

TUNNELING SPECTROSCOPY: SURFACE GEOMETRY AND INTERFACE POTENTIAL EFFECTS

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The dependence of the oscillations observed in scanning tunneling microscopy of the tunneling conductance with applied bias voltage on the tip's curvature and the interface potential has been studied. A trial and error analysis of the distance-voltage and conductance-voltage characteristics is used to determine both the radius of the tip and the parameters fixing the interface potential. The effect of asperities in the tip and sample on the oscillations and the dependence of the oscillations on the sign of the applied bias are also discussed.

1. Introduction

The scanning tunneling microscope [1] (STM) can be mainly used in two different modes; in one case the tip is displaced along the surface keeping the tunneling current, I , and the applied voltage, V_a , constant, in such a way that the z -dependence of the tip displacement yields the electronic surface topography [2,3] of the sample; in the second mode the tip is displaced in the perpendicular direction to the surface, keeping either I or V_a constant [4,5]. Using this second mode, constant current mode distance-voltage, s - V_a , and conductance-voltage, dI/dV_a - V_a , characteristics are typically measured [5,6] at a given point of the sample.

The first mode has been used abundantly to determine surface geometries [2,3]; the second one

provides a fine tool to measure electronic properties of the sample [5-9]. Recently, Bono and Good [7] have stressed that the information [4] obtained in the s - V_a and dI/dV_a - V_a characteristics could be used to calculate in detail the interface potential that an electron sees when it tunnels between the tip and the sample. Indeed, different researchers [4,7,10] have correlated the oscillations appearing in the dI/dV_a - V_a curves with the induced potential at the tip-sample interface.

The purpose of this paper is to extend previous [7-9] theoretical approaches of the s - V_a and dI/dV_a - V_a characteristics and to present an interpretation of the experimental data for the W-Au metal-metal interface [6], which includes not only a discussion of the interface potential but also of the interface tip-sample geometry. One of the aims of this paper is to show how interface geo-

metrical effects have to be included to have the appropriate interpretation of the experimental results: we shall show that inclusion of geometrical effects becomes more and more important to understand the experimental data for long tip-sample distances; moreover, an analysis of the $s-V_a$ and dI/dV_a-V_a characteristics allows us to deduce the tip radius of curvature.

In section 2 we consider tunneling between two planar metallic electrodes, using the jellium model to represent them, and we present simple results for some one-dimensional models of the tip-sample interface; most of these results are known from independent sources [7,10] and will be presented concisely. In section 3 we present our full analysis of the three-dimensional models, taking into account the tip-sample geometry, together with the calculations of $s-V_a$ and dI/dV_a-V_a characteristics of a W-Au interface. Finally, in section 4 we present our conclusions.

2. One-dimensional models

2.1. General considerations

In this section we analyse the $s-V_a$ and dI/dV_a-V_a characteristics for the following one-dimensional interface potentials, referenced to the vacuum level of the tip, which occupies the $z \leq 0$ half-space:

(a)

$$V(z) = \begin{cases} U_L, & z \leq 0, \\ -(V_a + \Phi_L - \Phi_R)z/s, & 0 \leq z \leq s, \\ U_R, & s \leq z, \end{cases} \quad (1)$$

(b, c)

$$V(z) = \begin{cases} U_L, & z \leq z_1, \\ -(V_a + \Phi_L - \Phi_R)z/s + V_{im}(z), & z_1 \leq z \leq z_2, \\ U_R, & z_2 \leq z, \end{cases} \quad (2)$$

and [11]

(d)

$$V(z) = \begin{cases} \frac{U_L}{A_L e^{B_L(z-z_L)} + 1}, & z \leq z_L, \\ -\frac{1}{4} \left[\frac{1 - e^{-\lambda_L(z-z_L)}}{z - z_L} + \frac{1 - e^{-\lambda_R(s-z_R-z)}}{s - z_R - z} \right] \\ + \frac{\mu}{4d} \left[2\gamma + \Psi\left(1 + \frac{z - z_L}{d}\right) + \Psi\left(2 - \frac{z - z_L}{d}\right) \right] \\ - (V_a + \Phi_L - \Phi_R) \frac{z}{s}, & z_L \leq z \leq s - z_R, \\ \frac{U_R}{A_R e^{B_R(s-z_R-z)} + 1}, & s - z_R \leq z. \end{cases} \quad (3)$$

Unless stated otherwise, atomic units are used throughout.

These potential barriers are shown in fig. 1 for $\Phi_L = 5.2$ eV, $\Phi_R = 4.4$ eV, $U_L = -13.2$ eV, $U_R = -9.9$ eV, $V_a = 4.0$ V and $s = 10$ Å. Cases (a), (b) and (c) are shown in fig. 1a. Case (a) corresponds to the simple case of a linear potential between the electrodes L and R; U_L , Φ_L and U_R , Φ_R represent the bulk inner potential and the work function of a W tip and a Au sample, respectively, V_a is the applied voltage, and s is the tip-sample distance. In cases (b) and (c) we have considered the image potential, $V_{im}(z)$; this image potential includes in case (b) just the contribution from the first image charge that the electron sees in each electrode:

$$V_{im}(z) = -s/4z(s-z). \quad (4)$$

Case (c) includes all the image charges following Simmon's approximation [12],

$$V_{im}(z) = -1.15 \ln 2 s/4z(s-z). \quad (5)$$

z_1 and z_2 in eq. (2) represent the positions where the interface potential and the bulk inner potentials become equal.

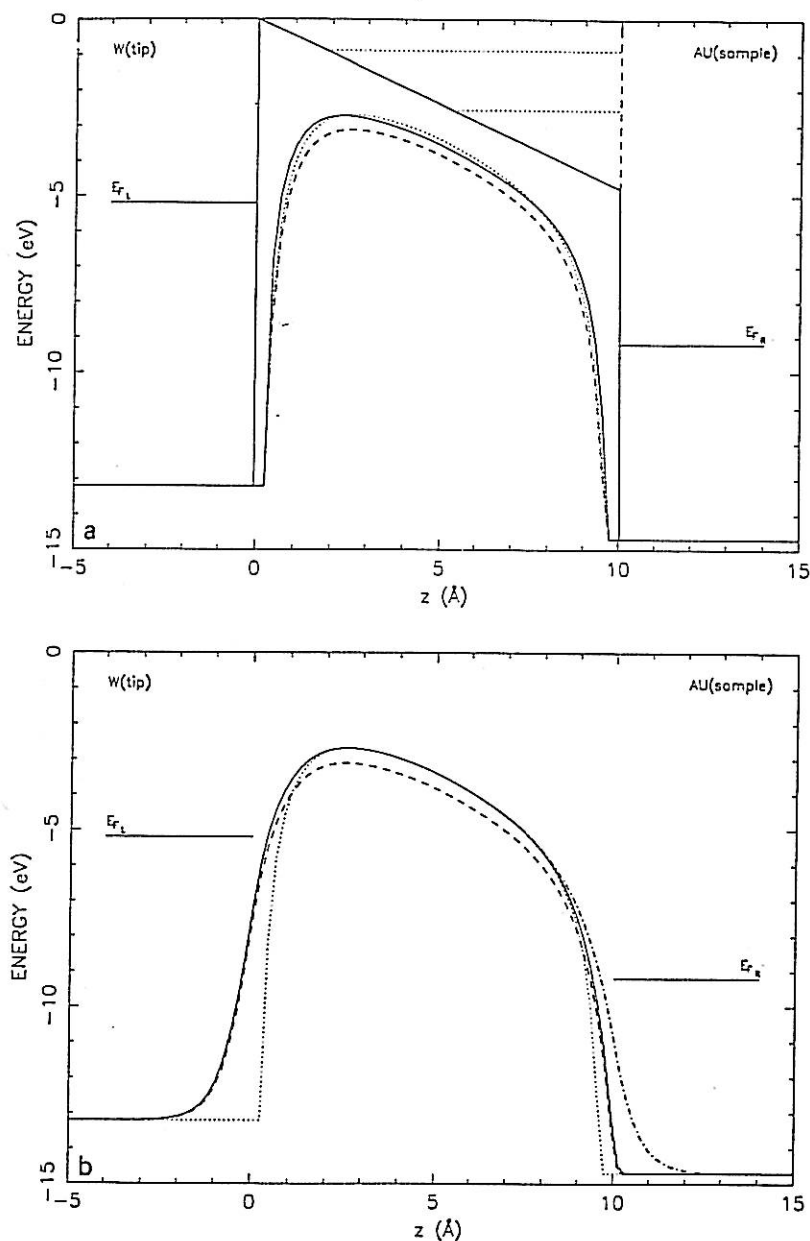


Fig. 1. Model potential barriers of eqs. (1), (2) and (3). (a) trapezoidal barrier of slope equal to the electric field between two planar electrodes, given by eq. (1), and image reduced potential barrier given by (2) and (5) (solid lines). Also shown are the image reduced potential barrier given by (2) and (4) (dashed curve), the exact classical image reduced potential barrier given by (2) and (6) (dotted curve), and the bound energy levels of the corresponding infinite barrier (horizontal dotted lines). (b) Potential barrier of eq. (3) with $\lambda_L = 1.0 a_0^{-1}$, and $\mu = 1.0$ (solid curve) and $\mu = 0.0$ (dashed curve). Also shown are the same potential barrier with $\lambda_L = 1.0 a_0^{-1}$, $\lambda_R = 0.9 a_0^{-1}$ and $\mu = 1.0$ (dashed-dotted curve), and the exact classical image reduced potential barrier of eqs. (2) and (6) (dotted curve).

The exact expression for the classical image potential between the electrodes may be written as [13]

$$V_{\text{im}}(z) = -\frac{1}{4} \left(\frac{1}{z} + \frac{1}{s-z} \right) + \frac{1}{4s} \left[\sum_{n=1}^{\infty} \left(\frac{2}{n} - \frac{1}{n+z/s} \right) - \sum_{n=2}^{\infty} \frac{1}{n-z/s} \right], \quad (6)$$

which, after some rearrangement, can be rewritten as

$$V_{\text{im}}(z) = -\frac{s}{4z(s-z)} + \frac{1}{4s} \left[2\gamma + \Psi\left(1 + \frac{z}{s}\right) + \Psi\left(2 - \frac{z}{s}\right) \right], \quad (7)$$

where γ is the Euler number, and $\Psi(x)$, the digamma function [14].

The interface potential appears as the result of different local and non-local effects [15–18]; non-local effects are reflected in the long-range image potential induced by both surfaces, while local effects, which can be obtained using a local density approach (LDA), are responsible for the fact that the effective potential barrier that the electron feels goes more smoothly to its bulk value than the classical image reduced barrier. On the other hand, although for large distance between the electrodes the effective potential barrier should converge in the middle of the gap to the barrier specified by (2) with the image potential given by (6), it has been shown [16] that for tip-sample distances of a few ångströms the barrier is substantially decreased by the overlapping of the metal electronic charges; in addition, dynamic effects tend to reduce the strength of the induced image potential, depending on the relation between the tunneling transversal time and the response time of both metals (typically given by the inverse of their plasmon frequencies). For very short tunneling transversal times the image potential would be reduced to zero, but theoretical calculations [19,20] show that for typical tip-sample

distances the image potential is just reduced to around 70% of its full classical value.

To take into account the physical basis of the problem discussed above we generalize the parametrized surface potential recently suggested by Jones et al. [21], and propose the barrier model specified by eq. (3). A_L , B_L , A_R and B_R in this expression are determined by matching $V(z)$ and its derivative at the image planes positions, $z = z_L$ and $z = s - z_R$, which represent the centre of mass of the induced charge distribution relative to the edge of the positive background; d is the gap distance, which is defined to be the distance between the image planes of the two metals. The quantities λ_L and λ_R in this expression determine the range of transition from the vacuum potential to its value in the bulk; for $d \geq 6$ Å they have a simple physical interpretation, since they tell us about the value of the potential at the image planes:

$$V(z_L) \approx -\left(\frac{\lambda_L}{4} + \frac{1-\mu}{4d} \right) - (V_a + \Phi_L - \Phi_R) \frac{z_L}{s}, \quad (8)$$

$$V(s - z_L) \approx -\left(\frac{\lambda_R}{4} + \frac{1-\mu}{4d} \right) - (V_a + \Phi_L - \Phi_R) \frac{s - z_R}{s}. \quad (9)$$

The parameter μ tells us about the dependence of the barrier height on the distance between the image planes, since

$$V\left(z_L + \frac{d}{2}\right) \approx -\frac{1-\mu(1-\ln 2)}{d} - \frac{1}{2}(V_a + \Phi_L - \Phi_R) \left(1 + \frac{z_L - z_R}{s}\right). \quad (10)$$

Consequently, $\mu(1 - \ln 2)/d$ represents the increase of the potential barrier height given by (3) over the one given by eqs. (2) and (4), so that in the tunneling region, far outside the image planes, the barrier of eq. (3) converges to the barrier of eq. (2) with the image potential specified by (4), (5) and (6) when $\mu = 0.0$, $\mu \approx 0.66$ and $\mu = 1.0$, respectively.

To clarify the origin of the experimentally observed oscillations appearing in the constant cur-

rent mode conductance-voltage characteristics and the dependence on the parameters defining the surface potential, first we consider the infinite potential barrier case. The energy levels are then given by

$$E_n = -(2F^{-2})^{-1/3} a_n, \quad (11)$$

referenced to the bottom of the barrier, where F is the field, and a_n , the zeros of the Airy function [14], the asymptotic expansion of which is

$$a_n = - \left[3 \frac{\pi}{2} \left(n - \frac{1}{4} \right) \right]^{2/3} \times \left\{ 1 + \frac{5}{48} \left[3 \frac{\pi}{2} \left(n - \frac{1}{4} \right) \right]^{-2} + \dots \right\}. \quad (12)$$

The bound levels of the infinite barrier, shown by dotted horizontal lines in fig. 1a, will broaden and turn into transmission resonances when the electrons tunnel the trapezoidal finite barrier. This happens because of the interference between incident and reflected electrons in the positive kinetic-energy region of the vacuum gap for energies close to the originally bound levels.

If the WKB approximation is used to calculate the transmission coefficient across potential barriers, no resonance phenomena as the ones described above will appear, because the effect of the partially reflecting interface at $z = s$ is neglected. In order to explain such resonances, we have to solve the Schrödinger equation across the potential barrier. The transmission probabilities for the trapezoidal barrier are calculated by connecting the Airy functions, exact solutions of the Schrödinger equation through this barrier, at both interfaces [22]; then, the peak energy positions for the transmission probabilities are obtained.

We find that the positions of the transmission resonances for the finite trapezoidal barrier coincides for energies below the vacuum level with the bound levels of the infinite barrier discussed in eq. (11). However, when the lowered trapezoidal barrier tangent to the image reduced barrier of eq. (2) is considered, the positions of the transmission resonances are lowered by $[1 - \mu(1 - \ln 2)]/s$, where μ is equal to 0.0, 0.66 and 1.0 depending on which image potential we consider, (4), (5) or (6), respectively.

In the case of the image reduced potential of eq. (2) the peak energy positions can be calculated solving numerically the Schrödinger equation, and it may be shown that the first transmission resonances are less pronounced and appear at smaller energies than those obtained for the corresponding lowered trapezoidal barrier; moreover, when the more realistic potential of eq. (3) is considered, one finds that the smoother the potential in the positive kinetic energy region is the less pronounced the transmission resonances become. Thus, the peak energy positions of the transmission probabilities, and consequently, the experimentally measured distance-voltage and conductance voltage characteristics are very sensitive not only to the height of the barrier, which is determined by the μ parameter in eq. (3), but also to the shape of the barrier in the positive kinetic energy region, which is determined by λ_R . Below we elaborate this further.

If the experimental conditions are such that the applied voltage and the tip-sample distance are modified simultaneously keeping the electric field constant, the number of transmission resonances appearing below the tip's Fermi level increases with the tip-sample distance. Then, an oscillation in the $dI/dV_a(V_a)$ curve will appear each time an additional resonant level crosses the tip's Fermi level as the applied potential increases. The experimentally observed oscillations in dI/dV_a , as well as the transmission resonances occurring in the region close to the sample-vacuum interface, are very sensitive to the detailed form of the potential barrier between the electrodes. In particular, the peak positions and the shape of the oscillations in the $dI/dV_a - V_a$ characteristics depend critically on the barrier height and on the shape of the barrier in the positive kinetic energy region, so that both parameters μ and λ_R in eq. (3) play an important role in the study of such oscillations. In addition, the dependence of the tunneling current on μ is, also, important, while the dependence of the current on λ_L and λ_R is negligible.

Therefore, we fix λ_L and consider λ_R and μ as two fitting parameters. Fig. 1b exhibits the model barrier of eq. (3) for $\lambda_L = 1.0 a_0^{-1}$ ($a_0 = 0.529 \text{ \AA}$) and different values of the λ_R and μ parameters,

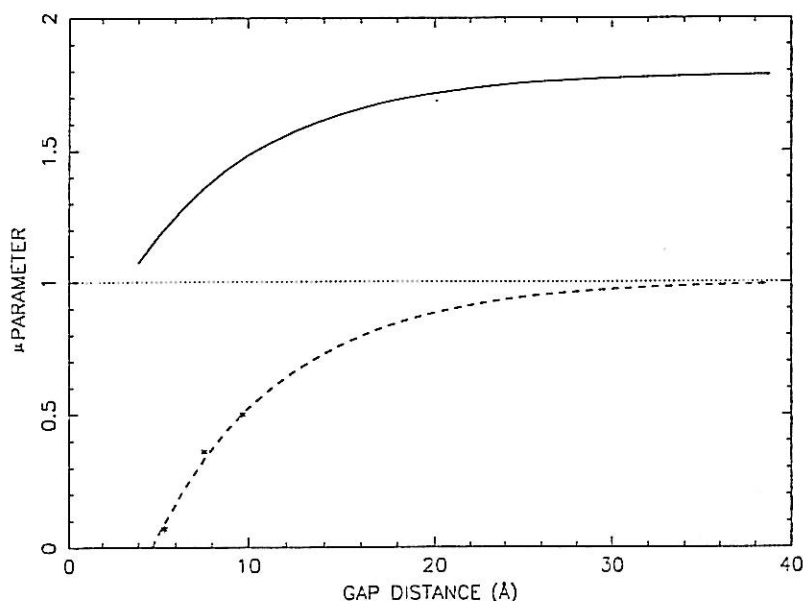


Fig. 2. Dependence on the gap distance of the parameter μ that makes the potential barrier of eq. (3) to fit at small distances the potential barrier of ref. [16] and to converge, at greater distances, to the exact image reduced classical barrier (dashed line); the cross points show the values of the parameter μ making the potential barrier of eq. (3) to fit the potential barrier of ref. [16] for gap distances of 5.4, 7.6 and 9.7 Å, and the solid curve represents the parameter μ dependence on the gap distance used in our calculations of figs. 4, 8 and 9, having assumed that the image potential is reduced by dynamical effects by 65%.

together with the exact classical image reduced potential barrier of eqs. (2) and (6).

The model barrier of eq. (3) fits the theoretical potential of De Andrés et al. [16], which includes both local and non-local effects, by appropriately choosing λ_L , λ_R and μ ; in particular, for $d \approx 9$ Å a good fitting is obtained between the theoretical potential of De Andrés et al. and the parametrized barrier of eq. (3) for $\mu = 0.5$. A good fitting is, also obtained using smaller values of μ when $d < 9$ Å, which shows that for small tip-sample distances the barrier is substantially decreased due to the local exchange and correlation energy, and it does not converge to the exact classical image reduced barrier of eqs. (2) and (6). However, these local effects depend on the tip-sample distance and become negligible for large distances, indicating that a dependence of the μ parameter on the gap distance should be included. For big gap distances μ should converge to unity, if dynamical effects are neglected. As pointed out above, the static μ parameter that fits the potential of ref. [16] should be enhanced in order to take into account these dynamical effects. Fig. 2 exhibits μ as a function

of the gap distance, for the static and dynamical cases, having assumed that the image potential is reduced to 65% of its full value due to dynamical effects.

We have found [23], also, that the apparent barrier height calculated with the model interface potential specified by eq. (3) is, for short tip-sample distances ($d < 6$ Å), in good agreement with Lang's calculation [15]; at greater distances, our potential yields the appropriate barrier height associated with image effects.

2.2. One-dimensional results

When planar electrodes are considered, the tunneling current is the product of the current density times the tunneling area. The current density is given by [13]

$$J = \frac{1}{2\pi^2} \left[V_a \int_{-U_L}^{-\Phi_L - V_a} dE_z T(E_z) + \int_{-\Phi_L - V_a}^{-\Phi_L} dE_z (-\Phi_L - E_z) T(E_z) \right]. \quad (13)$$

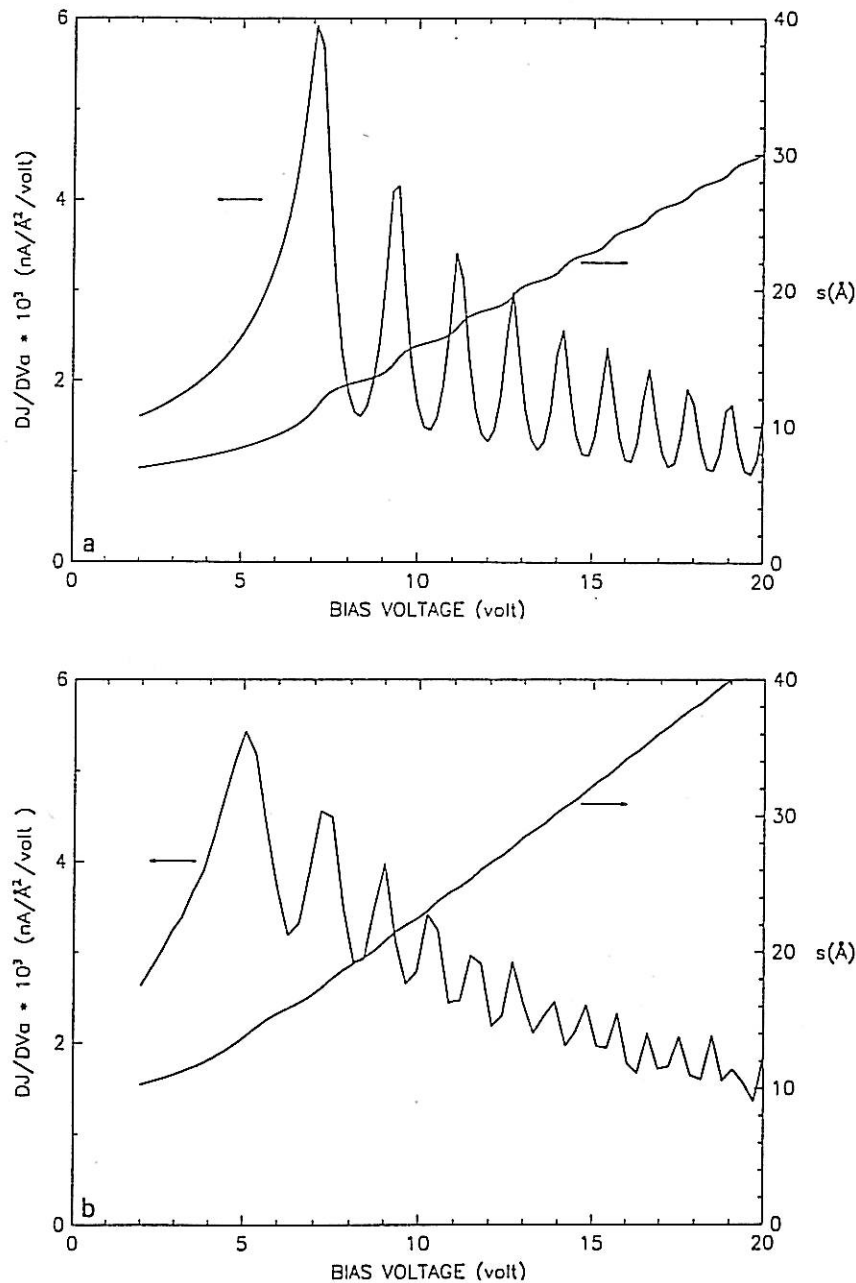


Fig. 3. Constant current density mode $s-V_a$ and dJ/dV_a-V_a tunnel characteristics for $J = 2.0 \times 10^{-3} \text{ nA}/\text{\AA}^2$, $\Phi_L = 5.2 \text{ eV}$ and $\Phi_R = 4.4 \text{ eV}$ and the potential barriers shown in fig. 1. The trapezoidal barrier of eq. (1) has been used in fig. 3a, and the image reduced potential barrier of eq. (2) with the image potential specified by (4) and (5), in fig. 3b and 3c, respectively. Fig. 2d shows the $s-V_a$ and dJ/dV_a-V_a characteristics for the potential barrier of eq. (3) with $z_L = s - z_R = 0.0$, $\lambda_L = 1.0 a_0^{-1}$, $\lambda_R = 1.3 a_0^{-1}$, and $\mu = 1.0$ (solid curves) and $\mu = 0.0$ (dashed curves); the results of fig. 3b are also shown (dotted curves), for comparison. Fig. 3e shows the tunneling characteristics for the potential barrier of eq. (3), with $\lambda_L = 1.0 a_0^{-1}$, $\lambda_R = 1.3 a_0^{-1}$, $\mu = 1.0$, and $z_L = s - z_R = 0.0$ (solid curves), $z_L = s - z_R = -0.5 \text{ \AA}$ (dashed curves) and $z_L = s - z_R = 0.5 \text{ \AA}$ (dashed-dotted curves).

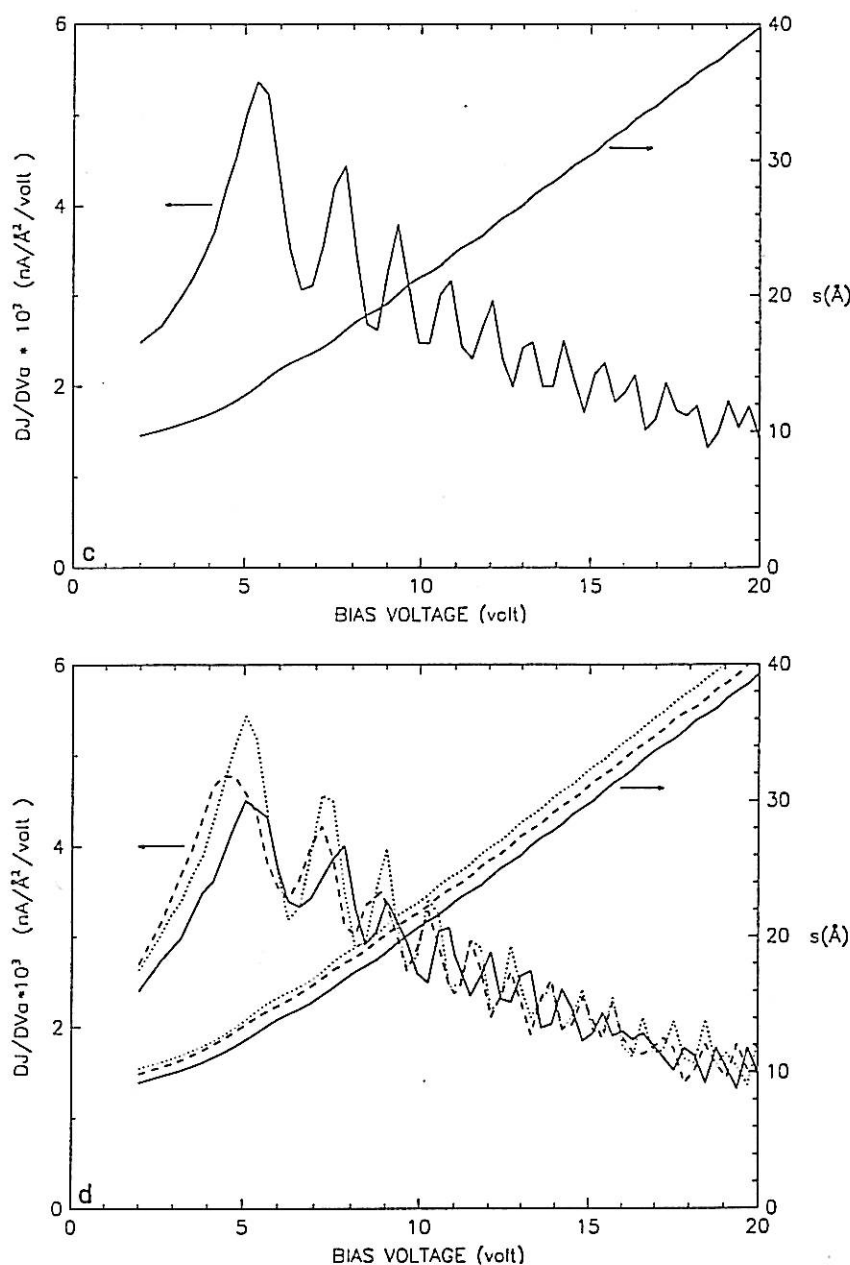


Fig. 3. Continued.

In this expression $T(E_z)$ is the electron transmission probability through the interface potential barrier, calculated for the incident electron energy E_z perpendicular to the surface. As pointed out above, the WKB approximation fails to explain the resonance phenomena, so that the transmis-

sion probabilities have been calculated solving numerically the Schrödinger equation; this has been done representing the solution as Chebyshev series [24].

Now, for a given area A , applied voltage V_a , and tip-sample separation s , the current can be

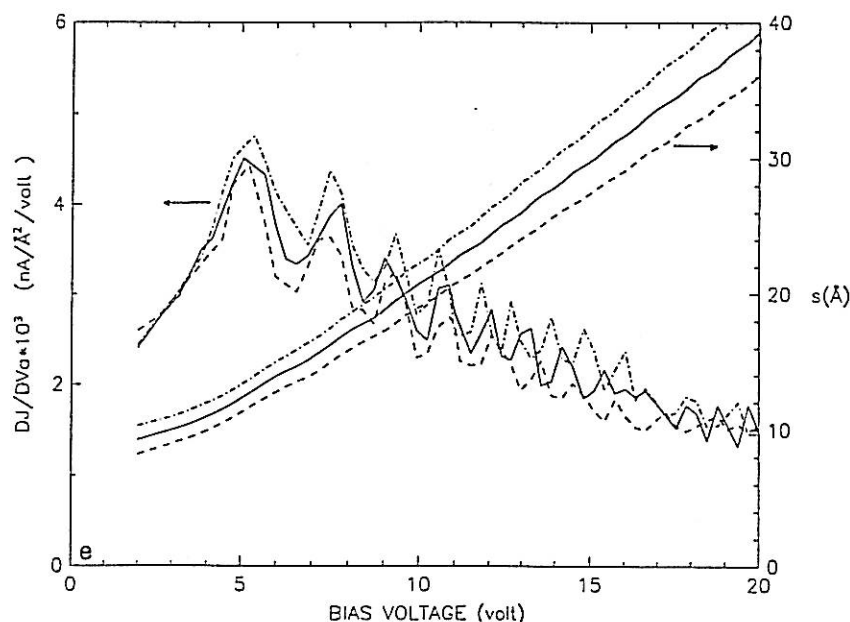


Fig. 3. Continued.

found from eq. (13). Since the experiment is done at constant current, we assume that the area is constant, and search for the value of s that makes the current density constant, as V_a is changed, obtaining the constant current density mode $s-V_a$ characteristics. Then, we evaluate dJ/dV_a numerically as a function of the bias voltage, along the calculated constant current density mode $s(V_a)$ curve.

The constant current density mode $s-V_a$ and dJ/dV_a-V_a characteristics have been calculated for a constant current of 1 nA and different constant areas from 10 to 10^3 Å^2 . The results of the calculation indicate that the peak positions of the oscillations in the conductance-voltage characteristics are very sensitive to the area. This happens as a consequence of the dependence of the resonant peak positions on the distance, which changes, in the constant current mode, with the area. Fig. 3 shows the distance-voltage and conductance-voltage characteristics calculated for $A = 500 \text{ Å}^2$ and the model barriers of eqs. (1), (2) and (3).

Comparing the results obtained in this way for the different models, we realize that the conductance-voltage characteristics show an important

dependence on the interface potential profile: in general, abrupt interface potentials, like the one given by eq. (1), tend to give sharper peaks, and the potential of eq. (3), representing a more realistic potential for ideal surfaces, yields smoother dI/dV_a-V_a characteristics, which become even smoother as λ_R gets smaller. The peak positions in the oscillations, however, do not depend appreciably on λ_R , since the gap distance changes very slightly with this parameter. The dependence on μ and image plane positions of dI/dV_a-V_a for the model potential of eq. (3) is shown in figs. 3d and 3e, respectively.

When the gap distance dependent dynamical parameter μ of fig. 2 and the image plane positions $z_L = s - z_R = 0$ are considered, a reasonable fit between the low voltage peak positions of dI/dV_a calculated with the realistic potential of eq. (3) and the experimental results of ref. [6] with $I = 1 \text{ nA}$ is obtained for a constant area of 500 Å^2 , as shown in fig. 4, where we have used, also, $\lambda_L = 1.0 a_0^{-1}$ and $\lambda_R = 1.3 a_0^{-1}$. However, two main discrepancies appear systematically, for any value of the area and the parameters, between our one-dimensional results and the experimental curves. First of all, the first experimental peak is sharper

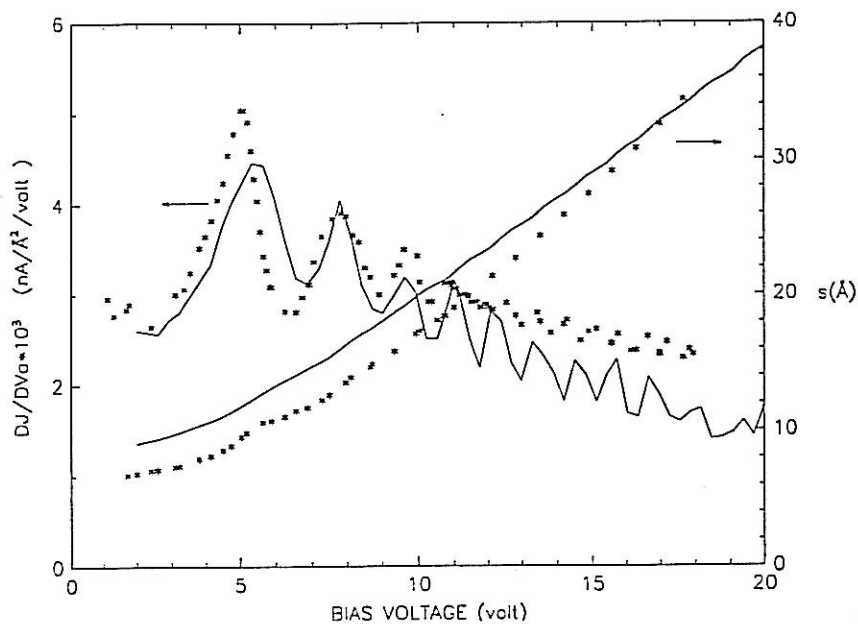


Fig. 4. Calculated results of the constant current density mode $s-V_a$ and dJ/dV_a-V_a characteristics for $J = 2.0 \times 10^{-3} \text{ nA/\AA}^2$, $\Phi_L = 5.2 \text{ eV}$, $\Phi_R = 4.4 \text{ eV}$ and the potential barrier of eq. (3) with $\lambda_L = 1.0 a_0^{-1}$, $\lambda_R = 1.3 a_0^{-1}$ and the gap-distance dependent parameter μ of fig. 2. Also shown are the experimental curves of ref. [6] obtained for $I = 1 \text{ nA}$ (cross points).

than the theoretical one, and, secondly, the theoretical dI/dV_a-V_a curve for high voltages is systematically lower than the experimental one and the oscillations are more spaced in voltage. In an alike manner, the theoretical $s-V_a$ characteristic does not fit appropriately the experimental behaviour at high voltages.

The first discrepancy can be related to the assumption made of having ideal and unrelaxed surfaces for both electrodes. As shown above, sharper peaks can be obtained at low voltages with more abrupt interface potentials; thus, it may be inferred that the experimental curve could be fitted, at low voltages, by the one-dimensional results, just assuming some inwardly relaxed surface geometry for Au, W or both, since this kind of relaxation would increase the electron density just near the last crystal layer, this effect creating a deeper electronic potential as needed in this case. The second discrepancy, appearing for high voltages, between the one-dimensional results and the experimental data, is related, however, to the three-dimensionality of the problem and the curvature of the tip. The analysis of this effect is

the subject of the next section, where a three-dimensional calculation of the $s-V_a$ and dI/dV_a-V_a characteristics is presented; in this calculation we will not include, however, any surface metal relaxation, which is assumed to be basically related to the low voltage behaviour of the conductance-voltage characteristics.

3. Three-dimensional models

3.1. Simple considerations

The tip-sample interface geometry plays an important role in the microscopic characteristics, as we will show below. In this section we discuss a simple model, in which the tip and the sample are assumed to have spherical and planar geometry, respectively. The spherical model of the tip has been used in other theoretical approaches and is a first approximation to its real shape.

In order to have a simple idea of the role played by the geometry, we argue in the following way. For tip voltages exceeding the sample work

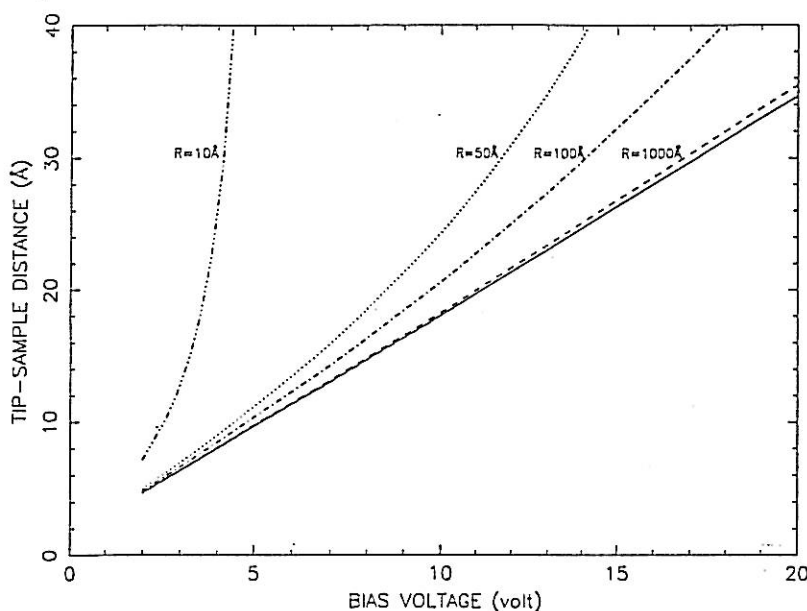


Fig. 5. Calculated tip-sample distances as a function of the applied voltage that makes the electric field at the tip apex constant and equal to $0.6 \text{ V/\AA}$, for a planar tip (solid line) and for a spherical tip with radius of $1000 \text{ \AA}$ (dashed line), $100 \text{ \AA}$ (dashed-dotted line), $50 \text{ \AA}$ (dotted line) and $10 \text{ \AA}$ (dashed-dotted-dotted-dotted line).

function, the electron propagates with positive kinetic energy through part of the vacuum gap, after having crossed the tunnel barrier. Thus, we are in the field emission regime, and the current is controlled approximately just by the electric field; consequently, assuming that the tunneling effective area keeps constant (this approximation will be improved later on) and noticing that the tunneling electrons are mostly crossing the interface from the tip apex, the constant-current mode $s-V_a$ characteristic can be determined, for high voltages, by imposing the condition of constant electric field at the tip apex. This is an elementary electrostatic problem that can be solved considering the arrangement of infinite point charges making equipotential the spherical tip and planar sample surfaces [25].

Fig. 5 shows the calculated constant-field $s-V_a$ characteristic for an electric field of $0.6 \text{ V/\AA}$ and several values of the tip radius. These results show that the $s-V_a$ curves are dramatically dependent on the tip radius, and comparing with the $s-V_a$ experimental data of ref. [6], we find that the bending up of that curve is similar to the one

found in fig. 5 for $R \approx 100 \text{ \AA}$. This is a very elementary argument to determine the tip radius, as we will see below; however, it shows the main effect that we will proceed now to analyse more carefully.

3.2. Effective tunneling area

The argument presented in the previous paragraph assumes that the effective tunneling area is constant. This is, however, an oversimplification that will be modified appropriately.

Fig. 6 exhibits some equipotential surfaces and electric field lines for a given tip-sample configuration, in the field emission regime. The equipotential surface with potential equalling the tip Fermi level is shown in this figure; this surface divides, neglecting image force effects, the tunneling region, that is the classically forbidden part of the vacuum gap, from the positive kinetic energy region of the most energetic electrons tunneling from the tip to the sample. Also shown is the tunneling region when the tip is made positive with respect to the sample, in such a way that

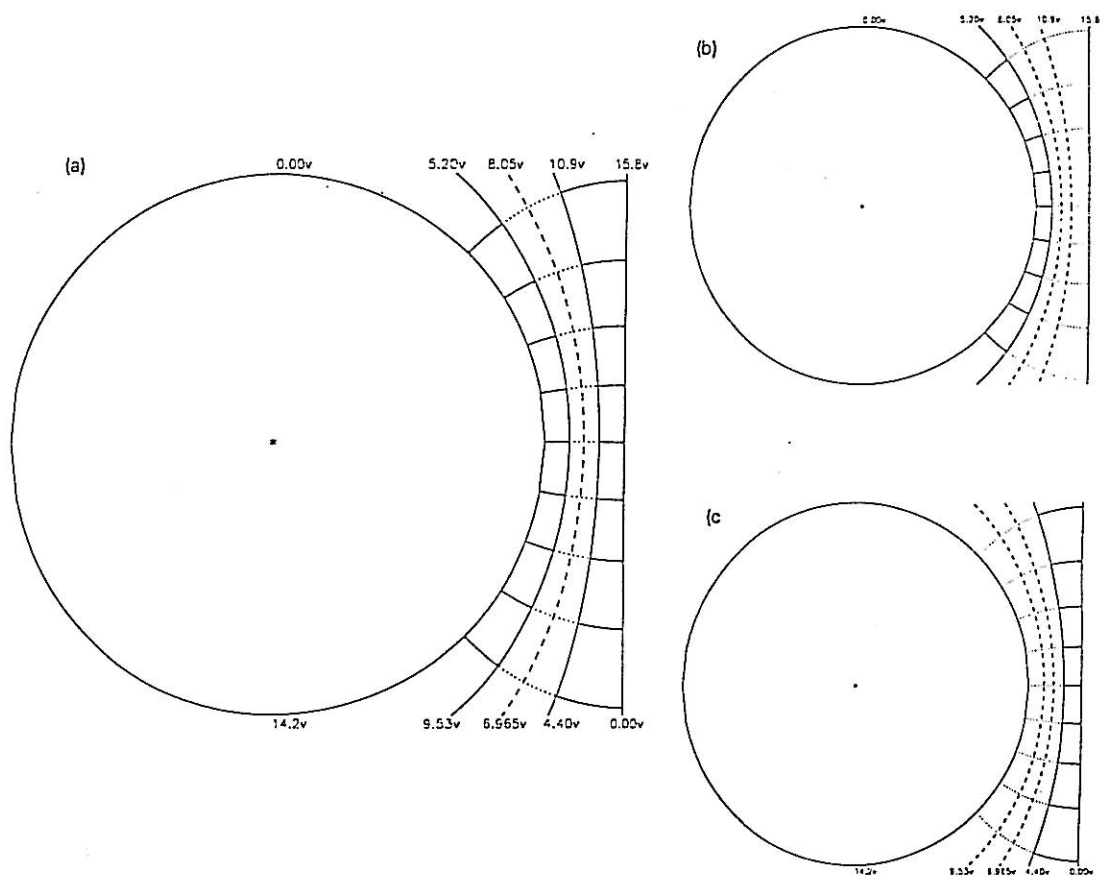


Fig. 6. (a) Equipotential surfaces and electric field lines for a planar Au sample and a spherical W tip of radius equal to 100 Å; work functions, applied voltage and tip-sample distance have been taken to be $\Phi_W = 5.2$ eV, $\Phi_{Au} = 4.4$ eV, $V_a = 15.0$ V and $s = 30$ Å. The equipotential surfaces where the most energetic electrons tunneling from the tip to the sample and from the sample to the tip have kinetic energy equal to zero are represented by solid lines. The numbers in the upper part of the figure represent the potential values corresponding to the positive sample bias, whereas the numbers in the base correspond to the positive tip bias case. Notice that the potential difference between the tip and the sample changes when the sign of the bias voltage is changed because of the difference between the work functions. Fig. 6a is split into halves in figs. 6b and 6c, so as to distinguish positive sample bias (fig. 6b) and positive tip bias (fig. 6c); the field lines are represented by solid lines along the tunneling region, and by dotted lines along the positive kinetic energy part of the vacuum gap.

electrons move from the sample to the tip and the tunneling region appears, at high voltages, close to the sample surface. Notice that there is an asymmetry in the problem, associated to the two very different tunneling regions: in the second case, that is, for a positive tip, the tip asperities should play a minor role, with the implications that will be discussed below.

In the field emission regime the tunnel current is approximately a function of the electric field alone, as it has been pointed out above, so that the

width of the tunnel barrier for electrons moving from the tip apex happens to be almost constant in the constant current mode, and it is at high voltages much less than the actual tip-sample separation. However, for the negative tip bias, the equipotential surface dividing the tunneling region from the positive kinetic energy region of the vacuum gap becomes more and more bended as the bias voltage gets bigger, and it becomes closer and closer to the negative bias electrode, no matter what the bias sign is.

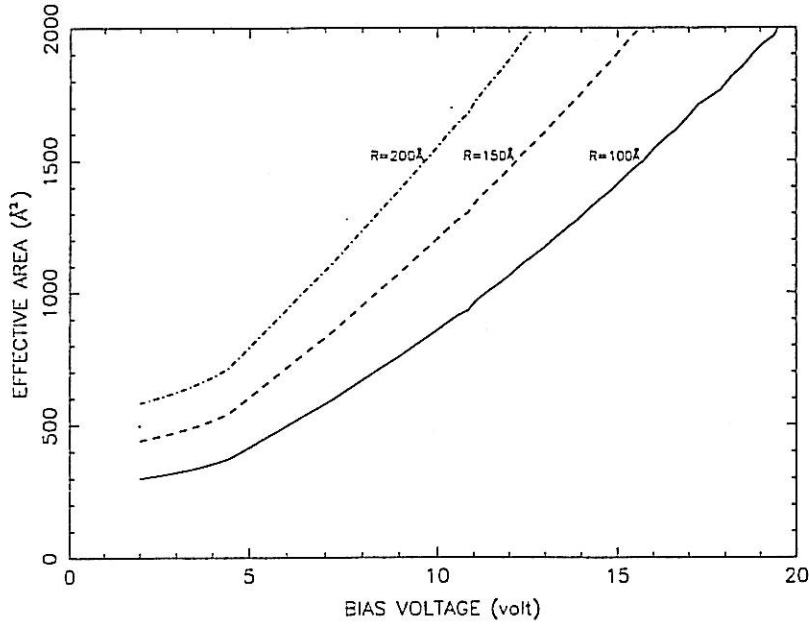


Fig. 7. Effective area calculated, in the constant current mode, from eq. (14), as a function of the applied voltage, for tip radius of 100 Å (solid curve), 150 Å (dashed curve) and 200 Å (dashed-dotted curve).

In a semiclassical approximation electrons must move along the electric field lines, in such a way that the maximum tunneling probability appears for the electrons leaving the tip apex, and this probability decreases as we move far from this point. As the tunneling probability decreases very quickly with the tunnel barrier width, we can expect the effective tunneling area to be reduced to a small area around the tip apex; moreover, it can be expected from the above discussion to increase with the applied voltage. We can define the effective tunneling area integrating over the equipotential surface where the most energetic electrons have kinetic energy equal to zero, S , as follows:

$$A_{\text{eff}} = \frac{1}{J_0} \int_S dS J(r), \quad (14)$$

where $J(r)$ represents the current density for electrons arriving at r in the equipotential surface S , J_0 being the current density for electrons tunneling just from the tip apex.

If the WKB approximation is used to evaluate the electron tunneling probabilities, the tunneling

current density can be written in the following way:

$$J(r) = \frac{1}{(2\pi)^2 \Delta s(r)^2} \left\{ \bar{\Phi}(r) \times \exp \left[-2\sqrt{2\bar{\Phi}(r)} \Delta s(r) \right] - (\bar{\Phi}(r) + V_a) \times \exp \left[-2\sqrt{2(\bar{\Phi}(r) + V_a)} \Delta s(r) \right] \right\}, \quad (15)$$

where $\bar{\Phi}$ represents the mean barrier height:

$$\bar{\Phi}(r) = \frac{1}{\Delta s(r)} \int_{l(r)} dl' (V(r') + \Phi_L), \quad (16)$$

and

$$\Delta s(r) = \int_{l(r)} dl'. \quad (17)$$

In these expressions $l(r)$ represents the electric field line going from the negative bias electrode to the equipotential surface S at r , and $V(r')$ is the electric potential at r' in the field line $l(r)$. In fig. 7 it is illustrated how the effective area obtained

after having introduced (15) into (14) changes for a negative tip bias for different tip radii ranging from 100 to 200 Å, in the constant current mode, when the electrostatic potential is included in eq. (16). It is obvious from this figure that for small voltages the area is practically constant, but it increases very quickly for voltages larger than the sample work function, as we have already discussed above.

The results of fig. 7 show that the effective area cannot be assumed to be constant, so that the current density is not constant in the constant current mode, neither is the electric field, not even at high voltages; the results of fig. 5 must be, therefore, reconsidered. Moreover, the tunneling effective area depends on the tip-sample distance, and the constant current mode distance depends, at the same time, on the effective area; consequently, both the effective area and the tip-sample distance must be evaluated self-consistently. This point is discussed in the next section, introducing an analysis of the tunneling intensity calculation through the barrier.

3.3. Three-dimensional calculations

In this section we shall be concerned with the calculation of the constant current mode distance-voltage and conductance-voltage characteristics. To calculate the tip-sample tunneling current for a given applied voltage, tip-sample distance and tip radius, we take the following steps:

First, we calculate the classical electrostatic potential, and determine the equipotential surfaces and the electric field lines. Then, we obtain the tunneling region, that is, the region between the negative bias electrode and the equipotential surface where the most energetic electrons have zero kinetic energy (see fig. 6), and we calculate the effective tunneling area integrating over this surface by means of eq. (14), as discussed above.

Secondly, we reduce the three-dimensional tunneling problem to a one-dimensional one, similar to the ones discussed in section 2, and we calculate the total tunneling current as the product of the effective area, obtained as shown above, and the current density, given by eq. (13), of electrons tunneling from/to the tip apex through the poten-

tial barrier of eq. (3) conveniently modified by the effect of the curvature; this has been done by replacing the linear potential $-(V_a + \Phi_L - \Phi_R)z/s$ in the region $0 < z < s$ by the true electrostatic potential along the trajectory of the electrons.

Next, we search for the tip-sample separation making constant the total current, that is, the product of the current density times the tunneling area, both calculated as outlined above.

Finally, once we have determined the constant current mode tip-sample separation for a given voltage, total current and geometry, we proceed to calculate the conductance-voltage characteristics. Thus, we evaluate numerically, along the calculated constant current mode $s(V_a)$ curve, dI/dV_a as a function of V_a , evaluating for each given value of the tip-sample distance the effective area specified by (14).

3.4. Three-dimensional results

Using the method discussed above, we have determined the constant current mode $s-V_a$ and

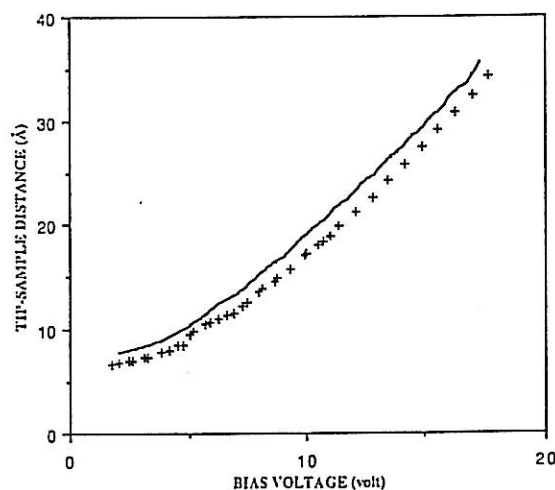


Fig. 8. Values of s plotted against V_a , obtained for a positive sample bias as shown in section 3.3, for $\Phi_L = 5.2$ eV, $\Phi_R = 4.4$ eV, $I = 1$ nA and the model barrier of eq. (3) with $\lambda_L = 1.0a_0^{-1}$, $\lambda_R = 1.3a_0^{-1}$, the gap-distance dependent parameter μ of fig. 2, $z_L = 0.8$ Å, $s - z_a = -0.2$ Å and the self-consistently calculated distance dependent effective area given by (14). Also shown is the experimental curve obtained in ref. [6] for $I = 1$ nA (cross points).

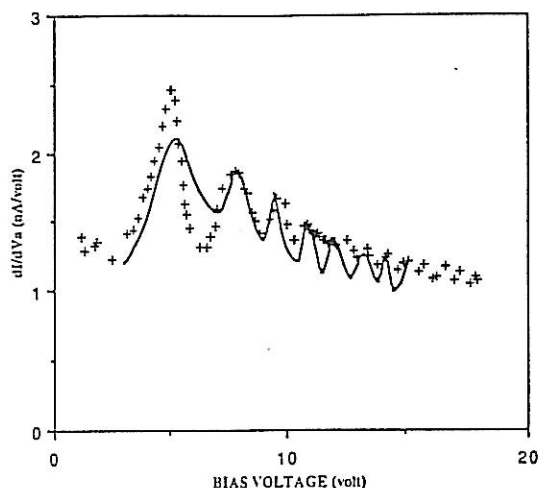


Fig. 9. Values of dI/dV_a plotted against V_a , obtained for a positive sample bias as shown in section 3.3, for $\Phi_L = 5.2$ eV, $\Phi_R = 4.4$ eV, $I = 1$ nA and the model barrier of eq. (3) with $\lambda_L = 1.0a_0^{-1}$, $\lambda_R = 1.3a_0^{-1}$, the gap-distance dependent parameter μ of fig. 2, $z_L = -0.8$ Å, $s - z_R = -0.2$ Å and the self-consistently calculated distance dependent effective area given by (14). Also shown is the experimental curve obtained in ref. [6] for $I = 1$ nA (cross points).

$dI/dV_a - V_a$ characteristics for different values of the tip radius, and for a tunneling current of 1 nA. Our first aim has been to determine the radius of the tip sphere yielding a good agreement between our calculations and the experimental data given by Becker et al. [6], and we have found that a radius of 150 Å yields the best agreement, although differences of 25 Å do not introduce important modifications to our results (see fig. 5 for the effects introduced by different tip radii of curvature). Figs. 8 and 9 show the calculated $s - V_a$ and $dI/dV_a - V_a$ characteristics for a tip sphere with a radius of 150 Å and a total current of 1 nA, together with the experimental data given in ref. [6] for a negative tip bias voltage. In these calculations we have considered the potential barrier of eq. (3) with $\lambda_L = 1.0a_0^{-1}$, $\lambda_R = 1.3a_0^{-1}$, the gap-distance dependent parameter μ of fig. 2, $z_L = -0.8$ Å and $s - z_R = -0.2$ Å. Main comments on these results are the following:

(i) The negative values of the parameters z_L and $s - z_R$ are consistent with the d-band character of W and Au, which will tend to displace the centroid of the induced density towards the

material, compared with the results of the jellium model.

(ii) The agreement between the theoretical and experimental values of the $s - V_a$ characteristics is very good (the displacement between both curves is not significant, since the experimental data do not give absolute distances). This agreement shows that the three-dimensional effects discussed above are necessary to explain the high voltage behaviour of the $s - V_a$ characteristics. Moreover, this analysis allows us to deduce the microscopic tip radius of curvature.

(iii) Good agreement is also found at high voltages for the $dI/dV_a - V_a$ characteristics, although some discrepancies appear around the first oscillation of this curve; as discussed above, this is probably due to some possible relaxation of the tip-surface layer. At high voltages we find, however, that the calculated oscillations of the $dI/dV_a - V_a$ curve are a little damped in the experimental curve. Bono and Good [7] have argued that this damping is due to the asperities of the tip that tend to produce interferences among the different parts of the tip protruding more or less from the mean tip surface. It is of crucial interest to remark that, if this were the right explanation, the same effects would disappear for the opposite bias, a positive tip bias, since in this case the tunneling region would be located, at high voltages, close to the sample surface, as discussed above (see fig. 6), and the tip asperities would play minor roles in the tunneling process.

4. Conclusions

In conclusion, we have shown that a reasonable fit to the interface potential can be made using a parametrized model potential, by a trial and error process to reproduce the experimentally determined $s - V_a$ and $dI/dV_a - V_a$ characteristics, which allows the tip radius to be determined and demonstrates the importance of taking account of the 3D tip-sample geometry, especially at high V_a .

Work on other tip-sample combinations and a detailed study of the role of tip asperities is in progress.

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