Exploring the non-equilibrium fluctuation relation for quantum mechanical tunneling of electrons across a modulating barrier

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Abstract

We experimentally explore the phenomenon of electron tunneling across a modulated tunneling barrier which is created between an STM tip and an Au film deposited on a vibrating piezo surface. Measurements of the time series of the quantum mechanical tunneling current across the modulating barrier show large fluctuations. Analysis of the average work done in establishing tunneling current in finite time interval shows a distribution of both positive and negative work events. The negative work events suggest tunneling against the bias voltage direction. We show that these distributions obey the Gallavotti Cohen Non-equilibrium Fluctuation Relations (GC-NEFR) valid for systems driven through a dissipating environment. Typically, while the GC-NEFR has been shown for non-equilibrium classical systems we show its validity for the quantum mechanical tunneling process too. The GC-NEFR analysis also gives us a way to measure the dissipation present in this quantum tunneling system. We propose the modulated barrier behaves like a lossy scattering medium for the tunneling electrons resulting in a tendency to randomize of the tunneling process.

Introduction

A phenomenon which distinguishes between classical from quantum behaviour is the quantum mechanical tunneling of particles across a finite potential barrier. While quantum tunneling across a static barrier is a popular textbook level problem [1], the study of tunneling across a periodically modulated barrier is rich and complex. The phenomenon of tunneling across a periodically modulated barrier came into focus with experiments showing the ionization of a neutral atom due to tunneling when placed in an alternating electric field [1,2]. It was found that the tunnel ionization probability
depends on both the frequency and amplitude of the drive. The tunneling across a modulated barrier is also seen in photon assisted tunneling phenomenon in Superconductor - Insulator – Superconductor junctions [3] and in quantum dots [4]. In the simple one-dimensional case, for a particle with total energy $E$ described as a plane wave, incident on a static rectangular barrier of height $V_0$ of width $w$, the tunneling probability exponentially decays with $w$ and $\sqrt{\frac{2m}{\hbar^2}}(V_0-E)$. Within the wave picture of the tunneling particle it has been suggested that for the periodically modulated barrier, a part of the wavefunction tunnels across the barrier while a part of it remains back within the barrier. The part left back evolves within the modulated barrier and part of it tunnels across the periodically modulated barrier [5]. Typically for a barrier modulated at a frequency $\Omega$, the solutions for the reflected and transmitted waves not only have the usual stationary wave solution, but also have waves which are reflected and transmitted at frequency $\Omega$ [2,5,6,7]. It has also been suggested that under certain conditions, a continuous drive may lead to localization and completely destroy coherent tunneling across the barrier [8,9,10]. Apart from the above issues it may be noted that periodically driven quantum systems are rarely studied in isolation, as they are continuously interacting with the environment like a thermal bath. Such situations lead to dissipation effects which affect the dynamics of the evolution of the wave function in such periodically driven systems [5,11,12,13]. Such open quantum systems can no longer be studied using the conventional Schrodinger’s equations [14,15,16,17] and have led to the study of interesting new phenomena like dissipative phase transitions in these open systems [18,19,20,21,22,23]. While dissipation is important in these system getting a measure of the dissipation in these system is not clear. All these described features make a dissipative driven tunneling system interesting and rich from the point of view of exploring its nonlinear and out-of-equilibrium features, and therefore important for more experimental investigations. It also turns out that experimental realization of these systems is quite sophisticated and complex.

Fluctuation theorems offer a way to quantitively explore systems far from equilibrium. For a system driven through a medium where it is dissipating and exchanging energy with the surrounding (for a steady state flow), Gallavotti Cohen derived a fluctuation theorem.
Here $s_\tau = \frac{1}{\tau} \int_0^{+\tau} s(t') dt'$ where $s_\tau$ denotes the rate at which entropy is produced in the non-equilibrium steady state. Here $P(+s_\tau)$ is the probability of entropy production, namely, of observing entropy-increasing events of magnitude $s_\tau$, and $P(-s_\tau)$ is the probability of observing an entropy decreasing event of magnitude $-s_\tau$ where $-s_\tau$ is the entropy consumed over a duration $\tau$. Here $\tau$ is the observation time interval. Note that for relatively small observation times $\tau$, the finiteness of $P(-s_\tau)$ suggests that one observes entropy decreasing events which is a violation of the second law of thermodynamics within the small time interval of observation. However for large observation time interval $\tau$, as expected (see eqn.1) the probability of an entropy-increasing event $+s_\tau$ dominates over the $P(-s_\tau)$. For non-equilibrium systems, the result of exchange of energy between the system and the environment results in the entropy of the system (within a finite time window) to either increase (the usual case) or even decrease. From eqn. 1 we get $\frac{1}{\tau} \ln \frac{P(+s_\tau)}{P(-s_\tau)} = \xi s_\tau$, where $\xi^{-1} \propto k_B T_{\text{eff}}$. Here $T_{\text{eff}}$ is an effective temperature scale for non-equilibrium systems, and it has no connections with the equilibrium temperature. The value of $T_{\text{eff}}$ has often been deduced to be as large as $10^{16}$ K [24, 25].

Different experiments have shown the validity of fluctuation relations in classical systems like dragging of a Brownian particle in an optical trap, electrical circuits, RNA stretching, Rayleigh-Bernard convection [26,27], pressure fluctuations on the surface kept in a turbulent flow [28], vertically shaken granular beads [29], Lagrangian turbulence on a free surface [30], liquid crystal electro-convection [31], vortices in superconductors [25] etc. While most of the studies exploring the Gallavotti Cohen Non-Equilibrium Fluctuation Relation (GC-NEFR) have been in classical systems, there have been few studies on exploring the validity of GC-NEFR in non-equilibrium quantum dissipating systems. In this work, we explore the tunneling across a modulated barrier using Scanning Tunneling Microscope (STM). Recently we have shown [32] that tunneling between the STM tip and a conducting surface (Au film) on a piezo vibrating at a frequency $\omega$, produces a modulation in the tunneling current at frequency $\omega$. The modulation in the tunneling current is effectively due to modulation in the tunneling barrier at

$$L \tau \to \infty \frac{P(+s_\tau)}{P(-s_\tau)} = e^{\xi s_\tau}$$ 

(1)
frequency $\omega$. In this paper we explore the fluctuations in the long time series of the tunneling current. We observe the presence of large excursions in the tunneling current which are both above and below the mean tunneling signal. We show that these distributions obey the well-known Gallavotti Cohen Non-Equilibrium Fluctuation Relations (GC-NEFR) valid for systems driven through a dissipating environment (steady state case). Typically, the validity of the GC-NEFR has usually been shown for non-equilibrium classical systems. The GC-NEFR analysis gives a way to measure the dissipation present in this quantum tunneling system. We propose the modulated barrier behaves like an inelastic scatterer for the tunneling electrons resulting in the observed features.

**Experimental details**

We use a Quazar Technologies make room temperature STM (NanoRev. 4.0). In fig. 1(a) we show the atomic arrangement in Highly Ordered Pyrolytic Graphite (HOPG) sample imaged using this STM. We also show in fig. 1(a) the schematic representation of our STM. This set up is used to measure the time series of the tunneling currents between the STM tip and a vibrating surface. Placed below the STM tip is a conducting gold film deposited on top of a vibrating piezoelectric crystal which has a diameter ~ 1.4 cm and thickness ~ 0.33 mm. The piezo is stuck to a glass substrate (1cm × 2cm) which is then stuck to the gold-coated metallic stub of the STM with double-sided adhesive tape (fig. 1(a)). The STM circuit is completed by shorting the top conducting surface of the piezoelectric crystal with the gold-coated STM metallic stub. For our STM tip, we use an electrochemically etched Pt-Ir alloy wire and maintain a constant dc bias $V_b = -1.5$ V between the tip and the gold film on top of the vibrating piezo surface. The STM in our experiment is operated in constant current mode. When the piezo crystal surface vibrates with frequency $f$, the tunneling gap between the STM tip and the conducting surface on top of the piezo also gets modulated with frequency $f$ (see schematic in fig. 1(a)). This results in periodic variations of the tunneling current predominantly at $f [32]$ (along with some higher harmonics). These
tunneling current modulations can be measured from the voltage drop across a 1 GΩ resistor \( V_T = I_{TC} \times 10^9 \, \Omega \) using an DSO (Digital Storage Oscilloscope). It may be mentioned here that in the constant current mode of STM operation, technically it is often easier to measure the modulations by measuring the time series of the feedback signal \( V_{FB} \). The feedback signal is a fraction of the tunneling signal \( V_T \), which is used to control the STM. We confirm this in fig. 2, which shows a linear relationship between the measured \( V_{FB} \) and \( V_T \) showing that the \( V_{FB} \) is proportional to the tunneling signal. The linear relationship shows that the \( V_{FB} \) is also a faithful representation of the tunneling signal. In our
measurement, the time series of the feedback signal \( \left( V_{FB}(t) \right) \) from the STM is measured using a DSO (Yokagawa DL 9000 series) with a sampling rate of 5 Giga-samples per second. We would like to mention that in order to avoid any AC electrical coupling between the bottom surface of the piezo (an oscillating voltage is applied to the bottom surface to vibrate the piezo) with the piezo’s top surface, the top surface of the piezo crystal (fig. 1(a)) is grounded through a 10 µF polar capacitor. The time-series signal \( (V_{FB}(t)) \) was captured for different frequencies \( f \) of the vibrating piezo surface, ranging from 100 kHz to 1000 kHz. Initially, before capturing the data, the piezo is not vibrated. In this condition, the STM is first set up to obtain an average finite dc \( V_{FB} \) level which is established due to tunneling. The modulations of the feedback voltage due to the vibrating piezo occur around this mean dc voltage level. After this setting, the piezo surface is vibrated by applying an AC signal to the piezo of amplitude 30 Volts. All measurements are performed in ambient conditions.

Figure 2: Shows the variation of the feedback signal with the tunneling voltage in the STM.

Figure 1(b) shows the time series of the feedback voltage for piezo excitation frequencies \( f \) of 500 kHz captured for a time duration of 500 seconds (note the signal shown is obtained after subtracting the mean dc \( V_{FB} \) level discussed above). The time-series data of the modulating feedback tunneling voltage \( V_{FB}(t) \) captured over tens of microseconds time interval has been explored in ref. [32]. Figures 1(b), 3(a), (d) and (g) show long time-series data of \( V_{FB}(t) \) spanning a few hundreds of seconds. Here we see large fluctuations which are associated with fluctuations in the tunneling current. Figure 1(d) (red data
points) shows the intrinsic electronic noise in $V_{FB}(t)$ signal when the tip is positioned over a non-vibrating thin Au film. It clearly doesn’t show any of the large fluctuations observed in fig.1(b). Also, in fig.1(d), we show as the black data points the time series of the bare STM electronics noise floor voltage signals when there is no tunneling current. Here too, we do not observe large fluctuations like that in fig.1(b). We must mention that all experiments discussed here are performed under identical ambient. While the fluctuations in the $V_{FB}(t)$ from a non-vibrating substrate are more than the bare electronic noise floor, they are still an order of magnitude less than that with a vibrating quartz surface below the STM tip (fig. 1(b)).

From the time series of $V_{FB}(t)$ data captured for different piezo excitation frequencies (500 kHz, 700 kHz and 1000 kHz) between the STM tip and vibrating piezo surface (fig 1(b)) we calculate the autocorrelation function using the expression $C(t) = \frac{\langle V_{FB}(t + t') V_{FB}(t) \rangle}{V_0^2}$ where $V_0$ is the mean of $V_{FB}(t)$ and $\langle .. \rangle = \frac{1}{T} \int_0^T .. dt'$ (see fig. 1(c)). The $C(t)$ shows the $V_{FB}(t)$ signals are uncorrelated beyond tens of millisecond. Inset of fig. 1(c) shows the behaviour of the Fourier transform of $|C(t)|^2$ of 500 kHz data in the main panel i.e. fig. 1(c) shows the power spectrum $P(f)$ as a function of frequency on a log-log scale. Inset of fig. 1(c) shows that the power spectrum of the noise has a non-shot noise type behavior, viz. $P(f) \propto \frac{1}{f^\alpha}$ where $\alpha = 0.7 \pm 0.1$ (recall that shot noise is characterized by $P(f) \propto \frac{1}{f}$). The absence of shot noise features suggests that the fluctuations are not to the discreteness of the tunneling of electronic charges.

In fig. 3(b) we analyze the time series in fig. 3(a) in terms of $P(W_\tau)$. Here the $P(W_\tau)$ is the probability of observing an event of magnitude $W_\tau$ within an observation time interval of $\tau$. From the $V_{FB}(t)$ signal (after subtracting the dc offset voltage), like the ones shown in fig. 3(a), the feedback voltage time series $V_{FB}(t)$ is broken up into a series of time bins each of width $\tau$ where we calculate $W_\tau$ as

$$W_\tau = \frac{1}{\tau} \int_t^{t+\tau} I_T(t') V_h \, dt'$$

$$= \frac{1}{V_h(I_T)} \int_t^{t+\tau} I_T(t') \, dt'$$

(2)
Here $I_T$ is the tunneling current, and $V_b$ is the constant dc STM bias voltage applied between the STM tip and base. Note $I_T \propto V_T$ where $V_T$ is the tunneling voltage and as $V_{FB} \propto V_T$ (fig. 2). Therefore,

$$W_\tau = \frac{1}{\tau} \int_0^{\tau} V_{FB}(t') V_b \, dt'$$

(3)

Figure 3: (a) Time series of the feedback voltages for a frequency of 500 kHz of the vibrating piezo. (b) The probability distribution functions (PDF) of the time series in (a). (c) The $R$ v/s $W_\tau$ for 500 kHz frequency. (d) Time series of the feedback voltages for a frequency of 700 kHz of the vibrating piezo. (e) The probability density functions of the time series in (d). (f) The $R$ v/s $W_\tau$ for 700 kHz frequency. (g) Time series of the feedback voltages for a frequency of 1000 kHz of the vibrating piezo. (h) The probability density functions of the time series in (g). (i) The $R$ v/s $W_\tau$ for 1000 kHz frequency.

Where $<V_{FB}>$ is the average value of the $V_{FB}(t)$ signal within an observation time interval $\tau$. $W_\tau$ is the average work done in binning time width $\tau$, for electrons tunneling from the STM tip onto the vibrating surface of the piezo.

Often for experiments, the Gallavotti Cohen Non-equilibrium fluctuation relation (GC-NEFR) is restated in terms of $W_\tau$ [24]

$$R = \frac{1}{\tau} \ln \left( \frac{P(W_\tau)}{P(-W_\tau)} \right) = s_\tau = W_\tau <s(t)>$$

(4)

With $s(t) = IV/T_{eff}$ where $\tau$ is the time window of observation and $T_{eff}$ is the effective temperature of the system. We determine $P(W_\tau)$ from the time series signal in fig. 3(a) and plot it in fig. 3(b). From fig. 3(b) we see that the $P(W_\tau)$ versus $W_\tau$ curve, also called the Probability Distribution Function (PDF), has
a non-Gaussian distribution around a positive mean peak of \(<W_\tau>\sim 1.1\). We see from the \(P(W_\tau)\) distribution that it not only has positive work events but also has a significant probability of negative work, \(-W_\tau\) events. The \(-W_\tau\) events arise from the excursions of \(V_{FB}(t)\) signal which fall below the mean \(V_{FB}\) signal as noted in fig.1(b) earlier. The positive \(W_\tau\) events represent entropy increasing events viz. work done within observation time interval \(\tau\) when tunneling is established with the bias voltage. The negative, \(-W_\tau\) event are unusual events, representing the average work done against the bias drive within time interval \(\tau\). For these negative work events it appears as if, within a finite observation interval \(\tau\), one catches glimpses of tunneling electron which seem to be swimming up against the bias voltage. All of the above features which are seen in fig. 3(b) for 500 kHz excitation of the vibrating piezo surface are reproduced at other excitation frequencies as well (see figs. 3(e) and 3(h)).

Figure 3(c) shows the \(R\) v/s \(W_\tau\) for different \(\tau\), scaled onto a single linear curve (eqn. 4) for 500 kHz frequency of the piezo. The scaling of \(R\) versus \(W_\tau\) (eqn.4) for all different values of \(\tau\) onto a straight-line fit, till significant values of \(W_\tau = 4\), demonstrates the validity of the GC-NEFR for the driven tunneling system where the tunneling barrier is modulated at a frequency of 500 kHz by vibration of the piezo surface below the STM tip. Figures 3(f) and (i) also exhibit linearity of \(R\) v/s \(W_\tau\) till significant values of \(W_\tau\), thereby confirming the validity of GC-NEFR for tunneling between the STM tip and piezo surface vibrating at 700 kHz and 1000 kHz.

Figure 4: \(F(W_\tau)\) curves for different \(\tau\) for (a) 500 kHz (b) 700 kHz and (c) 1000 kHz of the vibrating piezo. Solid lines in the curve represent the quadratic behaviour of \(F(W_\tau)\).
To analyze the shapes of the $P(W_\tau)$ curves in figs. 3(b), 3(e) and 3(h) we plot a function (also used to analyze the large deviation function [24,25])

$$F(W_\tau) \propto \ln [P(W_\tau)]$$ (5)

If $P(W_\tau)$ is gaussian then $F(W_\tau) \propto (W_\tau - \langle W_\tau \rangle)^2$ viz. $F(W_\tau)$ has a quadratic dependence on $W_\tau$. Using the $P(W_\tau)$ data of fig. 3(b) and eqn. 5 we determine $F(W_\tau)$, (see fig. 4(a)) for excitation frequencies of 500 kHz of the vibrating piezo. Note that for large $W_\tau$, the $F(W_\tau)$ is non-quadratic in nature. Similar feature is seen for 700 kHz data in fig 4(b). At 1000 kHz we see the $F(W_\tau)$ is completely non-quadratic in shape for all $W_\tau$, however it obeys GC-NEFR quite well (eqn. 4). A deviation of $F(W_\tau)$ from quadratic nature represents that the fluctuations in $W_\tau$ are not random events. Thus, it seems that the fluctuations in the tunneling current through the modulated barrier may be correlated. The correlations could be

Figure 5(a): Shows the variation of slope ($\propto \frac{1}{T_{\text{eff}}}$) with the frequency of the vibrating piezo. The solid line is a guide to the eye. (b) $I$-$V$ while tunneling onto the gold conducting surface on the piezo for different frequencies of vibration.

responsible for the deviation from the $\frac{1}{f}$ type of noise power spectrum seen in fig. 1(c) inset. It may be also noted that 1000 kHz is near resonance of the piezo. The deviation from Gaussian could be related to additional dissipation induced in the quantum tunneling occurring in the system near resonance. In the GC-NEFR, in the curve between $R$ v/s $W_\tau$ in eqn. (4), the slope is $\propto \frac{1}{T_{\text{eff}}}$. We use the slope of $R$ v/s $W_\tau$ curve as a measure of inverse of the dissipation ($\delta^{-1}$) in this driven quantum tunneling system. In fig.
5(a) we plot the slope ($\propto \delta^{-1}$) v/s the frequency of the piezo crystal’s vibration. It is clear that beyond 400 kHz of modulation frequency of the barrier, the dissipation in the tunneling quantum system increases. We show in fig. 5(b) that the local $I-V$ of the Au film (on top of the piezo) measured with the STM for different vibrating frequencies are identical, suggesting that the states of the Au film into which the electrons are tunneling into, are identical at different frequencies. Hence, any change in the density of electronic states of the film is not responsible for the observed features.

Although at present we don’t understand the mechanism of dissipation, we envisage the following scenario. It is known that a statistical description of the behavior of the tunneling current for a modulating barrier can be obtained by considering a part of the wave tunnels through and a part remains trapped within the barrier [5]. However, this is not sufficient to explain the dissipation and negative entropy events observed in our experiments. We believe the modulated barrier should possess internal quantized energy states associated with the part of the electron wave trapped within the barrier from prior tunneling events. With the modulating barrier due to changing STM tip to sample distance, the barrier energy level spacings are also periodically modulated. We believe that an electron impinging on the modulated barrier, it should scatter inelastically. Due to the modulating energy levels, it is possible that the impinging tunneling electron may cause resonant excitations within levels of the barrier and lead to losses in energy from the tunneling electron which go into causing excitations within the barrier. Some electrons may also be reflected back along with energy loss, and some may tunnel across with energy loss. The reflected electrons may account for the negative fluctuation events observed. Hence for quantum tunneling across a modulated tunneling barrier between the STM tip and piezo surface, the barrier at different frequencies behaves like a lossy medium, which appears to become impervious to some of the tunneling electrons while some tunnel through. The loss of this medium is a function of frequency.

In conclusion we show the validity of the Gallavotti Cohen Non-equilibrium fluctuation relation for a dissipating quantum tunneling system. We believe the analysis provides a useful way to quantify the dissipation in this quantum system. The validity of the GC-NEFR shows that the symmetries which govern the classical GC non-equilibrium fluctuation relations are valid in the quantum regime too. More
future theoretical and experimental investigations are needed to understand the complexity of electron tunneling across a modulated barrier.

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