

last time: production of HHG by evaluating

$$x(t) = \langle \psi(t) | x | \psi(t) \rangle$$

different HHG harmonics are phase-locked & form
at attosecond pulse

What does the as pulse look like?

→ note, short wavelength, so dipole approx. likely to

fail

$$\vec{E}(t, \vec{r}) = \frac{1}{2\pi} \int d\omega \vec{E}(\omega, \vec{r}) e^{-i\omega t}$$

↑
amplitude of each harmonic
& phase

Each harmonic obey the Maxwell's eqn.:

$$\nabla^2 \vec{E}(\omega, \vec{r}) + \frac{\omega^2}{c^2} \vec{E}(\omega, \vec{r}) = -\frac{1}{\epsilon_0} \frac{\omega^2}{c^2} \vec{P} - \frac{1}{\epsilon_0} \nabla(\nabla \cdot \vec{P}) \quad (1)$$

where $\vec{P}(\omega, \vec{r})$ is the polarization term:

$$\vec{P}(\omega, \vec{r}) = \epsilon_0 \chi^{(1)}(\omega, \vec{r}) \vec{E}(\omega, \vec{r})$$

↗ linear susceptibility (may vary in space)

$$\vec{P}(t, \vec{r}) = \frac{1}{2\pi} \int d\omega \vec{P}(\omega, \vec{r}) e^{-i\omega t}$$

⇒ Note, $\chi^{(1)}(\omega, \vec{r})$ may have a more complicated
evolution in time at peak intensities $> 10^{16} \frac{W}{cm^2}$ (such as in
XFEL)

(2)

inserting $\vec{E}(\omega, \vec{r}) \propto e^{i\phi(\omega, x)} \hat{e}$ into Eqn. (1), we get

$$i \frac{\partial^2 \phi}{\partial x^2} - \left(\frac{\partial \phi}{\partial x} \right)^2 + \left(\frac{\omega}{c} \right)^2 = - \frac{\omega^2}{c^2} \chi^{(1)}$$

For slow variation of material optical properties:

$$\left| \frac{\partial^2 \phi}{\partial x^2} \right| \ll \left| \frac{\partial \phi}{\partial x} \right|^2 \Rightarrow \frac{\partial \phi}{\partial x} \approx \frac{\omega}{c} \sqrt{1 + \chi^{(1)}}$$

$$\phi(\omega, x) = \frac{\omega}{c} \int_{-\infty}^x dx' \sqrt{1 + \chi^{(1)}(x')} \approx \frac{\omega}{c} \int_{-\infty}^x dx' \underbrace{n(x')}_{\substack{\text{local refractive} \\ \text{index}}} \quad \begin{matrix} \text{assumes material that} \\ \text{is not dispersive} \end{matrix}$$

asymptotic phase shift due to propagation

through a medium: $\delta(\omega) = \lim_{x \rightarrow 0} \frac{\omega}{c} \int_{-\infty}^x dx' [n(x') - 1]$

delay in propagation,
as compared to vacuum: $\tau = \delta/\omega$

$$\underbrace{E_{\text{free}}(t-\tau, x)}_{\text{shifted wave-packet}} = \frac{1}{2\pi} \int d\omega E_{\text{free}}(\omega, x) e^{-i\omega t} \underbrace{e^{i\omega\tau}}_{\text{phase-shift}}$$

absorbing medium: $e^{i\delta} = \underbrace{e^{iR(t)}}_{\text{phase-shift}} \underbrace{e^{-\text{Im}\delta}}_{\text{damping term}}$

Suppose an a.c. pulse passing through material of length, L , with constant refractive index, n

$$\delta = \frac{\omega}{c} (n-1)L \Rightarrow \tau = \frac{(n-1)L}{c} \Rightarrow \text{linear scaling with both } L \text{ \& } n$$

For dispersive material $\Rightarrow n(\omega, x) \Rightarrow$ refractive index is frequency-dependent

$$\vec{E}(t, x) = \frac{1}{2\pi} \int d\omega \underbrace{E_{free}(\omega, x)}_{\text{free propagation}} e^{-i\omega t} e^{i\delta(\omega)}$$

\Rightarrow wave-packet is deformed w.r.t. free propagation

Expanding around the central freq of the pulse:

$$\delta(\omega) = \delta(\omega_0) + \sum_{n=1}^{\infty} \frac{1}{n!} \left. \frac{\partial^n \delta}{\partial \omega^n} \right|_{\omega_0} (\omega - \omega_0)^n$$

δ
over-all phase
of the pulse

$n=1$ term determines the delay of the pulse

$\tau_{GD} = \frac{\partial \delta}{\partial \omega} \Rightarrow$ delay in the propagation of a harmonic

frequency ω

Similar to the Eisenbud-Wigner time delay

derived for mono-chromatic wave-packets:

$\Rightarrow \tau_{EW}$ used to calculate delays for single photon ionization

\Rightarrow Gives a shift (delay) of the peak of the wave-packet due to an interaction with a potential

Wigner, Physical Review, V. 98 (1), 1955

Eisenbud - Wigner, τ_{EW} , time delay derivation
(see below)

First, Let's take XUV wave-packet traveling through plasma of length L :

$$n_p(\omega) = \sqrt{1 - \left(\frac{\omega_p}{\omega}\right)^2} \quad \text{for } \omega \gg \omega_p$$

$$n_p(\omega) \approx 1 - \frac{1}{2} \left(\frac{\omega_p}{\omega}\right)^2 \quad \omega_p = \sqrt{\frac{Ne^2}{\epsilon_0 m}} \quad \text{is plasma frequency}$$

N - concentration of free electrons in plasma

$$S_p(\omega) = \frac{\omega}{c} [n_p(\omega) - 1] L \approx -\frac{1}{2} \frac{\omega_p^2}{c} \frac{L}{\omega}$$

\Rightarrow negative asymptotic phase seems to imply superluminal propagation, because refractive index $n_p(\omega) < 1$

$$\Rightarrow \text{However: } \tau_{GD} = \frac{\partial S_p}{\partial \omega} \approx \frac{1}{2} \frac{\omega_p^2}{c} \frac{L}{\omega^2} \quad \Rightarrow$$

light pulse is delayed by plasma as travels slower than c .

For phase-matching, plasma density can be altered by changing the intensity of the laser (changes N)

spherical potential:

Incident wavepacket:

$$\psi_{inc} = r^{-1} \left(e^{-i(k+k')r - i(v+v')t} + e^{-i(k-k')r - i(v-v')t} \right)$$

$k' \ll k$; $v' \ll v \Rightarrow$ two spherical waves of nearly identical momentum, $k \pm k'$ & energy $v \pm v'$

peak of the wave-packet: $2k'r + 2v't = 0$

Group velocity: $\frac{dv}{dk} = \frac{v'}{k'}$

Outgoing components incur phase-shifts $\eta + \eta'$ and $\eta - \eta'$, respectively.

$$\psi_{out} = r^{-1} \left(e^{i(k+k')r - i(v+v')t + 2i(\eta + \eta')} + e^{i(k-k')r - i(v-v')t + 2i(\eta - \eta')} \right)$$

peak of the wave-packet: $2k'r + 2v't + 4\eta' = 0$

$$r = -2\eta'/k' + (v'/k')t = \underbrace{-2 \frac{d\eta}{dk}}_{r_{free}} + \underbrace{\frac{dv}{dk}}_{v} t$$

\Rightarrow The outgoing wave is retarded by $-2 \frac{d\eta}{dk}$

additional time delay: $2\tau_{EW} = \frac{\Delta r}{v} = \underbrace{\frac{2d\eta}{dk}}_{\Delta r} \cdot \underbrace{\frac{1}{\hbar k/m}}_v = 2\hbar \frac{\partial \theta}{\partial E}$
 incurred for incoming & outgoing wave-packet

$$\tau_{EW} = \hbar \frac{\partial \theta}{\partial E}$$