SPECIFICS OF INITIAL AND FINAL STATES IN HIGH HARMONIC GENERATION

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OUTLOOK

- Ideas of the rescattering mechanism
- High Harmonic Generation (HHG) within the rescattering mechanism
- Specifics of initial/final wave function manifested in HHG
  - Angular part: Degenerate Combinational HG
  - Radial part: sensitivity of HHG rates
- Another process described by rescattering mechanism: Above-Threshold Detachment with Excitation
- Conclusion

THREE-STEP OR RESCATTERING MECHANISM

The three steps are:

1. Above-Threshold Ionization (ATI) (neutral atoms)

   or Above-Threshold Detachment (ATD) (anions)

2. Propagation of electron in the field of a laser wave

3. Recollision with residual atomic core in laser field

   (Specifics of the last step depends on the process considered)

Example: High Harmonic Generation (HHG)

$$\mathrm{A} + m\omega \xrightarrow{\mathrm{ATI}} \mathrm{A}^{+} + e + (N - m)\,\omega \xrightarrow{\mathrm{LAR}} \mathrm{A} + \{N\omega\}$$

The last step is Laser Assisted Recombination (LAR)

Three-step mechanism is known also under names 'atomic antenna', or 'recollision', or 'two-step', or 'simpleman model'

M. Yu. Kuchiev, Pis'ma Zh. Eksp. Teor. Fiz. **45**, 319 (1987) [JETP Letters **45**, 404 (1987)]; J. Phys. B **28**, 5093 (1995); Phys. Lett. A **212**, 77 (1996).

P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).

Processes effectively described by three-step mechanism

ONE-ELECTRON PROCESSES

High Harmonic Generation (HHG)

$$\mathrm{A} + m\omega \to \mathrm{A}^{+} + e + (N-m)\,\omega \to \mathrm{A} + \{N\omega\}$$

High Above-Threshold Ionization (HATI)

$$\mathrm{A} + m\omega \to \mathrm{A}^{+} + e + (N-m)\,\omega \to \mathrm{A}^{+} + e(\vec{p}_N)$$

TWO-ELECTRON PROCESSES

Double ionization (DI)

$$\mathrm{A} + m\omega \to \mathrm{A}^{+} + e + (N-m)\,\omega \to \mathrm{A}^{++} + 2e$$

Above-Threshold Ionization with Excitation (ATIE) or

Above-Threshold Detachment with Excitation (ATDE)

$$\mathrm{A} + m\omega \to \mathrm{A}^{+} + e + (N-m)\,\omega \to \left(\mathrm{A}^{+}\right)^{*} + e$$

Since A^{+*} is rapidly ionized in the laser field this process effectively contributes to yield of A^{++} ions

V. N. Ostrovsky, J. Phys. B **36**, 2647 (2003)

Typical Three-Step Process: High Harmonic Generation

Rate:

$$\mathcal{R}_N = \frac{\Omega^3}{2\pi c^3} \left| d_N^+ \right|^2$$

Amplitude:

$$d_N^+ = -\frac{i}{T} \int_0^T dt \int_{-\infty}^t dt' \langle \phi_a(t) | \exp(i\Omega t) U G(t,t')\, V(t') | \phi_a(t') \rangle$$

$$U = \vec{\epsilon} \cdot \vec{r} \qquad V_F(t) = -\,\vec{r} \cdot \vec{F}(t) \qquad \Omega = N\omega$$

$G(t,t')$ is Green function; approximately

$$G_{\mathrm{Volk}}(\vec{r},t;\vec{r}',t') = \int \frac{d^3p}{(2\pi)^3}\, \phi_{\vec{p}}(\vec{r},t)\, \phi_{\vec{p}}(\vec{r}',t')$$

M. Yu. Kuchiev and V. N. Ostrovsky. Phys. Rev. A **60**, 3111 (1999):

$$d_N^+ = 2 \sum_m d_{Nm}^+ \qquad d_{Nm}^+ = A_{m\mu_0}(\vec{K}_m)\, B_{M\,m\mu_0}(\vec{K}_m)$$

$$K_m = \sqrt{2\,(m\omega - U_p + E_a)} \qquad \vec{K}_m \parallel \vec{F}$$

$A_{m\mu_0}(\vec{K}_m)$ - ATI amplitude;

$C_{Nm}(\vec{K}_m)$ - Laser Assisted Recombination (LAR) amplitude;

$B_{N\,m\mu_0}(\vec{K}_m)$ - Propagation-LAR (PLAR) amplitude

$$B_{N\,m\mu_0}(\vec{K}_m) \approx \frac{1}{R_{m\mu_0}}\, C_{Nm}(\vec{K}_m) \qquad R_{m\mu_0} = -\frac{F}{\omega^2}\, \cos\omega t'_{m\mu_0}$$

$$d_{Nm}^+ = A_{m\mu_0}(\vec{K}_m)\, \frac{1}{R_{m\mu_0}}\, C_{Nm}(\vec{K}_m)$$

The first step: Above-Threshold Detachment

Steady (quasienergy) state, rate calculation, no depletion

Keldysh scheme

$$A_m(\vec{p}_m) = \frac{1}{T}\int_0^T dt\,\langle \phi_{\vec{p}_m}(t)|V_F(t)|\phi_a(t)\rangle$$
$$V_F(t) = -\,\vec{r}\cdot\vec{F}(t)$$

$\phi_{\vec{p}_m}(t)$ - Volkov wave function;

$\phi_a(\vec{r},t) = \phi_a(\vec{r})\,e^{-iE_a t}$ - initial bound state wave function

$E_a + m\omega = \frac{1}{2}p_m^2 + U_p$; $U_p = F^2/(4\omega^2)$ - ponderomotive potential

Saddle-point calculation of integral over time - adiabatic process; the number of absorbed photons m is large (Gribakin and Kuchiev 1997)

$$A_m(\vec{p}_m) = \sum_\mu A_{m\mu}(\vec{p}_m)$$
$$= -\frac{(2\pi)^2}{T}A\Gamma\left(1+\frac{\nu}{2}\right)2^{\nu/2}\kappa^\nu \sum_\mu Y_{\ell m_\ell}\left(\frac{\vec{p}_\mu}{p_\mu}\right)\frac{\exp(-iS_\mu)}{\sqrt{2\pi(-iS''_\mu)^{\nu+1}}}$$

$$\phi_a(\vec{r}) \approx Ar^{\nu-1}\exp(-\kappa r)\,Y_{\ell m_\ell}\left(\frac{\vec{r}}{r}\right) \qquad (r \gg 1/\kappa) \qquad \nu = \frac{Z}{\kappa}$$
$$E_a = -\frac{1}{2}\kappa^2 \qquad \vec{k}_t = \int^t \vec{F}(t')\,dt'$$
$$S(t) = \frac{1}{2}\int^t \left(\vec{p}+\vec{k}_t\right)^2 dt' - E_a t$$

Saddle points $t_{m\mu_0}$ are solution of equation $S'(t) = 0$

Technical details

$$\int_{-\infty}^{t} dt' \ \exp(iEt')\, f(t')$$
$$= -i \sum_{m=-\infty}^{\infty} \frac{1}{T} \int_0^T dt'\, f(t')\ \exp\{i[(E-m\omega)t + m\omega t']\}\ \frac{1}{E - m\omega - i0}$$

$$\begin{aligned}
\vec{R}(\vec{r},\vec{r}';t,t') &= \int\int_t^{t'} \vec{k}_t\, d\tau - \vec{r}' + \vec{r} \\
&= \frac{\vec{F}}{\omega^2}\,(\cos\omega t - \cos\omega t') + \vec{r} - \vec{r}' \\
R &\approx R_0 + (\vec{r} - \vec{r}')\,\frac{\vec{R}_0}{R_0} \\
\vec{R}_0(t,t') &= \frac{\vec{F}}{\omega^2}\,(\cos\omega t - \cos\omega t') \\
\frac{F}{\omega^2} &\gg 1
\end{aligned}$$

PLAR amplitude

$$B_{Nm\mu} = -\frac{1}{2\pi T} \int_0^T \frac{dt}{R_0(t, t'_{m\mu})}\ \langle\phi_a(t)|\exp\left(i\,\tilde{\Omega}_M t\right) U|\phi_{\vec{p}}(t)\rangle$$

LAR amplitude

$$C_M(\vec{p}) = \frac{1}{T} \int_0^T dt\,\langle\phi_a(t)|\exp\left(i\,\tilde{\Omega}_M t\right) U|\phi_{\vec{p}}(t)\rangle$$

Saddle point approximation for integration over t' and

(not necessarily) t

The Third Step: Laser Assisted Recombination

A.Jaron, J.Z.Kaminski, and F.Ehlotzky, Phys. Rev. A 61, 023404 (2000)

M.Yu.Kuchiev and V.N.Ostrovsky, Phys. Rev. A 61, 033414 (2000)

Amplitude

$$
\begin{aligned}
C_M(\vec{p}) &= \frac{1}{T}\int_0^T dt\,\langle\phi_a(t)|\exp\left(i\,\tilde{\Omega}_M t\right)U|\phi_{\vec{p}}(t)\rangle \\
\tilde{\Omega}_M &= \frac{1}{2}p^2 + \frac{F^2}{4\omega^2} - \epsilon_a + M\omega \\
U &= \vec{\epsilon}\cdot\vec{r}
\end{aligned}
$$

$$
\begin{aligned}
C_M(\vec{p}) &= -\frac{1}{T}\int_0^T dt\exp\{i[m+\eta)\omega t - S(t)]\}\,\tilde{\phi}_a^{(\vec{\epsilon})}(-\vec{p}-\vec{k}_t) \\
S(t) &= \frac{1}{2}\int^t d\tau\,(\vec{p}+\vec{k}_t)^2 - E_a t \\
\tilde{\phi}_a^{(\vec{\epsilon})}(\vec{q}) &= i\,(\vec{\epsilon}\cdot\nabla_{\vec{q}})\;\tilde{\phi}_a(\vec{q}) \\
\tilde{\phi}_a(\vec{q}) &= \int d^3r\,\exp(-i\vec{q}\cdot\vec{r})\,\phi_a(\vec{r})
\end{aligned}
$$

SPECIFICS OF INITIAL/FINAL WAVE FUNCTION IN HHG

Conventional an active s-electron is considered and its wave functions is taken in the form of a mere exponent, $\phi_a = N \exp(-\kappa r)$

What occurs if

- Initial state orbital momentum ℓ is non-zero ?
- Radial wave function has more complicated (and realistic) form ?

Conventional HG Combinational HG Degenerate Combinational HG (DCHG)

The m_ℓ-changing transitions are of interest since

- the emitted harmonic differs in polarization from that of the original laser wave;
- the harmonics produced by the conventional and DCHG processes are not coherent, although have the same frequency

DCHG process is forbidden by the Pauli Exclusion Principle if the active electron belongs to a filled shell, like np^6 shells in the noble gases Ne – Xe. The same applies to halogen anions F$^-$ – I$^-$.

Within the rescattering mechanism for HHG initial $m_\ell = 0$ sublevel is effectively selected on the first (ATI) step

There seems to be no obvious general reason to conclude what is more efficient on the last (LAR) step: m_ℓ-conserving (conventional HHG) or m_ℓ-changing (DCHG) process. Therefore numerical calculations are required. Two examples were considered: excited hydrogen atom H(2p) and B(1s^{2}2s^2 2p) ground state atom

CHOICE OF RADIAL WAVE FUNCTIONS

- Asymptotic (large-r) approximation together with length gauge for electromagnetic field work well for ATI (ATD) (Gribakin and Kuchiev; Kjeldsen and Madsen).
- For the LAR amplitude all range of r is important.
- Therefore realistic (non-asymptotic) wave functions might be important for calculations of HHG rates.

Realistic wave functions (Clementi and Roetti)

$$\begin{aligned} \phi_{n_i \ell m_\ell}(\vec{r}) &= \sum_i C_i \chi_{n_i \ell m_\ell}(\vec{r}) \ , \\ \chi_{n_i \ell m_\ell}(\vec{r}) &= R_{n_i,\ell}(r)\, Y_{\ell m_\ell}(\hat{\vec{r}}) \ , \qquad \hat{\vec{r}} = \vec{r}/r \ , \\ R_{n_i,\ell}(r) &= N_i r^{n_i - 1} \exp(-\zeta_i r) \ . \end{aligned}$$

The sum contains 4 or 5 terms; the parameters ζ_i, C_i, N_i are tabulated.

Asymptotic wave function

$$\phi_{n_i \ell m_\ell}(\vec{r}) \approx A_a r^{\nu - 1} \exp(-\kappa r)\, Y_{\ell m_\ell}(\hat{\vec{r}}) \qquad (r \gg 1/\kappa)$$

$$\nu = \frac{Z}{\kappa} \ , \qquad E_a = -\frac{1}{2}\kappa^2$$

Calculations with asymptotic wave functions:

V. N. Ostrovsky and J. B. Greenwood, J. Phys. B **38**, 1867 (2005)

Multiphoton detachment with atom excitation

$$\mathrm{A}^- + N\omega \to \mathrm{A}^*(\nu) + e(\mathbf{p}_N) \ ,$$

$$\text{example}: \ \mathrm{H}^- + N\omega \to \mathrm{H}(2\mathrm{s},\, 2p) + e(\mathrm{p}_N) \ ,$$

$$\frac{1}{2}\, p_N^2 = N\omega - \varepsilon_a - \Delta E_\nu - U_p \ ,$$

$$\frac{dw_N}{d\cos\theta} = 2\pi\, W_N(\theta) \ , \qquad W_N(\theta) = \frac{1}{(2\pi)^2}\, p_N \mid A_N(\mathbf{p}_N) \mid^2 \ \ ,$$

$$w_N = 2\pi \int_0^\infty W_N(\theta)\, \sin\theta\, d\theta \ .$$

$$A_N(\mathbf{p}_N) = \frac{1}{T} \int_0^T dt \int_{-\infty}^t dt' \, \langle \phi_{\mathbf{p}_N}(t)\, \phi_\nu(t) \mid V_a\, G(t,t')\, V_F(t') \mid \phi_0(t') \rangle \ ,$$

$$A_N(\mathbf{p}_N) = \sum_\sigma A_{N\sigma}(\mathbf{p}_N) \ ,$$

$$A_{N\sigma}(\mathbf{p}_N) = \sum_m \sum_\mu A_{m\,\mu}^{(\mathrm{sp})}(\mathbf{K}_m)\, \mathcal{V}_{N\,m\mu}(\mathbf{p}_N,\, \mathbf{K}_m) \ ,$$

$$\mathcal{V}_{N\,m\mu}(\mathbf{p}_N,\, \mathbf{K}_m) = -\frac{1}{2\pi T} \int_0^T dt\, \frac{1}{R_0(t, t'_{m\mu})} \langle \phi_{\mathbf{p}_N}(t)\, \phi_\nu(t) \mid V_a \mid \phi_{\mathbf{K}_m}(t)\, \phi_0(t) \rangle \ ,$$

$$\mathcal{V}_{N\,m\mu}(\mathbf{p}_N,\,\mathbf{K}_m)\,=\,-\frac{1}{2\pi}\tilde{V}_\nu\left(\mathbf{p}_N-\mathbf{K}_m\right)\,G_{N\,m\mu_0}\left(\mathbf{p}_N-\mathbf{K}_m\right)\;,$$

$$\tilde{V}_\nu(\mathbf{q})\,=\,\int d^3\mathbf{r}_1\,d^3\mathbf{r}_2\,\exp(-i\mathbf{q}\mathbf{r}_1)\,\phi_\nu(\mathbf{r}_2)\,V_a(\mathbf{r}_1,\mathbf{r}_2)\,\phi_0(\mathbf{r}_2)\;,$$

$$G_{N\,m\mu}(\mathbf{q})\,=\,\frac{1}{T}\int\limits_0^T dt\,\frac{1}{R_0(t,t'_{m\mu})}\exp\left[i\,(N-m)\,\omega t+i\int^t d\tau\,\mathbf{q}\cdot\mathbf{k}_\tau\right], \tag{1}$$

$$f_\nu^{\mathrm{B}}(\mathbf{q})\,=\,-1/(2\pi)\tilde{V}_\nu(\mathbf{q})\;,$$

$$\begin{aligned} f^{\mathrm{B}}_{\nu N_{\mathrm{abs}}}(\mathbf{q})\,&=\,\frac{1}{T}\int_0^T\,dt\,\langle\phi_{\mathbf{p}_f}(t)\,\phi_\nu(t)\mid V_a\mid\phi_{\mathbf{p}_i}(t)\,\phi_0(t')\rangle\\ &=\,-\frac{1}{2\pi}\tilde{V}_\nu\left(\mathbf{q}\right)C_{N_{\mathrm{abs}}}(\mathbf{q})\;,\end{aligned}$$

$$C_{N_{\mathrm{abs}}}(\mathbf{q})\,=\,\frac{1}{T}\int\limits_0^T dt\,\exp\left(iN_{\mathrm{abs}}\omega t+i\int^t d\tau\,\mathbf{q}\cdot\mathbf{k}_\tau\right),\qquad \mathbf{q}=\mathbf{p}_i-\mathbf{p}_f\;.$$

$$w_N=2\pi\int_0^\infty\,W_N(\theta)\,\sin\theta\,d\theta\;.$$

CONCLUSION

(recent novel results)

- m_ℓ-changing (DCHG) process is discussed, calculated for the first time and compared with conventional HHG process;
- m_ℓ-changing (DCHG) process is analyzed and calculated for the first time
- High sensitivity of HHG rates to details of final state radial wave functions is found