Quantum interference structures in the conductance plateaus of gold nanojunctions

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The conductance of breaking metallic nanojunctions shows plateaus alternated with sudden jumps, corresponding to the stretching of stable atomic configurations and atomic rearrangements, respectively. We investigate the structure of the conductance plateaus both by measuring the voltage dependence of the plateaus' slope on individual junctions and by a detailed statistical analysis on a large amount of contacts. Though the atomic discreteness of the junction plays a fundamental role in the evolution of the conductance, we find that the fine structure of the conductance plateaus is determined by quantum interference phenomenon to a great extent.

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The quantum nature of conductance is also reflected by quantum interference phenomenon to a great extent.

In both cases the contact has a single conductance channel with almost perfect transmission. Theoretical studies have pointed out that in gold the conductance of a monoatomic contact is not sensitive to the amount of stretching, which could explain the flatness of the last conductance plateau. In the experiments, however, the conductance plateaus always show a fine structure, which are different during each rupture (for examples see Ref. 6 and the inset in Fig. 1). This feature could be naturally explained by the atomic discreteness of the junction: as the electrodes are pulled apart the overlap between the central atoms changes, which alters the conductance of the contact. In this paper we show that this interpretation is not satisfactory, and the fine structure of the conductance plateaus is strongly affected by quantum interference phenomenon.

The basic idea behind quantum interference in atomic-sized junctions is illustrated in Fig. 1. The narrow neighborhood of the contact center can be considered as a ballistic region with a transmission probability, $T_0$. The

![Fig. 1: Representative conductance trace recorded during the break of a gold nanojunction. The inset shows a segment of the last conductance plateau demonstrating the fine structure of the conductance traces.](image-url)
electron wave that has travelled through the contact can be partially reflected by impurities or lattice defects farther away in the diffusive electrodes. This reflected wave goes back to the contact, and a part of it is reflected back again by the contact itself. This part of the wave interferes with the direct wave, modifying the conductance of the junction. The net transmission including the interference corrections can be written as:

\[ T(z, V) = T_0(z) \left[ 1 + \sum_j A_j \cos \left( \left( k_F + \frac{eV}{\hbar v_F} \right) L_j + \Phi_j \right) \right]. \]  

The total transmission is a function of the electrode separation, \( z \) and the bias voltage, \( V \). The bare transmission of the contact, \( T_0 \) is controlled by the shape of the junction and the overlap between the atomic orbitals, and accordingly it is dependent on the electrode separation, \( z \). It was shown that in the voltage scale of the measurement the voltage dependence of \( T_0 \) can be neglected. \(^7\) In the interference correction the sum runs over the various electron trajectories; \( L_j \) and \( \Phi_j \) are respectively the path length and the phase shift on a trajectory; and \( k_F \) is the Fermi wave number. The amplitude \( A_j \) is determined by the scattering cross section of the defect, the length of the path, and the reflection of the contact. The differential conductance of the system is obtained from the transmission as \( G(z, V) = G_0 \cdot T(z, V) \). (For the sake of simplicity, a single conductance channel is considered. The argumentation would be similar for multiple channels as well.)

Quantum interference results in fluctuations in the conductance when the interference conditions are tuned experimentally. If the wave number of the electrons is changed by the bias voltage, QI shows up as a small, random oscillation in the \( G(V) \) curve. The interference pattern can also be changed by tuning the phase factor of the electron paths with magnetic field. In atomic-sized contacts, however, a magnetic field of \( \geq 60 \text{ T} \) would be required to have a considerable influence on the interference, while a field of \( 1 \text{ T} \) already causes changes in the atomic arrangement of the contact due to magnetostriction effects. \(^8\) Here, we focus our attention on quantum interference due to the variation of the length of the electron paths. In nanojunctions the path length naturally changes with the separation of the electrodes. To have a complete period in the interference pattern the electrode separation should be changed by one wavelength of the electrons. Experimentally, such a displacement is not possible without a jump-like atomic rearrangement, which abruptly changes the interference pattern. From this reason, only shorter parts of the conductance plateaus can be studied, like that in the inset of Fig. 4. The fine structure of these short segments can originate both from the QI phenomenon and from the electrode separation dependence of the bare transmission, \( T_0(z) \). In the following we show experimental techniques, that can tell “to what extent these two phenomena are involved in the evolution of the plateaus”. To investigate the fine structure of the conductance traces, we have studied the local slope of the plateaus by two different methods.

The first approach examines the effect of bias voltage on the plateaus’ slope on individual junctions. Figure 3 shows the current (panel a), and the derivative of the current with respect to the electrode separation (panel c) recorded as a function of the bias voltage. The two curves were measured simultaneously on the same junction. The electrode separation was modulated by applying a sine-wave voltage on the piezo element. The oscillation of the separation had a typical amplitude of \( 0.1 \text{ Å} \). As the bias voltage was varied, the current was detected both by a current meter measuring the DC component and a lock-in amplifier recording the response to the modulation. The signal of the current meter provided the \( I(V) \) curve, whereas the lock-in measured the value of \( \partial I/\partial z \). The differential conductance, \( G(V) \) and the slope of the plateau, \( \partial G/\partial z \) was determined by numerical differentiation (Fig. 3b and 3d, respectively). These curves are reproducible to the very small details as long as the same...
contact is measured. When the junction is changed a completely new structures appear in the curves, as expected from QI phenomenon.

Assume that the dependence of the differential conductance on the electrode separation, \( z \) is attributed solely to the bare transmission \( T_0(z) \). In this case the slope of the conductance plateau can be written as:

\[
\frac{\partial G(z,V)}{\partial z} = \frac{1}{T_0(z)} \frac{\partial T_0(z)}{\partial z} G(z,V),
\]

i.e. the voltage dependence of \( \partial G/\partial z \) is simply proportional to \( G(V) \). This, however is disproved by the experimental results shown above. The oscillatory patterns of the \( G(V) \) curve and the \( \partial G(V)/\partial z \) curve in Fig. 4 do not coincide. Furthermore, in the \( G(V) \) curve the oscillations have a typical amplitude of 10% compared to the mean value of \( G = 0.96 \, G_0 \), while in the \( \partial G(V)/\partial z \) curve the relative amplitude of the oscillations is more than 10 times larger.

These observations can only be explained, if the change of the path lengths \( L_j \to L_j + dz \) is also taken into account as the electrode separation is varied by \( dz \). Then, the derivative of the transmission with respect to \( z \) is written as:

\[
\frac{\partial T(z,V)}{\partial z} \approx \frac{\partial T_0(z)}{\partial z} - T_0(z) \sum_j k_F A_j \sin \left( k_F + \frac{eV}{\hbar v_F} \right) L_j + \Phi_j.
\]

Based on this formula, \( \partial T_0/\partial z \) is well approximated with the mean value of the \( \partial G(V)/\partial z \) curve, which is \( \approx -0.023 \, \text{Å}^{-1} \). The amplitude of the interference correction is characterized by the standard deviation: \( \approx 0.022 \, \text{Å}^{-1} \). It shows, that the variation of the plateau’s slope due to QI is comparable to the separation dependence of the bare transmission. The comparison of the formulas (1) and (3) shows, that the amplitude of the oscillatory term changes by a factor of \( k_F \), while the constant term changes by \( \partial T_0/\partial z \) due to the differentiation. According to measurements on several contacts, \( \partial T_0/\partial z \) is typically below 0.05 Å\(^{-1}\), which is smaller by an order of a magnitude than \( k_F \approx 0.6 \, \text{Å}^{-1} \). This explains, that the contribution of QI is highly enhanced in the \( \partial G/\partial z \) curves, while in the \( G(V) \) curve it only gives a minor correction.

The above measurements were performed on individual contacts. In the following we present a second approach, investigating the statistical properties of the slope of the conductance plateaus. Independent atomic configurations with different set of the interference parameters \( (A_j, L_j \text{ and } \Phi_j) \) can be naturally created by repeating the break of the junction several times. The data set for the statistical analysis was obtained by recording \( \approx 15000 \) independent conductance vs. electrode separation traces at fixed bias voltage. The typical acquisition rate was 50 points/Å. The slope of the plateaus was determined by numerical differentiation. The derivative was calculated at each point of the conductance plateaus, however the jump-like changes between two plateaus – corresponding to sudden atomic rearrangements – were excluded from the analysis.

In the mean value of \( \partial G/\partial z \) the interference corrections cancel out due to their random distribution around zero, thus the average slope of the plateaus is only determined by the bare transmission:

\[
\langle \frac{\partial G}{\partial z} \rangle = G_0 \left( \frac{\partial T_0}{\partial z} \right).
\]

The proper quantity to study QI is rather the mean square deviation of \( \partial G/\partial z \), which contains the interference term beside the properties of the bare contact (see Eq. (3)):

\[
\frac{\sigma^2_{\partial G/\partial z}}{G_0^2} = \sigma^2_{\partial T_0/\partial z} + \frac{1}{2} \frac{T_0^2 k_F^2}{T_0} \sum_j \langle A_j^2 \rangle.
\]

The squared amplitude, \( A_j^2 \) is proportional to the probability that an electron is reflected back by the contact, \( R_0 = 1 - T_0 \). Therefore, the interference term in the mean square deviation vanishes both at \( T_0 = 1 \) and \( T_0 = 0 \).

Gold junctions with a few atoms (\( \leq 4 \)) in the cross section show the saturation of the channel transmissions, which means that a new channel only starts to open, if the previous ones are almost completely open. Due to this behavior at the quantized conductance values all transmission probabilities are close to unity or zero, thus the quantum interference is suppressed. If QI gives a detectable contribution to the slope of the plateaus, the \( \sigma^2_{\partial G/\partial z} (G) \) curves should also exhibit the quantum suppression at the multiples of \( G_0 \). This phenomenon is clearly resolved in our experiments: the mean square deviation of the plateaus’ slope exhibit pronounced minima accurately placed at 1, 2, and 3 \( G_0 \) (Fig. 4c). In contrast, the second and the third peak in the conductance histogram are significantly shifted from the integer values (Fig. 4d). It demonstrates, that the minima in \( \sigma^2_{\partial G/\partial z} \) are a consequence of a pure quantum phenomenon, and they are not related to the preferred atomic configurations shown by the peaks in the histogram.

The suppression of QI at the quantized values gives a possibility to estimate the contribution of the quantum interference term to the slope of the plateaus. According to Ref. 4 the magnitude of the quantum suppression is almost 100% at 1 \( G_0 \), while at higher quantized values it is decreasing. Therefore, we attribute the nonzero minimum value of \( \sigma^2_{\partial G/\partial z} \) at 1 \( G_0 \) purely to the scattering of the bare properties, \( \sigma^2_{\partial T_0/\partial z} \). The interference term in Eq. (3) \( \sigma^2_{QI} \) is approximated by subtracting \( \sigma^2_{\partial T_0/\partial z} \) which is considered as a constant background. The relative amplitude of QI in the slope of the plateaus can be characterized by the quantity
\[ \eta_{QI} = G_0 \cdot \sigma_{QI} / \sqrt{\langle \partial G / \partial z \rangle^2 + \sigma^2_{\partial G / \partial z}}. \]

This formula already treats a multichannel situation, where \( T_n \) is the transmission of the \( n \)-th channel, \( \gamma \) is the opening angle of the contact, and \( l_e \) is the elastic mean free path of the electrons. From the measured amplitude of \( \sigma^2_{QI} \) the elastic mean free path is estimated as \( \sim 5 \text{ nm} \), which is in good agreement with previous results.\(^{11}\)

Concluding, we have investigated the structure of the conductance plateaus in gold nanocontacts. We have studied the voltage dependence of the slope of the conductance plateaus on individual junctions. The \( \partial G(V) / \partial z \) curves have shown a strong oscillatory deviation from the mean value, which is an order of a magnitude larger than the conductance fluctuations in the \( G(V) \) characteristics. This feature could only be described by quantum interference due to the spatial modulation of the interference paths. In order to support these results we have performed a statistical analysis of the plateaus' slope for a large amount of junctions. The quantum suppression of \( \sigma^2_{\partial G / \partial z} \) at the quantized conductance values have provided an even stronger proof for the significant presence of QI. With our analysis the contributions of quantum interference and the strain dependence of the local atomic configuration to the plateaus' slope could be separated. The results have shown that the quantum interference phenomenon and the atomic discreteness of the junction have a similarly strong influence on the fine structure of the conductance plateaus.

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In the experiments the applied voltage is much smaller than the Fermi energy, so the term $eV/hv_F$ was neglected beside $k_F$. The term $\partial T_0/\partial z \sum_j A_j \cos(...) \cos$ was also neglected being an order of a magnitude smaller than $\partial T_0/\partial z$.

If $\sigma^2 \partial T_0/\partial z$ is assumed to be constant in the whole conductance range, the relative amplitudes of the minima in $\sigma_{QI}$ are similar to those in Ref. 4. This agreement supports our assumption. As a further verification, we have also performed measurements on polyvalent metals, for which no suppression of QI occurs at the quantized values, thus any special structure in the $G$ dependence of $\sigma_{QI}/\partial z$ should come from the bare properties. In these measurements the and variation of $\sigma^2 \partial T_0/\partial z$ was found to smaller than $\sim 15\%$ of the total signal.