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Transition from the tunneling regime to point contact studied using scanning tunneling microscopy

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Using scanning tunneling microscopy, we have studied local surface modifications induced by point contact of a tunnel tip with metallic surfaces. Two distinct types of topographical modification are found which correlate directly with the chemical condition of the tip as determined by tunneling spectroscopy. Observation of the dynamics of the transition from the tunneling regime to contact permits an evaluation of the gap distance prior to point contact. At gap spacings of $s \lesssim 3$ Å a significant decrease in the apparent tunnel barrier height is observed just before touching. Contact was found to initiate with the intimate contact of several atoms only. The implications of these results for lithography on a nanometer scale, and on the investigation of the mechanical properties of surfaces over ranges of a few nanometers, are discussed.

The starting point of intimate contact between two surfaces involves the interaction of clusters of atoms in the closest proximity.[1] It is here that the interfacial separation is similar to an interatomic spacing. However, in previous microscopic point-contact experiments, the effect observed is some average of many single-cluster contact events, and consequently single interactions are not resolved.

Scanning tunneling microscopy (STM)[2] involves the interaction of a single cluster (tip) separated by a sufficiently thin vacuum gap to permit electron tunneling to or from a surface.

The tunneling regime and point contact are separated by a variation in gap spacing of a few angstroms only. In this Rapid Communication, we discuss the transition from the tunneling regime to a single point contact between a metallic tip and surface observed using STM and related techniques. We have measured the dynamics of the transition for contact and separation of the tip from the surface. In addition, the surface morphological changes induced locally by such a procedure were recorded.

A sharp transition from tunneling to contact is observed. Contact initiates with an area of atomic dimensions. Two distinct types of surface modification are seen which correlate with the chemical nature of the tip used. These results have direct implications for our understanding of adhesion on the atomic scale, and demonstrate a useful technique for two types of nanometer-scale surface modification.

The effect of deliberate tip-surface contact in STM has been reported by Van Kempen and Van de Walle[3] who sought to evaluate tip structure from large indentations induced by contact. Preliminary observations of "touching" the surface with the tip have been reported by Abraham, Mamin, Ganz, and Clarke.[4] Interestingly, they produced both protrusions and indentations on Au surfaces $\sim$300–400 Å across, or smaller structures with distortions extending up to 700 Å away from the structure. Soler, Baro, Garcia, and Rohrer,[5] first proposed nondestructive tip contact on graphite surfaces to explain the enhancement of the amplitude of atomic corrugation observed in STM pictures. Nondestructive contact imaging forms a basis of atomic-force microscopy[6] and repulsive forces have been demonstrated to exist even during apparently normal STM imaging.[7] From these initial results, it has become clear that the separation of contact and tunneling is not always clear cut, and to this end models have been proposed to incorporate both mechanisms to act simultaneously under special conditions.[5,8]

Our experiments were conducted using a standard STM[9] mounted in a ultrahigh-vacuum (UHV) chamber ($p = 5 \times 10^{-11}$ mbar) with sample transfer and preparation chamber described elsewhere.[10] The samples used here were thick (1000 Å) Ag films deposited on a Si(111) wafer by evaporation in UHV.

The variation of the tip-sample gap separation, s, in a controlled manner was achieved as follows: Using a sample-and-hold circuit which temporarily disconnected the feedback loop of the z piezoelectric regulation circuit, we superimposed a ramp voltage which produced a controlled excursion Δz with respect to the initial tip position. This technique is similar to spectroscopic measurements of tunneling current versus tunneling voltage (I-V) at constant s,[11] except here the gap is scanned at constant voltage and the current response recorded (I_s-s).

It is particularly important to note that for the I_s-s measurement, the tip voltage V_s during the approach can be set independently of the tip voltage during regulation (V_t). To discriminate effects arising from the applied voltage and power dissipated by contact, the voltage during I_s-s measurements, V_s was lowered to a few mV. In the results presented, we found no influence of polarity or voltage within the range $V_s = \pm 20$ mV. The tunneling gap s is determined by V_t, i_t, and the effective tunnel barrier height,[2] $\phi_{\text{eff}} \approx [(dI_t/ds)/I_t \delta s]^2$. The latter quantity was determined by phase-sensitive techniques. During the measurements, the topography and ϕ_{eff} were measured continuously to discriminate between changes induced in the tip and surface structure.

First, we discuss the transition from the tunneling regime to point contact for a *clean* metallic tip and surface. Our criteria for *cleanliness* are that the measured value of ϕ is sufficiently high ($\sim$3.5–5 eV) and that the I-V characteristics (s = const) are typical for a metal. Figure 1(a) shows such a characteristic. It was found necessary to clean the tip using field emission on a separate clean Ag

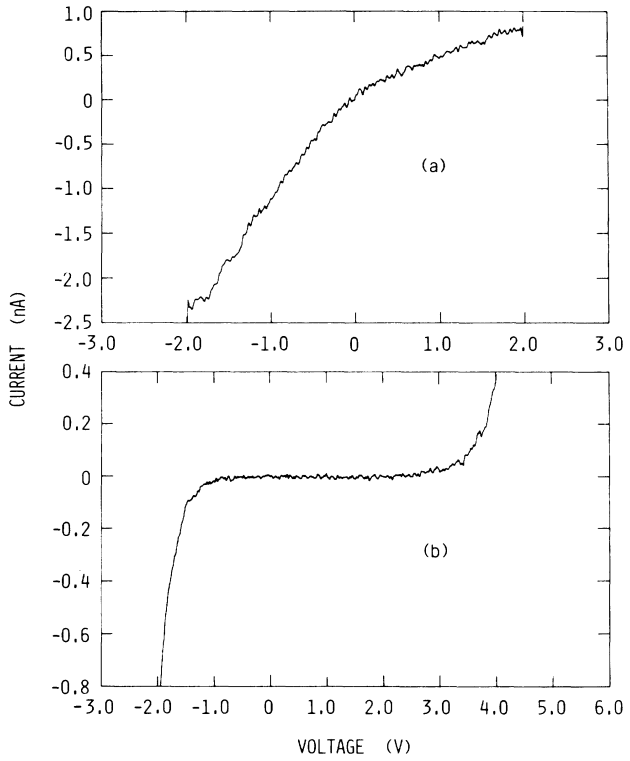

FIG. 1. Tunneling current vs voltage (I-V) for an iridium tip and polycrystalline Ag surface at constant tip-sample separation, corresponding to a constant tunnel current $i_t = 1$ nA and tunnel voltage $V_t = -1$ and -2 V. Curve (a) is for a freshly cleaned tip; curve (b) after several hours exposure to $p = 1 \times 10^{-10}$ mbar.

anode to obtain such a characteristic.

For metallic tip-sample configurations, the I-s characteristics for excursions $\Delta z = 6$ Å resulted in a current response similar to Fig. 2 ($V_t = -20$ mV, $V_s = -20$ mV, $i_t = 1.2$ nA). Compared to a typical tunneling condition with $V_t \sim 1$ V, the tunneling gap is reduced by about 2 Å because of the low tunneling voltage of -20 mV.

For $\Delta z < 4$ Å, the curve is typical for tunneling: $\Delta I = 10^3$ for $\Delta s = 4$ Å and varies to first order exponentially with the gap according to[2]

$$j \propto V e^{-A\phi_{\text{eff}}^{1/2} s} , \tag{1}$$

where ϕ_{eff} is the effective barrier height, $A = 1.025$ for ϕ_{eff} in eV, s in Å.

Upon closer inspection, Fig. 2 shows a distinct curvature. From the slopes at $z = 1$ and 3.5 Å we estimate $\phi_{\text{eff}} = (d \ln I/ds)^2$ to be 3.5 and 1.5 eV, respectively. However, at lower values of i_t, $\phi_{\text{eff}} \approx 3.5$–5 eV in the STM mode.

Although the real tunnel barrier is expected to show a marked reduction, even for $s = 10$ Å, due to the image potential, Binnig *et al.*[12] have demonstrated that at small enough values of V_t a measurement of $d \ln I/ds$ itself is nearly independent of s down to much smaller values of s, due to the fact that the $O(s^{-1})$ term in the expansion of $\sqrt{\phi_t(s)}$ cancels against s in the exponent of (1).

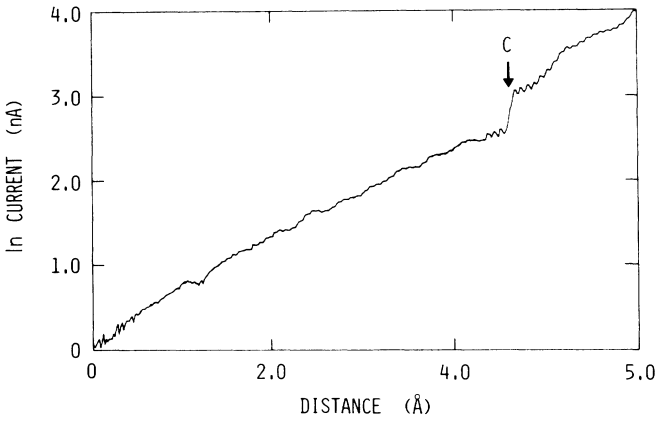

FIG. 2. Tunneling current vs distance z (I-s) for a *clean* iridium tip and polycrystalline Ag surface at constant $V_s = -20$ mV.

On the other hand, our observation of a decreased $d \ln I/ds$ is consistent with an extension of their calculations to $s \leq 3$ Å, where the electron densities at E_F overlap significantly and we reach the onset of metallic conduction.

Figure 2 is the first reported observation of this region for metal-vacuum-metal tunneling. Previous measurements involved distances $s \approx 7.4$–3.4 Å, whereas our experiments cover a much wider range $s \approx 0$–10 Å. Our estimate of s based on the position of the contact point (see below) is in overall agreement with the experiment of Binnig *et al.* ($s_0 = 7.4$ Å) when we normalize our data to his tunneling conditions ($R_t \approx 10^8$ Ω, $s_0 = 8.1$ Å). In contrast, for excursions beyond $\Delta z > 4$ Å a discontinuity is observed in the current response (see Fig. 2, point C). Typical changes in the gap resistance ΔR_{gap} are $\sim$20–1 kΩ.

For contact apertures with radius a less than the electron mean free path l ($l/a \gg 1$), the electrical resistance is given by the Sharvin resistance,[13,14]

$$R_s = 4\rho l/3\pi a^2 . \tag{2}$$

R_s is independent of the resistivity ρ since $\rho \propto 1/l$. Using (2), we evaluate an initial contact radius,[13] a, of 1.5 Å ($R_s \sim 10$ kΩ) which corresponds to the electrical resistance of one or two atoms in close contact. Therefore, we attribute the change observed in resistance ΔR_{gap}, to the transition to point contact of the tunneling tip with the surface which, by natural selection, is localized to several atoms. In addition, the atoms that support the actual contacting atoms are separated from the adjacent surface by a vacuum gap, and consequently also contribute to the current observed via tunneling. The condition $l/a \gg 1$ implies that the actual nanometer-scale contact is close to ambient temperature and that thermal processes are not involved in the observations reported.

For clean-tip conditions, excursions of $\Delta z < 5$ Å produced reversible I_s-s characteristics, whereas for $\Delta z > 5$ Å a significant hysterisis was observed; the current in the retraction direction persisted at a relatively higher level. These results suggest that adhesion occurs between tip

and sample as is expected for clean metal-metal contacts.[15,16] Subsequent topographies [Figs. 3(a) and 3(c), and 3(b) and 3(d)] showed a pronounced protrusion at the contact point, localized to dimensions of 30–100 Å full width at half maximum widths and heights of 20 Å for $\Delta z = 30$ Å. Surprisingly, the surrounding surface exhibited no observable modification or offset compared to the previous topography. In Figs. 3(c) and 3(d), fine stepped structure can be observed on the edge of the topography with heights of ~ 3 Å. These monatomic steps have a similar distribution before and after contact. This illustrates that the localized topographic changes observed are associated with the surface as expected for a hard tip material and soft surface, which show no extensive mutual solubility. We wish to include the possibility that the tip itself may contain a thin overlayer of silver previously deposited by field-emission cleaning. This may behave rather like a liquid-metal source reforming directly after contact and perhaps jumps toward the surface just before contact due to van der Waals interaction forces.

Through experience, we found that such hillocks could be used to fabricate nanometer-scale structures. However, for smaller excursions where the tip penetrated the surface by only a few angstroms, smaller structures tended to anneal out over a period of up to several minutes, probably because of surface diffusion and the nonequilibrium nature of the surface structure formed.

The results discussed so far were obtained with a freshly cleaned tip. However, when the tip was exposed about 6 h or longer to UHV, the I-V characteristics degraded to a type similar to Fig. 1(b), which we take as indicative of a contaminated tip covered by an overlayer. The characteristics are similar to a quasisemiconductor or graphitic overlayer probably caused by oxygen and/or carbon-containing species. The change in I-V characteristic is related to the tip. The Ag surface remains metallic since subsequent tip cleaning results in a return to I-V characteristics similar to Fig. 1(a), when remeasured on the same Ag sample. Hence, the changes observed in I-V characteristics were the direct result of degradation of the tip condition. It is possible that the contamination is linked to a contamination flux by gas exposure. However, adatom or admolecule diffusion along the tip shank may also be responsible for the changes observed.

For such *contaminated* tips with I-V characteristics similar to Fig. 1(b), deliberate contact with the surface produced a distinctly different type of surface modification. Figure 4 shows an area of the surface before [4(a) and 4(c)] and after [4(b) and 4(d)] a tip excursion of $\Delta z = 30$ Å at point I. The resulting nanometer-scale structure appears as an indentation rather than a protrusion. This was always found to be the case for such tips. Although the surrounding area of the image appears undisturbed, close inspection of the surface directly around the indentation suggests that material is pushed outwards locally from the indentation point. Figures 4(c) and 4(d) show enlarged areas of the modified region. It is reasonable to assume that the depression observed has partially relaxed after impact. Indeed, smaller indentations produced with $\Delta z < 10$ Å, although present during subsequent line scans, annealed out before rescanning the same area.

On the microscopic scale, it is known that the presence of thin oxide layers can dramatically lower the adhesion properties of metal-metal contacts in UHV.[17] There, however, mechanical failure at atomic clusters resulting in metal-metal adhesive contact under load complicates the interpretation. In our single-contact experiments, I-V

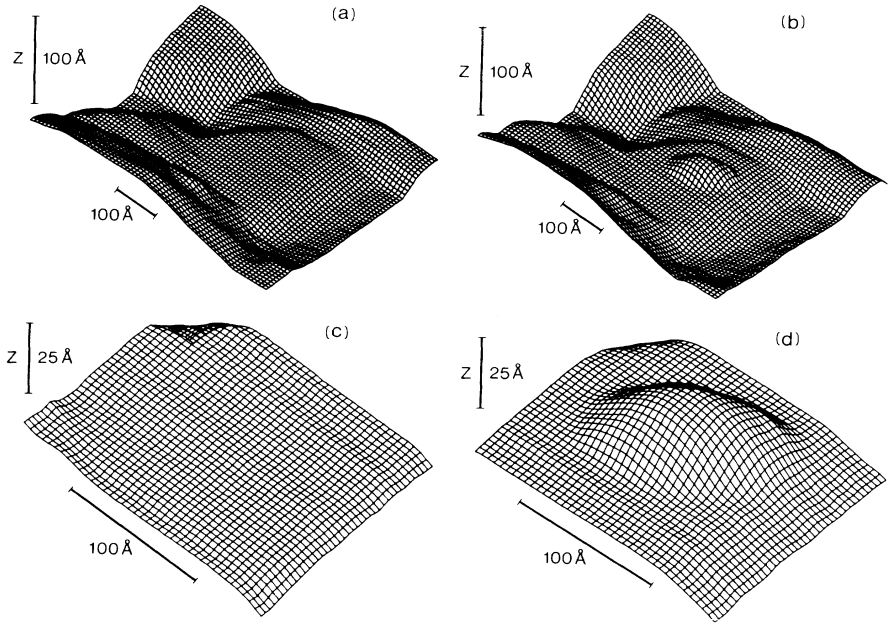

FIG. 3. An STM topograph of polycrystalline Ag with expanded-z axis: (a) before contact, (b) after contact with $\Delta z = 30$ Å. Image recorded at $V_t = 0.1$ V, $i_t = 1$ nA with I-V characteristics similar to Fig. 1(a). (c) and (d) are blowups of contact area in (a) and (b).

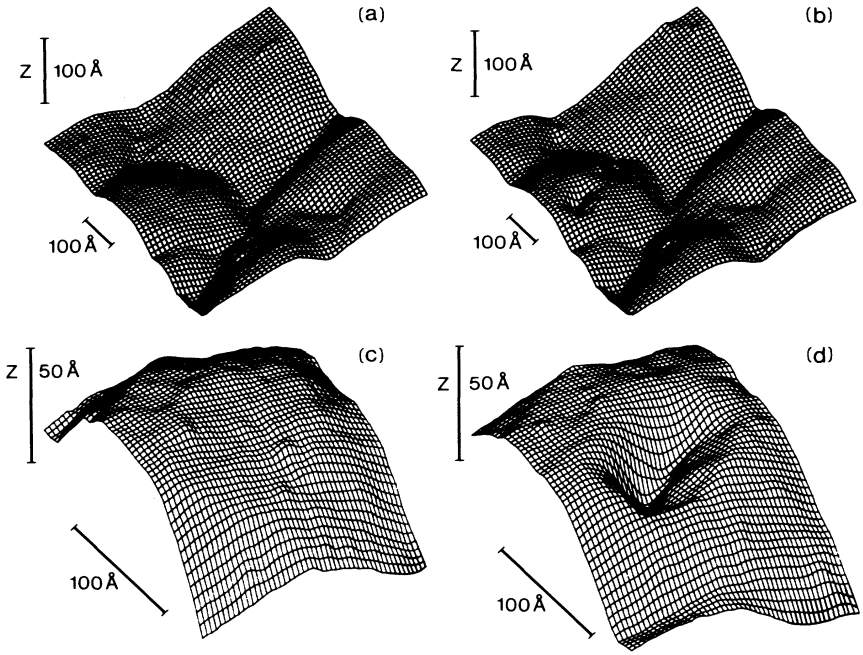

FIG. 4. An STM topograph of polycrystalline Ag with expanded-z axis: (a) before contact, (b) after contact with $\Delta z = 30$ Å. I-V characteristics similar to Fig. 1(b). (c) and (d) are blowups of contact area in (a) and (b).

characteristics taken immediately after producing nanometer-scale indentations generally showed that the *contamination overlayer* remained intact for gentle contact.

In this work, we have demonstrated two fundamental modes of surface deformation induced by single-point contact on the nm scale. Both initiate with contacts of atomic dimensions, and involve plastic (and possibly elastic) deformation. The two modes correlate directly with the chemical nature of the tip apex, as determined using tunneling spectroscopy, and are preceded by a collapse in ϕ_{eff} as determined from the measurement of $d \ln I/ds$ due to considerable overlap of the electron densities of tip and sample at E_F.

These data lead us to propose two distinct mechanisms for single atomic-scale point contacts.

For individual clean metal-metal nanometer-scale contacts, cohesive bonding between tip and sample occur. Such a result is expected theoretically[15] and found experimentally[17] for macroscopic contact. Upon subsequent tip retraction, the contact is in tensile stress, forming a neck of material which breaks, forming a localized protrusion. In contrast, for a contaminated tip such adhesive bonding is much weaker. Consequently, the tip plastically (and elastically) deforms the surface and, upon retraction, the surface forces are insufficient to pull material from the surface resulting in the indentation observed.

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