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Dynamical image interaction in scanning tunneling microscopy

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We consider the effect of the motion of the electron on the image interaction, a problem investigated earlier by Persson and Baratoff [Phys. Rev. B **38**, 9616 (1988)]. If one is interested in the semiclassical description of the image interaction for the tunneling process, arising from the interaction of the charge of the electron with the surface plasmons, then one should make use of a trajectory for the electron such that the plasmon system is not excited as a result of tunneling. We show how such a trajectory may be obtained. Our analysis leads to the conclusion that the trajectory obeys boundary conditions different from the ones used by Persson and Baratoff. Calculations of the tunneling exponent using this trajectory indicate that dynamical effects can modify the image interaction to a much larger extent than found by them.

In an interesting paper, Persson and Baratoff[1] (PB) have investigated the influence of the motion of the electron on image effects in tunneling. They consider tunneling in which at least one of the electrodes is a metal and consequently one has to account for the lowering of the barrier for tunneling, caused by the image interaction. They take the image as arising from the interaction of the charge of the electron with the surface plasmons. Following PB, we denote the tunneling time in the absence of image interaction by τ_0 and the frequency of the plasmons as ω_s. If τ_0 is large compared to the response time ω_s^{-1} of the plasmons then one expects the image interaction to be given by the usual static expression $-e^2/4z$. On the other hand, if $\omega_s\tau_0 \sim 1$, one expects dynamical effects to be important and the effective barrier height to be different from the static one. Persson and Baratoff[1] have developed a self-consistent theory for this situation. They find dynamical effects to be important and conclude that the static image potential overestimates the tunneling exponent by 20–30 % for a typical scanning tunneling microscopy (STM) experiment. These dynamical effects have been experimentally studied by Guéret.[2] It is the aim of this paper to argue that the approach of PB, though in the correct direction, has to be modified in its philosophy if one is interested in description of the image interaction in STM. This results in a change in the trajectory that is to be used for the calculation of the tunneling probability and leads to stronger dynamical effects than found by PB.

PB make use of path-integral techniques for their derivation. Their approach is the one adopted by Caldeira and Leggett[4] (CL) for calculating tunneling probabilities in cases where the tunneling particle is coupled to a heat bath, which consists of a collection of harmonic oscillators, coupled linearly to the coordinate of the tunneling particle. The harmonic oscillators have a continuum of frequencies ranging from zero to a maximum cutoff. Consequently, it is possible for the tunneling particle to excite several of them during the tunneling. Strictly speaking, one should find one path for the description of excitation to each final state, but as in the problem, there is a continuum of excitations and anyway one is only interested in the tunneling probability and not in the final state of the heat bath; one finds an average trajectory, which is suited for the description of the motion of the particle, irrespective of what the final state of the bath is. This average trajectory is a closed loop—i.e., if the particle starts on the left-hand side of the barrier, it goes to the right until its momentum becomes equal to zero and then goes back to where it started. This is a result of attempting to calculate the *probability* of tunneling, rather than the amplitude. Notice further that the equation of motion that results couples the motion in the left and right directions. As is obvious, in the tunneling process, only half of this (the portion from left to right) is actually traversed by the particle. One can, if one desires, demonstrate this explicitly by analyzing the probability amplitude for any given process. As probability is the square of the absolute value of amplitude, it is clear that its calculation shall require two such halves, which one may imagine to form a closed trajectory. So if one tries to find an average trajectory, for the calculation of probability of tunneling, the result is a closed trajectory, with coupling between different portions of it.

If one takes half of the closed trajectory (from left to right) and analyzes it, it is possible to show that in such a description, the energy of the particle on the left-hand side is not the same as its energy on the right-hand side.[3] That is, the particle loses energy in the process of tunneling, which is not surprising, as it would have excited the harmonic oscillators in the tunneling process.

The problem addressed by PB is quite similar in that the electron is coupled to the plasmons. But plasmons have a rather high frequency ω_s, and unless the voltage between the two electrodes is larger than ω_s, there is no actual excitation of the plasmons during the tunneling process. If one makes use of the semiclassical description, following CL, one would end up with a trajectory that would have energy loss to the plasmons, because in such a description, there is no constraint that the energy transfer is quantized. So it is possible for any amount of energy to be transferred to the plasmons. Therefore, one does not have a consistent description of the image interactions, if one adopted such an approach. It may be thought that the correct trajectory, which does not have these spurious inelastic effects in it, should not be much different from the one caused by PB, and consequently, the tunneling exponents obtained by PB should not be changed much. Our calculations indicate that this is not so, and in the range of parameters studied by PB, the difference could be large (see below for actual values).

The modification in the formalism that is necessary is the following: PB are interested in the semiclassical description of the tunneling particle, irrespective of what excitation has been created in the metal. For an STM experiment where there is no possibility of exciting a plasmon, the best path to choose for the description of tunneling is the one which obeys the condition that the final state after the tunneling process is the ground state $|0\rangle$ of the plasmon system. This means that one considers the scattering matrix element given by

$$\mathcal{S}_{0\to 0}=\int D\mathbf{x}(t)\exp\{iS_0[\mathbf{x}(t)]/\hbar\}\langle 0|\mathcal{U}[\mathbf{x}(t)]|0\rangle \ . \quad (1)$$

In the above

$$S_0[\mathbf{x}(t)]=\int dt\{(m/2)[d\mathbf{x}(t)/dt]^2-U(\mathbf{x}(t))\} \ . \quad (2)$$

$U(\mathbf{x}(t))$ is the potential due to the barrier in the absence of the image interaction and m is the mass of the electron. $\mathcal{U}[x(t)]$ is the time development operator for the plasmons, given that they are subject to the force from the electron, which follows the trajectory $\mathbf{x}(t)$. For an application of a similar method to atom-surface scattering, see the paper of Newns.[5] See also Feynman and Hibbs,[6] Pechukas,[7] Mohring and Smilansky,[8] and Miller and co-workers[9-11] for details on this type of approach.

In the case of image interactions, the coupling to plasmons can be taken to be linear in the coordinates of plasmons (see PB), and one can calculate $\langle 0|\mathcal{U}[\mathbf{x}(t)]|0\rangle$ exactly. Further, in order for tunneling to occur, one has to allow time to become imaginary ($t\to -i\tau$), following Miller and George.[9] This leads to the effective action

$$S_{\text{eff}}[x(\tau)]=\int_0^T d\tau\{(m/2)[d\mathbf{x}(\tau)/d\tau]^2+U(\mathbf{x}(\tau))\} +S_1[\mathbf{x}(\tau)] \ , \quad (3)$$

where the potential $U(\mathbf{x}(t))$ appears with a positive sign because one has made time imaginary, and in imaginary time the potential gets inverted.[12] The modification of the action, due to the interaction of the particle with the plasmons is given by

$$S_1[\mathbf{x}(t)]=-\frac{e^2}{2\pi\hbar}\int_0^T d\tau\int_0^T d\tau'\int_0^\infty dq\int_0^\infty d\omega\,\text{Im}g(q,\omega)\exp[-\omega|\tau-\tau'|-q\{z(\tau)+z(\tau')\}] \ . \quad (4)$$

$g(q,\omega)$ is the surface-linear response function, $q=|\mathbf{q}|$, where $\mathbf{q}$ is the two-dimensional wave vector, and ω is the frequency. Note that we follow the definitions of PB for q and $g(q,\omega)$. This equation is similar to that of PB even though they consider the probability of tunneling, while we consider the probability amplitude. An estimate of the magnitude of the tunneling amplitude is obtained as $\exp(-S_m/\hbar)$ where S_m is the value of S_{eff} for the path that makes S_{eff} a minimum, subject to conditions discussed below. The tunneling probability itself would be $\exp(-2S_m/\hbar)$. T is the time needed to cross the barrier in one direction, and is not the period of the bounce, which would be approximately $2T$. (For PB, T is the period of the bounce.)

We now follow PB and consider their model (a), in which one has a barrier of constant height ($=U_0$) in the region $0<z<L$. The tunneling time $\tau_0=L\sqrt{(m/2U_0)}$ and action for the barrier without image effects, $S_0=L\sqrt{2U_0m}$. Further, we introduce dimensionless variables, $\bar{z}=z/L$, $\bar{\tau}=\tau/\tau_0$ and measure action in units of S_0, by defining $\bar{S}=S/S_0$. Then, for the model (a) of PB, we get the action $\bar{S}$ to be

$$\bar{S}=\int_0^{\bar{T}}d\bar{\tau}\frac{1}{2}\left[\left(\frac{d\bar{z}}{d\bar{\tau}}\right)^2+R(\bar{z})\right] -\tfrac{1}{2}\xi\eta\int_0^{\bar{T}}d\bar{\tau}\int_0^{\bar{T}}d\bar{\tau}'\frac{\exp[-\xi|\bar{\tau}-\bar{\tau}'|]}{2-\bar{z}(\bar{\tau})-\bar{z}(\bar{\tau}')} \quad (5)$$

with $\bar{T}=T/\tau_0$, $R(\bar{z})=1$ if $0\le\bar{z}\le 1$ and 0 otherwise. $\xi=\omega_s\tau_0$ and $\eta=e^2/4LU_0$. Minimizing the action in the above equation, one finds the equation of motion of the particle to be

$$\frac{d^2\bar{z}}{d\bar{\tau}^2}=-\xi\eta\int_0^{\bar{T}}d\bar{\tau}'\frac{\exp[-\xi|\bar{\tau}-\bar{\tau}'|]}{[2-\bar{z}(\bar{\tau})-\bar{z}(\bar{\tau}')]^2} \ . \quad (6)$$

The conditions on the path and $\bar{T}$ are (1) the particle should start on the left-hand side of the barrier at time $\bar{\tau}=0$, i.e., $\bar{z}(0)=0$; (2) the initial momentum of the particle is given by

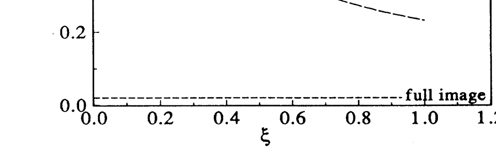

FIG. 1. The results of calculations using Eqs. (5)–(7). Se text for details.

$$\left[\frac{d\overline{z}}{d\overline{\tau}}\right]_{\overline{\tau}=0} = \left[1-2\xi\eta\int_0^T d\tau' \frac{\exp[-\xi\overline{\tau}']}{2-\overline{z}(\overline{\tau}')}\right]^{1/2} ; \quad (7)$$

(3) the value of $\overline{T}$ must be such that either $(d\overline{z}/d\overline{\tau})_{\overline{T}}=0$, or if the particle hits the right-hand side of the barrier before this condition is satisfied, then $\overline{T}$ should be taken to be given by $\overline{z}(\overline{T})=1$. In comparison with this, the condition used by PB is $\overline{z}(\overline{T})=0$ because they look for a closed trajectory. In actual calculations we have, following PB, replaced the 2 in the Eqs. (5)–(7) with 2.2.

In Fig. 1 we have plotted the results of calculations using the above equations. It shows $\overline{B}=S_m/S_m(g=0)=S_m/S_0$, the ratio of the exponent for tunneling probability to the exponent in the absence of image interaction, as a function of ξ for two different values of η. The upper three curves are for $\eta=0.1$ and the lower three are for $\eta=1.0$. Notice that the two horizontal lines (with short dashes) indicate $\overline{B}$ in the presence of full image interaction. The dashed lines (with longer dashes) are the results obtained using the method of PB, while the full lines are the results of our approach. It is seen that the value of $\overline{B}$ that we obtain is always greater than that of PB. The physical reason for this is the following: In the approach of PB, the electron loses its energy during the tunneling process and slows down, in contrast to our approach in which there is no spurious energy loss. Consequently, dynamical effects would be stronger in our approach. If $\xi<1$, then the time spent by the electron in the barrier is smaller than the time needed for the plasmon system to respond. Hence, the barrier height seen by the electron is larger than the one if the full image interaction is present. It is clear that for the smaller value of η $(=0.1)$ where the image interaction itself is not of much importance, the dynamical effect, can also be neglected. On the other hand, in the case $\eta=1$ and $\xi=1$, then $\overline{B}$ with the full image interaction is 0.024, while its value obtained by our method is 0.38. In comparison PB's theory gives 0.23. For a typical STM experiment, for which one might choose $\hbar\omega_s \sim U_0 \sim 5$ eV and $L \sim 4$ Å, one gets $\xi \sim 2$ and $\eta \sim 0.2$. Using these values, we find for the relative exponent of the tunneling probability, $\overline{B}=0.84$. In comparison with this, if one had the full image interaction, $\overline{B}$ would be 0.70, while using the approach of PB, one gets 0.77.

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