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Dyre, Jeppe; Jacobsen, Jacob M.

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Displacement in Extremely Rugged Energy Landscapes
with Equal Minima**

By: Jeppe C. Dyre and Jacob Jacobsen

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IMFUFA, Roskilde Universitetscenter, Postboks 260, 4000 Roskilde

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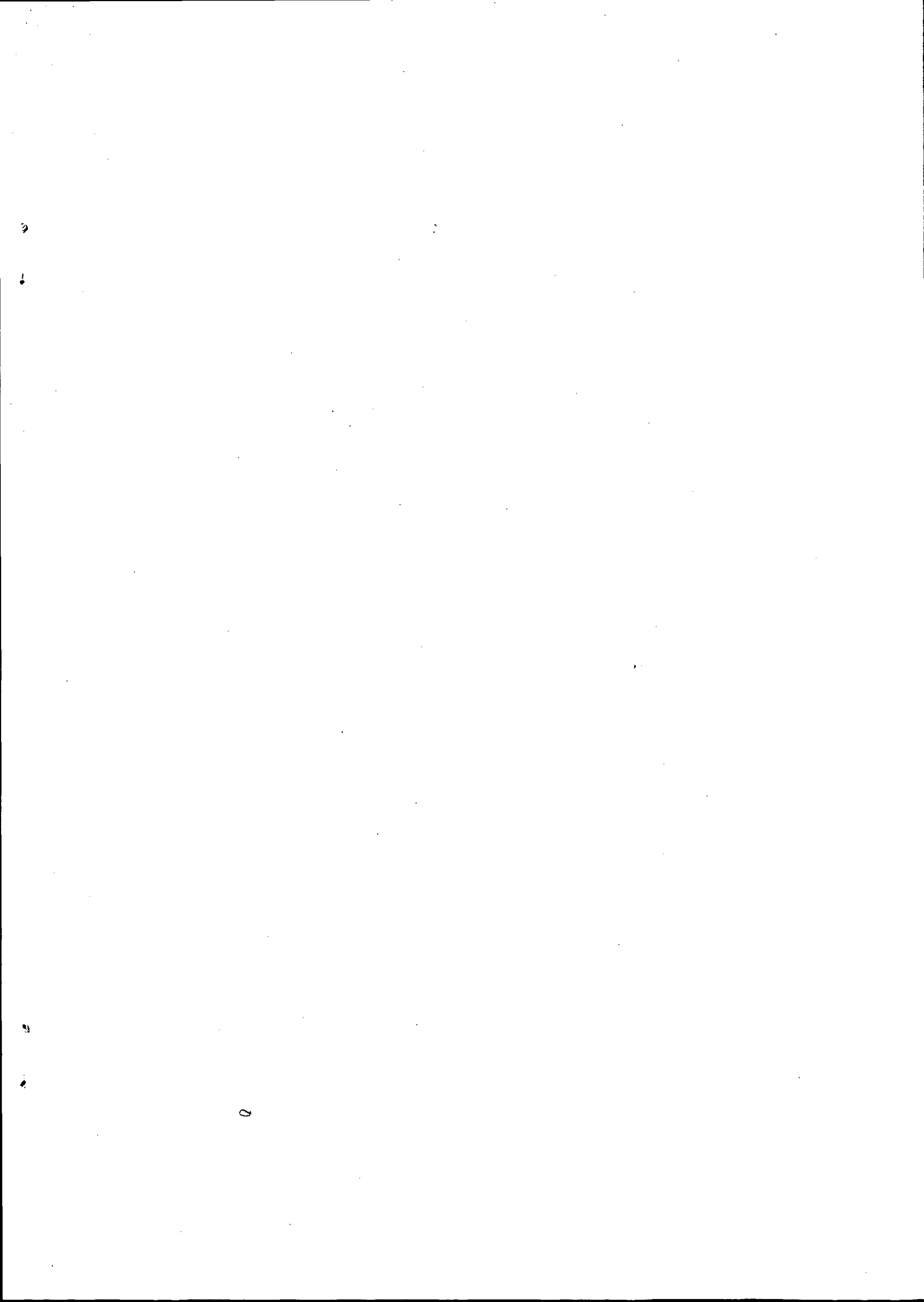
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Abstract

This paper presents a calculation of the time-dependence of the mean-square displacement for symmetric random energy barrier hopping models. The calculation is valid at low temperatures where the frequency-dependence of the normalized diffusion constant \tilde{D} becomes universal, i.e., independent of the energy barrier probability distribution [J. C. Dyre, Phys. Rev. B **49**, 11709 (1994)]. The universal time-dependence of the mean-square displacement is calculated from the effective medium approximation (EMA) universality equation, $\tilde{D} \ln \tilde{D} = \tilde{s}$, where \tilde{s} is the dimensionless imaginary frequency, as well as for the approximation $\tilde{D} \cong \tilde{s} / \ln(1 + \tilde{s})$. At long times the universal mean-square displacement is linear in time, corresponding to ordinary diffusion, whereas it at short times t varies as $2 / \ln(t^{-1})$.

PACS: 05.40.+j; 05.60.+w



I. INTRODUCTION

The study of stochastic motion in a rugged energy landscape is relevant in a number of contexts [1]. Examples include models for AC conduction in disordered solids [2–5], protein dynamics [6], viscous liquids close to the glass transition [7], diffusion in random flows [8], or plasma heat conduction in stochastic magnetic fields [8]. To be specific, consider the Langevin equation of motion [9] for a system with d degrees of freedom subject to the potential $U(X_1, \dots, X_d)$,

$$\dot{X}_i = -\mu \frac{\partial U}{\partial X_i} + \xi_i(t), \quad (1)$$

where μ is a constant and $\xi_i(t)$ is a Gaussian white noise term with variance given by $\langle \xi_i(t) \xi_j(t') \rangle = 2\mu k_B T \delta_{i,j} \delta(t-t')$. In the study of motion in a complex energy landscape, the potential is usually assumed to be random in some specific sense. For instance the potential could be chosen according to a Gaussian functional probability with a specified spatial correlation going to zero at long distances.

For the dynamics defined by Eq. (1) it is possible to monitor the ensemble average energy as function of time, as well as the average displacement as function of time. As an example relating to energy relaxation, the temperature may be an arbitrarily varying function of time and one may calculate how the average energy varies in time. Thus, energy relaxations in viscous liquids close to the glass transition may be modelled [7]. Also, the equilibrium energy time auto-correlation function may be calculated, giving the linear frequency-dependent specific heat [10,11]. In both cases, it is convenient in numerical simulations to use the Smoluchowski equation [9,12] for the probability instead of the noisy Langevin equations.

When the quantity of interest is the average displacement as function of time, the focus is on the mean-square displacement in some fixed axis direction i , $\langle \Delta X_i^2(t) \rangle$. In terms of the canonical equilibrium probability $P_o(X) \propto \exp[-\beta U(X)]$ and the Green's function $G(X \rightarrow X'; t)$, the mean-square displacement is given by (assuming isotropy)

$$\langle \Delta X_i^2(t) \rangle = \frac{1}{d} \int P_o(X) G(X \rightarrow X'; t) (X - X')^2 dX dX'. \quad (2)$$

The mean-square displacement is always an increasing function of time. In disordered systems the mean-square displacement increases linearly with time only asymptotically as $t \rightarrow \infty$. At shorter times, the mean-square displacement varies more rapidly with time leading to a negative curvature of $\langle \Delta X_i^2(t) \rangle$:

$$\frac{d^2}{dt^2} \langle \Delta X_i^2(t) \rangle \leq 0. \quad (3)$$

The faster average displacement at short times is easy to understand. The particle spends most of its time in a potential energy minimum. The most likely displacement is to overcome a low energy barrier to another energy minimum. This is a relatively fast process. From this new position the most likely jump is often to go back to the starting point. Thus, at longer times the displacement is smaller than expected from an extrapolation of the short time displacement.

If s denotes the imaginary ('Laplace') frequency, $s = i\omega$, the frequency-dependent diffusion constant $D(s)$ is defined [2] as follows

$$D(s) = \frac{s^2}{2} \int_0^\infty \langle \Delta X_i^2(t) \rangle e^{-st} dt. \quad (4)$$

It is understood that there is a convergence factor $\lim_{\epsilon \rightarrow 0} e^{-\epsilon t}$ in the integral. For ordinary diffusion, where $\langle \Delta X_i^2(t) \rangle = 2Dt$, one has $D(s) = D$. It is convenient to regard $D(s)$ as an analytic function of s that may be studied also for non-imaginary Laplace frequencies. It is possible to show that Eq. (3) implies that $D(s)$ is an increasing function of s for real s . Writing for real frequencies ω , $D(i\omega) = D'(\omega) + iD''(\omega)$, it can also be shown [13] that the real part $D'(\omega)$ is always an increasing function of ω .

If the particle moves in three dimensions and carry a charge, it is characterized by a frequency-dependent conductivity, $\sigma(\omega)$. By the fluctuation-dissipation theorem $\sigma(\omega)$ is proportional to the frequency-dependent diffusion constant. In fact, Eq. (4) may be derived from the Kubo formula for $\sigma(\omega)$ by two partial integrations [2] (utilizing the fact that the second time derivative of the mean-square displacement is twice the velocity time auto-correlation function).

One way to simplify Eq. (1) is to put it on a lattice. In this way one arrives at a hopping model [3,14–16]. A particularly simple case is when all energy minima have the same energy. If the minima correspond to the sites of the lattice, the problem is reduced to the study of a hopping model with symmetric transition rates for nearest-neighbor jumps. Each transition rate is given by an energy barrier. A further simplification of the model is arrived at, if it is assumed that the transition rates are uncorrelated from link to link; the model is then completely specified by the energy barrier probability distribution.

Symmetric hopping models have been studied extensively particularly with respect to evaluating their frequency-dependent conductivity. These apparently simple models are actually quite complex and cannot be solved analytically, even in one dimension. However, a useful approximation exists for evaluating $D(s)$ (or equivalently $\sigma(s)$), the effective medium approximation (EMA) [4,17]. The EMA is based on old ideas similar to those used in the derivation of the Clausius-Mosotti formula for the dielectric constant of a mixture [18]; in the solid state physics of disordered media the same idea is used in the successful coherent potential approximation (CPA) [19]. Before giving the EMA equation it is convenient to switch to a unit system in which the diffusion constant on a homogeneous lattice with link jump rate Γ is given by $D = \Gamma$. If the dimension is d , $p(\mathbf{k}) = \frac{1}{d} \sum_{i=1}^d \cos(k_i)$ and one defines the following integral over the first Brillouin zone ($-\pi < k_i < \pi$),

$$s\tilde{G} = \int_{1-BZ} \frac{s}{s + 2dD[1 - p(\mathbf{k})]} \frac{d\mathbf{k}}{(2\pi)^d}, \quad (5)$$

the EMA equation for determining $D(s)$ is an equation for the following average over the jump rate probability distribution [3,4]

$$\left\langle \frac{\Gamma - D}{dD + [1 - s\tilde{G}(\Gamma - D)]} \right\rangle_{\Gamma} = 0. \quad (6)$$

The EMA is a mean-field theory which becomes exact as $d \rightarrow \infty$. As recently shown [16], the EMA equation becomes rather simple in the extreme disorder limit, i.e., where the temperature goes to zero and the span of jump frequencies consequently covers more and more decades. In this limit, the EMA equation becomes universal, i.e., independent of the

energy barrier probability distribution. Introducing the normalized dimensionless diffusion constant $\tilde{D} = D(s)/D(0)$ and the dimensionless Laplace frequency $\tilde{s} = i\omega\tau$, where τ is a characteristic time marking the onset of frequency dispersion (the precise value of which is of little interest here), the **EMA universality equation** (valid for $d \geq 2$) [16] is

$$\tilde{D} \ln \tilde{D} = \tilde{s}. \quad (7)$$

At any finite temperature this expression is only valid for a finite range of Laplace frequencies, since $\tilde{D}(\tilde{s})$ eventually becomes independent of \tilde{s} for sufficiently large \tilde{s} . However, as the temperature goes to zero, the range of validity of the EMA universality equation extends to infinity, and therefore the existence of a ‘large frequency cut-off’ is ignored below.

The numerical solution of Eq. (7) was discussed in Ref. [20], that also gave an accurate analytical approximation to $\tilde{D}(\tilde{s})$. Equation (7) implies that

$$\tilde{D} = \frac{\tilde{s}}{\ln \tilde{D}} \cong \frac{\tilde{s}}{\ln \tilde{s}} \quad (|\tilde{s}| \rightarrow \infty). \quad (8)$$

An approximate solution of the EMA universality equation is provided by the following expression (first derived as the continuous time random walk (CTRW) solution of the symmetric hopping model with a box distribution of energy barriers [21]),

$$\tilde{D} = \frac{\tilde{s}}{\ln(1 + \tilde{s})}. \quad (9)$$

The equations (7) and (9) both imply that $\tilde{D}(\tilde{s})$ follows an approximate power law for real Laplace frequencies, $\tilde{D} \propto \tilde{s}^u$ as $\tilde{s} \rightarrow \infty$, where $u = 1 - 1/\ln \tilde{s}$. For real frequencies, one finds for the real part of the diffusion constant that $\tilde{D}' \propto \tilde{\omega}^v$, where $v = 1 - 2/\ln(\tilde{\omega})$ [20].

II. CALCULATION OF THE UNIVERSAL TIME-DEPENDENCE OF THE MEAN-SQUARE DISPLACEMENT

The mean-square displacement is given by the inverse Laplace transform of Eq. (4), where the integration contour as usual is from $-i\infty$ to $i\infty$ to the right of all poles and branch cuts,

$$\langle \Delta X_i^2(t) \rangle = \frac{1}{2\pi i} \oint \frac{2 D(s)}{s^2} e^{st} ds. \quad (10)$$

Note that the boundary condition

$$\langle \Delta X_i^2(t=0) \rangle = 0 \quad (11)$$

is ensured because Eq. (8) implies that $D/s \rightarrow 0$ for $|s| \rightarrow \infty$. Henceforth, it is convenient to consistently adopt the 'rationalized' unit system where $D(0) = 1$ and the time unit is chosen so that $\tilde{s} = s$; this is done by writing $\tilde{D} = D$ and $\tilde{s} = s$. In the 'rationalized' unit system the quantities D , s and t are all dimensionless and Eq. (7) becomes $D \ln D = s$.

The calculation of the universal mean-square displacement is complicated by the fact that $D(s)$ is only given indirectly. We first evaluate the inverse Laplace transform of the approximate expression Eq. (9), which is simpler. Substituting Eq. (9) into Eq. (10) gives

$$\langle \Delta X_i^2(t) \rangle = \frac{1}{2\pi i} \oint \frac{2}{s \ln(1+s)} e^{st} ds. \quad (12)$$

The integrand has a pole at $s = 0$ and a branch cut along the negative real s -axis from $s = -1$ to $s = -\infty$. The integration contour is displaced to run slightly below the real axis from $s = -\infty$ to $s = 0$ and back to $-\infty$ slightly above the axis. The pole at $s = 0$ gives a contribution calculated by expanding at $s = 0$:

$$\begin{aligned} \frac{2e^{st}}{s \ln(1+s)} &= 2 \frac{1 + st + \dots}{s(s - \frac{s^2}{2} + \dots)} \\ &= \frac{2}{s^2} (1 + st + \dots) \left(1 + \frac{s}{2} + \dots\right) = \frac{2}{s^2} \left(1 + \left(t + \frac{1}{2}\right)s + \dots\right), \end{aligned} \quad (13)$$

so the contribution to the integral from this pole is $2t+1$. If one defines $F(x) = 2 e^{xt}/[x \ln(1+x)]$ the remaining part of the integral equals (where $\epsilon > 0$ is infinitesimal)

$$\frac{1}{2\pi i} \int_1^\infty [F(-u - i\epsilon) - F(-u + i\epsilon)] du. \quad (14)$$

Since $F(-u + i\epsilon)$ is the complex conjugate of $F(-u - i\epsilon)$ Eq. (14) becomes

$$\frac{2}{\pi} \int_1^\infty \text{Im} \frac{e^{-ut}}{(-u)[\ln(u-1) - i\pi]} du = -2 \int_1^\infty \frac{e^{-ut}}{\ln^2(u-1) + \pi^2} \frac{du}{u}. \quad (15)$$

To summarize, the mean-square displacement is in the approximation Eq. (9) given by

$$\langle \Delta X_i^2(t) \rangle = 2t + 1 - 2 \int_1^\infty \frac{e^{-ut}}{\ln^2(u-1) + \pi^2} \frac{du}{u}. \quad (16)$$

Since $\langle \Delta X_i^2(t=0) \rangle = 0$, Eq. (16) may be rewritten [22] as

$$\langle \Delta X_i^2(t) \rangle = 2t + 2 \int_1^\infty \frac{1 - e^{-ut}}{\ln^2(u-1) + \pi^2} \frac{du}{u}. \quad (17)$$

Equation (17) makes it possible to estimate the asymptotic behavior of the mean-square displacement at short times, as briefly sketched here: The term $2t$ is, it turns out, insignificant compared to the integral. The latter quantity is separated into two terms, one integral from 1 to t^{-1} and one from t^{-1} to ∞ . In the first integral the term $1 - e^{-ut}$ is smaller than ut and the denominator may be replaced by $\ln^2(t^{-1})$; thus this integral is of order $2/\ln^2(t^{-1})$. This quantity is small compared to the value of the second integral: Here the term $1 - e^{-ut}$ may be replaced by 1 and the denominator may be replaced by $\ln^2(u)$. Thus, for $t \ll 1$ one has

$$\langle \Delta X_i^2(t) \rangle \cong \int_{t^{-1}}^\infty \frac{2}{\ln^2(u)} \frac{du}{u} = \frac{2}{\ln(t^{-1})}. \quad (18)$$

This asymptotic behavior suggests the following analytical approximation to Eq. (16),

$$\langle \Delta X_i^2(t) \rangle \cong \frac{2}{\ln(1 + t^{-1})}, \quad (19)$$

since this expression has both the correct short time behavior given by Eq. (18) and, as is easy to show, the correct long time behavior, $\langle \Delta X_i^2(t) \rangle \cong 2t + 1$. Equation (19) gives an approximation to Eq. (16) which for any t deviate less than 7 percent.

We now turn to the Laplace inversion of the EMA universality equation. It is possible to show that the equation $D \ln D = s$ defines $D(s)$ for all complex s , except the negative real numbers between $-\infty$ and $-1/e$ (this is confirmed by Eq. (34) below). Consequently, we again choose as integration contour the one going from $s = -\infty$ slightly below the s -axis and back to $s = -\infty$ slightly above the axis, encircling all poles and branch cuts. There is a pole at $s = 0$, just as above, and the branch cut this time stretches from $s = -1/e$ to $s = -\infty$. The residue at $s = 0$ is by standard rules equal to

$$\frac{d}{ds} [2 D(s)e^{st}] |_{s=0} = 2(D'(0) + D(0)t). \quad (20)$$

The EMA universality equation Eq. (7) implies that $D(0) = 1$ and by differentiation that $D'(0)[\ln D(0) + 1] = 1$ or $D'(0) = 1$. Thus, the residue is equal to $2t + 2$. For the remaining integration it is convenient to change to D as integration variable. The EMA universality implies $ds = (1 + \ln D)dD$ and thus

$$\langle \Delta X_i^2(t) \rangle = 2t + 2 + \frac{1}{2\pi i} \oint 2 \frac{D + D \ln D}{(D \ln D)^2} e^{D \ln Dt} dD. \quad (21)$$

The integration contour in the D - plane is defined by $D \ln D$ being real and $\leq -1/e$. Writing $D = re^{i\theta}$, the equation defining the integration contour is $\text{Im}(D \ln D) = 0$ which, since $\ln D = \ln r + i\theta$, implies that [23] $\theta \cos \theta = -\sin \theta \ln r$, or

$$r = e^{-\theta \cot \theta} \quad (-\pi < \theta < \pi). \quad (22)$$

Equation (22) implies that on the integration contour the real number $D \ln D$ is given by

$$D \ln D = re^{i\theta}(\ln r + i\theta) = e^{-\theta \cot \theta} \theta (-\cos \theta \cot \theta - \sin \theta), \quad (23)$$

or

$$D \ln D = -E(\theta), \quad (24)$$

where

$$E(\theta) = \frac{\theta}{\sin \theta} e^{-\theta \cot \theta}. \quad (25)$$

The function $E(\theta)$ varies monotonically from $1/e$ to ∞ as $|\theta|$ varies from 0 to π . Next, the integration variable is changed to θ . The differential of D is given by $dD = e^{i\theta}(dr + ir d\theta)$. From Eq. (22) one finds $dr = r(-\cot \theta + \frac{\theta}{\sin^2 \theta})d\theta$, and thus

$$dD = D \left(-\cot \theta + \frac{\theta}{\sin^2 \theta} + i \right) d\theta. \quad (26)$$

Substituting Eqs. (24) and (26) into Eq. (21) one obtains

$$\langle \Delta X_i^2(t) \rangle = 2t + 2 + \frac{1}{2\pi i} \int_{-\pi}^{\pi} 2 \frac{r e^{i\theta} - E(\theta)}{E^2(\theta)} e^{-E(\theta)t} r e^{i\theta} \left(-\cot \theta + \frac{\theta}{\sin^2 \theta} + i \right) d\theta. \quad (27)$$

Since $r = \frac{\sin \theta}{\theta} E(\theta)$ (Eqs. (22) and (25)) the factor $E^2(\theta)$ cancels. The integrand is the real number $e^{-E(\theta)t}$ times the quantity

$$2 \left(\frac{\sin \theta}{\theta} (\cos \theta + i \sin \theta) - 1 \right) \frac{\sin \theta}{\theta} (\cos \theta + i \sin \theta) \left(-\cot \theta + \frac{\theta}{\sin^2 \theta} + i \right). \quad (28)$$

As is straightforward to show, this function of θ has an antisymmetric real part and a symmetric imaginary part equal to $-2F(\theta)$ where

$$F(\theta) = \left(\cos \theta - \frac{\sin \theta}{\theta} \right)^2 + \sin^2 \theta. \quad (29)$$

Since Eq. (28) is to be multiplied with the symmetric factor $e^{-E(\theta)t}$ and integrated from $-\pi$ to π , only the symmetric imaginary part of Eq. (28) gives a contribution. We thus finally arrive at

$$\langle \Delta X_i^2(t) \rangle = 2t + 2 - \frac{2}{\pi} \int_0^{\pi} F(\theta) e^{-tE(\theta)} d\theta. \quad (30)$$

Utilizing the fact that $\langle \Delta X_i^2(t=0) \rangle = 0$ Eq. (30) may be rewritten as

$$\langle \Delta X_i^2(t) \rangle = 2t + \frac{2}{\pi} \int_0^{\pi} F(\theta) (1 - e^{-tE(\theta)}) d\theta. \quad (31)$$

Figure 1 shows a log-log plot of the universal mean-square displacement (full curve) as well as the mean-square displacement according to Eq. (16) (dashed curve, deviation less than 30% from the full curve). At long times one has ordinary diffusion leading to a mean-square displacement that grows linearly with time. At short times, the mean-square displacement is much larger than expected by extrapolation from the long time behavior. A detailed analysis of the asymptotic behavior of Eq. (30) for $t \rightarrow 0$ is somewhat involved. However, the short time behavior of $\langle \Delta X_i^2(t) \rangle$ is determined by the behavior of $D(s)$ for large Laplace frequencies, and a detailed analysis is unnecessary since we can refer directly to Eq. (18): This result must be valid also for the mean-square displacement given by Eq. (30) because of the asymptotic behavior Eq. (8).

An analytic approximation to Eq. (30), which for any t is more accurate than 3.3% is given by the following formula

$$\langle \Delta X_i^2(t) \rangle \simeq \frac{2}{\ln(1 + \frac{1}{t}) - \ln[\ln(e + \frac{1}{t})]} - \frac{2t}{e-1}. \quad (32)$$

Finally, we note that Eq. (30) gives rise to an explicit integral expression for $D(s)$ using Eq. (4). The term $2t + 2$ in Eq. (30) is transformed into $1 + s$ and thus

$$D(s) = 1 + s - \frac{1}{\pi} \int_0^\pi F(\theta) \frac{s^2}{s + E(\theta)} d\theta. \quad (33)$$

Utilizing the fact that $\int_0^\pi F(\theta) d\theta = \pi$ (which follows from $\langle \Delta X_i^2(t=0) \rangle = 0$ but may also be shown by direct calculation), Eq. (33) may be rewritten

$$D(s) = 1 + \frac{1}{\pi} \int_0^\pi F(\theta) \frac{sE(\theta)}{s + E(\theta)} d\theta. \quad (34)$$

Thus, as a byproduct of the calculation of the time-dependence of the mean-square displacement we have derived an integral representation of the function $D(s)$ obeying the functional equation $D(s) \ln D(s) = s$. Equation (34) confirms the fact used above, that $D(s)$ is defined for all complex s except the negative real numbers from $-1/e$ to ∞ . For the practical numerical evaluation of $D(s)$, this integral representation is not as useful as the Newton-Raphson method [20].

III. CONCLUSION

An analytic expression for the time-dependence of the mean-square displacement for low-temperature hopping has been derived from the EMA universality equation Eq. (7). At short times the mean-square displacement varies as $2/\ln(t^{-1})$, indicating a considerably faster effective motion at short times than expected from the long time diffusive behavior $\propto t$.

The expression derived for the mean-square displacement is valid as the temperature goes to zero. At any finite temperature the mean-square displacement actually returns to diffusive behavior $\propto t$ at very short times, corresponding to the fact that at very high

Laplace frequencies $D(s)$ always becomes constant. This effect has been ignored here because the range of validity of Eq. (30) becomes larger and larger as the temperature is lowered.

The transition from 'logarithmic' diffusion to ordinary diffusion defines a characteristic time, which in the above used dimensionless units is of order one. In real units this characteristic time is thermally activated with an activation energy equal to the percolation energy, the lowest energy barrier met on a long optimal path [16]. It follows from the detailed EMA treatment of the problem [16], that the 'DC' diffusion constant in real units, $D(0)$, is Arrhenius with the same activation energy. This fact, which was confirmed by the computer simulations of Ref. [16], has been known for many years from experiments on AC conduction in ionically and electronically disordered solids (the 'BNN relation' [24,20,16]).

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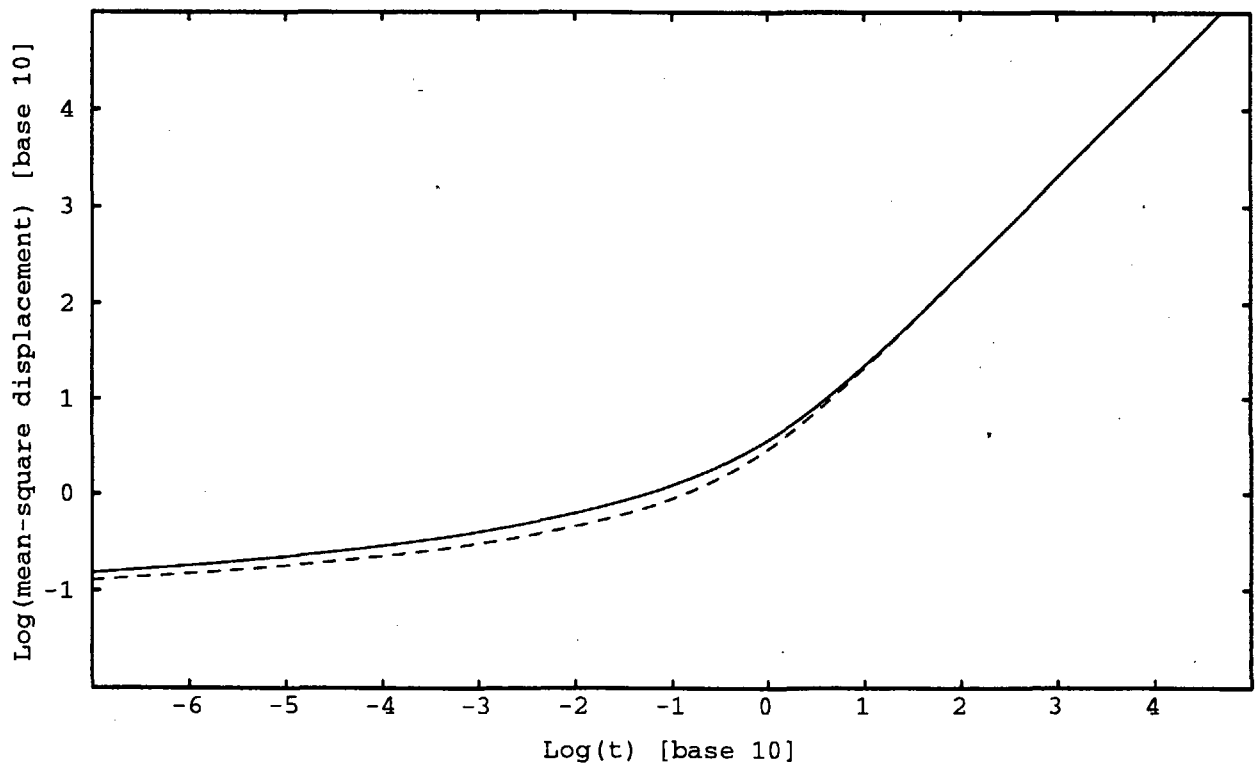
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FIGURES

FIG. 1. The universal mean-square displacement in dimensionless units in a log-log plot (base 10) according to the EMA (Eq. (31), full curve) and to the approximation to the EMA given by Eq. (17) (dashed curve). At short times the mean-square displacement varies as $1/\ln(t^{-1})$ which implies a much faster motion than expected from an extrapolation of the long time diffusive behavior, $\langle \Delta X_i^2(t) \rangle \propto t$. This is due to the fact that at short times it is mainly small barriers that are overcome, which is a fast process. At longer times the largest barrier on a 'percolation path' will have to be overcome in order to extend the diffusion to infinity.



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