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Coherent-medium approximation in the stochastic transport theory of random media

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An admittance for localized physical quantities is generally related to a random walk on the basis of linear-response theory. A coherent-medium approximation is introduced to solve a master equation which is assumed to govern the random walk. The general formalism is specialized to ac hopping conduction and applied to the bond-percolation model in one- and three-dimensional systems and to a lattice model for impurity conduction in doped semiconductors. For the one-dimensional bond-percolation model, the ac conductivity obtained by the coherent-medium approximation is in good agreement with exact results for both the frequency dependence and the critical behavior. The present method predicts a percolation transition and several critical behaviors of the ac conductivity at the transition point for the bond-percolation model in a simple cubic lattice. In particular, $\lim_{\omega\to 0}\mathrm{Re}[\sigma(\omega)-\sigma(0)]/\omega^{3/2}$ diverges as $(p-p_c)^{-3/2}$ when $p=p_c+0$ and $\lim_{\omega\to 0}\mathrm{Re}\sigma(\omega)/\omega^2$ diverges as $(p_c-p)^{-3}$ when $p=p_c-0$ and in the static limit $\mathrm{Re}\sigma(\omega)$ vanishes as ω^2 for $p<p_c$ and as $\omega^{1/2}$ at $p=p_c$. The present approximation also succeeds in reproducing the typical frequency dependence of the ac conductivity of the hopping conduction in doped semiconductors, namely, the transition from the dc behavior through an ω^s dependence to a plateau as the frequency ω is increased. The theoretical results are shown to be in good agreement with experiments.

I. INTRODUCTION

The transport of physical entities (hereafter referred to as electrons) such as electrons, excitons, and phonons in disordered media is affected greatly by the nature of randomness. If the degree of randomness is sufficiently large, the electronic wave function is localized in a small region in a sense described by Anderson[1] and transport of electrons takes place by hopping among these localized states with the assistance of other degrees of freedom. The hopping rate between two localized centers depends on several physical parameters associated with these centers, especially on the spatial distance between the centers and the energy difference between the initial and final states. These parameters fluctuate from center to center, since the environment of each center is statistically distributed. Because hopping is assisted by other degrees of freedom as well as environmental fluctuations, the site-to-site transition probabilities are random variables.

To obtain the ac conductivity for the stochastic transport described above, Scher and Lax[2] (hereafter referred to as SL) first constructed a tractable model system on a discrete lattice which simulates actual disordered systems and employed the continuous-time random walk (CTRW) formalism due to Montroll and Weiss.[3] Their result for the frequency and temperature dependence of the ac conductivity is in excellent agreement with experiments for doped semiconductors[4] and spinel-type MnCoNiCu complex oxide semiconductors[5] for a wide range of frequency. The SL procedure in the original form, however, treats all sites as equivalent and independent, and hence it may not be an appropriate approximation for a certain type of randomness in which the microscopic structure plays an important role in the problem.

In the present paper, we develop a new approximate procedure for the evaluation of the ac conductivity due to hopping in random media. In Sec. II, the relation between the ac conductivity and a random walk which SL have used as a basic equation is rederived from a general view point on the basis of linear-response theory. The frequency-dependent admittance is expressed in terms of the Laplace transform $P(\vec{s},u|\vec{s}_0)$ of the conditional probability $P(\vec{s},t|\vec{s}_0,0)$ of finding a moving particle at a point $\vec{s}$ at time t if it was at a point $\vec{s}_0$ at $t=0$. This relation holds quite generally if the physical quantities entering the admittance can be assumed to be diagonal for a set of local basis functions.

In the expression for the ac conductivity of a discrete lattice in terms of $P(\vec{s},u|\vec{s}_0)$, randomness appears only in the probability function $P(\vec{s},u|\vec{s}_0)$

which is assumed to obey a master equation. In Sec. III, we show that master equations can be solved by a generalized application of the coherent potential approximation,[6–9] i.e., a kind of scattering T matrix produced by a disordered unit embedded in a coherent medium is forced to be zero on the average to self-consistently determine the coherent medium and hence the coherent transition probability. In a sense, the coherent potential approximation constitutes a formal expression of earlier effective-medium approximations used to obtain the conductivity of a resistor network.[10,11]

The new method is applied to several representative examples for hopping conduction problems in Sec. IV: the bond-percolation model in one dimension and three dimensions and a model lattice system for topologically disordered systems. The present approximation gives fairly good agreement with exact results for the bond-percolation model in one dimension. Moreover, the present method predicts a percolation transition for the bond-percolation model in three dimensions as is physically expected. In this scheme, the critical bond-percolation probability p_c is $\frac{1}{3}$ for a simple cubic lattice. Several critical behaviors of the ac conductivity at the percolation point are also predicted.

For the topologically disordered system, our procedure also succeeds in reproducing the qualitative behavior of the frequency dependence of the experimentally observed ac conductivity.[4,5,12,13] Namely, the real part of the obtained ac conductivity shows a power law dependence ω^s on frequency between the static limit and the saturated value at higher frequencies and a gradual transition from one to another as the frequency is increased. The results will be compared with experiments for doped semiconductors.[4] Section V is devoted to discussion while in Appendix A we discuss the effective medium derived from the SL procedure.

II. FLUCTUATION-DISSIPATION THEOREM FOR LOCALIZED QUANTITIES

Suppose that an external field $F(t)=F_0\cos\omega t$ adds a perturbation $-AF(t)$ to the Hamiltonian of a system and we observe the response of the system through the change $\Delta B(t)$ of a physical quantity B. The response $\langle \Delta B(t)\rangle$ is written in the form

$$\langle \Delta B(t)\rangle = \mathrm{Re}\chi_{BA}(\omega)F_0 e^{i\omega t} \ . \tag{2.1}$$

The complex admittance $\chi_{BA}(\omega)$ is expressed in terms of the response function $\phi_{BA}(t)$ as

$$\chi_{BA}(\omega)=\int_0^\infty \phi_{BA}(t)e^{-i\omega t}dt \ . \tag{2.2}$$

The frequency ω should be understood to have an infinitesimal negative imaginary part to ensure convergence of this transformation.

In linear-response theory,[14,15] $\phi_{BA}(t)$ is given by

$$\phi_{BA}(t)=-\frac{\partial}{\partial t}\int_0^\beta \mathrm{Tr}[B(t)A(i\hbar\lambda)\rho]\,d\lambda \ . \tag{2.3}$$

If the time derivative of B is observed, the response function $\phi_{\dot{B}A}(t)$ is written as

$$\phi_{\dot{B}A}(t)=-\frac{\partial^2}{\partial t^2}\int_0^\beta \mathrm{Tr}[B(t)A(i\hbar\lambda)\rho]\,d\lambda \ . \tag{2.4}$$

In these expressions, $\beta=1/kT$ is temperature inverse, $B(t)=e^{iHt/\hbar}Be^{-iHt/\hbar}$ is the Heisenberg representation of B, $A(i\hbar\lambda)=e^{-\lambda H}Ae^{\lambda H}$, and $\rho=\exp(-\beta H)$ is the equilibrium density matrix, where H is the Hamiltonian of the system without the external field and k is Boltzmann's constant. By introducing $\beta-\lambda$ as a new variable the cumulative response can be shown to obey the identity

$$\int_0^\beta \mathrm{Tr}[B(t)A(i\hbar\lambda)\rho]\,d\lambda=\int_0^\beta \mathrm{Tr}[A(-i\hbar\lambda)B(t)\rho]\,d\lambda \tag{2.5}$$

Equations (2.3) and (2.4) can then be rewritten, respectively, into

$$\phi_{BA}(t)=\frac{\partial}{\partial t}\Psi_{BA}(\beta,t) \tag{2.6a}$$

and

$$\phi_{\dot{B}A}(t)=\frac{\partial^2}{\partial t^2}\Psi_{BA}(\beta,t) \ , \tag{2.6b}$$

where

$$\Psi_{BA}(\beta,t)=-\frac{1}{2}\int_0^\beta \mathrm{Tr}[|B(t)-A(-i\hbar\lambda)|^2\rho]\,d\lambda \tag{2.7}$$

differs by a constant from Eq. (2.5). Here, we have used the stationarity of the system. Insertion of Eqs. (2.3) and (2.4) with (2.6) and (2.7) into (2.2) and integration by parts yield

$$\chi_{BA}(\omega)=-\Psi_{BA}(\beta,0)+i\omega\int_0^\infty e^{-i\omega t}\Psi_{BA}(\beta,t)\,dt \tag{2.8a}$$

and a similar form for $\chi_{\dot{B}A}(\omega)$:

$$\chi_{\dot{B}A}(\omega)=-\frac{\partial}{\partial t}\Psi_{BA}(\beta,t)\bigg|_{t=0}-i\omega\Psi_{BA}(\beta,0)$$
$$-\omega^2\int_0^\infty e^{-i\omega t}\Psi_{BA}(\beta,t)\,dt \ . \tag{2.8b}$$

Now, let us assume that the physical quantities A and B are diagonal in the Hilbert space spanned by a set of orthonormalized local basis functions $|\vec{s}\rangle \equiv \phi_s(\vec{r}-\vec{s})$, where $\vec{s}$ denotes a localized site as well as a position vector. Explicitly, we assume

$$\langle \vec{s}|A|\vec{s}'\rangle = A_{\vec{s}}\delta(\vec{s},\vec{s}') \ , \tag{2.9a}$$

$$\langle \vec{s}|B|\vec{s}'\rangle = B_{\vec{s}}\delta(\vec{s},\vec{s}') \ , \tag{2.9b}$$

where $\delta(\vec{s},\vec{s}')$ is a Kronecker δ function. In addition, we assume the density matrix ρ is also diagonal with respect to the localized basis set;

$$\langle\vec{s}|e^{-\beta H}|\vec{s}'\rangle = f(\vec{s})\delta(\vec{s},\vec{s}') \ . \tag{2.10}$$

Then, the admittance $\chi_{BA}(\omega)$ and $\chi_{\dot{B}A}(\omega)$ can be rewritten as

$$\chi_{BA}(\omega) = -\frac{\beta}{2}\sum_{\vec{s}}(B_{\vec{s}}-A_{\vec{s}})^2 f(\vec{s}) + \frac{i\omega\beta}{2}\sum_{\vec{s},\vec{s}_0}(B_{\vec{s}}-A_{\vec{s}_0})^2 P(\vec{s},i\omega|\vec{s}_0)f(\vec{s}_0) \ , \tag{2.11}$$

$$\chi_{\dot{B}A}(\omega) = -\frac{\beta}{2}\sum_{\vec{s}}(B_{\vec{s}}-A_{\vec{s}})[2(\dot{B}_{\vec{s}}-A_{\vec{s}}) + i\omega(B_{\vec{s}}-A_{\vec{s}})]f(\vec{s}) - \frac{\beta\omega^2}{2}\sum_{\vec{s},\vec{s}_0}(B_{\vec{s}}-A_{\vec{s}_0})^2 P(\vec{s},i\omega|\vec{s}_0)f(\vec{s}_0) \ . \tag{2.12}$$

Here, $P(\vec{s},u|\vec{s}_0)$ is the Laplace transform of the absolute square of the Green's function $P(\vec{s},t|\vec{s}_0,0) \equiv |G(\vec{s},t|\vec{s}_0)|^2$;

$$P(\vec{s},u|\vec{s}_0) = \int_0^\infty e^{-ut}P(\vec{s},t|\vec{s}_0,0)\,dt \ , \tag{2.13}$$

where the Green's function $G(\vec{s},t|\vec{s}_0)$ is defined by

$$G(\vec{s},t|\vec{s}_0) = -i\theta(t)\langle\vec{s}|e^{-iHt/\hbar}|\vec{s}_0\rangle \tag{2.14}$$

with the Heaviside step function

$$\theta(t) = \begin{cases} 1, & t \geq 0 \\ 0, & t < 0 \end{cases} \ . \tag{2.15}$$

It should be remarked that the assumption Eq. (2.10) does not imply $G(\vec{s},t|\vec{s}_0) \sim \delta(\vec{s},\vec{s}_0)$, since we are concerned with low frequencies ω such that $\hbar\omega/kT \ll 1$ and a typical time scale we will deal with in Eq. (2.14) is $1/\omega$.

For the specific case of $B = A$, Eqs. (2.11) and (2.12) are reduced to simple forms

$$\chi_{AA}(\omega) = i\frac{\beta\omega}{2}\sum_{\vec{s},\vec{s}_0}(A_{\vec{s}}-A_{\vec{s}_0})^2 P(\vec{s},i\omega|\vec{s}_0)f(\vec{s}_0) \tag{2.16}$$

and

$$\chi_{\dot{A}A}(\omega) = -\frac{\beta\omega^2}{2}\sum_{\vec{s},\vec{s}_0}(A_{\vec{s}}-A_{\vec{s}_0})^2 P(\vec{s},i\omega|\vec{s}_0)f(\vec{s}_0) \ . \tag{2.17}$$

The quantity $P(\vec{s},t|\vec{s}_0,0)$ means the probability density of finding the particle at site $\vec{s}$ at time t if it started at site $\vec{s}_0$ at time 0, as stated before. The probability density describes the stochastic motion of the particle in a given sample. To get the admittance actually observed, we have to average Eq. (2.16) or (2.17) over the ensemble of samples. The ensemble is determined by what we are studying. In topologically disordered systems, the configuration of sites $\{\vec{s}\}$ differs from sample to sample and one must carry out an average over all possible configurations of sites $\{\vec{s}\}$. In cellular disordered systems, the set of sites $\{\vec{s}\}$ forms a fixed array of lattice points and hence the average is taken over transition probabilities rather than $\{\vec{s}\}$. The expressions (2.16) and (2.17) also contain a sum over the initial particle location through $\sum_{\vec{s}_0} f(\vec{s}_0)$. If the ensemble average is carried out, however, the averaged quantity $\langle(A_{\vec{s}}-A_{\vec{s}_0})^2 P(\vec{s},i\omega|\vec{s}_0)\rangle$ is independent of the initial site $\vec{s}_0$. Consequently, the term $\sum_{\vec{s}_0} f(\vec{s}_0)$ in Eqs. (2.16) and (2.17) simply gives unity and we have, for example,

$$\chi_{\dot{A}A}(\omega) = -\frac{\beta\omega^2}{2}\sum_{\vec{s}}\langle(A_{\vec{s}}-A_{\vec{s}_0})^2 P(\vec{s},i\omega|\vec{s}_0)\rangle \ , \tag{2.18}$$

where $\langle\cdots\rangle$ denotes the average over the distribution of random variables.[17] As shown in SL, a system with the fixed array of sites together with an appropriate distribution of other variables can serve as a useful and tractable model for topologically disordered systems. Therefore, in the succeeding part of the present paper we assume a fixed array of sites $\{\vec{s}\}$.

To be specific, Eq. (2.18) implies that the scalar ac conductivity $\sigma(\omega)$ for an isotropic random media with the fixed array of sites is given by the generalized Einstein relation as follows[2]:

$$\sigma(\omega) = \frac{ne^2}{kT}D(\omega) \ , \tag{2.19}$$

$$D(\omega) = -\frac{\omega^2}{2d}\sum_{\vec{s}}(\vec{s}-\vec{s}_0)^2\langle P(\vec{s},i\omega|\vec{s}_0)\rangle \ , \tag{2.20}$$

where d is the dimensionality of the system, e is the electronic charge, n is the number density of carriers, and $D(\omega)$ denotes a generalized diffusion constant. Here, use has been made in Eq. (2.18) of $A = \sum_{\vec{s}}|\vec{s}\rangle e s_\alpha\langle\vec{s}|$, s_α being a Cartesian component of $\vec{s}$, and $(s_\alpha - s'_\alpha)^2 = (\vec{s}-\vec{s}')^2/d$ in isotropic media.

III. MASTER EQUATION AND COHERENT MEDIUM APPROXIMATION

Although the expression for the ac conductivity (2.19) and (2.20) has a simple form, it is still difficult to evaluate $\langle P(\vec{s},i\omega|\vec{s}_0)\rangle$ on the basis of first princi-

ples. Instead, we set up a master equation which expresses the essential features of the actual stochastic time dependence of the probability density $P(\vec{s},t|\vec{s}_0,0)$.[16,17]

Let us assume now that $P(\vec{s},t|\vec{s}_0,0)$ obeys the following master equation:

$$\frac{\partial P(\vec{s},t|\vec{s}_0,0)}{\partial t} = -\Gamma_{\vec{s}} P(\vec{s},t|\vec{s}_0,0) + \sum_{\vec{s}'\neq\vec{s}} w_{\vec{s},\vec{s}'} P(\vec{s}',t|\vec{s}_0,0) \ , \tag{3.1}$$

where

$$\Gamma_{\vec{s}} = \sum_{\vec{s}'} w_{\vec{s}',\vec{s}} \ . \tag{3.2}$$

Then, the Laplace transform $P(\vec{s},u|\vec{s}_0)$ obeys

$$(u+\Gamma_{\vec{s}})P(\vec{s},u|\vec{s}_0) - \sum_{\vec{s}'\neq\vec{s}} w_{\vec{s},\vec{s}'} P(\vec{s}',u|\vec{s}_0) = \delta(\vec{s},\vec{s}_0) \ . \tag{3.3}$$

The transition rate $w_{\vec{s},\vec{s}'}$ from $\vec{s}'$ to $\vec{s}$ is a properly chosen random quantity which describes all of the stochastic nature of the process and is assumed for the sake of simplicity to satisfy $w_{\vec{s},\vec{s}'} = w_{\vec{s}',\vec{s}}$.

For later convenience let us introduce the operators[18]

$$\hat{P}(u) = \sum_{\vec{s},\vec{s}_0} |\vec{s}\rangle P(\vec{s},u|\vec{s}_0)\langle\vec{s}_0| \ , \tag{3.4}$$

$$\hat{H} = -\sum_{\vec{s}} |\vec{s}\rangle \Gamma_{\vec{s}} \langle\vec{s}| + \sum_{\vec{s}\neq\vec{s}'} |\vec{s}\rangle w_{\vec{s},\vec{s}'} \langle\vec{s}'| \ . \tag{3.5}$$

Then Eq. (3.3) can be written in a form

$$(u\hat{1}-\hat{H})\hat{P}(u) = \hat{1} \tag{3.6}$$

which makes $\hat{P}(u)$ behave as a propagator or resolvent, and $\hat{H}$ plays the role of the corresponding Hamiltonian. Of course, $\hat{H}$ bears no simple relationship to the Hamiltonian H of the underlying problem, as used following Eq. (2.4). We may call $\hat{P}(u)$ a random-walk propagator.

However, we exploit this analogy to the usual Hamiltonian formulation to construct a coherent-medium approximation for master equations. In particular, the formal solution for $\hat{P}$ is $(u\hat{1}-\hat{H})^{-1}$ so that $P(\vec{s},u|\vec{s}_0)$ is given by the $\vec{s}\,\vec{s}_0$ matrix element of the propagator:

$$P(\vec{s},u|\vec{s}_0) = \langle\vec{s}|(u\hat{1}-\hat{H})^{-1}|\vec{s}_0\rangle \ . \tag{3.7}$$

To evaluate the ac conductivity by Eqs. (2.19) and (2.20), we must calculate the ensemble average of $P(\vec{s},u|\vec{s}_0)$ which is written formally as

$$\langle P(\vec{s},u|\vec{s}_0)\rangle = \langle\vec{s}|(u\hat{1}-\hat{\Sigma})^{-1}|\vec{s}_0\rangle \ , \tag{3.8}$$

where the coherent Hamiltonian operator $\hat{\Sigma}$ is defined by

$$\langle(u\hat{1}-\hat{H})^{-1}\rangle \equiv (u\hat{1}-\hat{\Sigma})^{-1} \ . \tag{3.9}$$

So far, we have not specified the structure of the coherent medium described by $\hat{\Sigma}$. The structure of $\hat{\Sigma}$ should be determined by the nature of actual system which we treat. In the present paper, we restrict our discussion to a case where the set of sites $\{\vec{s}\}$ forms a regular lattice and the transition rate $w_{\vec{s},\vec{s}'}$ is zero unless two sites $\vec{s}$ and $\vec{s}'$ are nearest neighbor to each other. Though this model system is a very simplified one, it will work well if the hopping probabilities $w_{\vec{s},\vec{s}'}$ are given a suitable distribution.

Now, let us introduce an approximation to obtain the coherent medium. Suppose that in a coherent (average) medium all the transition probabilities associated with a given pair of nearest-neighbor sites (say 1 and 2) are given their specific rather than average values. The effective Hamiltonian $\hat{H}_A$ for this system is

$$\hat{H}_A = \hat{\Sigma} + \hat{V} \ , \tag{3.10}$$

where $\hat{\Sigma}$ is the coherent part, defined in Eq. (3.9) and

$$\hat{V} = |1\rangle(w_C - w_{21})\langle 1| + |1\rangle(w_{12}-w_C)\langle 2| + |2\rangle(w_{21}-w_C)\langle 1| + |2\rangle(w_C - w_{12})\langle 2| \ , \tag{3.11}$$

is the localized perturbation produced by using real rather than average values on the 1–2 bond. The as yet unknown coherent transition rate $w_C \equiv w_C(u)$ is introduced to describe the effective Hamiltonian as

$$\hat{\Sigma} = -zw_C \sum_{\vec{s}} |\vec{s}\rangle\langle\vec{s}| + w_C \sum_{\substack{\vec{s}'\neq\vec{s} \\ (\text{n.n.})}} |\vec{s}\rangle\langle\vec{s}'| \ , \tag{3.12}$$

where z is coordination number of the lattice. We determine the unknown parameter w_C in a self-consistent manner by the condition

$$\langle(u\hat{1}-\hat{H}_A)^{-1}\rangle = (u\hat{1}-\hat{\Sigma})^{-1} \ , \tag{3.13}$$

where $\langle\cdots\rangle$ denotes the average over the distribution of w_{12}. The situation is schematically shown in Fig. 1. In this figure, circles and bonds represent, respectively, the diagonal and off-diagonal matrix elements of $\hat{H}_A$ and $\hat{\Sigma}$. The diagonal element of a site is a sum of off-diagonal element of bonds connected to the site, which is designated by a partition of the diagonal element.

The explicit form of condition Eq. (3.13) is rewritten as a matrix equation:

$$\langle\bar{V}(\bar{1}-\bar{P}\bar{V})^{-1}\rangle = 0 \ , \tag{3.14}$$

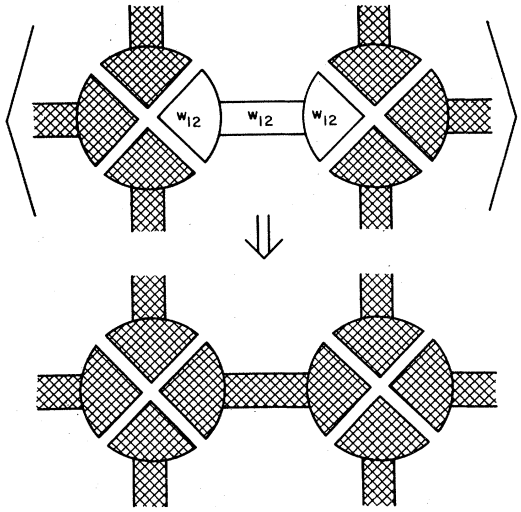

FIG. 1. Schematic illustration of the coherent-medium approximation. The hatched region denotes the coherent medium. The partition of the diagonal part (circle) denotes that the diagonal part is minus the sum of the off-diagonal parts (bonds) connected to it.

where

$$\bar{V}=\begin{pmatrix} w_C-w_{21} & w_{12}-w_C \\ w_{21}-w_C & w_C-w_{12}\end{pmatrix} , \tag{3.15}$$

$$\bar{P}=\begin{pmatrix} \bar{P}_{11} & \bar{P}_{12} \\ \bar{P}_{21} & \bar{P}_{22}\end{pmatrix} \tag{3.16}$$

with $\bar{P}_{ij}\equiv\bar{P}(\vec{i},u|\vec{j})=\langle\vec{i}|(u\hat{1}-\hat{\Sigma})^{-1}|\vec{j}\rangle$ and $\bar{1}$ denotes the 2×2 unit matrix. Since $\bar{P}_{ij}$ is a function of w_C, Eq. (3.14) must be understood as a self-consistent condition for w_C. A straightforward calculation shows that the matrix equation (3.14) ends up as a single condition

$$\left\langle\frac{w_C-w_{12}}{1-2(\bar{P}_{11}-\bar{P}_{12})(w_C-w_{12})}\right\rangle=0 . \tag{3.17}$$

Here, we have used the symmetry $\bar{P}_{12}=\bar{P}_{21}$, $\bar{P}_{11}=\bar{P}_{22}$, and $w_{12}=w_{21}$. From the definition of $\bar{P}_{ij}$, we have an identity

$$\bar{P}_{11}-\bar{P}_{12}=(1-u\bar{P}_{11})/zw_C . \tag{3.18}$$

It is easy to see that $\bar{P}(\vec{s},u|\vec{s}_0)$ is given by

$$\bar{P}(\vec{s},u|\vec{s}_0)=\frac{1}{N}\sum_{\vec{k}}\frac{e^{-i\vec{k}\cdot(\vec{s}-\vec{s}_0)}}{u+zw_C(1-f_{\vec{k}})} \tag{3.19}$$

with

$$f_{\vec{k}}=\frac{1}{z}\sum_{\vec{s}}e^{i\vec{k}\cdot(\vec{s}-\vec{s}_0)} , \tag{3.20}$$

where $\sum_{\vec{s}}$ sums over nearest neighbors of site $\vec{s}_0$ and N is the total number of sites in the system.

Specifically, it is apparent that the generalized diffusion constant $D(\omega)$ in the coherent medium approximation becomes

$$D(\omega)=a^2w_C(i\omega) , \tag{3.21}$$

where a is the lattice constant.

IV. APPLICATIONS

The present formalism for the admittance is quite general and applicable to various quantities. As a representative example, we shall consider the ac conductivity due to hopping which is governed by the master equation (3.1) with several kinds of distribution of $w_{\vec{s},\vec{s}'}$. We are interested in the frequency dependence of the ac conductivity $\sigma(\omega)$. Since $\sigma(\omega)$ always bears a factor $ne^2a^2w_0/kT$, we shall be concerned with the dimensionless ac conductivity $\tilde{\sigma}(\omega)\equiv\sigma(\omega)/(ne^2a^2w_0/kT)$ or equivalently the dimensionless diffusion constant $\tilde{D}(\omega)\equiv D(\omega)/a^2w_0$, where w_0 is a certain scaling factor of $w_{\vec{s},\vec{s}'}$.

A. Bond-percolation model in one-dimensional chain

Let us consider the bond-percolation model in a one-dimensional chain in which $w_{\vec{s},\vec{s}'}$ is distributed according to the following function:

$$P(w_{\vec{s},\vec{s}'})=p\delta(w_{\vec{s},\vec{s}'}-w_0)+(1-p)\delta(w_{\vec{s},\vec{s}'}) . \tag{4.1}$$

In other words, a bond is broken randomly with probability $(1-p)$ and loses its ability to transfer electrons through it.

Since the diagonal element $\bar{P}(\vec{s},u|\vec{s})$ of the propagator for the coherent medium of one dimension is given by

$$\bar{P}(\vec{s},u|\vec{s})=[u(u+4w_C)]^{-1/2} , \tag{4.2}$$

the self-consistency condition (3.17) with distribution function (4.1) yields

$$w_C/w_0=\frac{1}{\tilde{u}}\{\tilde{u}+2(1-p)^2-(1-p)\times[(\tilde{u}+2)^2+4p(p-2)]^{1/2}\} , \tag{4.3}$$

where $\tilde{u}=u/w_0$. Therefore, the dimensionless diffusion constant is given by

$$\tilde{D}(\omega)=\frac{1}{\tilde{\omega}}\{\tilde{\omega}-2(1-p)^2i+(1-p)\times[(2+i\tilde{\omega})^2+4p(p-2)]^{1/2}i\} \tag{4.4}$$

with $\tilde{\omega}=\omega/w_0$. The real and imaginary parts of $\tilde{D}(\omega)$ are plotted in Fig. 2 for various values of the

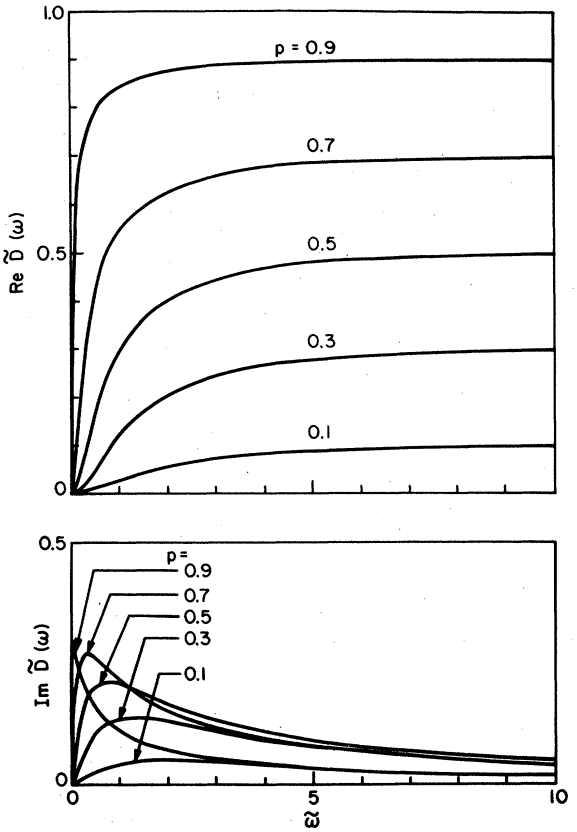

FIG. 2. Frequency and probability dependence of the real and imaginary parts of the dimensionless ac conductivity for the one-dimensional bond-percolation model.

probability p. As is expected, the static limit of the diffusion constant is always zero except for $p=1$. It is straightforward to observe the limiting behavior of $\tilde{D}(\omega)$, namely,

$$\tilde{D}(\omega) \to \frac{p(2-p)}{8(1-p)^4}\tilde{\omega}^2 + \frac{p(2-p)}{4(1-p)^2}\tilde{\omega} i \quad (\tilde{\omega} \to 0) \tag{4.5}$$

and

$$\tilde{D}(\omega) \to p - \frac{2p(1-p)(2-p)}{\tilde{\omega}^2} + \frac{2p(1-p)}{\tilde{\omega}} i \quad (\tilde{\omega} \to \infty) \ . \tag{4.6}$$

The dimensionless diffusion constant, Eq. (4.4), is in good agreement with exact results for both the frequency dependence and the critical behavior.[19]

B. Bond-percolation model in a simple cubic lattice

Next, let us apply the present method to the bond-percolation model in a simple cubic lattice. The transition rate $w_{\vec{s},\vec{s}'}$ between nearest-neighbor sites again obeys the distribution (4.1). In order to obtain simple analytic results we approximate $\bar{P}(\vec{s},u|\vec{s})$ for the simple cubic lattice by

$$\bar{P}(\vec{s},u|\vec{s}) = 2\{u + zw_C + [u(u+2zw_C)]^{1/2}\}^{-1} \ . \tag{4.7}$$

This approximation is equivalent to assuming that $f_{\vec{k}}$ [Eq. (3.20)] for the simple cubic lattice has a simplified density of states

$$\sum_{\vec{k}} \frac{\delta(\epsilon - f_{\vec{k}})}{N} = \frac{2(1-\epsilon^2)^{1/2}}{\pi} \tag{4.8}$$

with the appropriate behavior at the band edges. Then, the self-consistency condition to determine w_C is reduced to an algebraic equation of fourth order. We can easily find the physical solution which is a branch continuous to the proper solution at $p=1$. Figure 3 shows the frequency dependence of the diffusion constant $\tilde{D}(\omega)$. Using a series expansion, we obtain the limiting behavior of $\tilde{D}(\omega)$: (i) as $\tilde{\omega} \to \infty$:

$$\tilde{D}(\omega) = p + \frac{2p(1-p)}{\tilde{\omega}} i - \frac{p(1-p)(7p+4)}{\tilde{\omega}^2} \ , \tag{4.9}$$

and (ii) as $\tilde{\omega} \to 0$:

$$\tilde{D}(\omega) = \begin{cases} \frac{3}{2}(p-p_c) + \dfrac{F(1)(1-p)}{12(p-p_c)}\tilde{\omega} i + \dfrac{C(1-p)}{36\sqrt{2}(p-p_c)^{3/2}}\tilde{\omega}^{3/2}(1-i) & \text{for } p > p_c \quad (4.10) \\ \dfrac{\sqrt{F(1)}}{2\sqrt{6}}\tilde{\omega}^{1/2}(1+i) & \text{for } p = p_c \quad (4.11) \\ \dfrac{1-\sqrt{3}(p_c-p)^{1/2}}{9(p_c-p)}\tilde{\omega} i + \dfrac{[2+3(p_c-p)-3\sqrt{3}(p_c-p)^{1/2}](1-p)}{162(p_c-p)^3}\tilde{\omega}^2 & \text{for } p < p_c \ , \quad (4.12) \end{cases}$$

where $p_c = \frac{1}{3}(=2/z)$, and $F(1)=2$ and $C=2\sqrt{2}$. If we use the exact expression for $\bar{P}(\vec{s},u|\vec{s})$ of the simple cubic lattice, $F(1)$ and C must be replaced by 1.516 38,[20] and $3\sqrt{6}/2\pi$,[21] respectively, and Eq. (4.12) is rewritten as

$$\tilde{D}(\omega) = \alpha\tilde{\omega} i + \frac{\alpha^4(1-p)}{\alpha^2(p_c-p) + F'(1+1/6\alpha)/108}\tilde{\omega}^2 \ , \tag{4.13}$$

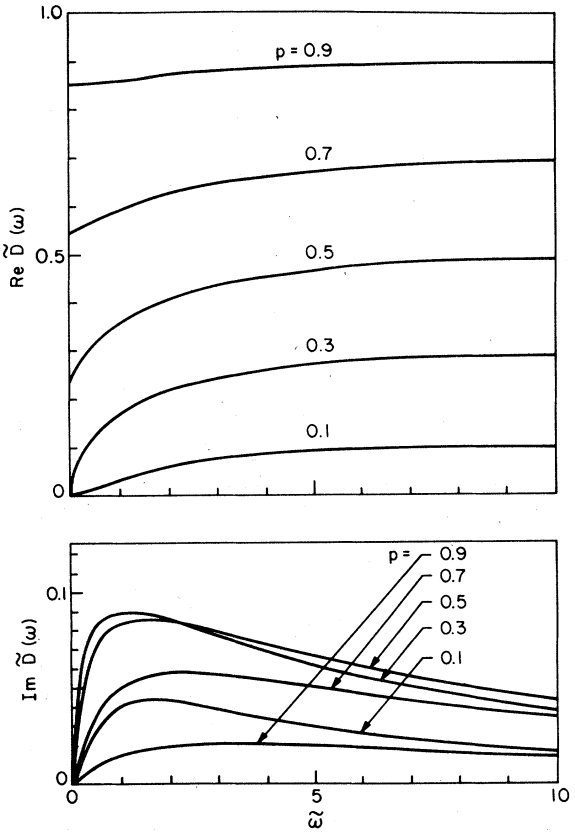

FIG. 3. Frequency and probability dependence of the real and imaginary parts of the dimensionless ac conductivity for the bond-percolation model in the simple cubic lattice.

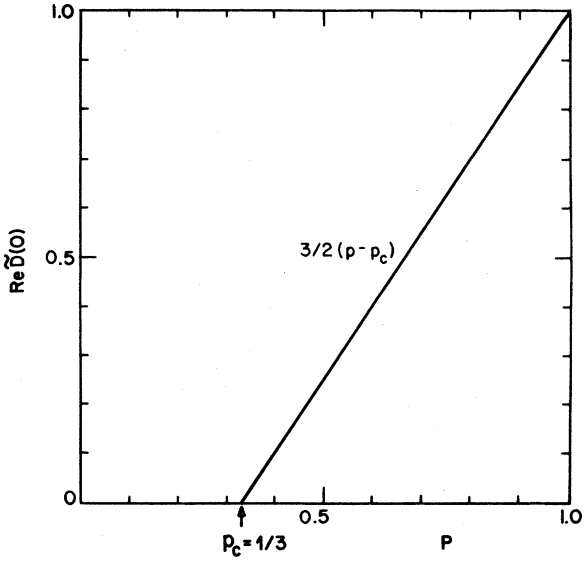

FIG. 4. Probability dependence of the static dimensionless conductivity for the bond-percolation model in the simple cubic lattice.

where α is a solution of the implicit equation

$$F(1+1/6\alpha)=18(p_c-p)\alpha \ . \tag{4.14}$$

Here, $F(\zeta)$ is defined by

$$F(\zeta)=\frac{1}{\pi^3}\int\int_0^{\pi}\int\frac{dx\,dy\,dz}{\zeta-\frac{1}{3}(\cos x+\cos y+\cos z)} \ , \tag{4.15}$$

and $F'(\zeta)=dF(\zeta)/d\zeta$. Near $p=p_c$, both coefficients in Eq. (4.13) become very large and their leading terms are again expressed in terms of $F(1)$:

$$\tilde{D}(\omega)\sim\frac{F(1)}{18(p_c-p)}\tilde{\omega}i + \frac{F(1)^2(1-p)}{324(p_c-p)^3}\tilde{\omega}^2 \quad (p\sim p_c-0) \ . \tag{4.16}$$

Obviously, the static diffusion constant $\tilde{D}(0)$ and hence the static conductivity is nonzero for $p>p_c\equiv\frac{1}{3}$, while it is identically zero for $p\leq p_c$, that is

$$\tilde{D}(0)=\begin{cases}3(p-p_c)/2 & (p\geq p_c)\\ 0 & (p\leq p_c)\end{cases} \ . \tag{4.17}$$

Therefore, the critical bond-percolation probability p_c for this model is $\frac{1}{3}$. The dependence of the static diffusion constant on p is shown in Fig. 4.

At $p=p_c$, the system shows several critical behaviors. First of all, the static conductivity vanishes linearly at $p\rightarrow p_c^+$, as in Eq. (4.17). In the static limit, we have

$$\lim_{\tilde{\omega}\rightarrow 0}\frac{\mathrm{Re}[\sigma(\tilde{\omega})-\sigma(0)]}{\tilde{\omega}^{3/2}}\sim\frac{1}{(p-p_c)^{3/2}} \quad (p>p_c) \ , \tag{4.18}$$

$$\lim_{\tilde{\omega}\rightarrow 0}\frac{\mathrm{Re}\sigma(\tilde{\omega})}{\tilde{\omega}^2}\sim\frac{1}{(p_c-p)^3} \quad (p<p_c) \ , \tag{4.19}$$

and

$$\lim_{\tilde{\omega}\rightarrow 0}\frac{\mathrm{Im}\sigma(\tilde{\omega})}{\tilde{\omega}}\sim\frac{1}{|p-p_c|} \ . \tag{4.20}$$

As we can see from Eqs. (4.10)–(4.12), the quantity $\mathrm{Re}[\sigma(\omega)-\sigma(0)]$ vanishes as $\omega^{3/2}$ above p_c and as ω^2 below p_c, and $\mathrm{Im}[\sigma(\omega)-\sigma(0)]$ vanishes linearly in frequency near the static limit. At $p=p_c$ these quantities approach zero as $\omega^{0.5}$ with frequency.

C. Impurity conduction

Let us now apply the present procedure to impurity conduction in an n-type compensated semiconductor at a low temperature where transport takes place by an electron hopping from a neutral donor to an ionized donor.

In order to simplify the problem, we first construct a lattice model which simulates the actual doped semiconductor. First, consider a simple cubic lattice whose lattice constant is $a \equiv (3/4\pi N_D)^{1/3}$, where N_D is the donor concentration. The nearest-neighbor transition rate $w_{\vec{s},\vec{s}'}$ is assumed to obey a certain distribution, which is derived by combining a transition rate

$$w_{\vec{s},\vec{s}'} \equiv w(|\vec{s}-\vec{s}'|) = w_0 e^{-|\vec{s}-\vec{s}'|/R_d} \tag{4.21}$$

and the Hertz distribution for the nearest-neighbor distance $r \equiv |\vec{s}-\vec{s}'|$ in the homogeneous random media

$$N(r) = \frac{4\pi N_D}{z} r^2 \exp\left(-\frac{4\pi N_D}{3z} r^3\right) . \tag{4.22}$$

Here, R_d is half the effective Bohr radius, z (=6) is the coordination number of the lattice and, w_0 is a function of the temperature.[2]

Now, we can easily apply the procedure described in Sec. III to obtain the ac conductivity. In fact, Eq. (3.17) reads as

$$\int_0^\infty \frac{N(r)[w_C - w(r)]}{1-2(\bar{P}_{11}-\bar{P}_{12})[w_C - w(r)]}\, dr = 0 . \tag{4.23}$$

Using Eq. (4.7) as the Laplace transform $\bar{P}(\vec{s},u|\vec{s})$ of the transition probability in the coherent medium, the self-consistent simultaneous Eqs. (3.18), (4,7), and (4.23) with the distribution (4.22) have been numerically solved. Figures 5 and 6 show the frequency

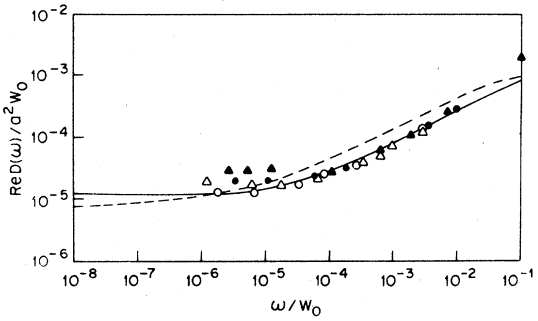

FIG. 5. Comparison of the theoretical and experimental values of $\mathrm{Re}\tilde{D}(\omega)$ for $N_D = 2.7\times10^{17}$ cm^{-3}. The solid line is the present result and the broken line is the SL result. The latter and experimental data for $\mathrm{Re}\sigma(\omega)$ are taken from Ref. 2. The dimensionless ac conductivity or diffusion constant $\tilde{D}(\omega) = \sigma(\omega)/(ne^2/kT)(a^2 w_0)$ with $n = 0.8\times10^{15}$ cm^{-3} and $a = (4\pi N_D/3)^{-1/3}$ is shown.

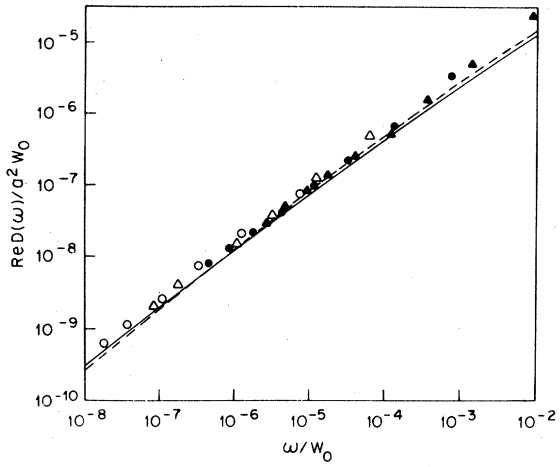

FIG. 6. Comparison of the theoretical and experimental values of $\mathrm{Re}\tilde{D}(\omega)$ for $N_D = 1.2\times10^{16}$ cm^{-3}. Symbols are the same as in Fig. 5. According to SL, a is assumed to be $0.9\ (4\pi N_D/3)^{-1/3}$ in this plot.

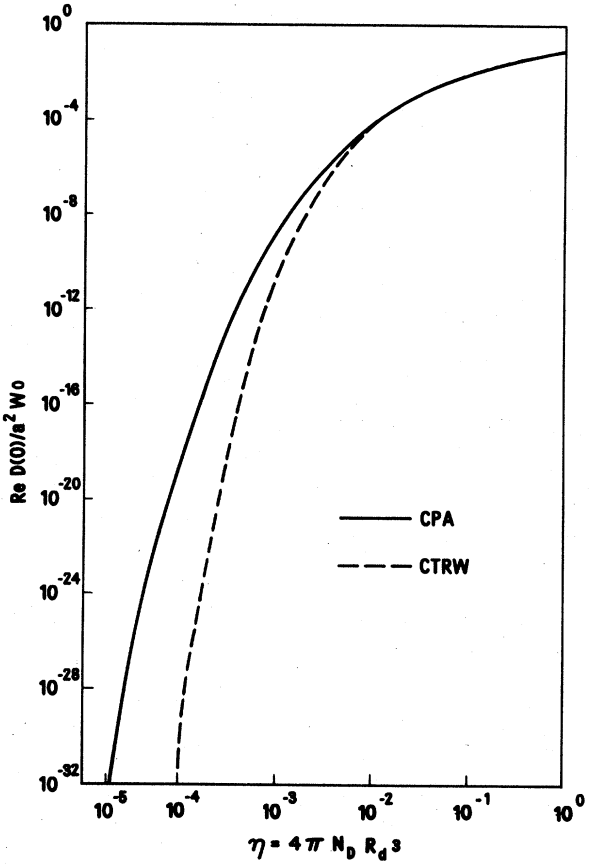

FIG. 7. Comparison of the present method (solid line) and the SL method (broken line) for the static dimensionless diffusion constant. $\mathrm{Re}D(0)/a^2 w_0 = \frac{1}{2} \times \exp[-(18\ln\frac{3}{2})^{1/3}\eta^{-1/3}]$ for the present method and $e^{\gamma}\eta^{1/4}\exp[-2\eta^{-1/2}/3]/6\sqrt{\pi}$ for the SL method with $\eta = 4\pi N_D R_d^3$ and $\gamma = 0.5772$.

dependence of the real part of the dimensionless ac conductivity or $\tilde{D}(\omega)$, for $N_D = 2.7 \times 10^{17}$ cm^{-3} and 1.2×10^{16} cm^{-3}, respectively. We have used $R_d = 12$ Å, as discussed in SL. In these figures, the solid line is the present result, the broken line is the result by SL and the small symbols are experiments for Si doped with P and B.[4] The present results are in an excellent agreement with experiments and reproduce the well-known ω^s dependence. The exponent s reads as 0.52 (for three decades of ω) in Fig. 5 and 0.77 (for seven decades of ω) in Fig. 6.

Simple algebra shows that the static diffusion constant $\tilde{D}(0)$ behaves as

$$\tilde{D}(0) \sim \tfrac{1}{2} \exp[-(18 \ln \tfrac{3}{2})^{1/3} \eta^{-1/3}] \qquad (4.24)$$

for small $\eta \equiv 4\pi N_D R_d^3$. A comparison between Eq. (4.24) and the SL formula is given in Fig. 7. The present method gives a higher static limit of $\tilde{D}(\omega)$ than that of the SL method for smaller values of η.

V. DISCUSSION

In the present paper, we have derived a general expression of the admittance for localized physical quantities. The expression is quite general and applicable to various systems. Then, the coherent-medium approximation has been first applied to solve the master equation which is assumed to govern the stochastic motion of the localized quantity. As a specific example, we have studied the hopping conduction in one- and three-dimensional systems.

The present approach essentially requires a cluster treatment, since the random distribution of $w_{\vec{s},\vec{s}'}$ in the Hamiltonian (3.5) cannot be expressed by single site disorder. It is well known that a certain type of the cluster CPA (coherent-potential approximation) is not analytic.[22] The cluster treatment employed here, however, is identical with the homomorphic cluster CPA,[23] the analyticity of which has been rigorously proved.[24] We have set one of the transition rates $w_{\vec{s},\vec{s}'}$ as a disordered unit, but it is easy to extend the present formulation to the multibond cluster.

The difference between the present approach and the SL method is apparent if one compares Eqs. (3.13) and (A5). In fact, the SL method treats all sites as equivalent and independent, and hence the reciprocity between two sites is not taken into account properly. On the other hand, the present method treats a pair of sites as a unit and the reciprocity is included correctly. These two methods have been compared in each application in Sec. IV. Both methods provide a qualitatively good frequency and probability dependence of the ac conductivity for one-dimensional bond-percolation model; a closer comparison is given elsewhere.[19]

As for the bond-percolation model in three-dimensional systems, the present method predicts a percolation transition at $p = p_c$ and various critical behaviors [Eqs. (4.17)–(4.20)] of the conductivity at the percolation threshold. The predicted value $p_c = \frac{1}{3}$ for simple cubic lattice is bigger than the critical bond-percolation probability 0.254 estimated by Monte Carlo methods,[25] though it coincides with the value given by the effective medium treatment of the resistor network.[10]

The critical behaviors of the conductivity in resistor networks[10,26,27] and composite media[28] have been discussed by making use of computer simulation and scaling theory. A direct comparison of the present result with those for resistor networks is possible only for the dc conductivity above the percolation threshold. The present method predicts that the dc conductivity of the bond-percolation model for the hopping conduction vanishes linearly with $p - p_c$. The critical exponent again coincides with the result obtained by the effective medium treatment of the resistor network.[10] The computer simulation yields $\sigma(0) \sim (p - p_c)^{1.6}$ near p_c for the bond-percolation model of the resistor network on the simple cubic lattice.[10,27]

The imaginary part of $\sigma(\omega)/\omega$ can be related to the real part of dielectric constant, so that the critical behavior (4.20) implies the divergence of the real part of dielectric constant at the percolation threshold. Actually, such a divergence of the dielectric constant has recently been observed in Ag-KCl random composite,[29] where a percolation model is supposed to be adequate. The critical index (=1.0) in Eq. (4.13) is somewhat larger than the observed value (=0.73) in the Ag-KCl composite system.

Although we have used a lattice model for topologically disordered systems, the coherent medium approximation as well as the SL method have produced a frequency dependence of the ac hopping conductivity which is in an excellent agreement with experiments for doped semiconductors. Both methods also give the same temperature dependence of the ac conductivity, since it is governed by the temperature dependence of w_0. However, the static conductivity given by these methods shows a big difference for low-concentration regions as one can see in Fig. 7.

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APPENDIX A: THE CTRW METHOD AND COHERENT MEDIUM

Let us divide the Hamiltonian operator $\hat{H}$ (3.5) into a purely diagonal part $\hat{\Gamma}$ and a purely off-diagonal part $\hat{W}$:

$$\hat{H}=\hat{\Gamma}+\hat{W} \ , \tag{A1}$$

where

$$\hat{\Gamma}=-\sum_{\vec{s}}|\vec{s}\rangle\Gamma_{\vec{s}}\langle\vec{s}|, \quad \hat{W}=\sum_{\vec{s}\neq\vec{s}'}|\vec{s}\rangle w_{\vec{s},\vec{s}'}\langle\vec{s}'| \ . \tag{A2}$$

Off-diagonal matrix elements of $\hat{\Gamma}$ and diagonal matrix elements of $\hat{W}$ are identically zero;

$$\langle\vec{s}|\hat{\Gamma}|\vec{s}'\rangle=-\Gamma_{\vec{s}}\,\delta(\vec{s},\vec{s}') \ , \tag{A3a}$$

$$\langle\vec{s}|\hat{W}|\vec{s}'\rangle=w_{\vec{s},\vec{s}'}[1-\delta(\vec{s},\vec{s}')] \ . \tag{A3b}$$

Now, let us expand the propagator $(u\hat{1}-\hat{H})^{-1}$ into a series

$$\frac{1}{(u\hat{1}-\hat{H})}=\frac{1}{(u\hat{1}-\hat{\Gamma}-\hat{W})}=\frac{1}{(u\hat{1}-\hat{\Gamma})}\left[\hat{1}+\sum_{k}[\hat{W}(u\hat{1}-\hat{\Gamma})^{-1}]^{k}\right] \ . \tag{A4}$$

In the SL procedure,[2,17] the average of Eq. (A4) is given in a Hartree approximation by

$$\left\langle\frac{1}{(u\hat{1}-\hat{H})}\right\rangle=\left\langle\frac{1}{(u\hat{1}-\hat{\Gamma})}\right\rangle\left[\hat{1}+\sum_{k}\left\langle\hat{W}\frac{1}{(u\hat{1}-\hat{\Gamma})}\right\rangle^{k}\right] \ . \tag{A5}$$

Since

$$\frac{1}{(u\hat{1}-\hat{\Gamma})}=\sum_{\vec{s}}|\vec{s}\rangle\frac{1}{(u+\Gamma_{\vec{s}})}\langle\vec{s}| \tag{A6}$$

and

$$\hat{W}\frac{1}{(u\hat{1}-\hat{\Gamma})}=\sum_{\vec{s}\neq\vec{s}'}|\vec{s}\rangle\frac{w_{\vec{s},\vec{s}'}}{(u+\Gamma_{\vec{s}'})}\langle\vec{s}'| \ , \tag{A7}$$

we have

$$\hat{\Phi}(u)\equiv\left\langle\frac{1}{(u\hat{1}-\hat{\Gamma})}\right\rangle=\sum_{\vec{s}}|\vec{s}\rangle\tilde{\Phi}(\vec{s},u)\langle\vec{s}| \tag{A8}$$

and

$$\hat{\psi}(u)\equiv\left\langle\hat{W}\frac{1}{(u\hat{1}-\hat{\Gamma})}\right\rangle=\sum_{\vec{s}\neq\vec{s}'}|\vec{s}\rangle\tilde{\psi}(\vec{s}-\vec{s}',u)\langle\vec{s}'| \ , \tag{A9}$$

where

$$\tilde{\Phi}(\vec{s},u)\equiv\langle 1/(u+\Gamma_{\vec{s}})\rangle \ , \tag{A10}$$

$$\tilde{\psi}(\vec{s}-\vec{s}',u)=\langle w_{\vec{s},\vec{s}'}/(u+\Gamma_{\vec{s}'})\rangle \ . \tag{A11}$$

These functions (A10) and (A11) are the same ones as SL defined. We can easily see that $\hat{\Phi}(u)$ and $\hat{\psi}(u)$ determine the following coherent medium $\hat{\Sigma}$, which is defined through $(u\hat{1}-\hat{\Sigma})^{-1}=\langle(u\hat{1}-\hat{H})^{-1}\rangle$:

$$\hat{\Sigma}=u\hat{1}-\hat{\Phi}(u)^{-1}+\hat{\psi}(u)\hat{\Phi}(u)^{-1} \tag{A12}$$

$$=\sum_{\vec{s}}|\vec{s}\rangle\left(u-\frac{1}{\tilde{\Phi}(\vec{s},u)}\right)\langle\vec{s}|+\sum_{\vec{s}\neq\vec{s}'}|\vec{s}\rangle\frac{\tilde{\psi}(\vec{s}-\vec{s}',u)}{\tilde{\Phi}(\vec{s}',u)}\langle\vec{s}'| \ , \tag{A13}$$

that is

$$\Sigma_{\vec{s},\vec{s}}=\frac{u\tilde{\psi}(u)}{\tilde{\psi}(u)-1}=-\left\langle\frac{\Gamma_{\vec{s}}}{u+\Gamma_{\vec{s}}}\right\rangle\Big/\left\langle\frac{1}{u+\Gamma_{\vec{s}}}\right\rangle \ , \tag{A14}$$

$$\Sigma_{\vec{s},\vec{s}'}=\tilde{\psi}(\vec{s}-\vec{s}',u)\frac{u}{1-\tilde{\psi}(u)}=\left\langle\frac{w_{\vec{s},\vec{s}'}}{u+\Gamma_{\vec{s}'}}\right\rangle\Big/\left\langle\frac{1}{u+\Gamma_{\vec{s}'}}\right\rangle \ . \tag{A15}$$

Here, we have used $\tilde{\Phi}^{-1}(\vec{s},u)=u/[1-\tilde{\psi}(u)]$, independent of $\vec{s}$, and $\tilde{\psi}(u)\equiv\sum_{\vec{s}}\tilde{\psi}(\vec{s}-\vec{s}',u)$ is the key function in the SL method. Since the generalized diffusion constant $\tilde{D}(\omega)$ is in proportion to $\Sigma_{\vec{s},\vec{s}}$ in CTRW and w_C in the present theory, the difference in the coherent medium for these methods is reflected in the different frequency dependence of $\tilde{D}(\omega)$. It should be remarked that Eqs. (A14) and (A15) are general relations between coherent medium and the Laplace transform of $\psi(\vec{s}-\vec{s}',t)$, a probability density that the time between steps occurs at time t resulting in a vector displacement $\vec{s}-\vec{s}'$. In other words, a coherent medium determines the Laplace-transformed distribution $\tilde{\psi}(\vec{s}-\vec{s}',u)$ through

$$\tilde{\psi}(\vec{s}-\vec{s}',u)=\Sigma_{\vec{s},\vec{s}'}/(u-\Sigma_{\vec{s},\vec{s}}) \ . \tag{A16}$$

The expressions (A14) and (A15) have also been derived by Klafter and Silbey.[30]

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