

Onset of Energy Dissipation in Ballistic Atomic Wires

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Electronic transport at finite voltages in free-standing gold atomic chains of up to seven atoms in length is studied at low temperatures using a scanning tunneling microscope. The conductance vs voltage curves show that transport in these single-mode ballistic atomic wires is nondissipative up to a finite voltage threshold of the order of several mV. The onset of dissipation and resistance within the wire corresponds to the excitation of the atomic vibrations by the electrons traversing the wire and is very sensitive to strain.

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The trend toward miniaturization in electronics will soon lead to devices of nanometer scale in which quantum effects become relevant. The ultimate quantum conductor is a perfect one-dimensional wire, such as an atomic chain [1,2] or semiconducting heterostructure [3]. In these wires the electrons are ballistic since there are no defects to inhibit resistance-free currents [3]. The limiting factor in the current-carrying capacity of a wire is dissipation, which results in heating. Two mechanisms contribute to the resistance of a metallic wire: elastic scattering with defects and impurities and inelastic scattering with the lattice vibrations [4]. In the absence of scattering, electrons can propagate freely and transport is said to be ballistic. This situation is possible in the nanoscale where the mean-free path of electrons can be much longer than the length of the device.

The two-terminal zero-bias resistance of a single-mode ballistic wire is the resistance quantum $h/2e^2$. This resistance is entirely associated with the connections of the wire to the electrodes [5], being the intrinsic resistance of the wire zero, as recently demonstrated in quantum wires fabricated from GaAs/AlGaAs heterostructures [3], and in agreement with Landauer framework [6,7]. Within this framework, the applied voltage serves to unbalance the chemical potentials for propagating electrons in each direction and drops entirely at the contacts and not within the wire. The Joule dissipation associated with this resistance is assumed to take place far away from the contact (at an inelastic relaxation length), where electrons and holes relax to the Fermi level of the electrodes. This picture is correct for bias voltages close to zero, which implies vanishingly small currents (note that the resistance-free currents in the experiment of Ref. [3] were smaller than 1 nA).

In this Letter we study transport at finite voltages and the mechanism of dissipation in ballistic wires. Our experiments are performed in freely suspended gold atomic wires of up to seven atoms in length, fabricated using a low-temperature scanning tunneling microscope (STM) [1,2]. Very recently the forces and conductance have been measured simultaneously [8] during the process of chain

formation giving insight into the formation mechanisms. The mechanical and electronic properties of these metallic nanostructures are of great interest not only from the point of view of the applications but also from a fundamental point of view, since the system has few atoms and should be amenable to detailed modeling.

Atomic wires of gold are fabricated at low temperatures (4.2 K), using an STM. Fabrication of an atomic wire is as follows [1]: a metallic contact is formed between a tip and a substrate of an STM, both made out of Au, with 99.99% purity. Elongation of the contact results in a decrease in its size. In the final stage a one-atom contact is formed, and as we continue elongating there is a certain probability that an atomic chain forms [1]. The length of the wire can be estimated from the length of the last plateau before rupture, whose conductance is approximately one in units of $2e^2/h$. The atomic wires are very stable at low temperature and the measured curves are completely reproducible as long as the tip position is not changed. Typically an experiment on a given atomic wire takes about one-half hour, and ends when the wire breaks as a result of further elongation. The differential conductance G of the wires as a function of voltage is measured using a lock-in technique, with a small modulation of 1 mV. The derivative of the differential conductance dG/dV is calculated numerically.

Experimentally, it is observed [1] that the zero-bias conductance of Au wires of up to seven atoms in length is close to $G_0 = 2e^2/h$, the quantum conductance unit, independently of their length, reflecting the fact that gold wires have a single almost completely open quantum channel, as confirmed by theoretical calculations [9–11]. This agrees with Picciotto *et al.* [3], showing that the resistance is at the contacts, not within the chain. These atomic wires do not show Ohmic behavior: the conductance is voltage dependent. We measure the differential conductance $G = dI/dV$ of atomic wires of different lengths (from one to seven atoms). Typical differential conductance curves for short and long atomic wires are shown in Figs. 1(b)–1(d). The differential conductance G shows a hump at zero bias, dropping about 1% in the range (20 mV). Often the

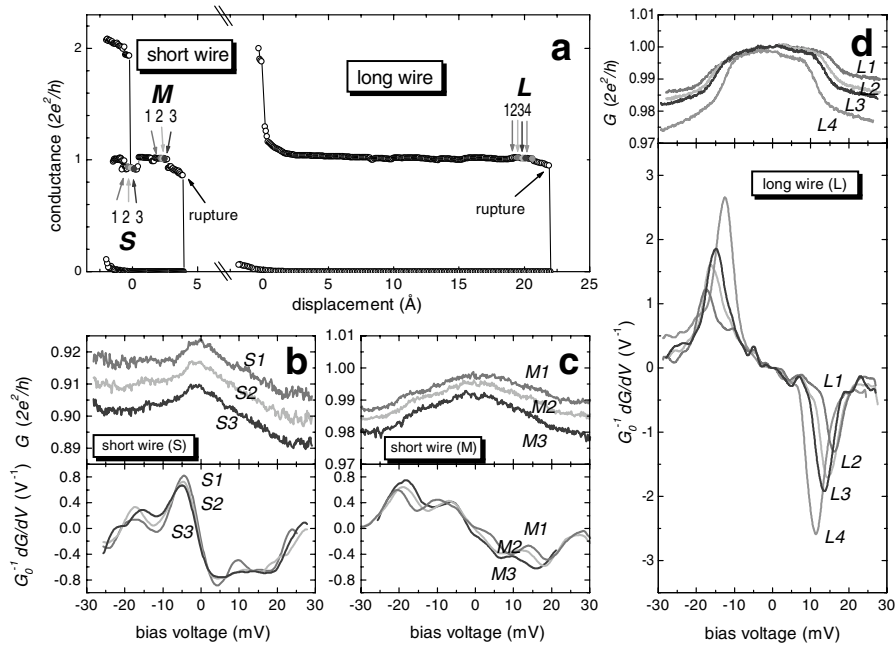


FIG. 1. (a) Short and long atomic wire, ~ 4 Å long and ~ 22 Å long, respectively. At the point of rupture, the atomic wire collapses and the conductance, which is negligible in the scale of this figure, corresponds to the tunneling regime. To reestablish contact, the electrodes must reapproach by a distance of the order of the chain length. Panels (b), (c), and (d) show the differential conductance and its derivative at points *S*, *M*, and *L*, respectively, marked by the arrows. The various curves in (b), (c), and (d) were acquired at intervals of 0.3, 0.3, and 0.5 Å, respectively. Note that the vertical scales are identical in these panels.

differential conductance curves show asymmetries and oscillations due to elastic scattering which results in interference effects [12,13].

These symmetric drops in the conductance are characteristic of inelastic scattering of electrons, and the range of voltages is typical of phonons. At cryogenic temperatures, the differential conductance of larger ballistic metallic contacts (100–1000 atoms in cross section) is also voltage dependent, and has been used to obtain energy-resolved information on the interaction of electrons with phonons and other elementary excitations. This well-established technique is known as point-contact (PC) spectroscopy [14–16]. When a voltage drop V is applied to a metallic contact, whose radius is smaller than the electron mean-free path, electrons are injected into the higher voltage electrode with excess energies of up to eV . These electrons have a finite probability of exciting a vibrational mode of the lattice, i.e., emitting a phonon. At these low temperatures the probability of phonon absorption is very low because the equilibrium phonon population is almost zero. The resulting dG/dV vs voltage curves, or PC spectra, are proportional to the phonon density of states (DOS) times the electron-phonon coupling strength. The spectra scale with the zero-bias conductance as $\propto G_0^{3/2}$, reflecting the fact that only electrons that are scattered close to the contact, i.e., within a distance to the contact of the order of the contact radius, have a finite probability of being backscattered and, consequently, of being detected as a reduction in conductance. The standard spectra for Au have peaks at 10 and 18 mV, corresponding to the maxima in the trans-

verse and longitudinal phonon DOS at 10 and 18 meV, respectively. The transverse peak being stronger than the longitudinal peak.

In contrast to the PC spectra of larger ballistic contacts, the spectra of atomic wires are very sensitive to the atomic configuration at the contact. Figures 1(b) and 1(c) show the differential conductance and spectra at two different situations during the elongation of a short atomic wire, positions labeled *S* and *M* in Fig. 1(a), respectively. At *S* the atomic wire is likely to be just one atom long, while at *M* the contact has been further stretched about 3 Å that is, the wire is about two atoms long. The evident broadening of the conductance hump is typically observed for atomic wires longer than one atom. The signal is about three times larger than that given by the semiclassical theory of PC spectroscopy [15,17]. Figure 1(d) corresponds to the long wire (about seven atoms long) in Fig. 1(a). For this long wire the conductance drop takes place quite sharply, which results in a sharp peak in the spectra. This dependence of the spectra on the length of the wire indicates that most of the measured signal comes from processes occurring within the wire itself.

The observed spectra for the longer atomic wires are a signature of their one dimensionality. In an infinite one-dimensional conductor, momentum is conserved, and, consequently, electrons can only excite longitudinal vibrations of the atomic chain whose wave number is twice the Fermi wave number k_F , hence only one phonon mode will show in the spectra. For a linear atomic chain with inter-atomic distance a and one conduction electron per atom,

k_F equals $\pi/2a$, and, consequently, electrons will only excite phonons of wavelength $2a$, that is, zone-boundary phonons. In this process an electron dissipates its energy, losing one quantum of vibrational energy $\hbar\omega_{2k_F}$ and being backscattered. Here, ω_{2k_F} is the frequency of the excited vibrational mode. In a ballistic conductor, this phonon emission process will be possible only for voltages V larger than $V_{\text{ph}} = \hbar\omega_{2k_F}/e$, since the chemical potentials for right-going electrons and for left-going electrons are unbalanced by eV . Consequently, the onset of dissipation is marked by a sudden decrease in the conductance, since the backscattered electrons do not contribute to the current. The position of the peak in the spectra V_{ph} gives the frequency of the $2k_F$ phonon, and its height A_{ph} , which is related to the conductance drop, is proportional to the probability of the phonon emission process. The magnitude of the conductance drop (about 1% for a chain of 20 Å in length) is consistent with an inelastic mean-free path of about 2000 Å in an infinite wire, which is reasonable for a metal at low temperatures. Experimentally, the sharpness of the conductance drop [see Fig. 1(d)] is limited by thermal smearing (~ 2 meV) and by the finiteness of the atomic wires. A linear monoatomic chain of N atoms will have N longitudinal vibrational modes. For an atomic chain of length L coupled to rigid electrodes, the wavelengths of the different modes n are simply given by $\lambda_n = 2L/n$. However, since the atomic chain is mechanically coupled to the bulk (the electrodes), the discrete modes of the chain will broaden into resonances, whose width will depend on the mechanical coupling between chain and electrodes. In addition, momentum conservation in the electron-phonon interaction will not hold strictly in a finite system, and, as a consequence, in a short atomic chain the electrons will be able to interact with phonons of wavelengths different from $2a$. This will lead not only to a smearing in the spectra but also to the possibility of observing other vibrational modes, especially in the shortest chains.

Experimentally, it is observed that the position and amplitude of the peak in the spectra of a given atomic wire are very sensitive to its state of strain. As shown in Fig. 1(d), stretching the wire results in a decrease of the frequency of the phonon and an increase in the emission probability. This phonon softening indicates that the elastic constant of an atomic chain decreases as it is stretched. The increase in the emission probability indicates an enhancement of the electron-phonon interaction. In Fig. 2, we have plotted the position of the peak in the spectra and its amplitude for many different atomic wires. The emission probability increases with the length of the wire [18], and the variations due to stretching are much larger than for short wires.

An atomic wire can be stretched elastically only a limited distance (~ 1 Å) before breaking or changing to a new configuration. These configurational changes result in stress relaxations correlated to abrupt conductance jumps [8], whose magnitude is much smaller than a conductance

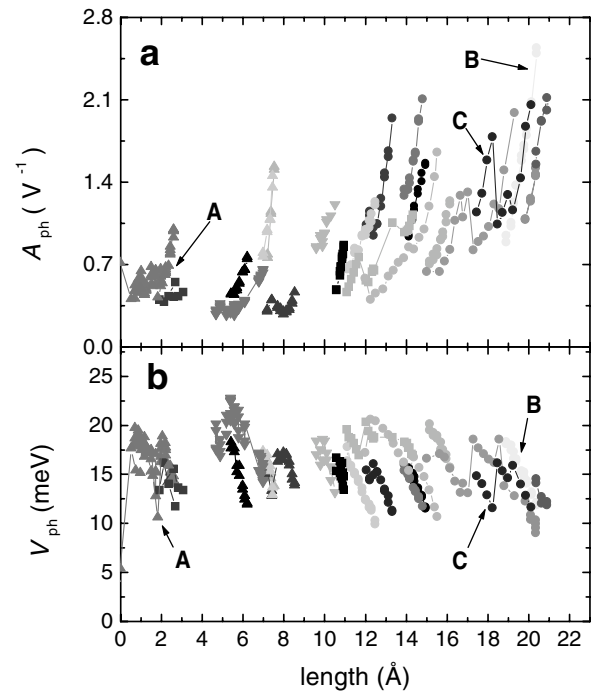


FIG. 2. (a) Magnitude A_{ph} and (b) position V_{ph} of the phonon peak in the PC spectrum as a function of length chain. For clarity, we present only 22 representative chains out of more than 100 studied. Each chain is represented by a different symbol. The chains labeled A and B are the short and long wires, respectively, of Fig. 1; C is the wire in Fig. 3. The length of the chain is estimated from the length of the last plateau.

quantum $2e^2/h$. Some of the stress relaxations correspond to the incorporation of an extra atom into the atomic wire, while other relaxations come from atomic rearrangements occurring in the electrode region close to the wire [8]. In Figs. 3(b)–3(d), we show the evolution of the conductance, and the amplitude and position (in energy) of the phonon peak as the wire is elongated along 3 Å before

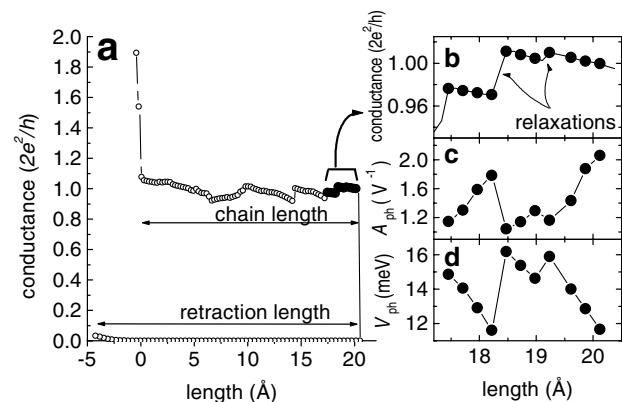


FIG. 3. (a) Evolution of the conductance for a 20-Å-long atomic wire. (b) Zoom of the last part of this evolution showing abrupt jumps in the conductance corresponding to force relaxations due to atomic rearrangements between the elastic stages. (c) and (d) show the magnitude A_{ph} and position V_{ph} , respectively, of the phonon peak in the PC spectra.

chain rupture. The final length of the wire was 21 Å [see Fig. 3(a)]. Note that jumps in the amplitude and position of the phonon peak in the spectra are correlated to conductance jumps and, consequently, to stress relaxations. The observed phonon softening with elastic deformation is consistent with the results of the *ab initio* calculations of Sánchez-Portal *et al.* [19] for linear and zigzag atomic wires. Our results are compatible with both types of wires. Detailed theoretical calculations would be necessary for a quantitative understanding of the electron-phonon interaction and its enhancement with elastic strain in metallic atomic wires.

As we have seen, dissipation in one-dimensional atomic wires occurs through the process of phonon emission by hot electrons. In contrast to what happens in a three-dimensional geometry, the hot electrons can excite a single vibrational mode of the atoms of the wire, and this requires electrons with sufficient energy. As a consequence, for voltages below V_{ph} , no dissipation occurs within the atomic wire. The maximum dissipationless current that an atomic wire can carry is then $G_0 V_{ph} \approx 1 \mu\text{A}$, which is an enormous current density $\sim 10^7 \text{ A/mm}^2$. For voltages above V_{ph} , the wire progressively heats up. For a given voltage a balance is established among the energy released by the electrons, the energy absorbed by the electrons (phonon absorption), and the energy lost by thermal conduction to the electrodes. In our experiments, atomic chains could sustain voltages of up to 500 mV (note that in the same conditions an atomic contact could stand 2000 mV). Phonon emission by hot electrons in a one-dimensional conductor was investigated previously in the context of carbon nanotubes [20]. In this case, the much longer length of the wires leads to qualitatively different results.

In summary, we have studied the voltage dependence of the conductance of metallic atomic wires at low temperatures, showing that inelastic scattering of electrons sets in at a finite voltage due to the excitation of the vibrations of the ions of the atomic chain. This well-defined threshold for dissipation is characteristic of the electron-phonon interaction in one-dimensional systems. We observe that the mechanical tensioning of the atomic chain results in bond softening, which is reflected in a decrease of the phonon

frequency and a dramatic enhancement of the electron-phonon interaction.

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