Starting from the Caldeira-Leggett (CL) model \cite{1}, we derive the equation describing the Quantum Brownian motion, which has been originally proposed by Dekker \cite{2} purely from phenomenological basis containing extra anomalous diffusion terms. Explicit analytical expressions for the temperature dependence of the diffusion constants are derived. At high temperatures, additional momentum diffusion terms are suppressed and classical Langevin equation can be recovered and at the same time positivity of the density matrix (DM) is satisfied. At low temperatures, the diffusion constants have a finite positive value, however, below a certain critical temperature, the Master Equation (ME) does not satisfy the positivity condition as proposed by Dekker.

The problem of Quantum Brownian Motion is a long-standing and challenging problem \cite{3, 4}, as it forms the underlying basis of non-equilibrium phenomena such as dissipative and relaxation dynamics of quantum systems \cite{5, 6}. Quantum dissipative dynamics has application in a wide variety of problems, starting from quantum cosmological models \cite{7, 8} to reaction rate theory \cite{9}. Unlike the classical nonequilibrium problem, quantum dynamics has additional complexity due to the Heisenberg Uncertainty Principle. Numerous attempts have been made to address this problem, in a variety of ways. These include semiclassical approaches using Wigner distribution function \cite{10}, phenomenological models \cite{2, 11, 12}, and using Boltzman’s collision terms \cite{13}. On the other hand, there have been attempts to obtain the Master Equation (ME) describing the time evolution of DM of open quantum systems through toy microscopic models \cite{1, 7, 14}, where the environment is modelled by noninteracting particles or fields in thermal equilibrium. The ME is obtained for the reduced density matrix (DM) of the system by integrating out the environment degrees of freedom. New form of ME’s are obtained from microscopic models where the diffusion constants have explicit time dependence \cite{4, 15}. Independently, a class of ME’s have been put forward, \cite{16} purely from mathematical consideration, which guarantee the positivity of the density matrix. In other words, the time evolution of the system takes place only through the physical states, and the ME is commonly known to belong to the Lindblad class. Most of the MEs describing the non-equilibrium quantum dynamics suffers from the fact that either they do not belong to Lindblad class or correct classical limit can not be recovered at high temperatures. The ME derived in the original paper by CL \cite{1}, does not satisfy the positivity condition at high temperatures, although it recovers the classical Langevin dynamics at high temperatures.

Independently a phenomenological model of quantum Brownian dynamics has been proposed by Dekker \cite{2}, which contains extra terms describing momentum diffusion apart from ordinary diffusion, and the diffusion constants have to satisfy some conditions in order to preserve the Heisenberg’s uncertainty condition as well as positivity of the DM \cite{17, 18}. However Dekker’s model of quantum Brownian motion is purely phenomenological and the microscopic origin of the extra momentum diffusion constants are not known. Moreover, most of the derivations of the ME’s rely on the fact that the autocorrelation function of the random force on the Brownian particle is short ranged at high temperature and that the system is assumed to be Markovian. But in practice, the effective action obtained from CL model has a memory kernel which decays as a power law at low temperatures \cite{19}. In fact, the random force in Quantum Brownian Motion is an operator and the force-force autocorrelation function has been evaluated exactly, which shows clear deviation from the Markovian limit \cite{20}. This non-locality makes the problem very complex. In this letter we take a new approach, to analytically derive Dekker’s form of quantum Brownian motion, starting from the CL model, where the analytical expressions of the diffusion constants are obtained for all temperatures. We show that at high temperatures, the ME satisfies the positivity condition and at the same time reproduces the classical Langevin dynamics. Although, the diffusion constants are finite and well behaved at all temperatures, the positivity of DM breaks down below a certain temperature depending on the damping rate of the system. This is not very unexpected, since we know that at low temperatures, the long range memory effect becomes very important. This form of quantum Brownian motion is applicable to describe the relaxation phenomena and nonequilibrium evolution of quantum systems, even at sufficiently low temperatures. We propose a new method to tackle the problem of nonlocality, using the equation of motion of the canonical coordinates. In this letter, we mainly focus on the quantum brownian dynamics in free space, however, our technique can, in principle, be extended to confined quantum particles.

A very well known description for the dissipative phenomena and relaxation dynamics of classical system is given by the Langevin equation:

\[ \ddot{q}(t) = -\gamma \dot{q}(t) + \theta(t) \] (1)
where, \( q(t) \) is the position of the Brownian particle, the dot denotes derivative with respect to time, \( \gamma \) is the damping constant and \( \theta(t) \) is related to the fluctuating force acting on the particle, whose autocorrelation function is given by,

\[
\langle \theta(t)\theta(t') \rangle = \Gamma \delta(t - t')
\]

(2)

where \( \Gamma \) and \( \gamma \) are related by the Fluctuation-Dissipation Theorem, which states that \( \Gamma = 2k_B T M \gamma \), where \( T \) is the temperature of the bath and \( M \) is the mass of the Brownian particle. However, in most cases, the microscopic details behind this dissipation are not well known.

The simplest microscopic model which describes the dissipative motion of a heavy particle in presence of a heat bath was first put forward by Ford, Kac and Mazur [21] and later applied to quantum systems by Caldeira and Legget [1]. In this model, full Hamiltonian has three different parts, system Hamiltonian \( H_A \), the Hamiltonian of the heat bath \( H_B \) and the Hamiltonian \( H_I \) describing the linear coupling between the system and the bath, as given below

\[
H_A = \frac{\gamma^2}{2M} + V(q)
\]

(3)

\[
H_B = \sum_i \left( \frac{P_i^2}{2m_i} + \frac{1}{2}m_i\omega_i^2 Q_i^2 \right)
\]

(4)

\[
H_I = q \sum_i C_i Q_i
\]

(5)

where, \( q \) and \( p \) are the position and momentum operators of the heavy particle having mass \( M \), \( V(q) \) is the potential in which the heavy particle is moving, \( Q_i \) and \( P_i \) are the position and momentum operators of the bath oscillators whose mass and frequency of oscillation are given by \( m_i \) and \( \omega_i \) respectively and \( C_i \) is the coupling strength between the heavy mass and the \( i^{th} \) oscillator in the bath. Following the work of Ford et. al., the time evolution of the position operator can be written as

\[
M\ddot{q} + \int_0^t dt' \alpha(t - t')\dot{q}(t') + V'(q) = F(t)
\]

(6)

where the prime denotes the derivative with respect to the position variable. The operator valued random force, \( F(t) \) is related to the statistical distribution of the bath variables \( Q_i \) and \( P_i \)’s. In order to recover the classical Brownian dynamics with short time memory effect at high temperatures, the frequency dependence of the coupling is typically taken to be \( C^2(\omega) / (m_i\omega_i^2) \propto M\gamma \), where \( \gamma \) is the damping constant, and the summation is then replaced by an integral over \( \omega \). According the Ehrenfest theorem, if the position and momentum operators are replaced by their classical equivalents, then Eq. 6 translates to the classical Langevin Equation in the limit of \( \hbar \rightarrow 0 \), where the force autocorrelation function takes the form of Eq. 2 with \( \Gamma = 2M\gamma k_B T \) obeying classical fluctuation-dissipation theorem.

The essential difference between the classical Langevin equation and the Quantum Master Equation is the fact that in the classical case, the fluctuations in canonical coordinates are controlled only by the scales set by the temperature and the dissipation constant. Whereas, in the Quantum case, there is an additional scale given by \( \hbar \) which appears due the Heisenberg Uncertainty Principle. Moreover, the evolution of the operators themselves have to be unitary. Apart from the semiclassical techniques, new methods have been proposed to take into account the constraints imposed by ‘uncertainty principle’ in the quantum case [22]. Among these methods, ME of the density matrix \( \rho \) is more suitable to describe dissipative dynamics of quantum systems. The quantum dynamics which is governed by the time dependent Schrödinger equation describes the pure state, whereas the dissipative dynamics introduces the concept mixed state, where trace of \( \rho^2 \) is less than unity. The evolution equation of the reduced density matrix of the system can be obtained by tracing out the bath degrees of freedom.

Using the Feynman-Vernon method [23], the dissipative dynamics of the reduced DM can be written in terms of the influence functional, which can be obtained after integrating out the bath degrees of freedom \( Q_i \). The main assumption of this method is that the subsystem is uncorrelated with the bath at the initial time. Hence the total density matrix at time \( t = 0 \) is given by \( \rho_T(0) = \rho_A(0) \otimes \rho_B(0) \), where \( \rho_A \), \( \rho_B \) and \( \rho_T \) are the density matrix of the subsystem, bath and the total system respectively. However, several authors have considered a correlated initial state, where the subsystem and bath density matrix cannot be factorized. For example, Hakim and Ambegaokar [24] have compared the two cases of uncorrelated and correlated initial conditions and shown that, for the correlated initial condition, different transient behaviours can be obtained at time scales larger than the inverse cutoff frequency of the bath, in contrast to the uncorrelated initial conditions. However, here we are following the usual procedure of Feynman and Vernon and we will show finally that we can obtain a consistent ME, describing the time evolution of the DM. The time evolution of the reduced DM is given by,

\[
\rho(q_1, q_2, t) = \int dq_1' dq_2' J(q_1, q_2, t; q_1', q_2', 0) \rho(q_1', q_2', 0)
\]

(7)

where the propagator \( J(q_1, q_2, t; q_1', q_2', 0) \) is given by,

\[
J(q_1, q_2, t; q_1', q_2', 0) = \int Dq_1 Dq_2 \exp\left( \frac{i}{\hbar} S_{EH}[q_1, q_2] \right)
\]

(8)

After integrating out the bath degrees of freedom, one obtains the nonlocal effective action corresponding to the
dissipative system:

\[
\frac{i}{\hbar}S_{\text{eff}} = \frac{i}{\hbar}(S_A[q_1] - S_A[q_2]) + \int_0^t (\Sigma_R + \Sigma_I) d\tau \quad (9)
\]

where \( S_A \) is the action corresponding to the noninteracting system, and,

\[
\Sigma_R = -\frac{1}{\hbar} \int_0^t [q_-(\tau)\alpha_R(\tau - s)q_-(s)] ds \quad (10)
\]

\[
\Sigma_I = -\frac{i}{\hbar} \int_0^t [q_-(\tau)\alpha_I(\tau - s)q_+(s)] ds
\]

where, \( q_\pm = q_1 \pm q_2 \) and the memory kernels are,

\[
\alpha_R(\tau) = \sum_i \frac{C_i}{2m\omega_i} \coth \left( \frac{\hbar\omega_i}{2k_B T} \right) \cos(\omega_i \tau)
\]

\[
\alpha_I(\tau) = -\sum_i \frac{C_i}{2m\omega_i} \sin(\omega_i \tau)
\]

It is assumed that the oscillator frequencies are continuously distributed from zero to a maximum frequency \( \omega_c \), value of which depends on a specific physical system. Cutoff frequency of the heat bath is chosen in such a way that the characteristic time scale of the dynamics of heavy particle is much larger than the collisional time scale \( 1/\omega_c \). Thus, the summations in Eq. 12 can be replaced by integrals by introducing an appropriate density of state, which will make the memory function analytically tractable. Hence, \( \Sigma_I \) can be replaced by \( \int d\omega F(\omega) \), where \( F(\omega) \) is the density of states of the bath. We choose a smooth Drude form for the density of states given by \( F(\omega) = \omega_c^2 / (\omega^2 + \omega_c^2) \) and \( F(\omega) C^2(\omega)/(2m\omega_i^2) = (2M\gamma / \pi)(\omega_c^2 / (\omega^2 + \omega_c^2)) \). At any non-zero temperature, using the Drude form of density of states the memory kernels can be evaluated analytically by using contour integrals in complex \( \omega \) plane and are given by:

\[
\alpha_R(\tau) = M\gamma\omega_c^2 \cot(\chi) \exp(-\omega_c \tau)
\]

\[
+ \frac{2}{\chi} \sum_{n=1}^{\infty} \frac{n\pi / \chi}{(n\pi / \chi)^2 - 1} \exp\left[-\left(\frac{n\pi}{\chi}\right)\omega_c \tau\right]
\]

\[
\alpha_I(\tau) = M\gamma\omega_c \frac{\partial}{\partial \tau} \exp(-\omega_c \tau)
\]

where \( \chi = \hbar\omega_c / 2k_B T \). From the above expression of \( \alpha_R \) we can clearly see the emergence of two time scales - the first being the microscopic collision time, given by \( 1/\omega_c \) and the other time scale given by temperature i.e. \( \hbar/k_B T \). It is interesting to note that in the high temperature regime, \( \alpha_R \) is short ranged over the thermal time scale while the timescale over which \( \alpha_I \) decays is given only by the collision time scale, which is independent of temperature. However, as the temperature is reduced, at some point, the thermal time scale dominates over the collision time scale, giving rise to a nonlocal memory kernel which has a power law decay.

We now proceed to simplify the nonlocal action, taking advantage of the above mentioned short-ranged kernel. The dynamics over a timescale larger than \( \hbar/k_B T \) can be studied by assuming smooth classical trajectories and expanding the dynamical variables in a Taylor series,

\[
q_{\pm}(s) = \sum_{l=0}^{\infty} \frac{q_{\pm}^{(l)}(\tau)}{l!} (s - \tau)^l
\]

where, \( q_{\pm}^{(l)} \) denotes the \( l \)th derivative of \( q \) with respect to time. We now insert Eq. 12 and Eq. 14 into Eq. 10. Neglecting the total derivative terms which generate the boundary terms in the action, thereby leaving the Lagrangian invariant, we obtain,

\[
\Sigma_R(\tau) = -\frac{1}{\hbar} \sum_{l=0}^{\infty} \frac{(-1)^l}{2l!} \left( q_{\pm}^{(l)}(\tau) \right)^2 \int_0^T \tilde{\tau}^2 \alpha_R(\tilde{\tau}) d\tilde{\tau}
\]

The transient term in the integral comes in the form of \( \exp(-(k_B T / \hbar) \tilde{\tau}) \). Thus, for \( \tau \gg \hbar/k_B T \), we obtain,

\[
\Sigma_R(\tau) = -\frac{M\gamma\omega_c}{\hbar\chi} \times
\]

\[
\left[ q_\pm^{(2)}(\tau) - 2 \sum_{l=1}^{\infty} \frac{q_{\pm}^{(l)}(\tau)^2}{\omega_c^2} \sum_{m=0}^{l} \frac{1}{\pi} \zeta(2m) \right]
\]

Similarly, one can evaluate \( \Sigma_I \) which gives,

\[
\Sigma_I(\tau) = -\frac{\gamma M}{\hbar} q_-(\tau)q_+(\tau)
\]

where, the higher order terms in the expansion of \( q_{\pm}(s) \) have been neglected, since they fall off as \( 1/\omega_c \).

We have thus transformed a highly nonlocal action into a local action containing higher derivative terms. While in general, this may seem to be a very complicated expression to work with, we show below that in at least the case of a particle in a harmonic potential attached to a heat bath and a Brownian particle there is considerable simplification of the expression which allows us to obtain exact results.

For the free particle, general dynamical equation for any order of time derivative of the position coordinate can be written as,

\[
q^{(n)} = \frac{(-\gamma)^{n-1}}{M} p
\]

Substituting Eq. 18 in Eq. 10, we obtain,

\[
\Sigma_R(\tau) = -\frac{M\gamma\omega_c}{\hbar\chi} q_-^{(2)}(\tau)
\]

\[
- \frac{M\gamma\omega_c}{\hbar\chi} \left[ q_-^{(2)}(\tau) \left\{ \frac{\hbar\omega_c}{\omega_c} \coth \left( \frac{\hbar\omega_c}{\omega_c} \right) - 1 \right\} \right]
\]
Thus, the effective action is given by,

\[
\frac{i}{\hbar} S_{\text{eff}} = \frac{i}{\hbar} \int_0^t dt \left[ \frac{M}{2} \dot{q}_+ \dot{q}_- - \gamma M q_- \dot{q}_+ \right] - \frac{2 k_B T \gamma M}{\hbar^2} \int_0^t dt \left[ q_-^2 + \alpha q_+^2 \right]
\]

(20)

where \( \alpha = \left[ \hbar \omega_c \coth(\hbar \omega_c / 2k_B T) \right] - 1 / \gamma^2 \), assuming \( \gamma \ll \omega_c \).

It is important to note that by using the dynamical equation of canonical coordinates and resummation method we have converted the non-local action into an effective action which is local and quadratic in canonical coordinates as well as independent of the cut-off frequency \( \omega_c \) of bath oscillators. Inserting the above form of the effective action into Eq. [5] the time evolution of the DM can be evaluated.

Following the prescription of CL [1], we consider the change of density matrix from \( t \) to \( t + \epsilon \) within a small time interval \( \epsilon \) in order to obtain the ME in differential form. To do so we expand both side of Eq. [7] up to leading order in \( \epsilon \). Within the small time \( \epsilon \), we approximate

\[
\dot{q}_1 = (x - x') / \epsilon = \beta_1 / \epsilon \quad \text{and} \quad \dot{q}_2 = (y - y') / \epsilon = \beta_2 / \epsilon.
\]

Now Eq. [7] reads:

\[
\rho + i \hbar \frac{\partial \rho}{\partial t} = \int d\beta_+ d\beta_- \exp \left[ \frac{i}{\hbar \epsilon} M \beta_+ (\beta_- - 2 \gamma x - \epsilon) \right] - \frac{2 k_B T \gamma M}{\hbar^2} (x^2 + \alpha \beta_+^2) \rho(x - \beta_1, y - \beta_2, t).
\]

(21)

where \( \beta_+ = (\beta_1 + \beta_2) / 2 \) and \( \beta_- = \beta_1 - \beta_2 \). We expand \( \rho \) up to second order in \( \beta \), which is equivalent to expanding up to first order in \( \epsilon \). After doing some algebra and performing the gaussian integrals we obtain the ME describing the time evolution of the density matrix:

\[
\frac{\partial \rho}{\partial t} = \frac{i}{2M} \left[ \frac{\partial^2 \rho}{\partial x^2} - \frac{\partial^2 \rho}{\partial y^2} - \gamma (x - y) \left( \frac{\partial \rho}{\partial x} - \frac{\partial \rho}{\partial y} \right) \right] + \frac{2n}{\hbar} D_{pp}(x - y) \left( \frac{\partial \rho}{\partial x} + \frac{\partial \rho}{\partial y} \right) + D_{qq} \left( \frac{\partial \rho}{\partial x} + \frac{\partial \rho}{\partial y} \right)^2 \rho
\]

\[\] - \frac{D_{pp}}{\hbar^2} (x - y)^2 \rho.

(22)

where, different diffusion constants are given by:

\[
D_{qq} = \frac{2 k_B T}{M \gamma} \left[ \frac{\hbar \gamma}{2 k_B T} \coth \left( \frac{\hbar \gamma}{2 k_B T} \right) \right] - 1
\]

(23)

\[
D_{pq} = 4 k_B T \left[ \frac{\hbar \gamma}{2 k_B T} \coth \left( \frac{\hbar \gamma}{2 k_B T} \right) \right] - 1
\]

(24)

\[
D_{pp} = 2 M \gamma k_B T \left[ \frac{2 \hbar \gamma}{k_B T} \coth \left( \frac{\hbar \gamma}{k_B T} \right) - 3 \right]
\]

(25)

The above form of the ME has been proposed earlier phenomenologically [2] and the diffusion constants have to satisfy some conditions, in order to preserve the Heisenberg Uncertainty Principle in quantum dissipative systems [17]. This form of the ME can also be recast into the Lindblad form [18] which maintains the positivity of DM during time evolution. Commonly known positivity condition is described by [17,18]:

\[
\Delta = D_{pp} D_{qq} - D_{pq}^2 - \frac{\hbar^2 \gamma^2}{4} \geq 0
\]

(26)

At high temperatures, the diffusion constants behave as,

\[
D_{pp} = 2 M \gamma k_B T \left[ \frac{2}{3} \left( \frac{\kappa}{T} \right) \right]^2, \quad D_{qq} = \frac{\hbar^2 \gamma}{6 M k_B T}
\]

and

\[
D_{pq} = \frac{\hbar^2 \gamma}{3 k_B T}.
\]

The forms of these diffusion constants are very similar to those obtained earlier [11]. However, we would like to point out that the diffusion constant \( D_{pq} \), in our case, is independent of the cutoff parameter \( \omega_c \), unlike the earlier result. It is also interesting to note that the diffusion constants \( D_{qq} \) and \( D_{pp} \) both originate from quantum effects and vanish at high temperatures as \( 1 / k_B T \). However, at high temperatures, \( \Delta \) approaches a value \( \hbar^2 \gamma^2 / 12 \) which preserves the positivity condition of the ME. At zero temperature, the diffusion constants are finite, positive and proportional to \( \hbar \), similar to the form proposed by Dekker [2]. However, \( \Delta \) becomes negative at zero temperature, violating the positivity criterion. We estimate a critical temperature \( T_0 \sim 0.2 \gamma \), below which above form of ME is not valid and transient behaviour as well as long range memory effects become crucial.

To summarise, within our scheme of calculation, we have shown that the ME obtained from the microscopic CL model satisfies the positivity condition and belong to the Lindblad class above a certain temperature. Analytical form for the diffusion constants have been obtained for any arbitrary temperature and are independent of the cutoff frequency of the heat bath. It is interesting to note that at high temperatures all the anomalous diffusion constants vanish as \( 1 / T \), which preserves the structure of classical Brownian motion. At the same time, the diffusion constants conspire in such a way that they satisfy the Dekker criterion \( \Delta > 0 \), hence the time evolution of the open quantum system takes place only through physical states. At low temperature, the diffusion constants have their origin from purely quantum effects. However, below the temperature \( T \sim \hbar \gamma \), Dekker’s positivity condition is violated, indicating that the long range memory effect may become important in the time evolution of the system.

We like to thank P. K. Panigrahi and S. Dattagupta for helpful discussions and critical reading of the manuscript.

[1] A. O. Caldeira and A. J. Leggett, Physica (Amsterdam) 121A, 587 (1983).
[2] H. Dekker, Phys. Rep. 80, 1 (1981).
[3] P. Hänggi, G.-L. Ingold, Chaos 15, 026105 (2005) and references therein.
[4] G. W. Ford, J. T. Lewis and R. F. O’Connell, Phys. Rev. A 37, 4419 (1988)
[5] S. Dattagupta and S. Puri, Dissipative Phenomena in Condensed Matter: Some Applications, Springer (Berlin), (2004).
[6] Ulrich Weiss, Quantum Dissipative Systems, World Scientific, Singapore (1993).
[7] W. G. Unruh and W. H. Zurek, Phys. Rev. D 40, 1071 (1989).
[8] Jonathan J. Halliwell, Phys. Rev. D 39, 2912 (1989).
[9] Peter Hänggi, Peter Talkner, Michal Borkovec, Rev. Mod. Phys. 62, 251 (1990)
[10] S. K. Banik, B. C. Bag, and D. S. Ray, Phys. Rev. E 65, 051106 (2002).
[11] L. Diosi, Physica (Amsterdam) 199A, 517 (1993); L. Diosi, Europhys. Lett. 22, 1 (1993).
[12] S. Gao, Phys. Rev. Lett. 79, 3101 (1997).
[13] B. Vacchini, Phys. Rev. Lett. 84, 1374 (2000).
[14] A. J. Leggett et. al., Rev. Mod. Phys. 59, 1 (1987).
[15] B. L. Hu, Juan Pablo Paz, and Yuhong Zhang, Phys. Rev. D 45, 2843 (1992).
[16] G. Lindblad, Commun. Math. Phys. 48, 119 (1976).
[17] M. C. Valsakumar and H. Dekker, Phys. Lett. A 104, 67 (1984).
[18] J G Peixoto de Faria et. al., J. Phys. A 31, 7095 (1998).
[19] A. O. Caldeira and A. J. Leggett, Ann. Phys. (N.Y.) 149, 374 (1983).
[20] G. W. Ford and R. F. O’Connell, Phys. Rev. A 61, 22110 (2000)
[21] G. W. Ford, M. Kac and P. Mazur, Jour. Mathe. Phys. 6, 504 (1965).
[22] I. R. Senitzky, Phys. Rev. 119, 670 (1960).
[23] R. P. Feynman and F. L. Vernon, Jr., Ann. Phys. (N. Y.) 24, 118 (1963).
[24] V. Hakim and V. Ambegaokar, Phys. Rev. A 32, 423, (1985).