

Accurate Electron-Phonon Interactions from Advanced Density Functional Theory

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Electron-phonon coupling (EPC) is key to understanding charge transport, band renormalization, and superconductivity in energy materials, including correlated transition-metal oxides, ferroelectric perovskites, optoelectronic semiconductors, and phonon-mediated superconductors. Although first-principles density functional theory (DFT)-based EPC calculations are used widely, their predictive power is limited by the accuracy, transferability, and efficiency of the underlying exchange-correlation functionals. These limitations become exacerbated in complex *d*- and *f*-electron materials, where beyond-DFT approaches and additional corrections, such as the Hubbard *U*, are commonly invoked. Here, using the examples of CoO and NiO, we show how the *r*²SCAN density functional correctly captures strong EPC effects in transition-metal oxides without requiring the introduction of Hubbard *U* parameters. We also find that *r*²SCAN successfully describes the subtle interplay between ionic and covalent bonding, and strong EPC effects in the low-temperature rhombohedral phase of the prototypical ferroelectric BaTiO₃, without requiring Hubbard *U* and intersite *V* corrections. We further demonstrate the ability of *r*²SCAN to accurately model the EPC of the main-group semiconductor GaAs and the phonon-mediated superconducting properties of MgB₂, with reliable electronic bands and phonons. Our study establishes *r*²SCAN-based EPC as a transferable, parameter-free, and computationally efficient framework for predictive material-specific modeling of EPC in energy materials, and opens a practical route toward high-throughput screening of superconductors, thermoelectrics, optoelectronic semiconductors, and oxide electrodes and catalysts.

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I. INTRODUCTION

Electron-phonon coupling (EPC) is fundamental to understanding a wide range of phenomena in condensed-

matter physics and materials science [1,2]. EPC directly controls key properties of functional materials central to energy applications, including phonon-mediated superconductivity [3,4], electrical resistivity in metals, carrier lifetime and mobility in semiconductors, and the temperature-dependent behavior of electronic structures [5,6]. The development of Kohn-Sham density functional theory (KS DFT) has enabled the quantitative prediction of EPCs from first-principles calculations for a wide range of materials [1], typically implemented through density functional perturbation theory (DFPT) [7] or the finite-difference method [8,9].

For complex materials containing *d* and *f* electrons, DFT employing the local-density approximation (LDA) or

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generalized-gradient approximations (GGAs) often fails to accurately predict EPCs, primarily due to inaccuracies in the computed electronic structures [10–13] and phonon dispersions [14–17]. For example, the standard Perdew-Burke-Ernzerhof (PBE) GGA incorrectly finds CoO as being metallic rather than insulating, leading to dynamic instabilities characterized by imaginary phonon modes.

To address these deficiencies, the DFT + U method incorporates a Hubbard U correction for localized d or f electrons, and it has been successfully extended to EPC calculations through DFPT + U , with demonstrated success in systems such as CoO [17,18]. Although the U parameter can be derived from first-principles calculations, it is sensitive to the local chemical environment [19], complicating its practical application in materials with competing bonding characteristics. For instance, DFT + U fails to capture the correct ground-state crystal structure of BaTiO₃ (BTO) [19], a prototypical ferroelectric resulting from a delicate balance between ionicity and covalency [20]. An intersite correction V can improve agreement with experiment in such challenging cases [19] but this requires detailed analysis or chemical intuition, thereby limiting its broader applicability. It should be noted that DFT methods incorporating U and/or V corrections generally are unsuitable for main-group compounds, even when the standard PBE functional inaccurately describes critical properties such as the carrier mobility in GaAs [21], which is typically overestimated due to the underestimation of EPC matrix elements and effective masses. To our knowledge, DFT + U + V approaches have not yet been explored in EPC calculations.

Recently, more sophisticated approaches combining beyond-DFT methods such as GW or dynamical mean-field theory (DMFT) with DFPT have gained traction for EPC calculations in correlated materials [22–24]. Despite their improved handling of electronic correlations, these methods are computationally intensive and usually depend on phonon modes derived from conventional density functionals, which limits them. Development of advanced and transferable density functionals that can reliably and efficiently predict EPCs in complex materials remains a challenge.

Meta-GGAs such as the strongly constrained and appropriately normed (SCAN) [25] density functional and its variant r^2 SCAN [26], which computationally scale like LDA and GGAs, have shown promise in providing accurate descriptions of total energies and electronic structures across diverse systems [25–27]. These functionals have proven effective in materials ranging from liquid water [28] to metal oxides [29], and complex materials with d and f electrons [12,30], such as the cuprates [10,11,31] and nickelates [13,32,33]. Recent work also indicates that r^2 SCAN can accurately obtain phonon dispersions of cuprates [14,16] and NiO [15], but the performance for EPC predictions remains unexplored.

Here, we apply the r^2 SCAN meta-GGA functional within the finite-difference approach [5,6] to calculate EPC in a set of representative energy-relevant materials: transition-metal oxides CoO and NiO, which are widely explored as model systems for oxide electrodes and catalysts [34,35], where polaronic transport is governed by EPC; the low-temperature rhombohedral phase of the prototypical ferroelectric BaTiO₃, where the interplay between electrons and phonons is essential for device performance; the III–V semiconductor gallium arsenide (GaAs), an important photovoltaic and optoelectronic material for which accurate EPC is required to capture carrier mobility and hot-carrier relaxation; and the phonon-mediated superconductor magnesium diboride (MgB₂), relevant for superconducting-magnet applications. We find that r^2 SCAN accurately captures electronic structures, phonon characteristics, and strong EPC effects in CoO, NiO, and the rhombohedral phase of BTO, significantly improving over GGAs without requiring the introduction of additional parameters such as the Hubbard U and the intersite V . r^2 SCAN also achieves high accuracy in modeling the EPC of the widely studied semiconductor GaAs and the phonon-mediated superconducting properties of MgB₂, offering reliable electronic and vibrational properties. Taken together, these results establish r^2 SCAN as a powerful functional that combines accuracy, efficiency, and transferability, and enables reliable EPC predictions for materials ranging from systems with itinerant s and p electrons to those with complex d -electron correlations. Our study thus represents a significant advancement in EPC calculations and paves the way for high-throughput screening of superconductors, thermoelectrics, and other materials for energy applications.

II. RESULTS AND DISCUSSION

A. Evaluation of EPCs in CoO and NiO

CoO and NiO are two fundamental transition-metal oxides that pose challenges for DFT [12,36]. In our simulations, we use the low-temperature antiferromagnetic (AFM) phase of CoO and NiO, characterized by alternating ferromagnetic planes along the [111] direction. Upon relaxation, the cubic rock-salt structures of CoO and NiO exhibit slight distortions due to the AFM order. Experimentally, CoO and NiO are polar semiconductors, which exhibit long-range dipole-dipole interactions arising from the polarization of collective ionic movements. These interactions cause splittings between the longitudinal optical (LO) and transverse optical (TO) phonon modes at the Γ point. To account for this effect, the nonanalytic contributions needed for calculating phonon dispersions and EPCs must be included [7,37], which depend on the Born effective charges (Z^*) and the ion-clamped macroscopic dielectric tensor (ϵ^∞).

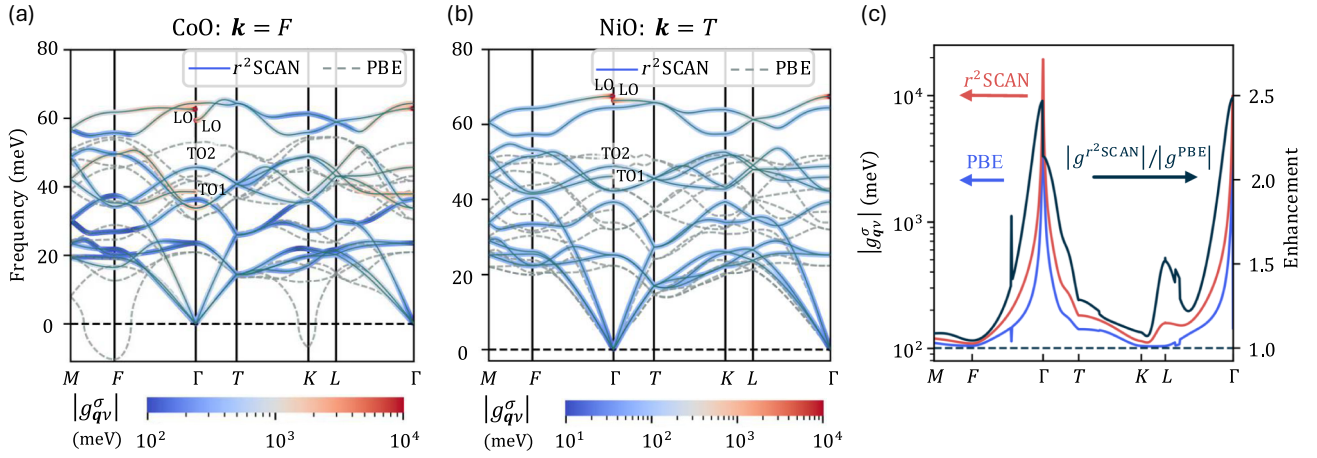


FIG. 1. Calculated phonon dispersions and electron-phonon couplings (EPCs) in CoO and NiO. (a) The phonon dispersion in G -type antiferromagnetic (AFM) CoO along the high-symmetry lines in the BZ, where the longitudinal optical (LO) and transverse optical (TO) splittings at the Γ point are marked. The color of markers along the $\mathbf{q}$ path shows the magnitude of the EPC matrix elements, calculated by summing over the bands (N_b) selected to include Co $3d$ and O $2p$ orbitals: $|g_{\mathbf{q}\nu}^{\sigma}| \equiv (\sum_{mn} |g_{mn,\nu}^{\sigma}(\mathbf{k} = \mathbf{K}, \mathbf{q})|^2 / N_b)^{1/2}$, where σ is chosen to be the spin-up channel, $\mathbf{q}$ follows the high-symmetry lines in the bulk BZ, and $\mathbf{k}$ is fixed at the K point. Note that the Perdew-Burke-Ernzerhof (PBE) EPC matrix elements are not shown for clarity. (b) The same as (a) but for NiO with $\mathbf{k} = T$. (c) A comparison of the EPC matrix elements for NiO between PBE and r^2 SCAN. The blue line represents the EPC matrix elements $|g_{\mathbf{q}\nu}^{\sigma}(\mathbf{k} = T)|$ calculated with the PBE functional, where $\mathbf{k}$ is fixed at the T point, with ν denoting the highest optical phonon mode. The red line shows the corresponding data but is calculated using the r^2 SCAN functional. The gray line indicates the ratio, $|g^{r^2\text{SCAN}}|/|g^{\text{PBE}}|$, along the high-symmetry lines in the BZ.

In Fig. 1(a), we present the phonon dispersions of CoO calculated using r^2 SCAN and PBE, along with the EPC matrix elements from r^2 SCAN. PBE predicts unphysical negative phonon frequencies along the M - F - Γ and T - K - L paths, indicating lattice instability in CoO, as reported in previous studies [17,18]. Consequently, PBE is unsuitable for calculating the EPC in CoO. The PBE + U method is often applied to eliminate these negative frequencies, allowing DFPT + U to provide accurate EPC predictions [17,18,38]. To further validate previous PBE + U calculations, we have also calculated the phonon dispersion of CoO using PBE + U (for details, see Sec. 3 of the Supplemental Material [39]). The U value of 5.9 eV for the Co $3d$ electrons used in our PBE + U calculations was obtained via the linear-response approach proposed by Cococcioni *et al.* [40]. This value is significantly larger than the 4.5 eV obtained using the DFPT method in Ref. [18], likely due to differences in the pseudopotentials employed in the Vienna *ab initio* simulation package (VASP) [41,42] and QUANTUM ESPRESSO [43–45]. No imaginary phonon frequencies are found in either our PBE + U results (see Fig. S3 in the Supplemental Material [39]) or those reported in Ref. [18]. Remarkably, r^2 SCAN resolves these lattice instabilities without requiring any parameters, as shown in Fig. 1(a). Furthermore, a comparison of phonon dispersions calculated by r^2 SCAN and PBE reveals that r^2 SCAN shifts the phonon frequencies upward, especially for the optical modes. This achieves better agreement with experimental

results, as shown in Fig. 1(a) (see also Fig. S3 in the Supplemental Material [39]).

In Fig. 1(a), our r^2 SCAN calculations reveal significant LO-TO splitting in CoO. Note that PBE does not capture this LO-TO splitting because it incorrectly predicts a metallic ground state [see Fig. S1(b) in the Supplemental Material [39]]. The predicted values of $\omega_{\text{LO}} = (59.4, 62.6)$ meV and $\omega_{\text{TO}} = (38.5, 45.7)$ meV from r^2 SCAN are close to the corresponding experimental values (see Fig. S3 in the Supplemental Material [39]). Additionally, we observe notable splitting between the TO1 and TO2 modes along the F - Γ - T direction [see Fig. 1(a)]. This splitting primarily results from the structural distortion induced by the AFM order, which breaks the equivalence along the $[111]$ directions (Γ - T), as noted in previous studies [18,46]. A similar, though smaller, LO mode splitting is seen at the Γ point. Similar results have been obtained in previous studies using PBE + U calculations [18], although the top two optical modes along the F - Γ - T direction are reversed in our r^2 SCAN results compared to earlier PBE + U calculations, likely due to the larger Born effective charge Z^* (2.25) and ion-clamped macroscopic dielectric function ϵ^{∞} (6.17) values predicted in these studies [18].

In Fig. 1(a), we also show that our r^2 SCAN calculations reveal substantial EPC contributions from both LO and TO modes near the Γ point, with maximum EPC matrix elements [g defined in Eq. (1)] exceeding 10^4 meV. This result aligns with previous DFPT + U findings [17]

TABLE I. The calculated band gap (E_g), diagonals of the Born effective charge tensor (Z^*), local magnetic moments (m), and ion-clamped macroscopic dielectric constants in the random-phase approximation (RPA) ($\epsilon_{\text{RPA}}^\infty$) and including f_{xc} (ϵ^∞) of CoO and NiO for the PBE, PBE + U , Heyd-Scuseria-Ernzerhof (HSE), and r^2 SCAN functionals, along with the experimental values.

	CoO					NiO				
	E_g (eV)	Z^*	m (μ_B)	ϵ^∞	$\epsilon_{\text{RPA}}^\infty$	E_g (eV)	Z^*	m (μ_B)	ϵ^∞	$\epsilon_{\text{RPA}}^\infty$
PBE	0	...	2.36	...	...	0.72	2.38	1.30	20.70	19.67
PBE + U	2.29	1.97	2.77	4.67	4.70	3.21	2.17	1.69	5.84	5.79
HSE	3.57	2.15	2.66	4.95	4.53	4.56	2.22	1.65	5.09	4.61
r^2 SCAN	0.85	1.83	2.58	5.48	5.44	2.30	2.23	1.58	7.52	7.10
Experiment	2.8 [50]	2.06 [51], 1.78 [52]	3.8 [53]	5.3 [52]		4.0 [54], 4.3 [55]	2.2 [56]	1.9 [57]	5.7 [52]	

quantitatively. In transition-metal oxides such as CoO and NiO, the polar nature of the bonds leads to strong interactions between the electrons and LO and TO phonons via the Fröhlich mechanism [47]. These interactions become particularly significant as the wave vector $\mathbf{q} \rightarrow 0$, causing a divergence in the EPC matrix elements for the LO and TO modes [see Fig. 1(a)]. These interactions are often substantial enough to form large polarons, which play a crucial role in charge transport and the dynamic behavior of electrons in polar materials. It is a significant advantage of r^2 SCAN that it can capture the Fröhlich interaction in polar materials without relying on additional Hubbard U parameters [17,48,49].

Phonon dispersions of NiO calculated using r^2 SCAN and PBE are shown in Fig. 1(b), where the phonon softening trend seen in going from r^2 SCAN to PBE is similar to that seen in CoO. Also, r^2 SCAN accurately captures the LO-TO mode splitting in NiO due to the inclusion of the nonanalytic term. Like CoO, r^2 SCAN calculations reveal a clear splitting of the TO1 and TO2 modes at the Γ point in NiO, although the LO mode splitting is comparatively smaller, as discussed in Ref. [46]. Notably, the TO mode splitting in NiO is smaller than in CoO [see Fig. 1(b)], which can be attributed to the smaller local magnetic moments of NiO [Table I], as pointed out in a previous study [18]. Importantly, the phonon characteristics computed with r^2 SCAN show strong consistency with those obtained using PBE + U (see Fig. S3 in the Supplemental Material [39]).

Like CoO, strong EPC contributions from the LO and TO modes in NiO are also predicted by r^2 SCAN [see Fig. 1(b)]. To illustrate the differences between PBE and r^2 SCAN in the EPC predictions, we plot the EPC matrix elements of NiO for the highest optical mode along the high-symmetry $\mathbf{q}$ path for these two functionals in Fig. 1(c). The EPC matrix elements are significantly enhanced in r^2 SCAN compared to PBE, particularly around the Γ point, with an approximate increase of 2.5, as shown in Fig. 1(c).

By considering the Fan-Migdal (FM) self-energy [Eqs. (12) and (13)], we have calculated the spectral function of the valence-band maximum (VBM) for both CoO and NiO,

as shown in Figs. 2(b) and 2(d), respectively. The results clearly reveal strong renormalization of the band structure in both materials: the original single-particle bands become significantly broadened. Note that more accurate spectral functions will require going beyond the one-shot FM approximation [58]. Such advanced calculations are beyond the scope of the present work. In addition, we estimate the zero-point renormalization (Δ_{ZPR}) of the band gap at the Γ point for NiO due to EPC at 0 K, using the one-shot special-displacement method [59,60]. The calculated Δ_{ZPR} values from PBE, PBE + U , and r^2 SCAN are -0.13 , -0.19 , and -0.25 eV, respectively. Notably, the larger magnitude of Δ_{ZPR} predicted by r^2 SCAN compared to PBE is consistent with the stronger g -matrix elements obtained from r^2 SCAN, as shown in Fig. 1(c). The pronounced spectral broadening and large Δ_{ZPR} are hallmarks of strong EPC and manifest as a redistribution of spectral weight, reflecting enhanced electron scattering by phonons. Our findings suggest that EPC plays a crucial role in shaping the transport and optical properties of correlated oxides.

B. Why does r^2 SCAN yield superior EPC predictions in CoO and NiO?

The preceding results demonstrate that r^2 SCAN significantly improves the accuracy of the EPC descriptions of CoO and NiO compared to PBE. Our previous analysis indicates that the superior performance of r^2 SCAN over PBE can largely be attributed to the more exact constraints that r^2 SCAN satisfies [25–27]. As a result, for transition- and rare-earth-metal compounds, r^2 SCAN exhibits reduced self-interaction error (SIE) compared to PBE, significantly improving the description of the electronic structures (Fig. 2; see also Fig. S1 in the Supplemental Material [39]) [30,31]. The significant improvement of r^2 SCAN over PBE in describing the phonon characteristics of NiO, particularly for the optical modes, has also been attributed to the reduction of the SIE [15]. SIE arises from the imperfect cancellation of the spurious classical Coulomb self-interaction by the approximate exchange-correlation self-interaction [61]. Since

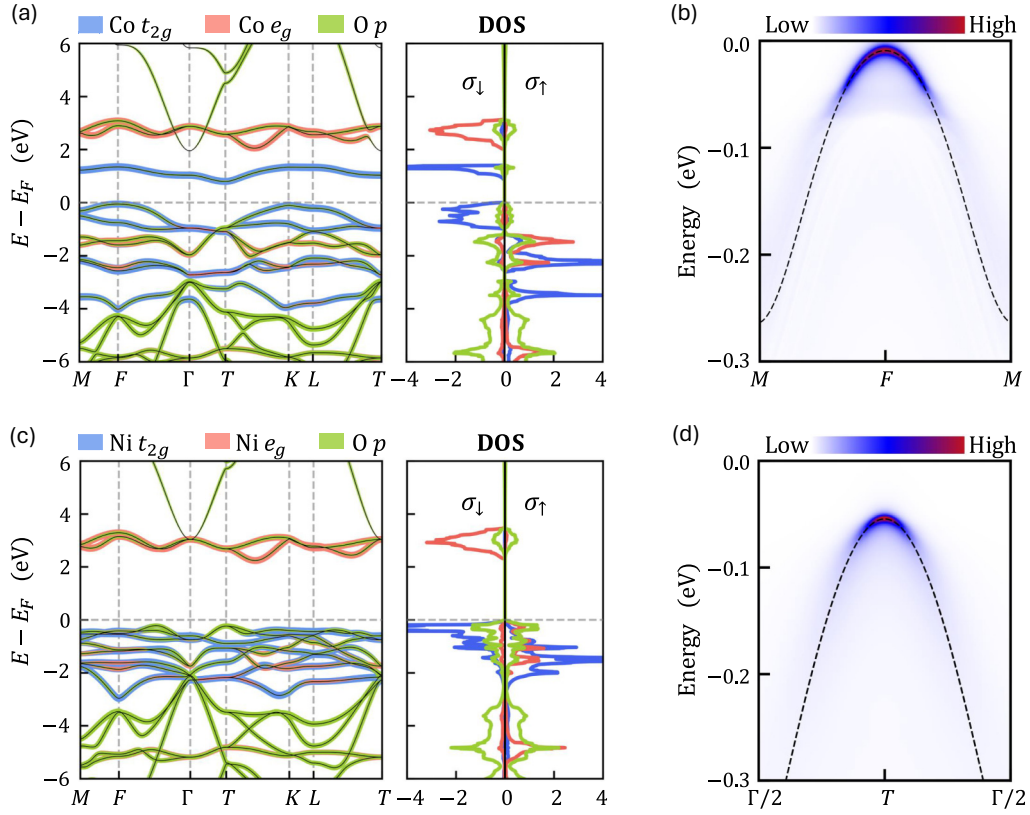


FIG. 2. The calculated electronic structure of the G -type AFM phase of CoO and NiO using the r^2 SCAN functional. (a) The orbital-projected band structure of CoO along the high-symmetry lines in the BZ, where the Co t_{2g} and e_g , and O p orbitals are marked with blue, red, and green colors, respectively. The side panel shows the partial density of states of the respective orbitals. (b) The calculated spectral function of the valence-band maximum (VBM) of CoO. The zero of the energy is set to the DFT-calculated Fermi level. The dashed blue line represents the bare DFT bands. (c) The same as (a) but for NiO. (d) The same as (b) but for NiO.

the repulsive self-Coulomb term exceeds the attractive self-exchange-correlation term, the net SIE is generally positive for semilocal functionals, causing orbitals to be underbound (orbital energies are too high) and wave functions to be excessively delocalized. This leads to electron densities that are overly spread out and easily perturbed, ultimately resulting in underestimated magnetic moments and band gaps. The reduction of the SIE in r^2 SCAN compared to PBE thus results in less delocalization error, more compact d and f orbitals, and fewer fractional occupations, which stabilize the magnetic moments [12,30] and increase the band gaps in NiO and CoO (see Table I).

In Table I, we also present the calculated Z^* and ϵ^∞ values for CoO and NiO based on various functionals, including PBE, PBE + U , Heyd-Scuseria-Ernzerhof (HSE), and r^2 SCAN, along with the experimental values. Both Z^* and ϵ^∞ are critical quantities used in the nonanalytic contributions needed for calculating phonon dispersions and EPCs. The Born effective charge Z^* measures the dynamic charge of an atom in a crystal, capturing how polarization changes in response to the atomic displacement and reflecting the redistribution of electronic charge under an applied electric

field [62–65]. The ion-clamped macroscopic dielectric tensor ϵ^∞ describes the long-range dielectric response of a material, considering only the electronic contribution without accounting for the ionic motion, reflecting how the electronic cloud in a crystal responds to an external electric field [62–65]. Z^* and ϵ^∞ are expected to be overestimated due to the SIE, which leads to overly delocalized and more easily perturbed electron densities. This can be seen by comparing the Z^* and ϵ^∞ values calculated from r^2 SCAN and PBE against the experimental data. For CoO, the r^2 SCAN predictions for Z^* and ϵ^∞ are in good agreement with experimental results [51,52]. However, PBE incorrectly predicts a metallic ground state for CoO, as shown in Fig. S1(b) in the Supplemental Material [39], resulting in the vanishing of the nonanalytic terms. In NiO, PBE overestimates ϵ^∞ by a factor of 3.77, whereas r^2 SCAN provides significant improvement by reducing the overestimation to around 30%. The calculated Z^* value using r^2 SCAN is 2.23, which is in excellent agreement with the experimental value of 2.2, while PBE overestimates Z^* by 8%. These results corroborate that r^2 SCAN reduces SIE in comparison with PBE in NiO and CoO.

We also find that the band gaps of CoO and NiO calculated using PBE + U are in close agreement with corresponding experimental values (see Table I). However, the ion-clamped macroscopic dielectric constant ϵ^∞ predicted by PBE + U for CoO is significantly underestimated relative to the experimental value. Although r^2 SCAN still underestimates the band gaps of CoO and NiO, it provides a substantial improvement in the predicted dielectric constants. In contrast, the nonlocal HSE functional overestimates the band gaps of both CoO and NiO, which in turn leads to an underestimation of ϵ^∞ .

The preceding analysis strongly indicates that the improvement of r^2 SCAN over PBE on EPC matrix elements originates from the SIE reduction. In the context of the KS DFT [1], the EPC matrix element is defined as

$$g_{mn,v}(\mathbf{k}, \mathbf{q}) = \langle \psi_{m\mathbf{k}+\mathbf{q}} | \partial_{\mathbf{q}v} v^{\text{KS}} | \psi_{n\mathbf{k}} \rangle, \quad (1)$$

where $\psi_{n\mathbf{k}}$ is the electronic orbital with the momentum $\mathbf{k}$ in the band n and $\partial_{\mathbf{q}v} v^{\text{KS}}$ is the derivative of the self-consistent KS potential with respect to a phonon of wave vector $\mathbf{q}$, band index v , and frequency $\omega_{\mathbf{q}v}$. Note that $\partial_{\mathbf{q}v} v^{\text{KS}}$ is calculated by the finite-difference approach here. By definition, $\partial_{\mathbf{q}v} v^{\text{KS}}$ is given by

$$\partial_{\mathbf{q}v} v^{\text{KS}} = \partial_{\mathbf{q}v} v^{\text{en}} + \partial_{\mathbf{q}v} v^H + \partial_{\mathbf{q}v} v^{\text{xc}}, \quad (2)$$

where v^{en} is the potential arising from electron-nuclei interactions, v^H is the Hartree potential, and v^{xc} is the exchange-correlation potential. This can be recast as [66]

$$\partial_{\mathbf{q}v} v^{\text{KS}} = (\epsilon^{\text{Hxc}})^{-1} \partial_{\mathbf{q}v} v^{\text{en}}. \quad (3)$$

Here, we have used the definition of the “test electron” dielectric matrix,

$$\epsilon^{\text{Hxc}} = 1 - (v^C + f^{\text{xc}}) \chi^0, \quad (4)$$

where v^C is the Coulomb kernel, $f^{\text{xc}}(\mathbf{r}, \mathbf{r}') \equiv \delta^2 E_{\text{xc}}[n] / \delta n(\mathbf{r}) \delta n(\mathbf{r}')$ is the exchange-correlation kernel, and $\chi^0(\mathbf{r}, \mathbf{r}') \equiv \delta n(\mathbf{r}) / \delta v^{\text{KS}}(\mathbf{r}')$ is the KS noninteracting linear-response function.

Equations (3) and (4) show that the change in the KS potential due to the motion of the nuclei originates from two contributions: (i) the change in the bare electron-nuclei potential and (ii) the response of the electronic degrees of freedom to screen these interactions at the density functional approximation (DFA) level. Thus, improvements of EPC matrix elements within the KS-DFT framework would come from two key contributions: χ_0 and f^{xc} .

The ion-clamped macroscopic dielectric constants ϵ^∞ are related to the head ($\mathbf{q} \rightarrow 0$) of ϵ^{Hxc} [67]. For a given density functional, ϵ^∞ can be calculated using the random-phase approximation (RPA), where f^{xc} is ignored, with the value denoted as $\epsilon_{\text{RPA}}^\infty$ [67,68]. In Table I, we show that the

difference between $\epsilon_{\text{RPA}}^\infty$ and ϵ^∞ of a density functional is much smaller than the difference between ϵ^∞ of different density functionals, suggesting that f^{xc} has a less important effect on ϵ^∞ than the χ_0 effect of the chosen density functional. We note that r^2 SCAN satisfies more exact constraints than PBE does with more accurate total energies in general, suggesting that r^2 SCAN can have a better f^{xc} [69].

The KS linear-response function χ_0 describes how the electron density n reacts to small changes in the KS potential v^{KS} . This linear-response function is often overestimated in magnitude as a result of SIE, which leads to an overly delocalized and easily perturbable electron density, as well as an underestimated band gap that is inversely proportional to the linear-response function.

In CoO, the failure of PBE to open a band gap [see Fig. S1(b) in the Supplemental Material [39]] leads to too large χ_0 , resulting in dynamical instabilities with the appearance of imaginary frequencies in the phonon calculations, as shown in Fig. 1(a). In contrast, due to the SIE reduction, r^2 SCAN predicts an indirect band gap of 0.85 eV, attributed to the splitting between the occupied t_{2g} orbitals and the unoccupied t_{2g} orbitals in the spin-down channel, as shown in Fig. 2(a). This undoubtedly improves χ_0 and thus provides an improved phonon dispersion as well as strong EPC effects for the LO and TO phonon modes around the Γ point, as shown in Fig. 1(a). Since a smaller gap generally correlates with a larger ϵ^∞ [70,71], the improved gaps from r^2 SCAN explain why our values of ϵ^∞ are closer to the experimental results (see Table I).

In NiO, r^2 SCAN predicts an indirect band gap of 2.30 eV, with the VBM at the T point and the conduction-band minimum (CBM) along the T - K direction. This value is significantly larger than the 0.72 eV gap predicted by PBE [see Fig. S1(d) in the Supplemental Material [39]], yet still smaller than the experimental estimates of 4.0–4.3 eV (see Table I). PBE significantly underestimates the band gap, yielding a larger χ_0 , which, in turn, produces smaller EPC matrix elements compared to r^2 SCAN [see Fig. 1(c)]. We also note that compared to the experimental values, the HSE hybrid functional overestimates considerably the band gaps of both CoO and NiO, which consequently leads to underestimations of ϵ^∞ , as shown in Table I.

Accurate EPC predictions can be expected within the KS-DFT framework if the SIE in the exchange-correlation approximation is minimized. This can be achieved by advancing up the Jacob’s ladder of DFT [72], moving from LDA to GGA, to meta-GGA, and then to the hybrid density functionals, the Perdew-Zunger self-interaction correction (PZ SIC) [61], etc., or by applying addition corrections to a chosen functional, such as DFT + U [73,74]. The HSE hybrid density functional [75] has become a standard for calculating band gaps in solids due to its reduced SIE. However, the computational cost of HSE is often 1–2 orders of magnitude higher than LDA, GGA, and

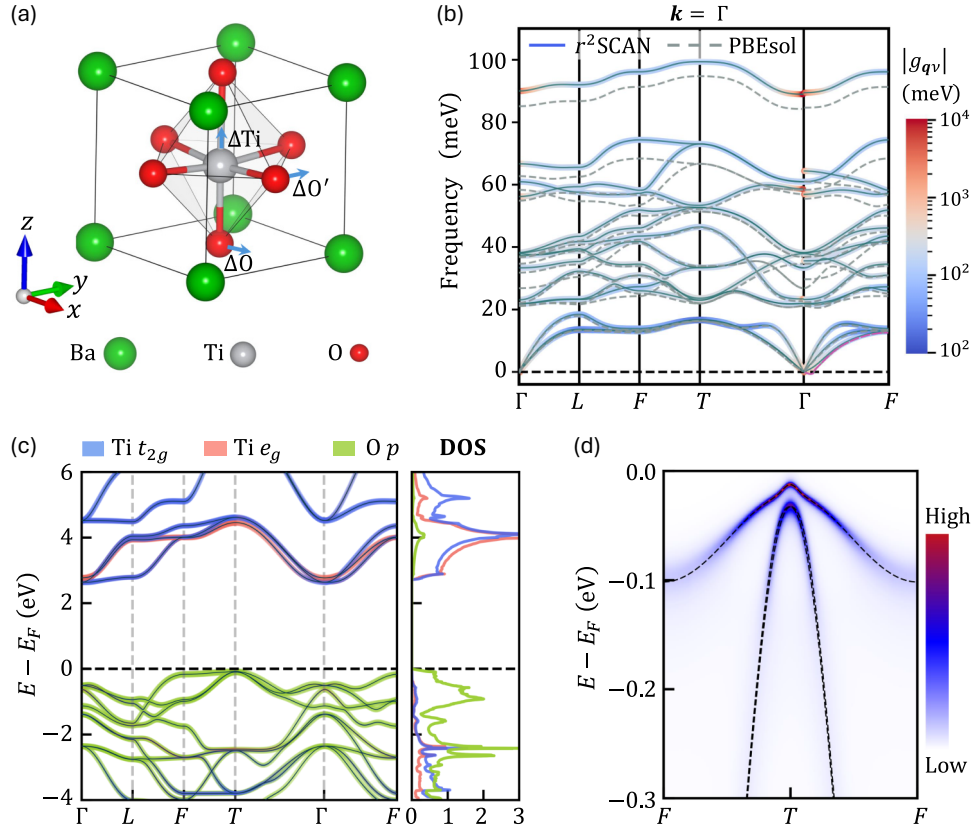


FIG. 3. The crystal structure, calculated electronic structure, phonon dispersion, and EPCs of BaTiO₃. (a) The crystal structure of rhombohedral BaTiO₃. The green, gray, and red balls represent Ba, Ti, and O atoms, respectively. The light-blue arrows indicate atomic displacements relative to the cubic phase. (b) A comparison of the phonon dispersions calculated using r^2 SCAN (solid blue line) and PBEsol (dashed gray line) along the high-symmetry lines in the BZ. The acoustic mode with imaginary frequencies along the Γ - F path in the PBEsol results is highlighted in purple. The color of the markers along the $\mathbf{q}$ path shows the magnitude of the EPC matrix elements, calculated by summing over bands (N_b) selected to include Ti $3d$ and O $2p$ orbitals: $|g_{qv}| \equiv (\sum_{mn} |g_{mn,v}^{\sigma}(\mathbf{k} = K, \mathbf{q})|^2 / N_b)^{1/2}$, where $\mathbf{q}$ follows the high-symmetry lines in the bulk BZ and $\mathbf{k}$ is fixed at the Γ point. (c) The orbital-resolved band structure of rhombohedral BaTiO₃ along the high-symmetry lines in the BZ from r^2 SCAN (left panel), where the O p , Ti t_{2g} , and e_g orbitals are indicated in green, blue, and red, respectively. The associated partial density of states for each orbital is shown in the right panel. (d) The calculated spectral function of the VBM of BaTiO₃ along the F - T - F line. The zero energy is set to the DFT-calculated Fermi level. The dashed black line represents the bare DFT band.

meta-GGA functionals, making it impractical for our current EPC calculations. DFT + U also offers a reasonable prediction of the band gap, Z^* , and ϵ^∞ for CoO and NiO. Nevertheless, in practice, the U parameter generally depends on both the specific material and the property of interest, and it is sensitive to the local chemical environment [19,73,74], as will be discussed in the following section.

C. Evaluation of EPC in the rhombohedral phase of BaTiO₃

In the above sections, the challenges of accurately modeling transition-metal compounds with density functionals have been exemplified by NiO and CoO, where the main difficulty lies in treating localized d electrons. Applying a U correction or using advanced functionals such as r^2 SCAN to reduce SIE significantly improves

their description. However, further complexity arises in transition-metal compounds from competing bonding characteristics with pd hybridization in, e.g., the prototypical ferroelectric BTO, which undergoes three ferroelectric phase transitions—from cubic to tetragonal, orthorhombic, and finally the rhombohedral phases—as the temperature decreases [76], where the spontaneous symmetry breaking results from a delicate balance between ionic and covalent bonding [20]. In Ref. [19], it is shown that PBEsol + U , where PBEsol is the revised version of PBE for solids [77], fails to capture the correct ground-state rhombohedral phase of BTO, and needs an additional intersite correction V to capture the delicate balance between ionicity and covalency. In this section, we examine the rhombohedral phase of BTO, with polarization along the [111] direction. We show that the r^2 SCAN functional describes the EPC, as well as the geometrical and electronic structure and

TABLE II. The calculated lattice parameter a , rhombohedral angle α , and the displacements of Ti and O atoms relative to the cubic phase illustrated in Fig. 3(a). Here, Δ_{Ti} denotes the displacement of the Ti atom along the z direction, while Δ_{O} and $\Delta_{\text{O}'}$ represent the displacements of O atoms along the x and y directions, respectively. Also given are the structural data obtained from previous PBEsol + U and PBE + $U + V$ calculations [19], along with experimental data.

	a (Å)	α (degrees)	Δ_{Ti}	Δ_{O}	$\Delta_{\text{O}'}$
PBEsol	4.007	89.86	−0.012	0.011	0.018
PBEsol + U	4.024	90	0.000	0.000	0.000
PBEsol + U [19]	3.990	90	0.000	0.000	0.000
PBEsol + $U + V$ [19]	4.017	89.77	−0.014	0.012	0.020
r^2 SCAN	4.036	89.82	−0.014	0.011	0.021
Experiment [78]	4.004	89.84	−0.013	0.011	0.019

phonon behavior of BTO, without the need for any free parameters.

As illustrated in Fig. 3(a), the rhombohedral phase of BTO exhibits spontaneous polarization along the [111] direction, with atomic displacements shown relative to the nonpolar cubic phase. In Table II, we summarize the structural parameters and atomic displacements of Ti and O calculated with various DFAs, alongside experimental data [78] and prior studies [19]. Although PBEsol yields structural parameters closely matching experimental values, it predicts imaginary phonon frequencies near the Γ point [see Fig. 3(b)], indicating dynamic instability. Additionally, PBEsol significantly underestimates the band gap compared to experiments with an exaggerated dielectric constant (see Table III), suggesting that its EPC prediction for BTO is underestimated (Sec. II B).

In this connection, Gebreyesus *et al.* have demonstrated that a Hubbard U parameter alone is not adequate to capture the ground state of BaTiO₃ and that an additional intersite V correction [19] is required; they have reported that PBEsol + U with a self-consistent $U = 5.21$ eV erroneously predicts a cubic ground state. We obtain a similar result by applying the same U to the Ti 3d states (see Table II). This failure arises because applying U exclusively to the Ti 3d orbitals increases their ionic character and reduces Ti-O hybridization, favoring the cubic phase. The inclusion of intersite V restores the correct rhombohedral ground state, with lattice constants and the band gap closely matching experimental values (see Table III), significantly improving dynamical stability, though minor

instabilities persist near the Γ point [19]. r^2 SCAN removes these instabilities without requiring Hubbard U or the intersite V correction. In Table II we show that r^2 SCAN also yields lattice constants and atomic displacements in good accord with the experimental data. Compared to PBEsol, r^2 SCAN corrects unstable modes near the Γ point, predicting similar acoustic branches but notably harder optical modes [see Fig. 3(b)]. The inclusion of nonanalytic corrections reveals clear LO-TO splitting at the Γ point, as in CoO and NiO (Sec. II A). The predicted Born effective charges for Ti and O sites from various DFAs significantly exceed nominal ionic values, which can be attributed to O 2p-Ti 3d hybridization [80]. Strong EPC between the electrons and the LO and TO phonon modes, driven by the Fröhlich mechanism, is observed with EPC matrix elements exceeding 10^4 meV, similar to CoO and NiO.

The electronic structure, presented in Fig. 3(c), shows that the VBM is primarily composed of O 2p states, while the CBM is mainly derived from Ti 3d states. Notably, flat bands are observed near the VBM and CBM along the F - T and Γ - L directions, indicating the potential for strong EPC in the rhombohedral phase of BTO. r^2 SCAN predicts an indirect band gap of 2.72 eV. Although this is still smaller than the experimental value of 3.4 eV, it represents a marked improvement over the PBEsol result of 2.24 eV (see Table III). The dielectric constant predicted by r^2 SCAN is 5.29, which closely matches the experimental value of 5.20. These results strongly indicate that r^2 SCAN can accurately capture the EPC in BTO

TABLE III. The calculated band gap (E_g), diagonals of the Born effective charge tensor (Z^*), and ion-clamped macroscopic dielectric constants (ϵ^∞) for rhombohedral BaTiO₃ using various DFAs, along with the measured experimental values.

	E_g (eV)	ϵ_{xx}^∞	ϵ_{yy}^∞	$\langle\epsilon\rangle$	Z_{Ba}^{*xx}	Z_{Ba}^{*zz}	Z_{Ti}^{*xx}	Z_{Ti}^{*zz}	$Z_{\text{O}\parallel}^*$	$Z_{\text{O}\perp}^*$
PBEsol	2.24	6.20	5.71	5.96	2.78	2.74	6.62	5.66	−5.12	−1.97
PBEsol + U	2.29	5.95	5.95	5.95	2.75	2.75	6.51	6.51	−5.19	−2.04
PBEsol + U [19]	2.26	5.88	5.88	5.88	2.77	2.77	6.57	6.56	−5.20	−2.07
PBEsol + $U + V$ [19]	3.46	5.54	5.00	5.36	2.80	2.74	6.18	5.08	−4.47	−1.95
r^2 SCAN	2.72	5.53	5.05	5.29	2.75	2.70	6.43	5.41	−4.95	−1.94
Experiment	3.4 [19]	...	...	5.20 [79]						

(see also Sec. II B). Notably, the calculated spectral function of the VBM reveals pronounced band-renormalization effects with significant broadening [see Fig. 3(d)], compared to the bare DFT bands. These features are clear signatures of EPC-induced many-body interactions and offer direct evidence of strong EPC in the rhombohedral phase of BTO, particularly in the low-energy electronic states near the Fermi level. The improved performance of r^2 SCAN over PBEsol and PBEsol + U in describing BTO arises primarily from its enhanced capability to identify and appropriately treat different chemical environments [25,27,81]. This capability stems from the incorporation of the kinetic energy density into meta-GGAs such as SCAN and r^2 SCAN [25,26,81], an ingredient absent in the GGAs such as PBE and PBEsol exchange-correlation functionals.

D. Evaluation of the EPC in GaAs

As a widely used semiconductor in electronic and optoelectronic devices, GaAs has attracted significant experimental and theoretical interest. EPC plays an essential role in understanding its electronic mobility and band-structure renormalization [21,49,82]. Recently, Zhong *et al.* have reported that electron mobility predicted by PBE is overestimated by approximately a factor of 3 compared to the experimental values, primarily due to underestimation of the g -matrix elements and effective mass [21]. In this section, we present an in-depth comparative study of EPC in GaAs with r^2 SCAN and PBE and show that the electronic and phonon structures obtained with r^2 SCAN are substantially improved over PBE, leading to a more accurate description of EPC effects in GaAs.

In Table IV, we show that the optimized PBE-based lattice constant is 5.763 Å, which is 1.89% larger than the experimental value of 5.656 Å. In contrast, the lattice constant predicted by r^2 SCAN is 5.672 Å, in good agreement with the experiment. In Fig. 4(a), we compare the band structures calculated with r^2 SCAN and PBE at the respective theoretical lattice constants. Both functionals yield similar orbital characteristics for the VBM and CBM: the VBM is primarily composed of As 4 p orbitals, while the CBM arises from strong hybridization between the Ga 4 s and 4 p orbitals. However, with the relaxed structure from PBE, the calculated band gap is

only 0.15 eV—significantly smaller than the experimental value of 1.42 eV [84] (see Table IV). Note that when using the experimental lattice constant, PBE predicts a slightly larger band gap of approximately 0.5 eV. The ion-clamped macroscopic dielectric function ϵ^∞ predicted by PBE is 16.77, which is substantially greater than the experimental value of 10.89 [85]. Although the PBE + U + V is often used to reduce SIE in the standard DFT, this approach is rarely applied to main-group semiconductors such as GaAs. Timrov and Cococcioni have shown that improvements in the electronic structure of GaAs using PBE + U + V are negligible compared to PBE [88].

Interestingly, without any free parameters, r^2 SCAN significantly improves over PBE the band-gap prediction in GaAs to 0.93 eV, which is partly a result of the smaller and more correct lattice constant. It can also be attributed to the partial incorporation by r^2 SCAN of the derivative discontinuity of the total energy with respect to the electron number in its effective potential—a feature of the exact functional that is explicitly accomplished in orbital-dependent functionals such as r^2 SCAN when implemented in the generalized KS scheme [89–93]. The calculated ion-clamped macroscopic dielectric function ϵ^∞ and Born effective charge Z^* calculated with r^2 SCAN are 11.64 and 2.15, both in close agreement with the experimental values of 10.89 [85] and 2.18 [86], respectively. These improvements strongly suggest that EPC predictions using r^2 SCAN are more accurate than those from PBE (see also Sec. II B).

In Fig. 4(c), we compare the calculated phonon dispersion of GaAs obtained using PBE and r^2 SCAN, along with experimental data. Notably, the phonon dispersion predicted by r^2 SCAN shows excellent agreement with experimental data for both the acoustic and optical branches, accurately capturing the phonon frequencies across the full BZ. In contrast, the PBE exhibits noticeable softening, particularly in the optical modes, due to the tendency of PBE to overestimate bond lengths and dielectric constants, as previously discussed in Ref. [15]. Strong g -matrix elements are found in the highest optical mode near the Γ point due to the Fröhlich mechanism [47]. Interestingly, our results show that the g -matrix elements for the acoustic modes in GaAs are generally larger than those for the optical modes, suggesting that electrons in

TABLE IV. The calculated lattice constant (a), band gap (E_g), ion-clamped macroscopic dielectric constant (ϵ^∞), Born effective charge (Z^*), and electron effective masses (m_e^*) of GaAs from the PBE and r^2 SCAN functionals, along with the experimental values.

	PBE	r^2 SCAN	Experiment
a (Å)	5.763	5.672	5.656 [83]
E_g (eV)	0.15	0.93	1.42 [84]
ϵ^∞	16.77	11.64	10.89 [85]
Z^*	2.29	2.15	2.18 [86]
$m_e^*(m_0)$ Γ - K	0.014	0.068	0.067 [87]
$m_e^*(m_0)$ Γ - L	0.012	0.063	

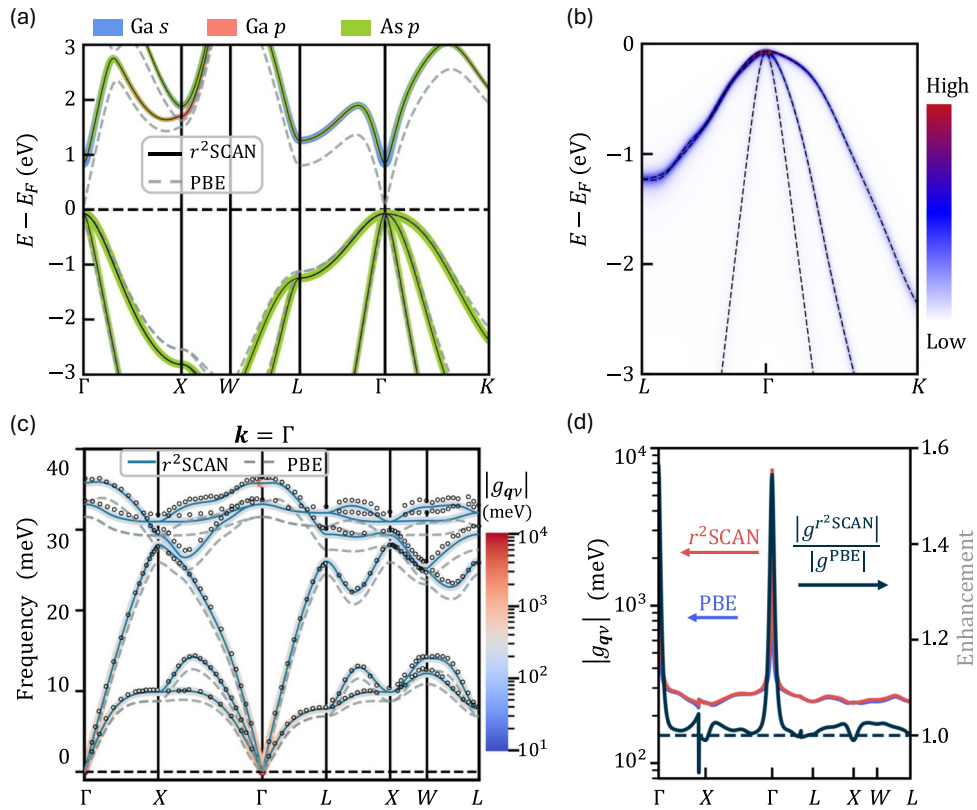


FIG. 4. The calculated electronic structure, phonon dispersion, and EPCs of GaAs. (a) The orbital-resolved band structure of cubic GaAs along the high-symmetry lines in the BZ from r^2 SCAN calculations, where Ga p and s , and the As p orbitals, are indicated in blue, red, and green, respectively. PBE results are also presented using dashed gray lines for comparison. (b) The calculated spectral function of the VBM of GaAs along the L - Γ - K line. The zero energy is set to the DFT-calculated Fermi level. The dashed black line represents the bare DFT band. (c) A comparison of the phonon dispersions calculated using r^2 SCAN (solid blue line) and PBE (dashed gray line) along the high-symmetry lines in the BZ. The color of markers along the $\mathbf{q}$ path shows the magnitude of the EPC matrix elements from r^2 SCAN, calculated by summing over the bands (N_b) selected to include Ga $4s$ and p , and As $4p$ orbitals: $|g_{qv}| \equiv (\sum_{mn} |g_{mn,v}^{\sigma}(\mathbf{k} = \Gamma, \mathbf{q})|^2 / N_b)^{1/2}$. (d) A comparison of the EPC matrix elements for the highest optical mode of GaAs between the PBE and r^2 SCAN. The blue line represents the EPC matrix elements $|g_{qv}^{\sigma}(\mathbf{k} = T)|$ calculated with the PBE functional, where $\mathbf{k}$ is fixed at the Γ point, with v denoting the highest optical phonon mode. The red line shows the corresponding results using the r^2 SCAN functional. The gray line indicates the ratio, $|g^{r^2\text{SCAN}}|/|g^{\text{PBE}}|$, along the high-symmetry lines in the BZ.

GaAs experience strong scattering from long-wavelength low-energy lattice vibrations, which could reduce electron mobility. In Fig. 4(d), we show that r^2 SCAN predicts stronger EPC elements than PBE for the highest optical mode along the high-symmetry lines, especially near Γ . In addition, the electron effective mass predicted by r^2 SCAN along the Γ - K direction is 0.068, in excellent agreement with the experimental value of 0.067, and notably larger than the PBE prediction of 0.014 (see Table IV). Taken together, these enhanced g -matrix elements and effective mass from r^2 SCAN are expected to reduce the electron mobility toward the experimental value in comparison to PBE [21]. Note that previous studies have shown that the predicted phonon dispersion and mobility of GaAs from LDA agree well with experimental data [94,95]; however, the accuracy of LDA in phonon dispersion primarily results from accidental error cancellation [15].

In Fig. 4(b), we show a modest band renormalization in GaAs with the inclusion of the FM self-energy. Unlike the strong band flattening seen in CoO, NiO, and BTO, no pronounced broadening appears near the VBM of GaAs, reinforcing its character as a weakly polar semiconductor. It is clear that r^2 SCAN provides a significant improvement over PBE for the structural, electronic, vibrational, and EPC properties of GaAs, which play key roles in determining the carrier mobility and optical properties of GaAs [21,49,59,60,82].

E. Performance of r^2 SCAN in the conventional superconductor MgB_2

Having examined the improvements that r^2 SCAN offers for predicting EPCs in complex compounds such as CoO, NiO, and BTO with d electrons, as well as GaAs, we now

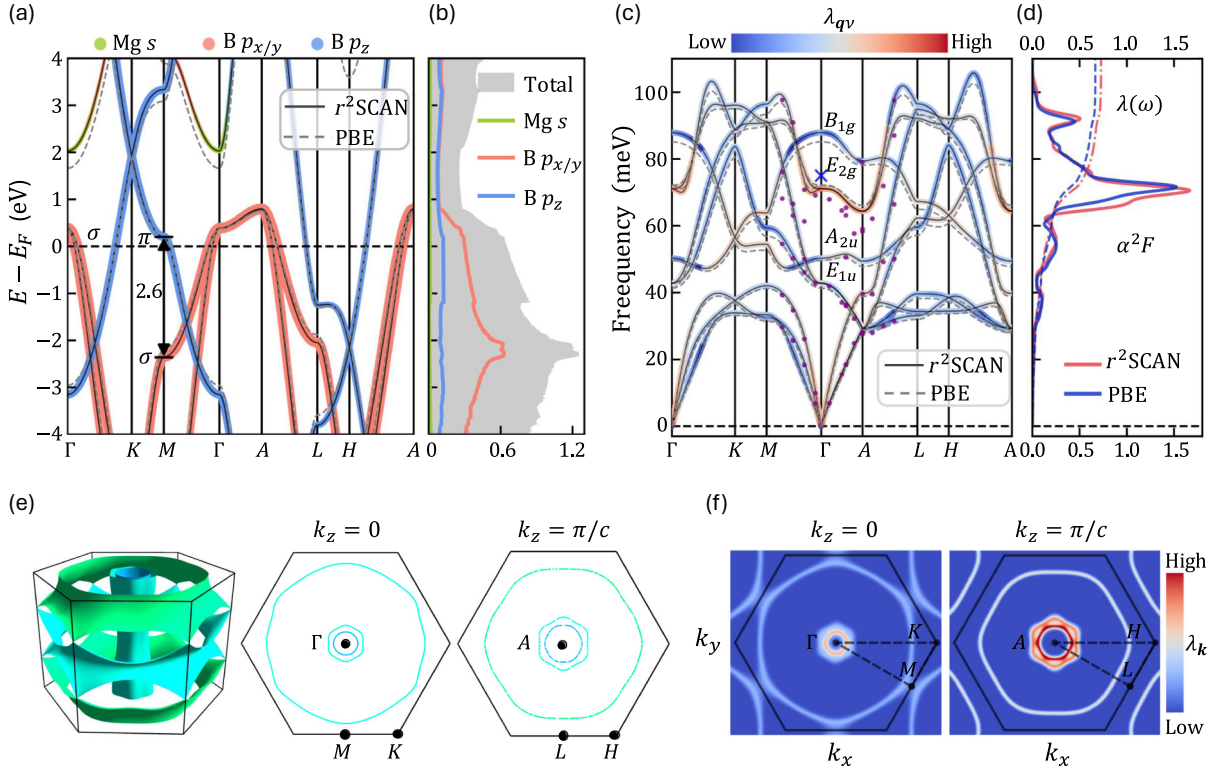


FIG. 5. The calculated electronic structure, phonon dispersion, and EPC of MgB_2 . (a) The band structure along the high-symmetry lines in the BZ from $r^2\text{SCAN}$ (solid black lines) and PBE (dashed gray lines). Orbital characters from $r^2\text{SCAN}$ are also shown, where the Mg s , B $p_{x/y}$, and B p_z orbitals are indicated in green, red, and blue, respectively. The splitting of π and σ bands from $r^2\text{SCAN}$ at the M point is highlighted. Orbital projections from PBE are omitted for clarity. (b) The total and partial density of states for various orbitals using $r^2\text{SCAN}$. (c) The phonon dispersion along the high-symmetry lines in the BZ, comparing $r^2\text{SCAN}$ (solid black lines) and PBE (dashed gray lines). The intensity of the marker color corresponds to the magnitude of the EPC strength, λ_{qv} , from $r^2\text{SCAN}$ calculations. EPC strength λ_{qv} values from PBE calculations are not shown. Experimental phonon data (solid magenta points) are extracted from Ref. [100]. The measured frequency of the E_{2g} mode at Γ from Raman spectroscopy (blue cross) is 75 meV [100,106,107]. (d) The calculated Eliashberg spectral functions $\alpha^2F(\omega)$ and cumulative EPC strength λ using $r^2\text{SCAN}$ (red lines) and PBE (green lines). A k mesh of $100 \times 100 \times 100$ and a q mesh of $50 \times 50 \times 50$ have been used for BZ sampling. (e) Left panel: the calculated three-dimensional Fermi surface obtained by using a Wannier model based on $r^2\text{SCAN}$ with a $200 \times 200 \times 200$ k mesh. Note that k_z ranges from $-\pi/c$ to π/c . Middle and right panels: Fermi-surface projections on the $k_z = 0$ and $k_z = \pi/c$ planes. (f) The distribution of k -resolved EPC strength $\lambda_{\mathbf{k}} = \sum_{mn,v\mathbf{q}} |g_{mn,v}(\mathbf{k}, \mathbf{q})|^2 \delta(\varepsilon_{m\mathbf{k}+\mathbf{q}} - \varepsilon_F) \delta(\varepsilon_{n\mathbf{k}} - \varepsilon_F) \times (2/\omega_{qv})$ from $r^2\text{SCAN}$ across the k_x - k_y plane at $k_z = 0$ (left panel) and $k_z = \pi/c$ (right panel).

demonstrate that $r^2\text{SCAN}$ also provides reliable predictions of the EPC strength λ [defined in Eq. (6)] in the main-group conventional superconductors. Here, we revisit the prototypical conventional superconductor MgB_2 , a well-known phonon-mediated multiband superconductor with a critical temperature (T_c) of approximately 39 K at ambient pressure [96–98]. MgB_2 has been extensively studied, both experimentally [96–100] and theoretically [101–105].

MgB_2 crystallizes in the tetragonal Bravais lattice with space group $P6/mmm$ (#191, 6/ mmm), where boron atoms form honeycomb lattices separated by hexagonal layers of magnesium atoms (see Fig. S3 in the Supplemental Material [39]). The optimized lattice parameters using $r^2\text{SCAN}$ are $a = 3.068$ Å and $c = 3.504$ Å, which are in excellent agreement with the corresponding experimental values of

$a = 3.086$ Å and $c = 3.524$ Å [96]. Electronic states at the Fermi level from $r^2\text{SCAN}$ are primarily derived from boron p states. Two σ -bonding bands, contributed by B $p_{x/y}$ orbitals, cross the Fermi level [Fig. 5(a)], forming two small cylindrical-hole Fermi surfaces along the Γ - A direction [see Fig. 5(e)]. These σ bands are confined to the boron planes, exhibiting a two-dimensional character. In contrast, the dispersive π -bonding bands, derived from the B p_z orbitals, exhibit a three-dimensional character [see Fig. 5(e)], resulting in one hole and one electron Fermi surface near the $k_z = 0$ and the $k_z = \pm\pi/c$ planes, respectively. Our findings here are generally consistent with previous calculations using the standard DFT [103,104].

Comparing $r^2\text{SCAN}$ and PBE, differences are noticeable in Fig. 5(a). For instance, the bandwidth predicted by

r^2 SCAN is generally larger than that predicted by PBE. At the M point, the band splitting between the upper π band and the lower σ band is approximately 2.6 eV with r^2 SCAN, compared to 2.4 eV with PBE. The r^2 SCAN prediction here matches the experimental value of 2.6 eV [108] and agrees with high-level modified HSE [109] and scQPGW calculations, all of which also yield 2.6 eV [109].

In Fig. 5(c), we compare the calculated phonon dispersion of MgB_2 using r^2 SCAN and PBE. As with other studies [100,104], r^2 SCAN finds four optical phonon modes at Γ : E_{1u} , A_{2u} , E_{2g} , and B_{1g} , with frequencies of 42.8, 50.2, 71.1, and 87.9 meV, respectively, in close agreement with the experimental values [100]. In Fig. 5(c), we show excellent agreement for acoustic modes between r^2 SCAN and PBE. The E_{2g} mode, involving in-plane stretching of B atoms, plays an important role for EPCs in MgB_2 [101,102,105] [see Figs. 5(c) and 5(f)]. Here, both r^2 SCAN and PBE provide highly accurate predictions for the E_{2g} mode frequency—71.1 and 72.1 meV, respectively—closely matching the experimental Raman value of 75 meV [100,106,107]. Notably, the discrepancy between DFT calculations and experiments has been a subject of controversy [100,104,105,110–113]. Frozen-phonon calculations using PBE have suggested that this discrepancy might be due to strong anharmonic effects in the E_{2g} mode along the Γ - A direction [104,105]. However, this conclusion has been challenged, as the frozen-phonon approximation does not account for all the terms in the anharmonic perturbation theory [100]. More advanced calculations incorporating anharmonic contributions suggest that the resulting frequency shift of the E_{2g} mode at Γ is very weak [110,111]. Several experimental studies show that anharmonic effects in MgB_2 are also relatively weak and anharmonic perturbation theory cannot fully explain the temperature dependence of the Raman E_{2g} peak [100].

In Fig. 5(d), we compare the calculated isotropic Eliashberg function $\alpha^2F(\omega)$ of MgB_2 , based on Eq. (9), using r^2 SCAN (red lines) and PBE (blue lines). As expected, the r^2 SCAN and PBE results between 0 and 50 meV are similar, since the phonon dispersions from the two functionals are nearly identical for low-frequency phonons [see Fig. 5(c)]. However, the optical modes predicted by PBE are slightly softer than those from r^2 SCAN, likely due to the overestimation of lattice constants in PBE. Both functionals yield dominant peaks in the range of 63–75 meV, with a secondary peak at 91 meV also well captured. The calculated EPC strength λ is 0.73 from r^2 SCAN, slightly higher than the PBE value of 0.67. Using Eqs. (10) and (11), the estimated ω_{log} from r^2 SCAN is approximately 58.1 meV, resulting in a predicted T_c ranging from 23.2 to 31.6 K, assuming that the adjustable Coulomb pseudopotential μ^* varies between 0.12 and 0.08. These results are consistent with previous studies [102]. Our r^2 SCAN results thus confirm that the strong EPC is mainly driven by the E_{2g} phonon branch along the Γ - A direction [see Fig. 5(c)],

indicating that the in-plane σ -bonding electrons strongly couple with the in-plane vibrations of the E_{2g} mode. This is further supported by the distribution of the EPC strength λ_{nk} in the $k_z = 0$ and $k_z = \pi/c$ planes, and clearly shows that the dominant contribution arises from σ bands [see Fig. 5(f)].

Our analysis in this section demonstrates that the r^2 SCAN functional also can provide a reliable description of the electronic structure, phonon dispersions, and EPC in MgB_2 . It will be interesting to solve the full superconducting-gap equation in MgB_2 , but this is beyond the scope of this study in this multigap superconductor [97,98,101,102,105,114,115].

III. CONCLUSIONS

Accurate and efficient modeling of EPCs in complex functional materials challenges first-principles methods, despite the critical role that these materials play in energy applications, from batteries and catalysts to ferroelectric and superconducting devices. In this study, we demonstrate that the efficient r^2 SCAN meta-GGA functional significantly improves EPC predictions compared to the standard GGAs for transition-metal compounds such as CoO, NiO, and BTO, alongside better descriptions of their electronic structures and phonon dispersions. Due to the SIE, PBE incorrectly predicts CoO as metallic with imaginary phonon frequencies, rendering it unsuitable for EPC calculations. In contrast, r^2 SCAN reduces the SIE, opens a band gap, stabilizes the phonon dispersion, and predicts strong EPCs for optical modes. Analysis of the dielectric screening for CoO and NiO also indicates that these improvements stem from the reduction of SIE in r^2 SCAN compared to PBE. We also demonstrate that r^2 SCAN accurately captures the delicate chemical bonding in the rhombohedral ground state of BTO, providing improved predictions of the electronic and dynamical properties as well as the EPC, without relying on any free parameters such as the Hubbard U and intersite V . Beyond the transition-metal oxides, we show that r^2 SCAN offers a more accurate representation of the band structure, phonon dispersion, and the EPC matrix elements than PBE in GaAs. In MgB_2 , r^2 SCAN also provides a reliable prediction of the phonon frequency of the key E_{2g} mode, which closely matches the Raman measurements. The EPC strength λ and the isotropic Eliashberg spectral function $\alpha^2F(\omega)$ from r^2 SCAN show good agreement with prior studies.

Our study highlights the accuracy, efficiency, and transferability of the advanced r^2 SCAN functional for EPC calculations in transition-metal oxides, ferroelectrics, semiconductors, and superconductors, which are all of key importance to current and emerging energy technologies. We thus establish that r^2 SCAN provides a robust

parameter-free basis for predictive material-specific modeling of EPC in energy materials, and a pathway toward high-throughput studies of superconductivity, thermoelectricity, carrier transport, and other EPC-driven phenomena in functional materials for energy applications.

IV. METHODS

All the calculations have been performed using the projector-augmented-wave (PAW) method [116], as implemented in VASP [41,42]. Both the r^2 SCAN and PBE density functionals have been employed in our calculations. For calculating the EPC, we have followed the Wannier-interpolation procedure outlined in Ref. [5], with the all-electron EPC matrix elements defined in Refs. [6,117]. The Wannier representation of the electronic band structure has been obtained using WANNIER90 [118] with the help of the VASP2WANNIER90 interface. We have calculated the EPC strength λ and the isotropic Eliashberg spectral function α^2F based on the following equations.

Within the Migdal approximation [119], the imaginary part of the phonon self-energy is expressed as

$$\Pi''_{\mathbf{q}\nu} = \text{Im} \sum_{m,n,\mathbf{k}} w_{\mathbf{k}} |g_{mn,\nu}(\mathbf{k}, \mathbf{q})|^2 \frac{f_{n\mathbf{k}} - f_{m\mathbf{k}+\mathbf{q}}}{\epsilon_{m\mathbf{k}+\mathbf{q}} - \epsilon_{n\mathbf{k}} - \omega_{\mathbf{q}\nu} - i\eta}, \quad (5)$$

where $f_{n\mathbf{k}}$ is the Fermi occupation of the single-particle state $|\psi_{n\mathbf{k}}\rangle$ associated with the eigenvalue $\epsilon_{n\mathbf{k}}$, $w_{\mathbf{k}}$ are the weights of the dense $\mathbf{k}$ points, and η is a real positive infinitesimal. The spin index is omitted for simplicity. The EPC strength λ is the full BZ average of the mode-resolved $\lambda_{\mathbf{q}\nu}$

$$\lambda = \sum_{\mathbf{q}\nu} w_{\mathbf{q}} \lambda_{\mathbf{q}\nu}, \quad (6)$$

where the $w_{\mathbf{q}}$ are the weights of the dense $\mathbf{q}$ points. The double-delta approximation [120] has been adopted for calculating the imaginary part of the phonon self-energy, under which Eq. (5) can be rewritten as follows:

$$\Pi''_{\mathbf{q}\nu} = \sum_{m,n,\mathbf{k}} w_{\mathbf{k}} \omega_{\mathbf{q}\nu} |g_{mn,\nu}(\mathbf{k}, \mathbf{q})|^2 \delta(\epsilon_{n\mathbf{k}}) \delta(\epsilon_{m\mathbf{k}+\mathbf{q}}). \quad (7)$$

Under the double-delta approximation, $\lambda_{\mathbf{q}\nu}$ can be expressed as

$$\lambda_{\mathbf{q}\nu} = \frac{1}{\pi N_F} \frac{\Pi''_{\mathbf{q}\nu}}{\omega_{\mathbf{q}\nu}^2}, \quad (8)$$

where N_F is the density of states at the Fermi level. Using this mode-resolved $\lambda_{\mathbf{q}\nu}$ and the phonon frequencies $\omega_{\mathbf{q}\nu}$,

the isotropic Eliashberg spectral function, α^2F , can be expressed as

$$\alpha^2F(\omega) = \frac{1}{2} \sum_{\mathbf{q}\nu} w_{\mathbf{q}} \omega_{\mathbf{q}\nu} \lambda_{\mathbf{q}\nu} \delta(\omega - \omega_{\mathbf{q}\nu}). \quad (9)$$

The phonon-mediated superconducting transition temperature, T_c , is evaluated with the McMillan-Allen-Dynes formula [121],

$$T_c = \frac{\omega_{\log}}{1.2} \exp \left[\frac{-1.04(1 + \lambda)}{\lambda(1 - 0.62\mu^*) - \mu^*} \right], \quad (10)$$

where the logarithmic average frequency $\omega_{\log}$ can be calculated from the following equation [2,122]:

$$\omega_{\log} = \exp \left[\frac{2}{\lambda} \int_0^\infty d\omega \frac{\log \omega}{\omega} \alpha^2F(\omega) \right]. \quad (11)$$

The FM term, which accounts for the electron-phonon interaction contributions to the electronic self-energy, is evaluated by using

$$\Sigma_{n\mathbf{k}}^{\text{FM}}(\omega, T) = \sum_m \sum_{\mathbf{q}\nu} w_{\mathbf{q}} |g_{mn,\nu}(\mathbf{k}, \mathbf{q})|^2 \times \left[\frac{1 - f_{m\mathbf{k}+\mathbf{q}}(T) + n_{\mathbf{q}\nu}(T)}{\omega - \epsilon_{m\mathbf{k}+\mathbf{q}} - \omega_{\mathbf{q}\nu} + i\eta} + \frac{f_{m\mathbf{k}+\mathbf{q}}(T) + n_{\mathbf{q}\nu}(T)}{\omega - \epsilon_{m\mathbf{k}+\mathbf{q}} + \omega_{\mathbf{q}\nu} + i\eta} \right], \quad (12)$$

where $f_{m\mathbf{k}+\mathbf{q}}(T)$ and $n_{\mathbf{q}\nu}(T)$ are the Fermi-Dirac distribution function and the Bose-Einstein distribution function for electrons and phonons of the system at the finite temperature T , respectively. The infinitesimal positive quantity η here is introduced to ensure physical causality and serves as an empirical tuning parameter in practical calculations [123].

Note that an accurate evaluation of the electron self-energy from EPC should include both the FM and Debye-Waller (DW) terms [1]. However, converging the real part of the FM self-energy and the DW term requires summing over a large number of empty states, which is not easily feasible within the Wannier basis. Therefore, for semiconductors, we have set the real part of the FM self-energy to zero and focused only on estimating the spectral function broadening in our calculations. In this connection, the electronic spectral function can be calculated via the following equation:

$$A_{\mathbf{k}}(\omega) = -\frac{1}{\pi} \sum_n \frac{\text{Im} \Sigma_{n\mathbf{k}}^{\text{e-ph}}(\omega)}{(\omega - \epsilon_{n\mathbf{k}})^2 + [\text{Im} \Sigma_{n\mathbf{k}}^{\text{e-ph}}(\omega)]^2}. \quad (13)$$

As an alternative, we have estimated the zero-point renormalization (Δ_{ZPR}) of the band gap due to EPC using the

one-shot stochastic displacement approach described in previous studies [59,60].

CoO and NiO. An initial *G*-type antiferromagnetic (AFM) structure with four atoms per unit cell has been used for structural relaxation. A high-energy cutoff of 600 eV has been applied to truncate the plane-wave basis set. The crystal structures and ionic positions have been fully optimized with a force-convergence criterion of 0.001 eV/Å for each atom and a total energy tolerance of 10^{-8} eV. A Γ -centered *k*-point mesh of $15 \times 15 \times 15$ has been employed for the structural relaxation. For the phonon calculations, a $4 \times 4 \times 4$ supercell with a Γ -centered *k*-point mesh of $3 \times 3 \times 3$ has been used, employing the finite-displacement method. A Gaussian smearing with a small σ of 0.01 eV has been applied in our phonon calculations. Transition-metal (Ni and Co) *d* states and oxygen *p* states have been included in constructing the Wannier functions. To estimate the zero-point renormalization (Δ_{ZPR}) of NiO, we have adopted a $4 \times 4 \times 4$ supercell with a Γ -centered *k*-point mesh of $3 \times 3 \times 3$ in our calculations, by using the stochastic displacement approach [59,60].

BTO. A high-energy cutoff of 520 eV has been applied to truncate the plane-wave basis set. The crystal structures and ionic positions have been fully optimized using a force-convergence criterion of 0.0001 eV/Å for each atom and a total energy tolerance of 10^{-9} eV. To relax the structures, we have adopted a Γ -centered *k*-point mesh of $20 \times 20 \times 20$ in the primitive bulk BZ. For phonon calculations, a $5 \times 5 \times 5$ supercell with a Γ -centered *k*-point mesh of $2 \times 2 \times 2$ has been employed, using the finite-displacement method. A Gaussian smearing with a small σ of 0.01 eV has been applied in our phonon calculations. Ti 3*d* states and O 2*p* states have been included in constructing the Wannier functions.

GaAs. A high-energy cutoff of 520 eV has been applied to truncate the plane-wave basis set. The crystal structures and ionic positions have been fully optimized using a force-convergence criterion of 0.0001 eV/Å for each atom and a total energy tolerance of 10^{-10} eV. To relax the structures, we have adopted a Γ -centered *k*-point mesh of $20 \times 20 \times 20$ in the primitive bulk BZ. For phonon calculations, a $8 \times 8 \times 8$ supercell with a single Γ point has been employed using the finite-displacement method. A Gaussian smearing with a small σ of 0.02 eV has been applied in our phonon calculations. Ga 4*p* and *s* states and As 4*p* states have been included in constructing the Wannier functions.

MgB₂. A high-energy cutoff of 520 eV has been applied to truncate the plane-wave basis set. The crystal structures and ionic positions have been fully optimized using a force-convergence criterion of 0.001 eV/Å for each atom and a total energy tolerance of 10^{-8} eV. To relax the structures, we have adopted a Γ -centered *k*-point mesh of $20 \times 20 \times 18$ in the primitive bulk BZ. For phonon calculations, a $6 \times 6 \times 6$ supercell with a Γ -centered *k*-point

mesh of $4 \times 4 \times 4$ has been employed, using the finite-displacement method. A Gaussian smearing with a small σ of 0.34 eV has been applied in our phonon calculations, following previous studies [124]. Mg *p* and *s* states, along with B *p* and *s* states, have been included in constructing the Wannier functions.

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R.Z., J.S., and A.B. designed the study. Y.W., M.E., and R.Z. performed first-principles computations and analyzed the data, with help from C.L., H.M., B.B., R.S.M., J.Z., G.K., A.B., and J.S. Authors A.B. and J.S. provided computational infrastructure. R.Z., Y.W., M.E., H.M., C.L., B.B., R.S.M., A.B., G.K., and J.S. wrote the manuscript, with input from all the authors. All authors contributed to editing the manuscript.

The authors declare no competing financial interests.

DATA AVAILABILITY

The data that support the findings of this paper are not publicly available. The data are available from the authors upon reasonable request.

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