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**EFFECTS OF TIP-BROADENING AND ASYMMETRY
ON SCANNING TUNNELING MICROSCOPE TOPOGRAPHS**

by

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Effects of tip size and asymmetry on scanning tunneling microscope topographs

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Abstract

We investigated broadening and skewing of scanning tunneling microscope topographs caused by tip size and symmetry. Model images were generated numerically by convoluting a gaussian response function for the tip with an idealized graphite image. Atomic-scale features were retained even for a 9.4 Å FWHM tip function, which demonstrated that single-atom tips are not required to obtain atomic resolution topographs. Model tips with an elliptical radial cross section produced many of the familiar skewed images of graphite. Such response functions could also be used to remove the skew from an experimental topograph, and thus provide an approximation for the shape of the physical tip.



Scanning tunneling microscope (STM) images with atomic scale features are now commonly found in both the scientific and popular literature. These STM topographs are obviously highly dependent on the nature of the tunneling tip. In most STM experiments, however, the tips used are neither well characterized nor reproducible. Tip structure characterizations before and after scanning are often possible [1-5] but impractical. Rather than characterizing the tips carefully, a typical STM experiment is often repeated with fresh tips (prepared by standard procedures [6-8]) until high resolution (usually atomic scale) features are clearly observed.

For the case of graphite images, Mizes et. al. [9] demonstrated that most of the commonly observed symmetric and skewed images could be modeled simply by manipulating three independent Fourier coefficients of a triangular lattice in reciprocal space. They conjectured that the physical reason behind the modification of the Fourier components was the presence of multiple tunneling sites on the STM tip, but did not provide any structural models of such a tip. We were interested in the maximum physical size of a single tip which could still yield atomic resolution and how the shape of the tip affects STM images.

To this end, we convoluted a model topograph of highly ordered pyrolytic graphite (HOPG) with cylindrically and elliptically symmetric tip functions which had gaussian profiles. In the case of cylindrically symmetric tips, we found that a tip blunt enough to have 20 atoms contributing substantially to the tunnel current (6 of the 20 accounting for 50% of the current flux) still yielded atomic resolution HOPG images. We also found that a single tip can produce many of the characteristic skewed images observed for graphite surfaces if the symmetry of the tip is lowered.

Finally, we applied this tip model to a real STM topograph of graphite. In particular, we deconvoluted a skewed HOPG topograph with an elliptical tip function that was chosen to produce a symmetric image and thereby obtained an approximate structural model of the tunneling portion of the tip.

We began by considering a parabolic tip that contains a reasonably large number of atoms, as illustrated in fig. 1. Here the z axis passes through the tip minimum and is perpendicular to the sample surface, and r is the radial coordinate perpendicular to the z axis. If the sample surface to be imaged is defined by $z = 0$, then the tip surface in the case of a cylindrically symmetric tip is defined by $z_t = \beta r^2 + z_0$, where z_0 is the height of the tip center above the surface (see fig. 1). The tip wave function in the region between the tip and sample can be approximated by the following (note that the complete wavefunction has an oscillatory part, which has been ignored for simplicity):

$$\begin{aligned} \Psi_{\text{tip}}(z) &\approx \exp(-\alpha(z_t - z)) && \text{for } 0 \leq z \leq (z_0 + \beta r^2) \\ &\approx \exp(-\alpha\beta r^2) \exp(-\alpha(z_0 - z)), \end{aligned} \quad (1)$$

where α is the inverse decay length ($1 \text{ \AA}^{-1}$ in this paper, which is standard for such considerations [10]). Furthermore, the density of states of the tip projected onto a surface parallel to the sample surface will have a gaussian profile, since

$$|\Psi(r)_{\text{tip}}|^2 \approx \exp(-2\alpha\beta r^2) = b(r) \quad (2)$$

for some fixed value of z between 0 and z_0 . Such a gaussian tip profile is also shown in fig. 1. Equation (3) is a generalization of the tip function for the case when the surface of the tip is a parabolic ellipsoid:

$$b(\mathbf{r}) = \exp\{-2\alpha\beta[r^2\sin^2(\theta + \delta) + r^2\sigma^2\cos^2(\theta + \delta)]\}, \quad (3)$$

where $\mathbf{r} = (r, \theta)$, δ is the angle between the tip axes and the scanning direction of the tip, and $\sigma^2 = A^2/B^2$, the square of the ratio of the semimajor to the semiminor axes of the elliptical tip cross-section.

Tersoff and Hamann derived a real-space theory for STM using a basis of spherical wave functions for the tip [10]. They derived the fortuitously simple result that the tunneling current is proportional to the state density of the surface evaluated at the center of curvature of the tip. The larger the radius of curvature, the further from the surface the state density $\rho(\mathbf{r}, E_f)$ is evaluated, and thus the smaller the corrugation that is observed in the image. If we express the state density of the tip with eqn. 2 above and derive an expression for the tunnel current as a function of position in a manner similar to Tersoff and Hamann [10], no such simple result is obtained. For heuristic convenience, we approximate the spatial dependence of the tunnel current in the following way:

$$I(\mathbf{r}_0, z_0) \propto \int \rho((\mathbf{r}_0 - \mathbf{r}), z_0, E_f) b(\mathbf{r}) d\mathbf{r}, \quad (4)$$

where $\mathbf{r}_0$ is the location of the center of the tip, $b(\mathbf{r})$ is the tip response function from eqn. (3) and $\rho((\mathbf{r}_0 - \mathbf{r}), z_0, E_f)$ is the sample state density at

the Fermi level and at a particular point (r_0, z_0) in the gap between the tip and sample.

Equation (4) should be recognized as an ideal image of the local surface charge density convoluted with a broadening function. Physically, $b(r)$ describes the width or extent of the tunneling portion of the tip. Taking the Fourier transform of eqn. (4) yields the familiar expression,

$$\begin{aligned}\mathfrak{I}(\mathfrak{I}(r_0)) &= \mathfrak{I}(\rho(r, E_f) * b(r)) \\ &= \mathfrak{I}(\rho(r, E_f)) \cdot \mathfrak{I}(b(r)),\end{aligned}\tag{5}$$

where $*$ denotes a convolution, $\mathfrak{I}$ is the Fourier transform and $\rho(r, E_f)$ is the state density at a constant height above the sample surface. Since $b(r)$ is a gaussian function in real space, $\mathfrak{I}(b(r))$ is also a gaussian in reciprocal space.

There are two limiting cases for $b(r)$, infinitely narrow and infinitely broad. In the narrow limiting case the tip approaches a discrete point probe, so that $b(r)$ approaches a delta function and the tunnel current is merely proportional to the local charge density from Fermi level states at the surface. This is exactly the result of Tersoff and Hamann for a point probe [10]. In the other limiting case the tip approaches a planar electrode and the tunnel current becomes a spatial average over the entire surface. In this paper we report the results of convolutions for gaussian tip functions of finite extent.

In order to eliminate most sources of computational noise and provide as simple a starting point as possible, a model STM topograph was synthesized for the numerical experiments in this paper. This model was a lattice of symmetric two-dimensional gaussian peaks placed so as to

represent the B sites of the HOPG surface plane [9]. Each gaussian had a full width at half maximum (FWHM) equal to 0.96 Å, which was the average FWHM of the features in the best experimental HOPG images collected at UCLA. This model starting image is shown in fig. 2A.

The tip functions used in the simulations are also two-dimensional gaussians. For the convolution calculations, various Fourier transformed gaussian tip functions were multiplied by the Fourier transform of the model topograph (each image was reduced to 64x64 pixels in order to use a standard fast Fourier transform procedure). This product of transforms was then inverse Fourier transformed back into real space to obtain the tip convoluted topographs in figs. 2 and 3.

The first set of simulations demonstrates the effect of increasingly broad, but cylindrically symmetric tip functions (i.e., gaussian tip functions with circular contours projected onto the sample plane). The spots in each image in figs. 2B-D broaden with increasing tip function breadth. The widest tip function used in our simulations broadened the ideal image (fig. 2A) until the spots began to run together noticeably but were still recognizable as features (figs. 2B-D). The corresponding gaussian tip function for fig. 2D had a FWHM of 9.4 Å. Upon integration of the tip state density function, we found that only 10% of the current passes through an area the size of one atom sitting at the center of curvature of this tip. An area the size of 6 atoms contributed 50% of the tunnel current, and an area with over 20 atoms is necessary to account for 90% of the current from the tip. This demonstrates that a single-atom tip is not a necessary requirement for atomic resolution imaging of a surface with periodic structure larger than the area of the tip through which tunneling occurs. For blunt tips, the observed atomic resolution is an artifact due to the fact

that the tunneling current from several atoms on a periodic surface can sum to yield local maxima and minima for translations of the tip corresponding to a fraction of a surface lattice spacing. In addition, it is seen from eqn. (5) that broadening the cylindrical tip function only decreases the magnitude of the Fourier coefficients of the surface image with respect to the low frequency spatial noise, but does not change the location of the Fourier coefficients in reciprocal space.

In the next set of simulations, the effects of lower symmetry tips on the ideal image of fig. 2A are investigated (i.e., tip functions with elliptical projections onto the sample plane). The tip shape was held constant with a minor axis of $B = 0.8 \text{ \AA}$ and major axis of $A = 3.1 \text{ \AA}$ at FWHM. The tip function was then rotated with respect to the substrate image in 10 degree increments before each convolution. The resulting image simulations are shown in fig. 3. Because the tip function is no longer cylindrically symmetric, the convolution images in figs. 3A-D contain rows of spots which are smeared together along a preferred direction. As the orientation of the elliptical tip changes, the images change from a nearly rectangular spot pattern to a zig-zag row pattern and finally to a straight row pattern. All of the model images in figure 3 resemble experimentally observed STM topographs collected for HOPG.

The cylindrical and elliptical tips can account for the experimentally observed close packed spot patterns, rectangular spot patterns and row-like patterns. It is seen from (5) that convoluting the original symmetric image with an elliptical tip response function is the same as changing the relative amplitudes of the Fourier coefficients of the image. In their paper, Mizes et. al. [9] changed both the amplitudes and phases of the Fourier coefficients, and thus they were also able to generate three-fold symmetric images from

an initial six-fold pattern. An elliptical tip cannot produce images with three-fold symmetry, and thus such images cannot arise from a simple linear distortion of the tip. Such images may result from more complex tip shapes, multiple tips, or other causes.

In our final demonstration we show how the effects of an asymmetric tip can be removed from a skewed experimental image of HOPG. Mathematically, removing tip effects involves a deconvolution, which is straightforward if one knows either the tip symmetry or the expected image symmetry. We have taken a typical constant height STM image of HOPG with a row-like pattern superimposed on a honeycomb-type structure and attempted such a deconvolution by assuming that the resulting deconvoluted image should be closer to a symmetric honeycomb image. The tip function used in the deconvolution was a gaussian with an elliptical cross section where the major axis had a FWHM of 7.3 Å and the value of σ in eqn. (3) was 4. This tip function was also rotated $\delta = 7.5$ degrees with respect to the scan direction. The original STM image is shown in fig. 4A and the deconvoluted image is shown in fig. 4B. Because the deconvolution with this tip function removed the row-like skew in the original image, we conclude that this function is a good approximation of the physical shape of the tunneling tip in the original experimental image. Values of σ and δ can be determined quite easily from symmetry considerations, but more quantitative information about the actual surface corrugation is required to determine β .

Therefore, if one knows the surface charge density for a particular regular surface, image deconvolutions can provide useful information about tip breadth and geometry. During STM experiments with deliberate tip-sample interactions (such as nanolithography experiments), the tip

may undergo many geometric changes during the course of an experiment due to contact or exchange of material with the sample. If such a surface were dominated by a regular surface mesh, the effects of tip asymmetries would be readily apparent in the regions of the unmodified surface. One could then deconvolute the image with an asymmetric tip function which would render the overall surface mesh symmetric again. In this way, the effects of tip asymmetries can be removed from experimental data.

In conclusion, we demonstrated that single atom tips are not a necessary requirement for atomic scale imaging in STM. Model tips with an area equivalent to 20 or more atoms contributing to the tunnel current can produce atomic scale features on large area surfaces with periodic structure. Experimental evidence for this assertion has recently been presented by van Loenen et. al. [11], in that they see ghost images of blunt STM tips at defects but can observe atomic scale features on the flat portions of silicon surfaces. Furthermore, elliptical tips were shown to reproduce many of the common skewed HOPG images observed experimentally, and thus tip asymmetries also provide a physical model for how the Fourier coefficients of an ideal STM image can be modified. Finally, deconvolutions with model tip functions to remove asymmetries from an experimental HOPG image can be used to determine an approximate structural model of the tunneling portion of the tip.

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FIGURE CAPTIONS

Figure 1 : Model of a tunnel junction with a parabolic tip function. The shape of the tip is described by a parabola, $z_t = \beta r^2 + z_0$, where z_0 is the height of the tip center above the surface. Since the tunnel current depends exponentially on tip-sample separation, the radial dependence of the density of states of the tip projected onto any plane parallel to the sample surface can be approximated by a gaussian :

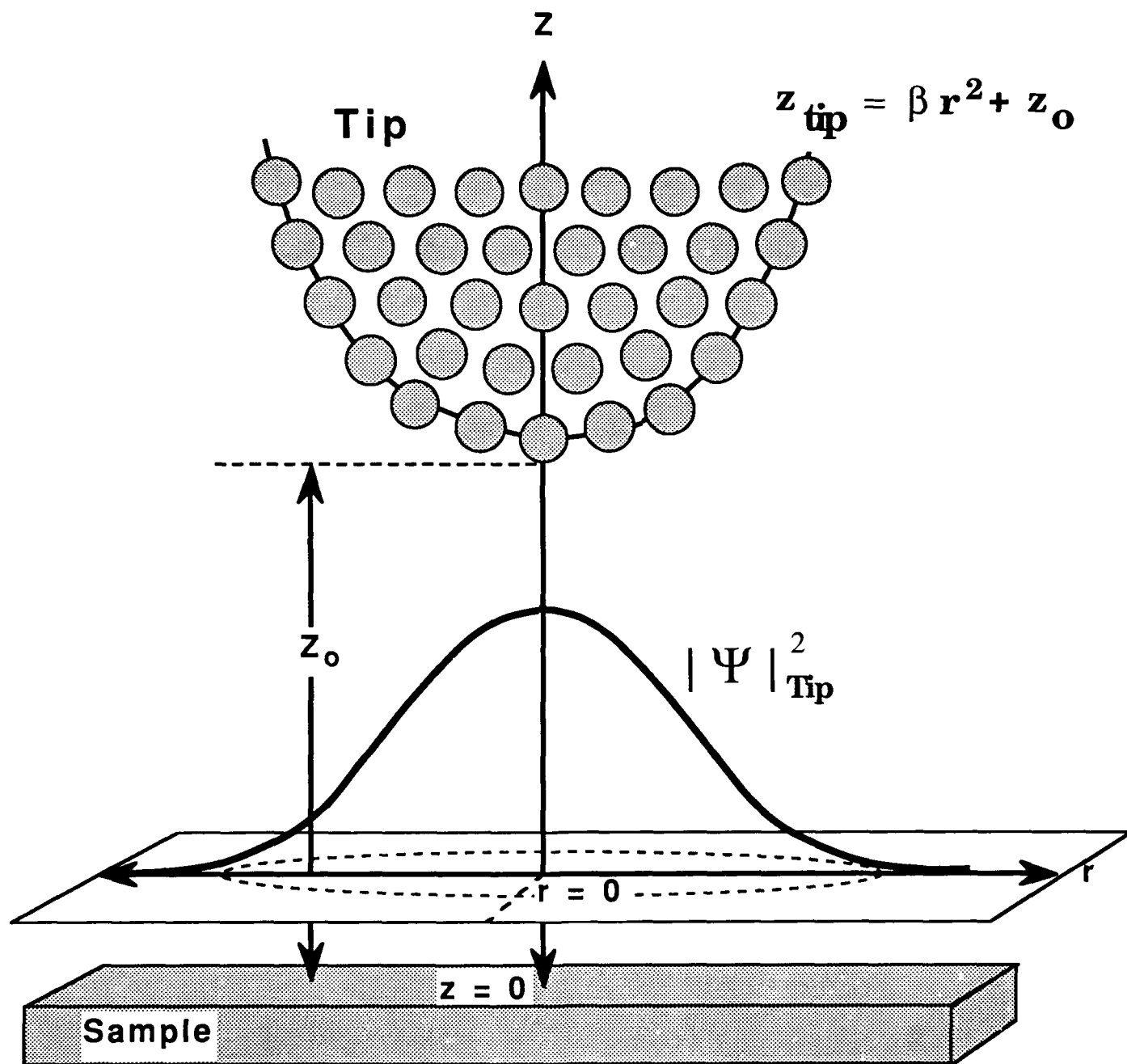
$$|\Psi(r)_{\text{tip}}|^2 \approx \exp(-2 \alpha \beta r^2),$$

where α is the inverse decay length of the tip wavefunction.

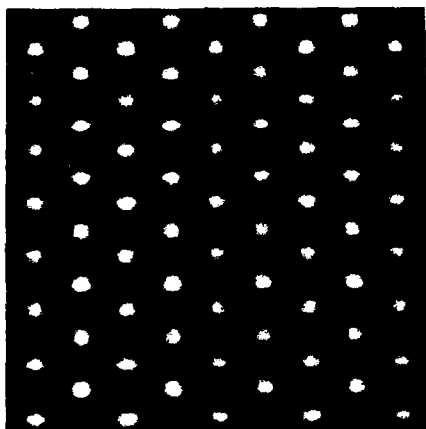
Figure 2: Original model HOPG starting image is shown in (a); (b-d) have been convoluted with a cylindrically symmetric gaussian tip function with a FWHM of 3.8Å, 7.5Å, and 9.4Å, respectively. The slight skewing of the spots and the intensity alternations are artifacts of the finite size of the 2-D Fourier transforms used in the calculations. The length scale is such that the distance between the spot centers is 4.92Å.

Figure 3: Image 2 (a) convoluted with an elliptical gaussian tip function with an angle of rotation $\delta = 0$ degrees with respect to the scanning direction and a ratio of semimajor to semiminor axes of $\sigma = 4$ is shown in (a). Images (b-d) are convolutions with the same tip rotated by 10, 20, and 30 degrees, respectively.

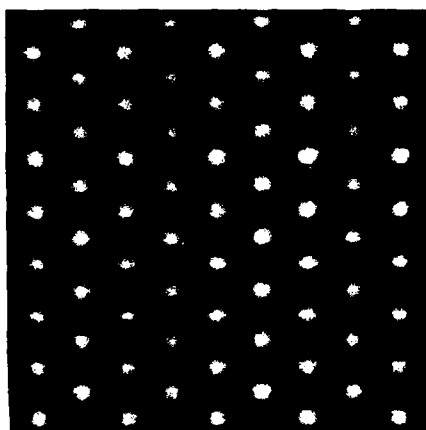
Figure 4 : Skewed experimental HOPG honeycomb image is shown in (a). The deconvolution of image (a) with an elliptical tip function (angle of rotation $\delta = -7.5$ degrees, ratio of semimajor to semiminor axes $\sigma = 4$) is shown in (b). The tip function for the deconvolution was chosen so that the resulting image would be symmetric.



a



b



c



d

