Part II

Interaction with Single Atoms
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Multiphoton Ionization
Tunneling Ionization
Ionization-Induced Defocusing
High Harmonic Generation in Gases

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Bohr model recap.

At the Bohr radius

\[ a_B = \frac{\hbar^2}{me^2} = 5.3 \times 10^{-9} \text{ cm}, \]

the electric field strength is:

\[ E_a = \frac{e}{a_B^2} \quad \text{(cgs)} \]

\[ \geq 5.1 \times 10^9 \text{ Vm}^{-1}. \]

This leads to the atomic intensity:

\[ I_a = \frac{cE_a^2}{8\pi} \quad \text{(cgs)} \]

\[ \simeq 3.51 \times 10^{16} \text{ Wcm}^{-2}. \]
Multiphoton Ionization

**Figure:** a) Multiphoton ionization (MPI): Electron with binding energy $E_{\text{ion}}$ simultaneously absorbs $n$ photons with energy $\hbar \omega$ and escapes from atom with minimal kinetic energy. b) Above-threshold ionization (ATI): electron absorbs more photons than necessary for ionization, acquiring momentum.
Above-threshold ionization (ATI)

- Experiments: distinct peaks in electron spectra beyond the ionization energy $E_{\text{ion}}$, separated by the photon energy $\hbar \omega$.
- Final kinetic energy of electron is given by an extended version of Einstein’s formula:

$$E_f = (n + s)\hbar \omega - E_{\text{ion}}, \quad (5)$$

where $n$ is the number of photons needed for multiphoton ionization; $s$ is the excess absorbed.
Above-threshold ionization (ATI): measurements of electron spectra

**Figure 2.** Electron spectra of eleven-photon MPI at 1604 nm for different pulse energies. The first peak vanishes at around 7 mJ. The maximum total count rate in these spectra was ten per laser shot.

Keldysh (1965) and Perelomov (1966): introduced a parameter \( \gamma \) separating the multiphoton and tunneling regimes, given by:

\[
\gamma = \omega_L \sqrt{2E_{\text{ion}}} \frac{I_L}{\ell} \sim \sqrt{\frac{E_{\text{ion}}}{\Phi_{\text{pond}}}}.
\]  

(6)

where

\[
\Phi_{\text{pond}} = \frac{e^2 E_L^2}{4m \omega_L^2}
\]

(7)

is the *ponderomotive potential* of the laser field.

\( \gamma < 1 \Rightarrow \) tunneling – strong fields, long wavelengths

\( \gamma > 1 \Rightarrow \) MPI
Figure: a) Schematic picture of tunneling or barrier-suppression ionization by a strong external electric field.
Tunnelling: barrier suppression model II

- Coulomb potential modified by a stationary, homogeneous electric field, see Fig. 10:

\[ V(x) = -\frac{Ze^2}{x} - e\varepsilon x. \]

⇒ suppressed on RHS of the atom, and for \( x \gg x_{\text{max}} \) is lower than the binding energy of the electron.

- If the barrier falls below \( E_{\text{ion}} \), the electron will escape spontaneously

⇒ over-the-barrier (OBT) or barrier suppression (BS) ionization.
Differentiate $V(x)$ to determine the position of the barrier,

$$x_{\text{max}} = \frac{Ze}{\varepsilon},$$

then set $V(x_{\text{max}}) = E_{\text{ion}}$ to get the threshold field strength for OTBI:

$$\varepsilon_c = \frac{E_{\text{ion}}^2}{4Ze^3}.$$  (8)
Barrier suppression model IV

- Equate critical field to the peak electric field of the laser – *appearance intensity* for ions created with charge $Z$:

$$I_{app} = \frac{c}{8\pi\varepsilon_0^2} = \frac{cE_{ion}^4}{128\pi Z^2 e^6}, \quad (9)$$

or:

$$I_{app} \simeq 4 \times 10^9 \left(\frac{E_{ion}}{\text{eV}}\right)^4 Z^{-2} \text{ Wcm}^{-2}. \quad (10)$$

- NB: $E_{ion}$ is the ionization potential of the ion or atom with charge $(Z - 1)$. 

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Appearance intensity: Hydrogen example

- Hydrogen: $Z = 1$

\[ E_{\text{ion}} = E_h = \frac{e^2}{2a_B} = 13.61 \text{ eV}. \]

- Making use of Eq. (??), the critical field for hydrogen is:

\[ \varepsilon_c = \frac{E_h^2}{4e^3} = \frac{e}{16a_B^2} = \frac{E_a}{16}, \]

- Appearance intensity:

\[ I_{\text{app}} = \frac{I_a}{256} \simeq 1.4 \times 10^{14} \text{ Wcm}^{-2}. \]
Appearance intensities of selected ions according to the BS ionization model – Eq. (10).

<table>
<thead>
<tr>
<th>Ion</th>
<th>$E_{ion}$ (eV)</th>
<th>$I_{app}$ (Wcm$^{-2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H$^+$</td>
<td>13.61</td>
<td>1.4 × 10$^{14}$</td>
</tr>
<tr>
<td>He$^+$</td>
<td>24.59</td>
<td>1.4 × 10$^{15}$</td>
</tr>
<tr>
<td>He$^{2+}$</td>
<td>54.42</td>
<td>8.8 × 10$^{15}$</td>
</tr>
<tr>
<td>C$^+$</td>
<td>11.2</td>
<td>6.4 × 10$^{13}$</td>
</tr>
<tr>
<td>C$^{4+}$</td>
<td>64.5</td>
<td>4.3 × 10$^{15}$</td>
</tr>
<tr>
<td>N$^{5+}$</td>
<td>97.9</td>
<td>1.5 × 10$^{16}$</td>
</tr>
<tr>
<td>O$^{6+}$</td>
<td>138.1</td>
<td>4.0 × 10$^{16}$</td>
</tr>
<tr>
<td>Ne$^+$</td>
<td>21.6</td>
<td>8.6 × 10$^{14}$</td>
</tr>
<tr>
<td>Ne$^{7+}$</td>
<td>207.3</td>
<td>1.5 × 10$^{17}$</td>
</tr>
<tr>
<td>Ar$^{8+}$</td>
<td>143.5</td>
<td>2.6 × 10$^{16}$</td>
</tr>
<tr>
<td>Xe$^+$</td>
<td>12.13</td>
<td>8.6 × 10$^{13}$</td>
</tr>
<tr>
<td>Xe$^{8+}$</td>
<td>105.9</td>
<td>7.8 × 10$^{15}$</td>
</tr>
</tbody>
</table>
Experimental appearance intensities

Figure 11. Comparison between the experimental ionization threshold intensities obtained in linear polarization and those predicted by the barrier-suppression model (full curve) versus $E_i/\sqrt{Z}$, where $E_i$ is the ionization potential and $Z$ the ionic charge state. All intensities are peak values.

Tunnelling ionization rate

- Keldysh formula for H-like ions (stripped down to the last 1s electron):

\[
\alpha_i = 4\omega_a \left( \frac{E_i}{E_h} \right)^{5/2} \frac{E_a}{E_L(t)} \exp \left[ -\frac{2}{3} \left( \frac{E_i}{E_h} \right)^{3/2} \frac{E_a}{E_L(t)} \right], \quad (12)
\]

where \( E_i \) and \( E_h \) are the ionization potentials of the atom and hydrogen respectively, \( E_a \) is the atomic electric field, \( E_L \) is the instantaneous laser field, and

\[
\omega_a = \frac{me^4}{\hbar^3} = 4.16 \times 10^{16} \, \text{s}^{-1} \quad (13)
\]

is the atomic frequency.

- Ammosov generalization (1986): more complex many-electron atoms & ions
Experimental ionization rates

**FIG. 1.** Approximate number of argon ions detected as a function of peak laser intensity. Similar graphs have been constructed for He, Ne, Kr, and Xe.

Ionization-induced defocussing

- Refractive index of plasma created after ionization given by:

\[
\eta(r, t) = \left(1 - \frac{n_e(r, t)}{n_c}\right)^{\frac{1}{2}},
\]

where \(n_e(r, t)\) is the local electron density and \(n_c\) the critical density for the laser, related to its frequency \(\omega_L\) by:

\[
\omega_L^2 = 4\pi e^2 n_c/m
\]

- More electrons at beam center \(\Rightarrow \eta(r)\) has \textit{minimum} at centre
- Defocussing lens for rest of beam.
- High gas pressure leads to \textit{deflection} of beam before it reaches nominal focus.
Ionization-induced defocussing: ray equation

- Trajectory of light ray $\mathbf{x}(t)$ in a refractive medium obeys the *ray equation* (Born & Wolf):

$$
\frac{d}{ds} \left( \eta(x) \frac{d\mathbf{x}}{ds} \right) = \nabla \eta(x),
$$

(15)

where $ds$ is an element of length along the ray.

- Apply *paraxial approximation*: $|\eta/\nabla \eta| \gg \lambda$, and $k_\perp \ll k_\parallel$

- Setting $\mathbf{x} = \mathbf{r} + \hat{z}z$, and taking $ds \approx dz$ then gives useful form:

$$
\frac{dr}{dz} = \frac{k_\perp}{k(z)},
$$

$$
\frac{dk_\perp}{dz} = k_0 \nabla_\perp \eta(r, z),
$$

(16)

where $k_0 = \omega_0 / c$ is now the vacuum wave vector of the laser and $k(z) = k_0 \eta(r, z)$. 
Define the divergence as $\theta = \frac{k_\perp}{k_\parallel}$, and assuming for a highly underdense plasma ($\frac{n_e}{n_c} \ll 1$), refractive index is approx.:

$$\eta(r) \simeq 1 - \frac{1}{2} \frac{n_e(r)}{n_c},$$

so

$$\frac{d\theta}{dz} \simeq -\frac{1}{2} \frac{\partial}{\partial r} \left( \frac{n_e(r)}{n_c} \right).$$

For a laser spot size $\sigma_L$, the total beam deflection scales as:

$$\theta_I \sim \frac{1}{\sigma_L} \int n_e(0) \frac{d}{dz} \frac{d}{dz},$$

$\Rightarrow$ rays bent away from regions of higher electron density
Density clamping

Gaussian beam focused in vacuum is ‘diffraction limited’:

\[ \theta_D = \frac{\sigma_L}{Z_R}, \]  

(18)

where \( Z_R = 2\pi\sigma_L^2/\lambda \) is the Rayleigh length. Find that ionization-induced refraction will dominate (\( \theta_I(z_R) > \theta_D \)) when

\[ \frac{n_e}{n_c} > \frac{\lambda}{\pi Z_R}. \]

Density \textit{clamped} at value \( O(\lambda/\pi Z_R) \), because no further focusing can occur.
Numerical propagation model

- Example: $\lambda = 1 \, \mu m$, $\tau_L = 80 \, \text{fs}$, vacuum focal spot size $\sigma_L = 4.5 \, \mu m$ and nominal peak intensity of $10^{15} \, \text{Wcm}^{-2}$.
- Initialized with a radial phase modulation corresponding to an $f/10$ lens; and enters a neutral H$_2$ gas at different pressures.

**Figure:** a) beam width; b) peak intensity; c) electron density at the pulse center.
Field-ionized electron may be sent back close to its parent ion, where it can *recombine*, emitting a single, high-frequency photon.

Cutoff energy $U_c$ – Krause (1992) given by:

$$U_c = I_p + 3.17 \ U_p,$$

(19)

where $I_p = E_{\text{ion}}$ and $U_p$ are the ionization potential of the atom and the ponderomotive potential (Eq. 7) respectively.
Recollision model I

Classical equations of motion for a linearly polarized laser $\mathbf{E} = \hat{x}E_0 \cos \omega t$:

$$
\begin{align*}
v &= v_{os} \sin \omega t + v_i, \\
x &= -\frac{v_{os}}{\omega} \cos \omega t + v_i t + x_i,
\end{align*}
$$

where

$$v_{os} \equiv \frac{eE_0}{m\omega} \tag{20}$$

is the *electron quiver velocity*, and $x_i$, $v_i$ are the electron’s position and velocity just after ionization.
Now suppose that this occurs at time $t = t_0$, and let $x(t_0) = v(t_0) = 0$: the electron is born with zero velocity close to the ion center. The orbit is then:

\[
\begin{align*}
    v(\phi) &= v_{\text{os}}(\sin \phi - \sin \phi_0), \\
    x(\phi) &= \frac{v_{\text{os}}}{\omega} \left\{ \cos \phi_0 - \cos \phi + (\phi_0 - \phi) \sin \phi_0 \right\},
\end{align*}
\]

(21)

where $\phi = \omega t$ and $\phi_0 = \omega t_0$.

Look for orbits where the electron returns to $x = 0$ (the ion center) at some later time $t_1$. 
Recollision model III

- Electron’s K.E. $U_c = \frac{1}{2}mv^2$ depends on $\phi_0$, the phase of the laser that the electron is born into.
- Max. velocity at the recrossing point $x = 0$ is at $v_m/v_{os} = \pm \sqrt{3.17/2}$
- Max. $U_c(x = 0)$ is $3.17U_p$ for $\phi_0 = 17^\circ$ and $197^\circ$