Generalized effective-medium approximation for particle transport

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We present here a generalization of the effective-medium approximation to include weighted initial conditions. The implementation of the method is demonstrated on a random-trap model with bias where some exact results are available. Our results provide a closed form and accurate interpolation between the short- and long-time regimes. The connection of these results with continuous-time random-walk theory is discussed.

The problems encountered in studies of particle transport in disordered media have been met with intense research.[1-3] This labor has concentrated on master-equation descriptions of the particle's motion with the transition rates, appearing as phenomenological coefficients, being random variables. This level of modeling has resulted in some exact results on ensemble-averaged transport properties for specific models.[3] Complete solutions for most models, especially for dimensions greater than unity, are still lacking.

To make further progress, it is important to have methods which can provide accurate estimates of average transport properties for a wide variety of models. For the so-called random-bond models, the effective-medium approximation[4] (EMA) has been successful in providing results which, although not in general exact, are accurate enough to be indistinguishable from numerical simulations of the models[5-7] in parameter regimes where perturbation theory fails completely, and in higher dimensions where other methods give no results.[4] Especially worthy of mention are the long-time tails of the velocity autocorrelation functions which behave as $t^{-(2+D)/2}$ in D dimensions.

The EMA procedure, as it was formulated in the past, must fail for problems where the initial state is weighted in some manner by the random transition weights appearing in the master equation. For instance, most models have stationary states whose occupation probabilities are determined by the transition rates. One of these models, the random-trap model, has a velocity autocorrelation function which has no frequency dependence at all,[3,8] and the usual averaging procedure would predict a complicated time dependence. It is the goal of this Brief Report to describe a scheme to generalize the EMA to apply to problems of this type.

A general procedure for providing an equivalence between a master-equation description of transport in disordered systems and a continuous-time random-walk theory is given by the projection-operator method. Klafter and Silbey[9] have established this equivalence *for the case of uniform initial conditions.* The projection operator method is a well-known tool which we do not develop in detail; instead we refer to the review of Haake.[10] The projection operator $\underline{D}$ is taken as an ensemble average over the set of disordered transition rates, for any quantity A, $\underline{D}A = \langle A \rangle$ and the brackets denote the disorder average. The master equation is symbolically written as

$$dP/dt = LP \ , \qquad (1)$$

where P is the conditional probability and L a matrix containing the transition rates. The disorder-averaged master equation can be written in the form[9,10]

$$d/dt\langle P(t)\rangle = \underline{D}L\underline{D}\exp(Lt)P(0) + \underline{D}L(\underline{1}-\underline{D})\exp(Lt)P(0) \ . \qquad (2)$$

The second term on the right-hand side can be expressed differently by using the solution of a suitable differential equation,

$$(\underline{1}-\underline{D})\exp(Lt)P(0) = \exp[(\underline{1}-\underline{D})Lt](\underline{1}-\underline{D})P(0) + \int_0^t dt'\exp[(\underline{1}-\underline{D})L(t-t')](\underline{1}-\underline{D})L\underline{D}\exp(Lt')P(0) \ . \qquad (3)$$

The first term vanishes when the initial conditions are uniform, and it was omitted by Klafter and Silbey. The final result of the procedure can be written as

$$\frac{d}{dt}\langle P(t)\rangle = \langle L\rangle\langle P(t)\rangle + \int_0^t dt' K(t-t')\langle P(t')\rangle + I(t) \ . \qquad (4)$$

The kernel K is the disorder average

$$K(t-t') = \langle L\exp[(\underline{1}-\underline{D})L(t-t')](\underline{1}-\underline{D})L\rangle \ , \qquad (5)$$

and the inhomogeneous term is

$$I(t) = \langle L\exp[(\underline{1}-\underline{D})Lt](\underline{1}-\underline{D})P(0)\rangle \ . \qquad (6)$$

We note that the averaged master equation contains an inhomogeneous term when the initial conditions are nonuniform. The generalized master equation with an inhomogeneous term is equivalent to a continuous-time random walk where the first transition is treated differently.[11]

The projection-operator method illustrates the necessity of considering the initial conditions; in practice this method is difficult to implement. A practical method is the effective-medium theory, generalized to include nonuniform initial conditions. With the exception of the random-barrier models, all other models will have nonuniform occupation of the sites in the stationary state. The generalized effective-medium approximation is demonstrated here by considering the random-trap model defined by the following set of master equations:

$$\frac{dP(\mathbf{n},t)}{dt}=\sum_{\hat{\mathbf{e}}}[b_{-\hat{\mathbf{e}}}\Gamma_{\mathbf{n}+\hat{\mathbf{e}}}P(\mathbf{n}+\hat{\mathbf{e}},t)-b_{\hat{\mathbf{e}}}\Gamma_{\mathbf{n}}P(\mathbf{n},t)] , \quad (7)$$

where $P(\mathbf{n},t)$ is the conditional probability of being on site $\mathbf{n}$ at time t, given that the particle was on site $\mathbf{0}$ at time zero; the dependence on the latter site index and the initial time is suppressed. The variable $\hat{\mathbf{e}}$ denotes the nearest-neighbor unit vectors. Here we discuss only simple Bravais lattices (i.e., chain, square, cubic, etc.). The $\{\Gamma_{\mathbf{n}}\}$ are independently distributed random transition rates and $b_{\hat{\mathbf{e}}}$ is the set of positive parameters characterizing the application of an external dc field, i.e., a bias. For definiteness, the bias is taken along the positive x axis, so that $b_{\hat{\mathbf{e}}}=1$ for $\hat{\mathbf{e}}=\pm\hat{\mathbf{e}}_j$, $j\neq x$, $b_{\hat{\mathbf{e}}_x}=b$, and $b_{-\hat{\mathbf{e}}_x}=b^{-1}$.

The master equations, Eq. (7), possess a simple steady-state solution given by $\rho_{\mathbf{n}}=\Gamma_{\mathbf{n}}^{-1}/N\langle\Gamma^{-1}\rangle$, where $\langle\Gamma^{-1}\rangle$ is the average of the transition rates over a suitable distribution $W(\Gamma)$ and N is the number of sites on the lattice.

In the EMA, a cluster of sites, whose nearest-neighbor transition rates are random, is embedded into an averaged medium with an associated time-dependent transition rate $\Gamma(t)$. The set of approximate master equations is now

$$\frac{dP(\mathbf{n},t)}{dt}=\sum_{\hat{\mathbf{e}}}\left\{\int_0^t d\tau\Gamma(t-\tau)[(1-\Delta_{\mathbf{n}+\hat{\mathbf{e}}})b_{-\hat{\mathbf{e}}}P(\mathbf{n}+\hat{\mathbf{e}},\tau)-(1-\Delta_{\mathbf{n}})b_{\hat{\mathbf{e}}}P(n,\tau)] + \Delta_{\mathbf{n}+\hat{\mathbf{e}}}b_{-\hat{\mathbf{e}}}\Gamma_{\mathbf{n}+\hat{\mathbf{e}}}P(\mathbf{n}+\hat{\mathbf{e}},t)-\Delta_{\mathbf{n}}b_{\hat{\mathbf{e}}}\Gamma_{\mathbf{n}}P(\mathbf{n},t)\right\} , \quad (8)$$

where

$$\Delta_{\mathbf{n}}=\begin{cases}1 \text{ if } \mathbf{n} \text{ has random transition rates,}\\ 0 \text{ otherwise.}\end{cases} \quad (9)$$

These equations are partially diagonalized by Fourier transforming with respect to $\mathbf{n}$ and Laplace transforming with respect to the time variable. The initial conditions are taken as $P(\mathbf{n},0)$; the solution may be written as

$$\tilde{\mathcal{P}}(\mathbf{k},s)=\mathcal{P}(\mathbf{k},0)+\sum_{\mathbf{n}}e^{i\mathbf{k}\cdot\mathbf{n}}\sum_{\hat{\mathbf{e}}}[\Delta_{\mathbf{n}+\hat{\mathbf{e}}}b_{-\hat{\mathbf{e}}}\delta\Gamma_{\mathbf{n}+\hat{\mathbf{e}}}\tilde{P}(\mathbf{n}+\hat{\mathbf{e}},s)-\Delta_{\mathbf{n}}b_{\hat{\mathbf{e}}}\delta\Gamma_{\mathbf{n}}\tilde{P}(\mathbf{n},s)]\Big/\left\{s+\tilde{\Gamma}(s)\sum_{\hat{\mathbf{e}}}b_{\hat{\mathbf{e}}}(1-e^{i\mathbf{k}\cdot\hat{\mathbf{e}}})\right\} , \quad (10)$$

where $\delta\Gamma_{\mathbf{n}}=\Gamma_{\mathbf{n}}-\tilde{\Gamma}(s)$, and all Laplace-transformed quantities are denoted by a tilde, and physical quantities in Fourier-transformed variables are written with script letters.

The quantities $\tilde{P}(\mathbf{n},s)$ appearing on the right-hand side of Eq. (10) can be obtained by inverse Fourier transformation and diagonalization of a set of inhomogeneous linear equations (i.e., equations for the finite set of sites in the embedded cluster). These results can then be substituted back into Eq. (10).

The average transition rate $\Gamma(t)$ must be determined by a self-consistency condition and it should not depend on the particular initial conditions. We can infer this property on the average transition rate by examining the kernel, $K(t)$, given by the projection-operator method. As in previous formulations of the EMA, $\Gamma(t)$ is determined by using uniform initial conditions, namely $\mathcal{P}(\mathbf{k},0)=1$ in Eq. (10). The conditional probability, averaged over the random transition rates, is called the Green's function.

We implement this procedure using site $\mathbf{0}$ as the cluster with its jump rate Γ_0 to nearest-neighbor sites. In this sample only $\Delta_0=1$ and the explicit form of Eq. (10) is now

$$\tilde{\mathcal{P}}(\mathbf{k},s)=\frac{1+\delta\Gamma_0\tilde{P}(\mathbf{0},s)\sum_{\hat{\mathbf{e}}}(b_{-\hat{\mathbf{e}}}e^{-i\mathbf{k}\cdot\hat{\mathbf{e}}}-b_{\hat{\mathbf{e}}})}{s+\tilde{\Gamma}(s)\sum_{\hat{\mathbf{e}}}b_{\hat{\mathbf{e}}}(1-e^{i\mathbf{k}\cdot\hat{\mathbf{e}}})} . \quad (11)$$

The Green's functions of the averaged medium in D dimensions are denoted by

$$\tilde{G}(\mathbf{m},s)=\frac{1}{(2\pi)^D}\int\frac{d^Dk e^{-i\mathbf{k}\cdot\mathbf{m}}}{s+\tilde{\Gamma}(s)\sum_{\hat{\mathbf{e}}}b_{\hat{\mathbf{e}}}(1-e^{i\mathbf{k}\cdot\hat{\mathbf{e}}})} , \quad (12)$$

and they satisfy a homogeneous generalized master equation.[3] Inverse Fourier transforming Eq. (10) yields the solution

$$\tilde{P}(\mathbf{0},s)=\frac{\tilde{G}(\mathbf{0},s)}{1-\delta\Gamma_0\sum_{\hat{\mathbf{e}}}b_{\hat{\mathbf{e}}}[\tilde{G}(\hat{\mathbf{e}},s)-\tilde{G}(\mathbf{0},s)]} . \quad (13)$$

The sum in the denominator is equal to $[s\tilde{G}(\mathbf{0},s)-1]/\tilde{\Gamma}(s)$. As discussed, we determine $\tilde{\Gamma}(s)$ by averaging Eq. (13) over the distribution of transition rates, $W(\Gamma_0)$. The left-hand side of Eq. (13) is the *Green's function* $\tilde{G}(\mathbf{0},s)$.

The self-consistency condition in D dimensions is written as

$$0=\left\langle \frac{\delta\Gamma_0}{\tilde{\Gamma}(s)+\delta\Gamma_0(1-s\tilde{G}(\mathbf{0},s))} \right\rangle . \tag{14}$$

This resembles the self-consistency condition of the random-barrier model in a single-bond approximation for one dimension. The noteworthy difference here is that it is valid for all dimensions. In the case where the inverse moments of Γ_0 exist, i.e.,

$$\int d\Gamma_0 \frac{1}{\Gamma_0^n} W(\Gamma_0) < \infty , \tag{15}$$

$\tilde{\Gamma}(s)$ can be expanded as a function of s, the form of the asymptotic expansion $s \to 0$ depends on the dimension and the presence of bias. However, in all dimensions and including bias, the leading term of this expansion is

$$\tilde{\Gamma}(0)=\bar{\Gamma}=\left\langle \frac{1}{\Gamma_0} \right\rangle^{-1} . \tag{16}$$

This gives an exact result for the diffusion coefficient of the unbiased random-trap model in any dimension.[8]

The quantity we are interested in is the disorder-averaged conditional probability where initial conditions are used which reflect the occupation probabilities of the different sites in a stationary state. For example, a deep trap site will have a larger-than-average occupation probability in the stationary state, and thus contribute more strongly to the average. The conditional probability, when averaged over the disordered transition rates and including the corresponding weighting of the initial states, will be called a response function. Following Ref. 1, we will use the notation $F(\mathbf{n},t)$ for this double average in the response function.

The response functions for the steady-state conditions can be calculated by inserting $\mathcal{P}(\mathbf{k},0)=\bar{\Gamma}/\Gamma_0$ into Eq. (10) and averaging over Γ_0. For this model, only $P(\mathbf{0},0)$ depends on the cluster transition rate Γ_0. In more general models the stationary occupation probabilities are affected by the transition rates of the cluster; these probabilities must be included in the average as well. A calculation of the right-hand side of Eq. (10) gives the following average after some algebra:

$$\langle \delta\Gamma_0 \tilde{P}(\mathbf{0},s)\rangle = \frac{\bar{\Gamma}-\tilde{\Gamma}(s)}{s} . \tag{17}$$

The response functions for the averaged medium are

$$\tilde{\mathcal{F}}(\mathbf{k},s)=\frac{1-\sum_{\hat{\mathbf{e}}} b_{\hat{\mathbf{e}}}(1-e^{i\mathbf{k}\cdot\hat{\mathbf{e}}})[\bar{\Gamma}-\tilde{\Gamma}(s)]/s}{s+\tilde{\Gamma}(s)\sum_{\hat{\mathbf{e}}} b_{\hat{\mathbf{e}}}(1-e^{i\mathbf{k}\cdot\hat{\mathbf{e}}})} . \tag{18}$$

The expression in Eq. (18) can be put into the form of a generalized master equation, Eq. (4). The 1 in the right-hand side's numerator represents an effective initial condition for the response functions, $F(\mathbf{0},0)=1$. The second term in the numerator of Eq. (18) is an inhomogeneous contribution in the generalized master equation. This term is solely due to the inhomogeneous occupation of the initial sites in the disordered configurations. It is interesting to note that for the random-trap model studied here, the inhomogeneity has the form required by the single-state continuous-time random-walk theory treated as a renewal process.[11]

The usefulness of the results can best be shown by deriving moments of the response functions and comparing expressions of the moments with exact asymptotic results. The first moments of the displacement and mean-square displacement calculated from the response functions are

$$\langle \mathbf{x}\rangle(s)=\frac{\hat{\mathbf{e}}_x\bar{\Gamma}}{s^2}(b-b^{-1}) , \tag{19a}$$

and

$$\langle |\mathbf{x}|^2\rangle(s)=\frac{2(D-1)\bar{\Gamma}}{s^2}+\frac{\bar{\Gamma}}{s^2}(b+b^{-1}) + \frac{2\bar{\Gamma}\tilde{\Gamma}(s)(b-b^{-1})^2}{s^3} . \tag{19b}$$

The first moment is a linear function of time as required for stationary initial conditions and the expression is exact. The mean-square displacement may have a complicated time dependence due to the appearance of $\tilde{\Gamma}(s)$ in the last term. The application of bias removes the symmetry of the transition rates and the velocity autocorrelation function can be a smooth function of t for $t>0$. For times when the drift terms are dominant in Eq. (19b), i.e., $t>t_d=2(b+b^{-1})/(b-b^{-1})^2\bar{\Gamma}$, the velocity autocorrelation function has no algebraic long-time dependence. At intermediate times $1 \ll \bar{\Gamma}t \ll \bar{\Gamma}t_d$, the velocity autocorrelation function exhibits an algebraic time dependence proportional to $(b-b^{-1})^2 t^{-(4+D)/2}$, which is weaker than that for the random-barrier model. This result was found in $D=1$ by Lehr, Machta, and Nelkin;[12] they provided the first analysis of the biased model. Furthermore, we note that the amplitude of the velocity autocorrelation function is proportional to $(b-b^{-1})^2$ so that it would be more difficult to detect if the fields were small. Of course, the fields must be small to give a reasonably long-intermediate-time regime. Nevertheless, when the bias is removed, this expression is a linear function of time in all dimensions, which is also an exact result for this model.[8]

It is instructive to calculate the initial site occupation probability ($b=1$):

$$\tilde{F}(\mathbf{0},s)=\tilde{G}(\mathbf{0},s)+\frac{[s\tilde{G}(\mathbf{0},s)-1]}{\tilde{\Gamma}(s)}\frac{[\bar{\Gamma}-\tilde{\Gamma}(s)]}{s} . \tag{20}$$

The leading corrections to $\tilde{\Gamma}(s)$ are

$$\frac{\tilde{\Gamma}(s)}{\bar{\Gamma}}=\begin{cases} 1+\frac{\kappa_2}{2}\left[\frac{s}{\bar{\Gamma}}\right]^{1/2} , & D=1 , \\ 1+\frac{\kappa_2}{8\pi}\frac{s}{\bar{\Gamma}}\ln\left[\frac{s}{4\bar{\Gamma}}\right] , & D=2 , \\ 1+\tilde{G}(\mathbf{0},0)\left[\frac{s}{\bar{\Gamma}}\right]-\frac{\kappa_2}{4\pi}\left[\frac{s}{\bar{\Gamma}}\right]^{3/2} , & D=3 , \end{cases} \tag{21}$$

where $\kappa_2=\langle[(\bar{\Gamma}/\Gamma_0)-1]^2\rangle$.

The asymptotic time dependence for $F(\mathbf{0},t)$ is

$$F(\mathbf{0},t)=(1+\kappa_2)(4\pi\bar{\Gamma}t)^{-D/2} , \tag{22}$$

in agreement with the results of Denteneer and Ernst.[1] This result is a consequence of the disorder in the medium, and the traps create a larger-than-expected occupation of the initial site at long times. The result obtained here is a direct consequence of the previously discussed inhomogeneous term generated from the generalized EMA. It cannot be obtained by only using the Green's function. Our results are accurate for long and short times and provide a useful interpolation between these two regimes.

The generalized effective-medium approximation discussed here can be applied to a wide variety of problems. Two examples of applications of this method are the random-barrier model with bias and the model with randomly blocked sites. The method systematically includes an inhomogeneous term in the generalized master equations. Although for the random-trap model this term has the form of a renewal process, this need not be the case for more general models. In summary, the procedure outlined here should provide an accurate representation of more general models for transport in disordered media.

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