Partitioning interatomic force constants for first-principles phonon calculations: Applications to NaCl and twisted bilayer graphene

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First-principles phonon calculations have been widely performed for studying vibrational properties of condensed matter, where the dynamical matrix can be constructed via supercell force-constant calculations or the linear response approach. With different manners, a supercell needs to be introduced in both methods. Unless the supercell is large enough, the phonon property highly depends on the shape and size of the supercell and the imposed periodicity could give unphysical results that can be easily overlooked. Along this line, the concept of partition of force constants is discussed, and addressed by a model structure, NaCl, and twisted bilayer graphene as examples for illustrating the effects of the imposed supercell periodicity. To diminish the unphysical effects, a simple partition of force constants, which relies only on the translational symmetry and interatomic distances, is demonstrated to be able to deliver reasonable results. The partition method is also compatible with the mixed-space approach for describing LO-TO splitting. The proper partition is especially important for studying moderate-size systems with low symmetry, such as two-dimensional materials on substrates, and useful for the implementation of phonon calculations in first-principles packages using atomic basis functions, where symmetry operations are usually not applied owing to the suitability for large-scale calculations.

I. INTRODUCTION

First-principles phonon calculations based on density functional theory have been successful in describing various vibrational and thermodynamic properties of condensed matter, owing to the accurate description of total energy surfaces against atomic displacements. Thanks to the computational power available nowadays, not only is the amount of phonon calculations from first principles rapidly increasing but also building phonon databases via high-throughput calculations becomes essential for large-scale exploration of new materials. The underlying theoretical foundation for the most performed phonon calculations relies on the harmonic approximation, which makes the physics easily accessible, and can be straightforwardly extended for exploring anharmonic effects including the progress in the field of information science, for example, the compressive sensing method in studying phonons with sufficient training data. In actual implementation, the dynamical matrix, whose eigenvalues deliver the phonon dispersion, can be constructed via the real-space force constants obtained directly from the supercell calculations by displacing atoms from equilibrium through the finite-difference method or the self-consistent calculations based on the density functional perturbation theory (DFPT) in the framework of linear response theory where the response, and therefore the dynamical matrix, can be calculated at any arbitrary wave vector accurately.

As long as the harmonic approximation is valid, both the supercell force-constant and linear response approaches should in principle reach the same result on the condition that the supercell is large enough allowing the forces decaying into negligible values away from the displaced atom and the dynamical matrices are calculated by DFPT at all the studied q points, respectively. However, a large-scale supercell calculation for systems having imperfection, such as defects and the presence of surfaces, could be difficult. The difficulty still remains in the adoption of linear response approach, where the required self-consistent calculations are time-consuming and the implementation with complicated functionals could be challenging. To reduce the computational effort for the DFPT calculation where a q solution can be obtained, the dynamical matrices need to be calculated only at the q grids that can be used for Fourier transform, leading to the real-space force constants. Note that the dynamical matrix can be considered as the reciprocal-space force constants regardless of the atomic mass. In this context, a denser q-mesh corresponds to a larger supercell and therefore having more complete long-range interatomic force constants that can be used for interpolating full phonon dispersion with higher accuracy.

Although the linear response approach can deliver accurate results, which can be considered as the exact theoretical solutions for the harmonic approximation, the supercell force-constant calculations have still been extensively adopted for studying phonon properties. This is because the concept of force constants in real space is intuitive. Meanwhile, the implementation of DFPT is not available in most of the first-principles packages using atomic basis functions which are efficient for large-scale calculations. Moreover, the constructed dynamical matrices via the supercell force-constant approach are expected to be consistent with those obtained by DFPT for the wave vectors commensurate with the adopted supercell. In this case, the requirement for having negligible forces away from the displaced atom becomes ir-
relevant. Although the commensurability offers a luring way to investigate phonons at the high-symmetry points in the Brillouin zone, the number of those \( q \) vectors is quite limited. To describe thermodynamic properties via phonon density of states, the phonon dispersion in the entire Brillouin zone is needed. Recently, there are several important efforts that have led to great improvements of determining phonon properties associated with the incommensurate \( q \) vectors, such as the compressive sensing lattice dynamics method\(^\text{[23]}\) for better fitting phonon dispersion by modeling the force constants and the mixed-space method\(^\text{[23,25]}\) for better interpolating the frequency splitting between the longitudinal optical and transverse optical (LO-TO) modes due to the dipole-dipole interactions. Along this line, we will address an issue of partitioning force constants, which can improve the interpolated phonon properties associated with the incommensurate \( q \) vectors while keeping those with the commensurate ones unaffected, for the phonon calculations using the supercell force-constant approach.

The paper is organized as follows. The dynamical matrix, including the non-analytic contribution, and how the mixed-space method can be used to interpolate the phonon dispersion with LO-TO splitting are given in Sec. III. The concept of partition of interatomic force constants and the related formulae are discussed in Sec. III A. In Sec. III B, a simple but useful partition method is proposed. The calculated phonon dispersions of NaCl and twisted bilayer graphene using the proposed partition method are presented in Sec. III C and Sec. III D, respectively. Finally, a summary is given in Sec. IV.

II. DYNAMICAL MATRIX

The dynamical matrix, \( D \), whose eigenvalues give the phonon frequencies (more precisely, the squares), can be constructed via the interatomic force constant, \( C \):

\[
D_{\alpha\beta}^{st}(q) = \frac{1}{\sqrt{M_s M_t}} \sum_{i=1}^{N} C^s_{\alpha\beta}(0, \vec{R}_i) \epsilon^{i\vec{q} \cdot \vec{R}_i},
\]

where \( M_s \) and \( M_t \) denote the mass of atoms \( s \) and \( t \) belonging to the primitive unit cell, respectively, and \( \alpha \) and \( \beta \) index the Cartesian components \((x, y, \text{and } z)\). \( \vec{R}_i \) labels the primitive lattice vector and \( i \) sums over the primitive unit cells in the supercell, so \( N \) indicates the number of the primitive unit cells in the supercell.

The non-analytic contribution, \( D_{\alpha\beta}^{na} \), which should be added to Eq. 1 to account for the LO-TO splitting in the long-wavelength limit \((q \to 0)\)\(^\text{[23]}\) is expressed as

\[
D_{\alpha\beta}^{na} = \frac{4\pi e^2}{V} \left( \frac{q \cdot Z_s^*}{q \cdot \epsilon^{\infty} \cdot q} \right)_{\alpha(\beta)},
\]

where \( V \) denotes the volume of the primitive unit cell. \( Z^* \) and \( \epsilon^{\infty} \) denote the Born effective charge and the high-frequency dielectric constant, respectively. Since the expected weight of the non-analytic contribution is 1 at \( \Gamma \) (more precisely, at \( q \to 0 \)) and 0 at the commensurate \( q \) points, an adjustable damping function, for example, \( e^{-\alpha q^2} \), can be introduced to the weight for interpolating the phonon dispersion\(^\text{[22]}\). However, it should be noted that a Gaussian function cannot be entirely reduced to zero by a finite \( q \). On the other hand, the weight has been chosen as the factor,

\[
f(q) = \frac{1}{N} \sum_{i=1}^{N} e^{i\vec{q} \cdot \vec{R}_i},
\]

in the mixed-space method\(^\text{[23,25]}\) which guarantees the required values of 1 and 0 at \( \Gamma \) and the commensurate \( q \) points, respectively. As a result, the term,

\[
\frac{4\pi e^2}{NV} \left( \frac{q \cdot Z_s^*}{q \cdot \epsilon^{\infty} \cdot q} \right)_{\alpha(\beta)}.
\]

can be added together with the force constant to share the same phase factor, \( e^{i\vec{q} \cdot \vec{R}_i} \), described in Eq. 1. The mixed-space method has been widely implemented in most of the phonon packages adopting the supercell force-constant approach\(^\text{[23,25]}\).

To construct the dynamical matrix of a system, in principle, all of the primitive-cell atoms in the supercell need to be displaced, and the complete force constants can then be obtained from the \( 3 \times N_{\text{atom}} \) supercell calculations, where \( N_{\text{atom}} \) denotes the number of atoms in the primitive unit cell. For systems with low symmetry, the required \( 3 \times N_{\text{atom}} \) calculations cannot be reduced significantly and the so-called supercell could be the primitive unit cell itself, for example, in the case of studying the slab system with a surface reconstruction where the primitive unit cell is large enough for exploring desired phonon properties and doubling the in-plane unit cell is already unfavorable for computation due to the thickness of the slab. One of the good strategies for studying this system is to fit the forces using the compressive sensing lattice dynamics method\(^\text{[23]}\) by performing a smaller number \((< 3 \times N_{\text{atom}})\) of supercell calculations. In this study, we focus on how to improve the obtained force constants from a supercell that is not large enough allowing the forces decaying into negligible values away from the displaced atom.

III. PARTITION OF FORCE CONSTANTS

A. Concept and formulation

A model structure of two-dimensional material is presented in Fig. 1(a). For clarity, the substrate is not shown and the reference atom A is plotted at the center of the unit cell, which is always possible owing to the periodic boundary condition. It can be observed that there are four periodic repeats of the corner atom and two pairs of periodic repeats at the boundary. Obviously, there is only one force that can be obtained for each set of
with and where the real-space force constants are obtained via the interpolation procedure in the linear response approach, respectively. The partition is also needed for the force constants and four for A-C, A-C' counterparts will be overlooked. The resulting force constants of the shorter-distance atoms should be smaller than the A-C ones due to the longer distance, another choice of partition is to divide the A-C force by two instead of four. At this step, the sum rule is still satisfied because the weight becomes 0.5, 0.5, 0, and 0 for the four atoms by treating the A-D and A-D' weight as 0 and the total sum is still 1. However, after applying rotational symmetry to further redistribute the force constants between atoms B and C, there is no guarantee that the phonon properties remain the same at the commensurate q vectors since atoms B and C do not belong to the same periodic set. Although the symmetry operation does recover the rotational symmetry if the system has the symmetry, it should be noted that symmetry operations need to be applied with caution; otherwise, all the phonon properties could become the interpolated ones. In Sec. III B, we will focus on how to improve the interpolation while avoiding affecting the solutions at the commensurate q vectors.

We now consider an adatom B, located at the hollow site as shown in Fig. 1(b). Clearly, the atoms C and D, which are the periodic counterparts of adatom B, are outside the supercell but cannot be neglected in analyzing the force constants associated with the reference atom A. For this case, atoms C and D can be found by applying the translational symmetry or rotational symmetry. For the supercell shown in Fig. 1(c), the adatom B is assumed to deviate from the perfect hollow-site position. If one considers only the force constants of the atoms inside the supercell, the force constants of the shorter-distance A-C and A-D counterparts will be overlooked. The result force constants can be unphysical depending on the strength of the A-B force constants, and the obtained vibrational modes could be incorrect even though the frequencies might still be close to the exact solutions. In this case, to obtain the periodic counterparts, what we need to do is to apply the translational symmetry, not the rotational symmetry. Alternatively, the supercell shown in Fig. 1(c) can be reshaped to the rectangle one shown in Fig. 1(d). However, the seemingly more symmetric distribution of the force constants cannot deliver the exact solutions at the originally commensurate q points, for example, the high-symmetry points in the Brillouin zone of the primitive unit cell, since selecting a supercell is equivalent to the selection of the exact q points.
FIG. 2. A supercell contains 16 atoms. The reference atom has been shifted to the supercell center for easy consideration of the associated force constants. The inner circle centered at the supercell center is defined as the largest circle inside the supercell. The outer circle is the smallest circle with the whole supercell being enclosed. The radii of the two circles are denoted as \( r_{\text{inner}} \) and \( r_{\text{outer}} \), respectively. Three regions, I: \( r < r_{\text{inner}} \), II: \( r_{\text{inner}} \leq r \leq r_{\text{outer}} \), and III: \( r > r_{\text{outer}} \), can be defined. The \( r_{\text{inner}} \) and \( r_{\text{outer}} \) can also be treated as tunable parameters, not being dictated by the largest and the smallest circles, respectively. For the three-dimensional case, the circles become spheres, where \( r_{\text{inner}} \) and \( r_{\text{outer}} \) can still be defined. Note that the lattice constant \( a \) might not be the same as \( b \).

B. Partition method by distance

We have introduced the weight for partitioning force constants in Eqs. 6 and 7. Here, we propose a simple way to choose the weight based on the interatomic distance. The supercell shown in Fig. 2 where three regions, I: \( r < r_{\text{inner}} \), II: \( r_{\text{inner}} \leq r \leq r_{\text{outer}} \), and III: \( r > r_{\text{outer}} \), can be defined, is adopted to illustrate how to perform the proposed partition. The weight is set to 1 for the atoms in the region I. All of the experienced forces in this region caused by displacing the reference atom are not going to be partitioned and are expected to be the leading contribution to the dynamical matrix. The force constants between the reference atom and the atoms in the region III are set to zero since they are far away from each other. In this context, only the atoms in the region II are relevant to the partition. In this region, a force among the periodic atoms that satisfy the translational symmetry described by the supercell lattice vectors can be redistributed based on the interatomic distance. The weight for each atom can be assumed to be inversely proportional to \( r^d \), where \( r \) denotes the radius of the corresponding circle, that is, the atom-reference atom distance. We treat \( d \) as a tunable parameter, whose value depends on the studied materials. Considering the rapidly decaying nature of the strength of force constants against distance, a value of \( d \) between 3 and 5, similar to those described by the dipole-dipole interactions, could be a reasonable choice. For a larger value of \( d \), the effect of the partition simply recovers the contribution of the force constants with the shortest atom-reference atom distance in the region II. After applying the \( r^{-d} \) weight for the atoms, the weight should be normalized to 1 to guarantee the unaffected solutions at commensurate \( q \) vectors. In addition, it is worth mentioning that the partition method is compatible with the mixed-space method because there exist degrees of freedom in performing Eq. 3 for a periodic system. Therefore, the same weight can be used to partition the non-analytic contribution.

C. Phonon dispersion in NaCl

NaCl, crystallizing in the rock salt structure, is adopted as an example for illustrating the applicability of the proposed partition method. The first-principles calculations were performed using OpenMX code, where the generalized gradient approximation (GGA), norm-conserving pseudopotentials, and optimized pseudatomic basis functions were adopted. Three, two, and one optimized radial functions were allocated for the \( s \), \( p \), and \( d \) orbitals, respectively, for the Na atom with a cutoff radius of 9 Bohr, denoted as Na9.0-s3p2d11. For the Cl atom, Cl7.0-s2p2d1 was used. A \( 12 \times 12 \times 12 \) \( k \)-point sampling was adopted for the primitive unit cell. A cutoff energy of 400 Ha was used for the numerical integrations and for the solution of the Poisson equation. The structure, including the unit cell and atomic positions, is fully relaxed relying on the chosen numerical grids without imposing the \( Fm\overline{3}m \) symmetry. The relaxed lattice constant was found to be 5.72\( \text{Å} \). The \( 6 \times 6 \times 6 \) supercell, containing 432 atoms, was chosen for the phonon calculation, where the force constants were obtained by the adoption of a displacement of 0.05\( \text{Å} \).

After displacing one of the Na or Cl atoms towards its first neighbor in the \( 6 \times 6 \times 6 \) supercell, say, along the \( x \) direction, the experienced forces of the neighboring atoms along the same direction are expected to decay with the increasing interatomic distance. As shown in Fig. 3 (a), the strength of the forces drops significantly beyond \( \sim 6 \text{Å} \). However, the forces are not converged to zero but oscillate around zero even at a long distance of \( \sim 12 \text{Å} \), which suggests that a larger supercell and denser real-space grids in describing the charge density and atomic basis functions are needed. In Fig. 3 (b), without including LO-TO splitting, the phonon dispersion obtained from the partitioned force constants using the traditional method is presented by the red curves. Several expected degeneracies in NaCl are lifted, for example, the two transverse acoustic branches along the path between \( \Gamma \) and \( X \) are not degenerate except those at the commensurate \( q \) points indicated by the arrows. The unaffected degeneracies at \( \Gamma \), \( X \), \( L \), and the other commensurate \( q \) points demonstrate the powerfulness of the supercell force-constant approach in calculat-
FIG. 3. (a) Force decay in NaCl along one dimension, calculated in the $6 \times 6 \times 6$ supercell containing 432 atoms using OpenMX code. Only the $x$-component forces of the $x$-direction neighboring atoms of Na or Cl atom that is displaced by 0.05 Å along the $x$ direction are analyzed here. The force constants, obtained by dividing the forces by 0.05 Å, decay rapidly with the distance but still oscillate around zero at a long distance ($\sim 12$ Å). (b) Phonon dispersion of NaCl calculated using the $6 \times 6 \times 6$ supercell. The red curves present the result using the traditional partition method while the black dashed curves present the result using the proposed partition method with $d = 5$. The arrows indicate two degenerate frequencies at the commensurate $q$ points between $\Gamma$ and $X$. LO-TO splitting is not considered here.

To improve the phonon dispersion at the non-commensurate $q$ points, the proposed partition method based on the interatomic distance, not on the number of the periodic atoms at the supercell boundary, is applied. Regarding the rapidly decreasing strength of the force constants against the distance found in Fig. 3 (a), $d = 5$ has been chosen for this partition. The resulting phonon dispersion is presented by the black dashed curves in Fig. 3 (b). Expectedly, the frequencies at the commensurate $q$ vectors are unaffected. Without explicitly applying the rotational symmetry, the degeneracy for the two transverse acoustic branches between $\Gamma$ and $X$ are recovered, and the oscillating behavior at the $q$ points close to $\Gamma$ is diminished and averaged to degenerate bands. Note that this oscillation can exist in a larger supercell calculation in reflecting the suppressed LO-TO splitting in the long-wavelength limit.

To account for the LO-TO splitting in a polar system from first principles, the mixed-space approach or the linear response approach can be adopted. The mixed-space method proposes a consistent way to incorporate the non-analytic contribution with the interatomic force constants, where the Born effective charges and dielectric constants can be obtained from the linear response methods or via other calculations. The linear response approach allows for the calculations of dielectric constants, Born effective charges, and the dynamical matrices using the same framework. For NaCl, the linear response solutions can be obtained without heavy computations and the result can be used to compare with those obtained from the $2 \times 2 \times 2$ supercell calculations.

For consistency, the DFPT and the $2 \times 2 \times 2$ supercell calculations were performed using ABINIT code where norm-conserving pseudopotentials plane-wave basis functions, and GGA were adopted. The cut-off energy of 120 Ha and a $12 \times 12 \times 12$ $k$-point sampling were adopted for the calculations. The structure was fully relaxed by imposing $Fm\bar{3}m$ space group symmetry. The relaxed lattice constant was found to be 5.69 Å. The linear response solutions at the selected $q$ points are presented by the red circles in Fig. 4 which can be treated as the exact solutions for reference. In the supercell calculations, the force constants were obtained using a displacement of 0.05 Å. The dielectric constant, 2.487, and the Born effective charges, Na: 1.106 and Cl: -1.106, obtained from
DFPT were used for constructing the non-analytic term of dynamical matrix. Since the \(2 \times 2 \times 2\) supercell is small, the directly calculated force constants must encounter a highly asymmetric distribution due to the shape of the supercell. The phonon dispersion using the traditional partition method shown by the green curves in Fig. 4 reflects the effects, for example, the degeneracy for the transverse acoustic branches between \(\Gamma\) and \(X\) has been lifted significantly. More seriously, imaginary frequencies are found around \(\Gamma\) even though the mixed-space approach has been adopted.

The black curves shown in Fig. 4 present the result after applying the proposed partition method with \(d = 5\). First, it can be observed that the frequencies remain unaffected at the commensurate \(q\) vectors in comparison with those delivered by the traditional partition method. Secondly, the dispersion calculated by the supercell force-constant method agrees well with the exact solution at the commensurate high-symmetry points. The tiny deviation between these two independent calculations can be attributed to the numerical noise, for example, the error can be introduced by the finite-difference method for the anharmonic potential energy. Finally, the interpolated phonon dispersion has been largely improved by the partition and is in good agreement with the DFPT calculation. It should be emphasized that the proper partition is important for improving not only the phonon dispersion but also the vibrational modes.

D. Phonon dispersion in twisted bilayer graphene

The phonon dispersion of twisted bilayer graphene (T-BLG) has been studied using the Bornvon Karman model and the Lennard-Jones potential for the intralayer and interlayer interactions, respectively.\cite{ref1} Assisted by the first-principles frozen-phonon method, the breathing and shear vibrational modes in T-BLG have also been investigated.\cite{ref2} Although the applications of frozen-phonon methods are limited, the shear modes carrying almost zero frequencies in T-BLG, which are difficult to be accurately analyzed by the general supercell force-constant and linear response approaches, can be studied by treating the graphene monolayer as a rigid body in calculating the total energy against the shear vibration. Different from the direct calculation of dynamical matrix, the phonon frequencies are also reported using molecular dynamics simulations.\cite{ref3} Here, the first-principles phonon dispersion for T-BLG with a rotational angle of 21.8°, as shown in Fig. 3(a), is presented using the proposed partition method based on the functionals within the local density approximation (LDA) and the corrected GGA with the van der Waals interactions (DFT-D2).\cite{ref4}

The first-principles calculations were performed using OpenMX code. C7.0-s2p2d1 was chosen for the basis functions. A cutoff energy of 400 Ha was used for the numerical integrations and for the solution of the Poisson equation. A \(12 \times 12 \times 1\) \(k\)-point sampling was adopted for the primitive unit cell. In general, T-BLG could contain a large amount of atoms in the primitive unit cell. For the \(\theta = 21.8^\circ\) case, the unit cell contains 28 atoms, which can be studied without heavy computations. The experimental in-plane lattice constant \((a = 6.5190\,\text{Å})\) was used and \(c = 20\,\text{Å}\) was adopted to avoid the interactions between T-BLG images. The relaxed interlayer distances by LDA and DFT-D2 were found to be 3.4Å and 3.3Å, respectively. The same unit cell was used for the phonon calculations and the force constants were obtained by the adoption of a displacement of 0.01Å.

For the interlayer breathing mode shown in Fig. 5(b), the reported frequencies are 89.5 cm\(^{-1}\) and 94.56 cm\(^{-1}\)\cite{ref5} In this study, the frequencies obtained from LDA and DFT-D2 are 77.04 cm\(^{-1}\) and 95.53 cm\(^{-1}\), respectively. This indicates that to describe
the interlayer interactions, the inclusion of the van der Waals interactions, as implemented in DFT-D2, is essential for delivering the frequency that is consistent with experiments.\cite{m1}

Without adopting larger supercells for the phonon calculations, a reasonable distribution of force constants is important for the interpolation of phonon dispersion. Since the supercell contains a thick vacuum layer, the number of atoms in the region II is large according to the suggested \( r_{outer} \). To keep only the leading contribution of the force constants in this region and allow the interlayer interactions similar to the Lennard-Jones potential, a large value of \( d \) should be chosen. The phonon dispersions using the traditional partition method and the proposed partition method with \( d = 9 \) are presented in Figs. 2(c) and (d) for the LDA and DFT-D2 functionals, respectively. The effect of the proposed partition can be found to improve the low-frequency part of phonon dispersion, for example, turning the imaginary frequencies obtained from the traditional method into the positive ones, while the high-frequency part of phonon dispersion described by the short-range interactions remains unaffected. As shown in Fig. 2(c), all of the imaginary frequencies become positive at \( d = 9 \) for the LDA case. For the DFT-D2 case, the imaginary frequencies around \( \Gamma \) cannot be further improved by the partition method with a larger value of \( d \) due to the numerical noise encountered in the calculation. Even with the adoption of the primitive unit cell for the phonon calculations, the phonon dispersion between \( \Gamma \) and \( K \) obtained from the properly partitioned force constants is in good agreement with the dispersion between \( \Gamma \) and \( K \) obtained from the properly partitioned force constants in this region and allow the interlayer interactions similar to the Lennard-Jones potential.\cite{m2} Although the frequency of the breathing mode is well described by DFT-D2, we note that not all the frequencies obtained by DFT-D2 are closer to the reported frequencies. For the modes with the frequency of 896.36 cm\(^{-1}\) at \( \Gamma \) both LDA and DFT-D2 give lower frequencies, namely, 860.48 cm\(^{-1}\) and 850.26 cm\(^{-1}\), respectively. The calculated LDA and DFT-D2 phonon dispersions together with the reported frequencies using different approaches are expected to be useful for the interpretation of experimental data in the future.

IV. Summary

We have addressed the importance of proper partition of force constants in phonon studies using the supercell force-constant approach. Without a proper partition, the resultant phonon frequencies and vibrations could be unphysical. Using a simple model structure, we have shown that applying symmetry operations can improve the phonon properties but should be used with caution. Otherwise, all of the phonon properties, including those at the commensurate \( q \) points, are affected. While there are abundant ways to partition the force constants as can be confirmed by the discussed formulation, we propose a simple way to partition the force constants based only on the interatomic distance and translational symmetry. The phonon dispersion of NaCl calculated by the \( 2 \times 2 \times 2 \) supercell with the properly partitioned force constants is demonstrated to be consistent with that obtained by the linear response method. The partition method is compatible with the mixed-space method for delivering accurate LO-TO splitting. We also show that the phonon frequencies could become imaginary even using the mixed-space approach if the interatomic force constants are not properly partitioned in NaCl. Finally, we present the phonon dispersion of twisted bilayer graphene with a rotational angle of 21.8° calculated from first principles. The obtained phonon dispersions from the primitive-cell calculations are consistent with the reported result using the Born von Karman model and the Lennard-Jones potential. The partition method can also be guided by symmetry operations and/or force-constant models as long as the sum rule (Eq. 7) is satisfied.

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