Extreme NLO: high-order harmonic generation (HHG)

Free-electron response:
no damping, no resonance

$$\ddot{x} = -e/m\ E_0 \cos \omega t$$
$$\dot{x} = -(e/m\omega) E_0 \sin \omega t + v_0 = v_{osc}(t) + v_0$$
$$\rightarrow x(t) = \frac{e}{m\omega^2} E_0 \cos \omega t + \underbrace{v_0 t + x_0}_{\text{free motion w/o field}}$$

Ponderomotive energy (wiggle, quiver energy)

$$U_p = \langle \tfrac{1}{2} m v_{osc}^2 \rangle = \frac{1}{4}\frac{e^2}{m\omega^2} E_0^2 \quad \propto \text{intensity}$$

in Gaussian units, $I = \dfrac{c E_0^2}{8\pi} \longrightarrow U_p = \dfrac{1}{4}\dfrac{e^2}{m\omega^2}\cdot\dfrac{8\pi I}{c}$

$U_p = \dfrac{2\pi e^2}{m c \omega^2} I$ w/ $\omega = \dfrac{2\pi c}{\lambda}$, $U_p = \dfrac{2\pi e^2 \lambda^2}{m c^3 4\pi^2}$

recall $\dfrac{e^2}{mc^2} = r_e$ classical electron radius $= 2.8 \times 10^{-15}$ m

$U_p = \dfrac{r_e \lambda^2}{2\pi c} I$ works in any units

calc. U_p in eV, λ in μm, I in W/cm^2

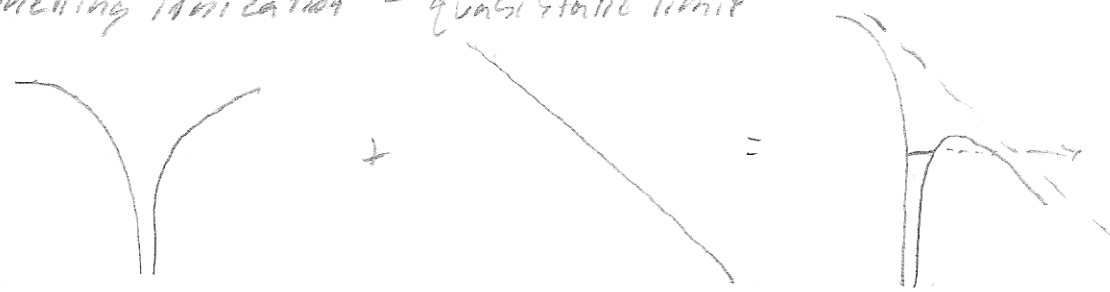
$$U_p = 9.4 \times 10^{-14} I \lambda^2$$

so at $\lambda = 1\mu m$ $I \sim 10^{14}$ W/cm^2 $U_p \sim 10$ eV

High-order harmonic generation.

"Simple-man" model - semiclassical.

1) tunnelling ionization - quasistatic limit



electron tunnels through barrier.

"ADK" ionization rate avg rate $W \propto I^{8}$

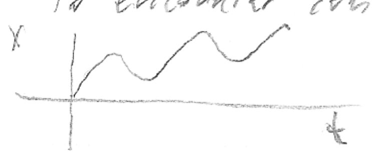
(multiphoton ionization)

$W \propto I^{n_p}$

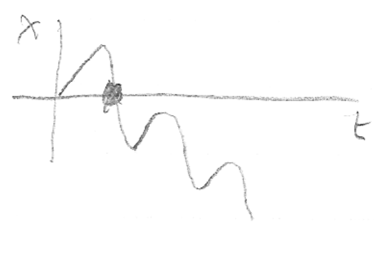
in tunnelling ionization electrons are released in bursts. probability of ionization varies strongly with time within cycle.

2) electron is released into field at $x=0$, $v_x=0$

- oscillates in field
- depending on when in cycle it is released, it may return to encounter ion.



or

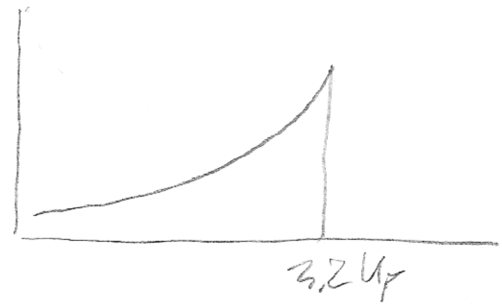


If electron reattaches, photon is given off w/
$\hbar\omega = \frac{1}{2} m_e v^2 + I_p$

range of collision energies
- maximum cutoff $\hbar\omega = 3.2\,U_p + I_p$

combine P(ionization) with KE at collision

→ P(KE)



3) recombination of electron w/ ion.
- calc. σ
- depends on velocity and amt of electron wavepacket spread during transit.

Note that ellipticity in polarization → electron can miss ion.

Nonlinear wave propagation.

- go from microscopic to macroscopic.

As before, we use Maxwell eqns to get wave eqn.

$$\underbrace{\nabla\times\nabla\times\vec{E}}_{\nabla(\nabla\cdot\vec{E})-\nabla^2\vec{E}} + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\vec{E} = -\frac{4\pi}{c}\frac{\partial^2\vec{P}}{\partial t^2}$$

$$\nabla\cdot\vec{D} = \nabla\cdot(\epsilon\vec{E}) = 0$$

We'll assume ϵ ~ spatially constant, even though there is nonlinearity.
Next separate $\vec{P} = \vec{P}^{(1)} + \vec{P}^{NL}$, bring linear part over.

$$\nabla^2\vec{E} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\underbrace{(\vec{E}+4\pi\vec{P}^{(1)})}_{=\vec{D}^{(1)} = \epsilon^{(1)}\vec{E}} = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\vec{P}^{NL}$$

assume isotropic

assume this is t-indep.

$$\nabla^2\vec{E} - \frac{\epsilon^{(1)}}{c^2}\frac{\partial^2\vec{E}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2\vec{P}^{NL}}{\partial t^2}$$

solutions:

linear eqn has wave solutions.

we will make the assumption that there will be several diff't waves at distinct frequencies.

e.g. $$\vec{E}_n(\vec{r},t) = E_n(\vec{r})\, e^{-i\omega_n t} + c.c.$$

$$\vec{E}(\vec{r},t) = \sum_n \vec{E}_n(\vec{r},t) \qquad n \geq 0$$

similar for $\vec{P}_n^{NL}$, and $\vec{D}_n^{(1)} = \epsilon_n^{(1)}\vec{E}_n^{(1)}$

$$\rightarrow -\nabla^2 \vec{E}_n(r) - \frac{\omega_n^2}{c^2}\varepsilon^{(1)}(\omega_n)\vec{E}_n(\vec{r}) = \frac{4\pi\omega_n^2}{c^2}\vec{P}_n^{NL}(\vec{r})$$

What if material is birefringent?

$\vec{D}_n^{(1)} = \overleftrightarrow{\varepsilon}^{(1)}(\omega_n)\cdot\vec{E}_n^{(1)}$ since ε is a tensor

$\therefore$ change $\varepsilon \rightarrow \overleftrightarrow{\varepsilon}$ and dot it with $\vec{E}$

Notes: we have several coupled equations, each at diff't ω_n

- coupling is through $\vec{P}^{NL}$

e.g. $P^{(2)} = \chi^{(2)} E_m E_n^*$

each of these equations is for 3 vector components.

Application: SFG (sum freq. gen.)

- CW
- 2 inputs at ω_1, ω_2
- outputs at $\omega_3 = \omega_1 + \omega_2$
- plane waves prop in z direction

- ignore polarization effects for now

$$E_n(z,t) = A_n e^{i(k_n z - \omega_n t)} + c.c.$$

with no nonlinearity, no coupling of separate, linear w.e.

$\therefore$ all A_n's are constant

with nonlinearity, RHS $\rightarrow$ P^{NL} terms, e.g.

$$P_3^{(2)} = 4 d_{eff} E_1 E_2 = 4 d_{eff} A_1 A_2 e^{i(k_1+k_2)z}$$

recall $\chi^{(2)} \equiv 2 d_{eff}$

second 2x from $\omega_1+\omega_2$ or $\omega_2+\omega_1$

to put this into the NLWE, we have to account for $A_n(z)$, no dependence on x, y

$$-\frac{d^2}{dz^2}\vec{E}_3(z) - \frac{\omega_3^2}{c^2}\varepsilon_3\vec{E}_3(z) = \frac{16\pi\omega_3^2}{c^2} d_{eff}\vec{E}_1(z)\vec{E}_2(z)$$

Non-depleted pump approximation:

- input beams have powers $\propto A_1^2, A_2^2$
- anticipate growth of A_3 from zero, no initial change (much) of pump beams. A_1, A_2 const.

$\therefore$ equations are decoupled.

$$\rightarrow -\left(\frac{d^2}{dz^2}A_3\right)e^{ik_3z} - 2ik_3\left(\frac{d}{dz}A_3\right)e^{ik_3z} + \cancel{k_3^2 A_3 e^{ik_3z}} - \cancel{\frac{\epsilon_3\omega_3^2}{c^2}A_3 e^{ik_3z}}$$

$$= \frac{16\pi\omega_3^2}{c^2} d_{eff} A_1 A_2 e^{i(k_1+k_2)z}$$

cancellation from $k_3^2 = \epsilon_3\omega_3^2/c^2$

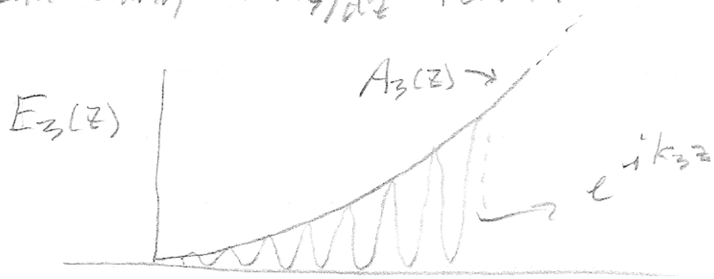
notice where e^{ik_3z} is: only on LHS

divide it out:

$$-\frac{d^2A_3}{dz^2} - 2ik_3\frac{dA_3}{dz} = \frac{16\pi\omega_3^2}{c^2} d_{eff} A_1 A_2 e^{i\Delta kz}$$

where $\Delta k \equiv k_1 + k_2 - k_3$ = phase mismatch.

Can we drop d^2A_3/dz^2 term?



let scale length for growth of A_3 be l

$\rightarrow \frac{dA_3}{dz}$ scales as $\frac{1}{l}A_3$ e.g. $A_3 = A_{30}e^{z/l}$

compare terms:

$\frac{1}{l^2}A_3 \; : \; \frac{2\pi}{\lambda l}A_3$ if $\lambda l \ll l^2$ or $\lambda \ll l$

we can drop d^2A_3/dz^2 $\rightarrow$ slowly varying envelope approx.

$$\rightarrow \frac{dA_3}{dz} = 8\pi i d_{eff} \frac{\omega_3^2}{k_3 c^2} A_1 A_2 e^{i\Delta k z}$$

$$= \frac{2\pi i \omega_3}{n_3 c} p_3 e^{i\Delta k z} \qquad p_3 \equiv 4 d_{eff} A_1 A_2$$

Account for pump depletion:

- let $A_1(z)$, $A_2(z)$
- eqn for dA_3/dz is unchanged.
- now do same for others - main work is getting RHS = driving term

$$P_1^{(2)} = 4 d_{eff} E_2^* E_3 \qquad \omega_1 = \omega_3 - \omega_2$$

conjugate E_2

$$\rightarrow \frac{dA_1}{dz} = 8\pi i d_{eff} \frac{\omega_1^2}{k_1 c^2} A_2^* A_3 \underbrace{e^{i(k_3-k_2-k_1)}}_{e^{-i\Delta k z}}$$

$$\frac{dA_2}{dz} = 8\pi i d_{eff} \frac{\omega_2^2}{k_2 c^2} A_1^* A_3 e^{-i\Delta k z}$$

here, 3 eqns 3 unknowns (actually 6 eqns, 6 unk. since A_n's are complex)

We'll come back to solution later.

Deal with simpler cases first:

1) non depletion (1 eqn) $\Delta k = 0$

2) non depl. $\Delta k \neq 0$

3) depletion (all 3) $\Delta k = 0$

4) " $\Delta k \neq 0$

5) add dispersion...

Effect of phase mismatch

- assume $\Delta k = 0 \rightarrow A_3(z) \propto z \quad I_3(z) \propto z^2$
- for $\Delta k \neq 0$:

$$\frac{dA_3}{dz} = s\, e^{i\Delta k z}$$

integrate directly:

$$A_3(z) = s \int_0^L e^{i\Delta k z}\, dz = s \left(\frac{e^{i\Delta k L} - 1}{i\Delta k} \right)$$

$$= s\, e^{i\Delta k L/2} \frac{\left(e^{i\Delta k L/2} - e^{-i\Delta k L/2} \right)}{2i\Delta k L/2}$$

$$= s\, e^{i\Delta k L/2} \frac{\sin(\Delta k L/2)}{\Delta k L/2} = s\, e^{i\Delta k L/2} \operatorname{sinc}\left(\frac{\Delta k L}{2}\right)$$

Intensity is $\propto |A_3|^2$

$$I_i = \frac{n_i c}{8\pi} |\mathcal{E}|^2 \qquad E = \mathcal{E} \cos(kz - \omega t)$$

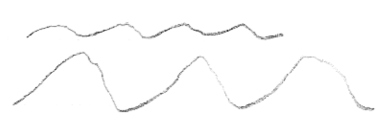
$$= A e^{i(kz - \omega t)} + c.c.$$

$$= \frac{n_i c}{2\pi} |A|^2 \qquad A = \mathcal{E}/2$$

$$\rightarrow I_3(L) = \frac{512\pi^5 d_{eff}^2 I_1 I_2}{n_1 n_2 n_3 \lambda_3^2 c} L^2 \operatorname{sinc}^2(\Delta k L/2)$$

notes:
- $I_3 \propto d_{eff}^2$ magn. of deff is impt.
- I_3 is linear in I_1, I_2
- for $\Delta k \neq 0$, $I_3 \propto \frac{\sin^2(\Delta k L/2)}{(\Delta k)^2}$

power osc. along length.
- as $\Delta k \rightarrow 0$ period $\uparrow$
ampl. $\uparrow$



- $\Delta k \neq 0$ fixed L vary Δk

$I_3 \propto \operatorname{sinc}(\Delta k L/2)$