Phonon dispersion of binary alloys with auxiliary coherent potential approximation

Zihan Cheng, Mankun Sang, Jianxiong Zhai, and Youqi Ke[*]
School of Physical Science and Technology, ShanghaiTech University, Shanghai, 201210, China

(Received 1 September 2019; revised manuscript received 21 November 2019; published 27 December 2019)

We report the auxiliary coherent potential approximation (ACPA) for calculating the phonon dispersion of three-dimensional alloys with both mass and force-constant disorders. To obtain the average spectra function of disordered alloys, the average coherent scattering structure factors are derived from the auxiliary coherent medium in single-site approximation. We provide an analytical proof of the sum rule in the auxiliary coherent medium, which ensures the analyticity of physical properties. To demonstrate the accuracy and applicability of the ACPA method, we apply the ACPA to calculate phonon dispersion of several alloys, including CuPd CuAu, PdFe, and NiPt with different disorder concentrations. We find the ACPA phonon dispersion results agree very well with the itinerant coherent potential approximation calculations and experimental measurements. The approximate separable force-constant model used in the ACPA can very well represent the first-principles disordered force constants, presenting minor or even negligible influence on the phonon dispersion of the alloy. The ACPA model features easy implementation and high computational efficiency, providing an effective method for simulating vibrational properties of realistic alloys.

DOI: 10.1103/PhysRevB.100.214206

I. INTRODUCTION

Understanding disordered lattice vibration is important for many key properties of realistic materials containing inevitable disorders, including light adsorption, thermodynamics, superconductivity, phase transition, thermal conduction, and heat capacity [1]. However, effectively treating atomic disorders has been a longstanding challenge for theoretical simulation of disordered lattice vibrations, making many important phenomena induced by disorders poorly understood or even unexplored [2–4]. As known, the mean-field-type method, coherent potential approximation (CPA) [5,6], has achieved important success in calculating the electronic properties of disordered materials by combining with first-principles methods [7–16] and its various extensions to include off-diagonal disorders [17–25] and cluster effects and nonlocal correlations [26–28], and nonequilibrium statistics [29–34]. However, extending CPA to disorder lattice vibration faces an important difficulty: the diagonal and off-diagonal disorder in the vibrational Hamiltonian is connected by the force-constant sum rule $\Phi_{ii} = -\sum_{j\neq i}\Phi_{ij}$ (where Φ is the force-constant matrix). For the lattice vibration, satisfaction of the force-constant sum rule is required by the law of momentum conservation and is thus important for correctly calculating vibrational properties [35]. Presently, the conventional CPA method [5] can only be applied to calculate the effects of mass disorder, such as isotopes without force-constant randomness [36]. Therefore, the approach to effectively handle the force-constant disorder, as one of the key disorders responsible for many important phenomena [37], is key for simulating disordered lattice vibration.

With great theoretical efforts in past decades, considerable progress has been achieved for treating both mass and force-constant disorders. Based on the augmented-space formalism (ASF) proposed by Mookerjee [38], the augmented-space recursion method (ASR) [24] and the itinerant coherent potential approximation (ICPA) [23] have been developed. Presently, the ASR method has been applied with reasonable success to different alloys [39–42], and the ICPA method has reproduced the phonon dispersion for several alloys [43,44,46]. As an important advantage, ASR and ICPA methods can provide the exact representation of the disordered force constants for vibrational systems with the sum rule obeyed. However, in comparison with the wide applications of CPA in electronic systems, applications reported with ASR and ICPA are rather limited, due to the fact that ASR and ICPA are not easy to implement and computationally expensive. Very recently, the authors proposed a new method, called the auxiliary coherent potential approximation (ACPA) [25], to treat both the mass and force-constant disorder. The ACPA is based on an approximate force-constant decomposition that transforms the force-constant disorder (correlating both the diagonal and off-diagonal elements) into a diagonal-like (single-site-dependent) disorder of an auxiliary system, and then a conventional CPA algorithm can be applied to obtain the auxiliary coherent medium to give averaged physical properties. The ACPA thus features advantages of easy implementation and high computational efficiency, similar to the conventional CPA used for simulating electronic system. Due to the feature of single-site disorder, the ACPA is feasible to combine with diagrammatic techniques and various cluster approximations to further account for the effects of clusters and nonlocal correlations. Presently, the ACPA method has been demonstrated only for a 1D disorder atomic chain in which molecular ACPA can produce results close to the exact results as the cluster size increases.

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[*] keyq@shanghaitech.edu.cn

However, there are still important concerns remaining, including the applicability of the ACPA for realistic three-dimensional alloys, and how well the results compare with ICPA and experimental measurements, and whether the sum rule is satisfied in the auxiliary coherent medium. In this work, we will present our formulation and implementation of the ACPA for calculating the phonon dispersion of alloys in 3D and compare our results for different alloys with ICPA calculations and experiments. For realistic alloys, we show the ACPA results in single-site approximation agree well with the ICPA and experimental measurements, demonstrating the important applicability of the ACPA. We also provide an analytical proof of the sum rule in the auxiliary coherent medium, ensuring the analyticity of the physical system.

The rest of the paper is organized as follows: In Sec. II, we present the formulation and implementation of the auxiliary CPA for simulating the three-dimensional realistic materials with both mass and force-constant disorders. In Sec. III, we provide a derivation of the coherent scattering structure factor to give the phonon spectra function of alloys with the ACPA. Section IV provides a proof of the sum rule for the coherent medium in auxiliary CPA. In Sec. V we apply auxiliary CPA to calculate the phonon dispersion curves for several alloy materials and compare our results with previous ICPA calculations and experimental measurements. We conclude in Sec. VI and provide additional information in Appendix A.

II. AUXILIARY COHERENT POTENTIAL APPROXIMATION FOR DISORDERED VIBRATION

For the completeness of our discussion, we will review the basic ideas in the ACPA [25] and present the formulation and implementation of the ACPA for simulating realistic materials with atomic disorders. We consider a random alloy A_xB_{1-x} in which the mass and force constants are random quantities, e.g., the mass takes m_i^Q, $(Q = A, B)$ and the force constant takes the possible values $k_{ij}^{QQ'}(Q, Q' = A, B)$ determined by the atomic occupations Q and Q' of sites i and j. To introduce an auxiliary CPA for realistic binary alloys, we adopt an approximate separable 3D force-constant model for the random force constant

$$k^{Q,Q'}_{i\alpha j\beta} = x_i^Q S_{i\alpha j\beta} x_j^{Q'} + \lambda_{i\alpha j\beta} \quad (i \neq j), \tag{1}$$

where $S_{i\alpha j\beta}$ and $\lambda_{i\alpha j\beta}$ with α and β denoting the bases are independent of the atomic occupations on sites i and j, and the information of the atomic occupant on the ith site is contained in the quantity x_i^Q $(Q = A, B)$. Then, the force-constant matrix for a specific random system can be given by

$$\Phi_{i\alpha j\beta} = -k^{Q,Q'}_{i\alpha j\beta}, \quad (i \neq j), \tag{2}$$

$$\Phi_{i\alpha i\beta} = \sum_{j\neq i} k^{Q,Q'}_{i\alpha j\beta}, \tag{3}$$

satisfying the sum rule that correlates the diagonal and off-diagonal disordered elements.

In Eq. (1), the introduced force-constant model combines geometric-scaled (first term) and the virtual-crystal (second term) models to overcome the shortcomings of both models, featuring higher accuracy and more general applicability. As we will show in Sec. V, the model in Eq. (1) can provide a rather accurate representation of the disordered force constants for realistic alloys. Note that, for present applications, we use the transferable force-constant model (TFC), which takes the symmetric form [47]

$$k = \begin{bmatrix} k_s & 0 & 0 \\ 0 & k_b & 0 \\ 0 & 0 & k_b \end{bmatrix}, \tag{4}$$

where k_s and k_b denote the respective force constants describing the bond stretching and bending. The adopted TFC [47] model features important transferability of force constants, providing a simplistic approach to determine the averaged force constants for alloys. However, for complex environments [48], force constants have a strong dependence on the local chemical environment, limiting the applicability of the TFC model. It should be mentioned that Eq. (1) is a general model and is not limited only to TFC with the form of Eq. (4). For the study of real alloys, the parameters x, S, λ in Eq. (1) can be obtained by fitting to the TFC, namely, k^{AA}, k^{AB}, and k^{BB} from first principles, with a numerical optimization method as shown in Appendix A.

As an important result for using the separable force-constant model of Eq. (1), the force-constant matrix of a specific system can be rewritten as

$$\Phi = XK, \tag{5}$$

with the matrix element explicitly given by $\Phi_{i\alpha j\beta} = X_{ii}K_{i\alpha j\beta}$. Here, X is a diagonal matrix $X_{ij} = x_i\delta_{ij}$, and the K matrix is defined as, for an off-diagonal element,

$$K_{i\alpha j\beta} = -\left(S_{i\alpha j\beta}x_j + \frac{\lambda_{i\alpha j\beta}}{x_i}\right) \quad (i \neq j), \tag{6}$$

and for a diagonal element,

$$K_{i\alpha i\beta} = \frac{1}{x_i}\sum_{j\neq i} k_{i\alpha j\beta} = \sum_{j\neq i}\left(S_{i\alpha j\beta}x_j + \frac{\lambda_{i\alpha j\beta}}{x_i}\right). \tag{7}$$

It is clearly seen that K matrix satisfies $\sum_j K_{i\alpha j\beta} = 0$. This sum rule of K ensures that the force-constant sum rule is always satisfied in our further derivation. Since the terms containing x_i and x_j are linearly independent in Eqs. (6) and (7), we can further decompose K into a sum of single-site quantities, namely,

$$K = \sum_i \widetilde{K}^i, \tag{8}$$

where the matrix $\widetilde{K}^i$ contains all the contributions only associating with x_i. For a site i with Z nonzero force-constant neighbors, the $\widetilde{K}^i$ matrix is of the size $d(Z+1) \times d(Z+1)$, where d is the dimension of the system (hereafter the quantities with a tilde are all $d(Z+1) \times d(Z+1)$ matrices). For an explicit form, $\widetilde{K}^i$ reads

$$\widetilde{K}^i_{j\alpha j\beta} = S_{j\alpha i\beta}x_i, \quad (j \neq i); \tag{9}$$

$$\widetilde{K}^i_{j\alpha i\beta} = -\,S_{j\alpha i\beta}x_i, \quad (j \neq i); \tag{10}$$

$$\widetilde{K}^i_{i\alpha j\beta} = -\,\frac{\lambda_{i\alpha j\beta}}{x_i}, \quad (j \neq i); \tag{11}$$

$$\widetilde{K}^i_{i\alpha i\beta} = \sum_j \frac{\lambda_{i\alpha j\beta}}{x_i}; \tag{12}$$

$$\widetilde{K}^i_{j\alpha j'\beta} = 0, \quad (j, j' \neq i). \tag{13}$$

It is clear that $\widetilde{K}_i$ satisfies the sum rule, namely, $\sum_J \widetilde{K}^i_{I\alpha J\beta} = 0$, ensuring the force-constant sum rule.

As an important consequence, the Green's function, describing the displacement-displacement correlator, can be written as

$$G = (m\omega^2 - D)^{-1} = gX^{-1}, \tag{14}$$

with the auxiliary Green's function defined as

$$g = (X^{-1}m\omega^2 - K)^{-1}. \tag{15}$$

In this form, we can define the corresponding auxiliary Hamiltonian matrix $P \equiv X^{-1}m\omega^2 - K$. Importantly, $P = \sum_i \widetilde{P}^i$, and the single-site quantity $\widetilde{P}^i$ is given by

$$\widetilde{P}^i_{I\alpha J\beta} = x_i^{-1} m_i \omega^2 \delta_{Ii}\delta_{Ji} - \widetilde{K}^i_{I\alpha J\beta}. \tag{16}$$

In such a form, the general disorders in lattice vibration are reduced to a diagonal-like disorder problem, enabling the implementation of the conventional CPA to calculate the disorder-averaged auxiliary Green's function g to derive physical properties of a disordered system.

To implement the CPA for vibrations in realistic materials with general disorders, we introduce a effective medium with coherent function $\mathcal{P} = \sum_i \widetilde{\mathcal{P}}^i$ to give the averaged auxiliary Green's function, namely, $\mathscr{g} = \mathcal{P}^{-1}$. Here, the single-site coherent function $\widetilde{\mathcal{P}}^i$ has the same size of $d(Z+1) \times d(Z+1)$ as the quantity $\widetilde{K}^i$ and $\widetilde{P}^i$, differing from the site-diagonal coherent function in the conventional CPA method [5,6].

Under the single-site approximation, the CPA condition for obtaining $\widetilde{\mathcal{P}}^i$ reads [49]

$$\langle \widetilde{t}^i \rangle = \sum_{Q=A,B} c^Q \widetilde{t}^{i,Q} = 0, \tag{17}$$

where

$$\widetilde{t}^{i,Q} = (\widetilde{\mathcal{P}}^i - \widetilde{P}^{i,Q})[\mathbf{1} - \widetilde{\mathscr{g}}_{ii}(\widetilde{\mathcal{P}}^i - \widetilde{P}^{i,Q})]^{-1}. \tag{18}$$

Note that considering the size of $\widetilde{\mathcal{P}}^i - \widetilde{P}^{i,Q}$, $\widetilde{\mathscr{g}}_{ii}$ refers to the part in $\mathscr{g}$ containing site i and its neighbors with size $d(Z+1) \times d(Z+1)$, different from the site-diagonal element of $\mathscr{g}$ used in conventional implementations. To stably solve the single-site CPA equations, we adopt the method with a coherent interactor $\widetilde{\Omega}^i$ [50], whose size is also $d(Z+1) \times d(Z+1)$. We summarize our procedures for implementing CPA and the computation of related quantities as follows:

(1) Initialize a $\widetilde{\mathcal{P}}^i$ with the average-t-matrix approximation (ATA) [51].

(2) Use $\mathcal{P} = \sum_i \widetilde{\mathcal{P}}^i$ to obtain the coherent function $\mathcal{P}$ with lattice translational invariance.

(3) Apply the lattice Fourier transformation to obtain

$$\mathcal{P}_{\boldsymbol{b}\boldsymbol{b}'}(\boldsymbol{k}) = \sum_{\boldsymbol{T}} \mathcal{P}_{\boldsymbol{b},\boldsymbol{b}'+\boldsymbol{T}} e^{i\boldsymbol{k}\cdot\boldsymbol{T}} \tag{19}$$

for $\boldsymbol{k}$ in the Brillouin zone (BZ), where $\boldsymbol{b}, \boldsymbol{b}'$ refer to the basis vectors in the unit cell, $\boldsymbol{T}$ denotes the translational vectors, and $\mathcal{P}_{\boldsymbol{b},\boldsymbol{b}'+\boldsymbol{T}}$ refers to the element of full matrix $\mathcal{P}$ [the size of $\mathcal{P}(\boldsymbol{k})$ is $ds \times ds$, where s refers to the number of sites in a unit cell].

(4) Calculate $\widetilde{\mathscr{g}}_{ii}$ by using the relations

$$\mathscr{g}(\boldsymbol{k}) = \mathcal{P}(\boldsymbol{k})^{-1} \tag{20}$$

and

$$\widetilde{\mathscr{g}}_{ii,\boldsymbol{b}+\boldsymbol{T},\boldsymbol{b}'+\boldsymbol{T}'} = \frac{1}{\Omega}\int d\boldsymbol{k}\, \mathscr{g}_{\boldsymbol{b}\boldsymbol{b}'}(\boldsymbol{k}) e^{-i\boldsymbol{k}\cdot(\boldsymbol{T}'-\boldsymbol{T})}, \tag{21}$$

where Ω is the volume of Brillouin zone, and $\boldsymbol{b}+\boldsymbol{T}, \boldsymbol{b}'+\boldsymbol{T}'$ denotes the lattice sites including the i site and its nonzero force-constant neighbors.

(5) Calculate the coherent interactor $\widetilde{\Omega}^i$ using

$$\widetilde{\Omega}^i = \widetilde{\mathcal{P}}^i - \widetilde{\mathscr{g}}_{ii}^{-1}. \tag{22}$$

(6) Calculate an output $\widetilde{\mathcal{P}}^i$ with

$$\widetilde{\mathcal{P}}^i = \left[\sum_Q c_i^Q (\widetilde{P}^{i,Q} - \widetilde{\Omega}^i)^{-1}\right]^{-1} + \widetilde{\Omega}^i. \tag{23}$$

(7) Obtain a new input by properly mixing the input $\widetilde{\mathcal{P}}^i$ and output $\widetilde{\mathcal{P}}^i$ and then repeat the above calculations from the second step until $\widetilde{\mathcal{P}}^i$ is converged.

III. COHERENT SCATTERING STRUCTURE FACTOR

In this section, based on the formalism of the ACPA, we derive the averaged coherent scattering structure factor to determine the phonon dispersion of alloys, aiming to make direct comparison with experimental measurements. The averaged coherent scattering structure factor can be defined as [24]

$$\langle S_\lambda(\boldsymbol{k}, \omega)\rangle_{\text{coh}} = -\frac{1}{\pi}\sum_{QQ'} d^Q d^{Q'} \operatorname{Im}\big\langle \mathcal{G}_\lambda^{QQ'}(\boldsymbol{k}, \omega)\big\rangle, \tag{24}$$

where λ is a normal-mode branch index, and d^Q is the coherent scattering length for the species Q. The partial Green's function $\langle \mathcal{G}^{QQ'}(\boldsymbol{k}, \omega)\rangle$ corresponding to species Q and Q' is given by the lattice Fourier transformation

$$\big\langle \mathcal{G}^{QQ'}_{\boldsymbol{b}\boldsymbol{b}'}(\boldsymbol{k}, \omega)\big\rangle = \sum_{\boldsymbol{T}} \big\langle \mathcal{G}^{QQ'}_{\boldsymbol{b},\boldsymbol{b}'+\boldsymbol{T}}(\omega)\big\rangle e^{i\boldsymbol{k}\cdot\boldsymbol{T}}, \tag{25}$$

where $\boldsymbol{b}, \boldsymbol{b}'$ refers to the basis vectors in the unit cell, $\boldsymbol{T}$ denotes the translational vector, and $\langle \mathcal{G}^{QQ'}_{ij}\rangle$ is related to the conditionally averaged Green's function by

$$\big\langle \mathcal{G}^{QQ'}_{ij}\big\rangle = c^Q\big\langle G^Q_{ii}\big\rangle \delta_{ij}\delta_{QQ'} + c^Q c^{Q'}\big\langle G^{QQ'}_{ij}\big\rangle(1 - \delta_{ij}), \tag{26}$$

where $\langle G^{QQ}_{ii}\rangle$ corresponds to the disordered system with Q fixed on the i site, and $\langle G^{QQ'}_{ij}\rangle$ corresponds to the disordered system with Q and Q' fixed on the respective sites i and j.

According to Eq. (14), the conditionally averaged Green's function is connected to the conditionally averaged auxiliary Green's function by the relations

$$\big\langle G^{QQ}_{ii}\big\rangle = \big\langle g^Q_{ii}\big\rangle \big(x^Q_i\big)^{-1}, \tag{27}$$

$$\big\langle G^{QQ'}_{ij}\big\rangle = \big\langle g^{QQ'}_{ij}\big\rangle \big(x^{Q'}_j\big)^{-1} (i \neq j). \tag{28}$$

Here, we adopt the occupation-operator technique to calculate the conditionally averaged auxiliary Green's functions $\langle g^{Q}_{ii}\rangle$ and $\langle g^{QQ'}_{ij}\rangle$ [52]. (For completeness, we include a derivation in Appendix B; here we summarize the major results.) For the quantity $\langle g^{Q}_{ii}\rangle$, we can find, in single-site approximation (SSA),

$$\langle g^{Q}_{ii}\rangle = (\mathscr{g} + \mathscr{g}\widetilde{t}^{i,Q}\mathscr{g})_{ii}, \tag{29}$$

which describes a system containing a site i with a fixed Q atom embedded in a coherent medium after averaging. Furthermore, for $\langle g^{QQ'}_{ij}\rangle$, describing an averaged system in which i, j sites are fixed to the respective elements Q, Q', we obtain the SSA result

$$\langle g^{QQ'}_{ij}\rangle = [(1 + \mathscr{g}\widetilde{t}^{i,Q})\mathscr{g}(1 + \widetilde{t}^{j,Q'}\mathscr{g})]_{ij}. \tag{30}$$

It is clear that, after obtaining the conditionally averaged auxiliary Green's functions with $\mathscr{g}$, we can calculate averaged coherent scattering structure factors, namely, $\langle S_{\lambda}(\boldsymbol{k},\omega)\rangle_{\mathrm{coh}}$ for each $\boldsymbol{k}$, to obtain the phonon dispersion curves.

IV. SUM RULE IN COHERENT MEDIUM

In the above, we have presented the formalism of the ACPA for computing the disorder averaged Green's function. Here, we provide an analytical proof of the fact that the auxiliary coherent medium satisfies the sum rule, guaranteeing the momentum conservation which is important for correctly simulating physical properties [53]. To do so, we can rewrite the coherent potential $\widetilde{\mathcal{P}}^{i}$ into two parts: a coherent matrix $\widetilde{\mathcal{K}}^{i}(\omega)$ and a term with effective $\widetilde{\mathcal{M}}^{i}(\omega)$ times ω^2, similar to Eq. (16):

$$\widetilde{\mathcal{P}}^{i} = \widetilde{\mathcal{M}}^{i}\omega^2 - \widetilde{\mathcal{K}}^{i}. \tag{31}$$

As we have shown for a system, the force-constant sum rule is ensured by the fact that the $\widetilde{K}^{i}$ matrix elements satisfy the important relation $\sum_J \widetilde{K}^{i}_{IJ} = 0$ for each site. A similar sum rule for the coherent matrix $\widetilde{\mathcal{K}}^{i}$ should promise the satisfaction of momentum conservation law in the coherent medium. Therefore, we need to prove the existence of such $\widetilde{\mathcal{M}}^{i}$ and $\widetilde{\mathcal{K}}^{i}$ with the relation

$$\sum_J \widetilde{\mathcal{K}}^{i}_{IJ} = 0, \tag{32}$$

where I, J denote the i site and its neighbors. We are going to test the relations in Eqs. (31)–(32) with various approximations, including a virtual crystal approximation (VCA), the average-t-matrix approximation, and then the coherent potential approximation.

To proceed, we start from the VCA, namely,

$$\widetilde{K}^{i}_{\mathrm{VCA}} = -\widetilde{P}^{i}_{\mathrm{VCA}} + \widetilde{M}^{i}_{\mathrm{VCA}}\omega^2 = \sum_Q c^{Q}\widetilde{K}^{i,Q}, \tag{33}$$

where $\widetilde{M}^{i}_{\mathrm{VCA},IJ} = \langle x_i^{-1}m_i\rangle\delta_{Ii}\delta_{Ji}$. By considering Eqs. (9)–(12), we have, for each element Q at site i,

$$\sum_J \widetilde{K}^{i,Q}_{IJ} = 0, \tag{34}$$

and together with Eq. (33), we can find

$$\sum_J \widetilde{K}^{i}_{\mathrm{VCA},IJ} = 0, \tag{35}$$

which ensures the sum rule for a medium with VCA.

Taking the $\widetilde{P}^{i}_{\mathrm{VCA}}$ as the reference, we have the $\widetilde{P}^{i}_{\mathrm{ATA}}$ as follows:

$$\widetilde{P}^{i}_{\mathrm{ATA}} = \widetilde{P}^{i}_{\mathrm{VCA}} - (1 + \langle\widetilde{t}^{i}\rangle\widetilde{g}_{\mathrm{VCA},ii})^{-1}\langle\widetilde{t}^{i}\rangle, \tag{36}$$

where $\langle\widetilde{t}^{i}\rangle = \sum_Q c^{Q}\widetilde{t}^{i,Q}$, and

$$\widetilde{t}^{i,Q} = \big[1 - \big(\widetilde{P}^{i}_{\mathrm{VCA}} - \widetilde{P}^{i,Q}\big)\widetilde{g}_{\mathrm{VCA},ii}\big]^{-1}\big(\widetilde{P}^{i}_{\mathrm{VCA}} - \widetilde{P}^{i,Q}\big), \tag{37}$$

which can be decomposed as

$$\widetilde{t}^{i,Q} = -\widetilde{s}^{i,Q} + \widetilde{p}^{i,Q}\omega^2, \tag{38}$$

where $\widetilde{s}^{i,Q} = [1 - (\widetilde{P}^{i}_{\mathrm{VCA}} - \widetilde{P}^{i,Q})\widetilde{g}_{\mathrm{VCA},ii}]^{-1}(\widetilde{K}^{i}_{\mathrm{VCA}} - \widetilde{K}^{i,Q})$ and $\widetilde{p}^{i,Q} = [1 - (\widetilde{P}^{i}_{\mathrm{VCA}} - \widetilde{P}^{i,Q})\widetilde{g}_{\mathrm{VCA},ii}]^{-1}(\widetilde{M}^{i}_{\mathrm{VCA}} - \widetilde{M}^{i,Q})$ and $\widetilde{M}^{i,Q}_{IJ} = (x_i^{Q})^{-1}m_i^{Q}\delta_{Ii}\delta_{Ji}$.

To proceed, we introduce a lemma: If matrix A satisfying $\sum_j A_{ij} = 0$, for any B matrix, the matrix product BA always satisfies

$$\sum_j (BA)_{ij} = 0. \tag{39}$$

Since $\sum_J(\widetilde{K}^{i}_{\mathrm{VCA}} - \widetilde{K}^{i,Q})_{IJ} = 0$, the lemma Eq. (39) leads to

$$\sum_J \widetilde{s}^{i,Q}_{IJ} = 0. \tag{40}$$

Thus we can write $\widetilde{K}^{i}_{\mathrm{ATA}}$ as

$$\widetilde{K}^{i}_{\mathrm{ATA}} = \widetilde{K}^{i}_{\mathrm{VCA}} - (1 + \langle\widetilde{t}^{i}\rangle\widetilde{g}_{\mathrm{VCA},ii})^{-1}\langle\widetilde{s}^{i}\rangle, \tag{41}$$

where $\langle\widetilde{s}^{i}\rangle = \sum_Q c^{Q}\widetilde{s}^{i,Q}$. With Eqs. (35) and (40), the lemma promises the sum rule for $\widetilde{K}^{i}_{\mathrm{ATA}}$:

$$\sum_J \widetilde{K}^{i}_{\mathrm{ATA},IJ} = 0. \tag{42}$$

With Eq. (36), the corresponding $\widetilde{M}^{i}_{\mathrm{ATA}}$ is given by

$$\widetilde{M}^{i}_{\mathrm{ATA}} = \widetilde{M}^{i}_{\mathrm{VCA}} - (1 + \langle\widetilde{t}^{i}\rangle\widetilde{g}_{\mathrm{VCA},ii})^{-1}\langle\widetilde{p}^{i}\rangle, \tag{43}$$

where $\langle\widetilde{p}^{i}\rangle = \sum_Q c^{Q}\widetilde{p}^{i,Q}$.

For the medium in CPA, by using Eq. (22), Eq. (23), and $\mathscr{g} = \mathcal{P}^{-1}$, an iterative equation similar to Eq. (36) can be obtained:

$$\widetilde{\mathcal{P}}^{i}_{N+1} = \widetilde{\mathcal{P}}^{i}_{N} - \big(1 + \langle\widetilde{t}^{i}_{N}\rangle\widetilde{g}_{N,ii}\big)^{-1}\langle\widetilde{t}^{i}_{N}\rangle, \tag{44}$$

where $\widetilde{t}^{i,Q}_{N} = [1 - (\widetilde{\mathcal{P}}^{i}_{N} - \widetilde{P}^{i,Q})\widetilde{g}^{i}_{N}]^{-1}(\widetilde{\mathcal{P}}^{i}_{N} - \widetilde{P}^{i,Q})$, and N denotes the Nth iteration step.

Similarly, from Eq. (44), $\widetilde{\mathcal{K}}^{i}_{N}$ and $\widetilde{\mathcal{M}}^{i}_{N}$ can be given as

$$\widetilde{\mathcal{K}}^{i}_{N+1} = \widetilde{\mathcal{K}}^{i}_{N} - \big(1 + \langle\widetilde{t}^{i}_{N}\rangle\widetilde{g}_{N,ii}\big)^{-1}\langle\widetilde{s}^{i}_{N}\rangle, \tag{45}$$

where $\widetilde{s}^{i,Q}_{N} = [1 - (\widetilde{\mathcal{P}}^{i}_{N} - \widetilde{P}^{i,Q})\widetilde{g}_{N,ii}]^{-1}(\widetilde{\mathcal{K}}^{i}_{N} - \widetilde{K}^{i,Q})$, and

$$\widetilde{\mathcal{M}}^{i}_{N+1} = \widetilde{\mathcal{M}}^{i}_{N} - \big(1 + \langle\widetilde{t}^{i}_{N}\rangle\widetilde{g}_{N,ii}\big)^{-1}\langle\widetilde{p}^{i}_{N}\rangle, \tag{46}$$

where $\widetilde{p}^{i,Q}_{N} = [1 - (\widetilde{\mathcal{P}}^{i}_{N} - \widetilde{P}^{i,Q})\widetilde{g}_{N,ii}]^{-1}(\widetilde{\mathcal{M}}^{i}_{N} - \widetilde{M}^{i,Q})$.

We usually choose ATA as the initialization of CPA, i.e., $\widetilde{\mathcal{P}}^{i}_{1} = \widetilde{P}^{i}_{\mathrm{ATA}}$, $\widetilde{\mathcal{K}}^{i}_{1} = \widetilde{K}^{i}_{\mathrm{ATA}}$, and $\widetilde{\mathcal{M}}^{i}_{1} = \widetilde{M}^{i}_{\mathrm{ATA}}$. Then, with the lemma in Eq. (39), it is easy to obtain the sum rule for $\widetilde{\mathcal{K}}^i_N$ of any iteration:

$$\sum_J \widetilde{\mathcal{K}}^i_{N,IJ} = 0. \tag{47}$$

As an important result, when the iteration converges, the coherent potential $\widetilde{\mathcal{P}}^i$ can be decomposed as $\widetilde{\mathcal{P}}^i = \widetilde{\mathcal{M}}^i\omega^2 - \widetilde{\mathcal{K}}^i$, with $\widetilde{\mathcal{K}}^i$ satisfying the sum rule.

Specifically, when $m^A/x^A = m^B/x^B$, namely, only K has disorder, we have $\widetilde{M}^{i,A} = \widetilde{M}^{i,B} = \widetilde{M}^i_{\mathrm{VCA}}$. Then, with Eqs. (43) and (46), we can find $\widetilde{\mathcal{M}}^i = \widetilde{M}^i_{\mathrm{ATA}} = \widetilde{M}^{i,A}$, and the sum rule of $\widetilde{\mathcal{K}}^i$ for the coherent medium directly corresponds to the sum rule of $\widetilde{K}^i$ for a specific system.

As a direct consequence, $\mathcal{K}$ satisfying the sum rule ensures the existence of one acoustic mode at $(\omega = 0, \boldsymbol{k} = 0)$ for the coherent medium. To show this, we consider the secular equation of the coherent medium

$$\det|\mathcal{K}_{\alpha\beta\boldsymbol{b}\boldsymbol{b}'}(\boldsymbol{k}) - \mathcal{M}\omega^2\delta_{\alpha\beta}\delta_{\boldsymbol{b}\boldsymbol{b}'}| = 0, \tag{48}$$

where $\boldsymbol{b}, \boldsymbol{b}'$ denotes the basis vectors of the unit cell.

When $\boldsymbol{k} = \boldsymbol{0}$, one may obtain, for any $\boldsymbol{b}$,

$$\sum_{\boldsymbol{b}'} \mathcal{K}_{\alpha\beta\boldsymbol{b}\boldsymbol{b}'}(\boldsymbol{0}) = \sum_{\boldsymbol{b}',\boldsymbol{T}} \mathcal{K}_{\alpha\beta\boldsymbol{b},\boldsymbol{b}'+\boldsymbol{T}} = 0, \tag{49}$$

and as a result, $\det|\mathcal{K}_{\alpha\beta\boldsymbol{b}\boldsymbol{b}'}(\boldsymbol{0})| = 0$ [54]. Therefore, $\omega = 0$ is a solution to the secular equation at $\boldsymbol{k} = 0$. If the sum rule of $\mathcal{K}$ is broken, there will be no zero-frequency mode at $\boldsymbol{k} = \boldsymbol{0}$. Thus the satisfaction of the sum rule is significant for the correct simulation of phonon dispersion of disordered materials.

In addition, we note that the auxiliary Hamiltonian $P \equiv X^{-1}m\omega^2 - K$ in general does not conserve the symmetry $P_{ij} = P_{ji}$ for a specific system. However, we have numerically checked another important symmetry, namely, the averaged physical Green's function $\langle G_{i\alpha,j\beta}\rangle = \langle G_{j\beta,i\alpha}\rangle$ [computed by using the relation $\langle G_{i,j}\rangle = \sum_{QQ'} c_i^Q c_j^{Q'} \langle G_{i,j}^{QQ'}\rangle$ and Eqs. (28) and (30)], which is strictly satisfied for a disordered alloys, and this symmetry reflects the symmetry of the corresponding effective physical Hamiltonian after disorder averaging.

V. NUMERICAL RESULTS AND DISCUSSIONS

In this section, to demonstrate the applicability of the ACPA method for the disordered lattice vibrations in realistic alloys, we calculate the phonon dispersion for different alloys including fcc $\mathrm{Cu_{0.715}Pd_{0.285}}$, $\mathrm{Cu_{0.75}Au_{0.25}}$, $\mathrm{Pd_{0.96}Fe_{0.04}}$, $\mathrm{Ni_{0.95}Pt_{0.05}}$, $\mathrm{Ni_{0.25}Pt_{0.75}}$, $\mathrm{Ni_{0.30}Pt_{0.70}}$. We compare ACPA results with the calculations of ICPA [43–45] and experimental measurements [55–61]. In our simulation, we did not calculate force constants by ourselves, and we use the same TFCs as ICPA calculations in Refs. [43–45], which are obtained from first principles with density functional perturbation theory [62]. The reliability of the TFC model for the alloys we investigate here has been well demonstrated in Refs. [43–45].

To enable ACPA calculation, we use an optimization method to obtain the parameters, including $x_{A/B}$, S, and λ, in the approximate force-constant model of Eq. (1) for different alloys, by fitting to the first-principles force constants (for optimization procedures, see Appendix A). In the present optimization, we use the equal weighting for k_s and k_b to minimize relative errors of the optimized force constants. In Table I, for all six alloys, we present the optimized parameters and the obtained approximate force constants together with the original first-principles results. It is clearly seen that the approximate separable force-constant model, namely, Eq. (1), can provide a very good representation of the first-principles force constants $\widetilde{k}^{Q,Q'}_{s/b}$ for different Q, Q' pairs of all six alloys. In particular, for all the alloys, the relative error between the optimized force constants $\widetilde{k}^{Q,Q'}$ and the first-principles result $k^{Q,Q'}$ is about a few percent for both stretching and bending components, providing an important basis for the applicability of the ACPA. For example, for the alloy $\mathrm{Cu_{0.75}Au_{0.25}}$, the optimized $\widetilde{k}^{\mathrm{CuCu}}_s$, $\widetilde{k}^{\mathrm{CuAu}}_s$, and $\widetilde{k}^{\mathrm{AuAu}}_s$ are the respective values of 22 707.20, 39 762.02, and 66 962.44 (dyn/cm), fitting very well to the respective original first-principles results 22 639.54, 40 107.48, and 66 595.32 (dyn/cm) [44]. It is also worth mentioning that the force constants of alloys with the same compositions have important dependence on the concentrations, as shown in Table I, and the force-constant model of Eq. (1) can work well for alloys with different concentrations. After the parameters in the separable force-constant model are determined, we can carry out the ACPA to obtain the physical properties of the alloys. It should be mentioned here that, as we will show in the following, the small error in the optimized force constants presents minor or even negligible effects in the phonon dispersion of different alloys.

The theoretical phonon dispersion $\omega(\boldsymbol{k})$ is obtained as the frequencies corresponding to the peaks of the averaged coherent scattering structure factor $\langle S_\lambda(\boldsymbol{k},\omega)\rangle_{\mathrm{coh}}$ in Eq. (24) for each $\boldsymbol{k}$. (For the used coherent scattering length of each element, see the Ref. [63]). We present the phonon dispersion curves along all the symmetry directions [including $(\zeta, 0, 0)$, $(\zeta, \zeta, 0)$, and (ζ, ζ, ζ)] in Fig. 1 for the $\mathrm{Cu_{0.715}Pd_{0.285}}$, $\mathrm{Cu_{0.75}Au_{0.25}}$, $\mathrm{Pd_{0.96}Fe_{0.04}}$, and $\mathrm{Ni_{0.95}Pt_{0.05}}$ and in Fig. 2 for $\mathrm{Ni_{0.25}Pt_{0.75}}$ and $\mathrm{Ni_{0.70}Pt_{0.30}}$ fcc alloys. For a fair comparison, we have also implemented the ICPA in our code, and ICPA calculations are carried out for both the original and optimized force constants (with the same simulation settings as the ACPA). We find our ICPA calculations reproduce very well the ICPA results in Refs. [43–45] for all six alloys. As shown in Figs. 1 and 2, despite the very different algorithm of the ACPA and the ICPA methods, results of the ACPA method (in red) present very good agreement with the calculations by ICPA (in black) for all the binary alloys, demonstrating the important applicability of the ACPA method for disordered lattice vibration. For example, in $\mathrm{Pd_{0.96}Fe_{0.04}}$ and $\mathrm{Ni_{0.95}Pt_{0.05}}$ with low concentration of disorder, the ACPA and the ICPA produce almost the same phonon dispersion. For the alloys with high disorder concentration, only minor deviation between the ACPA and the ICPA can be found in some small area of $\boldsymbol{k}$, for example, around [0.48,0,0] and [0.48,0.48,0] in $\mathrm{Cu_{0.75}Au_{0.25}}$, at about [1.0,0,0] in $\mathrm{Ni_{0.25}Pt_{0.75}}$ and $\mathrm{Ni_{0.70}Pt_{0.30}}$, as shown in Fig. 2. Here we attribute the small deviation between the ACPA (red) and the ICPA (black) to the difference in the two theoretical methods but not to the small difference between the original and approximate force constants used in the respective ICPA and ACPA calculations.

TABLE I. Force constants $k_{s/b}$ (in units of dyn cm^{-1}) for six alloys with first-principles methods using the TFC model [43–45], the optimized force constants $\widetilde{k}_{s/b}$, relative errors $e_{s/b}$, and the corresponding parameters in the separable force-constant model [25] ($x_A = 1.0$).

A_cB_{1-c}	Pair type	k_s	k_b	$\widetilde{k}_s$	$\widetilde{k}_b$	e_s	e_b	x_B	S_s	S_b	λ_s	λ_b
$Cu_{0.715}Pd_{0.285}$	Cu-Cu	23981.67	−1289.29	24378.00	−1251.18	1.7%	3.0%	1.94430	12628.74	−379.57	11749.26	−871.61
	Cu-Pd	37793.93	−1528.49	36303.32	−1609.61	3.9%	5.3%					
	Pd-Pd	58285.63	−2372.90	59489.72	−2306.50	2.1%	2.8%					
$Cu_{0.75}Au_{0.25}$	Cu-Cu	22639.54	−1537.81	22707.20	−1535.19	0.3%	0.2%	1.59488	28669.37	−3418.67	−5962.18	1883.48
	Cu-Au	40107.48	−3546.24	39762.02	−3568.89	0.9%	0.6%					
	Au-Au	66595.32	−6844.91	66962.44	−6812.39	0.6%	0.5%					
$Pd_{0.96}Fe_{0.04}$	Pd-Pd	45925	−2424	45685.48	−2435.54	0.5%	0.5%	2.30555	−7491.07	433.14	53176.55	−2868.68
	Pd-Fe	35698	−1880	35905.51	−1870.05	0.6%	0.5%					
	Fe-Fe	13366	−566	13357.31	−566.30	0.7%	0.1%					
$Ni_{0.95}Pt_{0.05}$	Ni-Ni	32600	−378	32225.83	−378.88	1.1%	0.2%	1.576578	62185.18	−3561.68	−29959.35	3182.80
	Ni-Pt	65604	−2495	68080.44	−2432.47	3.8%	2.5%					
	Pt-Pt	128283	−5550	124608.02	−5670.11	2.9%	2.2%					
$Ni_{0.70}Pt_{0.30}$	Ni-Ni	23872	−482	23685.30	−483.12	0.8%	0.2%	1.75997	38508.94	−2611.84	−14823.64	2128.72
	Ni-Pt	51584	−2516	52950.94	−2448.04	2.6%	2.7%					
	Pt-Pt	106572	−5867	104457.59	−5961.44	2.0%	1.6%					
$Ni_{0.25}Pt_{0.75}$	Ni-Ni	11816	−608	11732.66	−610.41	0.7%	0.4%	2.05765	19313.41	−1767.08	−7580.75	1156.67
	Ni-Pt	31291	−2542	32159.49	−2479.36	2.8%	2.5%					
	Pt-Pt	75860	−6203	74190.75	−6325.01	2.2%	2.0%					

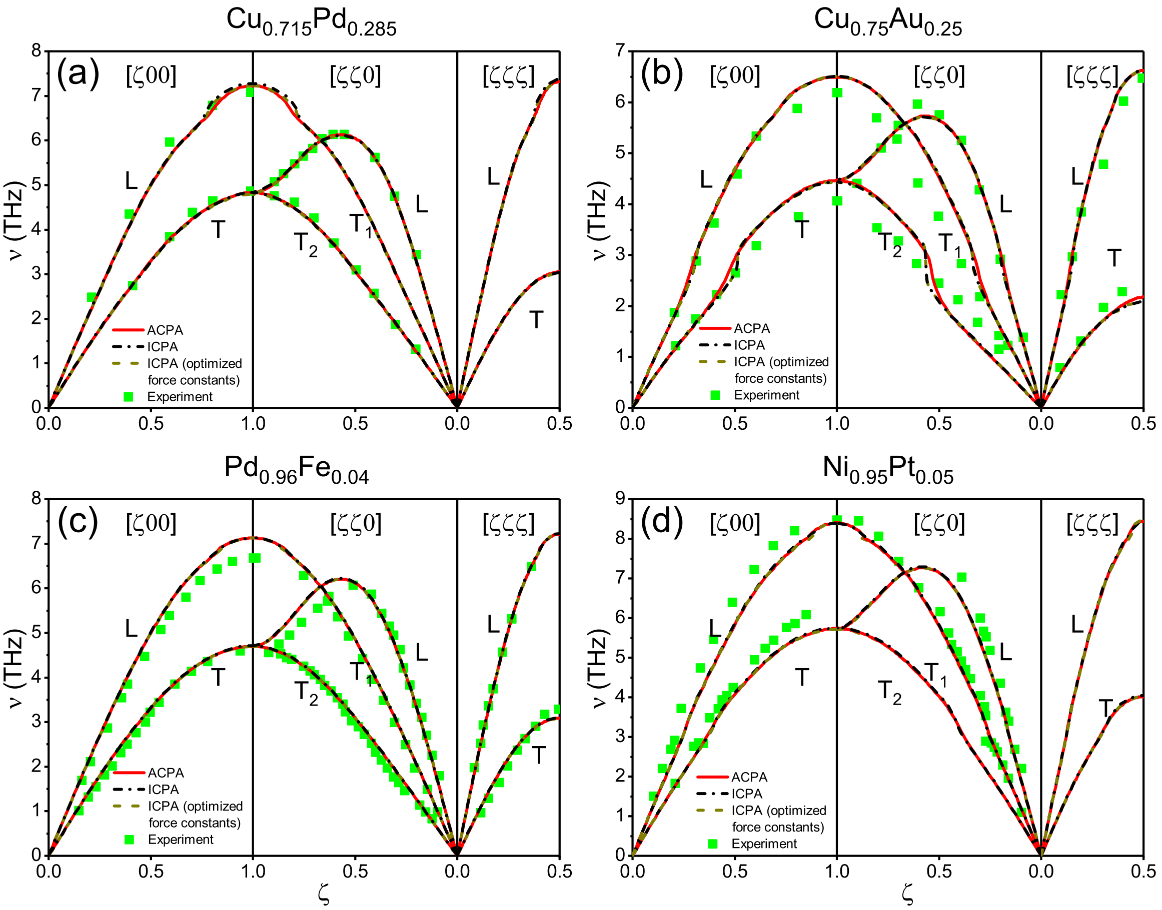

FIG. 1. Dispersion curves (frequency ν vs reduced wave vector ζ, where $\zeta = \frac{|\vec{k}|}{|\vec{k}_{\max}|}$ and $\vec{k}$ is the phonon wave vector) for four alloys including $Cu_{0.715}Pd_{0.285}$ in (a), $Cu_{0.75}Au_{0.25}$ in (b), $Pd_{0.96}Fe_{0.04}$ in (c), and $Ni_{0.95}Pt_{0.05}$ in (d). Red solid line: ACPA results; black dash dot lines: ICPA results with the first-principles force constants from Refs. [43–45]; brown dash lines: ICPA results with optimized force constants given by Table I. The green squares refer to the experimental data from Refs. [55–61].

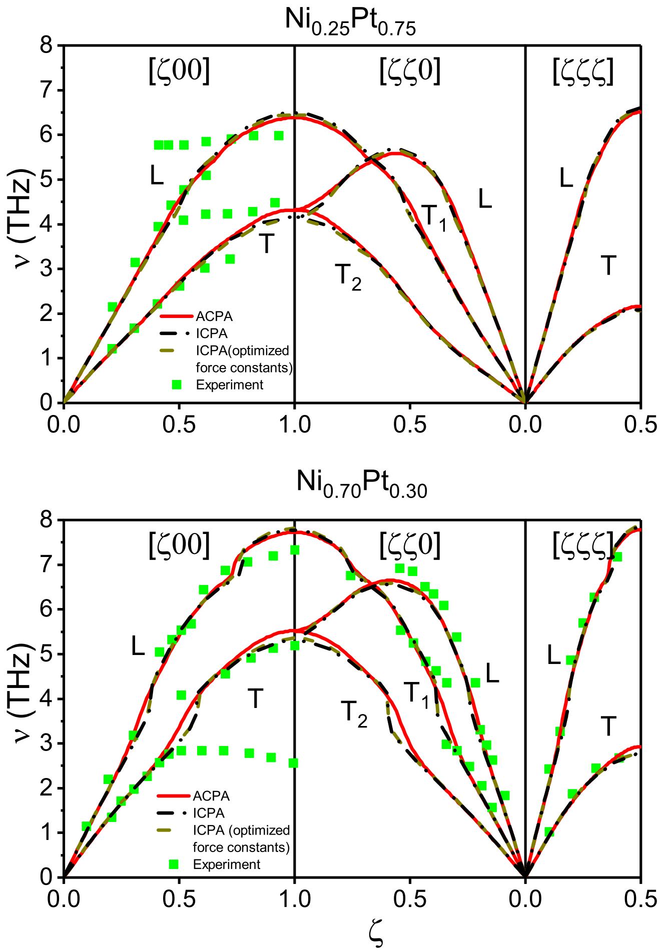

FIG. 2. Dispersion curves for $Ni_{0.25}Pt_{0.75}$ and $Ni_{0.70}Pt_{0.30}$ alloys (frequency ν vs reduced wave vector ζ, where $\zeta = \frac{|\vec{k}|}{|\vec{k}_{\max}|}$, and $\vec{k}$ is the phonon wave vector). Red solid line: ACPA results; black dash dot lines: ICPA results with the first-principles force constants from Ref. [45]; brown dash lines: ICPA results with optimized force constants given by Table I. The green squares refer to the experimental data from Ref. [61].

As shown for all six alloys, the ICPA calculations with the original and optimized approximated force constant produce almost the same phonon dispersion, demonstrating the fact that the separable force-constant model of Eq. (1) can very well represent the disordered force constants obtained from first principles. For a disordered system, the disorder average can smear out the small difference in the used force constants. Although, as we find, the ACPA and the ICPA are completely equivalent for mass-only disorder, the different treatments of the force-constant disorder in the CPA self-consistency give rise to the small deviations in the phonon dispersion of the ICPA and the ACPA. Despite that the ICPA and the ACPA are very close in accuracy, it should be mentioned that the ACPA calculation is computationally cheaper than ICPA calculations with our present implementation, and moreover, the ACPA is easy to implement. After Fourier transformation, for each k point in the first Brillouin zone, the matrix $\mathcal{P}(\boldsymbol{k})$ in the ACPA [see Eq. (19)] for fcc alloys has the size 3×3, which is much smaller than the matrix $V(\boldsymbol{k})$ with size 39×39 (the number of the nearest-neighboring sites $Z = 12$) in ICPA (see Eq. (A5) in Ref. [23]). Thus, for the matrix multiplication and inversion, whose computational cost scales as $O(n^3)$ (where n is the size of the matrix), the computation with ACPA is about 13^3 times faster than that with ICPA. After including all numerical procedures, the overall calculation of ACPA is about 1–2 orders magnitude faster than ICPA.

In Figs. 1 and 2, the available neutron-scattering measurements are included to compare with the theoretical results. As shown in Fig. 1, for the alloys $Cu_{0.715}Pd_{0.285}$, $Cu_{0.75}Au_{0.25}$, $Pd_{0.96}Fe_{0.04}$, and $Ni_{0.95}Pt_{0.05}$, both the ACPA and the ICPA calculations can provide a very good description of the experimental phonon dispersion. Therefore, we can see the ACPA method presents important predictive capability for simulating lattice vibration of realistic alloys. However, as reported in Ref. [45], there are some anomalous features of phonon dispersion in $Ni_{0.25}Pt_{0.75}$ and $Ni_{0.70}Pt_{0.30}$ that cannot be produced with ICPA. As shown in Fig. 2, the experimental dispersion curve of $Ni_{0.25}Pt_{0.75}$ features a double-peaked behavior for both the transverse and the longitudinal modes around [0.55,0,0], while the result of $Ni_{0.70}Pt_{0.30}$ shows a double-peaked behavior for the transverse modes from [0.5,0,0] to [1.0,0,0]. Both the ACPA and the ICPA results cannot produce these splitting features and resonance branches in the experimental results. These anomalous features might arise from the cluster or local environment effect of the disorders, which is beyond the ICPA and the ACPA in the single-site approximation. Although the ACPA or the ICPA in SSA have an important deficiency in the symmetry direction $[\zeta, 0, 0]$, the theoretical results agree well with the experimental measurements in the other two symmetry directions, as shown for $Ni_{0.70}Pt_{0.30}$. We thus have demonstrated the important applicability of the ACPA method (in SSA) by applying it to several alloys and comparing with ICPA and experimental results. Here, due to the single-site representation of the disorders, the ACPA model for both mass and force-constant disorders poses an important potential to combine with the cluster approximations, and diagrammatic corrections to account for effects of clusters and nonlocal correlation of disorder scattering.

Moreover, as shown in Fig. 3, we compare the averaged coherent scattering structure factor of the ACPA and ICPA models for several k points and the associated disorder-induced phonon broadening for the $Ni_{0.25}Pt_{0.75}$ alloy. It is clear that, for the different k points, peak positions of the ACPA and ICPA models are almost the same, giving rise to the good agreement in the phonon dispersion curves shown in Figs. 1 and 2. However, there are important differences in the phonon broadening, i.e., the linewidth of the averaged coherent scattering structure factor, between the ACPA and ICPA results. For example, for the state $[0.5, 0.5, 0]$L, the ACPA linewidth is 0.27 THz whereas the ICPA linewidth is 0.48 THz; and for the state $[0.3, 0.3, 0.3]$L, the ACPA linewidth is 0.26 THz whereas the ICPA linewidth is 0.49 THz. This difference in phonon broadening reflects the major theoretical difference between the ACPA and the ICPA approaches on dealing with force-constant disorders. A detailed comparison of the major differences of ACPA and ICPA formalisms is going to be reported in our next work [64].

Here we have reported the phonon dispersion results for several binary alloys. However, it is worth mentioning that,

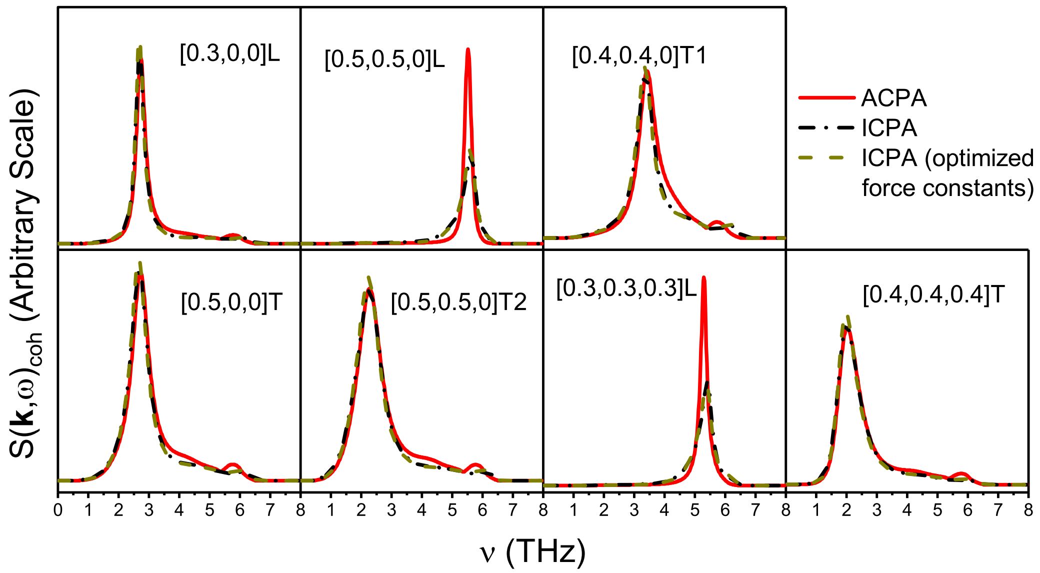

FIG. 3. Averaged coherent scattering structure factors for $\mathrm{Ni}_{0.25}\mathrm{Pt}_{0.75}$ alloy at several different k points. Red solid line: ACPA results; black dash dot lines: ICPA results with the first-principles force constants from Ref. [45]; brown dash lines: ICPA results with optimized force constants given by Table I.

since the ACPA formalism features the single-site characteristics, the ACPA method can be extended to multiple-component alloys similar to the conventional CPA, providing wider applicability for simulating disordered lattice dynamics.

VI. CONCLUSIONS

In this paper we demonstrate the accuracy and applicability of the ACPA approach for calculating the phonon dispersion of three-dimensional realistic binary alloys with both mass and force-constant disorders. We derive the coherent scattering structure factor to obtain the average phonon spectra and prove the sum rule in the auxiliary coherent medium to ensure the analyticity of physical properties. We have applied the ACPA to calculate the phonon dispersion of several realistic alloys and find the results agree well with results of ICPA and experimental measurements. The ACPA features the advantage of a higher computational efficiency and the potential to go beyond single-site approximation. Thus, we conclude that the ACPA provides an effective approach to simulate the vibrational properties of real materials and explore various features arising from disorders.

ACKNOWLEDGMENTS

Y.K. acknowledges financial support from the ShanghaiTech University startup fund, the Thousand Young Talents Plan, and Shanghai Shuguang Plan.

APPENDIX A: OPTIMIZATION DETAILS

For arbitrary force-constant parameters k_s and k_b, the separable force-constant model cannot provide an exact representation. Thus we need to optimize the parameters $x_{A/B}$, $S_{s/b}$, and $\lambda_{s/b}$ by fitting to the force constants obtained by first-principles calculations. In this case, we use the Gauss-Newton method to solve the nonlinear least-squares problem. The residuals are given by

$$r_i = k_i - f_i(\boldsymbol{\beta}), \tag{A1}$$

where k_i takes $k_{s/b}^{QQ'}$ for all kinds of $Q, Q', s/b$, and $f_i(\boldsymbol{\beta}) = x^Q S_{s/b} x^{Q'} + \lambda_{s/b}$ represents the corresponding separable force constant given in Eq. (1) as a function of the parameters $\boldsymbol{\beta} = (x^{A/B}, S_s, S_b, \lambda_s, \lambda_b)$. To solve the nonlinear least-squares problem, the sum of squares of residuals $S = \sum_i W_i r_i^2$, where W_i represents the weight of residuals, needs to be minimized, which gives five equations in the form of

$$\frac{\partial S}{\partial \beta_j} = 2\sum_i W_i r_i \frac{\partial r_i}{\partial \beta_j} = 0. \tag{A2}$$

To solve these equations to obtain optimized $\boldsymbol{\beta}$, we should start with an initial guess $\boldsymbol{\beta}^1$ and refine it iteratively as

$$\beta_j \approx \beta_j^{N+1} = \beta_j^N + \Delta\beta_j, \tag{A3}$$

where N denotes the Nth iteration step.

To obtain the proper shift vector $\Delta\boldsymbol{\beta}$, we expand the residuals $r_i \approx k_i - f_i(\boldsymbol{\beta}^N + \Delta\boldsymbol{\beta})$ as a first-order Taylor polynomial:

$$r_i = \Delta k_i - \sum_l J_{il}\Delta\beta_l, \tag{A4}$$

where

$$\Delta k_i = k_i - f_i(\boldsymbol{\beta}^N) \tag{A5}$$

and

$$J_{ij} = \frac{\partial f_i}{\partial \beta_j} = -\frac{\partial r_i}{\partial \beta_j}. \tag{A6}$$

Substituting Eqs. (A4) and (A5) into Eq. (A2) and rearranging it, we obtain

$$\sum_i \sum_l J_{ij} W_i J_{il} \Delta\beta_l = \sum_i J_{ij} W_i \Delta k_i, \tag{A7}$$

or in a matrix form,

$$J^T W J \Delta \boldsymbol{\beta} = J^T W \Delta \boldsymbol{k}. \tag{A8}$$

Then we can obtain

$$\Delta \boldsymbol{\beta} = (J^T W J)^{-1} J^T W \Delta \boldsymbol{k} \tag{A9}$$

to update parameters $\boldsymbol{\beta}$ as Eq. (A3) until $\boldsymbol{\beta}$ converges.

APPENDIX B: CONDITIONALLY AVERAGED AUXILIARY GREEN'S FUNCTIONS

For the completeness of introduction, we provide the derivation details with the occupation-operator technique for the conditionally averaged auxiliary Green's function. (Details can be found in the Ref. [52]). To evaluate $\langle g_{ii}^{Q} \rangle$ and $\langle g_{ij}^{QQ'} \rangle$, we first introduce the occupation matrix [50] $\widetilde{\eta}^{i,Q}$, where $\widetilde{\eta}^{i,Q} = 1$ if a Q atom is at the i site, and $\widetilde{\eta}^{i,Q} = 0$ otherwise. Then $\langle g_{ii}^{Q} \rangle$ and $\langle g_{ij}^{QQ'} \rangle$ can be expressed respectively as

$$\langle g_{ii}^{Q} \rangle = \frac{1}{c_i^Q} \langle \widetilde{\eta}^{i,Q} g \rangle_{ii} \tag{B1}$$

and

$$\langle g_{ij}^{QQ'} \rangle = \frac{1}{c_i^Q c_j^{Q'}} \langle \widetilde{\eta}^{i,Q} g \widetilde{\eta}^{i,Q'} \rangle_{ij}. \tag{B2}$$

From the definition of $\widetilde{\eta}^{i,Q}$, we can define the random quantity $\widetilde{P}^i$ as

$$\widetilde{P}^i = \sum_Q \widetilde{\eta}^{i,Q} \widetilde{P}^{i,Q} \tag{B3}$$

and have the relation

$$\sum_Q \widetilde{\eta}^{i,Q} = 1. \tag{B4}$$

With Eq. (B3), Eq. (B4), and the definition of $\widetilde{\eta}^{i,Q}$, we can obtain

$$\widetilde{\eta}^{i,A} = -(\Delta \widetilde{P}^i)^{-1} (\widetilde{P}^{i,B} - \widetilde{P}^i) = -(\widetilde{P}^{i,B} - \widetilde{P}^i)(\Delta \widetilde{P}^i)^{-1}, \tag{B5}$$

$$\widetilde{\eta}^{i,B} = (\Delta \widetilde{P}^i)^{-1} (\widetilde{P}^{i,A} - \widetilde{P}^i) = (\widetilde{P}^{i,A} - \widetilde{P}^i)(\Delta \widetilde{P}^i)^{-1}, \tag{B6}$$

where $\Delta \widetilde{P}^i = \widetilde{P}^{i,A} - \widetilde{P}^{i,B}$.

To eliminate the random quantity $\widetilde{P}^i$, we use the definitions $g = P^{-1}$ and $\mathcal{g} = \mathcal{P}^{-1}$, and obtain

$$P = \mathcal{P} + g^{-1} - \mathcal{g}^{-1}, \tag{B7}$$

which directly gives three matrix identities:

$$\langle P g \rangle = \mathcal{P} \mathcal{g}, \tag{B8}$$

$$\langle g P \rangle = \mathcal{g} \mathcal{P}, \tag{B9}$$

$$\langle P g P \rangle = \mathcal{P} \mathcal{g} \mathcal{P} + \langle P \rangle - \mathcal{P}. \tag{B10}$$

Substituting Eqs. (B5), (B6), and (B7) into Eqs. (B1) and (B2), respectively, and employing the three identities, we obtain

$$\langle g_{ii}^{Q} \rangle = (\widetilde{f}^{i,Q} \mathcal{g})_{ii} \tag{B11}$$

and

$$\langle g_{ij}^{QQ'} \rangle = (\widetilde{f}^{i,Q} \mathcal{g} \widetilde{h}^{j,Q'})_{ij}, \tag{B12}$$

where

$$c_i^A \widetilde{f}^{i,A} = -(\Delta \widetilde{P}^i)^{-1} (\widetilde{P}^{i,B} - \widetilde{\mathcal{P}}^i), \tag{B13}$$

$$c_i^A \widetilde{h}^{i,A} = -(\widetilde{P}^{i,B} - \widetilde{\mathcal{P}}^i)(\Delta \widetilde{P}^i)^{-1}, \tag{B14}$$

$$c_i^B \widetilde{f}^{i,B} = (\Delta \widetilde{P}^i)^{-1} (\widetilde{P}^{i,A} - \widetilde{\mathcal{P}}^i), \tag{B15}$$

$$c_i^B \widetilde{h}^{i,B} = (\widetilde{P}^{i,A} - \widetilde{\mathcal{P}}^i)(\Delta \widetilde{P}^i)^{-1}. \tag{B16}$$

With the CPA condition $\langle \widetilde{t}^i \rangle = 0$, $\widetilde{f}^{i,Q}$ and $\widetilde{h}^{i,Q}$ can be rewritten in a more compact form [52] as follows:

$$\widetilde{f}^{i,Q} = 1 + \widetilde{\mathcal{g}}_{ii} \widetilde{t}^{i,Q}, \tag{B17}$$

$$\widetilde{h}^{i,Q} = 1 + \widetilde{t}^{i,Q} \widetilde{\mathcal{g}}_{ii}, \tag{B18}$$

and then Eqs. (B11) and (B12) become

$$\langle g_{ii}^{Q} \rangle = (\mathcal{g} + \mathcal{g} \widetilde{t}^{i,Q} \mathcal{g})_{ii}, \tag{B19}$$

as given in Eq. (29), and

$$\langle g_{ij}^{QQ'} \rangle = [(1 + \mathcal{g} \widetilde{t}^{i,Q}) \mathcal{g} (1 + \widetilde{t}^{j,Q'} \mathcal{g})]_{ij}, \tag{B20}$$

as given in Eq. (30).

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