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Weighted two-particle Green's functions in the coherent-potential approximation

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We extend the two-particle theory of disordered systems within the coherent-potential approximation to obtain weighted contributions to averaged two-particle resolvents which arise from separate alloy components. Starting from first principles in a model of diagonal disorder and the single-site approximation for a binary substitutional alloy A_cB_{1-c} we extend the approach of a fundamental paper by Velický to evaluate various weighted forms of a general class of two-particle Green's functions. Applications in a wide range of linear response theory are discussed in detail as well as the behavior of the weighted functions in a strong disorder limit. To exemplify our analytic calculations the optical absorption in a disordered model alloy is studied numerically.

I. INTRODUCTION

In recent years the understanding of the effects of disorder in the physics of metals, semiconductors, and amorphous systems has made a tremendous progress. This vigorous development was motivated to a great extent by a thorough understanding of how strongly disorder effects determine the behavior of real physical systems. The success of the various analytical descriptions which have been considered, however, has always been decided by the relative simplicity to which approximations could be reduced, in order to keep the theory tractable analytically, without losing its capability to account for the most important physical aspects. One of the most successful approximations to match these requirements has been the coherent-potential approximation (CPA) developed by Soven,[1] Taylor,[2] and extended by Leath,[3–5] Velický *et al.*,[6–9] and many others (see Ref. 10 for a review).

In contrast to other approximations for the procedure of configurational averaging in disordered systems, the CPA is capable of interpolating correctly between the limits of weak[11] and strong disorder[12] as well as low and high impurity concentrations. Therefore it is also able to predict accurately the formation of impurity bound states turning into split-off impurity bands as the impurity concentration is increased. The inclusion of more detailed knowledge of the electronic band structure and atomic pseudopotential shapes as in the Korringa-Kohn-Rostoker (KKR) and tight-binding (TB) linear muffin-tin orbital (LMTO) CPA's (Refs. 13–17) as well as extensions of the single-site CPA to include scattering from clusters of impurities[18] remain the only existing theories that allow for a calculation of the density of states in disordered systems such that a reasonable verisimilitude is attained. The main difficulty with the CPA is the relative complexity of the self-consistent equations which have to be solved in more accurate extensions of the theory.

In the present paper we propose a more differentiated analysis of two-particle properties within the CPA building on the single-site approximation (SSA) in which it was developed originally. Our treatment should be especially useful for applications to binary substitutional alloys in all regimes of disorder. Following an earlier paper by Velický,[7] in which the regular two-particle theory within the CPA was introduced and which will be referred to hereafter as I, the theory of weighted single-particle resolvents, discussed originally for the case of lattice vibrations,[19] is extended to a properly weighted two-particle theory. Weighting of a Green's function in this context means that through the application of appropriate operators to the unaveraged resolvent, restrictions are made on the type of alloy component of either or both of the sites on which the particle starts and terminates its motion. Upon averaging, this results in a statistical weight being attributed to the averaged unrestricted resolvent with possibly separate additive terms.

On the average, therefore, contributions to a full two-particle Green's function in terms of constituent components can be resolved. This allows for a better understanding of how several physical processes contribute to a cumulative behavior as the principal parameters of the system are varied and it can therefore be used in an analysis of further effects which differentiates between these components. In more recent work, restricted single-particle Green's functions have been used in the KKR and TB-LMTO versions of the CPA,[20–23] whereby it was notable that in some of the papers which also dealt with two-particle aspects single-particle decouplings or other forms of approximation had to be used as the properly weighted two-particle averages were not available.[22,24]

We have structured the paper as follows: In Sec. II the important features of the single-particle CPA for diagonal disorder are recapitulated and relations which are important to the calculation of the corresponding two-particle Green's functions are established. In Sec. III and the Appendix we calculate the Fourier transforms of a class of weighted two-particle functions which are kept as general as possible. For that reason only a representative choice of weightings are calculated explicitly, since other weights can be obtained in an analogous fashion. Section IV is devoted to possible applications of weighted two-particle functions in linear response theory, and the peculiarities of two different classes of such functions are discussed in detail, which we have selected to cover a large range of conceivable applications.

In Sec. IV C the behavior of one class of functions discussed before is examined in a split band limit (strong dis-

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order). Section V is devoted to a numerical study of the splitting into several components of an interband absorption spectrum of a disordered binary alloy. Section VI in conclusion discusses the implications and possible further applications of the results obtained throughout this work.

II. SINGLE-PARTICLE PROPERTIES

We consider a binary substitutional alloy A_cB_{1-c} on a simple totally disordered monoatomic lattice on which each site is occupied by either an A or a B component with probabilities c and $1-c$, respectively. By symmetry it is only necessary to consider concentrations between $0 \leqslant c \leqslant 0.5$, and therefore we restrict our investigation to this region and define A as the impurity and B as the host component of the alloy. Since it is likely that this theory will find most applications in the calculation of electronic properties of binary solid solutions, we therefore cast the problem into a notation appropriate for this case, although, as has been stressed by Ref. 10, the algebra is essentially the same for all elementary excitations. Although it has proved possible in the past to also formulate the CPA for special cases of off-diagonal disorder, the only analytical solutions are obtained for multiplicative off-diagonal disorder,[21,22,25] where the problem is effectively solved by casting it into a model, where it can be treated in an essentially equivalent way as for the diagonal disorder case. We assume that the same can be achieved in the present problem, and in order to make the calculation of the weights and their properties more tractable and transparent, we shall confine our treatment to diagonal disorder and the single-site approximation from the outset.

We assume a Hamiltonian of the form

$$H = H_0 + U = H_0 + \sum_n U_n . \tag{1}$$

H_0 represents the periodic part and the U_n are single-site contributions to the disorder potential U. We assume that

$$H_0 = \sum_{\alpha,k} \varepsilon_\alpha(k) c_\alpha^\dagger(k) c_\alpha(k), \tag{2}$$

where the sum is over a series of bands or channels α and k values spanning the first Brillouin zone. It is often convenient to view the problem in a tight-binding approximation where the bands are built from a restricted set of orbitals s, centered on each site n of the medium, so that

$$H_0 = \sum_{n,n',s,s'} w_{s,s'}(n-n') c_s^\dagger(n) c_{s'}(n'). \tag{3}$$

Equally it is assumed that the single-site disorder potential has a similar structure

$$U_n = \sum_{n,s,s'} u_{s,s'}(n) c_s^\dagger(n) c_{s'}(n), \tag{4}$$

where $u_{s,s'}(n)$ assumes either the value $u_{s,s'}^A(n) = v_{s,s'}$ or $u_{s,s'}^B(n) = 0$, such that $U_n^A = V$, with $V = \Sigma_{n,s,s'} v_{s,s'} c_s^\dagger(n) c_{s'}(n)$, and $U_n^B = 0$ with probabilities c and $1-c$, depending on whether the site n is occupied by an A or B atom. In principle one could also have introduced a symmetric model for the disorder, but as it turns out the amount of algebra is somewhat reduced by the asymmetric definition, while switching from one form to the other does not present any difficulty. In general it may be convenient to diagonalize at least one, the periodic band structure as in (2) or the disorder potential, with respect to the orbital indices or bands, but usually it is impossible to diagonalize them both simultaneously. In the following formalism we therefore retain the most general form and all quantities should be regarded as matrices with site and orbital indices.

The propagator of the disordered medium G relates to the one of a pure host medium (pure B phase) g through the Dyson equation

$$G = g + gUG = g + gUg + gUGUg, \tag{5}$$

where summations over all occurring indices are implied as matrix multiplications. The CPA for a disordered medium is introduced by the usual method[1,2] of placing the impurities in a self-consistent medium such that the averaged propagator of this effective medium fulfills the relation

$$\bar{G} = g + g\Sigma\bar{G} = g + \bar{G}\Sigma g, \tag{6}$$

where Σ is the CPA single-particle self-energy. Σ itself is determined by the self-consistency condition that the average of the total scattering of a particle in the effective medium be zero. This total scattering is described by the equation

$$G = \bar{G} + \bar{G}T\bar{G}, \tag{7}$$

where T is corresponding T matrix of the problem and the self-consistency condition is therefore $\langle T \rangle \equiv 0$. In the SSA an additional requirement is made through the condition that the total scattering off a single site n be zero. This scattering is described by the single-site contribution to the T matrix T_n which is defined as

$$T_n = (U_n - \Sigma_n)[1 + FT_n], \tag{8}$$

where F is the site-diagonal average propagator and U_n and Σ_n are the single-site decompositions of U and Σ. The average of the disorder potential U alone amounts to $\langle U \rangle = cV$. This along with the average of (8) set to zero determines Σ to be

$$\Sigma = [1 - (V - \Sigma)F]^{-1} cV. \tag{9}$$

One can now define conditional or weighted propagators[19] which explicitly describe the propagation of a particle between partly or completely specified types of sites by multiplying them with a normalized version of the random potential. For example,

$$G^{i0} = V^{-1}UG, \quad G^{0i} = GUV^{-1} \tag{10}$$

describe the motion of a particle commencing on an impurity site and ending at an arbitrary other site in the medium (and vice versa), since U will be zero if the first site of the function is a host. Similarly,

$$G^{ii} = V^{-1}UGUV^{-1} \tag{11}$$

describes a situation where both sites are required to be impurities for the function not to be zero. Upon averaging over all configurations, the Green's functions become translationally invariant and the following relationships between the averaged weighted and unweighted functions can be obtained:

$$\bar{G}^{i0}=V^{-1}\Sigma\bar{G}, \quad \bar{G}^{0i}=\bar{G}\Sigma V^{-1}, \tag{12}$$

$$\bar{G}^{ii}=V^{-1}\{\Sigma\bar{G}\Sigma+(\Sigma-cV)\}V^{-1}. \tag{13}$$

The second term corrects the site-diagonal elements when $\bar{G}^{ii}=\bar{G}^{i}$ and uses identity (9). From here on, other weighted functions can be calculated by probability conservation. It is found that

$$\bar{G}^{h0}=\bar{G}-\bar{G}^{i}=(1-V^{-1}\Sigma)\bar{G}, \quad \bar{G}^{0h}=\bar{G}(1-\Sigma V^{-1}), \tag{14}$$

$$\bar{G}^{ih}=\bar{G}^{i0}-\bar{G}^{ii}=V^{-1}\{\Sigma\bar{G}(V-\Sigma)-(\Sigma-cV)\}V^{-1}, \tag{15}$$

$$\bar{G}^{hi}=\bar{G}^{0i}-\bar{G}^{ii}=V^{-1}\{(V-\Sigma)\bar{G}\Sigma-(\Sigma-cV)\}V^{-1}, \tag{16}$$

$$\bar{G}^{hh}=\bar{G}^{0h}-\bar{G}^{ih}=V^{-1}\{(V-\Sigma)\bar{G}(V-\Sigma)+(\Sigma-cV)\}V^{-1}. \tag{17}$$

It is our goal in the present paper to establish the two-particle analogs of these weighted Green's functions, i.e., jointly averaged products of such functions including the coherent scattering which induces correlations in the joint propagation of two particles.

In order to deduce these jointly averaged weighted functions, we first need to obtain the nonaveraged weighted functions, starting with G^{i0}, G^{0i}, and G^{ii}, in a representation such that no products between the disorder potential U and the unweighted single-particle function G occur. To accomplish this, one can simply employ Eq. (5) which yields

$$G^{i0}=V^{-1}(g^{-1}G-1), \quad G^{0i}=(Gg^{-1}-1)V^{-1}, \tag{18}$$

$$G^{ii}=V^{-1}[g^{-1}Gg^{-1}-g^{-1}-U]V^{-1}, \tag{19}$$

$$\tag{20}$$

and by means of (7), these go over to

$$G^{i0}=V^{-1}[g^{-1}\bar{G}+g^{-1}\bar{G}T\bar{G}-1], \quad G^{0i}=[\bar{G}g^{-1}+\bar{G}T\bar{G}g^{-1}-1]V^{-1}, \tag{21}$$

$$G^{ii}=V^{-1}[g^{-1}\bar{G}g^{-1}+g^{-1}\bar{G}T\bar{G}g^{-1}-g^{-1}-U]V^{-1}. \tag{22}$$

A further single-particle identity resulting from (6) which will prove to be very useful is

$$g^{-1}\bar{G}=\Sigma\bar{G}+1, \quad \bar{G}g^{-1}=\bar{G}\Sigma+1. \tag{23}$$

III. TWO-PARTICLE THEORY

A. Multichannel case

In this section a general weighted two-particle theory involving particles moving in two different subspaces a and b, which are possibly of different dimension, e.g., multiple conduction and valence bands of different angular momentum origin, is established. From this, results for the case of two particles moving in the same subspace can also be immediately obtained. The calculation for the unweighted two-particle function has been done in I. As a main result of that work the appropriate vertex corrections for the CPA were obtained which account for the coherent scattering processes of two particles that arise in the otherwise noninteracting two-particle function through the averaging process.

We follow the outline of Velický's reasoning to obtain the proper weights for two choices of the functions $\langle G_a^{\mu\nu}CG_b^{\mu'\nu'}\rangle$. The labels $\mu,\nu,\mu',\nu'\in\{i,h,0\}$ indicate the kind of weight—either an impurity, a host or no weight—which is attributed to the first and second sites of the respective function. C represents a generalized operator coupling the two single-particle functions and a and b label two possibly different bands on which the respective single-particle resolvents are defined. The positions of the weights within one single Green's function are thereby important, since the disordered medium before averaging is neither homogeneous nor isotropic and thus the nonaveraged Green's functions depend nontrivially on both arguments. In principle there would now be 80 possible ways of applying specific weights to these functions before averaging. However, in most cases, even if different bands are involved, only two-particle functions having an equal kind of weighting on its single-particle constituents will be needed in most applications. Later, we will consider two-particular examples of two-particle functions which find frequent use in linear response theory where the operator C is diagonal in real space and also only diagonal elements (or sums of diagonal elements) of the functions defined above are used. For both cases considered, the total choices of weightings reduce to only five different ones, since the first and the second sites of the first function will be the same as the second and first sites of the second function, respectively, which implies that they must also pairwise bear the same weighting label.

As an example we calculate for a most general choice of

C only the two functions $\langle G_a^{ii} C G_b^{ii}\rangle$ and $\langle G_a^{i0} C G_b^{0i}\rangle$ in this section, since they will prove to be the most useful types of weightings for the cases discussed thereafter and all other ones could be obtained in complete analogy to the calculations presented here.

Using the identities (21) and (22), the single-weighted two-particle function can be written in terms of the single-particle functions as

$$\langle G_a^{i0} C G_b^{0i}\rangle = V_a^{-1}\langle [g_a^{-1}\bar{G}_a + g_a^{-1}\bar{G}_a T_a \bar{G}_a - 1] C [\bar{G}_b T_b \bar{G}_b g_b^{-1} + \bar{G}_b g_b^{-1} - 1]\rangle V_b^{-1} \tag{24}$$

and the double-weighted one as

$$\langle G_a^{ii} C G_b^{ii}\rangle = V_a^{-1}\langle [(g_a^{-1}\bar{G}_a g_a^{-1} - g_a^{-1}) + g_a^{-1}\bar{G}_a T_a \bar{G}_a g_a^{-1} - U_a] C' [(g_b^{-1}\bar{G}_b g_b^{-1} - g_b^{-1}) + g_b^{-1}\bar{G}_b T_b \bar{G}_b g_b^{-1} - U_b]\rangle V_b^{-1}, \tag{25}$$

where for convenience we introduced $C' = V_a^{-1} C V_b^{-1}$ (note that C' would also be useful in the calculation of $\langle G^{0i} C G^{i0}\rangle$). We calculate the double-weighted function first, since it proves the more difficult task and from its solution it is straightforward to derive the single-weighted function as well. The problem of evaluating (25) is divided into two parts, the first one involving all terms not containing the matrix U, and the second one containing the remainder, i.e.,

$$V_a \langle G_a^{ii} C G_b^{ii}\rangle V_b = \mathscr{K} + \mathscr{M}, \tag{26}$$

$$\mathscr{K} = [g_a^{-1}\bar{G}_a g_a^{-1} - g_a^{-1}] C' [g_b^{-1}\bar{G}_b g_b^{-1} - g_b^{-1}] + g_a^{-1}\bar{G}_a \langle T_a \bar{G}_a g_a^{-1} C' g_b^{-1} \bar{G}_b T_b\rangle \bar{G}_b g_b^{-1}, \tag{27}$$

$$\mathscr{M} = \langle U_a C' U_b\rangle + [g_a^{-1} C' \langle U_b\rangle - g_a^{-1}\bar{G}_a g_a^{-1} C' \langle U_b\rangle - g_a^{-1}\bar{G}_a \langle T_a \bar{G}_a g_a^{-1} C' U_b\rangle + (a \leftrightarrow b)], \tag{28}$$

where in $\mathscr{K}$ the terms involving an average over a single T matrix have vanished, from the standard CPA condition, and $(a \leftrightarrow b)$ indicates that the labels are exchanged and the corresponding expressions reflected around the operator C'. We evaluate $\mathscr{K}$ first since it is the term needed in the wider range of applications.

With the identity (23) we find that

$$[g_a^{-1}\bar{G}_a g_a^{-1} - g_a^{-1}] C' [g_b^{-1}\bar{G}_b g_b^{-1} - g_b^{-1}] = g_a^{-1}\bar{G}_a \Sigma_a C \Sigma_b \bar{G}_b g_b^{-1}. \tag{29}$$

The T matrix can be decomposed into its single-site contributions T_n as

$$T = \sum_n T_n + \sum_{n\neq m} T_n \bar{G} T_m + \sum_{n\neq m\neq l} T_n \bar{G} T_m \bar{G} T_l + \cdots. \tag{30}$$

Thereby the characteristic exclusions in the sums prevent the particle from scattering twice in sequence on the same site and T_n satisfies Eq. (8).

As shown by Velický, T can then be replaced in two ways by a closed set of equations, namely,

$$T = \sum_n Q_n = \sum_n \tilde{Q}_n, \tag{31}$$

where

$$Q_n = T_n \left(1 + \bar{G} \sum_{n\neq m} Q_m\right) \tag{32}$$

and

$$\tilde{Q}_n = \left(1 + \sum_{n\neq m} \tilde{Q}_m \bar{G}\right) T_n. \tag{33}$$

Due to the requirement that $\langle T\rangle = 0$ and the single-site decomposition of T from (30) it is possible to decompose averages on different sites to give

$$0 \equiv \langle T\rangle = \sum_n \langle Q_n\rangle = \sum_n \langle T_n\rangle \left(1 + \bar{G} \sum_{m\neq n} \langle Q_m\rangle\right). \tag{34}$$

This also implies that $\langle Q_n\rangle = \langle \tilde{Q}_n\rangle = 0$. A vertex function Γ can now be defined similar to I such that

$$\mathscr{K} = g_a^{-1}\bar{G}_a (\Sigma_a C' \Sigma_b + \Gamma) \bar{G}_b g_b^{-1}, \tag{35}$$

where now

$$\Gamma = \langle T_a \bar{G}_a g_a^{-1} C' g_b^{-1} \bar{G}_b T_b\rangle. \tag{36}$$

Γ can be manipulated along the lines of I by using (31) to yield

$$\Gamma = \sum_n \sum_m \langle Q_n^a \bar{G}_a g_a^{-1} C' g_b^{-1} \bar{G}_b \tilde{Q}_m^b\rangle. \tag{37}$$

By means of (32)–(34) this can be cast into the form

$$\Gamma_n = \left\langle T_n^a \bar{G}_a g_a^{-1} \left(C' + g_a \sum_{p\neq n} \Gamma_p g_b\right) g_b^{-1} \bar{G}_b T_n^b \right\rangle, \tag{38}$$

where now $\Gamma = \Sigma_n \Gamma_n$, since from the decoupling introduced in (34) one gets $\langle Q_n^a \bar{G}_a g_a^{-1} C' g_b^{-1} \bar{G}_b \tilde{Q}_m^b\rangle = \langle Q_n^a \bar{G}_a g_a^{-1} C' g_b^{-1} \bar{G}_b \tilde{Q}_n^b\rangle \delta_{n,m}$. The only difference to the corresponding expression in I, [cf. (47) there] is that $\Sigma_{p\neq n}\Gamma_p$ is surrounded by the propagators of the pure medium $g_{a/b}$ here. We can then use (35) and (38) to obtain

$$\begin{aligned}\Gamma_n = {} & \langle T_n^a \bar{G}_a g_a^{-1} C' g_b^{-1} \bar{G}_b T_n^b\rangle + \langle T_n^a g_a \mathscr{K} g_b T_n^b\rangle \\ & - \langle T_n^a \bar{G}_a \Sigma_a C' \Sigma_b \bar{G}_b T_n^b\rangle - \langle T_n^a \bar{G}_a \Gamma_n \bar{G}_b T_n^b\rangle.\end{aligned} \tag{39}$$

To this point our model describes a situation where every occurring quantity in each subspace a and b is a matrix in a pair of channel (orbital or angular momentum) indices, as in the Hamiltonian (1)–(4). However, a solution of Eq. (39) for this general case is quite involved and not easy to handle for further applications. We shall therefore assume for the further calculations in this section that all Green's functions and the disorder potential are scalars in the channel indices. On the other hand we believe that the solution of the general problem could be of great potential interest to a whole spectrum of work, which uses a multichannel CPA as in the KKR-CPA or LMTO-CPA methods.[20–23] The only work known to us which addressed a simple version of the multichannel problem so far is a paper by Sen[26] where a case of band mixing was considered in the calculations of electrical conductivities. Although Sen seems to have solved the corresponding matrix vertex equations at least in part for his numerical calculations, the excerpts of the analytic solution given in his work do not seem to be very general. A proper formal solution of the most general multichannel problem is therefore given in the Appendix.

B. Scalar theory

With the restriction to scalar quantities (with respect to the channel indices) Γ_n and therefore also $\mathcal{K}$ are found from (39) without difficulty. Note that we are now able to shift site-diagonal expressions like Σ and F at our convenience within the products. As we cast our model into a site representation we obtain $T_n = |n\rangle t_n \langle n|$ and hence

$$\Gamma_n = |n\rangle \Lambda \langle n| [g_a \mathcal{K} g_b + \bar{G}_a g_a^{-1} C' g_b^{-1} \bar{G}_b - \Sigma_a \Sigma_b \bar{G}_a C' \bar{G}_b] |n\rangle \langle n|, \tag{40}$$

where Λ is the irreducible vertex part derived by Velický,

$$\Lambda(z_1, z_2) = \frac{\langle t_n^a(z_1) t_n^b(z_2) \rangle}{1 + F_a(z_1) \langle t_n^a(z_1) t_n^b(z_2) \rangle F_b(z_2)}. \tag{41}$$

The vertex Λ can be regarded as being intrinsic to the CPA, since it does not depend on the particular form of the operator C'. Substituting (40) into (35) yields

$$\mathcal{K} = \Sigma_a \Sigma_b g_a^{-1} \bar{G}_a C' \bar{G}_b g_b^{-1} + \Lambda g_a^{-1} \bar{G}_a \sum_n |n\rangle \langle n| [g_a \mathcal{K} g_b + \bar{G}_a g_a^{-1} C' g_b^{-1} \bar{G}_b - \Sigma_a \Sigma_b \bar{G}_a C' \bar{G}_b] |n\rangle \langle n| \bar{G}_b g_b^{-1}. \tag{42}$$

Multiplying by g_a and g_b from the left and right, we solve for the diagonal elements $\langle m| g_a \mathcal{K} g_b |m\rangle$,

$$\langle m| g_a \mathcal{K} g_b |m\rangle = \Sigma_a \Sigma_b \langle m| \bar{G}_a C' \bar{G}_b |m\rangle + \Lambda \sum_n \langle m| \bar{G}_a |n\rangle \langle n| [g_a \mathcal{K} g_b + \bar{G}_a g_a^{-1} C' g_b^{-1} \bar{G}_b - \Sigma_a \Sigma_b \bar{G}_a C' \bar{G}_b] |n\rangle \langle n| \bar{G}_b |m\rangle, \tag{43}$$

and hence also solve (42). At this point it is helpful to visualize the form of the operator C', in the site representation, which in the most general case can be written as

$$C' = (V_a V_b)^{-1} \sum_{l,m} \gamma_{lm} |l\rangle \langle m|. \tag{44}$$

For convenience we introduce the short notation

$$F_{n-m} = \langle n| \bar{G} |m\rangle, \tag{45}$$

$$\mathcal{F}_{n-m} = \langle n| g^{-1} \bar{G} |m\rangle, \tag{46}$$

where $F_0 \equiv F$ as already defined in (9). It should be noted that F_{n-m} and $\mathcal{F}_{n-m}$ are of different dimensions and their definition has been chosen to reduce the algebra as much as possible. Furthermore, these expressions show that Eq. (43) only contains translationally invariant quantities and hence it can be solved by Fourier transformation. The following Fourier transforms are introduced:

$$a_k = \Sigma_a \Sigma_b \sum_m e^{-ikR_m} \langle m| \bar{G}_a C' \bar{G}_b |m\rangle, \tag{47}$$

$$\begin{aligned} \alpha_k &= \sum_m e^{-ikR_m} \langle m| g_a^{-1} \bar{G}_a C' \bar{G}_b g_b^{-1} |m\rangle \\ &= \sum_m e^{-ikR_m} \Big\{ \Sigma_a \Sigma_b \langle m| \bar{G}_a C' \bar{G}_b |m\rangle + V_a^{-1} V_b^{-1} \\ &\quad \times \Big[\gamma_{mm} + \Sigma_a \sum_n \gamma_{nm} F^a_{m-n} + \Sigma_b \sum_n \gamma_{mn} F^b_{n-m} \Big] \Big\}, \end{aligned} \tag{48}$$

$$A_k = \sum_m e^{-ikR_m} F^a_m F^b_{-m}, \tag{49}$$

$$\mathcal{A}_k = \sum_m e^{-ikR_m} \mathcal{F}^a_m \mathcal{F}^b_{-m}, \tag{50}$$

$$b_k = \sum_m e^{-ikR_m} \langle m| g_a \mathcal{K} g_b |m\rangle, \tag{51}$$

$$c_k = \sum_m e^{-ikR_m} \langle m| \mathcal{K} |m\rangle. \tag{52}$$

The units of $\mathcal{A}_k$ and c_k are unity, and those of a_k, α_k, b_k, and A_k are J^{-2}. Inserting into (43) yields

$$b_k=\frac{a_k+\Lambda A_k(\alpha_k-a_k)}{1-\Lambda A_k}, \tag{53}$$

and hence by means of (42)

$$c_k=\alpha_k\frac{\Sigma_a\Sigma_b+\Lambda(\mathscr{A}_k-\Sigma_a\Sigma_b A_k)}{1-\Lambda A_k}=\alpha_k\frac{\Sigma_a\Sigma_b+\Lambda D}{1-\Lambda A_k}, \tag{54}$$

where

$$D=\mathscr{A}_k-\Sigma_a\Sigma_b A_k=[1+\Sigma_a F_a+\Sigma_b F_b], \tag{55}$$

where D is independent of k.

Similarly, one can evaluate the term $\mathscr{M}$ from (28), but it turns out that little simplification can be made until an explicit form of C is known and therefore we postpone its evaluation to the next section.

For now, we proceed to calculate the single weighted function $\langle G_a^{i0}CG_b^{0i}\rangle$. From (24) we get

$$\langle G_a^{i0}CG_b^{0i}\rangle=g_a^{-1}\bar{G}_aC'\bar{G}_bg_b^{-1}+C'-g_a^{-1}\bar{G}_aC' \\ -C'\bar{G}_bg_b^{-1}+g_a^{-1}\bar{G}_a\langle T_a\bar{G}_aC'\bar{G}_bT_b\rangle\bar{G}_bg_b^{-1}. \tag{56}$$

Using (23), this can be recast into

$$\langle G_a^{i0}CG_b^{0i}\rangle=\Sigma_a\Sigma_b\bar{G}_aC'\bar{G}_b \\ +g_a^{-1}\bar{G}_a\langle T_a\bar{G}_aC'\bar{G}_bT_b\rangle\bar{G}_bg_b^{-1}, \tag{57}$$

which can be readily solved since the term $\langle T_a\bar{G}_aC\bar{G}_bT_b\rangle$ exactly corresponds to the vertex part of the unweighted function which is known from I,

$$\langle T_a\bar{G}_aC\bar{G}_bT_b\rangle=\sum_n |n\rangle\Lambda\langle n|\langle G_aCG_b\rangle|n\rangle\langle n|. \tag{58}$$

Thus, the Fourier transform

$$d_k=\sum_m e^{ikR_m}\langle m|\langle G_a^{i0}CG_b^{0i}\rangle|m\rangle \tag{59}$$

can be obtained using the solution for the unweighted two particle function and Eqs. (47), (49), and (50) to yield

$$d_k=a_k+\Lambda\mathscr{A}_k\frac{a_k}{\Sigma_a\Sigma_b(1-\Lambda A_k)}. \tag{60}$$

Using (55) this can be written as

$$d_k=\frac{a_k}{\Sigma_a\Sigma_b}\,\frac{\Sigma_a\Sigma_b+\Lambda D}{1-\Lambda A_k}. \tag{61}$$

It is important to indicate at this point that the solution for a single-weighted function in which the weights have been swapped to the inside, i.e., $\langle G_a^{0i}CG_b^{i0}\rangle$, will not be the same as the one just obtained. A similar type of calculation yields instead of (60)

$$d'_k=b_k, \tag{62}$$

where $d'_k=\Sigma_n e^{ikR_m}\langle m|\langle G_a^{0i}CG_b^{i0}\rangle|m\rangle$.

Since we have now obtained expressions for the Fourier-transformed site-diagonal elements of some of the impurity-weighted two-particle functions, we are also able to find their respective off-diagonal parts by inserting into the appropriate Bethe-Salpeter equations [(42) in the case of the double-weighted function]. Other types of weighted functions can be calculated from here by setting up the corresponding equations in analogy to (24) and (25) from the single-particle functions. In particular, to obtain the corresponding host-weighted functions one can apply the weights $(1-U_{a/b}/V_{a/b})$ to the single-particle resolvents from either side, but the form of this weight already suggests that host-weighted properties can always be deduced by adding or subtracting corresponding unweighted and impurity-weighted functions. For the most general case the set of closed equations will be quite large and for that reason we will in the following deduce these equations only in some more specific cases.

IV. APPLICATIONS IN LINEAR RESPONSE THEORY

The most obvious application of a two-particle theory as introduced above is in linear response theory, i.e., in calculating weighted susceptibilities. The study of weighted functions in this context helps to determine how such quantities as susceptibilities and transport coefficients are constituted (on the average) from processes on different components of the alloy. A further advantage of such a differentiation is that it allows for a more refined treatment of further renormalizations to the considered quantities once further interactions are introduced into the problem. The standard expression employed in linear response theory is a generalized Kubo formula

$$\chi_{C^{(1)},C^{(2)}}(z)=\int d\xi\, d\eta\, S_0(z,\xi,\eta)Z^{-1}\mathrm{Tr}_b\{e^{-\beta H}C^{\dagger(2)} \\ \times\langle\hat{\delta}^{\mu\nu}(\xi-H_a)C^{(1)}\hat{\delta}^{\mu\nu}(\eta-H_b)\rangle\}. \tag{63}$$

Thereby $\chi_{C^{(1)},C^{(2)}}$ stands for a generalized type of susceptibility characterizing the linear response of an observable $C^{(1)}$ to an external perturbation coupling into the Hamiltonian through the operator $C^{(2)}$. S_0 denotes the zero-order associated two-particle Matsubara function in the Lehmann representation[27] and $\hat{\delta}^{\mu\nu}(\xi-H_{a/b})$ is the corresponding spectral function of the full resolvent $G_{a/b}^{\mu\nu}(\xi)=\Xi_{a/b}^{\mu}[\xi-H_{a/b}]^{-1}\Xi_{a/b}^{\nu}$, defined on the subspace (band) a/b, with the weighting operators $\Xi_{a/b}^{\mu}$ applied to it which assume the values $\Xi_{a/b}^{i}=U_{a/b}/V_{a/b}$, $\Xi_{a/b}^{h}=(1-U_{a/b}/V_{a/b})$, and $\Xi_{a/b}^{0}=1$. Tr_b denotes the trace over the subspace (band) b. Our particular interest focuses on the operators $C^{(1)}$ and $C^{(2)}$ which we now take to have the diagonal form

$$C^{(1)/(2)}=\sum_m \gamma_m^{(1)/(2)}(\xi,\eta)|m\rangle\langle m|. \tag{64}$$

To examine the behavior of the weighted functions further under such a constraint it is useful to consider the two cases

$$\gamma_m=\text{const}\times\delta_{m,r}, \tag{65}$$

where r is an explicitly specified site, and

$$\gamma_m=\text{const.} \tag{66}$$

The first case arises in a treatment of the response from local interactions such as for example the space-dependent exchange interaction between spin impurities (RKKY interaction[28]) submerged in a system otherwise containing nonmagnetic disorder.[29] The second case is widely used in calculations of the optical response in metals and semiconductors,[30,31] since the exciting optical fields can be taken as uniform over space with approximate momentum $q=0$ and the optical matrix elements describing transitions between bands of different angular momentum symmetry are usually well approximated as constants.

In both cases the restriction to only consider diagonal elements of two-particle functions reduces the set of closed equations for all possible weightings to

$$\langle G_a^h C G_b^h\rangle = \langle G_a C G_b\rangle - \langle G_a^i C G_b^i\rangle, \tag{67}$$

$$\langle G_a^{ih} C G_b^{ih}\rangle = \langle G_a^{hi} C G_b^{hi}\rangle = \langle G_a^i C G_b^i\rangle - \langle G_a^{ii} C G_b^{ii}\rangle, \tag{68}$$

$$\langle G_a^{hh} C G_b^{hh}\rangle = \langle G_a^h C G_b^h\rangle - \langle G_a^{ih} C G_b^{ih}\rangle. \tag{69}$$

It follows that for this case the calculation of only $\langle G_a^{ii} C G_b^{ii}\rangle$ and $\langle G_a^i C G_b^i\rangle$ is sufficient to also obtain the remaining mixed and host-weighted functions. We start to consider the case of (65) first.

A. Electronic susceptibility

The response of a system at some point r_1 in space to a perturbation applied at point r_2 has a wide range of applicability. Disregarding the coupling constants which simply scale the result we assume that $C^{(1)}=|2\rangle\langle 2|$ and $C^{(2)}=|1\rangle\langle 1|$, where the choice of the origin is arbitrary. In a slightly different notation the calculation of the double-weighted function corresponds to evaluating $\langle G_a^{ii}(r_1,r_2;t)G_b^{ii}(r_2,r_1;t')\rangle$.

For the RKKY interaction mentioned before, which couples spins at a distance r_1-r_2 through the electronic spin susceptibility of electrons in the conduction band, the two-particle time-dependent function is to be Fourier transformed and to be taken at two identical single-particle energies.[28] If the spins are at impurity sites, the weighted functions must be used to describe the problem adequately. Earlier treatments[29] neglected the vertex corrections and it is proposed to investigate the effect of their inclusion in a separate publication.

In this as in other problems the self-interaction $r_1=r_2$ can be neglected for most purposes, which implies that it will be sufficient to calculate only the term $\mathscr{K}$ from (27), since all terms occurring in $\mathscr{M}$ from (28) will vanish for $r_1\neq r_2$, since the disorder potential U is a diagonal matrix which vanishes identically for off-diagonal terms. To clarify the meaning of the quantity which is obtained through this special choice of $C^{(1)}$ and $C^{(2)}$, we look at how the corresponding Bethe-Salpeter (BS) equation can be rewritten for the case of a unweighted two-particle Green's function as introduced in Eqs. (22) of Ref. 4. In the single-site approximation the BS equation for the unweighted function reads

$$\langle G_a(1,2)G_b(1,2)\rangle \equiv \langle G_{ab}^{(2)}(1,1;2,2)\rangle = \langle G_a(1,2)\rangle\langle G_b(1,2)\rangle + \Lambda \sum_n \langle G_a(1,n)\rangle\langle G_b(1,n)\rangle\langle G_{ab}^{(2)}(n,n;2,2)\rangle, \tag{70}$$

where we have rewritten

$$\langle 2|\bar{G}_a|1\rangle\langle 1|\bar{G}_b|2\rangle \equiv \bar{G}_a(1,2)\bar{G}_b(1,2), \tag{71}$$

and

$$\langle 2|\langle G_a|1\rangle\langle 1|G_b\rangle|2\rangle \equiv \langle G_{ab}^{(2)}(1,1;2,2)\rangle. \tag{72}$$

For the Fourier transforms from before (47)–(50) this amounts to

$$a_k = \frac{\Sigma_a \Sigma_b}{V_a V_b}\sum_m e^{-ikR_m}\bar{G}_a(m)\bar{G}_b(m), \tag{73}$$

$$\alpha_k V_a V_b = \sum_m e^{-ikR_m}\langle m|g_a^{-1}\bar{G}_a|1\rangle\langle 1|\bar{G}_b g_b^{-1}|m\rangle = \sum_m e^{-ikR_m}[\Sigma_a\Sigma_b\bar{G}_a(m)\bar{G}_b(m) + \delta_{m,1}(1+\Sigma_a F_a + \Sigma_b F_b)] = \mathscr{A}_k. \tag{74}$$

Using (73) and (74) in connection with (60) and (62) the result for the single-weighted functions is independent of whether the impurity weights are applied to the interior or to the exterior of the single-particle resolvents, such that $\langle G_a^{i0}(1,2)G_b^{0i}(1,2)\rangle = \langle G_a^{0i}(1,2)G_b^{i0}(1,2)\rangle$. Indeed it is not hard to show that other arbitrary distributions of two impurity weights also give the same results as long as there is one weight applied to each resolvent. The same is of course true for host related properties. Accordingly, b_k from (53) in the previous section goes over to

$$b_k = a_k \frac{\Sigma_a\Sigma_b + \Lambda V_a V_b(\alpha_k - a_k)}{\Sigma_a\Sigma_b - \Lambda V_a V_b a_k} \tag{75}$$

and similarly c_k from Eq. (54) to

$$c_k = \Sigma_a \Sigma_b \alpha_k \left(1 + \Lambda \frac{V_a V_b \alpha_k}{\Sigma_a \Sigma_b - \Lambda V_a V_b a_k} \right). \tag{76}$$

We remember that the Fourier transform of the unweighted two-particle Green's function is

$$\sum_{R_{1,2}} e^{ikR_{1,2}} \langle G_a(1,2) G_b(1,2) \rangle = \frac{a_k V_a V_b}{\Sigma_a \Sigma_b - \Lambda V_a V_b a_k}. \tag{77}$$

Phenomenologically, one can scrutinize the uncorrelated limit where $\Lambda \to 0$ for which in (76) the k-dependent part of c_k behaves as

$$\lim_{\Lambda \to 0} c_k = \Sigma_a \Sigma_b a_k + \cdots, \tag{78}$$

which is the correct limiting result for a product of two averaged double-impurity-weighted single-particle functions $\overline{G}_a^{ii}(1,2)\overline{G}_b^{ii}(1,2)$ or the weighted two-particle function without coherent corrections.

However, taking this limit is reasonable only in special cases since generally the vertex Λ depends on the self energy Σ. It will be appropriate to take in a weak disorder limit (virtual crystal limit) since for $V_{a/b} \to 0$ the CPA predicts that $\Sigma_{a/b} \to c V_{a/b}$ and $\Lambda \to c(1-c) V_a V_b$ which means that Λ approaches zero faster than $\Sigma_{a/b}$ in this limit.

One should also note at this point that although it is apparent that the term $(1 + \Sigma_a F_a + \Sigma_b F_b)$, which arises in the difference of the Fourier transforms α_k and a_k, originates from the difference in the diagonal parts of their respective real-space Green's functions, it cannot be neglected in this treatment. When Λ is finite, this term is multiplied with other k-dependent quantities and thus contributes to the off-diagonal elements as well. Written in terms of a_k, (76) can be cast into

$$c_k = \frac{a_k}{\Sigma_a \Sigma_b - \Lambda V_a V_b a_k} \left(\Sigma_a \Sigma_b + \frac{\Sigma_a \Sigma_b}{a_k V_a V_b} (1 + \Sigma_a F_a + \Sigma_b F_b) \right) [\Sigma_a \Sigma_b + \Lambda (1 + \Sigma_a F_a + \Sigma_a F_b)]. \tag{79}$$

If again diagonal terms in real space are neglected, i.e., k-independent quantities in k space, it can be shown that (79) can be cast into the compact form

$$\zeta_k = \frac{a_k}{\Sigma_a \Sigma_b - \Lambda V_a V_b a_k} [\Sigma_a \Sigma_b + \Lambda (1 + \Sigma_a F_a + \Sigma_b F_b)]^2, \tag{80}$$

where we have introduced $\zeta_k \equiv c_k + c_0$, whereby c_0 is independent of k. According to the definition of ζ_k, this finally relates the weighted off-diagonal real-space two-particle Green's function to the unweighted one as

$$\langle G_a^{ii}(1,2) G_b^{ii}(1,2) \rangle = \langle G_a(1,2) G_b(1,2) \rangle \frac{[\Sigma_a \Sigma_b + \Lambda (1 + \Sigma_a F_a + \Sigma_b F_b)]^2}{(V_a V_b)^2}. \tag{81}$$

Equally, the relation for the single-weighted function $\langle G_a^{i}(1,2) G_b^{i}(1,2) \rangle$ can be obtained almost immediately if (60) is modified for this choice of $C^{(1)}$ and $C^{(2)}$, which goes over to

$$d_k = a_k + \Lambda \alpha_k \frac{a_k V_a V_b}{\Sigma_a \Sigma_b - \Lambda V_a V_b a_k} = \frac{a_k}{\Sigma_a \Sigma_b - \Lambda V_a V_b a_k} [\Sigma_a \Sigma_b + \Lambda (1 + \Sigma_a F_a + \Sigma_b F_b)], \tag{82}$$

thus relating the real-space functions in this case as

$$\langle G_a^{i}(1,2) G_b^{i}(1,2) \rangle = \langle G_a(1,2) G_b(1,2) \rangle \frac{\Sigma_a \Sigma_b + \Lambda (1 + \Sigma_a F_a + \Sigma_b F_b)}{V_a V_b}, \tag{83}$$

where no diagonal contributions were omitted.

The fact that in the single-weighted case the same weighting factor occurs once as opposed to twice for the double-weighted one is structurally equivalent to the results for the single-particle theory. Although retrospectively that might not be surprising, it is quite interesting in view of the fact that for the most general case of the previous section the weighting factors in (54) and (61) almost look alike were it not for the difference in the prefactors, i.e., α_k in the former and $a_k / \Sigma_a \Sigma_b$ in the latter case.

In principle all other functions can be derived now using the set of relations (67)–(69). However, one can save a considerable amount of algebra by recalling the following relation which holds in the transition from impurity to host related properties in the CPA:

$$-\Sigma \to (V - \Sigma), \quad c \to (1-c). \tag{84}$$

Starting from the definition of the vertex Λ in (41) and the CPA condition $\langle T_n \rangle = 0$ one finds that

$$\Lambda = \left[\frac{1-c}{(V_a - \Sigma_a)(V_b - \Sigma_b)} + \frac{c}{\Sigma_a \Sigma_b} \right]^{-1}, \tag{85}$$

which in a single band case $(a=b)$ readily simplifies to the form first introduced by Leath,[4]

$$\Lambda_{a=b} = \frac{\delta \Sigma}{\delta F} = \frac{\Sigma (V - \Sigma)}{1 - (V - 2\Sigma) F}. \tag{86}$$

With the relation (9) between F and Σ the weighting factor for the single-weighted impurity function from (83) reduces to

$$\frac{\Sigma_a\Sigma_b+\Lambda(1+\Sigma_aF_a+\Sigma_bF_b)}{V_aV_b}=\frac{(1-c)\Lambda}{(V_a-\Sigma_a)(V_b-\Sigma_b)}. \tag{87}$$

Correspondingly, the factor for the double-weighted function is the square of this quantity. One can now employ (67) and (68) to find the weights for the single host- and the impurity-host-weighted functions, respectively. We find

$$\langle G_a^hG_b^h\rangle=\langle G_aG_b\rangle\frac{c\Lambda}{\Sigma_a\Sigma_b} \tag{88}$$

and

$$\langle G_a^{ih}G_b^{ih}\rangle=\langle G_aG_b\rangle\frac{c(1-c)\Lambda^2}{\Sigma_a\Sigma_b(V_a-\Sigma_a)(V_b-\Sigma_b)}, \tag{89}$$

which by means of (69) gives

$$\langle G_a^{hh}G_b^{hh}\rangle=\langle G_aG_b\rangle\left(\frac{c\Lambda}{\Sigma_a\Sigma_b}\right)^2. \tag{90}$$

Note that this could also have been expected from the transformation property (84). The impurity and host weights as represented in (87) and (88) are seen to be the two contributions summing to Λ^{-1} in (85). Thus, even though there are many different possible representations of the two-band vertex, the representation in (85) shows immediately that the transition from impurity to host properties leaves Λ invariant.

A notable feature about the weights calculated in this section is that they are independent of any wave vectors and only multiply the unweighted function as scalar energy-dependent factors. This is a direct consequence of the single-site approximation.

B. Theory of absorption

In this section we evaluate a form of the two-particle functions needed in the calculation of the linear response absorption in a disordered solid. We take the operators $C^{(1)}$ and $C^{(2)}$ in the characteristic form of dipole operators similar to (66). Furthermore, we assume that the dipole matrix elements are essentially constant, such that $C^{(1)/(2)}=\gamma^{(1)/(2)}\Sigma_m|m\rangle\langle m|$. This choice corresponds to applications for the description of processes involving transitions between bands of different angular momentum symmetry such as required by the selection rule for optical processes at zero total momentum.[4,11]

A special example of this case is the photoelectric effect studied in alloys as in Refs. 22,24. In the theoretical discussion there, it is noted that weighted susceptibilities were required. However, since the final continuum state of the transition is intrinsically nondisordered, the weighting reduces to single-particle weights for the valence bands. Furthermore, as also often pointed out in this context, it is possible that the optical matrix elements are different for different types of atoms.[22,24,32] Such a situation would make the operators $C^{(1)}$, $C^{(2)}$ random as well, but as it turns out, the weighted two-particle functions are an excellent tool for dealing with such a situation. More about this is postponed to the numerical section (Sec. V) [cf. (131) there]. At the end of this subsection we will also give for completeness a short account of the calculation of linear response conductivities in disordered solids.

For the above choice of $\gamma^{(1)/(2)}$ the calculation of the term $\mathscr{K}$ in (42) is greatly simplified compared to before, but it will be necessary now to also consider total diagonal terms, since the sums over all states in $C^{(1)/(2)}$ couple all sites and hence all contributions coming from term $\mathscr{M}$ in (28) have to be included. As a consequence of the introduction of the dipole operator the main change arising in the result for $\mathscr{K}$ is that the site-diagonal elements $\langle n|\mathscr{K}|n\rangle$ and $\langle n|G_aC^{(1)}G_b|n\rangle$ as well as $\langle n|G_ag_a^{-1}C^{(1)}g_b^{-1}G_b|n\rangle$ are now actually independent of n.[30] Since $C^{(1)}$ now couples the functions to its left and right like a matrix product, the BS equations (42) and (43) have a very simple solution in terms of their Fourier transforms. Introducing $a,b,c\equiv a_k,b_k,c_k|_{k=0}$ as the zero-momentum elements of the respective transforms from last section, we find $c=\langle n|\mathscr{K}|n\rangle$, $a=\Sigma_a\Sigma_bV_a^{-1}V_b^{-1}\langle n|\bar{G}_a\bar{G}_b|n\rangle$, where the omission of $C^{(1)}$ indicates that the two single-particle resolvents are now simply multiplied as matrices. Equation (53) now reduces to

$$b=a\frac{\Sigma_a\Sigma_b+\Lambda V_aV_b(\alpha-a)}{\Sigma_a\Sigma_b-\Lambda V_aV_ba} \tag{91}$$

and Eq. (54) to

$$c=\Sigma_a\Sigma_b\alpha\left(1+\Lambda\frac{\alpha V_aV_b}{\Sigma_a\Sigma_b-\Lambda V_aV_ba}\right), \tag{92}$$

which can be recast as

$$\begin{aligned}c=&\frac{a}{\Sigma_a\Sigma_b-\Lambda V_aV_ba}[\Sigma_a\Sigma_b+\Lambda(1+\Sigma_aF_a+\Sigma_bF_b)]^2\\&+(1+\Sigma_aF_a+\Sigma_bF_b)[\Sigma_a\Sigma_b+\Lambda(1+\Sigma_aF_a\\&+\Sigma_bF_b)]V_a^{-1}V_b^{-1},\end{aligned} \tag{93}$$

where the term independent of a which was discarded in the previous section has to be kept in this case since the contributions of the diagonal elements become important.

In calculating $\mathscr{M}$ one uses the fact that U is a matrix which just has a random occupation of its diagonal. The sum over all sites in the operators $C^{(1)}$ and $C^{(2)}$ will hence just pick out the sum of all total diagonal parts $\langle n|\mathscr{G}|n\rangle\times\langle n|U|n\rangle$ where $\mathscr{G}$ is a generalized product of several Green's functions in the same band (the case where U and $\mathscr{G}$ are swapped is analogous). From (28) we get immediately

$$\begin{aligned}\mathscr{M}=c+[&cg_a^{-1}V_a^{-1}-cg_a^{-1}\bar{G}_ag_a^{-1}V_a^{-1}\\&-g_a^{-1}\bar{G}_a\langle T_a\bar{G}_ag_a^{-1}U_b\rangle V_a^{-1}V_b^{-1}+(a\leftrightarrow b)],\end{aligned} \tag{94}$$

where the only term giving slight complications is $\langle T_a\bar{G}_ag_a^{-1}U_b\rangle$. However, by means of (23) we obtain

$$\langle T_a\bar{G}_ag_a^{-1}U_b\rangle=\Sigma_a\langle T_a\bar{G}_aU_b\rangle+\langle T_aU_b\rangle. \tag{95}$$

In this expression, the second term presents more complications, since for the first one we remember from (7) that

$$T\bar{G}=\bar{G}^{-1}G-1 \tag{96}$$

and $U_b=U_aV_b/V_a$ such that

$$\langle T_a\bar{G}_aU_b\rangle=\langle\bar{G}_a^{-1}G_aU_b\rangle-cV_b=\frac{\Sigma_aV_b}{V_a}-cV_b. \tag{97}$$

Subsequently, the second term can be decoupled by means of (31)–(33),

$$\langle T_aU_b\rangle=\sum_n\langle\tilde{Q}_n^aU_b\rangle=\sum_n\left\langle\left(1+\sum_{m\neq n}\tilde{Q}_m^a\bar{G}_a\right)T_n^aU_b\right\rangle. \tag{98}$$

Applying (34) and using the fact that U is diagonal yields

$$\langle\tilde{Q}_m^a\bar{G}_aT_n^aU_b\rangle=\langle\tilde{Q}_m^a\rangle\bar{G}_a\langle T_n^aU_b\rangle=0, \tag{99}$$

since $(m\neq n)$ and $\langle\tilde{Q}_m\rangle=0$, and hence we find

$$\langle T_aU_b\rangle=\sum_n\langle T_n^aU_n^b\rangle=\frac{c(V_a-\Sigma_a)V_b}{1-(V_a-\Sigma_a)F_a}. \tag{100}$$

Collecting all terms for $\mathscr{M}$ and some more algebraic manipulation finally yields

$$\mathscr{M}=c-(1+\Sigma_aF_a)\Sigma_aV_a^{-1}-(1+\Sigma_bF_b)\Sigma_bV_b^{-1}. \tag{101}$$

The sum of all diagonal parts, i.e., $\mathscr{M}$ and the ones from the second term in (93), can be shown to assume the very compact form

$$\begin{aligned}\mathscr{M}&+\frac{1+\Sigma_aF_a+\Sigma_bF_b}{V_aV_b}[\Sigma_a\Sigma_b+\Lambda(1+\Sigma_aF_a+\Sigma_bF_b)]\\&=\Lambda F_aF_b.\end{aligned} \tag{102}$$

The final result for $\langle G_a^{ii}G_b^{ii}\rangle$ thus amounts to

$$\langle G_a^{ii}G_b^{ii}\rangle=\langle G_aG_b\rangle\left[\frac{(1-c)\Lambda}{(V_a-\Sigma_a)(V_b-\Sigma_b)}\right]^2+\frac{\Lambda F_aF_b}{V_aV_b}. \tag{103}$$

Here the first term has been rewritten in the same way as already derived in the last subsection for the finite-range susceptibility. From there it is also seen that the single-weighted function will have the same weight as calculated in (87) for the corresponding function in the exchange coupling case. We find

$$\langle G_a^iG_b^i\rangle=\langle G_aG_b\rangle\frac{(1-c)\Lambda}{(V_a-\Sigma_a)(V_b-\Sigma_b)} \tag{104}$$

and by means of (67)–(69)

$$\langle G_a^hG_b^h\rangle=\langle G_aG_b\rangle\frac{c\Lambda}{\Sigma_a\Sigma_b}, \tag{105}$$

$$\langle G_a^{ih}G_b^{ih}\rangle=\langle G_aG_b\rangle\frac{c(1-c)\Lambda^2}{(V_a-\Sigma_a)(V_b-\Sigma_b)\Sigma_a\Sigma_b}-\frac{\Lambda F_aF_b}{V_aV_b}, \tag{106}$$

$$\langle G_a^{hh}G_b^{hh}\rangle=\langle G_aG_b\rangle\left[\frac{c\Lambda}{\Sigma_a\Sigma_b}\right]^2+\frac{\Lambda F_aF_b}{V_aV_b}. \tag{107}$$

Comparing the results of the last two subsections, it becomes clear that the several weights obtained are essentially universal. The main difference in the absorption case as compared to the susceptibility one comes from the diagonal terms which have to be kept in the double-weighted functions. The single-weighted analogs are void of this difficulty and the weighting factors are identical for both cases.

So far we have omitted to consider a form of the two-particle functions which is needed for conductivity calculations. However, Velický[7] showed for the unweighted functions that the vertex Γ vanishes in the single-band case in the corresponding expression for the conductivity, due to the antisymmetry of the dipole matrix elements in k space if they are taken between Bloch states of a noninteracting Fermi system in a crystal with inversion symmetry. The same is also true for the weighted case and effectively the weighted functions which would have to be used for such calculations would just consist of products of the corresponding single-particle quantities.

This aspect turns out to be a general deficiency of the CPA in the single-site approximation, since due to the multiple scattering exclusions only ladders of nested diagrams are used in calculating the total contribution of the coherent scattering. The CPA therefore neglects higher-order two-particle correlations which are in fact nonzero and contribute markedly to the conductivity. Langer and Neal[33] have shown that the so called "maximally crossed" diagrams, i.e., diagrams which have a maximal crossing of coherent particle-particle scattering lines, actually contribute the leading part—in the order of the expansion considered—to the full two-particle disorder vertex for the conductivity in an otherwise noninteracting system. There are, however, two cases, where even the CPA vertex gives a finite contribution to the conductivity. One is the case of interacting Fermi systems, where the presence of interactions is sufficient to destroy the aforementioned antisymmetry and thus also the terms already included in the vertex of a single-site two-particle CPA as discussed here will give a finite contribution to the conductivity in real systems. The other is the case of multiple conduction bands with band mixing, discussed by Sen,[26] where there is a finite contribution from the mixing part of the vertex.

C. Split band limit

As already indicated, in contrast to many other theories of disorder, the CPA interpolates correctly to the limits of strong disorder and high concentrations. In this situation each band splits into two components of strength c and $1-c$, respectively, which represent largely separate A- and B-type excitations. An intuitive consideration of the underlying physics in this limit suggests that the correct description of an absorption process should predict that the overlap integral for transitions between sites pertaining to different alloy components will gradually decrease and thus, in reverse, that transitions between sites of the same type will be more and more favored. In the following, we prove that the CPA of weighted two-particle functions predicts this behavior correctly which makes it useful for a better quantitative understanding of absorptive and dispersive processes in strongly disordered alloys. The corresponding single-particle theory must fail in this respect, since it will weigh transitions only with the products of concentrations of sites involved.

To illustrate this we assume that our material components A and B have corresponding single site energies ε_a^A, ε_b^A and ε_a^B, ε_b^B for the two bands, respectively, and the carriers have become totally localized; i.e., their effective mass goes to infinity or vice versa the bandwidths involved go to zero. The potentials V_a and V_b are then defined as

$$V_a=\varepsilon_a^A-\varepsilon_a^B, \quad V_b=\varepsilon_b^A-\varepsilon_b^B, \tag{108}$$

and the single-particle site-diagonal Green's functions go over to

$$F_\lambda(z)=\frac{c}{z-\varepsilon_\lambda^A}+\frac{1-c}{z-\varepsilon_\lambda^B}\equiv\frac{1}{z-\varepsilon_\lambda^B-\Sigma_\lambda}, \tag{109}$$

where λ labels the corresponding band. Thus, if a two-particle theory is constructed from single-particle properties only, and coherent terms in the two-particle scattering are neglected, this leads to peaks in the absorption spectrum as shown in Table I. The energies at which the peaks are centered are shown in row 2 and their relative weight for the uncorrelated average is shown in row 3.

TABLE I. Energies of poles at which interband transitions may occur versus center of gravity weights in an asymptotically strong disorder limit before and after the inclusion of vertex corrections.

Transition	$A_b\rightarrow A_a$	$B_b\rightarrow A_a$	$A_b\rightarrow B_a$	$B_b\rightarrow B_a$
Energy of pole	$\varepsilon_a^A-\varepsilon_b^A$	$\varepsilon_a^A-\varepsilon_b^B$	$\varepsilon_a^B-\varepsilon_b^A$	$\varepsilon_a^B-\varepsilon_b^B$
Weight without vertex	c^2	$c(1-c)$	$c(1-c)$	$(1-c)^2$
Weight including vertex	c	0	0	$1-c$

For this limit of strong disorder we find, upon introducing the vertex corrections in conjunction with the appropriate weighting factors for the two respective components, that the expected transitions are given correctly with their appropriate transition strengths and the spurious crossed terms are suppressed as shown in row 4 of Table I. Defining

$$z-\varepsilon_\lambda^B\equiv x, \tag{110}$$

F and Σ can be rewritten as

$$F(x)=\frac{1}{x-\Sigma}=\frac{x-(1-c)V}{x(x-V)}, \tag{111}$$

$$\Sigma(x)=\frac{cVx}{x-V(1-c)}, \tag{112}$$

and the vertex Λ given by (85) can be written

$$\Lambda=\frac{(1-c)cV_aV_bx_ax_b(x_a-V_a)(x_b-V_b)}{[x_a-V_a(1-c)][x_b-V_b(1-c)][cx_ax_b+(1-c)(x_a-V_a)(x_b-V_b)]}. \tag{113}$$

The impurity weight $\xi\equiv(1-c)\Lambda/(V_a-\Sigma_a)(V_b-\Sigma_b)$ can be represented as

$$\xi=\frac{cx_ax_b}{cx_ax_b+(1-c)(x_a-V_a)(x_b-V_b)} \tag{114}$$

and equally the host weight $\eta\equiv c\Lambda/\Sigma_a\Sigma_b$ as

$$\eta=\frac{(1-c)(x_a-V_a)(x_b-V_b)}{cx_ax_b+(1-c)(x_a-V_a)(x_b-V_b)}. \tag{115}$$

It is now evident that the impurity-weighted quantities are proportional to c and the host-weighted ones to $1-c$ and not the other way around as the appearance of the weighting factors, e.g., (104) and (105), might superficially suggest. The correction factor coming from the diagonal terms in the double-weighted functions can be recast into

$$\gamma\equiv\frac{\Lambda F_aF_b}{V_aV_b}=\frac{(1-c)c}{cx_ax_b+(1-c)(x_a-V_a)(x_b-V_b)}. \tag{116}$$

Furthermore, the unweighted but vertex corrected two-particle propagator $K=\langle GG\rangle$ assumes the form

$$K=\frac{cx_ax_b+(1-c)(x_a-V_a)(x_b-V_b)}{x_ax_b(x_a-V_a)(x_b-V_b)}. \tag{117}$$

After further algebra one can show, that in this limit the weighted functions can be expressed as

$$\langle G^{ii}G^{ii}\rangle=K\xi^2+\gamma=\langle G^iG^i\rangle=K\xi=\frac{c}{(x_a-V_a)(x_b-V_b)}, \tag{118}$$

$$\langle G^{hh}G^{hh}\rangle=K\eta^2+\gamma=\langle G^hG^h\rangle=K\eta=\frac{(1-c)}{x_ax_b}, \tag{119}$$

$$\langle G^{ih}G^{ih}\rangle=0, \tag{120}$$

which is exactly what is expected to happen physically in this limit. The crossed terms in the transition are canceled out—hence the crossed function in (120) goes to zero—and the double-weighted functions become identical to the single-weighted ones, since now only the totally site diagonal element $K_D\equiv\langle G^{\nu\mu}(l,l)G^{\nu\mu}(l,l)\rangle$ still contributes to the transitions, which implies that only two possibilities for weighting the two-particle functions remain, namely, as $\langle G_a^iG_b^i\rangle$ and $\langle G_a^hG_b^h\rangle$. As could be expected from a theory which properly describes the strong disorder limit the transition strengths now distribute with the concentrations c and $1-c$ between the $A_b \rightarrow A_a$ and $B_b \rightarrow B_a$ transitions, respectively, such as shown in row 4 of Table I. This feature may in reverse be used to derive the total diagonal element K_D for all ranges of disorder. By requiring $K_D^{ih}=0$ we find

$$K_D=\frac{\gamma}{\xi\eta}=\frac{F_aF_b}{V_aV_b}\left(\frac{\Sigma_a\Sigma_b}{c}+\frac{(V_a-\Sigma_a)(V_b-\Sigma_b)}{(1-c)}\right). \tag{121}$$

The weighted versions of this element are obtained by just multiplying the corresponding single weights from (104) and (105) onto it. Moreover, K_D is equivalent to the $r_{1,2}=0$ component of the two-particle function calculated for the finite-range susceptibility in the last subsection. In terms of the notation introduced there it reads

$$K_D=\sum_k \frac{a_kV_aV_b}{\Sigma_a\Sigma_b-\Lambda_{a,b}V_aV_ba_k}, \tag{122}$$

which would have been harder to evaluate starting from that representation. The total site-diagonal element thus decouples into the corresponding site-diagonal single-particle functions with an appropriate correction term accounting for the coherent processes.

V. NUMERICAL RESULTS

In this subsection, in order to exemplify the general results, we discuss numerical results obtained for the optical absorption in a binary semiconductor alloy with a completely filled valence and empty conduction band for a simple model density of states. We are thus able to show how a CPA type of polarization, including vertex corrections, decomposes into contributions originating from single-alloy components as the strength of the disorder is increased, thus eventually causing the joint density of states to split into several components (up to three different ones for the double-weighted case).

In all our calculations we have used a semi elliptic density of states for a pair of three-dimensional (3D) conduction and valence bands as introduced in Ref. 6 for the single particle CPA, i.e.,

$$\rho_\lambda(E)=\frac{2}{\pi w_\lambda^2}\sqrt{w_\lambda^2-E^2}, \quad |E|\leqslant w_\lambda,$$
$$\rho_\lambda(E)=0, \qquad |E|\leqslant w_\lambda, \tag{123}$$

where λ labels either the conduction $\lambda=a$ or valence $\lambda=b$, and w_λ is the half-width of the band considered. This has the advantage that the self-consistent CPA equation for the self energy $\Sigma(E)$ is a third-degree polynomial which can be solved analytically.

To understand the effects that arise from genuine two-particle behavior as compared to those expected from the single-particle CPA, we recapitulate some of the features of the single-particle theory first, mainly building on the treatment presented in Ref. 6. It is established there that depending on the concentration and disorder strength relative to the bandwidth, an impurity band is eventually split off while in this split regime under some circumstances the CPA self-energy exhibits a pole. Figure 1 shows a reproduction of the "phase" diagram first presented there, indicating how the several regions are separated. It can be seen that for a disorder strength $|V_\lambda/w_\lambda|>1$ the bands always split into A and B components, whereas the splitting occurs earlier as the concentration c is reduced, going down to $|V_\lambda/w_\lambda|>0.5$ as $c\rightarrow 0$.

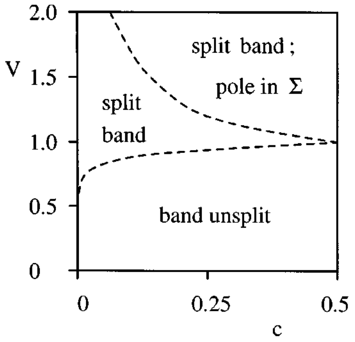

FIG. 1. Regimes for the CPA of the single-particle density of states (123) for $w_\lambda=1$ in a binary substitutional alloy depending on the impurity concentration c and the disorder strength V. In the lowest region the band is unsplit. Above the first dashed line the bands split into two components and above the second dashed line the CPA self-energy Σ exhibits a pole between these components.

We have calculated the linear polarizability of the medium by employing a Kubo formula as introduced in (63). Furthermore, we continue assume that the optical matrix elements are essentially constant and that such elements are the same for both alloy components and we hence normalize them to unity. The optical absorption is proportional to the negative imaginary part of the retarded polarizability of the disordered medium $-\mathrm{Im}\Pi(\omega)$, which can be formally written as

$$\Pi_r(\omega)=-\lim_{\substack{\beta\rightarrow\infty\\ i\omega\rightarrow\omega+i\delta}}\beta^{-1}\sum_{i\varepsilon}\int\frac{d^3k}{(2\pi)^3}$$
$$\times\langle G_a(k;i\varepsilon)G_b(-k;i\omega-i\varepsilon)\rangle, \tag{124}$$

whereby the k integration is understood to be carried out after the configurational average has been performed, since before that both resolvents would depend nontrivially on two momentum variables.

We consider our system at zero temperature and follow partly the method used in Ref. 30 for our calculations. At $T=0$, the polarization can be obtained as the energy convolution around the conduction band branch cut of the k-summed vertex-corrected two-particle function $K(z_1,z_2)=\int d^3k/(2\pi)^3\langle G_a(k;z_1)G_b(-k;z_2)\rangle$, such that

$$\Pi_r(\omega)=\oint_C K(z,\omega+i\delta-z)dz, \tag{125}$$

where we have taken over the following definitions from Ref. 30:

$$K(z_1,z_2)=\frac{R(z_1,z_2)}{1-\Lambda(z_1,z_2)R(z_1,z_2)}, \tag{126}$$

where $\Lambda(z_1,z_2)$ is the usual CPA vertex from (85) and

$$R(z_1,z_2)=\int\frac{d^3k}{(2\pi)^3}\,\bar{G}_a(k,z_1)\bar{G}_b(-k,z_2) \tag{127}$$

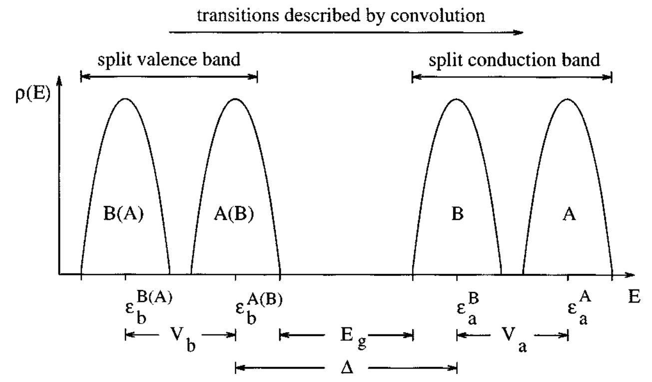

FIG. 2. Diagrammatic representation of the energy convolution in (125) effectively using the term $R(z_1,z_2)$ from (129) only. If R is taken at $z_1=z_2$ and c is about $c=0.5$, the conduction and valence bands are split and the imaginary part of R decomposes into four components centered about ε_a^A, ε_a^B and ε_b^A, ε_b^B as shown qualitatively. The components are separated by the band gap E_g in the middle and the single-particle splittings in the conduction and valence parts at the upper and lower ends, respectively. The energetic differences between the gravity centers are given by $V_a=\varepsilon_a^A-\varepsilon_a^B$ and $V_b=\varepsilon_b^A-\varepsilon_b^B$, as already introduced, as well as $\Delta\equiv\varepsilon_a^B-\varepsilon_b^{A(B)}$. Bracketed expressions denote antiparallel disorder.

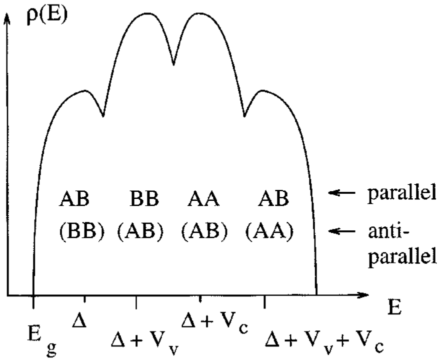

FIG. 3. Qualitative result of the convolution (125) as it can be expected if performed using the term $R(z_1,z_2)$ from (129) only. At about $c=0.5$ the joint density of states would be expected to distribute over regions in terms of their compositional origin as shown. The inclusion of the vertex is expected to primarily suppress the mixed AB transitions and favor the AA and BB transitions as the disorder strength is increased. For parallel disorder this leads to a suppression of the spectra at the flanks while for antiparallel disorder (in brackets) it causes a suppression of the central part. Both these features are well represented in the calculated spectra of Figs. 4 and 5.

is the average-decoupled two-particle function. Assuming that the conduction and valence band dispersion relations exhibit a similar shape such that they scale proportionally

$$\frac{\varepsilon_a(k)}{w_a}=\mp\frac{\varepsilon_b(k)}{w_b}, \tag{128}$$

(127) can be shown to simplify to

$$R(z_1,z_2)=\frac{w_aF_a(z_1)\pm w_bF_b(z_2)}{w_a[z_2-\Sigma_b(z_2)]\pm w_b[z_1-\Sigma_a(z_1)]}, \tag{129}$$

where $F_\lambda(z)$ are the site-diagonal single-particle functions first introduced in connection with (45). As usual we assume that the effective mass of an electron in the conduction band is positive and that of a hole in the valence band is negative. Accordingly we have chosen the upper choice of signs in (128) and (129) for our calculations.

To be able to analyze the obtained results with regard to the effect of the inclusion of vertex corrections we first consider qualitatively the features that would be expected from the transition process represented by the energy convolution in (125) in an intermediate regime of disorder, if the configurational average in the two-particle function is decoupled and effectively only single-particle properties are employed. This would correspond to replacing K from (126) by R from (127) in (125). We assume for now that the concentration is about 0.5 and the bands have just split by a finite amount. With the semielliptic bands used, the transition process can be represented as shown in Fig. 2.

The disorder strengths give approximately the distance between the centers of the single bands. The convolution of two separated finite bands, occurring in R, would yield a set of finite bands in the joint density of states (DOS) whose width is the sum of the widths of the contributing components. Two cases are considered where the band offsets of the A and B components of the alloy are in equal or opposite directions corresponding to parallel or antiparallel disorder.

In the case of parallel disorder, this would amount to the $A_b\rightarrow A_a$ and $B_b\rightarrow B_a$ transitions lying in the center of the joint DOS, framed by the contributions from the $B_b\rightarrow A_a$ and $A_b\rightarrow B_a$ transitions at the upper and lower ends, respectively, as shown in Fig. 3. At $c=0.5$ these regions would have relative distribution of weighted states of 1:2:1 from lower end : center : higher end. In the case of antiparallel disorder the picture should be similar with the only difference that the spectrum is turned inside out with the $B_b\rightarrow B_a$ and $A_b\rightarrow A_a$ components on the top and the bottom ends of the joint DOS and the mixed transitions in the center, again with a distribution of 1:2:1.

The calculations in the previous section for the split band case strongly suggest that the vertex corrections will increasingly suppress the cross transitions as the disorder strength is increased, which is verified in our numerical results. Indeed, our results show that this suppression is already displayed quite strongly in an intermediate disorder range, i.e., in a regime where the single bands just begin to split.

The plots in Figs. 4 and 5 show cumulative absorption spectra calculated from (125) as well as their single- and double-weighted components for parallel and antiparallel disorder of various strengths, covering both the joint and the split band regime. The conduction band half-width is normalized to unity and the valence band half-width is taken to be 0.8. The concentration of impurities is fixed to 0.35 in order to study the high-concentration behavior rather than dilute impurity effects.

In the transitional region when the disorder strengths start to exceed the single-particle half-bandwidths $|V_\lambda/w_\lambda|\geqslant 1$ and the conduction and valence bands start to split we observe the following behavior: In the case of parallel disorder shown in Fig. 4 the spectrum starts to exhibit a discontinuity

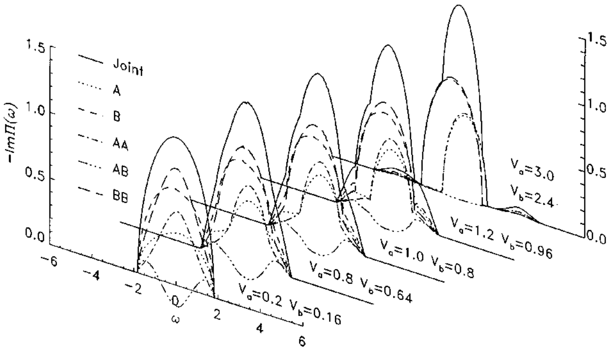

FIG. 4. Negative imaginary part of the polarizability of the disordered medium (joint DOS) for parallel disorder and its possible weightings into A and B or AA, AB, and BB components. The concentration of A atoms is fixed to $c=0.35$ and the band half-widths were taken to be $w_a=1.0$ and $w_b=0.8$ for the conduction and valence bands, respectively. The disorder was varied through a range of parameters, annotated on the bottom right of the plots, with particular focus on the splitting point $|V_\lambda/w_\lambda|=1$ of the single-particle bands. At the splitting point a pair of flank components starts separating sideways and gets completely isolated as $V_\lambda/w_\lambda \to 3$ in the last plot. These flanks are constituted of about 50% by the AB component, whereas the central region almost entirely consists of AA and BB components only.

in its derivatives at the flanks, accounting for a pair of mixed components splitting off sideways from the main contribution. At the transition point the contributions of the flanks relative to the central bulk part are approximately distributed in a ratio of 1:14:1. In the case of antiparallel disorder shown in Fig. 5 the suppression of the mixed transitions is even stronger so that as soon as the single bands split the crossed transitions cancel out entirely within the numerical accuracy and the joint density of states starts to exhibit a gap between two separate contributions which mainly consist of A and B transitions, respectively.

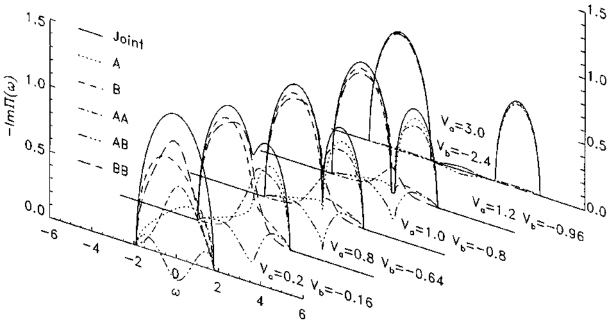

FIG. 5. Negative imaginary part of the polarizability for antiparallel disorder (reversed in the valence band) with otherwise equal model parameters to the parallel case shown in Fig. 4. The joint part starts to exhibit a gap as soon as the conduction and valence bands do. In the split regime the spectrum almost entirely builds up from single A and B components which in the interior of the two bands again coincide largely with the AA and BB components, indicating a strong breaking of the k-selection rule already in an intermediate disorder regime. In the last plot $V_a=3.0$, $V_b=-2.4$ it can be seen that the double-weighted functions are nonzero in the gap region. This means that in a case where the optical cross sections $\mu^{A/B}$ of the of A and B atoms in (131) are nonequal, the gap in the observed absorption spectrum would be less complete, containing either AA or BB states primarily.

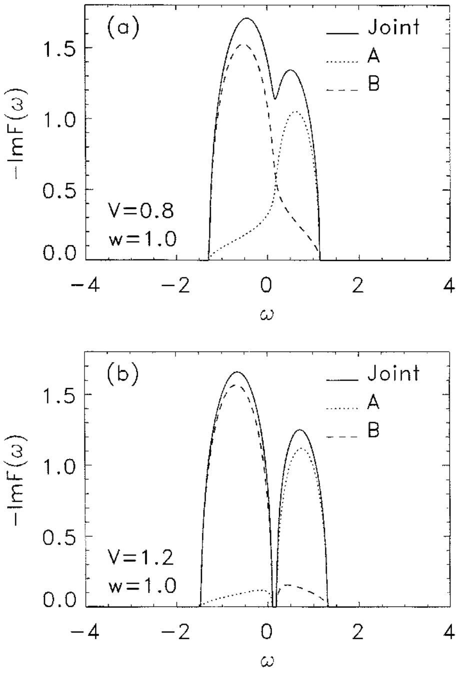

FIG. 6. Imaginary parts of the site-diagonal single-particle function F taken for the same concentration $c=0.35$, a band half-width of $w=1.0$, and disorder strengths of (a) $V=0.8$ and (b) $V=1.2$. By comparing the splitting behavior of this function with the one of the corresponding two-particle function for antiparallel disorder, shown in the third and fourth plots in Fig. 5, one finds that the behavior, particularly the one of the single-weighted A and B components, is strikingly similar.

In a regime of strong disorder, where there is a large splitting of the single-particle bands as shown in the last plots of Figs. 4 and 5, one finds that over large regions the single- and uniformly double-weighted contributions coincide almost exactly, implying that the spectrum is built almost entirely from the total diagonal element of the two-particle function, which we had calculated at the end of Sec. IV. The total diagonal element can be obtained as an independent k sum over the two single-particle resolvents involved. This situation represents a breakdown of the k-selection rule which holds in pure media.

If the splitting of Π into single-weighted components $\Pi=\Pi^A+\Pi^B$ is considered and the result is compared in appearance with the splitting into components of the site-diagonal single-particle function[6]

$$F_r(\omega)=\int \frac{d^3k}{(2\pi)^3}\bar{G}(k,\omega+i\delta), \qquad (130)$$

one finds that the single- and the two-particle behaviors appear to be strikingly similar in the case of antiparallel disorder, as can be seen if the plots for the imaginary part of the single-particle function shown in Figs. 6(a) and 6(b) are

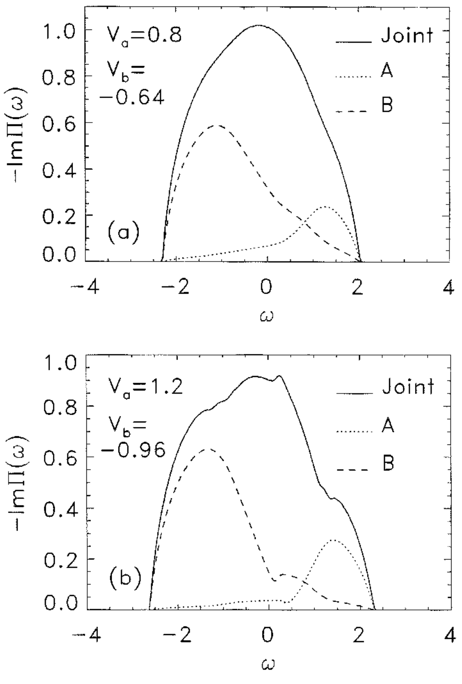

FIG. 7. Negative imaginary part of the polarizability without vertex corrections for antiparallel disorder. The parameters taken were (a) $c=0.35$, $V_a=0.8$, $V_b=-0.64$ and (b) $c=0.35$, $V_a=1.2$, $V_b=1.0$. The characteristic splitting of the joint DOS, which is displayed in the properly corrected function of Fig. 5 as the disorder strengths exceed the value $|V_\lambda/w_\lambda|=1$, does not occur and the shape of the joint DOS rather resembles the qualitative one of Fig. 3. The curves for the single-weighted components in both (a) and (b), which were obtained from correspondingly weighted uncoupled single-particle functions, do not satisfy the sum rule $\Pi=\Pi^A+\Pi^B$.

compared to the ones for the corresponding parameters in the two-particle case of Fig. 5 (third and fourth plots from the front).

In a system without disorder this similarity is evident if there are no further local interactions in the problem, since the noninteracting two-particle motion decouples into a center of mass and a relative coordinate, and while the center-of-mass motion can be set to zero, the relative one can be mapped onto a single-particle coordinate. Upon the addition of the disorder, this decoupling fails to work and it can only be regained by using an appropriate configurational averaging procedure. However, if in the presence of disorder an average is only performed on a single-particle level, thus omitting average induced two-particle correlations, the reduction obviously fails to work, as is seen through comparing the plots of Fig. 7 showing a spectrum calculated without the vertex corrections with the ones for the corresponding parameter values of Fig. 5, which properly include these corrections. The results show that in regimes of intermediate and strong disorder the influence of the vertex corrections is very substantial.

In comparison to the single-weighted (twofold) splitting the double-weighted (threefold) splitting exhibits a rather curious behavior. Even though the components $-\mathrm{Im}\Pi^{AA}$ and $-\mathrm{Im}\Pi^{BB}$ lie underneath their single-weighted complements $-\mathrm{Im}\Pi^{A}$ and $-\mathrm{Im}\Pi^{B}$ in some parts of the spectra, which one might expect to happen globally on first thought, they either coincide with them or even exceed them in other parts—sometimes to such an extent that they reach beyond the cumulative function. However, it has to be noted that these components, like the unweighted function, are always uniformly positive in sign, and therefore exhibit the correct analytic behavior that a function defined on this footing has to satisfy. It is required that these components be positive definite, because the net absorption in the medium must always be positive unless the system is excited out of equilibrium, which we do not consider here. In the preceding discussion we have assumed for convenience that the optical matrix elements between states of different components of the alloy are equal. If, as mentioned at the beginning of Sec. IV B, different optical cross sections (matrix elements) $\mu^{A/B}$ are distinguished for A and B atoms, the weighted contributions to yield the integrand of (125) would sum as

$$\tilde{K}=(\mu^A)^2K^{AA}+2\mu^A\mu^BK^{AB}+(\mu^B)^2K^{BB}. \qquad (131)$$

This shows that it would be possible to observe one of the functions Π^{AA} or Π^{BB} predominantly if either μ^A or μ^B happens to be much larger than the other. The mixed function Π^{AB}, however, will never be a separately observable quantity, no matter how the cross sections μ^A and μ^B scale relatively, and therefore not so rigid criteria for its analyticity apply as for the uniformly double-weighted functions.

VI. DISCUSSION AND CONCLUSION

In the previous sections we have obtained expressions for a wide class of weighted two-particle Green's functions. Additionally, in conjunction with the Appendix, we have managed to derive a general description for the solution of the two-particle CPA vertex equation, applicable for a case including weights as well as for one without, for an arbitrary number of channels within each of the two single-particle subspaces considered. The large choice for possible weightings is substantially reduced as restrictions are made to functions which would be useful in linear response theory. In both cases which are discussed for this kind of application, only five different weighted functions remain of which only two are genuinely independent.

The structure of the weighting process is closely related to that derived for the single-particle theory with the main difference that now the weights also depend significantly on the CPA vertex corrections. The calculation for the split band limit, the domain in which the CPA is superior to most of the other theories of disordered systems, gives direct insight into how the properly weighted CPA extracts the correct limiting behavior from different possible physical processes.

Some care is needed in interpreting the precise physical meaning of the weights, since they are obtained for the averaged functions, which describe the disordered medium as effectively homogeneous. The concept of the propagation of a particle between sites of different components is therefore lost in the effective medium as a consequence of averaging and the initial exclusion of specific propagation paths in the unaveraged function leads to the effective weights. These weights simply account for the average partition in probability for the simultaneous propagation of the particles between partly or completely specified site types at a given pair of energies z_1 and z_2.

Our numerical results show the general importance of the inclusion of vertex corrections into a properly self-consistent two-particle formalism. We find that, as a consequence of the inclusion of these average induced two-particle correlations, the center of mass and relative motion of the two-particle system effectively decouple to a large extent.

We believe that the general method developed here will find applications in various situations where two-particle motion is studied in a disordered medium. The effect of alloying on the electronic susceptibility and hence for example on the Ruderman-Kittel interaction has already been mentioned. In particular it can be extended for use in systems where the two particles have a direct interaction, such as the Coulomb interaction between carriers occurring in excitons in alloyed systems. In such a case the static correlations between particles, created by the disorder and accounted for by the vertex corrections, and the dynamic correlations introduced through the carrier-carrier interaction create additional static-dynamic correlations. Moreover it may be possible that the underlying disorder of the system gives rise to an induced disorder to the carrier-carrier interaction itself. Both of these effects can be treated within the method developed here and are discussed in a further publication.[34]

ACKNOWLEDGMENT

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APPENDIX

In this appendix we show how the site-diagonal vertex equation (39) can be solved even if all quantities in the a and b subspaces are matrices in an arbitrary choice of channel indices. First of all one can replace the average in (39) by writing

$$\Gamma_n = \langle T_n^a \{ \bar{G}_a g_a^{-1} C' g_b^{-1} \bar{G}_b + g_a \mathscr{K} g_b - \bar{G}_a \Sigma_a C' \Sigma_b \bar{G}_b - \bar{G}_a \Gamma_n \bar{G}_b \} T_n^b \rangle. \tag{A1}$$

This now represents a matrix equation for the site-diagonal vertex Γ_n, which has a definite solution. However, its solution cannot be obtained through a simple inversion of any combination of matrices which occur in the above expression, not even in a case when the two subspaces a and b coincide, as Γ_n on the right hand side is embraced by other matrices from *both* sides. It is therefore necessary to introduce some further algebraic tools. Equation (A1) is of the form

$$X = A + BXC, \tag{A2}$$

where a solution for X is required, A is an $N \times M$, B an $N \times N$, and C an $M \times M$ matrix, and B and C are regular. The matrix elements of all the matrices X, A, B, and C may be represented as x_{ij}, a_{ij}, b_{ij}, and c_{ij}, respectively. The quantity $B \otimes C$ shall be defined as the $N \cdot M \times N \cdot M$ matrix representation of the tensor product of B and C,

$$B \otimes C \equiv [b_{ij} \otimes c_{kl}]_{iM+l, jM+k}, \tag{A3}$$

as well as $\mathbf{I}$ the matrix representation of the unit tensor product,

$$\mathbf{I} \equiv [\delta_{ij} \otimes \delta_{kl}]_{iM+l, jM+k}, \tag{A4}$$

where $i,j \in \{1, \ldots, N\}$ and $k,l \in \{1, \ldots, M\}$ and every symbol representing an element of the new $N \cdot M \times N \cdot M$ tensor space is printed in boldface. Furthermore, the $N \cdot M$ vector $\underline{A}$ shall be defined as

$$\underline{A} \equiv [a_{ik}]_{iM+k}. \tag{A5}$$

This notation now allows us to rewrite the product BXC from (A2) as

$$\underline{BXC} = (B \otimes C) \underline{X}, \tag{A6}$$

and therefore (A2) is simply solved as

$$\underline{X} = \{ \mathbf{I} - B \otimes C \}^{-1} \underline{A}. \tag{A7}$$

Implicitly, this also immediately gives the solution for the matrix X, as the row index of its matrix elements is given by the integer value of division of the index of each element of $\underline{X}$ by M and their column index by the remainder of the division.

The solution of the vertex equation (A1) can therefore be written very similarly to (40),

$$\underline{\Gamma_n} = |n\rangle \hat{\Lambda} \langle n| [\underline{g_a \mathscr{K} g_b} + \underline{\bar{G}_a g_a^{-1} C' g_b^{-1} \bar{G}_b} - \underline{\bar{G}_a \Sigma_a C' \Sigma_b \bar{G}_b}] |n\rangle\langle n|, \tag{A8}$$

with the vertex $\mathbf{\Lambda}$ defined as

$$\mathbf{\Lambda} = \{ \mathbf{I} + \langle t_n^a F_a \otimes F_b t_n^b \rangle \}^{-1} \langle t_n^a \otimes t_n^b \rangle, \tag{A9}$$

and $T_n = |n\rangle t_n \langle n|$ was used again. Equations (42) and (43) generalize to

$$\underline{\mathscr{K}} = \underline{g_a^{-1} \bar{G}_a \Sigma_a C' \Sigma_b \bar{G}_b g_b^{-1}} + \sum_n ([g_a^{-1} \bar{G}_a |n\rangle] \otimes [\langle n| \bar{G}_b g_b^{-1}]) \mathbf{\Lambda} \langle n| [\underline{g_a \mathscr{K} g_b} + \underline{\bar{G}_a g_a^{-1} C' g_b^{-1} \bar{G}_b} - \underline{\bar{G}_a \Sigma_a C' \Sigma_b \bar{G}_b}] |n\rangle \tag{A10}$$

$$\langle m| \underline{g_a \mathscr{K} g_b} |m\rangle = \langle m| \underline{\bar{G}_a \Sigma_a C' \Sigma_b \bar{G}_b} |m\rangle + \sum_n (\langle m| \bar{G}_a |n\rangle \otimes \langle n| \bar{G}_b |m\rangle) \mathbf{\Lambda} | [\underline{g_a \mathscr{K} g_b} + \underline{\bar{G}_a g_a^{-1} C g_b^{-1} \bar{G}_b} - \underline{\bar{G}_a \Sigma_a C' \Sigma_b \bar{G}_b}] |n\rangle, \tag{A11}$$

as well as (47)–(52) to

$$\underline{a_k}=\sum_m e^{-ikR_m}\langle m|\underline{\bar{G}_a\Sigma_a C'\Sigma_b\bar{G}_b}|m\rangle, \tag{A12}$$

$$\underline{\alpha_k}=\sum_m e^{-ikR_m}\langle m|\underline{g_a^{-1}\bar{G}_a C'\bar{G}_b g_b^{-1}}|m\rangle, \tag{A13}$$

$$\mathbf{A}_k=\sum_m e^{-ikR_m}(F_m^a\otimes F_{-m}^b), \tag{A14}$$

$$\mathbf{B}_k=\sum_m e^{-ikR_m}(\langle m|g_a^{-1}\bar{G}_a|0\rangle\otimes\langle 0|\bar{G}_b g_b^{-1}|m\rangle), \tag{A15}$$

$$\underline{b_k}=\sum_m e^{-ikR_m}\langle m|\underline{g_a\mathscr{K}g_b}|m\rangle, \tag{A16}$$

$$\underline{c_k}=\sum_m e^{-ikR_m}\langle m|\underline{\mathscr{K}}|m\rangle, \tag{A17}$$

with which (A10) and (A11) can be solved as

$$\underline{b_k}=[\mathbf{I}-\mathbf{\Lambda A}_k]^{-1}[\underline{a_k}+\mathbf{\Lambda A}_k(\underline{\beta_k}-\underline{a_k})], \tag{A18}$$

$$\underline{c_k}=(\Sigma_a\otimes\Sigma_b)\underline{\alpha_k}+\mathbf{\Lambda B}_k[\underline{b_k}+\underline{\alpha_k}-\underline{a_k}]. \tag{A19}$$

As in the section for the scalar theory, $\underline{c_k}$ only provides a solution for the term $\underline{\mathscr{K}}$ and not the term $\underline{\mathscr{M}}$ which comprises the terms containing single products with U, for whose solution the structure of C' has to be known. Once this is the case, $\underline{\mathscr{M}}$ can be calculated exactly along the same lines as the generalized calculation for $\underline{\mathscr{K}}$ presented here. As before the solutions for the single-weighted functions, as, for example, $\langle G^{0i}CG^{i0}\rangle$, are void of this difficulty, and in analogy to before, the solution for the Fourier transforms of its diagonal elements can be obtained as

$$\underline{d_k}=\underline{b_k}, \tag{A20}$$

where $\underline{d_k}=\Sigma_m e^{ikR_m}\langle m|\langle\underline{G_a^{0i}CG_b^{i0}}\rangle|m\rangle$.

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[1] P. Soven, Phys. Rev. **156**, 809 (1967).

[2] D.W. Taylor, Phys. Rev. **156**, 1017 (1967).

[3] P.L. Leath, Phys. Rev. **171**, 725 (1968).

[4] P.L. Leath, Phys. Rev. B **2**, 3078 (1970).

[5] P.L. Leath, J. Phys. C **6**, 1559 (1973).

[6] B. Velický, S. Kirkpatrick, and H. Ehrenreich, Phys. Rev. **175**, 747 (1968).

[7] B. Velický, Phys. Rev. **184**, 614 (1969).

[8] B. Velický, S. Kirkpatrick, and H. Ehrenreich, Phys. Rev. **1**, 3250 (1970).

[9] B. Velický and K. Levin, Phys. Rev. B **2**, 938 (1970).

[10] R.J. Elliott, J.A. Krumhansl, and P. L. Leath, Rev. Mod. Phys. **46**, 465 (1974).

[11] Y. Onodera and Y. Toyozawa, J. Phys. Soc. Jpn. **24**, 341 (1968).

[12] F. Yonezawa and K. Morigaki, Prog. Theor. Phys. Suppl. **53**, 1 (1973).

[13] P. Soven, Phys. Rev. B **2**, 4715 (1970).

[14] B.L. Gyorffy and G.M. Stocks, J. Phys. (Paris) Colloq. **35**, C4-75 (1974).

[15] L. Schwartz and A. Bansil, Phys. Rev. B **10**, 3261 (1974).

[16] A. Bansil, L. Schwartz, and H. Ehrenreich, Phys. Rev. B **12**, 2893 (1975).

[17] G.M. Stocks, B.L. Gyorffy, E.S. Giuliano, and R. Ruggeri, J. Phys. F **9**, 1859 (1977).

[18] C.T. White and E.N. Economou, Phys. Rev. B **15**, 3742 (1977).

[19] R.N. Aiyer, R.J. Elliott, J.A. Krumhansl, and P.L. Leath, Phys. Rev. **181**, 1006 (1969).

[20] J.S. Faulkner and G.M. Stocks, Phys. Rev. B **21**, 3222 (1980).

[21] J. Kudrnovský and J. Mašek, Phys. Rev. B **31**, 6424 (1985).

[22] J. Kudrnovský and J. Mašek, J. Phys. F **16**, 943 (1986).

[23] J. Kudrnovský, V. Drchal, and J. Mašek, Phys. Rev. B **35**, 2487 (1987).

[24] H. Winter, P.J. Durham, and G.M. Stocks, J. Phys. F **14**, 1047 (1984).

[25] H. Shiva, Prog. Theor. Phys. **46**, 77 (1971).

[26] P.N. Sen, Phys. Rev. B **8**, 5613 (1973).

[27] G.D. Mahan, *Many Particle Physics* (Plenum, New York, 1990), p. 175.

[28] M.A. Ruderman and C. Kittel, Phys. Rev. **96**, 99 (1954).

[29] J.A. Blackman and R.J. Elliott, J. Phys. C **3**, 2066 (1970).

[30] S. Abe and Y. Toyozawa, J. Phys. Soc. Jpn. **50**, 2185 (1981).

[31] M.A. Kanehisa and R.J. Elliott, Phys. Rev. B **35**, 2228 (1987).

[32] B. Velický and A. Pieczonková, Phys. Scr. T **19**, 558 (1987).

[33] J.S. Langer and T. Neal, Phys. Rev. Lett. **16**, 984 (1966).

[34] N.F. Schwabe and R.J. Elliott, following paper, Phys. Rev. B **53**, 5318 (1996).