Conductance of tip–surface and tip–atom junctions on Au(111) explored by a scanning tunnelling microscope

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Abstract. The conductance between the tip of a scanning tunnelling microscope and a Au(111) surface is measured at tip–surface distances comprising tunnelling and contact regions. Contact between the tip and the flat sample surface as well as between the tip and an individual gold atom can be performed reproducibly without deteriorating the imaging capability of the instrument. Measurements performed on the face-centred cubic and hexagonal close-packed stacking domains of the Au(111) surface reconstruction lead to similar conductances of one quantum of conductance at contact.

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1. Introduction

Mechanical and electronic properties of molecules and atoms differ from those of macroscopic materials. The understanding, for instance, of the fundamentals of adhesion and friction, of photosynthesis and of signal transduction in molecular structures requires the knowledge of these properties. Moreover, electron transport through nanostructures may find applications in devices and is being investigated for semiconducting [1] and metallic [2, 3] constrictions, carbon nanotubes [4], single atoms and molecules [5]–[10].

Scanning tunnelling microscopy (STM) appears to be an ideal tool to study single atom or single molecule conductance in detail. The structure to be analysed can be imaged in direct space with atomic precision prior to and after taking conductance data. Thus, specific locations of the structure can be addressed and possible modifications can be easily detected. Another advantage of STM is the possibility to characterize to some extent the status of the second electrode, the microscope tip, by recording conductance data on clean metal areas. Consequently, STM can complement techniques like the mechanically controlled break junction.

Indeed, scanning probe techniques have been used to form point contacts between the tip and a metal surface whose quantized conductance was then investigated during forming and stretching of the contact [11]–[13]. A pioneering experiment by Joachim et al [9] reports on contacting an individual C₆₀ molecule with the tip of a scanning tunnelling microscope. Taking advantage of the imaging capability of STM recent experiments on individual adsorbed atom (adatom) [5]–[8] and on single molecule [10] contacts showed that these contacts can be formed reproducibly without structural changes of tip or sample.

In [6] contact between the tip of a scanning tunnelling microscope and Ag(111) and Cu(111) surfaces was studied. As a basic result, contact between the electrodes is established when chemical bonds between the surface and the tip apex start to weaken the adhesion of the atom to the tip structure. Upon further decrease of the tip–surface distance the conductance then in most of the cases changes—at the time resolution of the experiment (≈10⁻⁴ s)—discontinuously to a conductance of \( G \approx G_0 \) with \( G_0 = \frac{2 e^2}{h} \) (where \( e \) is the electron charge and \( h \) is Planck’s constant). The characteristics of contact to an individual atom, however, differ [6]. The transition from the tunnelling to the contact region upon decreasing the tip–atom distance occurs via a continuous conductance increase to \( \approx G_0 \) at the time resolution of data acquisition.

It is interesting to extend contact measurements with a scanning tunnelling microscope to different systems. The Au(111) surface is a particularly suitable candidate for several reasons. It exhibits a surface reconstruction, namely the \( 22 \times \sqrt{3} \) or herringbone reconstruction [14]–[16] which separates hexagonal-close packed (hcp) and face-centred cubic (fcc) stacking domains. Consequently, a natural platform is provided to test a possible influence of the second surface layer on the conductance. Further, in recent theoretical work forces between a tungsten tip and a Au(111) surface were calculated [17]. As a result, an onset of forces and relaxations was determined at tip–sample distances of 4–5 Å which were about 1 Å higher than earlier simulations for Cu(100) [18] and for Ni, Pt, Au single crystals [19]. The pertinent question arises as to when relaxations of tip and substrate material become visible in conductance measurements performed by a scanning tunnelling microscope. Another reason for choosing gold is that this material belongs to the archetypal metals used for contact measurements. The first results for Au(110) were presented by Brandbyge et al [20] who used a scanning tunnelling microscope. The tip was moved into contact with the substrate and conductance jumps were counted in a histogram. As a key result, integer multiples of \( G_0 \) were observed. Interestingly, using an atomic
force microscope Jarvis et al [21] reported evidence for half integer values of the conductance quantum when the gold-covered tip of the cantilever was brought into contact with a Au(111) surface. No explanation for this behaviour has been given so far. Finally, using standard low-temperature STM and mechanically controlled break junction techniques atomic-sized contacts were produced by moving the tip of the microscope out of the material or by breaking the electrodes apart, respectively. For gold it was found that the last conductance plateau (at a value of $G_0$) can often be stretched far beyond a length corresponding to an atomic diameter [22]. It is thus interesting to look at the formation of a diatomic wire when the tip approaches an individual gold atom on a gold surface.

The aim of this paper is to analyse conductance curves acquired on Au(111) and on individual gold atoms on this surface. Measurements were performed on hcp and fcc stacking domains of the $22 \times \sqrt{3}$ surface reconstruction. The transition from tunnelling to contact is signalled by a discontinuous change of the conductance. No significant influence of the stacking on the conductance was observed. Conductance-versus-displacement curves were likewise acquired on single gold atoms. We observed that the transition from tunnelling to contact was continuous within the time resolution of the experiment ($\approx 0.1$ ms).

2. Experiment

Measurements were performed with a custom-made scanning tunnelling microscope operated at 9 K and in ultrahigh vacuum with a base pressure of $10^{-9}$ Pa. The Au(111) surface and chemically etched tungsten tips were cleaned by argon ion bombardment and annealing. Controlled indentations of the tip into substrate material were repeated until adatoms appeared with nearly circular shape in constant-current images. As a consequence of this tip preparation technique the apex of the tungsten tip was most probably covered with several layers of gold. Single gold atoms were deposited on to the sample surface by controlled tip–surface contacts as previously described for silver and copper atoms in [6]. Prior to acquiring current-versus-displacement curves at a chosen surface location the tip–sample distance was fixed by opening the feedback loop of the microscope at a given current and sample voltage. Then the tip was approached towards the surface by $\approx 30 \text{ Å s}^{-1}$ and simultaneously the current was acquired. Typically a set of 20–50 conductance curves were recorded and averaged. This procedure was repeated for different orders of magnitude of the current. To this end the gain of the employed variable-gain transimpedance amplifier had to be changed. To rule out possible systematic errors in the presented conductance curves we ensured (i) that the relative gain error of the current-to-voltage converter is less than 1% for each applied gain and, (ii) that the integral nonlinearity for each gain is less than 0.01%. For conductance measurements tunnelling currents of the order of $10 \mu A$ are involved. Even at these elevated currents the voltage drop at the current-to-voltage converter is negligible. Conductances, $G = I/V$ (with $I$ the current and $V$ the sample voltage), can therefore be calculated using the sample voltage which is applied to the tunnelling and contact junction.

3. Results and discussion

In figure 1 we show a STM image of an area on Au(111) in order to introduce the two types of stacking domains, namely the fcc and hcp stacking domains, which are present on this surface.
Figure 1. Pseudo-three-dimensional constant-current STM image of an area on Au(111) showing discommensuration lines of the $22 \times \sqrt{3}$ surface reconstruction separating face-centered cubic fcc from hexagonal close-packed hcp stacking domains (size: 360 Å $\times$ 360 Å, sample voltage $V = -1.5$ V, current $I = 1.5$ nA). Image processing was performed by Nanotec WSxM [23].

These domains are separated from each other by discommensuration lines which appear as lines with higher corrugation in figure 1. Conductance measurements were performed with the tip placed above these domains in the middle of adjacent discommensuration lines.

Conductance curves acquired on the fcc domain are presented in figures 2(a) and (c). Displacement $\Delta z = 0$ marks the tip height when the feedback loop is opened at 200 mV and 50 pA. These representative curves exhibit three main characteristics. First, a wide range of displacements is observed where the conductance varies exponentially with the displacement (and thus appears as a linear curve on the logarithmic scale); secondly, a sudden change of the conductance occurs which at the time resolution of the experiment is detected as a discontinuous jump; thirdly, after the jump the conductance curves only weakly increase upon further tip approach. These subsequent regions are schematically illustrated in figure 2(b). The exponential behaviour is characteristic for the tunnelling region where $G \propto \exp(-1.025 \sqrt{\Phi} \Delta z)$ with $\Phi$ denoting the apparent barrier height (in eV) and $\Delta z$ the tip displacement (in Å). For the tunnelling region we obtain $\Phi \approx 4$ eV which is similar to the averaged tip and sample work functions. The discontinuous transition from the tunnelling region to a region where the conductance is nearly constant indicates contact between the tip and the surface via a single atom [6]. Within the tested voltage range of $-400$ mV to 400 mV no significant modifications of the conductance curves were observed. For instance, slopes of the conductance curves in the tunnelling and contact region as well as the displacement intervals in which the jump to contact takes place do not exhibit a dependence on the applied voltage. While the apparent barrier height varied at most by 5%, the jump to contact in 95% of the cases occurred between $-5.4$ and $-5.8$ Å. As shown in figure 2(c), which presents a close-up view of conductance curves in the interval $-6 \ Å \leq \Delta z \leq -5 \ Å$ where the discontinuous change of $G$ occurs, contact is established between $\approx -5.4 \ Å$ and $\approx -5.8 \ Å$.  

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Figure 2. (a) Conductance $G$ as a function of tip displacement $\Delta z$ on clean Au(111) acquired on a fcc stacking domain. $\Delta z = 0$ marks the displacement fixed by opening the feedback loop at 200 mV and 50 pA. The conductance in the tunnelling region appears as a linear curve on the logarithmic scale. Contact is signalled by a discontinuous conductance change. In contact the conductance increases slowly with increasing displacement. (b) Schematics of tip–surface geometry leading to conductance curves in (a). The displacement $\Delta z$ describes the movement of the tip and is—due to relaxations of tip and substrate material—not identical to actual modifications of the tip–surface distance. (c) Close-up view of (a) for displacements around the tunnelling-to-contact transition. (d) Statistical distribution of conductances in the contact region.

In most cases the conductance in the contact region is $G \approx G_0$. Sometimes, however, higher conductances can be reached. The probability distribution of $G$ in contact is shown in figure 2(d). In total, 30 conductance curves were evaluated for figure 2(d). Our findings for Au(111) are in accordance with the results of contact between the tip and Ag(111) and Cu(111) surfaces as recently reported in [6]. The random distribution of the displacements where the transition to contact occurs can be understood in the light of previously performed calculations [17]. The tunnelling-to-contact transition depends on where the tip approach is performed on the surface. When the tip is positioned on top of a surface atom—regardless of its chemical nature—the...
discontinuous change of $G$ should be detected $\approx 0.5 \text{ Å}$ earlier compared to a three-fold hollow position. All other locations on the surface exhibit the transition within this interval. Since with the tips used in the present experiments we did not resolve substrate surface atoms, the atomic location for conductance measurements is unknown. As a consequence, we expect the displacements of transition to be distributed within an interval of $\approx 0.5 \text{ Å}$. The representative conductance curves shown in figure 2(c) reveal that the majority of transitions occur within 0.4 Å. The statistical distribution of $G$ (figure 2d) indicates that configuration changes of the interface may affect electron transport properties quite substantially as recent transport calculations on Al(111) also pointed out [24]. Blanco et al [24] showed theoretically that the contact conductance between an aluminium tip and an Al(111) surface depends on the symmetry of the surface site which the tip is approached to.

As shown in figure 2(d), in the majority of cases the contact conductance between tip and sample is $\approx G_0$. In analogy to the results of [6] we propose that electrons are transported through a single atom which connects the two electrodes, i.e. the tip and the sample. It has been shown that for single metal atoms the number of observable conduction channels equals the number of valence orbitals [25]. Thus a monovalent metal such as gold has one conduction channel. The latter, as revealed by calculations [26, 27], is nearly open leading to a conductance of $\approx G_0$. Our proposed scenario of electron transport through a single Au atom is further corroborated by the observation of an individual atom when the area of contact is imaged after the conductance measurement. In accordance with [6] we propose that the imaged atom is expelled from the tip upon contact. The jump to contact occurs when chemical bonds between the surface and the tip apex start to weaken the adhesion of the atom to the tip structure. In this case, and over a relatively small distance variation of less than 0.1 Å [28] the atom will be transferred from the tip onto the sample. This atom is most probably a gold atom since in the course of tip preparation as discussed in the experiment section the tip apex is likely covered with substrate material. For contact conductances $G > G_0$ we observe clusters of (most probably) gold atoms at the contact location. In this case we propose that the contact between the tip and the surface was established by a neck whose width exceeded a single atom. This scenario is reasonable in terms of the Landauer–Büttiker expression for ballistic electron transport [29]:

$$G = G_0 \sum_{i=1}^{N} \tau_i,$$

where $N$ is the number of conduction channels supported by the contact and the $\tau_i$ denote the transmission probability of the $i$th channel. Experiments [13], [30]–[32] and simulations [26, 27] indicate that for gold contacts $\tau_i \approx 1$. In a combined STM and transmission electron microscopy experiment Ohnishi et al [33] related contact conductances between their microscope tip and a gold substrate to the number of adjacent Au rows bridging the electrodes. They found that a contact conductance of $G = nG_0$ ($n \leq 4$) was related to $n$ adjacent Au rows.

The slope of the conductance curve when both electrodes are in contact via a single atom is smaller than observed in the tunnelling region. We find that the conductance varies by $\approx G_0/10$ upon a displacement change of 1 Å. From experiments performed with the mechanically controlled break junction technique it is known that the slope of conductance curves in contact can be negative or positive. The sign of the slope reflects—according to Cuevas et al [27]—the participation of different atomic orbitals in electron transport. In the particular case of an individual Au atom the calculations [27] predict a fully open single conduction channel arising.
Figure 3. Left panel: conductance as a function of tip displacements on a single gold atom. Shown is the transition (2) from tunnelling (1) to contact (3) region. This transition is continuous and takes place within a displacement interval of $\approx 0.4$ Å. Inset: single gold atom deposited on a fcc domain. For conductance measurements the tip is placed above the centre of the atom. Right panel: sketch of tip–adatom contact. Tip and substrate surface are bridged via a diatomic Au wire through which the current is passed.

from the 6s orbital slightly hybridized with the 6p$_z$ orbital. As a result, the conductance in contact is nearly constant (in good agreement with our data).

For hcp stacking domains on Au(111) we observed a similar behaviour and we do not show analogous data characterizing the tip–surface contact here. Our data therefore do not reveal a dependence of the conductance on the surface reconstruction.

Before turning to the description of contacting an individual atom we compare our results of contacting a Au(111) surface with a gold-covered tungsten tip with simulations [17]. In these model calculations the Au(111) surface was mimicked by a five-layer Au(111) film while the tip was modelled as a tungsten tetrahedron mounted on the reverse of the Au(111) surface slab [17]. The calculations predict deviations from the exponential behaviour of the current starting from values as low as 5 nA. In our experiments, however, we observe the tunnelling region up to currents as high as 300 nA. A possible contribution to this marked difference could be the tip material which in the simulations is a pure W crystal while in our experiments the tungsten tip apex is covered by several layers of gold. Also force measurements using an atomic force microscope indicate that moving a tungsten tip [34] into contact with a Au(111) surface is different from using gold tips [35]. In a theoretical study of contact between an Al tip and an Al(111) surface [24] at a tip–surface distance of 5 Å the conductance was roughly three orders of magnitude higher than the conductance at the same distance between the W tip and the Au(111) surface as reported in [17]. Blanco et al [24] argue that part of this discrepancy may be due to the Bardeen model used in [17]. The Bardeen model does not take into account image potential effects which considerably reduce the tunnelling barrier and in turn increase the tunnelling current by one or two orders of magnitude [24].

We now turn to the discussion of contact between the tip and an individual gold atom. Since again results for fcc and hcp stacking domains are similar we discuss data obtained for single gold atoms residing on fcc domains. In figure 3 we show a typical conductance curve acquired on a single gold atom (see inset) which was deposited on the fcc stacking domain by the above described tip–surface contact. Figure 3 shows the transition region between
tunnelling and contact. Displacement $\Delta z = 0$ corresponds to the tip height at which the feedback loop of the microscope is frozen at 200 mV and 50 pA. As for the clean surface for a certain interval of displacements we observe an exponential behaviour of the conductance the tail of which can be seen in region 1 of figure 3. However, while the apparent barrier height of the conductance curve—as calculated from $\Phi = 0.952 (d \ln VG/d\Delta z)^2$ [36]—on the clean surface is $\approx 4 \text{ eV}$, the apparent barrier height on the gold atom is $\approx 5 \text{ eV}$ and thus higher than on the clean surface. This effect of an increased apparent barrier height has likewise been observed for Ag(111)–Ag and Cu(111)–Cu [6]. We suggest that an increased dipole moment at the atom site may be responsible for this effect. Moreover, on the atom no discontinuous jump to contact takes place. Rather than this, like for Ag(111)–Ag and Cu(111)–Cu [6], the transition is continuous at the time resolution of our experiment. Unlike the tip–surface contact, the tip–adatom contact does not lead to material being transferred from the tip. In the transition regions (see figure 3) the conductance curve exhibits different slopes. A displacement of 1 Å in the regions 1, 2 and 3 would imply conductance changes of $\approx G_0/8$, $\approx G_0/2$ and $\approx G_0/10$, respectively. Thus, while the conductance characteristics of tunnelling and transition regions for contact to an adatom (see sketch in the right panel of figure 3) differ from those obtained for contact to a flat surface (see figure 2), the conductance in contact exhibits the same magnitude and the same slope in both cases. As schematically shown in the right panel of figure 3, the most likely atomic arrangement of a tip–adatom contact may also be viewed as a two-atom wire. Interestingly, our data are in agreement with the almost length-independent conductance of short gold chains as reported, for instance, in [22, 33].

In agreement with [6] we observed that the probability distribution of $G$ has narrowed considerably around the mean value of $G \approx G_0$. Simulations of the atom contact [28] show that—in sharp contrast to the case of a flat surface—the atomic positions are only slightly relaxed during the transition from tunnelling to contact. The reason for this marked difference is the larger stiffness of the adatom bond to the surface. For Ag and Cu atoms on Ag(111) and Cu(111) an increase of the elastic constants to roughly double the value found on flat surfaces was reported [6, 28]. Most probably, a surface dipole induced by the Smoluchowski effect enhances the bonding of the adsorbed atom to the surface. It was further found that the tip–adatom interaction energy, contrary to the tip–surface one, remains well below 1 eV which was identified as the threshold value for jump to contact and atom transfer [6].

4. Conclusion

A scanning tunnelling microscope has been used to investigate contact conductances of structures at the nanometre scale. Tip–surface and tip–adatom contacts were formed reproducibly without deteriorating the structural integrity of the tip and thus the imaging capability of the instrument. Conductances on differently stacked surface layers are similar. While the tip–surface contact is signalled by a discontinuous conductance change, on an individual atom the tunnelling-to-contact transition occurs continuously at the time resolution of data acquisition. Contact conductances of mono- and diatomic gold wires reveal approximately one quantum of conductance.

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