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Disorder representations and self-consistent effective-medium theories for spatially extended quasiparticles in alloys

Abdoul Aziz Mbaye*
Laboratoire de Physique du Solide et Energie Solaire, Centre National de la Recherche Scientifique,
Parc Sophia Antipolis, Rue Bernard Grégory, F-06560 Valbonne, France

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Disorder effects on spatially extended quasiparticles such as free excitons in semiconductor alloys cannot be described within the usual single-site approximation [e.g., the coherent-potential approximation (CPA)]. From an analysis of the relationship between the spatial scale of alloy potential fluctuations and an alloy effective medium, a new self-consistent approach is derived from a quasiparticle-dependent alloy disorder representation. This permits identification of an alloy effective medium whose interaction with a given spatially extended quasiparticle is a function of composition, alloy scattering strength (U), and the quasiparticle extent (Ω). It is then proved that the obtained self-energy expression reduces to the usual CPA and virtual-crystal approximation, respectively, for site-localized (Frenkel-type) quasiparticles and for infinitely extended (Wannier-type) quasiparticles. Furthermore, an approximate, closed-form expression for the self-energy as a function of x, U, and Ω is obtained. Applications to the study of disorder effects on free excitons in semiconductor alloys are also reported.

I. INTRODUCTION

The introduction of the concept of an effective medium is the basis of most recent progress in the understanding of equilibrium properties of phonons, electrons, magnons, and Frenkel excitons in random alloys. This, in turn, largely rests on the development of single-site approximations (SSA's),[1-5] the self-consistent version of which is the coherent-potential approximation[3] (CPA). In these approximations the alloy disorder state is treated within a Bragg-Williams—type description,[3] i.e., based on single-site (or point-cluster) averaging. In this Bragg-Williams alloy disorder representation (ADR) only the alloy fluctuations on a given site are considered, this site being associated with an average Wigner-Seitz volume Ω_0. Therefore, although they describe bulk alloy properties reasonably well throughout the entire composition range, the single-site approximations do not account for the effects of clustering on elementary excitations, or for disorder effects on quasiparticles which are characterized by large spatial extent $\Omega \gg \Omega_0$, such as free Wannier excitons in semiconductors. These excitations, however, play a central role in the understanding of the optical spectroscopy of semiconductor alloys, where they act as disorder probes, i.e., the broadening thereby of their associated spectral line gives a measure of the state of disorder in the alloy. This failure of single-site approximations appears to be mainly due to the inadequacy of the alloy disorder representation (i.e., the Bragg-Williams—type description) rather than to the techniques and approximations used in the derivation of a particular alloy effective medium.

Hence an extension of the single-site approximation which accounts for the spatial properties of excitations is needed in order to describe the effects of alloy potential fluctuations (disorder) on the spectral properties of such excitations. To this end, a theory has recently been reported[5] which permits description of disorder effects on excitations as functions of the alloy composition, of the difference between the chemical potentials of the relevant atoms (the origin of the alloy potential fluctuations), and of the spatial characteristics of the excitations themselves. However, this theory[5] has been given only in its non-self-consistent version,[1-4] thus corresponding to an extension of the average—t-matrix approximation (ATA) to the problem. The purpose of the present paper is to derive the self-consistent version of this theory, which will then be compared to the self-consistent single-site approximation, i.e., to the coherent-potential approximation.

Now, let us introduce formal spatially extended quasiparticles (SEQ's) which, insofar as one is concerned with disorder effects, will be characterized only by their spatial extent: the volume Ω over which they experience the alloy fluctuations (e.g., the Bohr volume for a free Wannier exciton). This volume, in turn, is a characteristic of the strength of the particular interaction which leads to the quasiparticle formation (e.g., Coulombic interaction for a free Wannier exciton). From this viewpoint, the main difference between Frenkel and Wannier excitons, both interacting with a given alloy disorder state, appears to be their respective spatial extent $\Omega(\text{Frenkel})=\Omega_0$(an atomic volume) and $\Omega(\text{Wannier})=N_0\Omega_0 \gg \Omega_0$.[6] (Here N_0 is the number of Wigner-Seitz cells within Ω.) In assuming that SEQ's are affected only by the alloy fluctuations within their spatial extent,[7] it is expected that these two quasiparticles are differently affected by the alloy disorder state, i.e., they average differently the alloy potential fluctuations. This suggests that the usual concept of an alloy effective medium, which does not account for the spatial properties of the SEQ, needs to be generalized by introducing, for these excitations, a representation of the alloy statistical disorder which accounts for the specific potential fluctuations which actually affect their spectral properties. This representation therefore depends on the extent of the SEQ. From the viewpoint of statistical mechanics, it is equivalent to introducing a new statistical ensemble[8] over which the ensemble-average properties of alloys are calculated.

The construction of a quasiparticle extent-dependent alloy disorder representation and the averaging technique used in this work are reported in Sec. II, where the general expression for the self-consistent self-energy which describes disorder effects on spatially extended quasiparticles is given. In Sec. II we examine the latter formalism in the limits of site-localized and infinitely extended quasiparticles, where comparison is made between the present theory, the CPA, and the virtual-crystal approximation (VCA). Applications to the study of disorder effects on the spectral properties of free Wannier excitons in semiconductor alloys are discussed in Sec. IV. Conclusions are drawn in Sec. V about possible further uses of the present formalism.

II. ALLOY-DISORDER REPRESENTATION AND FORMALISM

Consider a binary alloy $A_{1-x}B_x$ and a free SEQ (i.e, one not bound to impurities or defects in the alloy, nor trapped by fluctuations[9]). Let Ω be the spatial extent of this SEQ. As discussed above, we assume that the spectral properties of the SEQ will be affected only by the fluctuations of the alloy potential within its volume Ω during its motion throughout the alloy.[7] Therefore, if one assumes that the mean free path of the SEQ is much larger than its characteristic dimensions, all permissible concentrations c of B-type atoms will appear in Ω, each with a given probability, denoted by $p(c)$, which depends on the nature of the solid solution under consideration. (This statement is valid for the free SEQ considered here, for which the disorder-induced spectral line broadening ΔE is small compared to the energy scale of their spectra, i.e., their lifetime $\hbar/\Delta E$, which is infinite in pure crystals, is still large in comparison to a scattering time. However, it would not apply to the case of fluctuation-localized quasiparticles which are associated with nonpropagating states and which have been recently observed in semiconductor alloys.[9]) For a finite size Ω, the permissible concentrations c in Ω are discrete and given by $c=0,1/N_0,\ldots,N_0/N_0=1$, where N_0 is the number of lattice sites (or equivalently of Wigner-Seitz cells) in Ω, $N_0=\Omega/\Omega_0$. Let R_Ω denote the ensemble of these sites in Ω.

The macroscopically observed disorder effects on the SEQ spectral properties result from the fluctuations about the average alloy composition (x) of the concentration $(c\neq x)$ of B-type atoms in its volume Ω during migration in the alloy. For each position of the SEQ, the alloy potential it experiences can be represented as the superposition of (i), inside Ω, the potential of a uniform medium of composition c occupying the volume Ω, and (ii) outside Ω, that of an embedding medium obtained from averaging over all the fluctuations outside Ω. Such a system, formed by a uniform medium of volume Ω and composition c, embedded in an as yet unknown effective medium, will be denoted S_c. Its probability of occurrence is the same as that of c and is given by $p(c)$. Therefore the ensemble $\{S_c,p(c)\}$ represents all the possible fluctuations of the alloy potential experienced by the SEQ's.

The statistical ensemble[8] over which the alloy potential fluctuations are averaged will be completely determined once the Hamiltonian used to describe each S_c is specified. We shall, for convenience of implementation, use a one-band tight-binding model Hamiltonian,[3] and write the Hamiltonian H_c for each S_c as

$$H_c=\sum_{i\in R_\Omega}|i\rangle e_c\langle i|+\sum_{i\notin R_\Omega}|i\rangle e_r\langle i| +\sum_{i}\sum_{j\,(\neq i)}|i\rangle t_{ij}\langle j|\ . \qquad (1)$$

The first sum in (1) is restricted to sites belonging to R_Ω (i.e., within Ω), at each of which is placed a potential e_c, related to the pure A and B constituent's on-site potentials e_A and e_B by $e_c=(1-c)e_A+ce_B$. [An assumption which is implicit in this expression for e_c is that the potential in Ω depends only on the composition of the uniform medium in Ω and not on the particular way the cN_0 B atoms and $(1-c)N_0$ A atoms are distributed in Ω. The physical meaning of this latter assumption is that the spectral properties of the SEQ's are assumed to be affected only by the deviation $\xi=c-x$ of the composition c in Ω from the average alloy composition x.]

The second sum in (1) is over all sites outside Ω, by our definition of S_c therefore characterized by an effective medium on-site potential $e_r=\bar{e}+W$, where $\bar{e}=(1-x)e_A+xe_B$ is the linear interpolation between e_A and e_B, i.e., the virtual-crystal-approximation on-site potential, and W is a yet-unknown potential describing the deviation of the average potential outside Ω from the VCA value $\bar{e}$.[3] Because W is an average quantity it is, in principle, determined only after averaging over all the systems S_c. The best description of the medium outside Ω (the reference medium) is thus obtained only in a self-consistent way. As previously shown for the case of single-site approximations,[3] the main difference between the CPA (which is self-consistent) and the ATA (which is non-self-consistent) arises from the fact that the latter assumes that W is a constant (specifically, $W=0$) and therefore the resulting description of disorder effects depends on this choice of W (i.e., this choice for W forces non-self-consistency), while the former makes no assumptions about W (it is an *a priori* unknown): Having first averaged over all fluctuations, it is determined from the condition of vanishing additional disorder scattering due to fluctuations. This requirement we may call the coherent-potential condition (CPC). Under the CPC it is clear that the properties of the alloy effective medium and related disorder effects (e.g., the self-energy) depend implicitly on the fluctuations selected to describe the alloy disorder state (i.e., on the alloy disorder representation). One purpose of the present work is to make explicit this dependence, in order to characterize the effective medium in which a spatially extended quasiparticle moves.

The third sum in (1) describes the propagation of the SEQ. We shall neglect the fluctuations in the transfer integral t_{ij} which arise from its dependence on the types of atoms occupying the sites i and j, i.e., we shall neglect "off-diagonal" disorder,[3] which is important only when the relative widths of the SEQ energy spectra in the pure-alloy end-point materials are considerably different. In our case, $t_{ij}=t$ and the bandwidth t will be taken as the energy unit.

Since fluctuations away from the effective-medium value are (by the definition of the system S_c) confined to the volume Ω, it is convenient to express the Hamiltonian H_c in S_c as the sum of a fluctuation-independent part H_r (evaluated for the reference effective medium, labeled r) and a fluctuation-dependent part U_c which describes the disorder in S_c, i.e.,

$$H_c \equiv H_r + U_c \ . \tag{2}$$

Here

$$H_r = \sum_i |i\rangle e_r \langle i| + \sum_i \sum_{j\,(\neq i)} |i\rangle t \langle j| \ ; \tag{3}$$

the fluctuation-dependent part U_c may be expressed as a function of the composition deviation $\xi = c - x$ and the on-site potential difference $U = e_B - e_A$ between atoms A and B as

$$U_c = \hat{P} * (\xi U - W) \ . \tag{4}$$

To proceed further we have introduced a new operator $\hat{P}$ which selects out all fluctuations $\{S_c, p(c)\}$ confined to Ω over which the macroscopic average quantities are to be calculated. $\hat{P}$ may be written as

$$\hat{P} = \sum_{i \in R_\Omega} |i\rangle\langle i| \ ; \tag{5}$$

$\hat{P}$ is a projection operator ($\hat{P}^2 = \hat{P}$) which does not have the translational symmetry of the average alloy lattice. The asterisk in Eq. (4) indicates that, when applied to a realistic potential, the product is a convolution product. [In the case of realistic atomic potentials, i.e., $U(\mathbf{r}) = e_B(\mathbf{r}) - e_A(\mathbf{r})$, $\hat{P}$ becomes a summation, over those sites τ of the average alloy lattice within the volume Ω, of Dirac delta functions $\delta(\mathbf{r} - \tau_i)$. The operator product $\hat{P} * U$ then stands for $\int d\mathbf{r}' \hat{P}(\mathbf{r}') U(\mathbf{r} - \mathbf{r}')$.] Here the triplets $\{c\} = \{S_c, H_c, p(c)\}$ constitute the alloy disorder representation over which the ensemble-average of the SEQ properties are calculated. For a given alloy $A_{1-x}B_x$, they are determined only by the volume Ω of the SEQ under consideration. The introduction of $\hat{P}$ ensures that only the disorder effects arising from the fluctuations within Ω are considered; those from the region outside Ω are included in the effective medium, i.e., in W.

Having specified the statistical ensemble, we now turn to the averaging procedure used to derive the macroscopic average effects of the alloy potential fluctuations on the SEQ spectral properties; these disorder effects may generally be represented as self-energy effects, related directly to the alloy coherent potential. To this end we shall use the exact self-energy expansion technique developed by Argyres.[10] This technique allows the application of the coherent-potential condition to any alloy disorder representation where the fluctuation-dependent Hamiltonian (such as H_c) can be split in the form of Eq. (2). When the CPC is applied the alloy effective medium, characterized by $S(E) \equiv \bar{e} + \Sigma(E, N_0)$, is uniquely determined; the parameter W of Eq. (4), since $\bar{e}$ is the VCA on-site potential, is equal to the energy-dependent self-energy contribution to the alloy potential, $\Sigma(E, N_0)$. In the present case, $W = \Sigma(E, N_0)$ depends also on Ω (or equivalently on $N_0 = \Omega/\Omega_0$) and is given by the self-consistent equation (see the Appendix)

$$\begin{aligned}\Sigma(E,N_0) &= \sum_{m=1}^{\infty} \hat{Q}\{\xi\hat{P}*U + \xi\hat{P}*U[1 - G_r(E)(\hat{Q}\xi\hat{P}*U - \hat{P}*W)]^{-1}\} G_r(E)\hat{Q}'\xi\hat{P}*U \\ &= \sum_{m=1}^{\infty} \sum_{L=0}^{m} \binom{m}{L} (-1)^{m-L} K_{2+L}(N_0;p) V_{m,L}(E) \ ,\end{aligned} \tag{6}$$

where

$$K_m(N_0;p) = \hat{Q}[\xi(\hat{Q}'\xi)^{m-1}] \tag{7}$$

and

$$\begin{aligned} & V_{m,L}(E) \\ & \quad = \hat{P}*U[G_r(E)\hat{P}*U]^L [G_r(E)\hat{P}*W]^{m-L} G_r(E)\hat{P}*U \ ;\end{aligned} \tag{8}$$

E is the energy of the SEQ. $\hat{Q}$ and $\hat{Q}' = 1 - \hat{Q}$ are projection operators which, respectively, effect the ensemble average and give the fluctuating part of any fluctuation-dependent quantity Z_c to their right:[10]

$$\hat{Q} Z_c = \sum_{\{c\}} p(c) Z_c \ . \tag{9}$$

The chemical factor $V_{m,L}(E)$ in Eqs. (6) and (8) describes the energy-dependent multiple-scattering processes experienced by the SEQ due to the fluctuation within Ω. $G_r(E)$ is the free Green function associated with the effective (reference, r) medium Hamiltonian.

The factorization of each term in the expansion (6) into the product of a purely statistical factor K_m and a purely chemical factor $V_{m,L}(E)$ is the main advantage of the representation used here. The statistical factor $K_m(N_0;p)$ depends on the SEQ spatial extent $\Omega = N_0\Omega_0$ and on the probability distribution $p(c)$ of the fluctuating composition c. It is a sum of products of moments of ξ with respect to the probability distribution $\{p(c)\}$. Note that at the present stage no specific assumption concerning this probability distribution has been made. Therefore, Eq. (6) provides the basis for calculation of disorder effects on SEQ in real alloys, where the strictly random probability distribution used in the derivation of single-site approximations (CPA or ATA) is not valid, in general. It is worth noting that Eq. (6) is exact, since no approximation is needed for its derivation once the ADR is chosen.

III. DERIVATION OF EFFECTIVE MEDIA

A. Limiting cases

We now consider two limiting cases: First, that of infinitely extended quasiparticles (IEQ's), for which $\Omega=\infty$ (i.e., $N_0 \rightarrow \infty$) and, second, the case of Frenkel (site-localized) quasiparticles, for which $\Omega=\Omega_0$.

(i) In the first case, the only possible composition within Ω is x, i.e., $p(c=x)=1$; $p(c\neq x)=0$, following the strong law of large numbers).[11] Therefore all K_m's vanish and hence $\Sigma(E,N_0=\infty)=0$. The VCA $[S(E)=\bar{e}]$ is thus obtained for these IEQ's.

(ii) For site-localized (Frenkel[6]) quasiparticles $\Omega=\Omega_0$, i.e., $N_0=1$, and the projection $\hat{P}$ becomes $\hat{P}=|0\rangle\langle 0|$. Under the random disorder approximation, the respective probabilities that the only site (0) in Ω is occupied by an A- or B-type atom are given by $p_0(A)=p_0(c=0)=1-x$ and $p_0(B)=p_0(c=1)=x$; the subscript 0 stands for the random disorder approximation. The statistical factors K_m are then given by $K_m(1;p_0)=x(1-x)(1-2x)^{m-2}$ for $m\geq 2$. In this case, the expectation value of $W=\Sigma(E,N_0)$ obtained from Eq. (6) is given by

$$\sigma(E,N_0=1) = \langle 0|\Sigma(E,N_0=1)|0\rangle = \frac{U^2 x(1-x)F(E,N_0=1)}{1-[U(1-2x)-\sigma(E,N_0=1)]F(E,N_0=1)}, \tag{10}$$

where

$$F(E,N_0)=\langle 0|G(E)|0\rangle=\int\frac{\rho(E')}{E-\bar{e}-\sigma(E,N_0)-E'}dE' . \tag{11}$$

In Eq. (11) $\rho(E)$ is the density of states, the same in pure crystals A and B (with neglect of off-diagonal disorder[3]). Equation (6), therefore, reduces to the single-site coherent-potential approximation[12] for site-localized quasiparticles which extend over a single unit cell ($\Omega=\Omega_0$ or $N_0=1$), i.e., for Frenkel quasiparticles. The major difference from previous derivations of the CPA is that in ours it appears as an exact solution of the alloy effective-medium problem once the local ADR is chosen ($\Omega=\Omega_0$). The usual neglect of clustering effects (i.e., the single-site approximation) required in the former derivations no longer enters the theory as an approximation, but rather as a characteristic of the ADR, where the only fluctuations involved in the scattering processes are selected by $\hat{P}=|0\rangle\langle 0|$ for $\Omega=\Omega_0$.

The study of these two limiting cases, the VCA ($\Omega=\infty$) and the CPA ($\Omega=\Omega_0$), shows the deep relationship between the concepts of alloy effective media [characterized by $\Sigma(E,N_0)$] and alloy disorder representations (defined by Ω or $N_0=\Omega/\Omega_0$). It also proves that the present approach reduces to the usual VCA (vanishing disorder effects) and to the CPA, respectively, in the limits of infinitely extended and of site-localized quasiparticles. It is hence expected that the present theory will offer a rigorous basis for the description of alloy effective-medium effects felt by spatially extended quasiparticles with an intermediate spatial extent $\Omega_0<\Omega<\infty$, as will be shown in the next subsection.

B. General case

For the general case of a SEQ with finite spatial extent Ω, we now derive a closed-form self-consistent self-energy expression for the disorder effects as functions of U $(=e_A-e_B)$, of composition x, $0\leq x\leq 1$, and as a function of Ω. Since the CPA ($N_0=1$) can be obtained when only multiple scattering from a single site is considered, we will seek an approximate self-energy expression which accounts for the same scattering process. This approximation on the $V_{m,L}(E)$, in turn, forces one, for self-consistency, to use an equivalent approximation for the $K_m(N_0;p_0)$. In this case, we have shown[5] that

$$K_m(N_0;p_0)=x(1-x)(1-2x)N_0^{-(m-1)} .$$

We then obtain a closed-form expression for the expectation value of $\Sigma(E,N_0)$,

$$\sigma(E,N_0)=\langle 0|\Sigma(E,N_0)|0\rangle = \frac{(U^2/N_0)x(1-x)F(E,N_0)}{1-[(U/N_0)(1-2x)-\sigma(E,N_0)]F(E,N_0)} . \tag{12}$$

Equation (12) reduces to the CPA [Eqs. (10) and (11)] for $N_0=1$ and to the VCA $[\Sigma(E,N_0)=0]$ for $N_0=\infty$. It therefore generalizes these two approaches, which are valid in the respective limits of site-localized and infinitely extended quasiparticles. In addition, it offers, for the first time, a rigorous basis for a self-consistent determination of the disorder effects "felt" by a given SEQ as functions of x, U, and its extent Ω.

IV. PHYSICAL APPLICATIONS

Optical characterization of semiconductor alloys has revealed the importance of disorder effects on the spectroscopy of free and impurity-bound quasiparticles.[13] Among these, free excitons play a central role in the interpretation of near-band-edge spectra of semiconductors. However, typical trends in the disorder effects on the spectra of these spatially extended quasiparticles are not accounted for by the usual single-site approximations, while they are well described by Eq. (12). For the sake of illustration, the present theory has been applied to the calculation of the absorption spectra of free Wannier excitons in semiconductor alloys as functions of their spatial extent Ω.[14,15] To this end a hydrogenic exciton model[13–16] has been used. In this model the spatial extent Ω ($\Omega=\langle r^3\rangle$) is given by $\Omega=10\pi a_B^3$, where the Bohr radius a_B is of the order $1/m_r$, where m_r is the reduced effective mass. From calculations of the absorption spectra for two free excitons with different spatial extents ($\Omega=125\Omega_0$ and

$1000\Omega_0$, i.e., $N_0=125$ and 1000, respectively) for $x=0.5$ and $U=0.4$ (in units of t), the trends can be summarized by the statement that, for decreasing N_0 or Ω, the following applies:

(i) the linewidth of the absorption peak associated with the exciton increases;

(ii) the intensity of the peak decreases; and

(iii) the high-energy absorption tail, generally attributed to disorder-assisted indirect transitions, increases (see Ref. 15).

From these results it can be inferred that when the exciton spatial extent N_0 increases, the associated absorption peak tends to a Dirac-type peak, which is characteristic of excitons in pure semiconductors or in alloys without measurable disorder effects. The important result that can be stated as a rule is that disorder effects on the spectral properties of a spatially extended quasiparticle decrease when its spatial extent (Ω) increases. This conclusion, although reasonable on purely intuitive grounds, has never previously followed from a rigorous calculation before the introduction of the approach reported in Ref. 5, whose self-consistent version is given here. Furthermore, in addition to the study of the limiting cases (Sec. III A) it follows from this conclusion that for general SEQ's ($\Omega\neq\Omega_0$ and $\Omega\neq\infty$), the CPA ($N_0=1$) will overestimate disorder effects while the VCA ($N_0=\infty$) will underestimate them.

The knowledge of such a trend in disorder effects on free excitons in semiconductor alloys also permits an understanding of the experimentally observed increase in disorder-induced effects on free Wannier exciton spectra (e.g., linewidth broadening) in going from direct-gap III-V alloys to indirect-gap III-V and II-VI alloys. The effective mass m_r^d in direct-gap III-V materials is much smaller than m_r^i in the indirect-gap material; since in a hydrogenic model the Bohr radius $a_B\sim 1/m_{\text{eff}}$, $\Omega(\textit{indirect}) \ll \Omega(\text{direct})$ so that, following the above-mentioned rule, smaller disorder effects are expected in the direct-gap material.

Another important property of these disorder effects as functions of the composition x of the alloy $A_{1-x}B_x$ is their asymmetry about $x=0.5$. This behavior, not explained by previous alloy theories, follows from Eq. (12) as a consequence of the difference between the SEQ spatial extents $\Omega(A)$ and $\Omega(B)$ in the pure constituents A and B, respectively: If $\Omega(A)>\Omega(B)$, i.e., $N_0(A)>N_0(B)$, disorder effects $[\Sigma(E,N_0)]$ are weaker in A-rich than in B-rich alloys, even in the weak-scattering limit ($U\sim 0$). In other words, Nordheim's "symmetry rule,"[1] which states that disorder effects are symmetric with respect to the interchange of A and B atoms, in this limit breaks down.

Finally, note that Eq. (12) yields a good basis for the study of the "persistence" (in which features of the pure A and B materials survive in the alloy SEQ spectrum, i.e., two modes persist) and "amalgamation" (where only a single alloy-averaged feature survives in the SEQ spectrum) regimes of SEQ spectral properties in alloys as functions of both alloy properties (x,U) and SEQ spatial properties (Ω or N_0). However, this extension of the previous work of Onedera and Toyozawa[17] is beyond the scope of the present paper.

V. CONCLUSIONS

In summary, we have explored the interconnection between the concepts of alloy disorder representation and alloy effective medium. Through the introduction of the quasiparticle extent-dependent alloy disorder representation we have demonstrated that both the virtual-crystal and the coherent-potential approximations are exact solutions of the alloy effective-medium problem in the respective limits of infinitely extended (IEQ's) and site-localized quasiparticles (SLQ's). An approximate self-consistent expression for the coherent potential which yields both these latter approximations (the VCA and the CPA) in the respective limits of IEQ and SLQ is obtained. This expression accounts for some general trends observed for real disordered alloys in the variation of disorder effects as functions of x, U, and Ω. The results presented here are therefore expected to bring a new understanding of the experimentally observed behavior of free spatially extended quasiparticles in alloys, particularly of free excitons in semiconductor alloys. Extensions of this approach to the case of short-range impurity-bound particles are presented elsewhere.[18]

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APPENDIX

The alloy effective medium, characterized by the self-energy $W=\Sigma(z,N_0)$, is defined by applying the no-additional-disorder-scattering condition to the fluctuations c within the SEQ volume Ω, such that the average Green function $G_r(z)$ is given by

$$G_r(z)=\hat{Q}g_c(z),\quad z=E+i0^+ \tag{A1}$$

where $\hat{Q}$ is the projection operator introduced by Argyres,[10] which gives the ensemble average of any quantity on its right. The configuration-dependent Green function is defined by

$$g_c(z)=(z-H_r-V_c)^{-1}\ . \tag{A2}$$

$G_r(z)$ can also be written as

$$G_r(z)=[z-H_r-\Sigma(z,N_0)]^{-1}\ , \tag{A3}$$

where $\Sigma(z,N_0)$ is the SEQ-extent-dependent self-energy, which describes the alloy effective medium "felt" by the SEQ under consideration and N_0 is Ω/Ω_0. From the averaging technique developed by Argyres,[10]

$$\Sigma(z,N_0)=\hat{Q}_0\{V_c+V_c[z-H_r-(\hat{Q}'\hat{V}_c-R)]^{-1}\hat{Q}'V_c\}\ , \tag{A4}$$

where $V_c=\hat{P}*\xi U$ and $R=\hat{P}*W$. Equation (A4) can be written as

$$\Sigma(z,N_0)=\hat{Q}\{V_c+V_c[1-G_r(z)(\hat{Q}'V_c-R)]^{-1} \times G_r(z)\hat{Q}'V_c\}\ . \tag{A5}$$

When the second term on the right-hand site is expanded, one obtains

$$[1-G_r(z)(\hat{Q}'V_c-R)]^{-1}=\sum_{m=0}^{\infty}[G_r(z)(\hat{Q}'V_c-R)]^m\ . \tag{A6}$$

Since $G_r(z)$ and R commute with $\hat{Q}'=1-\hat{Q}$ and V_c, each term in Eq. (A6) can be calculated from the usual commutative algebra laws

$$[G_r(z)(\hat{Q}'V_c-R)]^m=\sum_{L=0}^{m}(-1)^{m-L}\binom{m}{L}[G_r(z)\hat{Q}'V_c]^L \times[G_r(z)R]^{m-L}\ . \tag{A7}$$

Here $\binom{m}{L}$ has the usual meaning used in combinatorial analysis. Substituting for $\underline{V}_c$ and R we obtain

$$[G_r(z)(\hat{Q}'V_c-R)]^m=\sum_{L=0}^{m}(-1)^{m-L}\binom{m}{L}[G_r(z)\hat{P}*U]^L[G_r(z)\hat{P}*\Sigma(z,N_0)]^{m-L}(\hat{Q}'\xi)^L\ . \tag{A8}$$

To obtain (A8) we have used the commutativity of $\hat{Q}$ and $\hat{Q}'$ with any quantity independent of c (or ξ). Now substituting (A8) in (A4), we obtain

$$\Sigma(z,N_0)=\hat{Q}\xi\hat{P}*U\sum_{m=2}^{\infty}\sum_{L=0}^{m}(-1)^{m-L}\binom{m}{L}(Q'\xi)^L[G_r(z)\hat{P}*U]^L[G_r(z)\hat{P}\Sigma(z,N_0)]^{m-L}(\hat{Q}'\xi)[G_r(z)\hat{P}*U)]\ , \tag{A9}$$

the first term in (A4) being identically zero since $\hat{Q}(\xi)=0$.

The expression for $\Sigma(z,N_0)$ can be reorganized and written as

$$\Sigma(z,N_0)=\sum_{m=0}^{\infty}\sum_{L=0}^{m}\binom{m}{L}\hat{Q}[\xi(\hat{Q}'\xi)^{L+1}]\hat{P}*U[G_r(z)\hat{P}*U]^L[G_r(z)\hat{P}*\Sigma(z,N_0)]^{m-L}[G_r(z)\hat{P}*U]\ . \tag{A10}$$

From the definition of the statistical factor $K_m(N_0;p)$ used in Ref. 9, i.e.,

$$K_m(N_0;p)=\hat{Q}[\xi(\hat{Q}'\xi)^{m-1}]\ , \tag{A11}$$

we have

$$\Sigma(z,N_0)=\sum_{m=0}^{\infty}\sum_{L=0}^{m}(-1)^{m-L}\binom{m}{L}K_{L+2}(N_0;p_0)V_{L,m}(z)\ , \tag{A12}$$

where

$$V_{m,L}(z)=\hat{P}*U[G_r(z)\hat{P}*U]^L[G_r(z)\hat{P}*\Sigma(z,N_0)]^{m-L} \times G_r(z)\hat{P}*U\ . \tag{A13}$$

The symbol p in $K_m(N_0;p)$ indicates that K_m depends on the probability distribution law used in the alloy disorder representation, from which are calculated the moments of the fluctuating composition c. For a randomly disordered alloy $A_{1-x}B_x$ $(p=p_0)$,

$$p_0(c)=p_0(\xi)=\binom{N_0}{n}x^n(1-x)^{N_0-n}\ , \tag{A14}$$

where $n=cN_0$. Therefore, as shown in Ref. 9, the $K_m(1;p_0)$ for $N_0=1$ (Frenkel quasiparticles, $\Omega=\Omega_0$) are exactly given by

$$K_m(1;p_0)=x(1-x)(1-2x)^{m-2},\quad m\geq 2 \tag{A15}$$

while for $N_0\geq 2$ the $K_m(N_0;p_0)$ can be approximated by

$$K_m(N_0;p_0)=x(1-x)(1-2x)^{m-2}N_0^{-(m-1)},\quad m\geq 2\ . \tag{A16}$$

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[*] Present address: Solar Energy Research Institute, 1617 Cole Boulevard, Golden, CO 80401.

[1] R. J. Elliot, J. A. Krumhansl, and P. L. Leath, Rev. Mod. Phys. **46**, 475 (1974).

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

[3] H. Ehrenreich and L. M. Schwartz, in *Solid State Physics*, edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1976), Vol. 31.

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

[5] A. A. Mbaye, F. Raymond, and C. Vérié, Phys. Rev. B **30**, 909 (1984).

[6] Following Frenkel's exciton model [J. Frenkel, Phys. Rev. **37**, 17 (1931)], the electron and hole states involved in exciton formation are in the same atomic unit cell, of volume Ω_0 (a site-localized quasiparticle). This quasiparticle is, therefore, mainly sensitive to alloy fluctuations within this cell $\Omega_F=\Omega_0$, whereas for Wannier excitons [G. H. Wannier, Phys. Rev. **52**, 191 (1937)] the electron-hole pair occupies a volume Ω much larger than Ω_0. The limiting case for such a SEQ is $\Omega=\infty$.

[7] This assumption has been previously used by S. D. Baranovski *et al.* (Fiz. Tekh. Poluprovodn. **12**, 2233 (1978) [Sov. Phys.—Semicond. **12**, 1328 (1978)]) in carrying out their calculation on the localization of excitons in alloys. However, in the present work we are mainly concerned with effective-medium effects on free quasiparticles.

[8] R. C. Tolman, *Principles of Statistical Mechanics* (Dover, New York, 1979), p. 43.

[9] S. Lai and M. V. Klein, Phys. Rev. Lett. **44**, 1087 (1980).

[10] P. N. Argyres, Phys. Rev. B **23**, 2447 (1981).

[11] Y. A. Rozanov, *Probability Theory* (Dover, New York, 1969), p. 70.

[12] E. N. Economou, *Green Functions in Quantum Physics* (Springer-Verlag, Berlin, 1980).

[13] R. J. Nelson, *Excitons* (North-Holland, Amsterdam, 1982), p. 319.

[14] O. Goede, D. Henning, and L. John, Phys. Status Solidi B **96**, 671 (1979).

[15] A. A. Mbaye, F. Raymond, and C. Vérié, Solid State Commun. **50**, 479 (1984).

[16] G. Neu, A. A. Mbaye, and R. Triboulet, presented at the 17th International Conference on the Physics of Semiconductors, San Francisco, 1984 (unpublished).

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

[18] A. A. Mbaye and H. Mariette, in Proceedings of the 13th International Conference on Defects in Semiconductors, Coronado, 1986 (unpublished).