

## Unveiling thermal transport mechanisms in alumina laser crystal using machine learning potential

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Heat dissipation capability of laser gain media is critical for ensuring the performance and stability of high-power laser systems. However, the thermal transport mechanisms in these materials, particularly in rare-earth ion-doped laser crystals under high-power heating conditions, remain largely elusive. In this study, we employ molecular dynamics simulations combined with a neuroevolution machine learning potential to investigate the effects of temperature and Ce doping on thermal transport in alumina. Our simulations reveal a significant reduction in thermal conductivity under uniform elevated temperatures and finite Ce doping concentrations, providing quantitative insight into thermal transport in Ce-doped  $\text{Al}_2\text{O}_3$  under conditions relevant to laser operation. Simulation results demonstrate that elevated temperature causes a broad-spectrum suppression of thermal transport, while Ce doping predominantly affects specific low-frequency acoustic phonon modes and a few optical modes. Further analysis uncovers a mode-selective phonon-dopant scattering mechanism, where resonant and polarization-dependent coupling leads to the selective suppression of distinct phonon modes. This mechanism is in sharp contrast with the conventional understanding of phonon-dopant scattering observed in typical crystals. These findings provide valuable insights into the atomic-level mechanisms governing heat dissipation in alumina laser crystal, thus paving the way for the design of laser materials with optimized thermal properties for high-power applications.

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### I. INTRODUCTION

As one of the landmark inventions of the mid-20th century, the laser has developed rapidly and found widespread applications in various areas such as communications, modern medicine, and industrial precision processing [1–3]. However, thermal effects remain a major challenge in laser technology and application [4,5]. Excessive temperature rise can degrade the efficiency of power transfer and induce thermal lensing, which can impair performance and compromise device reliability [6,7]. This is particularly critical under high-power operation, where the highly concentrated laser energy generates significant localized hotspots [8,9]. While various external cooling methods have been employed to mitigate heating [10,11], the intrinsic thermal conductivity  $\kappa$  of the gain medium remains a critical factor in heat dissipation. Therefore, a comprehensive understanding of the thermal properties of laser materials is essential for optimizing thermal management in high-power devices.

Among various laser materials, alumina crystal (i.e.,  $\text{Al}_2\text{O}_3$ ) holds significant importance in laser technologies and related industries due to its relatively high  $\kappa$ , considerable mechanical strength, and strong electrical insulation [5,12–14]. For example,  $\text{Ti}:\text{Al}_2\text{O}_3$  and  $\text{Cr}:\text{Al}_2\text{O}_3$  [15–17] are widely utilized as effective gain media in ultrafast and high-energy lasers. However, the emission wavelengths of these transition-metal-doped lasers are largely confined to the near-infrared spectrum, offering limited spectral coverage. In contrast, rare-earth (RE) luminescent ions provide a wealth of electronic transitions, spanning a broad range of emission wavelengths from the visible to the midinfrared regions. This versatility has been demonstrated in RE-doped systems, such as  $\text{Pr}:\text{LiYF}_4$  [18],  $\text{Yb}:\text{CaAl}_2\text{O}_4$  [19],  $\text{Tm}:\text{GdScO}_3$  [20], and  $\text{Er}:\text{CaF}_2$  [21] crystals. Nevertheless, these gain media typically suffer from low  $\kappa$ , even falling below  $1 \text{ W m}^{-1} \text{ K}^{-1}$ , which poses severe challenges for heat dissipation during high-power operation. Meanwhile, with advancements in crystal growth and doping techniques, incorporating RE ions into high- $\kappa$  hosts, like  $\text{Al}_2\text{O}_3$ , has attracted significant research interest [22,23]. This approach combines the diverse spectral properties of RE ions with the favorable thermophysical attributes of  $\text{Al}_2\text{O}_3$ , showing great potential for laser applications.

However, an inevitable consequence of doping is its detrimental impact on thermal transport, often leading to a substantial reduction in  $\kappa$  [24–26]. In addition to the intrinsic mechanisms governing thermal transport, the impact of the extrinsic phonon-dopant scattering induced by the doping process on overall thermal transport remains poorly

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understood in RE-doped  $\text{Al}_2\text{O}_3$  crystal. Furthermore, under high-power laser operation, the significant thermal load induces a considerable temperature rise within the crystals. This elevated temperature strongly intensifies intrinsic phonon-phonon scattering, which drastically reduces phonon activities and thereby further suppresses the intrinsic thermal transport properties [27,28]. Unfortunately, its interplay with the aforementioned extrinsic phonon-dopant scattering remains elusive. Therefore, a systematic investigation into the effects of both temperature and doping under practical operating conditions on thermal transport is essential to balance the functional performance and thermal management for practical laser applications.

In this work, by using molecular dynamics (MD) simulations with a neuroevolution machine learning potential (NEP), we investigate the thermal transport properties of  $\text{Al}_2\text{O}_3$  over a range of temperatures and doping concentrations. To evaluate the effects of RE ion doping, we specifically focus on Ce-doped  $\text{Al}_2\text{O}_3$  because of its promising applications. The effects of elevated temperature and doping are further explored through modal-level decomposition and phonon lifetime analyses. Additionally, by correlating the observed unique phonon-dopant scattering behavior with phonon polarization vectors, we unveil a resonant and polarization-dependent scattering mechanism as the dominant effect of Ce doping on phonon dynamics. Our work provides a fundamental framework for understanding and tailoring thermal transport in Ce-doped laser systems.

The rest of this paper is organized as follows: Sec. II describes the methods used in this work, which include the training of machine learning potential, details of MD simulations, and the calculation details of phonon lifetime. In Sec. III, we present and discuss the main results. The accuracy of the NEP potential for doped  $\text{Al}_2\text{O}_3$  is validated in Sec. III A by comparing phonon properties. The distinct influences of temperature and doping to  $\kappa$  and spectral contributions are studied in Secs. III B and III C, respectively. We establish the relationship between polarization patterns and doping scattering sensitivity in Sec. III D. Finally, Sec. IV provides the conclusions of this work.

## II. MODEL AND METHODS

### A. Model

The atomic model of  $\text{Al}_2\text{O}_3$  (specifically  $\alpha$ - $\text{Al}_2\text{O}_3$ ) is constructed based on a rhombohedral primitive cell containing four aluminum atoms and six oxygen atoms in their ideal crystallographic positions, as shown in the left panel of Fig. 1. The optimized lattice parameters are  $a = 5.129 \text{ \AA}$  and  $\alpha = 55.3^\circ$ , showing good agreement with the experimental values [29] of  $a = 5.1284 \text{ \AA}$  and  $\alpha = 55.28^\circ$ . The right panel of Fig. 1 schematically illustrates the heat dissipation process in the laser crystal, starting with the absorption of pump energy, followed by device operation, the formation of localized hotspots leading to a temperature rise, and finally the dissipation of waste heat from these hotspots through the material. The upper-right inset provides an enlarged atomic-scale view of a dopant site, offering insights into the heat dissipation process. Green curved arrows indicate the phonon scattering processes

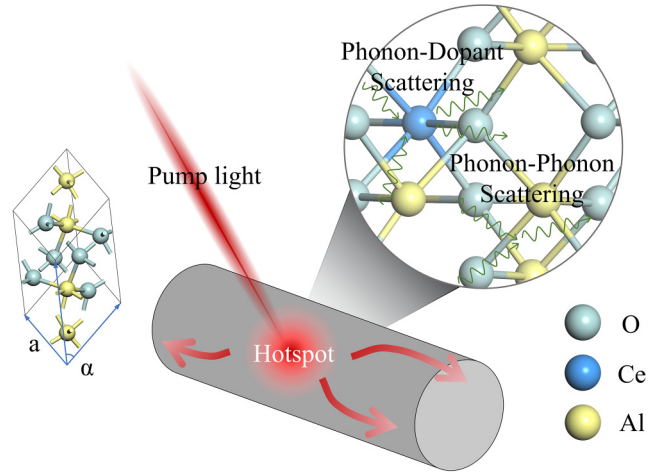


FIG. 1. Atomic structure of  $\text{Al}_2\text{O}_3$  on the left panel. A schematic illustration of laser operation on the right panel, where heat generation causes localized hotspots with temperature rise and thermal dissipation, along with a microscopic view depicting phonon-dopant and phonon-phonon scattering processes in laser crystals.

induced by both the dopants and other phonons, which in turn affect the thermal transport properties.

### B. Training of machine learning potential

To accurately describe the interatomic interactions in Ce-doped  $\text{Al}_2\text{O}_3$ , we have trained a NEP [30] following the protocol described in our previous work [31]. The training dataset is generated from *ab initio* molecular dynamics (AIMD) simulations performed using the Vienna Ab initio Simulation Package (VASP, version 6.1.2) [32]. The exchange-correlation functional is described by the Perdew-Burke-Ernzerhof (PBE) functional within the generalized gradient approximation [33]. The electron-ion interactions are treated using the projector augmented-wave (PAW) method with the PAW\_PBE O (08Apr2002), PAW\_PBE Ce (23Dec2003), and PAW\_PBE Al (04Jan2001) potentials. The corresponding valence configurations are  $2s^2 2p^4$  for O,  $5s^2 5p^6 5d^1 4f^1 6s^2$  for Ce, and  $3s^2 3p^1$  for Al. A plane-wave cutoff energy of 400 eV and a  $2 \times 2 \times 2$  Monkhorst-Pack  $k$ -point mesh are employed. The convergence criteria for the total energy and atomic forces are set to  $1 \times 10^{-6}$  eV and  $1 \times 10^{-5}$  eV  $\text{\AA}^{-1}$ , respectively. Electronic occupancies are treated using Fermi smearing (ISMEAR = -1) with a smearing width of 0.05 eV. Spin polarization is not included in the density functional theory (DFT) calculations. For the AIMD simulations used to generate the training dataset, simulations are performed in the isothermal-isobaric (NPT) ensemble using the Langevin molecular dynamics algorithm together with the Parrinello-Rahman barostat as implemented in vasp. The default thermostat and barostat coupling parameters (SMASS and PMASS) are adopted throughout the simulations. A time step of 0.5 fs is used throughout the simulations. These simulations employ a 270-atom supercell, in which 10 Al atoms at different sites are randomly substituted by Ce atoms to simulate the doping-induced interactions. The system is evolved over a temperature range of 100–700 K with a temperature

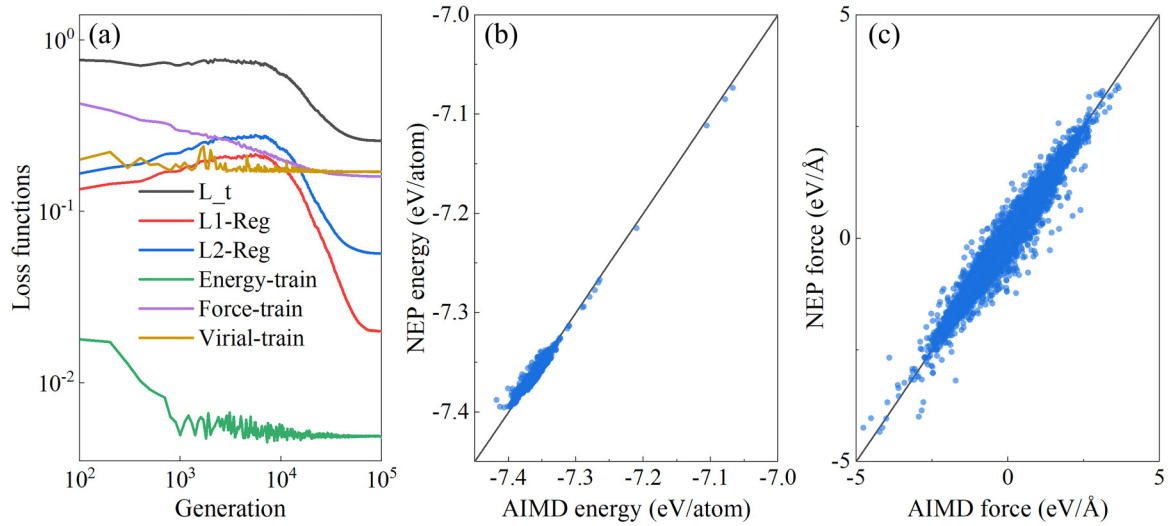


FIG. 2. Validation of the machine learned NEP for  $\text{Al}_2\text{O}_3$ . (a) Convergence of the loss functions (including total, L1, L2, energy, and force components) during the training process. Panels (b) and (c) compare the energy and force between the NEP predictions and AIMD calculations for the testing database, respectively.

interval of 50 K. In addition, three different doped structures are considered in the AIMD simulations. The final dataset, containing 5000 configurations, is subsequently divided into training (80%) and testing (20%) subsets.

To assess the validity of neglecting spin polarization in the DFT calculations, representative configurations extracted from the AIMD trajectory of the 9.3% Ce-doped  $\text{Al}_2\text{O}_3$  system at 300 K were recalculated using spin-polarized PBE while keeping the atomic structures fixed. As discussed in Appendix, the atomic forces obtained from the spin-polarized and non-spin-polarized calculations are in excellent agreement, with a coefficient of determination of  $R^2 = 0.994$  and a force root-mean-square error (RMSE) of  $0.121 \text{ eV } \text{\AA}^{-1}$ . These results indicate that introducing spin polarization has only a limited influence on the DFT forces, and therefore support the use of the non-spin-polarized calculations for constructing the NEP training dataset employed in the present work.

The NEP is trained using the graphics processing units molecular dynamics (GPUMD) package [34], which implements an explicit pairwise representation for many-body potentials [35,36]. The training process is conducted over 10<sup>5</sup> generations under default parameter settings. As shown in Fig. 2(a), the convergence of the loss function confirms the stability and reliability of the trained potential. Furthermore, Figs. 2(b) and 2(c) compare the energy and atomic forces predicted by the NEP with the AIMD reference values in the testing database, respectively. The trained NEP achieves a high accuracy: The RMSE for the energy and force in the testing database is  $4.63 \text{ meV atom}^{-1}$  and  $0.157 \text{ eV } \text{\AA}^{-1}$ , respectively. These error metrics, together with the phonon dispersion validation in Fig. 3(a), support the reliability of the trained NEP in describing the interatomic interactions and lattice vibrations of Ce-doped  $\text{Al}_2\text{O}_3$ .

The NEP describes the potential energy surface through local atomic environments within a cutoff radius of  $6 \text{ \AA}$ , rather than through the nominal dopant concentration of

the entire simulation cell. Although the training structures have a fixed Ce concentration, the random distribution of Ce dopants and AIMD sampling provide diverse local environments, including pristine Al–O coordination, isolated Ce-related configurations, and local regions containing multiple Ce dopants within the cutoff range (see Appendix). This local-environment coverage supports the use of the trained NEP for the Ce concentrations considered in this work.

To further evaluate the transferability of the trained NEP beyond the conventional training/testing procedure, additional independent AIMD validations are performed using pristine  $\text{Al}_2\text{O}_3$  and dilute Ce-doped  $\text{Al}_2\text{O}_3$  (1.9% Ce) structures that are not included in the training dataset. As discussed in Appendix, the NEP-predicted atomic forces are in excellent

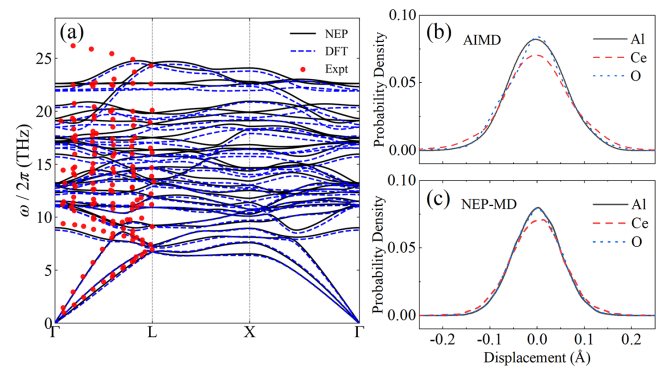


FIG. 3. Vibrational properties of  $\text{Al}_2\text{O}_3$  predicted by the NEP. (a) Phonon dispersion of pristine  $\text{Al}_2\text{O}_3$ . The solid and dashed lines represent calculation results using NEP and DFT, respectively, while the dots indicate experimental (Expt) results [52] for validation. Panels (b) and (c) show the normalized displacement probability distributions of Al, Ce, and O atoms derived from AIMD and NEP simulations, respectively.

agreement with the AIMD results, with overall force RMSE values of  $0.043 \text{ eV } \text{\AA}^{-1}$  for pristine  $\text{Al}_2\text{O}_3$  and  $0.112 \text{ eV } \text{\AA}^{-1}$  for the dilute-Ce structure, demonstrating that the trained NEP accurately describes the interatomic interactions over the composition range investigated in this work.

### C. Thermal transport simulations

Thermal conductivity  $\kappa$  and spectral thermal conductivity  $\kappa(\omega)$  are computed using homogeneous nonequilibrium molecular dynamics (HNEMD) simulations as implemented in the GPUMD package [34]. The modeled structure used in the thermal transport simulation is constructed by expanding the primitive cell into a cubic supercell containing 87780 atoms. The doping effect is simulated by randomly substituting Al atoms with Ce atoms at a certain doping concentration. The MD simulations are carried out with a 0.5 fs time step. During simulations, the system is first equilibrated in the isothermal-isobaric (NPT) ensemble at zero external pressure for 1 ns with periodic boundary conditions along all three directions. This is followed by a second equilibration in the canonical (NVT) ensemble for an additional 1 ns to ensure proper thermalization. Subsequently, the thermal transport and vibrational properties are computed during the production run. During laser operation, the generated hotspots can elevate the local temperature above room temperature. In the present simulations, we focus on temperature-dependent equilibrium thermal transport properties under uniformly elevated temperatures, rather than modeling nonequilibrium hotspot effects under practical operating conditions [37,38].

During the production stage, the HNEMD simulation is carried out in the NVT ensemble. An external driving force along the  $x$  direction is applied to generate a homogeneous heat current. Correspondingly,  $\kappa$  is calculated as [34]

$$\kappa(t) = \frac{\langle J(t) \rangle_{\text{ne}}}{TVF_e}, \quad (1)$$

where  $\langle J(t) \rangle_{\text{ne}}$  denotes the nonequilibrium ensemble average of the heat flux,  $T$  is the temperature, and  $V$  is the system volume. The external driving force  $F_e$  is set to  $1 \times 10^{-5} \text{ \AA}^{-1}$ . Following prior literature [34] and our additional linear-response tests for the present  $\text{Al}_2\text{O}_3$  system, this value is selected because  $1 \times 10^{-5}$  and  $1 \times 10^{-4} \text{ \AA}^{-1}$  give consistent thermal conductivities, whereas  $1 \times 10^{-6} \text{ \AA}^{-1}$  gives rise to large statistical fluctuations (see Appendix). The heat flux is recorded every 100 steps during the 1 ns production run for the calculations of  $\langle J(t) \rangle_{\text{ne}}$  and  $\kappa(t)$ . To assess the convergence of the  $\kappa$  calculations, we have evaluated both the time-dependent running thermal conductivity  $\kappa(t)$  and the statistical convergence over independent samples.  $\kappa(t)$  of pristine  $\text{Al}_2\text{O}_3$  reaches a stable plateau within the 1 ns production time (see Appendix), indicating that the simulation time is sufficiently large for the present  $\kappa$  calculations. For pristine  $\text{Al}_2\text{O}_3$ , five independent runs are performed. For each Ce-doped concentration, five random doped configurations are considered, and five independent runs are performed for each configuration, giving rise to 25 independent simulation samples in total for each doping concentration. The ensemble averaged  $\kappa$  gradually saturates with increasing sample number, supporting the statistical reliability of the calculated values (see Appendix).

To gain insight into the modal-level contributions, we further calculate the spectral thermal conductivity  $\kappa(\omega)$ . We first obtain the steady-state time correlation function of heat current, based on the following expression [34]:

$$\mathbf{K}(t) = \sum_i \sum_{j \neq i} \mathbf{r}_{ij}(0) \left\langle \left( \frac{\partial U_j}{\partial \mathbf{r}_{ji}}(0) \cdot \frac{\mathbf{p}_i(t)}{m_i} \right) \right\rangle_{\text{ne}}, \quad (2)$$

where  $\mathbf{r}_i$ ,  $m_i$ , and  $\mathbf{p}_i$  are the position, mass, and momentum of particle  $i$ . The position difference is defined as  $\mathbf{r}_{ij} = \mathbf{r}_j - \mathbf{r}_i$ .  $\mathbf{K}$  can be Fourier transformed to obtain the spectral heat current:

$$\mathbf{J}(\omega) = 2\tilde{\mathbf{K}}(\omega), \quad \langle \mathbf{J} \rangle_{\text{ne}} = \int_0^\infty \frac{d\omega}{2\pi} \mathbf{J}(\omega), \quad (3)$$

where  $\tilde{\mathbf{K}}(\omega)$  is the Fourier transform of the steady-state time correlation function. Then, the spectral thermal conductivity  $\kappa(\omega)$  along the same  $x$  direction is calculated according to Eq. (1), i.e.,  $\kappa(\omega) = \frac{J(\omega)}{TVF_e}$ . For the calculation of  $\kappa(\omega)$ , the heat current is sampled more frequently, namely, every 20 MD steps. With a time step of 0.5 fs, this corresponds to a sampling interval of 10 fs and a Nyquist frequency of 50 THz, which is sufficient to resolve the spectral range of doped  $\text{Al}_2\text{O}_3$ .

To assess the convergence of  $\kappa(\omega)$  calculations, the raw heat-current autocorrelation functions are provided in Appendix. In our calculations, the heat current is sampled every 20 MD steps, and 1000 correlation steps are used [34,35], corresponding to a maximum correlation time of 10 ps. The autocorrelation functions decay within this time window, supporting the convergence of the correlation integration. The production run is conducted for 1 ns to reduce statistical noise and achieve reliable spectral resolution, consistent with literature work [39]. In addition, uncertainty bands are added to  $\kappa(\omega)$  to quantify the statistical uncertainty from independent simulations.

### D. Phonon properties

The phonon dispersion of  $\text{Al}_2\text{O}_3$  is calculated by solving the dynamical matrix using the lattice dynamics method. The procedure begins with a full structural relaxation to obtain the equilibrium atomic configuration. The second-order interatomic force constants are then extracted by applying a set of finite atomic displacements within a symmetry-reduced supercell and then computing the resulting forces with the trained NEP, as performed by the phonopy software [40,41].

On the other hand, the phonon lifetimes are obtained for phonon scattering analysis considering the elevated temperature and doping effects [42,43]. The mode-dependent spectral energy density  $\Phi(\mathbf{k}, \nu)$  is first calculated from the normal mode velocities, as presented in Ref. [44], where  $\mathbf{k}$  and  $\nu$  indicate the wave vector and polarization branch, respectively. Meanwhile, the normal mode velocities are obtained by projecting the MD atomic velocities onto the reciprocal space at mode  $(\mathbf{k}, \nu)$ . The spectral energy density  $\Phi(\mathbf{k}, \nu)$  is then computed using the wave- and particlelike phonon transport (WPPT) package, which is a phonon-analysis code developed to extract mode-resolved phonon dynamics [45]. The source code is available in Ref. [46]. The obtained  $\Phi(\mathbf{k}, \nu)$  is fitted to

a superposition of Lorentzian functions as follows:

$$\Phi(\mathbf{k}, \nu) = \frac{I_0(\mathbf{k}, \nu)}{[\omega_0(\mathbf{k}, \nu) - \omega]^2 + \gamma^2(\mathbf{k}, \nu)}, \quad (4)$$

where  $\omega_0(\mathbf{k}, \nu)$  and  $\gamma(\mathbf{k}, \nu)$  are the central frequency and linewidth for mode  $(\mathbf{k}, \nu)$ , respectively, and  $I_0(\mathbf{k}, \nu)$  is a mode-dependent intensity. The phonon lifetime  $\tau(\mathbf{k}, \nu)$  is subsequently obtained from the linewidth via the relation  $\tau = 1/(2\gamma)$ . In the doped cases, the same crystal periodicity is applied in the projection process. Finally, the effects of elevated temperature and doping can be evaluated from the variation of the phonon lifetime  $\tau(\mathbf{k}, \nu)$ . In the randomly doped systems, translational symmetry is broken, so the wave vector and branch index are no longer exact quantum labels of the pristine crystal. Here, the atomic velocities of the doped systems are projected onto the phonon eigenmodes of pristine  $\text{Al}_2\text{O}_3$ , and the obtained mode-resolved quantities should be interpreted as projections onto the pristine phonon basis. Within this framework, the Ce-induced disorder is reflected by the lifetime reduction, spectral broadening, and suppression of the projected modal contributions [47,48].

### III. RESULTS AND DISCUSSION

#### A. Fundamental vibrational properties

In both semiconductors and insulators, such as the laser crystal  $\text{Al}_2\text{O}_3$  and its doped derivatives, lattice vibrations (i.e., phonons) play a fundamental role in governing the thermal transport properties of these materials in practical applications [49–51]. Furthermore, dopants are known to exert a significant influence on the vibrational characteristics at the atomic scale, thereby altering the thermal transport behavior.

To evaluate the accuracy of the NEP potential in capturing the fundamental vibrational properties of  $\text{Al}_2\text{O}_3$ , we first compare the phonon dispersion obtained from lattice dynamics calculations using NEP with experimental data for pristine  $\text{Al}_2\text{O}_3$ , as shown in Fig. 3(a). The predicted phonon dispersion shows overall good consistency with the experimental measurements. To further validate the applicability of the trained potential to pristine  $\text{Al}_2\text{O}_3$ , we also compare the NEP results with DFT calculations. The NEP and DFT results show good agreement with each other along the high-symmetry paths, especially for the acoustic and low-frequency optical branches that are relevant to thermal transport. These comparisons support the reliability of the trained NEP for describing the lattice vibrations of pristine  $\text{Al}_2\text{O}_3$ , corresponding to the low-concentration limit considered in this work. Some discrepancies remain for the highest-frequency optical modes near  $\Gamma$  point, partly due to the longitudinal-optical (LO) and transverse-optical (TO) splitting that is not explicitly captured by the present classical potential, which is expected to have minimal impact on thermal transport.

To further investigate the doping effect on lattice vibrations in Ce-doped  $\text{Al}_2\text{O}_3$ , we examine the local atomic displacements around Ce impurities. Figures 3(b) and 3(c) compare the displacement distributions of different atoms, expressed as normalized probability distributions of atomic displacements derived from AIMD and NEP-MD simulations, respectively. Figure 3(b) shows that the vibrations of the dopant Ce atoms are distinct from those of the pristine Al atoms, which

is expected to induce significant phonon-dopant scattering effect. Moreover, our NEP-based MD simulations also reproduce such a difference in the displacement distribution [see Fig. 3(c)]. The strong agreement between two methods demonstrates that the NEP effectively captures the local structural distortions and unique atomic vibrations introduced by Ce doping.

#### B. Effects of elevated temperature and doping on thermal conductivity

Figure 4(a) shows the predicted  $\kappa$  of pristine  $\text{Al}_2\text{O}_3$  from 100 to 700 K, together with literature results [53,54]. The overall trend exhibits a general decrease with increasing temperature, consistent with the dominance of Umklapp phonon scattering processes in this regime [55]. Our calculation results also agree well with the experimental data from Hofmeister [54] and with Boltzmann transport equation (BTE) calculations from Dongre *et al.* [53], particularly at elevated temperatures. However, below 200 K, the classical MD simulations yield significantly lower values than the BTE predictions. This discrepancy is largely attributable to the neglect of quantum effects in classical MD, which become important at low temperatures.

During laser operation, the generated hotspots can raise the temperature above room temperature, inducing a notable temperature effect [37,38]. The results in Fig. 4(a) show that increasing the temperature from 100 to 500 K sharply suppresses  $\kappa$  by as large as 70%, while the declining rate moderates above 500 K. Specifically,  $\text{Al}_2\text{O}_3$  exhibits a  $\kappa$  value exceeding  $33 \text{ W m}^{-1} \text{ K}^{-1}$  at 300 K and retains above  $12 \text{ W m}^{-1} \text{ K}^{-1}$  at around 500 K, which is the typical operating temperature under high-power laser conditions. This thermal performance surpasses that of other widely used laser materials [56–59], such as yttrium aluminum garnet (YAG), which exhibits a  $\kappa$  value [56,57] of  $10 \text{ W m}^{-1} \text{ K}^{-1}$  at 300 K and  $5 \text{ W m}^{-1} \text{ K}^{-1}$  at 500 K. The relatively high  $\kappa$  of  $\text{Al}_2\text{O}_3$  across this broad temperature range highlights its significant advantage for effective thermal management in laser applications.

On the other hand, as discussed in Figs. 3(b) and 3(c), Ce doping induces significant vibrational distortion compared to the pristine Al atoms. To further elucidate the effect of doping on thermal transport, we model the Ce doping effect by randomly substituting Al atoms with Ce in the  $\text{Al}_2\text{O}_3$  lattice. To prevent excessive structural distortion, typically associated with high doping concentrations, the Ce concentration is limited to below 2%. As shown in Fig. 4(b),  $\kappa$  decreases rapidly with increasing Ce concentration up to approximately 0.7%, beyond which the reduction rate slows down. Specifically,  $\kappa$  significantly drops by 70% and 58% as the doping concentration increases from 0% to 0.7% at temperatures of 300 and 500 K, respectively. In contrast, further increasing the doping level from 0.7% to 2% leads to a much smaller reduction of only 7% at both temperatures. These results show that  $\kappa$  decreases most rapidly at low doping concentrations. At higher doping levels, the reduction rate gradually slows down as phonon-dopant scattering approaches saturation, although the overall  $\kappa$  remains strongly suppressed. Within the typical operating temperature range of laser devices (e.g., 500 K), the corresponding  $\kappa$  of Ce-doped  $\text{Al}_2\text{O}_3$  falls between

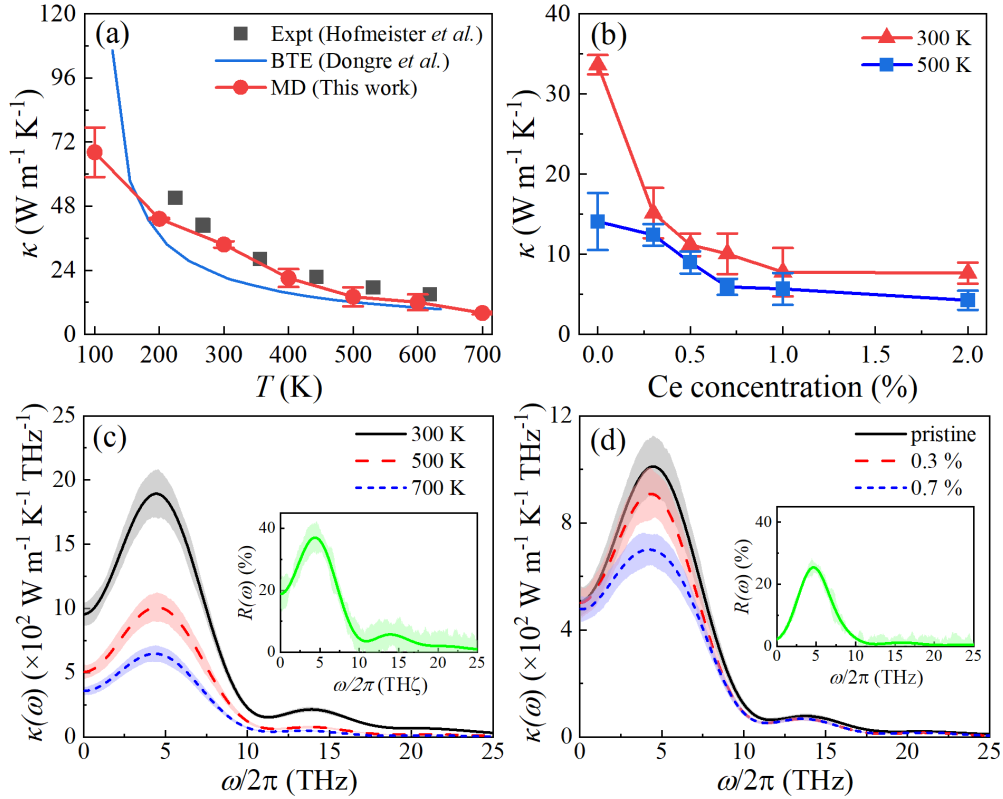


FIG. 4. (a) Thermal conductivity  $\kappa$  of pristine  $\text{Al}_2\text{O}_3$  vs temperature, comparing MD results (circles) with experimental data (squares) from Hofmeister [54] and BTE calculations (solid line) from Dongre *et al.* [53]. (b)  $\kappa$  of Ce-doped  $\text{Al}_2\text{O}_3$  vs Ce doping concentration at 300 and 500 K. (c) Spectral thermal conductivity  $\kappa(\omega)$  of pristine  $\text{Al}_2\text{O}_3$  at different temperatures. (d) Spectral thermal conductivity  $\kappa(\omega)$  of Ce-doped  $\text{Al}_2\text{O}_3$  at 300 K. The insets in panels (c) and (d) show the frequency-resolved reduction ratio  $R(\omega)$  from the temperature and doping effects, which represents the fractional reduction of  $\kappa$  at frequency  $\omega$  relative to the total  $\kappa$  of the pristine system. The shaded regions denote the uncertainty estimated from independent simulations. These results illustrate that heat transport is primarily governed by low-frequency phonons and is differentially suppressed by temperature (broad suppression) and doping (frequency-selective suppression).

approximately  $5$  and  $15 \text{ W m}^{-1} \text{ K}^{-1}$ , depending on the specific doping level, which demonstrates its viable heat dissipation window under practical conditions. In addition, compared to the reported  $\kappa$  of a typical Nd:YAG gain medium [60], the Ce-doped  $\text{Al}_2\text{O}_3$  system shows superior thermal performance, with  $\kappa$  values nearly double those of YAG-based systems (e.g., increasing from  $\sim 5.1$  to  $11.2 \text{ W m}^{-1} \text{ K}^{-1}$  at a 0.5% doping concentration). Furthermore, the converged  $\kappa$  values at higher doping levels indicate that  $\kappa$  of the Ce-doped  $\text{Al}_2\text{O}_3$  system remains substantially higher than those reported for other gain media under high doping. For instance, while high-doping concentration Yb:YAG often suffers from a drastic reduction in  $\kappa$  to below  $1 \text{ W m}^{-1} \text{ K}^{-1}$  [61], the  $\text{Al}_2\text{O}_3$  system maintains a much more robust heat dissipation capability even in the high-doping limit.

We note that Ce doping in the present work is described by a simplified substitutional model in which Ce atoms occupy Al sites, without explicitly introducing additional charge-compensating defect or defect complex. The AIMD-trained machine learning potential includes local electronic redistribution, polarization effects, and short-range electrostatic and chemical interactions within the local cutoff environment, while long-range electrostatics beyond the cutoff is

not explicitly treated in the production simulations. To assess this approximation, we have performed additional calculations with long-range electrostatic correction [62] for 0.7% Ce-doped  $\text{Al}_2\text{O}_3$  at 500 K, and obtained the calculated thermal conductivity of  $5.75 \pm 1.32 \text{ W m}^{-1} \text{ K}^{-1}$ . This result is very close to the predicted value of  $5.93 \pm 1.01 \text{ W m}^{-1} \text{ K}^{-1}$  without the long-range correction. This comparison indicates that the neglect of long-range electrostatic contribution has marginal impact on the calculated thermal conductivity and does not qualitatively affect the main conclusions.

### C. Modal-level decomposition of thermal conductivity

To further elucidate the underlying mechanism of thermal transport in  $\text{Al}_2\text{O}_3$  laser crystals, we decompose  $\kappa$  at the modal level, as shown by the spectral thermal conductivity  $\kappa(\omega)$  in Figs. 4(c) and 4(d). The  $\kappa(\omega)$  analysis reveals that low-frequency phonons, particularly those around 4.5 THz, predominantly contribute to the total  $\kappa$ . To quantify the spectral suppression, we introduce a frequency-resolved reduction ratio  $R(\omega)$  as

$$R(\omega) = \frac{[\kappa_{\text{pristine}}(\omega) - \kappa(\omega)]\Delta\omega}{\sum_{\omega} \kappa_{\text{pristine}}(\omega)\Delta\omega}, \quad (5)$$

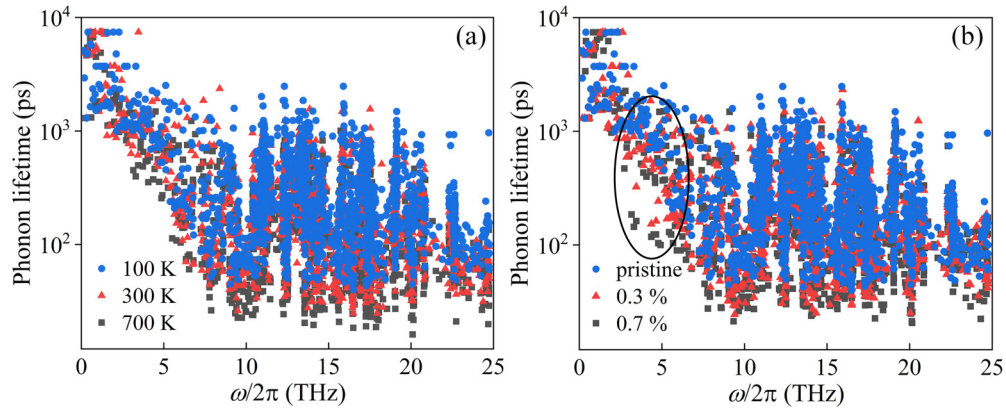


FIG. 5. (a) Phonon lifetime vs frequency for pristine Al<sub>2</sub>O<sub>3</sub> at different temperatures. (b) Phonon lifetime vs frequency for Ce-doped Al<sub>2</sub>O<sub>3</sub> at 300 K with various doping concentrations. The black circle in panel (b) highlights the predominant reduction occurring around 4.5 THz.

which represents the fractional reduction of  $\kappa$  at frequency  $\omega$  relative to the total  $\kappa$  of the pristine system, and  $\Delta\omega$  denotes the frequency interval in the discrete frequency grid.

The  $R(\omega)$  results for the temperature and doping effects are shown in the insets of Figs. 4(c) and 4(d), respectively. Moreover, with increasing temperature, a broad suppression of  $\kappa(\omega)$  is observed, especially across those low-frequency phonons. The inset in Fig. 4(c) confirms this broad suppression, where reduction ratio of  $\kappa$  remains significant across a wide frequency range. This broad suppression of the spectrum accounts for the decrease in  $\kappa$  with temperature, as shown in Fig. 4(a). In addition, these low-frequency phonons, corresponding to the acoustic phonons [see Fig. 3(a)], typically exhibit relatively long phonon mean free paths (i.e., MFPs) [63]. However, as the temperature rises due to laser operation, such as to 500 K, the phonon MFPs are significantly reduced, as indicated by the suppression of  $\kappa(\omega)$ , highlighting the critical constraints in laser design for effective heat dissipation [64].

In contrast, Fig. 4(d) and its inset show that Ce doping primarily suppresses the specific frequency contributions around 4.5 THz, while the remaining phonon modes are less affected by doping compared to the broad reduction of  $\kappa(\omega)$  caused by temperature effect. Notably, the suppression mechanism induced by Ce doping differs from previous reports on doping effects in thermal transport [49,50], which typically focus on dopants with small radii and lighter masses. Those studies highlight a strong suppression of high-frequency phonons with minimal impact on low-frequency phonons. For instance, Hahn *et al.* [65] reported that Ge doping in Si crystals primarily declines the high-frequency LO and TO phonons. The amplified suppression of specific frequency phonons in this work may arise from the unique structural and vibrational characteristics in Ce-doped Al<sub>2</sub>O<sub>3</sub>, which play a critical role in manipulating heat dissipation efficiency during the doping process.

#### D. Effects of elevated temperature and doping on phonon lifetimes

Figure 5 displays the frequency-dependent phonon lifetimes at varying temperatures and doping concentrations. A

consistent trend of decreasing lifetime with increasing frequency is observed, particularly for low-frequency regime, which aligns with the typical behavior reported in previous studies [66,67]. Meanwhile, the suppression of phonon lifetimes induced by the temperature effect is observed across the broad frequency spectrum, a typical characteristic of enhanced anharmonic phonon-phonon scattering at elevated temperature. This suppression contributes significantly to the reduction of  $\kappa$  and  $\kappa(\omega)$  in Figs. 4(a) and 4(c), respectively.

In contrast, the suppression induced by doping exhibits a different frequency dependence, as shown in Fig. 5(b). The weaker effect of doping on high-frequency phonons becomes evident when the phonon lifetimes are presented on a logarithmic scale, particularly as the doping concentration increases from 0.3% to 0.7%. Moreover, phonon lifetimes are notably suppressed around 4.5 THz (indicated by circle), which correlates well with the reduction of  $\kappa(\omega)$  shown in Fig. 4(d). Meanwhile, the suppression of lifetime  $\tau$  also differs from conventional impurity-phonon scattering, which generally produces a broadband Rayleigh-type scattering behavior with  $\tau^{-1} \propto \omega^4$  at low frequencies [48]. The present results show that Ce doping selectively suppresses specific acoustic and low-frequency optical phonon modes through frequency- and polarization-dependent coupling with the Ce-related localized vibration.

We have also quantified the averaged relative uncertainties of the phonon lifetimes. For the temperature-dependent results, the averaged uncertainties are 0.5%, 0.2%, and 0.6% at 100, 300, and 700 K, respectively, which are much smaller than the corresponding lifetime reductions of 17.5% from 100 to 300 K and 23.4% from 300 to 500 K. For the doping-dependent results at 300 K, the averaged uncertainties are 0.2%, 1.2%, and 2.6% for pristine, 0.3% Ce-doped, and 0.7% Ce-doped Al<sub>2</sub>O<sub>3</sub>, respectively. These values are also much smaller than the 11.9% reduction in lifetime from pristine to 0.3% Ce doping, and 17.5% reduction in lifetime from 0.3% to 0.7% Ce doping. Therefore, the observed lifetime suppression is much larger than the statistical uncertainty, which supports the conclusion that elevated temperature and Ce doping reduce phonon lifetimes.

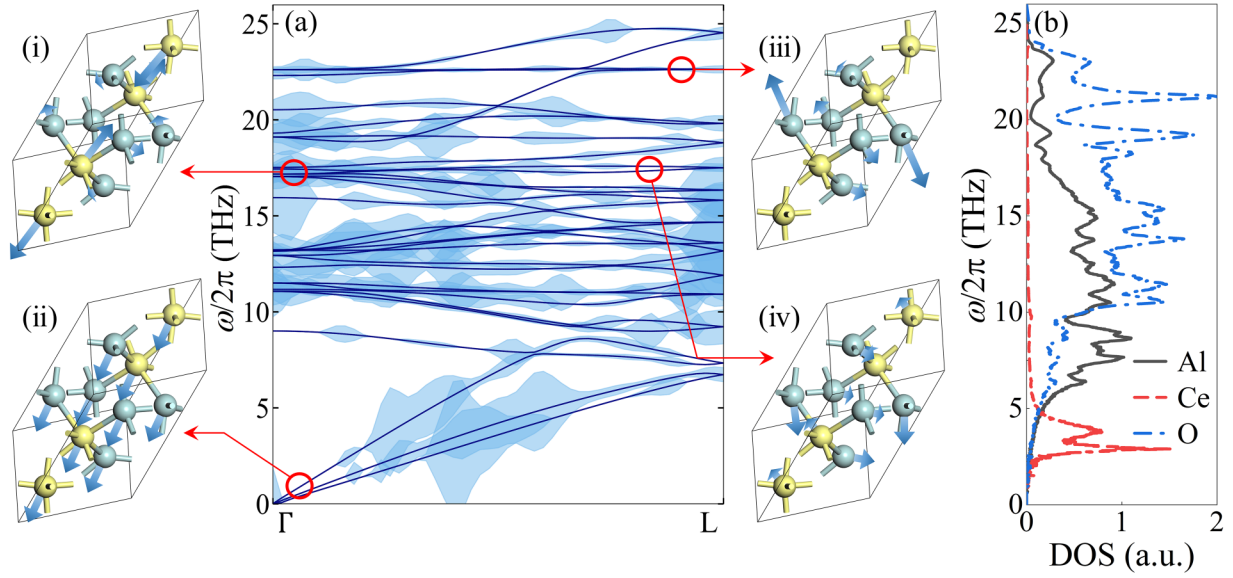


FIG. 6. (a) Phonon dispersion of  $\text{Al}_2\text{O}_3$  along  $\Gamma \rightarrow L$  path, with the projected linewidth representing the reduction in phonon lifetime from pristine to 0.7% Ce doping at 300 K. Four insets (i)–(iv) show the atomic polarization vectors of representative phonon modes, corresponding to the marked points on the dispersion curves. (b) Atom-projected DOS in Ce-doped  $\text{Al}_2\text{O}_3$ .

### E. Unique phonon-dopant scattering effect

We further investigate the unique Ce doping effect on phonon dynamics. As shown in Fig. 6, the linewidth of the phonon dispersion illustrates the reduction in phonon lifetime from pristine to 0.7% Ce-doped  $\text{Al}_2\text{O}_3$ . The most significant reduction in lifetime occurs at intermediate frequencies around 4.5 THz for acoustic phonons, with this effect being notably more pronounced compared to other acoustic and optical phonons, indicating a selective scattering phenomenon. Meanwhile, the phonon density of states (DOS) in Fig. 6(b) further reveals that the predominant contribution in the low-frequency range around 4.5 THz originates from the Ce atoms, which can be attributed to their large atomic mass of RE ions. This finding is consistent with the particular frequencies where the reductions in phonon lifetime and  $\kappa(\omega)$  are most significant. This agreement indicates that the vibrations of the doped Ce sites, along with their surrounding lattice, resonantly scatter the phonons in the pristine structure with similar intrinsic frequencies, leading to an enhanced phonon-dopant scattering in the selective specific frequency regime around 4.5 THz. Unlike conventional host-guest or rattling systems, where low-frequency resonant modes usually originate from weak bonding with cage-like structures [68,69], Ce-doped  $\text{Al}_2\text{O}_3$  is a dense crystalline oxide. The localized vibrational feature around 4.5 THz mainly arises from the large mass mismatch between substitutional Ce and host Al atoms, forming a narrow-band scattering channel for host phonons with compatible frequencies and polarizations. As a result, this resonance behavior particularly suppresses the phonon dynamics and thermal transport in the host  $\text{Al}_2\text{O}_3$ . In addition, the uncertainties of the mode-resolved linewidths are further projected onto the phonon dispersion (see Appendix). The relatively small uncertainty compared with the Ce-induced linewidth broadening indicates that the mode-selective scattering features identified in Fig. 6 are not dominated by statistical noise.

On the other hand, although high-frequency optical phonons are less important for thermal transport, their dynamics play a crucial role in hot electron-phonon scattering and nonradiative relaxation during the laser operation [70–72]. Our results in Fig. 6(a) also show that doping effects in  $\text{Al}_2\text{O}_3$  laser crystals induce nonuniform reductions in the lifetimes of high-frequency phonons, with some modes being significantly suppressed while others show relatively minor changes. This finding contrasts with previous work that reported a uniform reduction in optical phonon lifetimes across all wave vectors [65], confirming that the doping induces a mode-selective phonon scattering mechanism, rather than a uniform trend across frequency or wave vector.

Considering that Ce doping introduces structural distortions at the substituted sites, their vibrations localized around Ce sites should couple strongly with the propagating phonons of the host  $\text{Al}_2\text{O}_3$  matrix, thereby inducing phonon-dopant scattering [25,73]. The mode-selective phonon-dopant scattering mechanism is dependent on the microscopic vibrations of the Ce dopants and their couplings with the surrounding lattices. To further elucidate this point, we analyze the eigen-vibrations of  $\text{Al}_2\text{O}_3$  by examining the polarization vector of different phonon branches, as shown in the insets (i)–(iv) of Fig. 6(a). These insets illustrate the atomic polarization vectors of representative phonon modes.

In the acoustic branch [see inset (ii)], all atoms undergo collective vibrations, leading to strong couplings between dopants and surrounding atoms, which in turn results in pronounced phonon-dopant scattering and lifetime reduction around 4.5 THz. In contrast, the optical branches exhibit more localized vibrations, with distinct atomic participations. The mode shown in the inset (i) involves significant contributions from both O and Al atoms, leading to substantial scattering from Ce substitutions in the doped system. However, in insets (iii) and (iv), some specific vibrational modes are predomi-

nantly localized on O atoms, with minimal participation from Al atoms. Since Ce dopants specifically substitute for Al sites, these oxygen-dominated modes experience weaker scatterings, explaining their relatively preserved lifetimes despite the increasing doping concentration. This polarization-resolved analysis establishes a direct connection between the eigenstates of atomic vibrations and the Ce doping-induced phonon scattering, offering crucial insights into the selective suppression of specific phonon modes by point doping or defects in Ce-doped  $\text{Al}_2\text{O}_3$ . It is worth noting that the identified microscopic mechanisms primarily stem from the heavy mass of Ce ions and Ce doping-induced lattice distortions. The mechanisms identified in Ce-doped  $\text{Al}_2\text{O}_3$  may provide useful insight into thermal transport in other RE-doped laser crystals, including emerging high-entropy laser materials. However, their general applicability to different single-element or multielement RE dopants requires further validation.

#### IV. CONCLUSION

In summary, we have employed an atomic simulation approach with a machine learning potential to investigate the thermal transport mechanisms in Ce-doped  $\text{Al}_2\text{O}_3$ , a promising laser gain material. The neuroevolution machine learning potential trained in this work has shown reliable accuracy, enabling the modeling of temperature and doping effects in this laser crystal. Our simulation results show that both elevated temperature and Ce doping suppress the thermal conductivity of  $\text{Al}_2\text{O}_3$  crystal. Under elevated temperature, this suppression has manifested as a broad reduction across frequencies, whereas under doping, the suppression has been predominantly confined to a selective frequency regime. Additionally, we have identified a mode-selective phonon scattering mechanism with resonant and polarization-dependent couplings, which is responsible for the suppression of specific acoustic and optical phonon modes in Ce-doped  $\text{Al}_2\text{O}_3$ , leading to the promising thermal performance for  $\text{Al}_2\text{O}_3$  laser crystals. These findings have provided a solid foundation for the strategic design of doping schemes and material microstructures aimed at optimizing thermal performance in solid-state laser systems.

#### ACKNOWLEDGMENTS

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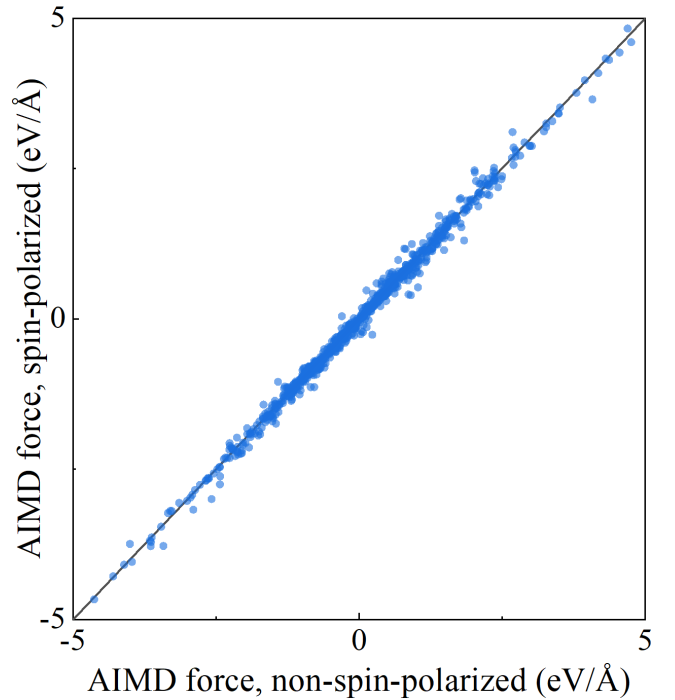


FIG. 7. Comparison of atomic forces obtained from non-spin-polarized and spin-polarized DFT calculations for representative configurations extracted from the AIMD trajectory of the 9.3% Ce-doped  $\text{Al}_2\text{O}_3$  system at 300 K. The atomic structures were kept fixed in both calculations. The solid line represents perfect agreement.

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#### DATA AVAILABILITY

The data supporting the findings of this study are available from the corresponding author upon reasonable request. The VASP input files used for the AIMD simulations and the final trained NEP model are openly available at [74].

#### APPENDIX

##### 1. Validation and transferability

The additional validation results for the first-principles calculations and the trained NEP are summarized in Figs. 7–9. First, we assess the influence of spin polarization on the DFT atomic forces used to construct the training dataset. Figure 7 compares the forces obtained from spin-polarized and non-spin-polarized PBE calculations for representative configurations extracted from the AIMD trajectory of 9.3% Ce-doped  $\text{Al}_2\text{O}_3$  at 300 K, with the atomic structures kept identical in these two calculations. The two sets of forces show good agreement, with a coefficient of determination of  $R^2 = 0.994$  and a force RMSE of  $0.121 \text{ eV}\text{\AA}^{-1}$ . This comparison indicates that the inclusion of spin polarization produces very limited changes in the atomic forces for the representative structures considered here, supporting the use of the

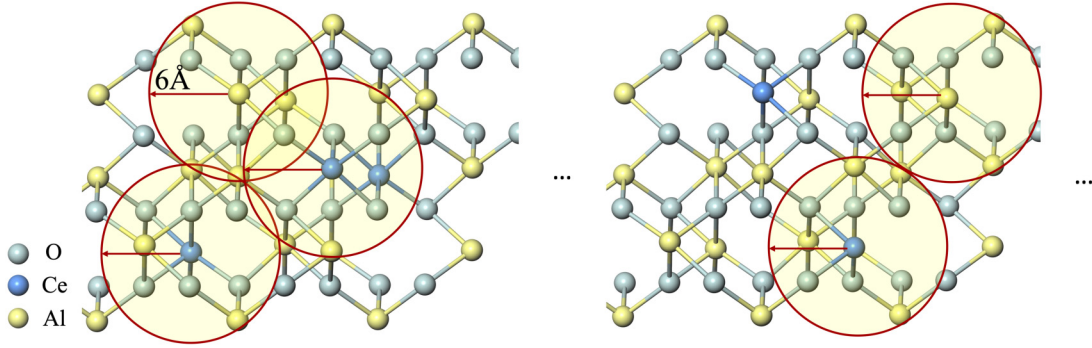


FIG. 8. Local atomic environments in the Ce-doped  $\text{Al}_2\text{O}_3$  training structure. The highlighted regions represent local coordination spheres within the 6 Å cutoff, including various configurations with zero, one, and multiple Ce dopants around selected atomic sites.

non-spin-polarized PBE calculations in constructing the NEP training dataset.

The applicability of the trained NEP to the doping concentration range investigated in the production simulations is further examined in Figs. 8 and 9. Because the NEP describes the potential-energy surface through local atomic environments within a cutoff radius of 6 Å, its applicability depends primarily on whether the relevant local coordination motifs are represented in the training dataset, rather than on the nominal Ce concentration of the entire simulation cell. As illustrated in Fig. 8, the random distribution of Ce dopants and the AIMD sampling generate diverse local environments, including pristine-like Al-O coordination, isolated Ce-related environments, and local regions containing multiple Ce dopants within the cutoff radius. These environments cover the principal local interaction motifs encountered in the pristine and 0 – 2% Ce-doped systems considered in the production simulations.

To provide a validation independent of the conventional training/testing partition, additional AIMD calculations are performed for pristine  $\text{Al}_2\text{O}_3$  and dilute Ce-doped  $\text{Al}_2\text{O}_3$  containing 1.9% Ce. These structures are not included in the training dataset. As shown in Fig. 9, the atomic forces predicted by the trained NEP agree well with the corresponding AIMD results, with overall force RMSE values of  $0.043 \text{ eV Å}^{-1}$  for pristine  $\text{Al}_2\text{O}_3$  and  $0.112 \text{ eV Å}^{-1}$  for the dilute Ce-doped structure. Together, Figs. 7–9 support the electronic-structure treatment used to generate the training data and the transferability of the trained NEP over the composition range examined in this work.

## 2. Convergence of the thermal transport calculations

The numerical convergence and statistical reliability of the thermal transport calculations are examined in Figs. 10–12. Figure 10 evaluates the dependence of HNEMD results on the magnitude of the external driving force. For pristine  $\text{Al}_2\text{O}_3$

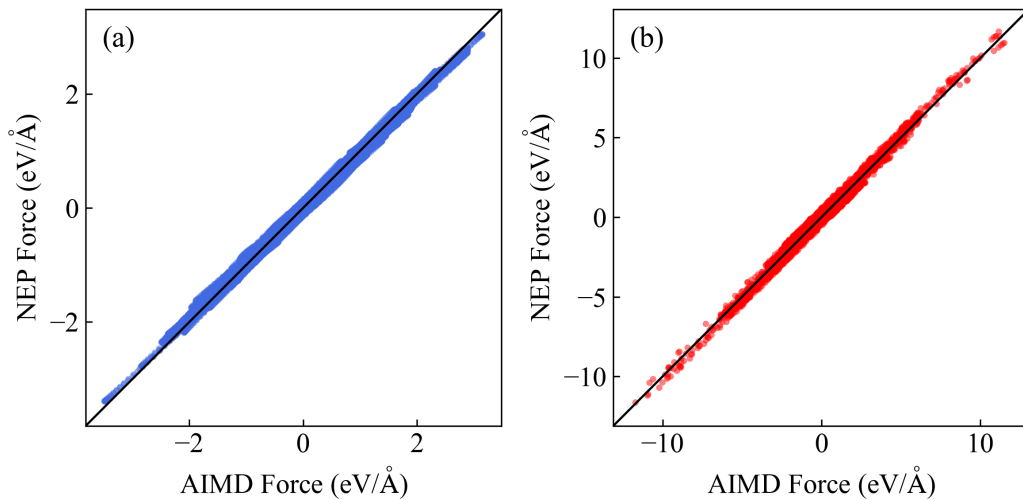


FIG. 9. Comparison of atomic forces predicted by the trained NEP model and AIMD for independent validation structures that are not included in the training dataset. (a) Pristine  $\text{Al}_2\text{O}_3$  and (b) dilute Ce-doped  $\text{Al}_2\text{O}_3$  (1.9% Ce) at 300 K. The solid line represents perfect agreement.

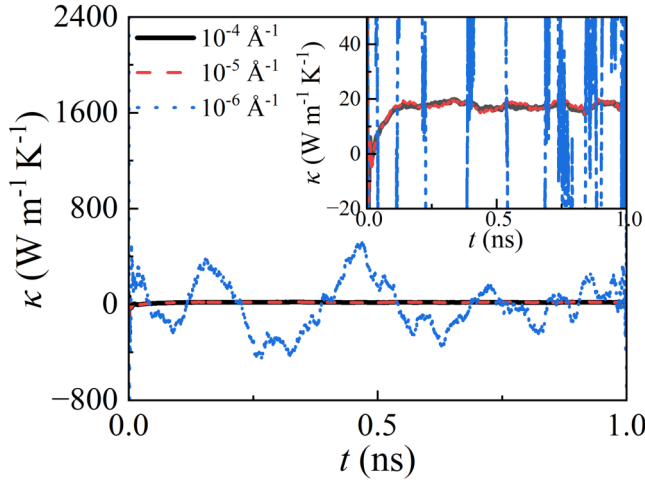


FIG. 10. Cumulative running thermal conductivity of pristine  $\text{Al}_2\text{O}_3$  at 500 K under different external HNEMD driving forces. The intermediate driving forces of  $1 \times 10^{-5}$  and  $1 \times 10^{-4} \text{ \AA}^{-1}$  give similar converged thermal conductivities, while  $1 \times 10^{-6} \text{ \AA}^{-1}$  shows large fluctuations due to the low signal-to-noise ratio. The inset shows a closed-up view around the plateau.

at 500 K, driving forces of  $1 \times 10^{-5}$  and  $1 \times 10^{-4} \text{ \AA}^{-1}$  yield similar converged thermal conductivities, indicating that the calculated response remains within the linear-response regime over this range. In contrast, the smaller driving force of  $1 \times 10^{-6} \text{ \AA}^{-1}$  produces substantially larger fluctuations because of its lower signal-to-noise ratio. On the basis of this material-specific test, an external driving force of  $1 \times 10^{-5} \text{ \AA}^{-1}$  is selected for the production simulations as a balance between linear-response behavior and statistical precision.

Figure 11 further examines convergence with respect to both simulation time and statistical sampling. As shown in Fig. 11(a), the running  $\kappa$  of pristine  $\text{Al}_2\text{O}_3$  from five inde-

pendent simulations reaches a stable plateau within the 1 ns production period, demonstrating that the production time is sufficient for the present calculations. For each Ce-doped concentration, five independently generated dopant configurations are considered, and five independent simulations are performed for each configuration, giving a total of 25 independent samples per concentration. The cumulative ensemble average shown in Fig. 11(b) gradually stabilizes as the number of samples increases, supporting the adequacy of both the configurational and statistical sampling. The convergence of the spectral  $\kappa(\omega)$  calculations is assessed independently in Fig. 12 using the normalized heat-current correlation functions. In these calculations, the heat current is sampled every 20 MD steps. With a time step of 0.5 fs and 1000 correlation steps, the maximum correlation time is 10 ps. As shown in Fig. 12, the correlation functions for both pristine and Ce-doped  $\text{Al}_2\text{O}_3$  decay within the selected 10 ps time window. Each independent trajectory contains a 1 ns production run, corresponding to a production-time-to-correlation-time ratio of 100 for each run. The use of five independent runs for pristine  $\text{Al}_2\text{O}_3$  and 25 independent simulations for each doped concentration provides additional statistical averaging. These results support the convergence of the correlation integration and the reliability of the calculated spectral  $\kappa(\omega)$ .

### 3. Uncertainty analysis

Figure 13 evaluates whether the mode-selective phonon-scattering features identified in the main text can be distinguished from statistical uncertainty. Figure 13(a) projects the reduction in  $\tau$  from pristine  $\text{Al}_2\text{O}_3$  to 0.7% Ce-doped  $\text{Al}_2\text{O}_3$  at 300 K onto the phonon dispersion, whereas Fig. 13(b) shows the corresponding uncertainty distribution. The uncertainty in the mode-resolved linewidths is generally smaller than the Ce-induced linewidth broadening, particularly for the modes exhibiting pronounced  $\tau$  suppression. This comparison

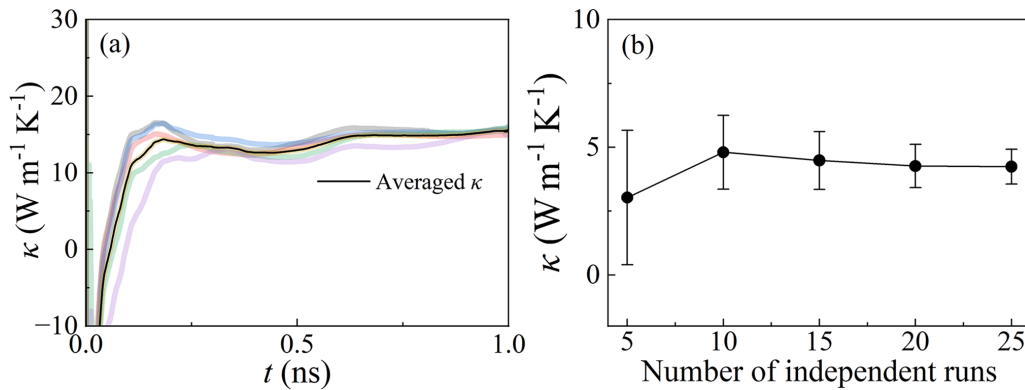


FIG. 11. Convergence tests for the thermal conductivity calculations. (a) Running thermal conductivity  $\kappa(t)$  from five independent MD runs and their ensemble average for pristine  $\text{Al}_2\text{O}_3$  at 500 K. (b) Ensemble averaged  $\kappa$  vs the number of independent samples for a representative 2% Ce-doped system at 500 K.

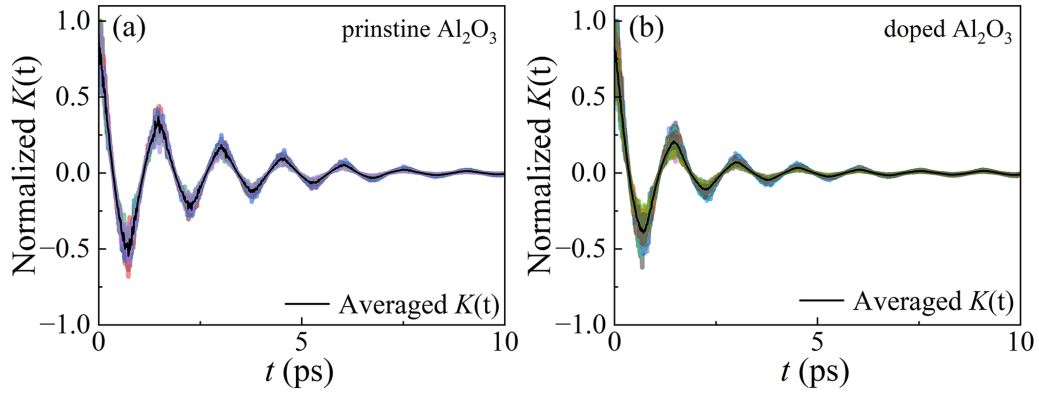


FIG. 12. Normalized heat-current autocorrelation functions used for the spectral thermal conductivity calculations at 500 K for (a) pristine  $\text{Al}_2\text{O}_3$  from five independent runs and (b) 0.7% Ce-doped  $\text{Al}_2\text{O}_3$  from five random configurations with five independent runs for each configuration. Each run is based on a 1 ns production simulation to reduce statistical noise. The correlation functions decay within the selected 10 ps correlation-time window, supporting the convergence of the calculated spectral thermal conductivity.

indicates that the mode-selective scattering pattern associated with Ce doping is not dominated by statistical noise and sup-

ports the robustness of the  $\tau$  analysis presented in the main text.

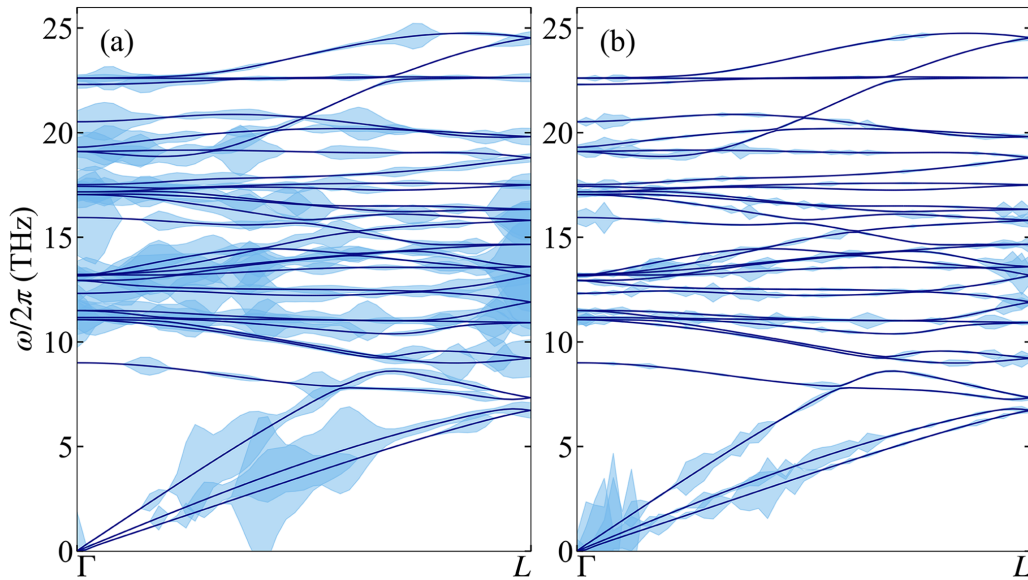


FIG. 13. Phonon dispersion of  $\text{Al}_2\text{O}_3$  along  $\Gamma \rightarrow L$  path, with the projected linewidth representing (a) the reduction in phonon lifetime from pristine to 0.7% Ce doping at 300 K and (b) the corresponding uncertainty distribution for comparison.

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