Examining the Information Content of Voltage-Dependent, Two-Dimensional Conductance Histograms

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I. INTRODUCTION

Conductance histograms [1] are a common way for experimental studies to characterize the conductance through nanometer-scale systems (*i.e.*, systems where quantum mechanical effects are crucial). In many cases, experimental techniques are unable to precisely determine the system being measured, so the experiment is performed many (usually thousands) of times and the conductance properties are determined statistically. The experimental data is compiled into a conductance histogram; a peak is interpreted as the "expected" conductance.

Recently, there have been several advances in probing the information content of conductance histogram peaks [2, 3]. All of these analyses assumed the conductance data was measured near zero applied bias (voltage), which is often the case. These studies showed that the peaks (i) qualitatively reflect the transport mechanism and (ii) quantitatively detail the electrode-channel coupling(s) and level alignment.

However, two-dimensional conductance histograms reporting conductance data at non-zero biases have been reported [4]. The main goal of this project is to extend the previous types of analyses [2, 3] to these voltage-dependent cases. The experimental data show interesting features that may reflect physical processes. For example, I hypothesize that asymmetry in the bias drop across the two interfaces is readily apparent in the experimental data.

II. PROJECT GOALS

1. Generalize the histogram simulation code to include the bias voltage.
2. Investigate the roles of various physical parameters (coupling strengths, level alignments, bias asymmetry) on two-dimensional conductance histograms. This entails using the simulation software from the previous point to generate many histograms and infer the underlying processes.
3. Correlate the simulated histograms to experimental histograms. Some are published in Ref. 4; that research group can also be contacted for additional data, if desired. Can we provide additional insight into the experimental data?

III. VOLTAGE-DEPENDENT ELECTRON TRANSPORT

The Landauer-Büttiker-Imry (coherent scattering) theory for electron transport will be assumed throughout. In this case, the key quantity is the transmission function,

$$T(E) = \frac{\Gamma^2}{(E-\varepsilon)^2 + \Gamma^2}, \tag{1}$$

where E is the energy of the electron, ε is the channel energy, Γ is the channel-electrode coupling (we assume the channel couples symmetrically to both electrodes [5]), and T is the probability of an electron transmits from one electrode to the other. T is called the transmission. The current is then given by

$$I(V) = \frac{2e}{h} \int\limits_{-\infty}^{\infty} \mathrm{d}E T(E;V) \left[f_{\mathrm{L}}(E;V) - f_{\mathrm{R}}(E;V) \right], \tag{2}$$

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where I is the current, V is the applied bias (voltage), e is the electron charge, h is Planck's constant, and $f_{\mathrm{L(R)}}$ is the Fermi function of the left (right) electrode. Note that $T(E)$, in principal, also depends on V since ε and Γ might be functions of V.

For simplicity, we will work in the zero-temperature limit, which is fine from the perspective of tunneling. This assumption essentially freezes out any inelastic effects, which are already neglected in the Landauer-Büttiker-Imry formalism. In this case,

$$f_{\mathrm{L}}(E;V) \to \Theta(-E + E_{\mathrm{F}} + \eta eV),$$
$$f_{\mathrm{R}}(E;V) \to \Theta(-E + E_{\mathrm{F}} + (\eta - 1)eV),$$

where Θ is the Heaviside step function, E_{F} is the Fermi energy of the system, and $0 \le \eta \le 1$ controls the relative voltage drop at each of the electrode-channel interfaces. As written, $\eta = 0$ means all voltage drops at the right electrode; $\eta = 1$ is at the left. Then, from Eq. (2),

$$\begin{aligned} I(V) &= \frac{2e}{h} \int_{-\infty}^{\infty} \mathrm{d}E T(E;V) \left[\Theta(-E + E_{\mathrm{F}} + \eta eV) - \Theta(-E + E_{\mathrm{F}} + (\eta - 1)eV)\right] \\ &= \frac{2e}{h} \int_{E_{\mathrm{F}}+(\eta-1)eV}^{E_{\mathrm{F}}+\eta eV} \mathrm{d}E T(E;V). \end{aligned} \tag{3}$$

Finally, we look at the differential conductance,

$$\begin{aligned} g(V) &\equiv \frac{\partial}{\partial V} I(V) \\ &= \frac{2e}{h} \int_{-\infty}^{\infty} \mathrm{d}E \left\{ \frac{\partial}{\partial V} T(E;V) \left[\Theta(-E + E_{\mathrm{F}} + \eta eV) - \Theta(-E + E_{\mathrm{F}} + (\eta - 1)eV)\right] \right. \\ &\qquad \left. + T(E;V) \left[\eta e \delta(-E + E_{\mathrm{F}} + \eta eV) + (1-\eta) e \delta(-E + E_{\mathrm{F}} + (\eta - 1)eV)\right] \right\} \\ &= \frac{2e^2}{h} \left[\eta T(E_{\mathrm{F}} + \eta eV; V) + (1-\eta) T(E_{\mathrm{F}} + (\eta-1)eV; V)\right] + \frac{2e}{h} \int_{E_{\mathrm{F}}+(\eta-1)eV}^{E_{\mathrm{F}}+\eta eV} \mathrm{d}E \frac{\partial}{\partial V} T(E;V). \end{aligned} \tag{4}$$

In the limit $V \to 0$, we recover the familiar expression

$$g(0) = \frac{2e^2}{h} T(E_{\mathrm{F}}; 0).$$

IV. SIMULATING CONDUCTANCE DATA

Equation (4) provides the differential conductance through a system with a particular level energy (ε), coupling strength (Γ), and bias drop symmetry (η). In experiment, each time the measurement is taken, these values will change slightly since the experiment is measuring a slightly different system. To simulate this effect, we consider each of these parameters to be a random variable. That is, each "experimental measurement" corresponds to a random choice of these parameters from some statistical distribution. For simplicity, we will assume each is normally distributed; as an example, Γ comes from a normal distribution with mean Γ_0 and standard deviation δ_Γ. We then simulate many (thousands) of experiments by choosing many sets of parameters and calculating the conductances.

I see this projecting evolving in a couple steps.

1. Until otherwise stated, we will assume

$$\frac{\partial}{\partial V} T(E;V) = 0$$

   so that the integral in Eq. (4) disappears. The first step is to write code to simulate the conductance data. This should be a seemingly straightforward extension of code for the zero-bias case. Code for binning conductance data into a histogram is already written.

2. Look at one-dimensional histograms for data collected from non-zero biases. I want to start developing an intuition for what these should look like.

3. Simulate two-dimensional conductance histograms. I will help write the code for binning two-dimensional conductance data into the histogram. Of particular interest here is the effect of bias drop asymmetry; that is $\eta \not\approx 0.5$. I think this may explain some of the experimental data.

4. Work in the case where

$$\frac{\partial}{\partial V} T(E; V) \neq 0.$$

This probably entails the supposition that ε_0 and/or Γ_0 are some functions of V. For simplicity, we'll start with linear functions. I hope this change will explain the nonlinearity in some of the experimental data.

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[1] B. Xu and N. J. Tao, Science **301**, 1221 (2003).
[2] M. G. Reuter, M. C. Hersam, T. Seideman, and M. A. Ratner, Nano Lett. **12**, 2243 (2012).
[3] P. D. Williams and M. G. Reuter, J. Phys. Chem. C **117**, 5937 (2013).
[4] S. Guo, J. Hihath, I. Díez-Pérez, and N. Tao, J. Am. Chem. Soc. **133**, 19189 (2011).
[5] Mathematically, only the product of the two couplings matters in the far off-resonance case. It is considerably easier to just define an effective coupling $\Gamma \equiv \sqrt{\Gamma_1 \Gamma_2}$ rather than account for any asymmetry.