DIFFUSION IN MODULATED MEDIA

Himadri S. Samanta
Department of Theoretical Physics,
Indian Association for the Cultivation of Science
Jadavpur, Calcutta 700 032, India
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We study the motion of Brownian particle in modulated media in the strong damping limit by using toy model, with special emphasis on the transition from localise to diffusive behavior. By using model potential we have seen the localised behavior when the number of minima of the potential is finite in the asymptotic time limit. In the limit of infinite number of minima we have seen the diffusive behavior. We calculate exactly the diffusion coefficient in periodic field of force. We have also studied the transport in commensurate and incommensurate media.

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I. INTRODUCTION

Thermally activated barrier crossing has been subject of research for many decades since the pioneering work of Kramers[1] on the subject. A fair amount of attention has recently been devoted to the study of complex non-equilibrium systems. These include the case of diffusion over a barrier in the presence of harmonic force[2] and the diffusion over a fluctuating barrier[3,4]. The hallmark of the former situation is the phenomenon of stochastic resonance. The problem of surmounting potential barriers[4,5] has gained importance in other field of science such as evolutionary computations and global optimization as well. In recent work[6] and extensive references therein), we have studied the barrier crossing of a time dependent potential which adiabatically evolves in time that sheds some light on the global optimization problem.

The transport and diffusion properties of Brownian particles continues to attract enormous interest and activity even though a century has passed since the appearance of the famous seminal work of Einstein on the subject[7]. The motion of atoms, vacancies, excitations, molecules, molecular clusters and colloidal particles on surfaces in an active area of research due to its theoretical interest[8] and modern technological relevance involving self-assembled molecular film growth, catalysis and surface bound nanostructures[9]. More recently, the problem of modeling molecular motors[10], i.e., microscopic objects moving unidirectionally along periodic structures, has renewed the interest in the field and stimulated much theoretical work devoted to the study of the directed motion in a fluctuating environment in the absence of bias forces. One of the more recent research foci concerns the experimental and numerical observation that even for large clusters of molecules, long jumps spanning many lattice sites may in some cases be the dominant contributor to the motion[11].

In recent work, the long, even Le\'vy-like, motions can be described by ordinary Langevin dynamics in the low friction regime. Another related work has dealt with the transport properties of particles in a symmetric periodic potential subject to thermal effects and external forces involving important physical applications that include Josephson junctions, superionic conductors, colloidal spheres and polymers diffusing at interfaces among many others[12]. Several different groups have studied the diffusion of Brownian particle in a periodic field of force[13]. However, these studies focus on the fact that the motion in periodic potentials is necessarily diffusive on very long time scales. Restricting ourselves to the one dimensional case, the mean-square displacement is given by $<[x(t) - x(0)]^2>$ = $2Dt$ for large times $t$. Where $D$ is the diffusion constant. One can map this problem onto a quantum system, representing the motion of a quantum particle in a modulated or random potential[14].

On this basis, the long time diffusive properties, including the discontinuous dependence of the diffusion constant on the ‘wavelength’ of the inhomogeneities in quasiperiodic media, is related to the leading low frequency behavior of the density of states in the associated quantum system. There are the connections between the classical diffusion, localization, and intermittency[15].

In the present work, we study the transport of a brownian particle in the field of force derivable from a potential with $N$ number of minima with same barrier heights by using Fokker-Planck dynamics. We calculate exactly the Kramers’ time that the time scale to approach to equilibrium in this case and study how it depends on number of minima. By taking different model potential with $N$ number of minima and of same barrier heights, we have seen that Kramers time is independent of the shape of the potential. We have seen the localised behavior of the particle in the asymptotic time limit in this case. Then we take the limit of $N \rightarrow \infty$ to get the periodic potential and study the diffusive behavior. We calculate exactly
the diffusion coefficient in this case in the asymptotic time limit. We also study the transport in commensurate and incommensurate media.

II. LOCALIZATION IN THE POTENTIAL WITH \( N \) MINIMA

Let us consider, a brownian particle is moving in an external potential \( V(x) \). The overdamped motion of the position variable obeys the Langevin equation

\[
\dot{x} = -\Gamma \frac{\partial V}{\partial x} + \eta \quad (1)
\]

Where, \( \Gamma \) is the inverse function. \( \eta(t) \) is the random force with Gaussian distribution and its correlation function \( \langle \eta(t_1)\eta(t_2) \rangle = 2\epsilon \delta(t_1 - t_2) \). The time dependent probability distribution \( P(x, t) \) of the random force \( x(t) \) obeys the Fokker Planck equation

\[
\frac{\partial P}{\partial t} = \frac{\partial}{\partial x} \left( \frac{\partial V}{\partial x} P \right) + \epsilon \frac{\partial^2 P}{\partial x^2} \quad (2)
\]

which, with substitution,

\[
P(x, t) = \exp \left(-\frac{V}{2\epsilon} \right) \phi(x, t) \quad (3)
\]

reduces to Schrodinger like equation

\[
-\frac{\partial \psi}{\partial t} = \left[ -\epsilon \frac{\partial^2}{\partial x^2} + \left( \frac{V''}{4\epsilon} - \frac{V'''}{2} \right) \right] \phi \quad (4)
\]

The time independent equation reads,

\[
\left[ -\epsilon \frac{\partial^2}{\partial x^2} + \left( \frac{V''}{4\epsilon} - \frac{V'''}{2} \right) \right] \psi = \lambda \psi \quad (5)
\]

where, \( \psi(x, t) = e^{it\phi}(x, t) \).

1. Kramers’ time

   a. Case I: Let us start with a model potential with number of minima is two. Then generalize to \( N \) number of minima and periodic potential.

   Consider, the potential

   \[
   V(x) = \begin{cases} 
   V_0(1 - \frac{x}{a}), & 0 \leq x \leq a \\
   V_0(1 + \frac{x}{a}), & -a \leq x \leq 0 \\
   V_0(\frac{x}{a} - 1), & a \leq x \\
   -V_0(\frac{x}{a} + 1), & x \leq -a 
   \end{cases}
   \]

   Now, the Schrodinger like eq. (5) becomes with the above form of potential

   \[
   -\epsilon \frac{d^2 \psi_n}{dx^2} + \frac{V_0}{a} [\delta(x+a) - \delta(x) + \delta(x-a)] \psi_n = -k^2 \psi_n \quad (7)
   \]

   where,

   \[
   \lambda_n = \frac{V_0^2}{4a^2\epsilon^2} - k^2 \quad (8)
   \]

   Consider,

   \[
   \psi_n(x) = \begin{cases} 
   A_0 e^{-kx} + B_0 e^{kx}, & -a \leq x \leq 0 \\
   A_1 e^{-kx} + B_1 e^{kx}, & 0 \leq x \leq a \\
   A_2 e^{-k(x-a)}, & x \geq a 
   \end{cases}
   \]

   Now, apply the boundary condition at \( x = 0, a \) namely

   i) \( \psi \) is symmetric around \( x = 0 \).

   ii) \( \psi' \) is ordinarily continuous i.e. it suffers a discontinuity proportional to the strength of the potential.

   We get the following relation between the coefficients \( A_2 \) at \( x \geq a \) and \((A_0, B_0)\) at \(-a \leq x \leq 0\).

   \[
   \begin{pmatrix} A_2 \\ 0 \end{pmatrix} = \begin{pmatrix} e^{-ka} & e^{ka} \\ \alpha e^{-ka} & (1 + \alpha)e^{ka} \end{pmatrix} \begin{pmatrix} 1 + \alpha & \alpha \\ -\alpha & 1 - \alpha \end{pmatrix} \begin{pmatrix} A_0 \\ B_0 \end{pmatrix} \quad (10)
   \]

   where, \( \alpha = \frac{V_0}{2\epsilon k} \). \( \psi_n(x) \) is symmetric around \( x = 0 \) gives the relation

   \[
   B_0 = \frac{1 + \alpha}{1 - \alpha} A_0 \quad (11)
   \]

   With the help of (10) and (11) we get the recursion relation of \( k \)

   \[
   e^{2ka} \frac{V_0 - 2ak\epsilon}{V_0} = 1 \quad (12)
   \]

   In order get the first excited state eigenvalue we use

   \[
   k = \frac{V_0}{2\epsilon} + \delta \quad (13)
   \]

   and from eq. (12) we get

   \[
   \delta = -\frac{V_0}{2\epsilon} e^{-V_0/\epsilon} \quad (14)
   \]

   Now, from (15), we get the first excited state eigenvalue.

   \[
   \lambda_1 = \frac{V_0^2}{2a^2\epsilon^2} e^{-V_0/\epsilon} \quad (15)
   \]

   In the asymptotic time limit \( \lambda_1 \) gives the dominant contribution in the time dependent probability distribution. Hence the Kramers’ time that the time scale to approach to equilibrium is

   \[
   \tau_0 = \frac{2a^2\epsilon^2}{V_0} e^{-V_0/\epsilon} \quad (16)
   \]
Now, consider the number of minima is three and intervening two maxima are of equal height $V_0$. As in the previous case, Schrödinger like equation becomes
\[
-\frac{d^2\psi_n}{dx^2} + \frac{V_0}{a}[\delta(x+2a) - \delta(x+a) + \delta(x)] - \delta(x-a) + \delta(x-2a)]\psi_n = -k^2\psi_n
\]
with $\lambda_n = \frac{V_0^2}{4ae^2} - k^2$.

Consider,
\[
\psi_n = A_0 e^{-ka} + B_0 e^{ka}, \quad -a \leq x \leq 0
\]
\[
= A_1 e^{-ka} + B_1 e^{ka}, \quad 0 \leq x \leq a
\]
\[
= A_2 e^{-k(x-a)} + B_2 e^{k(x-a)}, \quad a \leq x \leq 2a
\]
\[
= A_3 e^{-k(x-2a)}, \quad x \geq 2a
\]
(18)

From (19) and (21), we get the following recursion relation of $k$
\[
\left(\frac{V_0}{2ae}\right)^2 e^{2ka} = -\left(1 - \frac{V_0}{2ae}\right) \left[1 + \left(1 - \frac{V_0}{2ae}\right) \frac{V_0^2}{2ae^2k^2}\right] e^{4ka}
\]
(20)

By substituting $k = \frac{V_0}{2ae} + \delta$ into eq.(21), we get
\[
d = -\frac{V_0}{2ae} \frac{V_0}{\epsilon}
\]
(21)

Hence, the Kramers’ time in this case is
\[
\tau_1 = \frac{4a^2e^2}{V_0^2} e^{V_0/\epsilon}
\]
\[
= 2\tau_0
\]
(22)

Now, we generalize the potential with $N$ minima. The Schrödinger like equation takes the form
\[
-\frac{d^2\psi_n}{dx^2} + \frac{V_0}{a} \sum_{i=-N}^{N} (-)^{i+1} \delta(x-(i-1)a)\psi_n = -k^2\psi_n
\]
(23)

The $\psi_n$ takes the following form
\[
\psi_n(x) = A_0 e^{-ka} + B_0 e^{ka}, \quad -a \leq x \leq 0
\]
\[
= A_1 e^{-ka} + B_1 e^{ka}, \quad 0 \leq x \leq a
\]
\[\vdots\]
\[
= A_i e^{-k(x-(i-1)a)}, \quad x \geq (i-1)a
\]
(24)

As in the previous case, by applying boundary conditions at $x = 0, a, \ldots, (i-1)a$, we get the following relation between coefficients $(A_i, 0)$ at $x \geq (i-1)a$ and $(A_0, B_0)$ at $-a \leq x \leq 0$.
\[
\begin{pmatrix}
A_i \\
0
\end{pmatrix} =
\begin{pmatrix}
e^{-ka} & (1 + \alpha)e^{ka} \\
\alpha e^{-ka} & (1 + \alpha)e^{ka}
\end{pmatrix}
\begin{pmatrix}
1 - \alpha & -\alpha \\
\alpha & 1 + \alpha
\end{pmatrix}
\begin{pmatrix}
A_0 \\
B_0
\end{pmatrix}
\]
The ground state becomes degenerate and band appears in the periodic potential. Consider the potential with minima two is of the form $V(x) = V_0 (x^2 - 1)^2$. To evaluate the Kramers’ time, we have to evaluate the first excited state eigenvalue of the Hamiltonian of the form $-\epsilon \frac{d^2}{dx^2} + \left( \frac{V''}{4} - \frac{V'''}{2} \right)$. It can be evaluated by calculating the ground state eigenvalue of its super symmetric partner Hamiltonian of the form $-\epsilon \frac{d^2}{dx^2} + \left( \frac{V''}{4} + \frac{V'''}{2} \right)$. With the help of trial wave function we can evaluate the ground state eigenvalue of the partner Hamiltonian of the order of $e^{-V_0}$. Hence the first excited state eigenvalue of the original Hamiltonian is of the order of $e^{-V_0}$. Now the Kramers’ time, the time scale to approach to equilibrium is of the order of $e^{-V_0}$.

Now, consider the potential with three minima and each maxima is of the same height as double well potential. In the similar way we can evaluate the Kramers’ time of the order of $2e^{-V_0}$, where each maxima gives the same contribution to the Kramers’ time.

As a generalization, in case of potential with $N$’ number of minima and each of the $N - 1$ maxima is of the same height as double well potential, the Kramers’ time is of the order of $(N - 1)e^{-V_0}$.

Hence the Kramers’ time reads

$$\tau_N = N(\tau_0), \quad \text{when } N \gg 1$$

(29)

Where, $\tau_0 \sim e^{-V_0}$ is the Kramers’ time of the potential with minima two.

2. Localised to diffusive behavior

The probability distribution $P(x,t)$ is given by

$$P(x,t) = P_{eq} + c_1 \phi_1(x)e^{-\lambda_1 t} + \cdots$$

(30)

where $\lambda_1$ is the first excited state eigenvalue.

The diffusion coefficient is given by

$$D = \frac{1}{2} \lim_{t \to \infty} \frac{\partial}{\partial t} \langle x^2 \rangle (t)$$

$$= \frac{1}{2} \lim_{t \to \infty} \frac{\partial}{\partial t} \int dx x^2 P(x,t)$$

$$= \frac{1}{2} \lim_{t \to \infty} \left[ -c_1 \lambda_1 e^{-\lambda_1 t} \int dx x^2 \phi_1(x) + \cdots \right]$$

(31)

As long as $N$ is finite, $\lambda_1$ is constant. The diffusion coefficient goes to zero exponentially in the asymptotic time limit. Now, as $N \to \infty$, the potential passes over to periodic potential. We will see in that case, the transition from localised to diffusive behaviour as $t \to \infty$. Clearly, as $N \to \infty$, the excited states eigenvalues tend to zero. The ground state becomes degenerate and band appears in the periodic potential.

The probability distribution in this case is given by

$$P(x,t) = P_{eq}(x) + \sum_n \int dk a_n e^{ikx} \psi_n(x) e^{-Dk^2 t}$$

$$= P_{eq}(x) + \sum_n a_n \psi_n(x) e^{-\frac{2}{k^2} \sqrt{\frac{\pi}{D t}}}$$

(32)

Where, we have used eigenvalue $\lambda_n(k) \sim k^2$. $n$ and $k$ are the band and Bloch index respectively. Hence the approach to equilibrium distribution $P_{eq}$ is power law type in $t$ in case of periodic potential.

III. DIFFUSIVE BEHAVIOR IN THE PERIODIC POTENTIAL

3. Diffusion coefficient

The Schrödinger like equation becomes

$$-\epsilon \frac{d^2 \psi_n}{dx^2} + \frac{V_0}{a} \left[ \sum_{x=-\infty}^{\infty} (-1)^n \delta(x - na) \right] \psi_n = -k^2 \psi_n$$

(33)

where, $\lambda_n = \frac{V_0^2}{4a^2} - k^2$.

The general solution is

$$\psi(x) = Ae^{k(x-2a)} + Be^{-k(x-2a)}$$

(2a \leq x \leq 3a)

Equation (34)

According to Bloch’s theorem, the wave function in the cell $0 \leq x \leq a$ is

$$\psi(x) = e^{-2ik'a} \left[ Ae^{kx} + Be^{-kx} \right]$$

(35)

At $x = 0$, $\psi$ is continuous and $\psi'$ suffers the discontinuity proportional to the strength of the potential. Hence we get the following relations between coefficients.

$$F = e^{-ka} \left[ (1 - \frac{V_0}{2ak\epsilon})A - \frac{V_0}{2ak\epsilon}B \right]$$

(36)

and

$$G = e^{ka} \left[ \frac{V_0}{2ak\epsilon}A + (1 + \frac{V_0}{2ak\epsilon})B \right]$$

(37)

Similarly, matching at $x = 2a$, we get the following relations

$$F = e^{-2ik'a} \left[ (1 - \frac{V_0}{2ak\epsilon})Ae^{ka} - \frac{V_0}{2ak\epsilon}Be^{-ka} \right]$$

(38)

and

$$G = e^{-2ik'a} \left[ \frac{V_0}{2ak\epsilon}Ae^{ka} + (1 + \frac{V_0}{2ak\epsilon})Be^{-ka} \right]$$

(39)
From (36), (37), (38) and (39) we find the recursion relation of $k$

$$\cos 2k^*a = p^2 + (1 - p^2) \cosh 2ka$$ \hfill (40)

Where $k^*$ is the Bloch index and $k$ is the band index. $p$ is given by $\frac{\lambda_0}{2\pi \epsilon a}$. By substituting $k = \frac{\lambda_0}{2\pi \epsilon a} + \delta$ in (40) we find

$$\delta = -\frac{k'^2 a V_0}{e} \frac{1}{2 \sinh^2 V_0/2\epsilon}$$ \hfill (41)

From $\lambda_n = \frac{V_0^2}{4\pi^2 \epsilon a^2} - k^2$, we get $\lambda_1 = \frac{k'^2 V_0^2}{2\pi^2 \epsilon a^2} \sinh^2 V_0/2\epsilon$.

Hence, the diffusion coefficient $D$ is given by

$$D = \frac{V_0^2}{2\epsilon^2 \sinh^2 V_0/2\epsilon}$$ \hfill (42)

Which is independent of period of the potential and only depends on the height of the potential. It can be written in the following form

$$D = \frac{V_0^4/8\epsilon^4}{<e^{V/\epsilon}> <e^{-V/\epsilon}>}$$ \hfill (43)

where, $\sinh^{-1} \frac{V_0}{\epsilon} = \frac{V_0^2/4\epsilon^2}{<e^{V/\epsilon}> <e^{-V/\epsilon}>}$.

$$\frac{1}{2\pi} \int_{-a}^{a} e^{V/\epsilon} dx$$ where, in the range $0 \leq x \leq a$, $V(x) = V_0(1 - x/a)$ and in the range $-a \leq x \leq 0$, $V(x) = V_0(1 + x/a)$.

4. Transport in commensurate and incommensurate media

We consider the potential $V(x) = V_1(x) + V_2(x)$. Where, $V_2(x+2a) = V_2(x)$ and $V_1(x+na) = V_1(x)$. $V(x)$ has a period $2\pi n$. We will see how diffusion coefficient depends on $n$.

Consider, the potential $V_1(x)$ is of the following form

$$V_1(x) = V_0(1 - \frac{x}{a}), \quad 0 \leq x \leq a$$

$$= -V_0(1 - \frac{x}{a}), \quad a \leq x \leq 2a$$

$$= 3V_0(1 - \frac{x}{3a}), \quad 2a \leq x \leq 3a$$

$$= -3V_0(1 - \frac{x}{3a}), \quad 3a \leq x \leq 4a$$

$$\vdots$$

$$= -(m-2)V_0(1 - \frac{x}{(m-2)a}), (m-2)a \leq x \leq (m-1)a$$

$$= mV_0(1 - \frac{x}{ma}), \quad (m-1)a \leq x \leq ma$$ \hfill (44)

and $V_2(x)$ is of the following form

$$V_2(x) = V_0'(1 - \frac{x}{na}), \quad 0 \leq x \leq na$$

$$= V_0'(1 + \frac{x}{na}), \quad -na \leq x \leq 0$$ \hfill (45)

Now, $<e^V> <e^{-V}>$ takes the following form

$$<e^V> <e^{-V}> =$$

$$\frac{1}{2na} \left\{ \frac{1}{1 - \frac{V_0}{\alpha} + \frac{V_0'}{\alpha}} \right\} \left[ \frac{1 - e^{-2\frac{V_0'}{\alpha}}}{1 - e^{-2V_0'/n}} \left( \frac{2n - 2V_0'}{n} - e^{2\frac{V_0'}{\alpha} - e^{V_0' + \frac{2n - 1}{n} V_0'}} \right) + \frac{1}{\frac{V_0}{\alpha} - \frac{V_0'}{\alpha}} \left( \frac{1 - e^{-2\frac{V_0'}{\alpha}}}{1 - e^{-2V_0'/n}} \left( -e^{-\frac{2n + 2}{n} V_0'} + e^{V_0' + \frac{2n + 1}{n} V_0'} \right) \right) \right\}$$

$$\frac{1}{2na} \left\{ \frac{1}{\frac{V_0}{\alpha} + \frac{V_0'}{\alpha}} \right\} \left[ \frac{1 - e^{2\frac{V_0'}{\alpha}}}{1 - e^{2V_0'/n}} \left( e^{-\frac{2n + 2}{n} V_0'} - e^{-V_0' - \frac{2n - 1}{n} V_0'} \right) + \frac{1}{\frac{V_0}{\alpha} + \frac{V_0'}{\alpha}} \left( \frac{1 - e^{2\frac{V_0'}{\alpha}}}{1 - e^{2V_0'/n}} \left( -e^{-\frac{2n + 2}{n} V_0'} + e^{-V_0' - \frac{2n + 1}{n} V_0'} \right) \right) \right\} \hfill (46)$$

In the large $n$ limit, the potential $V(x)$ still periodic, and we find the finite diffusion coefficient.

When $n$ goes to zero, the potential varies very rapidly. Potential becomes quasiperiodic and localization of states occur. Particle becomes localised in the asymptotic time limit, hence the diffusion coefficient goes to zero.

IV. CONCLUSION

In conclusion, we have studied how Kramers’ time depends on the number of minima of the model potential. We have seen that it is independent of the shape of the potential. The localized behavior in this model potential is studied when the number of minima is finite. In this case we have seen the diffusion coefficient goes to zero exponentially in the asymptotic time limit. In the limit of
infinite number of minima in our model potential we have seen the transition from localized to diffusive behavior. Approach to equilibrium in the case of periodic potential is power law type in time. In the periodic field of force we have calculated exactly the diffusion coefficient of the Brownian particle. In case of rational periodic mixing we have seen the diffusive behavior of the Brownian particle. But in the case of irrational mixing we have seen the localized behavior of the Brownian particle.

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