New approach to numerical simulation of scanning tunnel microscopy

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(Submitted 16 May 1991)
Zh. Eksp. Teor. Fiz. **100**, 1640–1648 (November 1991)

We propose a simple and effective method of computing the tunnel current in scanning tunnel spectroscopy. The method reduces the problem of computing the tunnel current to that of determining the spectrum of the quasistationary state; it accounts also for the reaction of the tip to the spectrum of the crystal states on the surface. The method is illustrated by a number of simple examples in the context of the tight-binding method.

The discovery of tunnel microscopy[1] has provided science and technology with a most powerful tool for the study, at the atomic level, of the electron structure of the surface of a semiconductor or a metal. This method is based on a precise measurement of the tunnel current between a conducting crystal and a metallic tip. The sensitivity of modern instruments permits a direct study of the arrangement of the atom on the surface of a body. In addition, the variation of tunnel current with that of the voltage between the tip and the sample, and upon displacement of the tip, yields in principle extensive information on the electronic structure of a crystal.

Despite the abundance of experimental results for many greatly varying systems, no reliable foundation, based on a direct simulation of the experimental situation, exists for a theoretical interpretation of these results. The theoretical description of tunnel microscopy is made complicated mainly by the fact that the quantity to be calculated is kinetic, the tunnel current, whereas the existing calculation methods are aimed exclusively at a static electronic structure. In this respect, tunnel microscopy is in a less favorable situation then the atomic-force microscopy[2] derived later on its basis. Atomic-force microscopy can be directly simulated by using only static computations of the energy of a crystalline cluster (sample and tip) as a function of its mutual arrangement.[3]

Tunnel microscopy is described at present by two essentially different approaches. One, due to Bardeen,[4] is based on the notion of tunneling as a consequence of the overlap of the wave functions of the electron states of the sample and of the tip.[5] This approach accounts well for the principal, exponential dependences at large distance between the tip and sample, and can provide in practice only a qualitative description of the experiment.

The second, more consistent approach is based on calculation of the tunneling in the course of scattering, wherein an electron incident from the outside on the sample (or tip) surface and scattered by it penetrates partially into the tip (or sample) and produces there an outgoing wave. Although this scattering problem can in principle be formulated with sufficient rigor in the language of two weakly bound crystalline clusters, the need for classifying the wave functions in terms of incident, reflected, and transmitted waves makes the numerical computations extremely complicated. This approach is therefore more applicable to analytic investigations of simplified model problems[6,7] than for numerical simulation with allowance for the real microscopic structure of the sample and of the tip. A major shortcoming of this approach is the difficulty of accounting for the contribution of the localized surface states which are orthogonal to all current-carrying states that extend into the interior of the crystal.

An approach to the calculation of the tunnel current when the tunnel coupling between the tip and the crystal is not weak, was recently proposed.[8] Allowance was made there for the contribution made to the current by the surface states induced by the tip. The idea of this approach, however, is close to that of the tunnel-Hamiltonian method.

We propose here a fundamentally new approach to the computation of the tunnel current. Its central idea is to introduce formally into the Hamiltonian of the system, which contains two crystalline clusters—the sample and the tip—a drain for the electrons at the tip point located far from the sample. The tunnel current is determined by analyzing the spectrum of the complex eigenvalues of this Hamiltonian, which can be easily diagonalized by standard numerical methods. Our approach is universal and makes the problem of computing the tunnel current as simple as the computation of the electron spectrum of the system. In addition, it makes it possible to take rigorously into account the atomic structures of the sample and of the tip. We describe below the procedure in detail and illustrate it with simple examples within the scope of the tight-binding concept.

DESCRIPTION OF METHOD

We take first the sample to be a limited crystalline cluster. The cluster representing the tip will be first assumed to be infinite in one direction, so that the electrons incident from the crystal on the tip can go off unobstructed to infinity. The electrons in such a system can be described not only by stationary states but also in terms of states called quasistationary, whose energy $E - i\gamma$ has a small imaginary part—corresponding to an exponential decrease of the wave functions with time. At low amplitude of the electron hopover from the sample to the tip, the wave functions of the quasistationary states are close to stationary inside the crystal, and contain in the tip only the flux away from the sample.

The population of the quasistationary states should be described by a Fermi distribution with respect to the real parts of their energy. If the Fermi level μ_s in the sample is higher than that in the tip (μ_t), the only states that can really decay are those in the range $\mu_s > E > \mu_t$. Since the number of electrons going off per unit time from the sample as a result of decay of a separately taken state with damping

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γ is equal to $2\gamma/\hbar$, the total tunnel current is equal to the sum

$$J=2\hbar^{-1}\sum_{\mu_s>E_i>\mu_t}\gamma(E_i)$$

over the quasistationary states in the indicated energy interval.

There is no need to introduce an infinite sample and a bounded tip in order to consider the case when the Fermi level in the tip is higher than in the sample. The results can be obtained in the framework of the scheme described above by considering that the equilibrium reached when the chemical potentials μ_s and μ_t are equal means a mutual cancelation of the opposing currents from the sample and from the tip. To calculate the current from the tip into the sample at $\mu_s<\mu_t$ it suffices therefore to interchange the sample and tip Fermi levels and use the negative of the resultant current

$$J=-2\hbar^{-1}\sum_{\mu_t>E_i>\mu_s}\gamma(E_i).$$

ONE-DIMENSIONAL CASE

We demonstrate the approach using as an example one-dimensional problem having, furthermore, an exact analytic solution. Consider a one-dimensional chain. Let one orbital be centered on each atom. We equate the neighboring-atoms overlap integrals to unity, and the proper energy of an isolated orbital to zero. The left- and right-hand halves of the chain are weakly coupled, with an overlap integral ξ_0. Let us calculate the probability of tunneling between the left- and right-hand halves. Let a wave of unity amplitude be incident from the left infinity. A transmitted wave will pass then through the right-hand half, and a reflected wave will be added from the left. Only waves with energy in the continuum can go off to infinity. The wave function in the left-hand half is the sum of the incident and reflected waves

$$\Psi_n=e^{ikn}+R(k)e^{-ikn}, \tag{1}$$

R is the reflection coefficient and n is the site number. The right-hand half contains only the transmitted wave

$$\Psi_n=T(k)e^{ikn}, \tag{2}$$

where $T(k)$ is the transmission coefficient. The distance between atoms is set equal to unity.

The spectrum of a one-dimensional ideal chain has in the tight-binding approximation the form

$$E(k)=2\cos k. \tag{3}$$

If the energy is in the allowed part of the spectrum, we have

$$e^{\pm ikn}=[E/2\pm i(1-E^2/4)^{1/2}]^n. \tag{4}$$

The matching conditions lead in the tight-binding approximation to the equations

$$\begin{aligned}&E\Psi_1=\Psi_0+\xi_0\Psi_2,\\&E\Psi_2=\xi_0\Psi_1+\Psi_3.\end{aligned} \tag{5}$$

Taking (1) and (2) into account, we find that

$$\begin{aligned}&(Ee^{-ik}-1)R(E)-\xi_0e^{2ik}T(E)=1-Ee^{ik},\\&-\xi_0e^{-ik}R(E)+(Ee^{2ik}-e^{3ik})T(E)=\xi_0e^{ik}.\end{aligned} \tag{6}$$

Using (3) and (4) we obtain the final expressions for the transmission and reflection coefficients as functions of the energy:

$$\begin{aligned}&|T(E)|^2=\xi_0^2(4-E^2)/[(1-\xi_0^2)^2-\xi_0^2E^2],\\&|R(E)|^2=(1-\xi_0^2)/[(1-\xi_0^2)^2-\xi_0^2E^2].\end{aligned} \tag{7}$$

The total particle flux in the entire zone is

$$J=\int_0^\pi v(k)\,dk/2\pi, \tag{8}$$

where $v(k)=dE(k)/dk$ is the group velocity, we have (disregarding spin)

$$\begin{aligned}J=\int_{-2}^{2}|T(E)|^2\,dE&=(1/\pi)(1/\gamma'-\gamma')\ln|(\gamma'+1)/(\gamma'-1)|\\&\approx16/3\pi\gamma^2\approx16\xi_0^2/3\pi,\\&\gamma'=\gamma/2,\ 1/\gamma^2=\xi_0^2/(1+\xi_0^2)\ll1.\end{aligned} \tag{9}$$

The flux is thus proportional to the square of the overlap integral, which is in turn dependent exponentially on the distance.

Under equilibrium conditions in the absence of an applied voltage, the total flux is, naturally, zero since the fluxes from the left and from the right are equal.

When a voltage U is applied to the system, one can obtain the transmission coefficient (we assume for the sake of argument that the potentials of the right- and left-hand halves are U and zero, respectively). For the transmission coefficient we have

$$\begin{aligned}|T(\varepsilon)|^2=\xi_0^2(4-\varepsilon^2)/\{1-2\xi_0^2\varepsilon\varepsilon_u+\xi_0^4\\+2\xi_0^2[(1-\varepsilon^2)(1-\varepsilon_u^2)]^{1/2}\}.\end{aligned} \tag{10}$$

$$\varepsilon=E/2,\ \varepsilon_u=(E+U)/2.$$

We assume further that the bands are half-full, and that the Fermi level E_f is zero at $U=0$. For small ξ_0 (in view of symmetry it is enough to consider only the case $U=0$) we get for the flux

$$\begin{aligned}&J(U)\approx(\xi_0^2/2\pi)(4U-U^3/3),\ 0<U<2.\\&J(U)\approx(\xi_0^2/2\pi)[{}^{16}/_{3}-4(U-2)+(U-2)^3/3],\ 2<U<4,\\&J(U)=0,\ U>4.\end{aligned} \tag{11}$$

The current can be calculated as the rate of decay of the quasistationary states. Let us describe this procedure. Consider two weakly coupled (right- and left-hand) chains of finite but sufficiently large size (we call the right-hand one the crystal, and the left-hand one the tip). We produce a drain on the tip's end that is far from the crystal. For this, we add to the diagonal component of the Hamiltonian matrix of the system, in the tight-binding approximation, an imaginary increment to the energy of the orbital of the outermost atom of the tip. All the states in the system become then quasistationary and decay via the drain on the right-hand end of the tip. Owing to the presence of the weak-binding "bottleneck" the states in the crystal decay much more slowly than the states in the tip. This fact separates the weakly damped states belonging directly to the crystal. Note that separation of the energy imaginary parts pertaining to the

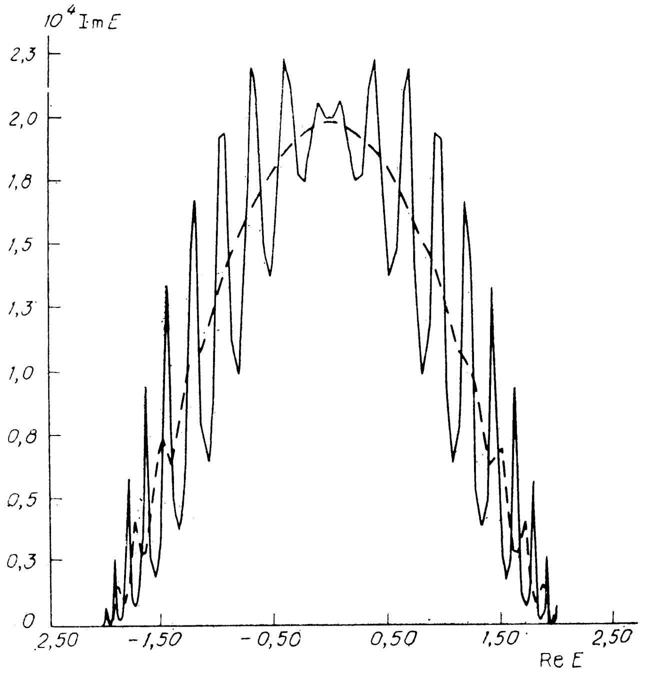

FIG. 1. Distribution of imaginary parts (transmission coefficient) as a function of the real parts of the crystal energy (no voltage is applied). Dashed line—with allowance for drain optimization, solid—without allowance.

states of the crystal is possible for any value of the overlap integral.

The imaginary increment to the energy of the orbital should be of the order of the bandwidth in the tip. The distribution of the imaginary spectrum of the crystal as a function of the real parts of the energy is shown in Fig. 1 (solid curve), which shows dips of the transmission coefficient at certain energies. The cause of the dips is that the drain is not ideal and also has a finite reflection coefficient. The drain must be designed to have a minimum reflection. This can be done by producing not one large drain at the outermost atom, but a set of gradually increasing drains at several extreme atoms. The drain sizes can be such that the states remaining in the tip have maximum damping, which is equivalent to minimum reflection from the drain.

Such an optimization was effected for an isolated tip chain of 20 atoms with a drain at five outermost atoms; the drain sizes were such that the imaginary energy increments to them ensured an integral damping maximum over all the remaining states in the tip. The optimal imaginary increments, starting with the atom farthest on the right, were the following:

$$\gamma_1=1{,}955,\quad \gamma_2=0{,}954,\quad \gamma_3=0{,}541,\quad \gamma_4=0{,}263,\quad \gamma_5=0{,}077.$$

The given optimal parameters were used next to calculate the damping of the states in a crystal (in a chain of 100 atoms). The distribution of the imaginary parts in the crystal following the optimization are shown in Fig. 1; it can be seen that the dips connected with reflection from the drain have practically vanished.

Calculation of the flux from the crystal into the tip over all the states (double the sum of the imaginary parts) yields $J = 0.16$ ($\xi_0 = 0.1$), which agrees well with the exact result [see Eq. (9)].

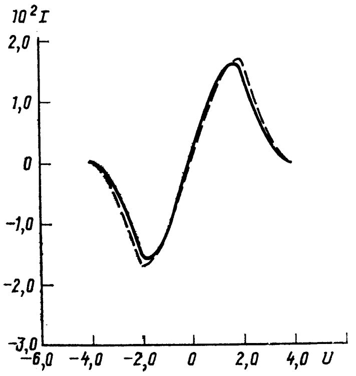

FIG. 2. Current-voltage characteristics. Exact analytic (dashed) and numerical calculation by the method of quasistationary states (solid).

All the figures below show in essence the dependences of the damping γ on the voltage; the current differs only by a coefficient $2e/\hbar$ (e is the electron charge). Bearing this in mind, we shall refer only to the current. The dependences of the current on the voltage, calculated for the one-dimensional case from analytic expressions and numerically, are shown in Fig. 2.

THREE-DIMENSIONAL CASE

We proceed now to the three-dimensional case. The tip was simulated, as before, by a one-dimensional chain of 20 atoms with optimized drain sizes. The crystal was simulated by a system of a finite number of layers. The overlap integrals and the interatomic distances in the crystal were chosen to be the same as in the one-dimensional case.

We calculated the current–voltage characteristics for clusters of 2×2, 3×3, 4×4, and 5×5 atoms in a plane and four layers thick. The tip was located directly above the crystal atom at a distance of three units, and the only overlap taken into account was between the outermost atom of the tip and the crystal atom beneath it. The dependence of the overlap integral on the distance was chosen in this calculation in the form $1/d^2$ (Ref. 9). To assess the convergence of the results we performed, with the same parameters, calculations for elementary cells measuring 2×2, 3×3, 4×4, and 5×5 with the tip over them; the calculations were repeated periodically along the surface. In this calculation of the current we integrated over a plane Brillouin zone over all the occupied states. The worst disparity for a 2×2 cell did not exceed 15%. Figure 3 shows the dependences of the tunnel current on the voltage for different cell dimension in the plane, and for periodically repeated cells along the surface. For a 5×5 cell (Fig. 4) the results of the two methods are indistinguishable. Analysis of the convergence of the results as a function of the crystal thickness shows that starting with 4–5 layers they become practically independent of the thickness.

In addition, we calculated the relief of the tunneling current $I(x,y)$, at $z = 2$ and at a voltage $U = 4$ V on the tip, along the [100], [010], and [110] directions, up to half the interatomic distance (see Fig. 5). We chose for this case the overlap of the outermost atom of the tip and all the atoms on the crystal surface. The dependence of the overlap integrals on the distance was chosen in the form $e^{-2(r-1)}$. The periodically repeated cell dimension was 5×5.

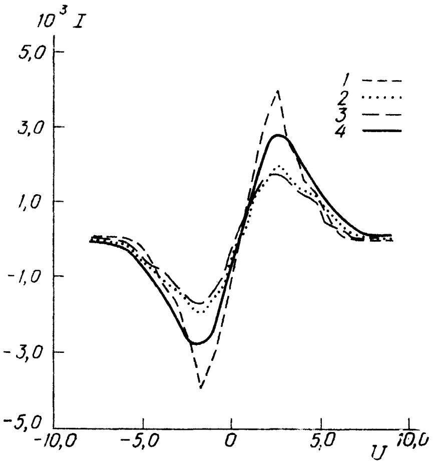

FIG. 3. Current-voltage characteristics for crystal clusters of various dimensions in the surface plane: 1—2×2, 2—3×3, 4—4×4, 5—5×5. Each cluster is 4 layers thick.

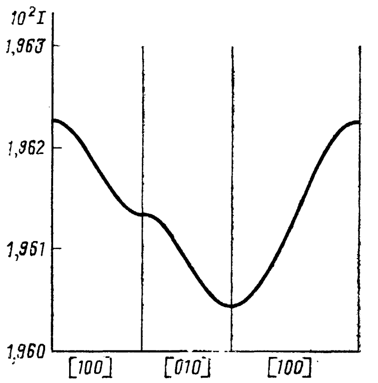

FIG. 5. Tunnel-current relief along individual directions in the surface plane, cell dimension 5×5 in the plane at a crystal thickness 5 layers.

The variations of the tunnel current (Fig. 5) are quite small. But this is connected with our illustrative model. More accurately speaking, the smallness of the current variation is due to the use of *s*-type orbitals in the crystal and in the tip for the spectrum construction. Since the overlap integral of the orbitals in the tip and in the crystal depends only on the mutual distance and is independent of angle, the current variations can be easily estimated analytically. Let us find the current variations when the needle is exactly above one of the surface atoms and in the case when it is located exactly above the center of a surface cell of a crystal (the center of a square with coordinates (1/2,1/2) the crystal surface-layer atoms at the vertices of the square). The difference between the currents in these two cases causes the maximum modulation amplitude of the tunnel current. For small overlap integrals the tunnel current is proportional to ξ^2. It is easy to obtain an expression for the current when the tip is above a point with coordinate (0,0) exactly above a surface atom, recognizing that $\xi^2 \simeq (e^{-2(r-1)})^2$, we get

$$I(0,0) \approx (e^{-2})^2 + 4\{\exp[-2(5^{1/2}-1)]\}^2 \approx 0{,}04812,$$

where the first term of the equation corresponds to the overlap integral between the tip atom and the surface atom directly beneath it. The second term accounts for the contribution to the tunnel current by the atom-orbital overlap on a crystal surface with coordinates ($\pm$1,0), (0, $\pm$1), with the orbital of the atom on the end of the tip. The overlaps with more remote neighbors is neglected everywhere (this is justified by the fast exponential decrease of the overlap integral with distance).

We can similarly obtain the tunnel current when the tip is located above a point (1/2, 1/2) of the crystal surface, viz.,

$$I({}^1/_2, {}^1/_2) \approx 4\{\exp[-2({}^9/_2)^{1/2}-1)]\}^2 \approx 0{,}04808,$$

where account is taken of atoms with coordinates (0,0), (1,0), (0,1), and (1,1). We obtain this

$$I(0,0) - I({}^1/_2, {}^1/_2) \approx 0{,}00004,$$

which is of the same order as the numerical results.

We emphasize once more that these small variations are connected with the use of *s*-orbitals in the tip and in the crystal. In fact, assume that the crystal spectrum is produced by p_x or p_y orbitals which are "dumbbells" parallel to the crystal surface. It is easy to verify that in this case the current variations are of the order of the current itself. Let the tip be located directly above one of the atoms; the contribution of this crystal atom to the tunnel is in this case zero (this follows from symmetry considerations). Since the overlap integral $\xi(s,p_{x,y})$ of the *s* and $p_{x,y}$ orbitals is an odd function of the coordinates in the plane, it follows that $\xi(s,p_{x,y}) \propto \cos(s;p_{x,y})$ (here $\cos(s;p_{x,y})$ is the cosine of the angle between the radius drawn from the outermost atom of the tip to the given surface atom, and the vector drawn to the same atom on the surface from the origin). The estimated tunnel current is

$$I(0,0) \approx \{\cos^2[s;\ p_x(1,0)] + \cos^2[s;\ p_x(-1,0)]$$
$$+\cos^2[s;\ p_y(0,1)]$$
$$+\cos^2[s;\ p_y(0,-1)]\}\{\exp[-2(5^{1/2}-1)]\}^2;$$

here $\cos[s;p_x(1{,}0)]$ corresponds to a p_x orbital centered on an atom with coordinates (1,0) and on equivalent atoms, and $(\cos^2[s;p_x(1{,}0)] = 1/5)$. Substitution yields

$$I(0,0) \approx {}^4/_5 e^{-4\cdot 1{,}25}.$$

Similar calculations can be made when the tip is above a surface point with coordinate (1/2,1/2). We obtain

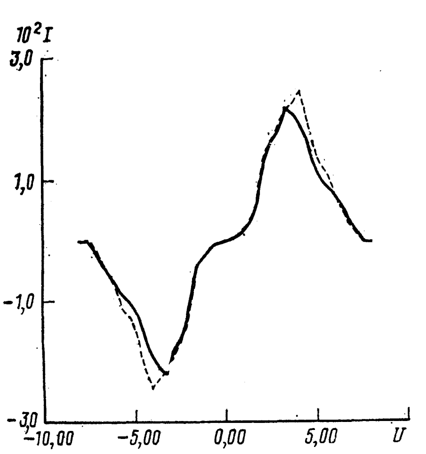

FIG. 4. Current-voltage characteristics for 5×5 cluster in a plane, 4 layers (solid) and 5 layers (dashed) thick.

$$\begin{aligned} I(^1/_2, {}^1/_2) \approx \{&\cos^2[s;\ p_x(0,0)] + \cos^2[s;\ p_y(0,0)] \\ &+\cos^2[s;\ p_x(1,0)] \\ &+\cos^2[s;\ p_y(1,0)] + \cos^2[s;\ p_x(0,1)] \\ &+\cos^2[s;\ p_y(0,1)] + \cos^2[s;\ p_x(1,1)] \\ &+\cos^2[s;\ p_y(1,1)]\}\{\exp[-2((^9/_2)^{1/2}-1)]\}^2. \end{aligned}$$

Recognizing that all the cosines are equal $[\cos^2(s;p_x(1,0)) = 1/9]$, we get

$$I(^1/_2, {}^1/_2) \approx {}^8/_9 e^{-4\cdot 1,15}.$$

The corresponding variation of the values of the tunnel currents takes the form

$$1 - I(0,0)/I(^1/_2, {}^1/_2) \approx 1 - {}^9/_{10} e^{-0,4} \approx 1 - (^9/_{10}) \cdot (1/1,8) \approx 0,5.$$

As follows from the approximate analysis, the current-modulation amplitude can be of the same order as the current itself. The directed valence orbitals are typical of covalent semiconductor compounds, where a strong modulation of the tunnel current is in fact observed.

CONCLUSION

The method proposed reduces thus the problem of calculating the critical value to determination of the spectrum of the quasistationary states. This problem can be solved by the usual methods of band theory. The approach takes into account, in natural fashion, of the atomic structure of both the crystal surface and the tip. We have demonstrated above the validity of the method using a tight-binding example. This circumstance, however, is not a constraint, and the approach can be used with other methods of calculating a band spectrum. From the formal point of view the method used to find the spectrum of the tip + crystal system is utterly unimportant. In all cases the problem reduces to construction of a Hamiltonian matrix into which the terms with damping are next inserted.

Strictly speaking, in the presence of an external electric field it is necessary to solve simultaneously an electrostatic problem and the problem of spectrum calculation. This requirement is not a constraint in our method. It suffices to recognize that the potential difference between one end of the tip and the outer side of the crystal is given, and determine in self-consistent manner the spectrum of such a cluster.

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Translated by J. G. Adashko