Analytic approximation for substitutional alloys

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It is shown that the terms in the cumulant expansion of the coherent potential for an uncorrelated substitutional alloy can be combined by partial summation in such a way that the Herglotz property can be studied directly. A consistency condition for the Herglotz property is found, which is satisfied by the single-site coherent-potential approximation (CPA) but not by the n-site CPA for $n \geq 2$. A natural generalization of the CPA [referred to as the "traveling-cluster approximation" (TCA)], satisfying the consistency condition, is developed in which graphs involving arbitrarily many sites are involved, but in such a way that overlaps of cumulant averages involve only limited sets of sites. A fixed-point theorem is developed that guarantees that iteration of the TCA equations for a broad range of physical systems converges to a unique self-consistent solution that preserves the Herglotz property of the mean resolvent. Calculations of the density of states using the nearest-neighbor TCA for a single-band tight-binding model are presented, and show a distinctly better fit to exact numerical results than the CPA, including some of the structure due to localized states.

I. INTRODUCTION

One of the standard methods for trying to understand the physical properties of waves in a random medium has been to study the mean resolvent [Eq. (1.4), below] for the system. For the case of electronic states and elastic modes in a random crystal,[1] the coherent-potential approximation (CPA) of Soven[2] and Taylor[3] has stood out[4] as the most effective single-site approximation and as the base for a large number of efforts to develop extensions involving clusters.

The problem of analyticity of the mean resolvent for a substitutional binary alloy has been raised by Nickel and Butler,[5] who found by direct computation that two well-known extensions[6,7] of the CPA fail to satisfy this basic condition. We present here a form for the perturbation expansion of the coherent potential, for the case of statistically independent scattering sites, that makes the study of analyticity properties quite straightforward and allows us to develop an approximation that correctly preserves these properties. We first review the problem of finding an effective, or "coherent" potential W, expressed as a sum of products of cumulant averages, with each term of the sum corresponding in the usual way to a Feynman-like graph. In Sec. II we show for a binary alloy that a partial summation of graphs, expressed in what we call "modified cumulant averages," reduces the mathematical expression for each graph to a much simpler form which permits (Sec. III) a term-by-term analysis of the set of properties, known collectively as the Herglotz property (HP), required for the coherent potential. The logic of this analysis bears some similarity to that of Schwartz and Bansil,[8] who, however, are tackling the problem within the framework of multiple-scattering theory rather than a partial summation of graphs, and come to conclusions that are not related in an obvious way to those obtained here. Certain sums of graphs satisfying a consistency condition are found to preserve the HP, so that it becomes possible to formulate (in Sec. IV) a class of approximations, the "traveling-cluster approximations," that preserve analyticity, at least if self-consistency is not imposed. When self-consistency is introduced (Sec. V), by the use of an appropriately modified mean resolvent for internal lines in each graph, then the resulting nonlinear equations no longer obviously preserve the HP, but a modified fixed-point theorem (proved in Appendix B and discussed in Sec. VI) shows that the HP is indeed preserved by the self-consistent equations and that iteration of the equations at complex energy always converges to a unique self-consistent solution of these equations. The formalism is applied in Sec. VII to the case of a separable potential, which includes as special cases the single-band model and the elastic-mode problem for a simple Bravais lattice with random masses. Finally, the results of some calculations of the nearest-neighbor TCA for electronic states in the single-band tight-binding approximation are shown and compared with the single-site CPA and with the exact numerical results of Alben *et al.*[9] The paper concludes with a brief discussion in Sec. VIII. The more mathematical parts of the presentation appear in the single-band formulation of Sec. VII and in Appendixes A and B.

In Appendix A, we set up a general formulation of the problem for an arbitrary number of species,[10] which is nontrivially different in form from the binary case and seems to provide a possible starting place for analysis of the problem of cor-

related systems, to which this approach has not yet been successfully applied.

In Appendix B, the fixed-point theorem is proved, and a complete demonstration of convergence and the HP is given.

We shall concentrate our attention in this paper on the behavior of an electron in a binary substitional alloy of N sites, with a single-particle Hamiltonian given by

$$H = H_0 + V, \tag{1.1}$$

$$V = \sum_x V_x . \tag{1.2}$$

Here H_0 is fixed and typically has the complete crystalline symmetry of the Bravais lattice, whose sites we label x, while V is random, the single-site terms V_x being statistically uncorrelated. Each V_x is thus a random operator, localized near the site x, and equal to V_x^A, with probability c_A, or to V_x^B, with probability $c_B = 1 - c_A$. No assumption is made here as to whether the potentials overlap. In general, H_0 and V are linear operators on a Hilbert space $\mathcal{H}$, and in a single-band model (see Sec. VII) they reduce to matrices with rows and columns labeled by the sites x. We define the "unperturbed resolvent" G_0 as the inverse of an operator L_0,

$$G_0 = (E - H_0)^{-1} = L_0^{-1}, \tag{1.3}$$

for a complex energy variable E, while the mean resolvent G and the coherent potential W are defined by

$$G = \langle (L_0 - V)^{-1} \rangle \tag{1.4}$$

$$= (L_0 - W)^{-1}, \tag{1.5}$$

where $\langle\ \rangle$ denotes an average over all configurations of the sample. For a more general class of systems, the necessary conditions on L_0 and V for the applicability of the fixed-point theorem are spelled out in Appendix B.

The resolvent defined by Eq. (1.4) and the coherent potential defined by Eq. (1.5) are well known to possess the Herglotz property, which, for an operator function $F(z)$ of a complex variable z, consists of the following three properties:

$F(z)$ is an operator-valued analytic function of z in the entire cut plane, $\mathrm{Im}z \neq 0$;

$$F(z^*) = \overline{F}(z) \quad \text{for } \mathrm{Im}z \neq 0; \tag{1.6}$$

$$\mathrm{Im}F(z) \leq 0 \quad \text{for } \mathrm{Im}z > 0 (z \in \mathcal{G}_+). \tag{1.7}$$

Here $\overline{F}$ is the Hermitian adjoint of F, and $\mathrm{Im}F$ refers to the anti-Hermitian part of F,

$$\mathrm{Im}F = (F - \overline{F})/2i. \tag{1.8}$$

In the usual definition of the Herglotz property $\mathrm{Im}F$ is negative definite in the upper half plane $\mathcal{G}_+$ rather than negative semidefinite as we have taken it. The weaker inequality is sufficient for our purpose and conveniently includes as a special case the situation in which $F(z)$ is a constant Hermitian operator. These properties are roughly equivalent to the assertion that $F(z)$ is derivable from a positive semidefinite Hermitian spectral function $f(\omega)$,

$$F(z) = a - bz + \int_{-\infty}^{\infty} \frac{f(\omega)}{z - \omega}\, d\omega , \tag{1.9}$$

where a and b are Hermitian, and b is non-negative. In particular, the spectral function for $G(E)$ is related to the density of states (per site) for the physical system,

$$n(E) = -(1/\pi N)\,\mathrm{Tr}[\mathrm{Im}G(E + i0+)] \quad (E \in \mathcal{R}), \tag{1.10}$$

so that a failure of the Herglotz property in some approximation gives rise to unphysical behavior of $n(E)$. In the examples studied by Nickel and Butler,[5] branch points were found in the complex plane, giving rise to multiple valuedness of $n(E)$.

II. MODIFIED CUMULANT EXPANSION

As a starting point for the analysis of the problem, we use the standard cumulant expansion method of Kubo,[11] Yonezawa and Matsubara,[12] Leath and Goodman,[13] and others and then show that a partial summation of terms yields an expansion formalism that is particularly suited to this problem. Following the discussion and notation of Mills,[14] we express the coherent potential W as a sum of combined cumulant averages (CA's):

$$W = \sum_{n=0}^{\infty} \langle V(G_0 V)^n \rangle^P . \tag{2.1}$$

Here $\langle\ \rangle^P$ indicates a sum of proper graphs, in the sense (Ref. 14) that all points on a given graph are interconnected by overlapping CA's. The CA of a product of n factors, which can be noncommuting operators, is defined as the nth-order residue when all combinations of CA's of lower order are subtracted from the actual configuration average of the product. Thus, using $\langle\ \rangle$ to denote a simple configuration average and $\langle\ \rangle_c$, a cumulant average, we have

$$\langle A \rangle = \langle A \rangle_c , \tag{2.2}$$

$$\langle AB \rangle = \langle A \rangle_c \langle B \rangle_c + \langle AB \rangle_c , \tag{2.3}$$

$$\langle ABC \rangle = \langle A \rangle_c \langle B \rangle_c \langle C \rangle_c + \langle AB \rangle_c \langle C \rangle_c + \langle A \langle B \rangle_c C \rangle_c + \langle A \rangle_c \langle BC \rangle_c + \langle ABC \rangle_c , \tag{2.4}$$

etc. If any one factor or group of factors is uncorrelated with the remaining factors, the CA of the product is equal to zero. The expression for the

CA is rather complicated, which makes a term-by-term analysis of the HP in W virtually impossible using the above expansion. We find, however, that there is a natural way of performing partial summations, at least in the absence of correlations, that makes such analysis quite feasible. The procedure which is similar, but not identical, to the "corrected cumulant average" procedure of Yonezawa,[15] Leath,[16] etc. is to group the graphs according to the overlap pattern of CA's involving the same site and then to sum all the terms in each group.

Consider, for example, the graph in Fig. 1(a), in which x, y, and z represent three different sites, and the CA's involving site x are emphasized. If the CA's involving sites y and z are disregarded, we see that those involving x form two "proper single-site subgraphs," as we may call them. That is, the first three x-site CA's overlap, forming one proper subgraph if all the other CA's are removed, and the last two x-site CA's form another. We characterize the graph of Fig. 1(a) by a sort of skeleton graph, in which those vertices that are linked by CA's into a proper single-site subgraph are joined in a single linkage, as in Fig. 1(b). We introduce a new graphical symbol for this purpose, which we refer to as a "modified cumulant average (MCA) linkage," and call the graph of Fig. 1(b) the "characteristic graph" of Fig. 1(a).

Now many CA graphs will have the same characteristic graph, and we now sum all such graphs to obtain a single expression corresponding to a given characteristic graph. Since the statistical factors associated with each CA, and hence with each MCA, are independent of the structure of the rest of the graph, we obtain in this way an expression, which we call a "modified cumulant average", for each MCA linkage. Out of these MCA's then, we can construct the contribution of the whole characteristic graph, just as one does with ordinary CA's.

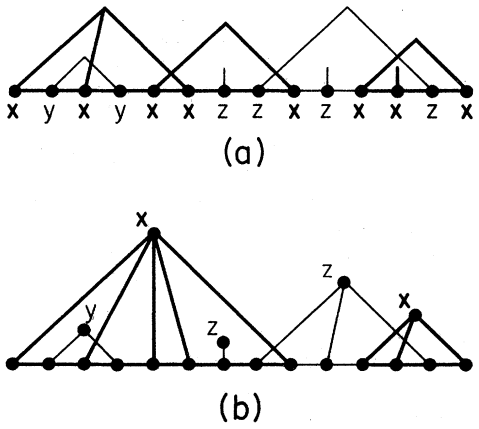

FIG. 1. Cumulant averages (CA) and modified cumulant averages (MCA). (a) Graphical expression in terms of CA's, with site x emphasized, for a typical term in the coherent potential W. (b) The corresponding MCA graph, in which overlapping CA's are summed to form MCA's.

In order to evaluate the nth-order MCA, we need to sum all the proper single-site subgraphs involving the same n vertices. To do this sum explicitly is difficult, since the expression for the CA itself is complicated, and the structure of the different graphs is hard to tabulate. There is, however, an easy way to get the answer without doing the work, as follows. As mentioned above, the statistical factors involved in a given proper single-site graph are independent of the rest of the graph, and in particular will be just the same if no other sites appear in the graph at all. In this case, the problem reduces to summing *all* proper graphs of nth order that involve just the one site in question. However, this is just the nth-order term in the coherent potential for a system consisting of just that one site, and this can be evaluated by using the original definition, Eq. (1.5), of the coherent potential. For the one-site problem,

$$G^{(1)} = (L_0 - W^{(1)})^{-1} = c_A(L_0 - V^A)^{-1} + c_B(L_0 - V^B)^{-1}, \tag{2.5}$$

from which it works out that

$$W^{(1)} = \overline{V} + V'(L_0 - \tilde{V})^{-1}V', \tag{2.6}$$

where

$$\overline{V} = c_A V^A + c_B V^B, \tag{2.7}$$

$$\tilde{V} = c_B V^A + c_A V^B, \tag{2.8}$$

$$V' = \sqrt{c_A c_B}(V^A - V^B). \tag{2.9}$$

(In $\overline{V}$, the bar refers to an average rather than the Hermitian adjoint.)

In the expansion for $W^{(1)}$, the first-order term is just $\overline{V}$, while for $n \geq 2$ the nth-order term is $V'(G_0\tilde{V})^{n-2}G_0V'$, a simple sequence of factors whose structure is independent of n. The statistical fators characteristic of the nth-order MCA are completely represented by the combinations $\overline{V}$, $\tilde{V}$, and V'.

The connecting factors G_0 are simply those connecting the factors V_x before averaging and may be replaced in the n-site problem by the appropriate factors involving other sites, but not involving site x. The MCA's involving other sites y, of course, take exactly the same form. As an example, the contribution to W of Fig. 1(b) is simply

$$W[1(b)] = V'_xG_0V'_yG_0\tilde{V}_xG_0V'_yG_0\tilde{V}_xG_0\tilde{V}_xG_0\overline{V}_zG_0 \times V'_zG_0V'_xG_0\tilde{V}_zG_0V'_xG_0\tilde{V}_xG_0V'_zG_0V'_x. \tag{2.10}$$

We can summarize the resulting MCA expansion with the symbolic equation

$$W = \langle V(1 - G_0 V)^{-1} \rangle_{\mathrm{MCA}} , \tag{2.11}$$

with the rule that all proper MCA graphs are to be included in which no two MCA's involving the same site are allowed to overlap. It will be noted that only the bare propagator G_0 appears; it seems impossible to do partial summations, as in the CA expansion, in such a way that G appears instead of G_0 and a restricted class of graphs is summed. It turns out, nevertheless, that when the sum over MCA graphs is restricted in order to construct an approximate theory, it then becomes possible to replace each factor G_0 by an appropriate self-consistent propagator, so that many of the graphs omitted in the approximation are restored and a sort of effective medium is used for each successive virtual propagation.

In order to make clear the overlap patterns involved in more complicated graphs, it is convenient for the analysis that follows to introduce a more streamlined graphical representation for the MCA expansion. Since the electron line carries no information, we eliminate it and replace the MCA linkage by a horizontal line, labeled by the site involved, with dots along the line to represent the successive interactions at that site, as in Fig. 2(a). This linear representation is appropriate to the simple product character of an MCA, with the dots representing the successive factors V_x' (at either end of an MCA line) and $\tilde{V}_x$ (not at an end). An isolated dot represents the first-order MCA, i.e., a factor $\overline{V}_x$. Several horizontal lines or single dots, as in Fig. 2(b) [corresponding exactly to Fig. 1(b) and thus to Eq. (2.10)], represent overlapping MCA's, with the order of factors being just the same as the order of the dots from left to right. The vertical location of an MCA line or dot can be used to suggest the location in the sample of the site involved, with neighboring lines representing neighboring sites.

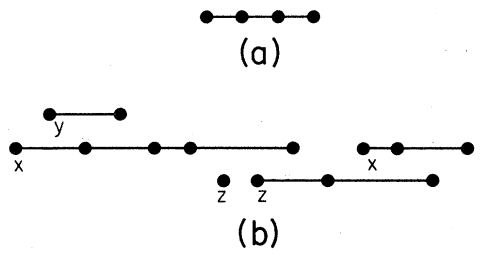

FIG. 2. Linear notation for MCA graphs. (a) Single MCA involving site x. (b) Graph of Fig. 1 (b) in the linear notation.

III. ANALYSIS OF Im W

We have seen that each term in the MCA expansion for $W(E)$ is directly expressible [as in Eq. (2.10)] as a product of factors $\overline{V}_x$, V_x', $\tilde{V}_x$, and G_0. The factors V are all Hermitian, and the dependence on the complex energy E is entirely in the factors $G_0(E)$. We now take the anti-Hermitian part of $W(E)$, following a procedure somewhat analogous to that of Schwartz and Bansil,[8] in order to find approximations that preserve the second Herglotz property [Eq. (1.6)]. We schematically represent an nth-order term in W, corresponding to a graph, γ, by

$$W(\gamma) = V_1 G_0 V_2 G_0 \cdots G_0 V_n , \tag{3.1}$$

where $V_1, V_2, \ldots, V_n$ represent the appropriate factors $\overline{V}$, V , $\tilde{V}$. The anti-Hermitian part of $W(\gamma)$ is

$$\mathrm{Im}W(\gamma) = (V_1 G_0 V_2 G_0 \cdots G_0 V_n - V_n \overline{G}_0 \cdots \overline{G}_0 V_2 \overline{G}_0 V_1)/2i . \tag{3.2}$$

The mirror image of the graph γ, which we call $\tilde{\gamma}$, gives rise to the same factors in reverse order, and as we shall consider only approximations in which mirror image graphs are always included, we can make a trade between $\mathrm{Im}W(\gamma)$ and $\mathrm{Im}W(\tilde{\gamma})$ in order to have the same sequence of factors in the two terms on the right-hand side of Eq. (3.2)

$$-[\mathrm{Im}W(\gamma) + \mathrm{Im}W(\tilde{\gamma})] = w(\tilde{\gamma}) + w(\gamma) , \tag{3.3}$$

$$w(\gamma) = (V_1 \overline{G}_0 V_2 G_0 \cdots G_0 V_n - V_1 G_0 V_2 G_0 \cdots G_0 V_n)/2i \tag{3.4}$$

$$= \sum_{r=1}^{n-1} (V_1 \overline{G}_0 V_2 \cdots V_r \overline{G}_0) \times Y(G_0 V_{r+1} \cdots V_{n-1} G_0 V_n) , \tag{3.5}$$

where

$$Y = \mathrm{Im}(G_0^{-1}) \tag{3.6}$$

$$= \mathrm{Im}E , \tag{3.7}$$

in the case we are considering.

The bracketed portions of Eq. (3.5) can be thought of, thanks to the factorizability of the MCA expansion, as "half graphs," obtained by cutting the original graph for $W(\gamma)$ between each pair of successive vertices. Thus, the simple W graph shown in Fig. 3(a) gives rise to the three cut graphs of Fig. 3(b). Each half graph can be characterized by the set S of sites associated with those MCA's that are divided by the cut. In Fig. 3(b), the three cut graphs are characterized by the sets $\{x\}$, $\{x, y\}$, and $\{y\}$, respectively.

In the course of taking the anti-Hermitian part of W, we see that all possible right-half graphs and all possible left-half graphs are generated and

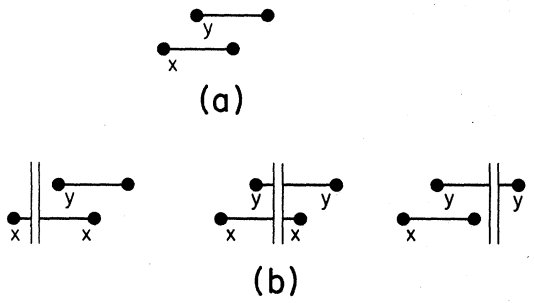

FIG. 3. Analysis of Im W. Fig. 3(b) shows the different ways the graph of Fig. 3(a) must be cut in calculating the anti-Hermitian part.

combined in all possible ways. If the sum of all right-half graphs characterized by the set S, as described above, is denoted by Γ_S, then the corresponding set of left-half graphs is $\bar{\Gamma}_S$, and we see that

$$\mathrm{Im}W = -\sum_S \bar{\Gamma}_S Y \Gamma_S , \tag{3.8}$$

where the negative definite character of ImW (ImE >0 will be assumed throughout) is plainly revealed.

We now have a way of constructing possible approximation schemes and testing at least for the second Herglotz property [Eq. (1.6)]. If we take as an approximation to W a subset of all the MCA diagrams, we can perform the same sort of analysis as above and see whether the cut graphs of ImW combine to give a negative definite form like Eq. (3.8). The second Herglotz property does not, of course, guarantee analyticity, but it is a necessary condition, and with luck the analyticity property will fall out, too. The third Herglotz property is automatic, since W is in any case a real functional of G_0, which has this property.

It is not hard to see that any graph or set of graphs included in W will tend to generate more graphs that also must be included if a negative definite form, like Eq. (3.8), is to be retained. In forming ImW, each way of cutting a given W graph gives a right- and a left-half graph. Then for each such term of the form $\bar{\Gamma}_1 Y \Gamma_2$, additional combinations must be included to complete the square, so to speak, into the form $(\bar{\Gamma}_1 + \bar{\Gamma}_2) Y (\Gamma_1 + \Gamma_2)$. These additional combinations, in this case $\bar{\Gamma}_1 Y \Gamma_1$, $\bar{\Gamma}_2 Y \Gamma_1$, and $\bar{\Gamma}_2 Y \Gamma_2$, must come from new W graphs, and the process must be repeated until it closes. Thus the W graph of Fig. 3(a) generates the cut graphs of Fig. 3(b), and in order to achieve the negative definite form, the nine additional cut graphs shown in Fig. 4(a) must be included, together with those of Fig. 3(b), giving rise to three negative definite expressions of similar form to Eq. (3.8). However, in order that these nine additional

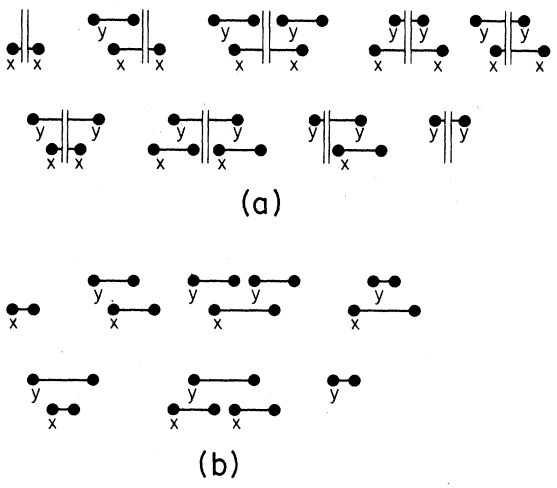

FIG. 4. Graphs generated by the consistency requirement. (a) Cut graphs that must be added to those of Fig. 3(b) to assure Im $W \leq 0$. (b) Graphs that must be added to Fig. 3(a) in order to obtain the cut graphs of Fig. 4(a).

cut graphs form part of ImW, it is necessary that W itself include additional terms of the form shown in Fig. 4(b), which in turn will generate a second generation of cut graphs. Except in the case of the first- and second-order graphs, this process generates an infinite number of W graphs which must be included to preserve the negative definite form for ImW. The associated sets S, though, are just those of the first generation of cut graphs; the process does not generate new ways for the MCA lines to overlap. Thus a consistent approximation can be achieved, typically, by including all graphs involving some limited number of overlap sets.

IV. NON-SELF-CONSISTENT TRAVELING CLUSTER

Now let us consider some of the approximations that can be generated in this way, but still without regard for self-consistency, i.e., without the use of a self-consistent propagator in place of G_0. The simplest such approximation is achieved by using for W the single term $\bar{V}_x$ for some chosen site x, represented graphically by a single dot [Fig. 5(a)]. This is, of course, Hermitian and independent of E, and trivially preserves the Herglotz property of G. This is not a translation-invariant approximation, and as we shall normally want our approximations to preserve the symmetries of the underlying Bravais lattice, we can extend the approximation by including all such terms:

$$W \approx \sum_x \bar{V}_x , \tag{4.1}$$

which is simply the virtual-crystal approximation[17]

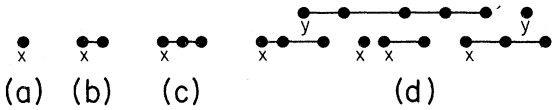

FIG. 5. Approximations to the coherent potential W. (a) First order: the virtual-crystal approximation. (b) Second order: satisfies the consistency requirement. (c) Third order: consistency requires the inclusion of all higher-order single-site terms and yields the average t-matrix approximation. (d) Typical graph involving just two sites.

in its simplest form.

The next stage is to include the second-order terms in W, represented by graphs of the form of Fig. 5(b). If we include also the first-order terms of Eq. (4.1) (reasonable, but not necessary for the Herglotz property), we have

$$W \approx \sum_x (\bar{V}_x + V_x' G_0 V_x') . \tag{4.2}$$

Thus the perturbative expansion of W preserves the Herglotz property up to second order.

If we go to third-order single-site terms, as in Fig. 5(c), we find we must include the single-site graphs of all orders. This gives a geometric series for each site, which sums to

$$W \approx \sum_x [\bar{V}_x + V_x'(L_0 - \tilde{V}_x)^{-1} V_x'] , \tag{4.3}$$

just as in Eq. (2.6), and corresponds exactly to the average-t-matrix approximation (ATA) of Korringa[18] and Beeby.[19]

We next turn to two-site terms, such as those generated from Fig. 3(a). These include all graphs involving as factors only the two expressions V_x' and V_y', a somewhat unnatural generalization of the one-site approximation represented by Eq. (4.2) (unsummed and without the first-order terms $\bar{V}_x$, however). We can immediately generalize this by simply including *all* graphs involving two given sites x and y, of which a typical term is displayed in Fig. 5(d). This clearly obeys the negative definiteness condition, since the process of cutting yields all possible left- and right-half graphs involving these two sites. This gives, in fact, the exact expression for W for a system consisting only of two sites, just as Eq. (4.3) is exact for the one-site system.

A difficulty arises if we now try to take a sum of two-site terms after the manner of the two-site CPA. If a term involving x and y and a term involving y and z are both included, we immediately find that three-site terms are also generated. This is illustrated in Fig. 6, where the W graphs shown in Fig. 6(a) generate, among others, the half graphs in Fig. 6(b), which in turn generate, among others, the new W graphs in Fig. 6(c).

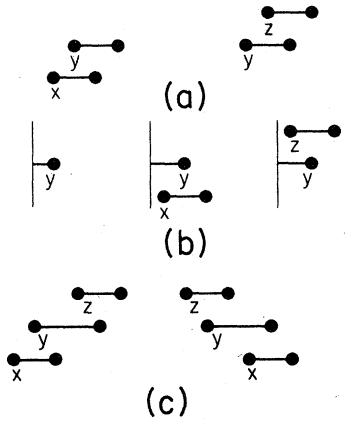

FIG. 6. Consistency in a two-site approximation. If both graphs of (a) are included, the half-graphs of (b) (among others) are generated, requiring for consistency the three-site term (c).

Thus we see a basic source of difficulty with the n-site CPA, which in the nearest-neighbor version, includes just such graphs as Fig. 6(a), without including the corresponding terms in Fig. 6(c). The same thing happens in every case except $n=1$, consistent with the observed fact that the single-site CPA displays the Herglotz property. The above examples do not constitute a proof that the second Herglotz property is violated since negative definite parts of ImW may still dominate if the parameters of the problem fall in the right domain, but indicate clearly the nature of the difficulty that is in fact observed. It should also be noted that we can avoid the difficulty by using only pairs without common members, that is, by including in W all terms involving sites one and two, all terms involving sites three and four, and so on. This corresponds exactly (when self-consistency is included) to the cluster CPA, considered by Tsukada,[20] Ducastelle,[21] and others, which is in logical structure equivalent to a single-site CPA on a sublattice of the original sample.

Let us go back now to the additional terms generated when overlapping pairs of sites are included. If the original W graphs involve only nearest-neighbor (nn) pairs, for example, then although the new graphs generated can involve many sites, the overlaps of successive MCA's involve only nn pairs, as in Fig. 6(c) [here, (x, y) and (y, z) are taken to be nn pairs]. As mentioned previously, no new overlap sets are generated by this process, where an "overlap set" is the set of sites whose MCA lines are divided when a W graph is cut. [We include also in the definition of "overlap set" the set that results from adding a single-site MCA $(\bar{V}_x)$.] This suggests an approximation to W con-

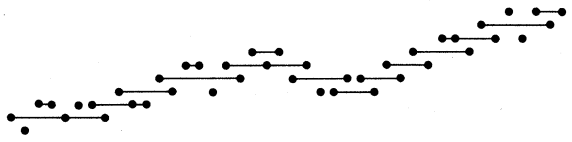

FIG. 7. Typical graph, in high order, of the nearest-neighbor traveling cluster approximation.

sisting of all graphs whose *overlap sets* are restricted to nn pairs, as exemplified, in a high order, by Fig. 7. The character of the MCA as a simple product of factors causes such an approximation to take the form of a geometric series, summable by straightforward algebra, as we shall see. While these graphs by no means depict literally the virtual processes actually taken into account, and the direct physical interpretation is rather lost in the formalism, it is still true that an MCA line roughly represents a memory of the identity of a scatterer as the electron progresses through the sample. In this approximation then, the spotlight of exact treatment, instead of remaining fixed on one cluster of sites as in the n-site CPA or the cluster CPA, is allowed to move across the sample, even to macroscopic distances. This approximation, which we therefore refer to as a "traveling cluster approximation" (TCA), is seen to be the completion of the n-site ATA, in the sense that we include all graphs generated from the n-site ATA by the requirement of negative definiteness of $\mathrm{Im}W$.

A particular TCA, referred to as the T approximation, is defined by a family T of allowed overlap sets S, the approximation consisting of the sum of all graphs consistent with this restriction. In the above example, T includes all single sites and all nn pairs. We would normally want T to have the symmetry of the lattice, though as we have seen this is not necessary for the negative definiteness condition. The TCA's are not the only approximations that can be constructed to satisfy this condition, but they seem the only natural ones. Since we construct a graph, moving from left to right, by successively adding or taking away single sites, we see that the family T, to be related to real graphs, must consist of sets that can be built up one site at a time from single-site sets without going outside the family T. A natural way to do this is to require that all the subsets of any set in T also belong to T, but this is not essential. For example, in a linear chain, T could include nn pairs and clusters of three, without including next-nearest-neighbor pairs. The calculations are sufficiently complicated that such options could prove useful.

The equations expressing the TCA can conveniently be written by expressing W as a sum of terms characterized by the initial and final scattering site:

$$W=\sum_{x,y}W^{xy}, \tag{4.4}$$

where

$$W^{xy}=\overline{V}_x\delta_{xy}+V_x'\Gamma^{xy}V_y' \tag{4.5}$$

and Γ^{xy} is one part of a generalized matrix propagator $\Gamma^{SS'}$, representing all subgraphs that start on the left with a factor G_0 and the overlap set S and that end on the right with a factor G_0 and the overlap set S'. The superscripts x, y in this case represent the single-site sets $\{x\}$ and $\{y\}$. This matrix propagator satisfies a Dyson-type equation,

$$\Gamma^{SS'}=G_0\left(\delta_{SS'}+\sum_{S''\in T}\Phi^{SS''}\Gamma^{S''S'}\right)\quad (S,S'\in T), \tag{4.6}$$

$$\Phi^{SS'}=\delta_{SS'}\left(\sum_{x\in S}\tilde{V}_x+\sum_{x\in\tilde{S}}\overline{V}_x\right)+\sum_{\substack{x\in S\\ x\cup S=S'}}V_x'+\sum_{\substack{x\in\tilde{S}'\\ x\cup S'=S}}V_x', \tag{4.7}$$

where the T approximation expresses itself in a restriction of sets S to the family T. The set $\tilde{S}$ consists of those sites not in S which do not take S outside the family T:

$$\tilde{S}=\{x\,|\,x\notin S, x\cup S\in T\}\ . \tag{4.8}$$

Combined with S, it constitutes what we may call the "neighborhood" S^+ of S:

$$S^+=S\cup\tilde{S}=\{x\,|\,x\cup S\in T\}\ . \tag{4.9}$$

What we have developed in this section is a generalization, or completion, of the n-site average ATA, satisfying the condition, at least in terms of the graphs included, that $\mathrm{Im}W$ be negative definite when $\mathrm{Im}E>0$. In fact, the entire Herglotz property is readily demonstrated since Eq. (4.6) allows $\Gamma^{SS'}$ to be expressed as the inverse of a simple operator. The vectors whose components are labeled by the sets S belonging to the family T constitute a vector space $\mathcal{S}_T$, and the operator-valued matrices $\Gamma^{SS'}$ and $\Phi^{SS'}$ can be regarded as linear operators Γ and Φ on the product space $\mathcal{S}_T\otimes\mathcal{H}$, where $\mathcal{H}$ is the Hilbert space of the original problem. Clearly Φ is Hermitian in $\mathcal{S}_T\otimes\mathcal{H}$, and Γ can be written

$$\Gamma=(\Gamma_0^{-1}-\Phi)^{-1}, \tag{4.10}$$

where

$$\Gamma_0^{SS'}=G_0\delta_{SS'}\ . \tag{4.11}$$

Since Γ_0 is Herglotz, it follows that Γ is also, and from Eqs. (4.5) and (4.4), the Herglotz property of W follows directly.

V. SELF-CONSISTENCY

We now turn to the question of self-consistency, i.e., of replacing G_0 in the equations developed above by a propagator that best represents the environment in which the virtual processes take place that are described by these equations. In graphical terms, this means including in this propagator as many as possible of the graphs not already included in the summation of overlapping MCA's within the approximation defined by the family T. In the conventional cumulant expansion (see Ref. 14) this self-consistency appears almost automatically through the summing of internal self-energy subgraphs and an associated reduction to irreducible self-energy graphs involving the true mean resolvent G. In the MCA expansion this does not happen, because certain internal self-energy subgraphs were needed in the formation of the MCA's, and the distinction between reducible and irreducible graphs cannot be made in a natural way in this case.

Nevertheless, there is a reasonably natural way of introducing self-consistency, by replacing each factor G_0 in Eq. (4.6) by a propagator G^S whose form depends on the overlap set S at the location of this propagator in the graph. The self-consistent propagator G^S is defined by

$$G^S = (L_0 - W^S)^{-1}, \tag{5.1}$$

where W^S is the sum of all W graphs that begin and end outside the family T. That is,

$$W^S = \sum_{x,y\in\hat{S}} W^{xy}, \tag{5.2}$$

where $\hat{S}$ is the set of sites for which $x\cup S$ lies outside of T:

$$\hat{S} = \{x \mid x\cup S \in T\}\ . \tag{5.3}$$

It is the complement of S^+ [Eq. (4.9)]. This definition of W^S is taken, of course, to avoid overcounting of graphs. There might seem to be a difficulty from the fact that internal portions of W^S are allowed to involve MCA's for sites belonging to the set S, as illustrated in Fig. 8(a), for a nn approximation. We see that two site-y MCA's overlap, in violation of the rules for the MCA expansion of Sec. II. However, the graph with which this would be redundant, shown in Fig. 8(b), is not included in the approximation, and so no overcounting has taken place. The original derivation of the MCA by summation of overlapping cumulant averages involving a single site y, in fact, remains valid even if iterations at site y are interspersed, so long as they are not involved in any of the CA's being summed. Not all of the CA graphs represented by Fig. 8(b) are included in the self-consistent nn approximation, the ones represented by Fig. 8(a) being those in which the two intermediate site-y interactions are not coupled by CA's to the initial and final ones. This reasoning applies quite generally to the graphs generated by use of the self-consistent G^S of Eqs. (5.1) and (5.2).

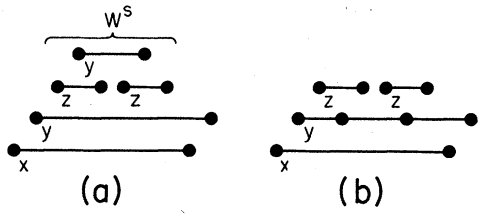

FIG. 8. Illustrating that use of the self-consistent propagation G^S does not produce overcounting. (a) Possible term in W^S, for $S=\{x,y\}$, in the nn approximation, showing overlap of MCA's involving site y. (b) MCA graph with which (a) would be redundant, but which is not included in the nn approximation.

We must now see what this modification of the theory does to the analysis of ImW of Sec. III. Our new W graphs differ from the one-self-consistent ones by having factors G^S instead of G_0 between successive interaction vertices. As we review the procedure of Sec. III for analyzing ImW, we find that all the steps go through trivially as before, except that ImG_0^{-1} [Eq. (3.6)] is replaced by Im$(G^S)^{-1}$ when the cut is associated with the overlap set S. From Eqs. (5.1) and (5.2), we see that

$$\mathrm{Im}(G^S)^{-1} = -\mathrm{Im}(L_0 - W^S) \tag{5.4}$$

$$= -(\mathrm{Im}E - \mathrm{Im}W^S)\,. \tag{5.5}$$

Thus if ImW^S is negative definite, then Im$(G^S)^{-1}$, and hence ImW, are also. However, ImW^S is subject to the same form of analysis as ImW itself, and its negative definiteness for all S is thus seen at least to be self-consistent; after any finite number of iterations (starting with W^S Herglotz for each S), the negative definite property is retained.

It is tempting to try to extend W^S to include graphs that might begin or end (or both) inside the family T [i.e., at a site x for which $x\cup S\in T$], but which go outside of T in internal MCA lines. The difficulty here is that the analysis of ImW^S gives rise to half graphs lying entirely in T, which require for consistency duplicate graphs to those already included explicitly (i.e., apart from the introduction of G^S). Thus the negative definite property is lost in such a scheme unless some graphs are overcounted.

The self-consistent TCA, which is the only approximation we shall seriously consider, is given then by Eqs. (4.4), (4.5), (4.7), together with

$$\Gamma^{SS'} = G^S\left(\delta_{SS'} + \sum_{S''\in T} \Phi^{SS''}\Gamma^{S''S'}\right) \quad (S, S'\in T)\,. \tag{5.6}$$

and Eqs. (5.1), (5.2), defining G^S.

This is the natural completion of the n-site CPA and reduces exactly to the regular CPA for the single-site case, where the equations take the following form:

$$W=\sum_x W^x, \tag{5.7}$$

$$G=(L_0-W)^{-1}, \tag{5.8}$$

$$W^x=\bar{V}_x+V_x'(G^{-1}-\tilde{V}_x+W^x)^{-1}V_x'. \tag{5.9}$$

To see that this is the CPA, look first at Eqs. (2.5) and (2.6), with L_0 replaced by

$$(G^x)^{-1}=G^{-1}+W^x \tag{5.10}$$

and $W^{(1)}$ identified as W^x. It follows that

$$[(G^x)^{-1}-W^x]^{-1}\equiv G=\langle[(G^x)^{-1}-V_x]^{-1}\rangle \tag{5.11}$$

$$=\langle(G^{-1}+W^x-V_x)^{-1}\rangle, \tag{5.12}$$

a standard form for the CPA. In fact, Eqs. (5.7)–(5.9) are exactly (though not transparently) equivalent to the "iterated average t-matrix approximation" introduced by Chen,[22] and found previously[21] to converge to a unique self-consistent analytic solution.

VI. PROPERTIES OF THE TRAVELING-CLUSTER APPROXIMATION

In the appendices the TCA is cast in a quite general form suitable for a broad range of problems, and a theorem is proved that shows quite generally that the TCA is a good self-consistent approximation, namely that, starting with any set of Herglotz operator functions W^S, iteration of the TCA equations for complex E converges to a unique, Herglotz solution of the self-consistent equations. This incidentally provides a proof for the conventional CPA that is somewhat more rigorous and more general in its application than that of Ducastelle[21] for the Cluster (or Molecular) CPA, which in turn is a generalization of the original proof of Müller-Hartmann[23] for the single-band case.

The method, and the theorem, are applicable to the locator formalism of Matsubara and Toyozawa[24,25] and others, to the elastic modes of a quasicrystalline solid with random masses, to the Blackman-Esterling-Berk[26] formalism for dealing with off-diagonal disorder, and to a variety of other problems with the same logical structure. The generalization to more than two species goes through, but is nontrivial, since the basic summation of cumulant averages, as in Eqs. (2.5) and (2.6), gives a quite different form in general, and an additional matrix structure must be introduced to deal with it. This formalism, which is developed in Appendix A and provides the necessary framework for proving the theorem in the case of non-Hermitian random terms in H, may also provide a somewhat better basis than the formalism developed above for the binary system for consideration of the difficult problem of correlated systems.

An important property that the TCA shares with the CPA is invariance with respect to choice of H_0. That is, if a nonrandom term,

$$\Delta V=\sum_x \Delta V_x, \tag{6.1}$$

is added to V and also to L_0 (i.e., subtracted from H_0), then G is unaltered in any given approximation. This can be seen by inspection of the equations [(4.4), (4.5), (4.7), (5.1), (5.2), (5.6)] which define the self-consistent approximation and Eqs. (2.7)–(2.9), which define $\bar{V}_x$, $\tilde{V}_x$, and V_x'. We find that ΔV_x is added to $\bar{V}_x$ and $\tilde{V}_x$, V_x' is unaltered, and $\Phi^{SS'}$ is augmented by $\delta_{SS'}\sum_{S^+}\Delta V_x$. If we assume $\Gamma^{SS'}$ is unaltered, then $\Delta V_x\delta_{xy}$ is added to W^{xy}, $\sum_{\hat{S}}\Delta V_x$ is added to W^S, and ΔV to W. Then $(G^S)^{-1}$ is augmented by $\sum_{S^+}\Delta V_x$, with the result that $\Gamma^{SS'}$ is unaltered, consistent with the assumption. Since the self-consistent solution is unique, the assumption is thus necessarily true, in fact, and L_0-W, and hence G, are unaltered. (It is interesting to note that it is not necessary for ΔV to be translation invariant.) The TCA, then, like the CPA is symmetric between host and impurity and is exact in both of the limits $c_A=1$ and $c_B=1$. H_0 can be chosen for convenience in any way; for example, it can be chosen to correspond to the virtual-crystal approximation, so that $\bar{V}_x=0$, thereby reducing (almost imperceptibly) the complexity of the calculations. It does not appear, incidentally, that the TCA is invariant, as is the CPA in many cases,[25] under the propagator-locator inversion.

VII. SINGLE-BAND MODEL–CALCULATIONS

There is a considerable variety of different cases which can be handled within the framework of a single-band model, including the elastic modes of a Bravais lattice with random masses, the electronic problem with separable potentials of random strengths and uniform shape, as well as the standard tight-binding model which we consider here by way of example.

Perhaps the most significant point in this calculation is the way in which the infinite family of sets S included in T is related by translation to a finite number of representative sets Σ, so that Fourier inversion reduces the equations to a finite set of linear equations for each value k of the crystal momentum.

The operators of the theory become $N\times N$ matrices (N is the number of sites in the sample),

with rows and columns labeled by x; V_x has only one nonzero element, equal to ϵ_A or ϵ_B, at the xth diagonal location, while $\bar{V}_x$, $\tilde{V}_x$, and V'_x have corresponding elements $\bar{\epsilon}$, $\tilde{\epsilon}$, and ϵ', at that location. The matrix W^{xy}, also, has just one nonzero element, at the (x, y) location, equal to W_{xy}, the corresponding element of the matrix W, given by

$$W_{xy} = \bar{\epsilon}\delta_{xy} + \epsilon'^2\Gamma^{xy}_{xy}. \tag{7.1}$$

Equations (5.1) and (5.6) do not simplify particularly, while Eqs. (4.7) and (5.2) can be rewritten to advantage as

$$\Phi^{SS'}_{xy} = \delta_{xy}\{\delta_{SS'}[\tilde{\epsilon}\theta(x\in S) + \bar{\epsilon}\theta(x\in\tilde{S})] + \epsilon'[\delta(S'; S, x) + \delta(S; S', x)]\}, \tag{7.2}$$

$$W^S_{xy} = W_{xy}\theta(x\in\hat{S})\theta(y\in\hat{S}). \tag{7.3}$$

In these equations we have introduced the logical functions θ ("statement"), which equals 1 if "statement" is true and equals 0 otherwise, and $\delta(S'; S, x)$, given by

$$\delta(S'; S, x) = \theta(x\in S)\theta(x\cup S = S'). \tag{7.4}$$

We shall be able to use the translation invariance of the problem (as $N\to\infty$) to handle sums over the whole sample by means of Fourier transform, so that only finite-matrix manipulations are left. To this end the matrix W^S [Eq. (7.3)] can be related to the translation-invariant matrix W by mean of a projection operator Λ^S,

$$\Lambda^S_{xy} = \delta_{xy}\theta(x\in S^+) \tag{7.5}$$

so that

$$W^S = (1-\Lambda^S)W(1-\Lambda^S). \tag{7.6}$$

In fact, the only components of $\Gamma^{SS'}_{xy}$ that are needed [Eqs. (5.6), (7.1)] are those for which $x\in S^+$ and $y\in S'^+$, so that only the portion of G^S within the subspace S^+ need be evaluated. We denote this submatrix by $\underline{G}^S$, and use the single underline in general to indicate a submatrix with row and column labels restricted to S^+. With the substitution (7.6), the inverse (5.1) works out to give

$$\underline{G}^S = [(1+\underline{WG})\underline{G}^{-1}(1+\underline{GW}) - \underline{WGW} - \underline{W}]^{-1}, \tag{7.7}$$

where G is the mean resolvent [Eq. (1.5)], and $\underline{G}^{-1}$ is the inverse of the submatrix $\underline{G}$. The $N\times N$ matrix products, WG, GW, and WGW, appearing here involve only translation-invariant matrices and can be done by Fourier inversions.

A form involving fewer Fourier inversions can be obtained in the case that the off-diagonal elements of L_0 are restricted to finite range, since Eq. (1.5) allows us to write

$$WG = L_0G - 1, \tag{7.8}$$

$$GW = GL_0 - 1, \tag{7.9}$$

and hence,

$$WGW = L_0GL_0 - L_0 - W. \tag{7.10}$$

Equation (7.7) then takes the form

$$\underline{G}^S = (\underline{L_0GG^{-1}GL_0} - \underline{L_0GL_0} + \underline{L_0})^{-1}, \tag{7.11}$$

where the unprojected matrix products involve only a finite neighborhood of S^+.

We now make use of the assumed translation invariance of the family of sets T which defines the particular approximation being considered to go to a Fourier-transformed representation. The crucial equation is Eq. (5.6), and we need to express the combination (S, x), where $x\in S^+$, in terms of an overall translation—a sort of center-of-mass coordinate—and a relative configuration of local character. To do this, we group the sets S into equivalence classes, where two sets are "equivalent" if they are related by a translation. Letting $S+\boldsymbol{r}$ represent the set obtained from S by translation through $\boldsymbol{r}$, i.e.,

$$S+\boldsymbol{r} = \{x \mid x-\boldsymbol{r}\in S\}, \tag{7.12}$$

we say that

$$S' \equiv S$$

if and only if

$$S' = S+\boldsymbol{r} \text{ for some } \boldsymbol{r}. \tag{7.13}$$

A choice of T involving only localized clusters would then comprise only a finite number of equivalence classes. Each equivalence class S can be characterized by one representative Σ of the class, and every member of the class can be expressed uniquely as a translation of the representative set Σ:

$$S = \Sigma_S + \boldsymbol{r}_S, \tag{7.14}$$

or

$$S_{\Sigma,\boldsymbol{r}} = \Sigma + \boldsymbol{r}. \tag{7.15}$$

In like fashion, the combination (S, x) can be expressed as a translation of a basic pair (Σ, ξ), consisting of the representative set Σ associated with S and the corresponding lattice point ξ:

$$S = \Sigma + \boldsymbol{r}, \tag{7.16}$$

$$x = \xi + \boldsymbol{r}, \tag{7.17}$$

$$\xi\in\Sigma^+. \tag{7.18}$$

The translation invariance of the theory now expresses itself in the fact that we can write

$$\Phi^{SS'}_{xx'} = \Phi^{\Sigma\Sigma'}_{\xi\xi'}(\boldsymbol{r}-\boldsymbol{r}'), \tag{7.19}$$

and

$$G^S_{xx'} = G^{\Sigma}_{\xi\xi'}(\boldsymbol{r}-\boldsymbol{r}'), \tag{7.20}$$

so that

$$\Gamma_{xx'}^{SS'} = \Gamma_{\xi\xi'}^{\Sigma\Sigma'}(r - r'). \tag{7.21}$$

Letting

$$\Gamma_{0\,\xi\xi'}^{\Sigma\Sigma'}(r - r') = G_{\xi\xi'}^{\Sigma}(r - r')\delta_{\Sigma\Sigma'}, \tag{7.22}$$

we can introduce a full matrix notation and write

$$\bar{\Gamma}(r - r') = \bar{\Gamma}_0(r - r') + \sum_{r_1, r_2} \bar{\Gamma}_0(r - r_1)\bar{\Phi}(r_1 - r_2)\bar{\Gamma}(r_2 - r'), \tag{7.23}$$

where the bar indicates a matrix of finite order, with rows and columns labeled by the pair (Σ, ξ) and with the restriction $\xi \in \Sigma^+$. The reduced matrix Φ takes the form:

$$\Phi_{\xi\xi'}^{\Sigma\Sigma'}(r) = \delta_{\Sigma\Sigma'}\delta_{\xi\xi'}\delta_{r,0}[\bar{\epsilon}\theta(\xi \in \Sigma) + \bar{\epsilon}\theta(\xi \in \bar{\Sigma})] + \delta_{\xi+r,\xi'}\epsilon'[\delta(\Sigma' - r; \Sigma, \xi) + \delta(\Sigma + r; \Sigma', \xi')], \tag{7.24}$$

and $\underline{G}^{\Sigma}(r)$ is given simply by

$$\underline{G}^{\Sigma}(r) = \delta_{r,0}\underline{G}^{\Sigma}, \tag{7.25}$$

where G^{Σ}, which is independent of r, is given by Eq. (7.7) or (7.11). The coherent-potential matrix $W_{xx'}$, from Eq. (7.1), now becomes a function only of $x - x'$:

$$W_{xx'} = W(x - x') = \bar{\epsilon}\delta_{x,x'} + \epsilon'^2\Gamma_{00}^{00}(x - x'), \tag{7.26}$$

where the representative single-site set is taken as the site $x = 0$. We can now perform the Fourier inversion, using, for any $F(y)$,

$$\hat{F}(k) = \sum_y F(y)e^{-ik\cdot y}, \tag{7.27}$$

$$F(y) = \frac{1}{\Omega}\int dk F(k)e^{ik\cdot y}. \tag{7.28}$$

Here k is restricted to the first Brillouin zone, whose volume is taken as Ω. The equations of the T approximation now become

$$\underline{\Gamma}_0^{\Sigma\Sigma'}(k) \equiv \underline{\Gamma}_0^{\Sigma\Sigma'} = \delta_{\Sigma\Sigma'}\underline{G}^{\Sigma} \tag{7.29}$$

(independent of k),

$$\hat{W}(k) = \bar{\epsilon} + \epsilon'^2\hat{\Gamma}_{00}^{00}(k), \tag{7.30}$$

$$\hat{\Phi}_{\xi\xi'}^{\Sigma\Sigma'}(k) = \delta_{\Sigma\Sigma'}\delta_{\xi\xi'}[\bar{\epsilon}\theta(\xi \in \Sigma) + \bar{\epsilon}\theta(\xi \in \bar{\Sigma})] + e^{ik\cdot(\xi - \xi')}\epsilon'[\delta(\Sigma' - \xi'; \Sigma - \xi, 0) + \delta(\Sigma - \xi; \Sigma' - \xi', 0)] \tag{7.31}$$

[note that $\delta(S'; S, x)$ is translation invariant, i.e., $\delta(S'; S, x) = \delta(S' - x; S - x, 0)$],

$$\hat{\bar{\Gamma}}(k) = [\hat{\bar{\Gamma}}_0^{-1} - \hat{\bar{\Phi}}(k)]^{-1}. \tag{7.32}$$

In fact, only the single element $\hat{\Gamma}_{00}^{00}(k)$ is needed to calculate $\hat{W}(k)$, so that only the ratio of two determinants is needed in a calculation, rather than the entire matrix inverse. The size of the matrix burgeons rather rapidly as the size of clusters included in T increases. A few cases are tabulated in Table I.

Computer calculations have been carried out on the IBM 361 facility at The Ohio State University, to test the nearest-neighbor TCA for the single-band tight-binding model on simple square and cubic lattices with site-diagonal disorder. For real E, of course, the theorem does not guarantee convergence, and, as might be expected, minor convergence problems were encountered close to band edges, but not elsewhere. In Figs. 9 and 10 the fit of the TCA to the numerically calculated density of states of Alben *et al.*[9] is shown for two choices of the parameters and compared with that of the CPA. Characteristically, the TCA does what a nearest-neighbor cluster approximation ought to do and does it reasonably well. The band edges are fit noticeably better than by the CPA, the overall fit is somewhat closer, and the structure due to localized states begins to appear. In Fig. 10 the satellite peaks in the minority band due to bonding and antibonding levels of a nearest-neighbor impurity pair show up in addition to the central peak due to isolated impurities.

VIII. CONCLUSIONS

By means of partial sums of overlapping cumulant averages, we have developed an expansion pro-

TABLE I. Order of Γ for various choices of T in one dimension (1D), two dimensions (2D) (square lattice), and three dimensions (3D) (simple cubic lattice). n is the maximum cluster size; d is the maximum diameter of cluster.

	1D			2D			3D			
	d			d			d			
n	1	2	3	1	$\sqrt{2}$	2	1	$\sqrt{2}$	$\sqrt{3}$	2
2	5	9	13	9	17	25	13	37	53	65
3		15	31		41	85		157	317	491
4			39		49	133		261	853	1579

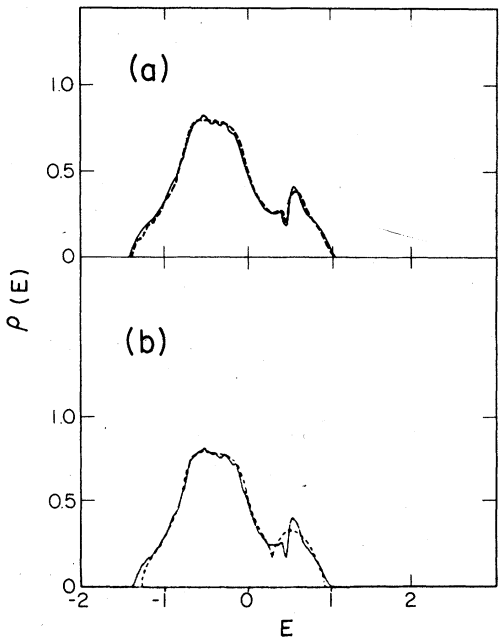

FIG. 9. Density of states for a three-dimensional alloy with $\epsilon_A = -\epsilon_B = 0.4$, $c_A = 0.16$. Solid curve: exact numerical calculation of Alben *et al.* (Ref. 8). (a) Broken curve: TCA(nn). (b) Broken curve: CPA.

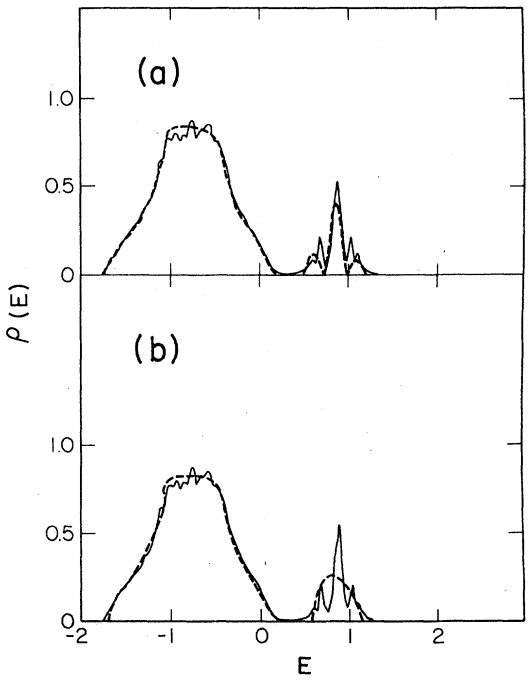

FIG. 10. Density of states for a three-dimensional alloy with $\epsilon_A = -\epsilon_B = 0.75$, $c_A = 0.1$. Solid curve; exact numerical calculation of Alben *et al.* (Ref. 8). (a) Broken curve: TCA(nn). (b) Broken curve: CPA.

cedure for the coherent potential W involving in each term only sequences of single factors, grouped into "modified cumulant averages," with an associated graphical representation. This method permits a straightforward term-by-term analysis of the Herglotz property of W and leads naturally to an approximation scheme, the "traveling-cluster approximation," that generalizes the CPA and correctly preserves the Herglotz property in a wide range of cases, as well as the translation symmetry (in the mean) of the original problem. The nearest-neighbor TCA gives a markedly improved fit to the density of states, over the CPA, for the simple systems studied numerically, but the complexity of calculation increases drastically with the order of approximation, as is true also of other self-consistent cluster approximations.

This approach has as yet shed no light on the problem of amorphous systems or of correlated substitutional systems, though the formalism developed here may well prove fruitful for further investigation of such problems.

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APPENDIX A

General formulation

We here present a formalism[10] adapted to a more general class of systems and suited, in particular, to substitutional systems with more than two species, where the crucial simplification of Eq. (2.6) does not occur. The configurations of the system will be specified by a set $\underline{\alpha}$ of parameters, which we take to be denumerable and shall shortly allow to be the assignment of species at the sites of a Bravais lattice. The probability of the configuration $\underline{\alpha}$ is $P(\underline{\alpha})$. We suppose that the generalized resolvent for the system is $[L^{\underline{\alpha}}(E)]^{-1}$, where the complex variable E may correspond to energy, squared frequency, or some other real physical

parameter, and $L^{\underline{\alpha}}(E)$, a linear operator on the Hilbert space of the physical system, is expressible as

$$L^{\underline{\alpha}}(E) = L_0(E) - V^{\underline{\alpha}}(E) . \tag{A.1}$$

Here $-L_0(E)$ is Herglotz, and $V^{\underline{\alpha}}(E)$, the random part of $L^{\underline{\alpha}}$, is either Herglotz, or Hermitian and independent of E. It is necessary for the application of our fixed-point theorem that $V^{\underline{\alpha}}(E)$ be a uniformly bounded linear transformation in any closed bounded domain in $\mathcal{G}_+$.

The mean resolvent

$$G(E) = \langle\langle (L^{\underline{\alpha}})^{-1} \rangle\rangle \tag{A.2}$$

can also be written as a scalar product in an augmented space, after the manner of Haydock *et al.*,[27] Mookerjee,[28] and others.[29] Let u be a unit vector in a Hilbert space $\mathcal{S}_c$ whose basis vectors are labeled by the configurations $\underline{\alpha}$. The components $u_{\underline{\alpha}}$ are taken as

$$u_{\underline{\alpha}} = [P(\underline{\alpha})]^{1/2} , \tag{A.3}$$

so that the mean resolvent can be written as

$$G = \sum_{\underline{\alpha}} |u_{\underline{\alpha}}|^2 (L^{\underline{\alpha}})^{-1} \tag{A.4}$$

$$= \bar{u}(L_0 - \mathfrak{V})^{-1} u , \tag{A.5}$$

where $\mathfrak{V}$ is a linear operator on the product space $\mathcal{S}$,

$$\mathcal{S} = \mathcal{S}_c \otimes \mathcal{K} , \tag{A.6}$$

and can be defined as an operator-valued matrix,

$$\mathfrak{V}_{\underline{\alpha}\underline{\alpha}'} = V^{\underline{\alpha}} \delta_{\underline{\alpha}\underline{\alpha}'} . \tag{A.7}$$

In what follows, for any operator on one of the spaces $\mathcal{S}_c$ or $\mathcal{K}$, we let the same symbol represent the corresponding operator on $\mathcal{S}$. Thus the symbol L_0 in Eq. (A.5) represents the operator $L_0 \otimes I_c$, where I_c is the unit operator on $\mathcal{S}_c$.

We note that $u\bar{u}$ is a projector on the "configuration space" $\mathcal{S}_c$, and define the complementary projector Q,

$$Q = I_c - u\bar{u} . \tag{A.8}$$

Starting with the separation

$$L_0 - \mathfrak{V} = L_0 - Q\mathfrak{V} - u\bar{u}\mathfrak{V} , \tag{A.9}$$

we perform some manipulations to obtain an explicit expression for the coherent potential W, as follows. First we multiply (A.9) on the left by $(L_0 - \mathfrak{V})^{-1}$ and on the right by $(L_0 - Q\mathfrak{V})^{-1}$ and rearrange to get

$$(L_0 - \mathfrak{V})^{-1} = (L_0 - Q\mathfrak{V})^{-1} + (L_0 - \mathfrak{V})^{-1} u\bar{u}\mathfrak{V}(L_0 - Q\mathfrak{V})^{-1} , \tag{A.10}$$

so that

$$G = \bar{u}(L_0 - \mathfrak{V})^{-1} u = L_0^{-1} + G\bar{u}\mathfrak{V}(L_0 - Q\mathfrak{V})^{-1} u \tag{A.11}$$

$$= [L_0 - \bar{u}\mathfrak{V}(1 - G_0 Q\mathfrak{V})^{-1} u]^{-1} . \tag{A.12}$$

Here

$$\bar{u}(L_0 - Q\mathfrak{V})^{-1} u = \bar{u} L_0^{-1} u \tag{A.13}$$

$$= L_0^{-1} , \tag{A.14}$$

since L_0 and Q commute and

$$\bar{u}Q = 0 . \tag{A.15}$$

From Eq. (A.12) and the definition of the coherent potential [Eq. (1.5)], we see that

$$W = \bar{u}\mathfrak{V}(1 - G_0 Q\mathfrak{V})^{-1} u \tag{A.16}$$

$$= \sum_{n=0}^{\infty} \bar{u}\mathfrak{V}(G_0 Q\mathfrak{V})^n u . \tag{A.17}$$

This expression represents W as a sum of products of operators, in a form that permits in general an analysis of the negative definiteness property [Eq. (1.7)]. The trick is to find a workable approximation to Q.

We now specialize to the substitutional alloy, whose random term V is a sum over sites of a Bravais lattice,

$$V^{\underline{\alpha}} = \sum_x V_x^{\alpha_x} , \tag{A.18}$$

and whose configuration is specified by the assignment of a species $\alpha_x = 1, 2, \ldots, \nu$ to each site x. We also keep to the case of an uncorrelated system, for which

$$P(\underline{\alpha}) = \prod_x P(\alpha_x) , \tag{A.19}$$

and

$$u_{\underline{\alpha}} = \prod_x [P(\alpha_x)]^{1/2} = \prod_x u_{\alpha_x} . \tag{A.20}$$

In this case $\mathcal{S}_c$ can be regarded as a product of "site spaces" $\mathcal{S}_x$:

$$\mathcal{S}_c = \prod_x \otimes \mathcal{S}_x , \tag{A.21}$$

$$I_c = \prod_x \otimes I_x , \tag{A.22}$$

$$u = \prod_x \otimes u_x , \tag{A.23}$$

and

$$I_x = u_x \bar{u}_x + Q_x . \tag{A.24}$$

In these terms the operator Q can be analyzed,

$$Q = \prod_x \otimes (u_x \bar{u}_x + Q_x) - \prod_x \otimes u_x \bar{u}_x \tag{A.25}$$

$$= \sum_{S \neq \phi} P_S , \tag{A.26}$$

where the projector P_S associated with each possible overlap set S is given by

$$P_S = \left(\prod_{x\in S} \otimes Q_x\right)\left(\prod_{y\in \bar{S}} \otimes u_y \bar{u}_y\right), \tag{A.27}$$

$\bar{S}$ being the complement of S. The projectors P_S are orthogonal, and a product $P_S \mathcal{V} P_{S'}$ vanishes unless S and S' differ at most by the site x.

We can now see, by using Eq. (A.26), how the expansion (A.17) duplicates the structure of the MCA expansion of Sec. II. A single term, corresponding to a product of factors of the form $\mathcal{V}_x$ and P_S, is represented by a single MCA graph. Here $\mathcal{V}_x$ is an operator on $\mathcal{S}_x \otimes \mathcal{H}$,

$$(\mathcal{V}_x)_{\alpha_x \alpha_x'} = \delta_{\alpha_x \alpha_x'} V_x^{\alpha_x}, \tag{A.28}$$

or may also represent the corresponding operator on $\mathcal{S}$. The factor P_S at any stage of the graph corresponds to an overlap set S, and the exclusion of $S = \Phi$, the null set, in Eq. (A.26) corresponds to the restriction to *proper* self-energy type graphs. From one factor P_S to the next, S must remain the same or vary by the addition or removal of one site. In a product $\mathcal{V}_x P_S \mathcal{V}_{x'}$ the factors $u_y \bar{u}_y$ and Q_y for sites other than x and x' are passive because of their character as projection operators and can be dropped, since for each site y the same factor must appear also to left and right. A factor Q_y represents a continuing MCA line, and a factor $u_y \bar{u}_y$ represents the absence of such a line. An isolated dot still represents the same factor as before, namely,

$$\bar{u}_x \mathcal{V}_x u_x = \bar{V}_x, \tag{A.29}$$

while $\bar{V}_x$ and V_x' are generalized to vectors and matrices as follows:

$$(u_x \mathcal{V}_x Q_x)_{\alpha_x} = V'_{x\alpha_x} \quad \text{(row vector)}, \tag{A.30}$$

$$(Q_x \mathcal{V}_x u_x)_{\alpha_x} = V''_{x\alpha_x} \quad \text{(column vector)}, \tag{A.31}$$

$$(Q_x \mathcal{V}_x Q_x)_{\alpha_x \alpha_x'} = V_{x\alpha_x \alpha_x'} \quad \text{(square matrix)}. \tag{A.32}$$

It is clear now that the traveling-cluster approximation is constructed, exactly as before, by restricting the overlap sets S to a family T, and including all graphs consistent with that constraint. Self-consistency is introduced as before, and the approximation is defined by Eqs. (4.4), (5.1), and (5.2), which are unaltered, together with the following:

$$W^{xy} = \bar{V}_x \delta_{xy} + \sum_{\alpha_x, \alpha_y} V'_{x\alpha_x} \Gamma^{xy}_{\alpha_x \alpha_y} V''_{y\alpha_y}, \tag{A.33}$$

$$\Gamma^{SS'}_{\alpha\alpha'} = G^S \left(\delta_{SS'} \delta_{\alpha\alpha'} + \sum_{\alpha''} \sum_{S''\in T} \Phi^{SS''}_{\alpha\alpha''} \Gamma^{S''S'}_{\alpha''\alpha'}\right), \tag{A.34}$$

$$\Phi^{SS'}_{\alpha\alpha'} = \delta_{SS'} \left(\sum_{x\in S} \bar{V}_{x\alpha_x \alpha_x'} \prod_{\substack{y\in S \\ y\neq x}} \delta_{\alpha_y \alpha_y'} + \delta_{\underline{\alpha}\underline{\alpha}'} \sum_{x\in S} \bar{V}_x\right) + \sum_x \left[\delta(S'; S, x) V'_{x\alpha_x'} \prod_{y\in S} \delta_{\alpha_y \alpha_y'} + \delta(S; S', x) V''_{x\alpha_x} \prod_{y\in S'} \delta_{\alpha_y \alpha_y'}\right]. \tag{A.35}$$

Here $\underline{\alpha}$ is the set of parameters α_x associated with sites $x \in S$.

In the case of a binary alloy, $\nu = 2$, and Q_x becomes a matrix of rank one which can be written as

$$Q_x = u_x' \bar{u}_x', \tag{A.36}$$

$$u'_{xA} = (c_B)^{1/2}, \tag{A.37}$$

$$u'_{xB} = -(c_A)^{1/2}. \tag{A.38}$$

The matrix elements of $\mathcal{V}_x$ between the appropriate combinations of u_x and u_x' then give rise to the original forms V_x and V_x' of Eqs. (2.8) and (2.9), and the remaining vectors u_x' and $\bar{u}_x'$ appearing in Eqs. (A.30)–(A.32) disappear as scalar products equal to one when all the factors of a full MCA graph are assembled. The simpler prescription of Sec. V is then recovered.

APPENDIX B

Fixed-point theorem and proof of analyticity

We now recast the equations of the TCA into a form suitable for analysis rather than calculation. We start with the formulation of Appendix A, where the basic expression (A.16) for W involves the projection operator Q, which is expanded [Eq. (A.26)] in terms of projectors P_S involving the different possible overlap sets S. The T approximation consists first of restricting the sum in Eq. (A.26) to the family T, i.e., replacing Q by Q_T, given by

$$Q_T = \sum_{S\in T} P_S. \tag{B.1}$$

Next, in Eq. (A.17), we replace G_0 by the self-consistent propagator G^S associated with each given overlap set S. Thus $G_0 Q$ in this equation is to be replaced by an operator $\mathcal{G}_T$ on $\mathcal{S}$, given by

$$\mathcal{G}_T = \sum_{S\in T} G^S P_S \tag{B.2}$$

$$= \{G^S\}, \tag{B.3}$$

where we introduce the notation $\{A^S\}$, defined for a family of operators A^S on $\mathcal{H}$ by

$$\{A^S\} = \sum_{S\in T} A^S P_S. \tag{B.4}$$

With this notation, Eq. (A.16) can be written as

$$W = \bar{u}\mathcal{V}(1 - \mathcal{G}_T\mathcal{V})^{-1}u \tag{B.5}$$

$$= \bar{u}\mathcal{V}(1 + \mathcal{G}\mathcal{V})u\,, \tag{B.6}$$

where $\mathcal{G}$ is the inverse, in the subspace defined by the projector Q_T, of $\mathcal{G}_T^{-1} - \mathcal{V}$. Because of the orthogonality of the subspaces defined by the projectors P_S and the definition [Eqs. (B.3),(5.1)] of $\mathcal{G}_T$, we can write this as

$$\mathcal{G} = Q_T\mathcal{L}^{-1}\,, \tag{B.7}$$

where

$$\mathcal{L} = L_0 - \mathcal{W} - Q_T\mathcal{V}Q_T\,, \tag{B.8}$$

and [Eq. (B.4)]

$$\mathcal{W} = \{W^S\}\,. \tag{B.9}$$

The individual terms W^{xy} needed to calculate W^S can be written as

$$W^{xy} = \bar{u}\mathcal{V}_x(\delta_{xy} + \mathcal{G}\mathcal{V}_y)u\,, \tag{B.10}$$

and W^S [Eq. (5.2)] takes the form

$$W^S = \bar{u}\mathcal{V}_{\hat{S}}(1 + \mathcal{G}\mathcal{V}_{\hat{S}})u\,, \tag{B.11}$$

where

$$\mathcal{V}_{\hat{S}} = \sum_{x\in\hat{S}} \mathcal{V}_x = \mathcal{V} - \mathcal{V}_{S^+}\,. \tag{B.12}$$

Equations (B.6)–(B.11) define the approximation, which can be expressed as the solution of a fixed-point equation,

$$\mathcal{F}(\mathcal{W}) = \mathcal{W}\,, \tag{B.13}$$

for a functional $\mathcal{F}(\mathcal{W})$ given by

$$\mathcal{F}(\mathcal{W}) = \{\bar{u}\mathcal{V}_{\hat{S}}(1 + \mathcal{G}\mathcal{V}_{\hat{S}})u\}\,, \tag{B.14}$$

with $\mathcal{G}$ given in terms of $\mathcal{W}$ by Eqs. (B.7) and (B.8).

In this appendix we prove that, for any nonreal E and any appropriately restricted initial choice of $\mathcal{W}$, iteration of Eq. (B.13) converges to a unique fixed point $\mathcal{W}(E)$ which is a Herglotz function of E. Furthermore, the coherent potential $W(E)$ determined by this $\mathcal{W}(E)$ through Eq. (B.6) is also Herglotz.

We need to assume the following. (i) Each $\mathcal{V}_x$ is a Herglotz operator function of E as defined in Sec. I. This includes the standard cases of electronic states in both the propagator form, where $\mathcal{V}_x$ is Hermitian, and the locator form, where the (Herglotz) locator g_x plays the role of V_x, and also the elastic-mode problem, where $V_x \propto E$. (ii) Each $\mathcal{V}_x(E)$, together with all the necessary derived quantities like $\mathcal{V}_S$ and $\mathcal{V}$, is a uniformly bounded operator on $\mathcal{S}$ in any closed finite domain $D \subset \mathcal{G}_+$ in the variable E. (iii) $-L_0(E)$ is a Herglotz operator function of E on $\mathcal{H}$, which need not be bounded. (iv) $\mathrm{Im}L_0$ has a uniform lowest bound $y > 0$ in D.

$$y = \inf_{E\in D} [\bar{\psi}(\mathrm{Im}L_0)\psi]/\bar{\psi}\psi > 0\,. \tag{B.15}$$

This property is trivial for the electronic problem [Eq. (1.3)] and for the elastic-mode problem and is in fact true in general if G_0 ($\equiv L_0^{-1}$) has a spectral representation of the form

$$G_0(E) = \int \frac{\rho_0(\omega)}{E - \omega}\, d\omega\,, \tag{B.16}$$

with $\rho_0(\omega)$ a positive semidefinite operator satisfying

$$\int \rho_0(\omega)\, d\omega = I\,. \tag{B.17}$$

This follows readily from the positive definiteness of the expression

$$\int\left(\frac{\bar{L}_0}{E^* - \omega} - 1\right)\rho_0(\omega)\left(\frac{L_0}{E - \omega} - 1\right) d\omega$$

$$= \frac{1}{\mathrm{Im}E}(\mathrm{Im}L_0) - 1\,. \tag{B.18}$$

Restricting our attention to the case $E \in D \subset \mathcal{G}_+$, we define $\mathfrak{D}_-$ to be the set of bounded linear transformations on $\mathcal{S}$ of the form (B.4) with negative semidefinite anti-Hermitian part:

$$\mathfrak{D}_- = \{A \,|\, A = \{A^S\},\ \mathrm{Im}A \leq 0,\ \|A\| < \infty\}\,, \tag{B.19}$$

and look for a solution $\mathcal{W}$ within this set.

Now supposing that $\mathcal{W} \in \mathfrak{D}_-$, let us find a bound for the norm of $\mathcal{F}(\mathcal{W})$, using the conventional operator norm:

$$\|A\| = \sup_\psi \|A\psi\|/\|\psi\|\,. \tag{B.20}$$

We find

$$\|\mathcal{F}(\mathcal{W})\| = \left\|\sum_{S\in T} P_S\bar{u}\mathcal{V}_{\hat{S}}(1 + \mathcal{G}\mathcal{V}_S)u\right\| \tag{B.21}$$

$$= \sup_{S\in T} \|\bar{u}\mathcal{V}_{\hat{S}}(1 + \mathcal{G}\mathcal{V}_{\hat{S}})u\| \tag{B.22}$$

$$\leq \sup_{S\in T} \|u\|^2\|\mathcal{V}_{\hat{S}}\|(1 + \|\mathcal{G}\|\,\|\mathcal{V}_{\hat{S}}\|)\,, \tag{B.23}$$

where u, being a vector in $\mathcal{S}_c$ is to be regarded as a linear transformation from $\mathcal{H}$ into $\mathcal{S}$. Equation (B.22) follows from the fact that the P^S's define orthogonal subspaces of $\mathcal{S}_c$. Now

$$\|u\|^2 = \sum_{\underline{\alpha}} P(\underline{\alpha}) = 1\,, \tag{B.24}$$

and

$$\|\mathcal{G}\| = \|Q_T\mathcal{L}^{-1}\| \leq \|\mathcal{L}^{-1}\| \tag{B.25}$$

$$\leq 1/\inf_\varphi(\bar{\varphi}\,\mathrm{Im}\mathcal{L}\varphi) \tag{B.26}$$

$$\leq 1/y\,, \tag{B.27}$$

from Eqs. (B.8) and (B.15), and the fact that $\mathrm{Im}\mathcal{V}'$ and $\mathrm{Im}\mathcal{W}$ are negative semidefinite. If we define

$$\beta = \sup_{s\in T} \|\mathcal{V}_{\hat{s}}\|, \tag{B.28}$$

then Eqs. (B.23), (B.27) give

$$\|\mathcal{F}(\mathcal{W})\| \leq \beta + \beta^2/y$$
$$= \gamma, \tag{B29}$$

say, and we see that $\mathcal{F}(\mathcal{W})$ is uniformly bounded for $\mathcal{W}\in\mathfrak{D}_-$ and $E\in\mathfrak{D}$.

Now we consider the difference $\Delta\mathcal{F}$ in $\mathcal{F}(\mathcal{W})$ for two different values of $\mathcal{W}$, say $\mathcal{W}_1$ and $\mathcal{W}_2$, and also, to deal with several cases of interest, for two different sets of values of $\mathcal{V}_x$. Letting

$$\chi_S = (1+\mathcal{G}\mathcal{V}_{\hat{S}})u, \tag{B.31}$$

$$\chi'_S = (1+\bar{\mathcal{G}}\mathcal{V}_{\hat{S}})u, \tag{B.32}$$

we have, after some manipulation,

$$\Delta\mathcal{F} = \mathcal{F}(\mathcal{W}_1) - \mathcal{F}(\mathcal{W}_2)$$
$$= \{\bar{u}\Delta\mathcal{V}_{\hat{S}}\chi_{S2}\}$$
$$- \{\bar{u}\mathcal{V}_{\hat{S}1}\mathcal{G}_1\Delta\mathcal{L}\mathcal{G}_2\mathcal{V}_{\hat{S}2}u\} + \{\bar{u}\mathcal{V}_{\hat{S}1}\mathcal{G}_1\Delta\mathcal{V}_{\hat{S}}u\} \tag{B.33}$$
$$= \{\bar{\chi}'_{S1}\Delta\mathcal{V}_{\hat{S}}\chi_{S2}\} - \{\bar{u}\mathcal{V}_{\hat{S}1}\mathcal{G}_1\Delta\mathcal{L}_{S^+}\mathcal{G}_2\mathcal{V}_{\hat{S}2}u\}, \tag{B.34}$$

where

$$\mathcal{L}_{S^+} = \mathcal{L} + Q_T\mathcal{V}_{\hat{S}}Q_T \tag{B.35}$$
$$= L_0 - \mathcal{W} - Q_T\mathcal{V}_{S^+}Q_T \tag{B.36}$$

[cf. Eq. (B.8)]. In particular, we can obtain $\mathrm{Im}\mathcal{F}$ as a special case of Eq. (B.34):

$$\mathrm{Im}\mathcal{F}(\mathcal{W}) = \{\bar{\chi}_S\,\mathrm{Im}\mathcal{V}_{\hat{S}}\chi_S\} - \{\bar{u}\bar{\mathcal{V}}_{\hat{S}}\bar{\mathcal{G}}(\mathrm{Im}\mathcal{L}_{S^+})\mathcal{G}\mathcal{V}_{\hat{S}}u\} \tag{B.37}$$
$$\leq 0, \tag{B.38}$$

where χ'_{S1} becomes χ_S because $\mathcal{G}_1 = \bar{\mathcal{G}}$ and $\mathcal{V}_{\hat{S}1} = \mathcal{V}_{\hat{S}}$. We see, then, that $\mathcal{F}$ maps into itself the closed subset of $\mathfrak{D}_-$ corresponding to norm $\leq\gamma$. It is thus clear already that $\mathcal{F}$ has at least one fixed point in that region and no fixed points in $\mathfrak{D}_-$ outside of it.

We now consider the iteration of $\mathcal{F}(\mathcal{W})$. Let $\mathcal{W}_0 \in \mathfrak{D}_-$ be the initial choice of $\mathcal{W}$, and let

$$\mathcal{W}_{n+1} = \mathcal{F}(\mathcal{W}_n), \quad n = 0, 1, 2, \ldots . \tag{B.39}$$

Then

$$\Delta\mathcal{W}_n = \mathcal{W}_{n+1} - \mathcal{W}_n \tag{B.40}$$
$$= \mathcal{F}(\mathcal{W}_n) - \mathcal{F}(\mathcal{W}_{n-1}) \tag{B.41}$$
$$= \{u\mathcal{V}_{\hat{S}}\mathcal{G}_n\Delta\mathcal{W}_{n-1}\mathcal{G}_{n-1}\mathcal{V}_{\hat{S}}u\}, \tag{B.42}$$

from Eqs. (B.34) and (B.36). It does not appear to follow from this that $\|\Delta\mathcal{W}_n\|$ is necessarily less than $\|\Delta\mathcal{W}_{n-1}\|$, so we have to modify the problem somewhat in order to see convergence. We make use of the similarity between (B.42) and the second term on the right-hand side of Eq. (B.37), which is limited by $\mathrm{Im}\mathcal{F}(\mathcal{W})$. We write

$$l_{Sn} = \mathrm{Im}\mathcal{L}_{S^+n} \geq y, \tag{B.43}$$

$$f_{Sn} = \bar{u}\bar{\mathcal{V}}_{\hat{S}}\bar{\mathcal{G}}_n l_{Sn}\mathcal{G}_n\mathcal{V}_{\hat{S}}u \geq 0, \tag{B.44}$$

$$f_n = \{f_{Sn}\} \leq -\mathrm{Im}\mathcal{F}(\mathcal{W}_n) = -\mathrm{Im}\mathcal{W}_{n+1}. \tag{B.45}$$

We can express f_{Sn} in the form

$$f_{Sn} = \bar{\psi}\psi, \tag{B.46}$$

where ψ is a transformation from $\mathcal{H}$ to $\mathcal{S}$ (like u), given by

$$\psi = (l_{Sn})^{1/2}\mathcal{G}_n\mathcal{V}_{\hat{S}}u, \tag{B.47}$$

where the operator square root $(l_{Sn})^{1/2}$ is taken as positive definite. Furthermore, we can relate ψ to a "unit vector" t_{Sn} by taking out a factor $(f_{Sn})^{1/2}$

$$\psi = t_{Sn}(f_{Sn})^{1/2}, \tag{B.48}$$

so that

$$\bar{t}_{Sn}t_{Sn} = I. \tag{B.49}$$

If f_{Sn} has zero eigenvalues, t_{Sn} becomes somewhat arbitrary, but can always be chosen to satisfy these equations. It follows from Eq. (B.49) that

$$\|t_{Sn}\| = 1. \tag{B.50}$$

In similar fashion we define f'_{Sn} and t'_{Sn} by

$$f'_{Sn} = \bar{u}\mathcal{V}_{\hat{S}}\mathcal{G}_n l_{Sn}\bar{\mathcal{G}}_n\bar{\mathcal{V}}_{\hat{S}}u, \tag{B.51}$$

$$t'_{Sn}(f'_{Sn})^{1/2} = (l_{Sn})^{1/2}\bar{\mathcal{G}}_n\bar{\mathcal{V}}_{\hat{S}}u, \tag{B.52}$$

so that

$$\bar{t}'_{Sn}t'_{Sn} = I, \tag{B.53}$$

$$\|t'_{Sn}\| = 1. \tag{B.54}$$

We now make these substitutions in Eq. (B.42), and get

$$\Delta\mathcal{W}_n = \left\{(f'_{Sn})^{1/2}t'_{Sn}\frac{1}{(l_{Sn})^{1/2}}\Delta\mathcal{W}_{n-1}\frac{1}{(l_{S,n-1})^{1/2}}t_{S,n-1}(f_{S,n-1})^{1/2}\right\} \tag{B.55}$$

$$= (f'_n)^{1/2}\left\{\bar{t}'_{Sn}\frac{1}{(l_{Sn})^{1/2}}\Delta\mathcal{W}_{n-1}\frac{1}{(l_{S,n-1})^{1/2}}t_{S,n-1}\right\}(f_{n-1})^{1/2} \tag{B.56}$$

$$= (f_n')^{1/2} A_n (f_{n-1})^{1/2}, \quad n \geq 1. \tag{B.57}$$

We find that the operators A_n do converge nicely to zero. We have, for $n \geq 2$,

$$A_n = \left\{ \bar{t}'_{Sn} \frac{1}{(l_{Sn})^{1/2}} (f'_{n-1})^{1/2} A_{n-1} (f_{n-2})^{1/2} \frac{1}{(l_{S,n-1})^{1/2}} t_{S,n-1} \right\}, \tag{B.58}$$

and find that

$$\left\| (f_{n-1})^{1/2} \frac{1}{(l_{Sn})^{1/2}} \right\|^2$$

$$= \sup_\varphi \left(\bar{\varphi} \frac{1}{(l_{Sn})^{1/2}} f_{n-1} \frac{1}{(l_{Sn})^{1/2}} \varphi \right) / \bar{\varphi}\varphi \tag{B.59}$$

$$\leq \sup_\varphi \left(\bar{\varphi} \frac{1}{(l_{Sn})^{1/2}} w_n \frac{1}{(l_{Sn})^{1/2}} \varphi \right) / \bar{\varphi}\varphi \tag{B.60}$$

$$= \sup_\psi \frac{\bar{\psi} w_n \psi}{\bar{\psi} l_{Sn} \psi} \tag{B.61}$$

$$\leq \sup_\psi \frac{\bar{\psi} w_n \psi}{y\bar{\psi}\psi + \bar{\psi} w_n \psi} \tag{B.62}$$

$$\leq \lambda, \tag{B.63}$$

where

$$\lambda = \gamma/(y+\gamma) < 1. \tag{B.64}$$

Here

$$w_n = -\mathrm{Im}\mathfrak{W}_n, \tag{B.65}$$

and Eq. (B.60) follows from Eq. (B.45). We use also the fact [Eq. (B.30)] that

$$\bar{\psi} w_n \psi / \bar{\psi}\psi \leq \|\mathfrak{W}_n\| \leq \gamma, \quad n \geq 1. \tag{B.66}$$

The same result holds, of course, for f'_n.

From Eqs. (B.50), (B.54), and (B.63), we now see immediately that

$$\|A_n\| \leq \lambda \|A_{n-1}\| \tag{B.67}$$

$$\leq \lambda^{n-1} \|A_1\|, \tag{B.68}$$

and

$$\|\Delta\mathfrak{W}_n\| \leq \|(f'_n)^{1/2}\| \, \|A_n\| \, \|(f_{n-1})^{1/2}\| \tag{B.69}$$

$$\leq \lambda^{n-1} \gamma \|A_1\|. \tag{A2.70}$$

Now for any m and $n > m$,

$$\|\mathfrak{W}_n - \mathfrak{W}_m\| = \left\| \sum_m^{n-1} \Delta\mathfrak{W}_p \right\| \tag{B.71}$$

$$\leq \gamma \|A_1\| \frac{\lambda^{m-1} - \lambda^{n-1}}{1-\lambda} \to 0 \quad (m, n \to \infty), \tag{B.72}$$

so that $\{\mathfrak{W}_n\}$ is a Cauchy sequence in the domain $\mathfrak{D}_-$. However, $\mathfrak{D}_-$ is complete, so that the sequence has a strong limit $\mathfrak{W}$ in $\mathfrak{D}_-$.

To show that $\mathfrak{W}$ is in fact a solution of Eq. (B.13), we observe that

$$\|\mathfrak{F}(\mathfrak{W}) - \mathfrak{W}_{n+1}\| = \|\mathfrak{F}(\mathfrak{W}) - \mathfrak{F}(\mathfrak{W}_n)\| \tag{B.73}$$

$$\leq \sup_S \|\bar{u}\mathfrak{V}_{\hat{S}} \mathfrak{g}(\mathfrak{W} - \mathfrak{W}_n) \mathfrak{g}_n \mathfrak{V}_{\hat{S}} u\| \tag{B.74}$$

$$\leq (\beta^2/y^2) \|\mathfrak{W} - \mathfrak{W}_n\| \to 0 \quad (n \to \infty), \tag{B.75}$$

making use of (B.24), (B.27) and (B.28). Thus

$$\mathfrak{F}(\mathfrak{W}) = \lim \mathfrak{W}_{n+1} = \mathfrak{W}, \tag{B.76}$$

as desired.

To see that this solution is unique we imagine $\mathfrak{W}_1$ and $\mathfrak{W}_2$ to be distinct solutions, and obtain, following the same pattern as above,

$$\Delta\mathfrak{W} = (f'_1)^{1/2} \left\{ \bar{t}'_{S1} \frac{1}{(l_{S1})^{1/2}} \Delta\mathfrak{W} \frac{1}{(l_{S2})^{1/2}} t_{S2} \right\} (f_2)^{1/2} \tag{B.77}$$

$$= (f'_1)^{1/2} A (f_2)^{1/2}, \tag{B.78}$$

with

$$A = \left\{ \bar{t}'_{S1} \frac{1}{(l_{S1})^{1/2}} (f'_1)^{1/2} A (f_2)^{1/2} \frac{1}{(l_{S2})^{1/2}} t_{S2} \right\}, \tag{B.79}$$

so that

$$\|A\| \leq \lambda \|A\|. \tag{B.80}$$

Since $\lambda < 1$, $\|A\|$, and hence $\Delta\mathfrak{W}$, must be zero, contrary to the assumed distinctness of the solutions.

Since γ and λ are fixed for the given closed domain $D \subset \mathfrak{g}_+$ of the variable E, we see that the sequence of functions $\{\mathfrak{W}_n(E)\}$ converges uniformly to $\mathfrak{W}(E)$ over D, provided only that $\mathfrak{W}_0(E)$ is uniformly bounded in D. Furthermore, if $\mathfrak{W}_0(E)$ is an operator-valued holomorphic function in D, then each element $\mathfrak{W}_n(E)$ is also, and it follows by a standard theorem of functional analysis[30] that the limit $\mathfrak{W}(E)$ is also analytic in D.

Now the arbitrariness of the domain D, the fact that $\mathfrak{W}(E) \in \mathfrak{D}_-$ for any $E \in \mathfrak{g}_+$, and the symmetry of the entire argument under $E \to E^*$ ensure that all of the requirements of Herglotzicity are satisfied by the solution $\mathfrak{W}(E)$. The pattern of argument leading to Eq. (B.38) can be applied almost exactly to Eq. (B.6) for the coherent potential W itself, so that

$$\mathrm{Im}W = \bar{\chi}(\mathrm{Im}\mathcal{V})\chi - \bar{u}\bar{\mathcal{V}}\mathcal{G}\,\mathrm{Im}(L_0 - \mathcal{W})\mathcal{G}\mathcal{V}u \qquad \text{(B.81)}$$

$$\leq 0\,, \qquad \text{(B.82)}$$

where

$$\chi = (1 + \mathcal{G}\mathcal{V})u\,, \qquad \text{(B.83)}$$

thus assuring that $W(E)$ is Herglotz if $\mathcal{W}(E)$ is. This completes the proof.

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