

## Generalized Einstein relation: A stochastic modeling approach

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For anomalous random walkers, whose mean square displacement behaves like  $\langle x^2(t) \rangle \sim t^\delta$  ( $\delta \neq 1$ ), the generalized Einstein relation between anomalous diffusion and the linear response of the walkers to an external field  $F$  is studied, using a stochastic modeling approach. A departure from the Einstein relation is expected for weak external fields and long times. We investigate such a departure using the Scher-Lax-Montroll model, defined within the context of the continuous time random walk, and which describes electronic transport in a disordered system with an effective exponent  $\delta < 1$ . We then consider a collision model which for the force free case may be mapped on a Lévy walk ( $\delta > 1$ ). We investigate the response in such a model to an external driving force and derive the Einstein relation for it both for equilibrium and ordinary renewal processes. We discuss the time scales at which a departure from the Einstein relation is expected. [S1063-651X(98)07907-0]

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### I. INTRODUCTION

Biased and unbiased anomalous diffusions are well established phenomena [1–8] found in many different systems. For unbiased processes, the mean and mean square displacements of a test particle interacting with some kind of thermal bath can behave like

$$\langle \mathbf{x}^2(t) \rangle_0 \sim t^\delta \quad \text{and} \quad \langle \mathbf{x}(t) \rangle_0 = 0. \quad (1)$$

The subscript zero in  $\langle \cdot \cdot \cdot \rangle_0$  denotes the case when no external driving force is applied to the particle. When  $\delta \neq 1$  the diffusion is anomalous, the case  $\delta < 1$  is called slow diffusion or subdiffusion, and  $\delta > 1$  is called enhanced diffusion. Such a behavior may be found in the absence of an external macroscopic driving force. When such an external force  $\mathbf{F}$  is applied the symmetry of the system is broken and then

$$\langle \mathbf{x}(t) \rangle_F \sim t^{\delta_F} \quad (2)$$

is found. In what comes we call a process for which  $\delta_F = \delta$  = 1 normal process, otherwise the process is considered to be anomalous. Normal processes are usually Gaussian while anomalous processes are as a rule non-Gaussian.

If the system is close to thermal equilibrium at a temperature  $T$ , the generalized Einstein relation [5,7,9–13] relates the fluctuations of the test particle position in the absence of an external field to its behavior under the influence of a constant (time and space independent) force field  $\mathbf{F}$ , according to [5]

$$\langle x_{\parallel}^2(t) \rangle_0 = 2 \frac{k_B T}{F} \langle x_{\parallel}(t) \rangle_F. \quad (3)$$

Here  $x_{\parallel}$  is the component of  $\mathbf{x}$  along  $\mathbf{F}$ . Equation (3) is strictly valid only in the linear response regime which might be found when  $F \rightarrow 0$ .

When the external field is finite though weak and for systems exhibiting normal diffusion and transport the Einstein relation between the diffusion and mobility coefficients [14,15] is useful in describing transport for both short and

long times (high and low frequencies). For anomalous transport systems this approach is not expected to work so well [to see this insert Eqs. (1) and (2) in Eq. (3) and assume  $\delta_F \neq \delta$ ]. As can be seen from Eq. (3) if the exponent  $\delta_F$  depends weakly on the external force then for any finite force and long enough time a large deviation from the generalized Einstein relation may be expected.

The Scher-Lax-Montroll (SLM) model [16–18] defined within the context of the continuous time random walk (CTRW) will be considered here. We show that for this model, describing subdiffusion of electrons in disordered materials, the effective exponent  $\delta_F$  is indeed force dependent. Once this exponent is calculated, it is then easy to find the time scales at which the deviations from the Einstein relation are expected to be large.

Bouchaud and Georges [5], starting from a Hamiltonian description of an unspecified system, give a proof of the generalized relation (3), based on a linearization procedure valid for short times. Their derivation is carried out for fixed disorder. In Sec. II we show that the generalized Einstein relation (3) can be derived from the well known linear response theory [15]. This derivation is not limited to a fixed disorder, however, it does assume that the underlying stochastic process is stationary. This inspired us to investigate both stationary and nonstationary stochastic processes. We have found that for a collision model under consideration here, the Einstein relation is valid (under certain conditions) also for nonstationary processes, provided that  $F \rightarrow 0$ .

Violations of the Einstein relation were found for several transport models [5,19,20]. Bouchaud and Georges [5] have already pointed out that for long times and finite forces the Einstein relation is not expected to be a useful approximation. Without limiting themselves to a specific model they predict a possibility of a crossover from a short time behavior with  $\langle x(t) \rangle_F \sim t^\delta$  to another long time regime with  $\langle x(t) \rangle_F \sim t$ . The subsection in Sec. III uses the SLM model to investigate a different type of departure from the Einstein relation and gives a detailed description of its nature for a specific model.

We were also motivated by recent experiments which have checked the validity of the Einstein relation for two different types of slow diffusions with  $\delta_F < 1$  and  $\delta < 1$ . Qing *et al.* [21] measured a non-Gaussian diffusion in semiconductors. The experiment indicated the correctness of Eq. (3) to within the prefactor of the order one which could not yet be determined exactly. Amblard *et al.* [22] measured both  $\langle \tilde{x}^2(t) \rangle_0$  and  $\langle \tilde{x}(t) \rangle_F$  for magnetic beads (diameter  $\mu m$ ) on a polymer network. We have found out [23] that the experiment of Amblard *et al.* is in agreement with the generalized Einstein relation. As far as we know this is the first direct experimental verification of the generalized Einstein relation for an anomalous system.

Section IV considers a stochastic collision model [24,25] resulting in an enhanced diffusion. In this model a particle of a mass  $M$  collides at random times with heat bath particles whose mass is  $m$ . An important parameter controlling the strength of the collisions is the mass ratio  $\epsilon \equiv m/M$ . We generalize Drude's approach to the case of a long tailed probability density function (PDF) of the independent time intervals between collision events. The dynamics of the particle between collision events are Newtonian. In the absence of an external force and under certain conditions [25], the collision model can be considered as belonging to the same universality class as the Lévy walks [1,2,26–28] since it produces the same asymptotic time behaviors of the mean square displacement.

Here we consider the influence of an external force and derive the Einstein relation (3) for this model. This type of a model allows us to discuss the limiting cases of strong  $\epsilon = 1$  or weak  $\epsilon \ll 1$  collisions. We show, *inter alia*, that unlike Gaussian transport processes, the mean displacement  $\langle x(t) \rangle_F$  in the long time limit does not depend on the strength of the collisions (i.e., it is independent of the mass ratio  $\epsilon = m/M$ ). However, the parameter  $\epsilon$  is important since it controls the transition from the short time behavior  $\langle x(t) \rangle_F \sim t$ , to an enhanced,  $\langle x(t) \rangle_F \sim t^{\delta_F}$ ,  $\delta_F > 1$ , behavior valid for long times.

## II. ANOMALOUS DIFFUSION, LINEAR RESPONSE THEORY, AND STATIONARITY

The linear response theory [15] is used here to show the conditions under which the generalized Einstein relation (3) holds. For simplicity we shall consider here a one dimensional case. A test particle, described by an unspecified Hamiltonian, moves under the action of a perturbation  $H_{\text{ext}} = -x F(t)$  where  $F(t)$  is an external time dependent force. Assuming  $F(t) = F \theta(t)$  where  $\theta(t)$  is a step function, linear response theory yields the average velocity of the test

$$\frac{d}{dt} \langle x(t) \rangle_F = (F/k_B T) \int_0^t \langle v(t') v(0) \rangle_0 dt', \quad (4)$$

where  $\langle v(t') v(0) \rangle_0$  is the canonical velocity correlation function at thermal equilibrium for  $F=0$ . The mean square displacement  $\langle x^2(t) \rangle_0$  is also related to the correlation function according to

$$\langle x^2(t) \rangle_0 = \int_0^t \int_0^t \langle v(t') v(t'') \rangle_0 dt' dt''. \quad (5)$$

For a stationary process, the function  $\langle v(t') v(t'') \rangle$  depends on the time difference  $|t' - t''|$  only. Using the stationary condition and the convolution theorem for Laplace transform it is easy to show that in the Laplace domain ( $t \rightarrow u$ )

$$\langle \hat{x}(u) \rangle_F = \frac{F}{2k_B T} \langle \hat{x}^2(u) \rangle_0. \quad (6)$$

Returning to the time domain we get the generalized Einstein relation (3). Equation (6) is assumed to work well both for normal and anomalous diffusions and for different types of disorder.

In deriving Eq. (6) it was assumed [15] that the perturbation has been switched on in the infinite past, when the system is described by a canonical density matrix. It was also assumed that the process is stationary, meaning that probabilities describing the process are invariant with respect to time shifts [15]. According to Ref. [15], the stationary condition is satisfied if the environment is in a stationary state with a constant temperature, pressure, etc., and if the particle spends long enough times interacting with its environment.

We may ask, how long is long enough? One expects that, if a relaxation time exists, one should wait for times long compared with this time scale. However, for some processes which result in anomalous diffusion no such time scale exists. Thus we believe that an additional insight into the validity of the Einstein relation is achieved by considering a kinetic approach which assumes a stochastic (non-Hamiltonian) description of the system.

## III. CTRW

In the decoupled version of the CTRW [3,8,29] which was introduced by Montroll and Weiss over 30 years ago, a walker hops from site to site and at each site it is trapped for a random time. For this well known model two independent PDFs describe the walk. The first is the  $\psi(t)$  PDF of the pausing times between successive steps. The second one is the  $\xi(\tilde{x})$  PDF for the displacement of the walker at each step. Shlesinger [30] showed that anomalous subdiffusion arises if  $\psi(t)$  is long tailed with its Laplace transform behaving like

$$\hat{\psi}(u) \approx 1 - A_F u^{\delta_F}, \quad \text{with } 0 < \delta_F < 1, \quad (7)$$

meaning that even the first moment of this PDF diverges.

The first two moments of the hopping length PDF,  $\xi(\tilde{x})$ , are assumed to exist, so that its Fourier transform for small  $q$  behaves like

$$\tilde{\xi}(q) = 1 + iq \langle \tilde{x} \rangle_F - \frac{1}{2} q^2 \langle \tilde{x}^2 \rangle_F + \dots \quad (8)$$

The parameters  $A_F$ ,  $\delta_F$ ,  $\langle \tilde{x} \rangle_F$ , and  $\langle \tilde{x}^2 \rangle_F$  determine the long time behavior of the mean linear  $\langle x(t) \rangle_F$ , and mean square  $\langle x^2(t) \rangle_F$ , displacements of the walker. They may, in principle, depend on the strength  $F$  of the external field. When the medium in which the walk is performed is on average isotropic and when  $F=0$ , it follows from the symmetry considerations that  $\langle \tilde{x} \rangle_0 = 0$ .

According to the results derived in [8] [see Eq. (2.82b) therein] the mean square displacement of the walker in the absence of the field is

$$\langle x^2(t) \rangle_0 \approx \langle \tilde{x}^2 \rangle_0 \frac{t^{\delta_0}}{A_0 \Gamma(1 + \delta_0)}. \quad (9)$$

When the walk is biased, we have [see Eq. (2.81b) in Ref. [8]]

$$\langle x(t) \rangle_F \approx \langle \tilde{x} \rangle_F \frac{t^{\delta_F}}{A_F \Gamma(1 + \delta_F)}. \quad (10)$$

We define now the dimensionless ratio

$$R(t) \equiv \frac{F}{2k_B T} \frac{\langle x^2(t) \rangle_0}{\langle x(t) \rangle_F}, \quad (11)$$

which according to the Einstein relation is expected to be time independent and satisfies  $R(t) = 1$ . However according to Eqs. (9) and (10)

$$R(t) = \frac{F}{2k_B T} \frac{\langle \tilde{x}^2 \rangle_0 A_F \Gamma(1 + \delta_F)}{\langle \tilde{x} \rangle_F A_0 \Gamma(1 + \delta_0)} t^{\delta_0 - \delta_F}. \quad (12)$$

For  $F \neq 0$  the quantity  $R(t)$  varies with time if  $\delta_F \neq \delta_0$ . Equation (12) shows that for any finite force, however small (i.e., even when  $F l / k_B T \ll 1$  with  $l$  being a characteristic microscopic scale), large deviations from the Einstein relation will be found for long enough times. Such a behavior is never found for Gaussian diffusion processes for which the ratio  $R(t)$  is time independent and for small fields  $R(t) \approx 1$ . Also notice that for Gaussian processes the ratio  $R(t)$  can be Taylor expanded in powers of the external force  $F$  around  $F = 0$  the coefficients being independent of time, whereas this is not possible for the anomalous processes with  $\delta_F \neq \delta_0$ .

Let us assume that the Einstein relation is valid when  $F \rightarrow 0$  and check what it implies for the relation between the microscopic parameters which enter the CTRW modeling. This means that we take  $\lim_{F \rightarrow 0} R(t) = 1$ , which implies  $\lim_{F \rightarrow 0} \delta_F = \delta_0$ ,  $\lim_{F \rightarrow 0} A_F = A_0$ , and

$$\lim_{F \rightarrow 0} \frac{F}{2k_B T} \frac{\langle \tilde{x}^2 \rangle_0}{\langle \tilde{x} \rangle_F} = 1. \quad (13)$$

Comparing between this equation and Eq. (3) we notice that Eq. (13) is an Einstein relation for the microscopic parameters of the CTRW model. In Appendix A we show in a straightforward way that Eq. (13) is valid for a model of symmetric random barriers thus giving some justification to the correctness of Eq. (13) and hence to the Einstein relation. The important assumption made in Appendix A is that the microscopic stochastic dynamics satisfies detailed balance which is the condition needed to ensure validity of the generalized Einstein relation.

It is interesting to emphasize that the two limits of long time and weak field in Eq. (12),

$$\lim_{F \rightarrow 0} \lim_{t \rightarrow \infty} R(t) \neq \lim_{t \rightarrow \infty} \lim_{F \rightarrow 0} R(t), \quad (14)$$

do not commute. The long time Einstein result is reached if we take first  $F \rightarrow 0$  and only then  $t \rightarrow \infty$ .

### Calculation of $\psi(t)$

This subsection shows, for a specific example, how a dependence of the exponent  $\delta_F$  on an external force may arise. Scher and Lax [16,17] were the first to calculate the function  $\psi(t)$  for an unbiased system and to use it within the CTRW for a description of hopping electron transport (i.e., current flow in semiconductors such as Si and Ge due to tunneling between impurities). They showed that for large times

$$\psi(\tau) \sim (\ln \tau)^2 / \tau^{1 + (\eta/3)(\ln \tau)^2}, \quad (15)$$

where  $\tau$  is a dimensionless time,  $\eta = 4\pi R_d^3 N_D$  with  $R_d$  being half of an effective Bohr radius and  $N_D$  the density of donors in the system. Unlike the PDF (7) the first moment of the PDF (15), exists. The logarithmic dependence of the exponent  $(\eta/3)(\ln \tau)^2$  on time guarantees that  $\psi(\tau)$  behaves like a power law for long times.

For calculations of transient photocurrent one can approximate Eq. (15) by  $\psi(\tau) \sim \tau^{-1-\delta}$  with [18]

$$\delta = \frac{1}{3} \eta (\ln \tau)^2, \quad \tau = O(\tau_t) \quad (16)$$

and  $\tau_t$  is a transient time. Scher and Lax have assumed the validity of the Einstein relation and with it calculated the complex ac mobility using Eq. (15). Thus their calculation of  $\psi(t)$  is for  $\mathbf{F} = \mathbf{0}$ .

An external field can strongly influence tunneling as observed in different physical effects (e.g., cold emission [31], Landau-Zener breakdown [32], and electron scavenging [33,34]). Here we shall calculate  $\psi(\tau)$  using the same procedure as used by SL for a system subject to a uniform bias. We shall show that when an external uniform field  $\mathbf{E}$  is switched on, the dimensionless density  $\eta$  is renormalized as

$$\eta \rightarrow \frac{\eta}{(1 - \beta^2)^2}, \quad (17)$$

with

$$\beta \equiv \left| \frac{e \mathbf{E} R_d}{k_B T} \right|, \quad (18)$$

the functional form of  $\psi(\tau)$ , Eq. (15), remaining unchanged.

Figure 1 shows the ratio between the PDF  $\psi_\beta(\tau)$  for  $\beta \neq 0$  and the field free result of SLM for the PDF  $\psi_0(\tau)$  (below the subscript  $\beta$  is suppressed). We see that for long times this ratio tends to zero. In the SLM theory the power law behavior of  $\psi(\tau)$  for large times gives the low frequency behavior of the anomalous ac conductivity. From Eq. (17) and Fig. 1 we conclude that there exist long times for which the sensitivity of  $\psi(\tau)$  on  $\beta$  becomes important and must be taken into account when analyzing experimental results obtained from finite field experiments.

When an external field reduces the potential barrier the charge carrier has to cross, the tunneling rate increases and this in turn may have a strong influence on the transport. This increase of the tunneling rate is consistent with our

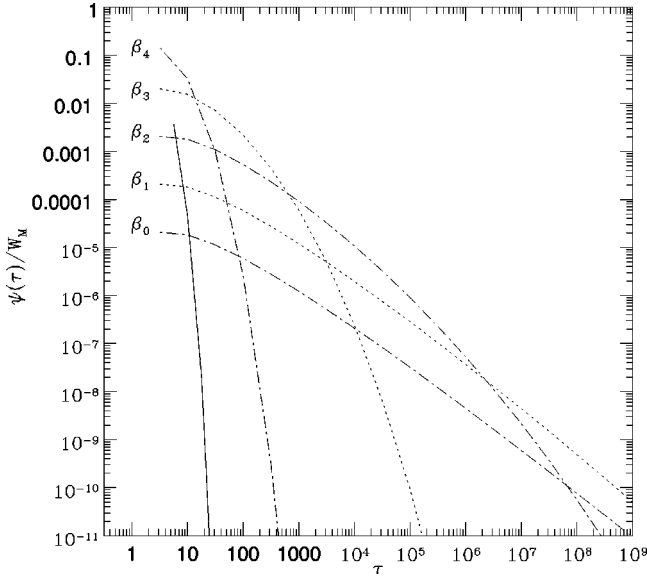


FIG. 1. The ratio  $\psi_\beta(\tau)/\psi_0(\tau)$  vs  $\tau$  for different values of  $\beta$  and  $\eta=10^{-2}$ . For  $D$  donors in Si SL use  $R_d=12 \text{ \AA}$  thus  $\eta=10^{-2}$  corresponds to  $N_D=4.610^{-17} \text{ cm}^{-3}$ . For  $T=3 \text{ K}$ ,  $\beta=0.15$  implies the field  $E=323 \text{ V/cm}$ . The intersection of the three curves,  $\psi_\beta(\tau)/\psi_0(\tau)=1$ , is explained in the text.

result (17), which implies that for fixed  $\eta$  and  $T$ ,  $\psi(\tau)$  decays faster to zero as the field  $\mathbf{E}$  increases. This behavior is in agreement with the numerical simulations [35]. Scher [36], in the context of recombination processes, pointed out that the exponents are functions of the external fields. Here the dependence of the PDF  $\psi(\tau)$  on the field is found and the approximation (17) is discussed.

According to the SLM procedure the probability  $Q(t)$  is introduced, of finding an electron in a given trap at time  $t$  if it has been there at time  $t=0$ . This probability decreases with time due to tunneling hopping to surrounding traps. Then according to Eq. (10) in Ref. [17],

$$\psi(t) = -\frac{d\langle Q(t) \rangle}{dt}, \quad (19)$$

with

$$\langle Q(t) \rangle = \exp\left(-N_D \int d^3r \{1 - \exp[-W(\mathbf{r})t]\}\right), \quad (20)$$

where  $W(\mathbf{r})$  is the transition rate between donors separated by  $\mathbf{r}$ .

The function  $W(\mathbf{r})$  has been calculated by Miller and Abrahams [37]. Using SLM notation

$$W(\mathbf{r}) \approx W'_0 \left(\frac{r}{R_d}\right)^{3/2} \Delta(\mathbf{r}) \exp(-r/R_d) \exp\left[-\frac{\Delta(\mathbf{r})}{k_B T}\right], \quad (21)$$

where  $\Delta(\mathbf{r})$  is the energy difference between the traps. Equation (21) was derived using the variational principle. It applies when the process of phonon absorption ( $\Delta > 0$ ) dominates. Equation (21) is an approximation valid when  $\Delta/k_B T \gg 1$ .

There are two contributions

$$\Delta(\mathbf{r}) = \Delta_0(\mathbf{r}) + e\mathbf{E} \cdot \mathbf{r}, \quad (22)$$

where the first term  $\Delta_0(\mathbf{r})$  is the energy difference due to the random potential and the second term is caused by the external field  $\mathbf{E}$ . We consider length scales at which

$$|\Delta_0| \gg |e\mathbf{E} \cdot \mathbf{r}|, \quad (23)$$

with  $|\Delta_0|$  being the variance of the fluctuating function  $\Delta(\mathbf{r})$ . The calculation is carried out assuming that the constant  $|\Delta_0|$  can replace the  $\mathbf{r}$  dependent function  $\Delta_0(\mathbf{r})$ . The agreement between theory and experiment found by SLM is the main justification for this approach. Equation (21) is replaced by the simple form

$$W(\mathbf{r}) = W_M \exp\left(-\frac{r}{R_d} - \frac{e\mathbf{E} \cdot \mathbf{r}}{k_B T}\right), \quad (24)$$

with

$$W_M = W'_0 \left(\frac{\bar{r}}{R_d}\right)^{3/2} \exp\left(-\frac{|\Delta_0|}{k_B T}\right). \quad (25)$$

$\bar{r}$  is an appropriate mean value. Equation (24) is valid for weak fields satisfying  $\beta < 1$ .  $W_M$  is an attempt frequency whose dependence on the external field is neglected.

A simple way to find the behavior of  $\psi(t)$  is to notice that the integrand in the exponent in Eq. (20) behaves like a step function,

$$1 - \exp[-W(\mathbf{r})t] = \begin{cases} 1, & W(\mathbf{r})t < 1 \\ 0, & W(\mathbf{r})t > 1 \end{cases} \quad (26)$$

and then using Eq. (24) to show that

$$-\ln\langle Q(t) \rangle = \frac{1}{3} (\ln \tau)^3 \frac{\eta}{(1-\beta^2)^2}, \quad (27)$$

with  $\tau = W_M t$ . An accurate approach given in Appendix B, shows that Eq. (27) is asymptotically exact to within a numerical factor.

The exact analysis shows that

$$\frac{\psi(\tau)}{W_M} \sim \frac{\eta e^\gamma}{(1-\beta^2)^2} (\ln e^\gamma \tau)^2 (e^\gamma \tau)^{-1 - (1/3)[\eta(1-\beta^2)^2](\ln e^\gamma \tau)^2}, \quad (28)$$

with  $e^\gamma \approx 1.78$ . This form replaces Eq. (15) derived for  $\mathbf{E} = \mathbf{0}$ . We see that the effective exponent is field dependent, and therefore the response to the field is nonlinear and as we have discussed in the preceding section the Einstein relation for long times and finite fields cannot be used to analyze the transport properties of the system. In Fig. 2 we show the PDF  $\psi(\tau)$  equation (28) vs  $\tau$  for fixed  $\eta$ . We observe a crossover from a power law behavior of  $\psi(\tau)$  valid for  $\beta = 0$  to an exponential decay found when  $\beta \rightarrow 1$ .

Using Eq. (28) we define now the ratio

$$\frac{\psi_\beta(\tau)}{\psi_0(\tau)} = \frac{1}{(1-\beta^2)^2} \exp\left\{-\frac{\eta \beta^2 (2-\beta^2)}{3 (1-\beta^2)^2} [\ln(e^\gamma \tau)]^3\right\} \quad (29)$$

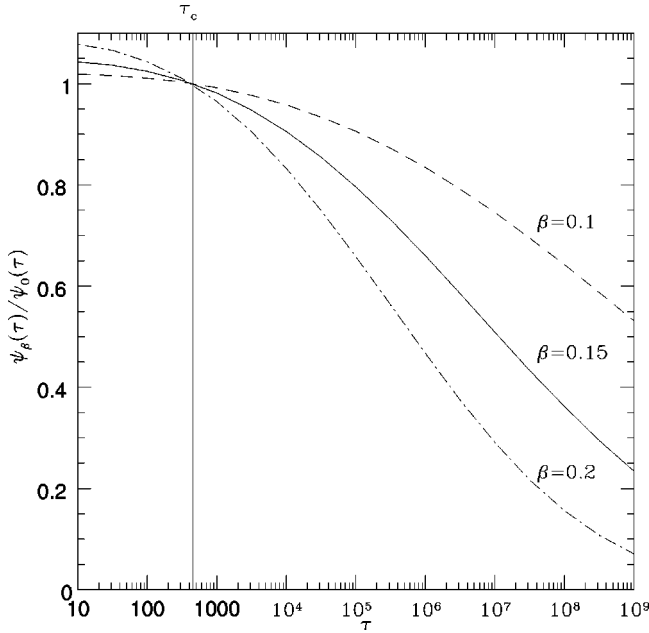


FIG. 2. Dimensionless PDF  $\psi(\tau)/W_m$  vs  $\tau$  for  $\eta = 2.1710^{-5}$  and different  $\beta$  values ( $\beta_0 = 0$ ,  $\beta_1 = 0.83$ ,  $\beta_2 = 0.95$ ,  $\beta_3 = 0.98$ ,  $\beta_4 = 0.99$ ). The solid curve presents the exponential PDF  $\psi(\tau)/W_m = \exp(-\tau)$ . Notice that the longed tailed behavior of  $\psi(\tau)$  found for  $\beta = 0$  switches into an exponential decay as  $\beta \rightarrow 1$ .

for  $e^\gamma \tau > 1$ . For a fixed short time this ratio is an increasing function of  $\beta$  while for a fixed long time this function is a decreasing function of  $\beta$ . The transition time  $\tau_c$  between these two behaviors is found, at  $\beta^2 \ll 1$ , to be

$$\tau_c = e^{-\gamma} \exp\left[\left(\frac{3}{\eta}\right)^{1/3}\right], \quad (30)$$

which is independent of  $\beta$ . It is easy to see that for  $\beta \ll 1$ ,  $\tau_c$  also satisfies the condition  $\psi_\beta(\tau_c)/\psi_0(\tau_c) = 1$ . All these features can be observed in Fig. 1 where we see (a) for  $\tau < \tau_c$  ( $\tau > \tau_c$ ) the ratio (29) increases (decreases) with  $\beta$ , (b) for all three choices of  $\beta$  the condition  $\psi_\beta(\tau_c)/\psi_0(\tau_c) = 1$  is fulfilled at the same point  $\tau_c$ .

At very large times the contribution to  $\psi(t)$  is from tunneling to very large distances. However, according to Eq. (23) for these large distances our analysis is not valid. Translating distances into times using Eq. (24), we find that our results are expected to be valid if

$$\tau < \exp\left(\frac{\Delta_0}{e|\mathbf{E}|R_d}\right). \quad (31)$$

For longer times the external field cannot be considered as a small perturbation.

We now discuss further the meaning of the approximations made above. Equation (24) considers the influence of the external field on the energy difference between sites (the  $\Delta/k_B T$  term) and neglected the temperature independent influence of the field on tunneling. Instead of Eq. (24) one may consider the tunneling rate

$$W(\mathbf{r}) = W_M \exp\left[-\left(\frac{r}{R_d}\right) - \left(\frac{e\mathbf{E} \cdot \mathbf{r}\mathbf{r}}{R_d U_0}\right) - \left(\frac{e\mathbf{E} \cdot \mathbf{r}}{k_B T}\right)\right], \quad (32)$$

where  $U_0$  is the characteristic energy barrier the charge carrier has to tunnel under (see details in [38]). The additional term  $e\mathbf{E} \cdot \mathbf{r}\mathbf{r}/R_d U_0$  appearing in Eq. (32) can be easily derived by calculating the action integral under a square potential barrier which is slightly tilted due to the external field. Equation (32) is valid for not too long distances  $r < U_0/e|\mathbf{E}|$ , the second term in the exponent being a perturbation to the first one. A calculation similar to that which has led to Eq. (27) shows that for  $\tilde{\epsilon} \equiv eR_d|\mathbf{E}|/U_0$  satisfying  $\tilde{\epsilon} \ll \beta$

$$-\ln\langle Q(t) \rangle = \frac{1}{3} \frac{\eta}{(1-\beta^2)^2} (\ln \tau)^3 + \eta \frac{\beta \tilde{\epsilon}}{(1-\beta^2)^2} \ln^4 \tau \dots \quad (33)$$

For weak fields the second term is a quadratic function of  $|\mathbf{E}|$ . This second term is smaller than the first one when

$$\beta \tilde{\epsilon} \ln \tau \ll 1. \quad (34)$$

Thus for not too long times our neglect of the field dependent corrections to the action integral is justified and Eq. (28) is a sound approximation.

#### IV. COLLISION MODEL

In the preceding section, the CTRW was used to investigate a system which exhibits subdiffusion. We shall now address a collision model which naturally leads to an enhanced diffusion with  $\delta > 1$  in Eq. (1) [24,25].

A classical test particle with a mass  $M$  moves in a one-dimensional space and interacts with bath particles of a mass  $m$ . At random, the test particle is elastically kicked by a bath particle. According to the energy and momentum conservation laws, the change of the test particle momentum due to the  $i$ th kick is described by the equation

$$p_i^+ = \mu_1 p_i^- + \mu_2 \tilde{p}_i, \quad (35)$$

where

$$\mu_1 = \frac{1-\epsilon}{1+\epsilon}, \quad \mu_2 = \frac{2}{1+\epsilon},$$

and  $\epsilon = m/M$ . Here  $p_i^-$  and  $p_i^+$  indicate the values of the momentum of the test particle just before and after the collision labeled  $i$  ( $i = 1, 2, \dots$ ).  $\tilde{p}_i$  is the momentum of the kicking bath particle. The coordinate of the test particle is not changed by the kick. Here we also assume that the duration of a collision event is much shorter than any other time appearing in the problem. An external uniform force  $F$  is supposed to act on the test particle. This force accelerates the test particle during the time intervals between collision events, according to Newton's laws of motion.

Our model assumes that the time intervals  $\tau_i$ , which elapse between the  $(i-1)$ th and  $i$ th collision events, are independent identically distributed random variables described by a yet unspecified PDF,  $q(\tau)$ . This PDF is assumed to be independent of the mechanical state of the test particle, and does not change in the course of the system's evolution. The momenta of the kicking bath particles  $\tilde{p}_i$  are also independent identically distributed random variables; their statistical

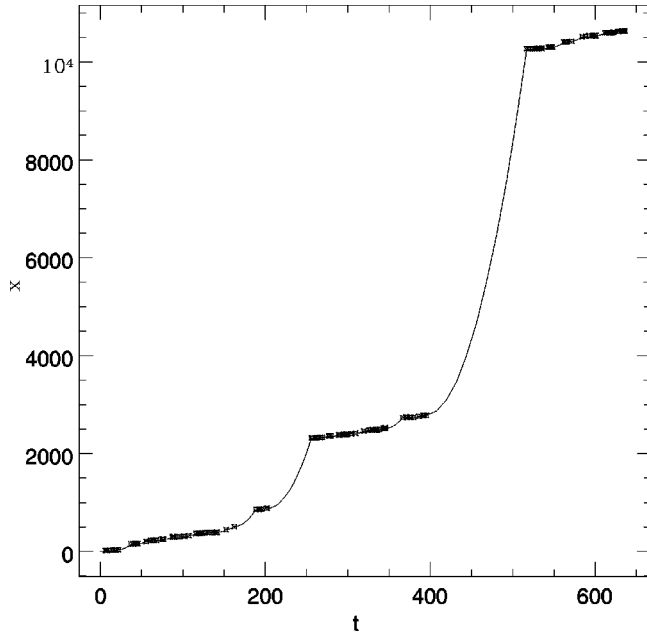


FIG. 3. Drift of a test particle which encountered 200 collisions. Dimensionless units with  $\epsilon=1$ ,  $F=1$ ,  $mk_B T=1$ , and an acceleration  $F/M=1$  are used. The dots denote the collision events. Notice the long time interval  $400 < t < 520$  in which no collision takes place; this time interval is roughly 20% of the observation time.

properties are determined by the Maxwell PDF  $f_m(\vec{p}_i)$  with a vanishing mean and a variance  $\langle \vec{p}_i^2 \rangle = mk_B T$ .

In the absence of an external force, with the exponential PDF,  $q(\tau)$ , and in the strong collision limit ( $\epsilon=1$ ) this model becomes the well known Drude model. The case of long tailed PDFs,  $q(\tau)$  and  $F=0$ , was investigated in our papers [24,25]. This leads to an enhanced diffusion with  $\delta > 1$  in Eq. (1). Here the version of this model considered is when the test particle is driven by an external space and time independent force  $F$ .

Figures 3–5 show trajectories of the test particle, when for long times

$$q(\tau) \sim \frac{2}{\pi} \tau^{-2}. \quad (36)$$

For this PDF all integer moments diverge and we are dealing with a situation very different from the classical Drude model. Appendix C describes an algorithm producing time intervals whose PDF decays algebraically with time, Eq. (36) being an example. Long time intervals in which no collision event takes place are shown in Figs. 3–5, a characteristic feature of the stochastic process. In Fig. 3 we see a drift of the test particle caused by the external force  $F$  in the strong collision limit  $\epsilon=1$ . One can observe that during the collisionless time intervals (e.g.,  $400 < t < 520$ ) the particle coordinate  $x$  increases quadratically with time as expected for an accelerating test particle. In time intervals in which many collisions have occurred the drift seems to increase linearly with time. As we shall show here the averaged drift generally follows  $\langle x(t) \rangle_F \sim t^{\delta_F}$  with  $1 \leq \delta_F \leq 2$ , meaning that the mean displacement behavior is intermediate between the quadratic and linear laws.

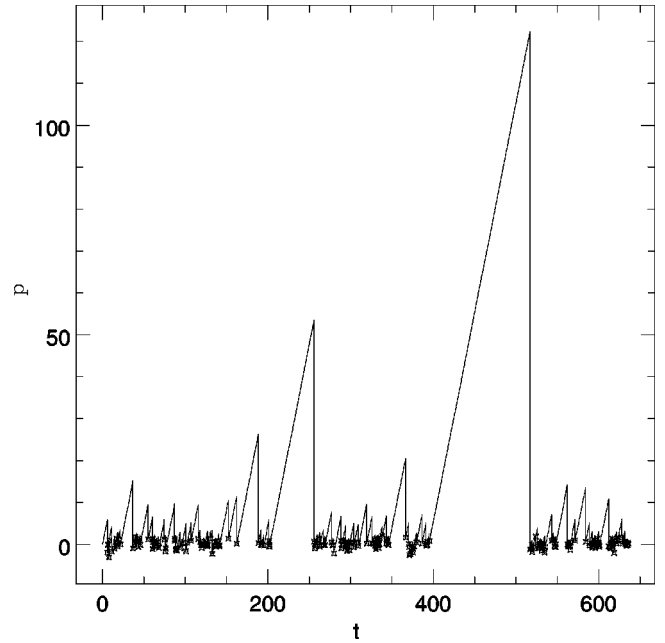


FIG. 4. Momentum of the test particle vs time for the same realization as in Fig. 1. Notice that in the time interval  $400 < t < 520$  the momentum strongly exceeds its thermal momentum which is set to be unity.

Figure 4 shows the momentum of the test particle vs time for  $\epsilon=1$ . One can see that the particle gains a momentum far beyond its thermal momentum  $p_{\text{th}} = \sqrt{mk_B T}$  which corresponds to  $p_{\text{th}}=1$  in the dimensionless units we use here. Notice that this deviation occurs only during the long collisionless time intervals, hence the departure from a close to

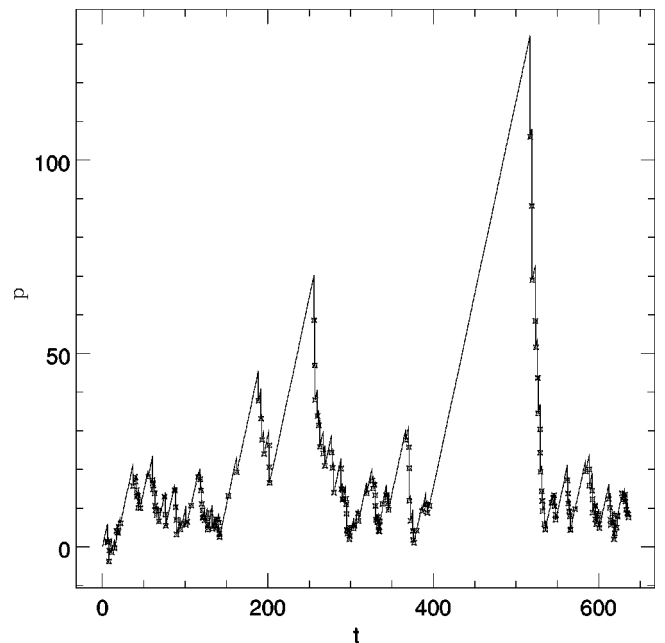


FIG. 5. Momentum of the test particle vs time in the weak collision limit  $\epsilon=0.1$ . All other parameters are identical to those used in Fig. 4. Notice that roughly ten collisions are needed to relax the test particle momentum from its maximal value, gained during the long collisionless time interval ( $400 < t < 520$ ). This should be compared to the single collision needed for the case  $\epsilon=1$ , Fig. 4.

equilibrium state would not have occurred had the collisions been distributed in a uniform way [e.g., had  $q(\tau)$  been an exponential function]. Figure 5 exhibits the same realization as in Fig. 4 but for the weak collision limit,  $\epsilon=0.1$ . We see that the relaxation of momentum occurs only after several collision events instead of a single collision for the strong collision case, Fig. 4.

### A. Definitions

The following definitions and mathematical tools will be used below. The sample space consists of (1) a non-negative integer  $s$  which is the number of collision events which occur during the time  $t$ . (2) For each  $s$  there exists a set of  $s+1$  real time intervals  $\tau_i$  ( $1 \leq i \leq s+1$ ;  $0 < \tau_i < \infty$ )

$$\{\tau_1, \tau_2, \dots, \tau_i, \dots, \tau_{s+1}\}.$$

The time interval  $\tau_1$  is the time elapsing between the start of observation ( $t=0$ ) and the first collision event. It is called the first waiting time.  $\tau_i$  ( $i \neq 1, i \neq s+1$ ) are time intervals between collision events called waiting times.  $\tau_{s+1}$  is the time between the  $s$ th collision (i.e., the last collision in the sequence) and the time of observation  $t$ . (3) For each  $s$  there exist a set of  $s$  real momenta  $\tilde{p}_i$  ( $-\infty < \tilde{p}_i < \infty$ ),

$$\{\tilde{p}_1, \tilde{p}_2, \dots, \tilde{p}_i, \dots, \tilde{p}_s\}$$

of the kicking bath particles.

The time intervals appearing in the problem are assumed to have the following properties.

(1) The waiting times (including the first one) and  $\tau_{s+1}$  are defined in the domain

$$\sum_{i=0}^s \tau_{i+1} = t. \quad (37)$$

(2) The first waiting time  $\tau_1$  is an independent random variable whose statistical properties are described by the PDF  $h(\tau_1)$ .

(3) The waiting times  $\tau_i$  ( $1 < i < s+1$ ) are assumed to be independent identically distributed random variables whose statistical properties are determined by the PDF  $q(\tau_i)$ .

(4) The probability that no collision event took place in the time  $\tau_{s+1}$  is, for  $s \geq 1$ ,

$$W(\tau_{s+1}) = 1 - \int_0^{\tau_{s+1}} q(t) dt. \quad (38)$$

(5) The probability that no collision event occurs in the interval  $(0, \tau_1)$  is

$$Z(\tau_1) = 1 - \int_0^{\tau_1} h(t) dt. \quad (39)$$

To calculate the average value of a physical quantity  $A(t, x, p)$  one has to consider the sequence

$$\{A_0(\tau_1), A_1(\tau_1, \tilde{p}_1, \tau_2), \dots, A_s(\tau_1, \tilde{p}_1, \dots, \tau_{s+1}), \dots\} \quad (40)$$

of functions over the state space. Here

$$A_s(\tau_1, \tilde{p}_1, \dots, \tau_{s+1})$$

is the quantity  $A$  calculated using the assumption that the particle has encountered a sequence of  $s$  collisions  $\{\tilde{p}_1, \dots, \tilde{p}_s\}$  with the bath particles and the time intervals are  $\{\tau_1, \dots, \tau_{s+1}\}$ . Then the average value is determined by the equation

$$\langle A(x, p) \rangle = \langle A_0 \rangle + \sum_{s=1}^{\infty} \langle A_s(x, p) \rangle \quad (41)$$

in which each term is the average value of  $A(x, p, t)$  calculated under the condition that  $s$  collision events have occurred. Then

$$\langle A_0 \rangle = A_0(t) Z(t) \quad (42)$$

and for  $s \geq 1$

$$\begin{aligned} \langle A_s(x, p) \rangle &= \int_{-\infty}^{\infty} \frac{dg}{2\pi} \int_0^{\infty} h(\tau_1) d\tau_1 \left[ \prod_{i=2}^s \int_0^{\infty} q(\tau_i) d\tau_i \right] \\ &\times \int_0^{\infty} W(\tau_{s+1}) d\tau_{s+1} \left[ \prod_{i=1}^s \int_{-\infty}^{\infty} f_m(\tilde{p}_i) d\tilde{p}_i \right] \\ &\times \exp \left\{ ig \left( \sum_{i=1}^{s+1} \tau_i - t \right) \right\} \\ &\times A_s \{ \dots, \tilde{p}_i, \tau_i, \dots, \tau_{s+1} \}. \end{aligned} \quad (43)$$

Here  $f_m(\tilde{p}_i)$  is the Maxwell-Gaussian PDF. The averaging procedure (41)–(43) implies summation over the number  $s$  of the collision events during the observation time  $t$ , as well as integrations over all PDFs of the time intervals between the kicks and over the momenta of the kicking particles. The representation

$$\delta \left( \sum_{i=1}^{s+1} \tau_i - t \right) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp \left\{ ig \left( \sum_{i=1}^{s+1} \tau_i - t \right) \right\} dg \quad (44)$$

of the  $\delta$  function in Eq. (43) ensures that the sum of all time intervals equals the observation time, Eq. (37).

The choice of the timing of the start of observation determines the function  $h(\tau_1)$ . This issue is discussed in the context of renewal theory by [39] and in the context of the CTRW in [6,40,41]. The SLM model predicts different conductivities depending on the choice of  $h(\tau)$  [40,41]. If a system is considered to be described by a constant rate  $1/\bar{\tau}$  of transitions (jumps, collisions, etc.), then for such a process, called an equilibrium renewal process, we have

$$h_{\text{eq}}(\tau_1) = \frac{1 - \int_0^{\tau_1} q(t) dt}{\bar{\tau}}. \quad (45)$$

An equilibrium renewal process can be defined only if at least the first moment of  $q(\tau)$  is finite. However, PDFs  $q(\tau)$  with diverging first moment are also to be considered, and for these an equilibrium renewal process cannot be defined. That is why we shall consider also the ordinary renewal process

$$h_{\text{or}}(\tau_1) = q(\tau_1) \quad (46)$$

for which the observation has started just after a collision event.

To choose the correct renewal process to model a physical process one needs detailed information on the way the system has been prepared. Thus, for example, for photo carrier experiments [18] (for which  $\delta < 1$ ) the ordinary renewal process is chosen since the electrons or holes are excited at  $t = 0$  when the process begins. If the process has been going on for a long time prior to the start of the observation of the process and if  $\bar{\tau}$  exists then it is an equilibrium process. For Gaussian transport systems and for the dc case these differences are of no practical importance, however, for the anomalous diffusion our collision model (for which  $\delta > 1$ ) indicates a strong sensitivity of the results to the way the system has been prepared initially. This may set limits on our abilities to predict the behavior of anomalous systems since the information on how the system has been prepared is not always sufficient to determine  $h(\tau)$ .

### B. Average momentum

We shall now calculate the mean momentum of the test particle moving under the action of an external force. The mean momentum allows one to find the mean displacement, which is our main goal in this section. According to the averaging procedure (41)–(43) we have to express first the momentum as a function of the time intervals between collision events  $\tau_i$  and momenta of the kicking bath particle  $\tilde{p}_i$ . This is done by using the stochastic map

$$p_{i+1}^+ = \mu_1(p_i^+ + Ma\tau_{i+1}) + \mu_2\tilde{p}_{i+1}, \quad (47)$$

which is obtained from Eq. (35) for the constant acceleration  $a \equiv F/M$ . This map relates between the momentum of the test particle just after the  $(i+1)$ th collision event and the momentum of the test particle just after the  $i$ th collision event. At a time  $t=0$ , the initial momentum of the test particle is  $p_0^+ = p_0$ .

The momentum of the test particle which encountered  $s$  collisions is

$$p_s = p_s^+ + Ma\tau_{s+1}, \quad (48)$$

where we have taken into account the driven motion of the test particle in the final time interval,  $\tau_{s+1}$ . Then using Eq. (47), Eq. (48) is rewritten as

$$p_s = p_s^i + p_s^f, \quad (49)$$

where the second term

$$p_s^f = \sum_{i=1}^s \mu_1^{s-i} (\mu_1 Ma\tau_i + \mu_2 \tilde{p}_i) + Ma\tau_{s+1} \quad (50)$$

does not depend on the initial momentum. This information is kept by the first term  $p_s^i = \mu_1^s p_0$ .

Now the Laplace transform of the average momentum

$$\langle p(t) \rangle = \langle p^f(t) \rangle + \langle p^i(t) \rangle \quad (51)$$

is calculated. Here according to Eq. (41)

$$\langle p^f(t) \rangle = \sum_{s=0}^{\infty} \langle p_s^f \rangle \quad \text{and} \quad \langle p^i(t) \rangle = \sum_{s=0}^{\infty} \langle p_s^i \rangle \quad (52)$$

and the meaning of the average  $\langle \rangle$  was explained in the preceding subsection. It is easy to see using the averaging procedure that all terms in Eq. (50) which depend on  $\tilde{p}_i$  will not contribute to the average since the bath particle momentum has zero mean.

Since we are interested in the averaged momentum,  $p_s^f$ , Eq. (50), is inserted in Eq. (43) with the identification  $A_s\{\dots, \tilde{p}_i, \dots\} = p_s^f\{\dots, \tilde{p}_i, \dots\}$ . Using the transformation  $ig = -u$ , carrying out the integrations over  $\tau_i$  and  $\tilde{p}_i$  as well as summing over  $i$  as appears in Eq. (50) one arrives after a straightforward calculation at the following results. For the realization in which the test particle experiences no collisions,  $s=0$ , one has

$$\langle p_{s=0}^f(t) \rangle = F \int_{-i\infty}^{i\infty} \frac{du}{2\pi i} \left\{ -\frac{d\hat{Z}(u)}{du} \right\} \exp(ut). \quad (53)$$

For  $s \geq 1$

$$\langle p_s^f(t) \rangle = \int_{-i\infty}^{i\infty} \frac{du}{2\pi i} \langle \hat{p}_s^f(u) \rangle \exp(ut), \quad (54)$$

with

$$\begin{aligned} \langle \hat{p}_s^f(u) \rangle = & F \left\{ \mu_1^s \left[ -\frac{d\hat{h}(u)}{du} \right] \hat{q}^{s-1}(u) \hat{W}(u) \right. \\ & + \mu_1 \left( \frac{1 - \mu_1^{s-1}}{1 - \mu_1} \right) \hat{h}(u) \left[ -\frac{d\hat{q}(u)}{du} \right] \hat{q}^{s-2}(u) \hat{W}(u) \\ & \left. + \hat{h}(u) \hat{q}^{s-1}(u) \left[ -\frac{d\hat{W}(u)}{du} \right] \right\}. \quad (55) \end{aligned}$$

Here the functions with hats denote the Laplace ( $t \rightarrow u$ ) transforms and the integration over  $u$  appearing in Eqs. (54) and (53) is identified as the inverse Laplace transformation (see [24] which elaborates on this point).



According to the formula (41) we sum over the number of collisions  $s$  and get the averaged momentum of the test particle in  $u$  space,

$$\begin{aligned} \langle \hat{p}^f(u) \rangle &= \sum_{s=0}^{\infty} \langle \hat{p}_s^f(u) \rangle \\ &= F \left\{ \left[ -\frac{d\hat{Z}(u)}{du} \right] + \left[ -\frac{d\hat{h}(u)}{du} \right] \hat{W}(u) \left( \frac{\mu_1}{1-\mu_1\hat{q}(u)} \right) \right. \\ &\quad \left. + \mu_1 \hat{h}(u) \left[ -\frac{d\hat{q}(u)}{du} \right] \hat{W}(u) \right. \\ &\quad \left. \times \frac{1}{[1-\hat{q}(u)][1-\mu_1\hat{q}(u)]} + \hat{h}(u) \frac{1}{1-\hat{q}(u)} \right. \\ &\quad \left. \times \left[ -\frac{d\hat{W}(u)}{du} \right] \right\}. \end{aligned} \quad (56)$$

The dependence of the mean momentum on its initial value  $p_0$  is calculated using the above procedure for  $p_s^i = \mu_1^s p_0$ . Equation (43) results in

$$\langle \hat{p}_{s=0}^i(u) \rangle = p_0 \hat{Z}(u) \quad (57)$$

for  $s=0$  and

$$\langle \hat{p}_{s \geq 1}^i(u) \rangle = p_0 \mu_1^s \hat{h}(u) \hat{q}^{s-1}(u) \hat{W}(u) \quad (58)$$

for  $s \geq 1$ . Summing over the number of collisions  $s$  [according to Eq. (41)] yields

$$\langle \hat{p}^i(u) \rangle = p_0 \left[ \hat{Z}(u) + \mu_1 \frac{\hat{h}(u) \hat{W}(u)}{1-\mu_1\hat{q}(u)} \right]. \quad (59)$$

Equations (56) and (59) give the mean of the test particle in  $u$  space,

$$\langle \hat{p}(u) \rangle = \langle \hat{p}^f(u) \rangle + \langle \hat{p}^i(u) \rangle. \quad (60)$$

This equation is quite general and can be used for various choices of the first waiting time PDF  $h(\tau_1)$ .

### C. Mean linear displacement

The mean linear displacement  $\langle x(t) \rangle_F$  is found using a relation between the Laplace transforms of the mean displacement and the averaged momentum

$$\langle \hat{x}(u) \rangle_F = \langle \hat{x}^i(u) \rangle_F + \langle \hat{x}^f(u) \rangle_F = \frac{\langle \hat{p}(u) \rangle}{uM} \quad (61)$$

in which Eq. (60) allows for a separation of the parts dependent and independent of the initial momentum  $p_0$ .

Considering the equilibrium renewal process, the Laplace transforms of Eqs. (38), (39), and (45) are found using the convolution theorem

$$\hat{W}(u) = \frac{1-\hat{q}(u)}{u}, \quad (62)$$

$$\hat{h}_{\text{eq}}(u) = \frac{1-\hat{q}(u)}{u\bar{\tau}}, \quad (63)$$

and

$$\hat{Z}_{\text{eq}}(u) = \frac{1}{u} \left\{ 1 - \left[ \frac{1-\hat{q}(u)}{u\bar{\tau}} \right] \right\}. \quad (64)$$

Inserting these three equations in Eq. (56) we have

$$\langle \hat{x}^f(u) \rangle_F = \frac{F}{Mu^3} \left\{ 1 - \frac{(1-\mu_1)}{u\bar{\tau}} \frac{[1-\hat{q}(u)]}{[1-\mu_1\hat{q}(u)]} \right\}, \quad (65)$$

then using Eqs. (59) and (62)–(64) we find

$$\langle \hat{x}^i(u) \rangle_F = \frac{p_0}{Mu^2} \left\{ 1 - \frac{(1-\mu_1)}{u\bar{\tau}} \frac{[1-\hat{q}(u)]}{[1-\mu_1\hat{q}(u)]} \right\}. \quad (66)$$

For the ordinary renewal process according to Eq. (46) we use

$$\hat{h}_{\text{or}}(u) = \hat{q}(u) \quad (67)$$

and

$$\hat{Z}_{\text{or}}(u) = \hat{W}(u). \quad (68)$$

Then using Eqs. (56) and (62) we find

$$\langle \hat{x}^f(u) \rangle_F = \frac{F}{Mu^3} \left\{ 1 + u \frac{d\hat{q}(u)}{du} \frac{1-\mu_1}{[1-\hat{q}(u)][1-\mu_1\hat{q}(u)]} \right\} \quad (69)$$

and

$$\langle \hat{x}^i(u) \rangle_F = \frac{p_0}{u^2 M} \left[ \frac{1-\hat{q}(u)}{1-\mu_1\hat{q}(u)} \right]. \quad (70)$$

A straightforward limiting case of these equations is when the bath particles are massless, namely,  $\epsilon=0$ . Then there is no interaction with the bath and the two results for the equilibrium renewal process equations (65), (66) and for the ordinary renewal process (69), (70) are identical. When  $\epsilon=0$ ,  $\mu_1=1$  and then for both processes we find the obvious result

$$\langle \hat{x}(u) \rangle_F = \langle \hat{x}^i(u) \rangle_F + \langle \hat{x}^f(u) \rangle_F = \frac{p_0}{u^2 M} + \frac{F}{Mu^3}, \quad (71)$$

which describes an accelerating particle with an initial momentum  $p_0$ . Generally for  $\epsilon \neq 0$  the two processes do not produce the same results even in the long time limit.

### D. Waiting time probability density function

Below the mean linear displacement is investigated for several special choices of the waiting time PDF,  $q(\tau)$ ,

$$\hat{q}(u) \approx \begin{cases} 1 - (Au)^\alpha + c_1(Au)^{2\alpha}, & 0 < \alpha < 1 \\ 1 + (Au)\ln(Au), & \alpha = 1 \\ 1 - \bar{\tau}u + c_\alpha(\bar{\tau}u)^\alpha + c_2(\bar{\tau}u)^2, & 1 < \alpha < 2 \\ 1 - \bar{\tau}u - c_1(\bar{\tau}u)^2\ln(\bar{\tau}u) + c_2(\bar{\tau}u)^2, & \alpha = 2 \\ 1 - \bar{\tau}u + c_1(\bar{\tau}u)^2 + c_\alpha(\bar{\tau}u)^\alpha, & 2 < \alpha < 3 \end{cases} \quad (72)$$

where  $c_\alpha$ ,  $c_1$ , and  $c_2$  are dimensionless constants. These PDFs have the property that for  $\alpha \leq 1$  all their moments diverge, for  $1 < \alpha \leq 2$  the first moment

$$\bar{\tau} = - \left. \frac{d\hat{q}(u)}{du} \right|_{u=0} \quad (73)$$

exists but all higher moments diverge, for  $2 < \alpha < 3$  the first and the second moment

$$\bar{\tau}^2 = \left. \frac{d^2\hat{q}(u)}{du^2} \right|_{u=0} = 2c_1\bar{\tau}^2 \quad (74)$$

exist but higher moments diverge. The use [1,2,5] of such PDFs in the framework of Lévy walks in a wide variety of systems is now well established and this gives us a motiva-

tion to investigate the driven motion using these types of functions with the collision model. Choosing PDFs with all converging moments leads according to the central limit theorem and law of large numbers to a Gaussian behavior; these PDFs are not discussed in this paper.

### E. Asymptotics of $\langle x(t) \rangle_F$

The long time (small  $u$ ) behavior of  $\langle x(t) \rangle_F$  for the equilibrium and ordinary renewal processes is now considered.

#### 1. Equilibrium renewal process

An equilibrium renewal process can be defined only if the average time  $\bar{\tau}$  between the collision events is finite. Therefore only PDFs with  $\alpha > 1$  are considered in this subsection. It will be shown later that the asymptotics of the mean linear displacement is determined only by the part which is independent of the initial momentum, so that

$$\langle x(t) \rangle_F \approx \langle x^f(t) \rangle_F \quad (75)$$

in the long time limit. The correction due to  $\langle x^i(t) \rangle_F$  depending on the initial momentum will be discussed in the next subsection.

Inserting the Laplace transformed PDF  $\hat{q}(u)$  (72) in Eq. (65), using the Tauberian theorem to  $u \rightarrow t$  transform the mean displacement, we find

$$\langle x(t) \rangle_F \approx \begin{cases} \frac{F}{M} \left\{ \frac{c_\alpha \bar{\tau}^2}{\Gamma(4-\alpha)} \left( \frac{t}{\bar{\tau}} \right)^{3-\alpha} + \left( c_2 + \frac{1-\epsilon}{2\epsilon} \right) \bar{\tau} t \right\}, & 1 < \alpha < 2 \\ \frac{F}{M} \left\{ (c_1 \bar{\tau} t) \ln \left( \frac{t}{\bar{\tau}} \right) + \left( c_2 + \frac{1-\epsilon}{2\epsilon} \right) \bar{\tau} t \right\}, & \alpha = 2 \\ \frac{F}{M} \left( c_1 + \frac{1-\epsilon}{2\epsilon} \right) \bar{\tau}, & 2 < \alpha < 3. \end{cases} \quad (76)$$

We notice that for  $1 < \alpha \leq 2$  the  $\langle x(t) \rangle_F$  increases faster than linearly with time (transport is enhanced) while when the first two moments of the times between collisions exist (i.e.,  $\alpha > 2$ )  $\langle x(t) \rangle_F$  increases linearly in time. A faster than linear transport is caused by the long time intervals when no collision event takes place. These time intervals can become longer for smaller values of the parameter  $\alpha$ . We also see from Eq. (76) that the test particle accelerates and gains energy in spite of the collisions with the bath particles.

For  $1 < \alpha \leq 2$  the leading term in Eq. (76) is independent of  $\epsilon$  characterizing the collision strength, for example, for very long times

$$\langle x(t) \rangle_F \approx \frac{F}{M} \frac{c_\alpha \bar{\tau}^2}{\Gamma(4-\alpha)} \left( \frac{t}{\bar{\tau}} \right)^{3-\alpha}, \quad 1 < \alpha < 2. \quad (77)$$

The fact that for long times  $\langle x(t) \rangle_F$  is independent of the mass ratio  $\epsilon$  is a unique feature indicating that the asymptotic

transport properties of the test particle are not controlled by the strength of the collisions but rather by long time intervals in which no collisions occur.

For classical collision models for which all the moments of the times between collision events exist and which are controlled by the mass ratio of the light bath particle to heavy Brownian particle (e.g., the Rayleigh piston [14]) the diffusion and the mobility coefficients always depend on the mass ratio. For our model this classical behavior corresponds to  $2 < \alpha$ , when the mobility, according to Eq. (76), is

$$\mu = \frac{1}{F} \frac{d\langle x(t) \rangle_F}{dt} = \frac{1}{M} \left( c_1 + \frac{1-\epsilon}{2\epsilon} \right) \bar{\tau} \quad (78)$$

and so unlike the enhanced case the transport coefficient  $\mu$  is indeed controlled by the mass ratio  $\epsilon$ , as well as by the (existing) first two moments of  $q(\tau)$ .

Notice also that  $\mu \geq 0$  since  $c_1 \geq 1/2$  and  $0 < \epsilon < \infty$ . To see why  $c_1 \geq 1/2$  we use Eqs. (73) and (74) to write  $\bar{\tau}^2 - \bar{\tau}^2 = (2c_1 - 1)\bar{\tau}^2 \geq 0$  and hence by definition  $c_1 \geq 1/2$ . The mobility decreases with  $\epsilon$  reaching its minimum (for constant  $c_1$ ) when  $\epsilon \rightarrow \infty$ . Then the test particle is surrounded by heavy bath particles and each collision just changes the particle velocity direction. When  $c_1 = 1/2$  and so  $\bar{\tau}^2 = \bar{\tau}^2$  meaning that there are no fluctuations of the time between collisions (i.e., the collisions follow one after the other with a constant time interval elapsing between collision events) and in addition when  $\epsilon \rightarrow \infty$  the particle bounces back and forward between two infinitely heavy walls and  $\mu = 0$  as expected.

Equation (76) for  $1 < \alpha \leq 2$  contains also linear in time corrections to the enhanced drift. The first, enhanced, term becomes larger than the second term only for a large enough time,

$$\left(\frac{t}{\bar{\tau}}\right) \gg \left[\frac{1 - \epsilon + 2\epsilon c_2}{2\epsilon} \frac{\Gamma(4 - \alpha)}{c_\alpha}\right]^{1/(2 - \alpha)}, \quad 1 < \alpha < 2 \quad (79)$$

$$\ln\left(\frac{t}{\bar{\tau}}\right) \gg \frac{1 - \epsilon + 2\epsilon c_2}{2\epsilon c_1}, \quad \alpha = 2.$$

Thus although the enhanced behavior  $\langle x(t) \rangle_F \sim t^{3 - \alpha}$  is independent of  $\epsilon$  as shown in Eq. (77), the parameter  $\epsilon$  controls the time of transition from the linear [ $\langle x(t) \rangle_F \sim t$ ] to the enhanced [ $\langle x(t) \rangle_F \sim t^{3 - \alpha}$ ] transport.

The limit  $\epsilon \ll 1$  can be considered as an example. In this case, corresponding to the Rayleigh limit of the model, the heavy test particle is kicked by light bath particles. The condition (79) reads now

$$\left(\frac{t}{\bar{\tau}}\right) \gg \left[\frac{\Gamma(4 - \alpha)}{2\epsilon c_\alpha}\right]^{1/(2 - \alpha)}, \quad 1 < \alpha < 2 \quad (80)$$

$$\ln\left(\frac{t}{\bar{\tau}}\right) \gg \frac{1}{2\epsilon c_1}, \quad \alpha = 2$$

and the transition time becomes long. It can become especially long when  $\alpha$  approaches the critical value 2, so that one should wait an extremely long time until the onset of the enhanced transport.

## 2. Ordinary renewal process

For the ordinary renewal process we use a procedure similar to that used for the equilibrium renewal process. The small  $u$  behavior of  $\langle \hat{x}^f(u) \rangle_F$ , Eq. (69), is found using  $\hat{q}(u)$  defined in Eq. (72). Then the transformation  $u \rightarrow t$  is invoked to find the mean displacement

$$\langle x(t) \rangle_F \approx \begin{cases} \frac{F}{2M} (1 - \alpha) t^2, & 0 < \alpha < 1 \\ \frac{F}{2M} \frac{t^2}{\ln(t/A)}, & \alpha = 1 \\ \frac{F}{M} \frac{c_\alpha \bar{\tau}^2 (\alpha - 1)}{\Gamma(4 - \alpha)} \left(\frac{t}{\bar{\tau}}\right)^{3 - \alpha}, & 1 < \alpha < 2. \end{cases} \quad (81)$$

Again we see that the asymptotic result is independent of  $\epsilon$ . For  $\alpha \geq 2$  the result coincides with that for the equilibrium renewal process (76). The behavior of  $\langle x(t) \rangle_F \sim t^2$  for  $0 < \alpha < 1$  can be understood by noticing that for this process, there exist long time intervals (of the order of the observation time  $t$ ) during which no collisions take place. During this interval according to Newton's law of motion  $x \sim t^2$  and the quadratic behavior of  $\langle x(t) \rangle_F$  is found. Comparing Eq. (81) with Eq. (76) for  $1 < \alpha < 2$  we see that the mean linear displacement  $\langle x(t) \rangle_F$  behaves differently for the equilibrium and ordinary renewal processes (the prefactors are nonidentical). This implies that the long time behavior of  $\langle x(t) \rangle_F$  is sensitive to the statistical properties of the first time interval in the sequence.

## 3. Dependence of $\langle x(t) \rangle_F$ on $p_0$

Now the dependence of the mean linear displacement on the initial velocity  $p_0/M$  of the test particle is discussed. For the equilibrium renewal process Eqs. (66) and (72) yield the long time behavior

$$\langle x^i(t) \rangle_F \approx \begin{cases} \frac{p_0}{M} \frac{c_\alpha \bar{\tau}}{\Gamma(3 - \alpha)} \left(\frac{t}{\bar{\tau}}\right)^{2 - \alpha}, & 1 < \alpha < 2 \\ \frac{p_0}{M} c_1 \bar{\tau} \ln\left(\frac{t}{\bar{\tau}}\right), & \alpha = 2 \\ \frac{p_0}{M} \left(c_1 + \frac{1 - \epsilon}{2\epsilon}\right) \bar{\tau}, & 2 < \alpha < 3. \end{cases} \quad (82)$$

For  $2 < \alpha < 3$ , the initial velocity  $p_0/M$  leads only to a small (time independent) displacement of the test particle whereas for  $1 < \alpha \leq 2$  the initial condition results in an averaged displacement which increases with time.

The strong influence of the initial condition  $p_0$  on  $\langle x(t) \rangle_F$  for  $\alpha \leq 2$  is due to samples where no collision event has taken place during the time of evolution  $t$ . To see this we rewrite Eq. (66) as

$$\langle \hat{x}^i(u) \rangle_F = \frac{p_0 \hat{Z}_{\text{eq}}(u)}{uM} + \frac{p_0 \mu_1}{Mu^3 \bar{\tau}} \frac{[1 - \hat{q}(u)]^2}{[1 - \mu_1 \hat{q}(u)]}, \quad (83)$$

where  $\hat{Z}_{\text{eq}}(u)$  is defined in Eq. (64). The first term in the right hand side describes processes for which no collision event takes place, namely,  $s = 0$ , in our averaging procedure [this term originates from Eq. (57)]. For the long tailed PDFs we are considering here the probability that no collision events occur in the time interval  $(0, t)$  [i.e.,  $Z(t)$ , Eq. (39)]

decays as a power law to zero. Inserting  $\hat{q}(u)$  for  $\alpha \leq 2$  in Eq. (83) the small  $u$  behavior of the mean displacement is easily shown to be

$$\langle \hat{x}^i(u) \rangle_F \approx \frac{p_0 \hat{Z}_{\text{eq}}(u)}{uM}.$$

Since the probability that no collision event takes place in the interval  $(0, t)$  is independent of the mass ratio  $\epsilon$  so is  $\langle x^i(t) \rangle_F$ . Needless to say, for classical Gaussian diffusion processes the realization for which no collision event takes place is negligible for long times.

For the ordinary renewal process Eqs. (70) and (72) yield

$$\langle x^i(t) \rangle_F \approx \begin{cases} \frac{p_0}{M} \frac{1+\epsilon}{2\epsilon} A \left( \frac{t}{A} \right)^{1-\alpha}, & 0 < \alpha < 1 \\ \frac{p_0}{M} \frac{1+\epsilon}{2\epsilon} A \ln \left( \frac{t}{A} \right), & \alpha = 1 \\ \frac{p_0}{M} \frac{1+\epsilon}{\bar{\tau} - 2\epsilon}, & 1 < \alpha < 3 \end{cases} \quad (84)$$

and if we compare this equation with Eq. (82), we see again that the two processes give different results due to the different statistical properties of the first time interval in the sequence.

Both for the equilibrium and ordinary renewal processes, the terms depending on the initial momentum  $p_0$  of the test particle equations (82) and (84), however large, are for long enough times smaller than the leading term of  $\langle x^i(t) \rangle_F$  in Eqs. (76) and (81). Nevertheless, Eqs. (82) and (84) indicate that the initial condition  $p_0$  decays slowly for anomalous transport. Its influence on  $\langle x^i(t) \rangle$  increases with time as a power law instead of the more normal situation where the initial condition  $p_0$  is responsible for long times only for a time independent shift of  $\langle x^i(t) \rangle$ .

### F. Generalized Einstein relation

We compare between the results derived here for the equilibrium (76) and ordinary (81) renewal processes and our results for the mean square displacement for the case  $F=0$ , published previously [25]. The Einstein relation is valid for this model both for the equilibrium and ordinary processes. This means that when we insert our results found in [25] in Eq. (11) and assume that the parameters  $\alpha, \tau, \dots$  are identical for both the force free process and the force driven process we find indeed  $R(t) = 1$ . Our results show that the Einstein relation holds for  $F \rightarrow 0$  even for nonstationary processes when there is no microscopic time scale.

We emphasize that if we compare between the two different processes, the ordinary and the equilibrium, the Einstein relation does not hold even when  $F \rightarrow 0$ . Thus departures from the Einstein relation (in the limit  $F \rightarrow 0$ ) will be found for long times when initial conditions are usually assumed to be of no importance. Thus if we take (wrongly)  $\langle x^2(t) \rangle_0^{\text{eq}}$  of an equilibrium renewal process and  $\langle x(t) \rangle_F^{\text{ord}}$  of an ordinary process the ratio  $R(t)$  defined in Eq. (11) is

$$\lim_{F \rightarrow 0} R(t) = \lim_{F \rightarrow 0} \frac{(F/2k_B T) \langle x^2 \rangle_0^{\text{eq}}}{\langle x(t) \rangle_F^{\text{ord}}} = (1 - \alpha)^{-1}$$

for  $1 < \alpha < 2$  instead of  $\lim_{F \rightarrow 0} R(t) = 1$ .

A strong sensitivity to the initial preparation of the system sets limitations on the way we can use the Einstein relation. If we know  $\langle x^2(t) \rangle_0$  derived either theoretically or measured experimentally this information can be used to predict  $\langle x(t) \rangle_F$  in the limit  $F \rightarrow 0$  only if we know that the two processes are of the same type. However, all these differences between the processes are of no importance for long times for systems exhibiting classical Brownian motion.

For Gaussian transport systems a criterion for the weakness of the external field, when the linear response approximation holds, is that the average drift velocity  $\langle \dot{x} \rangle_F$  of the test particle is much smaller than the thermal velocity. For stronger fields nonlinear effects become important and the Einstein relation is not valid even approximately. Usually when  $\langle \dot{x} \rangle_F > \sqrt{k_B T M}$  new types of dissipation mechanisms must be taken into account (e.g., inelastic collisions). For our model the velocity of the test particle is time dependent and therefore for any given force the condition

$$\langle \dot{x}(t) \rangle_F \ll \sqrt{k_B T M} \quad (85)$$

will be satisfied only during a finite interval of time. As follows from Eqs. (76) and (81) the condition (85) means

$$F \ll \begin{cases} \frac{M \sqrt{k_B T M}}{(1 - \alpha)t}, & 0 < \alpha < 1 \\ \frac{M \sqrt{k_B T M} \Gamma(4 - \alpha) \left( \frac{\bar{\tau}}{t} \right)^{2 - \alpha}}{(3 - \alpha)c_\alpha \bar{\tau}}, & 1 < \alpha < 2. \end{cases} \quad (86)$$

For any finite force  $F$ , however small, this criterion is violated for long enough times and then the generalized Einstein relation is not expected to be valid.

### G. Calculation of $q(t)$

An example of how an external field can influence  $q(\tau)$  is discussed. It is done for models which are slightly different from ours but maintain its most important feature, namely, the longed tailed PDF  $q(\tau)$ . Two such models are (a) the ordered Lorentz gas [42–44] in which a particle is reflected by equally spaced static spherical obstacles, centered on a hypercubic lattice and (b) anomalous Knudsen diffusion [45] where the reflections are from a random fractal which mimics a disordered porous medium. In both cases one can analyze the anomalous diffusion in the following approach. First calculate the PDF of the path lengths  $r$  (i.e., the length of the trajectories along which the test particle moves freely); for an anomalous system one finds  $p(r) \sim r^{-(1+\alpha)}$  [e.g., for the two-dimensional Lorentz gas  $\alpha=2$  and hence  $\langle x^2(t) \rangle_0 \sim t \ln t$ ]. Since between the collision events the particle moves with a constant velocity ( $F=0$ ) one finds after a simple transformation the PDF of times between the collision events,  $q(\tau) \sim \tau^{-(1+\alpha)}$ . Once the PDF  $q(\tau)$  is known the diffusion characteristics can be analyzed using the Lévy walk approach (see, e.g., [27]).

To take into account the influence of the force on the PDF  $q(\tau)$  we shall consider the transformation rule

$$r(t, \theta) = \sqrt{\left[ v \cos(\theta) + \frac{at^2}{2} \right]^2 + v^2 \sin^2(\theta)}, \quad (87)$$

where  $v = |\vec{v}|$  is the speed of the test particle after a collision event and the angle of reflection  $\theta$  is a random variable. This transformation rule simply relates the displacement  $r$  of a test particle accelerating in two dimensions with the time of free flight  $\tau$ . We shall assume for simplicity that  $v$  is a non-random variable which means that each collision thermalizes the test particle velocity (the case when  $v$  is distributed according to a Maxwell PDF can be easily calculated as well). The PDF  $q(\tau)$  is given by

$$q(\tau) = \frac{1}{2\pi} \int_0^{2\pi} p(r(\tau, \theta)) \dot{r}(\tau, \theta) d\theta, \quad (88)$$

where we have assumed that  $\theta$  is distributed uniformly. For short times we have  $r = vt$ , exactly like the case  $a = 0$  and then if  $p(r) \sim 1/(Ar^{1+\alpha})$  we have

$$q(\tau) \sim \frac{1}{Av^\alpha} \tau^{-(1+\alpha)} \quad (89)$$

for long times we have from Eq. (87)

$$q(t) \sim \frac{4^{(1+\alpha)}}{Aa^\alpha} t^{-(1+2\alpha)}. \quad (90)$$

We see that when a field is switched on the exponent describing the algebraic decay switches from  $1 + \alpha$  for  $F = 0$  to  $1 + 2\alpha$  for  $F \neq 0$ . The transition time from short to long time limit is determined by the short range behavior of PDF  $p(r)$  as well as by the strength of the field, and is not discussed here.

We see that very much like  $\psi(\tau)$  which describes the subdiffusive behavior in the Scher-Lax-Montroll model the longed tailed PDF  $q(\tau)$  may also depend on the external force. For both cases when a force is applied the processes become faster and hence  $q(\tau)$  and  $\psi(\tau)$  decay faster to zero. Like the CTRW the dependence of the exponent describing the long time behavior of  $q(\tau)$  on the field implies that for finite fields and long times a violation of the Einstein relation is naturally expected.

## V. SUMMARY AND DISCUSSION

Nonequilibrium phenomena close to thermal equilibrium are often described by means of the linear response theory [15] relating the response functions to equilibrium canonical correlators. van Kampen [46] put forward a sharp objection to this theory, claiming that it has only a very short range of validity. This objection was answered by Kubo *et al.* [15]. The linear response theory for normal systems was used successfully to analyze many experimental results and hence (we believe) from an experimental or phenomenological point of view the van Kampen objection to the derivation of linear response has no practical use. However, using a non-Hamiltonian approach, we have shown that when the expo-

nent controlling the algebraic decay of  $\psi(\tau)$  or  $q(\tau)$  depends on the strength of the applied external field, large deviations from the Einstein relation are found for long enough times and finite forces. Thus the van Kampen objection may find its applications for systems exhibiting anomalous diffusion.

These large deviations under ordinary situations are not found when the transport is Gaussian. For Gaussian systems the central limit theorem and the law of large numbers guarantees that both  $\langle x(t) \rangle_F$  and  $\langle x^2(t) \rangle_0$  increase linearly with time and hence  $R(t)$  is time independent. For the anomalous cases these laws are not valid. Rather an exponent  $\delta_F$  characterizes the drift which is directly related to the exponent controlling the algebraic decay of  $\psi(t)$  or  $q(t)$ . Hence if the exponent  $\alpha_F$  depends on the strength of the field so does  $\delta_F$ . For this case large deviations from the Einstein relation are expected for finite fields and long times.

A field dependence of  $\delta_F$  is calculated for the SLM model. Within the framework of CTRW the influence of the external field on  $\psi(t)$  was calculated. It was shown that the escape times of electrons from deep traps are reduced when a field is switched on. Two different field dependent mechanisms influence the escape from the trap. The first is temperature dependent and is caused by the energy shift in the different sites the electron can tunnel to. The second is due to a reduction of the action and is more directly related to quantum mechanical tunneling. The time scales at which the non-linear effects are of importance are discussed. Generally, unlike Gaussian transport mechanisms, these effects become important, as time passes by.

It is interesting to mention that an even stronger type of sensitivity of  $\psi(t)$  to an external bias has already been demonstrated in [47] in the context of chaos theory. It was shown that under certain conditions, a walker following a deterministic iteration rule  $x_{n+1} = g(x_n)$  can be described by the CTRW, the function  $\psi(t)$  being derived from the properties of the map  $g(x)$ . For nonbiased maps  $\psi(t)$  decays according to a power law [27,48], while for any finite bias and for long enough times  $\psi(t)$  decays exponentially.

Our collision model shows that when the second moment of the time between collisions diverges, the mean linear displacement is enhanced. Unlike Gaussian transport for large  $t$ , the mean linear displacement is independent of the strength of the collisions, controlled by the mass ratio  $\epsilon$ . It is an important parameter controlling the transition time before which  $\langle x(t) \rangle_F$  increases linearly with time and after which its growth is enhanced. The transport is dominated by long time intervals in which no collision event takes place. During these time intervals the particle accelerates and gains a high velocity. The energy gain during long time intervals between collisions is larger than the thermal energy and, hence, the perturbation acting on the velocity distribution is not weak. This means that linear response theory may be invalid for long times and a strong deviation from the Einstein relation is expected even for weak fields. Unlike the classical models with  $\langle x(t) \rangle_F \sim t$ , for our enhanced case the criterion for the weakness of the field is determined by an inequality (86) depending on time.

*Note added in proof.* Recently a related experimental study of anomalous hole transport in poly(phenylene vinylene) was published [50].

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## APPENDIX A

Consider a model of symmetric random barriers situated on a lattice. The transition rates from site  $n$  to site  $n+1$  and vice versa are

$$W_{n \rightarrow n+1} = W_n e^{lF/2k_B T}, \quad W_{n+1 \rightarrow n} = W_n e^{-lF/2k_B T}. \quad (\text{A1})$$

For this choice detailed balance is satisfied since  $W_{n \rightarrow n+1}/W_{n+1 \rightarrow n} = e^{lF/k_B T}$  with  $l$  being the lattice spacing. In a similar way  $W_{n-1 \rightarrow n} = W_{n-1} e^{-lF/2k_B T}$ . The probability that a particle initially at a site  $n$  will jump to the neighbor  $n+1$  before it jumps to site  $n-1$  is

$$P(n \rightarrow n+1) = \frac{W_{n \rightarrow n+1}}{W_{n \rightarrow n+1} + W_{n \rightarrow n-1}} \quad (\text{A2})$$

and similarly the probability to jump from  $n$  to  $n-1$  is

$$P(n \rightarrow n-1) = \frac{W_{n \rightarrow n-1}}{W_{n \rightarrow n-1} + W_{n \rightarrow n+1}}. \quad (\text{A3})$$

Therefore the displacement per jump is

$$\begin{aligned} \langle \tilde{x} \rangle_F &= l(\langle P_{n \rightarrow n+1} \rangle - \langle P_{n \rightarrow n-1} \rangle) \\ &= l \left\langle \frac{W_n e^{lF/2k_B T} - W_{n-1} e^{-lF/2k_B T}}{W_n e^{lF/2k_B T} + W_{n-1} e^{-lF/2k_B T}} \right\rangle. \end{aligned} \quad (\text{A4})$$

The average  $\langle \rangle$  is over the random variables  $W_n$  and  $W_{n-1}$ . We assume that the system is isotropic and so the right hand side of Eq. (A4) is independent of  $n$ . If we assume that the disordered system can be replaced by an ordered one we have

$$\langle \tilde{x} \rangle_F = l \tanh\left(\frac{Fl}{2k_b T}\right). \quad (\text{A5})$$

The effective medium approach we used to derive Eq. (A5) is similar in spirit to the CTRW approach where a disordered system is replaced by an ordered one [a single  $\psi(t)$  describes the evolution and not a family of  $\psi(t)$ 's which are dependent on the hopping site]. More generally we can consider also the disordered case assuming

$$\frac{Fl}{2k_b T} \ll 1, \quad (\text{A6})$$

then linearizing Eq. (A4)

$$\langle \tilde{x} \rangle_F \approx \frac{Fl^2}{2k_b T}. \quad (\text{A7})$$

This result is independent of the type of disorder. We now insert Eq. (A7) together with the identity  $\langle \tilde{x}^2 \rangle_0 = l^2$  in Eq. (13) and verify that  $\lim_{F \rightarrow 0} R(t) = 1$ .

## APPENDIX B

Using Eqs. (20) and (24) one can find

$$\begin{aligned} \langle Q(t) \rangle &= \exp\left(-\frac{\eta}{2} \int_{-1}^1 d \cos \theta \right. \\ &\quad \left. \times \int_0^\infty dx x^2 \{1 - \exp[-\tau e^{-x(1-\beta \cos \theta)}]\} \right), \end{aligned} \quad (\text{B1})$$

with  $\cos \theta = \hat{r} \cdot \hat{E}$ . Changing the variable,  $y = x[1 - \beta \cos \theta]$ ,

$$\begin{aligned} \langle Q(t) \rangle &= \exp\left(-\frac{\eta}{2} \int_{-1}^1 d \cos \theta \frac{1}{[1 - \beta \cos \theta]} \right. \\ &\quad \left. \times \int_0^\infty dy y^2 \{1 - \exp[-\tau e^{-y}]\} \right). \end{aligned} \quad (\text{B2})$$

The integration over  $y$  was done in SL,

$$\langle Q(t) \rangle = \exp\left\{-\frac{\eta}{(1-\beta^2)^2} \frac{\tau}{3} \left[ -\frac{d^3}{da^3} \tau^{-a} \gamma(a, \tau) \right] \Big|_{a=1} \right\}, \quad (\text{B3})$$

where  $\gamma(a, \tau)$  is the incomplete  $\Gamma$  function.

From Eq. (B3) we see that the renormalization rule (17) is valid for all times  $t$ . The asymptotic behavior of  $Q(\tau)$  derived from Eq. (B3) is [see Eq. (21) in Ref. [17]]

$$\begin{aligned} \ln \langle Q(t) \rangle &= -\frac{1}{3} \frac{\eta}{(1-\beta^2)^2} \\ &\quad \times [(\ln e^\gamma \tau)^3 + 3\xi(2)(\ln e^\gamma \tau) + 2\xi(3)], \end{aligned} \quad (\text{B4})$$

with  $\gamma = 0.5772 \dots$ ,  $\xi(2) = 1.645 \dots$ , and  $\xi(3) = 1.202 \dots$ . Using Eqs. (B4) and (19) we get Eq. (28).

## APPENDIX C

Ways to generate random variables described by different types of PDFs (e.g., the exponential, the Lorentzian, and the Gaussian PDFs), can be found in [49]. Here we show how to generate random variables whose PDF follows the rule

$$q(\tau) \sim \tau^{-(1+z)} \quad (\text{C1})$$

for  $\tau \rightarrow \infty$  and  $0 < \tau < \infty$ . We have in mind cases where the exact behavior of the PDF for small  $\tau$  is irrelevant. Two main methods are usually used [49] to generate random variables: (a) an accept-reject method which is not efficient and (b) a transformation method. Here we shall give a simple transformation rule which generates a random variable described by a longed tailed PDF.

We use a random number generator which manufactures a random variable  $u$  which is distributed uniformly in the interval  $0 < u < 1$ . Then we define the transformation (for  $\xi > 0$ )

$$\tau = \left[ \tan\left(\frac{u\pi}{2}\right) \right]^\xi \quad (\text{C2})$$

and hence

$$q(\tau) = p(u) \left| \frac{du}{d\tau} \right| = \left( \frac{2}{\pi\xi} \right) \frac{\tau^{(1-\xi)/\xi}}{(1 + \tau^{2/\xi})} \quad (\text{C3})$$

and  $p(u)$  is the uniform PDF. For long times we have

$$q(\tau) \sim \left( \frac{2}{\pi\xi} \right) \tau^{-(1+1/\xi)} \quad (\text{C4})$$

and hence if we identify  $1/\xi = z$  our goal is accomplished.

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