Nonlinear field dependence of the mobility of a charge subjected to a superposition of dichotomous stochastic potentials

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(Received 3 February 1998)

A general prescription is presented to address a large variety of forms of the nonlinear dependence of the static charge mobility on the applied electric field. The system consists of a classical charge subjected to an arbitrarily strong steady state electric field and to a stochastic potential consisting of a linear superposition of an unlimited number of dichotomous potentials in one-dimensional space. It is shown that the nonlinear mobility can be calculated for arbitrary forms of the density function of the individual dichotomous components of the stochastic potential. Specific cases of physical interest are analyzed. One of them provides a curious possibility for an explanation of the universally observed square root field dependence of the logarithm of the mobility of photoinjected charge carriers in molecularly doped polymers. [S1063-651X(98)09006-0]

PACS number(s): 05.20.−y, 05.60.+w

I. INTRODUCTION AND MODEL

In this paper we present a prescription to address the electric field dependence of the nonlinear static mobility of a charged particle moving under the action of a class of simple stochastic potentials having certain characteristics described below. While the contexts in which the calculation and the insights gained from our prescription can be useful are numerous, including the general theory of nonlinear response [1,2] and specific applications to phenomena in ceramic materials [3], our analysis was motivated by the near-universal behavior of the mobility of photoinduced charge carriers [4–8] observed in molecularly doped polymers. Indeed, it will be seen below that a curious correspondence exists between the so-called Poole-Frenkel behavior of the mobility [4–7] and the field dependence relevant to a simple particular case of our general result.

In the rest of this section we describe the model. In successive sections, we present our generalized prescription, which takes one from the density function of the stochastic superposition of potentials to the field dependence of the mobility; describe the Poole-Frenkel behavior, and its generalization, as simple particular cases emerging from our prescription; present other special cases of our formula; and provide a discussion.

We have shown recently [2,8] that a highly useful feature of the Kubo formalism [9], i.e., that the response of a system to an external stimulus may be expressed completely in terms of system correlation functions calculated in the absence of the external stimulus, can be retained for certain response situations *even in the fully nonlinear regime* which lies outside the validity of the Kubo formalism. One of our previously published results [2,7] describes the nonlinear mobility $\mu(E)$ of a classical particle of charge q and mass m moving in an infinite one-dimensional space spanned by the coordinate x, and subjected to a random stationary potential $U(x)$ and an external electric field E. The mobility, defined as the ratio of the velocity of the charge to the field E, is given simply in terms of the system correlation function

$$c(y)=\overline{\exp[U(x+y)/kT]\exp[-U(x)/kT]}. \qquad (1.1)$$

The overbar in Eq. (1.1) represents an ensemble average over realizations of the stochastic potential $U(x)$. The assumed stationarity of the stochastic process underlying the potential ensures that the correlation function depends only on the difference y in the coordinate values. If the Laplace transform of the correlation function $c(y)$, with $\varepsilon=qE/kT$ as the Laplace variable, is denoted by $\tilde{c}(\varepsilon)$, the mobility is given explicitly by

$$\mu(E)=\frac{\mu_\infty}{\varepsilon\tilde{c}(\varepsilon)}=\frac{\tau kT}{mE\int_0^\infty dy\ e^{-yqE/kT}c(y)}. \qquad (1.2)$$

Here μ_∞ equals the saturation value of the mobility, i.e., the value $q\tau/m$ (where τ is the relaxation time) which it would have in the absence of the potential.

Equation (1.2) can be obtained by Brownian motion analysis in the limit of high damping: the Langevin equation for the charge velocity is converted into a Smoluchowski equation which is then solved in the steady state [2]. This provides a straightforward way of calculating the *nonlinear* mobility from a given stochastic potential U through an evaluation of the correlation function $c(y)$, and thus has the advantages of a Kubo-like formalism despite the fact that the analysis is not restricted to the linear regime.

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In the present paper we consider stochastic potentials U which are constructed from arbitrary linear superpositions of independent dichotomous potentials:

$$U(x)=U_0+\sum_{i=1}^{N}\Delta_i(-1)^{n_i(x,0)}. \tag{1.3}$$

Here U_0 is a constant, Δ_i is the amplitude of the ith dichotomous component of the potential, and $n_i(x_2,x_1)$ is a random function characteristic of that component. This random function counts the number of jumps (whose distribution is assumed to be uniform) that the particular component of the potential makes between the value Δ_i and $-\Delta_i$ in the interval between x_1 and x_2. The ith component has the correlation length l_i, by which is meant that the ensemble average $\overline{n_i(x_2,x_1)}$ equals $|x_2-x_1|/l_i$. It is well known that the individual components of the potential are exponentially correlated [10]. We have shown elsewhere [2] that, for this case, the correlation function $c(y)$ appearing in Eq. (1.1) has a product form arising from the individual components:

$$c(y)=\prod_{i=1}^{N}c_i(y), \tag{1.4}$$

$$c_i(y)=1+(1-e^{-2y/l_i})\sinh^2(\Delta_i/kT). \tag{1.5}$$

This expression for the correlation function will serve as our starting point in the present paper.

II. GENERAL RESULTS

If we denote the amplitudes of the individual potential components by $\Delta_i/\sqrt{N}$ rather than Δ_i, and introduce the quantity $r_i=2/l_i$, we can rewrite Eq. (1.5) as

$$c_i(y)=1+(1-e^{-yr_i})\sinh^2(\Delta_i/\sqrt{N}kT). \tag{2.1}$$

We consider the limit of large N. Specifically, we assume that, for all i, $\Delta_i/\sqrt{N}kT\ll 1$. This allows the replacement of the hyperbolic sine in Eq. (2.1) by its argument:

$$c(y)=\prod_{i=1}^{N}\left[1+\frac{1}{N}\left(\frac{\Delta_i}{kT}\right)^2(1-e^{-yr_i})\right]. \tag{2.2}$$

Taking the logarithm of both sides of this equation and using the fact that N is large so that $\ln[1+(1/N)(\Delta_i/kT)^2(1-e^{-yr_i})]$ can be replaced by $(1/N)(\Delta_i/kT)^2(1-e^{-yr_i})$, we obtain

$$c(y)=\exp\left[\frac{1}{N}\sum_{i=1}^{N}\left(\frac{\Delta_i}{kT}\right)^2(1-e^{-yr_i})\right]. \tag{2.3}$$

We now assume that in the assembly of the dichotomous potentials which constitutes $U(x)$, all correlation lengths are possible, i.e., that we have a continuous variable r which extends from 0 to ∞. We take the limit of infinite N and introduce the quantity $\lambda(r)$ through

$$\lambda(r)=\lim_{N\to\infty}(1/N)\sum_{i=1}^{N}\Delta_i^2\delta(r-r_i). \tag{2.4}$$

Clearly, $\lambda(r)$ is a weighted density of states differing from the normalized density $g(r)=\lim_{N\to\infty}(1/N)\Sigma_{i=1}^{N}\delta(r-r_i)$ merely by a factor associated with the amplitude of the individual dichotomous components. We will call $\lambda(r)$ the density function. The correlation function can now be expressed as an integral over r space:

$$c(y)=\exp\left[(1/kT)^2\int_0^\infty dr\ \lambda(r)(1-e^{-yr})\right]. \tag{2.5}$$

Equation (2.5) can be expressed in terms of the density function $\lambda(r)$ through its Laplace transform with y as a Laplace variable,

$$c(y)=\exp\{(1/kT)^2[\tilde{\lambda}(0)-\tilde{\lambda}(y)]\}. \tag{2.6}$$

The tilde denotes the Laplace transform.

Equation (2.6) or (2.5), along with Eq. (2.4), is one of the primary results of the present paper. It gives a recipe for the calculation of the correlation function $c(y)$ from the density function $\lambda(r)$ which characterizes the system (i.e., the potential superposition). The correlation function, thus evaluated, can then be substituted into expression (1.2) to obtain the field dependence of the nonlinear mobility. The fully explicit prescription leading from the potential density function $\lambda(r)$ to the mobility $\mu(E)$ is

$$\mu(E)=\left(\frac{\tau kT}{mE}\right)\left[\int_0^\infty dy\ e^{-yqE/kT}e^{(1/kT)^2\int_0^\infty dr\ \lambda(r)(1-e^{-yr})}\right]^{-1}. \tag{2.7}$$

Although the model considered here is restricted to potentials which are linear combinations of dichotomous parts with amplitudes small enough to allow the limit $\Delta_i/\sqrt{N}kT\ll 1$, it has considerable generality conferred on it by the arbitrary nature of the density function $\lambda(r)$. The large N limit makes the potential a superposition of Ornstein-Uhlenbeck processes. The calculational task in obtaining the mobility for an arbitrary linear superposition of an infinite number of Ornstein-Uhlenbeck processes consists of the evaluation of two essentially successive direct Laplace transforms: that of the density function $\lambda(r)$ with the distance y as the Laplace variable to obtain the correlation function $c(y)$, and that of $c(y)$ with qE/kT as the Laplace variable to obtain the mobility $\mu(E)$.

III. POOLE-FRENKEL FIELD DEPENDENCE FROM AN EXPONENTIAL DENSITY FUNCTION

A near-universal observation in molecularly doped polymers is the Poole-Frenkel behavior of the mobility of photoinjected charge carriers: the logarithm of the mobility is found to be proportional to the square root of the electric field over a very large range of fields. The observation has received a great deal of attention for decades, and has been recently explained [7] satisfactorily on the basis of dipole disorder ideas developed over the years by a number of workers [4–6]. A remarkable consequence of our general formula (2.7) is that Poole-Frenkel dependence is found to be a consequence of simply assuming the density function of the stochastic potential superposition to have what is perhaps

the simplest decaying form: an exponential. No dipolar disorder is involved.

We take the density function to decay in r space with decay constant α, and to have the integral σ^2 over all r space:

$$\lambda(r)=\sigma^2\alpha\ \exp(-\alpha r). \tag{3.1}$$

We see from Eq. (2.4) that σ^2, which is given by

$$\sigma^2=\lim_{N\to\infty}(1/N)\sum_{i=1}^{N}\Delta_i^2=\lim_{N\to\infty}\overline{[U(y)-U_0]^2}, \tag{3.2}$$

is the mean squared amplitude of the dichotomous components of the stochastic potential, and is thus a measure of the disorder in the system. The correlation function is obtained from Eq. (2.6):

$$c(y)=\exp\left[\left(\frac{\sigma}{kT}\right)^2\left(\frac{y}{y+\alpha}\right)\right]. \tag{3.3}$$

By switching the integration variable to $y+\alpha$, the Laplace transform of Eq. (3.3) is written as

$$\tilde{c}(\varepsilon)=e^{\alpha\varepsilon}e^{(\sigma/kT)^2}\left[\left(\frac{2\sigma}{kT}\sqrt{\frac{\alpha}{\varepsilon}}\right)K_1\left(\frac{2\sigma}{kT}\sqrt{\alpha\varepsilon}\right)-\int_0^{\alpha}e^{-\varepsilon y-(\alpha/y)(\sigma/kT)^2}dy\right], \tag{3.4}$$

where K_1 is the modified Hankel function. The exact expression for the mobility is obtained from the reciprocal of Eq. (3.4) via the recipe in Eq. (1.2). Reduction of that exact expression to the Poole-Frenkel form is straightforward when, in Eq. (3.4), σ/kT is sufficiently large. In this limit, the integral may be neglected relative to the term proportional to K_1, because the former decreases exponentially with $(\sigma/kT)^2$, whereas the latter decreases exponentially only with σ/kT. Furthermore, when σ/kT is large, K_1 can be replaced by its asymptotic form to yield

$$\tilde{c}(\varepsilon)=\sqrt{\frac{\pi\sigma\sqrt{\alpha}}{kT\varepsilon\sqrt{\varepsilon}}}\exp\left[\left(\frac{\sigma}{kT}-\sqrt{\alpha\varepsilon}\right)^2\right]. \tag{3.5}$$

When the second term in the exponential is small with respect to the first, the mobility reduces to the Poole-Frenkel form

$$\mu(E)=\left(\frac{q\tau}{m}\right)\left[\frac{(kT)^3}{\pi^2\sigma^2qE\alpha}\right]^{1/4}e^{-(\sigma/kT)^2}e^{2\sigma\sqrt{qE\alpha/(kT)^3}}, \tag{3.6}$$

with the validity condition

$$\left(\frac{\sigma}{kT}\right)^2\gg\frac{qE\alpha}{kT}\gg\left(\frac{kT}{\sigma}\right)^2. \tag{3.7}$$

Both the mobility expression (3.6) and the validity condition (3.7) are seen to be identical to those derived in Ref. [7] on the basis of dipolar disorder arguments, if one makes a single correspondence. This correspondence is $\alpha\leftrightarrow a$, where a is the radius of the sphere introduced in Ref. [7] to take into account the finite size of a transport site, and the consequent inability of molecular dipoles to approach arbitrarily close to a charge located at such a site. While only the first of the two inequalities comprising Eq. (3.7) was mentioned explicitly in Ref. [7], the second inequality is also necessary for a derivation of Poole-Frenkel behavior from dipolar disorder, as is evident from the discussion in Ref. [7].

It is interesting to contrast this derivation of the Poole-Frenkel dependence of the mobility on the field with that given in Ref. [7]. Our present derivation makes no mention of dipoles, and does not have a cutoff in the correlation function arising from the finite size of a transport site (molecule). The cutoff length of Ref. [7] corresponds to the exponent of the r dependence of the density function here. Also, our derivation arrives directly at an expression for the full correlation function $c(y)$ rather than at the correlation of the quantity $U(y)-U(0)$, which was first obtained in Ref. [7] and then followed by an approximate Gaussian prescription to obtain $c(y)$. As a consequence of the central limit theorem, the stochastic potential in our present case is a Gaussian process, arising as it does from a superposition of independent Ornstein-Uhlenbeck pieces. Therefore, one obtains the exact result even if one follows the Gaussian approximation used in Ref. [7].

The present analysis does not provide a real ''explanation'' of the Poole-Frenkel behavior, since it does not ascribe the behavior to any physical source—only to an assumed dependence of $\lambda(r)$. Nevertheless, the analysis is intriguing, particularly in the light of the fact that Poole-Frenkel behavior appears to have been observed in some systems in which dipolar disorder is absent [11]. An understanding of what physical sources can give rise to an exponential dependence of the density function (or any related dependence capable of yielding the Poole-Frenkel behavior through an asymptotic analysis) should provide further insights into the Poole-Frenkel phenomenon.

IV. ARBITRARY POWERS IN THE FIELD DEPENDENCE FROM BIASED EXPONENTIAL DENSITY

If the correlation lengths of the dichotomous components of the potentials are concentrated at a nonzero value, the density vanishing at both very small and very large correlation lengths, the density function may be represented by a biased exponential

$$\lambda(r)=\sigma^2\frac{\alpha^{n+1}}{\Gamma(n+1)}r^ne^{-\alpha r}, \tag{4.1}$$

where Γ is the (complete) gamma function. The density function peaks at $r=n/\alpha$. It yields the correlation function

$$c(y)=\exp\left[\left(\frac{\sigma}{kT}\right)^2\left(1-\frac{\alpha^{n+1}}{(\alpha+y)^{n+1}}\right)\right]. \tag{4.2}$$

Exact evaluation of the Laplace transform of $c(y)$ in Eq. (4.2) does not appear possible. An asymptotic evaluation may be carried out as follows.

The Laplace transform of $c(y)$ is e^AI with $A=(\sigma/kT)^2$, and the integral I given by

$$I=\int_0^\infty dy\ \exp\left(-\varepsilon y-\frac{A\alpha^{n+1}}{(y+\alpha)^{n+1}}\right). \tag{4.3}$$

The value y_m at which the argument of the exponent is an extremum satisfies

$$y_m=\left[\frac{A\alpha^{n+1}}{\varepsilon}(n+1)\right]^{1/(n+2)}-\alpha. \tag{4.4}$$

If our interest is in the limit

$$y_m \gg \alpha, \tag{4.5}$$

Eq. (4.4) reduces to

$$y_m=\left[\frac{A\alpha^{n+1}}{\varepsilon}(n+1)\right]^{1/(n+2)}. \tag{4.6}$$

The definition $t=y/y_m$ reduces the integral to

$$I=y_m\int_0^\infty dt\ \exp\left(-\varepsilon y_m\left[t+\frac{A\alpha^{n+1}/\varepsilon y_m^{n+2}}{(t+(\alpha/y_m))^{n+1}}\right]\right)$$

$$=y_m\int_0^\infty dt\ \exp[-\varepsilon y_m g(t)], \tag{4.7}$$

where the last equality defines $g(t)$. We see from Eq. (4.6) and from condition (4.5) that

$$g(t)=t+\frac{1/(n+1)}{t^{n+1}}. \tag{4.8}$$

At the peak, $t=t_m=1$, $g(t_m)=(n+2)/(n+1)$, and $[d^2g(t)/dt^2]_{t_m}=n+2$. The replacement of $g(t)$ by $g(0)+(1/2)[d^2g(t)/dt^2]_{t_m}(t-1)^2$, along with the extension of the limits of the integral to $\pm\infty$, is possible if

$$\varepsilon y_m\left(1+\frac{n}{2}\right)\gg 1. \tag{4.9}$$

Under this condition,

$$I=y_m\sqrt{\frac{2\pi}{\varepsilon y_m(n+2)}}\exp\left(-\varepsilon y_m\frac{n+2}{n+1}\right). \tag{4.10}$$

With the definition $B(E,T)=[(qE\alpha/kT)^{(n+1)}(\sigma/kT)^2(n+1)]^{1/(n+2)}$, the mobility is then given by

$$\mu(\varepsilon)=\frac{q\tau}{m}\sqrt{\frac{n+2}{2\pi B(E,T)}}e^{-(\sigma/kT)^2+B(E,T)[(n+2)/(n+1)]}, \tag{4.11}$$

under the validity condition

$$\left(\frac{\sigma}{kT}\right)^2(n+1)\gg\frac{qEa}{kT}\gg\left[\left(\frac{kT}{\sigma}\right)^2\frac{1}{(n+1)\left(1+\frac{n}{2}\right)^{n+2}}\right]^{1/(n+1)}. \tag{4.12}$$

This validity condition is a combination of Eqs. (4.5) and (4.9).

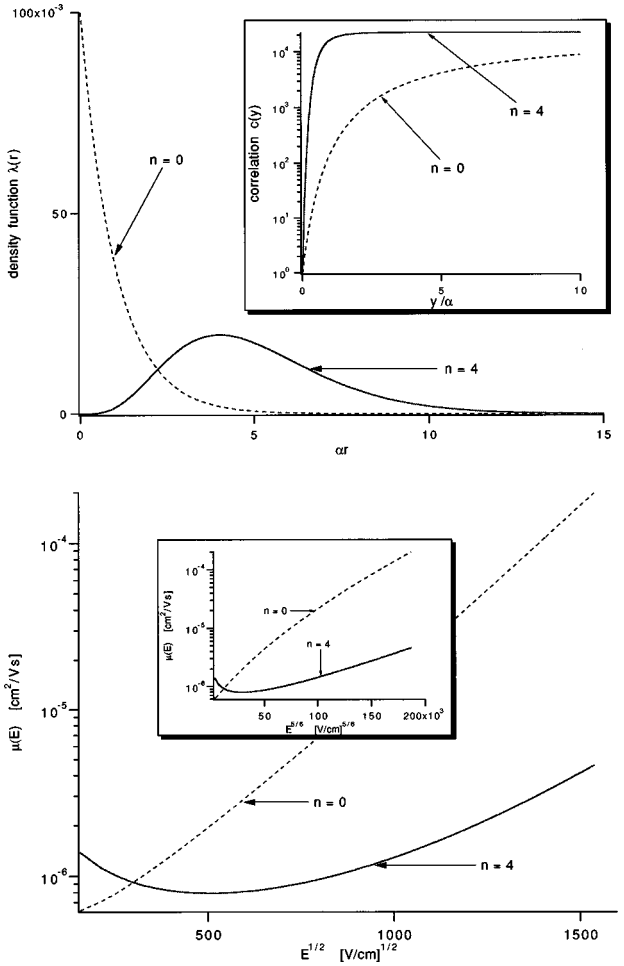

FIG. 1. The density function $\lambda(r)$, the correlation function $c(y)$, and the mobility $\mu(E)$ for two values of the exponent n (0 and 4) in the biased exponential case of Sec. IV. The case $n=0$ exhibits Poole-Frenkel behavior identical to that arising from charge-dipole interactions as in Ref. [7]. The case $n=4$ exhibits behavior identical to that arising from induced charge-dipole interactions also mentioned in Ref. [7]. The disorder parameter σ/kT is taken to be $\sqrt{10}$. The saturation mobility $q\tau/m$ is taken to be 7×10^{-3} cm^2/V s.

We see that, for the case $n=0$, for which the biased exponential density (4.1) reduces to the exponential density (3.1), the correlation function (4.2) reduces to Eq. (3.3), and the generalized dependence (4.11) of the mobility reduces to Poole-Frenkel behavior (3.6). The validity condition (4.12) reduces to Eq. (3.7) for this case.

The primary result we have obtained here is that, if the observed dependence of the logarithm of the mobility on the electric field E is of the power law form $\mu\sim\exp(E^c)$ with exponent c, we can ''explain'' this behavior by assuming the density function of the dichotomous components of the stochastic potential to be the biased exponential with $n=(2c-1)/(1-c)$:

$$\lambda(r)=\sigma^2\frac{\alpha^{c/(1-c)}}{\Gamma\left(\frac{c}{1-c}\right)}r^{(2c-1)/(1-c)}e^{-\alpha r}. \tag{4.13}$$

This density function starts at zero r with zero value, peaks at $r=(2c-1)/(1-c)\alpha$, and then decays to zero. In Fig. 1,

we display (a) density functions and correlation functions, and (b) the asymptotic dependence of the mobility on the field, for two different values of the exponent: $n=0$ and 4. The first represents Poole-Frenkel behavior, which arises from charge interactions with permanent dipoles as in Ref. [7]. The second exhibits behavior (an exponent of $\frac{5}{6}$ instead of $\frac{1}{2}$ in the field dependence) identical to that arising from induced charge-dipole interactions also mentioned in Ref. [7]. The values of the disorder parameter σ/kT and the saturation mobility are taken to be $\sqrt{10}$ and 7×10^{-3} $\mathrm{cm}^2/\mathrm{V\,s}$, respectively, and the square root of the field is displayed up to 1600 $(\mathrm{V/cm})^{1/2}$. These values are in keeping with experiments on molecularly doped polymers [4]. In Fig. 1(a), the density functions are displayed in the main part, and the correlation functions in the inset. The asymptotic dependence of the field is displayed versus $E^{1/2}$ in the main part of Fig. 1(b) (showing asymptotic linearity for $n=0$ but not for $n=4$), and versus $E^{5/6}$ in the inset (showing asymptotic linearity for $n=4$ but not for $n=0$). While the curvature for low fields arises from the prefactors becoming important in that range, the asymptotic formulas are not accurate in that region.

V. CORRELATION FUNCTION FOR OTHER SPECIFIC CASES

We present some additional specific cases of the correlation function $c(y)$. They are exact consequences of our formula (2.5).

A. Single correlation length

If the potential has a single correlation length l, the density function is given by

$$\lambda(r)=\sigma^2\delta(r-2/l), \tag{5.1}$$

and the correlation function is

$$c(y)=\exp[(\sigma/kT)^2(1-e^{-2y/l})]. \tag{5.2}$$

The mobility can be calculated explicitly for this case [2] and may be expressed in terms of the confluent hypergeometric function ${}_1F_1$ or, equivalently, in terms of the incomplete gamma function $\gamma(\alpha,x)=\int_0^x e^{-t}t^{\alpha-1}dt$:

$$\mu(E)=\left(\frac{2kT\tau}{Elm}\right)\left(\frac{\sigma}{kT}\right)^{qEl/kT}\left[\frac{e^{-(\sigma/kT)^2}}{\gamma(qEl/2kT,(\sigma/kT)^2)}\right]. \tag{5.3}$$

B. Biased Gaussian density function

Another density function which peaks at a noninfinite value of the correlation length is given by a biased Gaussian

$$\lambda(r)=\left(\frac{\sigma}{b}\right)^2 r\,\exp\left[-\frac{1}{2}\left(\frac{r}{b}\right)^2\right] \tag{5.4}$$

which peaks at $r=b$. The correlation function is given in this case by

$$c(y)=\exp\left[\left(\frac{\sigma}{kT}\right)^2\sqrt{\frac{\pi}{2}}bye^{(1/2)(yb)^2}\mathrm{erfc}\left(\frac{yb}{\sqrt{2}}\right)\right]. \tag{5.5}$$

C. Rectangular pulse density function

The mean value and dispersion of the correlation lengths inherent in the stochastic potential discussed in the above two cases are dependent on each other. In order to study the effect on the mobility of the independent variation of these two quantities, one might consider the density function to be given by a pulse starting at the value s of the variable r, and spanning a width w. Thus

$$\lambda(r)=\begin{cases}0 & \text{for } 0<r<s\\ \sigma^2/w & \text{for } s<r<s+w\\ 0 & \text{for } s+w<r.\end{cases} \tag{5.6}$$

The correlation function is given by

$$c(y)=\exp\left\{\left(\frac{\sigma}{kT}\right)^2\left[1-e^{-ys}\left(\frac{1-e^{-yw}}{yw}\right)\right]\right\}, \tag{5.7}$$

and reduces to the single-correlation-length case above in the limit $w\to0$ with $s=2/l$.

VI. APPROXIMATION TECHNIQUES

Exact analytical evaluation of the expression for the mobility is possible only in a few cases such as for the single-correlation-length density function. In the other cases it is straightforward to employ numerical procedures since, unlike the inverse Laplace transform which we do not require in our prescription, the direct transform does not suffer from numerical problems such as instability. Asymptotic methods of the analytical kind we have used in Sec. IV, which are based on the Laplace method [12], may also be developed in a general way as follows.

With the definition $h(y)=(1/kT)^2\tilde{\lambda}(y)$, we rewrite the general expression (2.6) as

$$c(y)=e^{h(0)-h(y)}, \tag{6.1}$$

and note that $\tilde{c}(\varepsilon)$, the Laplace transform of $c(y)$, equals $e^{h(0)}I$, with the integral I given by

$$I=\int_0^\infty dy\,\exp[-\varepsilon y-h(y)]. \tag{6.2}$$

The exponent in the integrand has an extremum at $y=y_m$, given by [13]

$$\left[\frac{dh(y)}{dy}\right]_{y=y_m}=-\varepsilon. \tag{6.3}$$

The definition $t=y/y_m$ leads to

$$I = y_m \int_0^\infty dt\ \exp\left(-\varepsilon y_m \left[t + \frac{h(t y_m)}{\varepsilon y_m} \right] \right)$$

$$= y_m \int_0^\infty dt\ \exp[-\varepsilon y_m g(t)], \tag{6.4}$$

where the last equality defines $g(t)$. We note that at the peak of $g(t)$, the respective values of t, $g(t)$, and $d^2g(t)/dt^2$ are 1, $1+h(y_m)/\varepsilon y_m$, and $y_m h''(y_m)/\varepsilon$. The asymptotic evaluation of the mobility is based on the replacement of $g(t)$ by its Taylor expansion around $t=1$ to second order. If this approximation is valid, one may extend the limits of integration to infinity, evaluate the Gaussian integral, and obtain

$$I = e^{-[\varepsilon y_m + h(y_m)]} \sqrt{\frac{2\pi}{h''(y_m)}}. \tag{6.5}$$

The first of the conditions of validity of this approximation is that the width of $\varepsilon y_m g(t)$ at the peak is small with respect to the location of the peak from the origin, i.e.,

$$y_m h''(y_m) \gg 1. \tag{6.6}$$

While Eq. (6.6) suffices for the asymptotic analysis to be valid in a number of instances (see, e.g., the discussion in Sec. IV), it is generally necessary to ensure that terms of order higher than the second make a negligible integration to the integral. This imposes the additional requirement

$$\tfrac{1}{2} \gg \left| \varepsilon y_m \delta + h[y_m(1+\delta)] - h(y_m) - \tfrac{1}{2} \right|, \tag{6.7}$$

where $1/\delta^2$ equals $y_m h''(y_m)$. The asymptotic mobility under the conditions (6.6) and (6.7), where the solution of Eq. (6.3) gives y_m, is

$$\mu = \frac{q\tau}{m\varepsilon} \sqrt{\frac{h''(y_m)}{2\pi}} \exp[\varepsilon y_m + h(y_m) - h(0)]. \tag{6.8}$$

We illustrate the application of the general asymptotic formula (6.8) by comparing the dependence it yields, with the exact expression in the single-correlation-length case (5.2). The analytical solution for the mobility is Eq. (5.3). The application of the asymptotic considerations developed above leads to

$$y_m = \frac{l}{2} \ln\left(\frac{2\sigma^2}{qElkT} \right) \tag{6.9}$$

for the peak value y_m. The asymptotic mobility expression is then

$$\mu = \frac{q\tau}{m} \left(\frac{kT}{\pi qEl} \right)^{1/2} \left(\frac{2\sigma^2}{qElkT} \right)^{qEl/2kT} e^{-(\sigma/kT)^2 + qEl/2kT}, \tag{6.10}$$

the validity conditions being

$$\frac{2kT}{qEl \ln^2\left(\dfrac{qElkT}{2\sigma^2} \right)} \ll 1 \ll \frac{qEl}{2kT} < \left(\frac{\sigma}{kT} \right)^2. \tag{6.11}$$

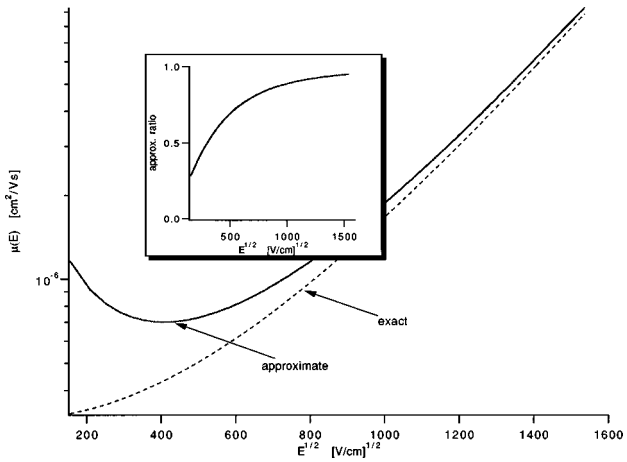

FIG. 2. Comparison of the exact [Eq. (5.3)] and asymptotic [Eq. (6.10)] expressions for the mobility $\mu(E)$ in the single correlation length case of Sec. V. The values of the disorder parameter and the saturation mobility are as in Fig. 1. The inset shows the field variation of the ratio of the exact mobility as given by Eq. (5.3) to the approximate value given by the asymptotic expression (6.10).

The first and second inequalities in Eq. (6.11) arise, respectively, from Eqs. (6.6) and (6.7), and the third from the fact that y_m in Eq. (6.9) is positive. This case provides an example where condition (6.7) has to be considered separately, as it is not implied by condition (6.6).

In Fig. 2 we compare the exact and the approximate mobilities, i.e., Eqs. (5.3) and (6.10), respectively [14]. As in Fig. 1, the disorder parameter σ/kT is taken to have the value $\sqrt{10}$, and the saturation mobility is 7×10^{-3} cm^2/V s in keeping with the experimental data on molecularly doped polymers [4]. The inset shows the field variation of the ratio of the exact mobility as given by Eq. (5.3) to the approximate value given by the asymptotic expression (6.10). As expected, the latter is valid only at high values of the field. To appreciate the manner in which Eq. (5.3) reduces to Eq. (6.10) in the high field limit provided the disorder parameter is large, notice that, for large enough values of σ/kT, the incomplete gamma function in Eq. (5.3) may be replaced by the complete gamma function. With $x = qEl/2kT$, one identifies in the denominator of Eq. (5.3) the expression $x\Gamma(x)$ as $\Gamma(x)$. For large enough x, the Stirling approximation allows its replacement by $e^{-x}x^x\sqrt{2\pi x}$. The asymptotic expression (6.10) follows.

The general form of the correlation function suggests that, in addition to the asymptotic analysis given above, one can use nonstandard procedures such as the one employed in polaron calculations by Silbey and Munn [15]. In the context of the present calculation, the approximation procedure would express the correlation function as

$$c(y) \approx 1 + [1/\tilde{\lambda}(0)][\tilde{\lambda}(0) - \tilde{\lambda}(y)][e^{(1/kT)^2\tilde{\lambda}(0)} - 1]$$

$$= e^{(1/kT)^2\tilde{\lambda}(0)} - [1/\tilde{\lambda}(0)][e^{(1/kT)^2\tilde{\lambda}(0)} - 1]\tilde{\lambda}(y). \tag{6.12}$$

The procedure is patterned after the well known relaxation time approximation in transport theory [16]. Its essence is that the evolution of $c(y)$ from its initial value to its infinite y value, written as

$$c(y)=c(\infty)+[c(0)-c(\infty)]g(y), \tag{6.13}$$

is approximated by choosing the function $g(y)$ to be the function $\tilde{\lambda}(y)/\tilde{\lambda}(0)$ rather than an exponential which is used in the relaxation time approximation [16]. Explicitly,

$$c(y)\approx c(\infty)+[c(0)-c(\infty)][1-\log_{c(\infty)}c(y)], \tag{6.14}$$

where $\log_{c(\infty)}c(y)$ is the logarithm of $c(y)$ to the base $c(\infty)$. The mobility formula reduces to the approximate version

$$\mu=\frac{\mu_\infty}{e^{(1/kT)^2\int_0^\infty dr\,\lambda(r)}-\left[\dfrac{e^{(1/kT)^2\int_0^\infty dr\,\lambda(r)}-1}{\int_0^\infty dr\,\lambda(r)}\right]\epsilon\int_0^\infty dr\,\dfrac{\lambda(r)}{\epsilon+r}}, \tag{6.15}$$

which can also be written as

$$\mu=\frac{\mu_0}{1-\dfrac{\mu_\infty-\mu_0}{\mu_0}\left[\dfrac{1}{\int_0^\infty dr\,\lambda(r)}\right]\epsilon\int_0^\infty dr\,\dfrac{\lambda(r)}{\epsilon+r}}, \tag{6.16}$$

with the linear response (Kubo) mobility $\mu_0=\lim_{E\to 0}\mu(E)$ being given by

$$\mu_0=\mu_\infty e^{-(1/kT)^2\int_0^\infty dr\,\lambda(r)}=\frac{\tau q}{m}e^{-(1/kT)^2\int_0^\infty dr\,\lambda(r)}. \tag{6.17}$$

Applied to the case of the rectangular pulse, this yields

$$\mu(E)=\frac{\mu_0}{1-[1-e^{-(\sigma/kT)^2}]\dfrac{qE/kT}{w}\ln\left[1+\dfrac{w}{s+(qE/kT)}\right]}, \tag{6.18}$$

with $\varepsilon=qE/kT$. Note that the numerator in Eq. (6.18) is the linear response (Kubo) mobility of the system. A study of Eq. (6.18) shows that reducing the width w and reducing the starting value s of the pulse on the r axis both make the mobility rise steeper as the field is increased.

VII. CONCLUDING REMARKS

Starting from our previous recipe for transforming the correlation function (1.1) into an expression for the mobility (1.2), in this paper we have presented a generalized prescription to calculate the field dependence from given characteristics of linear superpositions of dichotomous potentials. The prescription provides a useful technique for generating Gaussian random potentials having desired correlations, by adding the contributions of easily generated dichotomous potentials, the correlation lengths of individual dichotomous components being chosen from an appropriate density function. We have given analytic examples of this practical technique in the present paper. It is also possible to use it numerically to generate random potentials of desired characteristics.

Among the useful results that have emerged from our analysis is a derivation of the Poole-Frenkel behavior of the mobility observed in molecularly doped polymers, and its possible generalizations to arbitrary powers. An interesting question to pursue concerns the extent to which physically occurring stochastic potentials can be approximated accurately by superpositions of dichotomous components.

ACKNOWLEDGMENTS

This work was supported in part by the National Science Foundation under Grant Nos. DMR-9614848 and DMR-9614849.

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[14] As another example of the general asymptotic formula, we mention the rectangular pulse density function which results in Eq. (5.7) for the correlation function. Application of Eq. (6.3)

gives the transcendental equation

$$(y_m/\sigma)^2 wqEkT + e^{-(s+w)y_m}[1+(s+w)y_m] - e^{-sy_m}(1+sy_m) = 0,$$

whose solution y_m has to be obtained numerically. It is trivial, however, to calculate the mobility for any parameter values through a computer algorithm which first solves the above equation and substitutes the solution in Eq. (6.8) to obtain the mobility.

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