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Anderson localization in topologically disordered systems

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A self-consistent theory of localization in a tight-binding model of topologically disordered systems is investigated. A key element of the theory is use of a disordered reference system in which the structure of the medium is taken into account. The spatial disorder inherent in the Boltzmann center-of-mass distribution is taken as the explicit source of lateral disorder, and the effects of simultaneous site-diagonal disorder are also included. The theory is formulated in a manner which circumvents use of the so-called upper-limit approximation. The Anderson transition density predicted by the theory is estimated for transfer-matrix elements of the forms $V(R)\sim R^{-n}$, $V(R)\sim \exp(-R/a_H)$, and $V(R)\sim(1+R/a_H)\exp(-R/a_H)$. Full mobility-edge trajectories for power-law transfer-matrix elements are also determined, with particular emphasis on the cases $n=3$ and 5 corresponding, respectively, to dipolar and quadrupolar excitons. A connection is also made between the density of states determined from the self-consistent theory and that resultant from solution of the quantum mean spherical integral equation.

I. INTRODUCTION

The theory of localization in disordered systems, pioneered by Anderson[1] in 1958, continues to occupy a central role in condensed-matter physics. It was shown by Anderson[1] that there is a transition between localized and extended eigenstates in a one-particle tight-binding model with site-diagonal disorder (random site energies). There have since been numerous studies of this model on lattices.[2]

Systems such as liquids, gases, or glasses are, however, characterized by topological disorder. In recent years there has been interest in obtaining criteria for localization in this class of systems.[3-11] Attention has focused on analysis of random tight-binding models with off-diagonal (or lateral) disorder, in which the matrix element transferring the excitation from site to site is a random variable, reflecting the spatial disorder inherent in the problem. Particular emphasis has been given to the nature of electronic states near the metal-insulator transition in disordered materials, although the question of whether or not states at a given energy are localized is also relevant to phenomena such as the propagation of multipolar excitons in fluids and the propagation of phonons in amorphous materials.[12,13] The aim of this paper is to pursue a theory of localization in topologically disordered systems, in which one takes into account the structure of the medium in which the phenomenon occurs; both off-diagonal and site-diagonal disorder will be considered.

Abou-Chacra, Anderson, and Thouless[14,15] have developed a probabilistically based theory of localization for the Anderson model on a Bethe lattice. This is referred to as the self-consistent theory, and is centered on a self-consistent solution of the equation for the self-energy in second-order renormalized perturbation theory. Our approach to localization in the more complex case of topological disorder, which is also based on a noninteracting tight-binding model, follows the methods of Abou-Chacra *et al.*,[14] and yields a self-consistent theory in a manner which combines the methods of probabilistic quantum mechanics with elements of liquid-state theory. It need hardly be emphasized that the theory which we formulate is an approximate one for the systems considered, and also is in the nature of a mean-field theory.

Several authors[3,4] have studied the self-consistent theory for a completely random distribution of site positions, the structure of the medium thereby being ignored; furthermore, these studies have been confined to Anderson's so-called upper-limit approximation[1,14] to the basic equations of the self-consistent theory, whereby one neglects the influence of the real part of the self-energy. In a previous publication[13] we outlined a theory which alleviates the first problem, and examined the implications of the theory for lateral disorder alone and within the upper-limit approximation. Here we develop the theory further, to circumvent use of the upper-limit approximation and to include the effects of site-diagonal disorder.

In Sec. II we derive the general self-consistent equations. For a fixed center-of-mass configuration of the atoms, $\{\mathbf{R}_i\}$, we assume that the system is specified by a tight-binding model. The site energies are assumed to be drawn from a given probability distribution, while the topological disorder inherent in the Boltzmann distribution for the centers of mass is taken as the explicit source of randomness in the transfer-matrix element. To calculate the joint probability distribution, $f(E_j,\Delta_j)$, for the real and imaginary parts of the self-energy of atom j, $S_j=E_j-i\Delta_j$, we perform an average over a reference system deemed to consist of particles, each possessing a random self-energy and mutually interacting via an appropriate classical interaction potential, $U_0(\{\mathbf{R}_i\})$. For simplicity, we assume that the interaction potential is spherically symmetric and pairwise-additive, of the form

$$U_0(\{\mathbf{R}_i\})=\frac{1}{2}\sum_{i,j}{}' U_0(|\mathbf{R}_i-\mathbf{R}_j|) ;$$

in specific applications we will assume $U_0(R)$ to be that appropriate to a hard-sphere fluid with hard-sphere diameter σ. Introduction of a disordered reference system in which the structure of the medium is explicitly taken into account is a key element of the present theory.

Calculation of the average referred to above is formally equivalent to a determination of the excess chemical potential of atom i dissolved in the multicomponent reference fluid, wherein it interacts with the other atoms via a complex potential. We may therefore use suitable approximation schemes for calculating the "chemical potential" which are available to us from conventional liquid-state theory,[16] in order to render tractable the resultant integral equation for $F(k_1,k_2)$, the Fourier transform of $f(E_i,\Delta_i)$. Whether or not states at the chosen energy are localized depends on whether or not solutions to the derived equation exist. By examination of the asymptotic behavior of $F(k_1,k_2)$, we derive a linear homogeneous integral equation, the limits of stability of solutions to which indicate the transition to extended states at the determined energy and given number density. Analysis of this equation therefore yields the mobility edge as a function of number density.

In Sec. III we demonstrate a close connection between an approximate density of states resultant from the self-consistent theory, and the density of states obtained from solution of the quantum mean spherical integral equation for multipolar excitations.[17–22] The latter is essentially an effective-medium theory for the electronic states of topologically disordered systems, and has recently been the subject of considerable theoretical interest.[17–22]

In Sec. IV we estimate the Anderson transition density predicted by the theory, namely that density at which the mobility edges coalesce at the band center and below which all states in the band are localized. Transfer-matrix elements of the form $V(R)\sim R^{-n}$, applicable to a discussion of electronic multipolar excitons, are considered, and the relative influence of site-diagonal disorder is also investigated. Similarly, we investigate transfer-matrix elements of exponential form, applicable to triplet excitons or to a discussion of the nature of electronic states near the metal-insulator transition in disordered materials. In contrast to the majority of previous theories, and in agreement with the recent work of Puri and Odagaki,[5(b)] the predicted Anderson transition density for the case of exponential transfer-matrix elements depends on two length scales, namely a_H, an effective Bohr radius for the electron under consideration, and σ, the hard-sphere diameter; the relative influence of these competing length scales in determining the Anderson transition density is investigated.

Finally, in Sec. V we calculate the full mobility-edge trajectories for transfer-matrix elements of form $V(R)\sim R^{-n}$; particular emphasis is given to the cases $n=3$ and 5, corresponding, respectively, to dipolar and quadrupolar excitons.

II. SELF-CONSISTENT EQUATIONS

For fixed center-of-mass configurations $\{\mathbf{R}_i\}$, we assume that the system is specified by a tight-binding model. The excitation moves in the frozen configuration of atoms and has one possible characteristic energy, ϵ_i, associated with each atom i. When pertaining to electronic transport, ϵ_i is the energy of an electron bound to atom i, whereas in the excitonic case, ϵ_i is the difference in energy of the levels between which the excitation occurs. We assume that the site energies $\{\epsilon_i\}$ are random and independent, with some given probability distribution $P(\epsilon_i)$. In the limit in which site-diagonal disorder is absent, the site energies are the same for all i, $\epsilon_i=\epsilon_0$, corresponding to the trivial distribution $P(\epsilon_i)=\delta(\epsilon_i-\epsilon_0)$.

The matrix element which transfers the excitation from atom i to atom j is $-V(\mathbf{R}_i,\mathbf{R}_j)$, the range of which depends on the particular problem under investigation. For example, $V(R)\sim R^{-n}$ is applicable to multipolar excitons ($n=3$, 5, etc., corresponding to dipolar, quadrupolar, etc., excitons), and $V(R)\sim\exp(-\alpha R)$ is often deemed applicable to triplet excitons or to a discussion of electronic transport in the impurity band of a semiconductor. The source of off-diagonal disorder, or randomness in the transfer-matrix element, is taken to be the topological disorder inherent in the Boltzmann distribution for the centers of mass.

The Green function for the random tight-binding model satisfies

$$(E-\epsilon_i)G_{ik}(E)+\sum_j V(\mathbf{R}_i,\mathbf{R}_j)G_{jk}(E)=\delta_{ik} , \tag{2.1}$$

and the self-energy $S_i(E)$ is defined by

$$E-\epsilon_i-S_i(E)=[G_{ii}(E)]^{-1} . \tag{2.2}$$

In Feenberg (renormalized) perturbation theory,[23] the self-energy is written as a sum of terms, each of which corresponds to a nonrepeating path for the excitation transfer. The approach of Abou-Chacra *et al.*[14,15] is to consider solely the set of equations arising from the two-body, second-order perturbation terms (which are exact for a Bethe lattice), and to subsequently ignore the differences between the self-energies appearing on either side of any of these equations. One considers the approximate relation

$$S_i(E)=\sum_j V_{ij}[E-\epsilon_j-S_j(E)]^{-1}V_{ji} , \tag{2.3}$$

and enforces self-consistency by requiring that the probability distributions of S_i and S_j be self-consistent. We shall also adopt this approach for the topologically disordered systems under investigation. Elyutin,[4] and Fleishman and Stein,[5] have discussed the assumptions involved in applying the approximate relation (2.3) to topologically disordered systems. Elyutin[4] also points out that the self-consistent equation (2.3) is equivalent to a partial resummation of the class of graphs in the Matsubara-Toyozawa expansion[24] of the diagonal Green function, consisting of diagrams which have the topology of a Cayley tree with variable bonds that can, at each site, assume values from $K=0$ to $K=\infty$ (where K is the connectivity).

Localized and extended states are characterized by different distributions of $S_i(E)$ when E is complex and $\mathrm{Im}E$ tends to zero.[1,25] We examine the stability of localized states by looking for a solution to Eq. (2.3) in which $\mathrm{Im}[S_i(E)]$ is proportional to $\mathrm{Im}E$. Where such a solution exists the states are localized, and where no such solution exists the states are assumed to be delocalized. Equation (2.3) is separated into real and imaginary parts by writing

$$E=\tilde{R}+i\eta,\quad S_j(\tilde{R}+i\eta)=E_j-i\Delta_j\ . \tag{2.4}$$

We further take η to be very small and assume the states are localized, so that Δ_j is very small except on a set of measure zero. One obtains

$$E_i=\sum_j\frac{|V(\mathbf{R}_i,\mathbf{R}_j)|^2}{\tilde{R}-\epsilon_j-E_j}=\sum_j|V(\mathbf{R}_i,\mathbf{R}_j)|^2X_j\ , \tag{2.5a}$$

$$\Delta_i=\sum_j\frac{|V(\mathbf{R}_i,\mathbf{R}_j|^2(\eta+\Delta_j)}{(\tilde{R}-\epsilon_j-E_j)^2}=\sum_j|V(\mathbf{R}_i,\mathbf{R}_j)|^2Y_j\ , \tag{2.5b}$$

where X_j and Y_j are thus defined. Equation (2.5a) for E_i does not involve Δ_i, and the equation for Δ_i is a linear inhomogeneous equation which has a solution, provided the largest eigenvalue, λ, of the corresponding homogeneous equation,

$$\sum_j\left[\frac{|V(\mathbf{R}_i,\mathbf{R}_j)|^2}{(R-\epsilon_j-E_j)^2}-\lambda\delta_{ij}\right]\Delta_j=0\ , \tag{2.5c}$$

is less than unity; $\lambda=1$ corresponds to the limit of stability of localized states.

It is straightforward to obtain an expression for the joint probability distribution $f(E_i,\Delta_i)$ by averaging over the multicomponent reference system described in the Introduction. We specifically consider the Fourier transform $F(k_1,k_2)$ given by

$$F(k_1,k_2)=\int_{-\infty}^{+\infty}dx\int_0^{+\infty}dy\,f(x,y)\exp[i(k_1x+k_2y)]\ . \tag{2.6}$$

This is readily shown to be given by

$$F(k_1,k_2)=\left\langle\prod_j\int_{-\infty}^{+\infty}dX_j\int_0^{+\infty}dY_j\,Q(X_j,Y_j)\exp\left[i\sum_j|V(\mathbf{R}_i,\mathbf{R}_j)|^2(k_1X_j+k_2Y_j)\right]\right\rangle_\Gamma \tag{2.7a}$$

$$=\left\langle\exp\left[i\sum_j|V(\mathbf{R}_i,\mathbf{R}_j)|^2(k_1X_j+k_2Y_j)\right]\right\rangle_{\Gamma,\{X_j,Y_j\}}\ , \tag{2.7b}$$

where $\langle\ \rangle_\Gamma$ denotes an average over the phase space Γ of particles interacting via the chosen classical interaction potential; $Q(X_j,Y_j)$ is the joint probability distribution of the quantities X_j and Y_j defined by (2.5), and is given by

$$Q(X_j,Y_j)=\int_{-\infty}^{+\infty}d\epsilon_j\,P(\epsilon_j)f(\tilde{R}-\epsilon_j-X_j^{-1},\,Y_jX_j^{-2}-\eta)X_j^{-4}\ . \tag{2.8}$$

Equation (2.7b), written as an average over Γ and $\{X_j,Y_j\}$, is formally equivalent to[26]

$$F(k_1,k_2)=\exp\left[\frac{-\Delta\mu}{k_BT}\right]\ , \tag{2.9}$$

where T is the absolute temperature; $\Delta\mu$ is the excess interaction chemical potential for atom i dissolved in the multicomponent reference fluid, when it interacts with the atoms in the fluid via the complex "potential" $U(\mathbf{R}_i,\mathbf{R}_j)$ embodied in the argument of the exponential in (2.7b) and give explicitly by

$$U(\mathbf{R}_i,\mathbf{R}_j)=-ik_BT\,|V(\mathbf{R}_i,\mathbf{R}_j)|^2(k_1X_j+k_2Y_j)\ . \tag{2.10}$$

The interaction chemical potential $\Delta\mu$ may be calculated by the standard Onsager-Kirkwood strategy of liquid-state theory, whereby one "charges up" the interaction of atom i with the atoms in the reference fluid.[16] Assuming, for convenience, that $V(\mathbf{R}_i,\mathbf{R}_j)=V(|\mathbf{R}_i-\mathbf{R}_j|)$, one finds

$$\Delta\mu=\rho\int_0^1d\lambda\int d\mathbf{R}\int dX\times\int dY\,U(R)Q(X,Y)g(R;\lambda U(R))\ , \tag{2.11}$$

where ρ is the mean number density and $g(R;\lambda U(R))$ is the pair distribution function for two atoms, separated by a distance R in the reference fluid and interacting via $U_0(R)+U(R)$. Given the choice of reference system, Eq. (2.11) is exact. To proceed further we must specify the pair correlation function. Many elaborate approximations to this function have been devised in liquid-state theory (for a review, see Ref. 27), one of the simplest of which is the so-called primitive exponential (EXP) approximation,[16]

$$g(R;\lambda U(R))\cong g_0(R)\exp\left[-\frac{\lambda U(R)}{k_BT}\right]\ , \tag{2.12}$$

where $g_0(R)$ is the pair distribution function for particles interacting via $U_0(R)$ alone. The primitive EXP approximation has the virtue of being asymptotically exact at low densities, and we employ it as a matter of convenience.

Using (2.12), we therefore find

$$F(k_1,k_2)=\exp\left[4\pi\rho\int_0^\infty dR\,R^2g_0(R)[Q(k_1|V(R)|^2,k_2|V(R)|^2)-1]\right]\ , \tag{2.13a}$$

where $Q(k_1,k_2)$ is the Fourier transform of $Q(X,Y)$. It is a straightforward matter to show that

$$Q(k_1\,|\,V(R)\,|^2,k_2\,|\,V(R)\,|^2)=\int_{-\infty}^{+\infty}\frac{dk_1'}{2\pi}\int_{-\infty}^{+\infty}dx\,P(k_1')F(k_1';k_2\,|\,V(R)\,|^2/x^2)$$

$$\times\exp\left[-ik_1'(\tilde{R}-x)+ik_2\frac{|\,V(R)\,|^2}{x^2}\eta+ik_1\frac{|\,V(R)\,|^2}{x}\right], \qquad (2.13b)$$

where $P(k)$ is the transform of $P(\epsilon_i)$. Equations (2.13) constitute a nonlinear integral equation for $F(k_1,k_2)$. When solutions to this equation exist, the states are localized; when no solutions exist, the states are delocalized. If we employ Anderson's upper-limit approximation,[1,14] whereby one neglects the real part of the self-energy, Eqs. (2.13) reduce precisely to an integral equation for $F(k)$, the Fourier-transformed probability distribution of Δ_i, derived by us in a previous publication.[13] The critical density above which states at the chosen energy become delocalized for fixed parameters specifying $V(R)$ (or, equivalently, the value of the strength of the transfer-matrix element needed to destroy localized states at a given energy and number density) was found within the upper-limit approximation by studying the asymptotic small-k behavior of $F(k)$. Similarly, here we study the full integral equation for $F(k_1,k_2)$, for small values of k_2. Before proceeding with this analysis, note that if we entirely ignore the structure of the medium, by setting $g_0(R)=1$, Eqs. (2.13) reduce to that of Fleishman and Stein,[3] who consider the self-consistent theory of localization for a completely random distribution of site positions; for an exponential transfer-matrix element, these authors have discussed the Anderson transition for this model within the confines of the upper-limit approximation. In passing, we note that the relatively simple connection between (2.13), in which the structure of the medium is taken into account, and that pertaining to a completely random distribution of atoms, is, in essence, due to our use of the primitive EXP approximation. Systematic extension of the analysis beyond the EXP approximation is possible and does not appear to diminish the tractability of the resultant integral equation for $F(k_1,k_2)$. To study Eqs. (2.13), we assume[14] that for small k_2 we can write

$$F(k_1,k_2)\approx F_0(k_1)-k_2^{\beta}B(k_1)+\cdots\,,\quad \beta\in(0,1]\,, \qquad (2.14)$$

where

$$F_0(k_1)=F(k_1,k_2=0)=\int_{-\infty}^{+\infty}dE_i\exp(ik_1E_i)\left[\int_0^{+\infty}d\Delta_i\,f(E_i,\Delta_i)\right] \qquad (2.15)$$

is essentially the Fourier transform of the average probability distribution for the real part of the self-energy. The exponent β must be positive, as $\lim_{k_2\to 0}F(k_1,k_2)=F_0(k_1)$, and cannot be greater than unity, since an exponent greater than unity would imply that the mean value of Δ_i is zero. From Eqs. (2.13), we find that $F_0(k_1)$ satisfies the nonlinear integral equation:

$$F_0(k_1)=\exp\left[-4\pi\rho\int_0^{\infty}dR\,R^2g_0(R)\left[1-\int_{-\infty}^{\infty}\frac{dk_1'}{2\pi}\int_{-\infty}^{\infty}dx\,P(k_1')F_0(k_1')\exp[-ik_1'(\tilde{R}-x)+ik_1\,|\,V(R)\,|^2x^{-1}]\right]\right]. \qquad (2.16)$$

Substitution of (2.14) into (2.13) yields

$$F(k_1,k_2)=F_0(k_1)-4\pi\rho k_2^{\beta}F_0(k_1)\int_0^{+\infty}dR\,R^2g_0(R)\,|\,V(R)\,|^{2\beta}$$

$$\times\int_{-\infty}^{+\infty}\frac{dk_1'}{2\pi}P(k_1')B(k_1')$$

$$\times\int_{-\infty}^{+\infty}dx\,|\,x\,|^{-2\beta}\exp[-ik_1'(\tilde{R}-x)+ik_1\,|\,V(R)\,|^2x^{-1}]+\cdots\,, \qquad (2.17)$$

and comparison of (2.17) with (2.14) shows that $B(k_1)$ satisfies a linear homogeneous integral equation; this equation implies a relationship between ρ, β, $\tilde{R}$, and the parameters specifying $V(R)$ [analogously to the corresponding equation derived within the upper-limit approximation, namely Eq. (4) of Ref. 13]. The integral equation for $B(k_1)$ may be written in a more elegant form by defining the functions $A(x)$ and $W(x)$:

$$A(x)=\int_{-\infty}^{+\infty}\frac{dk}{2\pi}P(k)B(k)\exp[ik(x-\tilde{R})]\,, \qquad (2.18a)$$

$$W(x)=\int_{-\infty}^{+\infty}\frac{dk}{2\pi}P(k)F_0(k)\exp[ik(x-\tilde{R})]\,. \qquad (2.18b)$$

From (2.17) and (2.14) we find that $A(y)$ satisfies

$$A(y)=4\pi\rho\int_0^{+\infty}dR\,R^2g_0(R)\,|\,V(R)\,|^{2\beta}\left[\int_{-\infty}^{+\infty}dx\,|\,x\,|^{-2\beta}A(x)W(y+|\,V(R)\,|^2/x)\right], \qquad (2.19)$$

which is of the form

$$A(y)=4\pi\rho\int_{-\infty}^{+\infty}\mathscr{C}(y,x)A(x)dx\ , \tag{2.20a}$$

with

$$\mathscr{C}(y,x)=\int_0^{+\infty}dR\,R^2g_0(R)\,|\,V(R)\,|^{2\beta}W(y+|\,V(R)\,|^2/x)\,|\,x\,|^{-2\beta}\,. \tag{2.20b}$$

Analogously to the upper-limit analysis, there is a maximum value of ρ for which (2.19) has a solution, and that is the critical density, ρ_c, for given energy $\widetilde{R}$, given parameters specifying the transfer-matrix element $V(R)$, and for the particular distribution of site energies, $P(\epsilon)$. For $\rho>\rho_c$, states at the chosen $\widetilde{R}$ are delocalized (actually, this comment presumes that ρ_c is a monotonically increasing function of $\widetilde{R}$ above the Anderson transition density, which is indeed found to be the case). Following the method of Abou-Chacra *et al.*,[14] it can be shown that the critical value of the exponent β is $\frac{1}{2}$: If (2.19) possesses a solution for given β, ρ, and given parameters specifying $V(R)$, then so does the corresponding adjoint equation,

$$a(x)=4\pi\rho\int_0^{+\infty}dR\,R^2g_0(R)\,|\,V(R)\,|^{2\beta}\left[\int_{-\infty}^{+\infty}dy\,a(y)W(y+|\,V(R)\,|^2/x)\,|\,x\,|^{-2\beta}\right]. \tag{2.21}$$

The solution of the adjoint equation may be related to the solution of the original equation for a different value of β, but with the same values of ρ and the parameters specifying $V(R)$, by

$$a_\beta(x)=|\,x\,|^{-2\beta}\int_0^{+\infty}dR\,R^2g_0(R)\,|\,V(R)\,|^{2\beta}A_{1-\beta}(\,|\,V(R)\,|^2/x)\,. \tag{2.22}$$

Substitution of (2.22) into (2.21) gives back (2.19) with β replaced by $1-\beta$. The eigenvalue of (2.19) is thus an even function of $\beta-\frac{1}{2}$; the point $\beta=\frac{1}{2}$ is, in fact, a minimum, and thus ρ attains a maximum for $\beta=\beta_c=\frac{1}{2}$. Thus we must solve (2.19) with $\beta=\frac{1}{2}$ to find ρ_c for given $\widetilde{R}$, $P(\epsilon)$, and $V(R)$; from this solution we obtain ρ_c as a function of $\widetilde{R}$ [for fixed parameters specifying $V(R)$], or, equivalently, the mobility edge $\widetilde{R}_c$ as a function of density. To specify the kernel of the integral equation (2.19), we require knowledge of $F_0(k_1)$, as given by the solution of the nonlinear integral equation, (2.16), with an assumed form of the distribution $P(\epsilon)$. This problem, in itself, is complicated; in the following section, however, we obtain an approximate solution for $F_0(k_1)$ [or its Fourier transform $f_0(E_i)$ defined in (2.15)] which is asymptotically exact for small k_1 (large E_i).

III. APPROXIMATE SOLUTION FOR $F_0(k_1)$ AND CONNECTION WITH THE MEAN SPHERICAL INTEGRAL EQUATION

For convenience, we assume the distribution of site energies to be of Lorentzian form,

$$P(\epsilon)=\frac{\lambda\pi^{-1}}{\lambda^2+(\epsilon-\epsilon_0)^2}\ , \tag{3.1}$$

where λ determines the half-width of broadening due to site-diagonal disorder; $P(k)$ is then a Cauchy distribution, $P(k)=\exp(-\lambda\,|\,k\,|+ik\epsilon_0)$. Equation (2.16), to be solved for $F_0(k_1)$, becomes

$$\ln F_0(k_1)=-4\pi\rho\int_0^{+\infty}dR\,R^2g_0(R)\left[1-\int_{-\infty}^{+\infty}\frac{dk_1'}{2\pi}\int_{-\infty}^{+\infty}dx\,F_0(k_1')\exp[-\lambda\,|\,k_1'\,|-ik_1'(\widetilde{E}-x)+ik_1\,|\,V(R)\,|^2x^{-1}]\right], \tag{3.2}$$

where $\widetilde{E}=\widetilde{R}-\epsilon_0$. It may be shown that $F_0(k_1)$ asymptotically approaches a Cauchy distribution for small k_1; if we assume

$$F_0(k_1)=\exp(-\alpha\,|\,k_1\,|+ik_1x_0),\quad \alpha\geq 0 \tag{3.3}$$

the right-hand side of (3.2) is equal to

$$-4\pi\rho\int_0^{+\infty}dR\,R^2g_0(R)\left[1-\exp\left[[-(\alpha+\lambda)\,|\,k_1\,|+i(\widetilde{E}-x_0)k_1]\frac{|\,V(R)\,|^2}{(\alpha+\lambda)^2+(\widetilde{E}-x_0)^2}\right]\right]. \tag{3.4}$$

Assuming further that $V(R)$ falls off at large R faster than $R^{-3/2}$, we may expand the exponential appearing in (3.4), as all integrals of the form

$$\int_0^{+\infty}dR\,R^2g_0\,|\,V(R)\,|^{2n}\ \ (n>0)$$

are bounded. Retaining terms linear in k_1 only, (3.4) reduces to the form

$$[-(\alpha+\lambda)\,|\,k_1\,|+i(\widetilde{E}-x_0)k_1]\frac{J^2(\rho)}{(\alpha+\lambda)^2+(\widetilde{E}-x_0)^2}\ , \tag{3.5}$$

where

$$J^2(\rho)=4\pi\rho\int_0^{\infty}dR\,R^2g_0(R)\,|\,V(R)\,|^2\,. \tag{3.6}$$

Employing (3.3) on the left-hand side of (3.2), and equating it with (3.5), we show that $F_0(k_1)$ asymptotically approaches a Cauchy distribution, provided that the following pair of equations for α and x_0 are satisfied:

$$\alpha=\frac{(\alpha+\lambda)J^2(\rho)}{(\alpha+\lambda)^2+(\tilde{E}-x_0)^2}, \quad x_0=\frac{(\tilde{E}-x_0)J^2(\rho)}{(\alpha+\lambda)^2+(\tilde{E}-x_0)^2} . \tag{3.7}$$

In the following sections we will approximate $F_0(k_1)$ by the Cauchy form (3.3), with α and x_0 given by the solution of (3.7); we shall be specifically concerned with two limiting cases:

(1) $\tilde{E}=0$ ($\tilde{R}=\epsilon_0$), for which (3.7) yields

$$\alpha=\tfrac{1}{2}\{-\lambda+[\lambda^2+4J^2(\rho)]^{1/2}\}, \quad x_0=0 . \tag{3.8}$$

This case is of interest in respect of determining the critical density for the Anderson transition, ρ_A, namely that density at which the mobility edges coalesce at the band center and below which all states in the band are localized. For transfer-matrix elements of exponential form, and also for $V(R)\sim R^{-n}$ as applicable to multipolar excitons, we expect ρ_A to be sufficiently low that the band center occurs at $\tilde{E}=0$. We shall return to this problem in the following section.

(2) $\lambda=0$, corresponding to neglect of site-diagonal disorder $[P(\epsilon)=\delta(\epsilon-\epsilon_0)]$. For this case, (3.7) yields

$$\begin{aligned} &\alpha=\tfrac{1}{2}[4J^2(\rho)-\tilde{E}^2]^{1/2}, \quad |\tilde{E}|\leq 2J(\rho) \\ &x_0=\tfrac{1}{2}\tilde{E} , \end{aligned} \tag{3.9a}$$

$$f_0(E_i)=\frac{\alpha\pi^{-1}}{\alpha^2+(E_i-\frac{1}{2}\tilde{E})^2} . \tag{3.9b}$$

Equations (3.9) will be used in the final section to calculate mobility-edge trajectories for multipolar excitons with off-diagonal disorder alone.

The approximate analysis of $f_0(E_i)$ developed here provides a direct connection between the density of states resultant from the self-consistent theory and that predicted from solution of the quantum mean spherical integral equation for topologically disordered systems.[17–22] The density of states $\rho(\tilde{R})$ is defined by

$$\rho(\tilde{R})=-\frac{1}{\pi}\langle \operatorname{Im}G_{ii}^{+}(\tilde{R})\rangle , \tag{3.10}$$

where

$$G_{ii}^{+}(\tilde{R})=\lim_{\eta\to 0}G_{ii}(\tilde{R}+i\eta) .$$

For simplicity, we consider the case when lateral disorder alone is present $[P(\epsilon)=\delta(\epsilon-\epsilon_0)]$; from (2.2), (2.4), and (3.10), we obtain

$$\rho(\tilde{R})=\pi^{-1}\lim_{\eta\to 0}\int_{-\infty}^{+\infty}dE_i\int_0^{+\infty}d\Delta_i\frac{\eta+\Delta_i}{(\tilde{E}-E_i)^2+(\eta+\Delta_i)^2} \times f(E_i,\Delta_i) . \tag{3.11}$$

If $\tilde{R}$ corresponds to a localized state, the solution to (2.5b) for the imaginary part of the self-energy will be of the form $\Delta_i\propto\eta$; we therefore expect $f(E_i,\Delta_i)$ to take the form

$$f(E_i,\Delta_i)=f'(E_i)\delta(\Delta_i-c\eta) ,$$

and, from (2.15) (with η infinitesimal), we find

$$f'(E_i)=\int_0^{+\infty}d\Delta_i f(E_i,\Delta_i)=f_0(E_i) .$$

Thus, if $\tilde{R}$ corresponds to a localized state, we have

$$\rho(\tilde{R})=f_0(\tilde{E}) . \tag{3.12}$$

In particular, if we employ the approximate form of f_0, Eqs. (3.9), there results

$$\rho(\tilde{R})=\frac{2}{\pi}\frac{1}{4J^2(\rho)}[4J^2(\rho)-\tilde{E}^2]^{1/2}, \quad |\tilde{E}|\leq 2J(\rho) . \tag{3.13}$$

Equation (3.13) is a familiar semielliptic Hubbard density of states[28] with bandwidth

$$B=2J(\rho) . \tag{3.14}$$

From conventional liquid-state theory, it is well known that the low-density behavior of $g_0(R;\rho)$ is given by[16]

$$g_0(R;\rho)=\exp\left[-\frac{U_0(R)}{k_BT}\right]+O(\rho) , \tag{3.15}$$

where $U_0(R)$ is the chosen spherically symmetric, classical interaction potential employed in our multicomponent reference fluid. From (3.6) and (3.14), it is therefore clear that the spectral bandwidth B scales with $\rho^{1/2}$ in the low-density regime. As an illustration, consider the simple exponential transfer-matrix element applicable to a discussion of electronic transport,

$$V(R)=-V_0\exp(-R/a_H) , \tag{3.16}$$

where a_H is an effective Bohr radius for the electron under consideration. If $U_0(R)$ is that appropriate to a hard-sphere fluid with hard-sphere diameter σ, then $g_0(R;\rho)=\Theta(r-\sigma)+O(\rho)$, where $\Theta(x)$ is the unit step function. In this low-density regime, we thus find

$$B^2=4J^2(\rho)=4\pi V_0^2(\rho a_H^3)\left[2\left[\frac{\sigma}{a_H}\right]^2+2\left[\frac{\sigma}{a_H}\right]+1\right] \times\exp\left[-2\frac{\sigma}{a_H}\right] . \tag{3.17}$$

The Hubbard form of (3.13) need hardly surprise us when one recalls that the Hubbard Green function results from that appropriate to a Bethe lattice of connectivity K, in the limit that K tends to infinity with the Bethe-lattice bandwidth, $B=2V_0\sqrt{K}$, remaining constant.[28] Recall further, from Sec. II, that the average diagonal Green function resulting from the self-consistent equation (2.5) consists of a partial summation of all diagrams that have the topology of a Cayley tree. We can therefore use (3.17) to determine the effective connectivity, defined through $B=2V_0(K_{\text{eff}})^{1/2}$, for the topologically disordered system under consideration. It is apparent that $K_{\text{eff}}\propto\rho a_H^3$, and that $K_{\text{eff}}\ll 1$ for low concentrations; this conclusion is commensurate with the work of Elyutin.[4]

Consider now transfer-matrix elements of the form

$$V(R)=\gamma R^{-n}\ , \tag{3.18}$$

where $n=3$ (5) corresponds to dipolar (quadrupolar) electronic excitons. Assuming $U_0(R)$, as is appropriate for a hard-sphere (HS) fluid, we find

$$\begin{aligned} J(\rho^*)&=|\gamma^*|[\rho^* I_{2n}^{\mathrm{HS}}(\rho^*)]^{1/2}\ ,\\ I_{2n}^{\mathrm{HS}}(\rho^*)&=4\pi\int_0^\infty dy\, g_0(y;\rho^*)y^{2(1-n)}\ , \end{aligned} \tag{3.19}$$

where $\rho^*=\rho\sigma^3$ is the reduced density, $y=R/\sigma$, and $\gamma^*=\gamma\sigma^{-n}$; extended virial expansions for the integrals $I_{2n}^{\mathrm{HS}}(\rho^*)$ have been given by Larsen *et al.*[29] To make contact with the quantum mean spherical model for dipolar or quadrupolar excitons,[22] let

$$\begin{aligned} |\gamma^*|&=\frac{1}{\sqrt{2}}\alpha_0^*\epsilon_0 \ \text{ for } n=3\ ,\\ |\gamma^*|&=\sqrt{14}C_0^*\epsilon_0 \ \text{ for } n=5\ . \end{aligned} \tag{3.20}$$

Here, $\alpha_0^*=\alpha_0\sigma^{-3}$ ($C_0^*=C_0\sigma^{-5}$), where α_0 (C_0) is the isolated atom, zero-frequency dipole (quadrupole) polarizability for the particular dipolar (quadrupole) electronic transition under investigation, e.g., an $s\to p$ ($s\to d$) electronic transition.[22] For purposes of illustration, consider specifically the mean spherical integral equation for dipolar excitons.[17-21] There, one calculates the renormalized dipole polarizability $\alpha(\tilde{R})=\alpha'(\tilde{R})+i\alpha''(\tilde{R})$, which corresponds to the frequency-dependent renormalized polarizability of an atom in the condensed phase, the imaginary part of which is related to the dipolar optical-absorption spectrum of the fluid. At low densities one may legitimately neglect irreducible three-body contributions to $\alpha(\tilde{R})$ (this is tantamount to neglect of the irreducible three-body integral b in Ref. 20, which determines the spectral shift). $\alpha''^*(\tilde{R})=\alpha''(\tilde{R})\sigma^{-3}$ is then given from the mean spherical integral equation theory by

$$\alpha''^*(\tilde{R})=\frac{\alpha_0^*\epsilon_0}{4J^2(\rho)}\left\{4J^2(\rho)-\left[\frac{\epsilon_0}{2}\right]^2\left[\left[\frac{\tilde{R}}{\epsilon_0}\right]^2-1\right]^2\right\}^{1/2}\ . \tag{3.21}$$

Considering only the band in the vicinity of $\tilde{R}=+\epsilon_0$, it is clear that

$$\tilde{R}^2-\epsilon_0^2=(\tilde{R}+\epsilon_0)\tilde{E}\cong 2\epsilon_0\tilde{E}\ ,$$

whence

$$\alpha''^*(\tilde{R})\cong\rho(\tilde{R})\left[\frac{\pi\alpha_0^*\epsilon_0}{2}\right]\ , \tag{3.22}$$

where $\rho(\tilde{R})$ is given by (3.13) and $J(\rho)$ by (3.19). This direct connection between the mean spherical integral equation result for $\alpha''(\tilde{R})$, and the approximate density of states resultant from the self-consistent theory, is entirely due to the graphical structure of $\alpha(\tilde{R})$: Under neglect of irreducible m-body graphs ($m\geq 3$), $\alpha(\tilde{R})$ consists of a partial summation of all graphs that have the topology of a Cayley tree with variable connectivity.

IV. THE ANDERSON TRANSITION

To determine the Anderson transition density ρ_A, we may use Fredholm Theory[30] to solve the eigenvalue problem (2.19), with $\beta=\beta_c=\frac{1}{2}$ and $\tilde{E}=0$. To estimate ρ_A we consider here the first-order term in the Fredholm series corresponding to (2.19); a lengthly analysis of the integral equation, for the transfer-matrix element $V(R)\sim R^{-5}$, shows that the Anderson transition density determined from the first-order theory is in error by only ~15%.

First-order Fredholm theory applied to (2.19) yields

$$1=4\pi\rho_A\int_0^{+\infty}dR\,R^2g_0(R)\,|V(R)|\left[\int_{-\infty}^{+\infty}\frac{dx}{|x|}W(x+|V(R)|^2/x)\right]\ , \tag{4.1}$$

where $W(x)$ is given by (2.18b); for convenience, we regard $P(\epsilon)$ as given by the Lorentzian distribution, (3.1), and we take $F_0(k)$ and α as given by (3.3) and (3.8), respectively. Equation (4.1) becomes

$$1=4\rho_A(\alpha+\lambda)\int_0^{+\infty}dR\,R^2g_0(R)\,|V(R)|\left[\int_{-\infty}^{+\infty}\frac{dx}{|x|}\frac{1}{(\alpha+\lambda)^2+[x+|V(R)|^2/x]^2}\right]\ , \tag{4.2}$$

and with a simple transformation of variables, this may be cast in the form

$$1=16\rho_A(\alpha+\lambda)\int_1^{+\infty}dz\frac{Q(z)}{(z^2-1)^{1/2}}\ , \tag{4.3a}$$

$$Q(z)=\int_0^{+\infty}dR\,R^2g_0(R)\frac{|V(R)|}{(\alpha+\lambda)^2+4|V(R)|^2z^2}\ . \tag{4.3b}$$

We consider separately the solution of (4.3) for transfer-matrix elements appropriate to multipolar excitons, and transfer-matrix elements of exponential form.

A. Multipolar excitons

Consider transfer-matrix elements of the form (3.18). The pair distribution function $g_0(R)$ is assumed to be that appropriate to the low-density limit of a hard-sphere fluid, as discussed in Sec. III. [Alternatively, one could use the general form (3.15) with $U_0(R)$ given by, say, a Lennard-Jones (12-6) potential; this would introduce a temperature dependence into the problem which is absent in the case of a hard-sphere reference fluid.]

From (4.3b) it is clear that we require $n>3$ to avoid a large-R divergence in the $Q(z)$ integral. Thus one predicts that dipolar excitons (corresponding to $n=3$) lack an Anderson transition, so that states at the band center are always extended. As we discuss in the final section, one finds, in general, that dipolar excitons are predicted to be always delocalized.

For $n>3$ we find that

$$Q(z)=\frac{|\gamma^*|^{3/n}}{n}\sigma^3\int_0^{|\gamma^*|}\frac{V^{-3/n}}{(\alpha+\lambda)^2+4V^2z^2}dV\,, \tag{4.4}$$

where $\gamma^*=\gamma\sigma^{-n}$. Using the fact that[31]

$$\int_1^{+\infty}dz\frac{1}{(z^2-1)^{1/2}(a^2+z^2)}=F(1,1;\tfrac{3}{2};-a^2)$$
$$=\frac{\sinh^{-1}a}{a(1+a)^{1/2}}\,, \tag{4.5}$$

where $F(\alpha,\beta;\gamma;z)$ is the Gauss (nondegenerate) hypergeometric function, the first-order Fredholm equation, (4.3a), becomes

$$1=\frac{8\rho_A^*}{n}\left[\frac{\alpha+\lambda}{2|\gamma^*|}\right]^{-3/n}\int_C^{+\infty}y(\sinh y)^{(3-n)/n}dy,$$
$$n>3,\quad C=\sinh^{-1}\left[\frac{\alpha+\lambda}{2|\gamma^*|}\right]. \tag{4.6}$$

$\rho_A^*=\rho_A\sigma^3$ is the dimensionless reduced Anderson density. Consider now the case $\lambda=0$, where the site-diagonal disorder is absent and we have lateral disorder alone. Note that since

$$\left[\frac{\alpha}{2|\gamma^*|}\right]^2=\left[\frac{J(\rho)}{2|\gamma^*|}\right]^2=\pi\rho^*\int_1^{+\infty}dy\,g_0(y)y^{2(1-n)}\,, \tag{4.7}$$

ρ_A^* as determined by (4.6) is independent of γ^*. With negligible numerical error, we can set the lower limit of the integral in (4.6) to zero, and with $g_0(y)=\Theta(y-1)$ we find

$$1=\frac{2\pi}{n}\rho_A^*\left[\frac{\pi\rho_A^*}{2n-3}\right]^{-3/2n}B(3/2n;(n-3)/2n)\tan\left[\frac{3\pi}{2n}\right]\qquad n>3 \tag{4.8}$$

where $B(x,y)=\Gamma(x)\Gamma(y)/\Gamma(x+y)$ is the beta function.

Values for the reduced Anderson density predicted by (4.8) are shown in Table I for $n\geq 5$. As expected, on physical grounds, we see that as the transfer-matrix element becomes shorter ranged, the Anderson density progressively increases. It is straightforward to include the full $g_0(R)$ in the calculation of $J(\rho)$, as extended virial expansions for the $I_{2n}^{\rm HS}(\rho^*)$ integrals [cf. (4.7) and (3.19)] are available.[29] For $n=5$ and 7, correspondingly, respectively, to pure quadrupolar and octupolar excitons, inclusion of such has a minor effect on the predicted Anderson transition density, essentially because this occurs at a very low reduced density.

TABLE I. Anderson transition density ρ_A^* as a function of n for $V(R)\sim R^{-n}$, and for pure off-diagonal disorder.

n	ρ_A^*
5	0.0174
7	0.0825
9	0.1690
12	0.3120
14	0.4107

For $n>12$ (and, in particular, for the case $n=14$, which may be of relevance to the localization of high-frequency phonons in amorphous materials[12,13]), the predicted ρ_A is sufficiently large, so that a more realistic assessment of the effects of including fluid structure would likely be better achieved by using a more adequate representation of the pair correlation function $g_0(R)$ in (4.1).

To investigate the relative influence of site-diagonal disorder on the predicted Anderson transition density, we return to (4.6). If $\lambda/|\gamma^*|$ is large, it is not a good approximation to set the lower limit of the integral to zero; we have therefore solved (4.6) numerically. Further, note from (3.8) that

$$\frac{\alpha+\lambda}{|\gamma^*|}=\frac{1}{2|\gamma^*|}\left[\lambda+\left[\lambda^2+\frac{16\pi\rho^*|\gamma^*|^2}{2n-3}\right]^{1/2}\right], \tag{4.9}$$

whence ρ_A^* depends solely on the dimensionless parameter $D=\lambda/|\gamma^*|$. For all n ($n>3$) we find that, as the degree of site-diagonal disorder embodied in the parameter D increases, the Anderson density increases commensurately; explicit results are shown in Fig. 1 for $n=5$. In this case we may relate γ^* to the parameters of the mean spherical model for pure quadrupolar electronic excitons,[22] as given by (3.20). Typically $C_0^*\cong 10^{-4}$, and for an electronic transition, ϵ_0 is typically of the order of 10^5 cm^{-1}; thus γ^* is of the order of 40 cm^{-1}. The parameter λ is the half-width at half-height of the broadening of the electronic quadrupole-allowed resonance, arising from

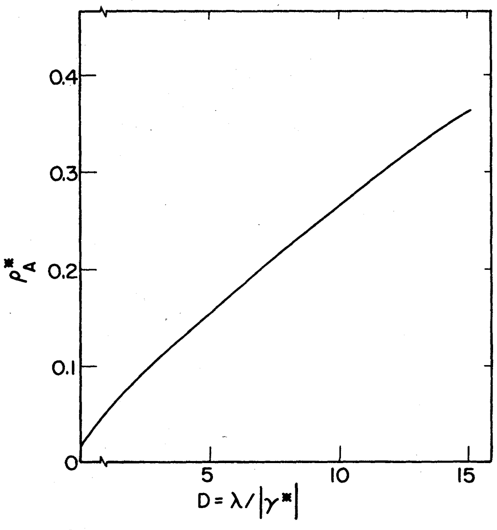

FIG. 1. Influence of site-diagonal disorder on the Anderson transition density ρ_A^* as a function of $D=\lambda/|\gamma^*|$ for $V(R)\sim R^{-5}$, corresponding to pure quadrupolar excitons. $D=0$ corresponds to the case of pure off-diagonal disorder.

mechanisms not involved in the excitation transfer under investigation, such as Doppler broadening or interatomic broadening processes other than those which arise from the quadrupolar transfer-matrix element. For a gas consisting of atoms with hard-sphere diameter of $\sigma=3.4$ Å (as appropriate to argon[32]), at a pressure of 1 atm and at $T=273$ K, the reduced density $\rho^*=10^{-3}$; thus the value of ρ_A^* predicted from (4.6) with $\lambda=0$, namely $\rho_A^*=0.0174$, corresponds to a pressure of about 17 atm. At such a pressure we expect the site-energy distribution to be determined primarily by collisional broadening, dominated by short-ranged, repulsive interatomic forces, and we typically expect λ to be of the order of 1—2 cm^{-1}; thus $D \ll 1$, and the influence of site-diagonal disorder on ρ_A^* should be very small.

In certain circumstances, of course, $\lambda/|\gamma^*|$ may be large. For example, if one considers an electronic transition in an atom of species A, embedded randomly at low concentration in a host matrix of species B, the influence of the host-matrix atoms on the distribution of site energies in species A may lead to large values of λ, thereby increasing the relative importance of site-diagonal disorder in determining ρ_A^*.

TABLE II. Calculated values of $\rho_A^{1/3}a_H=R_A^{-1}(0)$ for the pure exponential transfer-matrix element, (3.16), and for the modified exponential form, (4.13), with off-diagonal disorder alone and hard-sphere diameter $\sigma=0$.

	$\rho_A^{1/3}a_H$ (pure exponential)	$\rho_A^{1/3}a_H$ (modified exponential)
Present work	0.144	0.12
Elyutin[a]	0.076	0.09
Elyutin[b]	0.17 ± 0.02	
Ching and Huber[c]	0.37 ± 0.08	
Fertis *et al.*[d]		0.16
Puri and Odagaki[e]		0.25
Odagaki[f]	0.10	

[a] Reference 4.

[b] Reference 10.

[c] Reference 9.

[d] Reference 8. The form of the modified exponential transfer-matrix element used is, in fact, slightly different from our (4.13).

[e] Reference 5.

[f] Reference 6.

B. Transfer-matrix elements of exponential form

From Eqs. (4.3) and (4.6), and in the absence of site-diagonal disorder ($\lambda=0$), the first-order Fredholm equation may, in general, be cast in the form

$$1=8\rho_A\int_0^{+\infty}dR\,R^2g_0(R)\left[1+\left[\frac{J}{2|V(R)|}\right]^2\right]^{1/2}\times\sinh^{-1}\left[\frac{J}{2|V(R)|}\right]. \tag{4.10}$$

For the simple exponential transfer-matrix element given by (3.16), and with $g_0(R)=\Theta(R-\sigma)$, Eq. (4.10) becomes

$$1=8(\rho_A a_H^3)\int_C^{+\infty}y\frac{\{\ln[(2V_0/J)\sinh y]\}^2}{\sinh y}dy\,,\qquad C=\sinh^{-1}\left[\frac{J}{2V_0}\exp\left[\frac{\sigma}{a_H}\right]\right], \tag{4.11}$$

where $J^2(\rho)$ is given by (3.17). By virtue of the fact that J/V_0 is independent of V_0, we see that the Anderson transition density for exponential transfer, as given by the solution of (4.11), is also independent of V_0 (cf. the case of multipolar excitons where ρ_A is independent of γ). In agreement with Puri and Odagaki,[5(b)] and in contrast to other approaches,[3–10] our model has two dimensionless parameters, namely $\rho_A a_H^3$ and σ/a_H. The approximation inherent in most traditional approaches is that of implicitly setting $\sigma=0$, an approximation which is clearly justified if $\sigma/a_H \ll 1$. For example, in discussing the phenomenon of metallic condensation of a lattice of impurity centers in a host material,[33] $a_H \simeq \epsilon_{\text{st}}\hbar^2/m^*e^2$, where ϵ_{st} is the static dielectric constant of the host material, and m^* is the effective mass of an electron in the host conduction band. For many systems exhibiting a metal-insulator (M-I) transition, ϵ_{st} is very large and $\sigma/a_H \ll 1$ is indeed satisfied (for a comprehensive review of effective Bohr radii in systems exhibiting a M-I transition, see Ref. 34). For M-I transitions occurring in low-dielectric-constant host materials, however, the condition $\sigma/a_H \ll 1$ is not satisfied. For example, for the Xe:Hg system, $a_H=1.3$ Å,[35] for the Ar:Cu systems, $a_H \cong 0.9$ Å,[36] and for Ar:Na, $a_H \cong 2.08$ Å;[37] typically, we expect the hard-sphere diameter of the impurity species, σ, to be 3 or 4 Å. The formulation we have given enables us to assess the relative influence of the competing length scales, a_H and σ.

The solution to (4.11) may be cast in the form

$$\rho_A^{-1/3}=R_A(\sigma/a_H)a_H\,, \tag{4.12}$$

where $\rho_A^{-1/3}$ is essentially a measure of the mean separation between atoms at the Anderson transition; numerical solution of (4.11) gives the function $R_A(\sigma/a_H)$. For $\sigma=0$, we may compare our calculated value of $R_A^{-1}(0)=0.144$ with those obtained from a variety of other studies; the results are displayed in Table II. We see that our predicted value of $R_A^{-1}(0)$ falls fairly well within the domain of previously estimated values, although it is considerably less than that of Ching and Huber[9] obtained from a numerical study of the inverse participation ratio for a spatially disordered system with the exponential transfer-matrix element, (3.16). Elyutin,[10] however, suggests that the Ching-Huber result is too large, and points out that their numerical results are actually compatible with a lower value of R_A^{-1}, in the region of ~0.17.

In Fig. 2 we plot the function $R_A(\sigma/a_H)$ obtained from (4.11). For $\sigma/a_H \gtrsim 1$, the influence of σ becomes rather more important. For fairly large values of σ/a_H (as appropriate to M-I transitions occurring in low-dielectric-constant host materials), of the order of 3—5, the predicted ρ_A value differs by a factor of 2—3 over that obtained by setting $\sigma=0$; for example, with $\sigma/a_H=5$ we find $\rho_A a_H^3=1.56\times10^{-3}$, whereas for $\sigma=0$ we have $\rho_A a_H^3=2.96\times10^{-3}$. As pointed out by Puri and

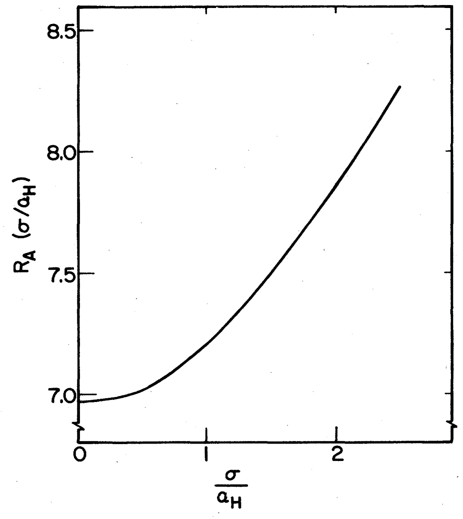

FIG. 2. $R_A = \rho_A^{-1/3} a_H^{-1}$ as a function of σ/a_H for the pure exponential transfer-matrix element $V(R) = -V_0 \exp(-R/a_H)$; a_H is the effective Bohr radius and σ is the hard-sphere diameter.

Odagaki,[5(b)] one should note that because of the hard-core condition the density is limited by its maximum value, given by $\rho^*_{\max} = 2^{1/2}$ ($\rho^*_{\max} = 3^{3/2}/4$) in the case of face-centered-cubic (body-centered-cubic) close packing. From (4.12) this condition imposes a minimum physically permitted value of $R_A(\sigma/a_H)$ for a given σ and a_H; the values of R_A displayed in Fig. 2, however, all lie well above the minimum permitted value.

We have also determined ρ_A for the frequently studied, modified exponential transfer-matrix element of the form

$$V(R) = -V_0 \left[1 + \left[\frac{R}{a_H} \right] \right] \exp(-R/a_H) \; . \tag{4.13}$$

To compare our results with previously estimated values, we consider explicitly the case where $\sigma = 0$; this should lead to a reasonable estimate of ρ_A, provided that $\sigma/a_H \ll 1$, a condition which will, for example, be satisfied in most doped semiconductors exhibiting a *M-I* transition. For this form of transfer-matrix element, the first-order Fredholm equation is given from (4.10) by

$$1 = 8\rho_A a_H^3 \int_0^{+\infty} dy \left\{ y^2 \sinh^{-1} \left[\frac{J\exp(y)}{2V_0(1+y)} \right] \bigg/ \left[1 + \left[\frac{J\exp(y)}{2V_0(1+y)} \right]^2 \right]^{1/2} \right\} , \tag{4.14}$$

where

$$J^2(\rho_A) = 7V_0^2 \rho_A a_H^3 \; . \tag{4.15}$$

Since J/V_0 is independent of V_0, the solution to (4.14) again depends solely on the dimensionless parameter $\rho_A a_H^3$. Numerical solution of (4.14) yields the result

$$\rho_A^{1/3} a_H = 0.12 \; , \tag{4.16}$$

which is lower than the value $\rho_A^{1/3} a_H = 0.14$ obtained for the simple exponential transfer-matrix element, reflecting the fact that the latter is essentially of shorter range than the modified exponential form (4.14). Equation (4.16) is the specific result for $\sigma = 0$, and, in general, the solution may be cast in the form (4.12). For the modified exponential transfer-matrix element, (4.13), Puri and Odagaki[5(b)] have calculated the function $R_A(\sigma/a_H)$ by numerical solution of a double-site version of the homomorphic coherent-potential approximation,[38] coupled with the $L(E)$ criterion[39] for the mobility edges. The qualititative dependence of R_A on σ/a_H found by Puri and Odagaki for the modified exponential form (4.13) is very similar to that shown in Fig. 2 [which applies to the simple exponential form (3.16)], although as seen from Table II the value of $R_A(0)$ found by these authors differs by a factor of ~2 from the present result, (4.16). In Table II we compare our calculated value, (4.16), with other previously estimated values of $\rho_A^{1/3} a_H$ obtained by setting $\sigma = 0$. From a detailed analysis of experimental data for a wide variety of systems exhibiting a *M-I* transition, Edwards and Sienko[34] have demonstrated the empirical validity of the relation $\rho^{1/3} a_H = 0.26 \pm 0.05$. For both forms of exponential transfer-matrix elements, our results, together with the majority of other estimates collected in Table II, are lower than the empirical value, implying that lateral disorder alone does not account for the observed *M-I* transition in impurity bands of uncompensated semiconductors. Fertis *et al.*[8] have observed that while the observed *M-I* transition in impurity bands of uncompensated semiconductors does not therefore appear to be a pure Anderson transition, it does not appear to be a pure Mott transition either, and may be of mixed Mott-Anderson type. Our discussion of exponential transfer-matrix elements has, however, been confined to the situation where site-diagonal disorder is absent. In the presence of diagonal disorder, ρ_A depends on V_0, solely through the dimensionless ratio λ/V_0 (analogously to the case of multipolar excitons), and the predicted Anderson transition density is an increasing function of λ/V_0 in a manner qualitatively similar to that shown in Fig. 1 for the case of quadrupolar excitons. Insofar as the *M-I*—transition density is an indication of the existence of a mechanism, the presence of site-diagonal disorder therefore enhances the relative role of the Anderson transition. For example, compensation of doped semiconductors will tend to increase the degree of site-diagonal disorder,[40] and, as pointed out by Fertis *et al.*,[8] it is possible that the observed *M-I* transition is some compensated semiconductors will be almost entirely of Anderson type.

Finally, we have also calculated the Anderson transition density in dimensionality 2 for both the pure exponential transfer-matrix element, (3.16), and the modified exponential form, (4.13). For lateral disorder alone, and with

$\sigma=0$, we find

$$\rho_A^{1/2}a_H=0.19 \text{ (pure exponential)}, \tag{4.17a}$$

$$\rho_A^{1/2}a_H=0.15 \text{ (modified exponential)}. \tag{4.17b}$$

The latter result is generally lower than previously estimated values obtained by Elyutin[10] ($\rho_A^{1/2}a_H=0.25\pm0.2$), Puri and Odagaki[5] ($\rho_A^{1/2}a_H=0.2$—$0.3$), and Debney[41] ($\rho_A^{1/2}a_H\cong0.36$).

V. MOBILITY-EDGE TRAJECTORIES FOR MULTIPOLAR EXCITONS

In this section we study the mobility-edge trajectories for transfer-matrix elements of the form (3.18), applicable to multipolar excitons. We consider the case where lateral disorder alone is present, and study the first-order Fredholm equation (4.1), with $F_0(k)$ given by (3.3) and $\alpha(\widetilde{E})$ and $x_0(\widetilde{E})$ given by (3.9a). The first-order Fredholm equation becomes

$$1=16\rho\alpha(\widetilde{E})\int_1^{+\infty} dz\frac{Q(z;\widetilde{E})}{(z^2-1)^{1/2}}\ , \tag{5.1a}$$

where

$$Q(z;\widetilde{E})=\tfrac{1}{2}\int_0^{+\infty} dR\, R^2 g_0\,|\,V(R)\,|\left[\frac{1}{\alpha^2(\widetilde{E})+[\frac{1}{2}\widetilde{E}-2\,|\,V(R)\,|\,z]^2}+\frac{1}{\alpha^2(\widetilde{E})+[\frac{1}{2}\widetilde{E}+2\,|\,V(R)\,|\,z]^2}\right]. \tag{5.1b}$$

For $\widetilde{E}=0$, Eqs. (5.1) reduce to (4.3) in the absence of site-diagonal disorder. For given ρ, solution of (5.1) gives the mobility edge $\widetilde{E}_c$, or, equivalently, for given $\widetilde{E}$, solution of (5.1) gives the critical density above which states at the chosen energy are delocalized.

Equations (5.1) are general for any $V(R)$ and $g_0(R)$; we now specifically consider $V(R)=\gamma R^{-n}$. From (5.1b) it is clear that we require $n>3$ to avoid a large-R divergence in the spatial integration. One therefore predicts that dipolar excitons (corresponding to $n=3$) are always delocalized; this conclusion is not an artifact of our use of first-order Fredholm theory, for, from (2.19) with $\beta=\frac{1}{2}$, we generally require $n>3$ to avoid a large-R logarithmic divergence in the spatial integration. The peculiarities of the dipolar problem have been known since Anderson's original paper.[1] Resolution of this issue may be complicated by shape-dependent effects associated with the long-ranged nature of the dipolar transfer-matrix element (which are familiar in dielectric theory[42]), and also by those effects arising from motion of the atoms in the liquid, which may be important in localizing states that would otherwise be extended. We note, however, that the simple introduction of an angular dependence into the dipolar transfer-matrix element does not appear to mitigate the conclusions reached above. For example, a subject of considerable current interest in infrared spectroscopy is that of resonant dipole-dipole transfer of vibrational excitations in a molecular liquid consisting of heteronuclear diatomics.[43] For this problem the appropriate transfer-matrix element takes the form

$$-V(\mathbf{R}_i-\mathbf{R}_j;\mathbf{\Omega}_i,\mathbf{\Omega}_j)=f^{(i)}f^{(j)}\mathbf{\Omega}_i\cdot\boldsymbol{T}^{(2)}(\mathbf{R}_i-\mathbf{R}_j)\cdot\mathbf{\Omega}_j\ ,$$

where $\mathbf{\Omega}_i$ denotes the orientation of molecule i, and $T^{(2)}(\mathbf{R})=-\nabla\nabla R^{-1}$ is the familiar dipole-dipole—interaction tensor.[42] The oscillator strength $f^{(i)}$ is proportional to $\langle 0\,|\,Q_i\,|\,1\rangle$, where Q_i is the vibrational coordinate of the molecule, and $|\,0\rangle$ ($|\,1\rangle$) is the ground vibrational state (first-excited vibrational state) of the molecule. With a minor generalization of the theory given in Sec. II, we arrive at an equation analogous to (2.19) in which $\int d\mathbf{R}$ is replaced by $\int d\mathbf{R}\int d\mathbf{\Omega}_1\int d\mathbf{\Omega}_2$, and $|\,V(R)\,|^{2\beta}$ by $|\,V(\mathbf{R};\mathbf{\Omega}_1,\mathbf{\Omega}_2)\,|^{2\beta}$. The fact that it is the *modulus* of the transfer-matrix element which occurs in the equation analogous to (2.19) means that, with $\beta=\beta_c=\frac{1}{2}$, we again predict that vibrational dipolar excitons are always delocalized.

For $n>3$, and with $g_0(R)=\Theta(R-\sigma)$, Eq. (5.1b) for $Q(z;\widetilde{E})$ may be cast in the form

$$Q(z;\widetilde{E})=\frac{|\,\gamma\,|^{3/n}}{n}\int_0^{|\gamma^*|} dV\,V^{-3/n}\left[\frac{1}{J^2(\rho)-2\widetilde{E}zV+4z^2V^2}+\frac{1}{J^2(\rho)+2\widetilde{E}zV+4z^2V^2}\right]=Q(z;-\widetilde{E})\ . \tag{5.2}$$

For $n\lesssim9$, numerical analysis shows that we may set the upper limit of (5.2) to $+\infty$ with negligible error. With this approximation, and employing the relation[31]

$$\int_0^{+\infty}\frac{x^{\mu-1}}{1+2x\cos t+x^2}dx=\frac{-\pi\sin[(\mu-1)t]}{\sin(t)\sin(\pi\mu)},\quad 0<\mu<2,\quad -1<\cos t<+1 \tag{5.3}$$

we find

$$Q(z;\widetilde{E})=\frac{|\,\gamma\,|^{3/n}}{2n}[J(\rho)]^{-(n+3)/n}(2z)^{(3-n)/n}[\mathscr{L}_n^{(+)}(\widetilde{E})+\mathscr{L}_n^{(-)}(\widetilde{E})]\ , \tag{5.4}$$

where

$$\mathscr{L}_n^{(\pm)}(E)=\frac{B(3/n;(n-3)/n)\sin[(3/n)\cos^{-1}(\pm\tilde{E}/2J)]}{[1-(\tilde{E}/2J)^2]^{1/2}}\ . \tag{5.5}$$

Using

$$\int_1^{+\infty} dz\frac{z^{(3-n)/n}}{(z^2-1)^{1/2}}=\tfrac{1}{2}B((n-3)/2n;\tfrac{1}{2}),\quad n>3 \tag{5.6}$$

the first-order Fredholm equation (5.1) becomes

$$1=\frac{2\rho}{n}\alpha(\tilde{E})(2\,|\,\gamma\,|\,)^{3/n}[J(\rho)]^{-(n+3)/n}B((n-3)/2n;\tfrac{1}{2})[\mathscr{L}_n^{(+)}(\tilde{E})+\mathscr{L}_n^{(-)}(\tilde{E})]\ . \tag{5.7}$$

Substituting for $\alpha(\tilde{E})$ from (3.9a) gives the final result,

$$1=\frac{2\pi\rho^*}{n}\left[\frac{J(\rho^*)}{2\,|\,\gamma^*\,|}\right]^{-3/n}B((n-3)/2n;\tfrac{1}{2})\csc\left[\frac{n-3}{n}\pi\right]\times\left\{\sin\left[\frac{3}{n}\cos^{-1}\left[\frac{\tilde{E}}{2J(\rho^*)}\right]\right]+\sin\left[\frac{3}{n}\cos^{-1}\left[\frac{-\tilde{E}}{2J(\rho^*)}\right]\right]\right\},\quad |\,\tilde{E}\,|\le 2J(\rho^*) \tag{5.8}$$

where we use $B(\lambda;1-\lambda)=\pi\csc(\lambda\pi)$. The requirement that $|\,\tilde{E}\,|\le 2J(\rho^*)$ is simply the condition that the half-width $\alpha(\tilde{E})$ in the Cauchy form of $F_0(k)$, Eq. (3.3), be positive [cf. (3.9a)]. Physically, this is simply the constraint that the state of energy $\tilde{E}$ lies within the band of the Hubbard density of states, (3.13), obtained by employing the Cauchy form of $F_0(k)$, or equivalently, that $\tilde{E}$ lies within the band obtained from the mean spherical integral equation theory at the "two-body" level [i.e., under neglect of irreducible m-body graphs ($m\ge 3$), as discussed in Sec. III]. The condition for localization at the band edges, $\tilde{E}_\pm=\pm 2J(\rho^*)$, is given from (5.8) by

$$1=\frac{2\pi\rho^*}{n}\left[\frac{J(\rho^*)}{2\,|\,\gamma^*\,|}\right]^{-3/n}B((n-3)/2n;\tfrac{1}{2}),\quad \tilde{E}=\tilde{E}_\pm\ . \tag{5.9}$$

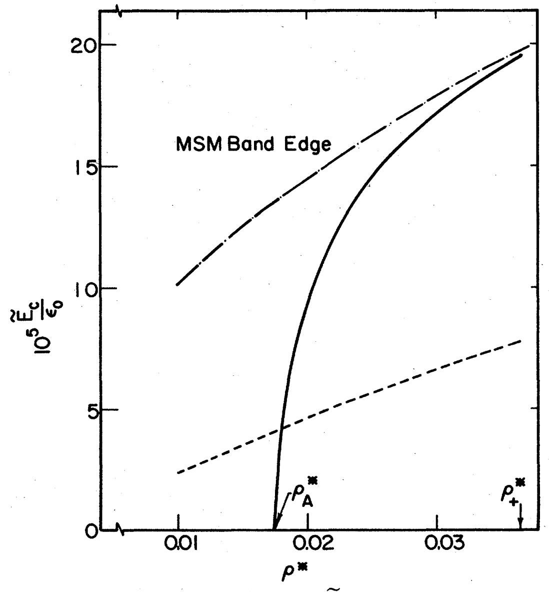

FIG. 3. Mobility-edge trajectory $\tilde{E}_c/\epsilon_0$ (solid line) as a function of reduced density ρ^* for $V(R)=\gamma R^{-5}$, with $|\,\gamma^*\,|=\sqrt{14}\times 10^{-4}\epsilon_0$; $\rho_A^*=1.75\times 10^{-2}$ and $\rho_+^*=3.66\times 10^{-2}$. We also plot the band edge obtained from solution of the mean spherical integral equation for quadrupolar excitons, and the mobility edge obtained from the upper-limit approximation to (2.13) (dashed line).

By virtue of the fact that $J(\rho^*)/2\,|\,\gamma^*\,|$ is independent of γ^* [see (3.19)], and, similar to (4.8) for the Anderson transition, we see that the solution of (5.8) is independent of γ^* and depends solely on ρ^*. For $\rho^*\ge\rho_+^*$, where ρ_+^* is given by the solution of (5.9), all states in the band resultant from the mean spherical theory are predicted to be extended.

In Fig. 3, for $n=5$ corresponding to pure quadrupolar electronic excitons, we plot the dimensionless mobility-edge trajectory, $\tilde{E}_c/\epsilon_0$, as a function of ρ^* and as determined by solution of the transcendental Eq. (5.8). γ^* is given by (3.20), we have chosen $C_0^*=10^{-4}$, and an extended virial expansion[29] for the integral $I_{10}^{\mathrm{HS}}(\rho^*)$, which determines $J(\rho^*)$, has been employed. The predicted Anderson transition density, $\rho_A^*=1.75\times 10^{-2}$, differs negligibly from that determined using the low-density-limit form of $I_{10}^{\mathrm{HS}}(\rho^*)$ (cf. Table I), reflecting the fact that the Anderson transition occurs at a low reduced density. We also plot the spectral band edge predicted from the "full" mean spherical theory for pure quadrupolar excitons,[22] in which one explicitly considers irreducible three-body graphs contributing to the density of states; the irreducible three-body graphs determine the shift and asymmetry in the density of states. The interesting range of reduced density from our present viewpoint is, however, $\rho^*\lesssim\rho_+^*$, and in this range the density of states resultant from the full mean spherical theory differs very little from that predicted at the two-body level [Eq. (3.13)], reflecting the fact that in this density regime the density of states is essentially symmetric with a maximum centered on $\tilde{E}=0$.

Below the Anderson transition density ρ_A^*, all states in the band are localized. From Fig. 3 we see that as the density is increased above ρ_A^*, the mobility edge initially moves very rapidly away from the band center; in fact, it is not difficult to show that, close to the Anderson transition density $[0<(\rho^*-\rho_A^*)\ll\rho_A^*]$, $\epsilon_0\tilde{E}_c\sim|\,\gamma^*\,|(\rho^*-\rho_A^*)^{1/2}$ for all $n>3$, giving rise to a characteristic square-root density dependence. For $\rho^* \geq \rho_+^* = 3.66\times 10^{-2}$, all states in the band resultant from the two-body mean spherical integral equation theory are extended. For a gas consisting of atoms with hard-sphere diameter $\sigma = 3.4$ Å at a temperature of 273 K, this means that over the relatively narrow pressure range of ~ 20 atm (cf. Sec. IV), we go from a situation where all states in the band are localized to one where essentially the reverse is true; in fact, a majority of states in the band become delocalized over a much narrower pressure range (~ 2 atm).

In Fig. 3 we also plot the mobility-edge trajectory obtained from the upper-limit approximation to (2.13), whereby one neglects the real part of the self-energy [this curve is obtained from the solution to (5a) of Ref. 13]. We see that for $\tilde{E}_c \lesssim 4.2\times 10^{-5}$ (or $\tilde{E}_c/2J \lesssim 0.3$), the upper-limit approximation makes it more difficult to localize states, and leads to an excess of extended states; in contrast, for $\tilde{E}_c/2J \gtrsim 0.3$, the upper-limit approximation leads to an excess of localized states. The argument given by Abou-Chacra *et al.*[14] to suggest that the upper-limit approximation makes it harder to localize the states is based on consideration of (2.5c) and is, in short, that inclusion of E_j increases the denominator of (2.5c) and therefore reduces the eigenvalue λ. Let us consider the case where site-diagonal disorder is absent, and, for convenience, assume that $\tilde{E} = \tilde{R} - \epsilon_0 \geq 0$. Inspection of (2.5c) shows that if (i) $E_j < 0$ or $E_j > 2\tilde{E}$, inclusion of E_j in (2.5c) indeed increases the denominator of that equation, thus reducing the eigenvalue λ; in contrast, if (ii) $0 \leq E_j \leq 2\tilde{E}$, inclusion of E_j decreases the denominator of (2.5c) and thus increases the eigenvalue λ.

Which of the cases (i) and (ii) above is most probable will be determined by the distribution of E_j, which, in our case, is essentially characterized by the Lorentzian probability distribution $f_0(E_j)$ of (3.9b). If, for $\tilde{E} = \tilde{E}_c$, a large fraction of the distribution $f_0(E_j)$ is concentrated in the ranges $E_j < 0$ and $E_j > 2\tilde{E}_c$, the upper-limit analysis is likely to overestimate the true mobility edge $\tilde{E}_c$, whereas if a large fraction of the distribution is concentrated in the range $0 < E_j < 2\tilde{E}_c$, the converse is expected to be true. From (3.9b) we see that $f_0(E_j)$ is a Lorentzian centered on $E_j = 1/2\tilde{E}$ with a half-width of $1/2[4J^2(\rho) - \tilde{E}^2]^{1/2}$, which decreases with increasing $\tilde{E}$. For $\tilde{E} \ll 2J(\rho)$, the vast majority of states in the distribution $f_0(E_j)$ fall into category (i) above; thus if the true mobility edge is close to the band center $[0 \leq \tilde{E}_c \ll 2J(\rho)]$, it is likely that use of the upper-limit approximation will result in an overestimation of the mobility edge and lead to an excess of extended states. In contrast, as $\tilde{E}_c$ moves away from the band center, a progressively smaller fraction of the probability distribution $f_0(E_j)$ falls with the regions $E_j < 0$ and $E_j > 2\tilde{E}_c$, and case (ii) above rapidly predominates; thus, as the true mobility edge moves away from the band center, use of the upper-limit approximation will eventually lead to an underestimation of the mobility edge and to an excess of localized states. This behavior is clearly manifested in Fig. 3.

Finally, we have not investigated in detail the influence of site-diagonal disorder upon the full mobility-edge trajectory. From the discussion given in Sec. IV on the influence of site disorder on the Anderson transition, however, it is clear that the presence of site-diagonal disorder will increase ρ_A^* and therefore also the density range of interest (in which a significant fraction of states in the band are localized). For the $n = 5$ example studied numerically in this section, it is likely that the influence of site-diagonal disorder upon E_c/ϵ_0 will be very small if we restrict ourselves to consideration of a quadrupole-allowed electronic transition in a pure gas. If, however, the atomic species of interest is diluted in a host liquid, the influence of site disorder on the mobility-edge trajectory may be quite pronounced.

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