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Review Article

Quantum Dissipation in a Spin Bath; Applications to Chemical Dynamics

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Dissipation is an integral part of a quantum system coupled to its environment. Traditionally in physical and chemical sciences, the environment is modeled by a reservoir of harmonic oscillators. However, there arise situations where the localized modes dominate in the dynamics of dissipation. Spin-bath has proven to be a successful alternative in such cases. In this review we have outlined a scheme for a universal description of an environment in terms of a generalized spin-bath. Making use of Holestein-Primakoff transformation one may realize the two limits of the reservoir, the spin-1/2 or fermionic bath and the harmonic or bosonic bath. The basis of the present analysis is the coherent state representation of the noise operators and thermal canonical distributions of Gaussian form. The appropriate quantum Langevin equation and its variant in c-numbers have been derived for spatial and momentum coupling between the system and the bath. The scheme has been applied to the problems of rate theory of chemical reactions, tunneling and spectroscopy with quantum dots.

Key Words : Quantum Dissipation; Spin Bath; Thermal Activation; Spectroscopy with Quantum Dots

Introduction

Preliminary Remarks; Classical Dissipation

It is our everyday experience that any macroscopic physical system cannot be completely isolated from its surrounding. As a consequence energy accumulated in a system must be given away to the environment — leading to energy dissipation. This energy loss is described in terms of the frictional force arising out of the coupling between the system and its surrounding. The open system is also subjected to a fluctuating force or noise of the same origin. The dissipation and noise are formally connected by fluctuation-dissipation theorem. This forms the basis for the theory of Brownian motion since the pioneering work of Einstein (Einste in, 1905; Einstein, 1906) and further advanced by Langevin (Langevin, 1908), Fokker (Fokker, 1914), Planck (Planck, 1917), Smoluchowskii (Smoluchowskii, 1915), Kramers (Kramers, 1940) and others in various directions (Grabert *et al.*, 1988; Hänggi *et al.*, 1990). The early work on Brownian motion was reviewed in an excellent article by Chandrasekhar (Chandrasekhar, 1943) and the theory of nonequilibrium stochastic processes grew as a major discipline in physical and chemical sciences (Lindenberg and West, 1990; van Kampen, 1992).

Approach to Quantum Dissipation; Historical Development

Several approaches to open quantum systems are known for the last forty years. One of the most acceptable and successful approaches among them relies on the system-reservoir model within the framework of a Lagrangian or a Hamiltonian (Ford *et al.*, 1965; Ullersma, 1966a; Ullersma, 1966b; Ullersma, 1966c; Ullersma, 1966d; Zwanzig, 1973; Dekker, 1977), where a dissipative system with a

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single degree of freedom remains in contact with infinitely many degrees of freedom acting as a bath or reservoir for the relevant system of interest. Both the system and the reservoir obey the standard rules of quantization and form the constituents of an energy conserving global system. In this picture, dissipation appears as a manifestation of transfer of energy from the system to the environment and the energy never goes back into the system within any physically relevant timescale.

The early development of quantum stochastic processes along the above mentioned line took place around the middle of twentieth century. A major thrust was the development of lasers in sixties followed by significant advances in the field of laser physics (Sargent *et al.*, 1974) and quantum optics (Meystre and Sargent, 1991). Two general and popular approaches based on system-reservoir model had been prescribed with wide applications. These are the generalized quantum master equations of the reduced density operator (Weidlich and Haake, 1965; Ullersma, 1966b; Agarwal, 1969; Carmichael, 1970) in the Schrodinger picture and the generalized Langevin equations (Ulersma, 1966a; Zwanzig, 1973)in the Heisenberg picture.

In the case of master equation approach, it is often impossible to analytically determine the time evolution of the reduced density operator for the system interacting with an environment. In such cases, one may adopt approximate schemes.The work of Schwinger (Schwinger, 1961) and Senitzky (Senitzky, 1960; Senitzky, 1961) on the dissipative effects in electromagnetic fields within Born-Markov approximation was an early attempt in this direction. Further, in a series of papers, Lax (Lax 1963; Lax 1966; Lax, 1967) analysed the problem of quantum noise, particularly the conditions that would permit Markovian description of the problem. Interestingly, in almost all physically relevant situations in quantum optics it is sufficient to work within weak coupling limit (Born approximation) and Markov approximation (i.e., the correlation time of the reservoir << characteristic time of the system). On the other hand, one may recast the same problem in the Heisenberg picture since the latter has the appealing feature over the Schrödinger picture that the equations of motion look like the corresponding classical equations of motion. But this also has a few drawbacks. The most important one is the ordering of operators. System operators commute with bath operators at equal times, but not necessarily at different times. Therefore the order of system and bath operators can no longer be interchanged. It is suggested that we must stick to one particular ordering of the operators. Any ordering namely normal, anti-normal, symmetrical will produce the same final results provided that it should be used consistently throughout the calculation. Although, final answers do not depend on the choice of the ordering, the physical interpretation of the results does (Milonni and Smith, 1975). After all this approach has the same limitations as the master equation approach (Louisell, 1973).

The validity of quantum theory of dissipation discussed so far is thus restricted to weak coupling and Markovian regime. Although some useful results in quantum optics, e.g., intensity-intensity correlation functions, calculation of spectra have been derived within Born-Markov approximation, but the validity and limitations of these approximations have been examined in the context of condensed matter and chemical physics. It was realized that the strength of friction as well as the nature of the noise correlation play a key role in the dynamics, particularly at low temperature. As an attempt to go beyond the weak coupling limit, theory of quantum dissipation emerged as a renewed subject of interest in early eighties when the problem of macroscopic quantum tunneling at zero temperature was addressed by Leggett and Caldeira (Caldeira and Leggett, 1981). The method that received major attention in a wide community of researchers is the real time functional integral (Feynman and Hibbs, 1965). This method had been shown to be an effective tool (Caldeira and Leggett, 1983a; Caldeira and Leggett, 1983b; Leggett *et al.*, 1987; Grabert *et al.*, 1988; Leggett *et al.*, 1995; Weiss, 1999) for the treatment of dissipative quantum coherence effects, quantum transition states, as well as incoherent quantum tunneling processes and many others. Numerical implementation of the schemes such as path integral Monte Carlo techniques (Binder and Heermann, 1988) has been popular in some of these issues.

Keeping in view of the aforesaid development it is worthwhile to ask is there any natural extension of the methods of classical theory of dissipation to quantum domain for arbitrary friction and temperature down to vacuum limit? An answer to this question has been addressed through quasi probability distributions. Since the uncertainty principle makes the concept of phase space in quantum mechanics problematic the functions which bear some resemblance to phase space distribution functions, are the quasi probability functions (Glauber, 2007). The whole class of distribution functions allows us to replace the solution of a density matrix equation by the solution of a classical differential equation; e.g., the quantum statistical equations of motion in the form of Fokker-Planck equation is the evolution equation for Wigner (Wigner, 1932; Hillery *et al.*, 1984) or Glauber-Sudarshan distribution function (Glauber, 1963; Sudarshan, 1963). Although quasi probability functions provide a route to understand quasi-classical correspondence, a number of points still remain to be addressed seriously. It becomes difficult to treat the equation of Wigner function as a quantum analogue of Fokker-Planck equation as the positivity of the Wigner function is never ensured by the occurrences of higher derivatives of distribution function in the equation. It is in this perspective and a few other relevant issues we would like to clarify for our basic motivation behind the present work.

The quantum dynamics of mesoscopic or macroscopic systems is always complicated by the coupling of different types of 'environmental' modes. In the overwhelming majority of the treatments dealing with quantum dissipative systems, the reservoir acts as a bosonic bath (Leggett *et al.*, 1987; Nitzan, 2006; Barik *et al.*, 2009) which is dominated by delocalized modes. However, there arise situations, particularly at low temperatures where one has to take care of the model of localized modes for, e.g., dynamics of a cavity mode damped by a bath of two level atoms suggested by Sargent, Scully, and Lamb in the seventies (Sargent *et al.*, 1974), dissipative quantum coherence in fermionic (spin-1/2) Hamiltonian (Guinea *et al.*, 1985), spontaneous and electron assisted tunneling (Vladar *et al.*, 1986), relaxation of magnetic crystals (Prokof'ev and Stamp, 1996; Prokof'ev and Stamp, 1998; Prokof'ev and Stamp, 2000), temperature dependent coherence in dissipative spin-1/2 bath (Shao and Hänggi, 1998), electric field induced optical conductivity and direct current resistivity (Ferrer *et al.*, 2006), dynamical localization (Ferrer and Smith 2007) and so on (Ronnow *et al.,*2005; Hanson *et al.,* 2008; Lange *et al.*, 2010; Lange *et al.*, 2012). In such situations spin-1/2 bath is a good description for the underlying stochastic dynamics, which makes the scenario very complicated and different from the oscillator bath.It need not be over emphasized that one cannot map the bosonic bath into a spin-1/2 bath model due to distinct universality class of quantum environment (Chang and Chakravarty, 1985). From an ion trapping experiment it has been demonstrated (Khaetskii *et al.,* 2002) that decoherence and dissipation rates exhibit characteristically distinct behavior from those of a bosonic bath. However, at very low energies, oscillator bath reduces to spin-1/2 bath (Caldeira *et al.,* 1993; Forsythe and Makri, 1999). Both spin-1/2 bath and bosonic bath thus appear in widely different contexts, and several formulations presenting the similarities and disimilarities are known (Suarez and Silbey, 1991; Gelman *et al.,* 2004; Schlosshauer *et al.,* 2008). Finally, the form of an interaction between the system and its environment demands a physical situation. For almost all practical purposes, the theory of classical and quantum dissipation assumes the form of a bilinear coupling between the spatial co-ordinates of a system and its environment. An opposite scenario arises when the momentum of the system and its environment get coupled and interesting physical situation may occur due to such momentum dissipation (Cuccoli *et al.,* 2001; Ankerhold and Pollok, 2007; Cuccoli *et al.,* 2010). Study of quantum dissipation thus continues to remain an active field of interest.

Scope and Plan of the Review

Keeping in view of the distinctive nature of the two kinds of bath, our object in this review is to discuss a unifying model for the environment. The universal realization of the environment in the form of a general spin bath also facilitates the implementation of the true canonical thermal distribution functions. The outlay of the review is as follows:

1. In the next section, we have proposed a system-reservoir Hamiltonian which can serve as a universal paradigm for the description of interaction between a quantum particle and its environment.
2. Based on spin coherent state representation we have presented in Sec III, a canonical formulation of c-number quantum noise and dissipation.
3. In Sec IV, we have illustrated the method for numerical simulation of c-number noise to solve quantum Langevin equation and an appropriate scheme for quantum correction to calculate the average values.
4. In Sec V, we have formulated the quantum analogues of Fokker-Planck, diffusion and Smoluchowski equations in terms of true probability distribution functions.
5. In Sec VI, we have investigated the problem of tunneling and thermal activation in spatial as well as momentum coupling cases for both spin-1/2 and bosonic bath.
6. In Sec VII, we have demonstrated the experimental realization of spin-1/2 bath in connection with absorption-emission experiments in quantum dots and in quantum optical context.

The quantum theory of dissipation encompasses a vast area of research. The work presented here is not an exhaustive review on the subject but an outline of some recent contributions (Sinha *et al.,* 2010; Sinha *et al.,* 2011a; Sinha *et al.,* 2011b; Ghosh *et al.,* 2011a; Ghosh *et al.,* 2011b; Ghosh *et al.,* 2012a; Ghosh *et al.,* 2012b; Sinha *et al.,* 2013a; Sinha *et al.,* 2013b; Ghosh *et al.,* 2014) made by the present group, particularly in the context of rate theory and a few related topics.

Dissipative Quantum Dynamics; Spin vs Boson Bath

The Model

Let us consider a quantum particle of unit mass coupled to a set of atoms considered as a bath. Each (k-th) atom of the bath has a mass m_k and spin S. The spin is characterized by the components of spin vector operators $\hat{S}_x, \hat{S}_y$ and $\hat{S}_z$ satisfying $[\hat{S}_x, \hat{S}_y] = i\hat{S}_z$ and their cyclic combinations. Indeed the mass m_k of the bath atoms are physically meaningless as the relevant dynamical variable of the bath particles is assumed to be their spin. But we keep it simply for dimensional reason and it has nothing to do with real mass of the atom carrying the spin. The state of a spin is denoted by $|S, \mathbf{m}_s\rangle$ which is the eigen state of operators $\hat{S}^2$ and $\hat{S}_z$ such that $\hat{S}^2 |S, \mathbf{m}_s\rangle$ = $S(S+1)|S, \mathbf{m}_s\rangle$ and $\hat{S}_z |S, \mathbf{m}_s\rangle = \mathbf{m}_s |S, \mathbf{m}_s\rangle$ respectively. $\mathbf{m}_s$ is the projection quantum number which takes values from $-S$, $-S+1$, ..., $S-1$, S. We further define $\hat{n} = S - \hat{S}_z$ as 'spin deviation' operator. The energy of the spin bath in terms of 'spin-deviation' operator can be expressed as $\hat{H}_{bath} = \sum_k \omega_k \hat{n}_k$ where the subscript k refers to the k^{th} atom of the bath so that $\hat{n}_k = S - \hat{S}_{zk}$. The operator algebra for each k^{th} atom obey the following commutation relations

$$[\hat{S}_k^\dagger, \hat{S}_k] = 2\hat{S}_{zk}; [\hat{S}_k^\dagger, \hat{S}_{zk}] = -\hat{S}_k^\dagger; [\hat{S}_k, \hat{S}_{zk}] = \hat{S}_k \quad (2.1)$$

where $\hat{S}_k^\dagger (\hat{S}_k)$ is the creation (annihilation) operator of the k-th spin and we have $\hat{S}_k^\dagger = \hat{S}_k^x + i\hat{S}_k^y$, $\hat{S}_k = \hat{S}_k^x - i\hat{S}_k^y$.

The quantum particle considered as the system is characterized by the Hamiltonian $\hat{H}_s = \frac{\hat{p}^2}{2} + V(\hat{q})$ where $\hat{p}$ and $\hat{q}$ are momentum and position operators for the particle which follow the usual commutation relation $[\hat{q},\hat{p}]=i\hbar$. $V(\hat{q})$ is an external force field acting on the particle.

The total Hamiltonian for the particle, bath and their interaction can be represented by the following Hamiltonian

$$\hat{H}=\frac{\hat{p}^2}{2}+V(\hat{q})+\hbar\sum_k \omega_k \hat{n}_k+\frac{1}{2}\sum_k c_k \hat{q}$$

$$\left[\frac{c_k}{m_k\omega_k^2}\hat{q}-\sqrt{\frac{\hbar}{Sm_k\omega_k}}\left(\hat{S}_k^{\dagger}+\hat{S}_k\right)\right] \tag{2.2}$$

Here we assume that the bath is coupled to the position of the particle. The coefficients $\{c_k\}$ characterize the detailed nature of the coupling. $\hat{q}^2$-containing term of the Hamiltonian ensures that in the resulting quantum dynamics the system potential does not get modified due to heat bath so that dissipation remains spatially homogeneous. In other words, the inclusion of this counter-term makes the model translationally invariant, otherwise the coupling between the position and the bath operators would lead to unphysical renormalization of the potential. Furthermore, to ensure the provision for an irreversible decay of energy of the particle and to avoid Poincare recurrences we require k to run from 1 to ∞, i.e., the bath to contain infinite degrees of freedom.

Mapping between Spin & Boson Operators; Holstein-Primakoff Transformation

We may note that for the state $|n_k\rangle$ which is an eigen state of $\hat{n}_k$ and $\hat{S}_{zk}$, the creation $\left(\hat{S}_k^{\dagger}\right)$ and annihilation $\left(\hat{S}_k\right)$ operators satisfy the following properties

$$\hat{S}_k^{\dagger}\,|n_k\rangle=(2S)^{1/2}\sqrt{1-\frac{n_k-1}{2S}}\sqrt{n_k}\,|n_k-1\rangle \tag{2.3}$$

$$\hat{S}_k\,|n_k\rangle=(2S)^{1/2}\sqrt{1+n_k}\sqrt{1-\frac{n_k}{2S}}\,|n_k+1\rangle \tag{2.4}$$

where

$$\hat{n}_k\,|n_k\rangle=n_k\,|n_k\rangle \tag{2.5}$$

Here n_k (= S – $\mathbf{m}_s$) is the eigen value of the 'spin deviation' $\hat{n}_k$ operator representing the difference between z-component of the k^{th} (atom) spin ($\mathbf{m}_s$) and its maximum value (S). Therefore n_k is allowed to run from 0 to 2S. Now, let the state with minimal projection (for which $\mathbf{m}_s = -S$) acts as a vacuum for a set of boson operators so that each subsequent state with higher projection quantum number can be considered as a boson excitation of the previous one. This can be achieved by considering the usual boson creation $\left(\hat{a}_k^{\dagger}\right)$/annihilation $\left(\hat{a}_k\right)$ operator as follows;

$$\hat{a}_k\,|n_k\rangle=\sqrt{n_k}\,|n_k-1\rangle \tag{2.6}$$

$$\hat{a}_k^{\dagger}\,|n_k\rangle=\sqrt{n_k+1}\,|n_k+1\rangle \tag{2.7}$$

Comparing Eqs. (2.3)-(2.5) and Eqs. (2.6)-(2.7) we have

$$\hat{S}_k^{\dagger}=(2S)^{1/2}\sqrt{1-\frac{\hat{a}_k^{\dagger}\hat{a}_k}{2S}}\hat{a}_k \tag{2.8}$$

$$\hat{S}_k=(2S)^{1/2}\hat{a}_k^{\dagger}\sqrt{1-\frac{\hat{a}_k^{\dagger}\hat{a}_k}{2S}} \tag{2.9}$$

where, the 'spin-deviation' operator $\hat{n}_k$ is identified as 'number' operator

$$\hat{n}_k=\hat{a}_k^{\dagger}\hat{a}_k \tag{2.10}$$

Equations (2.8)-(2.10) are precisely known as Holstein-Primak off transformation (Holstein and Primakoff, 1940) which sets up a mapping between the spin(-deviation) state and the bosonic state as follows:

$$|-S+\mathbf{m}_s\rangle_{Spin} \check{S}(a^\dagger)^n |0\rangle_{Boson} \; (\mathbf{m}_s = n) \quad (2.11)$$

This implies that each decrease in spin projection number ($\mathbf{m}_s$) in a spin state $|n_k\rangle$ corresponds to subsequent increase in boson excitation number. Eqs. (2.8)-(2.10) also ensure correct commutation relation for spin operators [Eq. (2.1)]. From Eqs.(2.8)-(2.10) it is apparent that the transformation is particularly useful for large S, when the square roots can be expanded as a Taylor series to obtain an expression in descending powers of S. In the high spin limit ($S >> 1$), the transformation therefore reduces to

$$\hat{S} \to (2S)^{1/2}\hat{a}^\dagger \text{ and } \hat{S}^\dagger \to (2S)^{1/2}\hat{a} \quad (2.12)$$

Let us point out an immediate consequence of the above relations. Using these on the Hamiltonian [Eq. (2.2)] and in view of Eq. (2.10) taking care of the zero point energy for the bosonic (harmonic) oscillator heat bath we obtain

$$\hat{H} = \frac{\hat{p}^2}{2} + V(\hat{q}) + \hbar\sum_k \omega_k \left(\hat{a}_k^\dagger \hat{a}_k + \frac{1}{2}\right)$$
$$+\frac{1}{2}\sum_k c_k \hat{q}\left[\frac{c_k}{m_k\omega_k^2}\hat{q} - \sqrt{\frac{2\hbar}{m_k\omega_k}}\left(\hat{a}_k^\dagger + \hat{a}_k\right)\right] \quad (2.13)$$

which, by defining $\hat{a}_k^\dagger = \frac{\hat{x}_k}{\sqrt{2\hbar/m_k\omega_k}} - \frac{i\hat{p}_k}{\sqrt{2\hbar m_k\omega_k}}$

and $\hat{a}_k = \frac{\hat{x}_k}{\sqrt{2\hbar/m_k\omega_k}} + \frac{i\hat{p}_k}{\sqrt{2\hbar m_k\omega_k}}$ further,

reduces to the Hamiltonian of Zwanzig (Zwanzig, 1973) form:

$$\hat{H} = \frac{\hat{p}^2}{2} + V(\hat{q})$$
$$+\sum_k \left[\frac{\hat{p}_k^2}{2m_k} + \frac{1}{2}\frac{c_k^2}{m_k\omega_k^2}\left(\frac{m_k\omega_k^2}{c_k}\hat{x}_k - \hat{q}\right)^2\right] \quad (2.14)$$

The above Hamiltonian eventually describes the interaction between a quantum particle and a harmonic oscillator heat bath $\{\hat{x}_k, \hat{p}_k\}$ which obey the usual commutation relation $[\hat{x}_k, \hat{p}_k] = i\hbar$, are the set of coordinates and momentum operators for oscillators with masses m_k. Holstein-Primakoff transformation thus sets up a correspondence between a spin bath [Eq. (2.2)] and a harmonic/bosonic bath [Eq. (2.14)]. Caldeira and Leggett were among the first who applied the model [Eq. (2.14)] to study the quantum mechanical tunneling of a macroscopic variable (Caldeira and Leggett, 1981). In the recent literature, the model described by Eq. (2.14) is usually referred to as Caldeira-Leggett model (Weiss, 1999).

Reduced Operator Langevin Equation

The Heisenberg equations of motion for the particle and the bath operators can be written down from the Hamiltonian [Eq. (2.2)] with the help of the commutation relations [Eq. (2.1)] as follows:

$$\dot{\hat{q}} = \hat{p} \quad (2.15)$$

$$\dot{\hat{p}} = -V'(\hat{q}) + \frac{1}{2}\sum_k c_k \left[\sqrt{\frac{\hbar}{Sm_k\omega_k}}(\hat{S}_k^\dagger + \hat{S}_k) - \frac{2c_k\hat{q}}{m_k\omega_k^2}\right] \quad (2.16)$$

$$\dot{\hat{S}}_k^\dagger = -i\omega_k\hat{S}_k^\dagger + \frac{ic_k}{\sqrt{Sm_k\hbar\omega_k}}\hat{S}_{zk}\hat{q} \quad (2.17)$$

$$\dot{\hat{S}}_k = i\omega_k\hat{S}_k - \frac{ic_k}{\sqrt{Sm_k\hbar\omega_k}}\hat{S}_{zk}\hat{q} \quad (2.18)$$

$$\dot{\hat{S}}_{zk} = \frac{ic_k\hat{q}}{\sqrt{S\hbar\omega_k}}(\hat{S}_k^\dagger - \hat{S}_k) \quad (2.19)$$

After formally eliminating the bath degrees of freedom as usual (Ghosh *et al.*, 2012b) we arrive at the reduced operator equation for the particle;

$$\ddot{\hat{q}} = -V'(\hat{q}) + \frac{1}{2}\sum_k c_k \sqrt{\frac{\hbar}{Sm_k\omega_k}}$$

$$\left\{\hat{S}_k^\dagger(0)e^{-i\omega_k t} + \hat{S}_k(0)e^{i\omega_k t}\right\} - \sum_k \frac{c_k^2}{m_k\omega_k^2}\hat{q}$$

$$+\sum_k \frac{c_k^2}{Sm_k\omega_k}\int_0^t dt'\hat{S}_{zk}(t')\hat{q}(t')Sin\omega_k(t-t') \tag{2.20}$$

The last term of the right-hand side contains the bath operators under time-integral and makes the equation difficult for an exact solution. However, an implification can be made at this stage by noting the time scale of evolution of the $\hat{S}_k^\dagger$ and $\hat{S}_k$ as compared to $\hat{S}_{zk}$. Eqs. (2.17)-(2.19) suggest that while the polarization operators ($\hat{S}_k^\dagger$ and $\hat{S}_k$) are governed by the characteristic time scale of free evolution (ω_k), the energy operators ($\hat{S}_{zk}$) follow the slower time scale of system bath interaction ($\sim c_k / \sqrt{\hbar m_k \omega_k}$). Therefore, we may approximate $\hat{S}_{zk}(t') \sim \hat{S}_{zk}(0)$ in Eq. (2.20). Furthermore, assuming that at $t = 0$, the spin bath consists of a set of spin-S atoms and is at thermal equilibrium and is decoupled from the system, we may set $\hat{S}_{zk}(0) = S$. The approximation of $\hat{S}_{zk}(0) \simeq S$ takes care of the fact that the particle feels the potential $V(\hat{q})$, which remains unaffected by the interaction during dynamical evolution of the particle. Based on these considerations and after carrying out the partial integration of the last term, we are led to the following equation

$$\ddot{\hat{q}} = -V'(\hat{q}) - \int_0^t dt'\dot{\hat{q}}(t')\sum_k \frac{c_k^2}{m_k\omega_k^2}cos\omega_k(t-t')$$

$$+\frac{1}{2}\sum_k c_k\sqrt{\frac{\hbar}{Sm_k\omega_k}}\left\{\hat{\sigma}_k^\dagger(0)e^{-i\omega_k t} + \hat{\sigma}_k(0)e^{i\omega_k t}\right\} \tag{2.21}$$

where $\hat{\sigma}_k(0)$ $\hat{\sigma}_k^\dagger(0)$ and are defined asthe shifted bath operators of following forms;

$$\hat{\sigma}_k(0) = \hat{S}_k(0) - \frac{c_k}{\omega_k}\sqrt{\frac{S}{\hbar m_k\omega_k}}\hat{q}(0) \tag{2.22}$$

$$\hat{\sigma}_k^\dagger(0) = \hat{S}_k^\dagger(0) - \frac{c_k}{\omega_k}\sqrt{\frac{S}{\hbar m_k\omega_k}}\hat{q}(0) \tag{2.23}$$

The approximation $\hat{S}_{zk}(0) \simeq S$ is consistent as it cancels out the counter term $\left(\frac{1}{2}\sum_k \frac{c_k^2}{m_k\omega_k^2}\hat{q}^2\right)$ and preserve the symmetry of the Hamiltonian. We, therefore, write down the reduced operator Langevin equation for the particle in the following form:

$$\ddot{\hat{q}} + \int_0^t dt'\dot{\hat{q}}(t')\kappa(t-t') + V'(\hat{q}) = \hat{f}(t) \tag{2.24}$$

where, the noise operator and the memory kernel are given by

$$\hat{f}(t) = \frac{1}{2}\sum_k c_k\sqrt{\frac{\hbar}{Sm_k\omega_k}}\{\hat{\sigma}_k(0)e^{i\omega_k t}$$

$$+\hat{\sigma}_k^\dagger(0)e^{-i\omega_k t}\} = \hat{F}(t) + \hat{F}^\dagger(t) \tag{2.25}$$

and

$$\kappa(t-t') = \sum_k \frac{c_k^2}{m_k\omega_k^2} cos\ \omega_k(t-t') \tag{2.26}$$

respectively.

Statistical Properties; Spin-1/2 vs Boson Bath

The reduced dynamical equation for the particle is described by the operator Langevin Eq. (2.24). It contains the usual dissipative term [$\kappa(t-t')$] as well as the noise term [$\hat{f}(t)$]. Statistical properties of the bath can then be calculated by thermal averaging over appropriately ordered noise operators. To this end we define

$$\left\langle \hat{A} \right\rangle_{qs} = \frac{Tr\hat{A}e^{-\hat{H}_{bath}/KT}}{Tre^{-\hat{H}_{bath}/KT}} \tag{2.27}$$

as quantum statistical average of any bath operator $\hat{A}$ where $\hat{H}_{bath} = \hbar\sum_k \omega_k \hat{n}_k$ at t = 0. Furthermore, without any loss of generality we may set $\left\langle \hat{q}(0) \right\rangle = 0$ which yields further simplification of Eqs. (2.22)-(2.23) as

$$\left\langle \hat{\sigma}_k(0) \right\rangle_{qs} = \left\langle \hat{S}_k(0) \right\rangle_{qs} \tag{2.28}$$

$$\left\langle \hat{\sigma}_k^\dagger(0) \right\rangle_{qs} = \left\langle \hat{S}_k^\dagger(0) \right\rangle_{qs} \tag{2.29}$$

At this point, we take a pause to note that Holstein-Primakoff transformation allows us to realize a mapping between a spin state and a bososnic state with finite number [Eq. (2.11)]. We utilize this correspondence to differentiate between two distinct situations for characterizing the statistical properties of the bath. Thus a bath can be treated either as a spin-1/2 bath or as abosonic bath and depending on its nature the occupancy of the bath states (with or without obeying Pauli Exclusion Principle) will have to be determined. We, therefore, consider the two cases separately.

(i) For spin-1/2 bath, the Hamiltonian in terms of the 'number' operator $\hat{n}_k^F$ is $\hat{H}_{bath}$ = $\hbar\sum_k \omega_k \hat{n}_k^F$. Here, the superscript 'F' stands for the fermionic (spin-1/2) character of the bath. Eq. (2.27) then gives the average thermal excitation number ofthe spin-1/2 bath as

$$\left\langle \hat{n}_k^F \right\rangle_{qs} = \frac{\sum_{n_k=0,1} n_k e^{-n_k\hbar\omega_k/KT}}{\sum_{n_k=0,1} e^{-n_k\hbar\omega_k/KT}} = \frac{1}{e^{\hbar\omega_k/KT}+1} = \overline{n}_F(\omega_k) \tag{2.30}$$

which results in

$$\left\langle (1-2\hat{n}_k^F) \right\rangle_{qs} = 1 - 2\overline{n}_F(\omega_k) = tanh\left(\frac{\hbar\omega_k}{2KT}\right) \tag{2.31}$$

and refers to the Fermi-Dirac distribution function.

Thus the noise properties of the spin-1/2 bath can be derived by using the canonical thermal distribution [Eq. (2.27)] of the noise operators. It is easy to verify that the spin-1/2 bath noise operator $\hat{f}(t)$ [Eq. (2.25)] is zero centered, i.e.,

$$\left\langle \hat{f}(t) \right\rangle_{qs} = 0 \tag{2.32}$$

and satisfies the relation

$$\begin{aligned} &Re\left\{\left\langle \hat{F}^\dagger(t)\hat{F}(t') - \hat{F}(t)\hat{F}^\dagger(t') \right\rangle_{qs}\right\} \\ &= \frac{\hbar}{2}\sum_k \frac{c_k^2}{m_k\omega_k}\left\langle (1-2\hat{n}_k^F) \right\rangle_{qs} cos\omega_k(t-t') \\ &= \frac{\hbar}{2}\sum_k \frac{c_k^2}{m_k\omega_k} tanh\left(\frac{\hbar\omega_k}{2KT}\right) cos\,\omega_k(t-t') \end{aligned} \tag{2.33}$$

where $\hat{n}_k^F = \hat{a}_k^{\dagger F}\hat{a}_k^F$. The negative sign on the left hand side of Eq. (2.33) carries the signature of anti-commutation relations obeyed by the fermionic (spin-1/2) operators, i.e., $\{\hat{a}_k^F, \hat{a}_k^{\dagger F}\} = 1$. Eq. (2.33) refers to the fluctuation-dissipation relation for the fermionic (spin-1/2) bath.

(ii) For bosonic bath on the other hand, the Hamiltonian reads as $\hat{H}_{bath} = \hbar\sum_k \omega_k \hat{n}_k^B$. The superscript '$B$' stands for boson for which the average thermal excitation number can be obtained as

$$\left\langle \hat{n}_k^B \right\rangle_{qs} = \frac{\sum_{n_k=0}^{2S} n_k e^{-n_k\hbar\omega_k/KT}}{\sum_{n_k=0}^{2S} e^{-n_k\hbar\omega_k/KT}} = \frac{x}{1-x} - \frac{2Sx^{2S}}{1-x^{2S}} \tag{2.34}$$

where, $x = e^{-\hbar\omega_k/KT}$ which lie between $0 < x < 1$. Hence, under Holstein-Primakoff transformation ($x^{2S} << 1$), neglecting the last term one can easily identify

$$\left\langle \hat{n}_k^B \right\rangle_{qs} = \frac{1}{e^{\hbar\omega_k/KT} - 1} = \overline{n}_B(\omega_k) \qquad (2.35)$$

as Bose-Einstein distribution. The above relation on the other hand gives

$$\left\langle (2\hat{n}_k^B + 1) \right\rangle_{qs} = 2\overline{n}_B(\omega_k) + 1 = coth\left(\frac{\hbar\omega_k}{2KT}\right) \qquad (2.36)$$

The noise properties for the bosonic bath can then be calculated similarly by using Eqs. (2.27)-(2.29) and Eqs. (2.34)-(2.36) as

$$\left\langle \hat{f}(t) \right\rangle_{qs} = 0 \qquad (2.37)$$

and

$$\begin{aligned} &Re\left\{\left\langle \hat{F}^{\dagger}(t)\hat{F}(t') + \hat{F}(t)\hat{F}^{\dagger}(t') \right\rangle_{qs}\right\} \\ &= \frac{\hbar}{2}\sum_k \frac{c_k^2}{m_k\omega_k}\left\langle (2\hat{n}_k^B + 1)\right\rangle_{qs} cos\omega_k(t-t') \\ &= \frac{\hbar}{2}\sum_k \frac{c_k^2}{m_k\omega_k} coth\left(\frac{\hbar\omega_k}{2KT}\right) cos\,\omega_k(t-t') \end{aligned} \qquad (2.38)$$

where, $\hat{n}_k^B = \hat{a}_k^{\dagger B}\hat{a}_k^B$. The positive sign in the left hand side of Eq. (2.38) signifies the commutation relation obeyed by the bosonic bath operators as $[\hat{a}_k^B, \hat{a}_k^{\dagger B}] = 1$. Eq. (2.38) represents the fluctuation-dissipation relation for a bosonic bath.

The distinctive features of the spin-1/2or fermionic bath incomparison to that of bosonic or harmoic bath are now quite imperative. Traditionally, the bosonic heat bath is supposed to be a universal description of the environment of an infinite collection of harmonic oscillators. For each bath oscillator, the possibility of thermally induced excitation is very large. On the other hand, for fermionic or spin-1/2 bath there is a severe restriction on the possibility for the thermal excitation of the environment due to Pauli Exclusion Principle. It is then expected that when a bosonic heat bath is replaced by a spin-1/2 bath, the thermal properties of the environment should differ quite significantly. Secondly, since fermions follow anti-commutation rules and have no classical analogue, the fluctuation-dissipation relation for spin-1/2 bath [Eq. (2.33)] does not reduce to classical fluctuation-dissipation relation similar to its bosonic counterpart [Eq. (2.38)] at high temperature. Thus the distinctive behavior of a spin-1/2 bath gets more prominentat high temperatures. However, the spin-1/2 bath is very much similar to a bosonic bath at zero temperature since both the hyperbolic tangent and cotangent factors in Eqs. (2.33) and (2.38) merge to unity (Caldeira *et al.*, 1993; Forsy the and Makri, 1999). In the next section we would further exploit this correspondence for a c-number description of spin-1/2 bath and bosonic bath.

Canonical Formulation of Quantum Dissipation in Coherent State Description

Radcliffe Spin Coherent States

We take a little digression at this point to summarize some of the properties of the spin coherent states. Existence of spin coherent states analogous to the harmonic oscillator coherent states had been proved a couple of decades ago by Radcliffe (Radcliffe, 1971). Let consider a single atom with spin S. The state with minimal projection of which is taken as the ground state $|0\rangle$ such that $\hat{S}_z|0\rangle = S|0\rangle$, where $\hat{S}_z$ stands for the z-component of the spin vector operator. Then the operator $\hat{S} \equiv \hat{S}_x - i\hat{S}_y$ as given in Eq. (2.9) creates spin deviations as follows

$$(\hat{S})^n|0\rangle = \left(\frac{n!2S!}{(2S-n)!}\right)^{1/2}|n\rangle \quad (0 \le n \le 2S) \qquad (3.1)$$

where, $|n\rangle$ is the eigen state of $\hat{S}_z$ with $\hat{S}_z|n\rangle$ = $(S-n)|n\rangle$.

The coherent state $|\mu\rangle$ is then defined as

$$|\mu\rangle = N^{-1/2}exp\left(\mu\hat{S}\right)|0\rangle$$

$$= N^{-1/2}\sum_{n=0}^{2S}\left(\frac{2S!}{n!(2S-n)!}\right)^{1/2}\mu^n|n\rangle \quad (3.2)$$

where, μ runs over the entire complex plane and N is the normalization factor such that

$$\langle\mu|\mu\rangle = N^{-1}\sum_{n=0}^{2S}\left(\frac{2S!}{n!(2S-n)!}\right)|\mu|^{2n}$$

$$= N^{-1}(1+|\mu|^2)^{2S} \quad (3.3)$$

Hence the normalized coherent state is

$$|\mu\rangle = (1+|\mu|^2)^{-S}exp\left(\mu\hat{S}\right)|0\rangle \quad (3.4)$$

The states $|\mu\rangle$ defined by Eq. (3.4) do form a complete set; although it is necessary to include an appropriate weight factor $\mathbf{W}(|\mu|^2)\geq 0$ in the integral, so that

$$\int d^2\mu|\mu\rangle\mathbf{W}(|\mu|^2)\langle\mu| = \sum_{n=0}^{2S}|n\rangle\langle n| = 1 \quad (3.5)$$

Defining $|\mu| = \rho$ and carry out the angular integrations, we find

$$\int d^2\mu|\mu\rangle\mathbf{W}(|\mu|^2)\langle\mu|$$

$$= 2\pi\sum_{n=0}^{2S}|n\rangle\langle n|\left(\frac{2S!}{n!(2S-n)!}\right)\int_0^\infty \rho\, d\rho\frac{\rho^{2n}}{(1+\rho^2)^{2S}}$$

$$\mathbf{W}(\rho^2) = \sum_{n=0}^{2S}|n\rangle\langle n|\left(\frac{2S!}{n!(2S-n)!}\right)\mathbf{I}(n,S) \quad (3.6)$$

where the integral

$$\mathbf{I}(n,S) = \pi\int_0^\infty d\beta\frac{\beta^n}{(1+\beta)^{2S}}\mathbf{W}(\beta), \text{ as } \left(\rho^2\equiv\beta\right) \quad (3.7)$$

The appropriate form of $\mathbf{W}(\beta)$ for which $\mathbf{I}(n,S) = \dfrac{n!(2S-n)!}{2S!}$ is given by

$$\mathbf{W}(\beta) = \frac{2S+1}{\pi}.\frac{1}{(1+\beta)^2} \quad (3.8)$$

So, finally the completeness relation can be expressed as

$$\frac{2S+1}{\pi}\int\frac{d^2\mu}{(1+|\mu|^2)^2}|\mu\rangle\langle\mu| = \sum_{n=0}^{2S}|n\rangle\langle n| = 1 \quad (3.9)$$

Eq. (3.9) can also be found by transforming back from a different parameterization of the states, namely, $\mu = tan(\theta/2)e^{i\phi}$. However, it is convenient to work with μ while drawing analogies with harmonic oscillator. The harmonic oscillator case can be recovered from Eq. (3.4) in the high-spin limit (S >> 1) of the Holstein-Primakoff transformation with the substitutions $\hat{S}\to(2S)^{1/2}\hat{a}^\dagger$ and $\mu\to\alpha/(2S)^{1/2}$. Thus, we have the normalized states $|\alpha\rangle_{(S)}$ as

$$|\alpha\rangle_{(S)} = \left(1+\frac{|\alpha|^2}{2S}\right)^{-S}exp\left(\alpha\hat{a}^\dagger\right)|0\rangle \quad (3.10)$$

But since

$$\lim_{S\to\infty}\left(1+\frac{|\alpha|^2}{2S}\right)^{-S} = exp\left(\frac{|\alpha|^2}{2}\right) \quad (3.11)$$

we may write

$$\lim_{S\to\infty}|\alpha\rangle_{(S)} = exp\left(-\frac{|\alpha|^2}{2}\right)exp\left(\alpha\hat{a}^\dagger\right)|0\rangle \quad (3.12)$$

which, apart from normalization constant is precisely the harmonic oscillator coherent state (Glauber, 1963). The spin coherent state approaches the harmonic oscillator coherent state in the same limit our starting Hamiltonian [Eq. (2.2)] reduces to Zwanzig Hamiltonian [Eq. (2.14)], as shown in the earlier section.

For the normalized coherent state [Eq. (3.4)] we now calculate the following matrix elements;

$$\langle\mu|\hat{n}|\mu\rangle=(1+|\mu|^2)^{-2S}\sum_{n=0}^{2S}\left(\frac{2S!}{n!(2S-n)!}\right)|\mu|^{2n} \tag{3.13}$$

The sum $\sum_{n=0}^{2S}\left(\frac{2S!}{n!(2S-n)!}\right)|\mu|^{2n}\,n$ can be expressed as $|\mu|^2\frac{\partial}{\partial|\mu|^2}(1+|\mu|^2)^{2S}$. Hence the matrix element becomes

$$\langle\mu|\hat{n}|\mu\rangle=\frac{2S|\mu|^2}{1+|\mu|^2}\equiv\tilde{C}(S,|\mu|)|\mu|^2$$

where

$$\tilde{C}(S,|\mu|)=\frac{2S}{1+|\mu|^2} \tag{3.14}$$

Similarly for other matrix elements we have

$$\langle\mu|\hat{S}^\dagger|\mu\rangle=(1+|\mu|^2)^{-2S}\frac{\partial}{\partial\mu^*}(1+|\mu|^2)^{2S}$$

$$=\frac{2S\mu}{1+|\mu|^2}\equiv\tilde{C}(S,|\mu|)\mu \tag{3.15}$$

and

$$\langle\mu|\hat{S}|\mu\rangle=(1+|\mu|^2)^{-2S}\frac{\partial}{\partial\mu}(1+|\mu|^2)^{2S}$$

$$=\frac{2S\mu^*}{1+|\mu|^2}\equiv\tilde{C}(S,|\mu|)\mu^* \tag{3.16}$$

We emphasize that the set of Eqs. (3.14)-(3.16) for the matrix elements of spin systems are equally applicable to bosonic systems. The only difference lies in the form of the thermal canonical distribution of the c-numbers μ,μ^* for the spin-1/2 and bosonic systems. Applications of such spin coherent states are well known in the context of ferromagnetic spin wave, phase transition in Dicke model of super-radiance, equilibrium statistical mechanics of radiation-matter interaction and so on (Klauder and Skagerstam, 1985).

C-number Quantum Langevin Equation

Now we construct a quantum Langevin equation with c-number noise. First we return to Eq. (2.24) and carry out its quantum mechanical average $\langle\ldots\rangle$ to obtain

$$\left\langle\ddot{\hat{q}}\right\rangle+\int_0^t dt'\left\langle\dot{\hat{q}}(t')\right\rangle\kappa(t-t')$$

$$+\langle V'(\hat{q})\rangle=\left\langle\hat{f}(t)\right\rangle \tag{3.17}$$

where, the quantum mechanical averages are taken over $|\chi\rangle|\mu_1\rangle|\mu_2\rangle\ldots|\mu_k\rangle\ldots|\mu_N\rangle$, i.e., the initial product separable quantum states of the particle and the bath at t = 0. Here, $|\chi\rangle$ refers to any arbitrary initial state of the particle and $\{|\mu_k\rangle\}$ correspond to the initial Radcliffe spin coherent states (Radcliffe, 1971) of the bath operators.

The idea behind using these spin coherent states for quantum mechanical averaging of the bath operators is to recast Eq. (3.17) as a classical-looking Langevin equation for the particle. We, then denote the following quantum mechanical averages as,

$$\langle\hat{q}(t)\rangle=q(t)\quad\langle\hat{f}(t)\rangle=\eta(t) \tag{3.18}$$

where,

$$\eta(t)=\frac{1}{2}\sum_k c_k\sqrt{\frac{\hbar}{Sm_k\omega_k}}\left[\langle\hat{\sigma}_k(0)\rangle e^{-i\omega_k t}+\langle\hat{\sigma}_k^\dagger(0)\rangle e^{i\omega_k t}\right]$$

$$=\frac{1}{2}\sum_k C_k(S,|\mu_k|)c_k\sqrt{\frac{\hbar}{Sm_k\omega_k}}\left\{\mu_k^*(0)e^{-i\omega_k t}+\mu_k(0)e^{i\omega_k t}\right\} \tag{3.19}$$

The last equality follows from Eqs. (2.28)-(2.29) with $\langle\hat{\sigma}_k(0)\rangle = \langle\hat{S}_k(0)\rangle = \widetilde{C}_k(S, |\mu_k|)\, \mu_k^*(0)$ [Eq. (3.15)] and $\langle\hat{\sigma}_k^\dagger(0)\rangle = \langle\hat{S}_k^\dagger(0)\rangle = \widetilde{C}_k(S, |\mu_k|)\, \mu_k(0)$ [Eq. (3.16)], where μ_k and μ_k^* are the associated c-numbers for general spin bath operators. Eq. (3.17) may then be written as

$$\ddot{q} + \int_0^t \dot{q}(t')\kappa(t-t')dt' + \langle V'(\hat{q})\rangle = \eta(t) \quad (3.20)$$

Eq. (3.20) may be expressed in a more convenient form for interpretation as follows;

$$\ddot{q} + \int_0^t \dot{q}(t')\kappa(t-t')\,dt' + V'(q) = \eta(t) + Q \quad (3.21)$$

where,

$$Q = V'(q) - \langle V'(\hat{q})\rangle \quad (3.22)$$

represents the quantum correction due to the system potential.We will return to its calculation in the next section.

Now to realize $\eta(t)$ as an effective c-number noise, we introduce the ansatz that $\mu_k(0)$ and $\mu_k^*(0)$ are distributed according to a canonical thermal distribution of Gaussian form. Depending on the nature of the bath, we assume the following distributions for fermions (spin-1/2) and bosons.

(a) For spin-1/2 bath (fermions), we assume a thermal canonical distribution function of the following form:

$$P_k^F\left(\mu_k(0), \mu_k^*(0)\right) = \mathrm{N}_F exp\left\{-\frac{|\mu_k(0)|^2}{2tanh\left(\frac{\hbar\omega_k}{2KT}\right)}\right\} \quad (3.23)$$

(b) For bosonic bath (or bosons), the corresponding distribution function is canonical thermal distribution due toWigner (Wigner, 1932);

$$P_k^B\left(\mu_k(0), \mu_k^*(0)\right) = \mathrm{N}_B exp\left\{-\frac{|\mu_k(0)|^2}{2coth\left(\frac{\hbar\omega_k}{2KT}\right)}\right\} \quad (3.24)$$

where, N_F and N_B are the normalization constants which take care of the factors $\widetilde{C}_k(|\mu_k|)$. The widths of the distributions in Eqs. (3.23) and (3.24) are defined by $tanh\left(\frac{\hbar\omega_k}{2KT}\right)$ (Ghosh *et al.*, 2012a; Ghosh *et al.*, 2012b) and $coth\left(\frac{\hbar\omega_k}{2KT}\right)$ respectively. For any arbitrary quantum mechanical mean value of a bath operator, $\langle\hat{A}_k\rangle$ which is a function of $\mu_k(0), \mu_k^*(0)$, its statistical average can then be calculated as

$$\langle\langle\hat{A}_k\rangle\rangle_s = \int \langle\hat{A}_k\rangle P_k^{F,B}\left\{\mu_k^*(0), \mu_k(0)\right\} d\mu_k^*(0)d\mu_k(0) \quad (3.25)$$

The ansatz Eq. (3.23) or Eq. (3.24) and the definition of statistical average Eq. (3.25) can be used to show that c-number noise $\eta(t)$ due to spin-1/2 bath or bosonic bath satisfies the following relations, respectively

$$\langle\eta(t)\rangle_s = 0 \quad (3.26)$$

$$\langle\eta(t)\eta(t')\rangle_s^F = \frac{\hbar}{2}\sum_k \frac{c_k^2}{m_k\omega_k} tanh\left(\frac{\hbar\omega_k}{2KT}\right) cos\omega_k(t-t') \quad (3.27)$$

$$\langle\eta(t)\eta(t')\rangle_s^B = \frac{\hbar}{2}\sum_k \frac{c_k^2}{m_k\omega_k}$$

$$coth\left(\frac{\hbar\omega_k}{2KT}\right)cos\omega_k(t-t') \quad (3.28)$$

Eqs. (3.26)-(3.28) imply that c-number noise $\eta(t)$ is such that it is zero-centered and follows fluctuation-dissipation relation as expressed in Eqs. (2.33) and (2.38). Therefore, Eq. (2.33) and Eq. (3.27) are equivalent. The same is true for Eqs. (2.38) and (3.28). The distinct advantage of the formulation of c-number noise is that the distribution function [Eq. (3.23) and Eq. (3.24)] remains valid as a pure state nonsingular distribution functions even at T = 0. Furthermore, the present formulation allows us to bypass the operator ordering prescription of Eqs. (2.33) and (2.38) for deriving the noise properties of the bath. We also point out that the temperature dependence is exactly like one found for noise correlation in terms of spectral functions in path-integral approach for both spin-1/2 and harmonic baths (Caldiera *et al*., 1993; Forsythe and Makri, 1999). It confirms the validity of the present generalized scheme for quantum noise processes both for spin 1/2 and bosonic heat bath. It becomes easy to identify $\eta(t)$ characterized by Eqs. (3.26)-(3.28) as a 'classical' looking noise with quantum mechanical content. Therefore, the formulation of c-number noise with the help of Radcliffe spin coherent states can be quite effective in implementing any standard techniques of classical non-equilibrium statistical mechanics in a straight-forward way to quantum domain (Lindenberg and West, 1990; van Kampen, 1992). It is also pertinent to note that for a traditional harmonic oscillator heat bath, one can proceed exactly in a similar fashion and use harmonic oscillator coherent states $|\alpha\rangle$, (Glauber, 1963; Barik *et al*., 2009) (which can also be derived from the Radcliffe's spin coherent states [Eq. (3.4)] in the high spin limit as a special case [Eq.(3.12)]). It is easy to recognize that the ansatz will be a canonical thermal Wigner distribution [Eq.(3.24)] which remains always a positive definite function in contrast quasi-probability Wigner distribution (Wigner, 1932)that may take negative values (Agarwal, 2013). The approach using this distribution has been extensively employed in several earlier occasions (Banerjee *et al*., 2002a; Banerjee *et al*., 2002b; Banerjee *et al*., 2003a; Barik *et al*., 2003b; Barik *et al*., 2003c; Banerjee *et al*., 2004; Barik *et al*., 2009).

Spectral Density Function; General Remarks

In order to characterize the properties of the thermal bath and to describe the dissipative mechanism, it is convenient to introduce, as usual, a spectral density function $J(\omega)$ associated with system-bath interaction (Caldeira and Leggett, 1983a) as follows

$$J(\omega)=\frac{\pi}{2}\sum_k\frac{c_k^2}{m_k\omega_k}\delta(\omega-\omega_k) \quad (3.29)$$

In terms of $J(\omega)$, one may rewrite the expressions for memory kernel [Eq. (2.26)] as

$$\kappa(t-t') = \frac{2}{\pi}\int_{-\infty}^{\infty}d\omega\frac{J(\omega)}{\omega}cos\omega(t-t') \quad (3.30)$$

and fluctuation-dissipation relations [Eqs. (3.27)-(3.28)] as

$$\langle\eta(t)\eta(t')\rangle_s^F = Re\left\{\left\langle\hat{F}^{\dagger}(t)\hat{F}(t')-\hat{F}(t)\hat{F}^{\dagger}(t')\right\rangle_{qs}\right\}$$
$$=\frac{2}{\pi}\int_{-\infty}^{\infty}d\omega\frac{J(\omega)}{\omega}\left[\frac{\hbar\omega}{2}tanh\left(\frac{\hbar\omega}{2KT}\right)\right]cos\omega(t-t') \quad (3.31)$$

for c-number noise for spin-1/2 (fermionic) bath and

$$\langle\eta(t)\eta(t')\rangle_s^B = Re\left\{\left\langle\hat{F}^{\dagger}(t)\hat{F}(t')+\hat{F}(t)\hat{F}^{\dagger}(t')\right\rangle_{qs}\right\}$$
$$=\frac{2}{\pi}\int_{-\infty}^{\infty}d\omega\frac{J(\omega)}{\omega}\left[\frac{\hbar\omega}{2}coth\left(\frac{\hbar\omega}{2KT}\right)\right]cos\omega(t-t') \quad (3.32)$$

for c-number noise for bosonic (harmonic) bath respectively. Eqs. (3.31) and (3.32) imply that the relaxation induced by the bath is determined by the properties of for example, a Lorentzian bath is characterized by $J(\omega)=\frac{\gamma^2}{\gamma^2+\omega^2}$, with constant γ.

The bath spectral density $J(\omega)$ in Eq. (3.30) actually provides a constraint on the choice of the coefficients $\{c_k\}$. When we choose $J(\omega)=\gamma\omega$, the corresponding classical kind of dissipation is Ohmic in nature. For a more generic form of spectral density $J(\omega)\propto\omega^m$. In this case, if $m>1$ the corresponding dissipation is 'super-Ohmic', while if $m<1$ it is 'sub-Ohmic' (Weiss, 1999).

Before closing this section we make some general remarks.

(i) From the above discussions it may appear that the Hamiltonian of the form [Eq. (2.2)] necessarily leads to a quantum Langevin Eq. (3.21). Since, in general, we do not know very much about the physical mechanism of dissipation, it is desirable that the model should be as general as possible subject to the condition that it leads to Eq. (3.21) in the appropriate limit. However, to make the subsequent calculation tractable it is necessary to impose one important restriction as emphasized by Caldeira and Leggett that "*any one environment degree of freedom is only weakly perturbed by its interaction with the system*" (Caldeira and Leggett, 1983a; Caldeira and Leggett, 1983b). For most practical cases for a geometrically macroscopic environment, the interaction of the system with each bath degree of freedom is proportional to the inverse of the volume of the reservoir. Hence, the coupling for each individual bath mode is weak. Therefore, it is reasonable for macroscopic global system to assume that system-bath coupling is a linear function of the bath variables (either coordinate or momenta). These criteria as noted by Leggett and Caldeira are favourable since it allows us to eliminate the bath degrees of freedom exactly. Importantly the condition of weak interaction of one degree of freedom does not imply that the interaction is weak when considered from the point of view of the system which, in principle, interacts with infinitely large degrees of freedom of the reservoir and the dissipative effect per degree of freedom adds up for the system. Thus the validity of the quantum Langevin equation is not restricted to weak interaction regime ($\gamma \ll \omega_0$); γ and ω_0 being the dissipation constant and the characteristic linear system frequency respectively. It applies equally well in the strong damping regime ($\gamma \gg \omega_0$).

(ii) A prime requirement of the theory of quantum dissipation is how the classical kind of dissipation can be deduced from a quantum mechanical point of view. To obtain, we carry out the statistical average over the bath states in Eq. (3.21) using the relation $\langle\eta(t)\rangle_s=0$. As a second step, we take $\hbar\to 0$ to recover the classical limit; then the term Q in Eq. (3.21) appearing due to quantum correction vanishes. As a result the effective equation of motion turns out to be

$$\ddot{q}=-V'(q)-\int_0^t \dot{q}(t')\kappa(t-t')dt' \quad (3.33)$$

Using Eq. (3.30) for Markovian baths, which do not keep memory of the interaction with the system and for Ohmic dissipation $J(\omega)=\gamma\omega$, the equation of motion [Eq. (3.33)] simplifies to the classical equation of motion of a particle with friction coefficient γ as;

$$\ddot{q}=-\gamma\dot{q}-V'(q) \quad (3.34)$$

Hence we see how the model Hamiltonian [Eq. (2.2)] fulfills the goal of obtaining classical dissipation within a c-number description.

(iii) Though the equation of motion for the system [Eq.(3.33)] is independent of the nature of the bath, the fluctuation-dissipation relations may provide interesting features. For bosonic bath the fluctuation-dissipation relation [Eq. (3.32)] reduces to well-known classical fluctuation-dissipation relation $\langle\eta(t)\eta(t')\rangle_s = \gamma KT$, while its fermionic counterpart [Eq. (3.31)] does not, as the spin does not have a classical analog. Since the temperature dependence of the spin-1/2 bath contains $tanh\left(\frac{\hbar\omega}{2KT}\right)$ factor which is different from that of bosonic bath expressed through $coth\left(\frac{\hbar\omega}{2KT}\right)$, one may encounter specific temperature dependence of the dissipative behavior of a particle in spin-1/2 bath. At $T = 0$ bosonic and spin-1/2 bath behave identically. At higher temperature because of the hyperbolic cotangent factor, the harmonic bath reaches the classical limit. On the other hand the temperature dependent factor for the spin-1/2 bath differs appreciably from unity and an expansion of the hyperbolic tangent factor results in additional low frequencies which modify the spectral density in an effective way. The thermal behavior of the spin-1/2 bath thus can be viewed under two distinct situations:

High Temperature Regime

From Eq. (2.31), it follows that $\frac{1}{2}tanh\left(\frac{\hbar\omega}{2KT}\right)$ = $\frac{1}{2}-\frac{1}{e^{\hbar\omega/KT}+1}$ = $-\frac{1}{2}\langle\hat{S}_z(0)\rangle_{qs}$,; where, $\langle\hat{S}_z(0)\rangle_{qs}$ is a measure of the population difference between the two levels of k-th bath atom. At high temperature $(e^{\hbar\omega/KT}+1)\rightarrow 2$, which implies, $\langle\hat{S}_z(0)\rangle_{qs}\rightarrow 0$ i.e., the bath atoms get thermally saturated and the effective system-bath coupling is largely suppressed as the noise correlation [Eq. (3.31)] tends to vanish with increase of temperature. This gives rise to emergence of coherent behavior in the system dynamics (Shao and Hänggi, 1998).

Low Temperature Regime

On the other hand, in the low temperature regime, i.e., much below the saturation temperature, we have $\langle\hat{S}_z(0)\rangle_{qs}\rightarrow -\frac{1}{2}$. This implies that the effective system-spin-bath coupling is large and the noise correlation [Eq. (3.31)] gets enhanced. Thus as $T\rightarrow 0$ quantum dissipation and the noise correlation of the spin-1/2 bath [Eq. (3.31)] tend to that for the bosonic bath [Eq. (3.32)]. We will elaborate this point further in the subsequent section, when we discuss numerical simulations of noise correlation functions.

Scheme for Numerical Simulation of Quantum Langevin Equation; Quantum Corrections and Average Values

Calculations of Quantum Correction

Were turn to the c-number quantum Langevin equation [Eq. (3.21)]

$$\ddot{q}+\int_0^t \dot{q}(t')\kappa(t-t')\,dt'+V'(q)=\eta(t)+Q \quad (4.1)$$

where

$$Q=V'(q)-\langle V'(\hat{q})\rangle \quad (4.2)$$

represents the quantum correction due to the system potential. Q can be expressed further in a more explicit and useful form by recognizing the operator nature of the system variables $\hat{q}$ and $\hat{p}$ as

$$\hat{q}(t)=q(t)+\delta\hat{q}(t) \quad (4.3)$$

$$\hat{p}(t)=p(t)+\delta\hat{p}(t) \quad (4.4)$$

where $\delta\hat{q}$ and $\delta\hat{p}$ are respectively the quantum corrections around the quantum mechanical mean values of $q(\equiv\langle\hat{q}\rangle)$ and $p(\equiv\langle\hat{p}\rangle)$ so that $\langle\delta\hat{q}\rangle=\langle\delta\hat{p}\rangle$ = 0 and by construction $[\delta\hat{q},\delta\hat{p}]$ = $i\hbar$. Using Eq. (4.3) in $V'(\hat{q})$ followed by a Taylor expansion around 'q', we may express Q as,

$$Q\left(q,\langle\delta\hat{q}^n\rangle\right)=-\sum_{n\geq 2}\frac{1}{n!}V^{(n+1)}(q)\langle\delta\hat{q}^n(t)\rangle \quad (4.5)$$

Here $V^{(m)}(q)$ is the m-th derivative of the potential $V(q)$ with respect to 'q'. For example, the lowest order quantum correction (n = 2) is given by

$Q=-\frac{1}{2}V'''(q)\left\langle\delta\hat{q}^2(t)\right\rangle$. The recipe for determination of Q therefore depends on $\left\langle\delta\hat{q}^n(t)\right\rangle$ which may be determined by solving quantum correction equations. To achieve this we first return to quantum operator Eq. (2.24) and use Eqs. (4.3)-(4.4). Furthermore, using Eq. (4.1) in the resulting equation, we obtain,

$$\delta\ddot{\hat{q}}+\int_0^\infty dt'\,\kappa(t-t')\,\delta\dot{\hat{q}}(t')+V''(q)\delta\hat{q}$$

$$+\sum_{n\geq 2}\frac{1}{n!}V^{(n+1)}(q)\left(\delta\hat{q}^n(t)-\left\langle\delta\hat{q}^n(t)\right\rangle\right)=\delta\hat{\eta}(t) \tag{4.6}$$

where, $\delta\hat{\eta}(t)=\hat{f}(t)-\eta(t)$.

Eq.(4.6) forms the basis for the calculation of quantum mechanical correction $\left\langle\delta\hat{q}^n(t)\right\rangle$. However, Eq. (4.6) is analytically untractable for an exact solution. Depending on the memory kernel and nonlinearity of the potential, systematic approximation schemes may be employed. We consider a special case, the over damped limit.

To this end we return to quantum correction Eq. (4.6) which in the overdamped limit (inertial term can be discarded compared to the damping term; Sinha *et al.*, 2011b) reduces to the following form

$$\gamma_0\delta\dot{\hat{q}}+V''(q)\delta\hat{q}+\sum_{n\geq 2}\frac{1}{n!}V^{(n+1)}(q)$$

$$\left(\delta\hat{q}^n-\left\langle\delta\hat{q}^n\right\rangle\right)=\delta\hat{\eta} \tag{4.7}$$

where, γ_0 is the dissipation constant in Markovian limit. With the help of operator Eq. (4.6) we obtain the equations for the quantum mechanical mean value of the corrections $\left\langle\delta\hat{q}^n\right\rangle$ in successive orders (after multiplying by $\delta\hat{q}$, $\delta\hat{q}^2$ ... and carrying out quantum mechanical averages with the initial product separable quantum state of the system and the coherent states of the noise operators $|\,\chi\rangle\{\,|\mu_k\rangle\}$ as before) as follows:

$$\langle\delta\dot{\hat{q}}^2\rangle=-\frac{1}{\gamma_0}\left[2V''(q)\langle\delta\hat{q}^2\rangle+V'''(q)\langle\delta\hat{q}^3\rangle\right] \tag{4.8}$$

$$\langle\delta\dot{\hat{q}}^3\rangle=-\frac{1}{\gamma_0}$$

$$\left[3V''(q)\langle\delta\hat{q}^3\rangle+\frac{3}{2}V'''(q)\langle\delta\hat{q}^4\rangle-\frac{3}{2}V'''(q)\langle\delta\hat{q}^2\rangle^2\right] \tag{4.9}$$

and so on. An examination of Eqs. (4.8)-(4.9) suggest that the quantum corrections are suppressed in successive orders as $\sim O(1/\gamma_0)$ by friction. Thus under overdamped condition for a minimum uncertainty state $\langle\delta\hat{q}^2\rangle\sim O(\hbar)$ the quantum correction diminishes as $\sim O(\hbar/\gamma_0)$ at each order. Thus with enhancement of the dissipation the system tends to loose its quantum character. To take into account of the leading order contribution due to quantum correction we may write Eq. (4.8) as

$$d\langle\delta\hat{q}^2\rangle=-\frac{2}{\gamma_0}V''(q)\langle\delta\hat{q}^2\rangle dt \tag{4.10}$$

The over-damped deterministic motion, on the other hand is given by

$$dq=-V'(q)dt \tag{4.11}$$

which when used in Eq. (4.10) yields after integration,

$$\langle\delta\hat{q}^2\rangle\ =\Delta\left[V'(q)\right]^2 \tag{4.12}$$

where, the quantum correction factor is given by $\Delta=\frac{\langle\delta\hat{q}^2\rangle_m}{\left[V'(q_m)\right]^2}$. q_m corresponds to the quantum mechanical mean position at which $\langle\delta\hat{q}^2\rangle$ is minimum (i.e., $\langle\delta\hat{q}^2\rangle_m \sim \frac{\hbar}{2\omega_0}$). With this the quantum correction in the leading order takes the form

$$Q\left(q,\left\langle\delta\hat{q}^n\right\rangle\right) = -\frac{1}{2}V'''(q)\left\langle\delta\hat{q}^2\right\rangle$$

$$= -\frac{\Delta}{2}V'''(q)\left(V'(q)\right)^2 \quad (4.13)$$

The modified potential is then given by

$$V'_{quan}(q) = V'(q) + \frac{\Delta}{2}V'''(q)\left(V'(q)\right)^2 \quad (4.14)$$

We emphasize that the leading order contribution from quantum correction does not contain any friction coefficient or temperature. Secondly, the inversion symmetry of the classical potential, if any, is therefore retained.

Calculation of Quantum Statistical Averages

Summarizing the discussions of the previous section, we see that the quantum Langevin dynamics in c-number can be calculated for a stochastic process by solving Eq. (4.1) for quantum mechanical mean values together with quantum correction equations which measure quantum dispersion around these mean values. In principle, quantum correction equations constitute an infinite set of hierarchy which must be tructated after a desired order in practice, to make the syetm of equations closed. Care must be taken to distinguish between the quantum mechanical mean of an operator $\hat{A}$ expressed as $\langle\hat{A}\rangle(= A)$ from the statistical average of the quantum mechanical mean A expressed as $\langle A\rangle_s$ and the quantum statistical average of $\hat{A}$, denoted as $\langle\hat{A}\rangle_{qs}$. Any physical averages which are evaluated with a thermal state for the bath is denoted by $\langle\ldots\rangle_{qs}$. In the present scheme they are calculated in two steps, i.e., as statistical averages $\langle\ldots\rangle_s$ of quantum mechanical averages $\langle\ldots\rangle$. This gives the averages of interest as $\langle\ldots\rangle_{qs} = \langle\langle\ldots\rangle\rangle_s$. To illustrate, we calculate, for example, the quantum statistical averages $\left\langle\hat{q}\right\rangle_{qs}$ and $\left\langle\hat{q}^2\right\rangle_{qs}$. Making use of Eq. (4.3), we may write,

$$\begin{aligned}\left\langle\hat{q}\right\rangle_{qs} &= \left\langle q+\delta\hat{q}\right\rangle_{qs} \\ &= \left\langle q\right\rangle_s + \left\langle\langle\delta\hat{q}\rangle\right\rangle_s = \left\langle q\right\rangle_s\end{aligned} \quad (4.15)$$

and,

$$\begin{aligned}\left\langle\hat{q}^2\right\rangle_{qs} &= \left\langle(q+\delta\hat{q})^2\right\rangle_{qs} \\ &= \left\langle q^2\right\rangle_s + \left\langle\delta\hat{q}^2\right\rangle_{qs} = \left\langle q^2\right\rangle_s + \left\langle\langle\delta\hat{q}^2\rangle\right\rangle_s\end{aligned} \quad (4.16)$$

Numerical Simulation of c-Number Noise

The fluctuation-dissipation relations [Eqs. (3.31)-(3.32)] play the key role for the generation of c-number noise. The left hand side of Eqs. (3.31)-(3.32) suggest that $\eta(t)$ is a 'classical' random number $\left\langle\eta(t)\eta(t')\right\rangle_s$ and is a correlation function which is 'classical' in form but quantum mechanical in its content. We now show that c-number noise $\eta(t)$ can be generated as a superposition of several Ornstein-Uhlenbeck noise processes (Uhlenbeck and Ornstein, 1930). We begin by expressing $\left\langle\eta(t)\eta(t')\right\rangle_s$ = $C(t-t')$ in the continuum limit as follows,

$$C^F(t-t') = \frac{2}{\pi}\int_0^\infty d\omega\,\frac{J(\omega)}{\omega}\left[\frac{\hbar\omega}{2}tanh\left(\frac{\hbar\omega}{2KT}\right)\right]cos\,\omega(t-t') \quad (4.17)$$

$$C^B(t-t') = \frac{2}{\pi}\int_0^\infty d\omega\,\frac{J(\omega)}{\omega}\left[\frac{\hbar\omega}{2}\coth\left(\frac{\hbar\omega}{2KT}\right)\right]cos\,\omega(t-t') \quad (4.18)$$

Eq. (4.17) suggests that it is possible to define an effective temperature-dependent spectral density function for the nonlinear/spin-1/2 bath as (Caldeira *et al*., 1993; Ferrer *et al*., 2006)

$$J_{eff}(\omega, T) = tanh\left(\frac{\hbar\omega}{2KT}\right) J(\omega) \tag{4.19}$$

The consequences of such nonlinear bath are captured in connection with optical conductivity in spin-1/2 bath (Ferrer *et al*., 2006), temperature dependent dissipation rate in two-level quantum dots (Ghosh *et al*., 2011a) and other systems (Sinha *et al*., 2013b) e.g., glasses (Leggett and Vural, 2013).

It is essential to know, a priori, the form of spectral density function $J(\omega)$. We assume it to be a Lorentzian as $J(\omega) = \frac{2}{\pi}\frac{\Gamma\omega}{1+(\omega-\omega_0)^2\tau_c^2}$ that yields an exponential memory kernel $\kappa(t) = (\Gamma/\tau_c)e^{-|t|/\tau_c}$ [Eq. (3.30)], where τ_c is the correlation time and Γ is the dissipation constant in the Markovian limit. ω_0 refers to the linearized static system frequency. For a given set of parameters Γ, τ_c and fixed temperature T, we first numerically evaluate the integrals [Eqs. (4.17)-(4.18)] as a function of time. The results are shown in Figs. 4.1(a)-(b) by discrete points. The calculated correlation function is then numerically fitted using a superposition of several exponentials with parameter set $\{D_i, \tau_i\}$ as

$$C^{F,B}(t-t') = \sum_i \frac{D_i}{\tau_i} exp\left(-\frac{|t-t'|}{\tau_i}\right), \quad i = 1, 2, 3.... \tag{4.20}$$

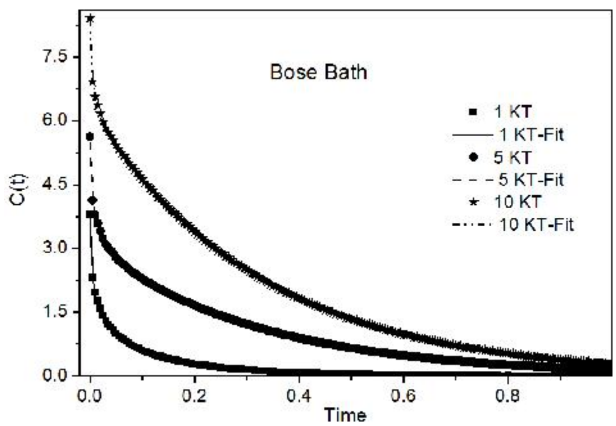
The fitted curves are shown by continuous lines in Figs. 4.1(a)-(b). The numerical fitting is done by Levenberg-Marquardt (LM) algorithm (Marquardt, 1963), which is very efficient for minimizing nonlinear function. The numerically obtained parameters $\{D_i, \tau_i\}$, are, in general, temperature dependent. Now, with the help of these parameters, a set of exponentially correlated color noise variables $\{\phi_i\}$ are generated by the following set of equations,

$$\dot{\phi}_i = -\frac{\phi_i}{\tau_i} + \frac{1}{\tau_i}\epsilon_i(t) \tag{4.21}$$

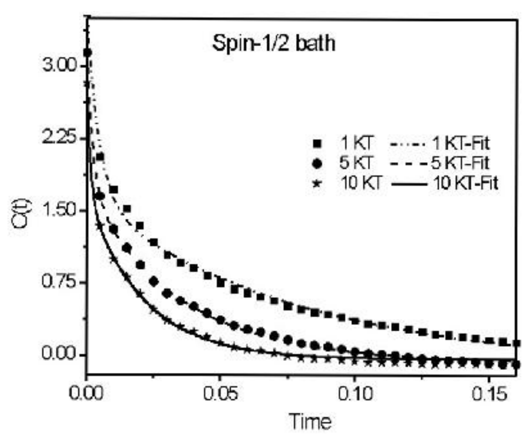
where,

$$\langle \epsilon_i(t) \rangle = 0 \tag{4.22}$$

$$\langle \epsilon_i(0)\epsilon_j(\tau) \rangle = 2D_i\delta_{ij}\delta(\tau) \quad i = 1, 2, 3... \tag{4.23}$$



(a)



(b)

Fig.4.1: Variation of normalized c-number noise correlation function *C*(*t*) with time at different temperatures for fixed set of parameters $\Gamma = 3.1, \tau_c = 1.0$. The symbols represent the analytical variation for (a) Harmonic bath and (b) Spin-1/2 bath, based on Eqs. (4.17)-(4.18) and the lines represent numerical fitting using Eq. (4.20)

where, $\epsilon_i(t)$ is a Gaussian white noise obeying Eqs. (4.22)-(4.23). Each ϕ_i is thus an Ornstein-Uhlenbeck noise with properties

$$\langle \phi_i(t) \rangle = 0 \tag{4.24}$$

$$\langle \phi_i(t)\phi_j(t') \rangle = \delta_{ij} \frac{D_i}{\tau_i} exp\left(-\frac{|t-t'|}{\tau_i} \right)$$

$$i = 1, 2, 3.... \tag{4.25}$$

Then c-number noise $\eta(t)$ due to spin-1/2 or harmonic bath is given by a superposition of several Ornstein-Uhlenbeck noise processes as follows;

$$\eta(t) = \sum_i^n \phi_i \tag{4.26}$$

where τ_i and D_i can now be interpreted as the correlation time and strength of the colour noise variable $\{\phi_i\}$. Having derived the scheme for generation of c-number noise $\eta(t)$ we may solve the quantum Langevin equation with quantum correlations (Sinha *et al.*, 2013a).

Probabilistic Description and Governing Equations

We have formulated the stochastic dynamics of a particle with quantum mechanical mean position q, and mean momentum p in terms of a c-number Langevin equation [Eq. (3.21)]. An equivalent description of the stochastic process can be formulated in terms of probability density function $P(q, p, t)$ of these variables. We begin by noting that the probability function we are considering here is distinctly different from that of quasi-probability function used to treat quasi-classical correspondence (for e.g., Wigner distribution or Glauber-Sudarshan distribution function (Glauber, 2007) which can frequently take negative values) in a sense that $P(q, p, t)$ is nonsingular and remains always positive definite function, even at $T = 0$. In what follows, we would like to enquire about the quantum analogues of Fokker-Planck, Smoluchowskii and diffusion equations and a few other relevant quantities like mean square displacement and decay rate from a metastable state.

Quantum Fokker-Planck Equation

To begin with, we consider the particle to be free from any external force field so that $V(q) = 0$. The c-number Langevin Eq. (3.21) then reduces to

$$\ddot{q}(t) + \int_0^t dt' \kappa(t-t')\dot{q}(t') = \eta(t) \tag{5.1}$$

where, $\kappa(t)$ is the memory kernel given by Eq. (3.30) and $\eta(t)$ is a zero centred c-number noise whose time correlation for spin-1/2 and bosonic bath are given by Eqs. (3.31)-(3.32) respectively. In the following we present a general prescription to construct Fokker-Planck equation (FPE) both for spin-1/2 and bosonic bath.

Let the formal solution of Eq. (5.1) is given by

$$q(t) = \langle q(t) \rangle_s + \int_0^t dt' \, H(t-t') \, \eta(t') \tag{5.2}$$

where

$$\langle q(t) \rangle_s = q(0) + p(0)H(t) \tag{5.3}$$

with $q(0)$ and $p(0)$ being, respectively, the initial values of the quantum mechanical mean position and momentum of the particle. $H(t)$ is the inverse Laplace transform of

$$\widetilde{H}(s) = \int_0^\infty dt \, e^{-st} H(t) = \frac{1}{s^2 + s\widetilde{\kappa}(s)} \tag{5.4}$$

where $\widetilde{\kappa}(s)$ denotes the Laplace transform of $\kappa(t)$. The time derivative of Eq. (5.2) gives

$$p(t) = \langle p(t) \rangle_s + \int_0^t dt' \, h(t-t') \, \eta(t') \tag{5.5}$$

where

$$\langle p(t)\rangle_s = p(0)\, h(t) \tag{5.6}$$

and

$$h(t) = \frac{d}{dt} H(t) \tag{5.7}$$

$H(t)$ and $h(t)$ are two relaxation functions. While $h(t)$ measures how the quantum mechanical mean momentum forgets its initial value, $H(t)$ measures how the quantum mechanical mean displacement forgets its initial momentum. We then make use of the solutions for $q(t)$, $p(t)$ and the symmetry property of the correlation function $\langle \eta(t)\eta(t')\rangle_s$ $[\equiv C(t-t')]$ to define the following variances as follows;

$$\sigma_{qq}^2(t) = \left\langle \left[q(t) - \langle q(t)\rangle_s \right]^2 \right\rangle_s = 2\int_0^t H(t_1)\, dt_1 \int_0^{t_1} H(t_2)\, C(t_1 - t_2)\, dt_2 \tag{5.8}$$

$$\sigma_{pp}^2(t) = \left\langle \left[p(t) - \langle p(t)\rangle_s \right]^2 \right\rangle_s = 2\int_0^t h(t_1)\, dt_1 \int_0^{t_1} h(t_2)\, C(t_1 - t_2)\, dt_2 \tag{5.9}$$

and

$$\sigma_{qp}^2(t) = \left\langle \left[q(t) - \langle q(t)\rangle_s \right]\left[p(t) - \langle p(t)\rangle_s \right] \right\rangle_s = \frac{1}{2}\dot{\sigma}_{qq}^2 = \int_0^t H(t_1)\, dt_1 \int_0^{t_1} h(t_2)\, C(t_1 - t_2)\, dt_2 \tag{5.10}$$

Assuming now that the statistical distribution of c-number noise $\eta(t)$ to be Gaussian [Eq. (3.23) or Eq. (3.24)], we define the joint characteristic function [$\tilde{P}(\mu,\rho,t)$] in terms of standard mean values and variances (these mean and variance are not to be confused with quantum mechanical mean of an operator and its dispersion as discussed in Sec. IV.B) as follows;

$$\tilde{P}(\mu,\rho,t) = exp \left[i\mu\langle q\rangle_s + i\rho\langle p\rangle_s - \frac{1}{2}\left\{ \sigma_{qq}^2\mu^2 + 2\sigma_{qp}^2\mu\rho + \sigma_{pp}^2\rho^2 \right\} \right] \tag{5.11}$$

Using standard procedure (Mazo, 1978a; Mazo, 1978b), we can write down the Fokker-Planck equation (FPE) obeyed by the joint probability distribution function $P(q, p, t)$ which is the inverse Fourier transform of characteristic function $\tilde{P}(\mu,\rho,t)$ as

$$\left(\frac{\partial}{\partial t} + p\frac{\partial}{\partial q} \right) P(q,p,t) = \Lambda(t)\frac{\partial}{\partial p}\, p\, P(q,p,t) + \phi(t)\frac{\partial^2}{\partial p^2} P(q,p,t) + \psi(t)\frac{\partial^2}{\partial q\partial p} P(q,p,t) \tag{5.12}$$

where,

$$\Lambda(t) = -\frac{\dot{h}(t)}{h(t)} \tag{5.13}$$

$$\phi(t) = \Lambda(t)\sigma_{pp}^2(t) + \frac{1}{2}\dot{\sigma}_{pp}^2(t) \tag{5.14}$$

and

$$\psi(t) = -\sigma_{pp}^2(t) + \Lambda(t)\sigma_{qp}^2(t) + \dot{\sigma}_{qp}^2(t) \tag{5.15}$$

Eq. (5.12) is the quantum analogue of non-Markovian classical Fokker-Planck equation (FPE) and valid for arbitrary temperature and friction. In the long time limit it can be shown that the non-Markovian coefficient $\psi(\infty) = 0$. The stationary solution of the Fokker-Planck Eq. (5.12) is then given by,

$$P_{st}(p_0) = \frac{1}{(2\pi\Delta_0)^{\frac{1}{2}}} exp\left[-\frac{p_0^2}{2\Delta_0} \right] \text{where,}$$

$$\Delta_0 = \frac{\phi(\infty)}{\Lambda(\infty)} \tag{5.16}$$

In the stationary state the system, as expected, therefore attains a Gaussian distribution whose half-width is determined by Δ_0.

Quantum Diffusion Equation

To formulate the quantum diffusion equation for the particle in spin-1/2 (Sinha *et al*., 2010) and bosonic bath we follow Mazo's procedure (Mazo, 1978a; Mazo, 1978b) for derivation of classical diffusion equation. We put $\rho = 0$ in Eq. (5.11) and consider the Gaussian distribution $P_{st}(p_0)$ of initial momentum to p_0 take average over the characteristic function as follows;

$$\begin{aligned}\widetilde{P}(\mu,t) &= \int \widetilde{P}(\mu,t)P_{st}(p_0)\, dp_0 \\ &= exp\left(-\frac{1}{2}\mu^2\sigma_{qq}^2 + i\mu q_0\right)\int e^{(i\mu p_0 H(t))}P_{st}(p_0)dp_0\end{aligned} \tag{5.17}$$

Using Eq. (5.16) in Eq. (5.17) and after performing inverse Fourier transform as before we arrive at the following diffusion equation after some algebra

$$\frac{\partial P(q,t)}{\partial t} = D_f(t)\frac{\partial^2 P}{\partial q^2} \tag{5.18}$$

where, $D_f(t)$ is the quantum diffusion coefficient which is given by,

$$D_f(t) = \sigma_{qp}^2(t) + \Delta_0 H(t)h(t) \tag{5.19}$$

The expression for quantum diffusion coefficient is characterized by the nature of relaxation function $H(t)$ and $h(t)$ as well as by the characteristic correlation function of c-number noise for the spin-1/2 bath [Eq. (3.31)] and bosonic bath [Eq. (3.32)] through $\sigma_{qp}^2(t)$. Since the key quantity that determines the variances is the relaxation function $H(t)$, it is necessary to incorporate correct well-behaved relaxation functions which are to be obtained from appropriate spectral density function. For a typical choice of Lorentzian distribution of frequency, we assume a spectral density $J(\omega)$ of the form:

$$J(\omega) = \frac{2}{\pi}\frac{\gamma\omega}{1+\omega^2\tau_c^2} \tag{5.20}$$

By virtue of Eq. (5.4) the relaxation function $H(t)$ is given by,

$$H(t) = \frac{1}{\gamma}\left[1 - Ae^{-t/2\tau_c}\sin(\lambda t + \alpha)\right] \tag{5.21}$$

which yields,

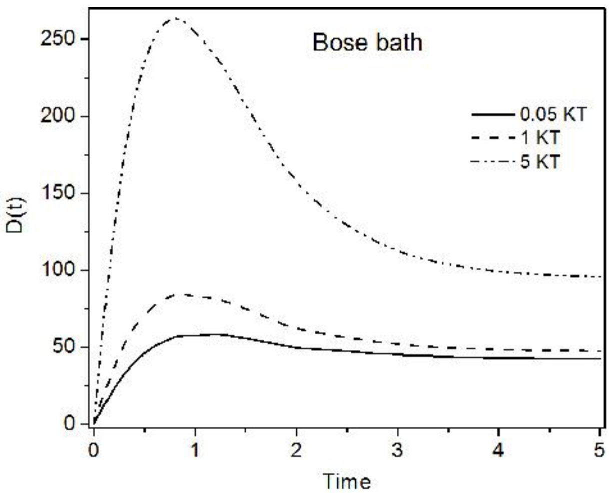
$$h(t) = \frac{dH(t)}{dt} \tag{5.22}$$

where,

$$A = \frac{\gamma}{\lambda}\lambda = \left(\frac{\gamma}{\tau_c} - \frac{1}{4\tau_c^2}\right)^{1/2}$$

$$\alpha = \tan^{-1}\left(\frac{2\tau_c\gamma}{1-2\tau_c\gamma}\right) \tag{5.23}$$

The time dependence of diffusion coefficient $D_f(t)$ is due to the relaxation functions $H(t)$ and $h(t)$ and the correlation functions of the respective bath [Eqs. (4.21)-(4.22)]. These functions govern the short time behavior of $D_f(t)$. To illustrate, the plot of $D_f(t)$ *vs* time for both spin-1/2 and bosonic bath at different temperatures is shown in Figs. 5.1(a)-(b). While the short time regime is characterized by a sharp increase in the asymptotic limit $D_f(t)$ settles down to a constant value. A notable conclusion of the present analysis is the temperature dependence of the steady state diffusion constant for fermionic (spin-1/2) bath in contrast to bosonic bath, which may be attributed to the widths of thermal distributions characterized by tanh and coth factors. The anomalous decrease of diffusion constant with increase in temperature had been observed earlier (Jorch, 1981) in positron diffusion of single crystal



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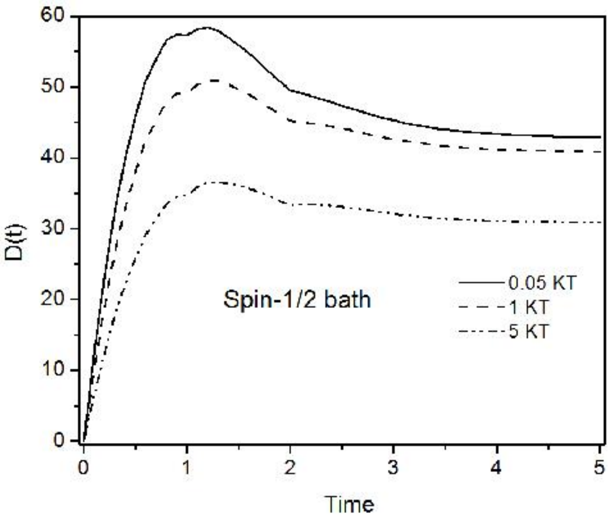


(b)

Fig. 5.1: Variation of quantum diffusion coefficient vs time at different temperatures for (a) Harmonic bath and (b) spin-1/2 bath at $\gamma = 2.83$ (Scale arbitrary)

Ge in one dimension. Thus in contrast to crossover from coherent to incoherent quantum tunneling characteristic of excitation of many levels of single degree of freedom in a bosonic reservoir, increase of temperature favours the coherent dynamics in a spin-1/2 bath (Shao and Hanggi, 1998; Rosch, 1999).The suppression of diffusion at higher temperature as observed here also points towards this coherence as a result of severe restriction on the excitations in spin-1/2 bath degrees of freedom. This is in conformity with earlier studies (Rosch, 1999) based on path integral approach to quantum transport of heavy particle in spin-1/2 bath.

It is now instructive to calculate the mean square displacement $\left\langle \hat{q}^2(t) \right\rangle_{qs}$ of the particle. From Eq. (4.16) we have

$$\left\langle \hat{q}^2(t) \right\rangle_{qs} = \left\langle q^2 \right\rangle_s + \left\langle \delta\hat{q}^2(t) \right\rangle_{qs} \qquad (5.24)$$

Here, $\left\langle q^2 \right\rangle_s$ can be calculated as an usual statistical average of q^2 with the probability density function $P(q, t)$ as follows;

$$\left\langle q^2 \right\rangle_s = \int q^2 P(q,t)dq = 2S(t) \qquad (5.25)$$

$P(q, t)$ is the solution of the quantum diffusion Eq. (5.18) subject to initial condition $P(q,t=0)$ = $\delta(q)$ and is given by $P(q, t)$ = $(1/\sqrt{4\pi S(t)})$ $exp[-q^2/4S(t)]$ with $S(t)$ = $\int_0^t dt' D_f(t')$. Now $\left\langle \delta\hat{q}^2 \right\rangle$ can be calculated for the free particle from Eq. (4.6), which assumes the following form;

$$\delta\ddot{\hat{q}} + \int_0^\infty dt'\ \kappa(t-t')\ \delta\dot{\hat{q}}(t') = \delta\hat{\eta}(t) \qquad (5.26)$$

Solving Eq. (5.26) by Laplace transform we have

$$\delta\hat{q}(t) = \delta\hat{q}(0) + H(t)\delta\hat{p}(0)$$

$$+\int_0^t dt' H(t-t')\delta\hat{\eta}(t') \qquad (5.27)$$

where, $H(t)$ is given by Eqs. (5.4) and (5.21). After squaring $\left\langle \delta\hat{q}^2(t) \right\rangle_{qs}$ can be expressed as

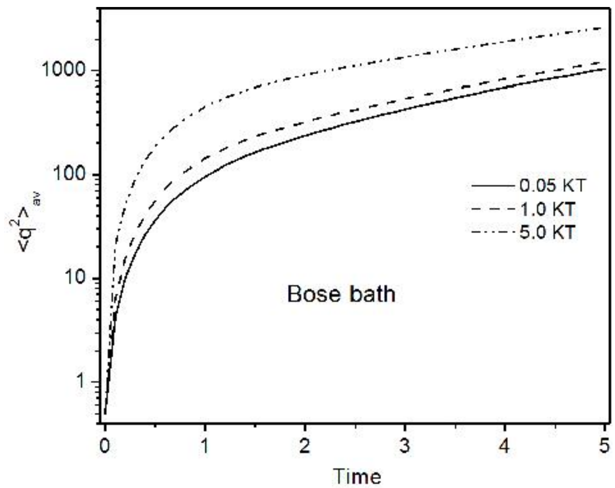
$$\left\langle \delta\hat{q}^2(t) \right\rangle_{qs} = \left\langle \delta\hat{q}^2(0) \right\rangle + H^2(t)\left\langle \delta\hat{p}^2(0) \right\rangle$$

$$+ H(t)\left\langle \delta\hat{q}(0)\delta\hat{p}(0) + \delta\hat{p}(0)\delta\hat{q}(0) \right\rangle$$

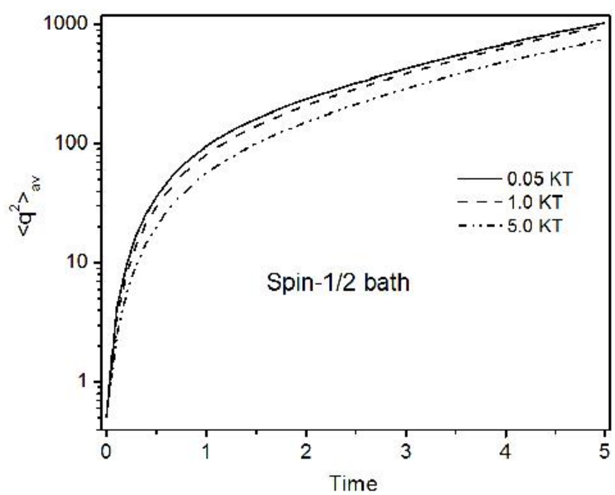
$$+2\int_0^t dt'\int_0^t dt''H(t-t')H(t-t'')$$

$$\left\langle \delta\hat{\eta}(t')\delta\hat{\eta}(t'')\right\rangle_{qs} \tag{5.28}$$

We now choose the initial conditions corresponding to minimum uncertainty state $\left\langle \delta\hat{q}^2(0)\right\rangle$ = $\left\langle \delta\hat{p}^2(0)\right\rangle$ = 0.5 and, $\langle\delta\hat{q}(0)\delta\hat{p}(0)$ + $\delta\hat{p}(0)\delta\hat{q}(0)\rangle$ = 1.0 so that the double integral in Eq. (5.28) can be evaluated numerically to calculate $\left\langle \delta\hat{q}^2(t)\right\rangle_{qs}$. We are, therefore, in a position to estimate the quantum mean square displacement $\left\langle \hat{q}^2(t)\right\rangle_{qs}$ [Eq. (5.24)]. The results are shown in Fig. 5.2(a)-(b) at different temperatures. It is apparent that in the balastic region, the rise is faster than quadratic while the mean square displacement is supper diffusive in the asymptotic limit. In general, one observes that higher temperature leads to suppression of the displacement for spin-1/2 bath (Sinha *et al*., 2010).



(a)



(b)

Fig. 5.2: Variation of quantum mean square displacement, $\left\langle \hat{q}^2(t)\right\rangle_{qs}$ *vs* time at different temperatures for (a) Harmonic bath and (b) Spin-1/2 bath at γ = 2.83 (Scale arbitrary)

Quantum Smoluchowski Equation

The diffusion of a particle in an external potential field $V(q)$ is described by the Langevin Eqs. (3.21)-(3.22). In the over damped limit we assume a Lorentzian spectral density $\dfrac{\pi}{2}\dfrac{\gamma_0\omega}{1+(\omega-\omega_0)^2\tau_c^2}$ which in the limit $\tau_c \to 0$ results in a damping kernel [Eq. (3.30)] as

$$\kappa(t-t') = \gamma_0\int_{-\infty}^{+\infty} d\omega\, cos\omega(t-t') = \gamma_0\delta(t-t') \tag{5.29}$$

where, γ_0 is the dissipation constant. ω_0 is the linearised static frequency of the system. Proceeding as before we derive the Markovian limit of the correlation function $\langle\eta(t)\eta(t')\rangle_s^{F,B}$ as

$$\langle\eta(t)\eta(t')\rangle_s^{F,B} = \int_{-\infty}^{+\infty} d\omega\frac{\pi}{2}\left[\frac{\gamma_0}{1+(\omega-\omega_0)^2\tau_c^2}\right]$$

$$D^{F,B}\times\cos\omega(t-t') \tag{5.30}$$

where, $D^{F,B}$ denotes the noise strength in the

Markovian limit for the spin-1/2 or harmonic bath, respectively. For vanishing τ_c, the distribution function around ω_0 becomes broad so that one may take the slowly varying quantity $D^{F,B}$ out of the integration over frequency centred at ω_0 with a value $\left[\frac{\hbar\omega_0}{2} tanh\left(\frac{\hbar\omega_0}{2KT}\right)\right]$ for spin-1/2 bath and $\left[\frac{\hbar\omega_0}{2}\coth\left(\frac{\hbar\omega_0}{2KT}\right)\right]$ for bosonic bath. We are then led to the following expressions for the correlation functions;

$$\langle\eta(t)\eta(t')\rangle_s^F = \gamma_0\left[\frac{\hbar\omega_0}{2}\tanh\left(\frac{\hbar\omega_0}{2KT}\right)\right]$$

$$\delta(t-t') = \gamma_0 D^F \delta(t-t') \tag{5.31}$$

$$\langle\eta(t)\eta(t')\rangle_s^F = \gamma_0\left[\frac{\hbar\omega_0}{2}\coth\left(\frac{\hbar\omega_0}{2KT}\right)\right]$$

$$\delta(t-t') = \gamma_0 D^B \delta(t-t') \tag{5.32}$$

Based on these considerations we may now construct a probabilistic description of the stochastic dynamics in the over damped limit. In this limit we drop the inertial term in Eq. (3.21) and make use of expression [Eq. (5.29)] to obtain (Sinha *et al.*, 2011b)

$$\gamma_0\dot{q} + V'_{quan}\left(q,\langle\delta\hat{q}^n\rangle\right) = \eta(t) \tag{5.33}$$

where,

$$V'_{quan}\left(q,\langle\delta\hat{q}^n\rangle\right) = V'(q) - Q\left(q,\langle\delta\hat{q}^n\rangle\right) \tag{5.34}$$

and $Q\left(q,\langle\delta\hat{q}^n\rangle\right)$ represents quantum correction due to system potential [Eq. (4.5)]. Since the system is thermodynamically closed the density is conserved, we have $\int\rho(q,t)dq = 1$, which ensures that any change in density with time is balanced by the divergence of a current $(\dot{q}\rho)$ by the continuity equation of the following form:

$$\frac{\partial\rho}{\partial t} + \frac{\partial(\dot{q}\rho)}{\partial q} = 0 \tag{5.35}$$

Following Zwanzig (Zwanzig, 2001) and using van Kampen's Lemma (van Kampen, 1992) by defining $\langle\rho(q,t)\rangle_s = P(q,t)$, where $P(q,t)$ is the probability density function, one arrives at the following equation

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial q}\left(\frac{V'_{quan}}{\gamma_0}\right)P + \frac{1}{\gamma_0^2}\int_0^t dt'$$

$$\langle\eta(t)\eta(t')\rangle_s^{F,B} e^{-(\phi(t)-\phi(t'))}\frac{\partial^2 P(q,t')}{\partial q^2} \tag{5.36}$$

where $\phi(t) = -\int_0^t \frac{\partial V'}{\partial q}\frac{1}{\gamma_0}dt'$.

The use of correlation function [Eqs. (5.31)-(5.32)] in Eq. (5.36) yields our desired Smoluchowski equation as follows:

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial q}\left(\frac{V'_{quan}}{\gamma_0}\right)P + D_f^{F,B}\frac{\partial^2 P}{\partial q^2} \tag{5.37}$$

Here the diffusion coefficients of the particle in a spin-1/2 bath and bosonic bath (in the Markovian limit) are given by,

$$D_f^F = \frac{\hbar\omega_0}{2\gamma_0} tanh\left(\frac{\hbar\omega_0}{2KT}\right) \tag{5.38}$$

$$D_f^B = \frac{\hbar\omega_0}{2\gamma_0}\coth\left(\frac{\hbar\omega_0}{2KT}\right) \tag{5.39}$$

Eq. (5.37) is the classical-looking Smoluchowski equation which describes the over damped quantum dynamics of the particle in terms of a probability density function $P(q, t)$ where q is the quantum mechanical mean value of position of the particle. Two important points may be noted. First, the potential $V_{quan}(q,\langle\delta\hat{q}^n\rangle)$ includes the quantum corrections, in principle, to all orders. Second, while $tanh\left(\frac{\hbar\omega_0}{2KT}\right)$ makes an imprint of the Fermi-Dirac character of the spin-1/2 bath [Eq. (5.38)], $\coth\left(\frac{\hbar\omega_0}{2KT}\right)$ makes an imprint of harmonic bath through the expression for diffusion coefficients [Eq. (5.39)] respectively. Lastly, we make a note that Smoluchowski Eq. (5.37) allows an equilibrium solution under zero current condition as $\sim exp\left(-\frac{V_{quan}(q,\langle\delta\hat{q}^n\rangle)}{D^{F,B}}\right)$. This is independent of the dissipation constant and thus ensures thermodynamic consistency of the present scheme.

A quantity of special interest here is the escape rate of an over damped particle from a metastable well which is subjected to quantum fluctuation due to spin-1/2 or bosonic bath. The time evolution of the particle is governed by the Smoluchowski Eq. (5.37) when the potential V'_{quan} in Eq. (5.37) is given by Eq. (4.14). One may derive the escape rate using flux-over-population method (Hanggi, 1990) by solving Smoluchowski Eq. (5.37) subject to the condition that the mean escape time from the metastable well is much larger compared to the time scale of the over-damped quantum dynamics of mean position of the particle and that the strength of c-number noise $\left(D^{F,B}\right)$ is smaller than the barrier height $\left(i.e., D^{F,B} < E^0\right)$; E^0 refers to the barrier height of the classical potential. The escape rate is then given by

$$k = D_f^{F,B} \frac{1}{\int_{q_0}^{A} e^{V_{quan}(q)/D^{F,B}} dq \int_{q_1}^{q_2} e^{-V_{quan}(q)/D^{F,B}} dq} \tag{5.40}$$

where, $\gamma_0 D = D_f$ and the limits of integration correspond to the points referred to a typical potential form $V(q) = \frac{1}{2}aq^2 - \frac{1}{3}bq^3$ as shown in Fig. 5.3. Upon integration the escape rate reduces to the following form (Sinha *et al.*, 2011b).

$$k = \frac{\omega_b\,\omega_0}{2\pi\gamma_0} exp(-E_0 / D^{F,B})$$

$$exp\left[-\frac{\Delta}{2D^{F,B}}(\phi_b - \phi_0)\right] \tag{5.41}$$

where ω_0 and ω_b are the harmonic frequencies associated with the left well and the inverted well, respectively. The quantum contribution is manifested through ϕ_b and ϕ_0 as

$$\phi_b = \int_0^{q_b} V'''(q)(V'(q))^2 dq \text{ and}$$

$$\phi_0 = \int_0^{q_0} V'''(q)(V'(q))^2 dq \tag{5.42}$$

For the given prototypical potential depicted in Fig. 5.3, ϕ_b and ϕ_0 are $\left(-2b\int_0^{a/b}(V'(q))^2 dq\right)$ and 0, respectively. The rate constant [Eq. (5.41)] therefore assumes the following form:

$$k = \frac{\omega_b\,\omega_0}{2\pi\gamma_0} exp(-E_0 / D^{F,B})$$

$$exp\left[\frac{\Delta b}{D^{F,B}}\int_0^{a/b} dq[V'(q)]^2\right] \tag{5.43}$$

At zero temperature $(0K)$ we have

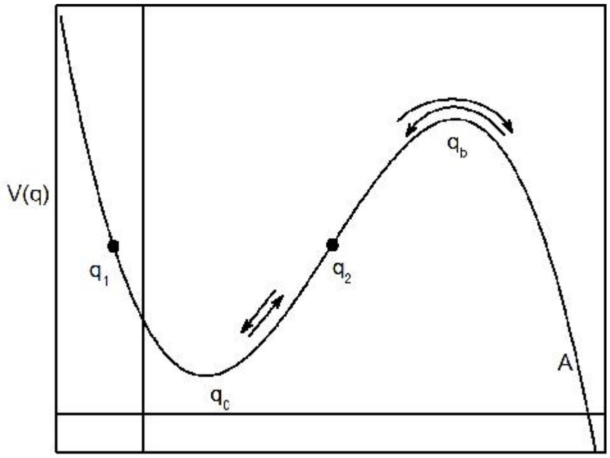


Fig. 5.3: A schematic plot of the cubic potential $V(q) = \frac{1}{2}aq^2 - \frac{1}{3}bq^3$, used for calculation of the decay of the metastable state

$D^{F,B} \to \frac{\hbar\omega_0}{2}$. The dissipative dynamics in spin-1/2 bath thus agrees well with the results obtained from bosonic bath. But at finite temperatures both the temperature dependent factors begin to differ from unity. The effective spectral density which contains a temperature dependent factor plays a significant role in the diffusion coefficient D_f as well as in the rate coefficient k of the particle. As the temperature rises there is an effective reduction of system-bath coupling due to thermal saturation in spin-1/2 bath.

Furthermore, it follows that all the quantities appearing in the square bracket in the expression [Eq. (5.43)] are positive and therefore the contribution from the quantum correction due to non-linearity of the potential enhances the rate.

Application to Chemical Dynamics; Spatial *vs* Momentum Coupling

In this section, we examine the decay of a quantum particle out of a metastable state which can tunnel and undergo thermal activation due to both spatial or momentum coupling.

Thermal Activation and Tunneling Rate; Spatial Coupling

We recall Eq. (3.21) which describes the dynamics of quantum mechanical mean motion of a particle in a thermal bath (spin-1/2 or bosonic) kept at a temperature T. The noise of the bath is described by c-number noise $\eta(t)$ [Eq. (3.19)]. The set of N-variables for bath degrees of freedom $\{\mu_k(0), \mu_k^*(0)\}$, $k = 1, 2, \ldots N$, can then be expressed in terms of momenta $\{\beta_k\}$ and coordinates $\{\alpha_k\}$ of N *fictitious* oscillators as $\beta_k = \frac{\tilde{C}(S,|\mu|_k)}{2i} \sqrt{\frac{\hbar m_k\omega_k}{S}}\left(\mu_k^* - \mu_k\right)$ and $\alpha_k = \frac{\tilde{C}(S,|\mu|_k)}{2} \sqrt{\frac{\hbar}{S m_k\omega_k}}\left(\mu_k^* - \mu_k\right)$. Using $\{\alpha_k, \beta_k\}$ the c-number Hamiltonian corresponding to Langevin Eq. (3.21)may be written down as;

$$H = \frac{p^2}{2} + \left[V(q) + \sum_{n\geq 2}\frac{1}{n!}V^{(n)}(q)\langle\delta\hat{q}^n\rangle\right]$$

$$+ \sum_k\left[\frac{\beta_k^2}{2m_k} + \frac{1}{2}\frac{c_k^2}{m_k\omega_k^2}\left(\frac{m_k\omega_k^2}{c_k^2}\alpha_k - q\right)^2\right] \quad (6.1)$$

C-number Langevin equation [Eq. (3.21)] can be derived directly from this c-number Hamiltonian after appropriate elimination of bath degrees of freedom $\{\alpha_k, \beta_k\}$ from equations of motion of the total system.Thus, the above Hamiltonian [Eq. (6.1)] is different from our starting operator Hamiltonian Eq. (2.2) because of the c-number nature of the bath variables.

Eq. (6.1) shows that it is possible to reduce the quantum dynamics of the particle in a dissipative bath (spin-1/2 orbosonic) to the dynamics of the particle in a *fictitious* harmonic bath. Furthermore, high spin limit of a spin coherent state merges to the harmonic oscillator coherent state. The spin-1/2 bath as a set of *fictitious* oscillators had been realized earlier by Makri (Makri, 1999) in a different context. Thus, although the spin-1/2 bath is nonlinear in nature, it can be conveniently described by linear response regime. The c-number variables $\mu_k(0), \mu_k^*(0)$ or $\alpha_k(0), \beta_k(0)$ follow characteristic canonical thermal distributions [Eqs. (3.23)-(3.24)] of corresponding to the spin-1/2 or bosonic bath as ensured by fluctuation-dissipation relations [Eqs. (3.31)-(3.32)]. Hamiltonian [Eq. (6.1)] is therefore the c-number equivalent of the Hamiltonian operator Eq. (2.2). Thermalization of the particle in either kind of heat bath can thus be conveniently understood in terms of this c-number Hamiltonian and the corresponding thermal canonical distribution.

The spectral density function corresponding to the Hamiltonian [Eq. (6.1)] can be defined as

$$J(\omega) = \frac{\pi}{2} \sum_k \frac{g_k^2}{\omega_k} \delta(\omega - \omega_k) \tag{6.2}$$

and $c_k = \sqrt{m_k}\, g_k$. Now we assume that at $q = 0$, the potential $V(q)$ has a barrier height $V^\#$ such that the harmonic approximation around $q = 0$ leads to:

$$V(q) = V^\# - \frac{\omega_b^2}{2} q^2 + V_2(q) \tag{6.3}$$

where, $-\omega_b^2 = V''(q)|_{q=0}$, refers to the square of the characteristic frequency of the system at barrier top and $V_2(q)$ is the nonlinear part of the classical potential which is given by

$$V_2(q) = \sum_{n \geq 3} \frac{1}{n!} V^{(n)}(q)|_{q=0}\, q^n \tag{6.4}$$

To diagonalize the Hamiltonian [Eq. (6.1)] we further define mass weighted coordinates and momenta of bath variables as

$$\bar{\alpha}_k = \sqrt{m_k}\,\alpha_k \bar{\beta}_k = \frac{\beta_k}{\sqrt{m_k}} \tag{6.5}$$

Using these newly defined bath variables the Hamiltonian [Eq. (6.1)] then may be rewritten as $H = H_0 + V_N(q)$, where

$$H_0 = \left[\frac{p^2}{2} + \sum_k \frac{\bar{\beta}_k^2}{2}\right] + \left[V_1^\# - \frac{1}{2}\omega_b^2 q^2 + \sum_k \frac{1}{2}\left(\omega_k \bar{\alpha}_k - \frac{g_k}{\omega_k} q\right)^2\right] \tag{6.6}$$

We neglect V_N, the nonlinear terms of the potential and assume

$$V_1^\# = V^\# \tag{6.7}$$

as the quantum correction to barrier height is small. As we are interested in the normal mode analysis of a system of N '*fictitious*' oscillators and one inverted system oscillator within harmonic approximation, a normal mode transformation to convert the quadratic Hamiltonian [Eq. (6.6)] into a diagonal form (Pollak, 1986; Levine *et al.*, 1988) is carried out. The c-number Hamiltonian of the unstable normal coordinate is then given by,

$$H_0 = \frac{1}{2}\dot{\rho}^2 + V^\# - \frac{1}{2}\lambda_b^2 \rho^2 + \frac{1}{2}\sum_{k=1}^{N}\left(\dot{x}_k^2 + \lambda_k^2 x_k^2\right) \tag{6.8}$$

where, λ_b and $\{\lambda_k\}$ are the characteristic frequencies of the unstable and stable normal modes of the system and the bath.These are given by,

$$\lambda_i^2 = -\frac{\omega_b^2}{1 + \sum_{k=1}^{N} \frac{g_k^2}{\omega_k^2(\omega_k^2 - \lambda_i^2)}} \tag{6.9}$$

and

$$\lambda_b^2 = \frac{\omega_b^2}{1+\sum_{k=1}^{N}\frac{g_k^2}{\omega_k^2(\omega_k^2+\lambda_b^2)}} \tag{6.10}$$

The time dependent friction coefficient $[\kappa(t)]$ of the system in continuum limit can be expressed using $J(\omega)$ [Eq. (6.2)] as

$$\kappa(t) = \frac{2}{\pi}\int_0^\infty d\omega\,\frac{J(\omega)}{\omega}\cos(\omega t) \tag{6.11}$$

which on Laplace transform gives

$$\tilde{\kappa}(s) = \frac{2}{\pi}\int_0^\infty d\omega\,\frac{J(\omega)}{\omega}\frac{s}{\omega^2+s^2} \tag{6.12}$$

where $\tilde{\kappa}(s) = \mathcal{L}(\kappa(t))$. Combining Eqs. (6.10) and (6.12) we have

$$\lambda_b^2 = \frac{\omega_b^2}{1+\sum_{k=1}^{N}\frac{g_k^2}{\omega_k^2(\omega_k^2+\lambda_b^2)}} = \frac{\omega_b^2}{1+\frac{2}{\pi}\int_0^\infty \frac{J(\omega)}{\omega}\frac{d\omega}{\omega^2+\lambda_b^2}} = \frac{\omega_b^2}{1+\frac{\tilde{\kappa}(\lambda_b)}{\lambda_b}} \tag{6.13}$$

Since, $J(\omega)\geq 0; \tilde{\kappa}(\lambda_b) > 0$, the effective barrier frequency is always less than ω_b for spatial coupling. An opposite scenario appears due to momentum coupling which leads to the phenomena of stochastic acceleration (Fermi, 1949; Sturrock, 1966). We will examine that in the following section.

Next, we proceed to normal mode analysis for N *fictitious* bath oscillators and one harmonic oscillator for the system mode.The harmonic Hamiltonian for the system mode can be obtained from Eq. (6.1) with the replacement of $V(q)$ by $\frac{\omega_0^2}{2}(q-q_0)^2$ (where the potential is linearized at $q = q_0$) and we neglect the nonlinear terms as before. After transformation to the normal mode, the Hamiltonian can be expressed as

$$H_0^{'} = \frac{1}{2}\dot{\rho}'^2 + \frac{1}{2}\lambda_0^2\rho'^2 + \frac{1}{2}\sum_{k=1}^{N}\left\{\dot{x}_{k'}^2 + \frac{1}{2}\Lambda_k^2 x_k'^2\right\} \tag{6.14}$$

where, $\lambda_0, \{\Lambda_k\}$ refer to the frequencies of the stable normal modes of the system and the bath, respectively. Here λ_0 and $\{\Lambda_k\}$ assume the following expressions

$$\lambda_0^2 = \frac{\omega_0^2}{1+\sum_{k=1}^{N}\frac{g_k^2}{\omega_k^2(\omega_k^2-\lambda_0^2)}} = \frac{\omega_0^2}{1+\frac{2}{\pi}\int_0^\infty \frac{J(\omega)}{\omega}\frac{d\omega}{(\omega^2-\lambda_0^2)}} \tag{6.15}$$

$$\Lambda_i^2 = \frac{\omega_0^2}{1+\sum_{k=1}^{N}\frac{g_k^2}{\omega_k^2(\omega_k^2-\Lambda_i^2)}} \tag{6.16}$$

It is also pertinent to note the two identities for the associated frequencies for the dynamical barrier top and the bottom of the well obey the following relations;

$$\begin{aligned}\omega_b^2\prod_{i=1}^{N}\omega_i^2 &= \lambda_b^2\prod_{j=1}^{N}\lambda_j^2\\ \omega_0^2\prod_{i=1}^{N}\omega_i^2 &= \lambda_0^2\prod_{j=1}^{N}\Lambda_j^2\end{aligned} \tag{6.17}$$

We then calculate the rate of escape of the particle trapped in a potential $V(q)$ depicted schematically in Fig. 5.3. In the normal mode description of (N+1) oscillator of the Hamiltonian [Eq. (6.8)], the system and bath modes are uncoupled. Considering the unstable reaction coordinate to be thermalized according to canonical thermal distribution due to spin-1/2 or bosonic bath [Eqs. (3.23) and (3.24)] we have

$$f_{eq}^{F,B} = Z_{F,B}^{-1} exp\left[-\frac{\frac{1}{2}\dot{\rho}^2 + V^{\#} - \frac{1}{2}\lambda_b^2\rho^2}{\hbar\lambda_0\left(1\mp 2\bar{n}(\lambda_0)\right)} \right]$$

$$\prod_{k=1}^{N} exp\left[-\frac{\frac{1}{2}\dot{x}_k^2 + \frac{1}{2}\lambda_k^2 x_k^2}{\hbar\lambda_k\left(1\mp 2\bar{n}(\lambda_k)\right)} \right] \quad (6.18)$$

where, $Z_{F,B}^{-1}$ is normalization constant. The procedure is the same as what is followed (Pechukas, 1981; Melnikov and Meshkov, 1986; Miller *et al.*, 1990) in multidimensional transition state theory (MDTST). As usual using the distributions [Eqs. (3.23) and (3.24)] it can be calculated even inside the reaction well. For this it is necessary to consider the normal mode Hamiltonian at the bottom of the well i.e., $H_0^{'}$. The corresponding distribution is given by

$$f_{eq}^{F,B} = Z_{F,B}^{-1} exp\left[-\frac{\frac{1}{2}\dot{\rho}'^2 + \frac{1}{2}\lambda_0^2\rho'^2}{\hbar\lambda_0\left(1\mp 2\bar{n}(\lambda_0)\right)} \right]$$

$$\prod_{k=1}^{N} exp\left[-\frac{\frac{1}{2}\dot{x}_k'^2 + \frac{1}{2}\Lambda_k^2 x_k'^2}{\hbar\Lambda_k\left(1\mp 2\bar{n}(\Lambda_k)\right)} \right] \quad (6.19)$$

These distribution functions may be normalized to give

$$Z_{F,B}^{-1} = \frac{\lambda_0}{2\pi\hbar\lambda_0\left(1\mp 2\bar{n}(\lambda_0)\right)}$$

$$\prod_{k=1}^{N} \frac{\Lambda_k}{2\pi\hbar\Lambda_k\left(1\mp 2\bar{n}(\Lambda_k)\right)} \quad (6.20)$$

Now using the identities Eq. (6.18) we obtain,

$$Z_{F,B}^{-1} = \frac{\omega_0}{2\pi\hbar\lambda_0\left(1\mp 2\bar{n}(\lambda_0)\right)}$$

$$\prod_{k=1}^{N} \frac{\omega_k}{2\pi\hbar\Lambda_k\left(1\mp 2\bar{n}(\Lambda_k)\right)} \quad (6.21)$$

Making use of the expression for normalization constant followed by integration over stable normal modes we obtain

$$f_{eq}^{F,B} = \frac{\omega_0}{2\pi\hbar\lambda_0\left(1- 2\bar{n}(\lambda_0)\right)} \frac{\lambda_b}{\omega_b} \chi^{F,B}$$

$$exp\left[-\frac{\frac{1}{2}\dot{\rho}^2 + V^{\#} - \frac{1}{2}\lambda_b^2\rho^2}{\hbar\lambda_0\left(1\mp 2\bar{n}(\lambda_0)\right)} \right] \quad (6.22)$$

where, $\chi^{F,B} = \prod_{k=1}^{N} \frac{\hbar\lambda_k\left(1\mp 2\bar{n}(\lambda_k)\right)}{\hbar\Lambda_k\left(1\mp 2\bar{n}(\Lambda_k)\right)}$

Rewritting total energy of the unstable mode as

$$E = \frac{1}{2}\dot{\rho}^2 + V^{\#} - \frac{1}{2}\lambda_b^2\rho^2 \quad (6.23)$$

and returning to the energy space from $\rho, \dot{\rho}$, the equilibrium distribution function is given by,

$$P_{eq}^{F,B}(E) = \frac{\omega_0}{2\pi\hbar\lambda_0\left(1\mp 2\bar{n}(\lambda_0)\right)} \frac{\lambda_b}{\omega_b} \chi^{F,B}$$

$$exp\left[-\frac{E}{\hbar\lambda_0\left(1\mp 2\bar{n}(\lambda_0)\right)} \right] \quad (6.24)$$

Eq. (6.25) is the required distribution we set out to search for which is valid for $E > V^{\neq}$ as well as $E < V^{\neq}$. The expression of in $\Gamma = \int_{V^{\neq}}^{\infty} P_{eq}(E)dE$ where $V^{\neq}$ refers to the activation barrier for the potential, yields

$$\Gamma_{QM} = \frac{\omega_0}{2\pi}\frac{\lambda_b}{\omega_b}\chi^{F,B}\, exp\left[-\frac{V^{\#}}{\hbar\lambda_0\left(1-2\bar{n}(\lambda_0)\right)}\right] \tag{6.25}$$

Use of Eq. (6.18) in the expression $\chi^{F,B}$ results in

$$\chi_{spin} = \frac{\omega_b\lambda_0}{\omega_0\lambda_b}\prod_{k=1}^{N}\frac{\tanh\left(\frac{\hbar\lambda_k}{2KT}\right)}{\tanh\left(\frac{\hbar\Lambda_k}{2KT}\right)} = \chi^F \tag{6.26}$$

$$\chi_{bosonic} = \frac{\omega_b\lambda_0}{\omega_0\lambda_b}\prod_{k=1}^{N}\frac{\coth\left(\frac{\hbar\lambda_k}{2KT}\right)}{\coth\left(\frac{\hbar\Lambda_k}{2KT}\right)} = \chi^B \tag{6.27}$$

Eq. (6.26) expresses the decay rate which includes both tunneling and thermal activation at a finite temperature. This is a product of four terms. The first factor $\frac{\omega_0}{2\pi}$ is due to classical transition state theory, while $\frac{\lambda_b}{\omega_b}$ corresponds to the Grote-Hynes modification of classical Kramersrate (Grote and Hynes, 1980).Among the other two factors, the last term is the Arrhenius factor where the usual thermal width is replaced by $\hbar\lambda_0\left[1\mp 2\bar{n}(\lambda_0)\right]$ that includes quantum effects due to spin-1/2 bath or bosonic bath for arbitrary temperature. As pointed out earlier, although both baths behave identically at 0K, the spin-1/2 bath does not reduce to usual classical limit at high temperature. In the high temperature Arrhenius factor for bosonic bath reduces to KT and one recovers the usual Boltzmann factor. While the other term χ is essentially quantum correction contribution to the Kramers-Grote-Hynes factor or more precisely vacuum corrected generalized Wolynes contribution or due to quantum transmission and reflection for the finite barrier (Wolynes, 1981).We consider the following two limiting cases separately;

(i) Tunneling at 0K

When T approaches 0K, i.e. $\frac{1}{KT}\to\infty$, both hyperbolic tangent and cotangent factors become unity. This reduces the factor $\chi = \frac{\omega_b\lambda_0}{\omega_0\lambda_b}$. Therefore the decay rate for both spin-1/2 and harmonic bath becomes

$$\Gamma_{at\,0K} = \frac{\omega_0}{2\pi}\left(\frac{\lambda_b}{\omega_b}\right)\left(\frac{\omega_b\lambda_0}{\omega_0\lambda_b}\right)exp\left(-\frac{V^{\#}}{\hbar\lambda_0}\right) \tag{6.29}$$

Now, we may note that the main effect of the bath is to modify the bare frequencies. By virtue of Eq. (6.15), dissipation always decreases λ_0 compared to ω_0. For a frequency-dependent bath, a Drude form of spectral density $J(\omega) = \frac{\pi}{2}\frac{\gamma_0\omega}{1+(\omega-\omega_0)^2\tau_c^2}$ (Pollok, 1986; Levine *et al.*, 1988) corresponds to a time dependent friction coefficient $\kappa(t) = \left(\frac{\gamma_0}{\tau_c}\right)e^{-\frac{t}{\tau_c}}$, where τ_c is the characteristic correlation time. Then for small τ_c, the modified well frequency [Eq. (6.15)] becomes $\lambda_0 \sim \omega_0\left(1-\frac{\gamma_0^3\tau_c}{4\omega_0^2}\right)$. Furthermore by choosing a cubic potential of the form $V(q) = \frac{a}{2}q^2 - \frac{b}{3}q^3$ [Fig. 5.3], the barrier height $V^{\#}$ can be expressed as $V^{\#} = \frac{2}{27}\omega_b^2(\Delta q)^2$ where Δq is the tunneling length under harmonic barrier and also we have $\omega_b = \omega_0$. The decay rate can then be calculated as

$$\Gamma_{at\,0K} = \left(\frac{\lambda_0}{2\pi}\right) exp\left[-\frac{A\omega_0(\Delta q)^2}{\hbar}\left(1+\frac{\gamma_0}{\omega_0}\phi\right)\right] \tag{6.30}$$

where $A = \frac{2}{27}$ and $\phi = \frac{\gamma_0^2 \tau_c}{4\omega_0}$. Eq. (6.30) implies that the tunneling rate at 0K is multiplied by a factor $exp\left[-A(\Delta q)^2 \gamma_0 \phi / \hbar\right]$ because of dissipation. For small τ_c and large γ_0 such that $\tau_c \gamma_0 \sim 1$, it is apparent that ϕ which is a function of (γ_0 / ω_0) is of the order of unity. Thus the tunneling probability is exponentially damped by friction as it goes as $exp\left[-A(\Delta q)^2 \gamma_0 / \hbar\right]$. This is in conformity with the seminal work by Caldeira and Leggett in eighties who first predicted exponential damping of tunneling at 0K in a harmonic bath (Caldeira and Leggett, 1981) The reason for this agreement may be traced to the basic that at 0K both the harmonic bath and the spin-1/2 bath behave identically.

(ii) Finite Temperatures

We first note that at higher temperature, the leading order contribution to the rate is mainly controlled by the χ factor. Therefore we consider following two cases;

1. Spin-1/2 Bath

We may rewrite χ_{spin} in the form

$$\chi_{spin} = \frac{\omega_b}{\omega_0}\frac{\lambda_0}{\lambda_b}$$

$$\prod_{k=1}^{N} \frac{sinh(\hbar\beta\lambda_k/2)}{cosh(\hbar\beta\lambda_k/2)} \frac{cosh(\hbar\beta\Lambda_k/2)}{sinh(\hbar\beta\Lambda_k/2)} \tag{6.30}$$

After expanding in powers of $\hbar$ we obtain after some algebra (Sinha *et al.*, 2011a)

$$\chi_{spin} = \left(\frac{\omega_b \lambda_0}{\omega_0 \lambda_b}\right)^2$$

$$\left[1 - \frac{\hbar^2\beta^2}{12}\left(\lambda_0^2 - \sum_k \Lambda_k^2\right) + \frac{\hbar^2\beta^2}{12}\left(\lambda_b^2 - \sum_k \lambda_k^2\right)\right] \tag{6.31}$$

Making use of the relation $\sum_k (\Lambda_k^2 - \lambda_k^2)$ = $(\omega_0^2 + \omega_b^2)$ – $(\lambda_b^2 + \lambda_0^2)$ we finally obtain the high temperature expression for χ_{spin} as follows;

$$\chi_{spin} = \left(\frac{\omega_b \lambda_0}{\omega_0 \lambda_b}\right)^2 \left[1 + \frac{\hbar^2\beta^2}{12}\left(\omega_0^2 + \omega_b^2 - 2\lambda_0^2\right)\right] \tag{6.32}$$

Eq. (6.32) can now be used in Eq. (6.26) with $tanh\left(\frac{\hbar\lambda_0}{2KT}\right) \sim \frac{\hbar\lambda_0}{2KT}$ to obtain,

$$\Gamma_{T\,high} = \left(\frac{\omega_0}{2\pi}\right)\left(\frac{\lambda_b}{\omega_b}\right)$$

$$\left\{\left(\frac{\omega_b\lambda_0}{\omega_0\lambda_b}\right)^2 \left[1 + \frac{\hbar^2\left(\omega_0^2 + \omega_b^2 - 2\lambda_0^2\right)}{12(KT)^2}\right]\right\}$$

$$exp\left[-\frac{V^{\#}}{(\hbar\lambda_0)^2 / 2KT}\right] \tag{6.33}$$

For cubic potential of the form chosen earlier, we have $\omega_0 = \omega_b$. This reduces Eq. (6.33) to a simple form

$$\Gamma_{T\,high} = \left(\frac{\lambda_0}{2\pi}\right)\left(\frac{\lambda_0}{\lambda_b}\right) exp[B/T^2]$$

$$exp\left\{\frac{-V^{\#}}{(\hbar\lambda_0)^2 / 2KT}\right\} \tag{6.34}$$

where $B = \hbar^2\left(\omega_0^2 - \lambda_0^2\right)/6K^2$. χ_{spin} is therefore exponentially enhanced as the temperature is lowered.Thus not only the T^2-dependent χ factor, but also the thermal activation term which contains an explicit quantum contribution $\sim O(\hbar^2)$. This indicates that unlike classical thermal activation, thermal activation by a spin-1/2 bath includes explicit quantum character.While the thermal activation term for spin-1/2 bath decreases, χ exhibits exponential enhancement with decrease in temperature.

2. Bosonic Bath

Similarly, from Eq. (6.28) $\chi_{bosonic}$ can be expressed as (Wolynes, 1981; Barik *et al.*, 2009).

$$\chi_{bosonic} = \left[1 + \frac{\hbar^2\left(\omega_0^2 + \omega_b^2\right)}{24(KT)^2}\right] \tag{6.35}$$

The decay rate for bosonic bath at high temperature therefore takes the following form:

$$\Gamma_{T\,high} = \frac{\omega_0}{2\pi}\left(\frac{\lambda_b}{\omega_b}\right)\exp\left[\frac{\hbar^2\left(\omega_0^2 + \omega_b^2\right)}{24(KT)^2}\right]$$

$$\exp\left[-\frac{V^{\#}}{KT}\right] \tag{6.36}$$

The tunneling rate is found to be exponentially enhanced at finite temperature. As the temperature is lowered this factor increases rapidly. Since the harmonic bath approaches the classical limit at high temperature, thermal activation factor reduces to standard Arrhenius form.

Thermal Activation and Tunneling Rate; Momentum Coupling

The total Hamiltonian for the particle, bath and their interaction in case of momentum coupling can be described by the following Hamiltonian (Ghosh *et al.*, 2014).

$$\hat{H} = \frac{\hat{p}^2}{2} + V(\hat{q}) + \hbar\sum_k \omega_k \hat{n}_k$$

$$+\frac{1}{2}\sum_k c_k\hat{p}\left[m_k c_k \hat{p} + i\sqrt{\frac{\hbar m_k\omega_k}{S}}\left(\hat{S}_k^{\dagger} - \hat{S}_k\right)\right] \tag{6.37}$$

where the operators carry their usual notations as discussed in Sec. IIA. The Hamiltonian here also includes $\hat{p}^2$-containing counter-term. The presence of such a term leads to stochastic acceleration (Fermi, 1949; Sturrock, 1966) with a reduction of effective mass. After formally eliminating the bath degrees of freedom as usual (Ghosh *et al.*, 2012b) the reduced operator equations for the particle with an effective mass $M = \left(1 + \sum_k m_k c_k^2\right)^{-1}$ takes the following form

$$M\ddot{\hat{q}} + M\int_0^t dt'\dot{\hat{p}}(t')\alpha(t-t') + V'(\hat{q}) = M\hat{f}(t) \tag{6.38}$$

and

$$\dot{\hat{p}} = -V'(\hat{q}) \tag{6.39}$$

where $\alpha(t-t')$ and the noise operator $\hat{f}(t)$ are given by

$$\alpha(t-t') = \sum_k c_k^2 m_k\omega_k \sin\omega_k(t-t') \tag{6.40}$$

$$\hat{f}(t) = \frac{1}{2}\sum_k c_k\omega_k\sqrt{\frac{\hbar m_k\omega_k}{S}}$$

$$\{\hat{\sigma}_k(0)e^{i\omega_k t} + \hat{\sigma}_k^{\dagger}(0)e^{-i\omega_k t}\} \tag{6.41}$$

respectively. The shifted bath operators similar to Eqs. (2.22)-(2.23) are given by

$$\hat{\sigma}_k(0) = \hat{S}_k(0) + ic_k\sqrt{\frac{m_k S}{\hbar\omega_k}}\hat{p}(0) \tag{6.42}$$

$$\hat{\sigma}_k^\dagger(0) = \hat{S}_k^\dagger(0) - ic_k\sqrt{\frac{m_k S}{\hbar\omega_k}}\hat{p}(0) \qquad (6.43)$$

On the basis of the quantum mechanical average $\langle \ldots \rangle$ taken with the initial product separable quantum states of the particle and the bath at t = 0, $|\chi\rangle|\mu_1\rangle|\mu_2\rangle\ldots|\mu_k\rangle\ldots|\mu_N\rangle$ as before, Eqs. (6.38)-(6.39) can be cast into the form of a set of generalized c-number pseudo-quantum Langevin equations

$$M\ddot{q} + M\int_0^t \dot{p}(t')\alpha(t-t')dt' + V'(q)$$

$$= M\eta(t) + Q(q, \langle\delta\hat{q}^n\rangle) \qquad (6.44)$$

$$\dot{p}(t) = -V'(q) + Q(q, \langle\delta\hat{q}^n\rangle) \qquad (6.45)$$

The properties of the thermal bath and the dissipative mechanism can be described in a convenient way by using a momentum spectral density function $J_P(\omega)$ associated with system-bath interaction as $J_P(\omega) = \frac{\pi}{2}\sum_k c_k^2 m_k \omega_k^3 \delta(\omega - \omega_k)$.

In terms of this momentum spectral density function $J_p(\omega)$, one may define a momentum function

$$\gamma_p(t) = \frac{2}{\pi}\int_{-\infty}^{\infty} d\omega \frac{J_p(\omega)}{\omega}\cos\omega(t-t') \quad (6.46)$$

The definition of statistical average Eq. (3.25) together with the ansatz Eq. (3.23) or Eq. (3.24) can be used to show that c-number noise $\eta(t)$ due to spin-1/2 bath or bosonic bath satisfies the following relations, respectively

$$\langle\eta(t)\rangle_s = 0 \qquad (6.47)$$

$$\langle\eta(t)\eta(t')\rangle_s^F = \frac{2}{\pi}\int_{-\infty}^{\infty} d\omega \frac{J_p(\omega)}{\omega}$$

$$\left[\frac{\hbar\omega}{2}\tanh\left(\frac{\hbar\omega}{2KT}\right)\right]\cos\omega(t-t') \qquad (6.48)$$

$$\langle\eta(t)\eta(t')\rangle_s^B = \frac{2}{\pi}\int_{-\infty}^{\infty} d\omega \frac{J_p(\omega)}{\omega}$$

$$\left[\frac{\hbar\omega}{2}\coth\left(\frac{\hbar\omega}{2KT}\right)\right]\cos\omega(t-t') \qquad (6.49)$$

respectively. Finally in Eq. (6.40) can be rewritten as

$$\alpha(t) = \int_0^t dt'\gamma_p(t') = \sum_k c_k^2 m_k \omega_k \sin\omega_k t \quad (6.50)$$

The c-number Hamiltonian corresponding to the pseudo-Langevin Eqs. (6.44)-(6.45) for a particle with an effective mass can be constructed as before

$$H = \frac{p^2}{2} + \left[V(q) + \sum_{n\geq 2}\frac{1}{n!}V^{(n)}(q)\langle\delta\hat{q}^n\rangle\right]$$

$$+\frac{1}{2}\sum_k\left[m_k\omega_k^2 x_k^2 + m_k\left(\frac{p_k}{m_k} - c_k p\right)^2\right] \quad (6.51)$$

where, $x_k = \frac{\widetilde{C}(S,|\mu|_k)}{2}\sqrt{\frac{\hbar}{Sm_k\omega_k}}\left(\mu_k^* + \mu_k\right)$ and $p_k = \frac{\widetilde{C}(S,|\mu|_k)}{2i}\sqrt{\frac{\hbar m_k\omega_k}{S}}\left(\mu_k^* - \mu_k\right)$. The spectral density function corresponding to the Hamiltonian [Eq. (6.51)] is $J(\omega) = \frac{\pi}{2}\sum_k \bar{c}_k^2\omega_k^3\delta(\omega-\omega_k)$ with $\bar{c}_k = c_k\sqrt{m_k}$. We now assume that at q = 0, the potential $V(q)$ feels a barrier height $V^\#$ around $q = 0$ so that

$$V(q) = V^\# - \frac{1}{2}\omega_b^2 q^2 + V_2(q) \qquad (6.52)$$

where, $V(q)$ contains the usual anharmonic terms. To diagonalize the Hamiltonian first we may similarly define the frequency-mass weighted coordinates and momenta as follows

$$\bar{q} = \omega_b q \qquad \bar{p} = \frac{p}{\omega_b}$$

$$\bar{x}_k = \sqrt{m_k}\omega_k x_k \qquad \bar{p}_k = \frac{p_k}{\sqrt{m_k}\omega_k} \tag{6.53}$$

In terms of these newly defined mass and frequency weighted variables the Hamiltonian [Eq. (6.51)] may be rewritten as,

$$H = H_0 + V_N(q) \tag{6.54}$$

where,

$$H_0 = \frac{1}{2}\omega_b^2 \bar{p}^2 + V_1^{\#} - \frac{1}{2}\bar{q}^2 + \frac{1}{2}$$

$$\sum_k \left[\bar{x}_k^2 + \omega_k^2 \left(\bar{p}_k - \frac{\bar{c}_k}{\omega_k}\omega_b \bar{p} \right)^2 \right] \tag{6.55}$$

and V_N contains of the nonlinear terms of the potentialas discussed in the spatial coupling case. The c-number Hamiltonian for the unstable system normal mode for the system is then given by

$$H_0 = \frac{1}{2}\lambda_b^2 \dot{\rho}^2 + V^{\#} - \frac{1}{2}\rho^2 + \sum_{k=1}^{N} \left\{ \frac{1}{2}\lambda_k^2 \dot{x}_k^2 + \frac{1}{2}x_k^2 \right\} \tag{6.56}$$

where λ_b^2 and $\{\lambda_k^2\}$ are respectively the square of the frequencies of the unstable and stable normal modes of the system and bath. They are given by the following expressions

$$\lambda_i^2 = -\frac{\omega_b^2}{1 - \omega_b^2 \sum_{k=1}^{N} \frac{\bar{c}_k^2}{(\omega_k^2 - \lambda_i^2)}} \tag{6.57}$$

and

$$\lambda_b^2 = \omega_b^2 \left(1 - \omega_b^2 \sum_{k=1}^{N} \frac{\bar{c}_k^2}{(\omega_k^2 + \lambda_b^2)} \right) \tag{6.58}$$

Taking the Laplace transformation of the time dependent memory function of the system $\gamma_p(t)$ [Eq. (6.46)] we have

$$\tilde{\gamma}_p(s) = \frac{2}{\pi}\int_0^{\infty} d\omega \frac{J_p(\omega)}{\omega} \frac{s}{\omega^2 + s^2} \tag{6.59}$$

where, $\gamma_p(s) = L\ [\gamma(t)]$. Combining Eq. (6.58) and Eq. (6.59) we obtain

$$\lambda_b^2 = \omega_b^2 \left[\frac{1}{M} - \frac{\tilde{\gamma}(s)}{s} \right] \tag{6.60}$$

where, M is an effective mass $M = \left(1 + \sum_k m_k c_k^2\right)^{-1}$. Next we proceed to a normal mode analysis for N *fictitious* bath oscillators plus one harmonic oscillator for the system mode. The harmonic Hamiltonian for the system mode can be obtained from Eq. (6.51) with the replacement of $V(q)$ by $\frac{1}{2}\omega_0^2(q - q_0)^2$ (where the potential $V(q)$ is linearized at $q = q_0$) and the nonlinear terms are neglected as before. After a similar transformation the normal mode Hamiltonian in this case can be expressed as,

$$H_0^{'} = \frac{1}{2}\lambda_0^2 \dot{\rho}'^2 + \frac{1}{2}\rho'^2 + \sum_{k=1}^{N} \left\{ \frac{1}{2}\Lambda_k^2 \dot{x}_{k'}^2 + \frac{1}{2}x_k'^2 \right\} \tag{6.61}$$

where, $\lambda_0, \{\Lambda_k\}$ refer to the frequencies of the stable normal modes of the system and the bath, respectively. Here λ_0 and $\{\Lambda_k\}$ are given by the following forms,

$$\lambda_0^2 = \frac{\omega_0^2}{1 + \omega_0^2 \sum_{k=1}^{N} \frac{\bar{c}_k^2}{(\omega_k^2 - \lambda_0^2)}} = \omega_0^2 \left[\frac{1}{M} - \frac{\tilde{\gamma}(s)}{s} \right] \tag{6.62}$$

$$\Lambda_k^2 = \frac{\omega_b^2}{1+\omega_0^2 \sum_{k=1}^{N} \frac{\bar{c}_k^2}{(\omega_k^2 - \Lambda_k^2)}} \tag{6.63}$$

Now we calculate the escape rate of a particle trapped in a potential well due to momentum coupling. We assume that the particle is equilibrated in the spin-1/2 bath or harmonic bath described by N *fictitious* oscillators. Thus the unstable mode of the inverted oscillator (for the particle) is thermalized according to a distribution of (N + 1) independent normal modes, given by

$$f_{eq}^{F,B} = Z_{F,B}^{-1} \exp\left[-\frac{\frac{1}{2}\lambda_b^2 \dot{\rho}^2 + V^{\#} - \frac{1}{2}\rho^2}{\hbar\lambda_0 \left(1 \mp 2\bar{n}(\lambda_0)\right)} \right]$$

$$\prod_{k=1}^{N} \exp\left[-\frac{\frac{1}{2}\lambda_k^2 \dot{x}_k^2 + \frac{1}{2}x_k^2}{\hbar\lambda_k \left(1 \mp 2\bar{n}(\lambda_k)\right)} \right] \tag{6.64}$$

where, $Z_{F,B}^{-1}$ are the respective normalization constants. Similarly for the stable normal modes for (N + 1) oscillators we may write,

$$f_{eq}^{F,B} = Z_{F,B}^{-1} \exp\left[-\frac{\frac{1}{2}\lambda_0^2 \dot{\rho}'^2 + \frac{1}{2}\rho'^2}{\hbar\lambda_0 \left(1 \mp 2\bar{n}(\lambda_0)\right)} \right]$$

$$\prod_{k=1}^{N} \exp\left[-\frac{\frac{1}{2}\Lambda_k^2 \dot{x}_k'^2 + \frac{1}{2}x_k'^2}{\hbar\Lambda_k \left(1 \mp 2\bar{n}(\Lambda_k)\right)} \right] \tag{6.65}$$

Normalizing the distributions and with $\chi^{F,B}$ defined as in Eq. (6.23) we have total energy of the unstable mode as

$$E = \frac{1}{2}\lambda_b^2 \dot{\rho}^2 + V^{\#} - \frac{1}{2}\rho^2 \tag{6.66}$$

Thus by going over to the energy space from $\rho, \dot{\rho}$, the equilibrium distribution functions are given by,

$$P_{eq}^{F,B}(E) = \frac{\omega_0}{2\pi\hbar\lambda_0 \left(1 \mp 2\bar{n}(\lambda_0)\right)} \frac{\lambda_b}{\omega_b} \chi^{F,B}$$

$$\exp\left[-\frac{E}{\hbar\lambda_0 \left(1 \mp 2\bar{n}(\lambda_0)\right)} \right] \tag{6.67}$$

The rate of barrier crossing is then given by

$$\Gamma^{F,B} = \frac{\omega_0}{2\pi} \frac{\lambda_b}{\omega_b} \chi^{F,B} \exp\left[-\frac{V^{\#}}{\hbar\lambda_0 \left(1 \mp 2\bar{n}(\lambda_0)\right)} \right] \tag{6.68}$$

where,

$$\chi_{spin} = \frac{\omega_b \lambda_0}{\omega_0 \lambda_b} \prod_{k=1}^{N} \frac{\tanh\left(\frac{\hbar\lambda_k}{2KT}\right)}{\tanh\left(\frac{\hbar\Lambda_k}{2KT}\right)} = \chi^F \tag{6.69}$$

and

$$\chi_{bosonic} = \frac{\omega_b \lambda_0}{\omega_0 \lambda_b} \prod_{k=1}^{N} \frac{\coth\left(\frac{\hbar\lambda_k}{2KT}\right)}{\coth\left(\frac{\hbar\Lambda_k}{2KT}\right)} = \chi^B \tag{6.70}$$

Eqs. (6.73)-(6.75) look similar in form as obtained for spatial coupling case. However, their explicit analytical expressions differ quite appreaciably leading to completely different situations. We examine the consequences in the following two limits as done in the spatial coupling case.

(i) Tunneling at 0K

When $T \sim 0$, i.e., $\frac{1}{KT} \to \infty$, both hyperbolic tangent and cotangent factors become unity. This results in χ to be $\chi = \frac{\omega_b \lambda_0}{\omega_0 \lambda_b}$. Therefore the decay rate for both kind of bath becomes,

$$\Gamma_{at\,0K} = \frac{\omega_0}{2\pi}\left(\frac{\lambda_b}{\omega_b}\right)\left(\frac{\omega_b \lambda_0}{\omega_0 \lambda_b}\right)\exp\left(-\frac{V^{\#}}{\hbar \lambda_0}\right) \quad (6.71)$$

As we have noted earlier that the main effect of the bath is to modify the bare frequencies. By virtue of Eq. (6.62), it follows that dissipation in momentum coupling increases λ_0 compared to ω_0 instead of decreasing it, as found in case of spatial coupling. To make this point clear, we consider a specific form of a momentum spectral density $J_p(\omega)$ with a cut off ω_c as;

$$J_p(\omega) = \frac{\pi}{2}\gamma\omega^3\theta(\omega_c - \omega) \quad (6.72)$$

where, $\theta(x)$ is the Heaviside function. One then finds from Eq. (6.59) that the Laplace transform of the momentum function is given by

$$\tilde{\gamma}_p(s) = \gamma s\left[\omega_c - s\tan^{-1}\left(\frac{\omega_c}{s}\right)\right] \quad (6.73)$$

and the effective mass M as

$$M = (1+\gamma\omega_c)^{-1} \quad (6.74)$$

Eq. (6.74) implies that in the limit $\omega_c \to \infty$, the effective mass goes to zero, i.e., a clear indication of stochastic acceleration (Sturrock, 1966). Now, substituting Eqs. (6.73)-(6.74) in Eq. (6.62) the stable normal mode frequency for the system mode reduces to

$$\lambda_0 = \omega_0(1+\gamma\omega_c)^{1/2} \quad (6.75)$$

Eq. (6.75) suggests that same thing happens also for the normal mode λ_b at the barrier top indicating that the momentum coupling leads to a thinning of the barrier, compared to the usual broadening of the barrier for spatial coupling (Pollak, 1986; Levine *et al*., 1988). Momentum coupling therefore causes an enhancement of the tunneling rate. By choosing a cubic potential of the form $V(q) = \frac{a}{2}q^2 - \frac{b}{3}q^3$, the barrier height $V^{\#}$ is given by $V^{\#} = \frac{2}{27}\omega_b^2(\Delta q)^2$ where Δq refer to the as usual tunneling length under harmonic barrier and also $\omega_b = \omega_0$. The decay rate is then given by

$$\Gamma_{at\,0K} = \frac{\lambda_0}{2\pi}\exp\left[-\frac{A\omega_0(\Delta q)^2}{\hbar}\left(1-\frac{\gamma\omega_c}{2}\right)\right]$$
$$= \frac{\lambda_0}{2\pi}\exp\left[-\frac{A\omega_0(\Delta q)^2}{\hbar}\right]\exp\left[\frac{A\omega_0(\Delta q)^2\gamma\omega_c}{2\hbar}\right]$$
$$(6.76)$$

where, $A = \frac{2}{27}$. Eq. (6.76) reveals that because of dissipation the tunneling rate at 0K is multiplied by a factor $\exp\left(\frac{A\omega_0(\Delta q)^2\gamma\omega_c}{2}\right)$. Tunneling at 0*K* is therefore exponentially enhanced by dissipation in the case of momentum coupling as compared to spatial coupling where we find the decay rate exponentially damped due to dissipation (Caldeira and Leggett, 1981).

(ii) At Finite Temperature

At higher temperatures the leading order contribution to the decay rate due to quantum transmission and reflection at the finite barrier is governed by the χ factor. Therefore we consider again the two cases separately;

1. Spin-1/2 Bath

The decay rate at finite temperature for spin-1/2 bath can be calculated similarly [Eqs. (6.30)-(6.35)] as

$$\Gamma_{T\,high} = \left(\frac{\omega_0}{2\pi}\right)\left(\frac{\lambda_b}{\omega_b}\right)$$

$$\left\{\left(\frac{\omega_b\lambda_0}{\omega_0\lambda_b}\right)^2\left[1+\frac{\hbar^2\left(\omega_0^2+\omega_b^2-2\lambda_0^2\right)}{12\left(KT\right)^2}\right]\right\}$$

$$\times\exp\left[-\frac{V^{\#}}{(\hbar\lambda_0)^2/2KT}\right] \tag{6.77}$$

For cubic potential of the form chosen earlier, we have to set $\omega_0 = \omega_b$. This reduces Eq. (6.82) to a simple form after a little bit of rearrangement as

$$\Gamma_{T\,high} = \frac{\lambda_0}{2\pi}\left(\frac{\lambda_0}{\lambda_b}\right)\exp\left[-\frac{\gamma\omega_c\hbar^2\omega_0^2}{6(KT)^2}\right]$$

$$\exp\left[-\frac{V^{\#}}{(\hbar\lambda_0)^2/2KT}\right] \tag{6.78}$$

where, we have used the relation Eq. (6.75). For momentum coupling χ_{spin} is therefore exponentially damped at finite temperature. Eq. (6.83) is the high temperature decay rate for the spin-1/2 bath. An hallmark of the above expression is that the χ factor increases with increase of temperature and also the thermal activation term contains an explicit quantum contribution ~ $O(\hbar^2)$. The Arrhenius term decreases with increase of temperature which arises due to thermal saturation of the bath. Unlike classical thermal activation, spin-1/2 bath contains explicit quantum character at high temperature.

2. Bosonic Bath

Similarly, from Eq. (6.75) $\chi_{bosonic}$ can be expressed as

$$\chi_{bosonic} = \left[1-\gamma\omega_c\frac{\hbar^2\left(\omega_0^2+\omega_b^2\right)}{24(KT)^2}\right] \tag{6.79}$$

The decay rate for bosonic bath at high temperature therefore takes the following form:

$$\Gamma_{T\,high} = \frac{\omega_0}{2\pi}\left(\frac{\lambda_b}{\omega_b}\right)\exp\left[-\gamma\omega_c\frac{\hbar^2\left(\omega_0^2+\omega_b^2\right)}{24(KT)^2}\right]$$

$$\exp\left[-\frac{V^{\#}}{KT}\right] \tag{6.80}$$

The tunneling rate is found to be exponentially damped at finite temperature. As the temperature is lowered this factor decreases rapidly. Since the harmonic bath approaches the classical limit at high temperature, thermal activation factor reduces to usual Arrhenius form.

Application; Experimental Realization of Spin Bath

We begin this section with a note that our theoretical findings are applicable to reservoir of independent spins interacting with a properly chosen system coordinate. Because of small size and large surface-to-volume ratio nano-electro-mechanical devices are good candidates for studying noise and dissipation in the regions ranging from classical to quantum domain. A nano-mechanical resonator e.g., a cantilever of nano-scale dimension which can vibrate at GHz frequencies (Craighead, 2000) may serve as an interesting device. The low frequency oscillation which arises from the anharmonic excitation of impurities and disorder on the amorphous surface of the cantilever itself may be viewed (Liu *et al*., 1999) as arising from two-level systems corresponding to two lowest energy eigen states. The dissipation of oscillation at various temperatures can be studied as possible application in these nano-mechanical devices and it is expected to reveal the differences from a bosonic bath. Another area of application is the spin dynamics of an electron in semi conducting nano structures. A typical example is an electron spin in an isolated GaAs semiconductor quantum dot in which each nucleus carries a spin, interacts through hyperfine interaction. Such investigations leads to understanding of spin decoherence (Khaetskii *et al.*, 2002). Since quantum dots serve as 'artificial' two-level atoms in many situations and the varying system size results in a distribution of frequencies, a reservoir of two-level quantum dots can be used for studying quantum dissipation in a spin bath at low temperature. When the number density of dots, say, is of the order of 10^5cm^{-3} as often found in experiments, the system experiences practically a Fermi sea with infinite degrees of freedom. Finally, we mention the thermally induced coherence as manifested in several photo-absorption emission experiments in quantum dots (Bras *et al.*, 2002) can be modeled to revalidate our formulation. Ideally the typical system-spin bath model for dissipation we have in mind for the demonstration of temperature assisted coherence in quantum dots is a two-level quantum dot kept in a suitably designed environment of two-level quantum dots with characteristic size distribution. The important findings include decrease of integrated absorption coefficient with temperature (Ghosh *et al*, 2011a), anomalous narrowing of line width (Ghosh *et al.*, 2011a; Ghosh *et al.*, 2011b) and formation of thermally induced Mollow triplet in presence of weak light field (Ghosh *et al.*, 2011b). The efficacy of the present scheme can also be ascertained from the recovery of Caldeira-Leggett's celebrated result of exponential suppression of tunneling by dissipation at 0K (Caldeira and Leggett, 1981) as discussed in the last section.

Temperature Assisted Coherence in Quantum Dots; An Experimental Verification

We consider the following Hamiltonian for a two-level system in a spin-1/2 bath;

$$\hat{H} = \frac{1}{2}\hbar\Omega\hat{\sigma}_z + \frac{1}{2}\sum_k \hbar\omega_k\hat{\sigma}_{zk} + \hbar\sum_k (g_k\hat{\sigma}_k^\dagger\hat{\sigma} + g_k^*\hat{\sigma}^\dagger\hat{\sigma}_k) \tag{7.1}$$

Hamiltonian for the system is specified by the Pauli operators $\hat{\sigma}^\dagger$, $\hat{\sigma}$ and $\hat{\sigma}_z$. The Pauli operators for the system follow the usual commutation relations as follows;

$$\begin{gathered}[\hat{\sigma}^\dagger,\hat{\sigma}] = \hat{\sigma}_z;[\hat{\sigma}^\dagger,\hat{\sigma}_z] = -2\hat{\sigma}^\dagger;[\hat{\sigma},\hat{\sigma}_z] = 2\hat{\sigma} \\ \hat{\sigma}^\dagger\hat{\sigma} = \frac{1}{2}(1+\hat{\sigma}_z);\hat{\sigma}\hat{\sigma}^\dagger = \frac{1}{2}(1-\hat{\sigma}_z)\end{gathered} \tag{7.2}$$

Similar relations also hold good for bath operators. Particularly, using the commutation and anti-commutation rules obeyed the spin-1/2 bath operators we have

$$\begin{gathered}\hat{\sigma}_k^\dagger\hat{\sigma}_k - \hat{\sigma}_k\hat{\sigma}_k^\dagger = \hat{\sigma}_{zk} \\ \hat{\sigma}_k^\dagger\hat{\sigma}_k + \hat{\sigma}_k\hat{\sigma}_k^\dagger = 1\end{gathered} \tag{7.3}$$

So, $\hat{\sigma}_{zk} = 2\hat{n}_k - 1$ where $\hat{n}_k$ is the number operator $\hat{\sigma}_k^\dagger\hat{\sigma}_k$. Proceeding as in Ref (Ghosh *et al.*, 2011a) we arrive at the Bloch equations for dissipative dynamics of a two-level atom in a spin-1/2 bath as follows;

$$\dot{U} = -\frac{1}{T_2}U \tag{7.4}$$

$$\dot{V} = -\frac{1}{T_2}V \tag{7.5}$$

$$\dot{W} = -\left(\frac{W - W_0}{T_1}\right) \tag{7.6}$$

where U, V and W are the Bloch vector components (Ghosh *et al.*, 2011a) and we have

$$\frac{1}{T_2} = \gamma_0 \tanh\left(\frac{\hbar\Omega}{2KT}\right) \tag{7.7}$$

and

$$\frac{1}{T_1} = 2\gamma_0 \tanh\left(\frac{\hbar\Omega}{2KT}\right) \tag{7.8}$$

The steady state value of W is W_0 given by

$W(t=\infty)=W_0=-\frac{1}{2}$. A characteristic feature ofthe spin-spin (T_2) and spin-lattice relaxation (T_1) times for the spin-1/2 bath is their temperature dependence. The origin of this temperature dependence is due to the nonlinear nature of the two-level bath atoms in contrast to harmonic bath as pointed out in Sec. IVC.

To gain further insight into the nature of the spin-1/2 bath for the damped two-level system, let us now specialize to the case of the two level atoms is subjected to optical excitation. The dynamics is modified to include the energy of interaction between the system and the field. One arrives at the following optical Bloch equations:

$$\dot{U}=-\frac{U}{T_2}-\delta V \tag{7.9}$$

$$\dot{V}=-\frac{V}{T_2}+\delta U+\mathcal{R}_0 W \tag{7.10}$$

$$\dot{W}=-\frac{1}{T_1}\left(W+\frac{1}{2}\right)-\mathcal{R}_0 W \tag{7.11}$$

Here $\delta(=\Omega-\omega)$ denotes the detuning frequency and R_0 refers to the Rabi flopping frequency $d\mathcal{E}/\hbar$. $\mathcal{E}$ is to the slowly varying amplitude of the classical radiation field with frequency and is the dipole moment matrix element between the two states of the system. The above description allows us to calculate the steady state absorption coefficient (α). We first set $\dot{U}$ = $\dot{V}$ = $\dot{W}$ = 0 and find the expressions for the steady state values of Bloch vector components U_s, V_s and W_s as

$$U_s=-\frac{\delta\mathcal{R}_0 T_2^2}{2(1+I+\delta^2T_2^2)} \tag{7.12}$$

$$V_s=-\frac{\mathcal{R}_0 T_2}{2(1+I+\delta^2T_2^2)} \tag{7.13}$$

$$W_s=-\frac{1}{2}\left(\frac{1}{1+I\mathcal{L}(\delta)}\right) \tag{7.14}$$

Here I stands for the dimensionless intensity $(I=\mathcal{R}_0^2T_1T_2)$ and $\mathcal{L}(\delta)$ is the dimensionless Lorentzian of the form $\mathcal{L}(\delta)=\frac{1}{1+\delta^2T_2^2}$. The steady state polarization P of N number of two-level atoms per unit volume can then be linked with the steady state Bloch components (Meystre and Sargent, 1991) as

$$\mathcal{P}=Nd(U_s-iV_s) \tag{7.15}$$

Furthermore, use of Eqs. (7.12)-(7.13) in Eq. (7.15) results in the expression

$$\mathcal{P}=-\frac{i}{2}\left(\frac{d^2}{\hbar}\right)\left(\frac{N\mathcal{D}(\delta)}{1+I\mathcal{L}(\delta)}\right)\mathcal{E} \tag{7.16}$$

with $\mathcal{D}(\delta)=\dfrac{1}{\gamma_0\tanh\left(\frac{\hbar\Omega}{2KT}\right)+i\delta}$. Comparing Eq. (7.16) with Maxwell equation relating polarization P and electric field amplitude E (Meystre and Sargent, 1991), i.e.,

$$\mathcal{P}=i\left(\frac{2}{\nu}\right)\alpha\mathcal{E} \tag{7.17}$$

where, ϵ is the permittivity of the medium and ν is the wave number, we have the expression for complex susceptibility as

$$\alpha=\alpha_0\frac{\gamma_0\tanh\left(\frac{\hbar\Omega}{2KT}\right)\mathcal{D}(\delta)}{1+I\mathcal{L}(\delta)} \tag{7.18}$$

where, the linear (I = 0), resonant $(\delta=0)$ susceptibility α_0 is given by

$$\alpha_0 = -\kappa \frac{d^2 N}{4\epsilon\hbar\gamma_0 \tanh\left(\frac{\hbar\,\Omega}{2KT}\right)} \tag{7.19}$$

which is positive for an uninverted medium ($N < 0$). The nonlinear absorption coefficient is then given by

$$Re(\alpha) = \alpha_0 \frac{\left[\gamma_0 \tanh\left(\frac{\hbar\,\Omega}{2KT}\right)\right]^2}{\left[\gamma_0 \tanh\left(\frac{\hbar\,\Omega}{2KT}\right)\right]^2 (1+I) + \delta^2} \tag{7.20}$$

We note this expression for absorption coefficient is Lorentzian; both the peak and the width are dependent on temperature. Ideally the peak of the Lorentzian at a given incident power corresponds to a zero-phon online since the bath considered here is devoid of any contamination of phonon modes. Taking care of these modes results in a simple modification of the line width $\left[\gamma_0 \tanh\left(\frac{\hbar\,\Omega}{2KT}\right)\right]$, by $\left[\gamma_{ph} + \gamma_0 \tanh\left(\frac{\hbar\,\Omega}{2KT}\right)\right]$ where γ_{ph} is the additional dissipation constant due to phonon modes. The line width varies with incident power, (I) as, $(1+I)^{1/2}$ where I can be written as a product of temperature-dependent term $\left[\tanh\left(\frac{\hbar\,\Omega}{2KT}\right)\right]^{-2}$ and $I_0 = \frac{1}{2}\frac{\mathcal{R}_0^2}{\gamma_0^2}$. It is convenient further to express Eq. (7.20) into the following form

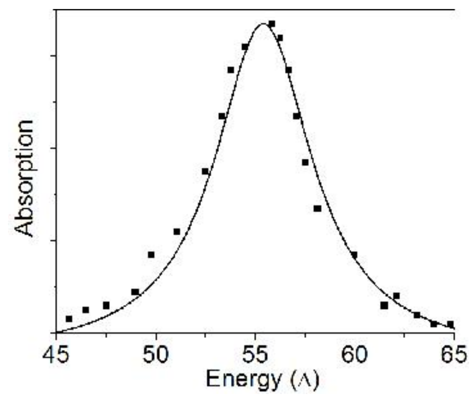
$$Re(\alpha) = c\frac{\tanh\left(\frac{\hbar\Omega}{2KT}\right)}{\left[\tanh\left(\frac{\hbar\Omega}{2KT}\right)\right]^2 \left[1 + I_0\left[\tanh\left(\frac{\hbar\Omega}{2KT}\right)\right]^{-2}\right] + \Delta^2} \tag{7.21}$$

Here c is a temperature independent constant that can be expressed in terms of quantities in Eq. (7.34). $\Delta^2 = \frac{\delta^2}{\gamma_0^2}$ is the dimensionless detuning parameter and I_0 is the dimensionless incident intensity or Rabi frequency. These two parameters and the dimensionless ratio $\frac{\hbar\Omega}{KT}$ are three basic quantities to correlate theoretical finding with experimental observations.

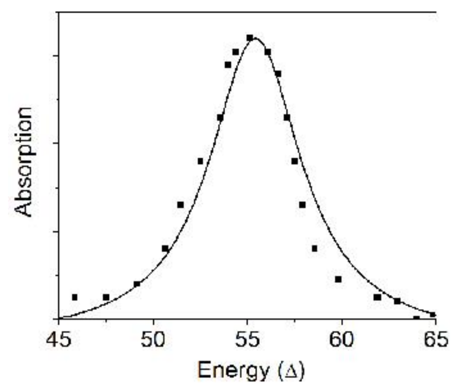
The theoretical results can be correlated with experiments, particularly on photo-absorption and emission experiments with single quantum dots to examine the relaxation processes. We focus on situations at low temperature where the quantum dot optical spectrum is mainly determined by zero-phonon line and the environment of two-level artificial atoms with characteristic distribution of frequencies can serve as a reservoir to induce relaxation effect. In what follows we consider particularly three aspects; 1. Anomalous temperature dependence of absorption coefficient; 2. Incident power dependence of absorption coefficient and 3. Narrowing of line width with temperature.

Temperature Dependence of Absorption Coefficient

The optical transitions or inter-sublevel transitions between conduction confined states or valance confined states of quantum dots can be observed experimentally by charge modulation spectroscopy, photo-induced spectroscopy or by direct spectroscopy. We consider the results reported by Bras *et al.* (Bras *et al.*, 2002) on inter-sublevel absorption as a function of temperature in n-doped InAs/GaAs self-assembled quantum dots for transition between s-type ground state to first p-type excited states. Two such typical absorption curves are shown (black squares) in Fig. 7.1 (a) and (b) at two different temperatures (15K and 60K). Also a decrease by a factor 4 of the integrated absorption is observed from low temperature to room temperature. This is illustrated in Fig. 7.2. The onset of the decrease occurs ~ 70K. In order to correlate the above observations with nonlinear absorption coefficient we make use of the expression [Eq. (7.21)] for $Re(\alpha)$. We fix c = 1.008 for the entire calculation. The parameters used are I_0 = 98.0 and $\hbar\Omega$ = 55.4 meV. The solid lines in Fig. 7.1 (a) and (b) represent the theoretical curves. It is evident that the theory agrees with experimental data at very low temperature. With increase of temperature the experimental absorption profile progressively deviates from the expected Lorentzian profile with the appearance of low temperature acoustic phonon side bands. As the origin of the width of the profile at higher temperature is uncertain due to various factors like, dislocations, defect etc., in addition to the factor due to spin-1/2 bath, we have used the experimental width for simulating the absorption profile for computing integrated absorption coefficient shown in Fig. 7.2 (solid line). The anomalous decrease of integrated absorption intensity with increase of temperature has been reported by several groups (Onishchenko, 2005) in other quantum dot systems in the recent past.



(a)



(b)

Fig. 7.1: A comparison of inter-sublevel absorption profiles at (a) *T* = 15*K* and (b) *T* = 60*K* between theory (solid line) and experiment (Bras *et al.*, 2002) (black squares) for dimensionless incident intensity I_0 and $\hbar\Omega$ = 55.4 meV. [Reproduced with permission from "A. Ghosh, S. S. Sinha and D. S. Ray, Langevin–Bloch equations for a spin bath, J. Chem. Phys. 134, 094114 (2011)". Copyright 2011 AIP Publishing LLC]

Variation of Line-width with Incident Power

A typical experimental variation of line-width with incident power is shown by black squares in Fig. 7.3 for a single quantum dot system at 20K (Favero, 2007). The expression for absorption coefficient [Eq. (7.21)] shows that the line-width varies with incident power as $(1+I)^{1/2}$ for a given temperature. This is depicted by a continuous line in Fig. 7.3 for $T = 20K$ and $\hbar\Omega = 1.96$ eV. The theoretical curve corroborates closely to the experimental data.

Narrowing of Line-width with Temperature

A characteristic feature of the Bloch equations for spin-1/2 bath is the temperature dependence of relaxation times T_1 and T_2. This is reflected in the expression for line-width in Eq. (7.21). It is apparent that for a given incident power (I_0) line width decreases with increase in temperature. The variation of line-width with temperature is shown in Fig. 7.4 for several values of incident power. Due to the limited accessibility of the states of a spin-1/2bath as compared to availability of infinite number of states of a harmonic bath. Therefore temperature favours coherence or leads to suppression of line-width. It may also be mentioned that the narrowing has been observed when the incident power is low so that thermal effect comes into play. The effect is likely to disappear at higher incident power due to power broadening.

Thermally Induced Spectral Splitting in Mollow Triplet

Next we consider the system driven by a classical electromagnetic field $v(t)$ so that the total

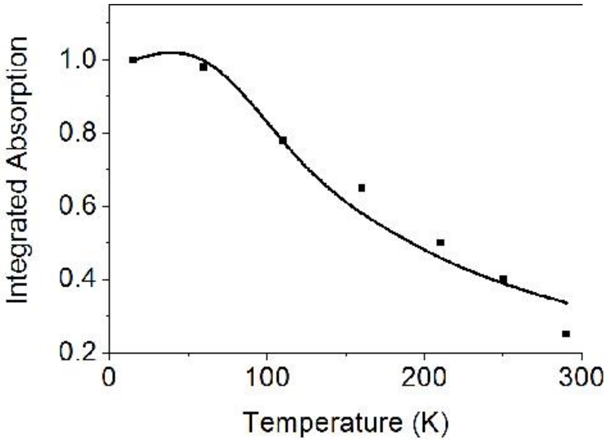


Fig. 7.2: **A comparison between theory (solid line) and experiment (Bras *et al*., 2002) (black squares) of normalized integrated absorption as a function of temperature for dimensionless incident intensity I_0 and $\hbar\Omega = 55.4$ and meV. [Reproduced with permission from "A. Ghosh, S. S. Sinha and D. S. Ray, Langevin–Bloch equations for a spin bath, J. Chem. Phys. 134, 094114 (2011)". Copyright 2011 AIP Publishing LLC]**

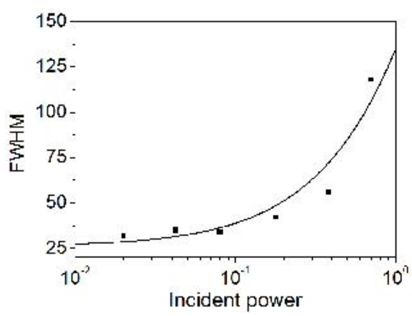


Fig. 7.3: **Variation of dimensionless line width (FWHM) with dimensionless incident intensity I_0 at 20K (theory-solid line; experiment (Favero *et al*., 2007)—black squares) for $\hbar\Omega = 1.96$ eV. [Reproduced with permission from "A. Ghosh, S. S. Sinha and D. S. Ray, Langevin-Bloch equations for a spin bath, J. Chem. Phys. 134, 094114 (2011)". Copyright 2011 AIP Publishing LLC]**

Hamiltonian can be expressed as

$$\hat{H} = \frac{1}{2}\hbar\Omega\hat{\sigma}_z + \frac{1}{2}\sum_k \hbar\omega_k \hat{\sigma}_{zk}$$

$$+ \hbar\sum_k (g_k \hat{\sigma}_k^\dagger \hat{\sigma} + g_k^* \hat{\sigma}^\dagger \hat{\sigma}_k)$$

$$+ \hbar\left(v(t)\hat{\sigma}^\dagger + v^*(t)\hat{\sigma}\right) \tag{7.22}$$

The last term refers to the interaction between

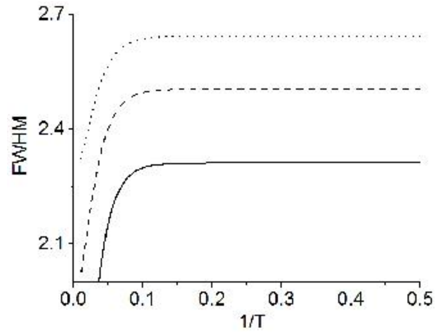


Fig. 7.4: **Variation of dimensionless line width (FWHM) with inverse of temperature for three different values of dimensionless incident intensity I_0 = 0.125 (solid line); (dashed line); (doted line) for $\hbar\Omega = 5.0$ meV. [Reproduced with permission from "A. Ghosh, S. S. Sinha and D. S. Ray, Langevin-Bloch equations for a spin bath, J. Chem. Phys. 134, 094114 (2011)". Copyright 2011 AIP Publishing LLC]**

the system and the classical field $v(t)$ which is given by $v(t) = V_2 \exp[-i\omega_0 t]$, $V_2 = {d\mathcal{E}}/{2\hbar}$. d is the transition dipole moment and E is the complex classical field amplitude. ω_0 is the frequency of the field. The Hamiltonian [Eq. (7.22)] is based on rotating wave approximation. As in other physically relevant situations in quantum optics it is sufficient to work within weak coupling (Born approximation) for the two-level atom and the bath. However, the coupling between the two-level system and the driving field is arbitrary. The working range of intensity is such that the Rabi frequency is either larger than the damping rate for coherent regime or much less than it for the incoherent regime. Proceeding as in Ref (Ghosh *et al*., 2011b) we calculate the resonance fluorescence spectrum and its intensity from driven atom in a spin-1/2 bath.

Scattered Intensity of Resonance Fluorescence

Since the far field emitted by a dipole is proportional to the dipole, the scattered intensity is proportional to the single-time first-order correlation function of the dipole i.e.,

$$I = \alpha\int_0^T dt\langle \hat{S}^\dagger(t)\hat{S}(t)\rangle_{qs} = \alpha T\langle S_a\rangle_s \tag{7.23}$$

where, α is a proportionality constant. In deriving Eq. (7.23) we have also used the relation

$$\hat{S}^{\dagger}(t)\hat{S}(t) = |a\rangle\langle b||b\rangle\langle a| = |a\rangle\langle a| \equiv \hat{S}_a(t) \tag{7.24}$$

Here, a and b denote the upper and lower levels of the two-level system, respectively. The spectrum can therefore be calculated by recognizing the slowly varying operator nature of the system variables $\hat{S}^{\dagger}(t), \hat{S}_z(t)$ and $\hat{S}(t)$ such that $\hat{S}(t)$ = $\hat{\sigma}(t)\exp(i\Omega t)$, $\hat{S}^{\dagger}(t) = \hat{\sigma}^{\dagger}(t)\exp(-i\Omega t)$ and $\hat{S}_z(t) = \frac{\hat{\sigma}_z(t)}{2}$. We further denote $\delta\hat{S}^{\dagger}(t), \delta\hat{S}_z(t)$ and $\delta\hat{S}(t)$ as the corresponding fluctuation operators so that $\hat{S}^{\dagger}(t) = S^{*}(t) + \delta\hat{S}^{\dagger}(t)$ and so on. By construction, $\langle\delta\hat{S}^{\dagger}(t)\rangle = \langle\delta\hat{S}_z(t)\rangle$ = $\langle\delta\hat{S}(t)\rangle$ = 0 and $[\delta\hat{S}^{\dagger}(t), \delta\hat{S}(t)] = 2\delta\hat{S}_z(t)$; $\{\delta\hat{S}^{\dagger}(t), \delta\hat{S}(t)\}_+$ = 1. Equation (7.24) therefore yields

$$I = \alpha\int_0^T dt\,|\langle S^{*}(t)\rangle_s|^2 + \alpha\int_0^T dt \;\langle\langle\delta\hat{S}^{\dagger}(t)\delta\hat{S}(t)\rangle\rangle_s \tag{7.25}$$

or,

$$I \equiv I_{coh} + I_{inc} \tag{7.26}$$

The scattered intensity consists of two terms. The first one originates from the mean motion of the dipole $\left(\langle S^{*}\rangle_s\right)$ driven by the applied field, i.e., the coherently scattered intensity I_{coh} while the incoherent contribution I_{inc} is due to the fluctuations of the dipole motion. Comparing Eq. (7.23) and Eq. (7.25), we find that the incoherent intensity is given by

$$I_{inc} = \alpha T(\langle S_a\rangle_s - |\langle S^{*}\rangle_s|^2) \tag{7.27}$$

Using the steady state values we have

$$I_{coh} = \alpha T\,|\langle S^{*}\rangle_s|^2 = \alpha T\frac{\frac{1}{2}I_2\mathcal{L}_2\Gamma_0}{2\gamma_0(1+I_2\mathcal{L}_2)^2} \tag{7.28}$$

$$I_{inc} = \alpha T\frac{\frac{1}{2}I_2\mathcal{L}_2}{1+I_2\mathcal{L}_2} - I_{coh} \tag{7.29}$$

Here, the dimensionless incident intensity is I_2 and L_2 is a Lorentzian in form. As usual, the coherently scattered intensity vanishes for the strong driving field $I_2 >> 1$. This is because the coherent part is proportional to the squared magnitude of the steady state dipole $\left[\;\langle S^{*}\rangle_s^2\right]$, which bleaches to zero in strong driving field limit. On the other hand, the incoherent part is due to spontaneous emission from the upper level, whose probability of occupation is approximately ½ in strong field. In the weak field limit, the atom remains essentially in the ground state as no spontaneous emission takes place. These radiative properties of the two-level system closely correspond to the usual case of harmonic bath and are illustrated in Fig. 7.5 which shows the coherent and incoherent contributions to the scattered light intensity as a function of dimensionless incident intensity I_2. The signature of the spin-1/2 bath, however, is markedly manifested when the coherent and incoherent scattered intensities are plotted at different temperatures as shown in Fig. 7.5. In the weak field limit, I_{coh} at higher temperature (T = 100K; dotted line) is larger than that at lower temperature (T = 30K; continuous line). This essentially indicates that increasing temperature assists coherence. From Eqs. (7.28) and (7.29) it appears that by equating them one may obtain a transition temperature T_{tr} for lower value of I_2 as given by $I_2 = \left[\tanh\left(\frac{\hbar\Omega}{2KT_{tr}}\right)\right]^2$ for zero detuning ($\delta_0 = 0$). In Fig 7.6 T_{tr} is plotted against I_2 to

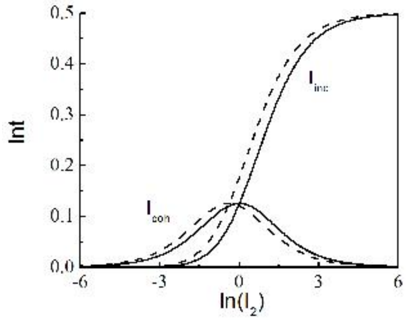


Fig.7.5: Variation of coherent and incoherent intensity with dimensionless incident intensity for $\hbar\Omega = 20$ meV at two different temperatures T = 30K (continuous line) and T = 100K (dotted line) with zero detuning ($\delta = 0$). [Adapted from "A. Ghosh, S. S. Sinha and D. S. Ray, Dissipation in a spin bath; Thermally induced coherent intensity and spectral splitting, Phys. Rev. E 134, 094114 (2011)". Copyright 2011 by the American Physical Society]

illustrate the dividing line between coherent and incoherent regime. It therefore, follows that by tuning the temperature of the spin-1/2 bath it is possible to realize a cross over between coherent and incoherent regime at a particular strength of the driving field. This result is a further confirmation of an earlier observation that spin-spin-bath model exhibits distinctly different physics at non-zero temperature as compared to spin-boson model. Higher field masks the coherence effect due to power broadening.

Calculation of Resonance Fluorescence Spectrum; Thermally Induced Splitting

For an ergodic and stationary process, the spectrum $\mathbb{S}(\varpi)$ (Louisell, 1973) can be expressed following Wiener-Khintchine theorem as

$$\mathbb{S}(\varpi) \propto \int_0^\infty d\tau \langle E^-(\tau)E^+(0)\rangle \exp(-i\varpi\tau)+c.c \tag{7.64}$$

Then using the slowly varying operators $\hat{S}(t)$ and $\hat{S}^\dagger(t)$ as before one may rewrite Eq. (7.64) in the following form:

$$\mathbb{S}(\varpi)=\beta\int_0^\infty d\tau \langle \hat{S}^\dagger(\tau)\hat{S}(0)\rangle_{qs}$$

$$\exp[i(\omega_0-\varpi)\tau]+c.c \tag{7.65}$$

where, β is a proportionality constant. We may derive the light spectrum into coherent and incoherent parts as in the case of scattered intensity;

$$\mathbb{S}(\varpi) \equiv \mathbb{S}_{coh}(\varpi)+\mathbb{S}_{inc}(\varpi)$$

$$=\beta|\langle S^*(t)\rangle_s|^2\int_0^\infty d\tau \exp[i(\omega_0-\varpi)\tau]$$

$$+\beta\int_0^\infty d\tau \langle\langle \delta\hat{S}^\dagger(\tau)\delta\hat{S}(0)\rangle\rangle_s$$

$$\exp[i(\omega_0-\varpi)\tau]+c.c \tag{7.66}$$

The coherent part, i.e., the Rayleigh peak can readily found to be

$$\mathbb{S}_{coh}(\varpi)=2\pi\beta|\langle S^*\rangle_s|^2\delta(\omega_0-\varpi)$$

$$=\frac{\frac{1}{2}\pi\beta I_2\mathcal{L}_2\Gamma_0}{\gamma_0(1+I_2\mathcal{L}_2)^2}\delta(\omega_0-\varpi) \tag{7.67}$$

It consists simply of a delta-function centered at ω_0 ($=\Omega$ for $\delta_0=0$). To evaluate the incoherent part, we need to estimate the two-time correlation functions of the atomic fluctuations operators, which

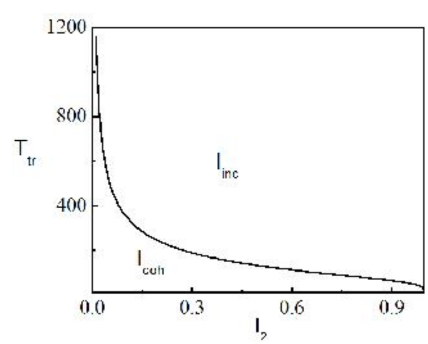


Fig. 7.6: Variation of T_{tr} forsmaller value of dimensionless incident intensity I_2 with $\hbar\Omega = 20$ meV for zero detuning $(\delta = 0)$. [Adapted from "A. Ghosh, S. S. Sinha and D. S. Ray, Dissipation in a spin bath; Thermally induced coherent intensity and spectral splitting, Phys. Rev. E 134, 094114 (2011)". Copyright 2011, American Physical Society]

can be calculated from the quantum correction equations (Ghosh *et al.*, 2011b). Explicit evaluation of the above for central pump detuning $(\delta_0 = 0)$ yields the three-peaked in coherent spectrum with $\Delta = (\omega_0 - \bar{\omega})$ as;

$$\mathbb{S}(\Delta)|_{\text{inc}} \simeq \frac{\beta}{2\gamma_0 \tanh\left(\frac{\hbar\Omega}{2\text{KT}}\right)} \left[\frac{\gamma_0^2\left[\tanh\left(\frac{\hbar\Omega}{2\text{KT}}\right)\right]^2}{\gamma_0^2\left[\tanh\left(\frac{\hbar\Omega}{2\text{KT}}\right)\right]^2 + \Delta^2}\right]$$
$$+\frac{\beta}{2\gamma_0 \tanh\left(\frac{\hbar\Omega}{2KT}\right)} \left[\frac{\gamma_0}{\Gamma_0+\gamma_0}\frac{\frac{1}{4}(\Gamma_0+\gamma_0)^2\left[\tanh\left(\frac{\hbar\Omega}{2KT}\right)\right]^2}{(\mathcal{R}_0 \pm \Delta)^2 + \frac{1}{4}(\Gamma_0+\gamma_0)^2\left[\tanh\left(\frac{\hbar\Omega}{2KT}\right)\right]^2}\right] \tag{7.73}$$

The incoherent part of the scattered spectrum consists of three peaks, as expected from the dressed atom picture. The central peak at $\bar{\omega} = \Omega$, has a width $\gamma_0 \tanh\left(\frac{\hbar\Omega}{2KT}\right)$, while the side bands centered about the frequency $\bar{\omega} = \Omega \pm \mathcal{R}_0$, have a width $\frac{1}{2}(\Gamma_0+\gamma_0)\tanh\left(\frac{\hbar\Omega}{2KT}\right)$. $\mathcal{R}_0$ is the Rabi flopping frequency defined by $\frac{d\mathcal{E}}{\hbar}$. Though at zero Kelvin the situations are completely identical for both bosonic and spin-1/2 bath, we obtain an interesting result for spin-1/2 bath at nonzero finite temperature which largely deviates from its bosonic counterpart. The substantial reduction of linewidth with rise of temperature in quantum dot two-level systems as reported in a number of photo-luminescence experiments by several groups lends support to this observation (Bras *et al.*, 2002; Onishchenko *et al.*, 2005). Fig. 7.7 shows that at low field i.e., for small Rabi frequency R_0, one may obtain three-peak Mollow triplet by tuning temperature to a higher value. This result demonstrates that with increase in temperature the single peak splits into three in a low driving field regime, because of emergence of coherence behaviour as a result of suppression of width at relatively higher temperature in a spin-1/2 bath. This result is a strong departure from what one observes in conventional quantum optics.

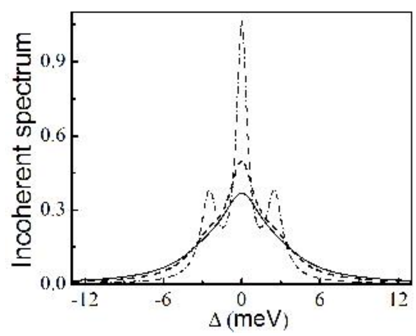


Fig. 7.7: Thermally-induced Mollow triplet:At low field the single peak splits into three by increase of temperature; T = 70K (continues line); T = 160K (dashed line); T = 460K (dashed-dot line) for I_2 and $\hbar\Omega = 20$ meV

Conclusions

In this review we have outlined a simple approach to quantum dissipation based on a universal realization of an environment in the form of a general spin bath. The basis of this approach is the spin coherent state representation of quantum noise operators and a thermal canonical distribution of the Gaussian form. We have shown it is possible to construct a stochastic differential equation in the form of a c-number Langevin equation with quantum correction terms which can be solved with sufficient degree of accuracy. Based on Langevin formalism we have derived the quantum analogues of Fokker-Planck, diffusion and Smoluchowski equations for both spin-

1/2 and harmonic bath and the formulation is applied to rate processes in chemical dynamics. Practical realization of spin-1/2 bath has been discussed in connection with absorption-emission experiment with quantum dots and in quantum optical context. We now summarize the main conclusions of the study:

(i) We have proposed a system-generalized spin bath Hamiltonian which can serve as a universal paradigm for description of interaction between an environment and a quantum particle. Two special limits of this general spin bath correspond to spin-1/2 bath and to bosonic bath. A mapping between spin operators and boson operators for a finite basis using Holstein-Primakoff transformation plays a key role in realization of the two limits within a unified description. The Hamiltonian reduces to Zwanzig Hamiltonian describing a particle interacting with a harmonic bath in the large spin limit. The reduced equationof motion for the particle assumes the form of a classical Langevin equation with quantum noise correlation.

(ii) Our emphasis has been to look for a natural extension of the classical theory of stochastic processes to quantum domain. Since the states of the general spin bath used are the spin coherent states, they facilitate the use of c-numbers for the description of the bath. The statistical character of the bath is reflected through the canonical thermal distributions of the c-numbers for bosons and fermions. For spin-1/2 bath this distribution is a fermionic counterpart of Wigner canonical thermal distribution and is a new content of the present analysis.

(iii) We have presented a scheme to generate quantum noise as 'classical' numbers which follow quantum fluctuation-dissipation relations. Hence, it is easy to comprehend that the classical numerical techniques for generation of noise can be used in a straightforward way to solve quantum Langevin equation under the present scheme. The procedure is easier to implement than path integral Monte Carlo techniques. The present scheme is equipped to deal with quantum dynamics for a wide range of noise correlation and temperature.

(iv) The probability distribution functions bear the true notion of probability rather than quasi-probability employed for describing Wigner, Glauber-Sudarshan or Q-functions. This enables one to explore these equations in full quantum domain, e.g., in the zero temperature tunneling. The behaviour of the diffusion constant and the mean square displacement of the particle in a spin-1/2 bath as a function of temperature have been found to differ significantly from their bosonic counterparts.The generic consequence due to restricted options for thermal excitations in a spin-1/2 bath is that the temperature favors suppression of diffusive motion, although quantization, in general, enhances mean-square displacement. We have shown that although quantization has a significant effect on the decay of the metastable state the rate constant is finite at 0K and exhibits the predominance of coherent behaviour at low but finite temperatures.

(v) We have considered the problem of quantum decay of a particle out of a metastable state which can interact with its environment either by spatial or momentum coupling. Both tunneling and thermal activation can be treated on equal footing by taking care of thermalization of the stable and unstable normal modes within an unified description by using the aforesaid canonical thermal distributions for spin-1/2 and bosonic bath. At 0K for both harmonic and spin-1/2 bath dissipation due to spatial and momentum coupling multiplies the tunneling probability by a factor $exp\left[\mp A\kappa_0\left(\Delta q\right)^2/\hbar\right]$, respectively, where $\left(\Delta q\right)^2$ is the tunneling length, κ_0 is the dissipation constant and the numerical constant A depends on the parameters of the potential. For spatial coupling this corresponds to the seminal result of Caldeira and Leggett (Caldeira and Leggett, 1981) and is a vindication of the fact that the spin-1/2 bath and the harmonic bath behave identically at 0K. We found that at low but finite temperatures both spin-1/2 and harmonic bath induces thermal activation and tunneling and the rate goes as $exp\left(B/T^2\right)$ for spatial coupling while exponential suppression of rate as $exp\left(-B/T^2\right)$ takes place for momentum coupling.

(vi) We have explored several practical examples of spin bath in connection with photo absorption-emission experiments with two-level single quantum dots (spin-1/2 bath). It has been found that since two-level bath behaves as a nonlinear system in contrast to a harmonic oscillator bath, the spin-spin and spin-lattice relaxation times behave differently from their bosonic counterparts. Because of limited options for excitation due to Pauli exclusion principle spin-1/2 bath assists temperature induced coherence. This is manifested in anomalous temperature dependence of integrated absorbance and variation of linewidth with power of excitation at a given temperature. An important offshoot is the prediction of anomalous narrowing of spectral linewidth with increase of temperature. We have further explored the role of spin-1/2 bath in the quantum optical context of an excited two-level system to demonstrate how Mollow triplet in resonance fluorescence can be realized as a thermally-induced process, rather than as a field-induced effect as observed in traditional quantum optics.

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