Data analysis of nonlinear systems: Application to Au nanowires

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A general scheme for analyzing nonlinear systems is presented, with measurements of the nonlinear \(I–V\) characteristics of a gold atomic contact with a mechanically controllable break junction used as an example. Simultaneous measurements of \(I(V)\) and \(\partial I/\partial V(V)\) were analyzed to extract the unbiased conductance from the normalized functional form of \(I(V)\). This allows us to remove the error in the conductance measurements associated with fluctuations in the atomic structure of the system (about 5\%). This demonstrates the potential of this technique to discover and subsequently understand new phenomena in nanometer scale systems, where measurements are often challenging due to noise, difficult-to-control quantities such as the atomic structure of a contact lead, and the lack of adequate, generally accepted theoretical descriptions. A discussion of the tradeoff between higher-order measurements and averaging artifacts due to nonlinearity is also included. © 2002 American Institute of Physics. [DOI: 10.1063/1.1498908]

I. INTRODUCTION

The ideal measurement requires that every variable remains constant except for one (the control variable). Obviously, this is not true in practice, but one aims to minimize the impact of “undesirable” variables. The error in most experiments is assumed to be random and is analyzed using Gaussian error calculus. The method that follows does not rely on mathematical fits or statistical analysis, and does not presuppose a physical model to extract the desired information from individual sets of data. This method reduces the systematic error due to the undesirable variables, which in turn adds strength to any mathematical fit, statistical analysis, or physical model that one may want to apply.

As a tangible example, let us consider the measurement of current (or conductance) as a function of applied bias voltage for a gold atomic contact formed with a mechanically controllable break junction at room temperature. The \(I–V\) characteristics of atomic scale wires is currently the subject of much debate.\(^1\) Figure 1 shows typical data of the differential conductance \(\partial I/\partial V\) and the dc conductance \(I/V\) over the course of a 5 s voltage sweep from positive to negative voltage bias. Both polarities share a common overall shape, but seem to vary significantly in the details of their conductance behavior. However, for each polarity, one notices that there seem to be similar details between the dc and differential conductance. There are some key questions: how much are these fluctuations due to changes in voltage, and how much are due to changes in atomic structure? Furthermore, what is the value of the observed nonlinearity in \(I(V)\)? One needs to extract the answers to these questions from experimental data such as presented in Fig. 1 in order to compare them to theoretical predictions.

II. FORMULATION

Let us begin with a very general formulation of the problem. This will allow us to answer these questions without using a mathematical fit or having to assume a theoretical model describing the physics. The measured current \(I\) depends on two things: voltage \(V\) and everything else \(X\). With this in mind, consider the following definitions:

\[
\begin{align*}
I(X,V) &= g(X) f(X,V), \\
\partial I/\partial V(X,V) &= g(X) \frac{\partial f}{\partial V}(X,V), \\
g(X) &= \frac{\partial I}{\partial V}(X,0).
\end{align*}
\]

While these equations remain completely general, they still allow for physical interpretation. In our particular system, we can interpret \(X\) as a parameter that reflects the atomic structure of the junction. It should be stressed that this interpretation has no effect on the analysis that follows. No explicit knowledge of \(X\) will be necessary. \(g(X)\) is defined as the unbiased conductance [from Eq. (3)], which makes \(f(X,V)\) the normalized functional form of the voltage dependence and \(\partial f/\partial V(X,V)\) the normalized differential conductance. It is easiest to see the convenience of this form by considering familiar examples: for Ohmic behavior, \(f(X,V) = V\) and \(g(X) = G\) since \(I = GV\). In mesoscopic conductance \(g(X) = G_0 \geq T\) according to Landauer formalism.\(^8\)\(^9\)

Let us define a quantity we will call the shape function \(S\):

\[
S = \frac{\partial I}{\partial V} \frac{V}{I}.
\]

Before discussing the importance and interpretation of \(S\), let us compute it using the data shown in Fig. 1. Figure 2 shows \(S\) plotted for both positive and negative bias; the similarity of the overall shape as well as the details (shown in the inset of
To emphasize the importance of this point, let us perform the following gedanken experiment: we measured $I$ and $\delta I / \delta V$ through a variable, Ohmic potentiometer as a function of applied voltage (the control parameter). Suppose there were drastic changes in the temperature during the measurements, and at the same time some troubleshooter turned the knob of the variable potentiometer without telling anyone (the undesired variables). Glancing at the measured $I(V)$ and $\delta I / \delta V(V)$ alone, one might wrongly conclude nonlinear behavior. Both the temperature and knob of the variable potentiometer are represented by the parameter $X$ in this case, and they strongly affect $g(X)$. However, since $f=V$ and $\delta f / \delta V = 1$ are not changing along with $g(X)$, the shape function ($S$) would also not be affected. Thus, we would be able to determine that $f(X,V)$ was not changing from trial to trial from the fact that $S$ remained unchanged. In this simple example, $S=1$, which is indicative of linear behavior. We could even determine the variable conductance $g(X)$ since we could deduce that $f=V$.

In a more general case, $S$ is a more complicated function of $V$, but from the previous arguments, we can understand some of our results presented in Fig. 2. As in our potentiometer example, if $S(X,V)$ is identical in two trials, then $f(X,V)$ is identical in both trials. This would mean that any differences in $I(X,V)$ between the trials are attributable to changes in $g(X)$. Since $S$ (shown in Fig. 2) for positive and negative bias is very similar despite the stronger differences in conductance in Fig. 1, we can see that these discrepancies between the conductance curves are predominantly due to the fluctuations in $g(X)$. Without considering the shape function, we might not have guessed that $S(X,V)$ [and thus $f(X,V)$] would be so weakly affected by changes in $X$ despite the stronger changes in $g(X)$.

Using this example, we would also like to stress a subtle but important point: our shape function analysis distinguishes between a quantity being a function of $V$ and being merely voltage driven. For instance, the change in temperature of the atomic contact, and thus $g(X)$, can depend on the history of how we ramp up the bias voltage from zero to a finite value $V$ at which the $I(X,V)$ is measured: one could increase $V$ so slowly that it does not destroy thermal equilibrium; or the troubleshooter could monitor the applied dc voltage while turning the knob so that changes in $g(X)$ were correlated with $V$. Despite the fact that both of these voltage-driven processes would be related to $V$ and lead to repeatable $I(X,V)$ vs $V$ curves, neither would be mistakenly attributed to a functional dependence of $V$ by our analysis. This is a purely mathematical consequence of the definition of $g(X)$ and the fact that $X$ is not a proper function of $V$ for these voltage-driven events that depend on the process history. Any functional dependence on $V$ is included in $f(X,V)$ by definition.

To get a better understanding of the interpretation of the shape function, let us again consider examples. Consider $I = g(V)^a$, $\delta I / \delta V = g a V^{(a-1)}$, which leads to $S = \alpha$. In the case of linear behavior ($\alpha=1$), this means $S=1$, as from the variable potentiometer example. As a slightly more complicated example, consider a current with a linear and cubic term: $I = g(V + BV^3)$ (see Fig. 3). At $V$ near 0, $S$ is close to 1 since

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**III. EXAMPLES AND INTERPRETATION OF S**

From Eqs. (1), (2), and (4) we can see that the shape function does not depend on the factor $g(X)$, which is common to the two simultaneously measured quantities $I$ and $\delta I / \delta V$:

$$S = \frac{\delta f}{\delta V} \frac{V}{f}.$$  

(5)

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![Fig. 1. Differential ($\delta I / \delta V$) and dc ($I/V$) conductance (upper and lower curves, respectively) of a Au junction vs $V$. The measurement was taken over the course of a 5 s voltage sweep from positive (gray) to negative (black) bias.](image1.png)

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![Fig. 2. Shape function ($S$) vs $V$ calculated from the data in Fig. 1. Inset shows the magnified portion of $S$ that illustrates shared detail between the two polarities. The positive bias is in gray and the negative bias is in black.](image2.png)
the linear term is dominant. As $V$ increases, $S$ approaches 3, with higher values of $\beta$ making a quicker approach to cubic behavior. These examples illustrate how the local behavior of $I$ is $V^5$. In other words, $S$ is the local slope of the log-log plot of $f(V)$ and $I(V)$ $[(\partial \log f)/(\partial \log V)$ or $(\partial \log I)/(\partial \log V)]$. This makes $S$ a useful measure of the strength of the nonlinearity since weak nonlinearity leads to $S$ close to 1; strong nonlinearity leads to $S$ deviating substantially from 1.

In passing, let us mention that it is beyond the scope of this article to analyze the physical origin of the observed shape function. In tunneling measurements, $S$ is approximately proportional to the electronic density of states.\textsuperscript{10,11} The origin of the nonlinearity in electronic transport observed in Figs. 1 and 2 is discussed elsewhere.\textsuperscript{7}

**IV. DATA ANALYSIS**

In the most general case, $I=I(X, V)$, and we can apply the chain rule to $I(X, V)$. Since we can measure $I$ and $\partial I/\partial V$, this allows us to directly calculate the changes in $I$ due to $V$ ($\int (\partial I/\partial V) dV$), and those due to $X$ ($\Delta I - \int (\partial I/\partial V) dV$). Going further, we would like to separate the changes in terms of variations in $g$ and $f$ so as to better compare with theoretical models. Within the framework of $g(X)$ and $f(X, V)$, the relationship between $S$ and $I$ will be easier to see. With these aims in mind, we manipulate the chain rule to yield

\[
\int \frac{dI}{I} = \int \frac{\partial f}{\partial V} \frac{1}{V} dV + \int \frac{\partial f}{\partial X} \frac{1}{V} dX, \tag{6}
\]

\[
\Delta \log(I) = \int \frac{\partial f}{\partial V} \frac{1}{V} dV + \int \frac{\partial f}{\partial X} \frac{1}{V} dX + \int \frac{\partial g}{\partial X} \frac{1}{g} dX. \tag{7}
\]

The first term in Eq. (7) determines the voltage-dependent change in $f$ (and thus $I$). At $V=V'$ relative to a point at $V=V_0$, we can conclude

\[
\frac{f(V')}{f(V_0)} = e^{\int_{V_0}^{V'} (\partial f/\partial V)(1/f) dV} = e^{\int_{V_0}^{V'} [S(V)/V] dV}, \tag{8}
\]

\[
\frac{\partial f}{\partial V}|_{V=V'} = \frac{S(V')}{S(V_0)} \frac{V_0}{V'} e^{\int_{V_0}^{V'} [S(V)/V] dV}, \tag{9}
\]

recalling Eq. (5). Equations 8 and 9 more clearly illustrate how $S$ is related to $f$, and $\partial f/\partial V$ due to changes in $V$. It should be noted that since these equations contain only the voltage dependence of $f$ (and thus $I$), they are of interest to compare with theories involving the electronic properties of the junction.

To be absolutely clear, let us provide the recipe for a discrete set of data points. From our $\partial I/\partial V$ measurement, we can use Eq. (3) as our boundary condition to determine $g$ at a point close to $V=0$. This, along with our measurement of $I$, allows us to determine $f$ from Eq. (1) at this point. By using a linear interpolation scheme between data points for $S$, we can evaluate Eq. (8) to determine $f$ at the next data point. $g$ and $\partial f/\partial V$ at this point can be determined from Eqs. (1) and (5), respectively. The process is repeated for each of the following points using each previous point as the boundary condition.

$\partial f/\partial V$ (Fig. 4) and $f$ (inset of Fig. 4) are plotted versus $V$. We can see that the positive and negative bias curves for $\partial f/\partial V$ and $f$ fall on top of each other as they did in the case of $S$ (Fig. 2). How much of an improvement is $\partial f/\partial V$ or $f/V$ compared with $\partial I/\partial V$ or $I/V$ (Fig. 1)? If we were to merely use $\partial I/\partial V$ or $I/V$ to compare with theory, we would be including the fluctuations of $g$ in our error. $g$ vs $V$ (Fig. 5) is a parametric plot that indicates what the unbiased conductance is at the time a particular measurement was made at $V$. Over the course of the 5 s voltage sweep, the fluctuations in $g$ were
roughly 5%. This represents the error that is removed when we use \( \partial f / \partial V \) and \( f \).

The above technique uses \( f(X,V) \rightarrow f(V) \) for mathematical convenience. This means that \( g \) encompasses all changes due to \( X \). This is justified when \( S \) is not strongly affected by the changes in \( X \). In our case, the strong overlap in \( S \) between the two polarities and the absence of abrupt changes in \( S \) between data points testifies to this. Another check of consistency is that the variations attributed to \( g \) (Fig. 5) do not correlate with \( S \) (Fig. 2).

If this is not the case, we can extend the ideas about analysis in terms of \( V \) and \( X \) by including higher-order partial derivatives of \( I \) with respect to \( V \). With these, we can deduce higher-order derivatives of \( S \). We can also deduce the shape function of \( S \) (let us call it \( S_2 \)):

\[
S_2 = \frac{\partial^2 I}{\partial V^2} V = -S + 1, \tag{10}
\]

and generalize this to an \( n \)th-order shape function:

\[
S_{n+1} = \frac{\partial S_n}{\partial V} V S_n. \tag{11}
\]

Under this convention, \( S_1 = S \) and \( S_0 = I \).

With these ingredients, it is straightforward to separate the \( V \) and \( X \) dependencies of \( I, S, S_2, \ldots, S_n \) to arbitrary order using the chain rule as outlined above. Now, if one suspects \( S \) is experiencing strong changes due to \( X \), we can use \( S_2 \) to separate these effects from those due to \( V \) to determine the components of \( f(X,V) \). We apply the exact same scheme used to separate \( I(X,V) \) into \( g(X) \) and \( f(V) \) using \( S \). If resorting to \( S_2 \) is still unsatisfactory, we can continue to \( S_3 \).

The higher-order derivatives also allow us to better define the local behavior of \( f \) and \( I \) by expanding \( S \) as a polynomial:

\[
S = a + bV + cV^2 + \cdots, \tag{12}
\]

\[
I = gV^{(1/2)}e^{(1/3)cV^3} + \cdots, \tag{13}
\]

which is more transparent than Eq. (8). We have thus derived higher-order correction terms as an improvement to the previous observation that \( I \) and \( f \) behave locally like \( V^5 \).

V. MEASUREMENT ARTIFACTS

Is there any limitation on the achievable precision by going to higher-order derivatives? One problem inherent with measuring derivatives is that we have to modulate our signal by modulating our control variable. Consider a Taylor expansion of our modulated signal:

\[
I[V_0 + \Delta V \sin(t)] = I(V_0) + \Delta V \sin(t) \frac{\partial I}{\partial V} \bigg|_{V = V_0} + \frac{(\Delta V)^2 \sin^2(t)}{2!} \frac{\partial^2 I}{\partial V^2} \bigg|_{V = V_0} + \cdots. \tag{14}
\]

Our measurements of \( I(X,V) \) are time averaged:

\[
\overline{I}(V_0) = I(V_0) + \frac{1}{2} \left( \frac{\Delta V}{V_0 - V_0} \right)^2 \frac{\partial^2 I}{\partial V^2} \bigg|_{V = V_0} + \frac{3}{8} \frac{\Delta V^4}{4!} \frac{\partial^4 I}{\partial V^4} \bigg|_{V = V_0} + \cdots. \tag{15}
\]

This shows how a systematic error \( [\overline{I}(V_0) - I(V_0) \neq 0] \) is measured in any nonlinear system as a function of the modulation amplitude \( \Delta V \). This rectified contribution from the ac (whether it is intended or due to noise) has proven to be a concern in other nanosopic systems.\(^{12}\)

Any wiggles in the \( I \sim V \) curve imply that there is at least one nonzero even-ordered derivative. The sharper the feature, the larger the derivative associated with them. The larger the modulation voltage, the stronger the effect these derivatives will have on the measurement. Another way of looking at this is that sharp features (having a small voltage scale in comparison to the modulation voltage) will be washed out, while broad features (having a large voltage scale in comparison to the modulation voltage) will not.

In a similar way, our lock-in measurements (which lead to \( \partial I / \partial V \)) are affected by \( \Delta V \) and the odd-ordered derivatives:

\[
\int_0^{2\pi} \sin(t) I[V_0 + \Delta V \sin(t)] dt = \frac{\pi}{2} \frac{\partial I}{\partial V} \bigg|_{V = V_0} + \frac{3 \pi}{8} \frac{(\Delta V)^3}{3!} \frac{\partial^3 I}{\partial V^3} \bigg|_{V = V_0} + \cdots, \tag{16}
\]
There is also a subtler problem associated with this phenomena: $f(\tilde{l}/dV)dV \neq \Delta \tilde{l}$ in the general case of a nonlinear dependence on the control variable ($V$). One obtains a systematic error of

$$\Delta \tilde{l} = \int \frac{\tilde{l}}{dV} dV = \frac{(\Delta V)^2}{8} \frac{\partial^2 I}{\partial V^2} + \cdots,$$

which would be interpreted as a change due to $X$ according to the chain rule. Note that these unexpected errors are present in any nonlinear, modulated system even in the absence of changes in $X$. It is a direct result of the nonlinearity of the system. In a similar way, modulation can lead to artifacts in $S$ that contribute to systematic errors in $f$.

This problem can be even more pronounced if higher-order derivatives are measured. To measure higher harmonics with good signal to noise with a lock-in amplifier, one needs to increase $\Delta V$. This compounds the problem since the systematic error increases along with $(\Delta V)^2$ for finite higher-order terms. One must then carefully evaluate the tradeoff of being able to measure higher-order terms (allowing better interpolation between discrete data points and the use of higher-order shape functions) and the reduction of these unavoidable artifacts related to the nonlinearity of the system.

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