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Generalizated effective-medium approximation for hopping transport in topologically disordered systems

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ABSTRACT

A generalized effective-medium approximation (EMA) for hopping transport in topologically disordered systems such as amorphous semiconductors or impurity bands of crystalline semiconductors is derived. In contrast with previous theories we are able to include the influence of closed loops in the path summation. These terms are responsible for the algebraic long-time tails of the velocity autocorrelation function which also show up as non-analytic terms in the low-frequency part of the ac conductivity. If the closed-loop contributions are neglected, the two-site EMA of Gochanour *et al.* and Movaghar *et al.* is re-obtained. We have tested the results of the present theory against simulations for an *r*-hopping network.

§1. INTRODUCTION

The effective-medium approximation (EMA) in the version suitable for structurally disordered systems, namely EMA-I (Gochanour *et al.* 1979, Movaghar *et al.* 1980a,b, Movaghar and Schirmacher 1981, Summerfield and Butcher 1982) has been proven to describe successfully dc and ac hopping conductivity phenomena in amorphous semiconductors and impurity bands of crystalline semiconductors. It has, however, been pointed out (Schirmacher and Wagener 1992) that the EMA-I version does not describe the long-time behaviour of the velocity autocorrelation function (VAF) correctly. Quite generally the VAF for single-particle motion in a disordered frozen environment has been shown to vary as $t^{-(d/2+1)}$ for $t \to \infty$ (van Beijeren 1982, Haus *et al.* 1983). This asymptotic behaviour manifests itself as a non-analytic low-frequency behaviour of the Laplace transform $D(\omega)$ of the VAF (generalized diffusivity) in the form $D(\omega) - D(0) \propto \omega^{d/2}$ (van Beijeren 1982, Haus *et al.* 1983). The existence of this non-analyticity is known to be a property of the exact dynamic conductivity $\sigma(\omega) \propto D(\omega)$ and is a consequence of the particle conservation law. Such a non-analyticity is not present in the dynamic conductivity as calculated in EMA-I. This is so because, in the path summation carried out to derive the EMA-I (Movaghar and Schirmacher 1981), paths with closed loops have been neglected.

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However, as pointed out by van Beijeren (1982), the long-time algebraic tails are just produced by recurrent walks, that is 'closed loops'. Schirmacher and Wagner (1992) made an attempt to repair the deficiency of the EMA-I, but this was done in a rather *ad hoc* and unsatisfactory way.

In the EMA version that we are going to derive in the present paper (a preliminary version of this paper (without derivation) was published previously (Ganter and Schirmacher 2000)), we have combined the elimination of repeated indices (renormalized perturbation expansion (Economou 1990)), applied to pairs of sites, with the path summation technique of Wertheim (1973), Logan and Winn (1988) and Winn and Logan (1989). With the help of the former the EMA is obtained as a decoupling approximation (Maraghar and Schirmacher 1981), and the latter takes care of closed paths in a systematic way.

§2. Derivation

We start with the master equation for the propagator $G_{ij}(t)$ giving the odds for the presence of the particle at site j at time t if it started initially at i:

$$\frac{\mathrm{d}}{\mathrm{d}t} G_{ij}(t) = -\sum_{\ell} w_{\ell i} G_{ij}(t) - \sum_{\ell} w_{i\ell} G_{\ell j}(t). \tag{1}$$

We treat here only the symmetric r-hopping case, that is $w_{ij} = w_{ji} = w(r_{ij})$, where r_{ij} is the distance between the sites i and j. The Laplace transform of $\mathbf{G}_{ij}$ is the ij matrix element of the resolvent $\mathbf{G}(z) = (z\mathbf{1} - \mathbf{H})^{-1}$, $z = \mathrm{i}\omega + 0$. The 'Hamiltonian' $\mathbf{H}$ is decomposed into sparse submatrices ('hopping matrices') $\mathbf{H} = \sum_{\xi} \mathbf{H}(\xi)$ associated with a single pair $\xi := (ij)$ of sites and the sum runs over all such pairs. The hopping matrices are defined as follows:

$$\mathbf{H}_{kl}(ij) := \begin{cases} -w(r_{ij}), & kl = ii \text{ or } jj, \\ w(r_{ij}), & kl = ij \text{ or } ji, \\ 0, & \text{else.} \end{cases} \tag{2}$$

We denote sites (bonds) in italic (Greek) letters.

We now proceed in two steps. First we derive a self-consistent set of equations for the averaged resolvent for which an unrenormalized locator expansion is performed. This yields a mean-field theory for the ac conductivity which does not take into account the repeated forward and backward hopping but includes closed loops in a systematic way. For a theory of hopping transport, however, it has been demonstrated to be crucial (Movaghar and Schirmacher 1981) to include the revisiting of the same site very carefully. This can be achieved by a renormalized locator expansion in which the repeated indices are eliminated systematically (Movaghar and Schirmacher 1981, Economou 1990). Therefore, in a second step we repeat the construction of the self-consistent equations for the renormalized locator expansion.

We now perform a (unrenormalized) locator expansion of the resolvent in the following way:

$$\begin{aligned} \mathbf{G}(z) &= (z\mathbf{1} - \mathbf{H})^{-1} \\ &= \frac{1}{z}\mathbf{1} + \sum_{m=1}^{\infty} \frac{1}{z^{m+1}} \sum_{\xi_1,\ldots,\xi_m} \mathbf{H}(\xi_1)\cdots\mathbf{H}(\xi_m). \end{aligned} \tag{3}$$

It is important to note that this is an expansion for the resolvent $\mathbf{G}(z)$ and not for a matrix element of the resolvent. The 'locators' are just $\mathbf{1}z^{-1}$, where $\mathbf{1}$ is the unit matrix. The configurationally averaged propagator, on the other hand, is defined in terms of diagonal and off-diagonal matrix elements of $\mathbf{G}(z)$:

$$\begin{aligned}\mathcal{G}(k,z) &:= \left\langle \frac{1}{N}\sum_{i,j} \exp(\mathrm{i}\mathbf{k}\cdot\mathbf{r}_{ij}) G_{ij}(z) \right\rangle \\ &=: \mathcal{G}(z) + \rho\bar{\mathcal{G}}(k,z) \\ &= \frac{1}{z} + \frac{1}{N}\sum_{i,j}\sum_{m=1}^{\infty}\frac{1}{z^{m+1}} \\ &\quad\times \sum_{\xi_1,\ldots,\xi_m}\int\prod_{p=1}^{N}\mathrm{d}\mathbf{r}_p p_N(\mathbf{r}_1,\ldots,\mathbf{r}_N)\exp(\mathrm{i}\mathbf{k}\cdot\mathbf{r}_{ij})[\mathbf{H}(\xi_1)\cdots\mathbf{H}(\xi_m)]_{ij}. \end{aligned} \tag{4}$$

$p_N(\mathbf{r}_1,\ldots,\mathbf{r}_N)$ is the configuration distribution function and N is the number of sites.

In order to evaluate the configuration average let us first consider complete spatial disorder (relevant for impurity bands of crystalline semiconductors):

$$p_N(\mathbf{r}_1,\ldots,\mathbf{r}_N) \approx V^{-N}, \tag{5}$$

where V is the system's volume. The terms on the right-hand side of equation (4) are of the following structure:

$$\frac{V^{-N}}{z^{m+1}}\int\prod_{p=1}^{N}\mathrm{d}\mathbf{r}_p\,\mathbf{H}(\xi_1)\cdots\mathbf{H}(\xi_{s-1})|\mathbf{H}(\xi_s)\cdots\mathbf{H}(\xi_m). \tag{6}$$

Now let X (Y) be the set of all points contained in $\xi_1,\ldots,\xi_{s-1}$ ($\xi_s,\ldots,\xi_m$). The right-hand side of equation of (6) factorizes into two independent integrals if and only if X and Y have only a single point in common:

$$X\cap Y = \{l\}. \tag{7}$$

Instead of equation (5) we now perform the generalized Kirkwood approximation in terms of the radial pair distribution function $g(r) = 1 + h(r)$:

$$\begin{aligned} p_N(\mathbf{r}_1,\ldots,\mathbf{r}_N) &\approx V^{-N}\prod_{i<j} g(r_{ij}) \\ &= V^{-N}\prod_{i<j}[1+h(r_{ij})]. \end{aligned} \tag{8}$$

After factorizing equation (8) and inserting it into equation (4) we have products of the form

$$\frac{V^{-N}}{z^{m+1}}\int\prod_{p=1}^{N}\mathrm{d}\mathbf{r}_p\,h(\eta_1)\cdots h(\eta_q)\mathbf{H}(\xi_1)\cdots\mathbf{H}(\xi_{s-1})|\mathbf{H}(\xi_s)\cdots\mathbf{H}(\xi_m). \tag{9}$$

Comparing this with equation (6) we conclude that | remains a factorization point if and only if there is no bond η_i connecting a point from $X\backslash\{l\}$ with a point in $Y\backslash\{l\}$.

Products without factorization points play a special role, because all general products may be composed by them in a unique manner.

Summing over all diagonal and off-diagonal products free of factorization points (denoted by 'no fp') leads us to the definition of the irreducible self-energies $\Sigma(z)$, given by

$$\Sigma(z) := \rho \sum_{m=1}^{\infty} \sum_{\substack{\eta_1,\ldots,\eta_q \\ \xi_1,\ldots,\xi_m}}^{\text{no fp}} \frac{V^{-(N-1)}}{z^{m-1}} \int \prod_{p=1}^{N} \mathrm{d}\mathbf{r}_p \, h(\eta_1) \cdots h(\eta_q) [\mathbf{H}(\xi_1) \cdots \mathbf{H}(\xi_m)]_{11}, \tag{10}$$

and $\Sigma(k,z)$, given by

$$\Sigma(k,z) := \rho \sum_{m=1}^{\infty} \sum_{\substack{\eta_1,\ldots,\eta_q \\ \xi_1,\ldots,\xi_m}}^{\text{no fp}} \frac{V^{-(N-1)}}{z^{m-1}} \int \prod_{p=1}^{N} \mathrm{d}\mathbf{r}_p \, h(\eta_1) \cdots h(\eta_q) \exp(\mathrm{i})\mathbf{k} \cdot \mathbf{r}_{12} [\mathbf{H}(\xi_1) \cdots \mathbf{H}(\xi_m)]_{12}. \tag{11}$$

We may use them to renormalize equation (4) exactly:

$$\mathcal{G}(z) = [z - \Sigma(z)]^{-1}, \tag{12}$$

$$\rho \bar{\mathcal{G}}(k,z) = \mathcal{G}(z) \Sigma(k,z) [\mathcal{G}^{-1}(z) - \Sigma(k,z)]^{-1}. \tag{13}$$

Logan and Winn (1988) and Winn and Logan (1989) emphasized that the representation of $\mathcal{G}(k,z)$ in terms of $\Sigma(k,z)$ is analogous to the representation of the static structure factor $S(k)$ in terms of the Ornstein–Zernike correlation function $c(k)$ in the theory of liquids.

To obtain a self-consistent theory for the liquid structure, one needs a 'closure relation' that connects $c(k)$ to the liquid potential and $S(k)$. The existing theories for an approximate evaluation of the locator expansions can be classified according to their 'closure relation' in terms of $\mathbf{H}(\xi)$ and the other quantities. The most sophisticated closure relation giving reliable results for the averaged band structure of topologically disordered systems has been formulated by Roth (1975). A somewhat simpler scheme is due to Ishida and Yonezawa (1973), which we are going to exploit:

$$\Sigma(k,z) = \rho w(k) + \rho^2 \int \frac{\mathrm{d}\mathbf{k}'}{(2\pi)^3} \times [w^2(\mathbf{k} - \mathbf{k}') - 2\, w(\mathbf{k} - \mathbf{k}')\, w(k')] \bar{\mathcal{G}}(k', z). \tag{14}$$

Here $\Sigma(z) = \Sigma(k = 0, z)$, and we have for the propagator

$$\mathcal{G}(k,z) = \frac{1}{z + D(k,z)\, k^2}, \tag{15}$$

with

$$D(k,z) k^2 = \Sigma(0,z) - \Sigma(k,z). \tag{16}$$

This constitutes a self-consistent theory for $D(\omega)$ for the r-hopping problem, but one can show that in the low-density limit the results become very poor, because multiple hops between a small group of sites and also percolative properties are not taken care of in this approximation.

We are now going to reformulate the locator expansion and the subsequent averaging procedure by successively eliminating repeated (pair) indices. We define $\mathbf{G}^{[\xi_1,\ldots,\xi_\ell]}$ to be the resolvent of the Hamiltonian in which the bonds $\xi_1, \ldots, \xi_\ell$ are missing. We further define renormalized hopping matrices by

$$\mathbf{S}(\xi, z) := \mathbf{H}(\xi)[\mathbf{1} - \mathbf{G}^{[\xi]}(z)\mathbf{H}(\xi)]^{-1} \tag{17}$$

and similar matrices, where the bonds $\xi_1, \ldots, \xi_\ell$ are missing by

$$\mathbf{S}^{[\xi_1,\ldots,\xi_\ell]}(\xi, z) := \mathbf{H}(\xi)[\mathbf{1} - \mathbf{G}^{[\xi_1,\ldots,\xi_\ell,\xi]}(z)\,\mathbf{H}(\xi)]^{-1}. \tag{18}$$

We now decouple in equations (17) and (18) all lower cross-correlations and write

$$\begin{aligned}&\langle \mathbf{S}(\xi_1, z) \cdots \mathbf{S}^{[\xi_1,\ldots,\xi_{m-1}]}(\xi_m, z)\rangle \\ &\approx \int \prod_{i=1}^{q} \mathrm{d}\mathbf{r}_{j_i}\, p_q(\mathbf{r}_{j_1}, \ldots, \mathbf{r}_{j_q}) \langle \mathbf{S}(\xi_1, z)\rangle_{[j_1,\ldots,j_q]} \cdots \langle \mathbf{S}^{[\xi_1,\ldots,\xi_{m-1}]}(\xi_m, z)\rangle_{[j_1,\ldots,j_q]}.\end{aligned} \tag{20}$$

In the same way we set (Movaghar and Schirmacher 1981)

$$\langle \mathbf{S}(\xi, z)\rangle_{[j_1,\ldots,j_q]} \approx \mathbf{H}(\xi)[1 - \langle \mathbf{G}^{[\xi]}(z)\rangle_{[j_1,\ldots,j_q]}\,\mathbf{H}(\xi)]^{-1}, \tag{21}$$

$$\langle \mathbf{G}^{[\xi]}(z)\rangle_{[j_1,\ldots,j_q]} \approx \langle \mathbf{G}^{[\xi]}(z)\rangle_{[\xi]}, \tag{22}$$

$$\langle \mathbf{S}^{[\ldots]}(\xi, z)\rangle_{[j_1,\ldots,j_q]} \approx \mathbf{H}(\xi)[\mathbf{1} - \langle \mathbf{G}^{[\ldots,\xi]}(z)\rangle_{[\xi]}\,\mathbf{H}(\xi)]^{-1}. \tag{23}$$

This can obviously be put into the form

$$\langle \mathbf{S}^{[\ldots]}(\xi, z)\rangle_{[\xi]} = \{\mathbf{1} + \langle \mathbf{S}(\xi, z)\rangle_{[\xi]}[\langle \mathbf{G}^{[\xi]}(z)\rangle_{[\xi]} - \langle \mathbf{G}^{[\ldots,\xi]}(z)\rangle_{[\xi]}]\}^{-1} \langle \mathbf{S}(\xi, z)\rangle_{[\xi]}. \tag{24}$$

It now can be shown (see appendix A) that in the thermodynamic limit the difference

$$\langle \mathbf{G}^{[\xi]}(z)\rangle_{[\xi]} - \langle \mathbf{G}^{[\ldots,\xi]}(z)\rangle_{[\xi]} \tag{25}$$

vanishes.

We therefore have

$$\langle \mathbf{S}^{[\ldots]}(\xi, z)\rangle_{[\xi]} = \langle \mathbf{S}(\xi, z)\rangle_{[\xi]} =: \tau(\xi, z). \tag{26}$$

Finally we obtain the following expression for $\mathcal{G}(k, z)$:

$$\begin{aligned}\mathcal{G}(k, z) = \frac{1}{z} + \frac{1}{N} \sum_{i,j} \sum_{m=1}^{\infty} \frac{1}{z^{m+1}} \sum_{\substack{\xi_1,\ldots,\xi_m \\ \xi_k \neq \xi_l}} \int \prod_{p=1}^{N} \mathrm{d}\mathbf{r}_p\, p_N(\mathbf{r}_1, \ldots, \mathbf{r}_N) \\ \times \exp(\mathrm{i}\mathbf{k} \cdot \mathbf{r}_{ij})\,[\tau(\xi_1) \cdots \tau(\xi_m)]_{ij},\end{aligned} \tag{27}$$

$$\tau(ij, z) := \mathbf{H}(ij)[\mathbf{1}1 - \langle \mathbf{G}^{[ij]}(z)\rangle_{[ij]}\mathbf{H}(ij)]^{-1}. \tag{28}$$

Equation (27) has the same form as equation (4) for the averaged unrenormalized propagator. Application of the path summation technique with subsequent Ishida–Yonezawa closure yields our generalized EMA equations which combine the merits of the path summation technique with those of the renormalized locator expansion:

$$\mathcal{G}(k, z) = \frac{1}{z + D(k, z)\,k^2}, \tag{29}$$

$$D(k, z)\,k^2 = \Sigma(0, z) - \Sigma(k, z), \tag{30}$$

$$\Sigma(k,z) = \rho\tau(k,z) + \rho^2 \int \frac{\mathrm{d}\mathbf{k}'}{(2\pi)^3} [\tau^2(\mathbf{k}-\mathbf{k}',z) - 2\,\tau(\mathbf{k}-\mathbf{k}',z)\tau(k',z)]\bar{\mathcal{G}}(k',z), \quad (31)$$

$$\tau(k,z) = \int \mathrm{d}\mathbf{r} \exp(\mathrm{i}\mathbf{k}\cdot\mathbf{r})\, g(r)\, \frac{w(r)}{1 + 2\,w(r)[\mathcal{G}(z) - \tilde{\mathcal{G}}(r,z)]}. \quad (32)$$

$\rho = N/V$ is the number density of sites, and $\bar{\mathcal{G}}(k,z)$ and $\tilde{\mathcal{G}}(k,z)$ are defined by

$$\bar{\mathcal{G}}(k,z) = \rho^{-1}\,\mathcal{G}(z)\,\Sigma(k,z)\mathcal{G}(k,z) \quad (33)$$

$$\tilde{\mathcal{G}}(k,z) = \rho^{-1}\,\mathcal{G}^2(z)\,\Sigma^2(k,z)\,\mathcal{G}(k,z). \quad (34)$$

$\mathcal{G}(z) = \langle \mathbf{G}_{ii} \rangle$ is the diagonal propagator and is given by

$$\mathcal{G}(z) = \frac{1}{z + \Sigma(0,z)}. \quad (35)$$

It has been shown by Ganter (1996) that this set of equations can be viewed as the 'amorphous' analogue of the two-site lattice coherent-potential approximation (Odagaki and Lax 1981, Summerfield 1981, Webman 1981).

§ 3. Results

As seen from equation (29), the k-dependent averaged propagator has a diffusion pole with diffusivity $D(z) = D(k=0,z)$ given by

$$D(z) = -\frac{\varrho}{2}\frac{\partial^2}{\partial k^2}\tau(k,z)\bigg|_{k=0} - \frac{\varrho}{3}\,\mathcal{G}(z) \int \frac{\mathrm{d}\mathbf{k}}{(2\pi)^3} \frac{[(\partial/\partial k)\,\tau(k,z)]^2 \Sigma(k,z)}{z + D(k,z)\,k^2}. \quad (36)$$

If all contributions from closed loops are dropped, the self-consistent set of equations simplifies considerably. Closed-loop contributions are the integral in equation (31) and the off-diagonal contribution $\tilde{\mathcal{G}}(r,z)$ in equation (32). We obtain

$$\begin{aligned}\Sigma(k,z) &= \rho\,\tau(k,z) \\ &= \rho \int \mathrm{d}\mathbf{r} \exp(\mathrm{i}\mathbf{k}\cdot\mathbf{r})\, g(r)\, \frac{w(r)}{1 + 2w(r)\,\mathcal{G}(z)}. \end{aligned} \quad (37)$$

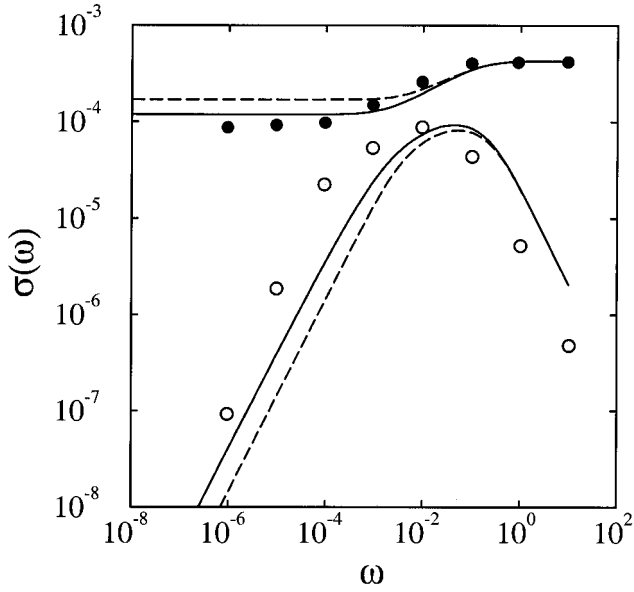
This is the version of EMA-I given by Gochanour *et al.* (1979).

The non-analytic behaviour of $D(\omega)$, which is present owing to the inclusion of the closed-loop terms is most clearly displayed in the low-frequency asymptotics of the so-called loss function $\mathrm{Re}\,[D(\omega) - D(0)]/\omega$. This contribution can be estimated as (Ganter 1996)

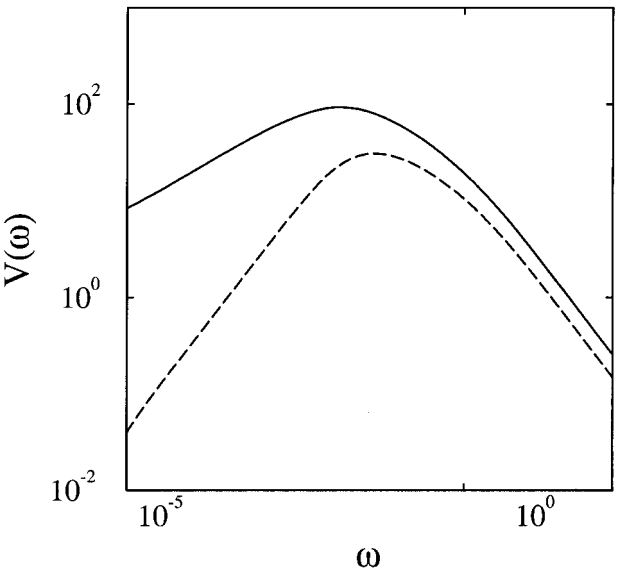
$$V(\omega) \approx \frac{1}{2^{1/2} \times 3\pi\rho} \frac{\omega^{1/2}}{[D(0)]^{3/2}}. \quad (38)$$

The structure of the integrals in the closed-loop terms shows that, in dimensions different from three, the non-analyticity takes the form $V(\omega) \propto \omega^{d/2-1}$.

To make sure that the incorporation of the closed-loop terms does not affect the good agreement of the EMA with the exact solution of the master equation we tested the results of the present theory against a simulation of an r-hopping system with hopping rates $w(r) = (\alpha r)^{3/2} \exp(-2\alpha r)$. The results for the normalized conductivity $\sigma(\omega) = (\rho/\alpha)D(\omega)$ are displayed in figures 1–3 (for different densities). We show also the loss function, in which the low-frequency non-analytic behaviour is displayed. In

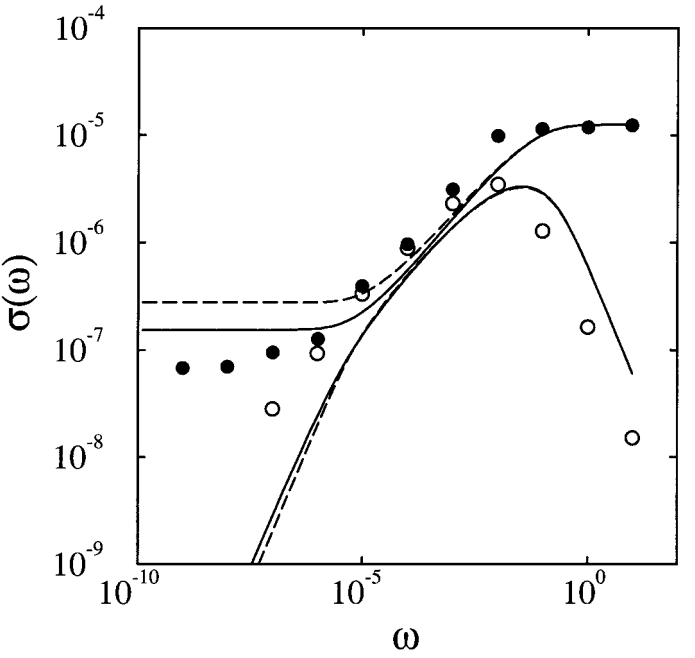


(*a*)

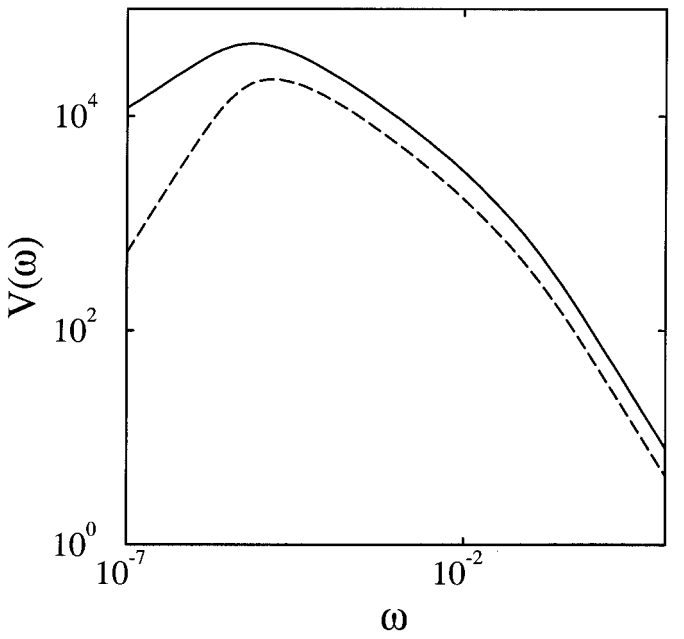


(*b*)

Figure 1. (*a*) Real part (●) and imaginary part (○) of the normalized conductivity $\sigma(\omega)$ of an r-hopping network with hopping rates $w(r) = (\alpha r)^{3/2} \exp(-2\alpha r)$ simulated by McInnes *et al.* (1980), compared with the result of the full EMA (equations (29)–(36)) (——) and that of the EMA-I (equation (37)) (· · · · · ·). (*b*) Loss function $V(\omega) = \mathrm{Re}\,[\sigma(\omega) - \sigma(0)]/\omega$ for both theories (same symbols as in (*a*)) with density $\rho = (\alpha/5)^3$.

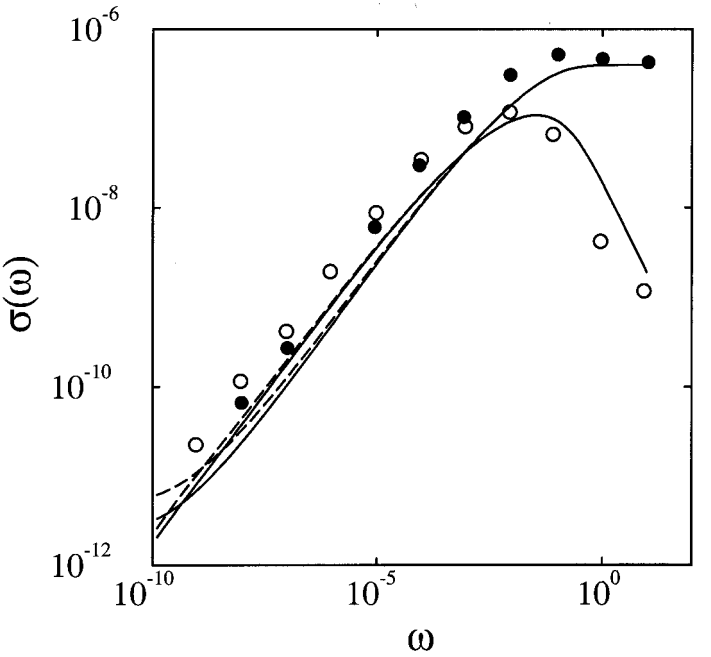


(*a*)

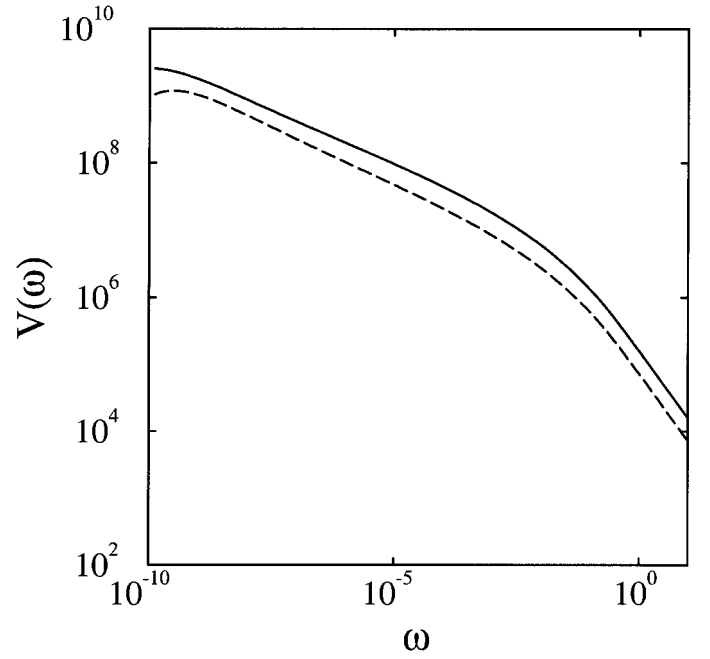


(*b*)

Figure 2. Same as in figure 1 with density $\rho = (\alpha/9)^3$.



(*a*)



(*b*)

Figure 3. Same as in figure 1 with density $\rho = (\alpha/16)^3$.

fact, the $V(\omega) \propto \omega^{1/2}$ frequency dependence has been observed experimentally (Long 1981) for amorphous silicon.

In conclusion we have obtained now an EMA for structurally disordered systems which compares well with computer simulations and has the correct non-analytic low-frequency asymptotics.

ACKNOWLEDGEMENT

Dedicated to M. Pollak on the occasion of his seventy-fifth birthday.

APPENDIX A
VANISHING OF $\langle \mathbf{G}^{[\xi]}(z)\rangle_{[\xi]} - \langle \mathbf{G}^{[\ldots,\xi]}(z)\rangle_{[\xi]}$

Let us consider the expression

$$\langle \mathbf{G}^{[\xi]}(z)\rangle_{[\xi]} - \langle \mathbf{G}^{[\ldots,\xi]}(z)\rangle_{[\xi]}, \tag{A 1}$$

with ... as usual specifying a given set of removed bonds.

The crucial expression is the following:

$$\langle \mathbf{G}^{[\xi]}(z)\rangle_{[\xi]} - \langle \mathbf{G}^{[\ldots,\xi]}(z)\rangle_{[\xi]} = \sum_{\zeta\in\{\cdots\}} \langle \mathbf{G}^{[\ldots,\xi]}(z)\,\mathbf{H}(\zeta)\,\mathbf{G}^{[\xi]}(z)\rangle_{[\xi]} \tag{A 2}$$

$$= \sum_{\zeta\in\{\cdots\}} \int \prod_{p=3}^{N} \mathrm{d}\mathbf{r}_p\, p(\mathbf{r}_1,\ldots,\mathbf{r}_N)\mathbf{G}^{[\ldots,\xi]}(z)\,\mathbf{H}(\zeta)\mathbf{G}^{[\xi]}(z). \tag{A 3}$$

Since at least one site in the bond ζ must be different from the sites which make up the bond ξ the integral over $\mathbf{r}_3$ is effectively confined to a volume $v \propto r_{\mathrm{eff}}^3$, limited by the finite range of $\mathbf{H}(\xi)$.

Therefore the integral must vanish as V^{-1} in the thermodynamic limit.

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