Phase Sensitive Electron Emission from Metal Nanostructures

by

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Physics of Nanostructures

Characteristic Parameters and time scale

Surface Plasma Oscillations/ Plasmons

Main Problem

Interaction of Strong Fields with Atoms

Theory and Equations

Results and Discussions
Interaction of Strong Fields with Atoms

Characteristic parameters and time scale:

average velocity of the valence electron:

\[ v_0 = \sqrt{\frac{2R_y}{m_e}} = \sqrt{\frac{27.2 \text{(eV)}}{0.511 \text{(MeV/c}^2)} = \alpha c \quad [\alpha \text{-Fine structure constant}] \]

\[ v_0 = \frac{c}{137} \]
Characteristic parameters and time scale:

average distance (Bohr radius):

$$
\langle V_{pot} \rangle = -2R_y = -\frac{e^2}{4\pi\varepsilon_0 a_0}
$$

$$
a_0 = \frac{e^2}{8\pi\varepsilon_0 R_y} \approx 0.529 \times 10^{-10} \text{ (m)}
$$
Characteristic parameters and time scale:

average orbit time:

\[ \tau_{\text{orbit}} = \frac{2\pi a_0}{\nu_0} = \frac{2\pi}{3 \times 10^8} \times 137 \]

\[ \approx 2\pi \times 24.2 \times 10^{-18} \text{ (s)} \]

[atomic unit of time]

\[ \tau_{\text{orbit}} \approx 150 \text{ (as)} \]
Characteristic times of quantum systems

<table>
<thead>
<tr>
<th>Characteristic State</th>
<th>$\Delta E$</th>
<th>$\tau$ (fs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>valence electron in atoms</td>
<td>13 eV</td>
<td>150</td>
</tr>
<tr>
<td>valence electron molecules</td>
<td>same as in atoms</td>
<td></td>
</tr>
<tr>
<td>vibrational motion of nuclei in molecules</td>
<td>100 meV</td>
<td>20</td>
</tr>
<tr>
<td>inner shell electrons</td>
<td>1 keV</td>
<td>2</td>
</tr>
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</table>

Attosecond Physics is the Physics of valence electrons

For the time being
Surface Plasma Oscillations

The electron charges on a metal-vacuum (or metal-dielectric) boundary can perform periodic oscillations of charge density which are called surface plasma oscillations (Langmuir waves or plasma waves). These oscillations, can be excited by external optical fields at the interface.

\[ \varepsilon_2 = \varepsilon'_2 + i\varepsilon''_2 \]
\[ \varepsilon'_2 > 0 \]

\[ \varepsilon_1 = \varepsilon'_1 + i\varepsilon''_1 \]
\[ \varepsilon'_1 < 0 \]
EM waves are quantized in to units having energy $E = \hbar \omega$ called **photons**.

Plasma waves are quantized in to units having energy $E_p = \hbar \omega_p$ called **plasmons**.

The **plasmon** is the quantization of plasma oscillations.

$$\omega_p = \sqrt{\frac{4\pi ne^2}{m}} \quad \text{(in cgs)}$$
Dispersion Relation of Surface Plasmons at a interface

\[ E_1 = (E_{x1}, 0, E_{z1})e^{i(k_{x1}x - k_{z1}z\omega t)}; \ z < 0 \]

\[ E_2 = (E_{x2}, 0, E_{z2})e^{i(k_{x2}x + k_{z2}z\omega t)}; \ z > 0 \]

\[ k_{zl} = \sqrt{\varepsilon_i \left(\frac{\omega}{c}\right)^2 - k_x^2}, \ i = 1,2 \]

where \( k_x = \frac{\omega}{c} \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}} \)
For metals;  \(|\varepsilon| > 1\), then

\[
k_x = \frac{\omega}{c} \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}} > \frac{\omega}{c}
\]

\[
k_{z_i} = \sqrt{\varepsilon_i \left(\frac{\omega}{c}\right)^2 - k_x^2} \Rightarrow \text{complex or imaginary}
\]

The fields have their maximum in the surface and decay in both z directions exponentially and localized in the z direction within the Thomas-Fermi length of about \(1\AA\).
Surface Plasmons

Plasmons play a significant role in the optical properties of metals.

Light waves, below the plasma frequency is reflected, because the electrons in the metal screen the electric field of the light.

Light waves, above the plasma frequency is transmitted, because the electrons cannot respond fast enough to screen it.

In most metals, the plasma frequency is in the UV range, making them shiny in the visible range.
Phase Sensitive Electron Emission
from Metal Nanostructures
Interaction of Strong Fields with Atoms

Ultrashort (*single or few oscillation*) laser pulse:

\[ \vec{E}(t) = E_a(t) \cos(\omega t + \phi) \hat{e}_z \]

- \text{amplitude envelop}
- \text{absolute phase}

\( T_0 \)
Classical laser field

High intensity $\sim 10^{12} (W/cm^2)$

Long wavelength $\sim 800 (nm)$

Photon energy $\hbar \omega \sim 1.5 (eV)$
Dipole approximation

Spatial variations of the field can be neglected. This is because of the extension of the interacting system is, in general, much smaller than the wave length of the laser.

The typical wave length of 800 nm to the Bohr radius of 0.05 nm!!
Ionization in a static electric field

Quasi-static ionization

For high field strength (~ $10^{12}$ W/cm$^2$) and long (800 nm) wavelength ionization can be considered as quasi-static.

\emph{i.e. at any moment in time the atom gets ionized at a rate like in a static electric field of strength $E(t)$}
Tunneling Ionization

For any non-vanishing static field, at sufficiently large distances the total potential energy \((atomic\ binding + dipole\ field)\) is below the bound state energy and the electron can leave by tunneling through the potential barrier.

We consider strong-field electron emission that takes place when the energy barrier separating the metal electrons from the surrounding space (whose height is defined by workfunction \(I_p\)) is slanted by a electric field causing the electron tunneling.
Tunneling Ionization

$E(t)=0$

$E(t)$
Tunneling Ionization

Such a process occurs for relatively small values of the Keldysh parameter \( \gamma \)

\[
\gamma = \sqrt{\frac{I_p}{U_p}} \leq 1
\]

where \( U_p \) is the electron quiver energy

\[
U_p = \frac{e^2 E^2}{2m \omega^2}
\]
The quasistationary emission is of advantage for our purpose since its dependence on the parameters of the problem. \textit{(in particular absolute phase)}
We can separate the electron emission process into two stages:

(1). Quantum tunneling stage.

(2). Subsequent almost classical motion in the free space.

(1). Quantum tunneling stage:

Ionization in a static electric field

Hamiltonian: 
\[
H = -\frac{\hbar^2}{2m} \nabla^2 - \frac{e^2}{4\pi\varepsilon_0} \frac{1}{r} - \vec{r} \cdot \vec{E}_0
\]
Exponential Dependence of Tunneling Ionization

\[
tunneling \text{ probability } \sim \exp \left[ 2 \int_0^{r_0} \frac{\sqrt{2m}}{\hbar} \sqrt{eI_p \left( 1 - \frac{x}{r_0} \right)} \, dx \right]
\]

\[
= \exp \left[ \frac{4}{3e\hbar} \frac{\sqrt{2mI_p^3}}{E(t)} \right]
\]

Strongly non-linear (exponential!) dependence on parameters

Wave mechanics of crystalline solids, R.A. Smith (1969)
Initial electron velocity after tunneling

Outside the binding potential, the electron more or less behaves like a free electron. We can easily solve the equation of motion (classically or quantum mechanically), if one can make a reasonable assumption about initial position and momentum.

At the end of tunnel

\[ E = -\frac{e^2}{4\pi\varepsilon_0} \frac{1}{r} - r_0E_0 \]

\[ -\frac{\hbar^2}{2m} \nabla^2 \Psi \bigg|_{r_0} = \left( -\frac{e^2}{4\pi\varepsilon_0} \frac{1}{r} - rE_0 - E \right) \Psi \bigg|_{r_0} \]

Expect velocity \( \sim 0 \) at \( r_0 \)

Electron velocity after the tunneling

\[ m\ddot{r} = eE_0(t)\cos(\omega t) \]

\[ m\dot{r} = \frac{eE_0(t)}{\omega} \sin(\omega t) \]

A(t)

\[ \frac{1}{c} \]

\[ v(t) = \frac{e}{mc} A(t) - \frac{e}{mc} A(t_0) \]

\[ v_d \]
Tunneling Probability per unit time

\[ W(t) = \frac{8\hbar}{m e^2} \varepsilon_f I_p^2 \nu_f \exp \left[ \frac{4}{3e\hbar} \sqrt{\frac{2mI_p^3}{E(t)}} \right] \theta[-E(t)] \theta[-A(t)] \]

Where:
- \( \theta[...] \) - unit step function
- \( \varepsilon_f \) - Fermi energy
- \( \nu_f \) - electron state density at Fermi surface
- \( E(t) \) - component of the time-dependent local electric field normal to the surface
- \( A(t) = \frac{c}{t} \int E(t) dt \)

*L.D. Landau and E.M. Lifshitz, Quantum Mechanics (1965)*
Current Density

\[ J = -env_d \int_{-\infty}^{\infty} n \exp \left( \frac{E_0}{E(t)} \right) \theta[-E(t)] \theta[-A(t)] dt \]

\( n \) – the density of electrons in the emitting layer

\( v_d \) – component of the drift velocity normal to the metal surface
Advantages of Metal (Silver) Nanosystem

The electron photoemission current exponentially depends on the time kinetics of the local optical electrical field \( E(t) \) at every point of the metal nanosystem. To find this field in an general case for a strong excitation field \( E \) would have been an extremely complicated, unrealistic task.

However, there are the following two properties of the metal nanosystems that make an approximate solution possible.

(1). The dielectric permittivity \( \varepsilon \) of metals is large and negative.

(2). We consider near-infrared spectral region. In this case, the quality factor of the metal plasmon resonances is high enough. i.e. \( \text{Im}(\varepsilon) \langle - \text{Re}(\varepsilon) \rangle \)
The Geometry of the Silver Nanostructures

V-shape RPC

Surface plasma (SP) life time as a function of SP eigenenergy $\hbar \omega_n$

Temporal dependencies of local electric optical field (the \( y \) component, in units of excitation field \( E_0 \)) at the apex of \( \text{V-shape} \) (at \( \hbar \omega = 1.55 \text{eV} \)). The red squares denote the temporal points contributing to the current, the blue triangles denote the points that do not contribute to the photocurrent.
The red squares denote the temporal points contributing to the current
**Major Results**

We have investigated strong optical-field emission of electrons from a metal nanostructure at relatively low intensity.

This originates from the first few oscillations of the local field due to the interference of the surface plasmons eigenmodes from a wideband packet excited by the ultrashort pulse.

This photoemission is shown to be highly sensitive to the absolute phase of the pulse.

Applications:

- to control ultrafast response of metal nanostructure.
- to measure the absolute phase of the laser pulse at relatively low intensity.
- and controllable photoinjection of electrons to free space or semiconductor.