Transition to Anomalous Relaxation: Localization in a Hierarchical Potential

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A model of classical relaxation in a one-dimensional potential with a hierarchical distribution of barriers is studied. The vanishing of an effective diffusion constant results in a transition of the low-lying states of the master equation from extended to localized, producing a transition from exponential to anomalous decay.

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Stretched exponential relaxation of the form $\exp[-(t/\tau)^\beta]$ has been observed in many experimental systems including glasses,[1,2] spin-glasses,[3] amorphous semiconductors,[4] charge-density waves,[5] and proteins.[6] Such relaxation is often equally well fitted by a sum of a few simple exponentials.[2] Thus one may question whether it is the true asymptotic behavior, or rather a convenient parametrization when the observed decay is dominated by a long intermediate time period characterized by several time constants. In fact, a crossover at very long times from stretched to simple exponential decay was observed in Ref. 5. Recently there has been much effort devoted to the construction of simple models of anomalous relaxation.[7-11] A common feature is the introduction of a distribution of microscopic time scales, often in a hierarchical fashion.[8-11] However, stretched exponentials result only for special choices of this distribution. A physical mechanism explaining how this distribution might vary, producing a transition from simple to stretched exponential behavior, is lacking. Simultaneously, there has been much work on the problem of diffusion in random systems with asymmetric hopping rates,[12,13] and the effects of localization.[13,14]

In this paper, these two approaches are connected to yield a simple physical mechanism for a transition to anomalous decay. I consider a model of relaxation in a one-dimensional harmonic potential with a self-similar distribution of barriers, specified by a parameter R. This model is motivated by the idea that relaxation in glassy materials may be qualitatively described in terms of the motion of the system in phase space on a free-energy surface with many local minima separated by barriers of all scales.[11,15] A transition is found which may be characterized in terms of the eigenstates of the master equation giving the dynamics. For $R > R_c$, the low-lying states are extended on the equilibrium length scale about the global minimum. An effective diffusion constant may be defined and relaxation is simple exponential for all time. For $R < R_c$, the low-lying states are localized at the most difficult barriers to cross. Relaxation is given by a sum of exponentials which dominate the decay over a long intermediate time. This sum may be approximated by a stretched exponential.

The model I consider is illustrated in Fig. 1. The state of the system sits on integer sites x in a one-dimensional potential $E(x) = \frac{1}{2}\kappa x^2$. To move one site to the left or right, the particle must hop a barrier; these are arranged in a hierarchical pattern. The dynamics is given by a master equation,

$$dP(x)/dt = W_{x+1,x}P(x+1) + W_{x-1,x}P(x-1) - (W_{x,x+1} + W_{x,x-1})P(x) \equiv -\sum_{x'} M(x,x')P(x'), \tag{1}$$

$P(x)$ is the probability to be at x, $W_{x,x\pm 1}$ is the rate to hop from x to $x \pm 1$, and $M(x,x')$ is the master-equation matrix. I choose the rates

$$\begin{aligned} W_{x,x+1} &= R^{n(x)} e^{[E(x)-E(x+1)]/2}, \\ W_{x+1,x} &= W_{x,x+1} e^{[E(x+1)-E(x)]}. \end{aligned} \tag{2}$$

$R^{n(x)}$ represents the barrier between sites x and $x+1$. $n(x) = n$ if $(2x+1)\mathrm{mod}(3^l) = 0$ for all $l \le n$. If we write $R^n = \exp[-n\Delta_0/T]$ in terms of a free-energy barrier $n\Delta_0$, as T varies from 0 to ∞, R varies from 0 to 1. The exponential factors in (2) give the asymmetry in the rates due to the potential $E(x)$. The choice is uniquely determined[13] by detailed balance and the requirement that for $R = 1$ (equal barriers), Eq. (1) reduce to a discretization

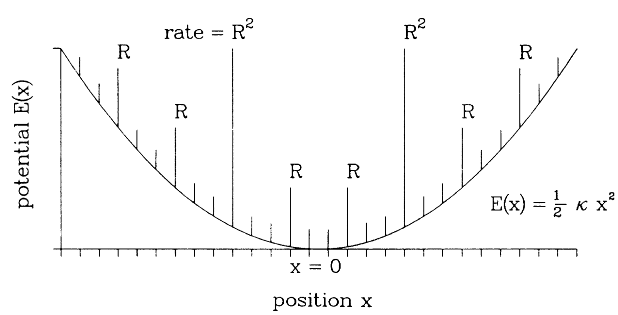

FIG. 1. Hierarchical barriers in a harmonic potential $E(x)$. The state of the system sits on integer sites x. To move right or left it must hop a barrier. These are arranged in a trifurcating hierarchical fashion, labeled by the rate $W = R^n$ to hop when $\kappa = 0$ [see Eq. (2)]. Unlabeled barriers have rate $W = 1$.

of the familiar diffusive dynamics in a potential, $dx/dt = -dE/dx +$thermal noise (temperature is absorbed in coupling κ).

Much work has been devoted to the case $\kappa = 0$; the potential is flat and hopping rates symmetric.[9,10] For any one-dimensional symmetric problem, the diffusion constant D for a system of size N is fixed by the rates,[16] $1/D(N) = (1/N)\sum_x 1/W_{x,x+1}$. For the rates (2) (with $\kappa = 0$), the sum as $N \to \infty$ gives a transition at $R_c = \frac{1}{3}$:

$$D(N \to \infty, R) = \tfrac{3}{2}(1 - 1/3R) \quad \text{for } R > R_c = \tfrac{1}{3}, \tag{3a}$$

$$D(N \to \infty, R) \sim N^{1+\ln R/\ln 3} \to 0 \quad \text{for } R < R_c. \tag{3b}$$

For $R > R_c$, asymptotic motion is diffusive. For $R < R_c$, $D \to 0$; a scaling argument,[10] $\langle x^2 \rangle \sim D(x)t$, gives asymptotic algebraic subdiffusive motion.

For $\kappa > 0$, we are interested in how the barriers effect the decay to $x = 0$. Making a standard transformation,[17]

$$\psi(x) = e^{E(x)/2} P(x),$$
$$\tilde{M}(x,x') = e^{E(x)/2} M(x,x') e^{-E(x')/2}, \tag{4}$$

we have $d\psi/dt = -\tilde{M}\psi$, where $\tilde{M}$ is symmetric. Its eigenvectors ψ_i, with eigenvalues λ_i, form a complete orthonormal basis, in terms of which the time evolution of the system is

$$P(x,t) = \textstyle\sum_i e^{-E(x)/2} \psi_i(x) \sum_{x'} \psi_i^*(x') P(x',0) e^{E(x')/2} e^{-\lambda_i t}. \tag{5}$$

For harmonic $E(x)$, and $R = 1$, the eigenvalues λ_i and eigenvectors ψ_i are those of the quantum harmonic oscillator with potential $V(x) = \frac{1}{2}\kappa[E(x) - 1]$.

The asymptotic behavior of the system is determined by the smallest nonzero eigenvalue λ_1. For the rates (2) I have numerically computed[18] λ_1 as a function of R and κ, for sizes $N = 3^6$ to 3^8. As N increased, λ_1 reached a limiting nonzero value. Thus the true asymptotic decay is exponential for all R (however, we will soon see that the observed decay over long intermediate times may be quite different). Results for this asymptotic time constant $\tau \equiv 1/\lambda_1$ are plotted in Fig. 2 versus κ, for several values of R. A least-squares fit (solid lines) by the form

$$\tau \sim B(R)\kappa^{-z(R)} \quad \text{as } \kappa \to 0 \tag{6}$$

gives excellent agreement, determining exponent $z(R)$ and scaling amplitude $B(R)$, shown in Figs. 3 and 4, respectively. In Fig. 3 a transition is clearly seen at $R_c = \frac{1}{3}$. For $R > R_c$, $z(R) = 1$. For $R < R_c$ I find empirically $z(R) = -\ln R/\ln 3$.

One can seek to explain the behavior of $z(R)$ by a scaling argument. Assume that the only effect of the barriers on relaxation in the potential is to provide an effective diffusion constant. Then $\tau = 1/D(L(\kappa))\kappa$ with $D(L)$ as in Eq. (3). $L(\kappa)$ is the effective size of the system sampled as the particle decays to $x = 0$. As $L(\kappa \to 0) \to \infty$, for $R > R_c$ Eq. (3a) gives $D \to$ const so that $z(R) = 1$, as for equal barriers. For $R < R_c$, however, Eq. (3b) gives $D(L \to \infty) \to 0$ as a power of $1/L$. So if we know how L scales with κ, we can compute the anomalous value of $z(R) > 1$. The dependence $L(\kappa)$ required to agree with the empirical result above is $L \sim 1/\kappa$. This is to be compared with the naive guess, the equilibrium length $L_{\rm eq} \sim 1/\sqrt{\kappa}$.

One can explain the above results for exponent $z(R)$, scaling amplitude $B(R)$, and length $L(\kappa)$ by a simple

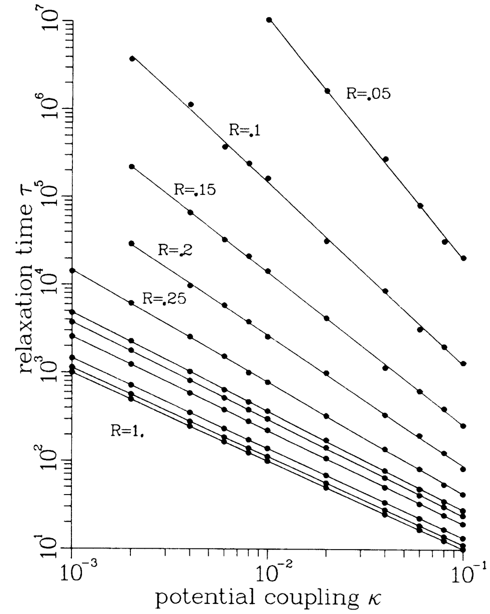

FIG. 2. Asymptotic relaxation time τ vs κ for several values of barrier parameter R. The curves between $R = 1$ and $R = 0.25$ are for $R = 0.8$, 0.6, 0.4, $R_c = \frac{1}{3}$, and 0.3. Dots are numerically computed. Solid lines are least-squares fits by the form $\tau = B(R)\kappa^{-z(R)}$.

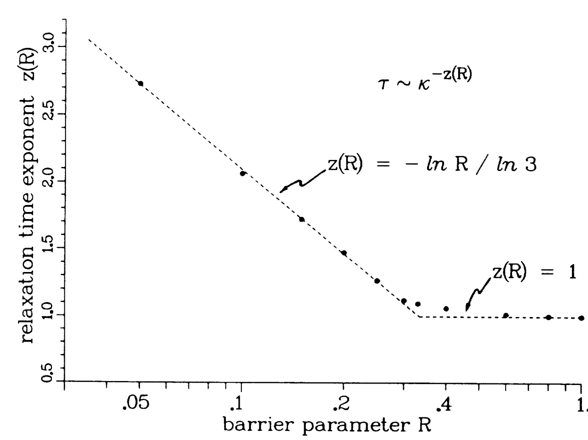

FIG. 3. Exponent $z(R)$ vs R. Dots are from least-squares fits to the data of Fig. 2. Dashed line is prediction, $z(R) = 1$ for $R > R_c = \frac{1}{3}$, $z(R) = -\ln R/\ln 3$ for $R < R_c = \frac{1}{3}$.

Ansatz. For $R > R_c$, the asymptotic decay is governed by the combined effect of all barriers within a region $\sim L_{\rm eq}$ of the global minimum, and given in terms of an effective diffusion constant. But for $R < R_c$, the asymptotic decay is determined by the single most difficult barrier for the system to cross in moving from large x to $x=0$. That is,

$$\tau = \max_{x>0}\,[1/W_{x+1,x}] \quad \text{for } R < R_c. \tag{7}$$

Performing the maximization in (7) one finds

$$\tau = \kappa^{\ln R/\ln 3} e^{-\kappa/4} \exp\{(-\ln R/\ln 3)[\ln(-4\ln R/\ln 3)-1]\} \tag{8}$$

due to the barrier at position

$$x_0 = -\frac{2}{\kappa}\frac{\ln R}{\ln 3}. \tag{9}$$

Thus, for $R < R_c$, one has the desired length $x_0 = L \sim 1/\kappa$, and the exponent $z(R) = -\ln R/\ln 3$. In Fig. 4 the scaling amplitude from Eq. (8) for $R < R_c$ (dashed line) is compared with the numerical results obtained from the fits in Fig. 1. For $R > R_c$ the prediction $B(R) = 1/D(2L_{\rm eq}, R)$ is also shown. Agreement is excellent.

I have also numerically computed[19] the eigenvector $\psi_1(x)$ for the eigenvalue λ_1. I find that for $R > R_c$, as $\kappa \to 0$, ψ_1 approaches the form expected for the equal-barrier case, i.e., the wave function for the first excited state of the harmonic oscillator,

$$\psi_1(x) \sim x \exp[-E(x)/2] \sim x\psi_0(x),$$

where ψ_0 is the equilibrium eigenvector with $\lambda_0 = 0$. However, for $R < R_c$, as $\kappa \to 0$, $\psi_1(x)$ is localized in a sharp spike at the most difficult barriers at $\pm x_0$ given by Eq. (9). The other low-lying eigenstates consist of localized states at the second, third, etc. most difficult barriers to cross, until the barrier of interest lies within a distance $\sim L_{\rm eq}$ of the origin.

To see how the above eigenstate structure effects the relaxation of the system, consider the dynamic correlation function $\langle x(t)x(0)\rangle$. Using Eq. (5) one can derive the result

$$\langle x(t)x(0)\rangle = \textstyle\sum_i a_i e^{-\lambda_i t}, \qquad a_i = \left[\textstyle\sum_x x\psi_0(x)\psi_i(x)\right]^2. \tag{10}$$

Only the antisymmetric ψ_i give nonvanishing a_i. For $R > R_c$, as $\kappa \to 0$, since $\psi_1(x) \sim x\psi_0(x)$, orthogonality implies that only a_1 is nonzero. Decay is pure exponential for all times. $R < R_c$, however, as $\kappa \to 0$, the low-lying eigenstates are localized. $x\psi_0(x)$ is no longer an eigenvector, and so many if not all the a_i's are nonzero. The smaller the eigenvalue λ_i, the farther from $x=0$ is the barrier causing the localization; hence the smaller the coefficient a_i which measures its contribution to the correlation function. The result will be very long intermediate times during which decay is described by a sum of several exponentials. If we approximate $\psi_i(x)$ for the lowest-lying states by a δ function at the localizing barrier, the time it takes for the contribution from the second-smallest antisymmetric eigenstate to die out compared to λ_1 is

$$t \gg [E(x_0) - E(x_0/3) - \ln 9]/(\lambda_2 - \lambda_1) \sim \kappa^{-[1+z(R)]}.$$

Thus the time t to reach asymptotic behavior diverges faster as $\kappa \to 0$ than the asymptotic relaxation time τ itself. The intermediate period before asymptopia sets in dominates the decay of the correlation function. I have computed $\langle x(t)x(0)\rangle$ by direct numerical simulation of the master equation (1), for values of $R > R_c$ and $R < R_c$. The simulations verify the picture presented above, and for $R < R_c$ the observed decay is well fitted by the stretched exponential form. Details will be presented in a longer paper.

I have also investigated a variation of the model presented here, in which the barriers are placed in a randomly shuffled configuration (but still symmetric about $x=0$). Preliminary results indicate the same qualitative behavior. Although the above model is one dimensional, the physical idea that anomalous relaxation is associated with localization of states in deep wells far from equilibrium may extend to more general geometries. Diffusion in random potentials in more dimensions can be anomalously slow.[20] Relaxation in the multidimensional phase space of a many-degree-of-freedom system may be dom-

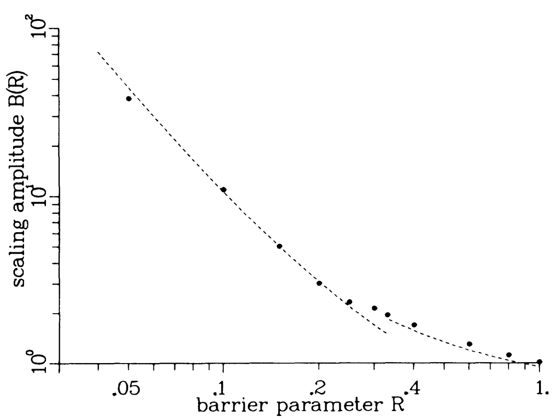

FIG. 4. Scaling amplitude $B(R)$ vs R. Dots are from least-squares fits to the data in Fig. 2. For $R < R_c = \frac{1}{3}$, the dashed line is the prediction from Eq. (8). For $R > R_c = \frac{1}{3}$, the dashed line is the prediction $B(R) = 1/D(2L_{\rm eq}, R)$ with $L_{\rm eq} = 1/\sqrt{\kappa}$ evaluated at $\kappa = 0.01$.

inated by effectively fewer-dimensional slow modes, such as the diffusion of domain walls. Thus one may hope that the picture of the transition to anomalous relaxation presented here may apply in many more general physical systems.

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