DECAY OF METASTABLE STATES - KRAMERS, FIRST PASSAGE TIME AND VARIATIONAL APPROACHES

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1. Introduction

The dynamics of a great variety of problems can be usefully studied on the basis of the Fokker-Planck equation (FPE), that builds in at the outset, a separation of macroscopic and microscopic time scales. The effect of macroscopic drives and dissipations enters through the drift terms, whereas the diffusion terms account for rapidly varying microscopic noise. An important question, conveniently answered in the FPE framework, is : what is the time required for the passage of a prepared initial state to a final stationary state . Physical examples where such questions find relevance include the nucleation rate of a liquid droplet from the vapour phase [1], decay of a supercurrent in a superfluid or superconductor [2] , spinodal decomposition in an alloy [3], optimum sweep rates for hysteresis in first order transitions [4], and so on. It is clear that deterministic forces and random noise will both play a role in determining the transition rates. Quite generally, therefore, one is interested in calculating the decay rate or the relaxation time of a metastable state through the natural tool of the FPE [5] .

Historically, a problem of this kind was first studied in a classic paper by Kramers[6]. He was interested in computing the rate of escape of a particle across a mechanical barrier due to thermal fluctuations. An FPE was used, with the stochastic variable being the position x, the thermal energy $k_B T$ being related to the diffusion constant, and the mechanical force and viscosity determining the drift term. The Kramers treatment is based on an ansatz that should be valid when the barrier height is larger than the thermal energy. A similar high barrier or weak noise assumption is invoked at different stages of other methods as will become clear later on.

An explicit example of a Kramers-like problem would be rotational Brownian motion of a single domain magnetic particle in a highly anisotropic potential[7]. Here the angle θ would replace the variable x. Another example, in a more general context, could be the dynamics of an equilibrium or nonequilibrium phase transition. Then the variable x would be an (single component) order parameter while the potential would be a coarse-grained free energy-like functional[8]. Extensions of the one-dimensional Kramers problem to many variables have also been made by several authors, using variants of the original Kramers ansatz[5]or other techniques[9]. Some comments on this will be made below.

The Kramers method is intuitively appealing and based on sound physical insight into the problem. An alternative approach is to estimate the mean time for a stochastic variable, within a given region, to first reach the boundary of that region. The

equation governing the dependence on the initial position of this mean 'First-Passage Time' (FPT), is derived from the FPE. In the high barrier/weak noise limit, the Kramers and FPT estimates for the decay rate of a metastable state are identical, if 'passage' is appropriately defined. The FPT formalism is instructive because it links the problem more directly and systematically to the machinery of stochastic problems [10], it can be formally generalized to the many-variable case and situations where detailed balance does not hold[9]. Physical applications of the FPT ideas include transient phenomena in optically bistable systems [11,12] and analysis of the problem of hysteresis versus jump behaviour in first order equilibrium and nonequilibrium transitions [4] .

Mathematically, the FPE is parabolic : it is a second order partial differential equation in the space derivatives and first order in the time derivative. The time-dependent probability can therefore be written as an eigenfunction expansion, with the eigenvalue $\{\lambda_n\}$ appearing in exponential decay factors associated with each eigenfunction. The zero eigenvalue corresponds to the stationary state, and the nontrivial eigenvalues, therefore, determine the decay rates for metastable states. This allows one to introduce a third, formally elegant approach to the problem, involving variational bounds on these eigenvalues. It turns out that the lowest non-trivial eigenvalue $\lambda_1 \sim \langle \tau_p \rangle^{-1}$, the inverse of the mean first passage time, in the large barrier limit. A physical application of this approach to the Brownian motion of a magnetic particle is discussed in detail in this volume [13].

The Kramers, FPT and variational approaches are different ways of looking at the same metastable state decay problem. These are all connected in the high barrier/low noise limit, as will be illustrated in the following sections. As mentioned earlier, generalizations of the one dimensional decay problem to many dimensions have been made in the context of Kramers treatment[5] and the FPT approach[9,14] . It is not surprising that similar ideas of saddle-point coordinate systems, small noise expansions, etc., have been independently introduced by mathematicians[9,14] and physicists[5,15] sometimes without knowledge of analogous work done elsewhere.

The rest of the article is organized as follows. In § 2, we outline the basic framework of the problem. The Kramers analysis for the one dimensional case is treated in § 3. We describe next the one dimensional FPT approach in § 4. The variational treatment for the one dimensional case is then included in § 5. The generalization to higher dimensions, in the FPT framework, as done by Schuss and Matkowsky, and its relationship to Kramers-like ideas[5], are discussed in § 6. Finally, some applications of the FPT ideas are considered in § 7.

2. Basic mathematical picture

For most part of the analysis (see, however, § 6), we shall restrict ourselves to a one-dimensional, multiplicative, but time-homogeneous stochastic process described by the FPE [16]

$$\frac{\partial P}{\partial t}(x,t) + \frac{\partial J(x,t)}{\partial x} = 0 \quad , \tag{2.1}$$

where the 'current'

$$J(x,t) = - A(x)\ P(x,t) - \frac{\partial}{\partial x}\left(D(x)\ P(x,t)\right) , \tag{2.2}$$

A(x) and D(x) being the drift and diffusion terms respectively. The stationary state solution (at $t = \infty$) is obtained by setting the current to zero, hence

$$P_o(x) = C \exp\left(- \Phi(x)\right) , \tag{2.3}$$

where C is a normalization constant, and

$$\Phi(x) = \ln D(x) + \int dx' \frac{A(x')}{D(x')} \quad . \tag{2.4}$$

We shall employ in (2.3) the natural boundary conditions :

$$P_o\ (\pm \infty) = 0 \tag{2.5}$$

It may be noted here that if one specializes to the case of an additive stochastic process, D(x) is a constant, and

$$P_o(x) = C' \exp\left(- \int dx'\ A(x')/D\right) \quad . \tag{2.6}$$

In addition, if the so-called 'potential condition' is satisfied, i.e. [10]

$$A(x) = \frac{\partial U(x)}{\partial x} \quad , \tag{2.7}$$

then,

$$P_o(x) = \eta \exp\left(-\ U(x)/D\right) \quad , \tag{2.8}$$

where η is yet another normalization constant. In the context of an 'equilibrium' problem, (2.8) has the familiar structure with D being proportional to the thermal energy $k_B T$.

Coming back to the general case of (2.3) and (2.4), we shall direct our attention to a bistable potential indicated schematically in Fig.1. The point X_s, in the one dimensional case, is a maximum of the potential. However, we use the subscript s to indicate that X_s is actually a saddle point in the more general context of a multidimensional process (§ 6). As mentioned in § 1, our analysis is restricted to high barrier/weak noise limit which implies that $A(x_s)/D(x_s)$ is 'suitably' small. The question we want to answer is : starting from an arbitrary initial distribution i.e. $P(x,t=o) = P_{init}(x)$, how long does one have to wait for the probability to evolve into $P_o(x)$, given by (2.3) ? The sequence of time development is expected to

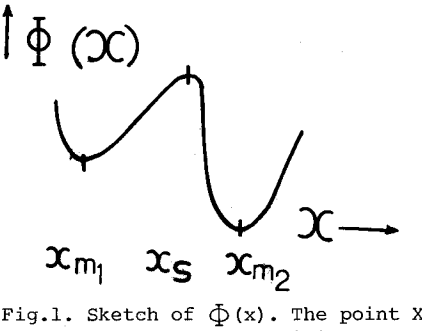

Fig.1. Sketch of $\Phi(x)$. The point X_{m_1} is a metastable minimum, x_{m_2} is a stable minimum, while X_s is an unstable maximum.

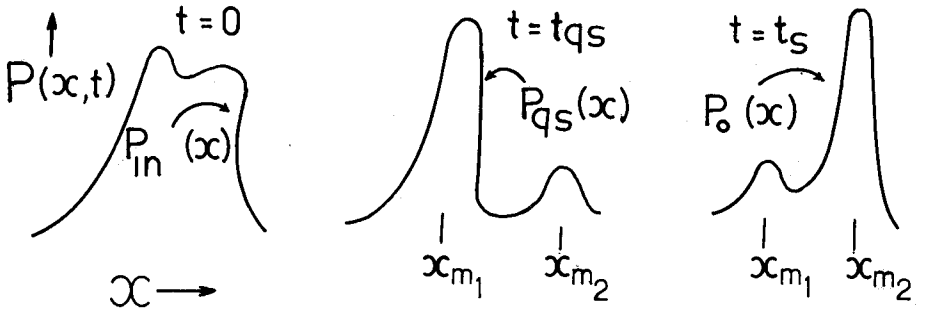

Fig. 2 The development of the probability. The quasistationary distribution $P_{qs}(x)$ is expected to be reached 'very quickly' ($t_s \gg t_{qs}$).

look like the one depicted in Fig.2. It is intuitively expected (and the expectation can be substantiated by WKB-type arguments [17]) that the time t_{qs} to reach the quasi-stationary distribution is much shorter than the time t_s to reach the final stationary distribution, in the large barrier/weak noise limit. We are, of course, interested in studying only the slow evolution of the probability between t_{qs} and t_s.

3. Kramers' argument

In the regime $t_{qs} \leqslant t \leqslant t_s$, the probability $P(x,t)$ is so slowly varying that its time derivative can be neglected. Hence from (2.1) and (2.2), the current can be replaced by J_{qs} which is independent of x and is a slowly varying function of t. Thus, in this domain of time-evolution, population of the state x_{m_1} is depleted while that of 'state' x_{m_2} is increased at an almost steady rate, in view of slow diffusion (or probability leakage) across X_s. Denoting by $P_{qs}(x,t)$ the probability in the quasi-stationary region, we have from (2.2) - (2.4),

$$J_{qs} \exp\left(\bar{\Phi}(x)\right) = -\frac{\partial}{\partial x}\left\{\exp\left(\Phi(x)\right) P_{qs}(x,t)\right\} \tag{3.1}$$

where

$$\bar{\Phi}(x) = \Phi(x) - \ln D(x) \quad . \tag{3.2}$$

Now, consider two 'small' regions $x_{m_1} - \Delta x_1 \leqslant x \leqslant x_{m_1} + \Delta x_1$ and $x_{m_2} - \Delta x_2 \leqslant x \leqslant x_{m_2} + \Delta x_2$, and define the 'density of points' in these regions by

$$n_1 \equiv \int_{x_{m_1} - \Delta x_1}^{x_{m_1} + \Delta x_1} dx \; P_{qs}(x,t) \quad . \tag{3.3}$$

A similar expression defines n_2. It is evident from Fig.2 that $P_{qs}(x,t)$ is sharply peaked around x_{m_1} and x_{m_2}. Accordingly,

$$P_{qs}(x,t) \approx P_{qs}(x_{m_1},t) \exp\left(-\left(\Phi(x) - \Phi(x_{m_1})\right), \; x_{m_1} - \Delta x_1 \leqslant x \leqslant x_{m_1} + \Delta x_1\right. \tag{3.4}$$

An analogous expression holds in the region around x_{m_2}. Combining (3.3) and (3.4)

$$n_1 \approx I_1 P_{qs}(x_{m_1},t)\exp(\bar{\Phi}(x_{m_1})), \tag{3.5}$$

where $$I_1 \equiv \int_{x_{m_1}-\Delta x_1}^{x_{m_1}+\Delta x_1} dx \exp(-\bar{\Phi}(x)). \tag{3.6}$$

The expression for n_2 is obtained from (3.5) upon replacing 1 by 2. Integrating (3.1) from x_{m_1} to x_{m_2}, and using (3.5), we have

$$J_{qs} I_s \approx n_1/I_1 - n_2/I_2, \tag{3.7}$$

where $$I_s \equiv \int_{x_{m_1}}^{x_{m_2}} \exp(\bar{\Phi}(x))\, dx. \tag{3.8}$$

Finally, we note that at this level of approximation, most of the system points are expected to be concentrated only around x_{m_1} and x_{m_2}. Thus

$$J_{qs} \approx \dot{n}_1 = -\dot{n}_2. \tag{3.9}$$

Combining (3.7) with (3.9), we arrive at the familiar rate equations

$$\dot{n}_1 = -\dot{n}_2 = -\omega_{12} n_1 + \omega_{21} n_2, \tag{3.10}$$

where

$$\omega_{12} \equiv \tau_{12}^{-1} = (I_s I_1)^{-1}, \quad \omega_{21} = \tau_{21}^{-1} = (I_s I_2)^{-1} \tag{3.11}$$

The times τ_{12} and τ_{21} are the so-called reaction times of Kramers which measure the times of passage from $x_{m_1} \longrightarrow x_{m_2}$ and $x_{m_2} \longrightarrow x_{m_1}$ respectively.

In the high barrier/weak noise limit, the integrals in (3.6) and (3.8) can be evaluated approximately by the method of steepest descents, and we obtain

$$\omega_{12} = \tau_{12}^{-1} \approx (2\pi)^{-1} \left(\bar{\Phi}''(x_{m_1}) \left|\bar{\Phi}''(\bar{x}_s)\right|\right)^{\frac{1}{2}} \exp\left(-(\bar{\Phi}(\bar{x}_s)-\bar{\Phi}(x_{m_1})\right) \tag{3.12}$$

The expression for ω_{21} can be written down from (3.12) by interchanging 1 and 2. Here $\bar{x}_s$ is defined by

$$\bar{\Phi}'(x=\bar{x}_s) = 0, \quad \bar{\Phi}''(x=\bar{x}_s) < 0 \tag{3.13}$$

It may be remarked that the analysis given above is more general than the original Kramers' treatment in that a multiplicative process or x-dependent diffusion has been considered. In the special case of an additive process for which the potential condition is satisfied (cf., (2.6)-(2.8)), (3.12) yields the familiar result [6]

$$\omega_{12} \approx D(2\pi)^{-1} (U''(x_{m_1})|U''(x_s)|)^{\frac{1}{2}} \exp(-(U(x_s)-U(x_{m_1}))/D). \tag{3.14}$$

4. First Passage Time (FPT) estimates

In this section we shall outline briefly the FPT method for one dimensional stochastic processes in order to put it at par with the Kramers treatment. Later, in § 6,7, we shall return to a more elaborate discussion of FPT calculations in the context of multi-dimensional processes and mention a few physical applications as well.

Let us focus our attention first to calculating τ_{12} (cf. (3.11)). The system point is assumed to be initially at x_o at time t = o, where $-\infty \leqslant x_o \leqslant x_s$. We imagine that an absorbing boundary is erected at x_s such that once the system point reaches x_s it is removed from any further consideration. We denote by $\tau(x_o)$ the time taken by the system point to reach x_s for the first time, having started from x_o at t = o. This so called first passage time (FPT) is clearly a random variable which varies from realization to realization. We shall later identify the mean FPT $\langle \tau(-\infty) \rangle$ with τ_{12} of § 3 provided 'passage' is defined in an appropriate manner.

Let $P(x,t|x_o,o)$ be the conditional probability that the random process is x at time t given that it was x_o at t=o. Then, the probability that at time t the system point is still within the interval $-\infty$ to x_s (not having reached x_s even once) is given by

$$G(x_o,t) = \int_{-\infty}^{x_s} P(x,t|x_o,o)\, dx. \tag{4.1}$$

The quantity $G(x_o,t)$ evidently equals $P_r(\tau(x_o) \geqslant t)$. Since the conditional probability by definition, is a delta function centred around x_o at t = o, it follows from (4.1) that

$$G(x_o,o) = P_r(\tau(x_o) \geqslant o) = 1, \quad -\infty \leqslant x_o \leqslant x_s \tag{4.2}$$

$$= o \qquad , \text{ elsewhere.}$$

On the other hand, if x_o happens to equal x_s, the absorbing boundary, the process 'dies' immediately, hence

$$G(x_s,t) = P_r(\tau(x_s) \geqslant t) = o \tag{4.3}$$

Alternatively

$$G(x_o, t=\infty) = o \tag{4.4}$$

as the system point is expected to reach x_s at least once when $t = \infty$.

Since $G(x_o,t)$ is the probability that passage has not occurred: $\tau(x_o) \geqslant t$, and $1-G(x_o,t)$ is the probability that passage has occurred, the mean FPT is given by

$$T_p(x_o) = \langle \tau(x_o) \rangle = -\int_o^\infty t\, \dot{G}(x_o,t)\, dt = \int_o^\infty G(x_o,t)\, dt, \tag{4.5}$$

where the last step follows upon integration by parts and from (4.4). Our next task is to derive an equation for $T_p(x_o)$. This can be done easily if one uses the backward FPE [10] which reads

$$\frac{\partial}{\partial t} P(x,t|x_o,o) = -A(x_o)\frac{\partial}{\partial x_o} P(x,t|x_o,o)+D(x_o)\frac{\partial^2}{\partial x_o^2} P(x,t|x_o,o). \tag{4.6}$$

Therefore, from (4.1) (4.5) and (4.6), we have

$$L^{\dagger}_{x_o} T_p(x_o) = -1 \tag{4.7}$$

where the adjoint operator $L^{+}_{x_o}$ is the one associated with the backward FPE:

$$L^{\dagger}_{x_o} \equiv D(x_o)\frac{\partial^2}{\partial x_o^2} - A(x_o)\frac{\partial}{\partial x_o}. \tag{4.8}$$

The solution of (4.7), consistent with the boundary conditions:

$G(x,t) = o$ at $x = x_s$ (absorbing boundary)

$$\frac{\partial}{\partial x} G(x,t) = o \text{ at } x = -\infty \text{ (reflecting boundary)}, \tag{4.9}$$

can be written as

$$T_p(x) = \int_x^{x_s} dx' \exp(\bar{\Phi}(x') \int_{-\infty}^{x'} dx'' \exp(-\bar{\Phi}(x'') \quad . \tag{4.10}$$

Equations for higher moments of $\tau(x)$ can also be derived from the distribution of FPT. It turns out [12] that in the high barrier/weak noise limit,

$$T_p^{(r)}(x) \equiv \langle \tau^r(x) \rangle \simeq r! \langle \tau(x) \rangle^r = r! (T_p(x))^r. \tag{4.11}$$

This corresponds to a distribution for τ that for large lifetimes is

$$P(\tau) \simeq [T_p(x)]^{-1} e^{-\tau(x)/T_p(x)} \tag{4.12}$$

Evaluating the integrals in (4.10) by the method of steepest descent, in the large barrier limit and setting $x = -\infty$, we obtain an answer for $\tau_{12} (= T_p(-\infty))$ which is one-half the Kramers estimate (cf.(3.12)). This discrepancy is, however, not serious and can be removed if the absorbing boundary X_s is taken at $+\infty$, which is of course the more appropriate limit, for a meaningful comparison with Kramers result.

5. Variational treatment

We may write the solution of the FPE (cf.(2.1) and (2.2) as

$$P(x,t) = a_o P_o(x) + \sum_{n>o} a_n P_n(x) \exp(-\lambda_n t), \tag{5.1}$$

where the eigenvalues $\lambda_n > o$ for $n > o$. The first term corresponds to zero eigenvalue which yields the stationary state solution (as $t \to \infty$). Comparing with (2.3), therefore,

$$a_o = c, \quad P_o(x) = \exp(-\bar{\Phi}(x)) \quad . \tag{5.2}$$

Combining (5.1) with (2.1) leads to the elliptic partial differential equation

$$\frac{\partial}{\partial x}\,(A(x)\;P_n(x)) + \frac{\partial^2}{\partial x^2}\,(D(x)\;P_n(x)) = -\lambda_n\;P_n(x)\quad . \tag{5.3}$$

Now, the substitution :

$$F_n(x) \;=\; P_n(x)\;/\;P_o(x)\quad , \tag{5.4}$$

transforms (5.3) into a <u>self-adjoint</u> eigenvalue equation of the Sturm-Liouville form [18] :

$$\frac{\partial}{\partial x}\;(D(x)\;P_o(x)\;\frac{\partial}{\partial x}\;)\;F_n(x) = -\lambda_n\;P_o(x)\;F_n(x)\quad . \tag{5.5}$$

Equation (5.5) is the Euler-Lagrange equation for the functional (with the density function $P_o(x)$) :

$$I\,[F_n(x)] \;=\; \int_{-\infty}^{\infty} dx\;P_o(x)\,[D(x)\Big(\frac{\partial F_n(x)}{\partial x}\Big)^2 \;-\;\lambda_n\;F_n^2(x)]\quad . \tag{5.6}$$

Following standard procedure [18], the eigenvalue λ_n can be shown to obey the Rayleigh-Ritz inequality

$$\lambda_n \leqslant\; K\,[f_n(x)]\;/\;H\,[f_n(x)]\quad , \tag{5.7}$$

where

$$K\,[f_n(x)] \;=\; \int_{-\infty}^{\infty} dx\;D(x)\;P_o(x)\;\left(\frac{\partial f_n(x)}{\partial x}\right)^2\quad , \tag{5.8}$$

$$H\;[f_n(x)] \;=\; \int_{-\infty}^{\infty} dx\;\;P_o(x)\;\;f_n^2\;(x)\quad . \tag{5.9}$$

and $f_n(x)$ is some general trial function.

In order to employ (5.7) for estimating the eigenvalue λ_n one normally assumes a particular form for the trial function $f_n(x)$ in terms of certain variational parameter(s) and minimizes the right hand side of (5.7) with respect to these parameter (s). As mentioned before, one requires only the lowest nontrivial eigen value for describing the passage from the quasistationary to the stationary state. Thus we shall use the inequality (5.7) for n=1 only, and drop the subscript n henceforth for the sake of brevity. The variational choice of the eigenfunction f(x) must be normalized and orthogonal to the lowest eigenfunction $F_o(x)$, i.e.,

$$\int P_o(x)\;\;f(x)\;dx\;\;=\;0\quad , \tag{5.10}$$

since $F_o(x)$ is unity in the present case (cf. (5.4)). Equation (5.10) implies that the trial function must change sign as the system point moves from the interval $x_{m_1} \leqslant x \leqslant x_s$ to $x_s \leqslant x \leqslant x_{m_2}$.

The first and only (to the best of our knowledge) attempt to correlate the variational treatment with the Kramers kind of approach was due to Brown[7,13]. While the Brown choice for the trial function is quite adequate for calculating upper bounds

to the eigenvalues from the Rayleigh-Ritz principle, it is not suitable for computing certain lower bounds given by Weinstein and Kamke[19] . The Weinstein-Kamke criteria, used in conjunction with the Rayleigh-Ritz principle, can obviously provide better bounds to the eigenvalues[20]. This turns out to be possible only if the trial function has finite first and second derivatives, a condition which the Brown choice does not satisfy. We shall present below an alternate form for the trial function which, in addition to providing Kramers-like result in the large barrier limit, yields also a lower bound to the eigenvalue λ [21] .

Guided by the fact that $\frac{\partial f}{\partial x}$ should be concentrated near X_s where $P_o(x)$ has its minimum in order to keep $K[f(x)]$ small (see (5.8)), we choose

$$f(x) = f_1\left[1+\exp(-a(x-x_s))\right]^{-1} + f_2\left[1+\exp\ (\ a(x-x_s))\right]^{-1} \quad , \tag{5.11}$$

where the constants f_1 and f_2 (which will turn out to be of opposite sign in view of (5.10)) can be determined from normalization and orthogonality conditions. Now, in anticipation of the fact that the Kramers - like formula should emerge in the large barrier limit, we take

$$a^2 = -b\ \Phi''(x_s) \quad , \tag{5.12}$$

where b is a variational parameter and $\Phi''(x_s)$ is the curvature of the potential at x_s. Details of the variational calculation will be reported elsewhere [21] . Here, we merely state that substitution of (5.11) into (5.7) and evaluation of the relevant integrals by the method of steepest descent yields

$$\lambda \leqslant \frac{\pi}{8}\, b\ (1-b)^{-\frac{1}{2}}\ \ (\omega_{12} + \omega_{21}) \quad , \tag{5.13}$$

where ω's are given by (3.12). Minimization of the b-dependent term in (5.13) leads to b=2, and hence the variational estimate for the lowest upper bound to the eigenvalue is

$$\lambda_v = \frac{\pi}{4}\ (\omega_{12} + \omega_{21}) \quad , \tag{5.14}$$

which is still lower than the Kramers estimate by a factor of $4/\pi$!

6. First passage time estimates in higher dimensions

We have presented in § 3 - § 5 three distinct approaches to the study of the decay of a metastable state. It has been demonstrated that for a one dimensional stochastic process the Kramers, FPT and variational methods all lead to essentially identical result for the decay rate, in the high barrier/weak noise limit. The equivalence is expected to hold also for higher dimensional stochastic processes which are important in certain phase transitions such as in superfluidity [2] and two-mode lasers [22] . In this section, we outline the extension of the FPT calculation to higher dimension, based on the work of Schuss and Matkowsky [9,14] .

We consider an n-component stochastic variable $\vec{x} = (x_1, x_2, \ldots\ldots x_n)$ taking an

initial value $\vec{x}_o$ somewhere within an n-dimensional volume Ω. Generalizing the concept introduced in § 4, 'first passage' is now defined by $\vec{x}$, within a given realization, first reaching the boundary $\partial\Omega$, an (n-1) dimensional surface. The mean FPT satisfies an equation analogous to (4.7) :

$$L^{+}_{\vec{x}_o} \; T_p(\vec{x}_o) = \left(D \, \vec{\nabla}^2_{\vec{x}_o} - \vec{A}(\vec{x}_o) . \vec{\nabla}_{\vec{x}_o} \right) T_p(\vec{x}_o) = -1 \; , \tag{6.1}$$

where, for simplicity, we consider an $\vec{x}$ - independent (additive noise) diagonal diffusion term [23] and restrict ourselves to the potential case $\vec{A}(\vec{x}) = \vec{\nabla} U(\vec{x})$. As in the one-dimensional case, the boundary condition imposed is (cf.(4.3))

$$T_p(\vec{x}_o \in \partial\Omega) = 0 \tag{6.2}$$

It may be noted that, unlike in the one dimensional problem, the equation for $T_p(\vec{x}_o)$ is now a partial differential equation in n variables, so obtaining a general solution is difficult. The strategy, therefore, would be to search for a single (or at least only a few) dominant variable and ignore the others. The Schuss-Matkowsky procedure employs just this in terms of a systematic high barrier/low noise expansion.

The first step is to scale (6.1) by ΔU, the smallest barrier height in the problem (defined below more precisely). Thus

$$(\epsilon \; \vec{\nabla}^2_{\vec{x}_o} - \vec{a}(\vec{x}_o) . \vec{\nabla}_{\vec{x}_o}) \, \tau_p(\vec{x}_o) = -1 \; , \tag{6.3}$$

where

$$\epsilon \equiv D/\Delta U \; , \quad \vec{a} \equiv \vec{A}/\Delta U \; , \quad \tau_p \equiv \Delta U \, T_p \; . \tag{6.4}$$

In the high barrier/weak noise limit, $\epsilon \ll 1$.

Since in the limit $\epsilon \longrightarrow o$ the FPT is expected to be infinitely large, the basic idea of Matkowsky and Schuss is to isolate the singular dependence of $\tau_p(\vec{x})$ on ϵ and assume the rest to be regular in ϵ, expandable in a power series. Thus

$$\tau_p(\vec{x}) = v(\vec{x}) \exp(K/\epsilon) \; , \tag{6.5}$$

with

$$v(\vec{x}) = v^{(o)}(\vec{x}) + \epsilon \, v^{(1)}(\vec{x}) + \ldots\ldots\ldots\ldots , \tag{6.6}$$

where K is a constant. Substituting (6.5) and (6.6) into (6.3),

$$(\epsilon \, \vec{\nabla}^2_{\vec{x}} - \vec{a}(\vec{x}) . \vec{\nabla}_{\vec{x}}) \, (v^{(o)}(\vec{x}) + \epsilon \, v^{(1)}(\vec{x}) + \ldots\ldots\ldots) = -\exp(-K/\epsilon) \tag{6.7}$$

Now, if we were to retain only the term manifestly independent of ϵ, we would write

$$\vec{a}(\vec{x}) . \vec{\nabla}_{\vec{x}} \, v^{(o)}(\vec{x}) = 0 \; , \tag{6.8}$$

which would imply that $v^{(o)}(\vec{x})$ is a <u>non-zero</u> constant(in regions where $\vec{a}(\vec{x}) \neq o$). This however, would immediately contradict the boundary condition (6.2), keeping in view (6.5). The paradox can be resolved satisfactorily if there is at least one direction along which $v^{(o)}(\vec{x})$ varies rapidly near the surface $\partial\Omega$ on a scale of $\epsilon^{\frac{1}{2}}$ in such a way that its second derivative times ϵ is actually independent of ϵ. This special

direction which we call z, is expected to lie along the path of the steepest descent from the saddle point $\vec{x}_s$ on $\partial\Omega$. It is convenient now to switch to a new set of axes $(\vec{\rho},z)$, centred on the point $\vec{x}_s$, as indicated schematically in Fig.3. The unit vector $\hat{z}$ points inwards from the boundary and the other n-1 directions $\vec{\rho} = (\rho_2 \ldots\ldots \rho_n)$ are normal to $\hat{z}$. For points near $\vec{x}_s$ i.e. $\hat{z}$ small, a consistent expression to zeroth order in ϵ would include only the steepest descent variation

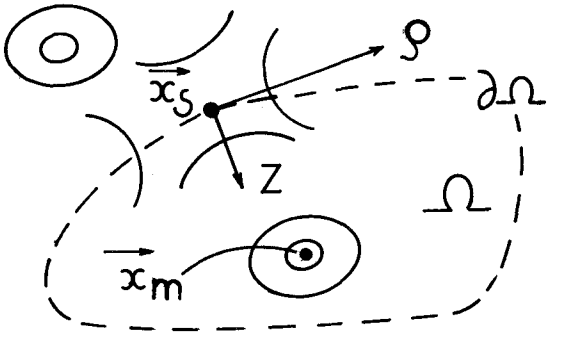

Fig.3. Schematic plot of equipotential (solid) lines and preferred axes $(z,\vec{\rho})$ at saddle point $\vec{x}_s$. $\vec{x}_m$ is the minimum within the boundary $\partial\Omega$ (dashed line).

$$\left(\epsilon \frac{\partial^2}{\partial z^2} - a_z(z,\vec{\rho}) \frac{\partial}{\partial z}\right) v^{(o)}(z,\vec{\rho}) = 0 \quad , \tag{6.9}$$

with terms first order in ϵ neglected :

$$\epsilon \vec{a}(z,\vec{\rho}) \cdot \vec{\nabla}_{\vec{\rho}} v^{(o)}(z,\vec{\rho}) \approx 0 \quad . \tag{6.10}$$

For the sake of simplicity we shall now assume that the potential condition (2.7) holds. From (6.4) then

$$a_z(z,\vec{\rho}) = \frac{1}{\Delta U} \frac{\partial U(z,\vec{\rho})}{\partial z} \quad . \tag{6.11}$$

Expanding around the saddle point $\vec{x}_s$ at $z=o, \vec{\rho}=o$

$$a_z(z,\vec{\rho}) \approx - \frac{z}{\Delta U} |U''(o)| \quad . \tag{6.12}$$

Here the primes denote partial derivatives with respect to z, and the argument is evaluated at the origin of $(z,\vec{\rho})$, i.e. the saddle point.

Substituting (6.12) into (6.9), and using (6.4), we have

$$v^{(o)}(z,\rho) \approx \xi \Delta U \int_o^z \exp\left[-\frac{1}{2}\frac{|U''(o)|}{D} z'^2\right] dz' \tag{6.13}$$

where ξ is a constant independent of $\vec{\rho}$ (cf.(6.10). From (6.4) and (6.5), the mean FPT in the high barrier/weak noise limit is then given by

$$T_p(z) = \xi \exp(K/\epsilon) \int_o^z \exp\left[-\frac{1}{2}\frac{|U''(o)|}{D} z'^2\right] dz' \tag{6.14}$$

The remaining task is to evaluate the constant ξ . To this end, we note from (6.3),

$$\int_{\Omega} d^n x \exp\left(-\frac{U(\vec{x})}{D}\right) = \int_{\Omega} d^n x \exp\left(-\frac{U(\vec{x})}{D}\right)\left[a(\vec{x})\cdot\vec{\nabla} - \epsilon\vec{\nabla}^2\right]\tau_p(x)$$

$$= -\epsilon\int_{\Omega} d^n x \; \vec{\nabla}_{\vec{x}} \cdot \left[\left(\vec{\nabla}_{\vec{x}} \tau_p(\vec{x})\right) \exp\left(-\frac{U(\vec{x})}{D}\right)\right] , \qquad (6.15)$$

where we have used the potential condition (6.11) in writing (6.15) as a divergence in the n-dimensional space. Employing now the Gauss theorem, and noting that the normal to the surface $\partial\Omega$ (at z=o) lies inward along $\hat{z}$, we have

$$\int_{\Omega} d^n x \exp\left(-\frac{U(\vec{x})}{D}\right) = \epsilon\int_{\partial\Omega} d^{n-1}\rho \left[\left(\frac{\partial}{\partial z}\tau_p(z,\vec{\rho})\right) \exp\left(-\frac{U(z,\vec{\rho})}{D}\right)\right]_{z=o}$$

$$= \epsilon\,\Delta U\,\xi \exp(K/\epsilon)\int_{\partial\Omega} d^{n-1}\rho \exp\left[-\frac{U(o,\vec{\rho})}{D}\right] , \qquad (6.16)$$

where the last step follows from (6.14) and the definition of τ_p. Equation (6.16) determines ξ and hence the mean FPT from (6.14). Noting that the mean FPT corresponds to the Kramers reaction time (§ 3) when the initial point z lies at infinity, we have from (6.14) and (6.16)

$$T_p \equiv T_p(z=\infty) = \frac{1}{D}\sqrt{\frac{\pi D}{2|U''(o)|}} \; \frac{\int_{\Omega} d^n x \exp(-U(\vec{x})/D)}{\int_{\partial\Omega} d^{n-1}\rho \exp\left[-\frac{U(o,\vec{\rho})}{D}\right]} \qquad (6.17)$$

Using sharp peaking arguments and assuming a minimum at $\vec{x}_m \in \Omega$, and a saddle point at $\vec{x}_s$, (6.17) can be given a more compact form

$$T_p = \frac{\pi \exp(\Delta U/D)}{\left[H_n(\vec{x}_n)\right]^{\frac{1}{2}} \left\{\left[H_{n-1}(\vec{\rho}=0)\right]^{-\frac{1}{2}} |U''(o)|^{\frac{1}{2}}\right\}} \qquad (6.18)$$

where ΔU, introduced earlier, is defined by

$$\Delta U \equiv U(\vec{x}_s) - U(\vec{x}_m). \qquad (6.19)$$

In (6.18), $H_n(\vec{x})$ is an nxn Hessian with its elements $\partial^2 U/\partial x_i \partial x_j$ evaluated at $\vec{x}_n$. The expression in curly brackers is evaluated at the origin ($\vec{x}_s$) of the coordinate system ($z,\vec{\rho}$), $H_{n-1}(\vec{\rho})$ being a similar (n-1) x (n-1) Hessian. It is easy to check that (6.18) reduces to the one-dimensional result discussed in § 4, upon replacing H_n by a second derivative and H_{n-1} by unity (see also (3.12)). For several saddle points on the boundary that are degenerate, i.e. have the same ΔU, a sum over the curly brackets is taken in the denominator of (6.18).

The result given above in (6.18) is for systems where the potential condition holds. We shall not deal with the 'non potential condition' case here except to remark that it can be done in terms of a series expansion in ϵ [23]. Graham and Schenzle [24] have developed similar ideas for the stationary solution of the FPE describing dispersive optical bistability, in the case where the potential conditions to not hold.

The generalization of the Kramers treatment to n dimensions has also been done [5]. An outline of the argument follows, pointing out the similarity to the FPT treatment above.

The quasistationary distribution, with steady probability flow across a saddle point at $\vec{x}_s$, is taken to be

$$P_{qs}(\vec{x}) = \omega(x)\, e^{-U(x)/D} \tag{6.20}$$

The probability current density describing this flow between wells is then

$$\vec{j}(\vec{x}) = -D(\vec{\nabla}\omega(\vec{x}))e^{-U(x)/D} \tag{6.21}$$

with the Fokker-Planck equation in the quasistationary limit, corresponding to $\vec{\nabla}.\vec{j}=0$. The $\omega(x)$ must have constant values in either well, to enhance/suppress the two peaks of the true stationary distribution $P_o(\vec{x}) = e^{-U(\vec{x})/D}$. By current conservation requirements, $P_o(\vec{x})$ falls off rapidly around $\vec{x}_s$, along the lines of current flow. $\omega(\vec{x})$ must therefore vary rapidly, near $\vec{x}_s$, to compensate. This is clearly reminiscent of the behaviour of $v(x)$ in the FPT treatment.

In fact, the equation for $\omega(x)$ coming from $\vec{\nabla}.\vec{J}(\vec{x}) = 0$ is seen to be similar to the homogeneous version of (6.3) (cf. also (5.4), (5.5), in the variational case). Once again, the arguments of a preferred coordinate system $(z,\vec{\rho})$, retention of only the steepest descent variable z etc., carry through for $\omega(\vec{x})$, just as for $v(\vec{x})$ [5].

With hindsight, there is even some conceptual similarity between the FPT and the Kramers treatments. In the latter, the decay rate is the (integrated) current divided by the initial density of the particles in the metastable well. This is like an initial decay rate, maintained by replenishment of the metastable population. The FPT treatment equates the metastable decay rate to the mean time of <u>first</u> passage of a given realization. Thus the similiarities of both the intermediate arguments and the final results, is not surprising.

7. <u>Applications:</u>

We now briefly discuss some applications of the first-passage time ideas. Applications of the variational approach are discussed elsewhere [13].

The first passage time can be explicitly calculated for models of physical systems. For example a ring laser [22] with two counter-propagating (complex) field modes E_1, E_2, obeys the Langevin equation

$$\frac{\partial E_{1,2}}{\partial t} = (a_{1,2} - |E_{1,2}|^2 - \xi|E_{2,1}|^2)\, E_{1,2} + q_{1,2}(\tau)\,. \tag{7.1}$$

Here a_1 and a_2 are pump parameters for the two modes, ξ is a mode-coupling constant $2 > \xi > 1$, and $q_1(t)$ and $q_2(t)$ are (complex) delta-correlated random forces, of scaled strength 2.

The potential conditions are satisfied, and the potential [22,4] depends only on the intensities $I_{1,2}$ and not on the phases $\theta_{1,2}$, where $E_{1,2} = \sqrt{I_{1,2}}\, e^{i\theta_{1,2}}$. It

is given by

$$U(I_1,I_2) = \frac{1}{2}a_1I_1 + \frac{1}{2}a_2I_2 - \frac{1}{4}I_1^2 - \frac{1}{4}I_2^2 - \frac{1}{2}\xi I_1I_2. \tag{7.2}$$

For the homogeneously broadened case, $\xi > 1$, there are wells at $I_1 = 0$, $I_2 = a_2$ and at $I_2 = 0$, $I_1 = a_1$, with a saddle point at $(I_{1s},I_{2s}) = (\xi a_2 - a_1)/(\xi^2-1)$, $(\xi a_1 - a_2)/(\xi^2-1)$. An estimate of the first passage time has been made [24]. The I_2 dependence of $P_o(I_1,I_2)$ is first integrated out and the one-dimensional T_p formula of (4.10) is used, with an I_1-dependent diffusion constant ansatz to allow for the higher dimensionality of the actual problem. This yields, for large pump parameters,

$$T_p \approx \frac{\pi^{\frac{1}{2}}}{2} \frac{e^{\frac{1}{4}(\xi^2-1)\ I_{1s}^2}}{(\xi^2-1)^{3/2}\ I_{1s}^2} \tag{7.3}$$

Similar results can be obtained, using the systematic Schuss-Matkowsky formalism applied to the four-dimensional case [25].

A direct measurement of 'dwell-times' within the wells centred at $I_1 = 0$ (off state) and $I_1 = a_1$ (on state) has been made [24]. The boundary is defined as at I_{1s}. A photodetector measures the intensity of the selected (I_1) laser mode, with $I_1 > I_{1s}$. A limiter changes the photodetector output to a series of rectangular pulses of variable duration τ_{ON}, that is the 'dwell time' in the well. The mean dwell time is equated to the mean first passage time from the 'on' to the 'off' state, $T_p = \langle \tau_{ON} \rangle$ T_p varies from milliseconds to minutes, depending on the parameters, and is in rough agreement with (7.3). The dwell-time statistics are in good agreement with (4.11).

First passage times also enter naturally in estimates of the extent of hysteresis phenomena analogous to superconducting and superheating [4]. Consider a first order phase transition, either dissipative or non-dissipative, described by a (one-component) order parameter x, with a drive parameter μ and a delta correlated random force $f(\tau)$ of strength 2D. The Langevin equation is

$$\dot{x} = -A(x,\mu) + f(t) = -\frac{\partial U}{\partial x}(x,\mu) + f(t) \tag{7.4}$$

The stationary states $\bar{x}(\mu)$, defined by

$$A(\bar{x},\mu) = 0 \tag{7.5}$$

have three branches if μ is within the range $\mu_2 < \mu < \mu_{c1}$. $\bar{x}(\mu)$ is an s-shaped curve on an $\bar{x}$-μ plot, with the backward bending branch unstable, corresponding to a maximum of the potential $U(x,\mu)$. The two forward-bending curves correspond to local minima of $U(x,\mu)$, that move relatively, up and down, as μ is varied. The higher, or metastable well disappears at the spinodal points $\mu = \mu_{c1}$ or μ_{c2}, defined by a vanishing relaxation rate $T_r^{-1} \propto \partial\, U(\bar{x},\mu)/\partial \bar{x}^2 = 0$ at the metastable minimum.

At some $\mu = \mu_x$, between μ_{c1} and μ_{c2}, the two well-depths are equal and thermodynamic 'Maxwell Construction' behaviour says that a system should jump

to the absolute minimum of $U(x,\mu)$ as soon as $\mu > \mu_x$. But hysteresis can occur. The question is, what determines when the jump actually takes place?

The answer turns out to be dependent on three relevant time scales. The rates are (i) $\dot{\mu}$ the rate of change of the control parameter; (ii) the 'hop-over' or first passage rate T_p^{-1}; and (iii) the roll-back or relaxation rate T_r^{-1} in the metastable well. The basic idea is that $\dot{\mu}$ must raise the metastable well too fast for a hop-over, but slow enough so the system sits near the moving well minimum (adiabatic following). These two requirements define the brackets of a 'hysteresis window' for $\dot{\mu}$.

For hysteresis to occur, $P_s^{-1} dP_s/dt > T_p^{-1}$ where $P_s \propto e^{-U/D}$ depends on time only through $\mu(t)$. For adiabatic following, the deviation from the minimum $\tilde{x}(t) = x(t) - \bar{x}(\mu(t))$ must be small, $\tilde{x}/\bar{x} \ll 1$, and not increase in time, $\dot{\tilde{x}} = o$. This yields the condition [4].

$$\frac{\bar{x}}{T_r^2} \left| \frac{\partial A(\bar{x},\mu)}{\partial \mu} \right|^{-1} > \dot{\mu} > \frac{D}{(\partial U(\bar{x},\mu)/\partial \mu)} T_p^{-1} \tag{7.6}$$

The first inequality, involving T_r, is a condition that the hysteresis state has a simple description, in terms of the most probable values, $\bar{x}(\mu)$, alone.

The second inequality, involving T_p, sets an upper bound on the degree of hysteresis that can occur. At the limit of metastability, the first passage time drops to zero, as the intervening barrier disappears [4]:

$$T_p \propto (\mu - \mu_{cl,2})^{\frac{1}{2}} \tag{7.7}$$

The inequality of (7.6) will be violated at some earlier μ, that is closer to $\mu_{cl,2}$ if $\dot{\mu}$ is larger. For $\dot{\mu} = 0$, infinitely slow variation, the hysteresis window shuts and the Maxwell construction obtains.

Numerical estimates of first passage times for condensed matter systems are often extremely large, when one-dimensional formulae corresponding to uniform states are used. The higher dimensional extensions of Section 6 could be used to include spatial variation and droplet formation, leading to improved estimates of T_p and the hysteresis window.

In conclusion, the Kramers, variational, and first-passage time formalism are three complementary and essentially equivalent ways of estimating metastable lifetimes, and can be usefully applied to a variety of systems in quantum optics and condensed matter physics.

References

1. R. Becker and W. Döring, Ann. Physik 24, 719 (1935); also J. Frenkel, "Kinetic Theory of Liquids", Chap.VII, Dover Publications, Inc., New York 1955.
2. J.S. Langer and M.E. Fisher, Phys. Rev. Letters 19, 560 (1967) and J.S. Langer and V. Ambegaokar, Phys. Rev. 164, 498 (1967).
3. J.S. Langer, Ann. Phys. (N.Y) 65, 53 (1971).
4. G.S. Agarwal and S.R. Shenoy, Phys. Rev. A23, 2719 (1981) ; See also R. Gilmore, Phys. Rev. A20, 2510 (1979).

5. R. Landauer and J.A. Swanson, Phys. Rev. 121, 1668 (1961) ; see also J.S. Langer, Ann. Phys. (N.Y.) 54, 258 (1969).
6. H.A. Kramers, Physica (utrecht) 7, 284 (1940).
7. W.F. Brown, Jr., Phys. Rev. 130, 1677 (1963).
8. H. Haken, "Synergetics", Chap. 7, Springer-Verlag, Berlin, 1977.
9. Z. Schuss, SIAM J. Review 22, 119 (1980).
10. R.L. Stratonovich, "Topics in the Theory of Random Noise", trans. by R.A. Silverman, Chap. 4, Vol. 1, Gordon and Breach, New York 1963. See also C.W.Gardiner," A Handbook of Stochastic Methods for Physics, Chemistry and Natural Sciences, Chap. 5, Springer- Verlag, Berlin 1982.
11. J.D. Farina, L.M. Narducci, J.M. Yuan and L.A. Lugiato, in "Optical Bistability", eds. C.M. Bowden et al., Plenum 1981, p.337.
12. R. Roy, R. Short, J. Durnin and L. Mandel, Phys. Rev. Lett. 45, 1486 (1980).
13. D. Kumar, this volume, p.
14. Z. Schuss and B.J. Matkowsky, SIAM J. Appl. Math. 35, 604 (1979); also B.J. Matkowsky and Z. Schuss, ibid, 33, 365 (1977).
15. R. Graham and A. Schenzle, Phys. Rev. A23, 1302, (1981).
16. G.S. Agarwal, this volume, p.
17. V. Srinivasan,this volume, p. ; see also the appendix in Ref. [13] .
18. See, for example, J. Mathews and R.L. Walker, "Mathematical Methods of Physics", Chap. 12, Benjamin, Inc., New York 1965.
19. D.H. Weinstein, Proc. Nat. Acad. Sci. U.S.A. 20, 529 (1934) and E. Kamke, Math. Z. 45, 788 (1939).
20. H. Brand, A. Schenzle and G. Schröder, Phys. Rev. A25, 2324 (1982).
21. G.S. Agarwal and S. Dattagupta, unpublished.
22. M.M. Tehrani and L. Mandel, Phys. Rev. A17, 677 (1978) ; 17, 694 (1978) ; F.T.Hioe, S. Singh and L. Mandel, ibid., 19, 2036 (1979).
23. The second of Ref. [14] contains a general treatment with off-diagonal, x-dependent diffusion, and without the 'potential condition', but within the usual $\epsilon \ll 1$ limit.
24. S. Singh and L. Mandel, Phys. Rev. A20, 2459 (1979); L. Mandel, R.Roy and S.Singh p.127 in "Optical Bistability", cited in Ref.[11].
25. G.S. Agarwal and S.R. Shenoy, unpublished.