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One-atom point contacts

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A mechanically controllable break junction is used to study the transition between vacuum tunneling and contact for three different metals. In the tunnel regime a faster-than-exponential behavior is seen at close distances, followed by a jump to contact, both interpreted as being due to metal bonding forces. We show that stable contacts of a single atom can be formed. For Cu, the conductance value for a one-atom contact is very close to $2e^2/h$. For Al this value is less well defined but of similar magnitude, while for Pt it is noticeably higher, implying that the electronic structure of the atoms is relevant to the one-atom conduction process.

The study of point contacts has provided a wealth of information on conduction properties, elementary excitations, and scattering mechanisms in metals, compounds, and alloys.[1,2] With traditional techniques of forming the contacts, such as the spear-anvil method, stable contacts can be produced up to resistance values of several tens of ohms, corresponding to a contact area of at least a thousand atoms. The advent of the scanning tunneling microscope (STM), and related techniques[3] made it possible, in addition to scan surface properties, to study vacuum tunneling between metals, and the transition from tunneling to contact. One of the first experiments of this kind[4] showed a jump to contact at approximately 10 kΩ by rapidly recording the resistance while the tip was driven into the surface. This work was followed by a number of theoretical analyses of the evolution of the conductance during contact formation.[5–8] Also the mechanical properties of atomic size contacts have been studied experimentally using STM-based techniques, and theoretically using large-scale molecular-dynamics calculations.[9–11]

An estimate for the conductance through one atom is obtained from the number N of conductance channels[12] in a one-atom-wide metallic point contact. We have $N \simeq (k_F a/2)^2$, where a is the atomic radius. For a simple metal such as Cu the Fermi wave vector $k_F = 1.4$ Å^{-1} and $a = 1.3$ Å, giving $(k_F a/2)^2 = 0.83$, i.e., there is probably one conduction channel in one atom. Ideally, the conductance would thus be given by $2e^2/h$, corresponding to a resistance of 12.9 kΩ. However, in view of the crudeness of the estimate, the small effective length of the point contact, and scattering of the wave functions from the irregular potential surrounding the contact, the conductance may be dramatically reduced below this value. Indeed, several model calculations[5–7] found values much below $2e^2/h$.

The recently developed mechanically controllable break (MCB) junction technique[13] is particularly well suited for the study of contact formation on an atomic scale. It is orders of magnitude less susceptible to vibrations than the STM and has the advantage that clean, freshly exposed surfaces can be brought into contact at low temperatures. The first experiments with MCB junctions[13,14] suggested that atomic size contacts can be formed and kept stable for long periods of time. For close approach of two metal surfaces an adhesive avalanche has been predicted[15] and the subsequent atomic dynamics was believed to make one-atom point contacts intrinsically unstable.[10] Here, we present a study of the process of contact formation and fracture by monitoring the junction resistance while displacing the electrodes by small fractions of an atomic distance. We present experimental evidence that one-atom contacts can be formed, and that the conduction through a single atom is very close to the conductance quantum. Our results are to a great extent corroborated by the recent calculations of Todorov and Sutton.[16]

The key element of the MCB technique, the sample mounting, is drawn schematically in the inset of Fig. 2 below. The sample, in the shape of a metal filament, is glued on a substrate (bending beam), which is made of phosphor bronze, covered with insulating kapton foil. Compared to a glass bending beam, as was used in earlier experiments, this substrate has the advantage that it is more flexible, making the preparation of the notch in the wire considerably less critical, and allowing a much wider range of metals to be studied. By bending the substrate at 4.2 K in high vacuum, the filament is broken. The electrodes, which are thus freshly exposed, are brought back into contact. The bending, from this point onward, is controlled by tuning the piezovoltage V_p allowing fine adjustment of the separation between the electrodes. The measurements presented are taken at 1.3 K in the case of Cu and Pt, and at 4.2 K in the case of Al. A magnetic field (2–8 T) is applied as this was found to further suppress mechanical vibrations in our system; field dependence of the measurements is neither expected

nor observed. Standard low-frequency ac resistance measurements were made at time constants of the order of 0.3 s. Some tunneling measurements were repeated using a dc current, and were found to reproduce the ac results. From the 1% noise level in the resistance measured in the tunnel regime at 1 MΩ with a bandwidth from dc to 10 kHz, we obtain an estimate of the level of vibrations in the distance between the two electrodes: the vibration amplitudes are typically less than 100 fm.

The evolution of the tunnel resistance that is observed when two electrodes are brought together is indicated for Pt in Fig. 1. The current modulation amplitude is 1.5 nA. The scan is recorded in 600 s and the drift in the resistance observed in a similar time interval for constant V_p is less than 5%. The piezovoltage V_p is proportional to the displacement of the electrodes with respect to each other. The proportionality factor is difficult to obtain: it depends on the mechanical configuration (displacement ratio) and the contact geometry. We estimate for our samples that 1 V corresponds to $\sim$ 0.1–0.2 Å. For resistance values above $\sim$ 500 kΩ, the resistance shows an exponential behavior with V_p, i.e., with vacuum barrier width. For decreasing V_p the tunnel resistance drops below the exponential line. At a tunnel resistance of $\sim$ 40 kΩ we find a jump to a resistance of about 8 kΩ, after which it remains more or less constant over a finite V_p range. The inset shows the tunnel-to-contact and contact-to-tunnel discontinuities for two successive cycles with Al electrodes. Hysteresis occurs in going from tunneling to contact and back, and the transitions are observed to reproduce fairly accurately, when scanning over a limited V_p range. We estimate that the difference in forward and backward jumps in this example is about 0.2 Å. The resistance value in the tunnel regime from

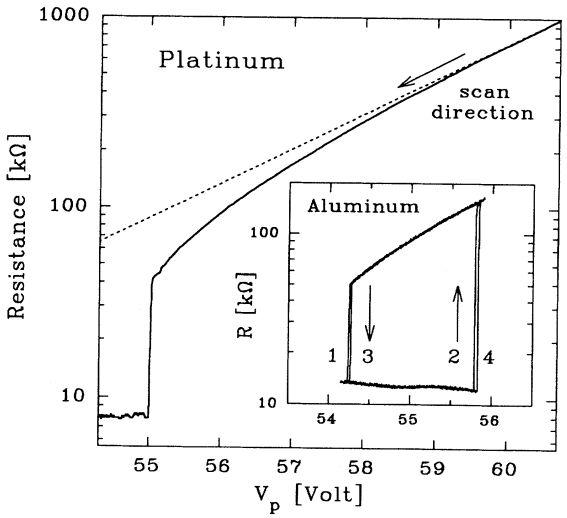

FIG. 1. Semilogarithmic plot of an ac resistance measurement of a Pt break junction at 1.3 K as a function of V_p, coming from the tunnel side. The dotted line is an extrapolation from larger V_p. Between $\sim$500 kΩ and $\sim$40 kΩ a downward deviation from exponential behavior is observed, followed by a jump to a stable value. The inset shows two successive cycles of an Al break junction at 4.2 K, illustrating the hysteresis of the jumps. The numbers indicate the sequential order of the jumps.

which the junction jumps to contact ranges between 30 and 150 kΩ and the difference in V_p for forward and backward jumps can be as small as 1 V, but values up to 30 V are also observed. A much larger hysteresis is observed for cycles that continue far into the contact regime.

After the jump to contact a resistance value is obtained of order $h/2e^2$. The systematics of these smallest contact values was investigated by carefully making or breaking contacts and recording the last resistance values on the contact side of the jump. Figures 2, 3, and 4 show measurements in the contact regime for Cu, Al, and Pt, respectively. In Fig. 2 two traces are shown of a Cu MCB junction. By moving the electrodes apart clear conductance discontinuities are observed as a result of the atom-by-atom reduction of the constriction.[14] As the electrodes are displaced over atomic distances between the two scans, some deformation in the contact region occurs. For this reason the two traces, which are measured on the same sample, do not reproduce in detail. There are, however, similarities between these two traces, which are characteristic for such measurements on Cu. The plateaus between two discontinuities show a constant negative slope. The general trend is that this slope decreases for point contacts of smaller dimensions until the plateau is horizontal at the smallest conduc-

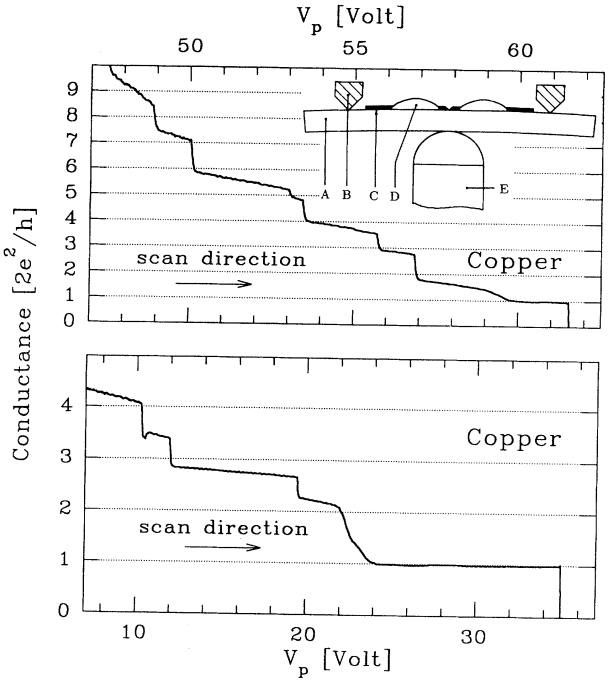

FIG. 2. Two examples of the conductance of a Cu junction at 1.3 K as a function of V_p. The transition between contact and tunneling was approached from the contact side. The conductance shows plateaus and decreases by sharp steps. The steps are interpreted as the atom-by-atom breakdown of the contact. The step size is not very reproducible, but of order $2e^2/h$. The last plateau, just before loosing contact, is nearly horizontal. The value of the last contact conductance is very close to $2e^2/h$. The sample mounting is drawn schematically in the inset, with A the bending beam, B the counter support, C the metal filament, D the epoxy adhesive, E the piezoelement.

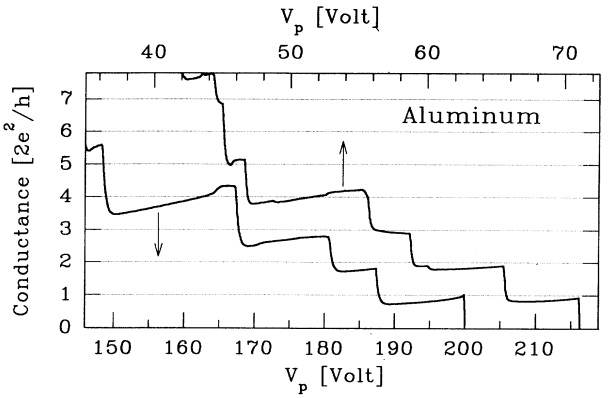

FIG. 3. Two examples of the conductance of an Al junction at 4.2 K as a function of V_p. The last plateau, just before loosing contact, has a positive slope: the conductance increases when the electrodes are moved apart. The value of the last contact conductance is close to $2e^2/h$. The scans are recorded for increasing piezovoltage.

tance value before the jump to tunneling occurs. Remarkably, for Cu this value is very close to $2e^2/h$, the conductance quantum for a single conductance channel.

For Al (Fig. 3) the last few plateaus are typically slightly curved and have a positive slope. Note that the positive slope signifies that the conductance *increases* when the electrodes are pulled apart. Again, the jump to the tunnel regime occurs at a conductance value close to $2e^2/h$.

For Pt the jump to tunneling is generally found at conductance values larger than $2e^2/h$. The two traces in Fig. 4 were recorded immediately after another, showing again hysteresis in going from contact to tunneling and back. The structure of the steps and plateaus is different

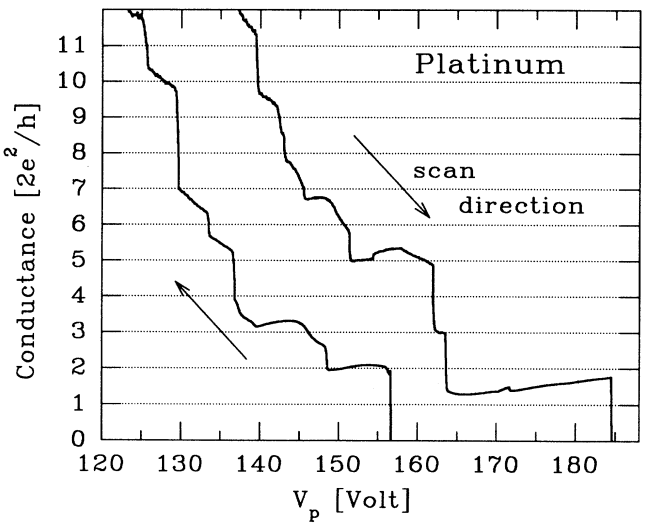

FIG. 4. The conductance of a Pt junction at 1.3 K as a function of V_p for two successive scans. In the first scan the transition was approached coming from the contact side, whereas in the second scan the transition was approached coming from the tunnel side. As for Al, the last plateau in this example has a positive slope. The value of the last contact conductance is larger than $2e^2/h$.

for the two scan directions. The plateaus for Pt have a more irregular behavior, compared to Cu and Al. For the smallest point contacts the plateaus often have an anomalous positive slope, similar as found for Al; for larger point contacts the slope becomes negative, as for all metals studied.

Figure 5 shows the conductance values at the point where the contact regime is left abruptly for a large number of measurements on Cu, Al, and Pt. For Cu the measurements show a narrow distribution centered close to $2e^2/h$. For Al the distribution is similarly centered at $2e^2/h$, with, however, a larger spreading compared to Cu. For Pt the center of the distribution is clearly located at a value larger than $2e^2/h$.

From the following experimental facts we infer that the last contact prior to the jump to tunneling consists of a single atom: (a) The interaction in the tunnel regime involves (with high probability) a single foremost atom. From experience with STM it is well known that atomic resolution can be achieved even with a "blunt" tip above a flat surface, as a result of the exponential dependence of the tunnel current with distance. (b) The hysteresis of the jump between contact and tunneling is of the order of 0.1 Å, an order of magnitude smaller than the size of an atom. (c) The steps in the conductance in the contact regime were analyzed[14] as being due to atom-by-atom changes of the contact area, and the size of the steps is comparable to that of the jump to tunneling.

Strong support for this interpretation comes from the elaborate calculations of Todorov and Sutton.[16] They find that a one-atom contact is stable (even at room temperature) and for the value of the conductance they obtain 0.93 ± 0.05 in units of $2e^2/h$. The atom was considered to be described by hydrogenlike wave functions. This is in excellent agreement with our observations for Cu, which is the element in our selection for which a sim-

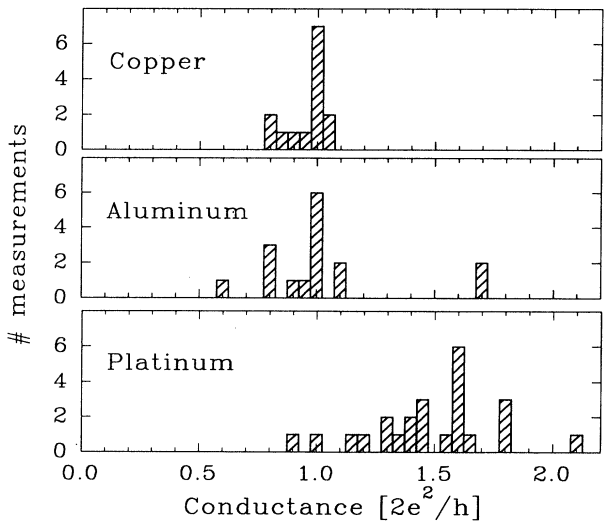

FIG. 5. Histograms of the conductance values just before the jump to the tunnel regime, in a series of measurements on Cu, Al, and Pt break junctions. Cu shows a narrow distribution centered at a value close to $2e^2/h$. Al and Pt show a larger spreading, centered at 1 and 1.6 times ($2e^2/h$), respectively.

ple s-wave function is a fair approximation. The explanation for the larger spread in the last contact values for Al and Pt, the positive slopes of the last plateaus, and, in particular, the last conductance value for Pt which is significantly larger than $2e^2/h$, needs to be sought in the atomic electronic structure. Qualitatively, one can imagine that s- and d-like wave functions for Pt constitute separate conduction channels, raising the conductance above unity. The slope of the plateaus may then result from a gradual evolution of the overlap between such states.

The remarkable fact that the electrodes can be pulled apart over a large fraction of an atomic distance, e.g., Fig. 2, with only small and gradual changes in the conductance, suggests that the last atom is suspended between the two electrodes at larger-than-bulk interatomic distances. This was also observed in the molecular-dynamics simulations.[16] It has been suggested[17] that a localized electronic state may develop at such an atom, and that resonant tunneling through this state results in a value $2e^2/h$ for the conductance.

Model calculations of the tunnel resistance as a function of electrode separation[5–8] predict a saturation of the resistance, observable at distances smaller than 3–4 Å. Such deviations from exponential behavior, due to the decrease of the effective tunnel barrier height, formed an explanation for the observations of Gimzewski and Möller[4] on Ir STM tips above a Ag surface. Later experiments, however, by Dürig, Züger, and Pohl[11] on Ir tips and surfaces, produced a purely exponential behavior. Here we find a deviation from exponential dependence which has the opposite sign compared to Ref. 4: the resistance changes faster than exponential.

A natural explanation for the rapid change in resistance is found when the metal bonding forces are taken into account, that the front atoms experience from the counter electrode.[9,10,15] This force pulls the front atoms out of their equilibrium positions, reducing the effective barrier width for tunneling, and thus decreasing the resistance with respect to the value anticipated from a strict proportionality between separation and piezovoltage. The instability is the result of the collective motion of tens of atoms around the contact: the apex atom makes the largest displacement, and the layers underneath undergo successively smaller, but non-negligible, shifts. The size of the jump and hysteresis depends sensitively on the steepness of the potentials involved. A possible saturation of the tunnel resistance found in the model calculations[5–8] may be hidden by this displacement effect.

In conclusion, with the MCB technique it is possible to follow the transition between tunneling and contact with unprecedented accuracy. The observed deviation from exponential tunnel behavior can be accounted for by considering displacements of the front atoms of the two electrodes due to their mutual attractive forces. The transition shows reproducible hysteresis. On the contact side of the transition single-atom contacts are formed. The conductance of such contacts depends on the type of metal. For a simple metal such as Cu, a well-defined value very close to $2e^2/h$ is observed, in excellent agreement with theory.[16]

We recently learned that B. Good, A. Banerjea, and J.R. Smith have extended previous calculations,[15] and confirm that one-atom contacts are stable, with the apex atom suspended at larger-than-bulk distance between the electrodes. The one-atom plateau in the conductance at $\approx 2e^2/h$ has also been found for Au contacts by Agraït, Rodrigo, and Vieira[18] using an STM-based technique.

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