Part II

Interaction with Single Atoms

2 Interaction with Single Atoms

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:

$$\begin{aligned} E_a &= \frac{e}{a_B^2} && \text{(cgs)} \\ &\simeq 5.1 \times 10^9 \text{ Vm}^{-1}. \end{aligned}$$

This leads to the *atomic intensity*:

$$\begin{aligned} I_a &= \frac{cE_a^2}{8\pi} && \text{(cgs)} \\ &\simeq 3.51 \times 10^{16} \text{ Wcm}^{-2}. \end{aligned} \tag{4}$$

Multiphoton Ionization

Intensity drops >10^10Wcm^-2

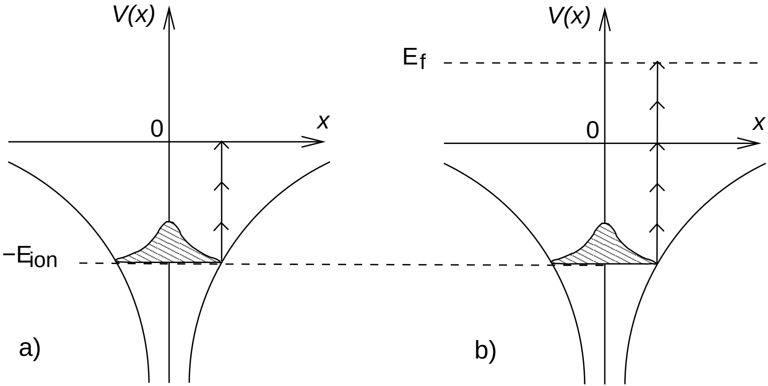


Figure: a) Multiphoton ionization (MPI): Electron with binding energy E_{ion} simultaneously absorbs n photons with energy $\hbar\omega$ and escapes from atom with minimal kinetic energy. b) Above-threshold ionization (ATI): electron absorbs *more* photons than necessary for ionization, acquiring momentum.

Above-threshold ionization (ATI)

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

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

where n is the number of photons needed for multiphoton ionization; s is the excess absorbed.

Above-threshold ionization (ATI): measurements of electron spectra

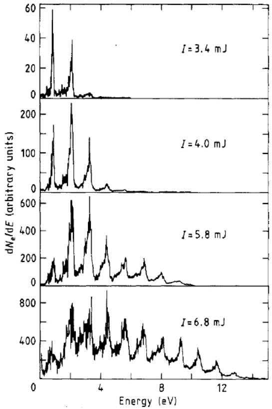


Figure 2. Electron spectra of eleven-photon MPI at 1604 nm for different pulse energies. The first peak vanishes at around 7 mJ. The maximum total count rate in these spectra was ten per laser shot.

The limit: the electron oscillation energy becomes larger than the photon energy

Source: Yergeau, Petite & Agostini, J. Phys. B (1986)

Tunneling ionization

I>I_a, when the laser field becomes strong enough to distort the Coulomb field felt by the electron

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

$$\gamma = \omega_L \sqrt{\frac{2E_{\mathrm{ion}}}{I_L}} \sim \sqrt{\frac{E_{\mathrm{ion}}}{\Phi_{\mathrm{pond}}}}. \tag{6}$$

where

$$\Phi_{\mathrm{pond}} = \frac{e^2 E_L^2}{4m\omega_L^2} \tag{7}$$

is the *ponderomotive potential* of the laser field.
$\gamma < 1 \Rightarrow$ tunneling – strong fields, long wavelengths
$\gamma > 1 \Rightarrow$ MPI

Tunnelling: barrier suppression model I

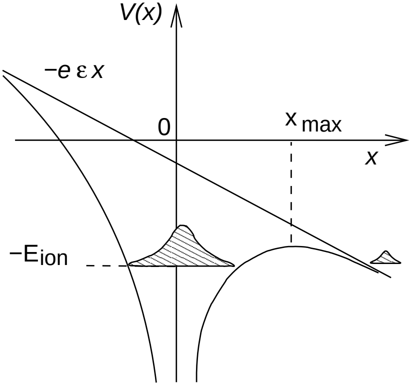


Figure: a) Schematic picture of tunneling or barrier-suppression ionization by a strong external electric field.

Tunnelling: barrier suppression model II

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

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

  $\Rightarrow$ suppressed on RHS of the atom, and for $x \gg x_{\mathrm{max}}$ is *lower* than the binding energy of the electron.

- If the barrier falls below E_{ion}, the electron will escape spontaneously
  $\Rightarrow$ *over-the-barrier* (OBT) or *barrier suppression* (BS) ionization.

Barrier suppression model III

Differentiate V(x) to determine the position of the barrier,

$$x_{\mathrm{max}} = (Ze/\varepsilon), \text{\textasciicircum}1/2 \qquad dV(x)/dx=0$$

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

$$\varepsilon_c = \frac{E_{\mathrm{ion}}^2}{4Ze^3}. \tag{8}$$

Barrier suppression model IV

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

$$I_{\rm app} = \frac{c}{8\pi}\varepsilon_c^2 = \frac{cE_{\rm ion}^4}{128\pi Z^2 e^6}, \tag{9}$$

or:

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

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

Appearance intensity: Hydrogen example

- Hydrogen: $Z = 1$

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

- 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:

$$I_{\rm app} = \frac{I_a}{256} \simeq 1.4 \times 10^{14} \text{ Wcm}^{-2}. \quad (11)$$

Appearance intensities of selected ions according to the BS ionization model – Eq. (10).

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

Experimental appearance intensities

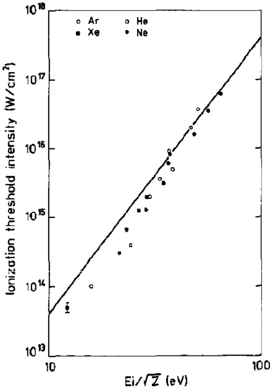


Figure 11. Comparison between the experimental ionization threshold intensities obtained in linear polarization and those predicted by the barrier-suppression model (full curve) versus $E_i/\sqrt{Z}$, where E_i is the ionization potential and Z the ionic charge state. All intensities are peak values.

Source: Auguste *et al.*, J. Phys. B (1992)

Tunnelling ionization rate

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

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

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

$$\omega_a = \frac{me^4}{\hbar^3} = 4.16 \times 10^{16}\ \mathrm{s}^{-1} \qquad (13)$$

is the atomic frequency.

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

Impact ionization (collisional ionization)

Experimental ionization rates

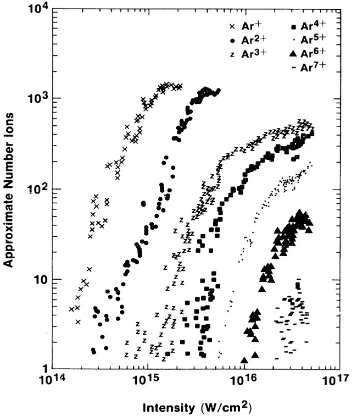


FIG. 1. Approximate number of argon ions detected as a function of peak laser intensity. Similar graphs have been constructed for He, Ne, Kr, and Xe.

Source: Auguste *et al.*, J. Phys. B (1992)

Ionization-induced defocussing

- Refractive index of plasma created after ionization given by:

$$\eta(r,t) = \left(1 - \frac{n_e(r,t)}{n_c}\right)^{\frac{1}{2}}, \tag{14}$$

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

$$\omega_L^2 = 4\pi e^2 n_c/m$$

- More electrons at beam center $\Rightarrow \eta(r)$ has *minimum* at centre
- Defocusing lens for rest of beam.
- High gas pressure leads to *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):

$$\frac{d}{ds}\left(\eta(\mathbf{x})\frac{d\mathbf{x}}{ds}\right) = \nabla\eta(\mathbf{x}), \tag{15}$$

  where ds is an element of length along the ray.
- Apply *paraxial approximation*: $\mid \eta/\nabla\eta \mid \gg \lambda$, and $k_\perp \ll k_\parallel$ (paraxial approximation)
- Setting $\mathbf{x} = \mathbf{r} + \hat{z}z$, and taking $ds \approx dz$ then gives useful form:

$$\begin{aligned} \frac{d\mathbf{r}}{dz} &= \frac{\mathbf{k}_\perp}{k(z)}, \\ \frac{d\mathbf{k}_\perp}{dz} &= k_0\nabla_\perp\eta(r,z), \end{aligned} \tag{16}$$

  where $k_0 = \omega_0/c$ is now the vacuum wave vector of the laser and $k(z) = k_0\eta(r,z)$.

Beam divergence

Define the divergence as $\theta = k_\perp / k_\parallel$, and assuming for a highly underdense plasma ($n_e/n_c \ll 1$), 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 σ_L, the total beam deflection scales as:

$$\theta_I \sim \frac{1}{\sigma_L}\int \frac{n_e(0)}{n_c} dz, \tag{17}$$

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

Density clamping

Gaussian beam focused in vacuum is 'diffraction limited':

$$\theta_D = \frac{\sigma_L}{Z_R}, \tag{18}$$

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 *clamped* at value $O(\lambda/\pi Z_R)$, because no further focusing can occur.

Numerical propagation model

- Example: $\lambda = 1\ \mu$m $\tau_L = 80$ fs, vacuum focal spot size $\sigma_L = 4.5\ \ \mu$m and nominal peak intensity of 10^{15} Wcm^{-2}.
- Initialized with a radial phase modulation corresponding to an $f/10$ lens; and enters a neutral H_2 gas at different pressures.

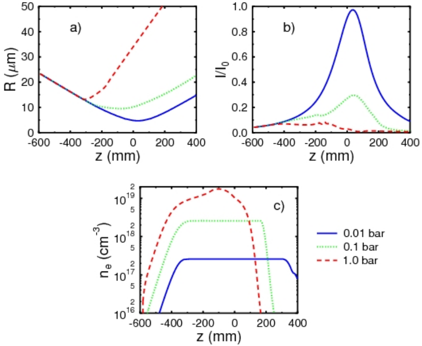
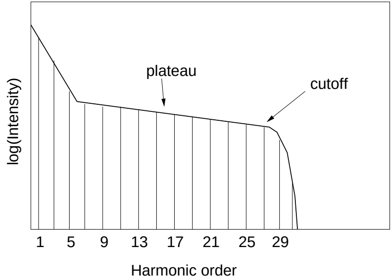


Figure: a) beam width; b) peak intensity; c) electron density at the pulse center

High-harmonic generation by atoms

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:

$$U_c = I_p + 3.17\ U_p, \tag{19}$$

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 $\mathbf{E} = \hat{x} E_0 \cos \omega t$:

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

where

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

is the *electron quiver velocity*, and x_i, v_i are the electron's position and velocity just after ionization.

Recollision model II

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:

$$\begin{aligned} v(\phi) &= v_{\rm os}(\sin\phi - \sin\phi_0), \\ x(\phi) &= \frac{v_{\rm os}}{\omega}\{\cos\phi_0 - \cos\phi + (\phi_0 - \phi)\sin\phi_0)\}, \end{aligned} \qquad (21)$$

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 ϕ_0, the phase of the laser that the electron is born into.
- Max. velocity at the recrossing point $x = 0$ is at $v_m/v_{\rm os} = \pm\sqrt{3.17/2}$
- Max. $U_c(x = 0)$ is $3.17U_p$ for $\phi_0 = 17^o$ and 197^o