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Pseudofermion approach to binary disordered systems. II. Goldstone systems with off-diagonal disorder

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A simple approximation is developed for the frequency spectrum of binary disordered Goldstone systems such as phonons in mixed crystals with both mass and force-constant disorder. This localized pseudofermion approximation is a simplification of the one-pseudofermion approximation previously discussed, which gives the coherent-potential approximation for systems with diagonal disorder. The approximation is shown to give properly analytic Green's functions, and to have the correct virtual-crystal, atomic, and dilute limits. Numerical results are presented for linear chains of atoms for comparison with the exact density of states, and results are given in the random-hopping electronic model for comparison with the theory of Blackman *et al*. It is found that the theory produces results very similar to those produced by the coherent-potential approximation for diagonal disorder. However, the neglect of pseudofermion propagation and certain overlap effects leads to somewhat incorrect locations of the impurity band edges. This effect vanishes when the disordered force constants superimpose linearly.

I. INTRODUCTION

A great deal of the literature on the theory of disordered systems has been concerned with extensions of the coherent-potential approximation (CPA).[1,2] Besides many attempts to include multisite scattering[3-14] and short-range correlations numerous efforts have been made to generalize the CPA to systems with off-diagonal disorder.[15-19] It is the purpose of this paper to show that the pseudofermion method we have recently developed[20] to derive analytical multisite generalizations of the CPA for diagonally disordered, binary systems can be easily extended to randomly disordered binary systems with off-diagonal disorder. In particular, we shall consider disordered Goldstone systems, such as the lattice vibrations in a site-disordered binary alloy $A_{c_A}B_{c_B}$ with mass and force-constant disorder, and spinwaves in site-disordered magnets. As a specific example for numerical calculations we shall use a disordered phonon system with random masses $m_i = m_A, m_B$ connected by force constants ϕ_{ij} which take the values $\phi_{ij}^{AA}, \phi_{ij}^{AB} = \phi_{ij}^{BA}, \phi_{ij}^{BB}$ depending on the occupation of sites i and j and with corresponding probabilities $c_A^2, c_A c_B$, and c_B^2 where c_A and $c_B = 1 - c_A$ are the concentrations of the two constituents A and B, respectively. For comparison of the results we shall also consider the random-hopping model of Blackman, Esterling, and Berk (BEB).[16] This is a tight-binding electronic model with random site energies $\epsilon_i = \epsilon_A, \epsilon_B$ and site-disordered hopping matrix elements $W_{ij} = W_{ij}^{AA}, W_{ij}^{AB} = W_{ij}^{BA}, W_{ij}^{BB}$.

The presence of off-diagonal disorder leads in general to two fundamental difficulties. First, the perturbation associated with a single impurity atom at a lattice site j, described by a single-impurity potential $V^{(j)}$, is not confined to the impurity site as in diagonally disordered systems, but extends to its neighborhood (the range of $V^{(j)}$). For simplicity, we shall restrict our calculations here to the case where the range of $V^{(j)}$ includes only site j and its nearest neighbors. In the case of Goldstone systems, the impurity-site diagonal element, the off-diagonal elements between the impurity and its neighbors, and the diagonal elements on neighboring sites are all changed by the presence of the impurity $V^{(j)}$. For the random-hopping model of BEB, on the other hand, the diagonal elements in the neighborhood of an isolated impurity remain unchanged. In this case, only s-wave scattering off the impurity site occurs as opposed to Goldstone systems where additionally p- and d-wave scattering must be treated.[21]

The second fundamental difficulty which one generally encounters is the loss of additivity of the impurity potential. In diagonally disordered systems the impurity potential V which describes the perturbation caused by a distribution of impurities can always be written as a sum $\sum_j V^{(j)}$ of single-impurity potentials $V^{(j)}$. Since in general $V^{(j)}$ extends to the neighborhood of site j, the regions where impurities on different lattice sites cause perturbations may overlap. This perturbation $V^{(j)}$ caused by each impurity j is generally a function of the number and location of any further impurities in its neighborhood, so that additivity is lost, except in the dilute limit where each impurity is, with high probability, in a pure host environment. Extended single-impurity potentials and the occurence of overlap are the characteristic features of

site-disordered systems with off-diagonal disorder. These overlap effects have been hard to handle and have made generalizations of the CPA to these systems difficult.

One previous method has been to ignore overlap effects by attributing to each impurity the perturbation it would cause in a pure host environment.[1,22-25] With this approximation a formal matrix generalization of the CPA can be easily obtained as was first shown by Takeno.[22] Such an approach was, for example, used by Harris *et al.*[23] and Holcomb[24] in their treatment of spin waves in diluted ferromagnets and antiferromagnets, respectively, and by Diehl and Biem[25] to describe the librational excitations in the orientationally ordered phase of solid mixtures of ortho and para hydrogen. Although this procedure is legitimate for sufficiently low impurity concentrations it clearly leads to incorrect self-energies in the limit where the host concentration goes to zero (unless the potential is additive as explained below). Since the overlap becomes more and more important for increasing impurity concentrations this "low-concentration CPA" becomes increasingly worse and may even completely break down for sufficiently large concentrations. Thus, it is important to get both dilute limits $c_A \to 0$ and $c_B \to 0$ in generalizations of the CPA right. Only then, and if the analytic structure of the self-energy, and the translational invariance of the average propagator are preserved, and both weak- and strong-scattering limits are correct, can one expect the theory to give a reasonable description for the whole concentration regime $0 \leq c_A \leq 1$ and for a wide range of scattering parameters. On the other hand, it can happen, for special choices of the potential parameters, that the impurity potential is additive. Then it is obvious that Takeno's method leads to a proper generalization of the CPA which preserves both dilute limits. For example, in the case of the phonon system described above this additivity occurs if, and only if the force constants ϕ_{ij} superimpose linearly, i.e., for $\phi_{ij}^{AB} = \frac{1}{2}(\phi_{ij}^{AA} + \phi_{ij}^{BB})$. Kaplan and Mostoller[18] have treated lattice vibrations in disordered lattices by this method.

Likewise the random-hopping model of BEB becomes additive if the hopping matrix elements superimpose linearly. Also, other simplifications occur in this model for the separable case $W_{ij}^{AB} = (W_{ij}^{AA} W_{ij}^{BB})^{1/2}$ as shown by Shiba.[15] (The simplifications caused by these special choices of the potential parameters were investigated in some detail by Niizeki.[17]) For the random-hopping model, the treatment of overlap effects can, however, by avoided quite generally by using the locator matrix expansion of BEB.[16] The advantage of this expansion is that the randomness is completely described by independently distributed single-site quantities, the locator matrices. Thus the system can be treated in a similar way as are diagonally disordered systems. We have recently generalized the theory of BEB by including multisite scattering processes.[26] In the special case where W_{ij}^{AB} is the arithmetic mean of W_{ij}^{AA} and W_{ij}^{BB} Takeno's method can again be applied to obtain the analog of Kaplan and Mostoller's phonon theory. Niizeki proved that this theory is equivalent to BEB theory (for the same special choice of W_{ij}'s).[17] Also, Blackman showed[27] that Shiba's theory which applies when W_{ij}^{AB} is the geometric mean of W_{ij}^{AA} and W_{ij}^{BB} can be recovered by specializing the equations of BEB.

In Goldstone systems the inverse of the propagator has diagonal elements depending on the occupation of neighboring sites. In our case the propagator is the displacement-displacement Green's function G with

$$(G^{-1})_{ij} = (m_i \omega^2 - \sum_{l(\neq i)} \phi_{il}) \delta_{il} + \phi_{ij} . \tag{1}$$

It is precisely this neighbor dependence of Goldstone systems which makes them more difficult to treat than the BEB model. In addition there exist cases other than Goldstone systems, such as electrons in alloys with charge transfer to neighboring sites or librons in solid mixtures of ortho and para hydrogen where the diagonal perturbation may depend upon the occupation of neighboring sites. In any of these cases the locators $g_i = (G^{-1})_{ii}^{-1}$ are no longer single-site quantities. To specify the value of g_i one must fix the occupation of the site i and all of its neighbors. Nevertheless, it is still possible to completely describe the randomness in terms of locator matrices whose diagonal elements correspond to the values g_i takes in the 2^{z+1} different configurations of the site i and its z neighbors. But due to the presence of overlap between the neighborhoods of two nearby sites these locator matrices are not statistically independent and one is faced with a kind of short-range order. (Averages of products of these locator matrices factorize only if the nearest-neighbor clusters of the sites to which they refer don't overlap.) Nevertheless one can thus generalize BEB theory[26] and extend the CPA to Goldstone systems by treating these short-range order effects using a method similar to the one of Watabe and Yonezawa.[28] Unfortunately the resulting equations are complicated mainly because of their high 2^{z+1} dimensionality in k space. It would seem that these complications are the price to be paid for properly including overlap effects.

In this paper we shall apply the pseudofermion

approach, which was described in our earlier paper[20] (hereafter called I). In this approach, which is based on Mookerjee's augmented-space method,[11,13] we introduce configuration states $|iA\rangle$ and $|iB\rangle$ at each lattice site i to describe the species occupation. Alloy configurations can then be represented in the form $|\cdots lS_l\cdots\rangle$ as a product of single-site configuration states $|lS_l\rangle$ where S_l $(=A,B)$ characterizes the species occupation of the site l.

In conventional methods where one works with configuration-dependent quantities such as m_i, ϕ_{ij}, and G_{ij}, the configuration dependence can be conveniently transferred to the indicator functions $\eta_l(A)$ and $\eta_l(B)$ which take the values 1 or 0 depending on whether the species occupation of site l is as indicated. Then we introduce the related operators $\hat{\eta}_l(S)$, as defined by their eigenvalue relations,

$$\hat{\eta}_l(S)\,|lS'\rangle = \delta_{SS'}|lS'\rangle . \tag{2}$$

For any configuration-dependent quantity, say Q, we then define a corresponding augmented-space operator by expressing Q in terms of η's (such that the disorder is completely contained in the η's and the coefficients are configuration independent) and making the replacements $\eta \to \hat{\eta}$. This gives, for example,

$$\hat{m}_i = m_A\hat{\eta}_i(A) + m_B\hat{\eta}_i(B) \tag{3}$$

and

$$\hat{\phi}_{ij} = \sum_{S,S'=A,B} \phi_{ij}^{SS'}\hat{\eta}_i(S)\hat{\eta}_j(S') . \tag{4}$$

[Strictly speaking, quantities like $\hat{\eta}_i(S)$, $\hat{m}_i$, and $\hat{\phi}_{ij}$ carrying site indices are operators only in the configuration space spanned by $\{|\cdots lS_l\cdots\rangle\}$ and not in the full augmented space which is a direct product of this space with the lattice space spanned by $\{|i\rangle\}$. To avoid confusion one may either think of, say $\hat{\phi}_{ij}$, as a matrix with respect to the omitted indices $\{lS_l\}$ or as an augmented-space operator defined by the matrix representation $\hat{\phi}_{ij}|i\rangle\langle j|$.]

We thus find, in particular, that the augmented-space operator $\hat{K}$ corresponding to $K = G^{-1}$ can be written in an obvious notation as

$$\hat{K} = \sum_i \left(\hat{m}_i\omega^2 - \sum_{l(\neq i)}\hat{\phi}_{il}\right)|i\rangle\langle i| + \sum_{i\neq j}\hat{\phi}_{ij}|i\rangle\langle j| . \tag{5}$$

The augmented-space Green's function is given by

$$\hat{G} = \hat{K}^{-1} . \tag{6}$$

Rather than using this A-B representation it is convenient to make a unitary transformation to the pseudo fermion representation defined by the states

$$|i0\rangle = c_A^{1/2}|iA\rangle + c_B^{1/2}|iB\rangle \tag{7}$$

and

$$|i1\rangle = c_B^{1/2}|iA\rangle - c_A^{1/2}|iB\rangle . \tag{8}$$

States in configuration space with lattice sites $lmn\cdots p$ in a $|1\rangle$ state and all other sites in a $|0\rangle$ state are denoted by $|f_{lmn\ldots p}\rangle$ and the augmented-space state $|i\rangle|f_{lmn\ldots p}\rangle$ by $|if_{lmn\ldots p}\rangle$, which is the more commonly used notation. Lattice sites in $|1\rangle$ ($|0\rangle$) states will be referred to as sites at which one (no) pseudofermion is present.

The convenience of the pseudofermion representation stems from the fact that the average $\bar{A}$ of a physical quantity A can be calculated simply as a matrix element of the corresponding augmented-space operator $\hat{A}$,

$$\bar{A}_{ij} = \langle if|\hat{A}|jf\rangle , \tag{9}$$

where $\langle if|$ is a state with no pseudofermions present. Hence $\bar{A}$ corresponds to the projection of $\hat{A}$ onto the subspace with no pseudofermions (which we shall call $S^{[0]}$) and we can represent $\hat{A}$ as a matrix in the block form

$$\hat{A} = \begin{pmatrix} \bar{A} & A' \\ A'^{\dagger} & \tilde{A} \end{pmatrix} \tag{10}$$

with

$$\bar{A} = P^{[0]}\hat{A}P^{[0]} , \tag{11a}$$

$$A' = P^{[0]}\hat{A}Q^{[0]} , \tag{11b}$$

and

$$\tilde{A} = Q^{[0]}\hat{A}Q^{[0]} , \tag{11c}$$

where we have introduced the projectors

$$P^{[0]} = \sum_i |if\rangle\langle if| \tag{12a}$$

and

$$Q^{[0]} = \hat{1} - P^{[0]} \tag{12b}$$

onto $S^{[0]}$ and its orthogonal complement $S_\perp^{[0]}$, respectively.

Finally, we can easily calculate the self-energy Σ in this representation in terms of the virtual-crystal propagator $G^{vc} = \bar{K}^{-1}$ by

$$\bar{G} = [(G^{vc})^{-1} - \Sigma]^{-1} . \tag{13}$$

The result is

$$\Sigma = K'\tilde{K}^{-1}K'^{\dagger} . \tag{14}$$

This formula is the analog of Eq. (23) in I which we used as a starting point for deriving generalizations of the CPA for diagonally disordered systems. We showed there that this equation gener-

ates an expansion for Σ in terms of agumented-space paths and that the usual single-site CPA can be recovered by restricting the paths to involve only those states which have at most one pseudofermion present at a time and by subsequently making the theory self-consistent. Likewise cluster generalizations or n-pseudofermion generalization of the CPA can be derived by including terms which involve at most n pseudofermions at a time.[20]

The analogous approximation for (Goldstone) systems with off-diagonal disorder is obvious. In particular, it seems to be obvious to attempt a generalization of the self-consistent one-pseudofermion approximation along these lines. The presence of off-diagonal disorder leads, however, to several complications. First, K' is no longer site diagonal as was H' in I for diagonal disorder. Thus, pseudofermion creation or destruction processes can take place simultaneously with real-space hops. Second (due to the term $\sum_{l(\neq i)}\hat{\phi}_{il}$), more than one (up to z, the number of nearest neighbors) pseudofermions can be created or destroyed at once. This feature will be discussed in detail in Sec. II where we show how Σ can be represented in terms of augmented-space paths and classify the various pseudofermion processes that occur. Nevertheless, a well defined, non-self-consistent form of the one-pseudofermion approximation is obtained by projecting out all states ($\in S_{\perp}^{[0]}$) with more than one pseudofermion. This approximation shows immediately a new feature: the (single) pseudofermion can propagate through the crystal by nearest-neighbor hops, since K' is off-diagonal. This approximation is the analog of the average-t-matrix approximation[1] for the case of diagonal disorder.

The third, and probably most severe new complication, is that there is no obvious, unique way to introduce self-consistency, even in the one-pseudofermion approximation. A reasonable scheme would be to include further pseudofermion processes of the same kind already taken into account, that is, one would include all processes which correspond to a similar constraint as that in diagonally disordered systems. In the diagonal case, the rule (for the self-consistent, one-pseudofermion approximation, corresponding to the CPA) is to restrict the contributions included by requiring that any pseudofermion can only be (finally) destroyed if all others created at a later time have been previously destroyed. [Processes in which a pseudofermion is only temporarily destroyed, i.e., destroyed and immediately reexcited are however allowed. (See I.)] If one tries to carry out this program for Goldstone systems with off-diagonal disorder one faces again "overlap problems;" since the presence of further pseudofermions in the vicinity of a single pseudofermion changes the augmented-space matrix elements of $\hat{K}$, one must keep track of all pseudofermions within the nearest-neighbor cluster of each lattice site. Such a theory would be very similar to our locator-matrix treatment of Goldstone systems[26] and correspondingly as complicated.

We propose a simpler self-consistent theory here, which is presented in Sec. III. First, we simplify the non-self-consistent equations by neglecting the pseudofermion propagation through the crystal. We shall call this the localized, one-pseudofermion approximation (LPF). We then have to calculate Green's functions only for a single pseudofermion at a particular lattice site. Clearly, a single pseudofermion then plays a role similar to an isolated impurity in conventional theories based on Takeno's method[22] and the self-energy is calculated self-consistently by associating with each pseudofermion the perturbation it would cause in an effective medium without pseudofermions.

The theory which then results has a structure like Takeno's matrix generalizations of the CPA[21] and some remarkable properties. First, we find that for either $c_A \to 0$ or $c_B \to 0$, the LPF approximation reverts to the corresponding low-concentration CPA. This means in particular that the self-energy is correct in both dilute limits. Second, the LPF theory reduces to that of Kaplan and Mostoller[18] if the force constants superimpose linearly (additive limit). The theory can also be applied to the BEB model and is similarly equivalent to BEB theory[16] in the additive case.

In Sec. IV we present numerical results for the density of states of one-dimensional alloys in the LPF approximation and compare them with corresponding exact results obtained by the Schmidt method.[29] Both our phonon model and the random-hopping model are investigated for a variety of potential parameters and concentrations. Concluding remarks are contained in Sec. V. In Appendix A we prove the analyticity of the LPF theory, in Appendix B we derive analytical formulas for the Green's functions of a linear chain with nearest- and next-nearest neighbor interactions, and in Appendix C we derive rigorous bounds on the phonon spectrum of alloys with mass and force-constant disorder.

II. AUGMENTED-SPACE PATHS

In this section we discuss the diagrammatic expansion of Σ in augmented-space paths. In order to obtain this expansion, we rewrite $\tilde{K}$ in the form $m^0\omega^2Q^{[0]} - (m^0\omega^2Q^{[0]} - \tilde{K})$ and expand $\tilde{K}^{-1}$ in powers

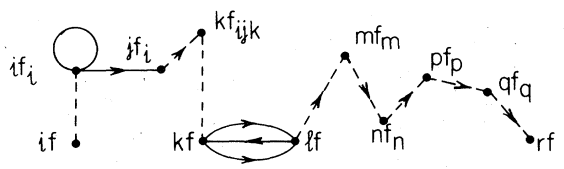

FIG. 1. An augmented-space path showing various pseudofermion creation and destruction processes (dashed lines). The states visited are indicated. In particular it is shown that pseudofermions can be created and destroyed simultaneously with real-space hops. Also an example for a process is given ($kf_{ijk} \rightarrow kf$) in which several pseudofermions are destroyed at once (i and j would both be neighbors of k) and the propagation of a single pseudofermion from site m to site g is shown.

of $(m^0\omega^2)^{-1}$ where m^0 is an arbitrary reference mass. Each individual term in this expansion can be represented diagrammatically as a path in the space $S_\perp^{[0]}$, an augmented-space path, in which at each instant at least one pseudofermion is present. A path is composed of lines connecting the dots that represent augmented-space states. Each visit of a state (i.e., everytime a line passes through the dot) is given a weight $(m^0\omega^2)^{-1}$ and each line connecting two augmented-space states, say $|if_\sigma\rangle$ and $|jf_{\sigma'}\rangle$, is given the weight $\langle if_\sigma | m^0\omega^2\hat{1} - \hat{K} | jf_{\sigma'}\rangle$. A typical augmented-space path diagram is shown in Fig. 1. This notation is the same as that used previously in I. The subscripts σ and σ' are the sets of sites with pseudofermions present. So for $\sigma = \{l, k, m, \ldots, p\}$, $|if_\sigma\rangle$ denotes the state $|if_{lkm\ldots p}\rangle$, and $|if\rangle = |if_\phi\rangle$ where ϕ is the null set. We note that states may be repeatedly visited; in particular, self-closing loops in which one immediately returns to the same state are allowed.

Furthermore, the (nonvanishing) matrix elements of K', namely, $\langle if | K' | jf_\sigma\rangle$ with $\sigma \neq \phi$, are represented graphically in the same way. Thus Σ_{ij} is given by the sum of all augmented-space paths which start from $|if\rangle$ go directly into the space $S_\perp^{[0]}$ (i.e., excite via K' at least one pseudofermion) and come directly (via $K'^\dagger$) back to $|jf\rangle$, with no intermediate visits to $S^{[0]}$.

The weights to be associated with the various lines (the link functions) can be expressed simply in terms of the matrix elements of $\hat{\phi}$ and $\hat{\mu} = \hat{m} - m^0\hat{1}$ of Eqs. (3) and (4). As a notation similar to $\bar{v}$, v', and $\tilde{v}$ of I, and that of Eq. (11) above, we introduce

$$\bar{\mu} = \langle if | \hat{\mu} | if\rangle = c_A m_A + c_B m_B - m^0 , \tag{15a}$$

$$\mu' = \langle if | \hat{\mu} | if_i\rangle = (c_A c_B)^{1/2}(m_A - m_B) , \tag{15b}$$

$$\tilde{\mu} = \langle if_i | \hat{\mu} | if_i\rangle = c_B m_A + c_A m_B - m^0 , \tag{15c}$$

and

$$\bar{\phi}_{ij} = \langle if | \hat{\phi}_{ij} | jf\rangle = c_A^2\phi_{ij}^{AA} + 2c_A c_B\phi_{ij}^{AB} + c_B^2\phi_{ij}^{BB} , \tag{16a}$$

$$\dot{\phi}_{ij} = \langle if_i | \hat{\phi}_{ij} | jf_i\rangle = c_A c_B(\phi_{ij}^{AA} + \phi_{ij}^{BB}) + (c_A^2 + c_B^2)\phi_{ij}^{AB} , \tag{16b}$$

$$\ddot{\phi}_{ij} = \langle if_{ij} | \hat{\phi}_{ij} | jf_{ij}\rangle = c_B^2\phi_{ij}^{AA} + 2c_A c_B\phi_{ij}^{AB} + c_A^2\phi_{ij}^{BB} , \tag{16c}$$

$$\phi'_{ij} = \langle if | \hat{\phi}_{ij} | jf_j\rangle = (c_A c_B)^{1/2}[c_A\phi_{ij}^{AA} + (c_B - c_A)\phi_{ij}^{AB} - c_B\phi_{ij}^{BB}] , \tag{16d}$$

$$\dot{\phi}'_{ij} = \langle if_i | \hat{\phi}_{ij} | jf_{ij}\rangle = (c_A c_B)^{1/2}[c_B\phi_{ij}^{AA} + (c_A - c_B)\phi_{ij}^{AB} - c_A\phi_{ij}^{BB}] , \tag{16e}$$

$$\phi''_{ij} = \langle if | \hat{\phi}_{ij} | jf_{ij}\rangle = \langle if_i | \hat{\phi}_{ij} | jf_j\rangle = c_A c_B(\phi_{ij}^{AA} - 2\phi_{ij}^{AB} + \phi_{ij}^{BB}) . \tag{16f}$$

In terms of this notation, each site carries the appropriate matrix element of $-(\hat{\mu}\omega^2 - \sum_j \hat{\phi}_{ij})$. For example, a self-closing loop at a lattice site with a pseudofermion, with n of its z nearest neighbors having pseudofermions, carries the weight

$$-\tilde{\mu}\omega^2 + n\ddot{\phi}_{ij} + (z - n)\dot{\phi}_{ij} .$$

III. ONE-PSEUDOFERMION APPROXIMATION

The non-self-consistent, one-pseudofermion approximation is defined by requiring that only augmented-space paths with one pseudofermion present at a time are allowed in the self-energy. The pseudofermion is created at a site in a process described by K' and can then either stay at that site or propagate via nearest-neighber hops. The possibility of pseudofermion propagation adds a new feature to the theory not present in diagonally disordered systems[20] and which clearly is a consequence of the overlap effects described above. These processes carry the weights $\pm\phi''_{ij}$ and thus do not contribute in the additive case where ϕ''_{ij} vanishes [see Eq. (16f)]. To derive Σ in this approximation we introduce the real-space matrix $K^{(1)}$ with elements

$$K_{ij}^{(1)} = -\langle if | \hat{K} | jf_i\rangle . \tag{17}$$

Then, using Eq. (14), we obtain

$$\Sigma = \sum_{ll'} K^{(l)} g^{(l)(l')} K^{(l')\dagger} , \tag{18}$$

where $g_{ij}^{(l)(l')}$ corresponds to $\langle if_l | \tilde{K} | jf_{l'}\rangle$ in the one pseudofermion approximation and is given by the sum of all one-pseudofermion walks from the state $|if_l\rangle$ with a pseudofermion at lattice site l to the state $|jf_{l'}\rangle$ with a pseudofermion at l'. This approximation is analogous to the average-t-matrix

approximation for diagonally disordered systems.

Those paths in which the initially created pseudofermion stays at the site l are particularly easy to sum. We denote their sum by $G^{\{l\}}$, the localized one-pseudofermion approximation. Since corresponding link functions are the same everywhere except in the neighborhood of the pseudofermion, $G^{\{l\}}$ is similar to a single-impurity Green's function and one finds

$$G^{\{l\}} = (G^{vc-1} - Y^{\{l\}})^{-1}, \tag{19}$$

where $Y^{\{l\}}$ describes the perturbation caused by the single pseudofermion at lattice site l in a virtual-crystal medium and is given by

$$Y_{ij}^{\{l\}} = \begin{cases} (\bar{m} - \tilde{m})\omega^2 - \sum_{m(\neq l)} (\bar{\phi}_{lm} - \dot{\phi}_{lm}), & \text{for } i=j=l \\ \bar{\phi}_{ij} - \dot{\phi}_{ij}, & \text{for } i=l\neq j \text{ or } j=l\neq i \\ \dot{\phi}_{il} - \bar{\phi}_{il}, & \text{for } i=j\neq l. \end{cases} \tag{20}$$

In the paths contributing to $g^{\{l\}\{l'\}}$ in which pseudofermion propagation occurs one can make a partial resummation to obtain $g^{\{l\}\{l'\}}$ in terms of $G^{\{l\}}$,

$$g^{\{l\}\{l'\}} = G^{\{l\}}\delta_{ll'} - \sum_m G^{\{l\}} R^{\{l\}\{m\}} g^{\{m\}\{l'\}}, \tag{21}$$

where

$$R_{ij}^{\{l\}\{m\}} = (1-\delta_{lm})\langle if_l | \hat{K} | jf_m \rangle. \tag{22}$$

[The results in Eqs. (18) and (22) can also be directly obtained from Eq. (14) by projecting onto the one-pseudofermion subspace, i.e., Eq. (18) is equivalent to $\Sigma = K' P^{[1]} (P^{[1]} \bar{K} P^{[1]})^{-1} P^{[1]} K'^{\dagger}$ where $P^{[1]}$ is the projector onto the one-pseudofermion subspace.] The evaluation of the above formula would presumably give results equally as good as does the average-t-matrix approximation for diagonal disorder. The evaluation is more difficult, however, because of the pseudofermion propagation.

To introduce self-consistency one would like to include further contributions from processes similar to those already included. By analogy with the pseudofermion derivation of the usual single-site CPA[20,30] one would thus allow that further pseudofermions are created but require that those created last are destroyed first. As indicated above this concept leads to difficulties because one must keep track of all pseudofermions which are excited in the neighborhood of a given lattice site in order to assign the correct weights to the processes which start at this partuclar site. This introduces short-range order into the calculation.

A very simple theory can, on the other hand, be obtained as follows: First we neglect the propagation of pseudofermions, i.e., make the localized pseudofermion approximation. Then, according to Eq. (18), Σ becomes a sum of single-site contributions $Q^{\{l\}} = K^{\{l\}} G^{\{l\}} K^{\{l\}}$ each of which is associated with a single pseudofermion at the site l. In each self-energy term involving a pseudofermion at the site l one can easily insert similar processes involving pseudofermions at all other sites if one neglects overlap effects. That is, one makes self-energy insertions corresponding to pseudofermions at all sites other than site l but ignores the different appropraite weighting for pseudofermions on neighbors of site l. If one neglects these overlap effects, G^{vc} in each of these terms gets renormalized by $[(G^{vc-1} - \sum_{m(\neq l)} Q^{\{m\}}]^{-1}$ and one finally obtains

$$\Sigma = \sum_l Q^{\{l\}}, \tag{23}$$

$$Q^{\{l\}} = K^{\{l\}} [\bar{G}^{-1} - (Y^{\{l\}} - Q^{\{l\}})]^{-1} K^{\{l\}\dagger}. \tag{24}$$

These equations, together with Eq. (13), define our self-consistent, localized one-pseudofermion approximation, hereafter called LPF approximation. The structure of these equations is similar to those of the usual matrix generalizations of the CPA which are based on Takeno's method.[22-25] This similarity becomes even more obvious if one realizes that $K^{\{l\}}$ and $Y^{\{l\}}$ are proportional to each other and can be written in terms of an effective single-impurity matrix $U^{\{l\}}$

$$K^{\{l\}} = -(c_A c_B)^{1/2} U^{\{l\}}, \tag{25a}$$

$$Y^{\{l\}} = (c_A - c_B) U^{\{l\}}. \tag{25b}$$

From Eqs. (17) and (20) one finds

$$U_{ij}^{\{l\}} = \begin{cases} (m_A - m_B)\omega^2 - \sum_{m(\neq l)} [c_A(\phi_{lm}^{AA} - \phi_{lm}^{AB}) - c_B(\phi_{lm}^{BB} - \phi_{lm}^{AB}], & \text{for } i=j=l \\ c_A(\phi_{ij}^{AA} - \phi_{ij}^{AB}) - c_B(\phi_{ij}^{BB} - \phi_{ij}^{AB}), & \text{for } i=l\neq j, \text{ or } j=l\neq i \\ c_B(\phi_{il}^{BB} - \phi_{il}^{AB}) - c_A(\phi_{il}^{AA} - \phi_{il}^{AB}), & \text{for } i=j\neq l. \end{cases} \tag{26}$$

Several important conclusions can be drawn from these results. First, the LPF approximations revert to the usual low-concentration (matrix) CPA in either one of the limits $c_A \to 0$ or $c_B \to 0$ and thus clearly preserves both dilute limits because $Y^{\{l\}}(c_B=0) = U^{\{l\}}(c_B=0)$ $[Y^{\{l\}}(c_A=0) = -U^{\{l\}}(c_A=0)]$ corresponds to the impurity potential for a single B (A) atom in an otherwise pure A (B) lattice and

because $(G^{vc})^{-1}$ to first order in $c_{B,A}$ can be written as $G^{A,B-1} - c_{B,A}\sum_j Y^{\{l\}}$ $(c_{B,A}=0)$, where $G^{A,B}$ are the Green's functions for a pure A and B lattice, respectively. So this theory can be regarded as one which interpolates between the low- $(c_B \ll 1)$ and high-concentration $(c_A \ll 1)$ CPA's.

Second, we note that, quite generally, the LPF approximation has the same formal structure as Takeno's matrix CPA applied to the Green's function $[(G^{vc})^{-1} - \sum_l V^{\{l\}}]^{-1}$, but with the "effective" random single-site potential

$$V^{\{l\}} = \eta_l(B) c_A U^{\{l\}} - \eta_l(A) c_B U^{\{l\}} . \tag{27}$$

This parallel can easily be seen by rewriting the CPA equations in the pseudofermion form where, by analogy with the quantities $\bar{v}, v', \tilde{v}$ we used in I for diagonally disordered systems, one introduces matrices $\bar{V}^{\{l\}}$, $V^{\{l\}'}$, and $\tilde{V}^{\{l\}}$. Specifically, from Eq. (27) it is clear that $\tilde{V}^{\{l\}}$ is given by $c_B(-c_B U^{\{l\}}) + c_A(c_A U^{\{l\}})$ and equal to $Y^{\{l\}}$, that $\bar{V}^{\{l\}}$ vanishes, and that $V^{\{l\}'}$ is equal to $K^{\{l\}}$. From these relations and since $U^{\{l\}}$ is seen to describe the effect of a single B impurity in an A lattice when $\phi_{ij}^{AB} = \frac{1}{2}(\phi_{ij}^{AA} + \phi_{ij}^{BB})$, one can also conclude that the LPF theory reduces in the additive limit to that of Kaplan and Mostoller.

Furthermore, the theory clearly preserves the weak-scattering or virtual-crystal limit, where the fluctuations of m_i, and ϕ_{ij} about their average values $\bar{m}$, and $\bar{\phi}_{ij}$ go to zero, since it is an expansion about this limit. In the electronic problem one also is concerned about the strong-scattering or atomic limit which in the case of only diagonal disorder is defined as the case where the hopping element W_{ij} goes to zero (on the scale of $\epsilon_B - \epsilon_A$) so that the density of states consists of δ functions at ϵ_A and ϵ_B. The atomic limit for the case of off-diagonal disorder is somewhat more complicated; here one defines it as the limit of the norm of the hopping element $\|W\|$ going to zero, where, for example,

$$\|W\| = \max_{S,S' = A,B} W^{SS'}, \tag{28a}$$

or

$$\|W\| = \left(\sum_{S,S' = A,B} |W^{SS'}|^2 \right)^{1/2} . \tag{28b}$$

It is well known that both the single-site CPA and the BEB theory preserve this limit.[1] Thus, the LPF theory also preserves it, since as $\|W\|$ goes to zero the additivity ceases to be an important factor. For Goldstone systems, such as phonon systems the analogous limit can be defined but is generally less important, except for optical modes where the Einstein approximation is useful. This limit is also, of course, correctly preserved by the LPF approximation.

Finally, the theory also preserves the correct analyticity properties, in particular, the important herglotz property

$$\mathrm{Im}\Sigma(z^2) = [\Sigma(z^2) - \Sigma(z^2)^\dagger]/2i > 0 \quad \text{for } \mathrm{Im}z^2 > 0 . \tag{29}$$

This property is satisfied because Σ satisfies Mills' symmetry conditions[30] since Σ can be represented by the diagrams topologically equivalent to those occurring in the pseudofermion formulation of the CPA (discussed in I). In Appendix A we show how it can be proven that the LPF equations have a unique analytical solution for all z^2 with $\mathrm{Im}\, z^2 \neq 0$.

IV. NUMERICAL RESULTS

In this section we apply the LPF approximation in detail to one-dimensional systems in order to evaluate its usefulness. We investigate both the phonon model and the BEB random-hopping model and present numerical results for the density of states in both cases. For the BEB model the density of states $\rho_e(\omega)$ is given by

$$\rho_e(\omega) = -\mathrm{Im}\, \bar{G}_{00}(\omega + i0)/\pi , \tag{30}$$

but for the phonon system[1] one has

$$\rho(\omega^2) = -\mathrm{Im}\langle m_0 G_{00}(\omega^2 + i0)\rangle/\pi \tag{31}$$

so that we must calculate $\langle m_0 G_{00}\rangle$. But this weighted Green's function can be written as

$$\langle m_0 G_{00}\rangle = \bar{m}\bar{G}_{00} + \mu' \langle 0 f_0 | \hat{G} | 0 f\rangle \tag{32}$$

[where μ' is defined in Eq. (15b)]. It then remains to calculate $\langle 0 f_0 | \hat{G} | 0 f\rangle$ in a way which is consistent with the LPF approximation for $\bar{G}$ (i.e., by including the same type of pseudofermion processes). This reasoning leads to

$$\langle 0 f_0 | \hat{G} | 0 f\rangle = \{[1 - \bar{G}(Y^{\{0\}} - Q^{\{0\}})]^{-1} \bar{G} K^{\{0\}} \bar{G}\}_{00} . \tag{33}$$

One can easily check that these equations (32) and (33) are also consistent with the usual CPA result

$$\langle m_0 G_{00}\rangle = c_A m_A \{[\bar{G}^{-1} - (V_A^{(0)} - Q^{(0)})]^{-1}\}_{00} + c_B m_B \{[\bar{G}^{-1} - (V_B^{(0)} - Q^{(0)})]^{-1}\}_{00} , \tag{34}$$

where $V_{A,B}$ are the matrix values of V provided $-K^{\{l\}} = (c_A c_B)^{1/2}(V_A^{\{l\}} - V_B^{\{l\}})$ and $-Y^{\{l\}} = c_B V_A^{\{l\}} + c_A V_B^{\{l\}}$, where we have taken $\bar{V} = 0$ or $m^0 = \bar{m}$.

For one-dimensional systems the (nonvanishing parts of the) matrices $K^{\{0\}}, Y^{\{0\}}, Q^{\{0\}}$ are 3×3 matrices which transform according to $2A_g + A_u$, where $A_{g,u}$ are the irreducible representations of the point group C_{2i}. Thus only 2×2 matrices need to be inverted in order to evaluate the LPF formula, Eq. (24).

Once the self-energy is known one also has to calculate the Green's function which is given by

$$\bar{G}_{0n}=\frac{1}{2\pi}\int_{-\pi}^{\pi}[\bar{m}\omega^2-2\bar{\phi}(1-\cos k)-\Sigma_k]^{-1}\cos(nk)\,dk,$$

$$\text{for } n=0,1,2 \quad (35a)$$

where

$$\Sigma_k=\sum_{n,n'}Q_{nn'}^{\{0\}}\cos[k(n-n')] \quad (35b)$$

is the Fourier transform of Σ. Since for nearest-neighbor force-constant disorder, $Q_{-1+1}^{\{0\}}$ is in general nonvanishing, the self-energy Σ has both nearest- and next-nearest neighbor elements. Nevertheless, Eq. (35) can be analytically evaluated (Appendix B). We first introduce the coefficients $a(z^2), b(z^2), d(z^2)$ by

$$\bar{G}_k^{-1}=a+b\cos k+d\cos 2k\,, \quad (36)$$

where $\bar{G}_k^{-1}$ is the expression inside the square brackets of Eq. (35a). The resulting analytical form (evaluated in Appendix B) is

$$\bar{G}_{0n}=2d^{-1}\sum_{\substack{i=1\\ |\zeta_i|<1}}^{4}\zeta_i^{n+1}\left(\prod_{j(\neq i)=1}^{4}(\zeta_i-\zeta_j)\right)^{-1}, \quad (37a)$$

with

$$\zeta_{1,2,3,4}=\tfrac{1}{2}[y_\pm^2-4)^{1/2}]\,, \quad (37b)$$

where

$$y_\pm=-(b/2d)\pm[(b/2d)^2-2(a/d)+2]^{1/2}. \quad (37c)$$

For the BEB model similar simplifications occur in one dimension. Since for this model we would like to compare the LPF results with the BEB approximation we also briefly recall some details of that theory.[16] In the BEB theory $\bar{G}_{00}$ is written as

$$\bar{G}_{00}=\gamma_A+\gamma_B\,, \quad (38)$$

where

$$\gamma_S=[c_S(\omega)-\epsilon_S-U_{SS}]^{-1}\,, \quad (39)$$

$(S=A,B)$ are elements of a 2×2 locator matrix

$$\gamma=\begin{pmatrix}\gamma_A & 0\\ 0 & \gamma_B\end{pmatrix} \quad (40)$$

and in which U_{SS} are the diagonal elements of the interactor matrix

$$U=\begin{pmatrix}U_{AA} & U_{AB}\\ U_{AB} & U_{BB}\end{pmatrix}. \quad (41)$$

U is determined from the condition that the $i=j$ block of the propagator matrix $\langle\eta_i(S)G_{ij}\eta_j(S')\rangle$, which describes the propagation of electrons from a lattice site i (occupied by a species S) to a site j (species S'), is equal to γ. In one dimension this yields

$$U/2=W(\gamma^{-1}+U/2)^{-1}W\,, \quad (42)$$

with $W=(W_{01}^{SS'})$, as is also shown in Appendix B.

The LPF equations and the BEB equations were solved numerically by the following procedure: We first chose complex values for $z^2=\omega_0^2+i\Delta$ (and $z=\omega_0+i\Delta$ in the electronic case) with $\Delta>0$ (usually $\Delta=0.04$) and a real part ω_0^2 which was bigger than the upper band edge and solved the equations iteratively. Then ω_0^2 or ω_0 was decreased by a finite amount δ (usually $\delta=0.1$) and the solutions at the points $\omega_0^2-n\delta+i\Delta$ were calculated using each time the results as the starting values in the iteration process for the following point. Finally, the limits $\Delta\to 0+$ were obtained by sequentially reducing Δ by one half of its previous value.

We present in Figs. 2 and 3, our numerical results for the phonon density of states $\rho(\omega^2)$ for various concentrations and force constants and compare them with the essentially exact results obtained by the Schmidt method.[29] In the additive case [Fig. 2(a)] the result agrees, as it should, with the CPA theory of Kaplan and Mostoller[18] when $\rho(\omega^2)$ is calculated according to Eqs. (24) and (32). (Actually, Kaplan and Mostoller used the approximation $\bar{m}\bar{G}_{ii}$ for $\langle m_iG_{ii}\rangle$ in their numerical calculations which introduces unnecessary errors. In particular, the A and B subbands are incorrectly weighted. Since $\bar{m}/m_B=\frac{7}{4}$ for $m_B=\frac{1}{2}m_A$ and c_B

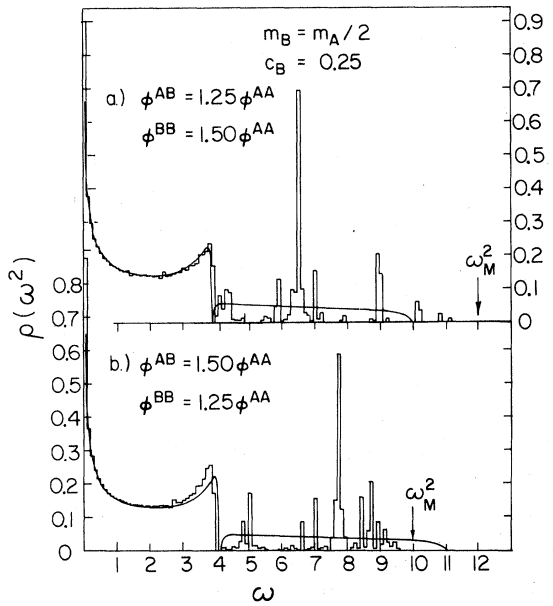

FIG. 2. Comparison of the phonon density of states calculated in the LPF approximation with the corresponding exact results (histogram) for $m_B=m_A/2$, $c_B=0.25$, and force-constants as given. ω^2 is given in units of ϕ^{AA}/m_A. In the case shown in (a) the force-constants are additive while they are not in (b). The arrow marks the exact upper band edge ω_M^2.

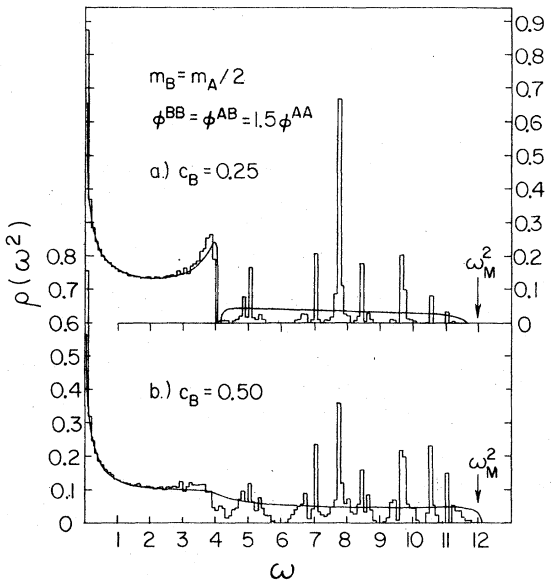

FIG. 3. LPF and exact results for the phonon density of states in a nonadditive case and for two different concentrations. At $c_B = 0.50$ and the LPF density of states extends beyond the exact upper band edge ($= 12$).

$= 0.25$ this approximation significantly overestimates the density of states in the B subband.) By and large the qualitative nature of the LPF results for nonadditive force constants seems to be almost as good as in the additive case (except for some shift of the position of the band edges as discussed below). As in the CPA for diagonally disordered systems, the LPF describes the host band rather well and gives a smooth density of states in the impurity region. The spiky structure of the minority band which originates from scattering by impurity pairs and clusters necessarily does not show up in this single-site theory.

For the diagonally disordered electronic model (Anderson's model) it is well known that all energy levels of any alloy configuration lie within the union of the pure bands of its constituents. The CPA preserves this property.[31,32] In Appendix C we prove the corresponding result for phonon systems with mass and force-constant disorder: The maximum phonon frequency ω_M of the alloy is the largest frequency of any of its pure configurations, namely, the two pure A and B lattices and the pure (two-sublattice) A-B alloy where each A atom is surrounded by B atoms and vice versa. In other words

$$\omega_M^2 \leq 2\max_{s,s'}[\phi^{ss'}(m_s^{-1} + m_{s'}^{-1})] . \tag{43}$$

Lifshitz[33] has pointed out that energy levels exist in disordered systems arbitrarily close to the pure band edges because arbitrarily large regions of these pure configurations occur with a nonvanishing, though exponentially small probability. This phenomenon gives rise to exponential bandtails in the vicinity of the pure band edges. A similar reasoning shows that the bound in Eq. (43) gives the correct upper band edge of the alloy. (The lower band edge which is realized in the Goldstone modes is obviously zero.) In each phonon case we have indicated the upper band edge ω_M^2 by an arrow in the figures.

It is clear from Fig. 2(b) that the LPF approximation violates somewhat the rigorous bound (43) in the case of nonadditive forces constants. Similar problems have occurred before. For example, the first attempts at self-consistent calculations (see, for example, Davies and Langer[34] and the discussion on p. 497 of Elliott *et al.*[1]) violated such bounds. These violations occurred because of overlap effects (multiple occupancy of sites by defects) which were corrected with the CPA. Later, matrix CPA theories were developed to handle dilute magnets[23,24] which worked well at low concentrations but produced lower band edges at slightly negative frequencies at high concentrations due to overlap effects. Two such errors are likewise giving rise to the band-edge shift here. The first occurs in the self-consistent insertions of the full Green's function into the self-energy; this allows the excitation of additional pseudofermions and ignores the overlap of the effects of these insertions with the original pseudofermions when the two are neighbors. The second error is the neglect of pseudofermion propagation, also a second-order effect.

In the first case, we have attributed to each pseudofermion, at, say, site j, the perturbation (the matrices $K^{(j)}$ and $Y^{(j)}$) it would cause in the absence of any other pseudofermion. But if, say, a second pseudofermion is excited at the nearest-neighbor site $j+1$, the matrix $Y^{(j)}$ replaces the bond $\bar{\phi}$ between sites j and $j+1$ by ϕ, but then $Y^{(j+1)}$ incorrectly adds again the correction $(\phi - \bar{\phi})$ instead of $(\ddot{\phi} - \bar{\phi})$, the correct version. This error is $2\phi - \bar{\phi} - \ddot{\phi} = (2\phi^{AB} - \phi^{AA} - \phi^{BB})(c_A - c_B)^2$ which, in the case of Fig. 2(b) is positive and thus is at least partly responsible for the high band edge. On the other hand, these errors vanish at $c_A = c_B = 0.50$ (as well as in the additive limit). Nevertheless, the LPF approximation [see Fig. 3(b)] still violates the rigorous bound in this case. This remaining error is caused by the neglect of pseudofermion propagation.

In order to explicitly evaluate the effect of the neglect of pseudofermion propagation, we calculate $R^{\{l\}\{m\}}$ of Eq. (22) for a linear-chain phonon system. The result is

$$R_{ij}^{\{l\}\{m\}} = \phi''(\delta_{l,m+1} + \delta_{l,m-1})[\delta_{il}\delta_{jm} + \delta_{im}\delta_{jl} - \delta_{ij}(\delta_{il} + \delta_{im})] . \quad (44)$$

For the parameters of Fig. 3(b), ϕ'' is negative, and thus from Eq. (21) it is clear that $R^{\{l\}\{m\}}$ decreases the effective force constant, in this case, by something of the order of ϕ''. Thus the inclusion of pseudofermion propagation reduces the band edge in this case as it must. Likewise in those cases where ϕ'' is positive the LPF theory underestimates the band edge, and in the additive limit, as $\phi'' \to 0$, these errors vanish linearly and the matrix CPA is recovered.

Both of these errors can be corrected but their correction introduces a kind of short-range order that makes the theory much more complicated to understand and numerically evaluate.

The LPF can also be applied to the random-hopping model. Just as in the phonon case, the LPF reduces, for additive hopping elements $W^{SS'}$, to a matrix CPA which is the analog of Kaplan and Mostoller's phonon theory.[18] Since for this model only s-wave scattering occurs, the part $Q_u^{\{0\}}$ of the single-site self-energy $Q^{\{0\}}$ which transforms according to A_u vanishes. Also, the CPA equation for the 2×2 s-wave block $Q_g^{\{0\}}$ is easily seen to be equivalent to Niizeki's equation.[17] Since Niizeki proved that these equations in the additive case have the same solutions as the BEB equations we can conclude that the LPF is precisely the BEB theory if the W's superimpose linearly. In Fig. 4, we give an example where the hopping elements are almost additive. The agreement is rather good as expected.

To exaggerate the differences between LPF and BEB theory we have chosen to investigate the extreme case where $W^{AB} = 0$ (the independent band limit). The BEB theory in this case reduces to a pair of self-consistent equations for the locators γ_A and γ_B that are independent.[16] A similar reduction does not occur in the LPF equations. That is, A and B subbands remain coupled. Figures 5(a)–5(c) show that the LPF (for these high A concentrations) gives a narrower majority subband while the minority subband while the minority subband can be either broader (at $c_B = 0.10$) or narrower (at $c_B = 0.30$) depending on the concentration. That the LPF gives a narrower majority subband for low c_B and $W^{BB} < W^{AA}$ can be qualitatively understood as follows. In the limit $c_B \to 0$ the LPF reverts to the low-concentration, additive CPA which, contrary to the BEB theory, does not appropriately treat the overlap effects of pairs of neighboring defects (which are pseudofermions in this limit). Clearly, the low-concentration, additive CPA and the LPF attribute a weaker bond to a pair of nearby (B) impurities (the low-concentration CPA gives even the negative value $-W^{AA}$) than the BEB theory which correctly uses the value $W^{BB} = 0.5W^{AA}$. Similarly the minority band for

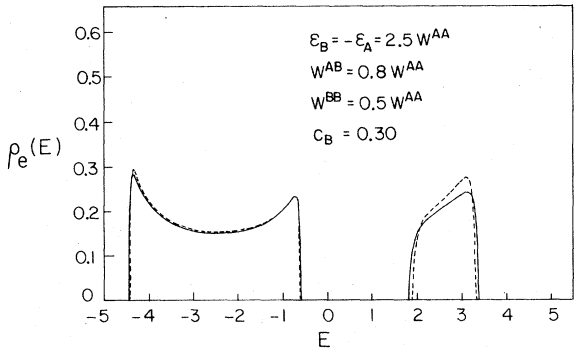

FIG. 4. Comparison between the LPF (full lines) and the BEB result (dashed lines) for the electronic density of states of the random-hopping model in a slightly nonadditive case. The energy E is given in units W^{AA}. The site energies used are $\epsilon_B = -\epsilon_A = 2.5\,W^{AA}$.

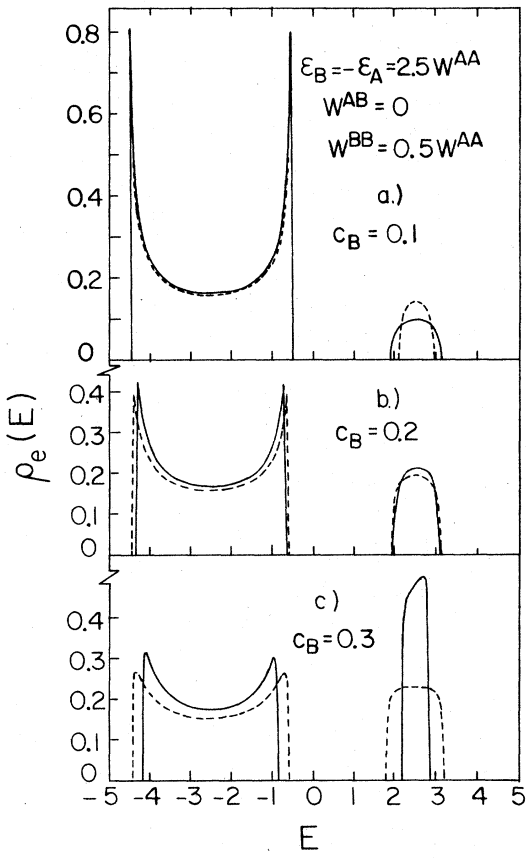

FIG. 5. Electronic densities of states calculated in the LPF theory (full lines) and in the BEB (dashed lines) theory as in Fig. 4 for $W^{AB} = 0$ and various concentrations. All other parameters are the same as in Fig. 4.

$c_B = 0.10$ seems to be broader than in the BEB theory because part of W^{AA} is incorrectly mixed with the B-B bond W^{BB}.

V. SUMMARY

We have shown that the pseudofermion approach as developed previously in I can be easily extended to Goldstone systems with off-diagonal disorder. In particular, the self-energy can be expanded in terms of pseudofermion, augmented-space paths as was done in the case of diagonal disorder. In the presence of off-diagonal disorder pseudofermions can, however, be created (or destroyed) simultaneously with real-space hops to neighboring sites and the pseudofermions can therefore propaate from site to site.

Also n-pseudofermion approximations can be developed as was done in I by including only those paths with at most n pseudofermions present at a time. For $n=1$, for example, the analog of the average-t-matrix approximation (ATA) is obtained. Because of the pseudofermion propagation, the single-site structure of the ATA equations is lost except for the additive case ($2\phi^{AB} = \phi^{AA} + \phi^{BB}$) where no pseudofermion propagation occurs.

We have derived a localized one-pseudofermion approximation by neglecting the pseudofermion propagation. To make the theory self-consistent we inserted self-energy parts corresponding to pseudofermions at all sites other than that of the localized pseudofermions but ignored the different appropriate weighting caused by pseudofermions on neighboring sites. This self-consistent localized one-pseudofermion approximation (LPF) is a proper generalization of the CPA in the sense that it preserves both dilute limits $c_A \to 0$ and $c_B \to 0$, is correct in the weak-scattering and the atomic limit and preserves the analytical structure (herglotz nature) of the self-energy as well as the translational invariance of the average propagator.

Furthermore, if the force constants superimpose linearly the LPF reduces to Kaplan and Mostoller's matrix CPA. Similarly, it is, in the case of the random-hopping (electronic) model, equivalent to the BEB theory if the hopping matrix elements are additive ($2W^{AB} = W^{AA} + W^{BB}$). For nonadditive force constants and hopping elements the LPF seems to give as good results as in the additive cases except near the band edges. Its less satisfactory description of the band edges in these cases is related to the occurrence of pseudofermion propagation and to short-range order in multiple-pseudofermion excitation. These effects are presently being investigated further.

In summary, we conclude that the pseudofermion approach seems to be useful both for systems with diagonal and off-diagonal disorder. In either case it gives rise to an expansion of the self-energy about the virtual-crystal approximation which is easily used to give analytical results and in which the symmetry in interchange of host and defect atoms is built in *a priori*. This is generally an important advantage since, particularly for systems with off-diagonal disorder, it seems to be rather difficult to preserve the high-concentration limit where the concentration of impurity atoms approaches one by expanding about the pure host crystal. (The exception are dilute systems where this may be advantageous in order to eliminate the unphysical degrees of freedom associated with the impurities.)

It is also very appealing that the same concepts which gave approximations for diagonally disordered systems, that is, the restriction of paths included to a certain pseudofermion subspace, seem to work also for models with off-diagonal disorder. For example, it is clear that one can define the analog of the non-self-consistent pair approximation explained in I by including all paths in which at any one time either a single or a pair of pseudofermions on nearest-neighbor sites is present. The corresponding equations can be likewise easily derived (as in I) by projecting onto the appropriate pseudofermion subspace. The difficult task which then remains to be done is, as in the one-pseudofermion approximation, to introduce self-consistency in an adequate way.

ACKNOWLEDGMENTS

We should like to thank Dr. T. Kaplan for providing the densities of states computed by the Schmidt method which were presented as histograms in Figs. 2 and 3. This work has been supported in part by the National Science Foundation through Grant No. DMR78-10813 which we gratefully acknowledge. One of us (H. W. D.) would also like to thank the German Academic Exchange Service (DAAD) for its support with a Nato Fellowship.

APPENDIX A: ANALYTICITY OF THE LPF GREEN'S FUNCTIONS

The similarities between the LPF theory and CPA theory strongly suggest that the LPF approximation also preserves the analytical structure of the self-energy. For the CPA analyticity was proven in various ways. Müller-Hartmann's[35] proof applies only for diagonally disordered systems. Ducastelle[36] gave a different proof and showed that Tsukada's molecular CPA[7] is also analytical. By similar reasoning one can also prove analyticity for Takeno's matrix CPA.[37]

In our case it is most convenient to proceed according to the procedure of Mills[30] who proved analyticity for this pair and cluster generalizations of the CPA. In particular, it is easy to see that the anti-Hermitian part of Σ

$$\mathrm{Im}\,\Sigma(z^2)=[\Sigma(z^2)-\Sigma(z^2)^\dagger]/2\pi i \tag{A1}$$

is positive definite for $\mathrm{Im}z^2>0$. This follows because the LPF approximation satisfies Mills' symmetry criterion[30] which guarantees that Σ is herglotz[11] but one can also easily prove that $\mathrm{Im}\,\Sigma(z^2)$ is positive definite directly from the LPF equations. Equation (24) implies

$$\mathrm{Im}\,Q^{\{i\}}=-J^{\{i\}}\,\mathrm{Im}\Big((G^{\mathrm{vc}})^{-1}-\sum_{l(\neq i)}Q^{\{l\}}-Y^{\{i\}}\Big)J^{\{i\}\dagger}, \tag{A2}$$

where

$$J^{\{i\}}=K^{\{i\}}\Big((G^{\mathrm{vc}})^{-1}-\sum_{l(\neq i)}Q^{\{l\}}-Y^{\{i\}}\Big)^{-1}.$$

But since $\mathrm{Im}\,(G^{\mathrm{vc}})^{-1}$ is positive definite for $\mathrm{Im}\,z^2>0$, then we find that (A2) gives $\mathrm{Im}\,Q^{\{i\}}<0$ if we assume that all the $Q^{\{l\}}$ on the right side are negative definite. Thus the iteration scheme in Eq. (24) preserves the sign of $\mathrm{Im}\,\Sigma$.

We now introduce a notation similar to that of Mills[30] and define column matrices (in the index i')

$$(\mu_i)_{i'}=\sum_{l(\neq i)}\delta_{li'}K^{\{l\}} \tag{A3}$$

and operator-valued matrices $\mathcal{U},\mathcal{W},\mathcal{G},\dot{\mathcal{G}},\mathcal{G}^{\mathrm{vc}}$

$$\mathcal{U}^{\{i\}\{i'\}}=Y^{\{i\}}\delta_{ii'}, \tag{A4a}$$

$$\mathcal{W}^{\{i\}\{i'\}}=\delta_{ii'}\sum_{l(\neq i)}Q^{\{l\}}=W^{\{i\}}\delta_{ii'}, \tag{A4b}$$

$$\mathcal{G}^{\{i\}\{i'\}}=\delta_{ii'}G^{\{i\}}, \tag{A4c}$$

$$\dot{\mathcal{G}}^{\{i\}\{i'\}}=\delta_{ii'}((G^{\mathrm{vc}})^{-1}-W^{\{i\}})^{-1}, \tag{A4d}$$

$$\mathcal{G}^{\mathrm{vc}}=\delta_{ii'}G^{\mathrm{vc}}. \tag{A4e}$$

Then the LPF equations can be written in the form

$$W^{\{i\}}=\mu_i^\dagger\mathcal{G}\mu_i \tag{A5}$$

$$\mathcal{G}^{-1}=\dot{\mathcal{G}}^{-1}-\mathcal{U}=(\mathcal{G}^{\mathrm{vc}})^{-1}-\mathcal{W}-\mathcal{U}. \tag{A6}$$

Equation (A5) and (A6) are analogous to Mills' equations and it is easy to see that one can prove existence, uniqueness, and analyticity of the LPF solutions by following his proof step-by-step from this point forward.

APPENDIX B: CALCULATION OF GREEN'S FUNCTIONS AND THE INTERACTOR

As explained in Sec. III we need to calculate Green's functions of the form

$$\bar{G}_{0n}=\frac{1}{2\pi}\int_{-\pi}^{\pi}e^{ink}(a+b\cos k+d\cos 2k)^{-1}\,dk \tag{B1}$$

where a,b,d are functions of z^2, and $a(z^2)=O(z^2)$ for $z^2\to\infty$. We now change variables by the substitution $\zeta=e^{ik}$ and $\bar{G}_{0n}$ as an integral in the complex ζ plane. This gives

$$\bar{G}_{0n}=\frac{1}{2\pi i}\oint\frac{\zeta^{n-1}d\zeta}{a+\frac{1}{2}b(\zeta+\zeta^{-1})+\frac{1}{2}d(\zeta^2+\zeta^{-2})}$$
$$=(2/d)\,\mathrm{Res}_{|\zeta|<1}\,[\zeta^{n+1}/P(\zeta)], \tag{B2}$$

where we integrate counterclockwise along the boundary of the unit circle $|\zeta|=1$, $\mathrm{Res}_{|\zeta|<1}$ denotes a sum over the residues of all poles with $|\zeta|<1$ (we assume that no poles lie on the boundary of the unit circle) and $P(\zeta)$ is defined by

$$P(\zeta)=\zeta^4+1+(b/d)(\zeta^3+\zeta)+2(a/d)\zeta^2. \tag{B3}$$

One easily verifies that the quantities $\zeta_i (i=1,2,3,4)$ defined in Eq. (37b) are the four roots of $P(\zeta)=0$. Then Eq. (37a) follows (for any nonnegative integer n).

Next, we will derive Eq. (42) for the interactor U in the BEB equations. U is determined by the requirement that the 2×2 i-i block of the matrix

$$\begin{array}{c} \\ i \\ i+1 \\ \\ \end{array}\begin{array}{c} \begin{array}{cccc} \cdots & i & i+1 & 0 \end{array} \\ \begin{bmatrix} \ddots & & & \\ & \gamma^{-1}+U & W & \\ & W & \gamma^{-1}+U & \\ 0 & & & \ddots \end{bmatrix} \end{array}^{-1} \tag{B4}$$

equals γ. We evaluate this block by using the continued-fraction method (as explained by Butler[38]). This gives

$$\gamma=(\gamma^{-1}+U-2WLW)^{-1}, \tag{B5}$$

where

$$L=(\gamma^{-1}+U-WLW)^{-1}. \tag{B6}$$

Equation (B5) then shows that

$$U=2WLW \tag{B7}$$

which together with Eq. (B6) yields the desired result in Eq. (42).

APPENDIX C: UPPER BOUND FOR MAXIMUM PHONON FREQUENCY

In this Appendix we shall prove that the maximum phonon frequency $\omega_{\max}$ in a one-dimensional, site-disordered, binary alloy with masses m_S and nearest-neighbor force constants $\phi^{SS'}$ $(S,S'=A,B)$ is bounded by

$$\omega_{\max}^2\leq 2\max_{S,S'}[\phi^{SS'}(m_S^{-1}+m_{S'}^{-1})]. \tag{C1}$$

We note that for $\phi^{AA}=\phi^{BB}=0\neq\phi^{AB}$ the bound yields exactly the maximum (optical) frequency $[2\phi^{AB}(m_A^{-1}+m_B^{-1})]^{1/2}$ for an ideal two-sublattice alloy with A particles in one and B particles in the other sublattice.

To prove (C1) we use that the density of states $\rho(\omega^2)$ for any alloy configuration can be written as

$$\rho(\omega^2)=-(1/\pi)\,\mathrm{Im}\{[(\omega^2+i\eta)1-M^{-1}\}_{00}\,, \tag{C2}$$

where $\eta>0$ is infinitesimal and M is a matrix

$$M_{ij}=(m_i m_j)^{-1/2}\left(\sum_{l(\neq i)}\phi_{il}\delta_{ij}-\phi_{ij}\right). \tag{C3}$$

In second-quantized phonon notation we rewrite M in the form

$$\begin{aligned}M&=\frac{1}{2}\sum_{i\neq j}\phi_{ij}\left(\frac{a_i^\dagger}{\sqrt{m_i}}-\frac{a_j^\dagger}{\sqrt{m_j}}\right)\left(\frac{a_i}{\sqrt{m_i}}-\frac{a_j}{\sqrt{m_j}}\right)\\&=\sum_{i\neq j}\phi_{ij}(m_i^{-1}+m_j^{-1})a_i^\dagger a_i\\&\quad-\frac{1}{2}\sum_{i\neq j}\phi_{ij}\left(\frac{a_i^\dagger}{\sqrt{m_j}}+\frac{a_j^\dagger}{\sqrt{m_i}}\right)\left(\frac{a_i}{\sqrt{m_j}}+\frac{a_j}{\sqrt{m_i}}\right).\end{aligned} \tag{C4}$$

Here, the second operator on the right-hand side is positive semidefinite. Consequently, the eigenvalues of M are bounded by the largest eigenvalue of the first operator. But

$$\sum_{j(\neq i)}\phi_{ij}(m_i^{-1}+m_j^{-1})\leq\max[2\phi^{SS'}(m_S^{-1}+m_{S'}^{-1})]$$

for any alloy configuration which proves (C1).

[1]R. J. Elliott, J. A. Krumhansl, and P. L. Leath, Rev. Mod. Phys. 46, 465 (1974).
[2]F. Yonezawa and K. Morigaki, Prog. Theor. Phys. 53, Suppl. No. 1 (1973).
[3]R. N. Aiyer, R. J. Elliott, J. A. Krumhansl, and P. L. Leath, Phys. Rev. 181, 1006 (1969).
[4]B. G. Nickel and J. A. Krumhansl, Phys. Rev. B 4, 4354 (1971).
[5]P. L. Leath, Phys. Rev. B 5, 1643 (1972).
[6]F. Ducastelle, J. Phys. F 2, 468 (1972).
[7]M. Tsukada, J. Phys. Soc. Jpn. 26, 684 (1969); 32, 1475 (1972).
[8]W. H. Butler, Phys. Lett. A 39, 203 (1972); Phys. Rev. B 8, 4499 (1973).
[9]J. Zittartz, Solid State Commun. 14, 51 (1974); Z. Phys. 267, 243 (1974).
[10]F. Brouers, F. Ducastelle, F. Gautier, and J. vander-Rest, J. Phys. F 3, 2120 (1973).
[11]A. Mookerjee, J. Phys. C 6, L205 (1973); 6, 1340 (1973).
[12]A. R. Bishop and A. Mookerjee, J. Phys. C 7, 2165 (1974).
[13]A. Mookerjee, J. Phys. C 8, 29 (1975).
[14]J. Mertsching, Phys. Status Solidi B 59, 227 (1973); 63, 241 (1974).
[15]H. Shiba, Prog. Theor. Phys. 46, 77 (1971).
[16]J. A. Blackman, D. M. Esterling, and N. F. Berk, Phys. Rev. B 4, 2412 (1971).
[17]K. Niizeki, Prog. Theor. Phys. 53, 74 (1975); 54, 1648 (1975).
[18]T. Kaplan and M. Mostoller, Phys. Rev. B 9, 1783 (1974).
[19]T. Kaplan and L. J. Gray, Phys. Rev. B 14, 3462 (1976).
[20]H. W. Diehl and P. L. Leath, preceding paper, Phys. Rev. B 19, 587 (1979).
[21]R. Bass and P. L. Leath, Phys. Rev. B 9, 2769 (1974).
[22]S. Takeno, Prog. Theor. Phys. 40, 942 (1968).
[23]A. B. Harris, P. L. Leath, B. G. Nickel, and R. L. Elliott, J. Phys. C 7, 1693 (1974).
[24]W. K. Holcomb, J. Phys. C 7, 4299 (1974).
[25]H. W. Diehl and W. Biem, Z. Phys. B 27, 7 (1977).
[26]H. W. Diehl and P. L. Leath (unpublished).
[27]J. A. Blackman, J. Phys. F 3, L31 (1973).
[28]M. Watabe and F. Yonezawa, Phys. Rev. B 11, 4753 (1975).
[29]H. Schmidt, Phys. Rev. 105, 425 (1957).
[30]R. L. Mills (unpublished). [Mills' work has been summarized and expanded in a recent paper by R. Mills and P. Rabanavararahsa, Phys. Rev. B 18, 5291 (1978); a preliminary version of which we recently received.]
[31]D. J. Thouless, J. Phys. C 3, 1559 (1970).
[32]F. Ducastelle, J. Phys. C 8, 3297 (1975).
[33]I. M. Lifshitz, Adv. Phys. 13, 483 (1964).
[34]R. W. Davies and J. S. Langer, Phys. Rev. 131, 163 (1963).
[35]E. Müller-Hartmann, Solid State Commun. 12, 1269 (1973).
[36]F. Ducastelle, J. Phys. C 7, 1795 (1974).
[37]H. W. Diehl (unpublished).
[38]W H. Butler, Phys. Rev. B 8, 4499 (1973).