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Frequency- and temperature-dependent mobility of a charged carrier on a randomly interrupted strand

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A randomly interrupted strand model of a one-dimensional conductor is considered. An exact analytical expression is obtained for the temperature-dependent ac mobility for a finite segment drawn at random, taking into account the reflecting barriers at the two open ends. The real part of mobility shows a broad resonance as a function of both frequency and temperature, and vanishes quadratically in the dc limit. The frequency (temperature) maximum shifts to higher values for higher temperatures (frequencies).

Spectral diffusion of an excitation in general, and of a charged carrier in particular in disordered one-dimensional conductors has received much attention in recent times.[1] Most theoretical treatments of the problem envisage a model of the disordered one-dimensional conductor as a chain of sites, connected by bonds, between which the charged carrier or the excitation hops incoherently and instantaneously. Other lattices without closed loops such as Bethe lattices, have also been considered.[2] Only the nearest-neighbor elementary hops are usually considered. Disorder is then introduced by making the associated intersite jump rates spatially random but symmetrical. The mobility, or the diffusion constant, is then calculated by solving a classical master equation governing the random walk process. The low-frequency (or the long-time) behavior turns out to be mathematically related to the low-lying eigenvalue spectrum of the random coefficient matrix, and much of the analysis to date has gone into this limiting aspect of the problem.[1] Very recently, however, Odagaki and Lax[3] have obtained an exact expression for the ac conductivity valid over the entire frequency range for a nontrivial realization of randomness. The latter corresponds to the so-called bond percolation model (BPM) in which the jump rates have a nonzero probability of being strictly zero, thus partitioning the chain into a class of disjoint sets of connected sites (clusters) of random lengths. The ac mobility of a finite cluster is then averaged over the statistics of cluster size. A notable feature of their treatment is that it takes into account exactly the boundary conditions appropriate to the open termination of the finite clusters. The results are significantly different for the other simpler but approximate choice of the boundary condition, namely, the periodic-boundary condition.[3,4]

In this work, we present the exact solution of yet another nontrivial model of one-dimensional disordered conductors. This is the interrupted strand model (ISM) and may be regarded as a continuum analog of BPM, where the discrete sites and intersite bonds have been replaced by the segments of a continuous strand. In our mesocopic treatment, the stochastic motion of the carrier on a finite segment is described by the Langevin equation.[5] Again the point crucial to the treatment is that of the termination of the segment. The boundary conditions appropriate to the two fixed open ends of a segment are those of the reflecting barriers. These we have treated exactly by the method of images[6] generalized to the case of the bivariate (position and velocity) distribution function. In addition to the expected frequency dependence, the calculated mobility also shows a pronounced temperature dependence explicitly. It must be noted in this connection that in the earlier treatments,[1] any temperature dependence had to be introduced implicitly through the phenomenological jump rates which were taken to be thermally activated with an assumed distribution of barrier heights. In the following we give the main steps of our derivation.

Consider a segment of length L drawn at random. The stochastic motion of the carrier is described by the Langevin equation[5]

$$\ddot{X}=-\gamma\dot{X}+f(t)\ ,\quad 0<X<L \tag{1}$$

with the reflecting boundary condition at $x=0$ and $x=L$. The friction coefficient γ and the concomitant Gaussian white noise $f(t)$ are related as[5]

$$\langle f(t)f(t')\rangle_T=\frac{2k_BT\gamma}{m}\delta(t-t') \tag{2}$$

so as to be consistent with the condition of thermal equilibrium with the substratum (bath) at temperature T. Here k_B is the Boltzmann constant and m is the mass of the carrier.

An important quantity in terms of which the carrier mobility can readily be expressed is the conditional probability density $W_L(x,u,t;x_0,u_0,t_0)$ such that $W_L(x,u,t;x_0,u_0,t_0)dx\,du$ is the probability of finding the particle in the phase-space element $dx\,du$ at time t, given that it was at the phase point x_0,u_0 at time $t_0\le t$. Here $u\equiv(dx/dt)$ is the velocity. This will normally involve solving an initial-boundary-value problem. However $W_L(x,u,t;x_0,u_0,t_0)$, subject to the above reflecting boundary conditions can be readily expressed in terms of $W_\infty(x,u,t;x_0,u_0,t_0)$, the well-known fundamental solution[5] of the Fokker-Planck equation associated with Eq. (1) for an infinitely long segment. The expression for $W_\infty(x,u,t;x_0,u_0,t_0)$ is too long to be reproduced here, but we should note the normalization:

$$\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty}W_\infty(x,u,t;x_0,u_0,t_0)dx\,du=1\ . \tag{3}$$

Now, we invoke the method of images[6] and note that every time the position is "reflected" at a boundary, the velocity gets reversed in sign. We get at once

$$W_L(x,u,t;x_0,u_0,t_0)=\sum_{n=-\infty}^{+\infty}W_\infty(x+2nL,u,t;x_0,u_0,t_0)+\sum_{n=-\infty}^{+\infty}W_\infty(-x+2nL,-u,t;x_0,u_0,t_0)\ . \tag{4}$$

We see from Eqs. (3) and (4) that $W_L(x,u,t;x_0,u_0,t_0)$ is correctly normalized:

$$\int_0^L dx\int_{-\infty}^{+\infty}du\ W_L(x,u,t;x_0,u_0,t_0)=1\ . \tag{5}$$

We note here that the infinite series on the right-hand side of Eq. (4) is absolutely, and uniformly convergent and thus interchange of summation and integration (term-by-term integration of the series) is permitted.

Now, the frequency- (ω) and temperature- (T) dependent complex mobility $\mu_L(\omega,T)$ for the segment of length L is given by

$$\begin{aligned}\mu_L(\omega,T)&=\frac{q}{k_BT}\int_0^\infty\langle u(t)u(0)\rangle_Te^{i\omega t}dt\\&=\frac{q}{k_BT}\int_0^L dx\int_0^L dx_0\int_{-\infty}^{+\infty}du\int_{-\infty}^{+\infty}du_0uu_0W_L(x,u,t;x_0,u_0,0)f_L(x_0,u_0,0)\ ,\end{aligned} \tag{6}$$

where q is the charge on the carrier. Here $f_T(x_0,u_0,0)$ is the initial position-velocity distribution function which must be taken to be the equilibrium distribution function as the process is assumed to have been ongoing from the infinitely remote past. Indeed, one verifies from Eq. (4) and the explicit form[5] of $W_\infty(x,u,t;x_0,u_0,t_0)$ that

$$\begin{aligned}f_L(x_0,u_0,0)&=\lim_{t_0\to-\infty}W(x,u,0;x_0,u_0,t_0)\\&=\frac{1}{L}f_\infty(u_0)\ ,\end{aligned} \tag{7}$$

where $f_\infty(u_0)$ is the Maxwellian velocity distribution function at temperature T. Substituting from Eqs. (4) and (7) into Eq. (6) and using the known expression for $W_\infty(x,u,t;x_0,u_0,t_0)$ from Ref. 5, we get, after some reduction, for the real part of mobility

$$\frac{\mathrm{Re}\mu_L(\Omega,\Theta)}{\mu_\infty}=8\int_0^\infty dx \sum_{n=1,3,5,\ldots}\left[\frac{e^{-x}}{\pi^2 n^2}-\Theta(1-e^{-x})^2\right]e^{-n^2\pi^2\Theta(x-1+e^{-x})}\cos\Omega x \tag{8a}$$

$$=8\int_0^\infty dx \sum_{n=1,3,5,\ldots}\frac{\Omega^2}{\pi^4 n^4}\frac{1}{\Theta}e^{-\pi^2 n^2\Theta(x-1+e^{-x})}\cos\Omega x\ , \tag{8b}$$

where $\mu_\infty = q/m\gamma$, the classical dc mobility on an infinitely long segment. Here we have introduced dimensionless frequency $\Omega=\omega/\gamma$ and temperature $\Theta=k_{BT}/m\gamma^2L^2$. The physical mobility, of course, is obtained by averaging $\mu_L(\Omega,\Theta)$ over the probability distribution $P(L)$ of lengths L, i.e.,

$$\mathrm{Re}\mu(\Omega,\Theta)=\int_0^\infty \mathrm{Re}\mu_L(\Omega,\Theta)P(L)dL\ . \tag{9}$$

The continuum analog of the uncorrelated interruptions of BPM will be

$$P(L)=\frac{1}{L_0}e^{-L/L_0}\ , \tag{10}$$

where L_0 is the mean length. We note again that the series in Eq. (8) is uniformly convergent as a simple M test shows, and hence both the x integration as well as the L integration involved in averaging can be performed term by term. Thus the problem is reduced to quadrature.

The essential features of our exact solution are all contained in Eq. (8). These features remain unaltered by the averaging over L so long as $P(L)$ has finite support. First, from Eq. (8b), in the dc limit ($\Omega=\omega/\gamma\to 0$) the mobility vanishes quadratically with frequency as also found by Odagaki and Lax.[3] From Eq. (8a) the mobility is also seen to vanish in the high-frequency limit ($\Omega\to\infty$) as a direct consequence of the Riemann-Lebesgue theorem. Physically, this is expected since in this limit the inertia m of the particle dominates the motion and the system behaves inductively. This is not so in the case of the BPM where jumps are taken to be instantaneous and inertial effects are not incorporated. In point of fact, the treatments based on the master rate equation[1,3] go over to the diffusion equation in the continuum limit which shows no inertial effects. On the contrary, Eq. (1) describes an Ornstein-Uhlenbeck process[5] and has a ballistic regime dominated by inertia. In the limit $\Theta\to 0$ (i.e., $L\to\infty$, or $T\to 0$) for fixed Ω, it follows from Eq. (8a) that mobility tends to its well-known classical value

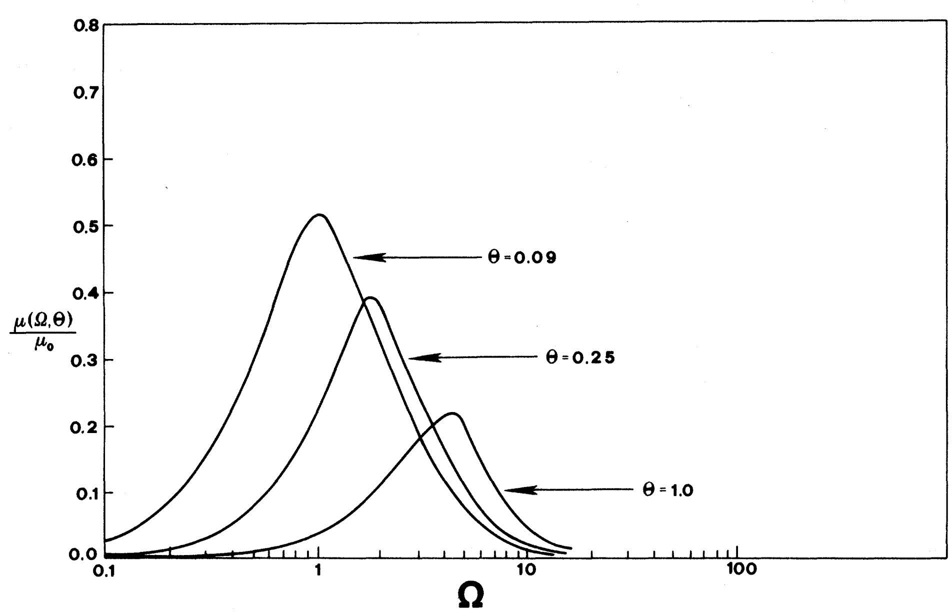

FIG. 1. Plot of real part of normalized mobility $\mathrm{Re}\mu_L(\Omega,\Theta)/\mu_\infty$ against normalized frequency $\Omega=\omega/\gamma$ for different values of normalized temperature $\Theta=k_BT/m\gamma^2L^2$.

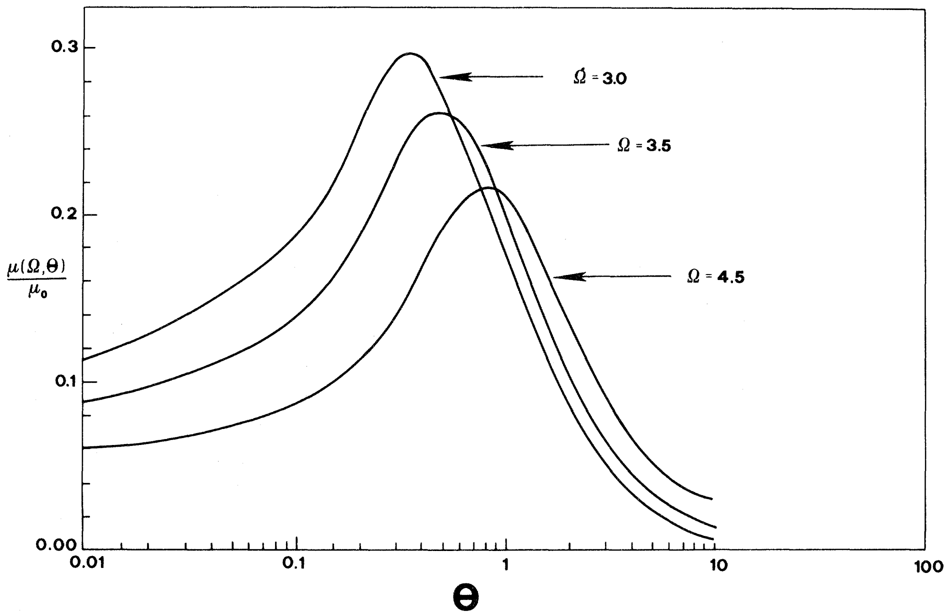

FIG. 2. Plot of real part of normalized mobility $\text{Re}\mu_L(\Omega,\Theta)/\mu_\infty$ against normalized temperature $\Theta = k_BT/m\gamma^2L^2$ for different values of normalized frequency $\Omega = \omega/\gamma$.

$$\text{Re}\mu_\infty(\omega) = \frac{q}{m\gamma}\left[\frac{1}{1+(\omega^2/\gamma^2)}\right] . \tag{11}$$

For this we have noted that $\sum_{n=1,3,5,\ldots} 1/n^2 = \pi^2/8$. This is understandable since in this limit the particle is unable to make an excursion to the boundary, and thus interrogate the boundary conditions, within the period $2\pi/\omega$ of the ac probe. In Figs. 1 and 2 we have plotted, respectively, the frequency and the temperature dependences of $\text{Re}\mu_L(\Omega,\Theta)$ obtained from Eq. (8b) numerically. The series is indeed very rapidly convergent. The plots show the frequency (temperatures) maxima that shift to higher frequencies (temperatures) for higher temperatures (frequencies). This feature is absent in the BPM referred to above. For $\Theta, \Omega >> 1$ it is possible to approximate the exponent $(x - 1 + e^{-x})$ occurring on the right-hand side of Eq. (8b) by $x^2/2$ and then integrate the series term by term. We get

$$\frac{\text{Re}\mu_L(\Omega,\Theta)}{\mu_\infty} \simeq \sum_{n=1,3,5,\ldots} \frac{2^{5/2}}{\pi^{9/2}}\frac{1}{n^5}\frac{\Omega^2}{\Theta^{3/2}} e^{-\Omega^2/2\Theta\pi^2 n^2} . \tag{12}$$

The series is again highly convergent and one may simply retain the first term which shows the frequency and temperature peaks as discussed above.

Finally, we expect the ISM, and hence our solution, to be relevant to such nondegenerate electronic or even superionic conductors where the mobility is unactivated and the carriers are delocalized, just as the BPM is relevant to the low-mobility systems with activated hopping conduction via localized sites.

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