# Observation of Metallic Adhesion Using the Scanning Tunneling Microscope

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(Received 29 January 1990)

The interaction between an Ir tip and an Ir sample was investigated during normal tunneling operation of the scanning tunneling microscope. Force gradients compatible with metallic adhesion were observed within a range of 2 Å before making contact. Based on the theory of metallic adhesion, a scaling relation is derived which allows the tip geometry and the characteristics of the interaction to be accounted for in a systematic manner.

PACS numbers: 61.16.Di, 73.40.Jn, 81.40.Pq

Investigation of tip-sample interaction forces in scanning tunneling microscopy (STM) allows adhesion to be studied on an atomic level. The first experiments to characterize the interaction were performed using W tips and Ag samples.[1] As they were influenced by effects of adsorbates, the results were not easy to interpret but served to establish the potential of the method employed. Very recently, Ciraci, Baratoff, and Batra[2] carried out an *ab initio* calculation of the interaction for an Al/Al STM junction. In the present Letter, however, we shall report on measurements of the interaction for an Ir tip and an Ir sample. The material was selected since it is well suited for experimental work. In particular, fabrication of usable STM tips has been established. Moreover, the crystal lattice is extremely rigid which is reflected by the high value of the elastic modulus ($\simeq 5.2\times 10^{11}$ N m$^{-2}$). Tip and sample surfaces thus remain stable even at a gap width of less than 1 Å.

The interaction between a metal tip and a metallic sample arises from the standard quantum-mechanical description of the conduction electrons. Two regimes are distinguished according to the separation of the bodies. At large distances, electron wave functions do not overlap and the interaction is dominated by Van der Waals (VdW) forces. The leading term has a power-law dependence on distance and its exponent is dependent on geometry. At a gap width of the order of 1 Å, the electron wave functions overlap appreciably and electron exchange becomes important. This gives rise to a fundamentally different type of attractive interaction, the metallic adhesion. The corresponding forces decay exponentially over a distance comparable to the screening length.

The experimental method to study metallic adhesion is the same as in Ref. 1, namely, the use of a flexible cantilever beam (CB) as sample stage in the STM. The tip serves for tunneling and as a force probe. Varying the tunnel conditions allows the distance between tip and sample to be adjusted. The interaction influences the resonance frequency $\nu_R$ of the CB according to the force gradient $C_i$ acting between tip and sample. $\nu_R$ is determined from the spectrum of tunnel current fluctuations induced by vibrations of the CB. The vibration amplitude was restricted to 0.1 Å in order to keep anharmonic effects at a negligible level. Details concerning experimental technique as well as fabrication of the Ir CB and tip have been published in Refs. 3 and 4. It should be emphasized that the experiment was conducted in ultrahigh vacuum ($p \leq 10^{-10}$ Torr) after *in situ* preparation of the tip and the sample surfaces.

We concentrated on measuring the tunnel resistance $R_T$ and the interaction force gradient $C_i$ as a function of tunnel gap width. First, the tip was brought into tunneling contact. Initial conditions were a tunnel current of $I_T = 1$ nA and a tunnel voltage of $V_T = 20$ mV. The latter proved to be sufficiently small for electrostatic forces to produce negligible effects. The STM feedback loop was then disconnected while the tip was ramped towards the surface. $I_T$ and $\nu_R$ were simultaneously recorded during approach. The tip was retracted to its initial position as soon as the tunnel current exceeded a threshold value $I_{\max}$. Selecting this value accordingly kept the surface from being touched. After each approach cycle, the feedback loop was turned on for 100 ms to compensate for possible drifts.

The results of one such experiment are shown in Fig. 1. The curves represent averages of 64 ramp cycles. Tip excursion $z$ is measured relative to the initial tip position, i.e., $R_T = 2\times 10^7$ Ω, with positive $z$ corresponding to decreasing gap width. Reversible and nearly exponential $R_T$-$z$ characteristics were consistently observed for $z \leq 2.5$ Å. In a second experiment the gap width was decreased further, thereby disregarding the current limit $I_{\max}$. The tunnel resistance then dropped abruptly to an asymptotic value of $\simeq 10$ kΩ. In contrast to the first regime ($z \leq 2.5$ Å), reproducibility was poor and some hysteresis was observed in the retraction direction. The curve in Fig. 1(a) was complemented for $z > 2.5$ Å using data from a single scan only. A set of corresponding curves is shown in the inset, demonstrating the degree of reproducibility in this regime.

Reliable interaction-force-gradient ($C_i$) data [Fig. 1(b)] were recorded in the reversible $R_T$-$z$ regime. The value of $C_i$ was derived from measured frequency shifts by solving the differential equations for the lowest eigenmode of a spring-loaded CB. The interaction force gradient turned out to be negative over the entire range $0 \leq z \leq 2.5$ Å. For $z < 0$ Å, frequency shifts were below

 

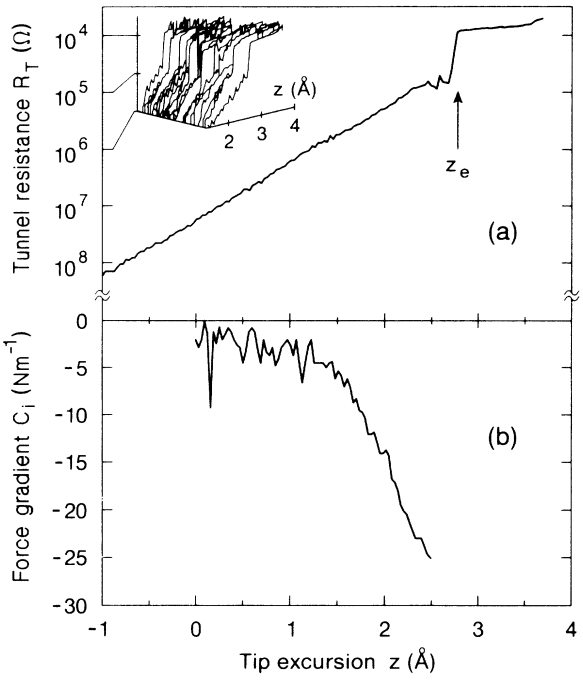


FIG. 1. (a) Tunnel resistance vs tip excursion $z$ measured on an Ir surface using an Ir tip. Positive $z$ corresponds to decreasing gap width. Transition to point contact is indicated by the arrow ($z_e$). Inset: Degree of reproducibility at the transition. (b) Interaction force gradient vs tip excursion measured simultaneously with tunnel resistance.

the detection limit corresponding to $|C_l| \simeq 1$ N m$^{-1}$. This implies that the interaction force was attractive. Integration of the $C_l$ curve yields a maximum force of $2\times10^{-9}$ N at $z=2.5$ Å.

We first discuss the resistance characteristics. The regime of small gap width has recently been investigated experimentally by Gimzewski and Möller using polycrystalline Ag as the sample material.[5] Consistent with their findings we also observe the onset of a discontinuity in the resistance-distance curve at $R_T \simeq 30$ kΩ. They suggested that the abrupt change of the resistance arises from the transition from tunneling to normal electron conduction. Correspondingly, the residual resistance $R_{\text{contact}}$ is that of a narrow point contact. $R_{\text{contact}}$ is approximately 10 kΩ in our experiment which implies a radius of contact of about $r_{\text{contact}} \simeq 1.5$ Å.[6] The latter suggests that the tip apex is formed by one atom.

A theory appropriate for simple metals[7] predicts that the slope of the $\log R_T$-$z$ curve, i.e., the apparent barrier height $\Phi$, substantially decreases as the point of contact is approached. This has been verified for Ag.[5] In the present experiment, however, $\Phi$ is approximately constant. This behavior might be the result of an accidental cancellation of the counteracting influences of barrier collapse and increasing participation of $d$ electrons in the conduction process at small distances. The latter process is likely to occur in view of the 9:1 mixture of $d$- and $p$-type states near the Fermi level.[8]

For the discussion of the force-gradient data the readily detectable position $z_e \simeq 2.8$ Å of the electrical discontinuity may be used as a convenient reference. It should be noted that, at the length scales of relevance in our experiment, the points of mechanical and electrical contact are not readily identified. Their exact positions indeed are a matter of definition. Their separation, however, cannot be much larger than the screening length.

The smallest gap width in Fig. 1(b) is $z_e - z \simeq 0.3$ Å. At this distance the electron wave functions at the apex and the sample surface overlap intensely, giving rise to short-range metallic adhesion forces. Hence, VdW-type interaction is irrelevant when we consider only the topmost atomic layer of the tip. Further from the tip apex the adhesive interaction is eventually dominated by VdW forces. Neglecting finite-size effects, the VdW force acting between the $n$th ($n \geq 2$) atomic layer of the tip and the sample surface is $F_{\text{VdW}} = H\sigma_n/6d_n^3$, where $H \simeq 2$ eV is the Hamaker constant,[9] $\sigma_n$ designates the exposed surface of layer $n$, and $d_n$ is the respective distance from the sample. Irrespective of the values of $H$ and $\sigma_n$ one can infer from the shape of the measured $C_l$ curve that the contribution of the VdW interaction is less than 10% in the range $z \geq 1.5$ Å [note that $d_n \simeq z_e - z + n\times 2.4$ Å for the most densely packed (111) planes]. Henceforth the VdW interaction will be neglected.

Comparison of our results with the theory of metallic adhesion is not readily possible since the structure of the tip determines the magnitude and distance dependence of the measured interaction. The problem can be approximately solved, however, if the following assumptions are made: In terms of a jellium model, the atomic structures of tip and sample are neglected. The relevant length scale $l_{\text{sc}}$ for the adhesion interaction is close to the screening length[10] which for most metals is small compared to the size of an atom. Hence, with regard to local variations of the electronic state, volume elements of size $l_{\text{sc}}^3$ may be considered independent entities. As a consequence, the electronic properties of curved and flat surfaces are only slightly different as long as the radius of curvature is $R_c \gg l_{\text{sc}}$.

In addition, we suppose that the interaction can be treated in terms of a two-body force model. This is a rather strong assumption which can be justified only for short-range interactions in our case. However, it allows us to relate the interaction force $F_{\text{int}}(a)$ between tip and sample to the energy of adhesion per unit area $W_{\text{ad}}(a)$ of two interacting parallel half spaces. For cylindrical symmetry, one gets[3]

$$F_{\text{int}}(a) = -W_{\text{ad}}(a)\frac{\partial\sigma}{\partial x}(0) - \int_0^\infty W_{\text{ad}}(x+a)\frac{\partial^2\sigma}{\partial x^2}dx + \frac{\partial W_{\text{ad}}}{\partial a}(a)\sigma(0). \quad (1)$$

Here the gap width $a$ is defined as the distance between

the positive background charges of tip and sample, respectively. Henceforth we use $a=0$ to define the mechanical contact, $z=z_{mc}$, and $a\equiv z_{mc}-z$. The $x$ axis is oriented normal to the sample surface and has its origin at the tip apex. $\sigma(x)$ denotes the tip cross section parallel to the sample surface. In deriving Eq. (1) we assumed $\partial\sigma/\partial x \geq 0$. This excludes tip shapes with a waist near the apex. Note that no specific assumptions concerning the nature of the two-body force need to be made. In fact, Eq. (1) is rather general, the first term having been known as the so-called Deryagin[11] approximation in the case of particle adhesion.

Rose, Ferrante, and Smith[10] calculated $W_{ad}(a)$ for simple metals using density-functional theory. They discovered that the binding energy as a function of interface separation can be mapped onto the Rydberg function using two scaling parameters, namely, the energy of adhesion $E_{ad}$ and $l_{sc}$. Later the theory was extended in a semiphenomenological way to include all metallic elements.[12,13] Based on this work we put for the adhesion interaction

$$W_{ad}(a)=2E_{ad}(1+a/l_{sc})\exp(-a/l_{sc}). \quad (2)$$

A good approximation of $E_{ad}\simeq 0.15$ eV Å$^{-2}$ is given by the solid-vapor surface tension of Ir.[14] The length scale $l_{sc}\simeq 0.42$ Å is taken from Ref. 12.

The interaction force gradient $C_i$ is calculated using Eq. (1) and taking the derivative with respect to $a$. $W_{ad}$ is provided by Eq. (2). Partial integration yields

$$C_i(\tilde{a})=2E_{ad}\exp(-\tilde{a})(2\tilde{M}_0-\tilde{M}_1-\tilde{a}\tilde{M}_0), \quad (3)$$

where linear dimensions are scaled with $l_{sc}$ (the tilde denotes scaled variables). $\tilde{M}_0$ and $\tilde{M}_1$ are the zeroth and first moments of the exponentially weighted tip cross section,

$$\tilde{M}_n=\int_0^\infty \tilde{\sigma}(\tilde{x})\tilde{x}^n\exp(-\tilde{x})d\tilde{x}, \quad n=0,1. \quad (4)$$

The exponential weighting factor indicates the sensitivity of metallic adhesion to the structure of the foremost atomic layer of the tip. Hence, force microscopy operating in the regime of adhesive forces is potentially capable of providing lateral resolution comparable to that of STM.

According to (3) a plot of $C_i/[2E_{ad}\exp(-\tilde{a})]$ vs $\tilde{a}$ should yield a straight line intersecting the $z$ axis at a point $z_0$ which is independent of the choice of $z_{mc}$. Such a plot for which the electrical discontinuity was taken as reference ($z_{mc}=z_e$) is shown in Fig. 2. The agreement with predicted values is satisfactory, which indicates that the above scaling is adequate for our experiment. Deviations from linearity for large $\tilde{a}$ suggest the beginning of a crossover into the VdW regime.

The slope and position of the straight line fitted to the experimental data provide values for $\tilde{M}_0$ and $\tilde{M}_1$. For the choice made above, $z_{mc}=z_e$, one finds $\tilde{M}_0=8.0$ and $\tilde{M}_1=21$. For comparison, $\tilde{M}_0=15$ and $\tilde{M}_1=23$ were

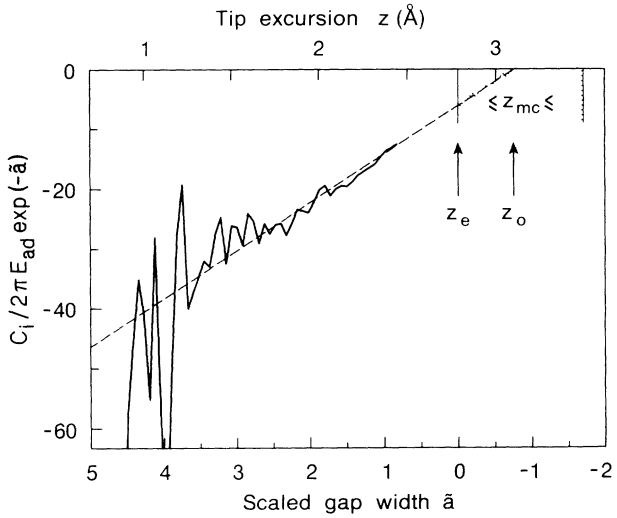


FIG. 2. Scaled interaction force gradient $C_i/[2E_{ad}\times\exp(-\tilde{a})]$ as a function of scaled gap width $\tilde{a}$ with reference to the point of electrical contact $z_e$. A straight-line fit to the data (dashed line) intersects the $z$ axis at a point $z_0$ irrespective of the choice of the reference point. The interval for the point of mechanical contact $z_{mc}$ is indicated in the figure.

calculated for a single Ir atom using the model of densely packed hard spheres. On that account the respective experimental values are somewhat on the small side. Larger values are obtained if $z_{mc}=z_e+\Delta$ is placed to the right of $z_e$ in Fig. 2. The respective geometrical parameters transform as follows:

$$\tilde{M}_n(z_e+\Delta)=\tilde{M}_n(z_e)\exp(\tilde{\Delta})\times\begin{cases}1, & n=0,\\ 1-\tilde{\Delta}\tilde{M}_0(z_e)/\tilde{M}_1(z_e), & n=1.\end{cases}$$

The upper limit $\tilde{\Delta}_{max}=[\tilde{M}_1(z_e)-\tilde{M}_0(z_e)]/\tilde{M}_0(z_e)\simeq 1.65$ follows from $\partial\sigma/\partial x\geq 0$ which implies $\tilde{M}_1\geq\tilde{M}_0$. Thus, one obtains for the point of mechanical contact $z_e\leq z_{mc}\leq z_e+0.7$ Å. It coincides with the position of the discontinuity of $R_T$ within less than $2l_{sc}$. The analysis suggests that electrical contact is established at a slightly positive gap width. In addition, we find that effective tip dimensions are compatible only with a single-atom tip, irrespective of the exact choice of $z_{mc}$ within the above interval.

As a further check of our model we also analyzed the data calculated by Ciraci, Baratoff, and Batra for the adhesion of a single-atom Al tip on a (111)-oriented Al surface.[2] Their data cover the range $0\leq\tilde{a}\leq 3$ with reference to the equilibrium lattice spacing for (111) planes. Two configurations were considered by the authors, namely, the tip placed on a top ($T$) and a hollow ($H$) site with respect to the lattice formed by the surface atoms. The $H$ geometry closely relates to the adhesion of two (111) planes in registry for which the interaction potential Eq. (2) was calculated[15] ($E_{ad}=0.715$ N m$^{-1}$ and $l_{sc}=0.66$ Å in Refs. 15 and 12, respectively).

Indeed we find that $C_i/[2E_{ad}\exp(-\tilde{a})]$ falls perfectly on a straight line for $a \geq 1$, yielding geometrical parameters $\tilde{M}_{0,1}$ compatible with a single-atom tip. The simple potential Eq. (2) does not sufficiently account for interaction effects due to the ionic cores. Deviations from the linear scaling are expected for gap widths close to equilibrium and, in particular, also when the coordination number of the tip atom with respect to the sample is different from the ideal crystal. The $T$ geometry provides an illustrative example of the latter. In fact, substantial deviations from the linear scaling law are found. Note that the effective gap width $\tilde{a}_{\text{eff}}$ defined on the basis of charge density is smaller in this case ($-1 \leq \tilde{a}_{\text{eff}} \leq 2$). In terms of the interaction the difference between the $H$ and $T$ geometries should become negligible as the gap width increases and scaling is expected to hold independent of the position of the tip.

In summary, we have investigated the metallic adhesion interaction of an Ir tip with an Ir sample under clean conditions. Based on adhesion theory a scaling relation was found which reduces the distance dependence of the interaction force gradient $C_i$ to a linear function. The latter provides information with regard to tip geometry which enters as the zeroth and first moments of the exponentially weighted tip cross section. Experimental values of $C_i$ tie in well with our model. Geometrical tip parameters require the assumption of a single-atom tip. The position of the discontinuity observed in the tunnel resistance coincides with the mechanical point of contact within 0.7 Å. The measured contact resistance suggests a contact size of atomic dimensions in agreement with the tip shape inferred from the interaction data. The above scaling also proved successful in describing the results of an *ab initio* calculation of the interaction of a single-atom Al tip with a (111) Al surface in the case where the tip was positioned on top of a hollow site.

It is a pleasure to acknowledge fruitful discussions with E. Courtens, A. Baratoff, and S. Ciraci. We thank U. Maier and U. Beck for their competent technical assistance and A. Stalder for his valuable contributions to the development of the data-acquisition software.

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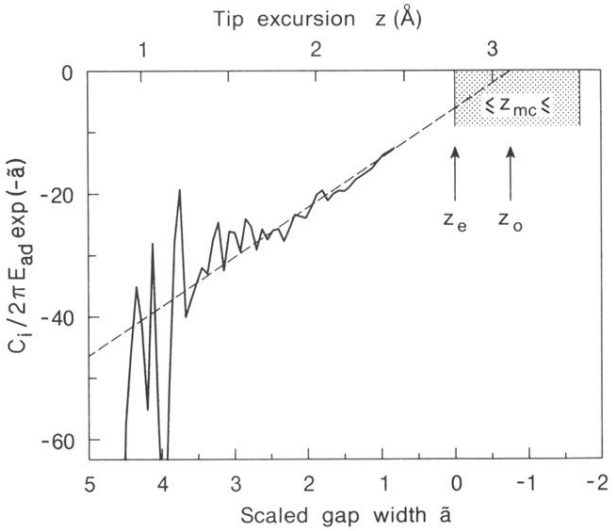


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