Exact solution of the 1D time-dependent Schrödinger equation for the emission of quasi-free electrons from a flat metal surface by a laser

Joel Lebowitz

Rutgers University

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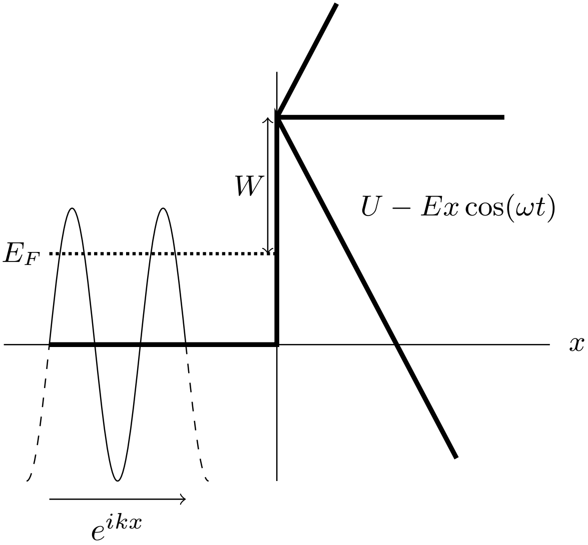
Introduction

- There have been many advances in recent years in the development and application of short intense laser pulses to produce femto-second and even atto-second beams of electrons from metallic surfaces.
- A full microscopic description of the short-time behavior of the emission process is therefore highly desirable.
- Here, I will describe our recent work where we obtained, for the first time, an exact solution for the time-dependent Schrödinger equation describing the emission of electrons from a flat metal surface by an oscillating electric field.

- We use the Sommerfeld model of quasi-free electrons with a Fermi distribution of energies, confined by a step potential $U = \mathcal{E}_F + W$, where $\mathcal{E}_F$ is the Fermi energy and W is the work function of the metal.
- This setup was first used by Fowler and Nordheim in 1928 for a time-independent field, and is commonly used as a model for the process of emission, both for a constant and an oscillating field.
- In both cases one imagines the metal occupies the half space $x < 0$, and focuses attention on electrons, part of the Fermi sea, moving from the left towards the metal surface at $x = 0$.
- These are described by a wave function e^{ikx}, $k > 0$, $x < 0$ and have energy $\frac{1}{2}k^2$ (in atomic units).



W
$U - Ex\cos(\omega t)$
E_F
x
e^{ikx}

- The time evolution of the wave function of an electron in such a beam subjected to an oscillating field for $x \geqslant 0$, is described by the one dimensional Schrödinger equation: for $x \in \mathbb{R}$ and $t > 0$,

$$i\partial_t \psi(x,t) = -\frac{1}{2}\Delta\psi(x,t) + \Theta(x)(U - Ex\cos(\omega t))\psi(x,t) \qquad (1)$$

- $\Theta(x) = 0$ for $x < 0$ and $\Theta(x) = 1$ for $x > 0$, E is the electric field perpendicular to the surface, $\frac{\omega}{2\pi}$ is the frequency and we are using atomic units $\hbar = m = e = 1$.

- Fowler and Nordheim considered the case of a constant field ($\omega = 0$) and assumed that $\psi(x,t)$ has the form $e^{-\frac{1}{2}k^2 t}\varphi_E(x)$, so that $\varphi_E(x)$ satisfies the equation

$$-\frac{1}{2}\Delta\varphi_E + \Theta(x)(U - Ex)\varphi_E = \frac{1}{2}k^2\varphi_E. \tag{2}$$

- The solution φ_E is then $\varphi_E(x) = e^{ikx} + R_E e^{-ikx}$ for $x < 0$ and has an Airy function expression for $x > 0$.
- The computation of the tunneling current

$$j(x,t) := \mathcal{I}m(\psi^*(x,t)\partial_x\psi(x,t)) \tag{3}$$

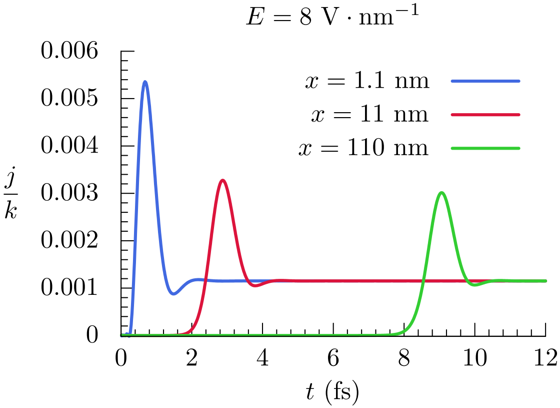
from the FN solution, is still the basic ingredient for the analysis of experiments at present.

- In a previous work we solved the time-dependent Schrödinger equation for a constant field starting from a realistic initial state.
- In particular, we took as an initial state the "stationary" solution in which there is no field $E = 0$, $\psi(x,t) = e^{-\frac{k^2}{2}t}\varphi_0(x)$.
- The requirement of continuity of ψ and of $\partial_x\psi$ at $x = 0$ then gives

$$\varphi_0(x) = \begin{cases} e^{ikx} + \dfrac{ik+\sqrt{2U-k^2}}{ik-\sqrt{2U-k^2}}e^{-ikx} & \text{for } x<0 \\ \dfrac{2ik}{ik-\sqrt{2U-k^2}}e^{-\sqrt{2U-k^2}x} & \text{for } x>0 \end{cases} \tag{4}$$

- $\varphi_0(x)$ consists of
  - a reflected beam of the same energy and intensity as the incoming beam e^{ikx},
  - an evanescent, exponentially decaying tail on the right.

- We showed that $\psi(x,t)$ converges, as $t \to \infty$, to the FN solution, with a rate of convergence $t^{-\frac{3}{2}}$.
- Surprisingly, the deviation of the current from the FN solution becomes quickly very small.



Time evolution of the current for $\omega = 0$ starting from $\varphi_0(x)$

Solution of the Schrödinger equation for an oscillating field

- The solution of the Schrödinger equation with $E > 0$, $\omega > 0$ covers a wide range of physical situations, depending on ω and E, ranging from mechanically produced oscillating fields to those produced by lasers of high frequency.
- As the Keldysh parameter $\gamma := \frac{2\omega}{E}\sqrt{W}$ increases, the process goes from tunneling to multi-photon emission.
- Solving the time-dependent Schrödinger equation with initial condition φ_0 turns out to be much more difficult than the constant field case.
- The very existence of physical solutions, which are bounded at infinity, and are continuously differentiable at $x = 0$, is not mathematically obvious.

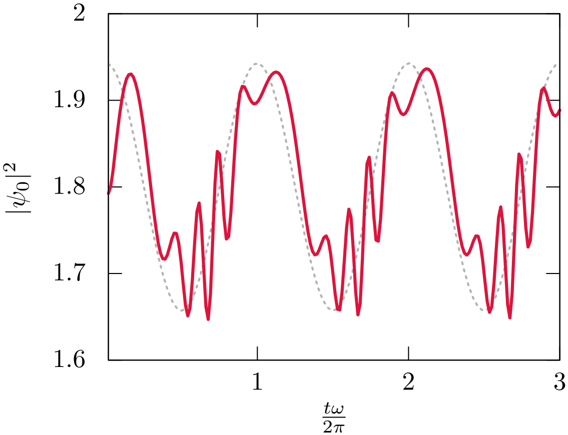
- We prove the existence of such solutions $\psi(x, t)$ for all physical initial conditions $\psi(x, 0)$.
- The solution is expressed in terms of convergent integrals.
- The solution converges for long time, at a rate $t^{-\frac{3}{2}}$, to a periodic solution considered by Faisal et al.
- Using the exact solution, we compute $\psi(x, t)$ for $t > 0$ via an exponentially convergent algorithm, taking as an initial condition $\varphi_0(x)$.

Behavior of solutions

The following figures show the results of the solution of the Schrödinger equation (1) for

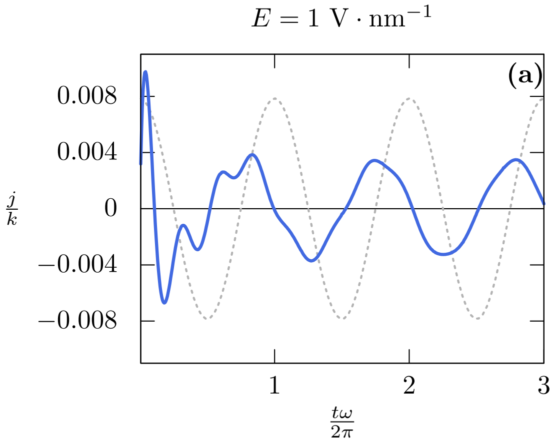
- $\frac{k^2}{2} = \mathcal{E}_F = 4.5\,\text{eV}$
- $W = 5.5\,\text{eV}$
- unless otherwise specified, $\omega = 1.55\ \mathrm{eV}$
- $\hbar = m = e = 1.$
- the laser period $\frac{2\pi}{\omega}$ is then equal to $2.7\ \mathrm{fs}$.

Surface Density



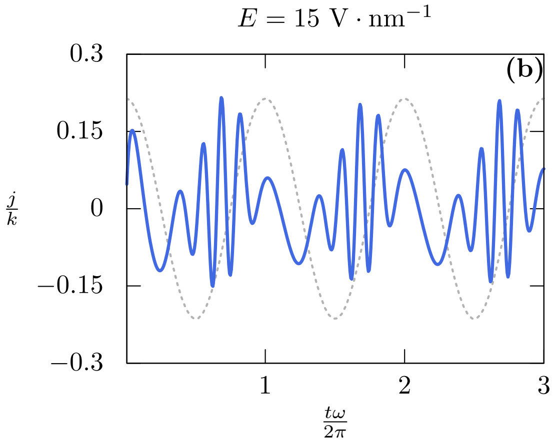
- $E = 15\ \mathrm{V} \cdot \mathrm{nm}^{-1}$.
- The dotted line is the graph of $\cos(\omega t)$ (not to scale).

Surface current



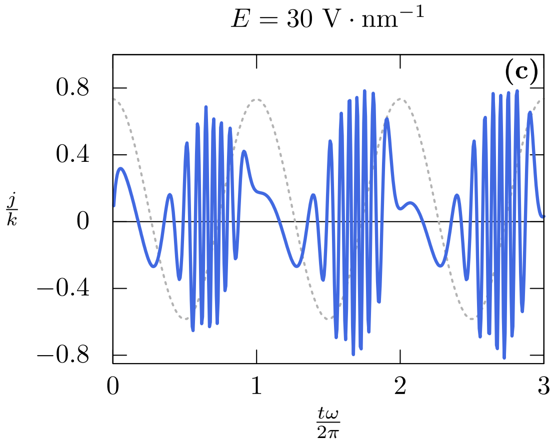
- The Keldysh parameter is $\gamma = 18.6$.

Surface current



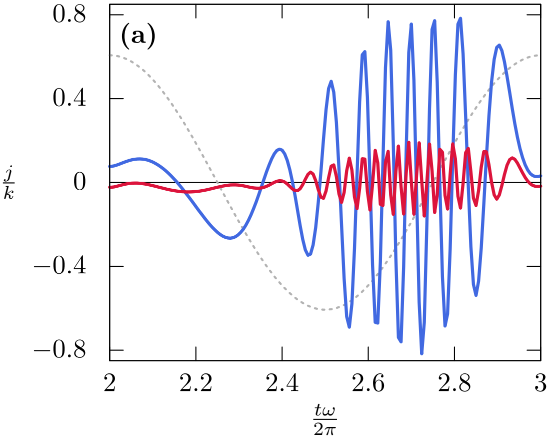
- The Keldysh parameter is $\gamma = 1.24$.

Surface current



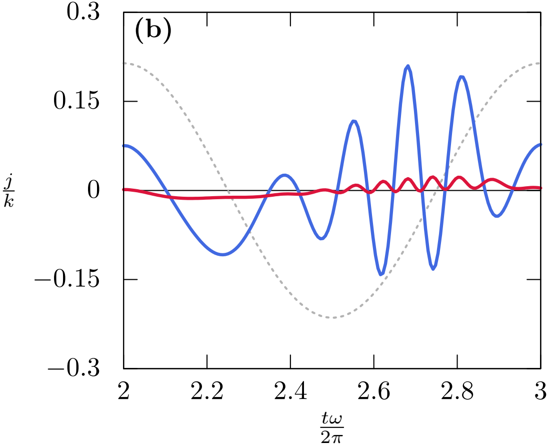
- The Keldysh parameter is $\gamma = 0.621$.
- For large values of E, fast oscillations appear as the field is increasing, which become faster and larger as E grows.

Surface current



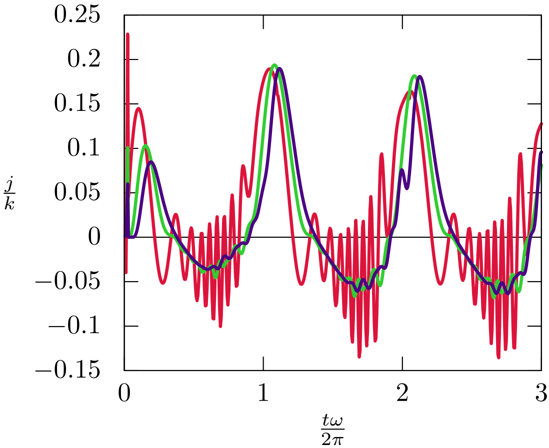
- Keldysh: $\gamma = 0.621$.
- Blue: $E = 30\ \mathrm{V} \cdot \mathrm{nm}^{-1}$ and $\omega = 1.55\ \mathrm{eV}$.
- Red: $E = 15\ \mathrm{V} \cdot \mathrm{nm}^{-1}$ and $\omega = 0.755\ \mathrm{eV}$.
- The frequency of the fast oscillations does not only depend on γ.

Surface current



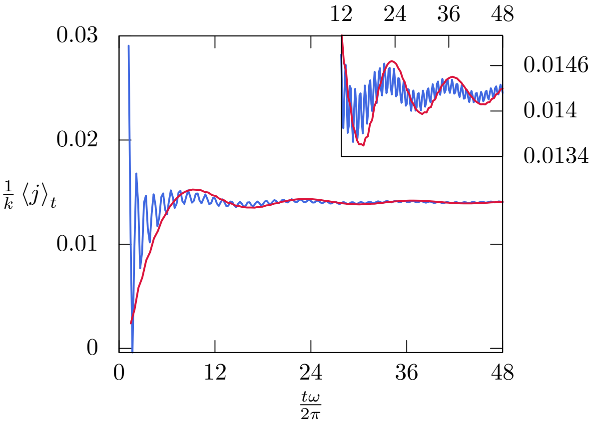
- Keldysh: $\gamma = 1.24$.
- Blue: $E = 15\ \mathrm{V} \cdot \mathrm{nm}^{-1}$ and $\omega = 1.55\ \mathrm{eV}$.
- Red: $E = 7.5\ \mathrm{V} \cdot \mathrm{nm}^{-1}$ and $\omega = 0.755\ \mathrm{eV}$.
- At fixed E, the frequency of the oscillations decreases with ω.

Current at $x \neq 0$



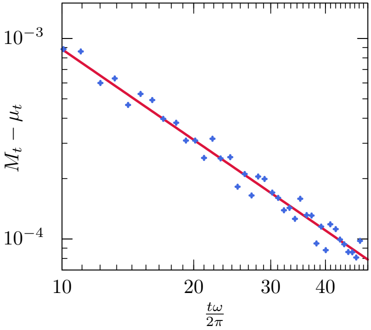
- $x = 0.12$ nm (red), $x = 0.24$ nm (green), $x = 0.37$ nm (purple).
- The fast oscillations die down quickly as x gets larger.

Average current as a function of time



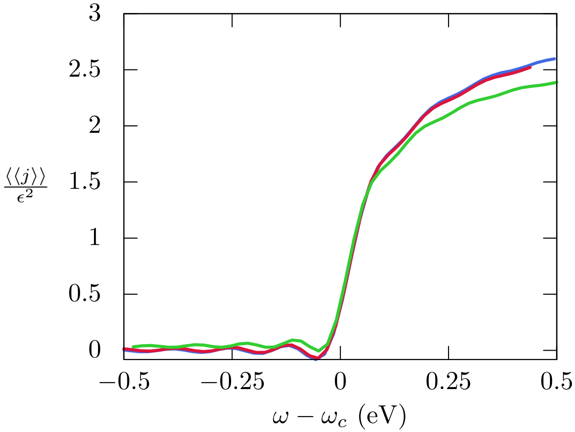
- $E = 10\ \mathrm{V}\cdot\mathrm{nm}^{-1}$, $\omega = 6\ \mathrm{eV}$.
- The blue curve is the surface current ($x = 0$), and the red curve is at $x = 0.37\ \mathrm{nm}$.

Convergence rate of the average current



- $E = 10\ \mathrm{V} \cdot \mathrm{nm}^{-1}$, $\omega = 6\ \mathrm{eV}$.
- The blue dots show the difference between the maximum and minimum of the average current within one period as a function of time, and the red line is $0.0030(\frac{t\omega}{2\pi})^{-\frac{3}{2}}$, which confirms that the rate of convergence is $t^{-\frac{3}{2}}$.

Critical frequency $\omega_c = W + \frac{E^2}{4\omega^2}$



- $\frac{E^2}{4\omega^2}$ is the ponderomotive energy of the electron in the field.
- $E = 3\ \mathrm{V \cdot nm^{-1}}$ (blue), $E = 10\ \mathrm{V \cdot nm^{-1}}$ (red), $E = 30\ \mathrm{V \cdot nm^{-1}}$ (green).
- There is a "sharp" transition at ω_c, which corresponds to the Einstein relation for the photoelectric effect $\omega_c = W$ when E is small.

Future plans

- The model described above is one of the simpler ones used to study electron emission.
- We plan to extend our results to metallic tips, such as are used in experimental setups.
- We plan to study situations where the laser field has a more realistic spatial structure, inside a carrier wave.
- We plan to include the Shottky correction to the potential, which rounds off the triangular barrier.

Solution of the Schrödinger equation

We solve the Schrödinger equation by using the one sided Fourier transforms $\hat{\psi}_-(\xi, t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{0} \mathrm{e}^{-i\xi x} \psi(x, t)\, dx$,
$\hat{\psi}_+(\xi, t) = \frac{1}{\sqrt{2\pi}} \int_{0}^{\infty} \mathrm{e}^{-i\xi x} \psi(x, t)\, dx$.
These satisfy

$$i \frac{\partial \hat{\psi}_-(\xi, t)}{\partial t} - \frac{\xi^2}{2} \hat{\psi}_- = \frac{1}{\sqrt{2\pi}} \frac{\partial \psi}{\partial y}(0, t) - i\xi \frac{1}{\sqrt{2\pi}} \psi(0, t) \tag{5}$$

and

$$\begin{aligned} i \frac{\partial \hat{\psi}_+(\xi, t)}{\partial t} + i \frac{E}{2} \cos(\omega t) \frac{\partial \hat{\psi}_+}{\partial \xi} - \left(\frac{\xi^2}{2} + U \right) \hat{\psi}_+(\xi, t) = \\ = \frac{1}{\sqrt{2\pi}} \frac{\partial \psi}{\partial x}(0, t) + i\xi \frac{1}{\sqrt{2\pi}} \psi(0, t) \end{aligned} \tag{6}$$

These have explicit solutions for initial values $\hat{\psi}_\pm(\xi, 0)$ and specified boundary values $\psi(0, t)$ and $\partial_x \psi(0, t)$.

- The continuity conditions for ψ and $\partial_x \psi$ at $x = 0$ then lead to an integral equation for $\psi(0, t)$ of the form

$$\psi(0, t) = h(t) + L\psi(0, t) \tag{7}$$

- L is some compact integral operator whose expression is rather involved, and $h(t)$ is a function of the initial condition $\psi(x, 0)$.
- We prove the existence and uniqueness of a physical solution of (7) for all $t > 0$, by showing that L is a contraction.
- Given that solution $\psi(0, t)$, we obtain $\psi(x, t)$ for all x by direct integration.
- To evaluate the solution numerically for our initial condition, we expand $\psi(0, t)$ in a Chebyshev polynomial series and identify the coefficients of this expansion.

References

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