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Anomalous transient-time dispersion in amorphous solids—A comment*

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The random-walk non-Markoffian equations of Scher and Montroll are subjected to two approximations: first, the continuum limit is taken and second, the diffusive term, as defined in the text, is neglected (this means that the carriers are drifted by the electric field). It is shown that this approach does preserve the dispersive character of the transport process and leads to, essentially, the same results as deduced by Scher and Montroll.

I. INTRODUCTION

In an important recent paper, Scher and Montroll[1] (SM) discussed the problem of transport of a space-charge packet in amorphous dielectrics under the action of an externally (high) applied field. Experimental current-time curves exhibit a long tail, indicative of dispersion of the carriers, and SM have proposed to explain this by electronic hopping through randomly distributed sites. This feature of the medium was incorporated in the general theory of the random walk by the use of a convenient hopping distribution function, with the result that the transport process is no longer Markoffian. SM undertook the (formidable) task of solving this new problem, approximating the theoretically derived hopping distribution function[2] by a mathematically more treatable function.

However, the computational work involved in this kind of calculation foresees the practical impossibility of treating other important experimental situations—for instance, the high-signal (space-charge limited) case. Therefore, it would be desirable to have a more simple theory, embracing the main features of the hopping process. The first step in this direction—already mentioned by Kenkre, Montroll, and Schlesinger[3]—is to take the continuum limit of the pertinent random-walk equations, for which more developed mathematical methods may be applied to find the solutions. The second one, more radical, is to disregard the diffusive component in these equations. The diffusive component we call that component of the transport equation allowing motion of the carriers in both directions. This amounts to saying that the hopping carrier is drifted by the electric field. The physical reason for this approximation is that in many experiments in "crystalline" materials (that is not displaying the long-tail current) the spreading of the packet seems to be unimportant. It should be stressed, however, that the proposed simplification does not eliminate the dispersive character of the hopping process and for this reason it helps us to understand more clearly its nature.

In this article the proposed steps will be undertaken and a calculation carried out to show that they also lead essentially to the same results as obtained by SM. However, small differences still remain but we think that future discussion about these points shall explain them (see Sec. V).

II. THEORY

We start with the generalized master equation as given by Kenkre, Montroll, and Schlesinger[3] assuming a cubic lattice of parameter a_0 (mean distance between hops) whose lattice points (cells) are characterized by the vector $\vec{l}$:

$$\frac{dP}{dt}=\int_0^t \phi(t-t')\sum_{\vec{l}'}[p(\vec{l}-\vec{l}')\tilde{P}(\vec{l}'t') - p(\vec{l}'-\vec{l})\tilde{P}(\vec{l},t')]\,dt' , \quad (1)$$

where $\tilde{P}(\vec{l},t)$ is the probability of finding a carrier in the cell $\vec{l}$ at the time t if at $t=0$ it was in the cell $\vec{l}_0$; $p(\vec{l})$ is the probability of hop from a given cell to one a vector $\vec{l}$ distant. $\phi(t)$ is related to the hopping function $\psi(t)$ through a sequence of operations; calling $\psi^*(u)$ and $\phi^*(u)$ the Laplace transform of $\psi(t)$ and $\phi(t)$,

$$\phi^*(u)=\mu\psi^*(u)/[1-\psi^*(u)] . \quad (2)$$

Defining $p(\vec{l})$ as different from zero for

$$p(0,\pm 1,0)=p(0,0\pm 1)=\tfrac{1}{6} ,$$

$$p(1,0,0)=\tfrac{1}{6}+bE, \quad p(-1,0,0)=\tfrac{1}{6}-bE.$$

E being the electric field in the x direction and b a constant and proceeding to the continuum limit, the following equation is readily obtained:

$$\frac{\partial\rho(x,t)}{\partial t}=\int_0^t\phi(t-t')\left(\frac{a_0^2}{6}\ \frac{\partial^2\rho(x,t')}{\partial x^2}-2a_0b\times\frac{\partial\rho(x,t')E(x',t')}{\partial x}\right)dt' . \quad (3)$$

In going from Eqs. (1) to (3) we have assumed planar symmetry and changed from $\tilde{P}$ to ρ defined as the charge density. Equation (3) is a generalized Fokker-Plank equation and has already been considered in connection with Brownian motion.[4,5] For a Markoffian transport process $[\psi(t)=\lambda e^{-\lambda t}]$, Eq. (3) gives the usual Fokker-Plank equation. The term in the integrand depending on $\partial^2\rho/\partial x^2$ is what we call the diffusive term and will tentatively be supposed small as compared with the electric drift one. We see that this approximation does, in principle, preserve the dispersive nature of the process through the memory represented by the term $\phi(t-t')$. Besides this, we will take the small signal case (that is, the carriers are drifted by the external applied field), writing

$$\frac{\partial\rho}{\partial t}=-2a_0bE\int_0^t\phi(t-t')\frac{\partial\rho}{\partial x}\,dt' . \quad (4)$$

Suppose now that $\phi(t-t')$ is equal to $\lambda\delta(t-t')$. If carriers were created at $t=0$ at x_0, we know that the solution of Eq. (4) is proportional to $\delta(x-x_0-2\lambda a_0bEt)$. Let us take the Laplace transform of Eq. (4) with q_0 a constant:

$$u\rho^*(u,x)-q_0\delta(x-x_0)=-2a_0bE\phi^*(u)\frac{\partial\rho^*}{\partial x} . \quad (5)$$

Since now the integration of this equation does not depend on u we may take its solution as the Laplace transform of the δ function [corresponding to $\phi(t-t')=\lambda\delta(t-t')$] substituting λ for $\phi^*(u)$. So we have the solution of Eq. (5) as

$$\rho^*(u,x)=q_0\exp^{[-u(x-x_0)/2a_0bE\phi^*(u)]}/2a_0bE\phi^*(u) . \quad (6)$$

Therefore, the problem is reduced to finding the inverse Laplace transform of $\phi^*(u,x)$. We will use for the hopping function $\psi(t)$, the same as Scher and Montroll,[1]

$$\psi(t)=4W_m\exp(W_mt)i^2\operatorname{erfc}(W_mt)^{1/2} ,$$

leading to[2]

$$\phi^*(u)=W_m[1+2(W_m/u)^{1/2}]^{-1} . \quad (7)$$

Now inserting Eq. (7) in Eq. (6), the inverse Laplace transform may be found, and we get

$$\rho(x,t)=A\,\frac{\exp^{[-(y-y_0)^2/\tau-y+y_0]}}{(\tau-y+y_0)^{1/2}}\left(\frac{y-y_0}{2(\tau-y+y_0)}+1\right) , \quad (8)$$

with

$$\tau=W_mt,\quad y=x/2a_0bE,\quad A=q_0/(\pi)^{1/2}a_0bE . \quad (9)$$

The asymptotic ($\tau\gg1$) value of $\rho(x,t)$ is given by

$$\rho(x,t)=A\exp^{[-(y-y_0)^2/\tau]}/(\tau)^{1/2} . \quad (8')$$

III. EXTERNAL CURRENT

From Eq. (8) we may derive the external current density j in the following way. The external current j is

$$j=i+\epsilon\frac{\partial E}{\partial t} ,$$

where i is the conduction (or material) current density and the second term is the displacement current density. Integrating from 0 to l, the sample thickness, we get, for constant applied voltage,

$$jl=\int_0^l i\,dx .$$

After partial integration and use of the continuity equation

$$jl=xi\,|_0^l+\int_0^l x\frac{\partial\rho}{\partial t}dx$$
$$=li(l,t)+\frac{d}{dt}\int_0^l x\rho\,dx . \quad (10)$$

In our case the carriers are drifted by the electric field and therefore there is no conduction current leaving the sample at $x=0$. Hence we may say that

$$i(l,t)=-\frac{d}{dt}\int_0^l\rho\,dx . \quad (11)$$

Inserting Eq. (11) in Eq. (10) we finally get

$$j=-\frac{d}{dt}\int_0^l\rho dx+\frac{1}{l}\frac{d}{dt}\int_0^l x\rho dx. \quad (12)$$

IV. RESULTS

We will apply Eq. (8) for $y_0=0$, that is for all the carriers at $y=0$ at $\tau=0$. The sample thickness l, in reduced units, is $y_l=l/2a_0bE$ and will be supposed to be much larger than 1.

We could follow, using Eq. (8), the initial behavior of $\rho(x,t)$ but we expect the results of the continuum approximation to be meaningful for $\tau\gg1$. Therefore, we instead take the asymptotic formula, Eq. (8′), and obtain

$$\int_0^l\rho\,dx=W_mq_0\ \operatorname{erf}\ [y_l/(\tau)^{1/2}] , \quad (13)$$

$$\int_0^l\rho x\,dx=\frac{W_mq_0(\tau)^{1/2}}{y_l(\pi)^{1/2}}\,(1-e^{-y_l^2/\tau}) . \quad (14)$$

From Eqs. (13) and (14) we calculate the external current density j:

$$j=[W_m q_0/2y_l(\pi\tau)^{1/2}](1-e^{-y_l^2/\tau}) , \quad (15)$$

which agrees with the SM result [Ref. 1, Eq. (44)]. We have following asymptotic behavior for Eq. (15):

$$j=W_m q_0/2y_l(\pi)^{1/2}\tau^{1/2} , \quad \tau << y_l^2 ,$$

$$j=W_m q_0 y_l/2(\pi)^{1/2}\tau^{3/2}, \quad \tau >> y_l^2 .$$

However, in a $\log_{10} j$ vs $\log_{10}\tau$ plot, the transition between these two behaviors is more or less sharp.

V. FINAL REMARKS

The aim of this comment was to show that the main results obtained by SM are preserved even when radical simplifications are made. The two treatments are formally different. In the present case free boundary conditions are used, while SM assume periodic boundary conditions in addition to the presence of absorbing planes at $x=0$ and $x=l$. Therefore, the same expressions used here appear to be different from those of SM. This applies in particular to the expressions for the current density, given by Eq. (16) of Ref. 1, and Eq. (12) of this paper, and that for the first moment [Ref. 1, Eq. (42), and Eq. (12) of this paper]. Nevertheless, the final results are in agreement.

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