Deriving Generalized Born-Markov Optical Master Equation for Analysis of Inelastic Tunneling Under Non-Equilibrium Condition

Arash Tirandaz

Department of Chemistry, Bu-Ali Sina University, Hamedan, Iran

Corresponding author email: a.tirandaz@basu.ac.ir

Regular paper: Received: Jan. 19, 2023, Revised: April. 20, 2023, Accepted: April. 23, 2023, Available Online: April. 25, 2023, DOI: To be added soon

ABSTRACT— A generalized Born-Markov master equation for describing inelastic tunneling under non-equilibrium interaction is recommended. Rate equations are extracted and analyzed for reaching maximization in tunneling rates. Possible rooms for reviving quantum coherence despite the role of the environment have been surveyed. The scheme extended in this article can provide a general framework for the analysis of quantum tunneling in different realms of quantum optics and quantum biology. It is shown how the non-equilibrium character of the system-environment interaction may strengthen the chance of predominance probability of occurrence of inelastic tunneling against elastic tunneling despite the usual expectation.

KEYWORDS: Inelastic tunneling, Open quantum systems, Born-Markov master Equations, Non-Equilibrium Rate Transfers.

I.INTRODUCTION

Nowadays, the theory of open quantum systems is among the most powerful frameworks hired to investigate the dynamics of quantum systems in real conditions in interaction with their environments. Quiet often, the delicateness of quantum superposition hindered the isolation of its evolution independent of the effects of environments. Despite the trivial role of environments in classical mechanics just as a source of noise, in the quantum realm, the reciprocal role of interaction of system-surrounding does have a significant impact on the interpretation of quantum state both empirically and conceptually [1-3]. Decoherence theory tries to unravel various aspects of such interaction. Interaction with the environment suppresses the quantumness at the level of the system's evolution and provides us with an effective dynamical map to explain the quantum-to-classical transition without referring to collapse postulates [1]. In experience, the openness of quantum systems is the main source of the fragility of superposition [4]. A critical issue against making stable Qubits necessary for developing quantum computers is the next expected revolution in science [1ch7,5]. The focus of emerging field Quantum Control is extensively focused on engineering the quantum-environment interaction in a way that, the preservation of the quantum behavior of the system can be possible in spite of the destructive role of environments [6-8]. In quantum optics, wherever one tries to study dissipative phenomena, the implications of open quantum systems theory should be inevitably taken into account [9,10]. In quantum biology, the openness of biological systems demands that decoherence theory be included where the quantum origins of biological systems are aimed to be surveyed [11-13].

Master equations are effective dynamical maps that summarize the underlying physics of system-environment interaction which are widely used in quantum optics problems [14-16]. They add new terms to the well-known Liouville unitary equation that transforms it to a non-unitary evolution dynamic which ascribes the irreversibility of the disappearance of quantum interference when the quantum systems are monitored by an environment with uncountable degrees of freedom [1 ch4, 2 ch5-7]. There are different kinds of master equations among them Lindblad [17], Born-Markov [18] and Redfield equation [19] are the most customary.

Considering non-equilibrium effects in the dynamics of open quantum systems [20,21] is another vivid branch of quantum sciences categorized under the Quantum Thermodynamics theory [22-25] that efforts to search for the validity of thermodynamics rules at the microscopic level or for the systems by a few degrees of freedom. Since the temperature is not exclusively present in the Schrodinger equation, the only way to introduce it is to involve interaction with the environment which is considered to be a collection of harmonic oscillators at thermal equilibrium. The main claim of such studies is that the mutual effect of two or many baths with a quantum system can be stacked in trade for bringing Schrodinger's cat to life which will open new ways for reviving quantum superposition for use in computation and different optic techniques.

All problems which are modeled in standard quantum mechanics can be tackled in an open system framework, too. Among them, inelastic tunneling is our concern in this work. In inelastic tunneling, a quantum object tunnels from a barrier and the difference between the energy of the donor and the acceptor sites in which the object resides hires to excite vibration or electronic states of another object placed in the gap of the two terminals [26]. Inelastic tunneling has been surveyed widely in optical research [27-30]. Inelastic electron tunneling spectroscopy (IETS) uses inelastic tunneling for analyzing the vibration of molecular absorbers on metal oxides [31-32]. It is a powerful tool for understanding nano-scale and molecular junctions. Active optical antennas can be driven by inelastic electron tunneling [33]. Excitation of spin degrees of freedom of an adsorbed atom by inelastic tunneling electrons is surveyed in [34].

In quantum biology, inelastic tunneling of the electron is considered a possible explanation for the sensitivity of olfactory receptors on the structural properties of odorants [35-37]. Since the action at non-equilibrium conditions is a hallmark of the evolution of living systems, having a framework that addresses this trait for studying possible quantum effects could be worthwhile.

In this study, we present the dynamics of an open quantum system in the barriers undergoing inelastic tunneling of the electron at non-equilibrium conditions. A quantum optical master equation has been extracted that gives quantum rate equations between different levels of the open system. We try to elucidate in what manner the rate equations can be updated for non-equilibrium conditions. In Section I, we introduce the model. Details of the calculation of the master equation are given in Section II. The results of the quantum rate equations and the analysis of them is summarized in Section III. The model is experimentally verified in Section IV. The results and the future perspectives are summed up in Section V.

II. ANALYSIS METHOD

To present a model that absorbs the essential features of inelastic tunneling under non-equilibrium conditions, we suppose that a donor-acceptor pair exists with sites $|\mathbf{D}\rangle, |\mathbf{A}\rangle$ on which the electron resides. The donor and the acceptor are coupled respectively with two environments that are in thermal equilibrium at temperatures T_H, T_C. The environments consist of bosonic modes characterized by creation and annihilation operators $\mathbf{b}_i, \mathbf{b}_i^\dagger (\mathbf{c}_i, \mathbf{c}_i^\dagger)$ for a bath in T_H, T_C temperatures where the frequency of bosonic modes is ω_i. The object modes in the gap of the donor and the acceptor are represented by $\mathbf{a}, \mathbf{a}^\dagger$ with frequency ω_0. The donor site with energy ε_D is coupled to the left bath at a higher temperature with a coupling constant λ_{iD} and for the acceptor with the energy ε_A the corresponding constant is λ_{iA}. The tunneling coupling parameter is η. As a result, the Hamiltonian of the system can be defined as:

$$\begin{aligned}\hat{\mathbf{H}} &= \varepsilon_A|\mathbf{A}\rangle\langle\mathbf{A}| + \varepsilon_D|\mathbf{D}\rangle\langle\mathbf{D}| + \\ &\eta\left(|\mathbf{A}\rangle\langle\mathbf{D}| + |\mathbf{D}\rangle\langle\mathbf{A}|\right) + \\ &+\omega_0\mathbf{a}^\dagger\mathbf{a} + \sum_i \omega_i\mathbf{b}_i\mathbf{b}_i^\dagger + \sum_j \omega_j\mathbf{b}_j\mathbf{b}_j^\dagger + \\ &+\left(\lambda_A|\mathbf{A}\rangle\langle\mathbf{A}| + \lambda_D|\mathbf{D}\rangle\langle\mathbf{D}|\right)\left(\mathbf{a}^\dagger + \mathbf{a}\right) + \\ &+\sum_i \lambda_{iA}|\mathbf{A}\rangle\langle\mathbf{A}|\left(\mathbf{b}_i + \mathbf{b}_i^\dagger\right) + \\ &+\sum_j \lambda_{iD}|\mathbf{D}\rangle\langle\mathbf{D}|\left(\mathbf{c}_j + \mathbf{c}_j^\dagger\right)\end{aligned} \quad (1)$$

To confine the dynamics of the object to $|\mathbf{D},\mathbf{A}\rangle$ states, one requires that the tunneling frequency η be small compared to the other energies that are present in the system. In this limit we are allowed to transform the Hamiltonian Eq. 1 to a polaron-transformed picture with the help of a unitary operator $\mathbf{U} = e^{-t\left(\hat{\mathbf{\Pi}}_1 + \hat{\mathbf{\Pi}}_2\right)}$ by the following form:

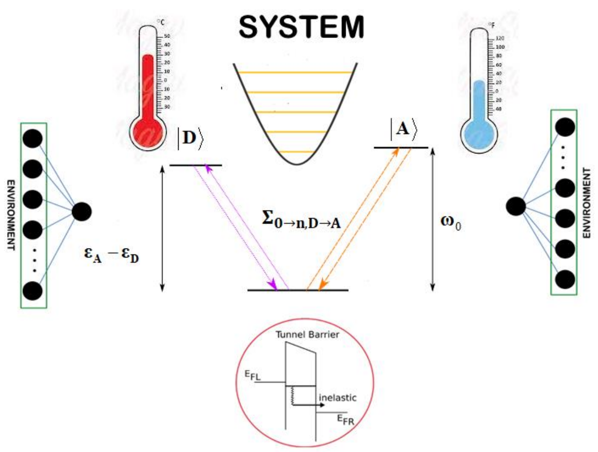

Fig. 1. Schematic representation of the inelastic tunneling under non-Equilibrium conditions. Top: The quantum object can be modeled by a harmonic oscillator interacting with a collection of bosonic modes, a quantum bath, at different temperatures. Bottom: Tunneling of electrons from donor to acceptor excites vibrational modes in the quantum system.

$$\begin{aligned}\hat{\mathbf{\Pi}}_1 &= \left(\kappa_1|\mathbf{A}\rangle\langle\mathbf{A}| + \kappa_2|\mathbf{D}\rangle\langle\mathbf{D}|\right)\left(\mathbf{a}^\dagger - \mathbf{a}\right), \\ \hat{\mathbf{\Pi}}_2 &= \sum_i \kappa_{iD}|\mathbf{D}\rangle\langle\mathbf{D}|\left(\mathbf{b}_i^\dagger - \mathbf{b}_i\right) \\ &+\sum_j \kappa_{jA}|\mathbf{A}\rangle\langle\mathbf{A}|\left(\mathbf{c}_i^\dagger - \mathbf{c}_i\right)\end{aligned} \quad (2)$$

where, $\kappa_{1(2)} = \dfrac{\lambda_{A(D)}}{\omega_0}$, $\kappa_{iA(D)} = \dfrac{\lambda_{iA(iD)}}{\omega_i}$. The polaron transformed Hamiltonian $\hat{\mathbf{H}}_{\mathbf{po}} = \mathbf{U}^\dagger\hat{\mathbf{H}}\mathbf{U}$ splits automatically to diagonal $\hat{\mathbf{H}}_{\mathbf{po},0}$ and non-diagonal $\hat{\mathbf{H}}_{\mathbf{po,int}}$ interaction Hamiltonians as follows:

$$\begin{aligned}\hat{\mathbf{H}}_{\mathbf{po},0} &= \tilde{\varepsilon}_A|\mathbf{A}\rangle\langle\mathbf{A}| + \tilde{\varepsilon}_B|\mathbf{B}\rangle\langle\mathbf{B}| + \\ &+\omega_0\mathbf{a}^\dagger\mathbf{a} + \sum_i \omega_i\mathbf{b}_i^\dagger\mathbf{b}_i + \sum_j \omega_j\mathbf{b}_j^\dagger\mathbf{b}_j\end{aligned} \quad (3)$$

By, $\tilde{\varepsilon}_{A(B)} = \varepsilon_{A(B)} - \omega_0\kappa_{1(2)}^2 - \sum_i \omega_i^2\kappa_{iA(D)}^2$ and,

$$\hat{\mathbf{H}}_{\mathbf{po,int}} = \\ = \eta\begin{pmatrix}|\mathbf{D}\rangle\langle\mathbf{A}|e^{(\kappa_1-\kappa_2)\mathbf{a}^\dagger - (\kappa_1-\kappa_2)\mathbf{a}}\mathbf{\Lambda}_{+h}\mathbf{\Lambda}_{-c} + \\ |\mathbf{A}\rangle\langle\mathbf{D}|e^{-(\kappa_1-\kappa_2)\mathbf{a}^\dagger + (\kappa_1-\kappa_2)\mathbf{a}}\mathbf{\Lambda}_{-h}\mathbf{\Lambda}_{+c}\end{pmatrix} \quad (4)$$

where,

$$\mathbf{\Lambda}_{\pm h(c)} = e^{\pm(\kappa_{iA}-\kappa_{iD})\left(\mathbf{b}^\dagger(\mathbf{c}^\dagger) - \mathbf{b}(\mathbf{c})\right)}.$$

Using time dependent interaction Hamiltonian $\hat{\mathbf{H}}_{\mathbf{po,int}}(t) = e^{\frac{it\hat{\mathbf{H}}_{\mathbf{po},0}}{h}}\hat{\mathbf{H}}_{\mathbf{po,int}}e^{\frac{-it\hat{\mathbf{H}}_{\mathbf{po},0}}{h}}$, one can generate the master equation which encapsulates the dynamics of the reduced density matrix of the quantum object $\boldsymbol{\rho}_s(t)$, The time dependent density matrix in the interaction pictures obtains as:

$$\begin{aligned}\dot{\boldsymbol{\rho}}_s(t) &= -\eta^2\int_0^\infty d\tau\Bigg\{e^{i(\tilde{\varepsilon}_A-\tilde{\varepsilon}_D)\tau} \times \\ &\times\begin{bmatrix}e^{-(\kappa_1-\kappa_2)\mathbf{e}^{i\omega_0 t}\mathbf{a}^\dagger + (\kappa_1-\kappa_2)\mathbf{e}^{-i\omega_0 t}\mathbf{a}}|\mathbf{A}\rangle\langle\mathbf{D}| \\ e^{(\kappa_1-\kappa_2)\mathbf{e}^{i\omega_0(t-\tau)}\mathbf{a}^\dagger - (\kappa_1-\kappa_2)\mathbf{e}^{-i\omega_0(t-\tau)}\mathbf{a}}|\mathbf{D}\rangle\langle\mathbf{A}|\boldsymbol{\rho}_s(t)\end{bmatrix} + \\ &+e^{-i(\tilde{\varepsilon}_A-\tilde{\varepsilon}_D)\tau} \times \\ &\times\begin{bmatrix}e^{(\kappa_1-\kappa_2)\mathbf{e}^{i\omega_0 t}\mathbf{a}^\dagger - (\kappa_1-\kappa_2)\mathbf{e}^{-i\omega_0 t}\mathbf{a}}|\mathbf{D}\rangle\langle\mathbf{A}| \\ e^{-(\kappa_1-\kappa_2)\mathbf{e}^{i\omega_0(t-\tau)}\mathbf{a}^\dagger + (\kappa_1-\kappa_2)\mathbf{e}^{-i\omega_0(t-\tau)}\mathbf{a}}|\mathbf{A}\rangle\langle\mathbf{D}|\boldsymbol{\rho}_s(t)\end{bmatrix}\Bigg\} \times \\ &\times\chi(\tau) + h.c\end{aligned} \quad (5)$$

where $\chi(\tau)$ stands for the correlation function of the environments and contains information about the partitioning of frequencies in bosonic baths:

$$\chi(\tau) = tr_{c,h}\left(\mathbf{\Lambda}_{+h}(t)\boldsymbol{\rho}_{\varepsilon_h}\mathbf{\Lambda}_{+h}(0)\times\mathbf{\Lambda}_{-c}(t)\boldsymbol{\rho}_{\varepsilon_c}\mathbf{\Lambda}_{-c}(0)\right) \tag{6}$$

where tr_ε means trace over environmental degrees of freedom. $\boldsymbol{\rho}_{\boldsymbol{\varepsilon},h(c)}$ represents the density matrix of two baths which are considered to be in thermal equilibrium.

Using $\mathbf{D}(\alpha_1)\mathbf{D}(\alpha_2) = e^{i\,\mathrm{Im}\,\alpha_1^*\alpha_2}\mathbf{D}(\alpha_1+\alpha_2)$ where $\mathbf{D}(\boldsymbol{\alpha})$ stands for displacement operator, and:

$$\left\langle \exp\left(\mu_i(t)\mathbf{a}_i^\dagger - \mu_i^*(t)\mathbf{a}\right)\right\rangle_{\boldsymbol{\rho}_{\boldsymbol{\varepsilon}_j}} = \exp\left(-\frac{|\mu_i(t)|^2}{2}\coth\frac{h\omega_i}{2k_BT}\right) \tag{7}$$

where k_B and T are Boltzmann constant and absolute temperature, respectively, the correlation function of Eq. 6 reduces to:

$$\chi(\tau) = e^{-\int_0^\infty d\tau \frac{J_H(\omega)}{\omega^2}\left((1-\cos(\omega\tau))\coth\left(\frac{h\omega}{2k_BT_H}\right)+i\sin(\omega\tau)\right)} \times e^{-\int_0^\infty d\tau \frac{J_C(\omega)}{\omega^2}\left((1-\cos(\omega\tau))\coth\left(\frac{h\omega}{2k_BT_C}\right)-i\sin(\omega\tau)\right)} \tag{8}$$

where $J(\omega)$ is the spectral density of the environment that reads as:

$$J(\omega) = \sum_k (\lambda_{iA} - \lambda_{iD})^2\,\delta(\omega-\omega_k) \tag{9}$$

There are several ways of defining spectral densities. Usually, the frequency dependence of $J(\omega)$ is taken to obey a power law of the form $J(\omega)\propto\omega^s$. The familiar choice is $s=1$ known as Ohmic spectral densities. Less important cases of interest are sub-Ohmic $s<1$ and super-Ohmic $s>1$. To be physically reasonable, most often, $J(\omega)\propto\omega$ multiplies by another function to avoid an increase in the frequency of bosonic modes without bound. Typically, linear dependency of spectral density holds just for lower frequencies and after crossing a cut-off frequency Γ, subsides at higher values of ω. A Lorentz-Drude form [38] $J(\omega) = \gamma_0\frac{\omega}{\Gamma}\frac{\Gamma^2}{\omega^2+\Gamma^2}$ or $J(\omega) = \gamma_0\frac{\omega}{\Gamma}e^{\frac{-\omega}{\Gamma}}$ are of the most interest. The coupling constant γ_0 is a measure of the interaction strength between bosonic modes of the bath and the quantum system.

Master Eq. 5 has been derived under two approximations and is known as the Born-Markov master equation [39,40]. According to the former, the interaction between the system environment is considered to be weak enough. Then, the combined density matrix of the system environment can be decomposed to for all the time intervals of interaction. Due to the latter, because of the large Hilbert space of the environment in comparison with the system, the memory effect in the environment can be considered to be negligible. As a consequence, the correlation function $\chi(\tau)$ decreases rapidly in time in comparison with natural frequencies in the system. However, one should keep in mind that, negligibility of memory effects does not omit the characteristics of the environment. Since, after tracing on bath, traits of the environment, remain in the functionality of $\chi(\tau)$ functions. Since the two environments have different temperatures, their non-equilibrium character of them affects the dynamics of the system eighter.

Introducing Born and Markov approximation transforms master equations into local in-time differential equations which can be tackled analytically.

III. RESULTS AND DISCUSSION

Let's assume that at the initial time, the electron resides on the donor and the quantum object, represented by a Harmonic potential, is in the ground state. Then we can follow the tunneling of electrons accompanied by excitation of the vibrational mode of the quantum object according to $|\mathbf{D},\mathbf{0}\rangle \to |\mathbf{A},\mathbf{n}\rangle$. We can define the general form of the density matrix of the Donor-Acceptor-Quantum object as:

$$\boldsymbol{\rho}_S(t) = \sum \boldsymbol{\rho}_{h,h',m,n}(t)|\mathbf{h},\mathbf{m}\rangle\langle\mathbf{h}',\mathbf{n}| \qquad (10)$$

where $h, h' \in \{\mathbf{D},\mathbf{A}\}$ and m, n stands for Fock states of QO. We search for populations where the tunneling of the electron has been completed. In other words, when populations of the acceptor have been taken into account. Accordingly, we first make $\langle\mathbf{A}|\dot{\boldsymbol{\rho}}_S(t)|\mathbf{A}\rangle$ due to the master Eq. 5 and then trace over the two environments to obtain the reduced state of the QO. Particularly, we assume that the energy splitting is large enough that hinders excitation to any other Donor-QO states. To be more specific we limit our analysis to only Donor-QO states with no QO excitation at the initial condition. As a result, the rate of transitions of the form $|\mathbf{D},\mathbf{0}\rangle \to |\mathbf{A},\mathbf{n}\rangle$ integrates as:

$$\Sigma_{\substack{0\to n,\\ D\to A}} = \eta^2 \frac{(\kappa_1-\kappa_2)^{2n} e^{-|\kappa_1-\kappa_2|^2}}{n!} \times \times \int_{-\infty}^{+\infty} e^{i(\tilde{\varepsilon}_A-\tilde{\varepsilon}_B+\gamma-n\omega_0)\tau-\Xi\tau^2} d\tau \qquad (11)$$

where $J_0 = \gamma_0^H + \gamma_0^C$ is the sum of coupling constants and Ξ is summarized to:

$$\Xi = \Xi_H + \Xi_C = \lambda_0^H \Gamma_H \left(\frac{k_B T_H}{h\Gamma_H}\right) + \lambda_0^C \Gamma_C \times \times \left(\frac{k_B T_C}{h\Gamma_C} + \frac{1}{6}\frac{h\Gamma_C}{k_B T_C} - \frac{1}{30}\left(\frac{h\Gamma_C}{k_B T_C}\right)^3 + \dots\right) \qquad (12)$$

In doing Eq. 12, $J(\omega) = \gamma_0 \frac{\omega}{\Gamma} e^{\frac{-\omega}{\Gamma}}$ is assumed and the Taylor's series of $\coth\left(\frac{h\omega}{2k_B T}\right)$ for the environment with lower temperature is taken. Also, it is supposed that the regime that we are presently interested, is where the correlation functions are progressively peaked around $\tau \approx 0$. Consequently, in doing time integrals we are allowed to time dependent function in integral Eq. 11 up to the second order in τ.

Figure 2 shows the change in parameter Ξ against temperature, for different proportions of T_H/T_C. As seen, it grows faster for negligible differences between temperatures of the environments for a fixed value of cut-off frequency.

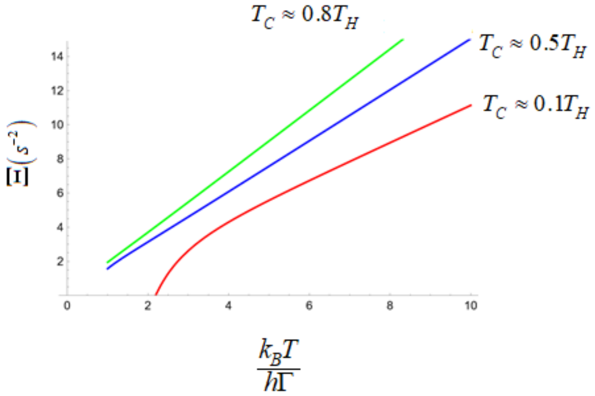

Fig. 2. The change in Ξ for different amounts of T_H/T_C is followed against $(k_B T)/(h\Gamma)$. Where T_H/T_C is lower Ξ raises up faster for a fixed value of cut-off frequency Γ.

Figure 3 depicts the rate of in-elastic (Blue-Red) and elastic (Dashed) tunneling rates against Ξ for different amounts of $J_0, \omega_0, \kappa_{1,2}$. It admits that in-elastic tunneling rates could be predominant in precise portion of the above parameters. In regions where the sum of coupling constants, natural frequency of the system and the difference between energy sites of the donor and the acceptor, represented by $J_0, \omega_0, \kappa_{1,2}$ respectively, have the same order of magnitudes, elastic tunneling rates can be overcome in all regions of Ξ. The Blue plots show inelastic rates for $|\mathbf{0}\rangle \to |\mathbf{2}\rangle$ and red for $|\mathbf{0}\rangle \to |\mathbf{1}\rangle$ excitation.

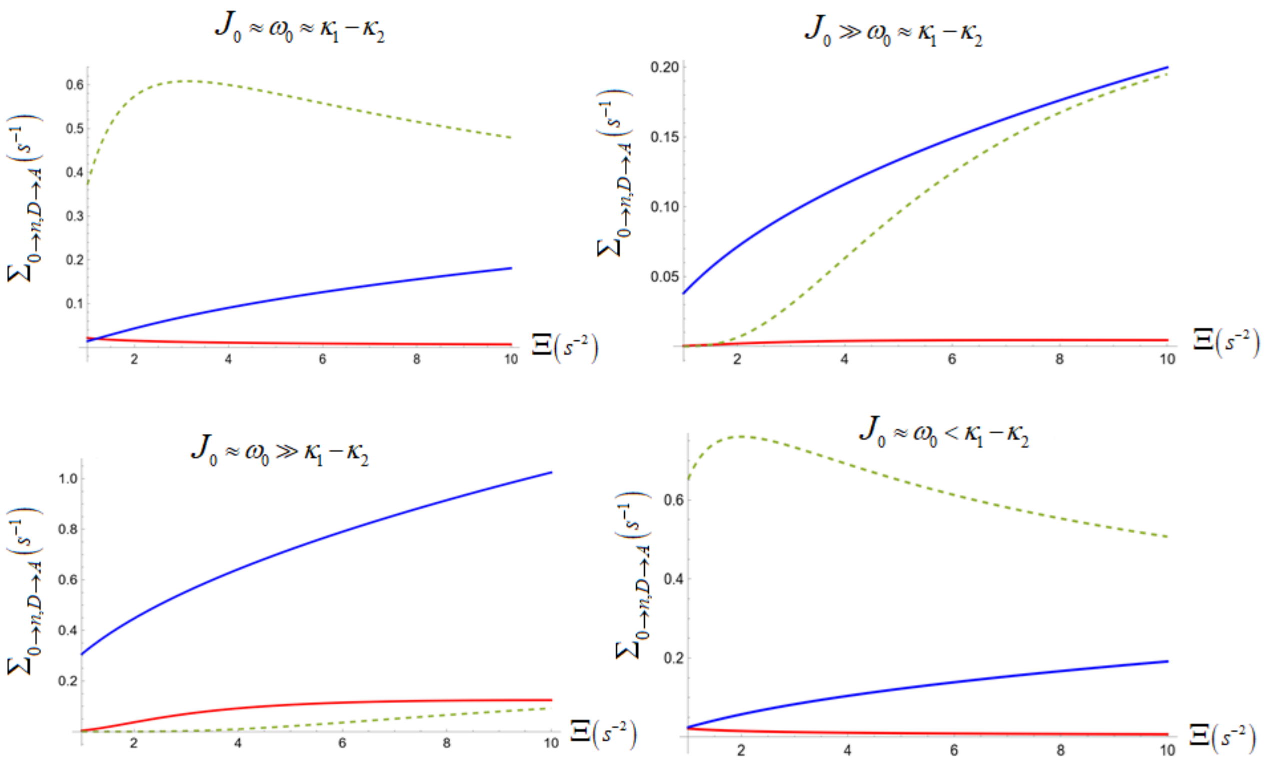

Fig. 3. Rate of inelastic tunneling is depicted for $|\mathbf{0}\rangle \to |\mathbf{1}\rangle$ and $|\mathbf{0}\rangle \to |\mathbf{2}\rangle$ excitation in vibrational mode of the quantum object for different amounts of $J_0, \omega_0, \kappa_{1,2}$ against Ξ. The dashed line describes elastic rates where the quantum object is absent. Inelastic rates are dominant for larger values of J_0 and lower Ξ.

As a noticeable result, excitation rates to higher modes of vibration of the QO, facilitates where the non-equilibrium character of the dynamics is taken into account. In the region in which coupling constants and natural frequency of QO are comparable and larger than the energy difference of donor-acceptor sites, inelastic rates are predominant for a different selection of temperature differences introduced by Ξ. For higher Ξ inelastic rate of $|\mathbf{0}\rangle \to |\mathbf{1}\rangle$ approaches to elastic rate where the QO is absent (Bottom-Left). The same occurs for $|\mathbf{0}\rangle \to |\mathbf{2}\rangle$ inelastic rates where the ω_0 and $\kappa_1 - \kappa_2$ are comparable and lower than coupling strength between system and the environment (Up-Right).

Figure 3 admits that larger values of J_0 is necessary for the dominancy of inelastic tunneling which implies that interaction with non-equilibrium environments with different coupling strength opens rooms for the augmentation of inelastic tunneling. Also, such dominancy is effectively present where the parameter Ξ accepts lower values. According to the analysis of Fig. 3, it is in correspondence with a larger difference between the temperatures of the two environments. As a consequence, an increase in the non-equilibrium character of the environments results in more chances for overcoming inelastic rates of tunneling.

In Fig. 4 we search for the importance of the role of coupling constants and temperature in priority of inelastic against the elastic rate of tunneling. As can be inferred, where the temperature difference is not significant, excitation in lower modes of vibration, $|\mathbf{0}\rangle \to |\mathbf{1}\rangle$ takes larger values in lower coupling strengths. As coupling becomes large, excitation to higher modes, for example $|\mathbf{0}\rangle \to |\mathbf{2}\rangle$, overcomes. The same is true where the temperature of the two baths approaches to gather. However, in this limit, the maximum rate for $|\mathbf{0}\rangle \to |\mathbf{2}\rangle$ excitation becomes larger than the corresponding rate for $|\mathbf{0}\rangle \to |\mathbf{1}\rangle$ transition. As a consequence, a very neat selection of coupling strength and temperatures is necessary for the maximization of inelastic tunneling rates.

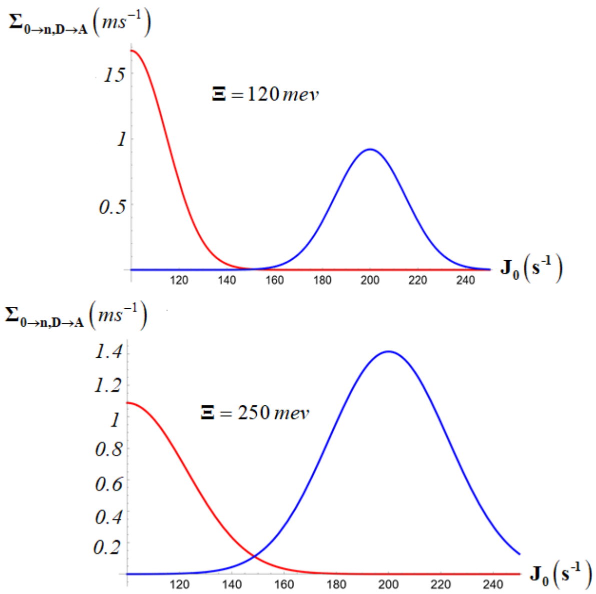

Fig. 4. Inelastic rates for $|0\rangle \to |1\rangle$ (Red) and $|0\rangle \to |2\rangle$ (Blue) excitation is depicted for different values of Ξ. Larger coupling strength needs for having effective inelastic tunneling that accompanies in excitation in a higher mode of vibration.

Investigation of tunneling under non-equilibrium conditions has been surveyed in recent years and attracted attention in the physics community. The general consistency that non-equilibrium condition is a determinant factor in amplification and maintenance of quantum behavior in microscopic systems can be reproduced according to the results discussed above.

In [41], the presence of non-equilibrium effects is introduced as the reason behind the anisotropic relaxations in inelastic tunneling processes. A many-body inelastic quantum tunneling under non-equilibrium conditions for the non-Maxwell-Boltzmann distribution of baths is discussed in [42]. It shows that long-range tunneling between arrays of cold atoms may be accessible under non-equilibrium conditions. Dushmukh and colleagues have shown that tunneling through metal nano particles can be mediated under non-equilibrium condition where many states of the acceptors are provided for the electron [43].

Facilitation in quantum transport in solid-state systems under the non-equilibrium condition is also extensively discussed in [44] and the references therein where in such studies the theory of open quantum systems under the approximations used above are assumed, too.

IV. Experimental Verification of the Model

Inelastic tunneling is one of the fascinating quantum phenomena that can be find its empirical realization in some physical contexts. Inelastic electron tunneling spectroscopy is among well-known examples [45]. In the lab, we can provide two metal plates where a bias voltage is applied to the two contacts. They are characterized by a density of states filled up the Fermi energy. The junction of the two metals is filled by a molecule, a quantum object in our terminology.

Fermi states can be in resonance with some states on the molecule in junction that roles as the donor and acceptor sites, named by D and **A** in our formalism. When the quantum object is absent, the electron on the metal terminal tunnel through the gap and the resulting current will be monitored against the change in the voltage of the source. In the presence of the molecule, some of the electrons lose their energy while tunneling those results in excitation in the vibrational modes of the molecule. As a consequence, it gives an additional current contribution to the aforementioned current that can be tracked in the second derivative of the current against the voltage. The intensity of the inelastic peak is directly dependent on the quantum rates obtained in equation (11). To provide non-equilibrium conditions, the metal terminal can be attached to the thermostat with definite different temperatures.

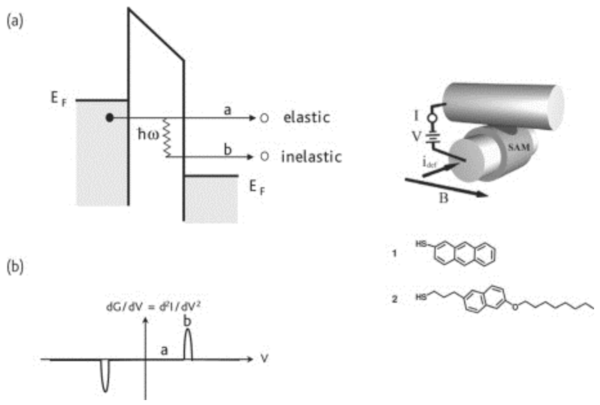

Fig. 5. Inelastic tunneling can be realized in experience by imposing a variable voltage on two metal terminals (Left Up) and monitoring the tunneling current against the voltage where the gap of terminals is filled by a molecule (Right). The inelastic current effect appears in the second derivative of current against voltage and its intensity corresponds with tunneling rates extracted in Eq. 11. The schematic of the figure is presented in [45]

Accordingly, the theoretical results presented above may be found in their empirical verification under experimental constraints.

V. CONCLUSION

In this study, we have generalized and proposed analysis for the examination of inelastic quantum tunneling under non-equilibrium conditions. Circumstances under which the inelastic tunneling can be overcome on elastic one is thoroughly scrutinized. It is verified that the non-equilibrium essence of the system-environment dynamics, summarized in larger values of J_0 and lower values of Ξ increases the chance of predominant inelastic tunneling. It is shown that excitation in a higher mode of vibration can be reached under non-equilibrium conditions for a detailed selection of J_0, Ξ amounts. Since inelastic tunneling is a basis for developing quantum microscopy techniques, this work hopefully shed new lights on extending the limit of applicability of such devices were working at non-equilibrium condition has been considered. We expect that our analysis can be used in experiments to examine the validity of the dissipative quantum model of inelastic tunneling.

Acknowledgment

This work would not have been possible without the support of the Chemistry department of Bu Ali Sina University. I am especially indebted to Dr. Basiri Parsa, Chairman of the Department of Chemistry, who have been supportive of my career goals and who worked actively to provide me with the protected academic time to pursue those goals.

APPENDIX

For the calculation of rate constants, the following integrals have been used:

$$\int_0^\infty \frac{\gamma_0}{\Gamma\omega} e^{\frac{-\omega}{\Gamma}} \sin(\omega\tau)\, d\omega = \frac{\gamma_0}{\Gamma}\tan^{-1}(\Gamma\tau) \qquad \text{(A.1)}$$

$$\int_0^\infty \frac{\gamma_0}{\Gamma\omega} e^{\frac{-\omega}{\Gamma}} \left(1-\cos(\omega\tau)\right)\times$$
$$\times\left(\frac{2k_BT}{h\omega}-\frac{1}{3}\frac{h\omega}{2k_BT}-\frac{1}{45}\left(\frac{h\omega}{2k_BT}\right)^3+\ldots\right)d\omega$$
$$=\frac{\gamma_0}{180h\Gamma\left(k_BT\right)^3}\times$$
$$\times\left(\begin{array}{l} -h^4\Gamma^3+30\dfrac{h^2\left(k_BT\right)^2\Gamma^3\tau^2}{1+\Gamma^2\tau^2} \\ +360\left(k_BT\right)^4\tau\tan^{-1}\left(\Gamma\tau\right) \\ +\dfrac{h^4\Gamma^3\cos\left[3\tan^{-1}\left(\Gamma\tau\right)\right]}{\left(1+\Gamma^2\tau^2\right)^{\frac{3}{2}}} \\ -\dfrac{180\left(k_BT\right)^4\ln(1+\Gamma^2\tau^2)}{\Gamma} \end{array}\right)$$

Also, in doing time integration introduced in Eq. 5, some approximations, in addition to Born-Markov approximations, are made to obtain analytical results. We have supposed that the regime we are presently interested in is where the correlation functions are progressively peaked around $\tau\approx 0$. Accordingly, time integration up to the second order in τ are enough in doing integrals. Hence,

$\tan^{-1}(\Gamma\tau) \approx \Gamma\tau$ and $1-\cos(\omega\tau) \simeq \frac{\omega^2 t^2}{2}$ is assumed:

$$\int_{-\infty}^{+\infty} e^{+iS\tau - P\tau^2} d\tau = \sqrt{\frac{\pi}{P}} e^{\frac{-S^2}{4P}} \quad (A.2)$$

References

[1] M. Schlosshauer, *Decoherence and the Quantum to Classical Transition*, Springer 2nd Ed., 2007.

[2] C. Kirham, *Decoherence: A Quantum Whodunnit*, Independently Published, 2020.

[3] E. Joos, *Decoherence and the Appearance of Classical World in Quantum Mechanics*, 2nd Ed., Springer, 1990.

[4] F. Petruccione and Heinz-Peter Breuer, *Theory of Open Quantum Systems*, Oxford University Press, 2nd Ed., 2007.

[5] T. Monz, K. Kim, A.S. Villar, P. Schindler, M. Chwalla, M. Riebe, C.F. Roos, H. Häffner, W. Hänsel, M. Hennrich, and R. Blatt, "Realization of Universal Ion-Trap Quantum Computation with Decoherence-Free Qubits," Phys. Rev. Lett., Vol. 103, pp. 200503-200510, 2009.

[6] D. Dong and R. Petersen, "Quantum Control theory and Applications: A Survey," IET Digital Library, Vol. 4, pp. 2651-2671, 2010.

[7] D. D'Alessandro, *Introduction on Quantum Control and Dynamics*, Chapman and Hall, 2007.

[8] S. Deffner and S. Campbell, "Quantum speed limits: from Heisenberg's uncertainty principle to optimal quantum control," J. Phys. A: Math. Theor., Vol. 50, pp. 453001-453053, 2017.

[9] M. Orszag, *Quantum Optics Including Noise Reduction, Trapped Ions, Quantum Trajectories, and Decoherence*, Springer, 2016.

[10] U. Weiss, *Quantum Dissipative Systems*, World Scientific Publisher, 3rd Ed., 2012

[11] M. Mohseni, *Quantum Effects in Biology*, Cambridge University Press, 2014.

[12] D. Abbott, *Quantum Aspects of Life*, World Scientific Publishing, 2008.

[13] J. Al. Khalili and J. Mac. Fadden, *Life on the Edge: The Coming age of Quantum Biology*, Transworld Publishing, 2014.

[14] V.N. Gorbachev and A.I. Zhiliba, "Master equations for quantum optics problems," Quantum Opt., Vol. 5, pp. 193-212, 1993.

[15] G.S. Agarwal, "Master Equation Methods in Quantum Optics," Progress Opt., Vol. 11, pp. 1-73, 1973.

[16] D. Manzano, "A Short Introduction to Lindblad Master Equation," AIP Adv., Vol. 10, pp. 025106-025114, 2020.

[17] A. Trushechkin, "Unified Gorini-Kossakowski-Lindblad-Sudarshan quantum master equation beyond the secular approximation," Phys. Rev. A., Vol. 103, pp. 062226-062235, 2021.

[18] C. Karlewski and M. Marthaler, "Time-local master equation connecting the Born and Markov approximations," Phys. Rev. B., Vol. 90, pp. 104302-104311, 2014.

[19] V. Timur and P. Brumer, "Partial Secular Bloch-Redfield master equation for incoherent excitation of multilevel quantum systems," J. Chem. Phys., Vol. 142, pp. 104107-104117, 2015.

[20] G. Schaller, *Open Quantum Systems far From Equilibrium*, Springer, 2014.

[21] P. Mazur, "Mesoscopic non-equilibrium thermodynamics for quantum systems," Physica A., Vol. 298, pp. 81-100, 2001.

[22] S. Deffner and S. Campbell, *Quantum Thermodynamics: An Introduction*, Springer, 2020.

[23] J. Gemmer, *Quantum Thermodynamics*, Springer, World Scientific Publishing, 2004.

[24] G. Kurizki and A.G. Kofmann, *Thermodynamics and Control of Open Quantum Systems*, Cambridge University Press, 2021.

[25] F. Binder, Luis A. Correa, C. Gogolin, J. Anders, and G. Adesso, *Thermodynamics in Quantum Regimes*, Springer, 2019.

[26] M. Razavi, *Theory of Quantum Tunneling*, World Scientific Publishing, 2003.

[27] A. Bayman, P. Hansma, and W. Kaska, "Shifts and dips in inelastic-electron-tunneling spectra due to the tunnel-junction environment," Phys. Rev. B., Vol. 24, pp. 2449-2453, 1981.

[28] J. Hahn, H. Lee, and W. Ho, "Electronic Resonance and Symmetry in Single-Molecule Inelastic Electron Tunneling," Phys. Rev. Lett., Vol. 85, pp. 1914–1927, 2000.

[29] M. Takashi, M. Tikhodeev, and S. Hiromu, "Spectral features of inelastic electron transport via a localized state," Phys. Rev. B., Vol. 68, pp. 205406-205411, 2003.

[30] A. Gupta and J. Lutz, "Single-Atom Spin Flip Spectroscopy," Science, Vol. 306, pp. 466-469, 2004.

[31] W. Wenyong, L. Takhee; I. Kretzschmar, I. Reed, and A. Mark, "Inelastic Electron Tunneling Spectroscopy of an Alkanedithiol Self-Assembled Monolayer," Nano Lett., Vol. 4, pp. 643-646, 2004.

[32] K.W. Hipps and U. Mazur, *Inelastic Electron Tunneling Spectroscopy, Handbook of Vibrational Spectroscopy*, Wiley Publishing, 2001.

[33] K. Braun, F. Laible, O. Hauler, X. Wang, A. Pan, M. Fleischer, and A.J. Meixner, "Active optical antennas driven by inelastic electron tunneling," Nanophoton., Vol. 7, pp. 1503-1516, 2018.

[34] N. Lorente and J.P. Gauyacq, "Efficient Spin Transitions in Inelastic Electron Tunneling Spectroscopy," Phys. Rev. Lett., Vol. 103, pp. 176601-176605, 2009.

[35] A. Tirandaz, F. Taher Ghahramani, and V. Salari, "Validity Examination of the Dissipative Model of Olfaction," Sci. Rep., Vol. 7, pp. 4432-4442, 2017.

[36] E.R. Bittner, A. Madalan, A. Czader, and G. Roman, "Quantum Origins of molecular recognition and Olfaction in Drosophila," J. Chem. Phys., Vol. 137, pp. 22A551-22A558, 2012.

[37] J. Brookes, "Quantum Effects in Biology: Golden rules in enzyme, olfaction, photosynthesis and magnetodetection," Proc. Royal Soc. A, Vol. 273, pp. 551-562, 2017.

[38] E. Wilner, H. Wang, M. Thoss, and E. Rabani, "Sub Ohmic to Super Ohmic behavior in non-Equilibrium Quantum Systems with Electron-Phonon interactions," Phys. Rev. B., Vol. 92, pp. 195143-195152, 2015.

[39] V. Vadimov, J. Tuorila, T. Orell, J. Stockburger, T. Ala-Nissila, J. Ankerhold, and M. Möttönen, "Validity of Born-Markov Master Equation for Single and two Qubit Systems," Phys. Rev. B., Vol. 103, pp. 214308-214317, 2021.

[40] M. Nakatani and T. Ogawa, "Quantum Master Equations for Composite Systems: Is Born-Markov approximation really valid?" J. Phys. Soc. Jpn., Vol. 79, pp. 084401-084412, 2010.

[41] B. Sothmann and J König, "Nonequilibrium current and noise in inelastic tunneling through a magnetic atom," New. J. Phys., Vol. 12, pp. 083028-0833038, 2010.

[42] W. Hou, Y. Wang, J. Wei, Z. Zhu, and Y. Yan, "Many body Tunneling and Non-Equilibrium Dynamics of Doublons in Strongly coupled quantum Dots," Sci. Rep., Vol. 10, pp. 2468-2476, 2017.

[43] M. Deshmukh, "Equilibrium and nonequilibrium electron tunneling via discrete quantum states," Phys. Rev. B. Cond. Matter, Vol. 7, pp. 65-69, 2002.

[44] G. Schaller, *Open Quantum Systems Far from Equilibrium*, Springer, 2017.

[45] M.A. Reed, "Inelastic Electron Tunneling Spectroscopy," Mater. Today, Vol. 11, pp. 46-50, 2008.

Arash Tirandaz was born in Shiraz, Iran, in 1983. He has completed Bachelor (1385), Master (1387) and PhD (1393) at Sharif University of Technology, Tehran, Iran. His field of Study is Pure Chemistry. His Major field of Study is physical chemistry specially foundation of quantum mechanics and the theory of open quantum systems.

Arash was the reasercher of the Research Institute in Fundamental Sciences (IPM) from 1393-1395 at Foundation of Physics group under the supervision of Dr. Mehdi Golshani.

Afterwards, he was a Postdoc Researcher at IPM in School of Biology and BioInformatics for three years until 1398. Then, he started a new position as a researcher in Quantum Technologies Center at Tehran until 1400. Now, he is a faculty member of Bu Ali Sina University in Departement of Chemistry, Hamedan, Iran. He has published 11 ISI journal papers till 2023.

THIS PAGE IS INTENTIONALLY LEFT BLANK.