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Auxiliary coherent medium theory for lattice vibrations in random binary alloys with mass and force-constant disorders

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The inevitable and random impurities or defects can significantly influence the lattice-vibrational properties of materials and devices. Thus, the capability of effectively treating disorder effects is indispensable for theoretical simulations. In this paper, we report an auxiliary coherent medium theory, in the framework of multiple scattering theory, to simulate disordered vibrational systems containing both mass and force-constant disorders. In this method, the physical Green's function is related to an auxiliary Green's function by introducing a separable force-constant model to describe disordered systems. As an important result, the force-constant disorder can be transformed to a diagonal-like disorder in the auxiliary Hamiltonian while maintaining the important force-constant sum rule. In combination with the single-site and cluster coherent potential approximation, the configurational average over the auxiliary Green's function can be performed to obtain the configuration-averaged physical properties. To demonstrate the effectiveness of this method, we apply it to a one-dimensional harmonic chain with atomic disorders and find our calculations agree very well with the exact results for a wide range of mass and force constants. Moreover, we show that the phonon transport property of disordered devices can be derived based on the auxiliary Green's function formalism in combination with vertex corrections. The auxiliary coherent medium theory features easy implementation and feasible incorporation with diagrammatic technique in many-body perturbation and various cluster approximations, providing an important approach to analyze disorder effects on the vibrational properties. Moreover, it is also straightforward to apply the present formalism to treat the general atomic disorder in electronic systems.

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I. INTRODUCTION

Inevitable atomic disorders possess profound and fundamental influences on the ground-state and excitation properties of materials and devices [1–3]. For example, the random impurities/defects in lattice vibrations can significantly change the thermal conductivity of materials [2,4,5]. However, understanding the disorder effects in materials/devices remains challenging for both experiment and theory. Over the past decades, considerable efforts have been spent to develop effective methods to derive the disorder averaged physical properties, to enable the simulation of disorders. The most important approximations include the conventional coherent potential approximation (CPA) for only the diagonal disorder (as first proposed by Soven for the electronic system [6] and Taylor for the vibrational system [7]), and its various extensions and further developments to include the off-diagonal disorder and the effects of clusters, as demonstrated with different model systems [8–15] and implemented with first-principle methods [16–25]. Within the first-principles KKR [16–20] and MTO [21,22] methods with the local muffin-tin potential approximation, the combined diagonal and off-diagonal disorders for general electronic problem can be transformed to a diagonal-like disorder problem by introducing the scattering-path-operator technique [18]. Presently, the first-principles CPA has seen many important applications in simulating disordered electronic materials [16–18,21,22] and quantum transport in nanoelectronics [26–29]. In addition, the separability of diagonal and off-diagonal disorders in general electronic problem allows the formulation of the Black-Esterling-Berk (BEB) locator CPA method [8,9] for disordered electronic structure simulation, for example, the attempts for implementing BEB-CPA in combination with LCAO first-principles method [30]. The wide applications of CPA in electronic system can be attributed to its important advantages of easy implementation and high computational efficiency, and moreover the advantage that conventional CPA can be systematically improved with diagrammatic techniques (vertex corrections) in many-body perturbation to account for nonlocal correlations [31] and various cluster approximations beyond single-site approximation [12,13]. These important advantages of conventional CPA are desirable for simulating the disordered vibrational systems.

However, for a general vibrational system, the translational invariance (or momentum conservation) requires that the dynamic matrix elements between site i and its neighbors j always satisfy the relation $D_{ii} = -\sum_{j\neq i} D_{ij}$, namely, the force-constant sum rule [32]. As an important consequence, unlike the general electronic problem, the diagonal and off-diagonal disorder in vibrational Hamiltonian are inseparable, making the conventional CPA and BEB locator-CPA unfeasible for simulating disordered vibrational systems (except for the special case of the diagonal only-mass disorder). However, to

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extend the CPA to include the force-constant disorders, there have been several reported attempts but only for some special cases, such as Kaplan-Mostoller's method [33] with the arithmetical-scaled force constant [$k^{AB} = (k^{AA} + k^{BB})/2$] and Grünewald's locator-CPA method [34] with the geometric-scaled force constant ($k^{AB} = \sqrt{k^{AA}k^{BB}}$). As far as we know, there are only a very limited number of general methods that can deal with both mass and force constant disorders. These approaches are based on the augmented-space formalism (ASF) proposed by Mookerjee [35], in which the disorder average in real space is mapped to an augmented space, such as the augmented-space recursion method (ASR) [15,36–38] and the itinerant coherent potential approximation (ICPA) [14]. As the important advantage, the state-of-art ASR and ICPA methods can provide the exact representation of the disordered force-constants for vibrational systems with the sum rule obeyed. The ASR method [15] is not self-consistent, but it can handle large cluster effects, while ICPA provides a self-consistent approach in a single-fluctuation approximation as a generalization of the traveling cluster approximation (TCA) [10,39]. Presently, the ASR method has been applied with reasonable success to different alloys [40,41] and the ICPA method has reproduced the phonon dispersions for several alloys [42,43]. However, ASR and ICPA are not easy to implement and are computationally expensive, compared to the conventional CPA [6,7]. For the self-consistent ICPA, it is not easy to go beyond the single-fluctuation approximation to include the important cluster effects and nonlocal correlation of disorders. Extending the conventional CPA to the disordered vibration is thus desirable for studying vibrational properties of realistic materials with inevitable disorders, especially important for material/device design for thermal control and management.

In this paper, we report an auxiliary coherent medium theory to treat both the mass and force-constant disorders in random binary alloys. In this method, an approximate force-constant decomposition technique is introduced to transform the force-constant disorder into a diagonal-like disorder problem in an auxiliary Hamiltonian. Then, the disorder averaged physical quantities, including density of states and phonon transmission function, can be calculated with an averaged auxiliary Green's function obtained by performing conventional CPA, enabling the simulation of the general disorders in lattice vibration. The applications to 1D disordered harmonic chains for a wide range of disordered mass and force-constants show good agreement with the exact numerical results, demonstrating the effectiveness of the auxiliary coherent medium theory. In addition to disordered vibrational problem, this decomposition technique can also be used in the electronic disorder problem with off-diagonal disorder.

The rest of the paper is organized as follows: In Sec. II, we introduce the force-constant decomposition and the associated auxiliary Hamiltonian and Green's function that gives the physical Green's function and properties. Section III introduces the conventional CPA to self-consistently calculate configurational average of the auxiliary Green's function, to obtain averaged physical phonon properties. Section IV presents the configurational averaged phonon transmission function based on the auxiliary Green's function. In Sec. V we apply our method to calculate the phonon density of states in a one-dimensional (1D) disordered harmonic chain with different mass and force-constant disorders, and we compare our results with the exact results and previous method. At last, we conclude in Sec. VI.

II. FORCE-CONSTANT DECOMPOSITION AND AUXILIARY GREEN'S FUNCTION

We consider the lattice vibration Hamiltonian in the harmonic approximation

$$H = \sum_{i\alpha} \frac{p_{i\alpha}^2}{2m_i} + \frac{1}{2}\sum_{i\alpha j\beta} u_{i\alpha} D_{i\alpha j\beta} u_{j\beta}, \tag{1}$$

where $p_{i\alpha}$ and $u_{i\alpha}$ are the respective momentum and displacement of the atom on site i along $\alpha = \{x, y, z\}$ direction, m_i is the atomic mass on site i, and $D_{i,j}$ denotes the dynamic matrix element between the sites i and j. The corresponding retarded Green's function, namely the displacement-displacement correlation function, can be defined as [44]

$$G^R_{i\alpha j\beta}(t) = -\frac{i}{\hbar}\theta(t)\langle[u_{i\alpha}, u_{j\beta}]\rangle, \tag{2}$$

where θ is the step function. By taking second-order time derivative and Fourier transformation to the frequency domain, one can obtain

$$m_i\omega^2 G^R_{i\alpha j\beta}(\omega) = \delta_{\alpha\beta}\delta_{ij} + \sum_{k\gamma} D_{i\alpha k\gamma} G^R_{k\gamma j\beta}(\omega), \tag{3}$$

or more concisely

$$G^R(\omega) = (m\omega^2 - D)^{-1}, \tag{4}$$

which is the basic equation that describes the vibrational properties of materials. For a random alloy, the Hamiltonian in Eq. (1) and corresponding Green's function in Eq. (4) become random quantities, depending on the atomic configuration of a specific sample. For a binary alloy A_xB_{1-x} with x denoting the concentration of A, the quantities m_i and D_{ij} become random and are determined by the disordered atomic species on the relevant lattice site(s), for example, $m_i = m_i^Q$ ($Q = A, B$) and $D_{ij} = D_{ij}^{QQ'}$ ($Q, Q' = A, B$). Therefore, to obtain the meaningful averaged physical quantities, we need to calculate the configurational average of the Green's function. However, the force-constant disorder in the diagonal (due to the sum rule) and off-diagonal parts of the dynamic matrix prohibits the implementation of conventional CPA method for lattice vibration of random alloys. To provide a solution to this problem, we introduce a force-constant decomposition technique and the associated auxiliary Green's function in the following.

For a random alloy A_xB_{1-x}, the force constants k_{ij} between the site i and j are generally determined by the atomic occupants and structure of the lattice. For example, k_{ij} can take the different possible values $k_{ij}^{AA}, k_{ij}^{AB}, k_{ij}^{BB}$ depending on the atomic occupations of sites i and j. To proceed, we introduce a new separable form to the force constants

$$k_{ij} = x_i S_{ij} x_j + \lambda_{ij} \quad (i \neq j), \tag{5}$$

where S_{ij} and λ_{ij} contains the structural information (including the distance and lattice geometry) independent of the atomic occupations on sites i and j, and the quantity $x_i = x_i^Q$ $(Q = A, B)$ contains the information of the atomic occupant on the i-th site. The first term in Eq. (5) is the geometric-scaled force-constant model proposed in Ref. [34] with very limited application due to the neglect of other contributions. Importantly, the form of the first term provides a way to account for the disorder information of the atomic occupants on the two sites. Without the first term, λ_{ij} only provides a virtual crystal approximation to the disordered force constant, losing the disorder information due to the randomness on the two sites. The combination of the virtual-crystal and geometric-scaled models in Eq. (5) overcomes the shortcomings of the two models, featuring higher accuracy and more general applicability in the description of disordered systems. As shown in the Appendix, we find Eq. (5) can provide the exact representation of the disordered force constants for the 1D chain with arbitrary values. The exact representation of 1D case provides an important validation for the separable force-constant model and gives the important basis for generalizing to higher dimensions. Extending to the systems of 2D and 3D can be realized by introducing the respective 2×2 and 3×3 tensors for the single-site quantity x_i and two-site quantities S_{ij} and λ_{ij}. As shown in the Appendix, the model in Eq. (5) can represent the disordered force-constants from first principles for face-centered-cubic PdFe alloy with very small error.

As an important consequence of the above force-constant model, the dynamic matrix can be rewritten as

$$D = XK, \tag{6}$$

with the matrix element explicitly given by $D_{i\alpha,j\beta} = X_{ii}K_{i\alpha j\beta}$. Here, X is a diagonal matrix, namely $X_{ii} = x_i$, and $X_{ij} = 0$ $(i \neq j)$, and the K matrix is given in the following form:

$$K_{ij} = -\left(S_{ij}x_j + \frac{\lambda_{ij}}{x_i}\right) \quad (i \neq j) \tag{7}$$

and

$$K_{ii} = \frac{1}{x_i}\sum_{j\neq i} k_{ij} = \sum_{j\neq i}\left(S_{ij}x_j + \frac{\lambda_{ij}}{x_i}\right). \tag{8}$$

Note that x_i and x_j are linearly separated in K, providing an important feature of K. It is clearly seen that K matrix satisfies $K_{ii} = -\sum_{j\neq i} K_{ij}$. This sum rule of K ensures that the force-constant sum rule is always satisfied in our further derivation. As an important result of Eqs. (6)–(8), we can further decompose K into a sum of single-site dependent quantities, namely,

$$K = \sum_i \mathcal{K}_i, \tag{9}$$

where $\mathcal{K}_i$ is a matrix and $\mathcal{K}_i$ contains all the contributions only associating with x_i. For example, for a 1D disordered chain of a binary alloy as illustrated in Fig. 1, the $\mathcal{K}_i$ of the site i takes the form (see the Appendix for the final result)

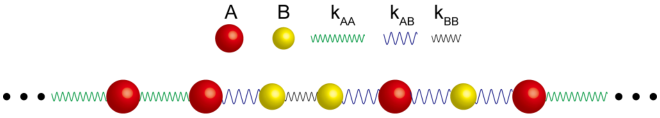

FIG. 1. Schematic illustration of a 1D atomic chain of a binary alloy A_xB_{1-x}.

$$\mathcal{K}_i = \begin{bmatrix} 0 & \dots & 0 & \dots & 0 \\ \dots & S_{i-1,i}x_i & -S_{i-1,i}x_i & 0 & \dots \\ \dots & -\frac{\lambda_{i-1,i}}{x_i} & \frac{\lambda_{i-1,i}+\lambda_{i,i+1}}{x_i} & -\frac{\lambda_{i,i+1}}{x_i} & \dots \\ \dots & 0 & -S_{i,i+1}x_i & S_{i,i+1}x_i & \dots \\ 0 & \dots & 0 & \dots & 0 \end{bmatrix}. \tag{10}$$

As we show in the Appendix, the decomposition formalism from Eqs. (5)–(10) present an exact representation of the dynamic matrix for the 1D chain with the nearest-neighbor coupling for arbitrary values of force-constants.

Importantly, using diagonal matrix X and Eq. (6), the Green's function in Eq. (4) can be rewritten as

$$G = gX^{-1}, \tag{11}$$

where the auxiliary Green's function is defined as

$$g = (X^{-1}m\omega^2 - K)^{-1} \tag{12}$$

and the corresponding auxiliary Hamiltonian $P \equiv X^{-1}m\omega^2 - K$, which yields a complete separation of single-site quantity from the dynamic matrix. Thus, P can be decomposed as $P = \sum_i P_i$, where the single-site quantity

$$P_i = x_i^{-1}m_i\omega^2 - \mathcal{K}_i. \tag{13}$$

In this form, the general disorders in lattice vibration is reduced to a diagonal-like disorder problem. The auxiliary Hamiltonian containing only single-site P_i is essential for the implementation of conventional CPA for calculating the disordered phonons in binary alloys. This is because propagator expansion in multiple-scattering description of disordered system requires the single-site scatters in conventional CPA, [45] as we show in next section. Moreover, Eq. (11) provides a simple connection between the physical G and the auxiliary g. Hence, the vibrational properties of the system can be related to the auxiliary Green's function, such as the phonon density of state discussed in Sec. V and the phonon transmission function derived in Sec. IV.

III. COHERENT POTENTIAL APPROXIMATION TO AUXILIARY *g*

For a disordered binary alloy, the single-site quantity P_i is a random quantity, resulting in the random g and G. Thus, for disordered systems, it is important to do the disorder average to obtain physically meaningful results. To do so, we first introduce CPA to calculate the disorder averaged auxiliary Green's function $\langle g\rangle$, from which the averaged physical property can be derived as we show for phonon transport in Sec. IV and density of state in Sec. V.

Generally, the coherent potential approximation constructs an effective medium with the Green's function same as the disorder averaged one. To start, we introduce a coherent function $\mathcal{P} = \sum_i \mathcal{P}_i$ to give the auxiliary Green's function of an effective medium, namely $\mathscr{g} = \mathcal{P}^{-1}$. Then, we consider a specific configuration of random binary alloy with the auxiliary Green's function given as $g = P^{-1}$.

By defining the deviation function $V = \mathcal{P} - P = \sum_i V_i$, that contains all the randomness, the auxiliary Green's function can be rewritten in a form of Dyson equation with reference to the effective medium

$$g = \mathscr{g} + \mathscr{g} V g. \tag{14}$$

By defining a T-matrix $T = V(\mathbf{1} - \mathscr{g} V)^{-1}$ describing the total scattering due to disorders, the auxiliary Green's function changes to

$$g = \mathscr{g} + \mathscr{g} T \mathscr{g}. \tag{15}$$

By taking the disorder average, we obtain

$$\langle g\rangle = \mathscr{g} + \mathscr{g}\langle T\rangle \mathscr{g}. \tag{16}$$

We can enforce $\langle g\rangle = \mathscr{g}$ to obtain the CPA condition namely $\langle T\rangle = 0$, providing a self-consistent equation to solve the effective medium $\mathcal{P}$. However, T is generally an intractable full matrix that contains all the complexity of a random system. As a result, directly solving the CPA equation $\langle T\rangle = 0$ is not practical, and thus further approximation to the average of T is required [45].

To make the CPA practical, one can introduce the single-site approximation (SSA) [45]. As known, the total T can be expressed with multiple scattering events, namely,

$$T = \sum_i t_i + \sum_{i\neq j} t_i \mathscr{g}_{ij} t_j + \sum_{i\neq j, j\neq k} t_i \mathscr{g}_{ij} t_j \mathscr{g}_{jk} t_k + \ldots., \tag{17}$$

with the single-site scattering matrix t_i defined as

$$t_i = V_i(\mathbf{1} - \mathscr{g}_{ii} V_i)^{-1}. \tag{18}$$

The single-site approximation neglects the correlation between all the scattering events on different sites and on the same site at different times. As an important result, the CPA condition $\langle T\rangle = 0$ is reduced to a single-site equation

$$\langle t_i\rangle = \sum_{Q=A,B} c^Q t_i^Q = 0. \tag{19}$$

By combining the above single-site equation and $\mathscr{g} = \mathcal{P}^{-1}$, the single-site quantity $\mathcal{P}_i$ can be self-consistently solved for each disordered site of the system. In such a way, the effective medium described by $\mathcal{P} = \sum_i \mathcal{P}_i$ can be efficiently obtained. To provide a stable solution to the single-site CPA equations, one usually introduces the coherent interactor Ω_i to describe the effects of the surrounding coherent medium to the site i [28], and then the site-diagonal auxiliary Green's function can be given as

$$\mathscr{g}_{ii} = (\mathcal{P}_i - \Omega_i)^{-1}, \tag{20}$$

and the conditionally averaged auxiliary Green's function, which corresponds to the system with the fixed Q atom on the site i, can be given as

$$\mathscr{g}_{ii}^Q = \mathscr{g}_{ii} + \mathscr{g}_{ii} t_i^Q \mathscr{g}_{ii} = \left(P_i^Q - \Omega_i\right)^{-1}. \tag{21}$$

In addition, we have the relation $\mathscr{g}_{ii} = \sum_Q c_i^Q \mathscr{g}_{ii}^Q$ (c^Q is the concentration of Q), thus we can obtain

$$\mathcal{P}_i = \left[\sum_Q c_i^Q (P_i^Q - \Omega_i)^{-1}\right]^{-1} + \Omega_i. \tag{22}$$

Equations (20)–(22) together with $\mathscr{g} = \mathcal{P}^{-1}$ form a closed set of self-consistent equations to solve $\mathcal{P}_i$ for all the disordered sites. The iterative calculation starts with an initial $\mathcal{P}_i$ obtained under average-t-matrix approximation, which is regarded as a good start in the self-consistent CPA calculation [46]. Then, with $\mathcal{P} = \sum_i \mathcal{P}_i$, we calculate $\mathscr{g} = \mathcal{P}^{-1}$ to obtain $\mathscr{g}_{ii}$, and thus obtain the coherent interactor Ω_i with Eq. (20). Finally, new $\mathcal{P}_i$ is calculated for using in next iteration with Eq. (22). Such calculation iterates till $\mathcal{P}_i$ is converged.

With the averaged auxiliary Green's function, various disorder averaged physical quantities can be derived to realize the simulation of disordered material/devices. For example, the averaged physical Green's function can be directly given as

$$\langle G_{ii}\rangle = \left\langle g_{ii} x_i^{-1}\right\rangle = \sum_Q c_i^Q \left\langle g_{ii}^Q\right\rangle \left(x_i^Q\right)^{-1}. \tag{23}$$

Furthermore, as we will show in Sec. IV, the averaged two-Green's-function correlator can also be formulated with the auxiliary Green's function formalism in combination with the vertex correction technique, thus enabling the simulation of thermal transport through disordered devices.

IV. FORMULATION OF TRANSMISSION FORMULA WITH THE AUXILIARY GREEN'S FUNCTION *g*

We consider the thermal transport through a device containing a central region sandwiched by two semi-infinite ordered thermal leads. In order to calculate the averaged thermal conductance [47] given by

$$\sigma = \int_0^\infty \frac{\mathrm{d}\omega}{2\pi} \hbar\omega \langle \mathcal{T}(\omega)\rangle \frac{\partial f}{\partial T}, \tag{24}$$

where f is the Bose-Einstein distribution function, we need the phonon transmission function given by the Caroli formula [47,48]

$$\mathcal{T}(\omega) = \mathrm{Tr}(G^R \Gamma_L G^A \Gamma_R), \tag{25}$$

where $G^{R/A}$ is the retarded/advanced Green's function of the central region of the device, the linewidth function $\Gamma_{L/R}$ describes the coupling of the central region to the left/right leads.

By using the relation $g^{R/A} = G^{R/A} X$, the averaged transmission function becomes

$$\langle \mathcal{T}(\omega)\rangle = \mathrm{Tr}(\langle g^R X^{-1} \Gamma_L g^A X^{-1} \Gamma_R\rangle) = \mathrm{Tr}(\langle g^R \tilde{\Gamma}_L g^A \tilde{\Gamma}_R\rangle), \tag{26}$$

where the auxiliary line-width function $\tilde{\Gamma}_{L/R} = X^{-1}\Gamma_{L/R}$. $\tilde{\Gamma}_{L/R}$ is independent of the atomic disorders in central device, because it only depends on the ordered left/right leads and ordered buffer-layers at left/right side of the central region. Then the nonequilibrium vertex correction (NVC) theory [26] can be introduced to treat the two-particle auxiliary

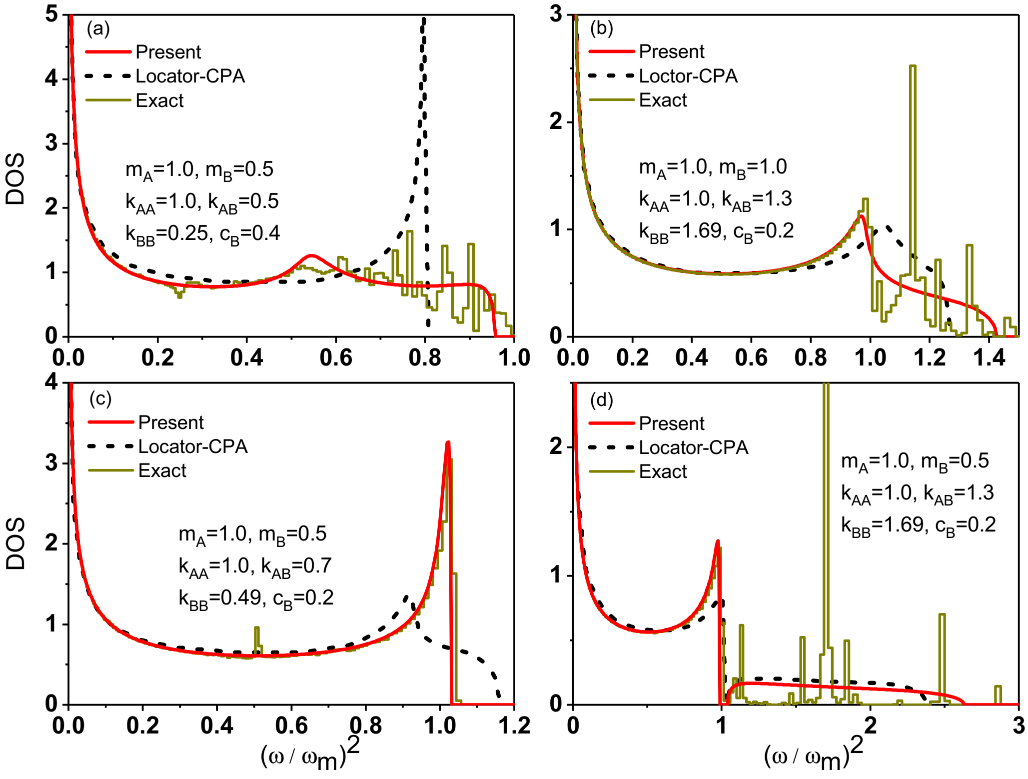

FIG. 2. DOS of 1D disordered harmonic chains in the single-site approximation vs $(\omega/\omega_m)^2$ where $\omega_m = 2\sqrt{k^{AA}/m_A}$. Specific parameters are shown for all cases. Red solid: results of our present method; black dashed: results of Grünewald's locator-CPA method [34]; brown histograms: exact results calculated by Dean's technique [50].

Green's function. Using the relations in Sec. III, $g^{R/A} = \mathscr{g}^{R/A} + \mathscr{g}^{R/A}T^{R/A}\mathscr{g}^{R/A}$ and $\langle T^{R/A}\rangle = 0$, we can obtain

$$\langle g^R\tilde{\Gamma}_L g^A\tilde{\Gamma}_R\rangle = \mathscr{g}^R\tilde{\Gamma}_L\mathscr{g}^A\tilde{\Gamma}_R + \mathscr{g}^R\Omega_{\text{NVC}}\mathscr{g}^A\tilde{\Gamma}_R, \tag{27}$$

where the NVC matrix Ω_{NVC} is defined as

$$\Omega_{\text{NVC}} = \sum_{i,j}\langle T_i^R\mathscr{g}^R\tilde{\Gamma}_L\mathscr{g}^A T_j^A\rangle, \tag{28}$$

where

$$T_i^{R/A} = (1 + T^{R/A}\mathscr{g}^{R/A})V_i = t_i^{R/A} + \sum_{j\neq i} T_j^{R/A}\mathscr{g}^{R/A}t_i^{R/A}. \tag{29}$$

In the SSA, for $i \neq j$ we have $\langle T_i^R\mathscr{g}^R\tilde{\Gamma}_L\mathscr{g}^A T_j^A\rangle = 0$. Thus, the Ω_{NVC} can be written as a sum of site-diagonal matrices

$$\Omega_{\text{NVC}} = \sum_i \Omega_{\text{NVC},i} = \sum_i \langle T_i^R\mathscr{g}^R\tilde{\Gamma}_L\mathscr{g}^A T_i^A\rangle. \tag{30}$$

Substituting Eq. (29) into Eq. (30), the terms with single t_i vanish in the SSA, and then we obtain a self-consistent equation of Ω_{NVC}

$$\Omega_{\text{NVC},i} = \langle t_i^R\mathscr{g}^R\tilde{\Gamma}_L\mathscr{g}^A t_i^A\rangle + \sum_{j\neq i}\langle t_i^R\mathscr{g}^R\Omega_{\text{NVC},j}\mathscr{g}^A t_i^A\rangle. \tag{31}$$

With the Ω_{NVC}, the averaged transmission function can be obtained easily by Eqs. (26) and (27), thus the thermal transport through disordered devices can be simulated.

V. NUMERICAL RESULTS AND DISCUSSIONS

The source of errors in conventional CPA arises from the neglect of the disordered-surrounding-induced local fluctuation away from the uniform effective medium in which the single-site or cluster is embedded. However, the relative local fluctuation due to surroundings is inversely proportional to the number of neighboring disordered sites [49] (given by the central limit theorem). The 1D chain features the smallest number of neighboring couplings and thus the largest local fluctuations, compared to the 2D and 3D systems. Thus, as an important test of the accuracy and applicability of the present method, we calculate the disordered 1D harmonic chain as shown in Fig. 1. For a general combination of possible force constants k^{AA}, k^{BB}, and k^{AB} between two neighboring sites, the $\mathcal{K}_i$ has the final form of Eq. (A7) in the Appendix, and CPA equations are solved self-consistently to obtain auxiliary $\mathscr{g}$. The local phonon density of state [45] of a harmonic chain is given by

$$\begin{aligned}\rho_i(\omega^2) &= -\frac{1}{\pi}\text{Im}\langle m_i G_{ii}(\omega)\rangle \\ &= -\frac{1}{\pi}\text{Im}\sum_Q \langle m_i^Q \mathscr{g}_{ii}^Q(\omega)x_i^{Q,-1}\rangle,\end{aligned} \tag{32}$$

which requires the conditionally averaged auxiliary Green's function [see Eq. (21)].

As the first application, we consider the alloys with geometric-scaled force constants, namely $k^{AB} = \sqrt{k^{AA}k^{BB}}$.

Our results are compared with the Grünewald's locator-CPA [34,51] calculations and exact results of Dean's technique [50] (with a chain of 1 000 000 random atoms) for four different cases, as shown in Fig. 2. To erase the influence of the absolute value of atomic parameters, we take the host $m_A = 1.0$ and $k^{AA} = 1.0$ for simplicity, and normalize the frequency by taking $(\omega/\omega_m)^2$, where $\omega_m = 2\sqrt{k^{AA}/m_A}$ is corresponding to the maximum frequency of the pure A lattice. It can be clearly seen that, in comparison with the exact result for all cases, our present results (solid) show significant improvement over the results of locator-CPA (dashed). The locator-CPA calculations present seriously wrong behavior in the DOS, and significant errors in the bandwidth for the four specific cases. In particular, locator-CPA for the case in Fig. 2(a) presents a divergence at about $(\omega/\omega_m)^2 = 0.8$ (like a virtual-crystal approximation), which is absent in the exact and our present calculations; for the case in Fig. 2(b), the expected peak of DOS at about $(\omega/\omega_m)^2 = 1.0$ has an observable shift in the result of locator-CPA, while our result gives a good match to the exact result; for the case in Fig. 2(c), the exact and our present results show a cutoff frequency at $(\omega/\omega_m)^2 = 1.0$ with a sharp peak, which is missing in the result of locator-CPA. Moreover, locator-CPA significantly underestimates the bandwidth for cases of Figs. 2(a), 2(b) and 2(d), and overestimates the bandwidth for the case of Fig. 2(c), while our results present better agreement with the exact results. The important accuracy of present method over the previous locator-CPA method [34,51] can be attributed to the fact that the force-constant sum rule is fully satisfied in our method, but not in the locator-CPA method.

However, because the single-site approximation neglects the local environmental effects, our results in the SSA cannot include the effects of localized modes corresponding to different specific local configurations, which contribute the sharp and localized peaks in the exact results. To involve the effects of different local structures, we combine molecular coherent potential approximation (MCPA) [11,12] with our method, where all single-site quantities in our method are replaced with single-cluster quantities. Although MCPA violates the single-site translational symmetry, it has the Herglotz analytic properties and can include the local environment effect (short-range order effect) systematically [45]. To show the improvement of our formalism with MCPA, we take the case in the Fig. 2(d) as an example and show the DOS for the cluster size changing from $N_c = 1$ to $N_c = 10$ atoms in Fig. 3 (note that we account for all the possible 2^{N_c} atomic configurations in each MCPA calculation). It is obvious that, as increasing the cluster size, more sharp features caused by local environmental effects emerge in the high-frequency region.

To demonstrate the general applicability of present method, we investigate the averaged DOS of 1D atomic chain for two cases with the respective light- ($m_B = 0.5m_A$) and heavy-mass ($m_B = 2.0m_A$) defects and force-constants chosen arbitrarily. Figure 4 shows the present CPA results for cluster sizes ranging from $N_c = 1$ to $N_c = 10$, and compare with the exact results [50]. It is clear that the SSA (namely, $N_c = 1$) works very well at low-frequency regime, but it fails to produce the localized peaks at high frequencies, illustrating the deficiency of SSA in comparison with the exact result. However,

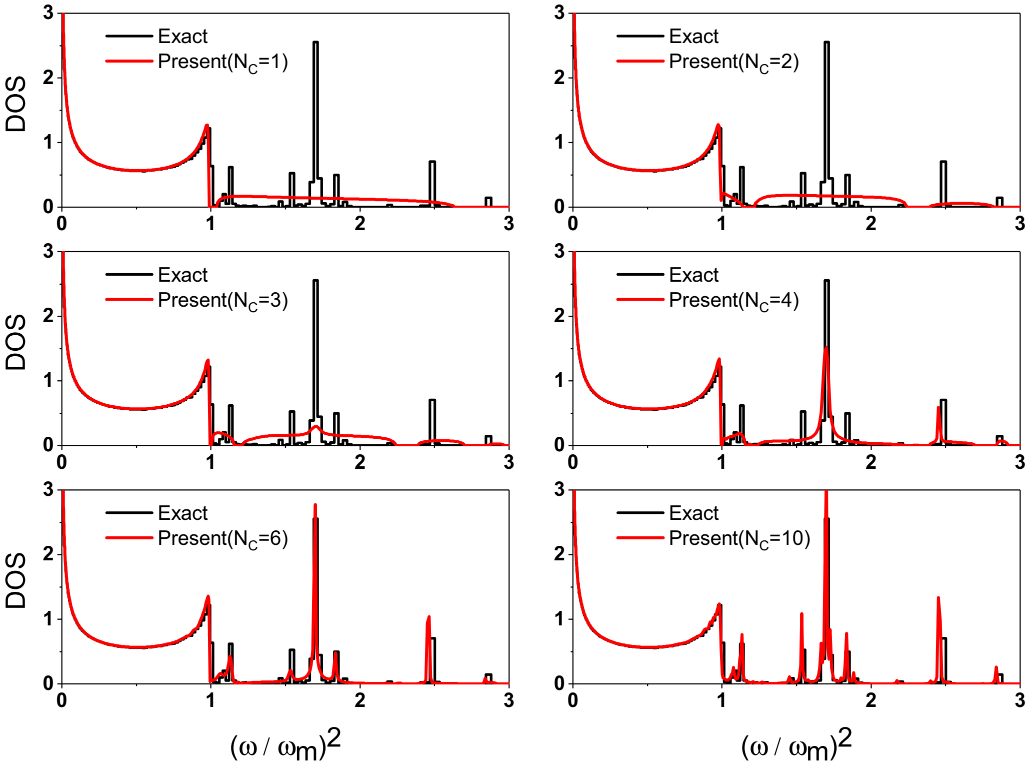

FIG. 3. DOS of 1D disordered harmonic chains for the cluster size $N_c = 1, 2, 3, 4, 6, 10$ with $m_A = 1.0$, $m_B = 0.5$, $k^{AA} = 1.0$, $k^{AB} = 1.3$, and $k^{BB} = 1.69$ with the defect concentration $c_B = 0.2$.

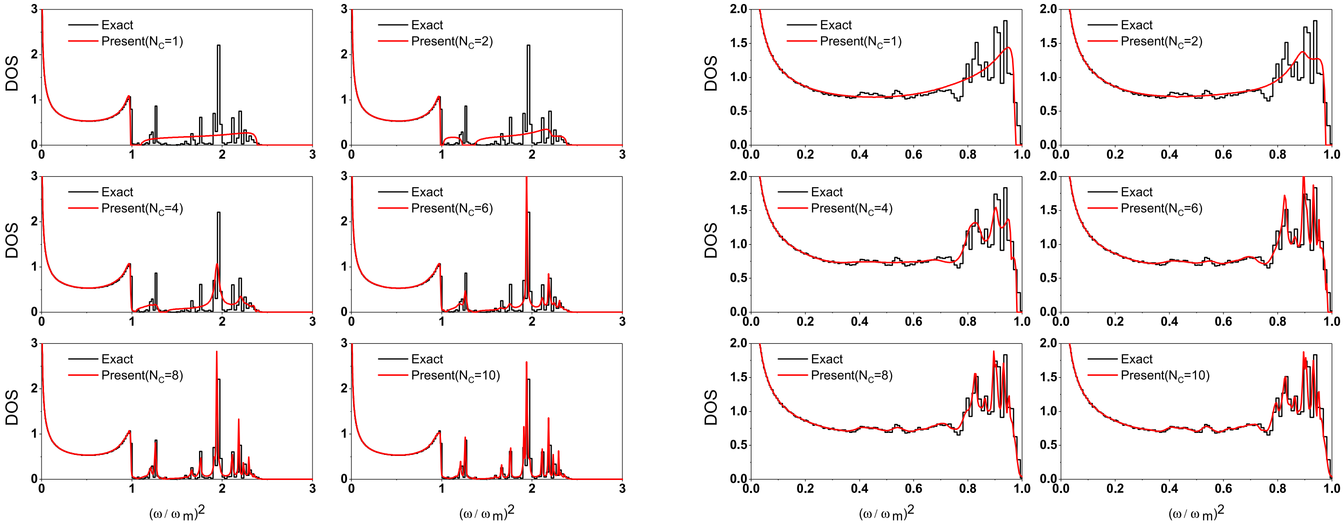

FIG. 4. DOS of 1D disordered harmonic chains for the cluster size $N_c = 1$, 2, 4, 6, 8, 10. Left side is for the chain with $m_A = 1.0$, $m_B = 0.5$, $k^{AA} = 1.0$, $k^{AB} = 1.5$, and $k^{BB} = 1.25$ with the defect concentration $c_B = 0.25$; right side is for the chain with $m_A = 1.0$, $m_B = 2.0$, $k^{AA} = 1.0$, $k^{AB} = 1.2$, and $k^{BB} = 1.5$ with $c_B = 0.50$.

accounting for the localized modes at high frequencies requires the MCPA method. As N_c reaches 6 and beyond, both cases shown in Fig. 4 exhibit the inclusion of local environmental effects in the CPA calculations and present good convergence to the exact results. The good agreement with the exact results in Figs. 3 and 4 illustrates the effectiveness of the present CPA method for treating disorder effects in lattice vibration. It is also worth mentioning that the translational symmetry, broken by the MCPA, can be restored by implementing with dynamic cluster approximation [13,52] (DCA). It is noted that DCA was used to address diagonal mass disorder and its cluster effects on phonon density of states properly [13,52] and the DCA simulation of force-constant disorders has not been shown to date. Our present single-site auxiliary method [Eq. (9)] provides a good jumping off point to implement the DCA to study the cluster effects of force-constant disorders. As found in Figs. 3 and 4, with the sum rule obeyed, the spectral calculated with the auxiliary CPA can approach the exact results as increasing the cluster size. We note that in Ref. [53] for magnetism (Ising model), it has been shown the CPA and DCA methods enforced to obey the sum rule (optical theorem in this case) can approach the exact result versus cluster size, and the CPA with sum rule obeyed gives closer result to the DCA result.

In above calculations, we have used the disordered 1D atomic chain to demonstrate the effectiveness of present auxiliary CPA. As one may notice that the disordered force constants $k_{ij}^{Q,Q'}$ are independent of the surrounding atomic configurations in a simulation of disordered system. For an effective medium calculation of disordered system, it is not realistic to account for the variance of the force constants induced by the completely random chemical environment. Thus, to realize the *ab initio* simulation of real disordered materials (for force-constant decomposition in 2D and 3D, please see the Appendix), we can use the force constants which are obtained by averaging over different surrounding atomic configurations calculated with first-principles super-cell method. Furthermore, a practical approach called transferable force-constant model [41,54] reported recently can be combined with our auxiliary CPA method to realize the *ab initio* modeling of disordered vibration (see more details in the Appendix). It is worth to mention that the implementation of present force-constant model and the auxiliary CPA for simulating the 3D realistic alloys is straightforward, and presently we have realized such an implementation and reproduced the phonon dispersions for several 3D alloys in comparison with ICPA results and experiments, which will be reported in our next work.

After presenting the effectiveness of present auxiliary CPA method in disordered lattice vibration, we want to mention that the separable form of Eq. (5) is also applicable to the general disordered electronic Hamiltonian. Thus, the auxiliary CPA method proposed above can be applied to simulate the electronic structure and electron transport in disordered systems with both diagonal and off-diagonal disorders. For electronic systems, we note that the first term in Eq. (5) is equivalent to the first-order TB-LMTO Hamiltonian with very good accuracy (see page 76 in Ref. [28]). Adding the λ_{ij} can make the representation of disordered electronic Hamiltonian with Eq. (5) more accurate.

VI. CONCLUSIONS

In this paper, we have reported an auxiliary coherent medium theory to treat both mass and force-constant disorders for lattice vibration in random binary alloys. Based on an approximate force-constant decomposition, the general force-constant disorder (correlating diagonal and off-diagonal disorders) is transformed to a diagonal-like disorder in an auxiliary Hamiltonian, enabling the use of coherent potential approximation. The applications of this method in the DOS calculation of 1D disordered harmonic chains show good agreement with the exact numerical results. Moreover, we show the averaged phonon transmission through a disordered device can be derived based on the auxiliary Green's function method in combination with vertex correction technique, enabling the thermal transport simulation of disordered devices. Hence, we conclude that this method provides an effective approach for simulating the disorder effects on the vibrational properties of random alloys/devices. The application of present auxiliary CPA to general disordered electronic systems is also straightforward.

Note added. Recently, we note that Ref. [55] just presented a study of effects of mass and force-constant disorders on phonon localization with typical medium theory (TMT) [56] in combination with DCA to treat cluster effects and BEB transformation for treating force-constant (off-diagonal) disorder. This work is an extension of the method TMT-DCA-BEB recently reported for electronic system [57]. However, due to the limitation of BEB transformation, the force-constant sum rule is hard to be fully satisfied for different finite size of clusters, and the corresponding induced error is unclear so far. Here we want to mention that the approach to effectively handle force-constant disorder is key for localization and other key phenomena for vibrational systems with disorders.

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APPENDIX: FORCE CONSTANT MODEL FOR 1D AND 3D BINARY ALLOYS

For a 1D binary alloy with nearest-neighbor force constants, force constants only can take three possible values k^{AA}, k^{AB}, and k^{BB}. Using the force constant model in Eq. (5) and defining a, b as

$$x_i = \begin{cases} a \text{ if A atom is at i site} \\ b \text{ if B atom is at i site} \end{cases},$$

we can obtain the separable forms of force constants

$$a^2 S + \lambda = k^{AA}, \tag{A1}$$

$$b^2 S + \lambda = k^{BB}, \tag{A2}$$

$$abS + \lambda = k^{AB}. \tag{A3}$$

We set $a = 1$ and then obtain the solution

$$\lambda = \frac{k^{AA}k^{BB} - k^{AB2}}{k^{AA} + k^{BB} - 2k^{AB}}, \tag{A4}$$

$$S = \frac{(k^{AB} - k^{AA})^2}{k^{AA} + k^{BB} - 2k^{AB}}, \tag{A5}$$

$$b = \frac{k^{BB} - k^{AB}}{k^{AB} - k^{AA}}. \tag{A6}$$

Then we can write the $\mathcal{K}_i$ explicitly

$$\mathcal{K}_i = \begin{bmatrix} 0 & \dots & 0 & \dots & 0 \\ \dots & Sx_i & -Sx_i & 0 & \dots \\ \dots & -\frac{\lambda}{x_i} & \frac{2\lambda}{x_i} & -\frac{\lambda}{x_i} & \dots \\ \dots & 0 & -Sx_i & Sx_i & \dots \\ 0 & \dots & 0 & \dots & 0 \end{bmatrix}. \tag{A7}$$

The above exact representation is based on the approximation of nearest-neighbor interaction. If considering the interaction beyond the nearest neighbor (usually significantly smaller than nearest-neighbor interaction), then we can obtain the optimal parametrization for the separable force-constant model by numerical optimization.

For a general 2D/3D lattices, k_{ij}, S_{ij}, λ_{ij} will be replaced by tensors with Cartesian coordinates $k_{ij,\alpha\beta}$, $S_{ij,\alpha\beta}$, $\lambda_{ij,\alpha\beta}$:

$$a^2 S_{\alpha\beta} + \lambda_{\alpha\beta} = k^{AA}_{\alpha\beta}, \tag{A8}$$

$$b^2 S_{\alpha\beta} + \lambda_{\alpha\beta} = k^{BB}_{\alpha\beta}, \tag{A9}$$

$$abS_{\alpha\beta} + \lambda_{\alpha\beta} = k^{AB}_{\alpha\beta}. \tag{A10}$$

To realize the phonon calculation in real materials with atomic disorder, the well-established transferable force-constant model can be adopted for the implementation of auxiliary CPA simulation. The transferable force-constant tensors take the form [42,54]

$$k = \begin{bmatrix} k_s & 0 & 0 \\ 0 & k_b & 0 \\ 0 & 0 & k_b \end{bmatrix}, \tag{A11}$$

where k_s represents the stretching stiffness along the bond and k_b represents the isotropic bending stiffness.

Unlike the 1D case in Eqs. (A1)–(A6), for arbitrary parameters k_s and k_b, an exact representation of the force-constant is difficult in the present separable form in Eqs. (A8)–(A10). Thus, we propose to use the optimization method to fit our separable force-constant model, namely the parameters a, b and tensors S_{ij} and λ_{ij}, to the results obtained by first-principles calculations or some transferable force fields. In such as optimization for a 3D lattice, the inclusion of a cluster of neighboring sites with direct coupling to the central sites are required, and this optimization can be realized by the conjugate-gradient method. For example, we consider the FCC $\mathrm{Pd}_{0.96}\mathrm{Fe}_{0.04}$ alloy with force constants given by the first-principles calculation in Ref. [42]: $k_s^{\mathrm{FeFe}} = 13366$, $k_s^{\mathrm{PdPd}} = 45925$, $k_s^{\mathrm{FePd}} = 35698$, $k_b^{\mathrm{FeFe}} = -566$, $k_b^{\mathrm{PdPd}} = -2424$, and $k_b^{\mathrm{FePd}} = -1879$ (in unit of dyn cm^{-1}). The optimized representation will give $S_s = -41191.8$, $\lambda_s = 54557.4$, $S_b = 2362.2$, $\lambda_b = -2935.9$, and $b = 0.4578$ with approximate force constants $\tilde{k}_s^{\mathrm{FeFe}} = 13365.6$, $\tilde{k}_s^{\mathrm{PdPd}} = 45924.4$, $\tilde{k}_s^{\mathrm{FePd}} = 35699.8$, $\tilde{k}_b^{\mathrm{FeFe}} = -573.7$, $\tilde{k}_b^{\mathrm{PdPd}} = -2440.8$, and $\tilde{k}_b^{\mathrm{FePd}} = -1854.5$, with accuracy significantly beyond the Shiba's and virtual-crystal models. Thus, with the separable force constant model and auxiliary CPA, the effect of the force-constant disorder together with mass disorder can be simulated for real materials, with the expense of the little error in some specific force constants.

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