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Theoretical study of conductance in stretched monatomic nanowires

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Abstract

Recent experiments showed that the last, single channel conductance step in monatomic gold contacts exhibits significant fluctuations as a function of stretching. From simulations of a stretched gold nanowire linked to deformable tips, we determine the distribution of the bond lengths between atoms forming the nanocontact and analyze its influence on the electronic conductance within a simplified single channel approach. We show that the inhomogeneous distribution of bond lengths can explain the occurrence and the 5% magnitude of conductance fluctuations below the quantum conductance unit $g_0 = 2e^2/h$.

Keywords: Gold; Green's function methods; Surface electrical transport (surface conductivity, surface recombination, etc.)

1. Introduction

Ballistic electronic conductance in short metallic nanowires and nanocontacts between tips has been widely analyzed both experimentally and theoretically in the last decade. The transport properties of the nanocontacts are known to be influenced by the geometrical structure of the contacts and by the electronic confinement [1]. Several experimental [2,3] and theoretical [4–9] papers discussed the interplay between geometry and conductance.

Mechanical properties of atomic contacts were accessed experimentally by scanning tunneling and atomic force microscopy, (STM/AFM) [2] and theoretically mostly by molecular dynamics simulations [3,5,9,10]. It was shown [11,12] that it is possible to pull stable monatomic gold wires, up to seven atoms in length between two gold electrodes, with a conductance close to $g_0 = 2e^2/h$. More recently, STM supplemented with a force sensor was used [13] to study the mechanical response under stretching of a low temperature ($T = 4$ K) chain of single Au atoms. Ab initio calculations of the breaking strength and of other mechanical

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properties of the nanowires demonstrated [13] a very considerable strength of bonds at these low coordinations relative to the high coordinations of the bulk metal.

Transport of electrons in metal nanowires was also analyzed [2] and abrupt conductance jumps were associated with atomic rearrangements in the wire. STM tips were used [12] to generate gold nanowires in situ in a high resolution transmission electronic microscope while recording the conductance. These results revealed the existence of suspended gold atom chains with a conductance equal to g_0, while another study [14] addressed the correlation between gold nanowire structure and the quantum conductance behavior within the same technique.

Theoretical approaches devoted to the understanding of the relation between electronic transport and the geometrical structure of metallic nanocontacts has been extensively developed. Tight binding models were proposed to analyze the conductance of gold wires in terms of electron standing waves due to the interference of electronic waves reflected at the extremities of the atomic constrictions [16] or to determine the transmitting channels in the atomic chain in terms of s,p,d orbitals [17]. The conductance of linear chains of four Au atoms suspended between two jellium electrodes was calculated vs. the distance between the electrodes within the ab initio local spin density functional (LSDF) approach. The conductance was determined using the recursion transfer matrix [18] and shown to decrease from 1 g_0 as the chain was stretched, similar to a Peierls transition.

More recently, the electronic transport in free standing gold atomic chains of up to seven atoms in length was studied at 4 K [15]. All along during the pulling of the monatomic nanowire, the behavior of conductance was characterized by two features. The first was a 5%-level fluctuation as a function of stretching, or between one pull and another. The second was a 1%-level conductance drop when the voltage across the nanocontacts exceeded about 15 meV. The latter was interpreted as a dissipative effect, quite similar to losses observed in point contact spectroscopy, this time due to inelastic scattering of electrons inside the nanowire, where longitudinal vibrations can be excited via electron–phonon interaction. The identification of the loss peak with a phonon inside the nanowire could be made thanks to the large phonon softening and strengthening of the loss intensity which is observed upon stretching.

That interpretation was directly supported by a recent DFT calculation accompanied by a molecular dynamics simulation [19]. There it was also shown that only about half the total stretch magnitude is absorbed by the nanowire itself, the other half being absorbed by the tip–wire junctions. The enhancement of inelastic scattering due to longitudinal phonon in the stretched nanowire was also theoretically addressed and found to be directly related to the stretching-induced phonon softening through perturbation theory [20].

In this paper, we provide a more specific rationalization to the zero-voltage ballistic conductance fluctuations around g_0. We do that by calculating explicitly, albeit approximately, the conductance in a nanocontact. To that end we first built a mechanical model to represent the gold nanowire suspended between two tips. Based on that, and on a one dimensional bond model between atoms of the wire connected to two infinite reservoirs, we calculated the wire conductance during the course of its stretching, as described by the mechanical model. Focusing on the distribution of interatomic distances in the gold stretched wire, we analysed the conductance by means of a Green's function operator formalism, that takes into account the changes in the hopping integrals with the distance between atoms.

2. Theoretical model

2.1. Model for stretched monatomic nanowire

In order to model the structural properties of a gold wire stretched between two tips, we use a semi-empirical effective potential, which is in turn based on tight binding theory in the second moment approximation (SMA). The potential experienced by the nth gold atom and due to the other gold atoms m separated by a distance r_{nm} is written as [21]:

$$V_n = \lambda \sum_m e^{-p((r_{nm}/r_0)-1)} - \epsilon \left(\sum_m e^{-2q((r_{nm}/r_0)-1)} \right)^{\alpha} \quad (1)$$

All the parameters entering in the above expression are calculated by fitting the experimental bulk and surface properties of gold. Their values are $\lambda = 0.4086$ eV, $p = 8.5624$, $\epsilon = 1.6332$ eV, $q = 3.6586$, $\alpha = 0.6666$, $r_0 = 2.88$ Å and we consider a cutoff function for distances $r_{nm} > r_c$ where r_c is the second nearest neighbor distance [19,21]. This potential was used to determine the mechanical properties (equilibrium spacing and cohesive energy) of a gold wire wedged between two gold tips. It was found to give results in close agreement with those calculated using a more accurate DFT approach [6,19].

The system is formed by some atoms which are allowed to move and other atoms that are fixed, the latter mimicking the connecting tips. The moving atoms are those forming the nanowire (we chose seven atoms at the beginning of the simulation) and the junction forming a part of the two tips. Each tip contains 13 moving atoms arranged in a rectangular (1 1 0) lattice. This moving system is blocked on each side by a rectangular plane of fixed gold atoms, forming the back sides of the tips, and the distance between the inner fixed planes is noted L.

As L is increased and the planes move apart, they stretch the nanowire and the tip–wire junctions in between. We simply optimize the total energy of the system submitted to a gradually increasing stress, without the need of thermal dynamics (the temperature of the experiments is very low (4 K)). All the moving atoms are free to find their equilibrium positions, at each stretching step.

Conductance calculations are then carried out at each step of the stretching simulation. We verify first that the geometry of the tips and of the wire–tip junctions is free to evolve, and thus to influence the behavior of the wire during stretching. We ran several calculations with a variety of initial conditions, including a wire attached to a top site of the rectangular tip, or to a bridge site, or to a hollow site. The results showed that the evolution from different starting configurations was significant, especially regarding the value of the total elongation of the wire. Accordingly we decided to concentrate on a single reasonable, but particular path. Starting from a perfect geometry of wire and tips ($L = 25.6$ Å, which corresponds to the equilibrium bond length for each gold atom in the system), we compress the wire by progressively moving one tip closer to the other (i.e. by decreasing the length L) and minimizing the total energy of the system. The compression stops when the energy reaches a minimum (distance between fixed tips equal to 9.8 Å).

At this point, we begin stretching the system to form a nanowire. These two preparatory steps in the simulation allow us to generate a complete a priori geometry for all the moving atoms, a geometry which was normally different from the initial arrangement. In this way, we may obtain results that are less dependent on the initial configuration chosen for the system.

The remaining problem concerns the choice on the number of moving atoms which participate in the stretching process. We are mainly concerned by the electronic transport in the nanowire and the tip geometry might appear a secondary problem. However, it is important to check how the geometry of the whole tip–wire junction influences the conductance. That aspect will be discussed in Section 3.

The elongation of the nanowire is simulated by slowly increasing the distance between the two tips, avoiding abrupt variations that could break the wire. For each stretching step, corresponding to an elongation of 0.02 Å, the equilibrium configuration is determined for all the moving atoms by minimizing the total energy. That minimum corresponds to a constrained equilibrium configuration for the tip-suspended nanowire subject to a given stretching length. The two tips are then progressively moved apart until the wire breaks.

2.2. Electronic transmission through a metallic nanowire

To study the quantum transport of electrons through the nanowire, we adopt a very simple

one-electron model. The whole tip–nanowire–tip system is treated as a one-dimensional (1D) system, made up of perfect semi-infinite left and right leads, connecting through an extended defect, formed by the monatomic nanowire, the junctions and the first piece of the tips. Inside the perfect semi-infinite 1D leads, or reservoirs, electron propagation is described by a single-band tight binding Hamiltonian characterized by an on-site energy E_0 and the electron hopping integral J between nearest neighbor sites.

The defect, which corresponds to the nanowire, is assumed to consists simply of a monatomic chain of N atoms connected to the reservoirs. If and when the distance between neighboring atoms fluctuates under stretching, the electron propagation will be modified. To further simplify the treatment, we assume these positional fluctuations to affect the hopping integrals J, but not the on-site energies, retaining their fixed value E_0. As a result, the electron propagation inside the nanowire will be characterized by a set of $N-1$ hopping integrals J_n, $n = 1,\ldots,N-1$ (see Fig. 1). They can be related to the distances and to the bulk integral J according to the empirical form [22]

$$J_n = J\exp(\beta(1 - r_n/r_{\mathrm{eq}})) \tag{2}$$

where r_n stands for the distance between the $(n-1)$th and nth atoms belonging to the nanowire and r_{eq} denotes the equilibrium distance (i.e. the lattice parameter of the perfect atomic chain $r_{\mathrm{eq}} = a_0 = 2.49$ Å). Note that when all the distances r_n are set to r_{eq}, the different hopping integrals J_n reduce to the bulk integral J yielding an ideal system without any defects. While the model is clearly oversimplified in many aspects, it does serve our purpose of pinpointing the effect of distance fluctuations on the conductance.

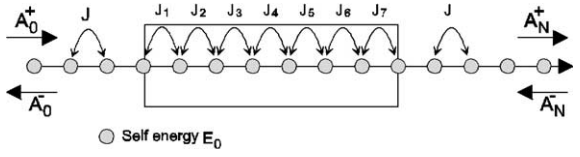

Fig. 1. One dimensional model for the calculation of the conductance of the stretched nanowire connected to semi-infinite reservoirs.

The Green's function formalism can be used to calculate the transmission coefficient in this model. We consider an incident electron propagating freely from the left tip with a wavevector q and an energy $E = E_0 + 2J\cos(q)$. The electron is scattered by the nanowire leading to transmitted and reflected waves. The transmission coefficient is given by

$$t(E) = 1 + \frac{\mathrm{i}}{2J\sin(q)}\sum_{n,m} T(n,m)\mathrm{e}^{\mathrm{i}q(x_n - x_m)} \tag{3}$$

where x_n denotes the position of the nth atom of the 1D nanowire and the T matrix is a $N \times N$ matrix operator

$$T = V(1 - G_0(E)V)^{-1} \tag{4}$$

In Eq. (4), $G_0(E)$ denotes the Green's function operator of the ideal perfect infinite chain. The perturbation operator V accounts for the modification of the hopping integrals in the nanowire due to the fluctuations of the interatomic distances inside the wire compared to their ideal values in the reservoirs [23]. This operator, whose dimension is again N, acts inside the N-atom subspace and is defined as

$$V(n,n') = (J_n - J)\delta_{n',n+1} + (J_{n-1} - J)\delta_{n',n-1} \tag{5}$$

where n and n' define two atoms in the wire. The conductance g through the region where scattering takes place is related to the transmission coefficient of the electron via the Landauer formula [24,25], expressed at low temperature and low voltage, and for a single channel, as

$$g = \frac{2e^2}{h}|t(E_{\mathrm{F}})|^2 \tag{6}$$

where E_{F} is the Fermi level of the electrons. Applying Eq. (6) to our situation requires that the left- and right-hand sides of the region where scattering takes place is connected to two reservoirs of electrons held at infinitesimally different electrochemical potentials. By assuming that the two semi-infinite chains play the role of reservoirs, our single-band model leads at half filling to a Fermi level E_{F} equal to E_0.

3. Results and discussion

Fig. 2 displays typical snapshots during the formation and the stretching of the wire. At the beginning of the simulation (Fig. 2a), all atoms in the wire and tips are at their equilibrium distance $a_0 = 2.49$ Å and $a = 2.88$ Å, respectively. The ideal nanowire contains seven atoms. Compression proceeds until the two tips are stuck together and the total energy is a local minimum with nearest neighbor distance between Au atoms approximately equal to $a = 2.80$ Å (Fig. 2b). At this point, the initial configuration is lost and the moving part of the system is disordered: the stretching process can start. Upon initial stretching, gold atoms tend to arrange themselves in twisted chains until one atom is extracted from a tip (Fig. 2c). Subsequently, a monatomic chain is formed by successive incorporation of atoms from the tips into the wire (Fig. 2d). At a total length corresponding to eight atoms long (one more than the starting wire), the nanowire breaks (Fig. 2e). These results obtained with the SMA potential are fully consistent with those found using an effective medium potential [13]. Note that, as already mentioned in a previous paper [19], incorporation of each new atom in the chain is obtained after an elongation ΔL of L nearly equal to 1.5 Å. This incorporation induces a reorganization of the wire, particularly by decreasing the bond lengths between the gold atoms. This model reproduces quite well the limit distance from which a nanowire can be elastically stretched without dramatic change (1 Å in the experiments [15]). Between two successive incorporations, the values of the distances between the atoms in the wire and between the wire extremities and the apex of the tips generally display a non-uniform distribution. The stretching tends to increase the bond lengths in the wire, the disturbance being stronger on those atomic bonds close to the wire ends than on the bonds in the wire center. In fact, we generally observe that the bonds linking the wire to the tips are weaker than the bonds between atoms of the wire. That also explains why a wire can be pulled out of tips and stretched out to such remarkable lengths without breaking in the middle.

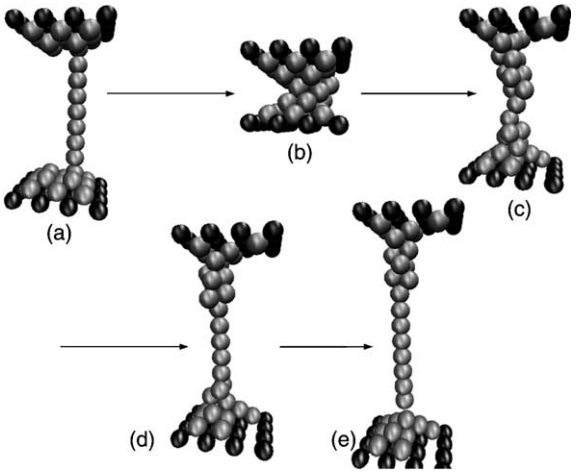

Fig. 2. Mechanical model. The distance L defined in the text represents the separation between the two black ball planes: (a) starting configuration of the simulation; (b) the two tips are approached to the closest position and (c) stretching has begun, and a single atom has been pulled out of the neck, initiating the monatomic nanowire. This is also the starting point of conductance calculations; (d) strained monatomic nanowire now reaching five atoms in length and (e) maximum elongation to an 8-atom long nanowire. At this point, the wire breaks and conductance drops to zero. The total energy of the system is calculated with Gamba's semi-empirical potential [21] and is minimized throughout the stretching process.

The changes of interatomic distances r_n in the nanowire are extracted from the simulation and turned into one dimensional effective distances x_n for conductance calculations. The x_n are the same as r_n except at the wire–tip junctions. There we assume conduction to proceed always via the smallest bond length. For the energy of the electrons generating the conduction in the nanocontact, we consider an energy E equal to and close to the Fermi level energy E_F of the bulk gold.

Fig. 3 shows the calculated behavior of the conductance of the total model nanocontact of Fig. 2 during stretching, and represents our main result. The conductance is close to g_0 as it should be for a single channel, the broad electronic band suffering only minor scattering from the weak perturbation represented by the distance fluctuations. The main characteristics we wish to focus upon here is precisely the conductance fluctuations that occur during stretching of the wire. These variations represent less than 5% of g_0 and are directly comparable with the value obtained in

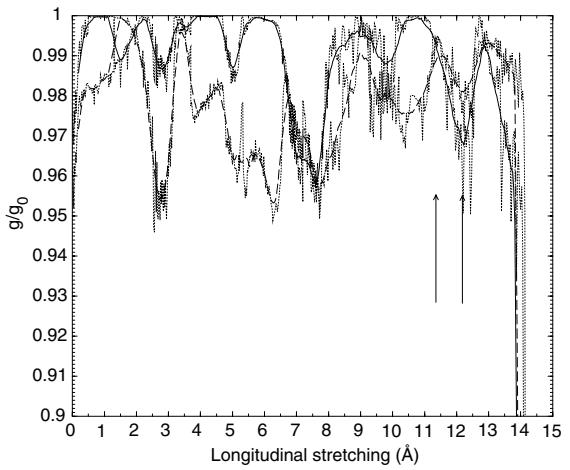

Fig. 3. Behavior of the reduced conductance vs the wire stretching (dotted curves) for two incident electron energies. The solid line represents a spline fit of the conductance fluctuations during stretching of the wire for an incident electron energy corresponding exactly to E_F. The dashed line represents a spline fit of the conductance fluctuations during stretching of the wire for an incident energy corresponding to $E_F + J$. Arrows correspond to maxima and minima for the nanowire containing seven atoms (see Fig. 4).

experiment [15]. Besides the 5% magnitude, another point of similarity of the conductance curves in Figs. 3 and 5 with the experimental curves under stretching is the sharp slope at the jumps. Incorporation of an additional atom in the wire or configurational change of the wire at its extremities (wire–tip junction) can interpret the constant distance (1.5 Å) between the conductance maxima.

The fluctuations occur in the form of rather sharp conductance jumps with minima and maxima that depend strongly on the wire stretching. These jumps are directly related to the fluctuations of the atomic bond distances in the nanocontact perturbing the electronic transmission inside the nanowire. If the nanowire length were infinite, the fluctuations, no matter how weak, would of course block conductance completely, and lead to an Anderson insulator [26]. The inverse Anderson localization length depends on disorder, roughly proportional to the square of the hopping fluctuations. In a nanowire of finite and short fixed length like we have here, the conductance simply shows small fluctuating drops from g_0. In general, conductance should drop exponentially with the inverse localization length. The exponential amplification expected on the conductance due to hopping disorder may explain the abruptness of the fluctuations evidenced in Fig. 3.

At finite voltage $W = \mu_L - \mu_R$ (μ_L and μ_R representing, respectively, the left and right electrochemical potentials), Landauer's ballistic conductance is the same as in Eq. (5), only with $|t(E_F)|^2$ replaced by an average of transmission $|t(E)|^2$ between μ_L and μ_R. We study in Fig. 3 the influence on transmission of the incident electron energy by drawing g for two distinct values of this energy (respectively, equal to E_F and $E_F + J$). Although the conductance presents globally a similar jagged profile, a detailed examination of the curves shows some differences. A first difference concerns the average amplitude of these variations. For $E = E_F$ the mean curve tends in fact to remain localized around 0.99 g_0 while for energy $E_F + J$ the fluctuations take place around 0.975 g_0.

This is relevant, for it implies a slight drop of *ballistic* conductance for finite and increasing voltage. The calculated decrease in fact corresponds to an increased tendency to Anderson localization as one moves away either sides from the center of the 1D band, the band being centered precisely at E_F, where the localization length is maximum. Because of that, the resulting predicted conductance decrease is monotonic and relatively *uniform* for increasing voltage, that is for increasing deviation of the electrochemical potential from E_F.

Interestingly, the experimentally observed conductance indeed does show a slight decrease with increasing voltage W [15]. However the part of the conductance decrease that is uniform with voltage (i.e., that has a roughly constant voltage derivative) is not major. The most interesting part is instead strongly peaked around a specific voltage, ranging from 18 to 12 meV depending on the strain. This peak voltage in fact corresponds to a longitudinal phonon frequency in the nanowire [15,19], and is evidence for the onset of non-ballistic, dissipative resistance in the nanowire.

Then, returning to Fig. 3 we note that in all cases the conduction minimum is around 0.95 g_0 in nice agreement with the experimental data. The maxima of all curves correspond to the situation where a new atom is freshly incorporated in the

wire thus permitting a general compression of the chain. The minima mark the limit stretch just before the atom pullout with incorporation and reorganization of the wire.

To correlate more quantitatively the conductance behavior with the atomic bond length fluctuations, we have drawn in Fig. 4 the distribution of the bond lengths in two wires formed by seven atoms at the corresponding maxima (just after atom incorporation) and minima (just before atom incorporation). We see that this distribution, fitted by Gaussian profiles, broadens by 40% during the stretching between two successive incorporations, and the maximum of the profile shifts from 2.48 to 2.51 Å. This feature is quite general for long wires, and for instance we obtained for a six atoms long wire the same type of bond length distribution.

We also observe an energy dependence of conductance connected to the wire length. When the nanowire is shorter than 8 Å (i.e., four atoms) the amplitudes of the fluctuations are much larger for $E = E_F + J$, so the shapes of the two distribution curves appear less similar than for longer nanowires (seven or eight atoms). Clearly, the stretching perturbs much more substantially the atomic bonds in the junctions than those amid the wire. The fluctuations are largely if not exclusively a junction effect. As such, they are more important for a shorter nanowire.

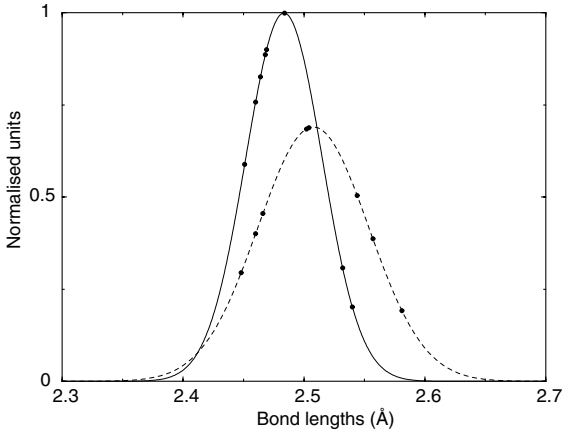

Fig. 4. Bond length distribution between nearest neighbor atoms in the 7-atom monatomic nanowire at the maxima (solid curve) and minima (dashed curve) of conductance (as shown by arrows in Fig. 3).

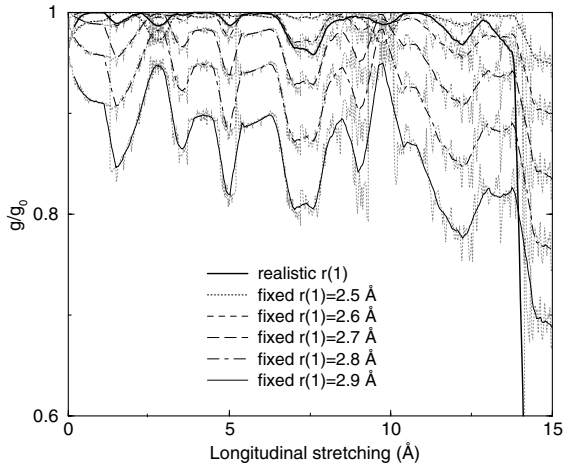

Fig. 5. Influence of the effective bond length $r(1)$ representing the junction between the nanowire and the tips during stretching.

Fig. 5 displays further evidence of the influence of the junctions between the nanowire and the two tips. In these calculations, we changed the bond length of the tip–wire junction, always assuming the current-carrying path to remain strictly one dimensional. The results show that while such a change does not modify the shape of the conductance vs the wire length, it does affect the absolute values and amplitudes. Stretching that effective bond further and further away from its ideal value leads to a considerable conductance decrease.

4. Conclusion

Motivated by recent experimental observations showing that a stretched monatomic gold wire between two tips presents systematic conductance fluctuations below g_0, we carried out simulations of the behavior of electronic conductance during the nanowire stretching up to the rupture of the contact. We find that the observed 5% conductance fluctuations around g_0 can be explained as the result of continuously distributed atomic bond lengths inside the stretched nanowire. The maxima of conductance are correlated to an atom incorporation in the wire which induces a compression

of the chain while the minima correspond to the maximum wire stretching, just before incorporation and/or reorganization.

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