# Detailed experimental investigation of the barrier-height lowering and the tip-sample force gradient during STM operation in air

S. C. Meepagala and F. Real
*Department of Physics, Polytechnic University, Brooklyn, New York 11201*
(Received 25 October 1993)

Using a combined scanning-tunnel-microscopy (STM)/atomic-force microscopy (AFM) setup, we have measured simultaneously the apparent barrier height and the tip-sample force gradient for various tip-sample distances of an STM configuration in air. Our results reconfirm the existence of repulsive tip-sample forces during the STM operation with inert metal systems in air. Also, the gradient of such forces is of the correct sign that is necessary to cause the lowering of the apparent barrier heights through elastic deformations. However, our results indicate that the model that is based on the elastic deformation due to the tip-sample forces does not provide a satisfactory explanation of the lowering of the barrier heights for the case of gold in air.

In scanning tunneling microscopy, the tunnel barrier height $\phi$, in eV, can be extracted (up to a good approximation) from the expression:

$$-\sqrt{\phi}=0.976d(\ln I)/ds \ , \tag{1}$$

where $I$ is the tunnel current and $s$ is the tip-sample distance in angstroms.[1] The barrier height is generally taken as the average of the work functions of the tip and of the sample. However, it is well known that when the barrier heights are measured with scanning tunneling microscopy (STM), unusually low values are obtained. As an example, Binnig *et al.*, in their pioneering work, reported a value of $\phi=0.6$ eV for the W-Pt system, under what they called a moderate vacuum.[1] The expected barrier height for this system is about 5 eV. The situation for some other materials such as graphite is even more drastic.[2] These extremely low values of barrier heights are not due to an intrinsic deficiency that is associated with the STM, since under clean UHV conditions, reasonable values for barrier heights have been observed.[2,3] Thus, the contamination that is inevitably present on surfaces under non-UHV conditions must be responsible. It is known that surface impurities can change work-function values. However, these changes are small compared to the drastic lowering of barrier-height values that are observed commonly in STM measurements.

Improving over the model by Soler *et al.*,[5] Coombs and Pethica,[4] and independently Mamin *et al.*,[2] put forward a model to explain the lowering of barrier-height values in STM measurements. In this model, which is originally suggested for the case of graphite, there exists a repulsive interaction between the tip and the sample. This repulsive interaction is mediated by the surface contaminants that are present between the tip and the sample. Under this situation, when the tip moves a distance $\Delta z$ away from the sample, the actual increase in the tip-sample distance is $\Delta s=\Delta z+\Delta F/k$, where $\Delta F$ is the increase in the force between the tip and the sample, and $k$ is the spring constant that characterizes the elastic deformation of the tip-sample system in the tunnel direction. What the STM measures is the tip motion $\Delta z$, not the actual change in tunnel distance $\Delta s$. Thus the STM measures an apparent barrier height $\phi_a$, which is related to the barrier height $\phi$ in Eq. (1) by

$$\sqrt{\phi_a}=\sqrt{\phi}[1+(1/k)(dF/dz)] \ . \tag{2}$$

If it is taken that $F$ increases as the tip moves closer to the sample (that is, $z$ decreasing), which is the case under many typically used potentials, then $dF/dz$ is negative and therefore, the apparent barrier height that is measured by the STM is lower than the true barrier height.

This model received some support by experiments which established the existence of both the repulsive tip-sample force and the negative force gradient ($dF/dz$), for the case of graphite samples in air.[6,7] Other experiments showed that similar situations occur for silver-tungsten,[8] and gold-iridium[9] in UHV and gold-gold in air.[10] However, experiments that relate in detail the apparent barrier heights with the tip-sample force gradients have not been reported in literature. In this paper, we report an experimental attempt to relate the variation of the apparent barrier height to the tip-sample force gradient for the gold-gold system in air. During the experiment, we measured simultaneously the apparent values of tip-sample force gradient and barrier height, while the tunnel conductance was varied over a decade. Then we obtained the best straight-line fit for $\phi_a^{1/2}$ vs $dF/dz$ as suggested by Eq. (2).

The experiment was performed on a home-made STM/AFM setup that was built around an inertial sample translation scheme.[11] The setup consisted of a gold-coated microcantilever that acted as the sample as well as the force-sensing element. The sharp gold tip was positioned carefully over the cantilever, forming a tunnel junction similar to a regular STM. The deflection of the cantilever was measured by using an optical lever detection scheme.[12,13] We used previously an essentially similar arrangement to detect forces with subnano Newton resolution during an STM operation in air.[10] In the present experiment, we used a commercially available $Si_3N_4$ cantilever. The cantilever was 0.6–$\mu$m-thick,

0163-1829/94/49(15)/10761(3)/$06.00  

"V"-shaped, and had an apex-base distance of 100 $\mu$m. The cantilever was coated with gold on both sides to a total gold film thickness of 400 nm. The spring constant of the cantilever was estimated to be abound 1 N/m. The tip was formed by cutting a piece of gold wire at an angle of about 45° with the wire axis. Much care was taken with electrical shielding, to minimize the noise coupling with the tunnel current.

During the experiment the tunnel current I was held at 0.1 nA by using a constant-current feedback mechanism. The bias voltage was adjusted to several values that were in the range of 0.02–0.5 V. This voltage range corresponds to a tunnel conductance range of $0.02\times10^{-9}\ \Omega^{-1}$ to $0.5\times10^{-9}\ \Omega^{-1}$. To make a measurement, the feedback was interrupted, then the tip was moved by $\Delta z = 1.25$ nm towards the sample, and $\Delta I$, the change in tunnel current, and $\Delta F$, the change in tip-sample force, were noted. Afterwards, the tip was moved back to its original position, and the feedback was enabled again. For each bias voltage value, this process was repeated 32 times in succession, and then the data were averaged. From the measured data, the apparent barrier height and the apparent force gradients were calculated.

Figure 1 shows one plot of the square root of the apparent barrier vs the tunnel conductance for the gold-coated cantilever sample and a gold tip. Two features can be seen in the figure. First, the obtained barrier-height values are much smaller than the typical STM values, which are a few tenths of eV, that are seen in air. This is understood easily in terms of the force gradients, as can be seen from Eq. (2). In the presence of a negative $dF/dz$, $\phi_a$ will be less than $\phi$. In the case of a cantilever sample, the spring constant $k$ in Eq. (2) will be replaced by the lower value $[kk_c/(k+k_c)]$, where $k_c$ is the spring constant of the cantilever. This lower effective spring constant, in turn, will make $\phi_a$ even lower. Second, $\phi_a$ tends to decrease towards higher conductances (that is, smaller tunnel distances). One may be tempted to interpret this decrease as arising from an increase in $-dF/dz$ as the tip gets closer to the sample. After all, potentials that yield such a force behavior have been used to model tip-sample interaction.[5] If this picture is correct, $\phi_a^{1/2}$ vs $-dF/dz$ should be a straight line, with a negative slope and with an intercept that is equal to $\phi^{1/2}$.

Figure 2 shows one plot of $\phi_a^{1/2}$ vs $-dF/dz$. The data that are plotted in Figs. 1 and 2 were measured simultaneously. In Fig. 2, the solid line is the best straight-line fit to the data, by the least-squares method. The best straight-line fit has a positive slope. This is contrary to the behavior that is predicted by Eq. (2). The data have much scattering. We have seen evidence that this scattering is not due to instrumentation noise but due to some changes that are happening at the tunnel junction. We will address this issue in detail somewhere else. Since the data have much scattering, we should look at the overall behavior of many runs rather than one particular measurement. In eight out of eleven measurements, we obtained a positive slope for the best straight-line fit to $\phi_a^{1/2}$ vs $-dF/dz$. Even for the few cases where a negative slope was obtained, the intercept was far lower than the expected value of $\phi^{1/2}$. The highest value of the intercept that we have observed is 0.038 $(\text{eV})^{1/2}$. Thus our data suggest that the variation of the apparent barrier height of the gold-gold system in air, when the tunnel conductance is varied from $0.02\times(10-9)\ \Omega^{-1}$ to $0.5\times10^{-9}\ \Omega^{-1}$, is not caused by a variation in the tip-sample force gradient. However, our data up to this point do not show whether or not the force gradient plays any significant role in lowering the apparent barrier height in STM measurements.

In order to look at the effect of force gradients in lowering the apparent barrier heights we performed the following experiment. Here, we prepared a set of six cantilever tips of varying spring constants, $k_c$, by bending a Pt-Rh wire of diameter 0.075 mm. Using these six cantilever tips, and one regular Pt-Rh tip on an STM, we measured the apparent barrier heights on a gold film in air. During this measurement, the tunnel current was about 0.1 nA and the tunnel bias was 100 mV. The cantilever spring constants were calibrated by pressing against a 5-mm-long, 1-mm-wide rectangular cantilever that was made out of a 12-$\mu$m thick aluminum foil. The spring

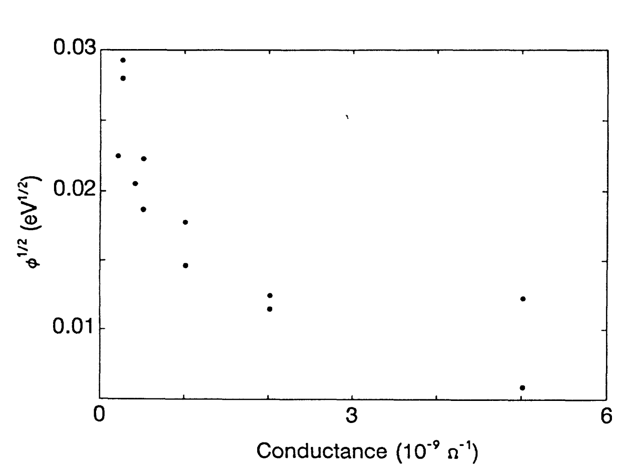


FIG. 1. The square root of the apparent barrier height vs the tunnel conductance for gold-gold system in air.

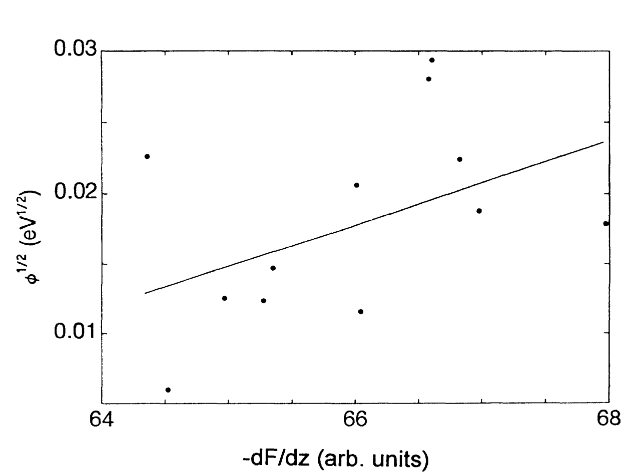


FIG. 2. The square root of the apparent barrier height vs the negative of the apparent tip-sample force gradient. The data of Figs. 1 and 2 were gathered simultaneously.

constant of this aluminum cantilever was calculated to be about 0.2 N/m. The results of this measurement are shown in Fig. 3 as a plot of $1/\phi_a^{1/2}$ vs $1/k_c$. The solid line is the best straight-line fit to the data. The relation between $\phi_a$ and $\phi$ is also given by

$$\frac{1}{\sqrt{\phi_a}}=\frac{1}{\sqrt{\phi}}\left[1-\frac{1}{k}\frac{dF}{ds}-\frac{1}{k_c}\frac{dF}{ds}\right]. \quad (3)$$

Equation (3) is analogous to Eq. (2) except that Eq. (3) involves $dF/ds$ instead of $dF/dz$. If we assume that all the tips have similar geometrical features in the tunneling vicinity, then we can assume that $dF/ds$ and $k$ are constant for all the tips, for a given tunnel conductance. Thus according to Eq. (3), $1/\phi_a^{1/2}$ vs $1/k_c$ should be a straight line with a slope that is proportional to $dF/ds$. The data in Fig. 3, even with the scattering present, conforms with this behavior with a negative value for $dF/ds$. This again confirms the existence of the negative tip-sample force gradients that can lower the apparent barrier heights through elastic deformation. However, if we take a typical value of 4 eV for $\phi$, then to fit the data, the spring constant $k$ that is associated with the sample deformation and $-dF/ds$ should be estimated to be about 15 and about 31 N/m, respectively. Following other authors, we can model the tip-sample system by a rigid punch and a sample system.[4,9] Then, by using the elastic constants of gold, the value of $k$ becomes approximately equal to $181a$ N/m, where $a$ is the radius of the punch in nanometers. Thus to obtain a $k$ value of 15 N/m, the punch radius has to be unreasonably small. Thus, although the tip-sample force gradient is of the correct sign that is necessary to lower the barrier heights through elastic deformations, the role that this force gradient actually plays must be minor for the case of gold in air. Although Lang has proposed an alternate explanation for the lowering of the apparent barrier heights, that theory is not completely satisfactory for the present case.[14] This is because Lang's calculations do not take into account a role played by the surface contaminants, while we know that the barrier-height lowering is quite drastic in the non-UHV environments. Thus we say that the occurrence of low apparent barrier heights during STM operation on inert metal systems such as gold in air is still an open problem.

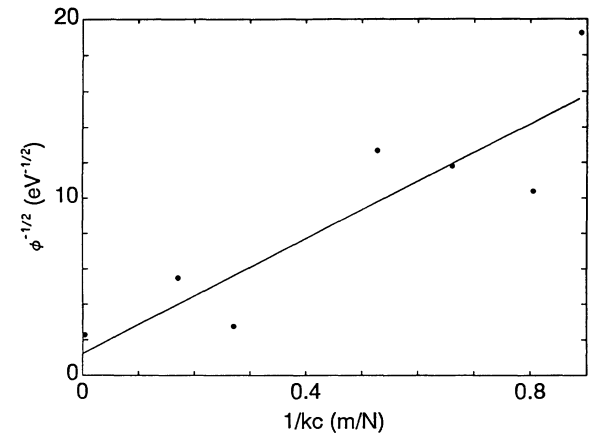


FIG. 3. The inverse of the square root of the apparent barrier height in air vs the inverse of the spring constant of cantilever-tips. The sample was a gold film and the cantilever tips were made of a wire of Pt-Rh.

In summary, we have measured experimentally the variation of the tip-sample force gradient with the apparent barrier height during STM operation with inert metal system in air. We used low tunnel conductances where we found a prominent variation of the barrier height with the tunnel conductance. Our results show that the tip-sample force gradient does not play a significant role in the lowering of the apparent barrier heights in STM measurements on gold in air.

We acknowledge useful discussions with E. L. Wolf. We are also grateful to C. B. Reyes for carefully proofreading the manuscript. This work was supported by the Office of Naval Research through Grant No. N00014-89-J-1800.

[1]G. Binnig, H. Rohrer, Ch. Gerber, and E. Weibel, Appl. Phys. Lett. **40**, 178 (1982).

[2]H. J. Mamin, E. Ganz, D. W. Abraham, R. E. Thomson, and J. Clarke, Phys. Rev. B **34**, 9015 (1986).

[3]J. K. Gimzewski and R. Moller, Phys. Rev. B **36**, 1284 (1987).

[4]J. H. Coombs and J. B. Pethica, IBM J. Res. Dev. **30**, 455 (1986).

[5]J. M. Soler, A. M. Baro, N. Garcia, and H. Rohrer, Phys. Rev. Lett. **57**, 444 (1986).

[6]H. Yamada, T. Fujii, and K. Nakayama, J. Vac. Sci. Technol. A **6**, 293 (1988).

[7]C. M. Mate, R. Erlandsson, G. M. McClelland, and S. Chiang, Surf. Sci. **208**, 473 (1989).

[8]U. Durig, J. K. Gimzewski, and D. W. Pohl, Phys. Rev. Lett. **57**, 2403 (1986).

[9]S. R. Cohen, G. Neubauer, and G. McClelland, J. Vac. Sci. Technol. A **8**, 3449 (1990).

[10]S. C. Meeagala, F. Real, and C. B. Reyes, J. Vac. Sci. Technol. B **9**, 1340 (1991).

[11]S. C. Meepagala, F. Real, C. B. Reyes, A. Nonoselskaya, Z. Rong, and E. L. Wolf, J. Vac. Sci. Technol. A **8**, 3555 (1990).

[12]G. Meyer and N. M. Amer, Appl. Phys. Lett. **53**, 1045 (1988).

[13]A. L. Weisenhorn, P. K. Hansma, T. R. Albrecht, and C. F. Quate, Appl. Phys. Lett. **54**, 2651 (1989).

[14]N. D. Lang, Phys. Rev. B **37**, 10 395 (1988).