PHYSICAL REVIEW B VOLUME 4, NUMBER 12 15 DECEMBER 1971

Percolation on a Continuum and the Localization-Delocalization Transition in Amorphous Semiconductors

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(Received 3 May 1971)

The concept of percolation has been set in a form which is more directly germane than the existing theory of percolation on lattices to the question of the localization-delocalization transition (mobility edge) in amorphous semiconductors. The problem of percolation on a continuum has been formulated in the context of motion in a random potential $V(\vec{r})$. An energy-dependent dimensionless density $\phi(E)$ is introduced which specifies the fraction of space satisfying $V<E$. Extended states appear above a critical density ϕ_c; this provides our working criterion $\phi(E_c)=\phi_c$ for the location of the percolation threshold E_c. In one dimension $\phi_c=1$, and in two dimensions we find that $\phi_c=\frac{1}{2}$ for an important class of random potentials. In three dimensions we obtain an estimate of $\phi_c=0.15$ from an empirical rule. The percolation criterion for the location of the mobility edge E_c has been applied to several types of disordered potentials. The oft-invoked Gaussian potential distribution has been treated and the results compared with those of several recent calculations. Random-walk techniques can be used to attack more general random potentials; we have used this approach to explicitly calculate E_c for the potential of an array of random dipoles, which is an initial model for an amorphous molecular solid. A way of including short-range order is briefly discussed.

I. INTRODUCTION

The mathematical analysis of percolation on a lattice is applicable to a variety of physical phenomena.[1,2] In a lattice in which each site is either occupied (with probability p) or unoccupied (with probability $1-p$), and in which adjacent (i.e., nearest-neighbor) occupied sites are regarded as linked, there exists a critical percolation probability (p_c) above which unbounded linked clusters appear. Below p_c, no filled sites belong to infinitely extended clusters; above p_c, a finite proportion of the filled sites do belong to such infinite clusters. A related but distinguishable problem is defined by considering the network of bonds connecting nearest-neighbor lattice sites. If each bond is either open (with probability p_b) or closed (with probability $1-p_b$) above a critical fraction ($p_{b,c}$) of open bonds, there appear infinitely extended unblocked paths. Nearly all extant percolation theory deals with *ordered* structures (periodic lattices); a typical application is the determination of the critical concentration of magnetic atoms in dilute ferromagnetic crystals.[3] Even for simple lattices the theory is difficult; except for two two-dimensional cases for which exact values are deducible, only approximate estimates (based on Monte Carlo and series-expansion techniques) have been obtained for p_c and $p_{b,c}$.

The presence in percolation processes of this critical behavior (at p_c) appearing with varying concentration (p) makes this theory a natural one to associate with the Mott transition[4] (sharp metal → insulator transition accompanying an abrupt correlation-induced transformation from delocalized to localized electronic wave functions, postulated to take place with decreasing density), as well as with the Anderson transition[5] (a sharp disorder-induced localization of electronic states). This paper is concerned with an Anderson-transition situation: the mobility edge[6,7] presumed to occur in the band tails of amorphous semiconductors. In several recent papers,[7-9] results of *lattice* percolation theory have been adopted in theoretical models for the mobility edge. In the work presented here our aim is twofold: to construct a useful formulation of the proper percolation problem appropriate to this class of phenomena, which is that of *percolation on a continuum;* and to connect the percolation results to the question of the localization-delocalization transition in amorphous semiconductors.

The question of the connection between percolation theory and the localization-delocalization transition was first addressed in an article by Ziman.[10] Ziman's approach, discussed in Sec. II B, involves an empirical rule based on critical probabilities for the bond problem. The approach developed in the present work makes use of an empirical rule for the site problem proposed in a recent note by Scher and Zallen (SZ).[11] This line of development has the advantage of invoking the concept of a critical percolation density by means of which it is straightforward to make contact with the parameters characterizing a random potential (such as the rms spatial fluctuation).

The analysis of percolation on a continuum is dealt with in Sec. II of the paper, and applications to amorphous materials are given in Sec. III. A brief outline will be helpful here. Continuum percolation is introduced in Sec. II A in the context of the percolation of a classical particle in a random potential $V(\vec{r})$. An energy-dependent dimensionless density $\phi(E)$ is defined which specifies the fraction of space accessible $(V<E)$ to particles of energy E. Empirical rules for a critical density ϕ_c, corresponding to a percolation threshold, are discussed in Sec. II B and compared to the different empirical rule observed and employed by Ziman. Estimates for ϕ_c in two and three dimensions are discussed in Secs. II C and II D. It is ϕ_c which provides our percolation criterion for the localization-delocalization transition in a random potential. In Sec. III A we note that the essential character of $V(\vec{r})$, needed to determine $\phi(E)$, is contained in the distribution function $\psi(V)$. This prescription is applied, in Sec. III B, to the location of the critical energy $E(\phi_c)$ for that class of situations in which $\psi(V)$ can be approximated by a Gaussian, and our results are then connected with two relevant recent treatments invoking Gaussian statistics. A method for handling more general distributions is outlined in Sec. III C, and is used in Sec. III D to calculate $E(\phi_c)$ for an assembly of randomly distributed dipoles. In Sec. III E we indicate a way in which short-range order may be included. Our principal results and conclusions are summarized in Sec. IV.

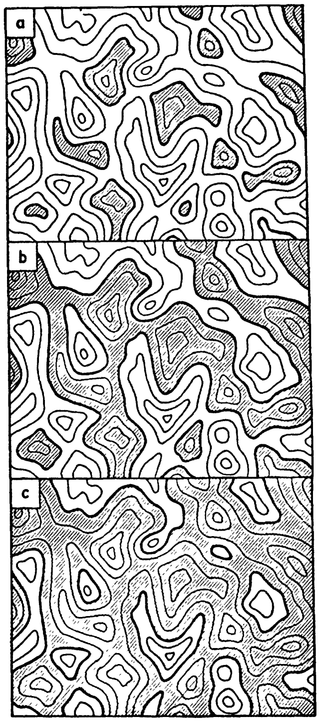

FIG. 1. Percolation in a two-dimensional potential. The contour lines represent equipotentials of $V(r)$. The shaded regions in (a), (b), and (c) indicate allowed $(V<E)$ regions for three successively increasing values of E.

II. CRITICAL PERCOLATION DENSITY

A. Classical Percolation in Random Potential

To construct in concrete terms the problem of percolation in a continuum, we consider the motion of a classical particle in a random potential $V(\vec{r})$. While our ultimate interest is, of course, in real (i.e., quantum-mechanical) systems, this classical model will elucidate the essential topological aspects of the problem. (Another situation embodying percolation in a continuum is that of conducting particles randomly dispersed in an insulating medium.[11]) The subtle problem of the spatial variation of the effective potential appropriate for the quantum-mechanical or semiclassical description of a given amorphous solid will not come in for detailed scrutiny in this paper; in Sec. III we will find that a statistical characterization of $V(\vec{r})$ suffices for our purposes. It is worth noting, however, that the effective potential will be more slowly varying than the one-electron potential because of the pseudopotential-theory cancellation (within the regions of the atomic cores) of the potential energy by the kinetic energy associated with orthogonalization-induced wave-function oscillations.[12] Procedures for constructing properly "smoothed" potentials have been described by Eggarter and Cohen (EC)[8] in connection with the mobility edge, and by Halperin and Lax (HL)[13] in connection with the band-tail density of states. The fact that the random potentials of interest are, therefore, rather well-behaved functions will be implicit in the mathematical discussions to follow in this section.

Given $V(\vec{r})$, we introduce a function $\phi(E)$ which defines the fraction of space accessible to particles of energy E. This is simply that fraction of space for which the total energy exceeds the potential energy V:

$$\phi(E) \equiv \int_{V(\vec{r})<E} d\vec{r} \Big/ \int d\vec{r} \; . \qquad (1)$$

The connection with percolation theory is revealed by the fact that for energies such that $\phi(E)$ exceeds a critical value ϕ_c, there are infinitely extended volumes of allowed $(V<E)$ space (i.e., "delocalized states" occur). While (1) is expressed in terms of the explicit spatial dependence of V, in Sec. III we will show that ϕ is simply obtainable as a statistical characterization of the potential.

The critical percolation behavior in a two-dimensional continuum is depicted in Fig. 1, in which the contour lines represent equipotentials of $V(\vec{r})$. The shaded areas in Figs. 1(a)–1(c) indicate regions satisfying $V(\vec{r})<E$ for three successively increasing values of E. Referring to

allowed ($V<E$) regions as water areas and forbidden ($V>E$) regions as land areas (the analogy is to a terrain with V represented by the altitude of the surface and E represented by a water level which is the same for all bodies of water), at low E the allowed areas comprise isolated lakes in an infinitely extended continent [as in Fig. 1(a)], while at high E there is the opposite situation in which isolated islands are embedded in an infinitely extended ocean of allowed territory [Fig. 1(c)]. Somewhere in between [Fig. 1(b)], at the critical coverage ϕ_c, there is a situation corresponding to the critical-percolation probability as the extended ocean makes its debut on the scene. (In two dimensions as we will see in Sec. II C, the appearance of the ocean coincides with the disappearance of the continent.) We can characterize the critical coverage ϕ_c in two ways:

$$\text{average lake size} \to \infty \text{ as } \phi \to \phi_c^- , \tag{2}$$

$$\text{fraction of space belonging to an extended ocean} \to 0 \text{ as } \phi \to \phi_c^+ . \tag{3}$$

In (2) the left-hand side approaches 0 as $\phi \to 0$, while in (3) the left-hand side approaches 1 as $\phi \to 1$. We will use E_c to denote the energy corresponding to the percolation threshold ϕ_c:

$$\phi(E_c) = \phi_c . \tag{4}$$

$\phi(E)$ and therefore E_c are well defined for any nonpathological $V(\vec{r})$. For example, for an attractive potential ($V<0$) produced by a single center or by a finite set of centers ($V \to 0$ as $r \to \infty$), $\phi(E)=0$ for $E<0$, $\phi(E)=1$ for $E>0$, and therefore $E_c=0$ (i.e., the critical percolation energy corresponds, quite naturally, to the bottom of the quantum-mechanical continuum). For a periodic potential, E_c is fixed by the lowest-energy set of mountain passes (saddle points) separating neighboring unit cells. The working hypothesis of this paper is that, for a broad class of physically realistic random potentials (i.e., which exhibit continuity and differentiability except at isolated point singularities, as well as *homogeneity on a macroscopic scale*), the value of ϕ_c is approximately constant—that is to say, insensitive to the detailed nature of $V(\vec{r})$. Empirical evidence for this, along with estimates of the critical densities in one, two, and three dimensions, is presented in Secs. II B–II D. With this basis, (4) becomes a useful criterion for the determination of the percolation threshold E_c, which in turn provides an important, and perhaps decisive, clue to the location of the mobility edge in real materials.

B. Empirical Rules Based on Lattice Percolation

Two inductive generalizations of results calculated for percolation on regular lattices have been proposed. (These papers are pertinent, albeit implicitly, to the problem of percolation in a continuum.) An empirical relation for the bond problem on a three-dimensional lattice, first observed by Vyssotsky *et al.*,[14] was recently rediscovered and applied to disorder-induced localization by Ziman[10]: $p_{b,c} n \cong 1.5$, where n is the lattice coordination number. Ziman indicates how to formally construct a quantity $N(E)$ specifying the "average number of unblocked paths" (saddle points with $V<E$) leading from any atom; his criterion for E_c is then $N(E_c)=1.5$, a result analogous to, but more cumbersome to apply than, that given in (4). More serious is the objection that his formulation in terms of nearest-neighbor coordination relies on the literal interpretation of $V(\vec{r})$ as the rapidly varying one-electron potential.

An empirical rule directly relevant to ϕ_c, based on the lattice site problem, was the subject of a recent note by SZ.[11] (Unlike Ziman's treatment, in which interatomic saddle points play the role of percolation bonds, SZ's formulation does not rely on coordination number and is more generally applicable.) SZ introduce a geometric interpretation of site-occupation probability p in terms of a *space-occupation probability density* ρ, corresponding to the packing of spheres (in three dimensions) or circles (in two dimensions). They find that in three dimensions the various critical site probabilities for different lattices ($p_c = 0.19$–0.43)[2] all correspond to the *same* (within a few percent) critical probability density (space-occupation fraction) of $\rho_c = 0.15$.[11] In two dimensions their result is 0.44, and in one dimension it is, of course, 1.00. These numbers offer first estimates for ϕ_c.

In one dimension the only possibility for percolation is for the allowed territory to occupy the *entire* space, $V<E$ everywhere. Thus $E_c = V_{\max}$ and $\phi_c(1_D) = 1 = \rho_c(1_D)$. At E_c the localization-delocalization transition is discontinuously sharp, since for $E<E_c$ none of the space belongs to an extended ocean, while for $E>E_c$ *all* of it does. This circumstance is unique to the one-dimensional case, a point underscored by Ziman in disputing the applicability of one-dimensional models[4] to real (three-dimensional) systems. Two other crucial distinctions between percolation in one- and three-dimensional continua are pointed out in Sec. II C. Our knowledge of ϕ_c in one dimension is exact, but of little interest. The critical densities in two and three dimensions are attacked below.

C. Critical Density in Two Dimensions

There is an important class of random potentials (examples are the Gaussian and Holtsmark distributions to be discussed in Secs. III B and II D) which exhibit symmetry between high and low energies in the following sense:

$$\phi(E_{1/2}+\Delta E)+\phi(E_{1/2}-\Delta E)=1 \ . \tag{5}$$

For such symmetric potentials the median energy $E_{1/2}$, defined by $\phi(E_{1/2})=\frac{1}{2}$, coincides with the average potential $\langle V(\vec{r})\rangle$. We will now show that, for this class of $V(\vec{r})$'s in two dimensions, $\phi_c=\frac{1}{2}$.

Unlike the one-dimensional case, in two dimensions an ocean may exist without filling the entire space (Fig. 1). However, it does remain true that only one such infinitely extended ocean or continent may exist at a time, since two extended networks cannot avoid intersection in 2_D.[15] (The extended oceans are weblike networks, not threadlike rivers, because the random potential must exhibit large-scale homogeneity.) The symmetry expressed in (5) means that the critical fraction of water coverage for the occurrence of an infinitely extended ocean must equal the critical fraction of land coverage for the occurrence of an infinitely extended continent. Since an ocean and a continent cannot coexist (a statement valid in one and two dimensions, invalid in three or more dimensions), ϕ_c cannot be less than 0.5 as otherwise this (impossible) coexistence would occur for $\phi_c<\phi<1-\phi_c$.

Figure 1 further suggests that a randomly produced 2_D map [i.e., a map generated by an equipotential of a random $V(\vec{r})$] cannot contain *only* bounded lakes and islands. Such a situation would require an infinite set of *isoenergetic* saddle points. Examples are a checkerboard with black and white squares representing island and lakes, and a Kagomé lattice with hexagonal areas as islands, triangular areas as lakes. These two cases illustrate the essential requirement, a nonvanishing number of saddle points per island (two for the checkerboard, three for the Kagomé). While such a situation automatically occurs for a periodic crystal potential, for which entire sets of saddle points (i.e., a finite number per unit cell) occur at *precisely* the same energy, it is inadmissible for a random potential since the disorder disperses the saddle-point energies into a continuous distribution.[16] Armed with the information that a random map without an extended ocean or continent is disallowed (this statement holds in two or more dimensions, does not hold in one dimension), we deduce that ϕ_c cannot exceed 0.5 by an argument analogous to that of the preceding paragraph.

The above arguments reveal that for a symmetric random potential in two dimensions, the critical percolation density is $\frac{1}{2}$:

$$\phi_c(2_D;\ V_{\rm sym})=0.50 \ . \tag{6}$$

We can compare this result with the empirically induced quantity $\rho_c(2_D)=0.44$ discussed in Sec. II B. This quantity was constructed from a situation with substantial asymmetry between filled regions (circles surrounding filled sites) and empty regions (circles surrounding empty sites, *plus* the interstitial spaces). The relatively small difference between these two numbers reflects the weak dependence of ϕ_c on gross characteristics of the random potential.

D. Critical Density in Three Dimensions

A simple derivation for $\phi_c(3_D;\ V_{\rm sym})$, analogous to the above analysis, cannot be given. The second half of the 2_D argument, based on the necessary occurrence of at least one ocean or continent, continues to apply to the 3_D case; but this merely yields an overly generous upper limit for the critical density:

$$\phi_c(3_D;\ V_{\rm sym})<0.5 \ . \tag{7}$$

In the absence of any better estimate for the critical density in a three-dimensional continuum, we will tentatively adopt the value of the empirically obtained quantity $\rho_c(3_D)$,[17]

$$\phi_c(3_D)\cong 0.15 \ . \tag{8}$$

[The two-dimensional results suggest that (8) may slightly underestimate $\phi_c(3_D;\ V_{\rm sym})$.[18] A balancing consideration may be that, for actual quantum-mechanical systems, the possibility of tunneling through narrow nonallowed $(V>E)$ regions means that the classical result overestimates ϕ_c.] In Sec. III we will make use of Eq. (4), with the critical density given by (8), as our working criterion for the location of the localization-delocalization energy E_c.

Before proceeding with applications of (4) and (8) we present, in Fig. 2, a diagrammatic summary of some of the key differences between continuum percolation processes in one, two, and three dimensions. In order to include one-dimensional E_c's in the comparison, the $\phi(E)$ shown corresponds to a random potential possessing upper and lower bounds. The quantity $R(E)$,[19] schematically plotted in Fig. 2, is the fraction of space belonging to infinitely extended oceans (i.e., accessible to delocalized electrons of energy E). The softening of the localization-delocalization transition with increasing dimensionality is evident.

III. CALCULATION OF DENSITY $\phi(E)$

A. Potential Distribution Function

Given the localization-delocalization criterion expressed in (4) and the value of ϕ_c stated in (8), we now wish to consider the calculation of $\phi(E)$ from a specific $V(\vec{r})$, since this is needed to determine E_c. To calculate $\phi(E)$ from the basic definition (1) would necessitate an analytic representation of the random potential $V(\vec{r})$, and then an integration over a function of $V(\vec{r})$ such as the Dirichlet unit step function. This would be a formidable task.

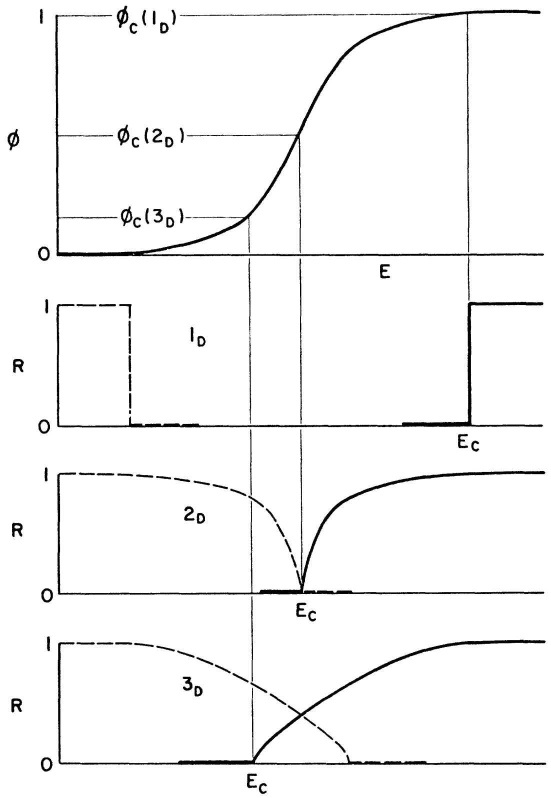

FIG. 2. Dimensionality dependence of continuum percolation processes. $\phi(E)$ is the fraction of allowed $(V<E)$ space and $R(E)$ is the fraction of space contained in infinitely extended allowed regions. [The dashed curves represent functions opposite to $R(E)$, specifying the fraction of space contained in infinitely extended forbidden regions.]

Furthermore, requiring an analytic expression for $V(\vec{r})$ would be overspecifying the problem for the following reason: It is only important to know the *fraction* of space satisfying $V(\vec{r})<E$; the *coordinates* of these valleys are unnecessary for our purpose. Thus a simple *statistical* characterization of $V(\vec{r})$ is all that is needed. It is this statistical interpretation (spelled out below), rather than the direct geometric definition of (1), which lends ϕ its primary usefulness in dealing with percolation processes.

Associated with every random $V(\vec{r})$ is the distribution $\psi(V)$ of values of V generated at an arbitrary position $\vec{r}$. The normalized potential distribution function can be interpreted in two ways: $\psi(V)\,dV$ specifies the probability of occurrence, at an arbitrary $\vec{r}$, of a potential value in the range $V \longrightarrow V + dV$; $\psi(V)\,dV$ specifies the relative frequency count of V values in this range obtained by sampling the potential in all of the infinitesimal volume elements of the space. With the latter interpretation, to determine $\phi(E)$ we simply count the relative frequency of regions with $V<E$,

$$\phi(E)=\int_{-\infty}^{E}\psi(V)\,dV\;. \tag{9}$$

Thus, once $\psi(V)$ becomes known for a specific random potential, the critical percolation energy is obtained from

$$\int_{-\infty}^{E_c}\psi(V)\,dV=\phi_c\;. \tag{10}$$

We now go on to a consideration of the determination of $\psi(V)$, dealing first with an important particular example and then with a more general approach applicable to an arbitrary $V(\vec{r})$.

B. Critical Energy for Gaussian Potential Distribution

A random potential $V(\vec{r})$ is generated by summing, at $\vec{r}$, the independent contributions from randomly placed centers. In other words, $V(\vec{r})$ is a sum of random variables. We know (from the central-limit theorem) that for a sufficiently high density of centers[13] Gaussian statistics apply, so that $\psi(V)$ obeys a Gaussian distribution:

$$\psi(V)=(2\pi V_{\mathrm{rms}}^2)^{-1/2}\,e^{-(V-\bar{V})^2/2V_{\mathrm{rms}}^2}\,, \tag{11}$$

where $\bar{V}$ is the mean potential and V_{rms}^2 is the variance. Inserting (11) into (10) yields

$$\mathrm{erf}\,[(\bar{V}-E_c)/2V_{\mathrm{rms}}^2]=1-2\phi_c\,, \tag{12}$$

where $\mathrm{erf}(z)$ is the error function. Since $\mathrm{erf}(z)$ is an odd function of its argument, for $2\phi_c<1$, E_c must be less than $\bar{V}$. Using $\phi_c(3_D)=0.15$ in (12) locates E_c for a Gaussian distribution:

$$E_c=\bar{V}-1.03\,V_{\mathrm{rms}}\;. \tag{13}$$

We now consider applications of (13) to two recent models invoking Gaussian statistics.

HL[13] have calculated the density of states $\rho(E)$ for an impurity band tail in the high-density limit. They consider in some detail the situation in which the random potential is produced by a superposition of screened Coulomb potentials of the form $(Z_ie^2/\epsilon_0 r)e^{-Qr}$, where Z_ie is the charge of the i-type impurity, ϵ_0 is the dielectric constant of the host semiconductor, and Q is the reciprocal screening length. They find that

$$V_{\mathrm{rms}}^2=(2\pi e^4/\epsilon_0^2 Q)\sum_i \bar{n}_i Z_i^2\,, \tag{14}$$

where $\bar{n}_i$ is the i-type impurity concentration. HL define a dimensionless parameter $\xi' \equiv (V_{\mathrm{rms}}/E_Q)^2$, where $E_Q=\hbar^2Q^2/2m^*$, and they present their tail-states results, for various values of ξ', with $\rho(E)$ plotted against energy from the band edge in dimensionless units, $\nu=(\bar{V}-E)/E_Q$. In terms of their dimensionless variables, (13) becomes

$$\nu_c=1.03(\xi')\;. \tag{15}$$

Only for large ξ' does (15) yield a critical energy high in the tail.

Recently Stern[20] has used the Halperin-Lax density of states to calculate the optical absorption edge of amorphous silicon. He obtained a best fit with experiment using $V_{\rm rms} = 0.89$ eV and $Q^{-1} = 6$ Å. With these values and $m^* = 0.3m_e$ substituted into the parameters appearing in (15), we obtain [using (15) and HL's curves] a states density at the conduction-band mobility edge of $\rho(\nu_c) = 5\times 10^{20}\,{\rm cm}^{-3}\,{\rm eV}^{-1}$ compared to Stern's result of $9\times 10^{20}\,{\rm cm}^{-3}\,{\rm eV}^{-1}$. Stern used the Anderson[5] localization criterion in his determination of the mobility edge.

A second application of (13) is in the reinterpretation of a criterion appearing in a simple model recently investigated by EC.[8] They consider a gas of hard-core scatterers. In a potential-smoothing procedure based on the uncertainty principle, they divide the system into cells of side $\hbar/p$ and treat the number N_i of scatterers per cell as a random variable (the cells are *not* infinitesimals as $\langle N\rangle \gg 1$). EC introduce a quantity $C(E)$ which is defined as the fraction of cells with cell potential $V_i < E$. As the wave function for the electron in each cell is a plane wave, $C(E)$ can be interpreted as the fraction of the total volume available to an electron of energy E, that is, their $C(E)$ corresponds to our $\phi(E)$. EC adopt, as their mobility-edge criterion, the critical percolation probability for percolation on a *simple cubic lattice*, $C(E_c) = p_c({\rm sc}) = 0.31$. Their result, assuming Gaussian statistics for N_i (and hence for V_i, since $V_i \propto N_i$), is

$$E_c = \bar{V} - 0.52\,\bar{V}/\langle N\rangle^{1/2} . \tag{16}$$

Since in this model $V_{\rm rms} = \bar{V}/\langle N\rangle^{1/2}$, this result is equivalent to (13) with the numerical factor 1.03 replaced by 0.52. The numerical difference is produced by *EC*'s particular choice of p_c(sc) as the percolation criterion playing the role which, as we have seen, properly belongs to $\phi_c(3_D)$. The use of 0.15 for $\phi(E_c)$ locates the mobility edge further down in the density-of-states tail (*EC*'s Fig. 1) than does (16).

C. Potential Distribution for Arbitrary Random Potentials

To obtain $\psi(V)$ for an arbitrary $V(\vec{r})$ generated by a random distribution of potential centers, we proceed as follows. The problem is to find the probability density for $V = \sum_i V_i$, where the V_i are random variables. This is equivalent to a one-dimensional random walk. The general k-dimensional random-walk problem has been solved using Markoff's method and is treated in the well-known review article of Chandrasekhar.[21] In the limit of a large system,

$$\psi(V) = (1/2\pi V_0)\int_{-\infty}^{\infty} dx\, e^{ixv - nC(x)} ,$$

where (17)

$$C(x) = \int dz\, p(z)(1 - e^{-ixv(z)}) .$$

Here v is a dimensionless potential V/V_0, with V_0 an appropriate scaling energy for the problem, z specifies the position and internal coordinates of a center, $p(z)$ is the probability distribution of the potential centers, $v(z)$ is the potential produced by a center characterized by z, and n is the density of centers. Distributions defined by (17) have been considered extensively in the theory of inhomogeneously broadened resonance lines in solids.[22] The essential aspect of (17) is that it relates the probability density of V, a sum of random variables, to the probability density $p(z)$ characterizing the individual random variable V_i.

D. Critical Energy for Random Dipole Distribution

A collection of randomly oriented dipoles can be considered as an initial model for an amorphous molecular solid. The potential due to each center is

$$V_i = P_i \cos\theta_i/\epsilon_0 r_i^2 , \tag{18}$$

where P_i is the dipole moment at $\vec{r}_i$ and $\cos\theta_i = \hat{P}_i \cdot \hat{r}_i$. If a typical molecular dimension is denoted by a, then $P_i \sim ea$ and we can choose $V_0 = e/\epsilon_0 a$ in (17). We assume a random isotropic distribution of the molecular dipoles,

$$p(z)\,dz = \beta(p')\,dp'\,r^2\,dr \sin\theta\,d\theta\,d\phi , \tag{19}$$

where $\beta(p')$ is the distribution of the magnitude of the dimensionless dipole moment $p' \equiv P/ea$. We shall assume $\beta(p') = \delta(p' - p)$, i.e., $P_i/ea = p$ for all i.

We now can calculate the spectral density factor $e^{-nC(x)}$ in (17):

$$nC(x) = n\int_0^{2\pi} d\phi \int_0^{\pi} d\theta\,\sin\theta \int_R^{\infty} dr\, r^2 (1 - e^{ixp\cos\theta\, a^2/r^2})$$

$$= 2\pi n a^3 \int_0^{\pi} d\theta\,\sin\theta \int_{R/a}^{\infty} d\rho\,\rho^2 \left[1 - \cos\left(\frac{xp\cos\theta}{\rho^2}\right) - i\,\sin\left(\frac{xp\cos\theta}{\rho^2}\right)\right] . \tag{20}$$

The radius R in (20) separates an inner region surrounding our sampling position from an outer region in which the random continuum approximation of (19) may be used. In the inner region we can specify details of short-range order; a way of doing this is indicated in Sec. III E. For the moment we proceed by setting $R=0$. In (20) it is readily seen that $\mathrm{Im}[C(x)]$ vanishes, since the term $\sin[xp(\cos\theta)/\rho^2]$ is odd in $\cos\theta$. Thus, from (17), $\bar{V}=0$; this is a consequence of the isotropic approximation. After a change in integration variables,

$$nC(x)=2\pi na^3\int_0^\infty dw\, w^{-5/2}\int_{-1}^1 du\,\tfrac{1}{2}(1-\cos xpwu)$$

$$=2\pi na^3(|x|p)^{3/2}\int_0^\infty dw\, w^{-7/2}(w-\sin w)$$

$$=\tfrac{4}{15}\,na^3(2\pi p\,|x|)^{3/2}\,. \tag{21}$$

We therefore have

$$\psi(V)=(\epsilon_0 a/\pi e)\int_0^\infty dx\cos xv\; e^{-Ax^{3/2}}\,,$$

where

$$A=\tfrac{4}{15}na^3(2\pi p)^{3/2}\,. \tag{22}$$

Substituting (22) into (9) yields

$$\phi(E)=\int_{-\infty}^E \psi(V)\,dV$$

$$=\pi^{-1}\int_{-\infty}^\epsilon dv\int_0^\infty dx\cos xv\; e^{-Ax^{3/2}}$$

$$=\int_{-\infty}^{\epsilon/A^{2/3}} d\beta\, H(\beta)\,, \tag{23}$$

where

$$H(\beta)=\pi^{-1}\int_0^\infty dt\cos\beta t\, e^{-t^{3/2}}$$

and

$$\epsilon=E/V_0\,,\quad V_0=e/\epsilon_0 a\,.$$

$H(\beta)$ is the Holtsmark distribution.[21]

While (23) cannot be integrated analytically, we can make use of the fact that the Holtsmark distribution is intermediate in behavior between a Gaussian and a Lorentzian to obtain a close approximation for ϵ_c:

$$\begin{aligned} &G(\beta)<H(\beta)<L(\beta)\quad \text{for } \beta<0.4 \text{ and } \beta>2.8\,,\\ &G(\beta)>H(\beta)>L(\beta)\quad \text{for } 0.4<\beta<2.8\,, \end{aligned} \tag{24}$$

where

$$G(\beta)=(4\pi)^{-1/2}e^{-\beta^2/4}$$

and

$$L(\beta)=\pi^{-1}(1+\beta^2)^{-1}\,.$$

Corresponding to each of the functions G, H, and L there is a critical value β_c such that the area under the curve from $-\infty$ to β_c is equal to ϕ_c; we designate these values as β_c^G, β_c^H, and β_c^L, respectively. In the tail of the Holtsmark distribution,

$$H(\beta)\xrightarrow[|\beta|\gg 1]{}\tfrac{3}{4}(2\pi)^{-1/2}|\beta|^{-5/2}\,, \tag{25}$$

falling off just slightly faster than the Lorentzian, so that

$$|\beta_c^G|<|\beta_c^H|\lesssim|\beta_c^L|\,. \tag{26}$$

Integrating $G(\beta)$ and $L(\beta)$, we obtain

$$|\beta_c^G|=1.46\,,\quad |\beta_c^L|=\cot\pi\phi_c=1.96\,. \tag{27}$$

[The Gaussian result also follows directly from (13) with $\bar{V}=0$ and $V_{\mathrm{rms}}=\sqrt{2}$.] From (26) and (27) it is clear that a good estimate for $|\beta_c^H|$ is 1.8. Using this value in (23) to obtain the critical energy,

$$\epsilon_c\cong-1.8A^{2/3}=-1.8\,(\tfrac{4}{15})^{2/3}(na^3)^{2/3}\,2\pi p$$

and (28)

$$E_c=\epsilon_c(e/\epsilon_0 a)=-4.7n^{2/3}P/\epsilon_0\,.$$

The result expressed in (28) is the critical energy for a random dipole potential; it is proportional to the magnitude of the molecular dipole moment P and to the two-thirds power of the molecular concentration n.

E. Short-Range Order

As a final consideration we indicate the way that short-range order can be incorporated in $\psi(V)$. Since $\psi(V)$ can be interpreted as the probability density for a given V to appear at a single arbitrarily chosen position equally as well as being interpreted as the frequency count for V over all of the volume elements of the space, the short-range correlations can be included by properly specifying the $p(z)$ in (17). Stoneham[22] has considered this problem in the context of the origin of satellite lines in the linewidth case. In our notation, one can write $\psi(V)$ as in (17) but with

$$nC(x)=\textstyle\sum_{\mathrm{I}} f_z[1-e^{-ixv_z}]+n\int_{\mathrm{II}} dz\, p(z)(1-e^{-ixv(z)})\,, \tag{29}$$

where region I is an inner region in which discrete structure is recognized and region II can be treated, as before, as a continuum. The decomposition in (29) adds further structure to the spectral density term in (17) and can be used to take into account the presence of short-range order in an amorphous system. Further details would be best entertained in the context of a specific model for a particular amorphous material, which we do not attempt here.

IV. SUMMARY

It is becoming increasingly apparent that percolation theory has something important to say about the localization-delocalization transition in amorphous semiconductors. It has been our purpose here to contribute to the implementation of the percolation concept in applications to random potentials of physical interest. Existing percolation theory deals almost entirely with percolation on lattices; the motion of an electron in a random potential corresponds to the process of percolation on a *continuum*. In our formulation of the continuum percolation problem, the central concept introduced is the energy-dependent dimensionless density ϕ defined by (1) and rendered calculable by (9). The critical density ϕ_c is the quantity which provides our working criterion $\phi(E_c) = \phi_c$ for the location of the percolation threshold energy E_c. In one dimension ϕ_c is immediately revealed to be unity, and in two dimensions we have shown that ϕ_c is $\frac{1}{2}$ for the class of symmetric random potentials satisfying (5). In three dimensions we are forced to rely on the empirically induced estimate of $\phi_c = 0.15$ proposed previously by the authors. The differences between continuum percolation processes in one, two, and three dimensions have been indicated in Fig. 2.

The percolation criterion for the location of the mobility edge E_c has been applied to several types of disordered potentials by relating $\phi(E)$ directly to a statistical characterization of these potentials. The frequently invoked Gaussian potential distribution has been treated and the results connected with the recent calculations of HL, EC, and Stern. Random-walk techniques can be used to attack more general random potentials. We have used this approach to calculate E_c for the potential distribution of an array of random dipoles, which is an initial model for an amorphous molecular solid. Finally, a way in which to include short-range order in the calculation has been discussed.

ACKNOWLEDGMENT

The authors are indebted to Scott Kirkpatrick for a critical reading of this paper and for valuable suggestions about the nature of the effective potential, as well as for bringing to our attention the papers mentioned in Ref. 15.

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[15] While this statement seems inescapable, we do not attempt here any rigorous proof. The lattice percolation analog *has* been proven by Fisher [J. Math. Phys. 2, 620 (1961)] in a generalization of a theorem of Harris [Proc. Cambridge Phil. Soc. 56, 13 (1960)]. Fisher showed that for all 2_D lattices with no crossing bonds (only nearest-neighbor coordination), an infinite cluster of filled sites (the lattice equivalent of a continent) cannot coexist with an infinite cluster of empty sites (lattice ocean). For the more difficult continuum percolation problem, the corresponding formal proof awaits future work.

[16] As a function of energy, the number of saddle points per unit area per unit energy is a sum of δ functions for a crystal potential, a continuous distribution for a random potential. [The latter is analogous to the distribution of potential minima discussed by Halperin and Lax (Ref. 13).] The point is that a disordered potential, unlike a crystal potential, does not permit a finite fraction of the saddle points to occur at any single energy value.

[17] A lower limit for the critical density can be given for a model in which the filled volume is specified by randomly placed penetrable spheres [B. I. Halperin and M. E. Fisher (private communication)]. Lattice percolation theory provides $1/z$ as a lower bound for p_c, where z (a generalization of lattice coordination n) is the number of neighboring sites directly connected to a given site. Applying this theorem, in appropriate fashion, to the random-sphere situation yields $\phi_c > 1 - e^{-1/8} = 0.117$.

[18] A speculated extrapolation to the degree of underestimation in (8) may be carried out by taking $\phi_c \approx p_c/f_{\max}$, where $f_{\max}$ is the filling factor for closest packing: $f_{\max} = 1.00$, 0.91, 0.74 in 1_D, 2_D, 3_D. This holds (trivally) for 1_D and (to a close approximation) for 2_D, and suggests that $\phi_c(3_D) \approx (0.15/0.74) \approx 0.20$.

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Spectral Dependence and Polarization Anisotropy of Optical-Transmission Modulation by Acoustoelectric Domains in CdS[†]

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(Received 2 August 1971)

The modulation by acoustoelectric domains of monochromatic light transmitted through platelets of CdS has been studied with emphasis on the spectral variation and polarization dependence of the effect near the band edge. Samples of several crystallographic orientations were used. It was found that for light propagating along the *c* axis of the crystal, the domain-induced modulation was strongest with the light polarized along the drift-field direction, in agreement with previously reported work on GaSb. The spectral variation of the transmission modulation observed with polarized light propagating perpendicular to the *c* axis was consistent with the double band edge of CdS. The modulation resembles a shift of the absorption spectrum to longer wavelengths. Recently published theoretical and experimental studies of the Franz-Keldysh effect, together with the observed spectral, polarizational, and orientational dependences of the optical modulation lead us to conclude that the modulation is produced by the Franz-Keldysh effect. Further, it is shown that the fluctuating high-frequency electric fields in the domain, rather than the "steady" field across the domain, produce the effect. Previously measured properties of the domain, such as current noise, induced birefringence, and induced light emission, corroborate this interpretation. Other proposed mechanisms are shown to be invalid.

I. INTRODUCTION

Several techniques have been used for probing acoustoelectric domains in piezoelectric semiconductors. These include direct electrical probing, microwave probing, and optical probing. One optical probing technique involves observing the transient change in optical transmission of the semiconductor sample as the domain traverses the optical probe. This technique was used by Kumar, Sliva, and Bray[1] in GaSb, and by Spears and Bray[2] in GaAs. It was demonstrated that the change in the optical-absorption coefficient correlated well with the domain strength as measured by other techniques, and that the modulation was strongest for wavelengths approaching the band edge of the semiconductor. More recently, Kumar and Hutchinson[3] and Yamamoto *et al.*[4] applied the technique to CdS and observed the characteristics of domain propagation in that material. The work being reported here is primarily concerned with the spectral variation of the transmission modulation produced by acoustoelectric domains in CdS at wavelengths near the band edge, and with the relationship between the orientational dependence of this spectral variation, including polarization anisotropy, and the band structure of CdS. These topics have been covered only briefly in the available literature. The results of the present work viewed in the light of recently published experimental and theoretical studies of the Franz-Keldysh effect, lead us to conclude that this effect is responsible for the observed modulation. However, it is the strong fluctuating electric fields in the domain, associated with the intense high-frequency phonon flux, rather than the "steady" field across the domain, which produce the effect. The presence of such strong fluctuating fields is adduced also from earlier studies of the acoustoelectric domains in CdS, namely, the high levels of current noise at current saturation, induced birefringence, and induced light emission.

II. DESCRIPTION OF EXPERIMENT

The system used for measuring transmission modulation in the presence of acoustoelectric domains is shown in Fig. 1. A Perkin-Elmer model 83 monochromator containing a quartz prism was used in conjunction with a pulsed xenon lamp. The peak pulsed-light intensity near the band edge of CdS was over 1000 times greater than that obtainable from CW arc lamps, and this high intensity was essential to the experiment. Furthermore, the xenon spectrum was relatively flat, a desirable characteristic for spectral measurements. The detector was an RCA 6217 photomultiplier which,