Channel saturation and conductance quantization in single-atom gold constrictions

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It is shown that the finite elasticity of atomic-sized gold constrictions allows for a continuous and reversible change in conductance. This is achieved by superposition of atomic-scale or subatomic-scale mechanical oscillations on a steadily retracting/approaching gold tip against a gold substrate. Through these perturbation studies, we report the direct observation of channel saturation and conductance quantization in stable, single-atom gold constrictions. In addition, the origin of peaks in conductance histograms is explained, and the peaks alone are shown to be insufficient in evaluating the stability of atomic configurations.

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I. INTRODUCTION

Realization of atomic-sized metal or metal-molecule electronics devices^{1–4} requires a fundamental understanding of conditions under which conductance quantization prevails,⁵ their electronic structure and regulation of electron transmission behavior,^{6–19} and their mechanical stability against disruptive forces of entropic thermal fluctuations.^{20–23} In addition, positioning and manipulation of individual atoms requires a basic understanding of operative forces^{24–27} and an ability to measure them.^{28–37} Quantization of conductance was first shown experimentally in a two-dimensional electron gas.^{38,39} Similar studies on atomic-sized metal point contacts pose experimental challenges owing to the difficulty in continuously varying the diameter of the constriction on the scale of the Fermi wavelength (on the same order of interatomic distances) without causing an atomic rearrangement. Simultaneous measurements of conductance and force in gold constrictions show that a stepwise change in conductance is accompanied by a stepwise change in force, indicating a concomitant atomic rearrangement within the constriction.^{36,37,40,41} To extract the signature of conductance quantization, conductance histograms were initially introduced.^{42,43} From such histograms, it was posited that even though the observed conductance may assume integer and noninteger values of G_o ($=2e^2/h$, the quantum of conductance; e is quantum of charge and h is Planck constant), the fact that peaks only occur at or near integers of G_o constitutes or at least strongly suggests conductance quantization.^{42,43} In addition, a characteristic sequence of peaks was reported in sodium conductance histograms (peaks at $1G_o$, $3G_o$, $5G_o$, and $6G_o$, with peaks at 2, 4, and $7G_o$ being absent) as a signature of conductance quantization⁴⁴ (even though a small but well-defined peak is observed at $2G_o$ in both Na and K histograms⁴⁵). Later, two of the authors in Ref. 44 questioned the use of histogram peaks as proof of conductance quantization, instead reinterpreting them as arising from preferred atomic configurations.⁴⁶ Nonetheless, these authors noted that their original interpretation of the characteristic sequence of peaks in Na histogram⁴⁴ as a signature of conductance quantization would be “difficult to explain by other models,” while adding that the role of preferred atomic arrangements has to be

considered in interpreting the histograms.⁴⁶ In subsequent studies, peaks in conductance histograms were construed as representing stable atomic configurations arising from electronic and shell effects.^{47–49} Recently, peaks in conductance histograms have been theoretically revisited using molecular-dynamics simulations and conductance calculations based on the tight-binding model.⁵⁰ These results show theoretically that the minimum cross-sectional area of the contacts is not the only factor in determining conductance. Contact geometry and disorder may also play a role, and contacts of different radii may have similar net conductance. *The above discussion shows that the current interpretation of conductance histograms recognizes the limitations of histogram peaks, although the origin of the histogram peaks remains unclear and their use remains prevalent in the literature.* In investigations related to conductance quantization, and aside from histograms, distinct trends of quantum suppression of shot noise have also been observed near integer multiples of G_o .^{51,52} In addition, the number of valence electrons has been shown to be equal to the number of available channels.⁵³ However, measurements of transmission coefficients have also shown that an observed conductance of NG_o may not necessarily arise from N channels.⁵⁴

The present study reports the direct observation of channel saturation and conductance quantization in single-atom gold constriction under mechanical perturbation. Combined with noise analysis of conductance plateaus, the origin of peaks in conductance histograms is also explained.

II. EXPERIMENTAL DETAILS

Gold films (200 nm thick) were magnetron sputtered on silicon substrates in an Ar partial pressure of 3 mtorr in a UHV chamber whose base pressure was $\sim 10^{-8}$ – 10^{-9} torr. The Au sputtering target was 99.999% pure. Atomic force microscope (AFM) cantilever tips (Si) were also sputter coated with Au films (60 nm thick) for force-elongation measurements. The cantilevers were periodically rotated relative to the sputtering gun for enhanced uniformity of the coating. The scanning tunnel microscope (STM) tips were prepared by electrochemically etching an annealed Au wire (250 μ m diameter, 99.999% purity). Etching was performed in a mixture of 50% HCl and 50% ethanol at 2.4 V. The sharpness of

the STM tip was critical to the formation of stable contacts.

A modified AFM (Ambios Q-Scope Nomad) was used for simultaneous measurement of conductance and force elongation at room temperature. The AFM assembly consisted of a dual piezo configuration, one for coarse and another for fine alignment of the substrate relative to the cantilever tip. With this configuration, the minimum step size was 4 pm and the noise was ~ 5 pm. The cantilevers had a spring constant of ~ 11 N/m (Veeco Probes), and they were also independently calibrated.⁵⁵ The photodetector was calibrated using the well-established optical deflection technique.

Mechanical perturbation measurements were made using a custom-built, inert atmosphere STM at room temperature, which also utilizes the dual piezo configuration. The perturbations were in the form of superimposed atomic-scale or subatomic-scale mechanical oscillations on a retracting/approaching gold tip against a gold substrate. The amplitude of oscillations ranged from 0.05 to 0.5 nm (peak-to-peak) at frequencies from 0.1 to 100 Hz, with most experiments done at 3 Hz. Use of distance modulation has been previously applied to detect formation of single-molecule junctions.⁵⁶ The motion of the fine piezo (Physik Instrumente, GmbH) was calibrated using an absolute method, viz., by forming a parallel-plate capacitor between the holders for the tip and substrate. The piezo motion was obtained by multiplying the piezo voltage by the calibration factor (an open loop circuit). For enhanced stability, the AFM and STM assemblies were mounted on a three-stage vibration isolation system to minimize the destabilizing effects of mechanical vibrations. Finally, they were enclosed in an acoustic chamber and a Faraday cage. Stable, single-atom contacts lasting for long periods (~ 25 s) could be readily formed without any feedback (no piezo motion).

Conductance traces were recorded at a bias voltage of 250 mV. The current was measured using an *IV* converter (Femto), whose series resistance was accounted for in the conductance calculations. Whereas in the literature the measured conductance values in a histogram have sometimes been further shifted,^{57–59} no such shifts were made in the present study. For all experiments, the piezo was extended at a rate of 1 nm/s until the conductance was $>20G_0$ to ensure a new geometry each time. For noise analysis and mechanical perturbation measurements, the piezo was then retracted at a rate of 0.25 nm/s until the conductance was $10G_0$ and then further retracted at a rate of 3.5 pm/s until the contact broke. The retraction rate of 3.5 pm/s was the slowest that could consistently break the contacts from a size of approximately ten atoms. Below this rate, atomic diffusion caused the contacts to grow faster than they could be consistently broken. The slowest possible retraction rate was chosen to minimize the effect of strain. For other measurements, the retraction rate varied from 3.5 pm/s to 5 nm/s (see figure captions).

III. RESULTS AND DISCUSSION

A. Histograms and load-elongation characteristics

Figure 1 shows a series of gold conductance histograms, where successive histograms represent the collective total of

FIG. 1. (Color online) A series of gold conductance histograms at room temperature, where successive histograms represent the collective total of all preceding experimental cycles (from 400 to 5000). The inset shows the zoom-in view of the histograms corresponding to the first three peaks. The retraction speed of the piezo is 1 nm/s. The bin size of the conductance histograms is $0.02G_0$. The data acquisition during retraction was arbitrarily cut off at $0.1G_0$ in order to separate counts for piezo elongation versus retraction. This leads to an artificial hump in counts at this conductance value.

all the preceding experimental cycles. The inset in Fig. 1 shows a zoom-in view of the first three peaks; distinct peaks can be seen close to integer multiples of G_0 . Notice that, except for an increase in the number of counts with additional experimental cycles, the position of the peaks remains unchanged. To gain a mechanistic understanding of the underlying conductance traces, consider the trace shown in Fig. 2(a), and the simultaneously measured force-elongation curve in Fig. 2(b). Figure 2(a) shows that the long plateau at $\sim 1G_0$, in fact, undergoes several atomic reconfigurations before rupture. For example, even the small but finite stepwise change in conductance from $1.05G_0$ to $0.99G_0$ represents an atomic reconfiguration, as indicated by a large stepwise change in force in Fig. 2(b). This highlights the fact that *a small stepwise change in conductance does not necessarily mean a small jump in force*. The latter is a function of the spring constant of the rupturing atomic configuration, its elasticity, and the state into which it transitions. Moreover, the force-elongation curves can only tell that an atomic reconfiguration has occurred *but not the number of atoms involved*. In Figs. 2(a) and 2(b), a series of atomic reconfigurations occur just prior to rupture, manifesting as telegraphic fluctuations in conductance and corresponding jumps in the measured force. The elongated plateau at $\sim 1G_0$ corresponds to a total extension of ~ 0.62 nm prior to rupturing (see note in Ref. 60), and it could be due to atomic chain formation. Chain formation, which is well established, is beyond the scope of the present study, and means of investigating them are reported elsewhere.^{61,62}

Pertinent to the present study, two-atom constrictions are later shown to frequently give rise to net conductance close to or even below $1G_0$. Therefore, when adding counts to the

FIG. 2. (Color online) (a) A conductance trace along with (b) the simultaneously measured force-elongation curve. The last plateau reveals several small stepwise changes in conductance, each of which registers corresponding stepwise change in force. (c) A conductance trace whose last conductance plateau is consistently below $1G_o$. (d) Its simultaneously measured force-elongation curve. Inset in (c) shows details of the conductance trace and force-elongation curve for the plateau below $1G_o$. The retraction speed of the piezo is 5 nm/s. The dotted lines selectively highlight correlation between change in conductance and measured force.

first peak in a histogram, it must first be ascertained whether the various plateaus, say, at $1.05G_o$, $0.99G_o$, and $0.84G_o$ in Fig. 2(a), actually correspond to a true single-atom constriction. Yet, in building a histogram, such counts are routinely added to the first peak at $\sim 1G_o$ without any distinction. Figure 2(c) shows another conductance trace where the last plateau is consistently below $1G_o$; Fig. 2(d) shows the simultaneously measured force-elongation curve. The inset in Fig. 2(c) shows a zoom-in view of the plateau at $\sim 1G_o$ and the corresponding force-elongation curve. Figures 2(c) and 2(d) show that telegraphic changes in conductance occur within the $1G_o$ plateau. Even these minimal changes in conductance (from $\sim 0.97G_o$ to $\sim 0.95G_o$ and back) are accompanied by changes in the measured force. The telegraphic change in conductance occurs when the constriction has been stretched by 0.31 nm, following which the constriction elongates by another 0.115 nm before rupturing, for a total elongation of

FIG. 3. (Color online) (a) An example of a conductance trace that undergoes random telegraphic noise, which registers corresponding changes in force, as shown in its simultaneously measured force-elongation curve in (b). The window emphasizes the correlation between RTN and corresponding changes in force. The retraction speed of the piezo is 5 nm/s. (c) An example of a two-level RTN and the corresponding changes in force. The dotted lines in (c) selectively highlight the close correlation between change in conductance and measured force corresponding to RTN. (d) An example of RTN that undergoes transition between more than two levels. The two (solid) windows in (d) highlight the close correlation between change in conductance and measured force corresponding to the complex RTN transitions. In the examples shown in (c) and (d), the piezo is stationary.

0.425 nm.⁶³ Although this plateau is consistently below $1G_o$, in terms of building a conductance histogram, *a priori* it cannot be assumed that it is due to a single-atom constriction.

Figure 3(a) is an example of a conductance trace that shows sustained random telegraphic noise (RTN) during elongation of the constriction, along with the simultaneously measured force-elongation curve in Fig. 3(b). The window in Fig. 3(a) highlights a section where the conductance telegraphically fluctuates back and forth between approximately $4G_o$ and $5G_o$. The corresponding section of the force-elongation curve in Fig. 3(b) shows a finite restoring force

for these two conductance levels, revealing the origin of these fluctuations to be the transition between two metastable atomic configurations; note that the use of the term “level” and its origins is in the context of accepted terminology (such as states, classes, and levels) of random telegraphic noise to describe discrete conductance values taken by various atomic configurations and should not be confused with “quantum-mechanical levels.” Figures 3(c) and 3(d) show two other examples of RTN. In these examples, the piezo is held stationary (no retraction or extension of the piezo), and thermal fluctuations cause instability in the constrictions. The dotted vertical lines in Fig. 3(c), and the highlighted sections in Fig. 3(d), selectively emphasize the correlation between change in conductance and measured force as the system transitions between different atomic configurations. Whereas Fig. 3(c) represents the simpler case of two-level fluctuations, Fig. 3(d) shows the more complicated case of multi-level fluctuations; see Ref. 64 for detailed discussion of RTN in metal constrictions. For other origins of RTN, see Refs. 6, 16, and 65–73. In particular, earlier work on larger diameter metal nanocontacts (where quantized conductance is not observed) by Ralls and Buhrman^{72,73} attributed RTN to fluctuations of metastable defects between discrete configurations. In contrast, in the present study, as well as in a recent study,⁶⁴ mechanical or thermal perturbations cause the contact geometry to fluctuate between different atomic configurations, leading to slightly different transmission probabilities, and manifesting as RTN. Nonmechanical origins of RTN are beyond the scope of the present study, and will be discussed in the future.

B. Noise analysis

An assessment of relative stability of atomic configurations requires a mechanistic understanding of how various atomic configurations respond to a perturbation. Description of mechanical stability of an atomic configuration requires knowledge of both the spring constant k and elongation x ; within the energy landscape, the change in elastic potential energy of an atomic configuration being $\Delta U = \int_x^x kx dx$. This information cannot be obtained from a histogram. Therefore, a question that naturally arises is: how are various atomic configurations represented or assimilated in a conductance histogram? One avenue is to analyze the characteristic noise of each conductance plateau. However, prior to making such an analysis, artifacts arising from stepwise changes in conductance (for example, those arising from RTN) must be eliminated. If such stepwise transitions are included, the resulting analysis will give spuriously high noise. This is illustrated in Fig. 4(a) where the constriction is seen to undergo RTN between two distinct levels. The inset in Fig. 4(a) shows a zoom-in view of the two levels (labeled as Level-1 and Level-2). The highlighted bins (green) show sections of the trace containing stepwise jumps between the two levels. Figure 4(b) is the conductance histogram of the entire trace in Fig. 4(a). It shows that (as expected) the conductance either lies in Level-1 or in Level-2, with no counts in between. Note that the finite spread in conductance of the two histogram peaks in Fig. 4(b) arises from the finite standard deviation

FIG. 4. (Color online) (a) A conductance trace consisting of RTN between two different atomic configurations. The inset shows a zoom-in view of the two levels in RTN. The dotted lines in the inset show the bins; bins that include stepwise transition are highlighted in green. (b) Conductance histogram of the trace shown in (a). (c) Standard deviation of all the bins from the trace in (a). The data points within the red and blue ellipses in (c) correspond to the true standard deviation of Level-1 and Level-2, respectively. All other data points in (c) are artifacts arising from bins that contain the stepwise change between Level-1 and Level-2.

tion (SD) of conductance for each level. Figure 4(c) is the SD of *all the bins* from the trace in Fig. 4(a), including those bins that contain a stepwise change in conductance. However, *only* the data points within the red ellipse (on the left) and blue ellipse (on the right) in Fig. 4(c) correspond to the true SD of Level-1 and Level-2, respectively. All other data points in Fig. 4(c) arise from bins that contain stepwise changes between Level-1 and Level-2. Inclusion of these bins not only gives rise to spurious SD data points (forming the arch) at conductance values that do not exist in the histogram in Fig. 4(b) but also gives rise to spuriously high SD for either level (for example, points enclosed in the dotted rectangle). Similarly, in experiments where conductance transitions between integer multiples of G_0 [for example, RTN in Fig. 3(a) between $4G_0$ and $5G_0$], artificially high SD would appear at noninteger values of G_0 . This is the reason why all stepwise changes in conductance have to be first eliminated in noise analysis, however laborious. (This type of artifact in SD can be easily demonstrated by binning a square wave from a function generator).

Figure 5(a) shows a plot of SD versus conductance where every bin containing a stepwise change in conductance was

FIG. 5. (Color online) (a) A dust map showing true standard deviation versus conductance that excludes any stepwise transitions in the recorded traces. The dust map consists of over 61 000 data points from 400 traces. Each data point was calculated from 1000 conductance data points. Only plateaus that lasted for >200 ms without discrete steps were used. The inset shows an intensity plot for the dust map to emphasize the nonuniform density of data points. The retraction speed for individual traces is 3.5 pm/s. (b) Zoom-in view of the dust map in the vicinity of $1G_0$. The red curve in (b) is the average standard deviation of the dust map whereas the green curve roughly outlines the lower bound of the standard deviation.

manually eliminated from the data. The striking feature of the “dust map” in Fig. 5(a) is the *quasicontinuous distribution* of conductance levels, corresponding to a multitude of atomic configurations. Moreover, each of these finely spaced conductance levels have a wide range of possible SD values (vertical axis). The inset in Fig. 5(a) is a color intensity plot corresponding to the dust map. It emphasizes the existence of a high density of conductance levels in the vicinity of integer multiples of G_0 . Figure 5(b) is a zoom-in view of the dust map in the vicinity of $1G_0$; the red curve in Fig. 5(b) is the average SD. Note that even with thousands of data points

FIG. 6. (Color online) (a) Average standard deviation of the conductance juxtaposed with (b) the histogram shown in Fig. 1 with 5000 traces. For (a), average SD was computed by binning 500 consecutive conductance values from the dust map. The position of the local minima did not change by slightly changing the bin size.

in the dust map, there is a distinct lower bound for the SD [green curve in Fig. 5(b)]. Figure 5(b) reveals the existence of numerous atomic configurations both above and below $1G_0$. Levels with net conductance just above $1G_0$, which correspond to constrictions with more than one atom, can have SD comparable to the SD of conductance traces at or even below $1G_0$. At the same time, levels with net conductance at or below $1G_0$ cannot necessarily be assumed to be arising from a single-atom constriction. Although Fig. 5 reveals the existence of numerous levels at a far higher resolution than a conventional histogram, neither provides any information on the number of atoms in each configuration. However, the value of the dust map lies in understanding the origin of peaks in a conductance histogram, as explained in the following.

Figures 6(a) and 6(b) juxtaposes, respectively, the average value of the SD [obtained from the dust map in Fig. 5(a)] with a histogram shown earlier in Fig. 1. The juxtaposition shows that the peaks in the histogram generally correspond to the minima of the SD plot. Similar correspondence has been previously reported for shot noise,^{51,52} and the SD of the voltage dependence of conductance.⁷⁴ However, these previous studies did not reveal the presence of numerous local minima, such as those shown in Fig. 6(a). Moreover, no peaks can be seen corresponding to these local minima in the histograms. As explained in the following, the smearing of these local peaks in the histograms results from (1) a finite SD for each conductance plateau, (2) a high density of plateaus that are narrowly separated in terms of their net con-

ductance, and (3) a finite strain dependence of conductance for each plateau. First consider the simple case of a two-level fluctuation shown earlier in Fig. 4(a). The nonzero SD of conductance at each level results in a finite width for its corresponding histogram peak, as shown in Fig. 4(b). In this example, the spread in conductance values for each level does not overlap since their average conductance values are sufficiently far apart. However, as seen from the dust map, there is a high density of finely spaced levels, each with a finite SD in conductance. When individual histograms of all these levels are compiled, the finite spread in conductance for each level overlaps, causing the local peaks to smear out. In addition, each level also has a finite strain dependence of conductance (as shown in the following section), which further smoothens these local peaks.

The dust map further explains the origin of the remaining peaks in the histogram. They simply arise from a nonuniform density of observed conductance levels; again recall that the use of the term level and its origins refer to discrete conductance values taken by various atomic configurations and should not be confused with quantum-mechanical levels. As seen from the intensity plot in the inset of Fig. 5(a), the highest density of levels occur in the vicinity of $1G_0$, with high densities also observed just below $2G_0$ and $3G_0$ (and fewer levels at intermediate values). This naturally causes more counts to be accumulated in regions with a higher density of conductance levels, causing the fortuitous appearance of amalgamated, monolithic peaks in a histogram. *They are not a result of any particularly stable atomic configuration, nor can they be used to identify the number of atoms in the constriction.* Moreover, the height and location of the peaks can be easily changed by altering the experimental conditions (see the following section).

C. Channel saturation and conductance quantization

The conductance of a single-atom gold constriction should saturate at $1G_0$ under a perturbation and its demonstration would constitute an unambiguous and direct signature of conductance quantization. In this regard, let us consider the effect of a mechanical perturbation on the observed conductance. Here the perturbation is in the form of a small mechanical oscillation of subatomic or atomic-scale amplitude that is superimposed on a retracting/approaching gold tip against a gold substrate, as shown schematically in the inset of Fig. 7(a). Histograms were built at different perturbation amplitudes (peak-to-peak), and are shown in Fig. 7(a); Fig. 7(b) shows a zoom-in view of the amplitude dependence of the peak around $1G_0$. For reference, the topmost curve in Fig. 7 is the histogram without any superimposed mechanical oscillation (from Fig. 1). Figure 7 shows that with increasing amplitude of oscillation, counts in each peak decrease and the peaks themselves shift to higher conductance values. For example, Fig. 7(b) shows that with progressively higher oscillation amplitudes, an increasing percentage of the first peak lies above $1G_0$. In other words, the first amalgamated peak has a progressively decreasing contribution from true single-atom constrictions.

The application of aforesaid mechanical perturbation enables us to separate counts arising from true single-atom con-

FIG. 7. (Color online) (a) Histograms built by superimposing various peak-to-peak amplitudes of mechanical oscillation on the retracting gold tip. Each histogram is built using 1000 traces. The bin size of the conductance histograms is $0.02G_0$. The inset shows the profile of the oscillations superimposed on the retracting tip. Retraction rate is 3.5 pm/s. (b) Zoom-in view of the first peak as a function of oscillation amplitude. In (a) and (b) count percentage (ordinate) is obtained by normalizing the counts between $0.5G_0$ and $10G_0$ to 100%. The zero-amplitude peak is the same trace as the one shown in Fig. 1 for 1000 traces. Thus for example, the counts at the first peak height corresponding to count percentage of ~ 0.0075 at zero amplitude are equal to 0.05×10^6 . Therefore, counts at 0.1 nm amplitude equals $0.05 \times 10^6 \times 0.0045/0.0075 = 0.03 \times 10^6$. The tunneling data below $0.5G_0$ was specifically left out so it did not distort the peak normalization.

strictions. First consider Fig. 8(a), which shows a prototypical example (representing the behavior of thousands of similarly behaving contacts) of a trace arising from a constriction whose net conductance is close to $1G_0$. Figure 8(a) shows that the *conductance changes continuously and reversibly below and above $1G_0$* as the constriction is elongated and retracted. Such traces cannot be attributed to a single-atom contact. In sharp contrast, the characteristics of a one-atom constriction under mechanical perturbation are seen in Fig. 8(b). It shows a conductance trace of a true single-atom contact that has a partially closed channel, which can be fully opened under mechanical perturbation to saturate at $1G_0$. *This is the hallmark of conductance quantization, viz., saturation at $1G_0$* due to the availability of only a single channel for conductance across a single gold atom. As shown in Fig. 8(b), the process of opening the conductance channel to saturation is reversible over repeated piezo cycling. Figure 8(c) emphasizes the reversible change in conductance with repeated cycling of the contact corresponding to Fig. 8(a) whereas Fig. 8(d) shows the same for the contact corresponding to Fig. 8(b). Although both show reversibility under perturbation, only a true single-atom contact shows conductance saturation at $1G_0$.

It is clear from above that these perturbation studies allow us to identify true single-atom contacts. Each trace used to build the histograms at various amplitudes in Fig. 7 was

FIG. 8. (Color online) (a) Continuous and reversible change in conductance below and above $1G_o$ as the piezo elongates and retracts for a contact whose net conductance is close to $1G_o$. (b) Saturation at $1G_o$ due to the availability of only a single channel for conductance across a single gold atom as the hallmark of conductance quantization. (c) and (d) Change in conductance versus elongation/retraction corresponding to (a) and (b), respectively. For (c) six contiguous cycles are plotted whereas for (d) three contiguous cycles are plotted.

scrutinized and the histograms were rebuilt after sorting for their size. The sorting criterion used in rebuilding the histograms is based on conductance. For example, contacts that saturate at $1G_o$ are true one-atom contacts whereas those that go continuously above $1G_o$ are not. Similarly, contacts whose conductance rises continuously above $2G_o$ are not two-atom contacts, etc. The perturbation approach allows us to unambiguously ascertain whether a constriction in the immediate vicinity of NG_o is due to N atoms or greater than N atoms. The resolved histograms for true one-atom contacts (blue traces) are shown in Figs. 9(a)–9(c) at three different amplitudes (0.1, 0.2, 0.3 nm peak-to-peak, respectively). In Figs. 9(a)–9(c), the red trace represents counts in the vicinity of $1G_o$ arising from two-atom contacts. It highlights the fact

FIG. 9. (Color online) Conductance histograms for true one-atom contacts (blue trace) at three different amplitudes (a) 0.1 nm, (b) 0.2 nm, (c) 0.3 nm peak-to-peak. For each amplitude 1000 traces were used. The red trace corresponds to two-atom histogram in the vicinity of $1G_o$. The dotted trace is the cumulative total of all counts in the histogram. In (a) and (c), all data were normalized to the maximum height for 0.1 nm amplitude peak, for which from Fig. 7, the number of counts is 0.03×10^6 . (d) Split conductance histograms to higher G_o values. For (d), counts (arbitrary units), all data were normalized to the height of the one-atom peak (blue trace). That is the peak height for the blue trace in (b). While it is possible to unambiguously ascertain counts in the vicinity of $1G_o$ from true one-atom contacts (blue traces), the tail of the trace from two-atom contacts (red traces) in the vicinity of $1G_o$ may have some contribution from, say, a three-atom configuration having a net conductance much less than $2G_o$; similar can be said for higher conductance values.

that a finite number of counts exist below $1G_o$ which do not arise from true single-atom contacts. The dotted (black) line is the summation of all counts, just as in a conventional histogram. Notice the theoretically expected abrupt drop in counts in histograms for true single-atom contacts (blue traces) when the conductance value exceeds $1G_o$ (the slight

FIG. 10. (Color online) (a) A profile of the tip oscillation for mechanical susceptibility ($\Delta G/\Delta d$) measurements of the contacts. (b) Example of corresponding conductance response to oscillation profile in (a). The subscripts “C” and “T” represent contact in compression (piezo elongation) and tension (piezo retraction), respectively.

overshoot in counts above $1G_o$ for one-atom contacts is due to the allowance for noise in the conductance signal). Figure 9(d) shows similar behavior for the entire resolved histogram after size sorting to higher G_o values. Figures 9(a)–9(c) shows that at higher amplitudes (≥ 0.3 nm), the counts to the first peak from single-atom contacts become negligible. In other words, a true single-atom contact rarely forms at high amplitudes. This explains the observed shift of peaks in Fig. 7(b) to higher conductance values.

Finally, consider the mechanical susceptibility of the contacts (defined as $\Delta G/\Delta d$; see Fig. 10). For these measurements, the profile of the distance modulation is shown in Fig. 10(a). In subsequent analysis, the sections of the profile with zero piezo elongation/retraction [see inset in Fig. 10(a)] help ensure that the conductance of selected traces had reversibly returned back to its initial state after each excitation cycle. It also enables an unambiguous determination of ΔG , as shown in Fig. 10(b). Using this profile of mechanical perturbation, Fig. 11(a) shows another prototypical example of a true single-atom contact with near perfect channel transmission. It again highlights channel saturation at $1G_o$ owing to the availability of only a single conductance channel across a gold atom. Figure 11(b) shows that ΔG for this contact is virtually unchanged, and represents the limiting case of $\Delta G/\Delta d$ approaching zero. Figure 11(c) shows a prototypical example of a contact whose conductance changes continuously across $1G_o$ without channel saturation. In contrast to a true single-atom contact, a noticeable change in conductance

FIG. 11. (Color online) (a) A prototypical example of a single-atom contact with near perfect transmission, highlighting saturation at $1G_o$. (b) Corresponding conductance versus tip oscillation trace. (c) A prototypical example of a contact with net conductance close to $1G_o$. The trace reveals that this contact is not a true single-atom constriction. (d) Corresponding conductance versus tip oscillation trace.

occurs with piezo elongation (or retraction), as shown in Fig. 11(d). A plot of all measured values of $\Delta G/\Delta d$ in the vicinity of $1G_o$ is shown in Fig. 12(a); the inset shows the SD of $\Delta G/\Delta d$ for single-atom contacts. A salient feature of this plot is the asymptotic drop in mechanical susceptibility to zero for true single-atom contacts, and this may be taken as a manifestation of the observed channel saturation; Fig. 12(b) shows similar behavior for larger sized contacts. The asymptotic drops in $\Delta G/\Delta d$ to zero near integer multiples of G_o shows that for atomic configurations with near perfect channel transmission, the applied tension or compression is

FIG. 12. (Color online) (a) A plot of mechanical susceptibility ($\Delta G/\Delta d$) versus conductance in the vicinity of $1G_0$ (blue solid dots for true one-atom contacts and red open dots for two-atom contacts) using 1000 traces from each of the following amplitudes: 0.1 nm, 0.15 nm, 0.2 nm, 0.25 nm, and 0.3 nm peak-to-peak. Inset shows standard deviation of the data for single-atom contacts. Note the asymptotic drop in $\Delta G/\Delta d$ to zero for single-atom contacts, and a large jump in mechanical susceptibility for two-atom contacts. (b) Similar behavior for larger sized contacts. Total number of measurements is ~ 2000 .

mostly accommodated by regions away from the constriction. This has to be the case as Au-Au bonds obviously cannot stretch to the full extent of the applied amplitude of

perturbation (up to 0.3 nm peak-to-peak). The picture that emerges is that the narrowest region of a constriction with a near perfect transmission is also structurally more ordered and less susceptible to deformation compared to (disordered) regions away from it. Transmission electron microscopy that combines *in situ* conductance measurements^{62,75,76} is needed to investigate the distribution of deformation in these contacts, and is beyond the scope of the present study.

IV. CONCLUSIONS

Using mechanical perturbations, the present study shows the hallmark of conductance quantization for a gold single-atom constriction, namely, saturation of conductance to $1G_0$ due to the availability of only a single channel across a single gold atom. The asymptotic drop in mechanical susceptibility to zero is a manifestation of channel saturation.

The dust map reveals a quasicontinuous distribution of conductance levels corresponding to numerous atomic configurations. Local peaks due to these configurations are smeared off in a conventional histogram due to a finite SD for each conductance level, a high density of levels that are narrowly separated in terms of their net conductance, and a finite strain dependence of conductance for each level.

Additionally, the dust map helps to explain the origin of peaks in a conventional histogram. They are shown to arise from a nonuniform density of observed conductance levels. This results in more counts being accumulated in regions with a higher density of conductance levels, leading to the fortuitous appearance of amalgamated, monolithic peaks in a conventional histogram. The position of these amalgamated peaks can be shifted by changing the mechanical perturbation conditions. Peaks are not a result of any particularly stable atomic configuration, nor can they be used to identify the number of atoms in the constriction.

The mechanical perturbation approach offers a way to identify the number of atoms in a given constriction.

The monotony of scrutinizing the characteristics of each individual trace cannot be substituted by a nondiscriminate statistics on a data set, however large.

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