Reliable lattice dynamics from an efficient density functional

Jinliang Ning,* James W. Furness, and Jianwei Sun†
Department of Physics and Engineering Physics,
Tulane University, New Orleans, Louisiana 70118, United States
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First principles predictions of lattice dynamics are of vital importance for a broad range of topics in materials science and condensed matter physics. The large-scale nature of lattice dynamics calculations and the desire to design novel materials with distinct properties demands that first principles predictions are accurate, transferable, efficient, and reliable for a wide variety of materials. In this work, we demonstrate that the recently constructed $r^2$SCAN density functional meets this need for general systems by demonstrating phonon dispersions for typical systems with distinct chemical characteristics. The functional’s performance opens a door for phonon-mediated materials discovery from first principles calculations.

I. INTRODUCTION

Each new age of human technology has been enabled by the discovery of new materials. From the knapped stone and simple metallurgy of history to the semiconductor revolution of recent decades, a new understanding of materials has expanded the horizons of possibility. This trend for materials to drive progress has not gone unnoticed and ever increasing effort is being devoted to using computational models to search the vast materials space for desirable new compounds.

Phonons are the quanta of lattice waves driven by the elementary thermal excitation of the atoms or molecules that make up a condensed matter system. Intuitively, long-wavelength phonons are perceived as sound. Phonons can interact with electronic structure and have a profound impact on a wide range of observed material phenomena, from thermal and electrical conductivity through to more exotic charge density waves and superconductivity, alongside their decisive role controlling the dynamic stability of materials. This position at the center of materials property design has driven prediction of phonon spectra to become an important aspect of materials space searches.

The connection between the vibrational frequency of a phonon, $\omega(k)$, and the wavevector, $k$, is known as the phonon dispersion. It can be measured experimentally by inelastic neutron or x-ray scattering. The phonon dispersion can also be predicted from theory using force constants calculated with computational models, though the cost of such calculations is generally high. This results in a simultaneous requirement for phonon calculations to efficiently scale for high-throughput workflows while maintaining sufficient accuracy to usefully guide experiments. Density functional theory (DFT) [1] using a semi-local exchange-correlation (XC) functional offers an appealing balance of these considerations and has become the workhorse computational method for high-throughput materials discovery. Efficient evaluation of phonon spectra can be obtained from density functional calculations using density functional perturbation theory [2], or through direct displacements of the atoms [3]. Indeed, density functional methods have already proved effective tools for identifying new phonon phenomena, with the discovery of high/room temperature hydrogen-based superconductors as a prominent example[4, 5].

High throughput calculations of thermodynamic properties [6] is vital for phase diagram predictions [7, 8], and discovery of new meta-stable materials [9] places a particularly high demand for simultaneous accuracy, transferability, efficiency, and reliability. While the accuracy of a DFT calculation is largely determined by the accuracy of the chosen XC functional, the high computational demand of phonon calculations largely excludes expensive nonlocal XC functionals, like hybrid density functionals [10]. The current choice for phonon calculations remains conventional density functionals, including local density approximation (LDA) and the Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation (GGA). While efficient and reasonably accurate, one problem with these conventional density functionals is their transferability in the materials space where different compounds can have very different chemical bonds. Recent progress has shown that semi-local meta-GGAs can maintain this efficiency while being accurate for a wide variety of materials [9, 11–22], exemplified by the strongly constrained and appropriately normed (SCAN) meta-GGA [11, 12]. Unfortunately, extensive use has shown that SCAN suffers numerical problems that are exaggerated in phonon calculations, making reliably obtaining accurate phonon spectra from SCAN calculations a challenging task. Here, we show that a revised version of SCAN which solves the numerical problems, called $r^2$SCAN [23], delivers accurate, transferable, and reliable lattice dynamics. This is demonstrated in a selected set of materials that have different bonding characteristics. We then further explain the origins of such excellent performance of $r^2$SCAN for phonon calculations.

* jning1@tulane.edu
† jsun@tulane.edu
FIG. 1. Well converged Phonon dispersions of (a) Si, (b) GaAs, (c) Fe, and (d) NiO, calculated by LDA, PBE, and r2SCAN, compared with available experimental data, of 1963 [24], 1972 [25], and 1994 [26] for Si, of 1990 [27] for GaAs, from Ref. [28] for Fe, and from Ref. [29] for NiO. The second 3 experimental acoustic band data points along Γ-K direction for NiO were directly taken from their figure of Ref. [29].

II. RESULTS

The selected small test set includes four solids. Two are industrially important semiconductors with covalent or mixed covalent-ionic interactions: covalent Si in the diamond structure, and GaAs with zinc blende structure. One magnetic metal: body-centered-cubic (bcc) Fe. We also include a magnetic oxide with covalent-ionic bonding NiO. These solids have been widely studied experimentally and theoretically and their phonon dispersions have been accurately determined from experiments. In all cases we are interested in establishing both the accuracy and numerical stability of the phonon spectra calculated by different density functionals.

A. Si and GaAs

Figures 1 a) and b) compare calculated phonon dispersions with experimental results for Si and GaAs respectively. The aim of these calculations is to establish the relative accuracy of the functionals under ideal conditions with high-accuracy computational settings tuned to ensure well converged results for all functionals. We find that LDA predicts a relatively accurate spectrum for GaAs but underestimates lower frequency phonon bands in Si, while PBE underestimates phonon frequencies across the board. The r²SCAN meta-GGA shows the most consistent accuracy across both materials and at all energy ranges, closely matching the experimental data.

To establish the relative efficiency of r²SCAN compared to its parent SCAN functional, we repeat the calculations of Figures 1 a) and b) using the default VASP computational settings that better reflect a high-throughput workflow. The resulting phonon spectra are shown in Figures 2 a) and b) for Si and GaAs respectively. For Si of Figure 2 a), SCAN and r²SCAN show similar accuracy across much of the spectrum, though SCAN’s error is significant for low frequency bands between the $L$ and $\Gamma$ points. For GaAs in Figure 2 b) however, the numerical problems of the SCAN functional are immediately apparent. Here spurious imaginary frequencies occur across the SCAN spectrum and the higher frequency bands show generally poor accuracy. Conversely, the r²SCAN functional remains well behaved under these cheaper settings, predicting an accurate and well converged spectrum for high and low frequencies. Note that imaginary frequencies are predicted by SCAN despite using a fully relaxed ionic structure that should be stable along all wave vectors. This prediction of spurious imaginary frequencies can be attributed to incomplete sampling of sharp oscillations in the SCAN XC potential as the ionic positions are displaced [23, 30]. With special
FIG. 2. Phonon dispersions of (a) Si and (b) GaAs calculated by SCAN and r2SCAN with default settings, compared with available experimental data from 1963 [24], 1972 [25], and 1994 [26] for Si, and from 1990 [27] for GaAs.

Figure 1 c) compares calculated and experimental phonon dispersions for bcc Fe. Here, the improvement from GGA to meta-GGA is less clear, with both showing regions of accuracy. The $N$ high-symmetry point of bcc Fe appears particularly challenging for all functionals. It is notable however, that $r^2$SCAN significantly improves over other functionals for the lower band along $\Gamma - N - P - N - H$, though this trend is reversed at higher frequencies with PBE showing greater accuracy for the middle and higher bands.

It has commonly been observed that SCAN tends to overestimate magnetic moments and the magnetisation energy of simple magnetic metals like Fe, Co, and Ni [43–49]. As we include bcc Fe we have calculated the local magnetic moments for the transition metal atoms and present the results in Table I. We see that $r^2$SCAN maintains the over-magnetisation of SCAN, as should be expected from $r^2$SCAN’s construction as a regularisation of SCAN. Figure 1 c) shows that the $r^2$SCAN calculated phonon dispersions are not unduly degraded by this over-magnetisation however.

C. NiO

Figure 1 d), compares the calculated and experimental phonon dispersions for NiO. The $r^2$SCAN meta-GGA shows significant improvements over LDA and PBE for this material. In particular, the high-frequency optical bands from LDA and PBE are qualitatively wrong, while $r^2$SCAN is reasonably accurate. Note we allow the crystal structure to be fully relaxed from the ideal AFM FCC structure, resulting in a small shrinkage of the lattice in the direction perpendicular to the ferromagnetic Ni planes. This symmetry breaking then leads to three optical bands. For this system we expect the main source of error to be self-interaction error, which causes the $d$ orbital electron density to become too diffuse and fractionally occupied. In comparison with LDA and PBE, SCAN and $r^2$SCAN reduce self-interaction errors and localize the $d$ electrons around the Ni ion to a greater degree [16, 17], stabilizing the magnetic moment as shown Table I. The self-interaction error of semi-local functionals can be remedied by including a Hubbard $U$ term. The $ad hoc$ nature of its parameterization limits predictive power however.

Polar bonds, such as those found in systems like NiO and GaAs, can cause the longitudinal optical and transverse optical (LO-TO) splitting in the experimentally observed phonon dispersion. A non-analytical correction to the phonon dispersion [50–52] based on the high-frequency dielectric constants $\epsilon_\infty$ and Born effective charge $Z^*$ must therefore be considered. Table I shows that $r^2$SCAN delivers slightly better $Z^*$ in comparison with LDA and PBE. As $\epsilon_\infty$ is related to the response of electrons to the external electric field and can be strongly affected by the self-interaction error, $r^2$SCAN significantly improves $\epsilon_\infty$ over LDA and PBE, although the discrepancy in $\epsilon_\infty$ from the experimental values is notable. In order to better illustrate the comparison of calculated force constants, we use the $r^2$SCAN $\epsilon_\infty$ and $Z^*$ for the non-analytical term corrections [50–52] of the phonon dispersions of NiO and GaAs for all functionals.

III. DISCUSSION

Viewing Figure 1 as a whole, we can see a broad trend of accuracy across the different materials: PBE < LDA < $r^2$SCAN. It is perhaps surprising that the inclusion of gradient information into the PBE GGA results in worse accuracy than the simpler LDA functional for the phonon dispersions. This effect can also be viewed from the other direction: it is surprising that LDA is as successful as it
is for the phonon dispersion, particularly given its well known tendency to underestimate bond lengths and lattice constants. The reason for this can be found in the force constant dynamic matrix \[2\],

\[
\frac{\partial^2 E(R)}{\partial R_i \partial R_j} = -\frac{\partial F_I}{\partial R_j} = \int \frac{\partial n_R(r)}{\partial R_j} \frac{\partial V_R(r)}{\partial R_i} dr + \int n_R(r) \frac{\partial V_R(r)}{\partial R_i} \frac{\partial^2 V_R(r)}{\partial R_j} dr + \frac{\partial^2 E_N(R)}{\partial R_i \partial R_j},
\]

(1)

where \(R_I\) is the position of nucleus \(I\), \(n_R(r)\) and \(V_R(r)\) are the ground state electron density and nuclear potentials respectively with nuclei in positions \(R\), and \(E_N(R)\) is the Coulomb repulsion between the nuclei at positions \(R\).

As the LDA bond lengths are too short, the second order derivative of the nuclear repulsion energy is overestimated (the final term of Eq. 1). This error is compensated however, by an overestimation of the linear response of electron density \(n_R(r)\) to the nuclear distortion in the first term of Eq. 1. Since the first and final terms of Eq. 1) have opposite signs, their errors are favorably cancelled. The overestimation of the linear response of electron density is a consequence of the self-interaction errors intrinsic to semilocal density functionals, including LDA, PBE, and r^2SCAN. PBE tends to overestimate bond lengths without correcting the linear response, so the favorable cancellation is lost. Like its parent functional r^2SCAN improves both these aspects, giving accurate lattice constants \cite{23} while simultaneously improving linear response characteristics \cite{12} as demonstrated in Table I for lattice constants and \(\epsilon_\infty\). This results in a more accurate phonon spectrum with greater transferability across different classes of materials.

As previously mentioned, Figure 2 shows how r^2SCAN improves on the SCAN functional by avoiding the numerical sensitivities that necessitate the expensive tuning of the fast Fourier transform grid. A full analysis of the origin of the numerical issues in SCAN, and their solution in r^2SCAN, is presented in Ref. \cite{23}. When calculating lattice dynamics from finite atomic displacements the smooth exchange-correlation potential of r^2SCAN is well sampled by a coarse grid while the sharp oscillations of the SCAN potential are not \cite{23, 30}. This poor sampling results in slow and unpredictable convergence of the SCAN phonon spectrum with grid density, and the appearance of spurious imaginary frequencies.

### IV. CONCLUSIONS

We have tested the performance of r^2SCAN for calculating the phonon dispersions of typical systems relative to experimental data and other commonly used functionals (LDA and PBE). Our results for these systems suggest that r^2SCAN can calculate accurate lattice dynamics for general systems with good transferability between different bonding characteristics. Across all the materials tested we find r^2SCAN is either the best choice, or competitive with the best choice in the case of magnetic metals. While we find that SCAN can be accurate when Fourier transform grid and atomic displacement settings are tuned, however its poor numerical stability makes identifying the ideal parameters burdensome. Additionally, the necessary use of expensive Fourier transform grids prevents the SCAN functional being truly useful to high throughput studies. When default low-cost computational settings are used we find that SCAN predicts spurious imaginary bands. These problems are avoided in the r^2SCAN functional which predicts accurate phonon spectra even from low-cost default parameters. While we find that while LDA and PBE can be quite accurate for some systems, they do not show the same generally transferable accuracy as r^2SCAN does. With these inspiring findings, we strongly recommend r^2SCAN to the community as an effective computational tool for future phonon dispersion studies.

### V. METHODS

DFT \cite{1} calculations with the LSDA, PBE\cite{53}, SCAN\cite{11}, and r^2SCAN \cite{23} XC functionals were performed using the Vienna Ab-initio Simulation Package (VASP) \cite{54}. The projector-augmented wave (PAW)

| Methods | \(Si\) | \(GaAs\) | \(Fe\) | \(NiO\) |
|---------|--------|--------|--------|--------|
| LDA     | 5.4029 | 5.6110 | 2.07   | 17.33  |
| PBE     | 5.4688 | 5.7505 | 2.32   | 92.83  |
| SCAN    | 5.4723 | 5.6670 | 2.16   | 12.67  |
| r^2SCAN | 5.4398 | 5.6688 | 2.15   | 11.63  |
| Expt.   | 5.4309 | 5.6556 | 2.18   | 10.89  |

a: \cite{31} b: \cite{32} c: \cite{33} d: \cite{34} e: \cite{35} f: \cite{36} g: \cite{37} h: \cite{38} i: \cite{39} j: \cite{40} k: \cite{41} l: \cite{42}
method was used to treat the core ion-electron interaction \cite{55,56}. An energy cutoff of 600 eV was used to truncate the plane wave basis. A Γ-centered mesh with a spacing threshold of 0.15 Å \(^{-1}\) was used for \(k\)-space sampling for unit cell relaxations of semiconducting systems Si, GaAs and NiO, and 0.1 Å \(^{-1}\) for metallic Fe. For supercell atomic force calculations, only a single Γ point is used for semiconducting systems Si, GaAs and NiO, and a \(2 \times 2 \times 2\) k-point mesh for metallic Fe. A Gaussian smearing with 0.02 eV is used for semiconducting systems Si, GaAs and NiO, and Methfessel–Paxton smearing with 0.2 eV for Fe. For atomic force calculations, \(3 \times 3 \times 3, 3 \times 3 \times 3, 5 \times 5 \times 5,\) and \(4 \times 4 \times 2\) supercells of the conventional unit cells (as shown in Table 1) are used for Si, GaAs, Fe and NiO, respectively. The ionic positions of all systems were relaxed for all functionals until the maximum ionic forces were below 1 meV Å \(^{-1}\). We used the Phonopy code \cite{57} to obtain the harmonic force constants from VASP atomic force calculations within finite displacement method (0.015 Å). For Figure 1, PREC = High; ENAUG = 2000 is specified for \(\text{r}^2\text{SCAN},\) while for Figure 2, we used the VASP officially recommended accurate defaults (PREC = Accurate) together with special tuned fast Fourier transform grid density for comparison. The full set of comparison is referred to the supplementary material.

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[1] W. Kohn and L. J. Sham, Self-consistent equations including exchange and correlation effects, Phys. Rev. 140, A1133 (1965).
[2] S. Baroni, S. De Gironcoli, A. Dal Corso, and P. Giannozzi, Phonons and related crystal properties from density-functional perturbation theory, Reviews of modern Physics 73, 515 (2001).
[3] K. Parlinski, Z. Li, and Y. Kawazoe, First-principles determination of the soft mode in cubic zro 2, Physical Review Letters 48, 1093 (1997).
[4] Y. Li, J. Hao, H. Liu, Y. Li, and Y. Ma, The metalization and superconductivity of dense hydrogen sulfide, The Journal of chemical physics 140, 174712 (2014).
[5] A. Drozdov, M. Eremets, I. Troyan, V. Ksenofontov, and S. I. Shylin, Conventional superconductivity at 203 kelvin at high pressures in the sulfur hydride system, Nature 525, 73 (2015).
[6] A. Jain, S. P. Ong, G. Hautier, W. Chen, W. D. Richards, S. Dacek, S. Cholia, D. Gunter, D. Skinner, G. Ceder, and K. a. Persson, The Materials Project: A materials genome approach to accelerating materials innovation, APL Materials 1, 011002 (2013).
[7] P. Spencer, A brief history of calphad, Calphad 32, 1 (2008).
[8] B. Sundman, B. Jansson, and J.-O. Andersson, The thermo-calc databank system, Calphad 9, 153 (1985).
[9] J. Ning, Y. Zhu, J. Kidd, Y. Guan, Y. Wang, Z. Mao, and J. Sun, Subtle metastability of the layered magnetic topological insulator mni 2 te 4 from weak interactions, npj Computational Materials 6, 1 (2020).
[10] A. V. Krukau, O. A. Vydrov, A. F. Izmaylov, and G. E. Scuseria, Influence of the exchange screening parameter on the performance of screened hybrid functionals, The Journal of chemical physics 125, 241106 (2006).
[11] J. Sun, A. Rużyniszcz, and J. P. Perdew, Strongly constrained and appropriately normed semilocal density functional, Physical review letters 115, 036402 (2015).
[12] J. Sun, R. C. Remsing, F. Zhang, Z. Sun, A. Rużyniszcz, H. Peng, Z. Yang, A. Paul, U. Waghmare, X. Wu, et al., Accurate first-principles structures and energies of diversely bonded systems from an efficient density functional, Nature chemistry 8, 831 (2016).
[13] R. C. Remsing, M. L. Klein, and J. Sun, Dependence of the structure and dynamics of liquid silicon on the choice of density functional approximation, Physical Review B 96, 024203 (2017).
[14] Y. Zhang, C. Lane, J. W. Furness, B. Barbieri, J. P. Perdew, R. S. Markiewicz, A. Bansil, and J. Sun, Competing stripe and magnetic phases in the cuprates from first principles, Proceedings of the National Academy of Sciences 117, 68 (2020).
[15] J. W. Furness, Y. Zhang, C. Lane, I. G. Buda, B. Barbieri, R. S. Markiewicz, A. Bansil, and J. Sun, An accurate first-principles treatment of doping-dependent electronic structure of high-temperature cuprate superconductors, Communications Physics 1, 1 (2018).
[16] Y. Zhang, J. Furness, R. Zhang, Z. Wang, A. Zunger, and J. Sun, Symmetry-breaking polymorphous descriptions for correlated materials without interelectronic u, Phys. Rev. B 102, 045112 (2020).
[17] Y. Zhang, J. W. Furness, B. Xiao, and J. Sun, Subtlety of ti02 phase stability: Reliability of the density functional theory predictions and persistence of the self-interaction error, The Journal of Chemical Physics 150, 014105 (2019).
[18] A. D. Kitchaev, H. Peng, Y. Liu, J. Sun, J. P. Perdew, and G. Ceder, Energetics of mm02 polymorphs in density functional theory, Phys. Rev. B 93, 045132 (2016).
[19] H. Peng and J. P. Perdew, Synergy of van der waals and self-interaction corrections in transition metal monoxides, Phys. Rev. B 96, 100101 (2017).
[20] Y. Zhang, J. Sun, J. P. Perdew, and X. Wu, Comparative first-principles studies of prototypical ferroelectric materials by LDA, GGA, and SCAN meta-GGA, Phys. Rev. B 96, 035143 (2017).
[21] C. Lane, J. W. Furness, I. G. Buda, Y. Zhang, R. S. Markiewicz, B. Barbieri, J. Sun, and A. Bansil, Antiferromagnetic ground state of la 2 cuo 4: A parameter-free ab initio description, Physical Review B 98, 125140 (2018).
[22] R. Zhang, B. Singh, C. Lane, J. Kidd, Y. Zhang, B. Barbieri, R. S. Markiewicz, A. Bansil, and
J. Sun, Understanding the quantum oscillation spectrum of heavy-fermion compound smb6, arXiv preprint arXiv:2003.11052 (2020).

[23] J. W. Furness, A. D. Kaplan, J. Ning, J. P. Perdew, and J. Sun, Accurate and numerically efficient $r^2$SCAN meta-generalized gradient approximation, J. Phys. Chem. Lett. 11, 8208 (2020).

[24] G. Dolling, Inelastic scattering of neutrons in solids and liquids (IAEA, Vienna, 1963).

[25] G. Nilsson and G. Nelin, Study of the homology between silicon and germanium by thermal-neutron spectrometry, Physical Review B 6, 3777 (1972).

[26] J. Kulda, D.Strauch, P. Pavone, and Y. Ishii, Inelastic-neutron-scattering study of phonon eigenvectors and frequencies in si, Physical Review B 50, 13347 (1994).

[27] B. Brockhouse, H. Abou-Helal, and E. Hallman, Lattice vibrations in iron at 296 k, Solid State Communications 5, 211 (1967).

[28] W. Reichardt, V. Wagner, and W. Kress, Lattice dynamics of nio, Journal of Physics C: Solid State Physics 8, 3955 (1975).

[29] W. Yim and R. Paff, Thermal expansion of aln, sapphire, silicon and germanium by thermal-neutron spectrometry, Physical Review B 98, 094413 (2018).

[30] A. J. Price, K. R. Bryenton, and E. R. Johnson, Requirements for an accurate dispersion-corrected density functional, The Journal of Chemical Physics 154, 230902 (2021).

[31] J. W. Furness, A. D. Kaplan, J. Ning, J. P. Perdew, and J. Sun, Accurate and numerically efficient $r^2$SCAN meta-generalized gradient approximation, J. Phys. Chem. Lett. 11, 8208 (2020).

[32] B. Brockhouse, H. Abou-Helal, and E. Hallman, Lattice vibrations in iron at 296 k, Solid State Communications 5, 211 (1967).

[33] W. Reichardt, V. Wagner, and W. Kress, Lattice dynamics of nio, Journal of Physics C: Solid State Physics 8, 3955 (1975).

[34] W. Yim and R. Paff, Thermal expansion of aln, sapphire, silicon and germanium by thermal-neutron spectrometry, Physical Review B 98, 094413 (2018).

[35] A. J. Price, K. R. Bryenton, and E. R. Johnson, Requirements for an accurate dispersion-corrected density functional, The Journal of Chemical Physics 154, 230902 (2021).

[36] X. Gonze, J.-C. Charlier, D. Allan, and M. Teter, Interatomic force constants from first principles, Physical Review B 82, 081104 (2010).

[37] P. Gielisse, J. Plendl, L. Mansur, R. Marshall, S. Mitra, R. Mykolajewycz, and A. Smakula, Infrared properties of nio and coo and their mixed crystals, Journal of Applied Physics 36, 2446 (1965).

[38] P. Hao, Y. Fang, J. Sun, G. I. Csonka, P. H. Philipsen, and J. P. Perdew, Lattice constants from semilocal density functionals with zero-point phonon correction, Physical Review B 85, 014111 (2012).

[39] G. Spencer, A. Ho, J. Menendez, R. Droopad, H. Fathol- lahnejad, and G. Maracas, Lattice-constant dependence of the dynamical effective charge in alas and gaas, Physical Review B 50, 14125 (1994).

[40] A. Togo and I. Tanaka, First principles phonon calculations in materials science, Scripta Materialia 108, 1 (2015).
Table 1 and Figures 1-2 contain the detailed information for the comparison of numerical stability between SCAN and \( r^2 \)SCAN with respect to specific input settings, including the basic accuracy setting (the PREC tag in INCAR), the FFT-grid and the size of small displacements used in atomic force calculations, being tested on Si and GaAs. Figure 3 shows the phonon dispersions of GaAs and NiO calculated by LDA, PBE and \( r^2 \)SCAN with the non-analytical correction using Born effective charge and high-frequency dielectric constant from the underlying density functional. In contrast, the Figures 1(b) and (d) in the main text use the \( r^2 \)SCAN Born effective charge and high-frequency dielectric constants for all LDA, PBE and \( r^2 \)SCAN results.

| Tag      | Basic Settings | Unit cell | Supercell | d (Å) |
|----------|----------------|-----------|-----------|-------|
| Acc      | ENCUT 2NGX     | 48 96     | 140 280   | 0.015 |
| AccNGX   | Specified 2NGX | 48 96     | 144 288   | 0.015 |
| HighNGX | ENCUT ENAUG 48 | 108 144   | 320       | 0.03  |

| Tag      | Basic Settings | Unit cell | Supercell | d (Å) |
|----------|----------------|-----------|-----------|-------|
| Acc      | ENCUT 2NGX     | 48 96     | 140 280   | 0.015 |
| AccNGX   | Specified 2NGX | 48 96     | 144 288   | 0.015 |
| HighNGX | ENCUT ENAUG 48 | 112 144   | 336       | 0.03  |

* jning1@tulane.edu
† jsun@tulane.edu
FIG. 1. Phonon dispersions of Si calculated by SCAN and $r^2$SCAN. See Table I for definition of legend tags.

FIG. 2. Phonon dispersions of GaAs calculated by SCAN and $r^2$SCAN. See Table I for definition of legend tags.
FIG. 3. Phonon dispersions of (a) GaAs and (b) NiO of LDA, PBE and $r^2$SCAN with the non-analytical correction using Born effective charge and high-frequency dielectric constants calculated by the underlying density functional, compared with experimental results.