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## Generalization of the Effective-Medium Approximation for Hopping Transport in Amorphous Materials

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We present a new version of the effective-medium approximation (EMA) for hopping in structurally disordered systems such as amorphous semiconductors or impurity-bands of crystalline semiconductors. In contrast to 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.

The effective-medium approximation in the version suitable for structurally disordered systems (EMA-I) [1 to 5] has been proven to successfully describe dc and ac hopping conductivity phenomena in amorphous semiconductors and impurity bands of crystalline semiconductors. It has, however, been pointed out [6] that this EMA 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  $t \rightarrow \infty$  [7, 8]. 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  $D(\omega) - D(0) \propto \omega^{d/2}$  [7, 8]. 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 [4] paths with closed loops have been neglected. However, as pointed out in Ref. [7] the long-time algebraic tails are just produced by recurrent walks, i.e. “closed loops”. In Ref. [6] an attempt was made to repair the deficiency of the EMA-I, but this was done in a rather ad-hoc and unsatisfactory way.

In the EMA version we are going to present in the present communication we have combined the elimination of repeated indices (renormalized perturbation expansion, [9]), applied to pairs of sites, with the path summation technique of Wertheim, Logan and Winn [10, 11]. With the help of the former the EMA is obtained as a decoupling approximation [4], and the latter takes care of closed paths in a systematic way. Because of lack of space the derivation [12] can only be outlined.

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  $i$ ,

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

We treat here only the symmetric  $r$ -hopping case, i.e.  $w_{ij} = w_{ji} = w(r_{ij})$ , where  $r_{ij}$  is the distance between the sites  $i$  and  $j$ . The Laplace transform of  $G_{ij}$  is the  $ij$  matrix element of the resolvent  $G(z) = [z1 - H]^{-1}$ ,  $z = i\omega + 0$ . The “Hamiltonian”  $H$  is decomposed into sparse submatrices (“hopping matrices”)  $H = \sum_{\xi} H(\xi)$  associated with a single pair  $\xi := (ij)$  of sites and the sum runs over all such pairs. The hopping matrices are defined as follows:

$$H_{kl}(ij) := \begin{cases} -w(r_{ij}) & : (kl = ii \text{ or } jj) \\ w(r_{ij}) & : (kl = ij \text{ or } ji) \\ 0 & : \text{else.} \end{cases} \quad (2)$$

We denote sites (bonds) in *italic* (greek) letters. We now define by  $G^{[\xi_1, \dots, \xi_\ell]}$  the resolvent of the Hamiltonian in which the bonds  $\xi_1, \dots, \xi_\ell$  are missing. We further define renormalized hopping matrices by

$$S(\xi, z) := H(\xi) [1 - G^{[\xi]}(z) H(\xi)]^{-1} \quad (3)$$

and similar ones, where the bonds  $\xi_1, \dots, \xi_\ell$  are missing,

$$S^{[\dots]}(\xi, z) := H(\xi) [1 - G^{[\dots, \xi]}(z) H(\xi)]^{-1}. \quad (4)$$

Then the renormalized perturbation expansion of the resolvent takes the form [9, 12]

$$G(z) = \frac{1}{z} 1 + \sum_{m=1}^{\infty} \sum_{\substack{\xi_1, \dots, \xi_m \\ \xi_i \neq \xi_j}} \frac{1}{z^{m+1}} S(\xi_1, z) \dots S^{[\xi_1, \dots, \xi_{m-1}]}(\xi_m, z). \quad (5)$$

We are now interested in the configuration-averaged  $k$ -dependent propagator  $\mathcal{G}(k, z)$

$$\mathcal{G}(k, z) := \left\langle \frac{1}{N} \sum_{ij} e^{i\mathbf{k}\mathbf{r}_{ij}} G_{ij}(z) \right\rangle. \quad (6)$$

In order to evaluate the configuration average in an approximate way we define self-energy matrices

$$\tau(ij, z) := H(ij) [1 - \langle G^{[ij]}(z) \rangle_{[ij]} H(ij)]^{-1}, \quad (7)$$

which constitute our effective medium. In (7)  $\langle \dots \rangle_{[\xi]}$  denotes a restricted average with the bond  $\xi$  not to be averaged over. It is shown in Ref. [12] that the averaged propagator can be written approximately as

$$\begin{aligned} \mathcal{G}(k, z) &= \frac{1}{z} + \frac{1}{N} \sum_{ij} \sum_{m=1}^{\infty} \frac{1}{z^{m+1}} \sum_{\substack{\xi_1, \dots, \xi_m \\ \xi_k \neq \xi_i}} \int \prod_{p=1}^N d\mathbf{r}_p p_N(\mathbf{r}_1, \dots, \mathbf{r}_N) e^{i\mathbf{k}\mathbf{r}_{ij}} [\tau(\xi_1) \dots \tau(\xi_m)]_{ij}. \end{aligned} \quad (8)$$

Here  $p_N(\mathbf{r}_1, \dots, \mathbf{r}_N)$  is the probability for the quenched configuration of  $N$  sites. The evaluation of this expression is performed in analogy to the path summation of Logan and Winn [11] with a generalized Ishida-Yonezawa closure [11 to 13], resulting in the following self-consistent set of equations:

$$\mathcal{G}(k, z) = \frac{1}{z + D(k, z) k^2}, \quad D(k, z) k^2 = \Sigma(0, z) - \Sigma(k, z), \quad (9)$$

$$\Sigma(k, z) = \varrho \tau(k, z) + \varrho^2 \int \frac{d\mathbf{k}'}{(2\pi)^3} [\tau^2(\mathbf{k} - \mathbf{k}', z) - 2\tau(\mathbf{k} - \mathbf{k}', z) \tau(k', z)] \bar{\mathcal{G}}(k', z), \quad (10)$$

$$\tau(k, z) = \int d\mathbf{r} e^{i\mathbf{k}\mathbf{r}} g(r) \frac{w(r)}{1 + 2w(r)(\mathcal{G}(z) - \tilde{\mathcal{G}}(r, z))}. \quad (11)$$

$\varrho$  is the number density of sites,  $g(r)$  is the pair distribution function, and  $\bar{\mathcal{G}}(k, z)$  and  $\tilde{\mathcal{G}}(k, z)$  are defined using the following notations:

$$\bar{\mathcal{G}}(k, z) = \varrho^{-1} \mathcal{G}(z) \Sigma(k, z) \mathcal{G}(k, z), \quad \tilde{\mathcal{G}}(k, z) = \varrho^{-1} \mathcal{G}^2(z) \Sigma^2(k, z) \mathcal{G}(k, z). \quad (12)$$

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

$$\mathcal{G}(z) = \frac{1}{z + \Sigma(0, z)}. \quad (13)$$

As seen from (9) the  $k$ -dependent averaged propagator has a diffusion pole with diffusivity  $D(z) = D(k=0, z)$  given by

$$D(z) = -\frac{\varrho}{2} \frac{\partial^2}{\partial k^2} \tau(k, z) \Big|_{k=0} - \frac{\varrho}{3} \mathcal{G}(z) \int \frac{d\mathbf{k}}{(2\pi)^3} \frac{\left[ \frac{\partial}{\partial k} \tau(k, z) \right]^2 \Sigma(k, z)}{z + D(k, z) k^2}. \quad (14)$$

If all contributions from closed loops are dropped, the self-consistent set of equations simplifies considerably. Closed-loop contributions are the integral in (10) and the off-diagonal contribution  $\tilde{\mathcal{G}}(r, z)$  (11). We obtain

$$\Sigma(k, z) = \varrho \tau(k, z) = \varrho \int d\mathbf{r} e^{i\mathbf{k}\mathbf{r}} g(r) \frac{w(r)}{1 + 2w(r)\mathcal{G}(z)}. \quad (15)$$

This is the version of the EMA-I of Gochanour et al. [1].

The non-analytic behaviour of  $D(\omega)$ , which is present due to the inclusion of the closed-loop terms is most clearly displayed in the low-frequency asymptotics of the so-

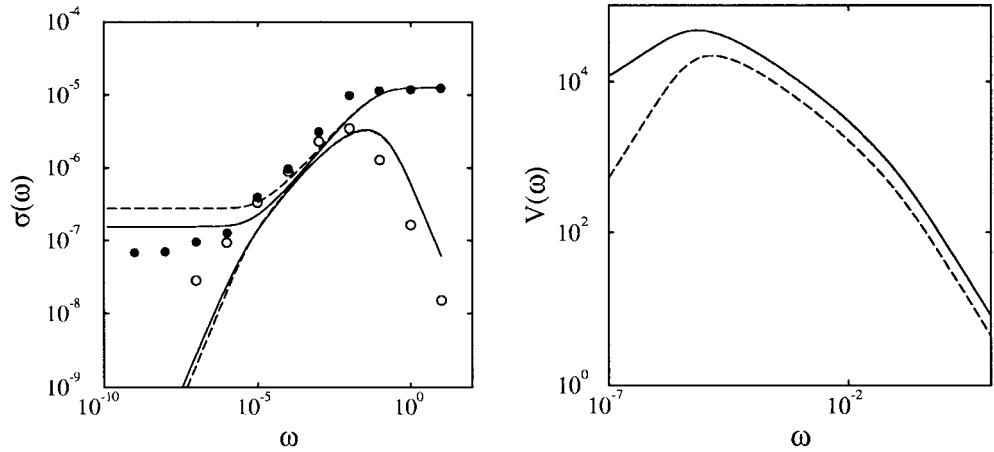


Fig. 1. Left: Real part (full circles) and imaginary part (open circles) of the normalized conductivity  $\sigma(\omega)$  of an  $r$ -hopping network simulated by McInnes et al. [14], compared with the result of the full EMA, Eqs. (9) through (14) (full line), and that of the EMA-I, Eq. (15) (dotted). Right: Loss function  $V(\omega) = \text{Re}\{\sigma(\omega) - \sigma(0)\}/\omega$  for both theories (same symbols as left-hand side) density:  $\varrho = [\alpha/9]^3$

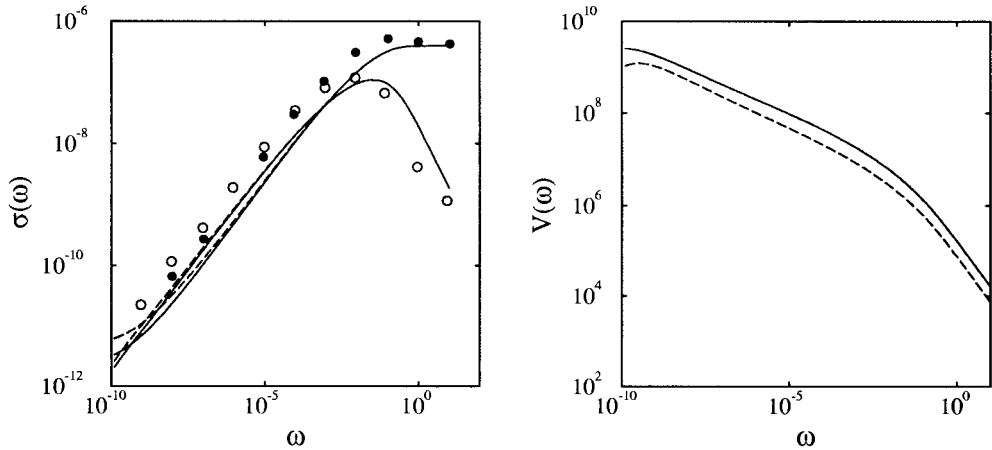


Fig. 2. Same as in Fig. 1 with density  $\varrho = [\alpha/16]^3$

called loss function  $[\text{Re}\{D(\omega) - D(0)\}]/\omega$ . This contribution can be estimated as [12]

$$V(\omega) \simeq \frac{1}{\sqrt{2}3\pi\varrho} \frac{\omega^{1/2}}{[D(0)]^{3/2}}. \quad (16)$$

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  $w(r) = [ar]^{3/2} e^{-2ar}$ . The results for the normalized conductivity  $\sigma(\omega) = (\varrho/\alpha) D(\omega)$  are displayed in Figs. 1 and 2 for two different densities. We also show the loss function, in which the low-frequency non-analytic behaviour is displayed.

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

## References

- [1] C. R. GOCHANOUR, H. C. ANDERSEN, and M. D. FAYER, *J. Chem. Phys.* **70**, 9 (1979).
- [2] B. MOVAGHAR, B. POHLMANN, and W. SCHIRMACHER, *Solid State Commun.* **34**, 451 (1980).
- [3] B. MOVAGHAR, B. POHLMANN, and W. SAUER, *phys. stat. sol. (b)* **97**, 533 (1980).
- [4] B. MOVAGHAR and W. SCHIRMACHER, *J. Phys. C* **14**, 859 (1981).
- [5] S. SUMMERFIELD and P. N. BUTCHER, *J. Phys. C* **15**, 7003 (1982).
- [6] W. SCHIRMACHER and M. WAGENER, *Phil. Mag. B* **65**, 607 (1992).
- [7] H. VAN BEIJEREN, *Rev. Mod. Phys.* **54**, 195 (1982).
- [8] J. HAUS, K. W. KEHR, and K. KITAHARA, *Z. Phys. B* **50**, 161 (1983).
- [9] E. N. ECONOMOU, *Green's Functions in Quantum Physics*, Springer-Verlag, Berlin/Heidelberg/New York 1990.
- [10] M. S. WERTHEIM, *Mol. Phys.* **25**, 211 (1973).
- [11] D. E. LOGAN and M. D. WINN, *J. Phys. C* **21**, 5773 (1988).
- [12] M. D. WINN and D. E. LOGAN, *J. Phys.: Condensed Matter* **1**, 1753 (1989).
- [13] C. GANTER, Doctoral dissertation, Techn. Univ. München 1996, unpublished.
- [14] Y. ISHIDA and F. YONEZAWA, *Progr. Theor. Phys.* **40**, 731 (1973).
- [15] J. A. MCINNES, P. N. BUTCHER, and J. D. CLARK, *Phil. Mag. B* **41**, 1 (1980).