation-type anode for high-performance LIBs [11]. Although its electrochemical performance has been well established, the underlying lattice dynamics and thermal transport behavior remain elusive, which severely limits a complete assessment of its practical viability and application prospects.

Electrode materials such as LiYTiO_4 are typically multielements compounds with complex crystal structures [12,13]. Their unit cells contain a large number of atoms interconnected through hierarchical bonding, which gives rise to a dense phonon band structure and strong anharmonicity [14–16]. These features make the theoretical prediction of thermal transport in these materials rather challenging. The conventional Boltzmann transport equation (BTE), based on the assumption of well-separated phonon branches, captures only particlelike phonon propagation but neglects the wavelike tunneling between phonon eigenstates [17,18]. As a result, it often fails to fully explain the thermal conductivity behavior in such systems [19,20]. To overcome this shortcoming, many efforts have been made to establish unified theories and numerical techniques beyond the conventional BTE scheme. Notable progress in thermal transport theory has now enabled explicit consideration of the wave-particle duality of phonons [21–24]. Among these advances, Simoncelli *et al.* developed a unified theory by reformulating the BTE within the Wigner formalism [22,25], using a phonon velocity operator whose diagonal and off-diagonal elements, respectively, describe propagative and diffusive contributions to heat transport. This approach has been successfully applied to evaluate the κ of numerous strongly anharmonic and structurally complex crystals [26–29], thereby providing a solid theoretical foundation for studying thermal transport in battery materials. Nevertheless, it should be noted that the Wigner formalism, being rooted in lattice dynamics, does not account for the effects of potential Li-ion migration, as observed in superionic systems such as Ag_9GaSe_6 [30], Ag_8SnSe_6 [31], and CsCu_2I_3 [32], which may exert a non-negligible influence on κ_L . In contrast, molecular dynamics (MD) simulations inherently capture such ion migration along with wavelike tunneling and higher-order anharmonicity [33,34], offering a powerful alternative for investigating thermal transport in LIB materials.

In this work, we present a systematic investigation of the anharmonic lattice dynamics and thermal transport in

LiYTiO_4 , combining experimental measurements with theoretical modeling. Our approach employs a machine-learning potential-based framework that integrates the temperature-dependent effective potential (TDEP) method with the Wigner thermal transport (WTT) formalism. The framework accounts for diagonal and off-diagonal contributions of the heat-flux operator, which include both three-phonon (3ph) and four-phonon (4ph) scattering processes. At zero temperature, we identify dynamical instabilities arising from opposing rotations of adjacent TiO_6 octahedra. These highly unstable optical modes are stabilized at finite temperatures via anharmonic renormalization. The lattice thermal conductivity is then predicted using the WTT formalism, which incorporates both particlelike phonon propagation and wavelike coherence contributions. The resulting room-temperature average κ_L of $3.6 \text{ W m}^{-1} \text{ K}^{-1}$ is qualitatively supported by the experimentally measured value of $3.2 \pm 0.08 \text{ W m}^{-1} \text{ K}^{-1}$ for polycrystalline samples. To further assess the influence of ion hopping on high-temperature thermal transport, we evaluate the κ_L using the neural evolution potential (NEP)-based Green-Kubo equilibrium molecular dynamics (GK-EMD) method, which naturally captures all-order phonon scattering and ionic hopping contributions. The GK-EMD results show improved consistency with the experimental measurements compared with the WTT results. These results provide a comprehensive understanding of heat transport in LiYTiO_4 , thus advancing the knowledge of thermal transport in LIB materials.

II. METHODOLOGY

A. Neuroevolution machine learning potential

To navigate the trade-off between computational accuracy and cost, we employed a neuroevolution machine learning potential (NEP) [35] to describe interatomic interactions in LiYTiO_4 . The training and testing database was constructed using first-principles molecular dynamics simulations based on density functional theory (DFT). Structural optimizations and *Ab Initio* molecular dynamics (AIMD) simulations were carried out using the VASP [36], employing the projector-augmented wave method [37]. The exchange-correlation functional was treated within the generalized gradient approximation using the Perdew-Burke-Ernzerhof functional [38]. For structural optimization of the unit cell, a $2 \times 2 \times 2$ Monkhorst-Pack $\mathbf{k}$ -point mesh [39] was used for Brillouin zone integration, and a plane-wave cutoff energy of 550 eV was applied. The convergence criteria for the electronic self-consistent iteration and ionic relaxation were set to 10^{-8} eV for energy and $0.01 \text{ eV/\AA}$ for forces, respectively. AIMD simulations were carried out on a $1 \times 2 \times 2$ supercell (112 atoms) in the canonical (NVT) ensemble to sample energies, forces, and stresses across atomic configurations. Temperatures ranging from 100 to 800 K were sampled

with a time step of 1 fs, using a Nosé-Hoover thermostat [40] for temperature control. To enhance structural diversity, additional configurations were collected at 300 K under the NPT ensemble. Furthermore, to improve the transferability of the NEP, we applied $\pm 2\%$ strains to the original configurations, generating four deformed structures. For each deformed structure, atomic trajectories were generated using the same AIMD settings. All AIMD simulations employed a Γ -centered $2 \times 2 \times 2$ $\mathbf{k}$ -point grid.

To reduce frame correlation and account for the data efficiency of the NEP model, configurations were sampled from the AIMD trajectories at intervals of 10 steps, yielding an initial set of 574 structures for the first training round. The training set was subsequently expanded through an active learning strategy: from the remaining data, the MaxVol algorithm [41] was used to select 500 configurations that were structurally most distinct from the initial training set. The NEP obtained from the first training round was used to evaluate the energies and forces of these configurations, which were then incorporated into the training set. This results in a final training set of 1074 structures for the second round of training. During training, the radial and angular descriptor cutoffs were set to 8.0 and 4.0 Å, respectively, and the model quality was evaluated using the root-mean-square error (RMSE). To validate the accuracy of the trained NEP, we compared the phonon spectra derived from the NEP with those computed directly from DFT. The harmonic interatomic force constants (IFCs) at the ground state (0 K) were calculated using the finite displacement method with a $2 \times 2 \times 2$ supercell, as implemented in the ALAMODE package [42].

B. Thermal conductivity based on Wigner formalism

To accurately predict the lattice thermal conductivity of LiYTiO_4 , we employ the linearized WTT equation that explicitly includes the contributions from particlelike phonon propagation (κ_L^P) and wavelike tunneling of phonons (κ_L^C). Under the single-mode relaxation time approximation, the equation is formulated as [22,26,43]

$$\kappa_L^{P/C} = \frac{\hbar^2}{k_B T^2 V N} \sum_{\mathbf{q}} \sum_{j,j'} \frac{\Omega_{\mathbf{q}j} + \Omega_{\mathbf{q}j'}}{2} u_{\mathbf{q}jj'} \otimes u_{\mathbf{q}j'j} \times \frac{\Omega_{\mathbf{q}j} n_{\mathbf{q}j} (n_{\mathbf{q}j} + 1) + \Omega_{\mathbf{q}j'} n_{\mathbf{q}j'} (n_{\mathbf{q}j'} + 1)}{4(\Omega_{\mathbf{q}j} - \Omega_{\mathbf{q}j'})^2 + (\Gamma_{\mathbf{q}j} + \Gamma_{\mathbf{q}j'})^2} (\Gamma_{\mathbf{q}j} + \Gamma_{\mathbf{q}j'}), \quad (1)$$

where j and $\mathbf{q}$ denote the mode index and wave vector of a phonon mode, respectively. k_B is the Boltzmann constant, T the temperature, V the primitive-cell volume, N the total number of $\mathbf{q}$ -points sampled in the first Brillouin zone, and $n_{\mathbf{q}j}$ the phonon occupation following the Bose-Einstein distribution. $\Omega_{\mathbf{q}j}$ represents the anharmonically renormalized phonon frequency, and $u_{\mathbf{q}j'j}$ is the generalized group

velocity that involves both intrabranch and interbranch components [44]. In this expression, the diagonal terms ($j = j'$) of the heat-flux operator correspond to the populations contribution (κ_L^P), whereas the off-diagonal terms ($j \neq j'$) of the heat-flux operator yield the coherences' contribution (κ_L^C). The total lattice thermal conductivity is given by the sum of these two components: $\kappa_L = \kappa_L^P + \kappa_L^C$. Here, the $\Gamma_{\mathbf{q}j}$ stands for the total scattering rate, accounting for scattering from 3ph and 4ph interactions, as well as isotope scattering. These rates were computed based on Fermi's golden rule [43] using the temperature-dependent effective potential (TDEP) approach with renormalized IFCs up to fourth order. The individual linewidths due to 3ph and 4ph scattering processes are given by [17,26,43]

$$\Gamma_{\mathbf{q}}^{3\text{ph}} = \sum_{\mathbf{q}', \mathbf{q}''} \left\{ \frac{1}{2} (1 + n_{\mathbf{q}'}^0 + n_{\mathbf{q}''}^0) \Lambda_{-} + (n_{\mathbf{q}'}^0 - n_{\mathbf{q}''}^0) \Lambda_{+} \right\}, \quad (2)$$

$$\Gamma_{\mathbf{q}}^{4\text{ph}} = \sum_{\mathbf{q}', \mathbf{q}'', \mathbf{q}'''} \left\{ \frac{1}{6} \frac{n_{\mathbf{q}'}^0 n_{\mathbf{q}''}^0 n_{\mathbf{q}'''}^0}{n_{\mathbf{q}}^0} \Lambda_{---} + \frac{1}{2} \frac{(1 + n_{\mathbf{q}'}^0) n_{\mathbf{q}''}^0 n_{\mathbf{q}'''}^0}{n_{\mathbf{q}}^0} \Lambda_{+-} + \frac{1}{2} \frac{(1 + n_{\mathbf{q}'}^0)(1 + n_{\mathbf{q}''}^0) n_{\mathbf{q}'''}^0}{n_{\mathbf{q}}^0} \Lambda_{++} \right\}, \quad (3)$$

where $\Lambda_{\pm}$ and $\Lambda_{\pm\pm}$ are defined as

$$\Lambda_{\pm} = \frac{\pi}{4N} |V^{(3)}(\mathbf{q}, \pm \mathbf{q}', -\mathbf{q}'')|^2 \Delta_{\pm} \frac{\delta(\Omega_{\mathbf{q}} \pm \Omega_{\mathbf{q}'} - \Omega_{\mathbf{q}''})}{\Omega_{\mathbf{q}} \Omega_{\mathbf{q}'} \Omega_{\mathbf{q}''}}, \quad (4)$$

$$\Lambda_{\pm\pm} = \frac{\pi^2}{8N^2} |V^{(4)}(\mathbf{q}, \pm \mathbf{q}', \pm \mathbf{q}'', -\mathbf{q}''')|^2 \Delta_{\pm\pm} \times \frac{\delta(\Omega_{\mathbf{q}} \pm \Omega_{\mathbf{q}'} \pm \Omega_{\mathbf{q}''} - \Omega_{\mathbf{q}'''})}{\Omega_{\mathbf{q}} \Omega_{\mathbf{q}'} \Omega_{\mathbf{q}''} \Omega_{\mathbf{q}'''}}. \quad (5)$$

Here the quantities $V^{(3)}(\mathbf{q}, \pm \mathbf{q}', -\mathbf{q}'')$ and $V^{(4)}(\mathbf{q}, \pm \mathbf{q}', \pm \mathbf{q}'', -\mathbf{q}''')$ represent the reciprocal-space forms of the third- and fourth-order IFCs. The Dirac delta function $\delta(\Omega)$ enforces energy conservation in 3ph and 4ph scattering processes, while the Kronecker symbols $\Delta_{\pm}$ and $\Delta_{\pm\pm}$, shorthand for $\Delta_{\mathbf{q}\mathbf{q}'-\mathbf{q}''}$ and $\Delta_{\mathbf{q}\mathbf{q}'\mathbf{q}''-\mathbf{q}'''}, \mathbf{q}$, respectively, impose the conservation of crystal momentum.

Solving the WTT equation numerically requires second-, third-, and fourth-order IFCs as inputs, which were obtained using the trained NEP as implemented in the LAMMPS package [45]. To incorporate phonon renormalization effects, finite-temperature IFCs were calculated via

the TDEP method [46]. To reduce the computational cost, the atomic trajectories for TDEP were generated from classical MD simulations using the trained NEP, rather than directly from DFT calculations. At different temperatures, equilibrium MD simulations were performed in the NVT ensemble on a $2 \times 4 \times 4$ supercell containing 896 atoms, running for 100 000 steps with a time step of 1 fs. From these simulations, the renormalized second-, third-, and fourth-order IFCs were extracted using cutoff radii of 9, 5, and 3 Å, respectively. To validate the reliability of our fitted force constants using the TDEP technique, we adopted both a hierarchical fitting strategy and a joint-fitting approach to extract force constants of different orders. The resulting thermal conductivities obtained from the two protocols exhibit only minor discrepancies, and the truncation convergence of the force constants is shown in Tables S2 and S3 in Supplemental Material [47], confirming the reliability of our fittings. Thermal conductivity calculations were then carried out using the modified ShengBTE code [48,49] with a $15 \times 15 \times 15$ $\mathbf{q}$ -mesh and a $2 \times 4 \times 4$ supercell, which ensures the convergence of κ_L , as verified in Fig. S3 in Supplemental Material [47]. Given the high computational expense associated with 4ph scattering calculations, we adopted a sampling-estimation approach [50], whereby 4ph scattering rates are estimated from a subset of all possible phonon scattering events. A sample size of 10^6 was used for this estimation.

C. Green-Kubo molecular dynamics

Once the NEP with a small error is constructed, the lattice thermal conductivity of LiYTio₄ is evaluated using the Green-Kubo equilibrium molecular dynamics (GK-EMD) method as implemented in the open-source GPUMD package [51,52]. Based on the Green-Kubo relation [53], the κ_L is obtained as

$$\kappa_L = \frac{1}{3Vk_B T^2} \int_0^\infty \langle \mathbf{J}(t) \cdot \mathbf{J}(0) \rangle dt, \quad (6)$$

where $\mathbf{J}(t)$ denotes the heat current and the angular brackets represent the autocorrelation function, defined as

$$\mathbf{J} = \mathbf{J}_{\text{pot}} + \mathbf{J}_{\text{kin}} = \frac{1}{V} \sum_i \mathbb{W}_i \cdot \mathbf{v}_i + \frac{1}{V} \sum_i \mathbf{v}_i \epsilon_i. \quad (7)$$

Here, $\mathbf{J}_{\text{pot}}$ and $\mathbf{J}_{\text{kin}}$ correspond to the potential (virial) and kinetic (convective) contributions, respectively. The per-atom energy of particle i is given by $\epsilon_i = \frac{1}{2} m_i \mathbf{v}_i^2 + U_i$, where m_i , $\mathbf{v}_i$, and U_i denote its mass, velocity, and site potential energy. V is the system volume, and $\mathbb{W}_i$ is the per-atom virial of particle i . Note that the convective term was not explicitly included in our calculations, as ionic diffusion is negligible, as confirmed later.

To mitigate finite-size effects, which are mainly governed by the phonon wavelength in the GK-EMD method,

the simulation cell was constructed with a length of approximately 4 nm along the heat-flow direction, corresponding to a $4 \times 8 \times 8$ supercell containing 7168 atoms. The convergence of thermal conductivity with respect to the supercell size is shown in the inset of Fig. 3(d), confirming that the chosen supercell size is well converged.

A time step of 1 fs was used in the GK-EMD simulations, with periodic boundary conditions applied in all three directions. The system was first equilibrated with a 0.1-ns run in the NVT ensemble, followed by a 1-ns run for stabilization in the microcanonical (NVE) ensemble, and then a production run of 1 ns was performed to compute the heat-current autocorrelation function. The maximum correlation time was set to 100 ps, long enough to capture the relevant fluctuation-dissipation behavior. To reduce statistical uncertainties inherent in the GK-EMD approach, ten independent simulations with different initial velocities were carried out and their results averaged. Besides, ionic diffusion was characterized using mean squared displacement (MSD) and radial distribution function analyses. The MSD was calculated following the established methodology in Ref. [54]. For these calculations, a supercell containing 896 atoms was first equilibrated in the NVT ensemble for 100 000 steps. Following equilibration, MSD data were accumulated over a run of 1000 000 steps in the NVE ensemble, while radial distribution functions were obtained from atomic trajectories spanning 100 000 steps.

D. Experimental method

1. Synthesis and characterization of LiYTio₄ samples

We utilized a sol-gel method coupled with an ion-exchange process to synthesize LiYTio₄. The synthesis began with the preparation of a sodium-based precursor, NaYTio₄, via the sol-gel route, followed by a lithium-sodium ion exchange reaction. Compared to conventional high-temperature solid-state calcination, which typically requires prolonged thermal treatment, this approach substantially reduces the calcination time and effectively suppresses high-temperature-induced phase separation and abnormal grain growth. The lithium and sodium contents were determined by inductively coupled plasma optical emission spectrometry (ICP-OES). As summarized in Table S1 in Supplemental Material [47], the experimental lithium mass fraction is 3.17%, close to the theoretical value of 3.34%, while the residual sodium content is below 0.5%, indicating that most Na ions have been successfully substituted by Li ions. Further synthesis details can be found in our earlier work [11].

LiYTio₄ crystallizes in an orthorhombic layered perovskite structure with space group *Pbcm* [11]. Refined lattice parameters are $a = 11.0196$ Å, $b = 5.3994$ Å, and $c = 5.3929$ Å, in good agreement with the optimized

values of $a = 11.27$ Å, $b = 5.46$ Å, and $c = 5.35$ Å from our DFT calculations. As shown in Fig. 2(a), the crystal exhibits a layered arrangement along the a axis, consisting of alternating layers of LiO_4 tetrahedra, YO_8 dodecahedra, and TiO_6 octahedra.

Given the difficulty in obtaining single crystals, polycrystalline LiYTiO_4 samples with grain sizes of approximately 1–2 μm were synthesized. High-magnification SEM and transmission electron microscopy (TEM) images [Fig. S5(c) in Supplemental Material [47]] show smooth, dense, and defect-free surfaces. High-resolution TEM reveals clear lattice fringes, and fast Fourier transform (FFT) analysis confirms an interplanar spacing of 0.3663 nm, corresponding to the (300) plane. TEM at both low and high magnifications [Fig. S5(d) in Supplemental Material [47]] shows that the particles exhibit uniform, pore-free surfaces. Energy-dispersive x-ray spectroscopy (EDS) mapping [Fig. S5(d) in Supplemental Material [47]] demonstrates homogeneous distributions of Y, Ti, and O, while time-of-flight secondary ion mass spectrometry mapping [Fig. S5(e) in Supplemental Material [47]] indicates uniform lithium distribution on the sample surface. Together with the ICP-OES results, these observations confirm that the compacted samples are highly dense, with nearly complete Li^+ substitution and minimal residual Na^+ , and that any porosity or minor compositional variations are unlikely to significantly influence the measured lattice thermal conductivity.

2. Thermal transport measurements

Compacting powders. The LiYTiO_4 powders were loaded into a graphite die. The powder samples in the die were densified at 850 °C for 12 min under an axial pressure of 50 MPa in a high vacuum using an SPS system (SPS2000).

Thermal conductivity measurements. A laser flash apparatus (LFA467 HyperFlash) was used to record thermal transport properties of the pressed pellet of powders. The pellet was disc-shaped with a diameter of 9.5 mm and a thickness of 2.956 mm. Both circular faces of the pellet were coated with graphite prior to measurement, and the analysis area was 2.8 mm in diameter. The thermal conductivity was calculated from the formula $\kappa_L = D_{\text{th}} C_p \rho$, where D_{th} is the thermal diffusivity, C_p is the constant-pressure heat capacity, and ρ is the mass density of the specimens. The ρ was determined to be 3.62 g cm⁻³ based on the geometrical dimensions and masses of the pellet, which amounts to 84% of the theoretical density (4.31 g cm⁻³). This relatively low relative density indicates that the pellets were not fully compacted during pressing and retain considerable porosity. Under such conditions, any macroscopic residual stress generated during cold pressing is expected to be minimal

and easily relaxed by the existing pores and thus does not significantly affect our thermal measurements.

III. RESULTS AND DISCUSSION

A. Machine-learning interatomic potentials

The NEP is trained and tested, and its reliability is summarized in Fig. 1. It is seen that the energies, forces, and stresses, along with the corresponding error metrics, converge to stable values, indicating excellent generalization ability. Moreover, the diagonal correlation plot exhibits high consistency between predicted and DFT values, further confirming the strong predictive power of the NEP. The RMSEs for energies, forces, and stresses on the test dataset are 1.10 meV/atom, 61.71 meV/Å, and 9.04 meV/atom, respectively, demonstrating the high accuracy of the potential. To further validate the NEP, we compare energy-volume curves obtained from both NEP and DFT under 2% strain, as provided in Fig. S1 in Supplemental Material [47], and find good agreement. Additionally, the phonon dispersion calculated using the NEP aligns well with DFT results in Fig. S1 in Supplemental Material [47], underscoring the reliability of the NEP in reproducing DFT-level potential energy surface.

To assess its thermodynamic accuracy, we further evaluate a crucial, experimentally accessible C_p . Using renormalized IFCs extracted from NEP-based MD simulations at finite temperatures, we calculate C_p and compare it with experimental measurements [Fig. S8 in Supplemental Material [47]]. The good agreement demonstrates that the trained NEP captures the full spectrum of

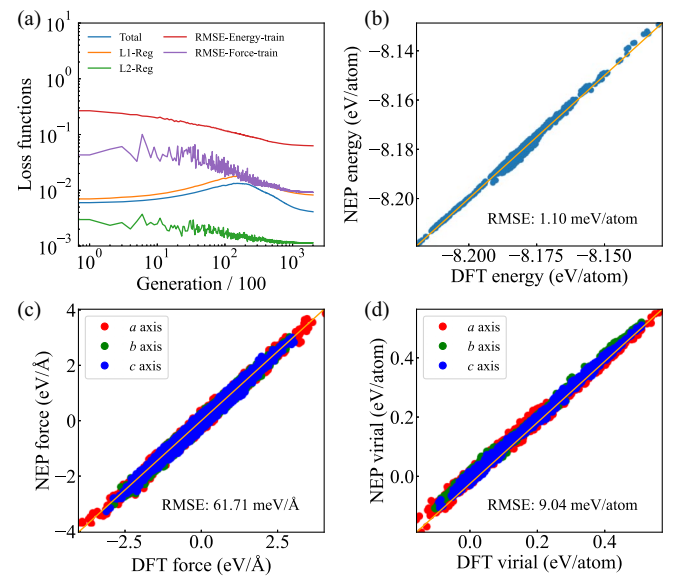


FIG. 1. (a) Evolution of the loss functions during the training process. (b) Energy, (c) force, and (d) virial as calculated from the NEP potential compared with the training data from DFT calculations.

lattice vibrations necessary to accurately predict macroscopic thermodynamic properties. These results collectively confirm that our trained NEP possesses high predictive accuracy, robustness, and thermodynamic reliability and can be confidently employed in subsequent simulations.

B. Anharmonic lattice dynamics

We begin by examining the lattice dynamics of LiYTio₄ within the harmonic approximation (HA) using the NEP. The calculated harmonic phonon dispersions at zero temperature, shown in Fig. 2(c), reveal several soft phonon branches with imaginary frequencies along high-symmetry directions in the Brillouin zone, indicating dynamical instability of the crystal structure at low temperatures. As evidenced by the atom-projected PDOS in Fig. 2(d), these soft modes arise primarily from vibrations of Y and O atoms. When temperature effects

are incorporated via the TDEP technique, the anharmonically renormalized phonon spectra become fully stabilized above room temperature. Apart from the hardening of the low-frequency soft modes with increasing temperature, we find that the high-frequency modes (>15 THz), primarily contributed by O and Li atoms, show significant softening, underscoring the strong anharmonicity in the system.

To attain deep insight into the lattice instability and anharmonicity in LiYTio₄, we calculate the PES of the lowest-frequency transverse acoustic (TA) mode at the X point, as shown in Fig. 2(e). At 0 K, the soft mode clearly exhibits a double-well potential, indicating strong lattice anharmonicity. This anharmonic double-well potential can be accurately reproduced by a polynomial fit that includes up to the sixth-order term. When the PES is decomposed into contributions of different orders, we find that the harmonic term of the soft mode has a negative coefficient, which accounts for the imaginary frequency and thus explains the failure of the conventional HA treatment. Although the PES of the soft mode displays anharmonic behavior up to the sixth order, the contribution from the sixth-order term is relatively minor. Therefore, in our anharmonic phonon renormalization calculations, only anharmonic contributions up to the fourth order are included. When finite-temperature effects are taken into account, the PES at 300 K transforms into a single U-shaped well with a positive curvature, underscoring the significance of quartic anharmonicity in the renormalization of phonon frequencies. This evolution indicates that the collective atomic dynamics, which are unstable at 0 K, become dynamically stable at finite temperatures. Vibrational mode analysis further reveals that the double-well U-shaped PES is associated with opposite rotations of adjacent TiO₆ octahedra along the crystallographic *b* axis, as illustrated in Fig. 2(f).

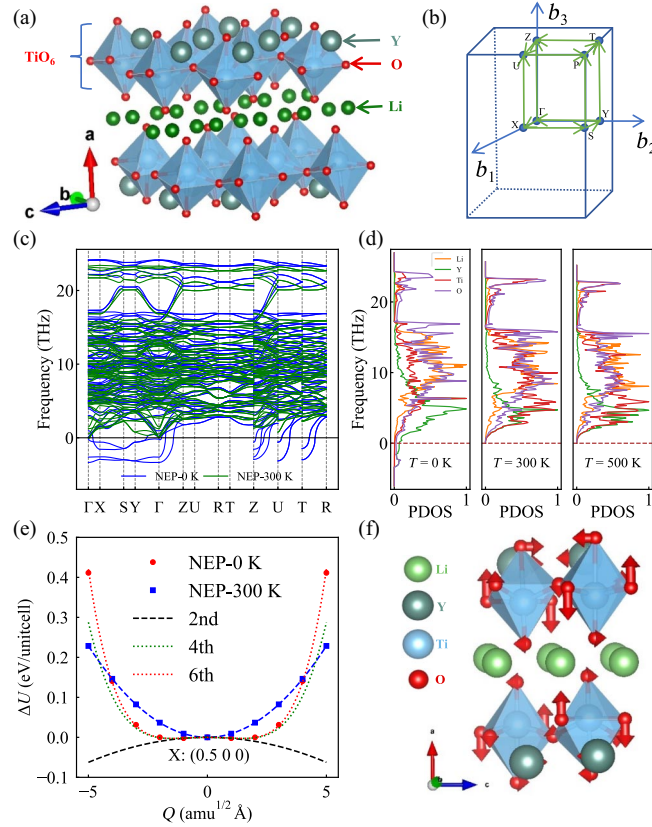


FIG. 2. (a) The crystal structure of layered perovskite LiYTio₄. (b) Brillouin zone of LiYTio₄. (c) Phonon spectra obtained from NEP at 0, 300, and 500 K. (d) Comparison of atom-decomposed partial phonon density of states (PDOS) at 0, 300, and 500 K. (e) Potential energy surfaces of the lowest TA mode at the X point calculated at 0 and 300 K. The dashed lines show the potential energy surface decomposed to the second, fourth, and sixth orders, respectively. (f) The crystal structure of LiYTio₄ with the displacement pattern of the soft phonon mode at the X point.

C. Li-ion diffusion dynamics

We have previously demonstrated, using nudged elastic band calculations, that Li ions in pristine LiYTio₄ migrate via a two-dimensional pathway within the *b*-*c* plane [11]. Such Li-ion diffusion may lead to the breakdown of the phonon picture and potentially induce pronounced phonon anharmonicity, which in turn may profoundly affect thermal transport behavior. It is therefore necessary to conduct an in-depth investigation of Li-ion diffusion dynamics at finite temperatures using machine-learning potential-based MD simulations. The hopping behavior of Li ions can be characterized via time-dependent mean square displacement (MSD) analysis, as shown in Fig. 3(a). At 300 K, the MSD of Li ions remains nearly constant over time, indicating localized oscillatory motion around their equilibrium positions. As the temperature increases to 700 K, Li ions begin to migrate, and their MSD grows approximately linearly with time, demonstrating the appearance of

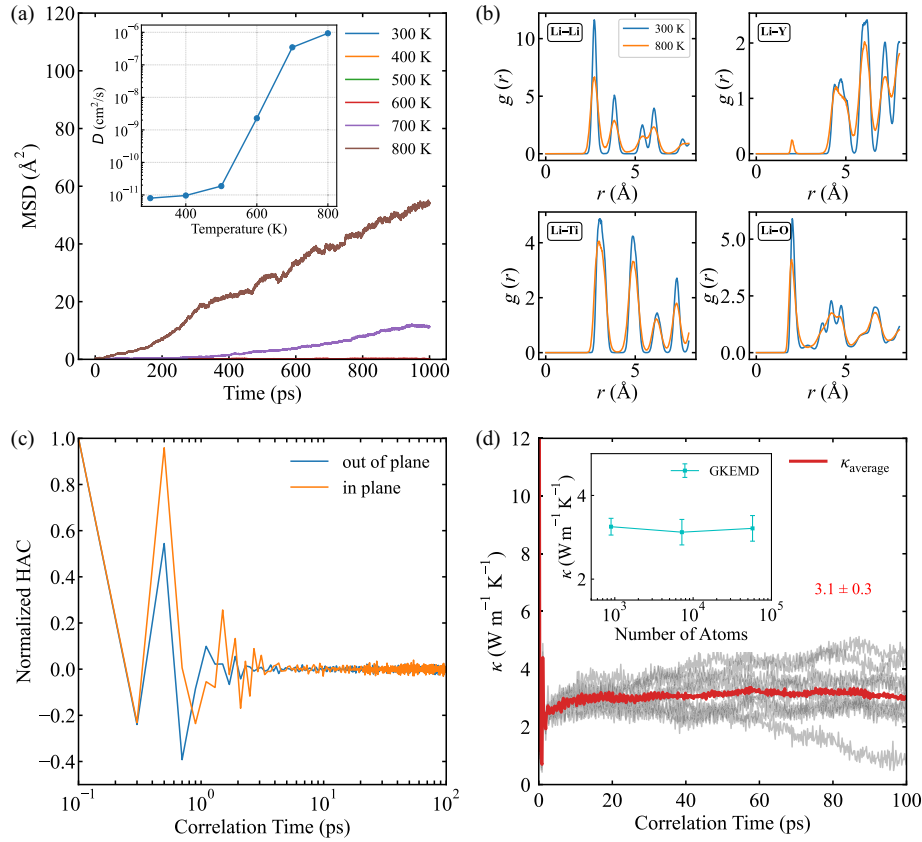


FIG. 3. (a) Time-dependent mean square displacement (MSD) of Li atoms from 300 to 800 K. (b) Comparison of radial distribution functions $g(r)$ at 300 and 800 K. (c) The heat-flux autocorrelation function at 300 K. (d) The running thermal conductivity averaged over three crystallographic directions, computed using the GK-EMD method at 300 K.

intralayer hopping between adjacent TiO_6 octahedral units at and above this temperature. Alongside Li ions, O atoms also exhibit noticeable diffusion at elevated temperatures, as revealed by their time-dependent MSD presented in Fig. S4 in Supplemental Material [47]. Based on the MSD data, we further calculate the Li-ion diffusion coefficients, as shown in the inset of Fig. 3(a). It is evident that, despite the occurrence of Li-ion diffusion at high temperatures, the diffusion coefficients remain below the characteristic threshold of $10^{-6} \text{ cm}^2/\text{s}$ typically associated with liquidlike ionic transport [55]. Consequently, it is justified in our GK-EMD calculations to consider only the conductive contribution while neglecting convective terms and the associated cross-correlations [56].

To further corroborate the Li-ion hopping behavior, we also compute the radial distribution functions $g(r)$ of relevant atomic pairs, which allow us to monitor the evolution of the local structure surrounding the Li ions. By comparing the cases at 300 and 800 K in Fig. 3(c), distinct changes in the peak width and shape are observed in the $g(r)$ for different atomic pairs. In particular, the $g(r)$ peaks for Li-Li and Li-O pairs become notably lower and broader at elevated temperatures. These results confirm

that Li-ion diffusion induces local structural relaxation at high temperatures.

This ion diffusion behavior may render the conventional phonon gas model inadequate or inaccurate for describing thermal transport in LiYTiO_4 . To overcome this limitation, we employ the NEP-based GK-EMD method, which provides a more comprehensive and accurate prediction of κ_L by inherently incorporating all-order anharmonic phonon interactions and explicitly accounting for ionic hopping effects. Figure 3(c) showcases the normalized heat-flux autocorrelation versus correlation time for the in-plane and out-of-plane directions at 300 K. The results indicate that a truncation time of 10 ps is sufficient to achieve well-converged values. At elevated temperatures, where phonon lifetimes are shorter, even faster convergence of the heat-flux autocorrelation is expected. Accordingly, Fig. 3(d) presents the calculated κ_L as a function of the truncation time, confirming that 10 ps adequately ensures convergence of the thermal conductivity. Based on ten independent EMD simulations, the thermal conductivity is statistically averaged over the three crystalline directions to be $3.1 \pm 0.3 \text{ W m}^{-1} \text{ K}^{-1}$ at 300 K. This value is close to our measured value of $3.2 \pm 0.1 \text{ W m}^{-1} \text{ K}^{-1}$ for polycrystalline

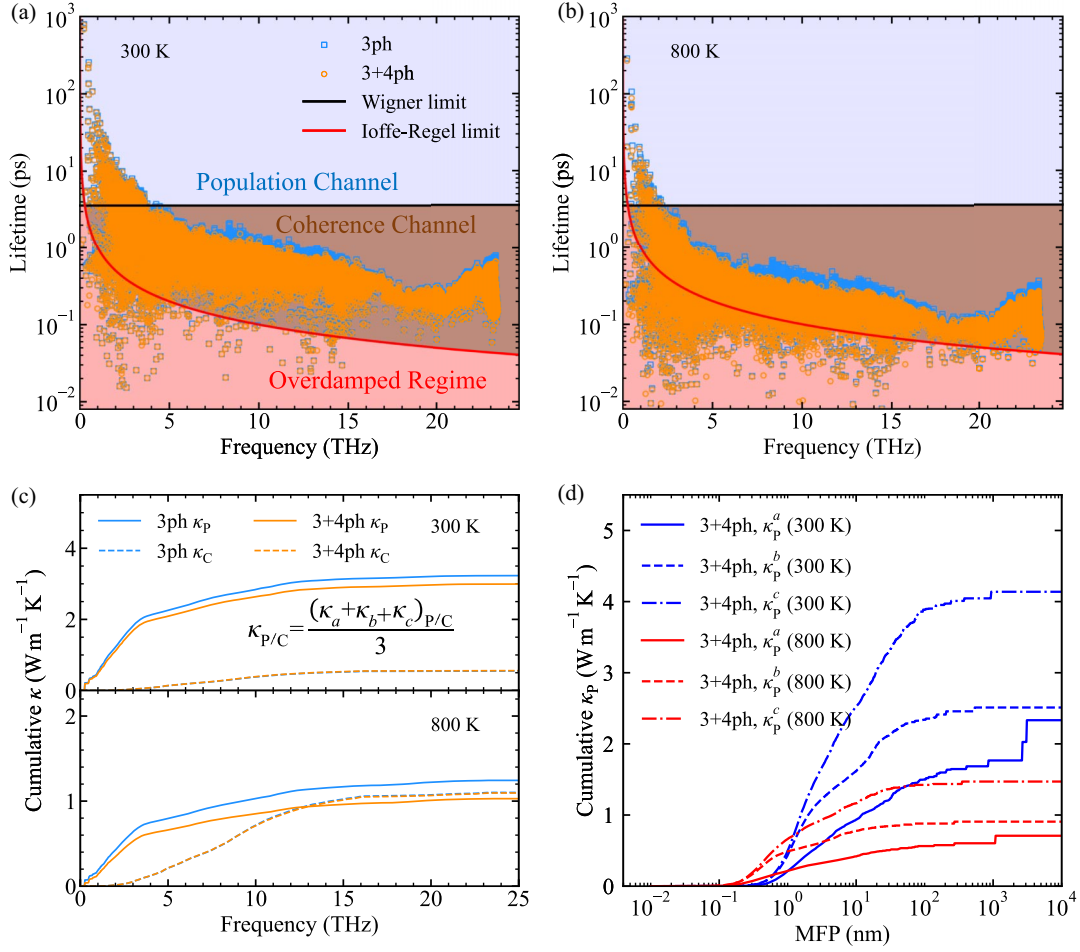


FIG. 4. Computed phonon lifetimes of LiYTiO_4 at (a) 300 K and (b) 800 K with and without including 4ph scattering. For comparison, the Ioffe-Regel and Wigner limits in time are provided. (c) Cumulative thermal conductivities (κ_P^P and κ_C^C) as functions of frequency averaged over different crystallographic axes at 300 and 800 K. (d) Cumulative thermal conductivity with respect to the phonon MFP along the in-plane (c axis) and out-of-plane (a axis) directions.

samples, thereby validating the reliability of the NEP-based GK-EMD description of thermal transport in LiYTiO_4 . Using the same computational setup, we further calculate the thermal conductivity at other temperatures.

D. Particlelike and wavelike contributions to lattice thermal conductivity

Given its complex unit cell, LiYTiO_4 exhibits an intricate phonon band structure with the large number of closely spaced optical phonon branches [Fig. 2(c)], along with strong anharmonicity as revealed by its large Grüneisen parameters [see Fig. S2 in Supplemental Material [47]]. These features collectively signal that phonon wave-particle duality plays a critical role in its thermal transport. To verify this inference, we plot in Fig. 4(a) the computed phonon lifetimes at 300 K against the Ioffe-Regel limit in time [57] (corresponding to a phonon scattering rate equal to the phonon frequency, i.e., $1/\tau_{\text{Ioffe-Regel}} = \omega$) and the

Wigner limit [22] (corresponding to a phonon scattering rate equal to the average interband spacing, i.e., $1/\tau_{\text{Wigner}} = \Delta\omega_{\text{ave}}$). Phonons with τ longer than the Wigner limit contribute to κ_L^P via particlelike propagation, while those with scattering rates between the Ioffe-Regel and Wigner limits contribute to κ_L^C via wavelike tunneling. Critically, our results show that the majority of phonons reside in this wave-tunneling regime, a trend that becomes even more pronounced when 4ph scattering is included, underscoring the pivotal role of coherent transport. It is clearly seen from Fig. 4(d) that when the temperature increases to 800 K, a greater number of phonons drop below the Wigner limit, indicating that the contribution of wave-like tunneling to κ_L grows significantly with temperature.

With finite-temperature IFCs obtained, we proceed to calculate the κ_L of LiYTiO_4 using the WTT equation, accounting for both phonon population and coherence contributions. It is evident from the cumulative κ_L with frequency in Fig. 4(c) that while particlelike transport

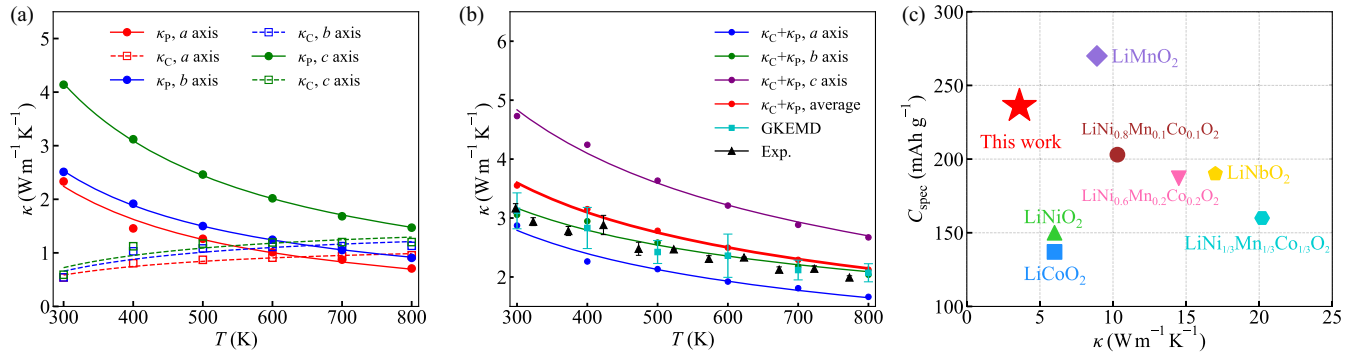


FIG. 5. (a) Calculated temperature-dependent κ_L^P and κ_L^C along different crystallographic axes, accounting for both 3ph and 4ph scattering processes. (b) The calculated total lattice thermal conductivity from the WTT equation (direction-dependent values and their average) and the GK-EMD method, compared with experimental measurements as a function of temperature. (c) Comparison of room-temperature thermal conductivity and typical specific capacity for various layered cathode materials [58–65].

dominates κ_L from 300 to 800 K, wavelike coherence also plays a substantial role, especially at high temperatures. Notably, phonons below 15 THz account for the majority of both transport contributions. Furthermore, we find that 4ph scattering noticeably suppresses the particlelike thermal conductivity at high temperatures, reducing κ_L^P by over 18% at 800 K, while having a negligible effect on the wavelike tunneling contribution.

From an experimental perspective, the phonon mean free path (MFP) spectra provide crucial insights into the influence of grain size on thermal conductivity. As shown in Fig. 4(d), the dominant phonon MFP along the in-plane directions (*b* axis and *c* axis) in LiYTio₄ at 300 K is below 1 μm and converges at a grain size of 1 μm , with phonons exceeding this threshold contributing only minimally. This indicates that the in-plane lattice thermal conductivity is effectively saturated at length scales relevant to our polycrystalline samples. For the out-of-plane direction (*a* axis), convergence requires a larger grain size of 3 μm . The difference in cumulative thermal conductivity between MFPs of 2 and 3 μm [corresponding to the sharp rise in Fig. 4(d)] is approximately 20% at 300 K. However, since the out-of-plane component represents only part of the total conductivity, the overall effect on the average thermal conductivity is limited to roughly 6% and decreases further at elevated temperatures. These observations confirm that grain boundaries in our synthesized polycrystalline samples have a negligible impact on lattice thermal transport. Therefore, direct comparison between our theoretical predictions and experimental measurements is physically justified and reasonable.

Figure 5(a) shows the calculated temperature-dependent thermal conductivities from the Wigner formalism for the κ_L^C and κ_L^P contributions along different crystallographic axes. It can be observed that particlelike conductivity (κ_L^P) exhibits significant anisotropy: its values along the in-plane directions (*c* and *b* axes) are significantly higher than those along the out-of-plane direction (*a* axis). This

pronounced anisotropy stems primarily from the orthorhombic layered structure of LiYTio₄, which features considerably stronger chemical bonding within the planes. In contrast, the corresponding coherent components (κ_L^C) show only minor variation across these directions. Moreover, we can see that at room temperature, κ_L^C along all three axes is substantially lower than κ_L^P , contributing only 19%, 18%, and 13% to the total thermal conductivity (κ_L), respectively. As the temperature rises to 800 K, the importance of κ_L^C increases markedly, accounting for 58%, 56%, and 45% of κ_L .

To validate our theoretically predicted lattice thermal conductivity of LiYTio₄, we compare the calculated total values ($\kappa_L = \kappa_L^P + \kappa_L^C$) with experimental measurements over the temperature range of 300–800 K, as shown in Fig. 5(b). Since the experiments were conducted on polycrystalline samples, the theoretical values are averaged over all three crystallographic axes to enable a direct comparison. The results indicate that the average thermal conductivity predicted by the Wigner formalism is slightly higher than the experimental data across the entire temperature range. This discrepancy likely arises from the neglect of temperature-induced structural changes, such as lattice expansion and distortion, as well as the omission of ion-hopping effects in the calculations. Notably, ion hopping resembles the rattling-like motion observed in certain superionic crystalline materials [13,66,67], which enhances anharmonicity. Increased anharmonicity tends to suppress particlelike thermal transport while simultaneously strengthening wavelike coherence contributions. Consequently, the net impact of ion hopping on κ_L reflects the interplay between particle-like and wavelike transport [20]. In contrast, the total thermal conductivity predicted by the GK-EMD method closely matches the experimental data across the full temperature range. The WTT predictions are slightly higher than the GK-EMD results, which is reasonable since MD simulations inherently account for all orders of anharmonic interactions and temperature-induced structural changes.

Therefore, the machine-learning-based GK-EMD approach offers a more complete framework for accurately predicting the thermal transport properties of LiYT₂O₄.

Combining theoretical predictions with experimental measurements, we find that the upper limits of the thermal conductivity of LiYT₂O₄ along the in-plane and out-of-plane directions are only 3.9 and 2.9 Wm⁻¹ K⁻¹ at 300 K, respectively. Although the maximum specific capacity of LiYT₂O₄ is highly prominent, surpassing that of most conventional layered cathode materials [58–65], its thermal conductivity remains nearly the lowest among these systems, as presented in Fig. 5(c). This implies that despite its outstanding electrochemical performance, the ultralow thermal conductivity of LiYT₂O₄ may constitute a major bottleneck to its practical application.

IV. CONCLUSION

In summary, we have established a comprehensive understanding of the lattice dynamics and thermal transport in pristine LiYT₂O₄ through combined experimental and theoretical investigations. Phonon calculations at 0 K reveal dynamical instabilities associated with opposite rotations of adjacent TiO₆ octahedra, which are stabilized at finite temperatures via anharmonic renormalization. Using the WTT formalism, which explicitly accounts for three-phonon and four-phonon scattering as well as both particlelike and wavelike contributions, we predict an orientation-averaged κ_L of 3.6 Wm⁻¹ K⁻¹ at room temperature. This prediction is qualitatively validated by the experimentally measured value of 3.2 ± 0.08 Wm⁻¹ K⁻¹ for polycrystalline samples.

Mean-square displacement analyses indicate that Li-ion diffusion begins above 700 K and becomes increasingly significant at higher temperatures. To assess the influence of such ionic hopping on thermal transport, we calculate κ_L using NEP-based GK-EMD simulations. The resulting values show qualitative consistency with both the experimental measurements and WTT predictions, suggesting that Li-ion mobility plays a negligible role in determining κ_L even at elevated temperatures, consistent with observations in Cu₃BiS₃ [20]. These results imply that, in pristine LiYT₂O₄, Li-ion diffusion has little effect on lattice thermal transport. By contrast, in Li-inserted LiYT₂O₄ during battery charge-discharge cycles, lattice dynamics and thermal conductivity are expected to evolve with Li content, rendering the correlation between ionic diffusion and lattice thermal transport an interesting topic for future studies.

Overall, our work demonstrates that the integration of anharmonic phonon renormalization, wavelike coherent phonon transport, machine-learning-based molecular dynamics simulations, and experimental measurements provides a comprehensive framework for assessing the thermal transport properties of complex electrode materials. The intrinsically low thermal conductivity identified in

LiYT₂O₄ poses a significant challenge for its practical application as an anode material in lithium-ion batteries.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available because they contain commercially sensitive information. The data are available from the authors upon reasonable request.

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