Lecture 5 15.1.13

Last time: HHG with a semiclassical model

1st step: Quantum ⇒ tunneling
2nd step: classical ⇒ propagation in laser field
3rd step: Quantum ⇒ recombination

showed that dominant contribution to HHG comes from electrons that: i) Return to the nucleus with zero velocity
ii) Appear in the continuum
iii) Have an appropriate KE to produce a given harmonic @ the time of return: $KE = (2n+1)\hbar - I_p$

Fully Quantum and exact theory of HG has been formulated (early '90s)
i) as a solution to TDSE
ii) or, equivalently in terms of solutions of time-indept. Floquet equations

Fully quantum approach: TDSE not so easy to solve, esp. for elliptical polarizations. Gives little physical insight

On the other hand: semiclassical ("simple man's model")
⇒ good physical insight
⇒ does not account for quantum diffusion/interference

Today: present an approximate analytical solution to the Schrodinger eqn. as an intermediary btwn the semiclassical & the exact quantum treatment

First: a simple example to see how radiation is produced when the electron transitions btwn different quantum states ⇒ Need dipole moment $\langle x \rangle$

Wavefunction of electron in a state of quantum number n & Energy E_n: $\Psi_n = \varphi_n e^{-iE_n t}$

$\Psi_n^* = \underbrace{\varphi_n^*}_{\text{time-indep. part}} e^{+iE_n t}$

Expectation value $\langle x \rangle$ of position of such an electron: $\underset{\text{x-component of dipole moment}}{\langle x \rangle} = \int_{-\infty}^{\infty} x \, \Psi_n^* \Psi_n \, dx$

$$\langle x \rangle = \int_{-\infty}^{\infty} x \, \varphi_n \varphi_n^* \, dx \Rightarrow \text{constant in time!}$$

⇒ electron does not oscillate & no radiation occurs

⇒ an electron in a specific quantum state produces no radiation.

Now, suppose the electron wave-function is a super-position of 2 states: $\Psi = a\Psi_n + b\Psi_m$

$|a|^2$ ⇒ probability the electron is in state a

$|b|^2$ ⇒ " " " " " " b

$$\langle x \rangle = \int_{-\infty}^{\infty} x \, (a^* \Psi_n^* + b^* \Psi_m^*)(a\Psi_n + b\Psi_m) \, dx$$

$$= \int_{-\infty}^{\infty} x \, (\underbrace{a^2 \Psi_n^* \Psi_n}_{\text{constant}} + \underbrace{b^* a \Psi_m^* \Psi_n + a^* b \Psi_n^* \Psi_m}_{\text{time-dep. terms}} + \underbrace{b^2 \Psi_m^* \Psi_m}_{\text{constant}}) \, dx$$

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time-dep. part: $\underbrace{b^* a \int_{-\infty}^{\infty} x \, \psi_m^* \psi_n \, dx}_{\text{time-indep}} \cdot e^{it(E_m - E_n)}$

$$+ a^* b \int_{-\infty}^{\infty} x \, \psi_n^* \psi_m \, dx \cdot e^{-it(E_m - E_n)}$$

Oscillation @ frequency: $\omega = \frac{E_m - E_n}{\hbar}$ ⇒ frequency of the emitted/absorbed photon in a $m \leftrightarrows n$ transition!

atomic dipole: $d_x = \int_{-\infty}^{\infty} x \, \Psi_m^* \Psi_n \, dx = \langle \Psi_m | x | \Psi_n \rangle$
for $n \to m$ transition

transitions where $d_x = 0$ are forbidden!

Hence transitions where $\int_{-\infty}^{\infty} x \, \Psi_n^* \Psi_m \, dx \neq 0$ are allowed

Ψ_n & Ψ_m can be calculated exactly for a hydrogenic atom, where the wavefunction is dictated by the quantum #'s n, l, m_l

n: energy; l: orbital ang. momentum; m_l: magnetic quantum #

condition $\int_{-\infty}^{\infty} \vec{r} \, \psi_{n,l,m_l}^* \psi_{n',l',m_l'} \, dV \neq 0$

determines allowed transitions ⇒ "dipole selection rules"

Selection rules: $\Delta l = \pm 1$; $\Delta m_l = 0, \pm 1$

Note: these rules result in an emission/absorption of a single photon of energy $|E_m - E_n|$, which carries off angular momentum $\pm\hbar$ (equal to the difference in angular momentum, l, between m & n states

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⇒ so to get radiation frequencies in HHG, we have to calculate the x-component of time-dep. dipole moment:

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

why only $x(t)$? Because electron osc. involved in HHG is along the $\hat{x}$-axis, since $\vec{E} = E\cos(\omega t)\hat{x}$

⇒ Approximate solution for $|\Psi(t)\rangle$ already obtained from SFA.

⇒ Note: $|\Psi(t)\rangle$ is made up of a continuum of states $|v\rangle$ + ground state $|0\rangle$:

$$|\Psi(t)\rangle = e^{iI_p t} a(t)|0\rangle + \int \underbrace{d^3v}_{\text{integration over a continuum of velocity states}} a_v(t)|v\rangle$$

i) In case of no depletion: $a(t) \simeq 1$

ii) Note: only ground state & continuum states contribute to $|\Psi(t)\rangle$ ⇒ no intermediate resonances.

iii) Neglect Coulomb field in the continuum (key SFA assumption)

where $a_v(t) = \langle v | \Psi(t) \rangle$ was derived previously using SFA for the following TDSE:

$$i\frac{\partial}{\partial t}|\Psi\rangle = \Big[\underbrace{-\frac{1}{2}\nabla^2 + V(x)}_{\hat{H}_0} - \underbrace{E\cos(t)\,x}_{\text{length gauge: } \hat{V}_L}\Big]|\Psi\rangle$$

$\hat{H}_0$ (field free Hamiltonian): $-\frac{1}{2}\nabla^2 + V(x)$
atomic potential

$\hat{V}_L$ (interaction with the laser field): $\hat{V}_L = \underbrace{E\cos(t)}_{\text{field strength}} \cdot \underbrace{\hat{x}}_{\text{dipole term}}$

Full solution of TDSE:

$$|\Psi(t)\rangle = i\int_0^t dt' \, e^{-i\int_{t'}^{t} \hat{H}(t'')\,dt''} \times \hat{V}_L(t') \, e^{\overbrace{-\int_0^{t'} \hat{H}_0(t'')\,dt''}^{+iI_p t'}} \times |0\rangle + e^{+iI_p t} |0\rangle$$

SFA: set $\hat{H} \approx \hat{H}_0 + \hat{H}_{Laser} \approx \hat{H}_{Laser} = \frac{1}{2} m \hat{v}^2 \Rightarrow$ KE of a free electron

$$a_v(t) = \langle v | \Psi(t)\rangle \approx i\int_{t_0}^{t} dt' \, e^{-i\int_{t'}^{t} \frac{(v'')^2}{2} dt'' + iI_p t'} \cdot \underbrace{\langle v(t') | x | 0\rangle}_{\text{dipole matrix element!}} \times E\cos(t')$$

From conservation of canonical momentum: $\vec{p} = v(t) + \vec{A}(t) = v(t'') + \vec{A}(t'')$

$$\frac{(v'')^2}{2} = \frac{\left[v(t) + A(t) - A(t'')\right]^2}{2}$$

Dipole moment: $x(t) = \langle \Psi(t) | x | \Psi(t)\rangle$

$$x(t) = \left(\langle 0 | e^{-iI_p t} + \int d^3v' \, a_{v'}^*(t) \langle v'|\right)(x)\left(e^{iI_p t}|0\rangle + \int d^3v \, a_v(t)|v\rangle\right)$$

(6)

again, only cross-terms $\langle 0|V\rangle$ & $\langle V|0\rangle$ contribute to the radiation

Note: continuum - continuum contributions: $\langle v'|v\rangle$ are neglected ⇒ one reason is that ionization probability is low (no saturation), hence $a_{v'}^* a_v \ll a_v$ ⇒ these terms only introduce higher order corrections

⇒ corresponds with the intuition that the main contribution to HHG come from recombination with the ground state, $|0\rangle$

$$x(t) \approx \int dv^3 \, a_v(t) \cdot \underbrace{\langle 0|x|v\rangle}_{d_x^*(t)} + \int dv^3 \, a_v^*(t) \underbrace{\langle v|x|0\rangle}_{d_x(t)}$$

$$= 2 \cdot \mathrm{Re}\left(\int dv^3 \, a_v(t) \cdot d_x^*(t) \right)$$

substituting for $a_v(t)$:

$$x(t) = 2\mathrm{Re}\left\{ i \int_0^t dt' \int d^3p \cdot E\cos(t') \underbrace{\langle v(t')|x|0\rangle}_{d_x(t')} \times d_x^*(t) \times e^{-iS(\vec{p},t,t')} \right.$$

where $S(\vec{p},t,t') = \int_{t'}^{t} dt'' \frac{[\vec{p} - \vec{A}(t'')]^2}{2} - I_p t'$

Note: t' corresponds to ionization time

As we learned in the last lecture, there is 1 particular trajectory for each ionization time, t' that contributes

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⇒ can use the saddle-point method to evaluate the integral $\int d^3p \ldots$ $S = \int p\,dq$

SPM: $\nabla_{\vec{p}} S(\vec{p}, t, t') = 0$

Note $\nabla_p S(\vec{p}, t, \vec{t}) = \vec{x}(t) - x(t')$ ⇒ hence

$\nabla_{\vec{p}} S(\vec{p}, t, t')$ corresponds to requiring that electron that appears in the continuum @ time t', returns to the same pt @ time t.

$$S(\vec{p}, t, t') = \frac{1}{2}\int_{t'}^{t} dt'' \{ (p_x - \underbrace{E\sin(t'')}_{A_x})^2 + p_y^2 + p_z^2 \} - I_p t'$$

$$\frac{\partial S}{\partial p_y} = \frac{\partial S}{\partial p_z} = 0 \Rightarrow p_y = p_z = 0$$ ⇒ justifies the semi-classical assumption of zero transverse velocity

$$\frac{\partial S}{\partial p_x} = \int_{t'}^{t} dt'' (p_x - E\sin(t'')) = p_x(t - t') - E\cos(t'')\Big|_{t'}^{t}$$

$$p_{st}(t, t') = \frac{E(\cos(t) - \cos(t'))}{t - t'}$$ ⇒ corresponds to classical trajectory

↗ trajectory that contributes to the harmonic emission for ionization & recombination times given by t' & t, respectively.

$t - t'$ ⇒ excursion time

Long trajectories, smaller p_{st}, ionization near the peak