Bistable Flows Driven by Colored Noise

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1. Introduction

The escape from a metastable state has long been a subject of theoretical atten-
tion. Such processes play an important role in physics, chemistry, engineering
sciences and bio-physics in problems such as thermoionic emmission, transport in
Josephson junctions and super-ionic conductors, modeling of chemical reaction
rates and ligand migration in proteins. Bistable systems often resemble the
model of a Brownian motion in a potential field with two or perhaps more adja-
cent wells and barriers in between which prevent the particles from jumping too
often. This is the viewpoint put forward by KRAMERS /1/ whose original work in
this field represents a milestone. In his approach, the relevant nonlinear mo-
tion is treated explicitly whereas the interaction with the residual degrees of
freedom are represented by noise and frictional forces. Kramers treated the mo-
tion in a one-dimensional potential field and described the heat bath interaction
by white Gaussian noise which satisfies an Einstein relation. Generalizations of
Kramers'work to Brownian motion in a multi-dimensional potential field are due to
BRINKMAN /2/ , LANDAUER and SWANSON /3/ and LANGER /4/.

Often, the influence of a finite correlation time τ of the thermal noise $\xi(t)$
plays a minor role on the dynamics such that a Markovian approximation for the
heat bath interaction is well justified. On the other hand, there exist situations
for which a non-Markovian heat bath interaction becomes essential. A well-known
example is given by the motional narrowing in magnetic resonance /5/ or also the
refined treatment of Brownian motion in water /6/. Our focus will be on the ex-
tension of Kramers's approach to a more general heat bath interaction described
by colored thermal noise (non-Markovian Brownian motion) and memory - damping
in overdamped regimes /7,8,9,10,11/ and energy-diffusion controlled underdamped
regimes /12,13,14/. The memory effect on the thermal activation rates has recently
been observed experimentally in the relaxation dynamics of migration of ligands in
heme proteins /9,15/ and in chemical isomerization reactions /16,17/.

The problem of escape can also be broadened to a dynamics in driven systems,
which in general do not obey the condition of manifest detailed balance /18/ and
which in general have a drift field which cannot be derived from a potential field.
Nevertheless, such systems can exhibit competing states of local stability. Early
discussions of driven nonlinear bistable systems include those by STRATONOVICH /19/
and LANDAUER /20/ . Over the last years, the interest in nonlinear driven systems
has been growing steadily in the context of describing non-equilibrium relaxation,
non-equilibrium instabilities and turbulent (chaotic) behavior, etc. There are
recent, clear , and detailed reviews of this field /21,22,23,24/. As a proto-
type for this kind of system we consider here the non-Markovian dynamics in a
bistable symmetric double well which is driven by a stationary colored Gaussian
noise , or by a telegraphic noise (a two-state Markov process) of variable cor-
relation time τ .

2. Thermal Activation Rates in Systems with Memory

A. Overdamped Motion

Inherent in Kramers's treatment is the assumption of a clear-cut separation between the time scales of particle and heat-bath motion. GROTE and HYNES /7/ and HANGGI and MOJTABAI /8,9/ treated the non-Markovian escape problem associated with the barrier dynamics; i.e., for moderate and heavy damping the rate determining step is controlled by the diffusion at the top of the potential barrier. The thermal equilibrium motion for a Brownian particle of unit mass can then be linearized around the barrier top. For the one-dimensional motion in a potential field $U(x)$, one obtains with an expansion of $U(x)$ around the barrier top $x_b = 0$,

$$U(x) = U(0) - \tfrac{1}{2} \omega_b^2 x^2 + \dots \quad , \quad \omega_b > 0 , \quad (1)$$

and the linearized (uniform) memory damping $\phi(t)$ of the thermal non-Markovian dynamics near $x = x_b = 0$, the linear generalized Langevin equation

$$\ddot{x} = \omega_b^2 x - \int_0^t \phi(t-s) \, \dot{x}(s) \, ds + \xi(t) . \quad (2)$$

$\xi(t)$ is the stationary, non-white Gaussian thermal noise source of vanishing mean which obeys the fluctuation-dissipation theorem

$$\langle \xi(t) \, \xi(s) \rangle = k_B T \, \phi(t-s) . \quad (3)$$

The relations (2) and (3) consistently /25c/ describe the non-Markovian Gaussian thermal equilibrium process near $x = x_b$. Its conditional probability $p_t(x,\dot{x}|x_o,\dot{x}_o)$ prepared initially at time $t_o = 0$ satisfies the time-convolutionless (but not memory-less) non-Markovian master equation /8/

$$\dot{p}_t = [- \dot{x} \frac{\partial}{\partial x} - \bar{\omega}^2(t) \, x \frac{\partial}{\partial \dot{x}}] \, p_t + \bar{\gamma}(t) \frac{\partial}{\partial \dot{x}} (\dot{x} \, p_t)$$

$$+ k_B T \, \bar{\gamma}(t) \frac{\partial^2}{(\partial \dot{x})^2} \, p_t + \frac{k_B T}{\omega_b^2} [\bar{\omega}^2(t) - \omega_b^2] \frac{\partial^2}{\partial x \partial \dot{x}} \, p_t \quad (4a)$$

$$\bar{\gamma}(t) = - \dot{a}(t) / a(t) \quad ; \quad \bar{\omega}^2(t) = -b(t) / a(t) \quad (4b)$$

$$a(t) = \dot{\rho}(t) [1 + \omega_b^2 \int_o^t \rho(s) ds] - \omega_b^2 \rho^2(t) \quad (4c)$$

$$b(t) = \omega_b^2 [\rho(t) \ddot{\rho}(t) - \dot{\rho}^2(t)] . \quad (4d)$$

The correlation $\rho(t)$ is given by the inverse Laplace transform (L^{-1}), $Lf(t) = \hat{f}(z)$

$$\rho(t) = L^{-1} [1/ (z^2 - \omega_b^2 + z \hat{\phi}(z))] \quad , \quad \rho(t_o=0) = 0 . \quad (4e)$$

There are of course arbitrary many different coupling schemes modeling the heat bath interaction, which result within the linearized dynamics in a uniform memory function $\phi(t)$ obeying (3). In this context, it should be noted that keeping the full nonlinearity for the conservative drift motion while keeping the memory damping uniform is in general inconsistent with an initial thermal equilibrium preparation of the total system /22,25/. In other words, writing with a memory damping $\phi(t)$ and a noise $\xi(t)$ obeying (3)

$$\ddot{x} = - \partial U/\partial x - \int_o^t \phi(t-s) \, \dot{x}(s) \, ds + \xi(t) \quad (5)$$

the initial canonical probabilty p_o

$$p_0 = \overline{p} = Z^{-1} \exp - (\tfrac{1}{2} \dot{x}^2 + U(x)) / k_B T \tag{6}$$

does in general not stay invariant under time evolution; i.e., $p_t \neq \overline{p}$, $t > t_0$. As an example of a consistent Markovian modeling of the heat bath interaction we consider a coupling to a set of bath variables $(z_1,....,z_n)$ which is of the form /26/

$$\dot{x} = p$$

$$\dot{p} = - \partial U / \partial x + z_1$$

$$\dot{z}_1 = - c_1 p - \gamma_1 z_1 + z_2 + \xi_1(t)$$

$$\dot{z}_2 = - c_2 z_1 - \gamma_2 z_2 + z_3 + \xi_2(t)$$

$$\vdots$$

$$\dot{z}_n = - c_n z_{n-1} - \gamma_n z_n + \xi_n(t) \tag{7a}$$

where $\quad \gamma_i \geqslant 0$, $\quad c_i > 0$, $\quad i = 1,...,n$, $\quad \gamma_n > 0$, $\tag{7b}$

and $\quad < \xi_i(t)\xi_j(s) > = 2 \delta_{i,j} k_B T \gamma_i (\prod_{m=1}^{i} c_m) \delta(t-s)$. $\tag{7c}$

The corresponding multi-dimensional Fokker-Planck equation is readily seen to have the thermal equilibrium probability

$$\overline{p} = Z^{-1} \exp - [U(x) + \tfrac{1}{2} p^2 + \tfrac{1}{2} z_1^2 / c_1 + ... + \tfrac{1}{2} z_n^2 / (c_1 c_2 \cdots c_n)]/k_B T \tag{8}$$

with no correlations among the variables. On contracting the dynamics in (7) onto the relevant variables $(x,\dot{x})$ one recovers for an initial preparation scheme/25/ consistent with (8) the generalized nonlinear Langevin equation (GLE) in (5). The uniform memory damping is given by its Laplace transform as /26/

$$\hat{\phi}(z) = \frac{c_1}{z + \gamma_1 +} \; \frac{c_2}{z + \gamma_2 +} \; .... \; \frac{c_n}{z + \gamma_n} \; . \tag{9}$$

Due to (7b), all the continued fraction coefficients in (9) are positive (including zero). In particular, note that the example of a two-term exponentially decaying memory $\phi(t)$

$$\phi(-t) = \phi(t) = \exp - t + \exp - 2t \quad , \quad t > 0 \tag{10}$$

which has the Laplace transform (written as a continued fraction)

$$\hat{\phi}(z) = \frac{2}{z + {}^3/_2 +} \; \frac{-\tfrac{1}{4}}{z + {}^3/_2} \quad , \text{ i.e. } c_2 < 0 \; , \tag{11}$$

is not a possible memory function for the above special heat bath coupling (7). However, within the linearized non-Markovian dynamics near the barrier $x_b = 0$, (1-3), the memory damping in (10) is permitted fully.

Next, let us generate a non-equilibrium current J by injecting particles at the locally stable well, say x_0, and removing them at the adjacent well, say x'. For moderate and heavy damping, the particle density n_0 around x_0 is given by the thermal equilibrium probability inside the well around x_0 /7-10/. Then, the thermal activation rate r equals at low noise (without this assumption the problem

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of escape is not well defined anyhow) the ratio

$$r = J / n_0 \; . \tag{12}$$

Evaluating the constant non-equilibrium current J via the non-Markovian dynamics in (4) /8,9,10/ one obtains for the escape rate in the moderate and heavy damping regime /7-11/

$$r = \frac{\alpha \;\; \omega_0}{2\pi \;\; \omega_b} \; \exp - E_b/k_B T \;\; = \;\; \frac{\alpha}{\omega_b} \; r^{TST} \; . \tag{13}$$

Hereby, E_b denotes the barrier height and $\omega_0 = U''(x_0)$ is the angular frequency in the bottom of the potential well. r^{TST} indicates the familiar transition state theory result /27,28/. The effective frequency α /8,9,10/

$$\alpha = \lim_{t\to\infty} \; [\; (\tfrac{1}{4} \overline{\gamma}^2(t) + \overline{\omega}^2(t) \;)^{\frac{1}{2}} - \tfrac{1}{2} \overline{\gamma}(t) \;] \tag{14}$$

is determined solely by ω_b and the memory damping $\phi(t)$, see (4b - 4e). Assuming that $\rho(t)$ (or $\phi(t)$) admits a representation in form of a meromorphic function (including a slight generalization thereof /9/) it has been shown first in Ref./9/, and later in /10,11/, that α equals the largest, real and positive pole, $z = \alpha$ of $\hat{\rho}(z)$ in (4e). Clearly, this is equivalent with α *being the largest positive solution* of

$$\alpha = \frac{\omega_b^2}{\alpha + \hat{\phi}(\alpha)} \; . \tag{15}$$

(15) is known as the Grote - Hynes relation /7/. However, those authors did not originally specify α as being the largest positive solution among possibly several positive solutions of (15). Also note that a continued fraction representation of $\hat{\phi}(z)$ of the type in (9), with γ_i and c_i not necessarily all positive, is equivalent to a meromorphic function representation in the form of a [n-1/n] Padé approximant /29/.

For the memory damping

$$\phi(-t) = \phi(t) = [\; A/ (c + t^{1-\kappa}) \;] \; \exp - (\omega^2 t/\gamma_s) \; , \; 0 \leq \kappa \leq 1 \; , \; t>0, \tag{16}$$

which occurs in the modeling of overdamped CO or O_2 migration in myoglobin/9,15/, the behavior of the effective frequency α versus solvent damping γ_s is sketched in Fig.1. The explicit fits to experimental data can be found in Ref. 15. The form of the memory damping in (16) is due to DOSTER /15/. In his pac-man model of dynamic friction, the memory damping $\phi(t)$ is modeled by a correlation of local defect fluctuations ($\propto t^{\kappa - 1}$) and a statistically independent coupling to global protein-solvent fluctuations ($\propto \exp - \omega^2 t/ \gamma_s$).
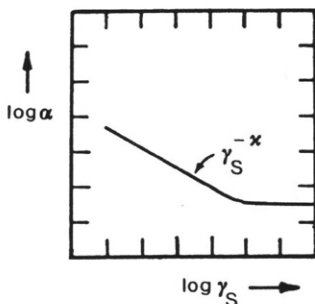

Fig. 1 Schematic sketch of the effective frequency α versus solvent damping γ_s for the memory damping in (16)

The arguments given above for a non-Markovian motion in a one-dimensional potential field $U(x)$ can be extended to a multi-dimensional potential field $U(\underset{\sim}{x})$ in which the transport occurs over sequential saddle points. The influence of the multi-dimensional potential field reduces then essentially to a pure phase-space factor. This factor equals the product of the frequencies in the locally stable potential well over the the product of stable frequencies at the saddle point. The multi-dimensional generalization of (13), valid for moderate and heavy damping, thus reads/9/

$$r = \frac{\alpha}{\omega_b} \; r_N^{TST} \quad , \tag{17}$$

where r_N^{TST} denotes the N-dimensional generalization of the transition state rate.

B. Underdamped Motion

The term "underdamped motion" refers to a situation for which the rate determining step is controlled by a slow particle diffusion up along the energy or action coordinate. At low damping, the motion is close to the conservative motion and escape up in energy becomes very difficult. Now, the population in the initial well can no longer be assumed to be in thermal equilibrium. If the memory damping is weak the appropriate physically relevant process is the energy diffusion. This was already realized by KRAMERS /1/, who obtained in the Markovian limit an escape rate proportional to the frequency-independent damping. In presence of memory damping, the energy diffusion can be modeled by an effective Fokker-Planck equation/12,13/ for the action variable J or energy variable E. Originally, this effective Fokker-Planck equation had been derived for an anharmonic oscillator by ZWANZIG /30/.

For extremely weak underdamping, the rate r can be identified with the inverse of the mean first passage time $\tau \, (E_b)$ to reach the absorbing top of the barrier /11,12,13/. This of course amounts to setting the nonequilibrium population density $n_o(E)$ at the barrier top equal to zero, $n_o(E_b) = 0$. A recent refinement of Kramers' original approach for the underdamped regime /31/ treats the uphill diffusion in energy just as Kramers did; but in addition allows in the range above the barrier for a simultaneous flow out of the well, across the barrier - thus implying $n_o(E_b) > 0$. The same idea can be generalized to the case of memory damping /14/. In the limit of a deep well one obtains for the thermal activation rate, valid in the underdamped regime /14/ ,

$$r = \left[\frac{(1 + 4/ \; D(E_b)\beta^2)^{\frac{1}{2}} - 1}{(1 + 4/ \; D(E_b)\beta^2)^{\frac{1}{2}} + 1} \right] \beta^2 \; D(E_b) \; r^{TST} \quad , \quad \beta = (k_b T)^{-1} \; . \tag{18}$$

In the Markovian limit we have ve $D(E_b) = J(E_b) \, \gamma \, k_b T$, and the result of (18) coincides with Ref. /31/. Moreover, on expanding the bracket in (18) in function of $D(E_B)$, the prefactor vanishes $\propto D(E_b) \, (1 - const.(D(E_b))^{\frac{1}{2}})$ /32/ which in the Markovian limit equals the behavior of the lowest non-zero eigenvalue /32,33/. Also, note that in the underdamped regime the rate in (18) incorporates via $D(E_b)$, or the action $J(E_b)$, information of the global shape of the potential well. For a *smooth* barrier region, $D(E_B)$ is approximately given by

$$D \, (E_b) = k_b T \, J \, (E_b) \, \hat{\phi} \, (z=0) \quad , \tag{19}$$

whereas for a *cusped-shaped* barrier (e.g.,a truncated harmonic oscillator potential) one has approximately

$$D \, (E_b) = k_b T \, J \, (E_b) \, \int_0^\infty \phi(t) \, \cos(\omega_0 t \, /2\pi) \, dt \, . \tag{20}$$

3. Non-Equilibrium Bistable Flow Driven by Colored Noise

As a prototype of a bistable flow driven by colored noise, which does not obey the fluctuation-dissipation theorem (3), we consider the set of Langevin equations

$$\dot{x} = ax - bx^3 + y \quad , \quad a > 0 \quad , \quad b > 0 \tag{21}$$

$$\dot{y} = -(1/\tau) y + \eta(t) . \tag{22}$$

$\eta(t)$ is a stationary Gaussian white noise of zero mean and correlation

$$< \eta(t)\, \eta(s) > = (2D/\tau^2) \; \delta\, (t-s) . \tag{23}$$

Upon integrating (22) from an initial time $t_0 = 0$, $y(0) = y_0$, one obtains by virtue of

$$< y_0 > = 0 \quad , \quad < y_0^2 > = D/\tau \tag{24}$$

a nonlinear Langevin equation driven by stationary colored noise $y(t)$

$$\dot{x} = ax - bx^3 + y(t) \tag{25a}$$

$$< y(t) > = 0 \quad , \quad < y(t)\, y(s) > = (D/\tau)\, \exp - |t-s| / \tau . \tag{25b}$$

Note that $y(t)$ is a *Gaussian only if* y_0 *has been prepared initially with a Gaussian consistent with* (24). Moreover, given at time $t_0 = 0$ the initial preparation $\rho_0(x,y)$, the variable $x(0)$ and the noise $y(0)$ are generally correlated

$$<x(0)\, y(0)> \neq 0 . \tag{26}$$

In the following we assume that the processes x_t and y_t are initially statistically independent (correlation-free initial preparation /25/). We also restrict the further discussion to the case of stationary Gaussian noise $y(t)$ obeying (25b). For a symmetric telegraphic noise $y(t)$ obeying (25b), the exact activation rates of bistable flows have been evaluated by HANGGI and RISEBOROUGH in Ref./34/. Those rates have been shown to exhibit an exponential enhancement with decreasing correlation time τ at fixed noise intensity D /34/. The exact dynamics and the rates of asymmetric telegraphic noise /35,36/ $y(t)$ obeying (25b), which contains as limits both a particular type of white shot noise and white Gaussian noise, has been the subject of another recent study /35/.

The joint process of (21) and (22) constitutes a two-dimensional Fokker-Planck process. However, it does not satisfy detailed balance /18,22/ and thus its stationary probability is not readily determined. For weak noise, one usually sets for the stationary probability $\overline{p}\,(x,y)$

$$\overline{p}\,(x,y) = a(x,y)\, \exp - (\, \psi_0\,(x,y)/D + 0\,(D)\,) . \tag{27}$$

Then, ψ_0 ($\neq$ const) obeys the first-order differential equation /37,38/

$$(\, ax - bx^3 + y\,)\, \partial\psi_0/\partial x - \tau^{-1} y\, \partial\psi_0/\partial y + \tau^{-2}(\partial\psi_0/\partial y)^2 = 0 \tag{28}$$

and $a(x,y)$ too, obeys a corresponding first-order differential equation /38/. With $p_x = \partial\psi_0/\partial x$, $p_y = \partial\psi_0/\partial y$, it is convenient to interpret ψ_0 as the *action* of a reversible system with a Hamiltonian

$$H = p_y^2/\tau^2 - \tau^{-1}\, y\, p_y + (ax - bx^3 + y)\, p_x . \tag{29}$$

Then (28) coincides with the Hamilton - Jacobi equation of a system with the Hamiltonian (29), moving on the hypersurface H = 0 /39/. Therefore, the characteristic system of (28) is given by the canonical equations of (29). Now, the rigorous

existence of ψ_0 independent of D is intimately connected with the integrability of the Hamiltonian H /40/. Integrability of a Hamiltonian is well known to be a non-generic special property. In particular, apart from the integration constant in (28), i.e.,H = 0, the author has not yet found a second constant of integration for (29). An explicit example, showing that ψ_0 does not exist independent of D,is given by the stationary probability of a flow driven by telegraphic noise (see relation (7) in Ref. /34/ or (2.11) in Ref./35/). This same stationary probability has also been used in the description of phase diagrams of telegraphic noise-induced transitions /41/. Although H is in general not integrable, the ansatz in (27) can still present a useful approximation away from repellers or saddle points of the deterministic motion /40/. With H non-integrable, (27) is however of limited use in an explicit evaluation of activation rates in multi-dimensional systems /39,42,43/, i.e., $\bar{p}$ must be found a priori by other methods /22,34,35,42/ or one uses numerical methods.

In view of such difficulties, one might as well directly focus on the *one-dimensional, but non-Markovian* dynamics x_t in (25). The rate of change of the probability $p_t(x)$ obeys the exact relation /44/

$$\dot{p}_t = - \frac{\partial}{\partial x} (ax - bx^3) p_t +$$

$$+ (D/\tau)\frac{\partial^2}{\partial x^2} \int_0^t \exp - \frac{(t-s)}{\tau} <[\delta x(t)/\delta y(s)] \delta(x(t) - x)> ds \qquad (30)$$

where $\delta x(t)/\delta y(s)$ is the functional derivative. This functional derivative obeys an integral equation /44/ which for (25) reads

$$\delta x(t)/\delta y(s) = \Theta(t-s) [1+ \int_s^t (a - 3bx^2(r)) \delta x(r)/\delta y(s) \ dr]. \qquad (31)$$

On expanding $(\delta x(t)/\delta y(s))$ around $s^- = t$, and iterating the relation in (31), we observe that p_t obeys a master equation which is of the Kramers-Moyal type. Moreover, infinitely many series with terms of the order $D^m \tau^n$, n >1 , m ⩽n , contribute both to higher order Kramers-Moyal moments K_n, n > 2, and to the drift and diffusion moments K_1 and K_2 /45/. In what follows, we neglect transients and sum up the terms of order $D\tau^n$ only. This results in an approximative Fokker-Planck structure /46/. Keeping the terms of order $D\tau^n$ only is not systematic /44, 45,47/; in particular, this Fokker-Planck approximation /46/ is not identical with the truncated (at second order) Kramers-Moyal expansion /45/. Nevertheless, due to its simplicity, we shall use this approximation for (25), which results in

$$\dot{p}_t = - \frac{\partial}{\partial x} (ax - bx^3) p_t + D \frac{\partial^2}{\partial x^2} G (x,\tau) p_t \qquad (32)$$

where with $f(x) = ax - bx^3$ /45,46/

$$G (x, \tau) = f(x) (1 + \tau f(x) \frac{\partial}{\partial x})^{-1} \frac{1}{f(x)} . \qquad (33)$$

$G (x, \tau)$ is not necessarily positive for all x. Therefore, we use for the diffusion: $\bar{G} (x,\tau) = G (x,\tau) \Theta(G(x,\tau))$.

Because of the neglect of transients in (32) (in this context, it cannot be over-emphasized that the (t→ ∞) - limiting master operator for $p_t(x)$ is in general different for different initial preparation schemes;the stationary probability, however, is of course independent of initial preparation), we look upon (32) as a small relaxation time approximation to the long time behavior of (25). With (32) we can readily express the rate as the inverse mean first passage time of the approximative Fokker-Planck equation (32). This procedure gives a rate $r^+ = r^- = r$ /45/

$$r = (a/\sqrt{2} \ \pi)[(1-2a\tau)/(1+a\tau)]^{\frac{1}{2}} \ \exp-(\Delta \psi/D) . \qquad (34a)$$

The Arrhenius factor $\Delta\psi$, which is determined solely by the values of the stationary probability at the extrema, reads explicitly

$$\Delta\psi = (a^2/4b)\ (1 - a^2\ \tau^2)\ \ + \ 0\ (\tau^3).\ \ \ \ \ \ \ \ \ \ \ \ \ \ (34b)$$

Note that with the approximation (32), $\Delta\psi$ does not exhibit a correlation time dependence in first order in τ. In view of this fact, we performed a numerical simulation /45/. In contrast to our forecasting in (34b), $\Delta\psi$ *is actually increasing proportional to* τ (see Fig.2) /45/. This clearly is bad news; it simply shows that the wings of the stationary probability $\bar{p}$ (x) are in leading order in τ not recovered from a short relaxation time Fokker-Planck approximation scheme.
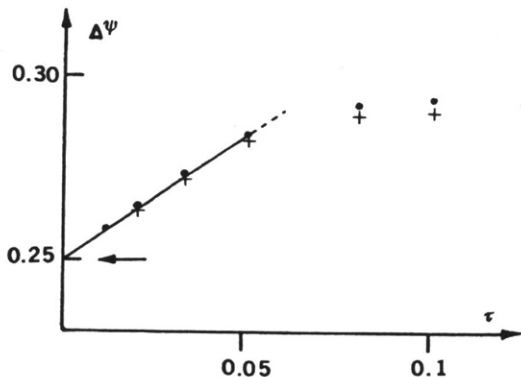

Fig. 2 Simulation results for the Arrhenius factor $\Delta\ \psi$ versus noise correlation time τ (after Ref. /45/). The parameter values are a = b = 1 , D = 0.1 ($\bullet$) and D = 0.05 (+). The maximum error bar of the numerical calculation is estimated to be about 10 %. The arrow denotes the white noise limit, $\Delta\ \psi\ (\tau=0) = (a^2/4b) = 0.25$

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