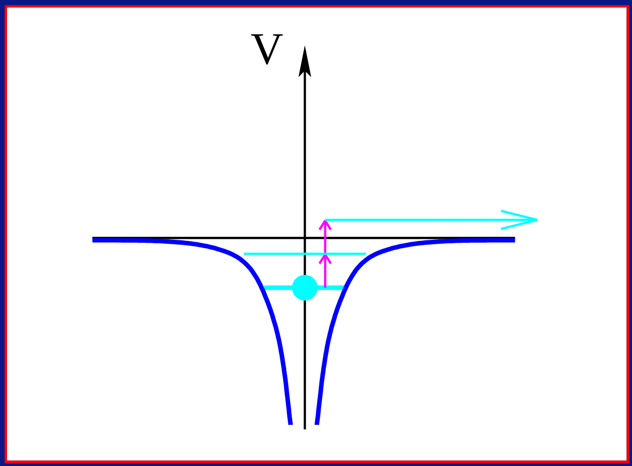
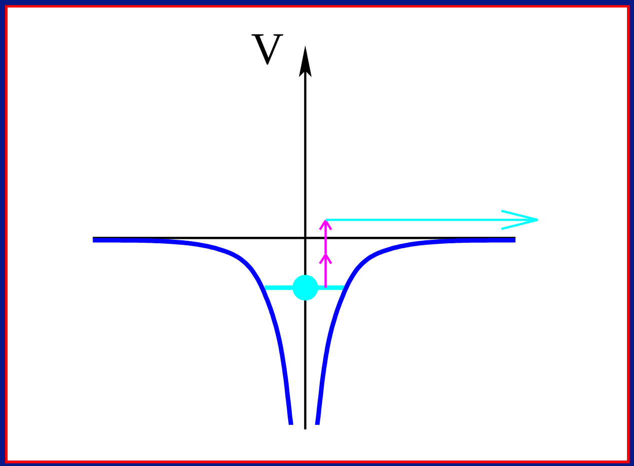
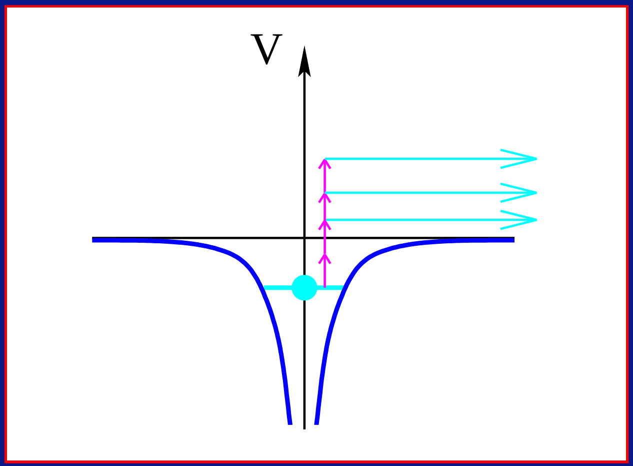
Qualitative strong-field ionization models



REMPI
Resonance-enhanced multiphoton ionization

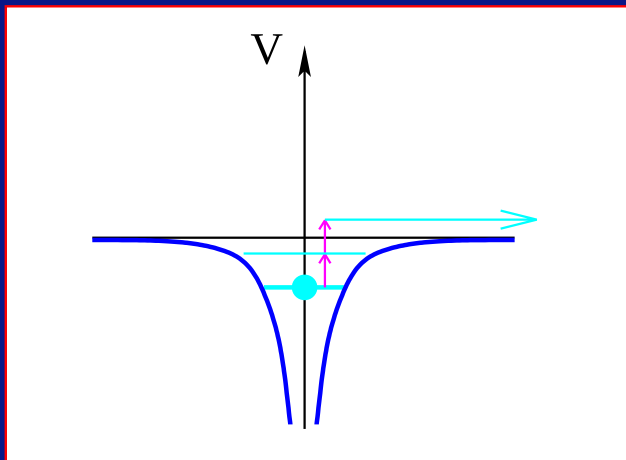


Multiphoton Ionization
Non-resonant multiphoton ionization



ATI
Above-threshold ionization

Qualitative strong-field ionization models



REMPI

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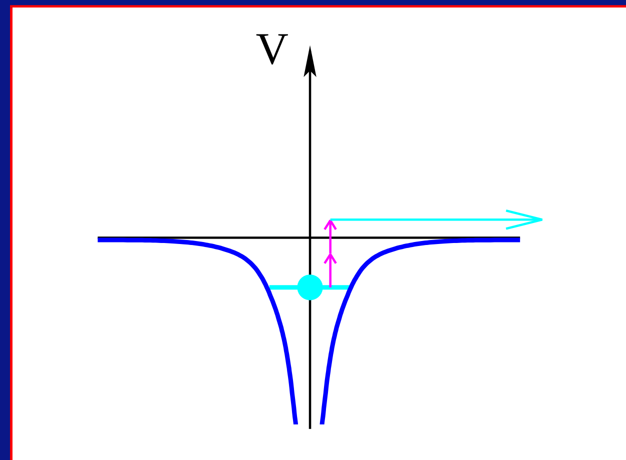

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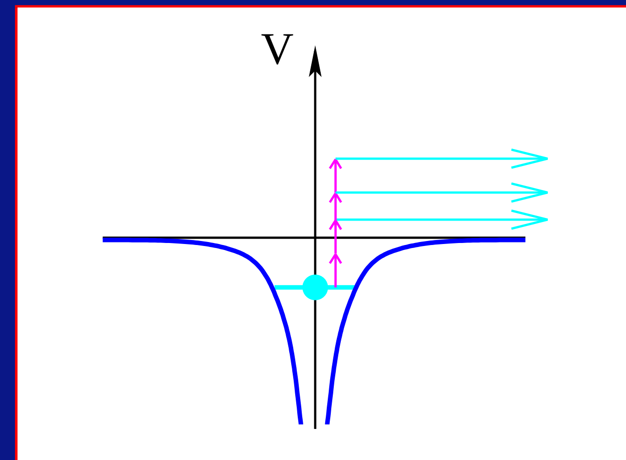

ATI

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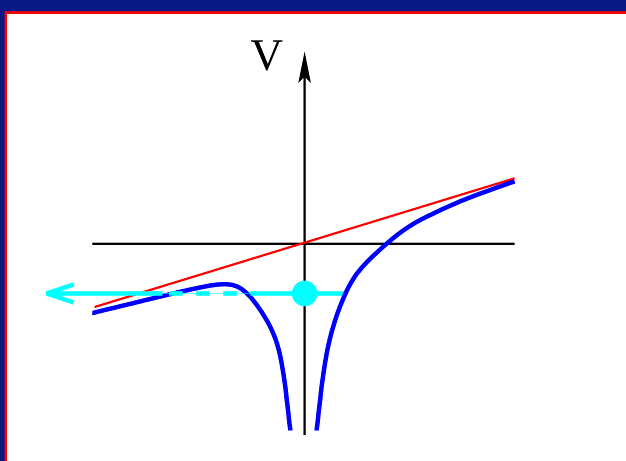
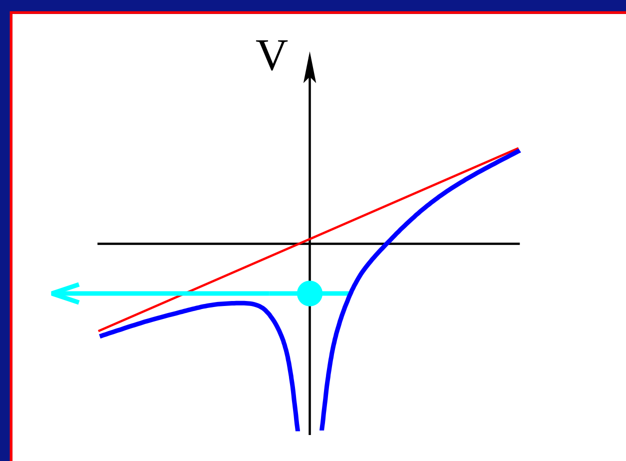


Tunnel ionization



Over-the-barrier ionization

Multiphoton regime

(upper row)

vs.

Quasi-static regime

(lower row)

Ionization models for atoms in fields

- **Lowest-order perturbation theory (LOPT)** (low laser intensities).

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  - ⋆ barrier-suppression models (none appears convincing)
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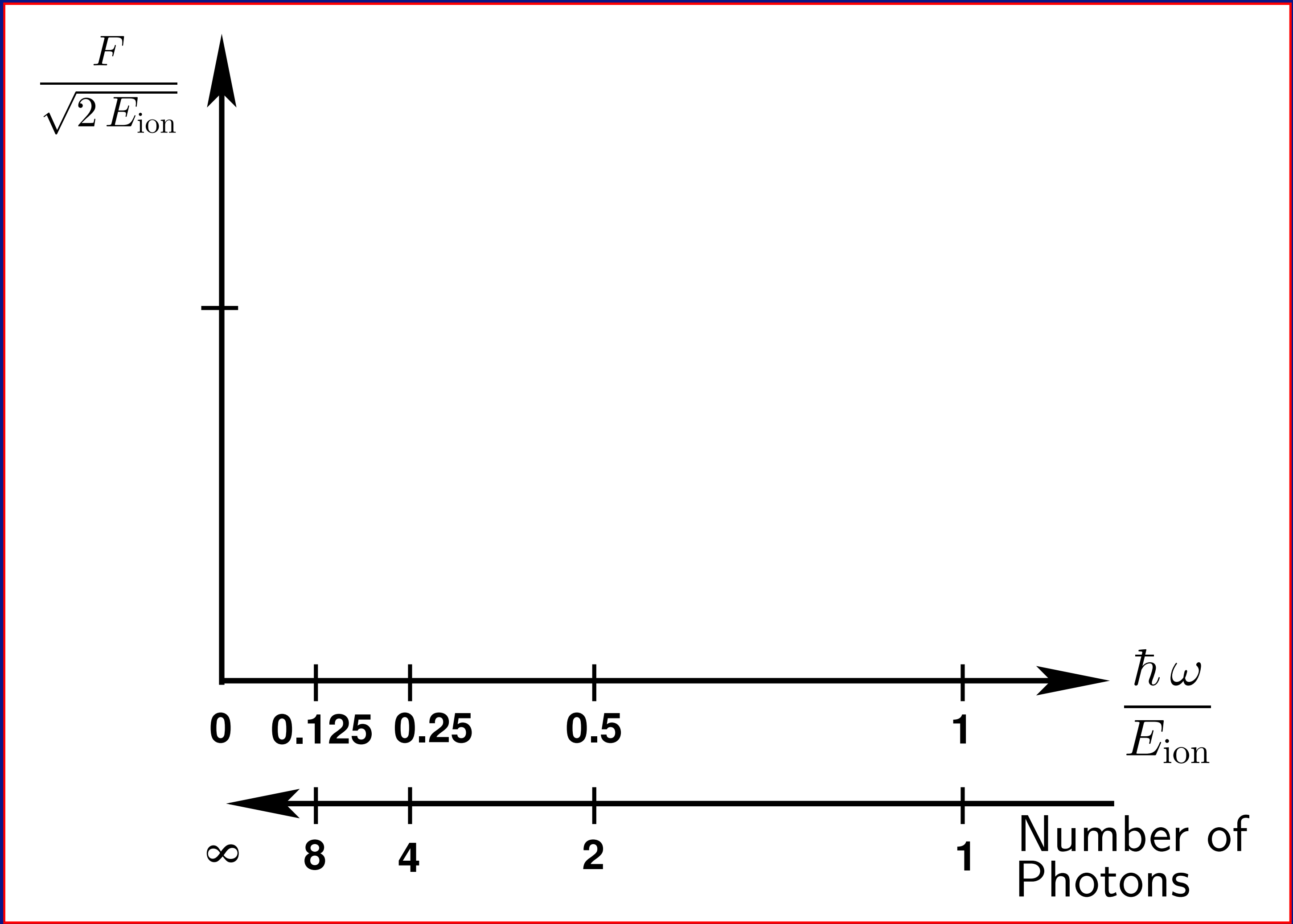
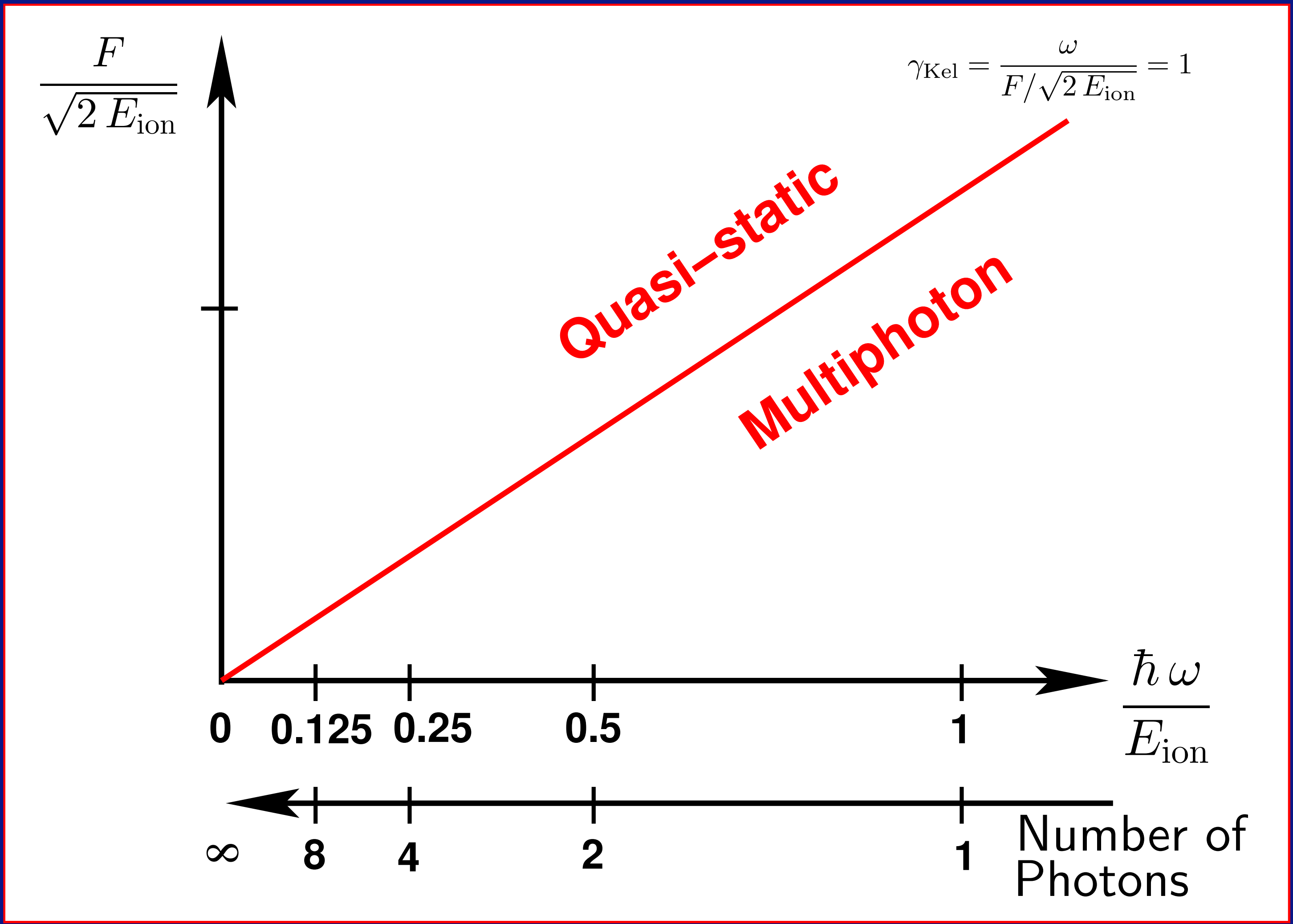
- Solution of the time-dependent Schrödinger equation (**TDSE**): exact.

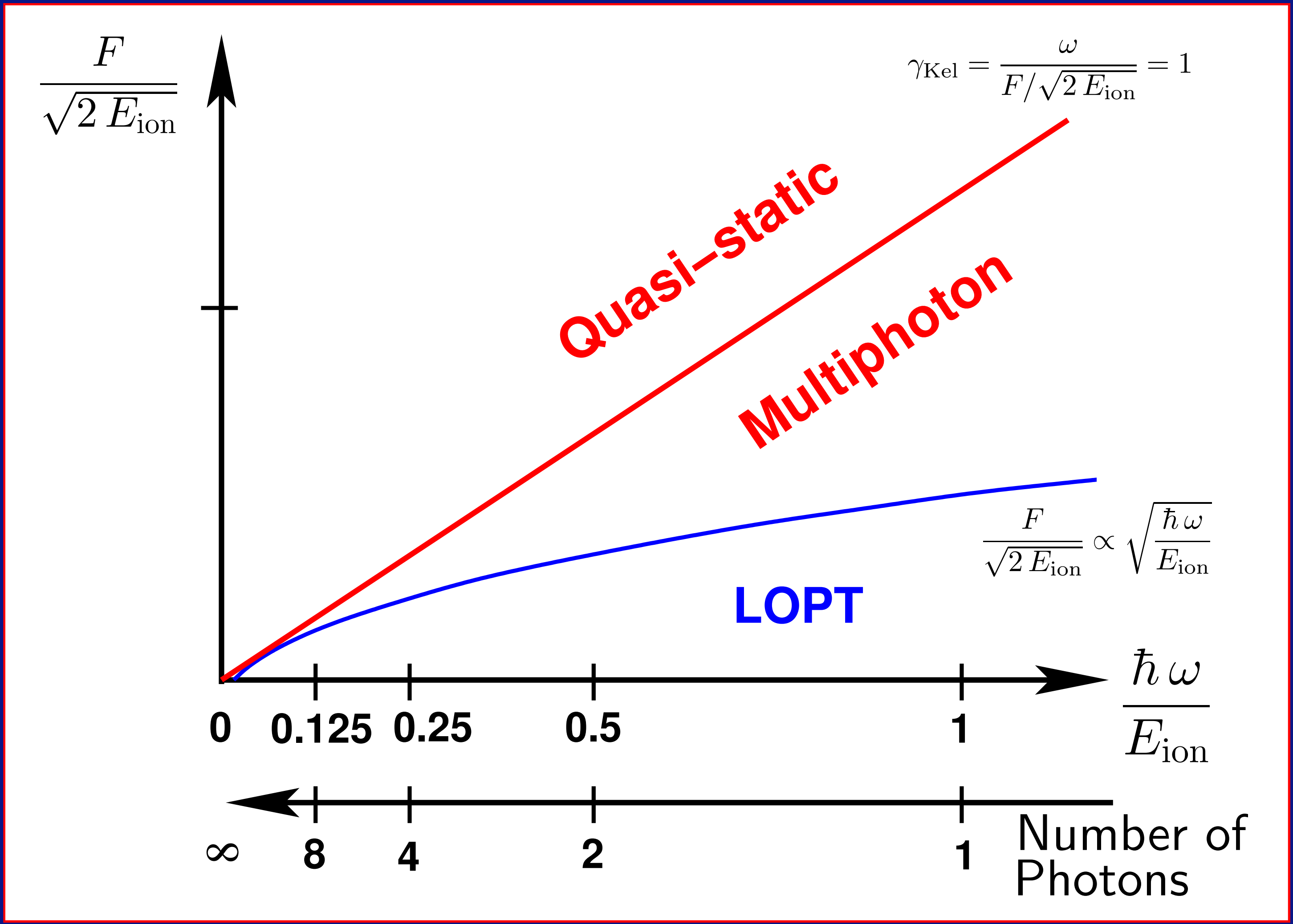
Validity regimes of approximations



Validity regimes of approximations



Validity regimes of approximations



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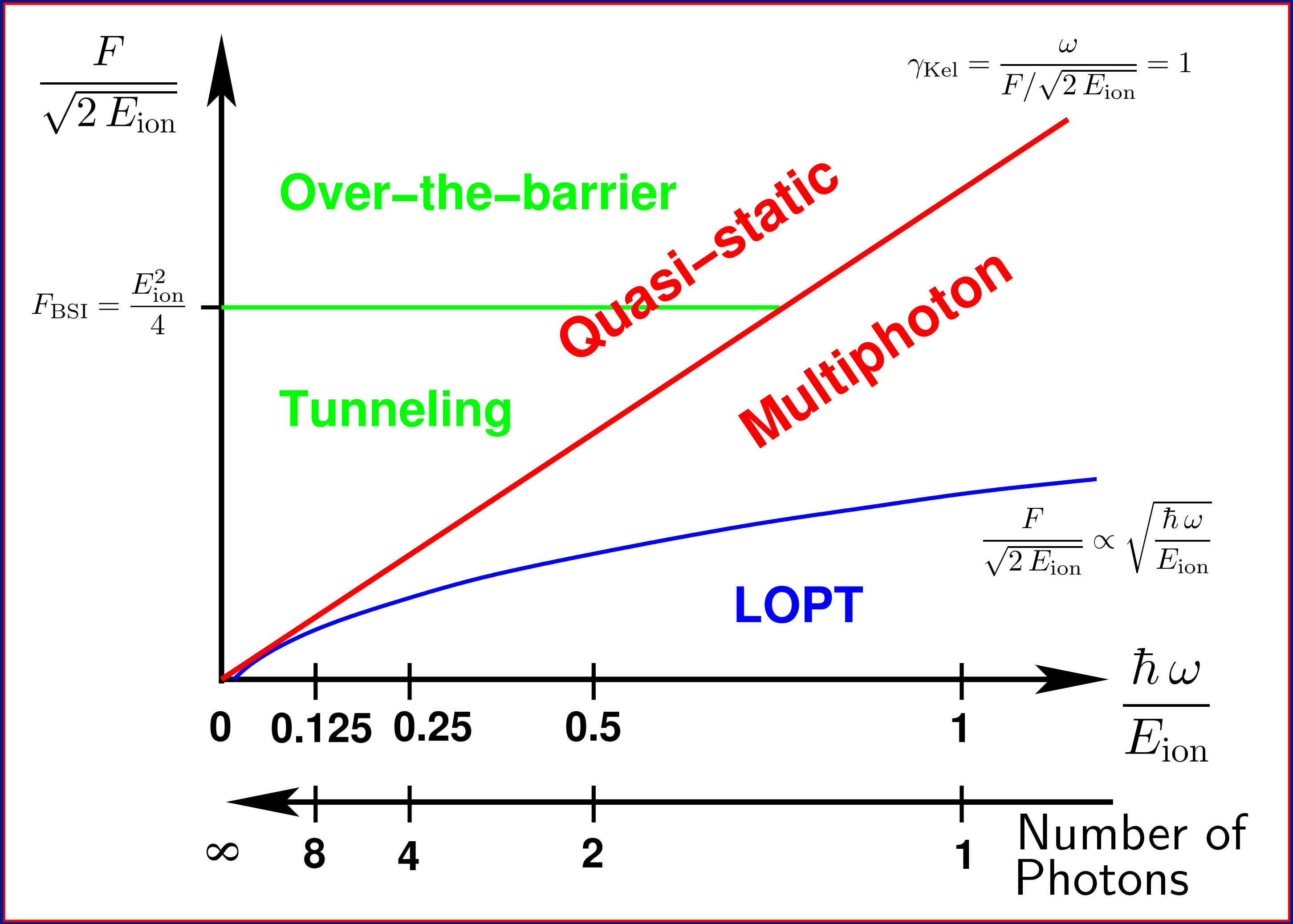

Lowest-order perturbation theory (LOPT)

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

- **Perturbative regime:** high photon energies, low intensity.
- Simple intensity dependence ($\Gamma_{\mathrm{LOPT}} \propto I^N$)

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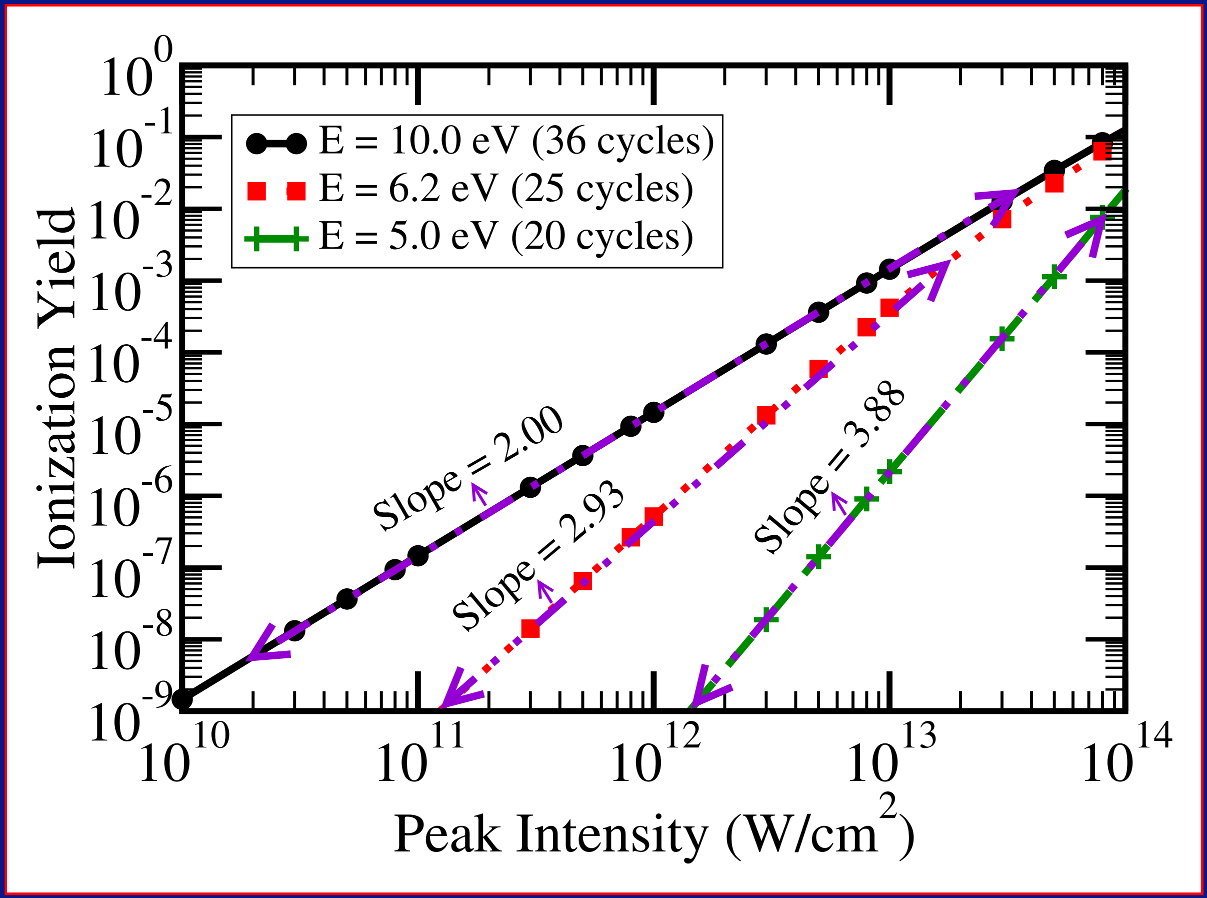
  - H_2^+ (vibrational and orientational dependence).

    A. Apalategui, A. Saenz und P. Lambropoulos, *J. Phys. B: At. Mol. Opt. Phys.* **33**, 2791 (2000).
  - H_2 (fully correlated, method comparison complex scaling vs. B splines).

    A. Apalategui und A. Saenz, *J. Phys. B: At. Mol. Opt. Phys.* **35**, 1909 (2002).
  - H_2 (including nuclear motion in BO approx.).

    A. Palacios, H. Bachau, F. Martín, *Phys. Rev. Lett.* **96**, 143001 (2006).

LOPT regime for H_2: intensity scan ($R = 1.4\,a_0$)

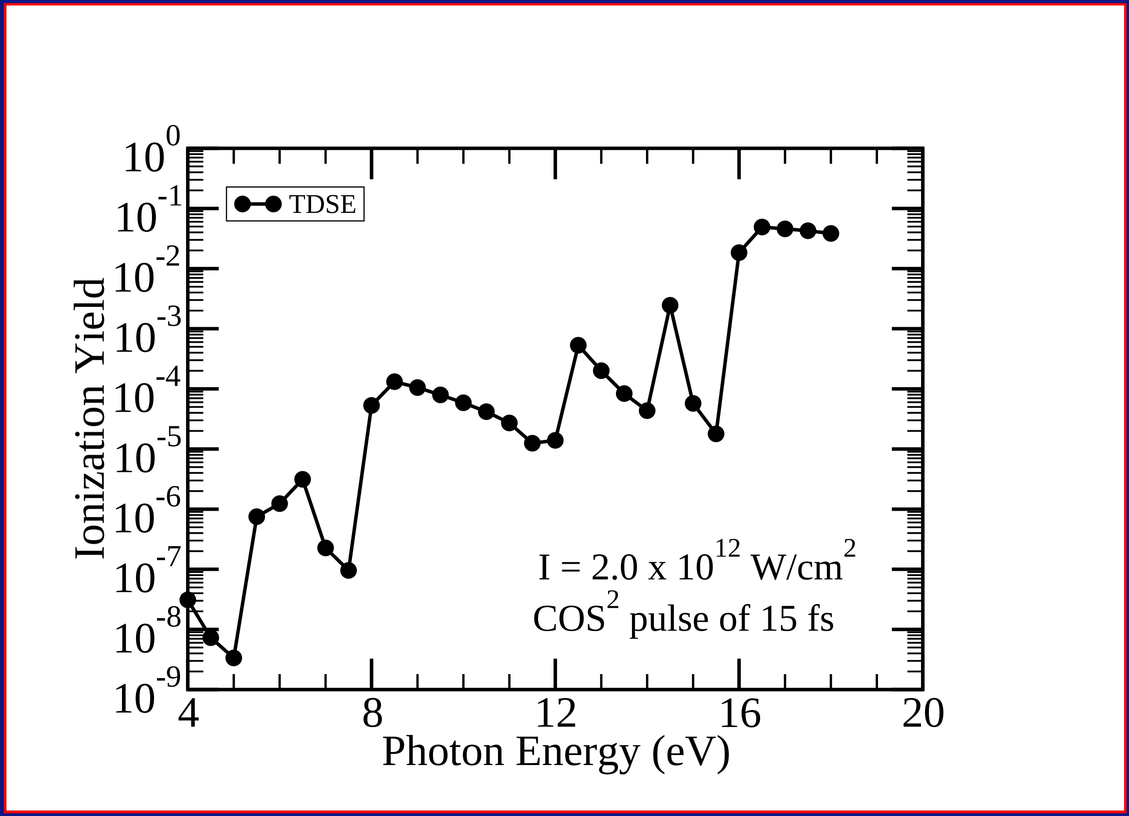


$$\Gamma^{(N)} = \sigma^{(N)}\left(\frac{I}{\hbar\omega}\right)^N$$

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

Ionization yield: $P_{\text{ion}} = \int_{\text{pulse}} \Gamma^{(N)} \mathrm{d}t$

LOPT regime for H_2: frequency scan ($R = 1.4\,a_0$)



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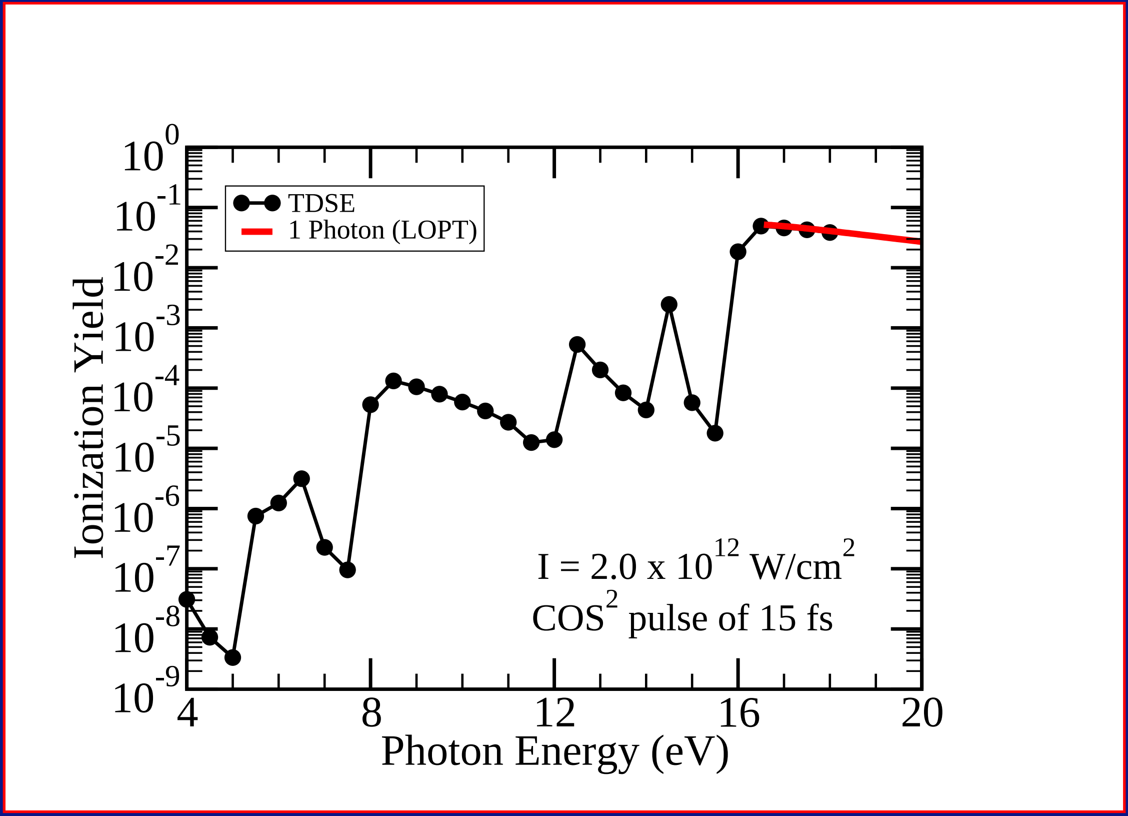

LOPT: Ionization yield $P_{\text{ion}} = \int \Gamma^{(N)}\,\mathrm{d}t$

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

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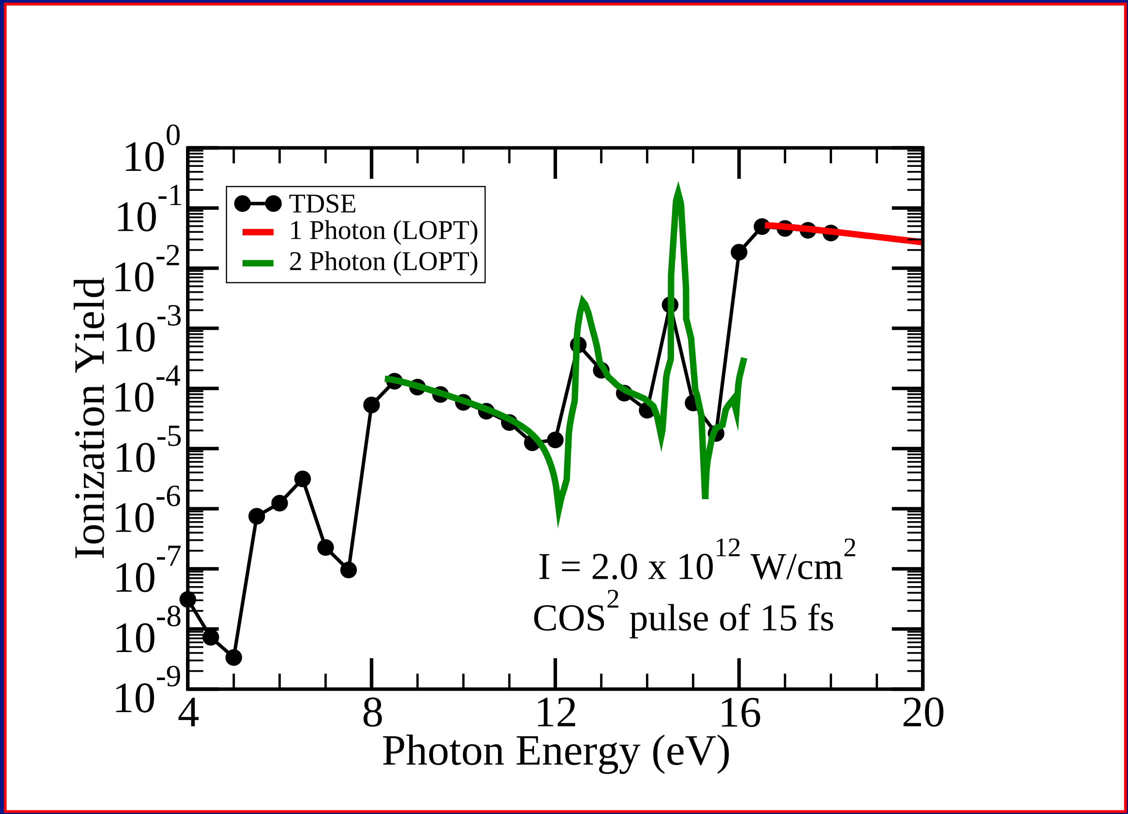

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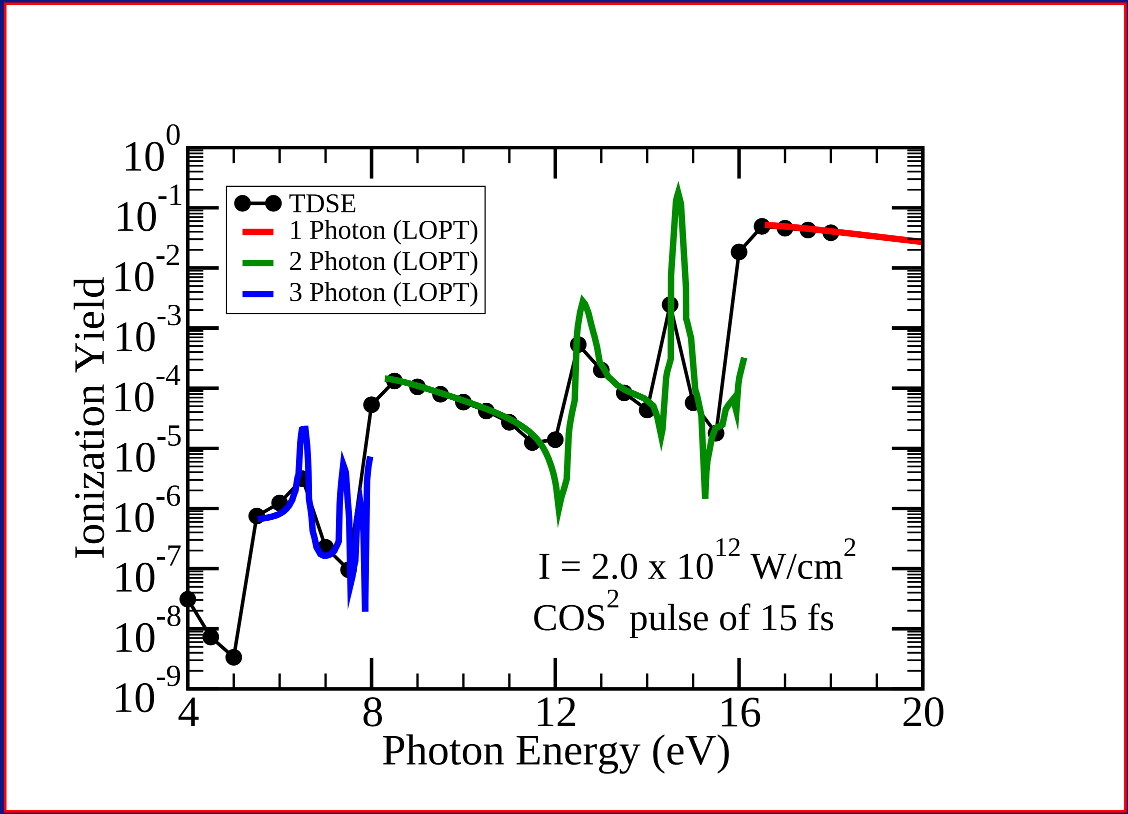

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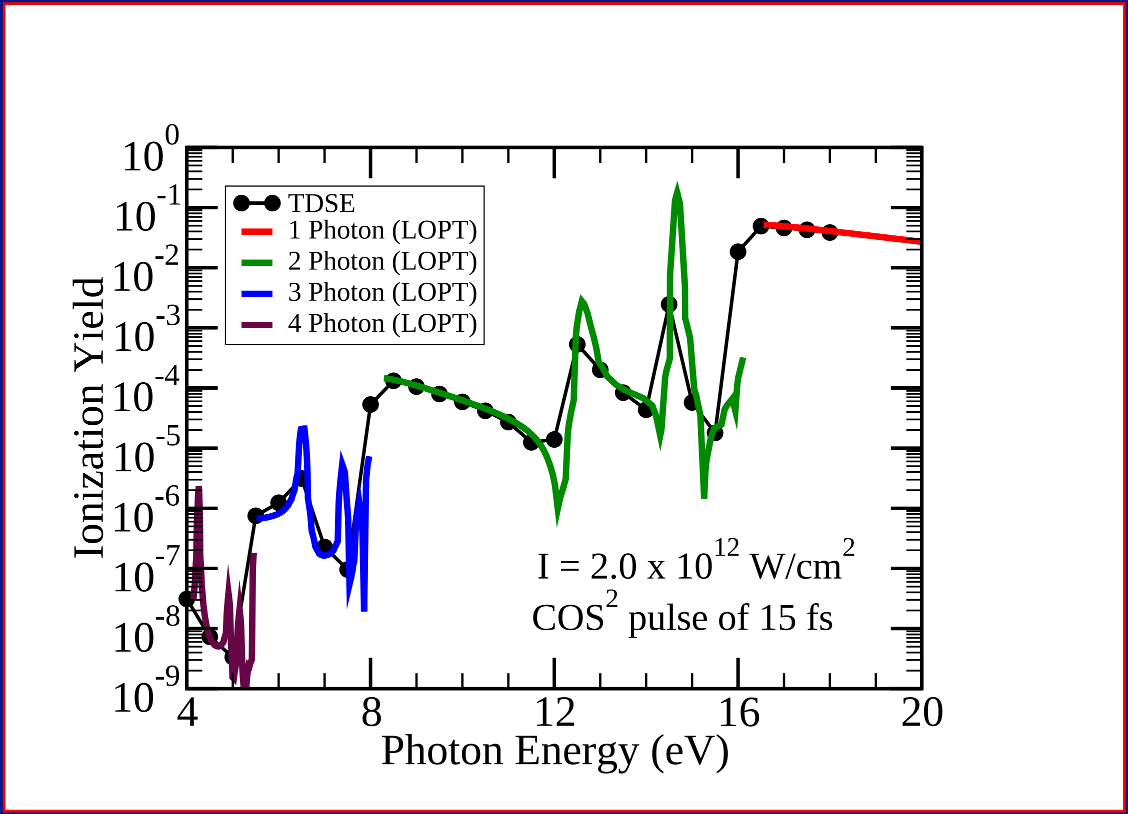

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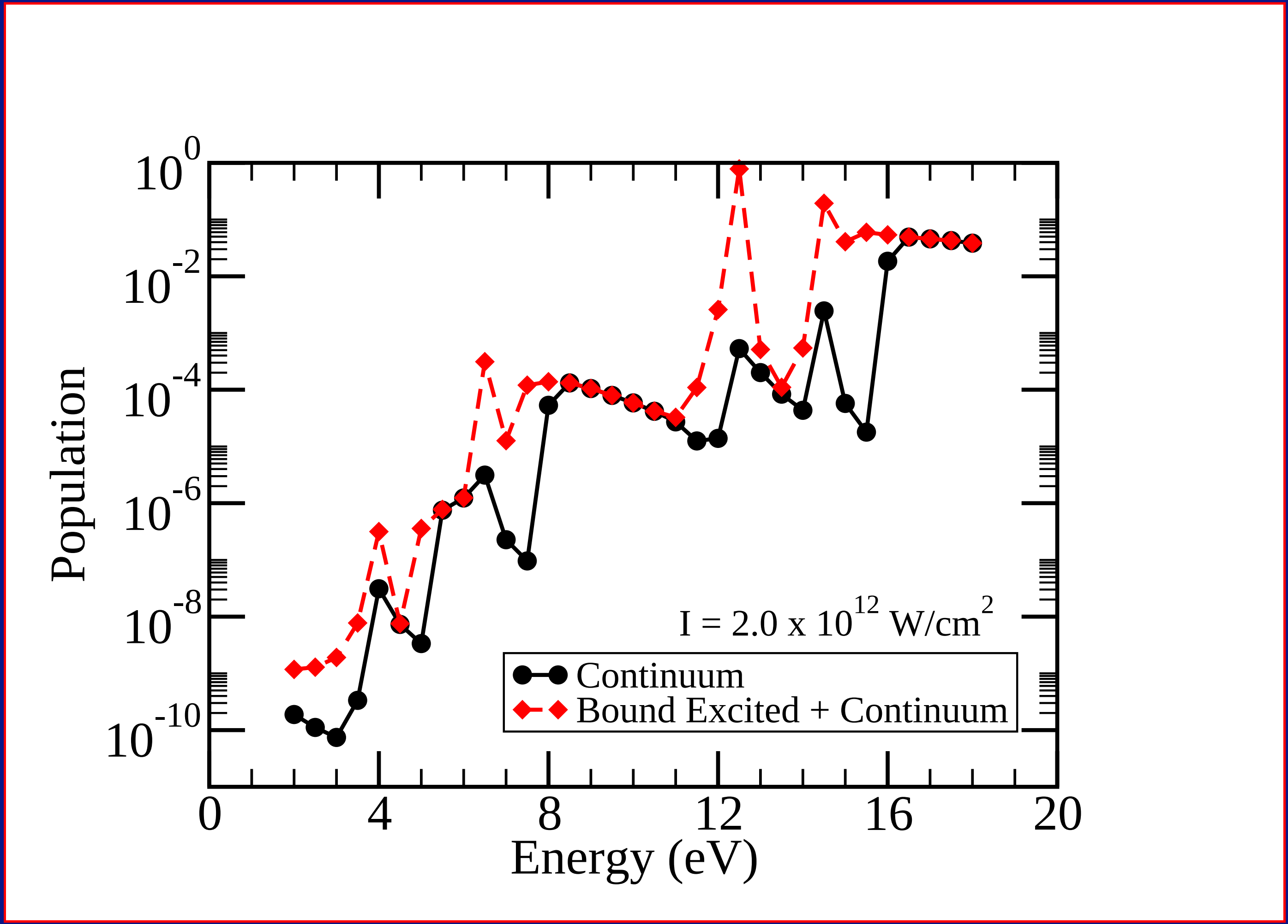
Resonances (REMPI) and thresholds (H_2, $R = 1.4\, a_0$)



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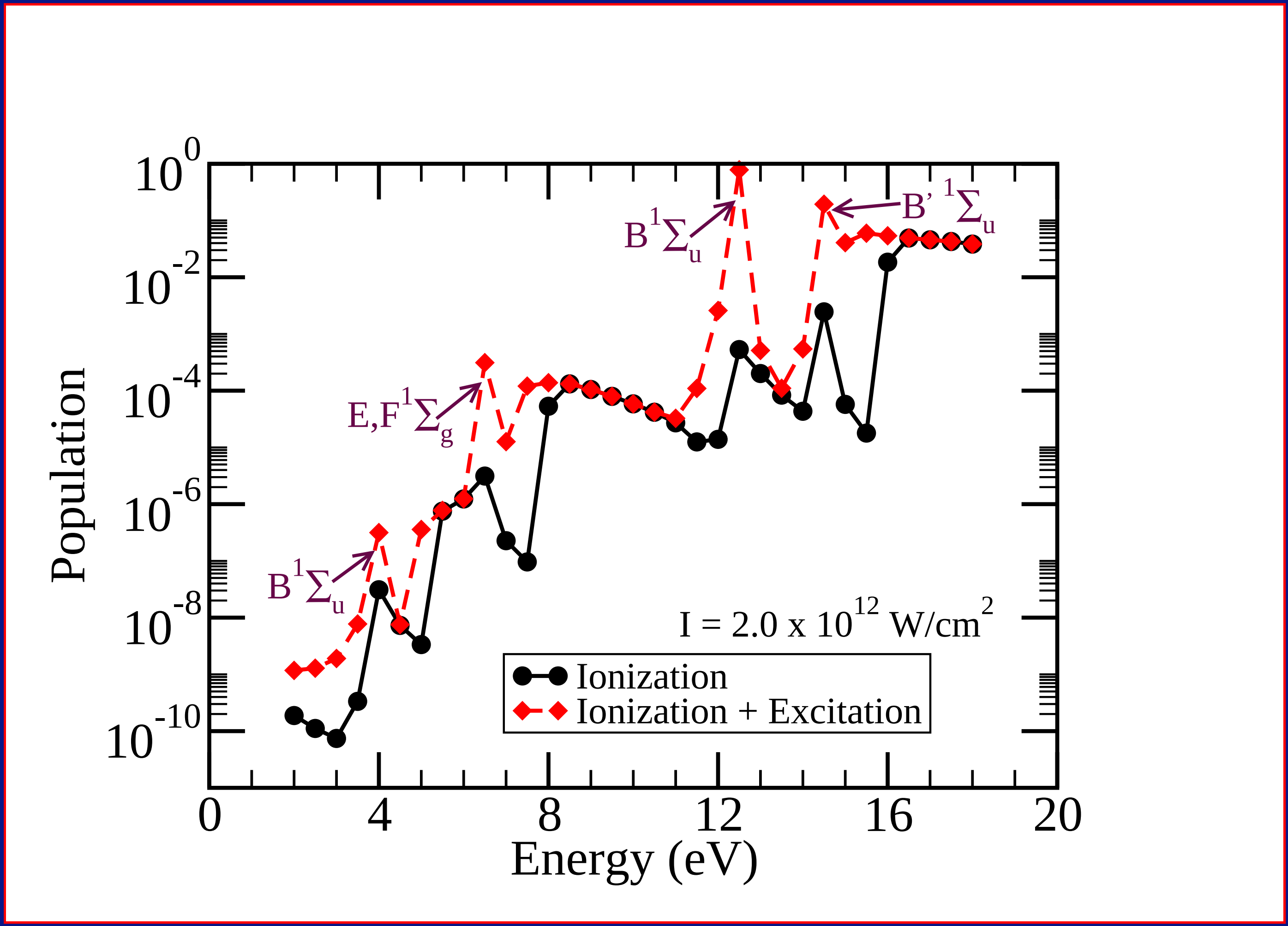

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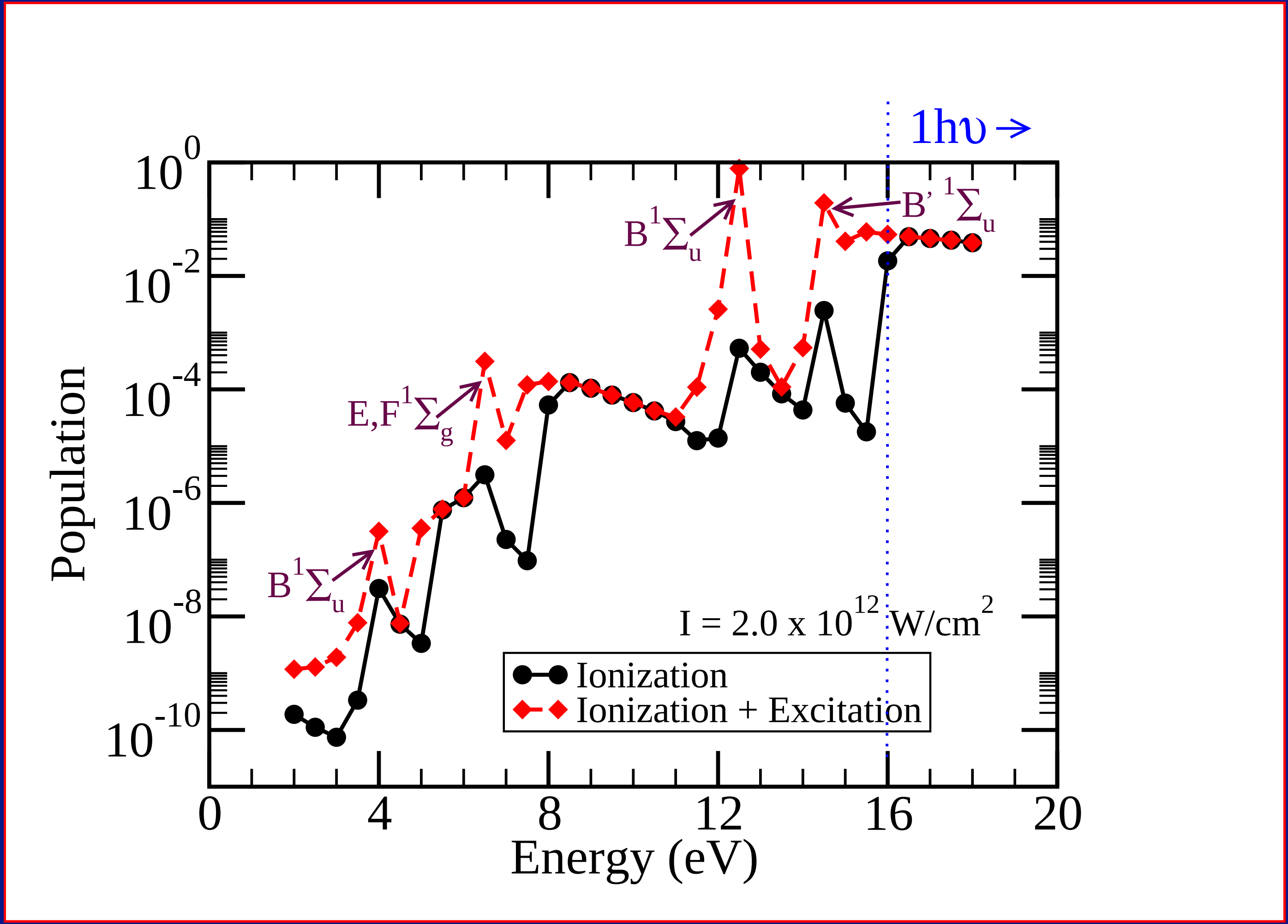

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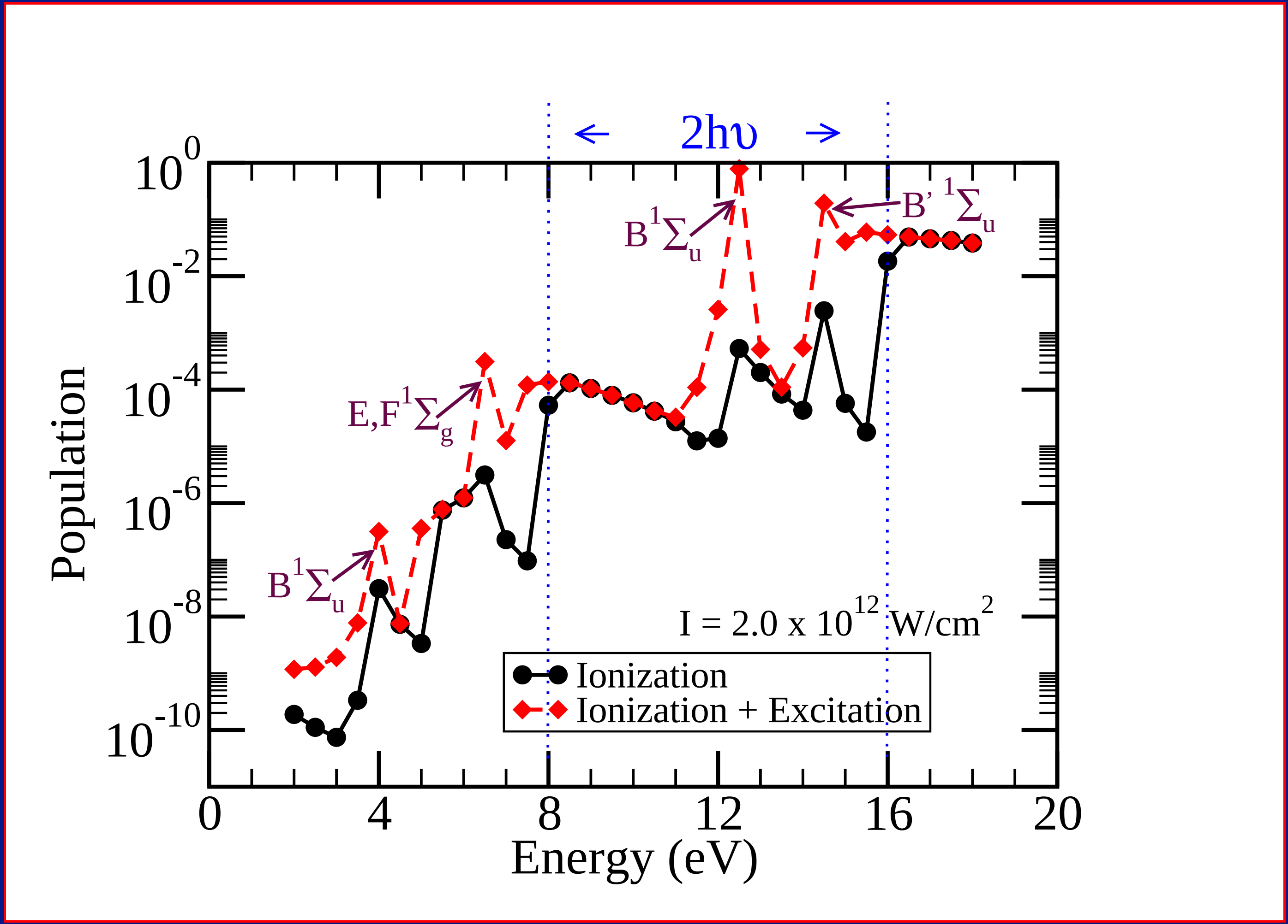

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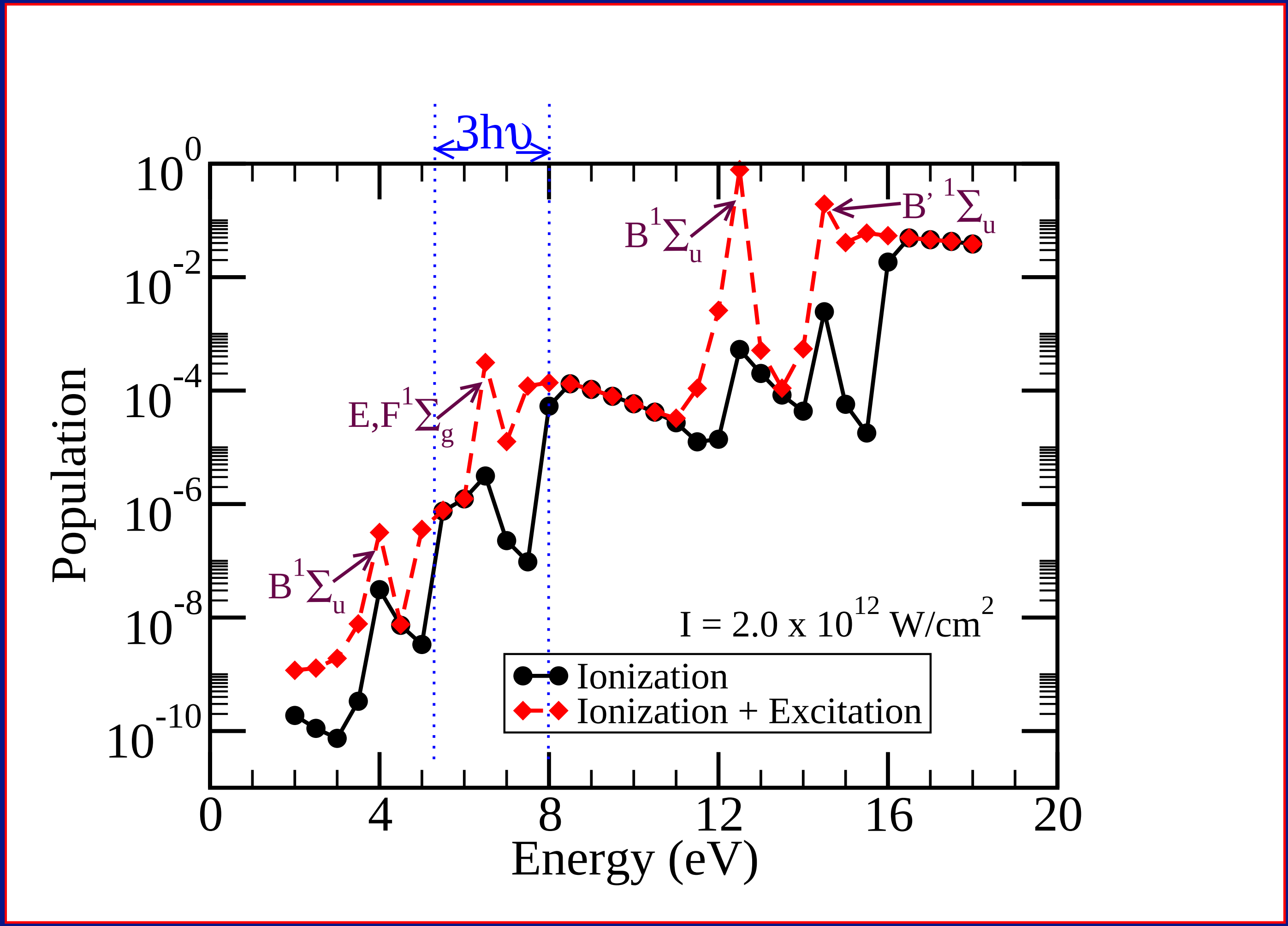
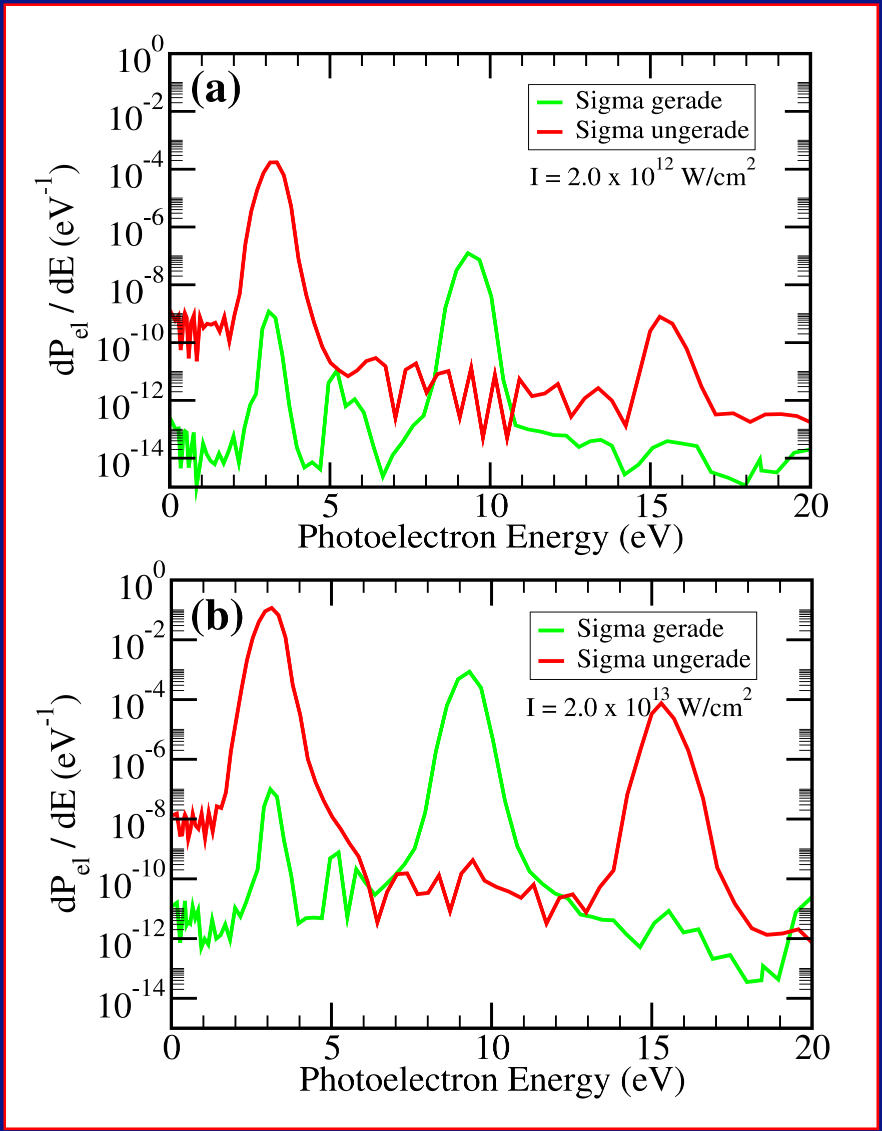


Photoelectron spectra for H_2



LOPT prediction:

only first Σ_u peak exists (3 photon process)!

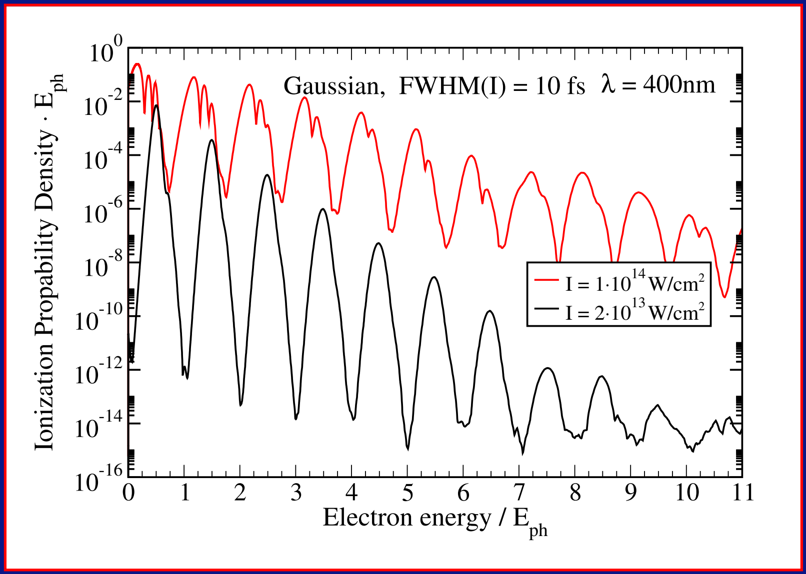
Additional peaks:
above-threshold ionization (ATI)

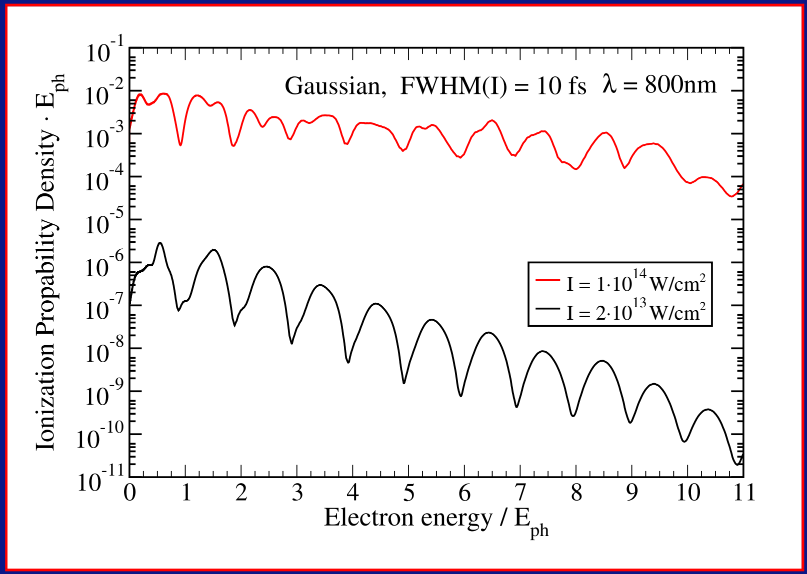
H_2 at $R = 1.40\, a_0$

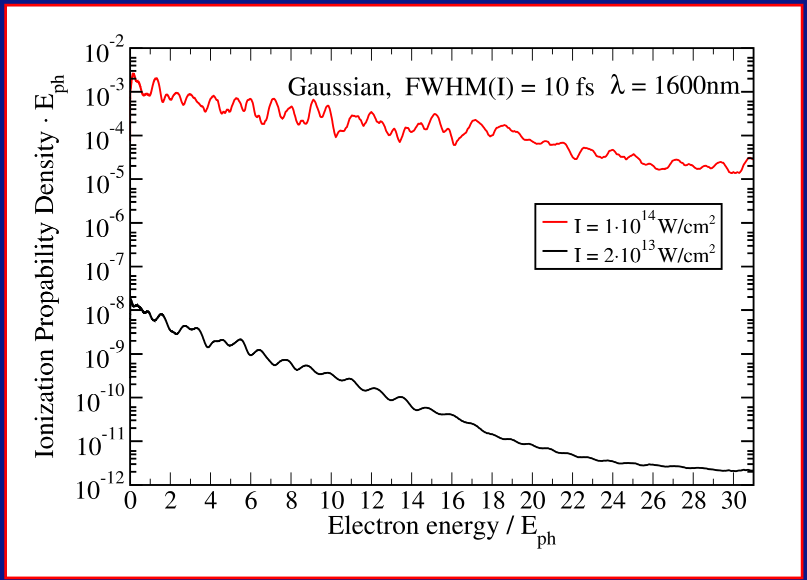
Photon energy: 6.2 eV

Pulse length: 25 cycles ≈ 16.5 fs

Photoelectron spectra (H atom)







H atom:

Ionization potential $I_P = 13.6\,\text{eV}$

400 nm (3.1 eV) $\longrightarrow N_{\text{ph,min}} = 5$

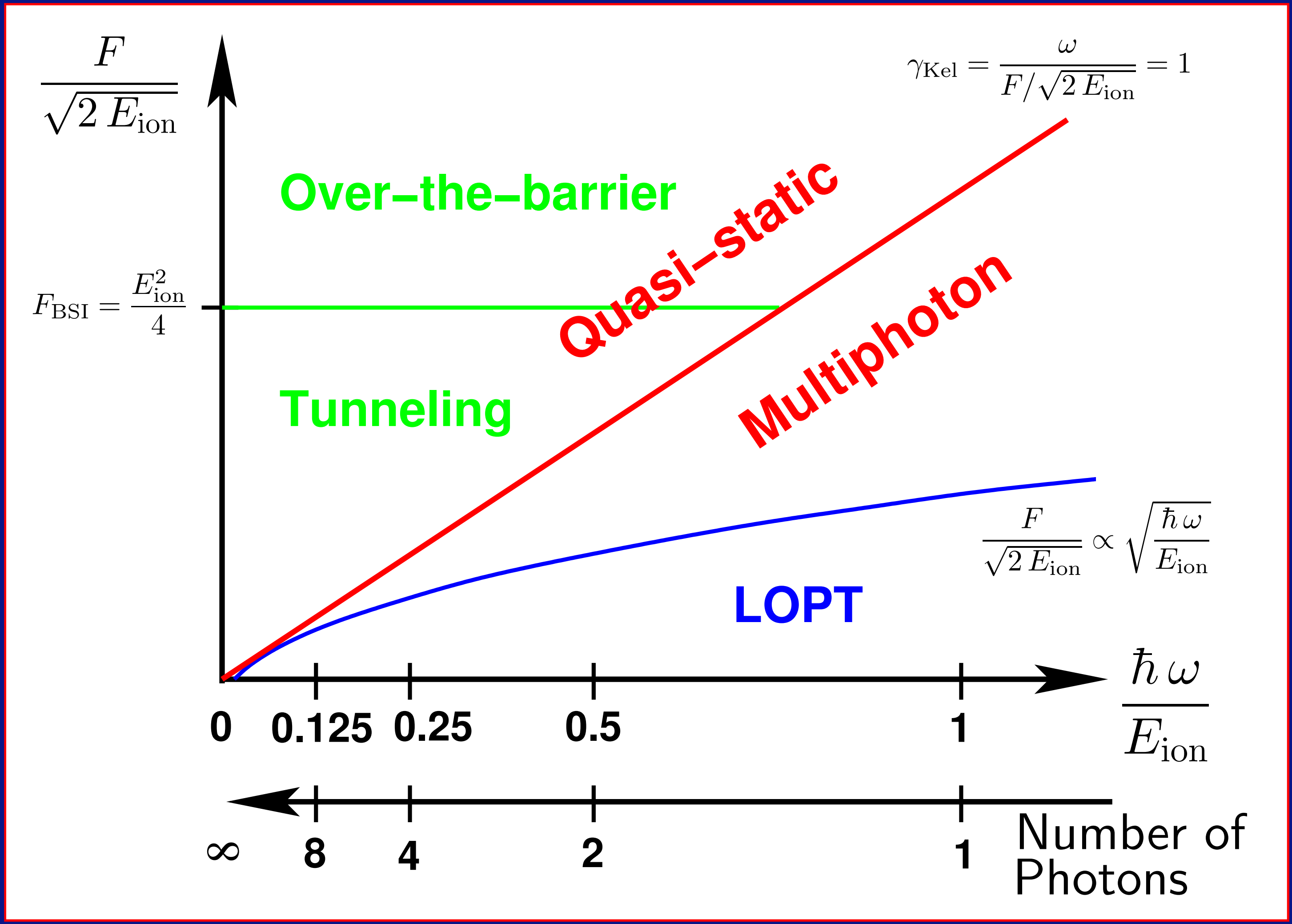
800 nm (1.55 eV) $\longrightarrow N_{\text{ph,min}} = 9$

1600 nm (0.775 eV) $\longrightarrow N_{\text{ph,min}} = 18$

Efficient solution of TDSE for H atom:

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

Validity regimes of approximations (reminder)



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).

$$\text{Ionisation rate} \qquad \Gamma_{\text{ac}} = \frac{1}{T} \int_{-T/2}^{+T/2} \Gamma_{\text{dc}}\left(F(t)\right) \mathrm{d}t$$

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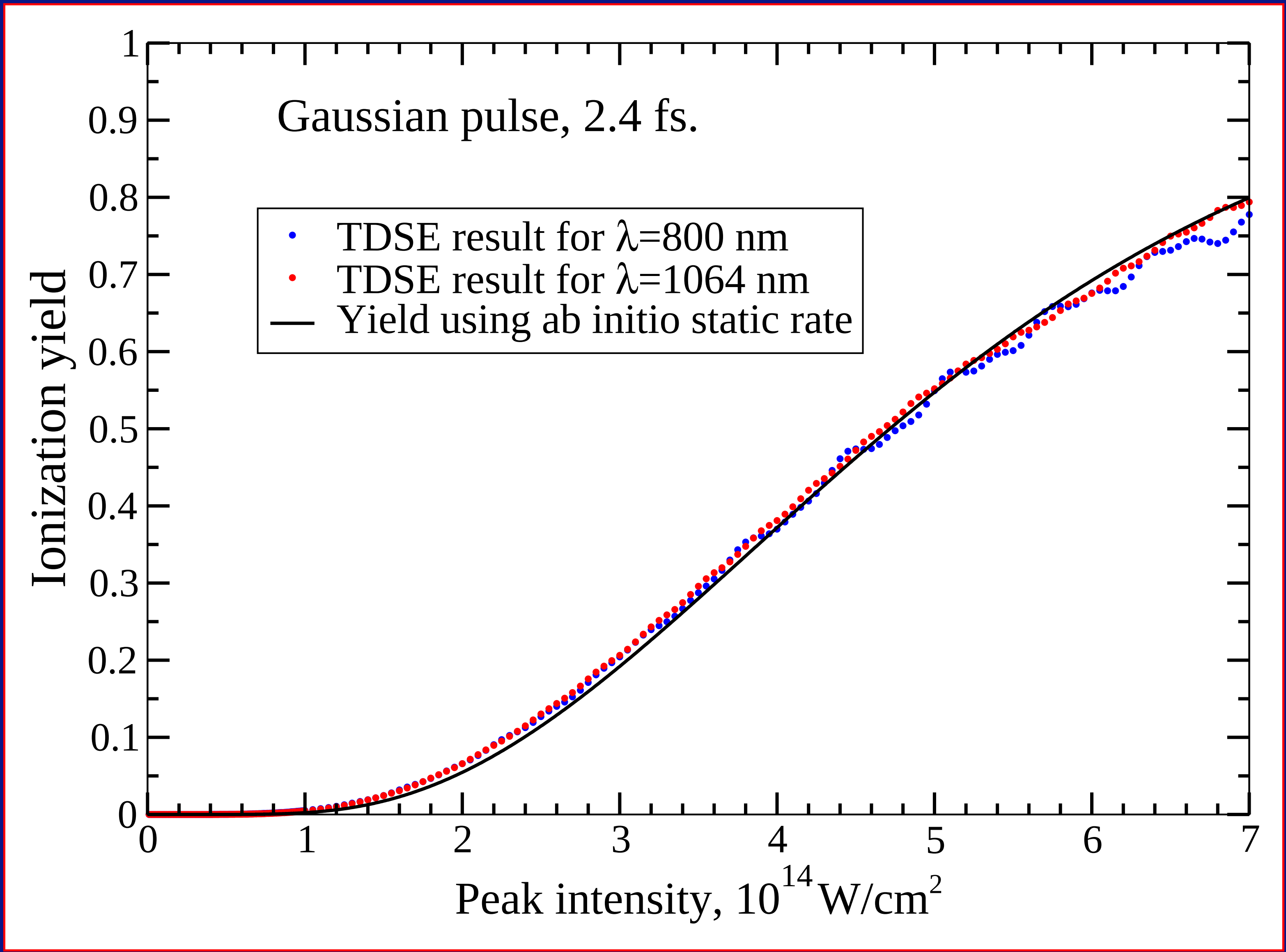
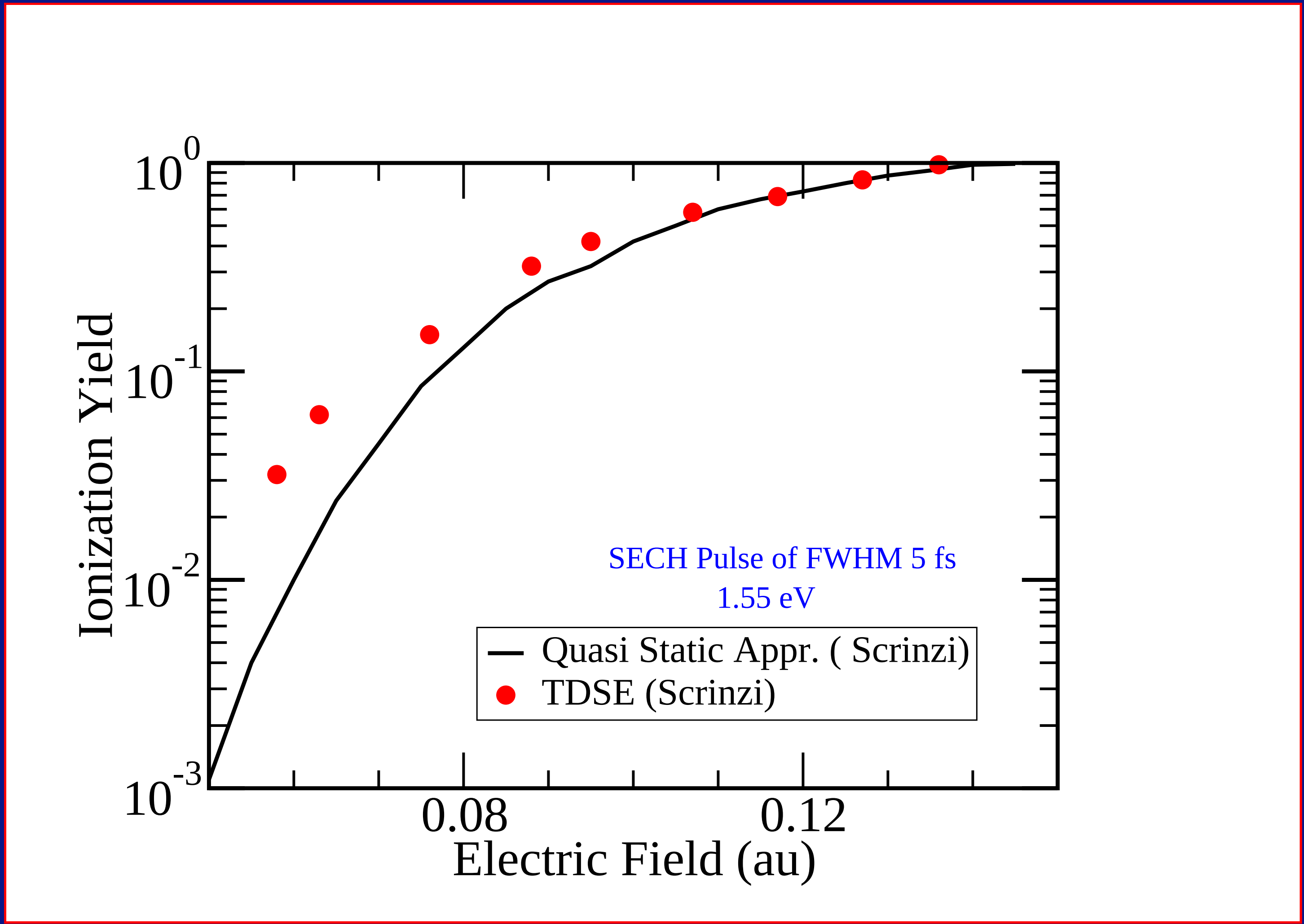
- **Quasistatic regime:** low frequency, high intensity.
- The static rate is (evidently) frequency independent.
- Tunnel regime (lower intensity) vs.
  over-the-barrier ionisation (higher intensity).
- Simple atomic one-electron tunnel models exist for a long time.
  Example: Ammosov-Delone-Krainov (ADK) model.
- Fully correlated 3D calculation of dc rates difficult !!!

  $\longrightarrow$ **so far ab initio dc rates exist only for H_2^+ and H_2!**

Validity of exact QSA for H atom (I)

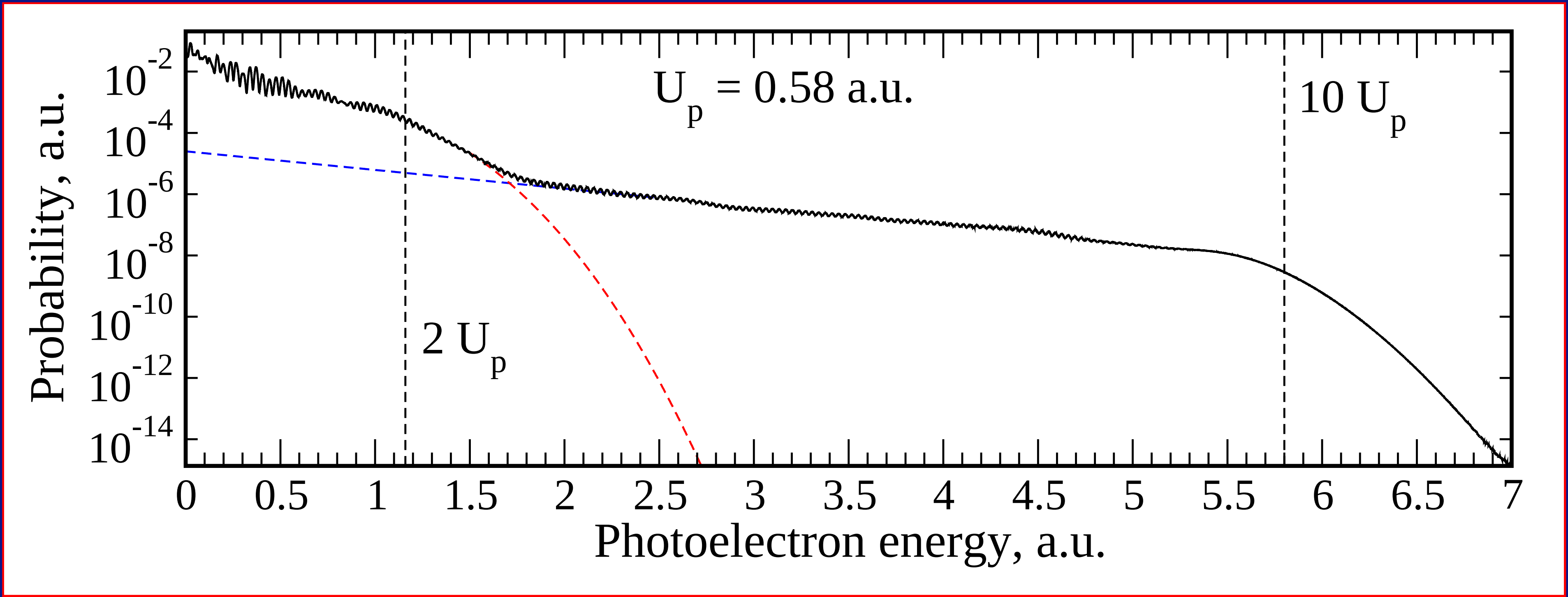


Validity of exact QSA for H atom (II)



Taken from A. Scrinzi, M. Geisler, T. Brabec, *Phys. Rev. Lett.* **83** (1999).

Example electron spectrum (ATI)



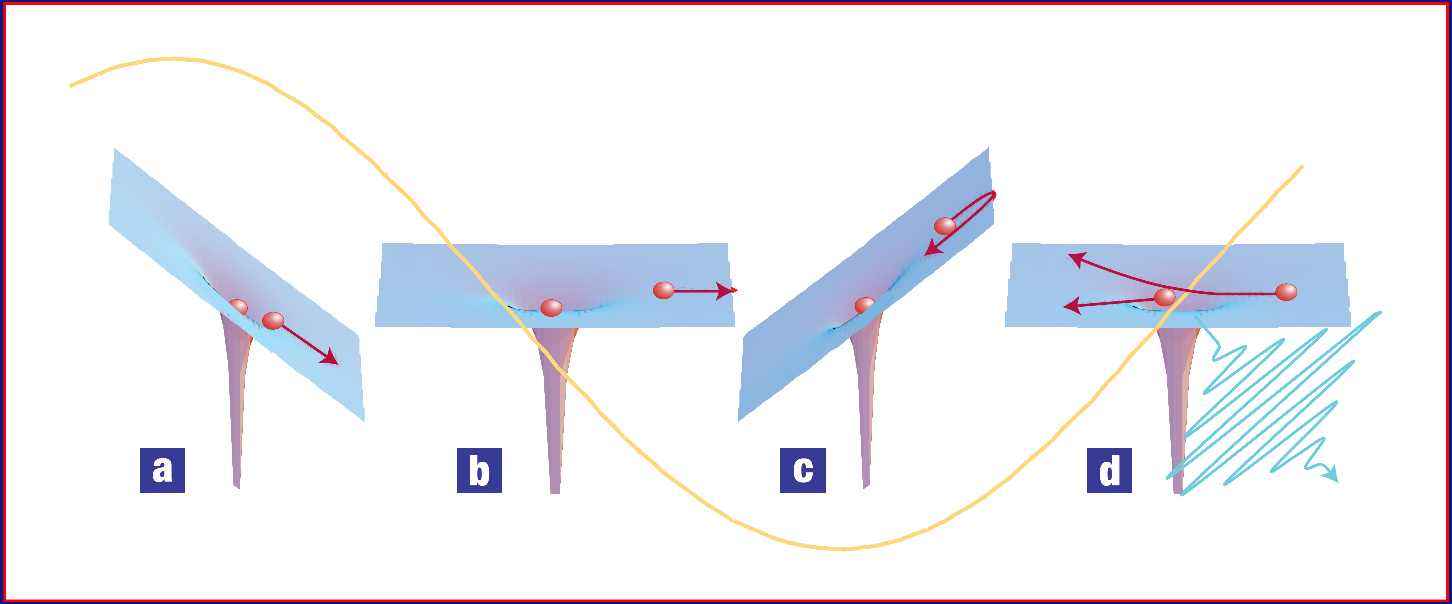
Technical details of the TDSE calculation:

Hydrogen atom

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

Direct electrons: 0 to about 2 times the ponderomotive energy U_p.

Corkum's 3-step model for imaging



from P. B. Corkum and F. Krausz, *Nature Phys.* **3**, 381 (2007)

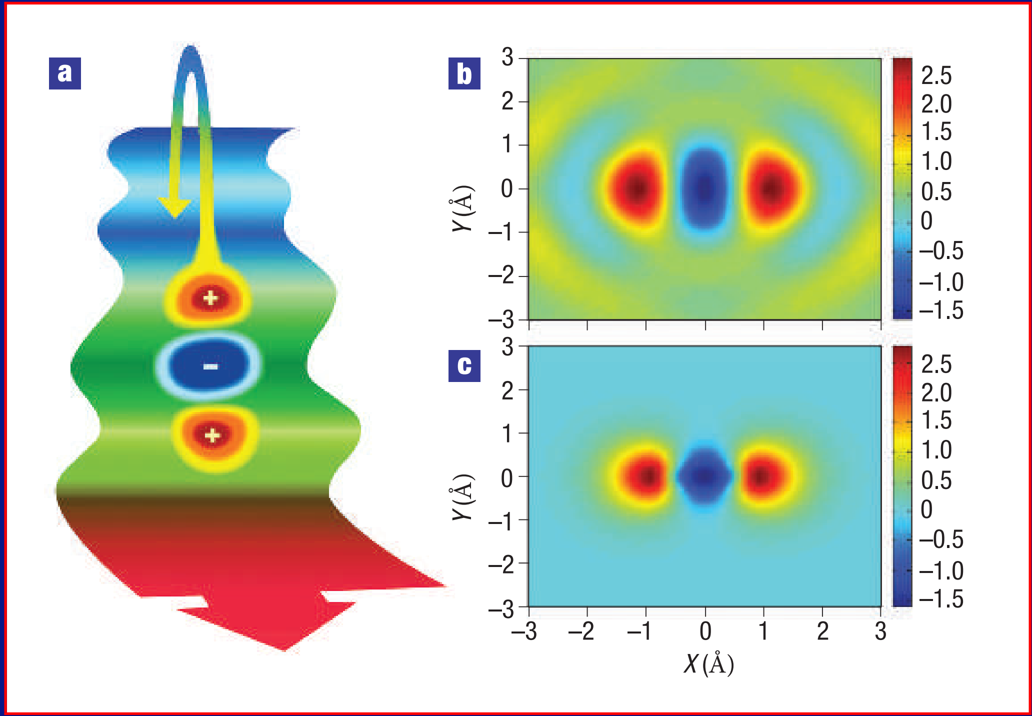
1. Electron escapes through or over the electric-field lowered Coulomb potential (a).
2. Electronic wavepacket moves away until the field direction reverses (b) and is (partly) driven back to its parent ion (c).
3. The returning electron may (d)
   - scatter elastically (electron diffraction)
   - scatter 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)]



graph from P. B. Corkum and F. Krausz, *Nature Phys.* **3**, 381 (2007)

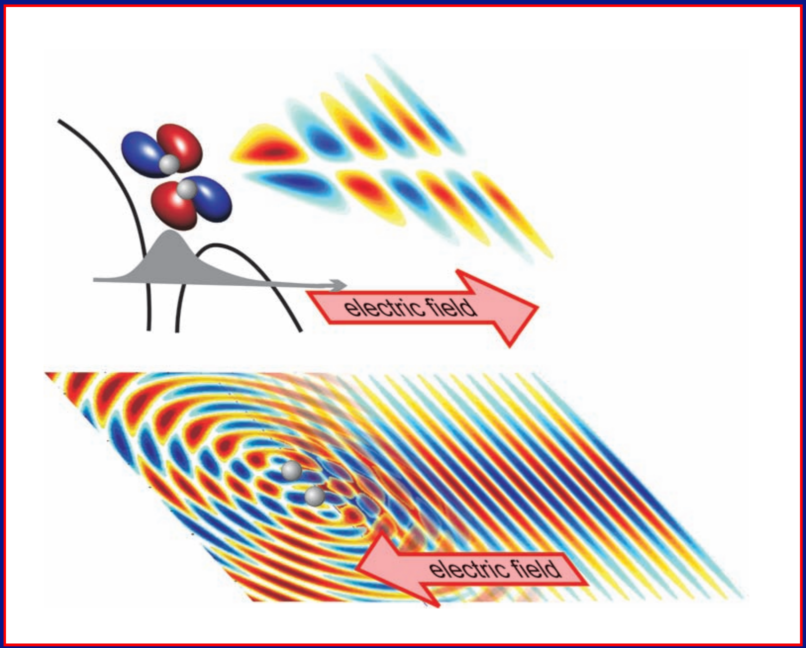
Tomographical reconstruction of the HOMO (highest-occupied molecular orbital) of N_2 using the high-harmonic generation (HHG):

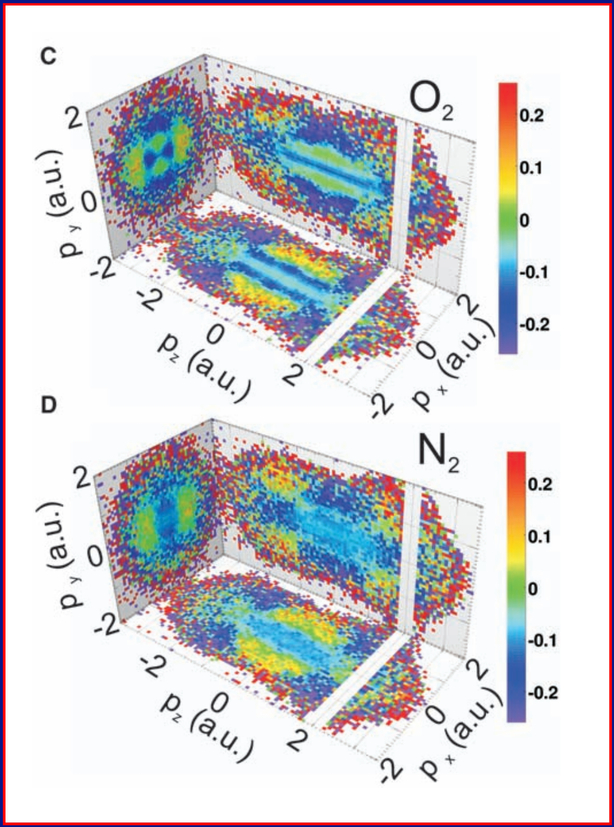
(a) Cartoon of the process: the interference pattern encoded in the HHG changes with electron wavelength and molecular orientation.
(b) Experimentally reconstructed orbital.
(c) Quantum-chemically calculated orbital (reference).

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 N_2 and O_2!