Ionization of surface-state electrons by microwave fields: Quantum treatment

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We calculated quantum mechanically the response of surface-state electrons to an applied microwave field. By solving an effective Schrödinger equation which takes into account both bound-bound and bound-continuum transitions to all orders, we show that the ionization proceeds via direct coupling to the continuum. This contrasts with the classical description, where ionization occurs as the result of a diffusive energy gain.

Jensen[1] analyzed recently the response of surface-state electrons (SSE) to an external microwave field. He showed that the classical dynamics of this system displays a transition from a regular to a stochastic regime and suggested that this transition could be experimentally observed by measuring the ionization rate as a function of the microwave-field parameters.

We report here on our quantum-mechanical (QM) studies of the microwave ionization of SSE for the same range of field parameters, for which the transition from regular to stochastic dynamics occurs classically, and for which Jensen proposed to conduct the actual experiment.

The time-dependent Hamiltonian governing the system is

$$H=H_0+V(x,t);\quad H_0=\frac{P^2}{2m}-\frac{Ze^2}{x};\quad V(x,t)=-eEx\sin(\omega t)\ . \tag{1}$$

The classical treatment of this problem suggested that in the chaotic regime ionization occurs as the result of a diffusive energy gain. The quantum analog of such a process would correspond to a multistep excitation process, which cannot be treated perturbatively. In order not to miss any important aspect of the quantum behavior of the SSE system, a nonperturbative calculation of the SSE dynamics governed by the Hamiltonian (1) is clearly necessary.

To this end, the wave function is expanded in the complete set (bound and continuum) of eigenfunctions of H_0. Using standard projection techniques, one obtains an exact description of the system in terms of the bound-state amplitudes $b_n(\tau)$ only:

$$i\frac{d}{d\tau}b_n(\tau)=-\frac{1}{n^2}b_n(\tau)+\gamma\sin(\Omega\tau)\sum_m\left\{\langle n|(\alpha x)|m\rangle b_m(\tau)-i\alpha\gamma\int_0^\tau d\tau' b_m(\tau')\sin(\Omega\tau')K_{nm}(\tau,\tau')\right\}\ . \tag{2}$$

Here, energies are measured in terms of the Rydberg constant R ($\approx 6.7\times10^{-4}$ eV for the SSE problem). τ is the time in $\hbar/R$ units and $\alpha=2m(Ze^2)/\hbar^2$. $\gamma=eE/\alpha R$ measures the coupling of the SSE to the rf field and $\Omega=\hbar\omega/R$. The time integral in (2) introduces the effects of the continuum on the bounded motion. The kernel $K_{nm}(\tau,\tau')$ is approximated by the Coulomb propagator which amounts to neglecting continuum-continuum transitions induced by the external field.[2] In this approximation, which is valid if the ionization rate is kept small, we obtain

$$K_{nm}(\Delta)=\tfrac{1}{2}\int_0^\infty d\chi\langle n|(\alpha x)|\chi\rangle e^{-i\chi^2\Delta}\langle\chi|(\alpha x)|m\rangle,\qquad \Delta=\tau-\tau'>0\ . \tag{3}$$

Here, $|n\rangle$ and $|\chi\rangle$ are properly normalized bound and continuum eigenfunctions of H_0, respectively. Since the bound-free matrix elements appearing in (3) are known analytically the K_{nm} integrals can be calculated explicitly.

The integrodifferential equation (2) is converted into a set of first-order differential equations by expanding each $K_{nm}(\Delta)$ into a finite series of decaying exponentials:

$$K_{nm}(\Delta)=\sum_{j=1}^{N_{nm}}[A_{nm}^{(j)}\exp(B_{nm}^{(j)}\Delta)-i\tilde{A}_{nm}^{(j)}\Delta\exp(\tilde{B}_{nm}^{(j)}\Delta)]\ . \tag{4}$$

Introducing the variables:

$$\begin{aligned}
y_{nm}^{(j)}(\tau)&=\exp(B_{nm}^{(j)}\tau)\int_0^\tau b_m(\tau')\sin(\Omega\tau')\exp(-B_{nm}^{(j)}\tau')d\tau'\ ,\\
\tilde{y}_{nm}^{(j)}(\tau)&=\exp(\tilde{B}_{nm}^{(j)}\tau)\int_0^\tau b_m(\tau')\sin(\Omega\tau')\exp(-\tilde{B}_{nm}^{(j)}\tau')d\tau'\ ,\\
z_{nm}^{(j)}(\tau)&=\exp(\tilde{B}_{nm}^{(j)}\tau)\int_0^\tau b_m(\tau')\tau'\sin(\Omega\tau')\exp(-\tilde{B}_{nm}^{(j)}\tau')d\tau'\ .
\end{aligned} \tag{5}$$

The set of Eqs. (2) transforms into

$$\begin{aligned}
i\dot{b}_n&=-\frac{1}{n^2}b_n+\gamma\sin(\Omega\tau)\sum_m\{\langle n|\alpha x|m\rangle b_m-i\gamma\sum_j[A_{nm}^{(j)}y_{nm}^{(j)}-i\tilde{A}_{nm}^{(j)}(\tau\tilde{y}_{nm}^{(j)}-z_{nm}^{(j)})]\}\ ,\\
\dot{y}_{nm}^{(j)}&=B_{nm}^{(j)}y_{nm}+b_m\sin(\Omega\tau),\quad \dot{\tilde{y}}_{nm}^{(j)}=\tilde{B}_{nm}^{(j)}\tilde{y}_{nm}+b_m\sin(\Omega\tau),\quad \dot{z}_{nm}^{(j)}=\tilde{B}_{nm}^{(j)}z_{nm}+b_m\tau\sin(\Omega\tau)\ .
\end{aligned} \tag{2'}$$

Details of the numerical accuracy and the practicality of this method will be given elsewhere.

The Eqs. (2′) do not conserve the quantity $P_B(\tau) = \sum_n |b_n(\tau)|^2$, i.e., the probability that the system remains bounded. Rather, we found that $P_B(\tau)$ decays exponentially, and we define the ionization rate by

$$I_R = -\frac{2\pi}{\omega}\left\langle \frac{d}{d\tau} \ln P_B(\tau) \right\rangle , \tag{6}$$

where the average is taken over a few field cycles to smooth over periodic modulations which appear in $P_B(\tau)$ when the rf field is in resonance with any of the dominant transition frequencies of the SSE spectrum.

In all the calculations reported here the SSE is initially in its ground state ($n=1$). The field strengths were taken to be smaller than the "static ionization threshold" defined by $\gamma = \frac{1}{16}$ ($E = 110$ V/cm). At this field, the ground-state energy coincides with the continuum threshold of the potential at $\omega t = \pi/2$. The frequencies span the interval from $\Omega = 0.2$ to $\Omega = 2.0$ which includes the region of classical stochasticity ($\Omega \approx 1$).

In Fig. 1 we compare the classical and QM predictions for the probability to remain bounded [$P_B(\tau)$]. The classical analog of the SSE wave function is constructed by a set of trajectories in (action-angle) phase space, all starting with an action variable $n=1$ and evenly distributed in angle. After an initial build-up phase where the chaotic trajectories diffuse towards the continuum threshold the classical probability to remain bounded decays exponentially and finally approaches a constant value which represents the fraction of regular orbits in the initial ensemble. Considering only the set of irregular trajectories,[3] we obtain a qualitative agreement between the classical and the exponential QM decay. This agreement, however, is accidental and does not imply any resemblance between the classical and the QM ionization processes. For $\Omega > 1$ this point is proven by the following observations. (a) The probability to excite any of the bound states with $n > 1$ was rather small, suggesting a direct transition from the ground state to the continuum. Indeed, there is no difference in the ionization rates between a full ten-states calculation and a calculation including only the ground state [see Fig. 2(b)]. The ionization rates depend quadratically on the field strength γ, which implies a first-order (direct) transition to the continuum.

For $\Omega < 1$ the number of chaotic trajectories in the initial set diminishes drastically and for rf fields lower than the static ionization threshold we observe negligible ionization. In contrast with this result the QM calculations predict appreciable ionization due to resonances and threshold effects as can be seen in Fig. 2(a). In order to get a better understanding of the mechanism which produces the complicated features in the ionization rates, we show in Fig. 2(b) ionization rates calculated with only one or two bound states (besides the continuum). These calculations are only relevant in the vicinity of the dominant features and therefore the curves shown in Fig. 2(b) do not cover the entire frequency domain.

The thresholds at $\Omega = 1, \frac{1}{2}, \frac{1}{3}$ ($f \simeq 163, 82, 55$ GHz) are due to the matching of the field frequency with the direct (1,2,3 photon) transitions from the ground state to the continuum. They occur also in a calculation in which only the

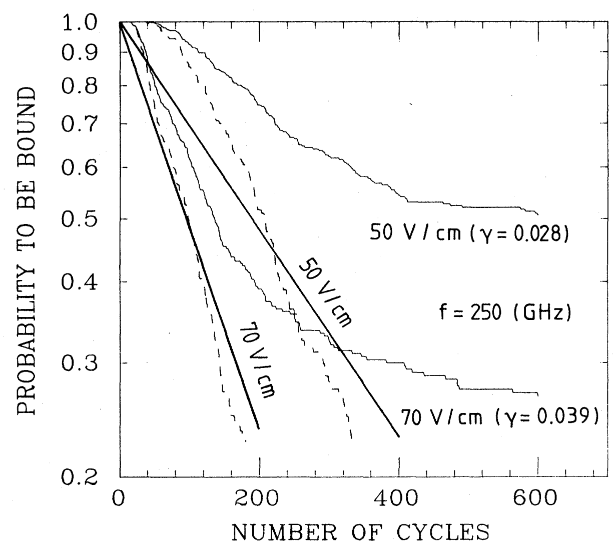

FIG. 1. Classical vs QM decay of the probability of the SSE to remain bounded. Full heavy line, QM. Thin line, classical. Dashed line, classical, considering only the subspace of irregular trajectories.

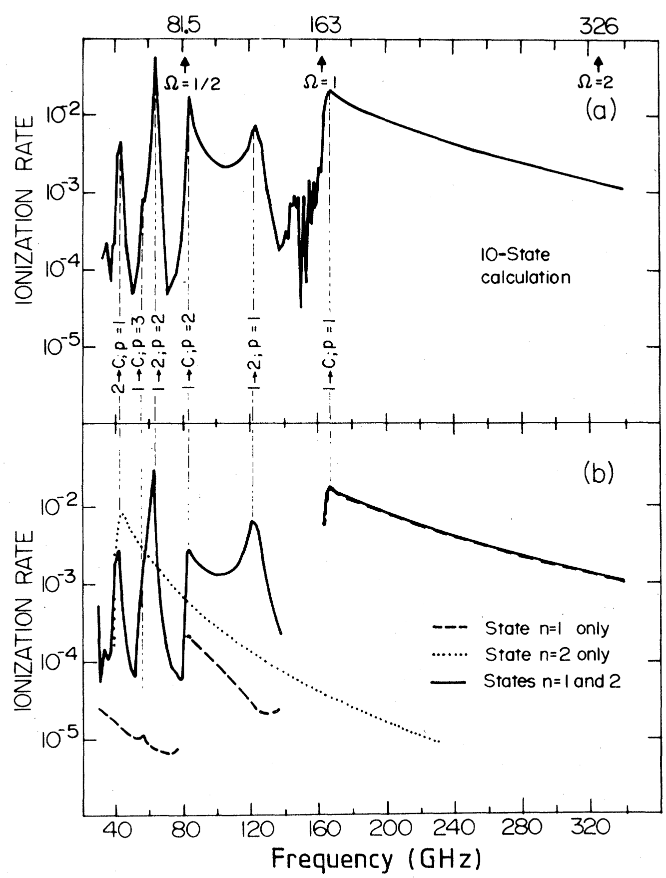

FIG. 2. QM ionization rates for $E = 50$ V/cm ($\gamma = 0.028$). The SSE is initially in its ground state. (a) Full-scale calculations, including ten bound states. (b) Calculations including selected bound states only. The vertical dashed lines mark the resonances, threshold, and their assignments.

ground state is considered [dashed line in Fig. 2(b)]. The peak at $\Omega=\frac{1}{4}(f\simeq 41$ GHz) occurs at the ionization frequency for the second state as can be attested by the dotted line which was calculated by considering the state with $n=2$ only. The two resonances at $\Omega\approx\frac{3}{4}$ and $\Omega\approx\frac{3}{8}(f\simeq 120,63$ GHz) correspond to one- and two-photon transitions between the $n=1$ and $n=2$ states, and they appear as prominent peaks in a calculation in which only these two states are considered [full line in Fig. 2(b)]. The spikes in the ionization rate near the $\Omega=1$ threshold correspond to resonances of the ground state with the $n\geqslant 3$ states.

The dependence of the ionization rates on the field strength reflects the nature of the transitions involved. In the domain $\Omega>1$, $I_R\sim\gamma^2$, manifesting the first-order (direct) nature of the transition. For lower frequencies I_R cannot be expressed as a power law of γ since the ionization is due to the interference of various excitation routes. Exceptions occur near the dominant resonances where $I_R\sim\gamma^{2n_p}$, n_p being the order of the relevant transition amplitude. Thus, e.g., $I_R(\gamma;\Omega\simeq 1/2)\simeq\gamma^4$.

Our main result is that classical stochasticity is suppressed in the present case. Moreover, the QM ionization rates can be used to assert the localized nature of the SSE wave function, and to stress other purely quantal effects (resonances), which dominate the SSE dynamics. This is consistent with previous QM studies, which were only able to provide an incomplete discussion of the SSE problem, either because the ionization channel (continuum) was not taken into account,[4] or because the external rf field was represented by a periodic $\delta(t)$ force.[5] This, however, does not preclude the possibility that a closer similarity between the classical and the quantal predictions may exist, e.g., for the ionization of Rydberg states.[4,6]

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