Qualitative strong-field ionization models

- REMPI: Resonance-enhanced multiphoton ionization
- Multiphoton Ionization: Non-resonant multiphoton ionization
- ATI: Above-threshold ionization
Qualitative strong-field ionization models

REMPI
Resonance-enhanced multiphoton ionization

Multiphoton Ionization
Non-resonant multiphoton ionization

ATI
Above-threshold ionization

Multiphoton regime
(upper row)

vs.

Quasi-static regime
(lower row)

Tunnel ionization

Over-the-barrier ionization
Ionization models for atoms in fields

- **Lowest-order perturbation theory (LOPT)** (low laser intensities).
Ionization models for atoms in fields

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  - Perelomov, Popov, Terent’ev (PPT): tunneling
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- Solution of the time-dependent Schrödinger equation (**TDSE**): exact.
Validity regimes of approximations

\[ \frac{F}{\sqrt{2} E_{\text{ion}}} \]

Number of Photons

\[ \frac{\hbar \omega}{E_{\text{ion}}} \]

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Validity regimes of approximations

Quasi-static

Multiphoton

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Validity regimes of approximations

\[ F = \frac{\gamma_{\text{Kel}}}{\sqrt{2} E_{\text{ion}}} = 1 \]

LOPT

Quasi-static

Multiphoton

Number of Photons

8 4 2 1

∞ 0 0.125 0.25 0.5 1
Validity regimes of approximations

\[ \frac{F}{\sqrt{2} E_{\text{ion}}} \]

\[ F_{\text{BSI}} = \frac{E_{\text{ion}}^2}{4} \]

\[ \gamma_{\text{Kel}} = \frac{\omega}{F/\sqrt{2} E_{\text{ion}}} = 1 \]

- Over-the-barrier
- Quasi-static
- Multiphoton
- Tunneling
- LOPT

Number of Photons

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Berlin, 07.11.2014
Lowest-order perturbation theory (LOPT)

\[ \Gamma_{\text{LOPT}} \propto I^N \left| \sum_{\nu, \mu \ldots \zeta} \frac{\langle \Psi_f | \hat{D} | \Psi_\nu \rangle \langle \Psi_\nu | \hat{D} | \Psi_\mu \rangle \cdots \langle \Psi_\zeta | \hat{D} | \Psi_i \rangle}{[E_\nu - E_i - (N - 1)\omega] [E_\mu - E_i - (N - 2)\omega] \cdots [E_\zeta - E_i - \omega]} \right|^2 \]

- **Perturbative regime**: high photon energies, low intensity.
- Simple intensity dependence (\( \Gamma_{\text{LOPT}} \propto I^N \))
  \( \rightarrow \) absolute cross-sections, energy dependence?
- **Problem**: Calculation requires sum over all virtual intermediate states, incl. continua.
Lowest-order perturbation theory (LOPT)

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- \(H_2^+\) (vibrational and orientational dependence).

- \(H_2\) (fully correlated, method comparison complex scaling vs. \(B\) splines).

- \(H_2\) (including nuclear motion in BO approx.).
LOPT regime for H$_2$: intensity scan ($R = 1.4 \, a_0$)

\[ \Gamma^{(N)} = \sigma^{(N)} \left( \frac{I}{\hbar \omega} \right)^N \]

- $N$: Number of photons
- $\Gamma^{(N)}$: Ionization rate
- $\sigma^{(N)}$: N-photon ionization cross-section
- $I$: Intensity
- $\hbar \omega$: Photon energy

Ionization yield: $P_{\text{ion}} = \int_{\text{pulse}} \Gamma^{(N)} \, dt$
LOPT regime for $\text{H}_2$: frequency scan ($R = 1.4 \, a_0$)

Ionization Yield

$\text{TDSE}$

$I = 2.0 \times 10^{12} \, \text{W/cm}^2$

$\text{COS}^2$ pulse of 15 fs

Photon Energy (eV)

$10^{-9} \quad 10^{-8} \quad 10^{-7} \quad 10^{-6} \quad 10^{-5} \quad 10^{-4} \quad 10^{-3} \quad 10^{-2} \quad 10^{-1} \quad 10^0$
LOPT regime for $H_2$: frequency scan ($R = 1.4 a_0$)

**LOPT**: Ionization yield $P_{ion} = \int \Gamma^{(N)} \, dt$

$$\Gamma^{(N)} = \sigma^{(N)} \left( \frac{I}{\hbar \omega} \right)^N, \quad \sigma^{(N)} \propto \sum_{\nu, \mu \ldots \zeta} \frac{\langle \Psi_f | \hat{D} | \Psi_\nu \rangle \langle \Psi_\nu | \hat{D} | \Psi_\mu \rangle \cdots \langle \Psi_\zeta | \hat{D} | \Psi_i \rangle}{[E_\nu - E_i - (N-1)\omega] [E_\mu - E_i - (N-2)\omega] \cdots [E_\zeta - E_i - \omega]} \bigg| 2$$

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$I = 2.0 \times 10^{12} \ \text{W/cm}^2$

$\text{COS}^2$ pulse of 15 fs

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$1 = 2.0 \times 10^{12} \ \text{W/cm}^2$

COS$^2$ pulse of 15 fs
Resonances (REMPI) and thresholds ($\text{H}_2$, $R = 1.4 \, a_0$)
Resonances (REMPI) and thresholds ($H_2$, $R = 1.4 \alpha_0$)

![Graph showing population of ionization and ionization + excitation as a function of energy.](Image)

- Population of ionization and ionization + excitation as a function of energy.
- Energy (eV) range from 0 to 20.
- Population values range from $10^{-10}$ to $10^0$.

Mathematical expressions:

- Ionization: $I = 2.0 \times 10^{12}$ W/cm$^2$
- Ionization + Excitation: $B^1\Sigma_u^+$

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Resonances (REMPI) and thresholds ($H_2$, $R = 1.4 \alpha_0$)
Resonances (REMPI) and thresholds ($H_2$, $R = 1.4 a_0$)

\[ I = 2.0 \times 10^{12} \text{ W/cm}^2 \]

- Ionization
- Ionization + Excitation

Energy (eV)
Population

$2\hbar \nu$
Resonances (REMPI) and thresholds ($H_2$, $R = 1.4 \alpha_0$)

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Photoelectron spectra for $\text{H}_2$

**LOPT prediction:**
only first $\Sigma_u$ peak exists
(3 photon process)!

Additional peaks:
above-threshold ionization (ATI)

$\text{H}_2$ at $R = 1.40\ a_0$
Photon energy: 6.2 eV
Pulse length: 25 cycles
$\approx 16.5\ \text{fs}$
**Photoelectron spectra (H atom)**

**H atom:**

Ionization potential $I_P = 13.6$ eV

- **400 nm (3.1 eV)** $\rightarrow N_{ph,min} = 5$
- **800 nm (1.55 eV)** $\rightarrow N_{ph,min} = 9$
- **1600 nm (0.775 eV)** $\rightarrow N_{ph,min} = 18$

**Efficient solution of TDSE for H atom:**

Y. V. Vanne and A. Saenz, to be published

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Validity regimes of approximations (reminder)

\[ F \frac{\sqrt{2}}{E_{\text{ion}}} \]

\[ F_{\text{BSI}} = \frac{E_{\text{ion}}^2}{4} \]

\[ \gamma_{\text{Kel}} = \frac{\omega}{F_{\text{BSI}} \sqrt{2} E_{\text{ion}}} = 1 \]

Over-the-barrier

Quasi-static

Tunneling

Multiphoton

LOPT

\[ \frac{F}{\sqrt{2} E_{\text{ion}}} \propto \sqrt{\frac{\hbar \omega}{E_{\text{ion}}}} \]

\[ \frac{\hbar \omega}{E_{\text{ion}}} \]

Number of Photons

8 4 2 1
Quasi-static approximation (QSA)

Concept: Atomic and molecular response to intense laser field is similar to the response to a slowly varying electric dc field (with strength $F$).

\[
\Gamma_{ac} = \frac{1}{T} \int_{-T/2}^{+T/2} \Gamma_{dc} (F(t)) \, dt
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- **Quasistatic regime:** low frequency, high intensity.
- The static rate is (evidently) frequency independent.
- **Tunnel regime** (lower intensity) vs. **over-the-barrier ionisation** (higher intensity).
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- **Quasistatic regime:** low frequency, high intensity.
- The static rate is (evidently) frequency independent.
- **Tunnel regime** (lower intensity) vs. over-the-barrier ionisation (higher intensity).
- Simple atomic one-electron tunnel models exist for a long time.
  Example: Ammosov-Delone-Krainov (ADK) model.
- Fully correlated 3D calculation of dc rates difficult !!!
  → so far ab initio dc rates exist only for $H_2^+$ and $H_2$!
Validity of exact QSA for H atom (I)

Gaussian pulse, 2.4 fs.

- TDSE result for \( \lambda = 800 \) nm
- TDSE result for \( \lambda = 1064 \) nm
- Yield using ab initio static rate

Ionization yield vs. Peak intensity, \( 10^{14} \) W/cm\(^2\)
Validity of exact QSA for H atom (II)

Ionization Yield vs. Electric Field (au)

- Quasi Static Appr. (Scrinzi)
- TDSE (Scrinzi)

SECH Pulse of FWHM 5 fs
1.55 eV


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Example electron spectrum (ATI)

Technical details of the TDSE calculation:

Hydrogen atom

Laser parameters: 1300 nm; 6 cycles; \( \cos^2 \); \( I_{\text{max}} = 10^{14} \text{ W/cm}^2 \)

Direct electrons: 0 to about 2 times the ponderomotive energy \( U_p \).
Corkum’s 3-step model for imaging


1. Electron escapes through or over the electric-field lowered Coulomb potential (a).
2. Electronic wavepacket moves away until the field direction reverses (b) and is (partly) driven back to its parent ion (c).
3. The returning electron may (d)
   - scatter elastically (electron diffraction)
   - scatter inelastically (excitation, dissociation, double ionization, . . . )
   - recombine radiatively (high-harmonic radiation).

Outcome of 3rd step reveals time-resolved structural information!
Examples for pioneering experiments (I)

Orbital tomography using high-harmonic radiation:
[J. Itatani et al., Nature 432, 867 (2004)]


Tomographical reconstruction of the HOMO (highest-occupied molecular orbital) of N\(_2\) using the high-harmonic generation (HHG):

(a) Cartoon of the process: the interference pattern encoded in the HHG changes with electron wavelength and molecular orientation.
(b) Experimentally reconstructed orbital.
(c) Quantum-chemically calculated orbital (reference).
Examples for pioneering experiments (II)

Laser induced electron tunneling and diffraction:
[M. Meckel et al., Science 320, 1478 (2008)]

Sketch:
Some electrons tunnel directly to the detector, others recollide and show thus diffraction. Both, direct and recolliding electrons may reveal structural information!

Experimental results:
indication (picture?) of the different orbital structures of the HOMO of N₂ and O₂!