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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. 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 investigation of the mechanical and electrical properties of atomic-sized metallic junctions has recently become an interesting topic of nanoscience (for a review see Ref. 1). A contact with a single atom in the cross section can be created by pulling a nanowire with a scanning tunneling microscope (STM) or the mechanically controllable break junction (MCBJ) technique. In such nanocontacts the coherent quantum phenomena always interplay with the atomic granularity of matter, as the wavelength of the electrons and the interatomic distance are in the same order of magnitude. The atomic nature of the junction is clearly demonstrated by the evolution of the conductance during the break of the contact showing plateaus alternated with sudden jumps (Fig. 1). Force measurements have shown that the conductance plateaus correspond to the stretching of stable atomic configurations, whereas the conductance jumps are related to atomic rearrangements.[2] On the other hand, the statistical analysis of a large amount of conductance vs electrode separation traces has shown signs of conductance quantization in metals with loosely bound *s* electrons.[3] The quantum nature of conductance is also reflected by the quantum interference (QI) phenomenon of the electron waves scattered on nearby impurities, which was reported in Refs. 4 and 9. These works investigated the interference patterns in the voltage dependence of the conductance. In this paper we demonstrate that QI has a definite influence on the structure of the conductance plateaus as well, which arises due to the spatial variation of the electron paths during the stretching of the junction.

The measurements were performed on high-purity gold samples at liquid-helium temperature with the MCBJ technique.[1] The conductance histogram of Au shows a sharp peak at the quantum conductance unit, $G_0=2e^2/h$. This peak arises from the frequent occurrence of plateaus that are accurately positioned at $1G_0$, as shown in Fig. 1. It was found that these plateaus are related to the conductance through a single gold atom[1] or through a chain of gold atoms in a row.[5] In both cases the contact has a single conductance channel with almost perfect transmission.[10,11] 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.[6] In the experiments, however, the conductance plateaus always show a fine structure, which are different during each rupture (for examples, see Ref. 9 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. 2. The narrow neighborhood of the contact center can be considered as a ballistic region with a transmission probability T_0. The electron wave that has traveled 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

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

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.[7,8] In the interference correction the sum runs over the various electron trajectories; L_j and Φ_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

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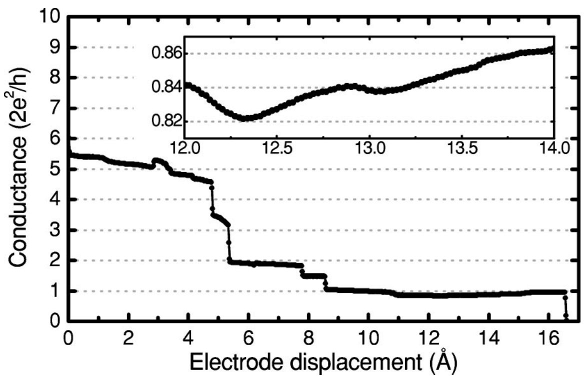

FIG. 1. Representative conductance trace recorded during the break of a gold nanojunction. The inset shows a segment of the last conductance plateau demonstrating the fine structure of the conductance traces.

differential conductance of the system is obtained from the transmission as $G(z,V)=G_0T(z,V)$. (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.[4,9] 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 $\gtrsim 60$ T would be required to have a considerable influence on the interference, while a field of 1 T already causes changes in the atomic arrangement of the contact due to magnetostriction effects.[12] 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 jumplike atomic rearrangement, which abruptly changes the interference pattern. From this reason, only shorter parts of the conductance plateaus can be studied, such as that in the inset of Fig. 1. 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

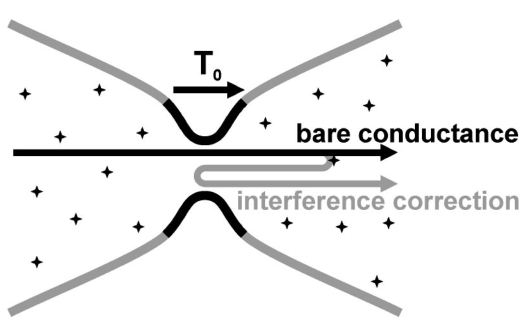

FIG. 2. Illustration for the quantum interference effect in nanojunctions, following the model in Ref. 4.

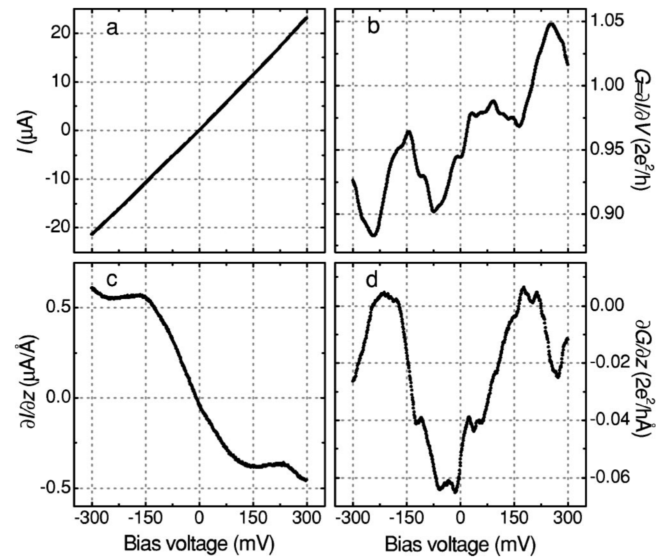

FIG. 3. The $I(V)$ curve (a), the $G(V)=\partial I/\partial V$ curve (b), the $\partial I(V)/\partial z$ curve (c), and the $\partial G(V)/\partial z$ curve (d) recorded on the same single-atom gold junction.

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 piezoelement. The oscillation of the separation had a typical amplitude of 0.1 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 [Figs. 3(b) and 3(d), respectively]. These curves are reproducible to the very small details as long as the same contact is measured. When the junction is changed 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

$$\frac{\partial G(z,V)}{\partial z}=\frac{1}{T_0(z)}\frac{\partial T_0(z)}{\partial z}G(z,V), \tag{2}$$

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. 3 do not coincide.

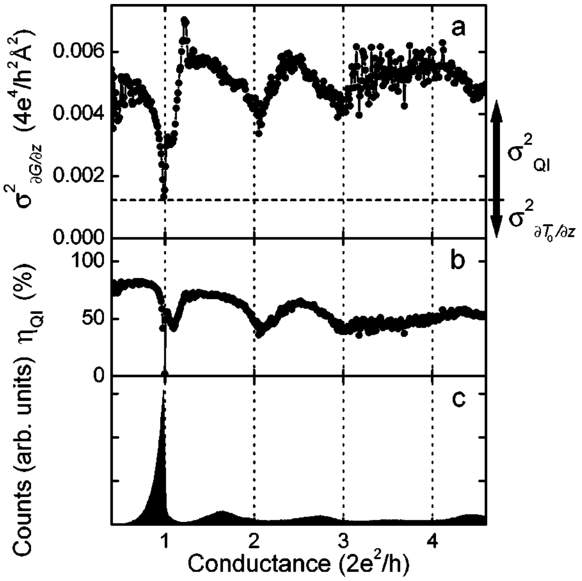

FIG. 4. Panel (a) shows the standard deviation of the plateau slope $\partial G/\partial z$ as the function of the conductance. The arrows indicate the separation of the QI term from $\sigma^2_{\partial T_0/\partial z}$. Panel (b) presents the relative contribution of QI to the plateaus' slope. In panel (c) the conductance histogram is presented for the same data set.

Furthermore, in the $G(V)$ curve the oscillations have a typical amplitude of 10% compared to the mean value of $G=0.96G_0$, while in the $\partial G(V)/\partial z$ curve the relative amplitude of the oscillations is more than ten times larger.

These observations can only be explained, if the change of the path lengths $L_j \rightarrow L_j + dz$ 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[13]

$$\frac{\partial T(z,V)}{\partial z} \simeq \frac{\partial T_0(z)}{\partial z} - T_0(z) \sum_j k_F A_j \times \sin\left\{ \left(k_F + \frac{eV}{\hbar v_F} \right) L_j + \Phi_j \right\}. \tag{3}$$

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 $\simeq -0.023$ Å^{-1}. The amplitude of the interference correction is characterized by the standard deviation $\simeq 0.022$ 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 $(\partial T_0/\partial z)/T_0$ 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 $k_F \simeq 0.6$ 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, and Φ_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 $\sim$15 000 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 jumplike 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

$$\left\langle \frac{\partial G}{\partial z} \right\rangle = G_0 \left\langle \frac{\partial T_0}{\partial z} \right\rangle. \tag{4}$$

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)]:

$$\frac{\sigma^2_{\partial G/\partial z}}{G_0^2} = \sigma^2_{\partial T_0/\partial z} + \underbrace{\frac{1}{2} T_0^2 k_F^2 \sum_j \langle A_j^2 \rangle}_{\sigma^2_{QI}}. \tag{5}$$

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 ($\leqslant 4$) 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.[4] 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 $1G_0$, $2G_0$, and $3G_0$ [Fig. 4(a)]. In contrast, the second and the third peaks in the conductance histogram are significantly shifted from the integer values [Fig. 4(c)]. 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 $1G_0$, while at higher quantized values it is decreasing. Therefore, we attribute the nonzero minimum value of $\sigma^2_{\partial G/\partial z}$ at $1G_0$ purely to the scattering of the bare properties, $\sigma^2_{\partial T_0/\partial z}$. The interference term in Eq. (5), σ^2_{QI}, is approximated by subtracting $\sigma^2_{\partial T_0/\partial z}$, which is considered as a constant background.[14] The relative amplitude of QI in the slope of the plateaus can be characterized by the quantity $\eta_{QI} = G_0\sigma_{QI}/\sqrt{\langle \partial G/\partial z\rangle^2 + \sigma^2_{\partial G/\partial z}}$. This curve takes values larger than 50% [Fig. 4(b)], which demonstrates that the influence of QI on the slope of the plateaus is dominating over the features due to the atomic arrangement of the bare contact.

For a more quantitative description of the observations we have performed a calculation following the model in Ref. 4. In the $V\rightarrow 0$ limit the standard deviation of the plateau's slope due to the QI terms can be written as

$$\sigma^2_{QI} = \frac{24}{\sqrt{\pi}(1-\cos\gamma)}\frac{1}{l_e^2}\sum_{n=1}^{N} T_n^2(1-T_n). \tag{6}$$

This formula already treats a multichannel situation, where T_n is the transmission of the nth channel, γ is the opening angle of the contact, and l_e is the elastic mean free path of the electrons. From the measured amplitude of σ^2_{QI} the elastic mean free path is estimated as ~ 5 nm, which is in good agreement with previous results.[4]

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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[14] 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 σ_{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 (Ref. 4), thus any special structure in the G dependence of $\sigma^2_{\partial G/\partial z}$ should come from the bare properties. In these measurements the variation of $\sigma^2_{\partial T_0/\partial z}$ was found to be smaller than $\sim$15% of the total signal.