Photostimulated field emission: A theoretical attempt to find rapid oscillations with applied field

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Using a surface-effect model of photoemission that includes coupling through the divergence of the radiation field, we have tried to find a theoretical reason for experimentally observed rapid oscillations in the yield as a function of applied static field. Model calculations reveal no plausible qualitative explanation.

Photostimulated field emission (PFE) is a process which combines features of both photoemission[1] (PE) and field emission[2] (FE). In PFE, as in FE, a static electric field, typically of order 0.1 V/Å, is applied to a metal deforming the shape of the surface potential into a tunneling barrier. In addition the metal is illuminated with a radiation field from which the electrons can absorb energy as in PE. The simultaneous presence of the static and radiation fields allows one to probe states that lie above the Fermi level yet below the unperturbed vacuum level. States in this energy range are ordinarily, inaccessible to either PE or FE alone, so there has been a natural interest in the development of the hybrid PFE process.

Several groups have reported experimental PFE studies on a variety of clean and adsorbate covered surfaces.[3–18] Although most of the data are amenable to at least a qualitative understanding in terms of simple models of bulk and/or surface electronic structure, we focus here on a feature that has so far defied a convincing explanation. It is that the total PFE current density j has been observed—not always, but at least several times[4–6,17]—to oscillate rapidly as a function of the applied static field F. Similar oscillations in both thermionic emission and photoemission were observed and explained previously[19] for values of F much smaller than those used in present PFE. Since the explanation then lay in oscillations of the electron transmission coefficient, analogous calculations have been tried for the recent PFE data.[20–22] However in neither WKB nor exact numerical solutions has one found in the transmission coefficient the rapid oscillations reported for j in experiments. A common feature of these calculations, as well as of most recent theories of the excitation process,[20–24] is that only the simplest surface-effect model is used: band-structure effects are neglected, the possibility of adsorbates ignored, and the radiation field is presumed to be spatially constant. Here we explicitly improve on the last approximation, but in the process also gain insight into the first two. However, we do not succeed in finding a theoretical explanation of the reported rapid oscillations.

In the PE literature, the spatial variation of the vector potential, $\vec{A}$, of the radiation field has recently received renewed interest.[25–28] All these works begin with the remark that within the textbook Fresnel treatment, the normal component of $\vec{A}$ (taken to be the z direction) is discontinuous at the metal surface. They then show how, within various models of surface electronic structure, this discontinuity is spread out over a distance comparable to a screening length. The precise spatial dependence of $\vec{A}$ is difficult to calculate, and we shall take here the extreme limit of retaining the step discontinuity. This certainly gives the correct integrated strength of $\vec{\nabla}\cdot\vec{A}$ and probably acts to enhance any oscillatory structure in the PFE yield. There remains only an ambiguity as to where the discontinuity should be located. We will treat this position $\tilde{z}$ as a free parameter.

This allowance for a spatial dependence in A_z is the only change we are making from previous model treatments. We continue to use a Sommerfeld treatment for the electronic structure. The only static potential energy variation an electron sees is at the surface where we use either one of two models (see Figs. 1 and 2): The triangular barrier (TB) for which

$$V(z) = (V_0 - eFz)\Theta(z) \ , \tag{1}$$

or the image rounded barrier (IR) for which

$$V(z) = (V_0 - eFz - e^2/4z)\Theta(z - z_0) \ . \tag{2}$$

In these equations Θ is the unit step function, e is the magnitude of an electron's charge, and z_0 in (2) is chosen so that V is continuous. The parameter V_0 is the depth of the unperturbed well confining the electrons to the metal ($z < 0$). It is the sum of the Fermi energy, E_F, and the work function. These three energies along with the plasma frequency, ω_p, are the free-electron parameters that we choose to mimic the properties of tungsten.[20]

It has been previously shown[21,22] that within this approach the total emitted current density in PFE is

given by

$$j = \int dE \left(\frac{dj}{dE} \right) , \tag{3}$$

where

$$\frac{dj}{dE} \propto \int_{\hbar\omega}^{E} dW D(W) |M|^2 [W(W - \hbar\omega)]^{-1/2} . \tag{4}$$

Here E is the total energy of the electron after absorption of a photon of energy $\hbar\omega$ and one must have $E - \hbar\omega < E_F < E$. The transmission coefficient of the surface barrier is D and M is the matrix element linking initial and final states. Both states have a plane-wave variation parallel to the surface, described by the common wave vector $\vec{K}$, and the normal energy W is defined by $E = W + \hbar^2 K^2/2m$, with m the electron mass. A more explicit form of the surface effect matrix element is

$$M \propto \int dz\, v(z) \left(A_z \frac{d}{dz} + \frac{1}{2} \frac{dA_z}{dz} \right) \phi(z) , \tag{5}$$

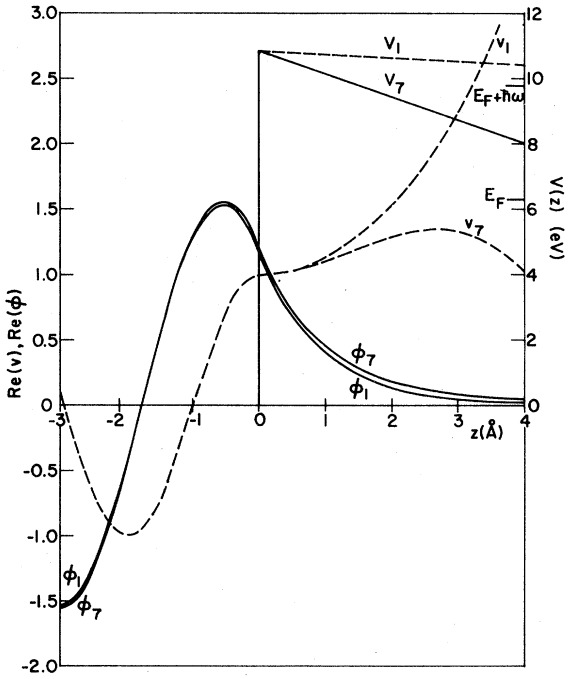

FIG. 1. The potential $V(z)$ and the real part of the wave functions $\phi(z)$ and $v(z)$ for the triangular barrier at the applied fields $F = 0.1$ and 0.7 V/Å. The broken curve labeled V_1 is the potential for $F = 0.1$ V/Å, while the solid curve labeled V_7 is the potential for $F = 0.7$ V/Å. The wave functions are similarly labeled by the appropriate subscript. The solid curve in each case is ϕ whose energy is the Fermi level $E_F = 6.3$ eV and the broken curve is v whose energy is $E_F + \hbar\omega = 9.8$ eV. By definition (Ref. 20), the state v is independent of F for $z \leq 0$ and the states v and ϕ have different normalizations.

where v and ϕ are solutions of the one-dimensional Schrödinger equation in the potentials of (1) or (2) at the energies W and $W - \hbar\omega$, respectively. In Figs. 1 and 2 we have plotted the real parts of v and ϕ for two choices of F which span the region where oscillations in j have been reported. The differences in the final states, v, between the two figures are easily understood. At the energy W used here, the excited electron in the TB model must tunnel through the barrier while in the IR model it can pass over the barrier. A more important feature to notice is the general weak dependence of all states on F. Furthermore, as one varies F between the two extremes shown, both v and ϕ change smoothly.

Previous evaluations of (3)–(5) used a spatially constant A_z.[20–25] With our new approximation we use in (5)

$$A_z = \Theta(z - \tilde{z}) + \epsilon^{-1} \Theta(\tilde{z} - z) , \tag{6}$$

where ϵ is the (Drude) dielectric function of the metal

$$\epsilon(\omega) = 1 - \omega_p^2/\omega^2 . \tag{7}$$

As we remarked earlier, (6) is a crude representation of A_z but gives us perhaps the best chance to find oscillations in j versus F. In this same spirit we have suppressed all factors in (4)–(6) that do not change the F dependence. Finally, we shall only present here calculations of $|M|^2$ versus F for an initial state

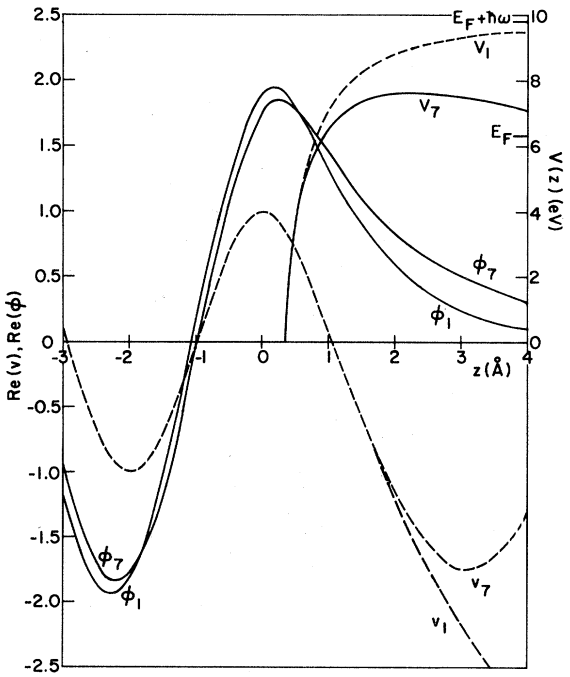

FIG. 2. The same as Fig. 1 but now for the image rounded barrier.

at the Fermi level with $\vec{K}=0$, i.e., for $W-\hbar\omega=E_F$. The combined cutoffs imposed by the Pauli principle and by the transmission coefficient, limit the integrals in (3) and (4) to essentially such states. Again this procedure would seem only to accentuate any possible oscillations.

In Figs. 3 and 4 we plot $|M|^2$ with $E=W=E_F+\hbar\omega$ as a function of F at fixed illumination conditions. The frequency of the radiation field is considerably smaller than the nominal plasma frequency; $\hbar\omega=3.5$ eV versus $\hbar\omega_p=14.9$ eV. The potential parameters are the same as in Figs. 1 and 2: $V_0=10.8$ eV and $E_F=6.3$ eV, and the various curves are labeled by the value of $\tilde{z}$. As F varies, $\tilde{z}$ is held fixed; except for the IR case where for $\tilde{z}>z_0$, it is $\tilde{z}-z_0$ that is held fixed. This exception, made for numerical convenience, should not matter since the total change in z_0 with F is only 0.006 Å. It is clear from the figures that $|M|^2$ is a monotonic function of F for both the TB and IR models and for any reasonable choice of $\tilde{z}$. In further calculations we found that varying the energy of the states in M had no qualitative effect. We can find no rapid oscillations in $|M|^2$.

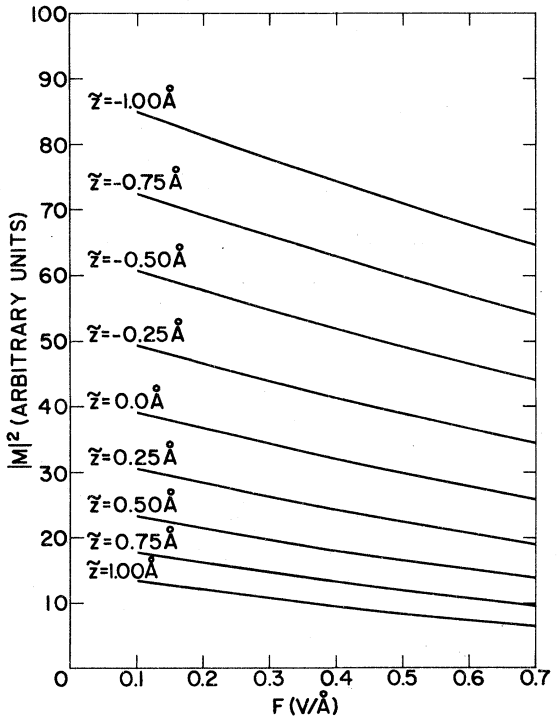

FIG. 3. Absolute square of the surface effect matrix element $|M|^2$ (in arbitrary units) for the triangular barrier as a function of the applied field F (in units of V/Å) for various locations $\tilde{z}$ of the discontinuity of the Fresnel field. In each case the vector potential is characterized by the frequency $\hbar\omega=3.5$ eV and $\hbar\omega_p=14.9$ eV.

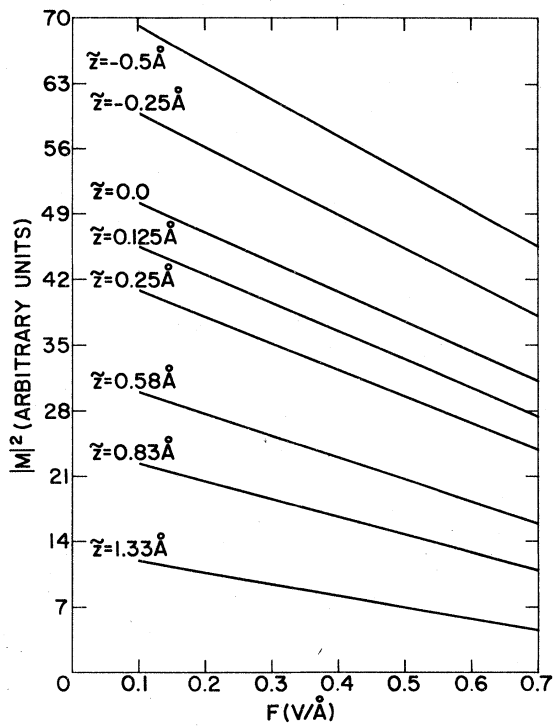

FIG. 4. The same as Fig. 3 but now for the image rounded barrier.

Combining these results for the excitation matrix element with earlier work on the transmission coefficient,[20,22] we conclude that surface effect models of PFE as presently formulated cannot explain rapid oscillations of the yield with applied static field. The experiments[4–6,17] show more than 10 oscillations over a range of F smaller than that considered here, while our calculations are essentially monotonic in F. The explanation of the theoretical results is clear from Figs. 1 and 2: The states involved smoothly and scarcely change with the large applied fields used here. In turn this behavior arises from the slight changes of the surface barrier with F. Only the transmission function shows a strong dependence on F, yet it too is monotonic.

This qualitative view of the limitations of the theory can be used to assess the importance of effects we have omitted. For instance, if we allow for adsorbed particles, then resonances can occur at isolated energies.[29] But to produce agreement with the PFE data, one would need to presume essentially as many resonances as there are oscillations.[6] This assumption would be physically unreasonable. Another possibility would be band-structure effects. While it is reasonable to suppose that evidence of the onset of bulk transitions should appear as shoulders or sudden changes of slope in j versus F data,[7–10] we are again stuck with the dilemma of too much implied band

structure in order to explain the rapid oscillations. Further one has the problem of how such sharp (supposed) bulk structure can be so sensitive to surface conditions.[4–6,17]

In summary, we have not been able to find a plausible theoretical explanation of the rapid oscillations occasionally seen in PFE data. Of course our theoretical model is rather crude; so our conclusion is not completely firm. Still the qualitative aspects elucidated here argue strongly that, if the rapid oscillations are not an experimental artifact, they owe their origin to a rather subtle cause.

ACKNOWLEDGMENT

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