ATOMS AND MOLECULES IN INTENSE ULTRASHORT LASER PULSES: IS THERE A BRIDGE BETWEEN LONG AND SHORT WAVELENGTH REGIMES?

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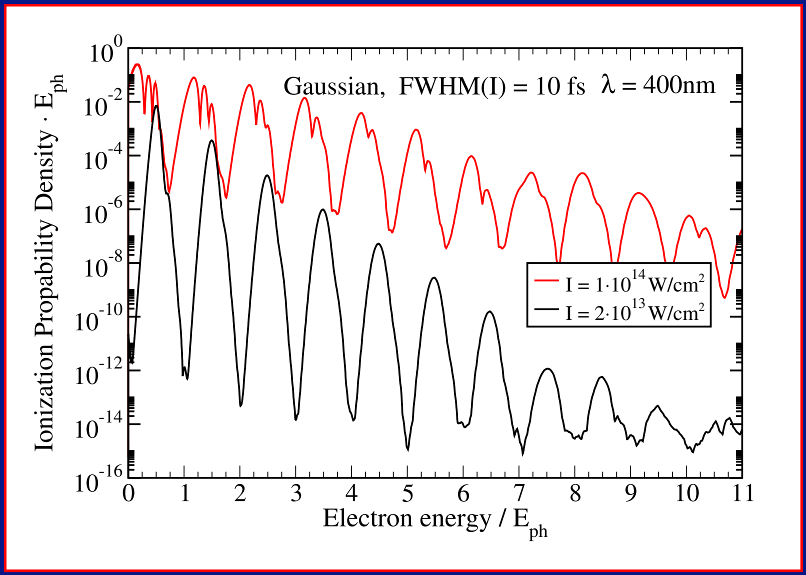
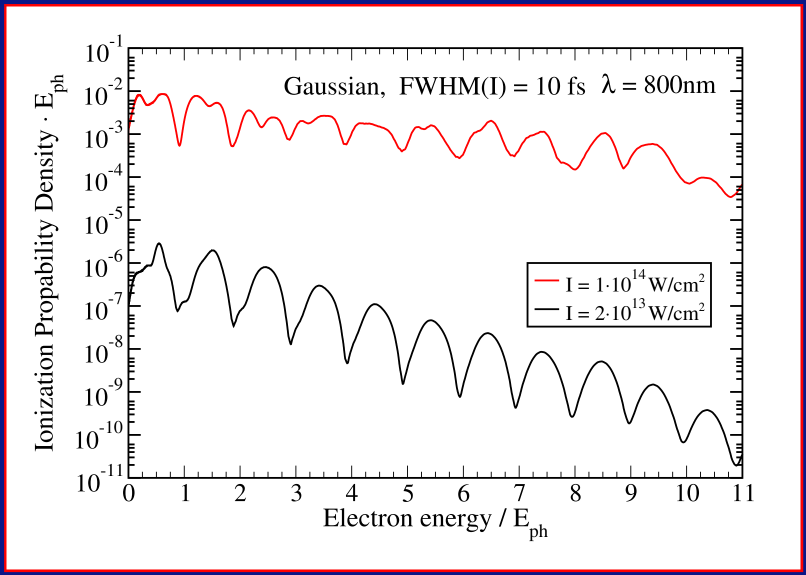
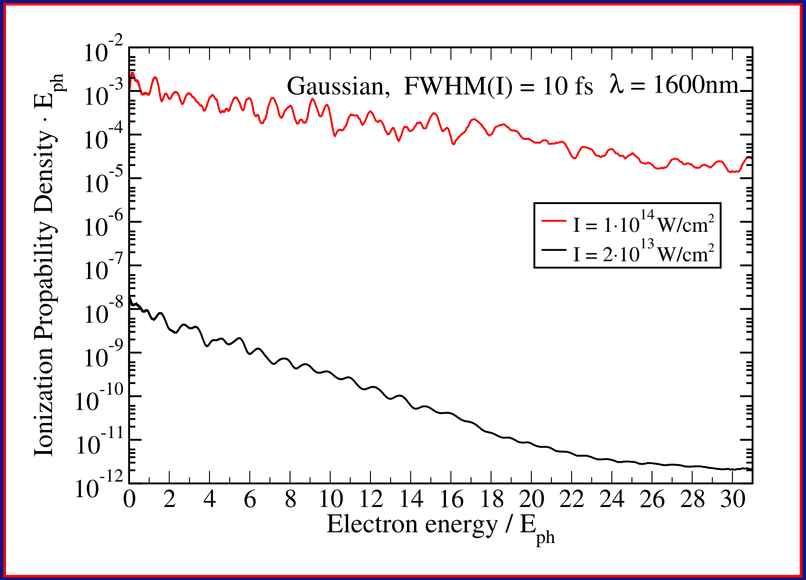
(Kavli-Institute for Theoretical Physics, 23.08.2010)

Quasistatic vs. multiphoton picture: electron spectra of 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

LOPT from helium to molecular hydrogen

J. Phys. B: At. Mol. Opt. Phys. **32** (1999) 5629–5637. Printed in the UK   PII: S0953-4075(99)07138-2

Theoretical two-, three- and four-photon ionization cross sections of helium in the XUV range

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‡ Foundation for Research and Technology-Hellas, Institute of Electronic Structure and Laser, PO Box 1527, Heraklion 71110, Crete, Greece
and
Department of Physics, University of Crete, Greece

J. Phys. B: At. Mol. Opt. Phys. **33** (2000) 2791–2807. Printed in the UK   PII: S0953-4075(00)50485-4

Effect of vibration and internuclear axis orientation on multiphoton ionization of H_2^+

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Department of Physics, University of Crete, Greece

INSTITUTE OF PHYSICS PUBLISHING   JOURNAL OF PHYSICS B: ATOMIC, MOLECULAR AND OPTICAL PHYSICS

J. Phys. B: At. Mol. Opt. Phys. **35** (2002) 1909–1928   PII: S0953-4075(02)33213-9

Multiphoton ionization of the hydrogen molecule H_2

Amalia Apalategui and Alejandro Saenz

Fachbereich Chemie, Universität Konstanz, Fach M 721, D-78457 Konstanz, Germany

$$\int_0^{\pi} |D_{fi}^{(2)}(\vartheta)|^2 \sin\vartheta \, \mathrm{d}\vartheta$$

$$= \frac{3}{15}\left|\sum_m \frac{\langle\Psi_i^{\Sigma_g}|\sum_s r_s Y_1^0(\Omega_s)|\Psi_m^{\Sigma_u}\rangle\langle\Psi_m^{\Sigma_u}|\sum_s r_s Y_1^0(\Omega_s)|\Psi_f^{\Sigma_g}\rangle}{(E_m - E_i - \omega)}\right|^2 \quad (7a)$$

$$+ \frac{8}{15}\left|\sum_m \frac{\langle\Psi_i^{\Sigma_g}|\sum_s r_s Y_1^{-1}(\Omega_s)|\Psi_m^{\Pi_u^+}\rangle\langle\Psi_m^{\Pi_u^+}|\sum_s r_s Y_1^{+1}(\Omega_s)|\Psi_f^{\Sigma_g}\rangle}{(E_m - E_i - \omega)}\right|^2 \quad (7b)$$

$$+ \frac{4}{15}\left|\sum_m \frac{\langle\Psi_i^{\Sigma_g}|\sum_s r_s Y_1^{-1}(\Omega_s)|\Psi_m^{\Pi_u^+}\rangle\langle\Psi_m^{\Pi_u^+}|\sum_s r_s Y_1^{-1}(\Omega_s)\Psi_f^{\Delta_g^+}\rangle}{(E_m - E_i - \omega)}\right|^2 \quad (7c)$$

$$+ \frac{2}{15}\left|\sum_m \frac{\langle\Psi_i^{\Sigma_g}|\sum_s r_s Y_1^0(\Omega_s)|\Psi_m^{\Sigma_u}\rangle\langle\Psi_m^{\Sigma_u}|\sum_s r_s Y_1^{-1}(\Omega_s)|\Psi_f^{\Pi_g^+}\rangle}{(E_m - E_i - \omega)}\right|^2 \quad (7d)$$

$$+ \frac{2}{15}\left|\sum_m \frac{\langle\Psi_g^{\Sigma_g}|\sum_s r_s Y_1^{-1}(\Omega_s)|\Psi_m^{\Pi_u^+}\rangle\langle\Psi_m^{\Pi_u^+}|\sum_s r_s Y_1^0(\Omega_s)|\Psi_f^{\Pi_g^+}\rangle}{(E_m - E_i - \omega)}\right|^2 \quad (7e)$$

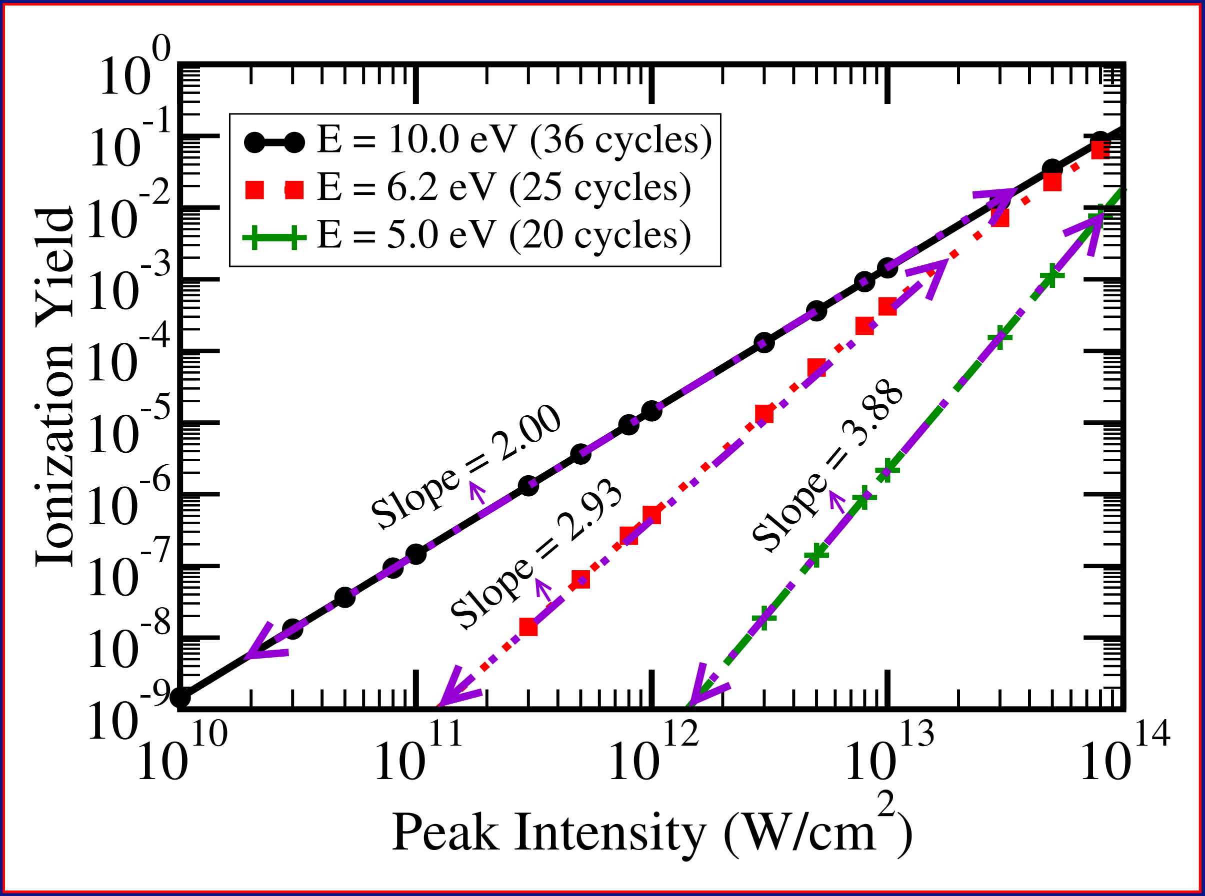
$$- \frac{4}{15}\,\mathrm{Re}\left\{\sum_m \sum_n \frac{\langle\Psi_i^{\Sigma_g}|\sum_s r_s Y_1^{-1}(\Omega_s)|\Psi_m^{\Pi_u^+}\rangle\langle\Psi_m^{\Pi_u^+}|\sum_s r_s Y_1^0(\Omega_s)|\Psi_f^{\Pi_g^+}\rangle}{(E_m - E_i - \omega)}\right.$$

$$\left. \times \frac{\langle\Psi_f^{\Pi_g^+}|\sum_s r_s Y_1^{+1}(\Omega_s)|\Psi_n^{\Sigma_u}\rangle\langle\Psi_n^{\Sigma_u}|\sum_s r_s Y_1^0(\Omega_s)|\Psi_i^{\Sigma_g}\rangle}{(E_n - E_i - \omega)}\right\} \quad (7f)$$

$$- \frac{4}{15}\,\mathrm{Re}\left\{\sum_m \sum_n \frac{\langle\Psi_i^{\Sigma_g}|\sum_s r_s Y_1^{-1}(\Omega_s)|\Psi_m^{\Pi_u^+}\rangle\langle\Psi_m^{\Pi_u^+}|\sum_s r_s Y_1^{+1}(\Omega_s)|\Psi_f^{\Sigma_g}\rangle}{(E_m - E_i - \omega)}\right.$$

$$\left. \times \frac{\langle\Psi_f^{\Sigma_g}|\sum_s r_s Y_1^0(\Omega_s)|\Psi_n^{\Sigma_u}\rangle\langle\Psi_n^{\Sigma_u}|\sum_s r_s Y_1^0(\Omega_s)|\Psi_i^{\Sigma_g}\rangle}{(E_n - E_i - \omega)}\right\} \quad (7g)$$

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

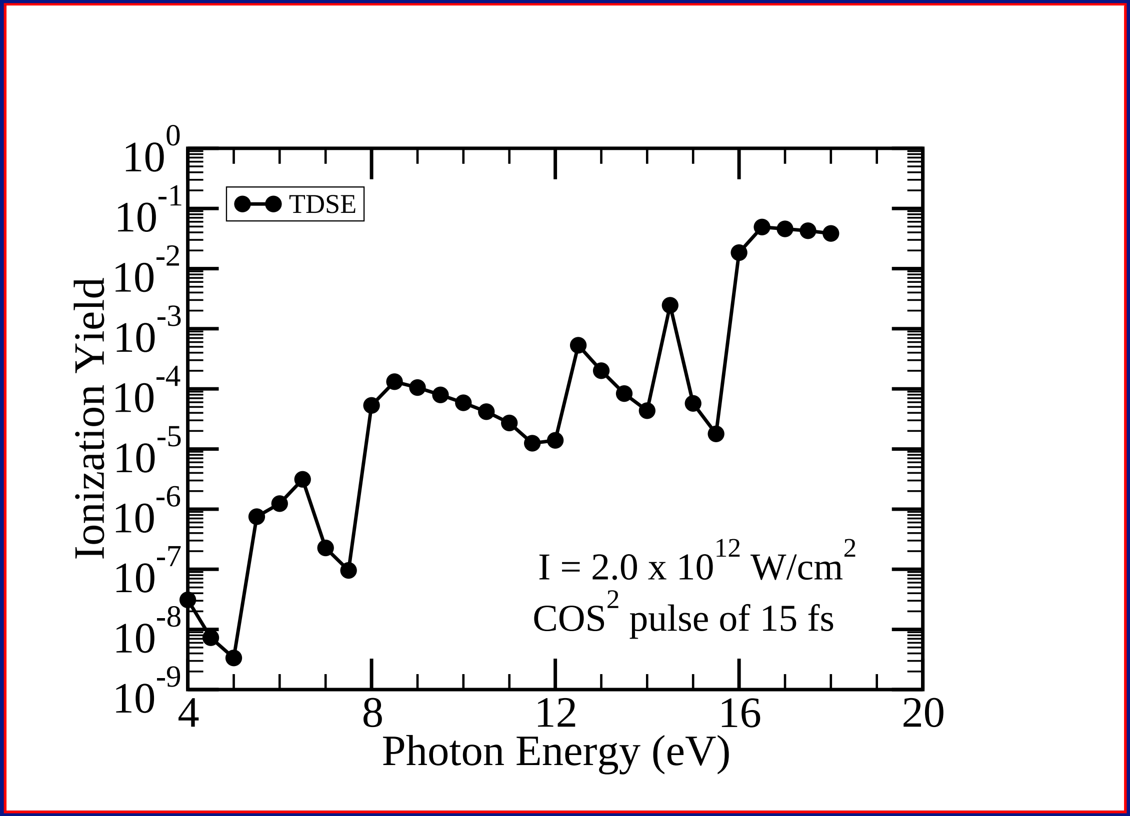


$$\Gamma^{(N)} = \sigma^{(N)} \left(\frac{I}{\hbar\omega}\right)^N$$

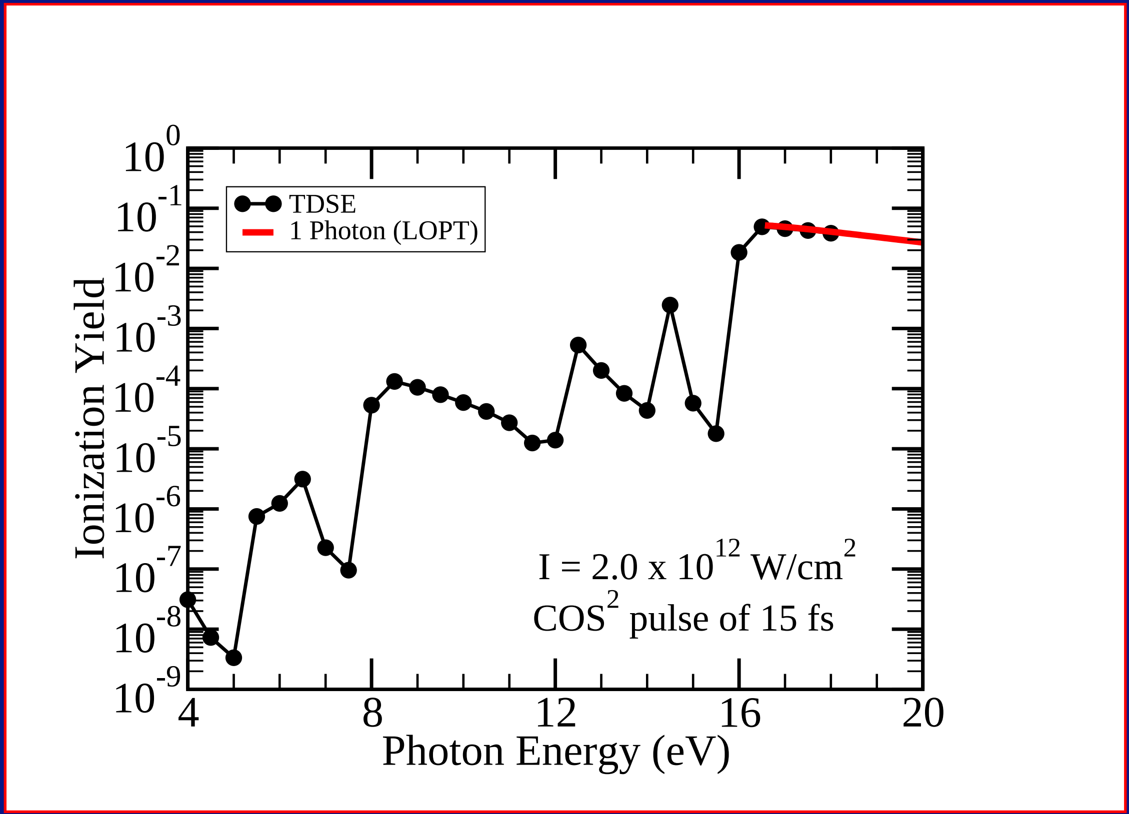
- N: Number of photons
- $\Gamma^{(N)}$: Ionization rate
- $\sigma^{(N)}$: N-photon ionization cross-section
- I: Intensity
- $\hbar\omega$: Photon energy

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

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



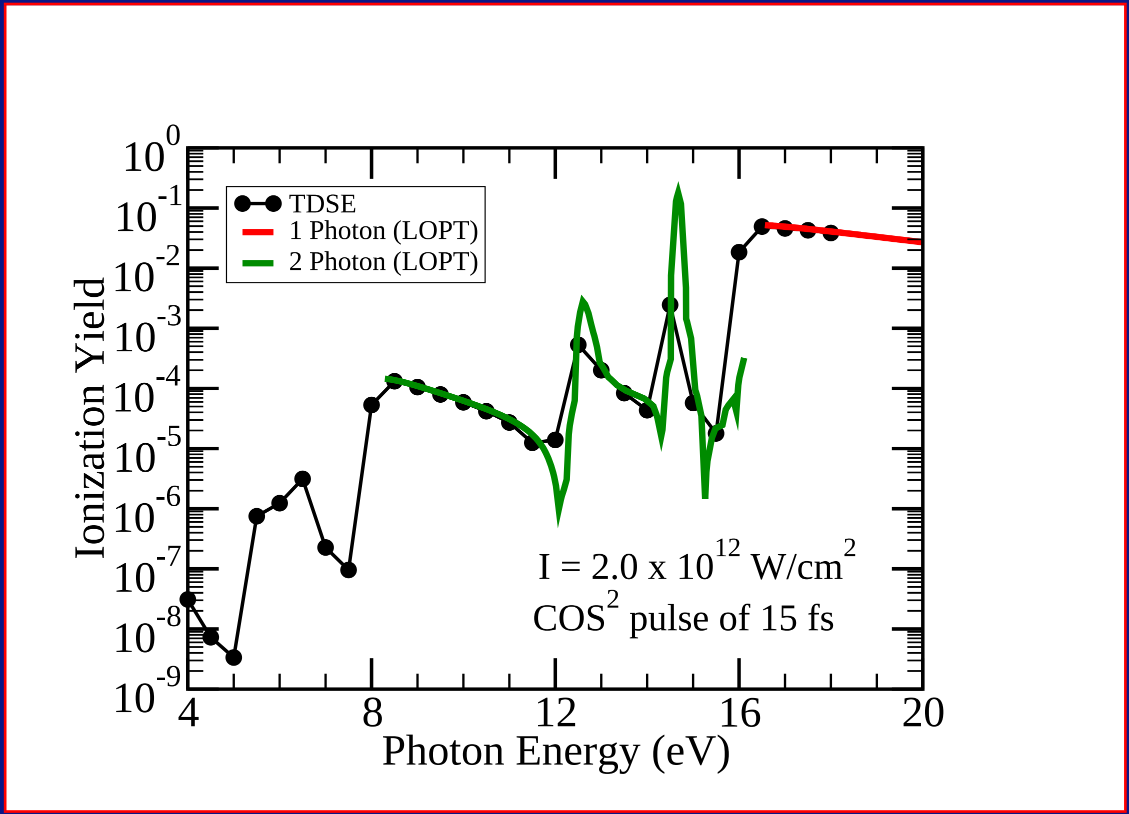
LOPT regime for H_2: frequency scan ($R = 1.4\,a_0$)



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

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

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



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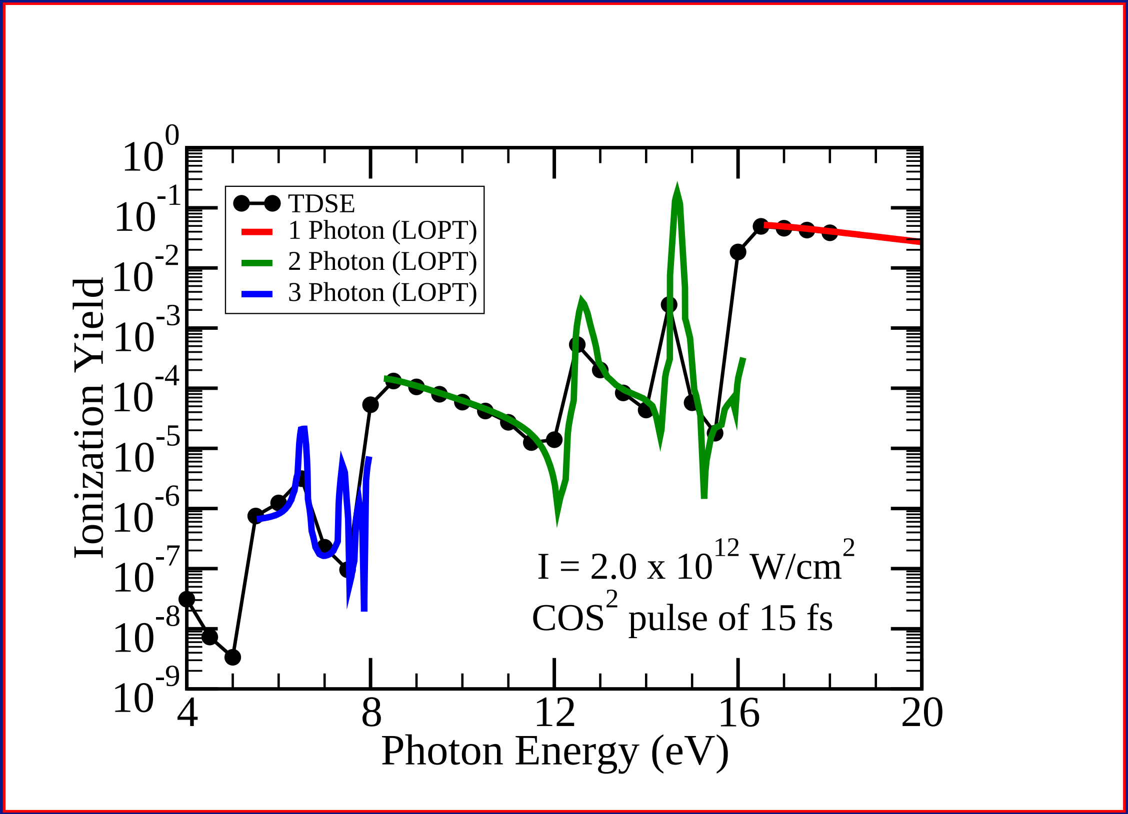
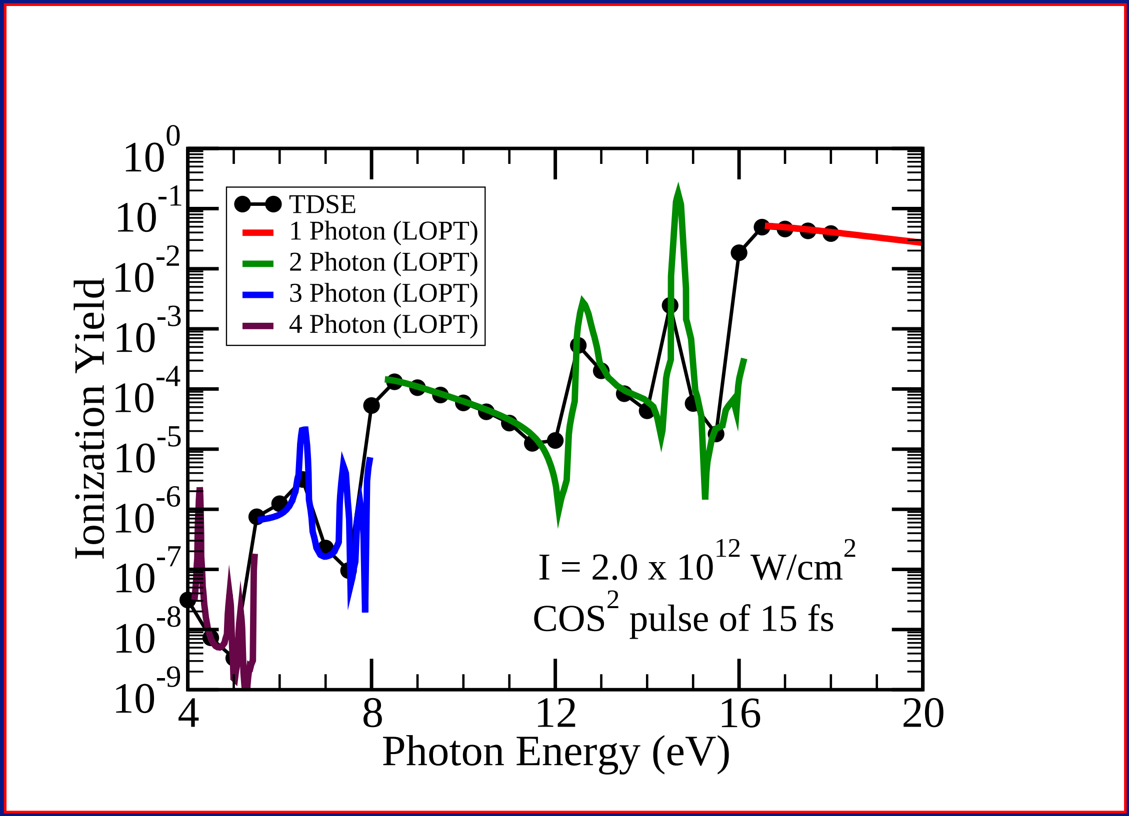
LOPT regime for H_2: frequency scan $(R = 1.4\, a_0)$



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

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

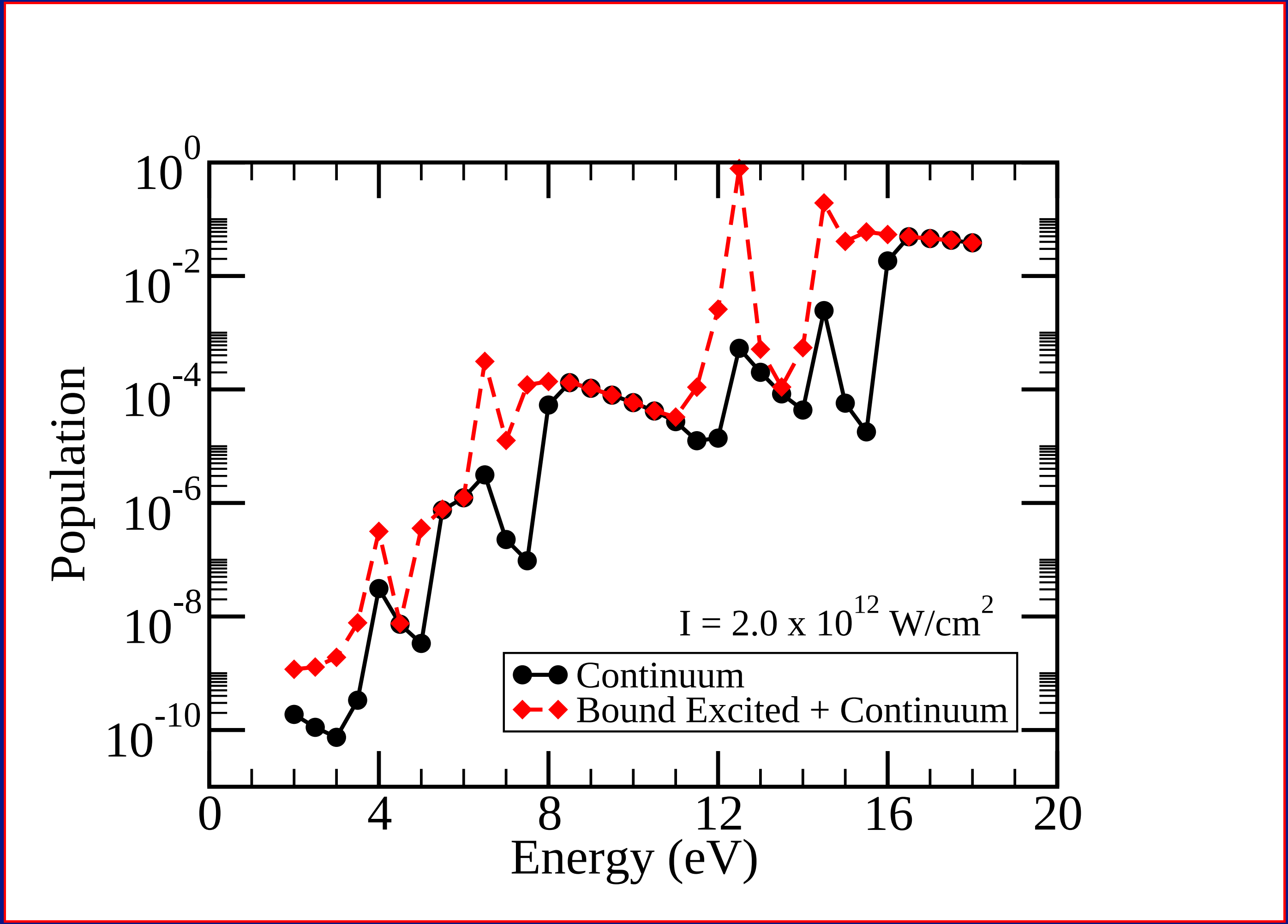
LOPT regime for H_2: frequency scan $(R = 1.4\,a_0)$



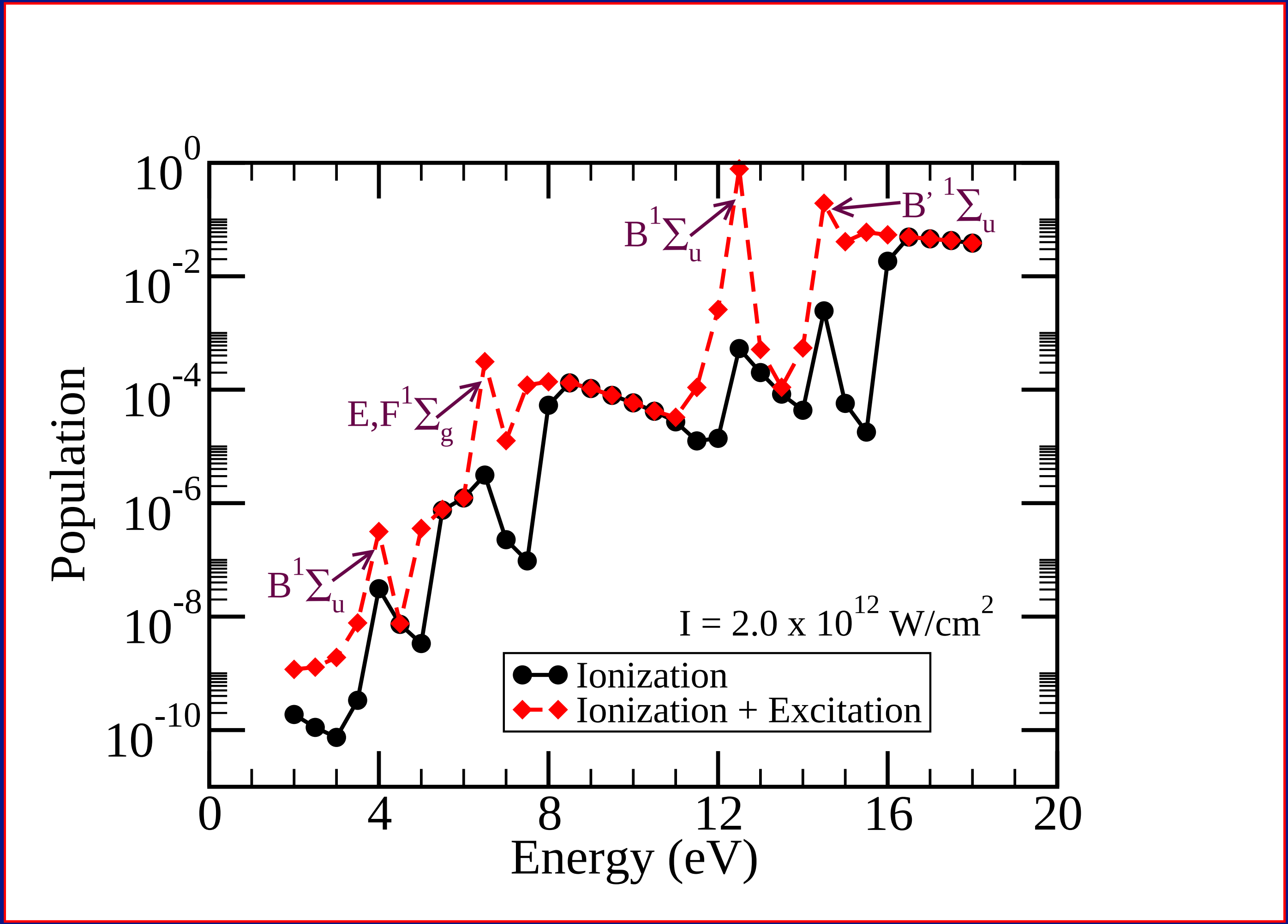
LOPT: Ionization yield $P_{\text{ion}} = \int \Gamma^{(N)}\,\mathrm{d}t$

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

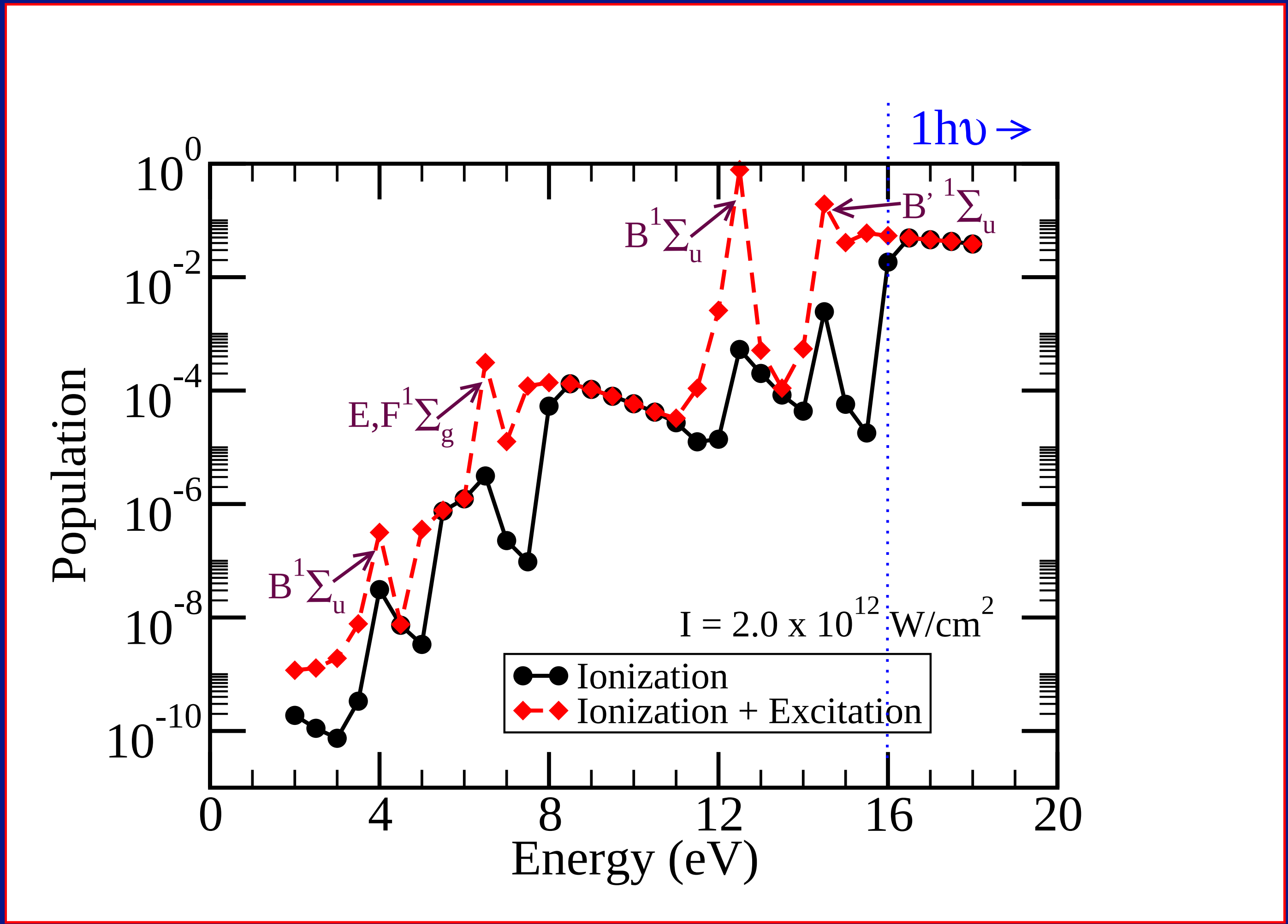
Resonances (REMPI) and thresholds (H_2, $R = 1.4\, a_0$)



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



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



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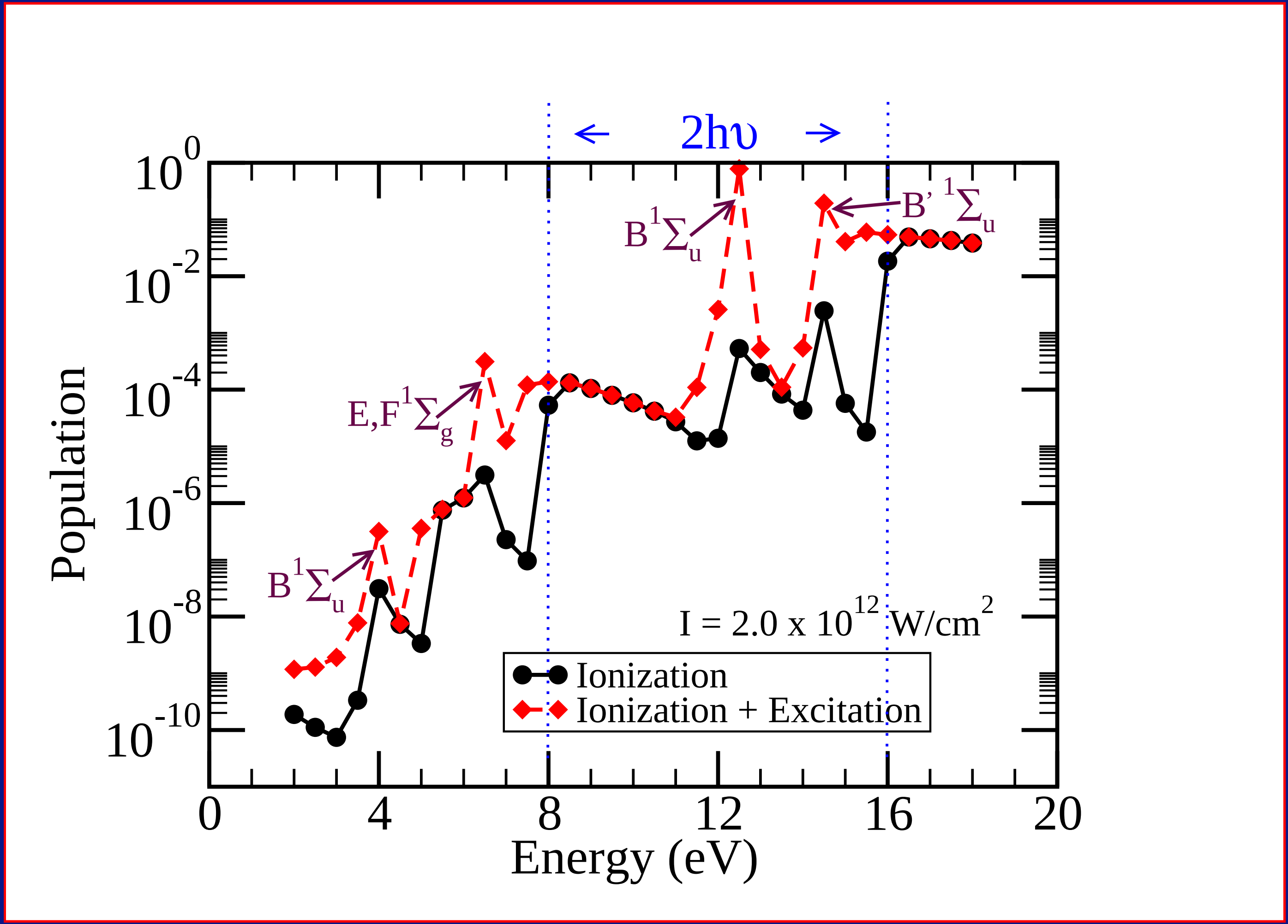

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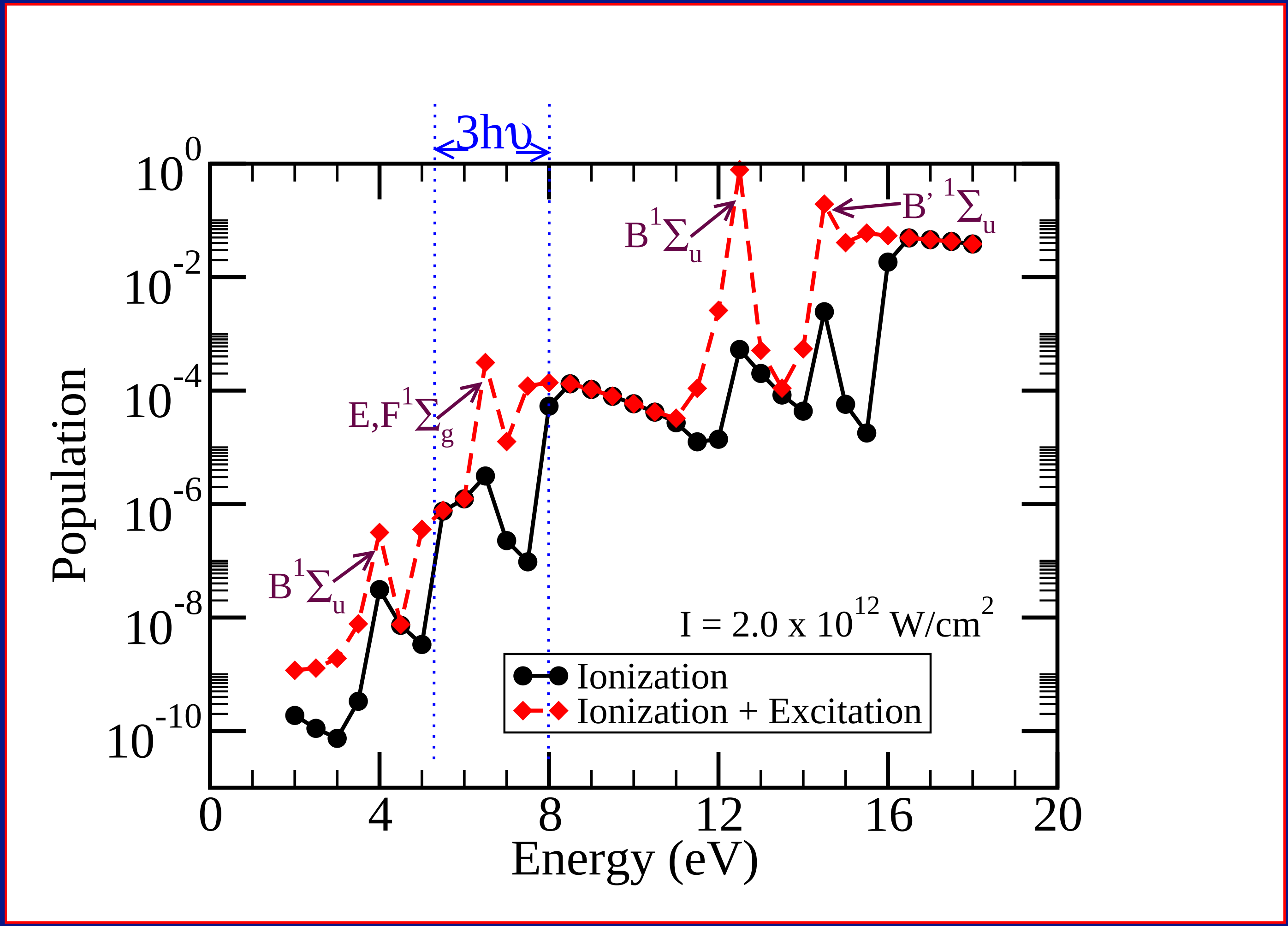
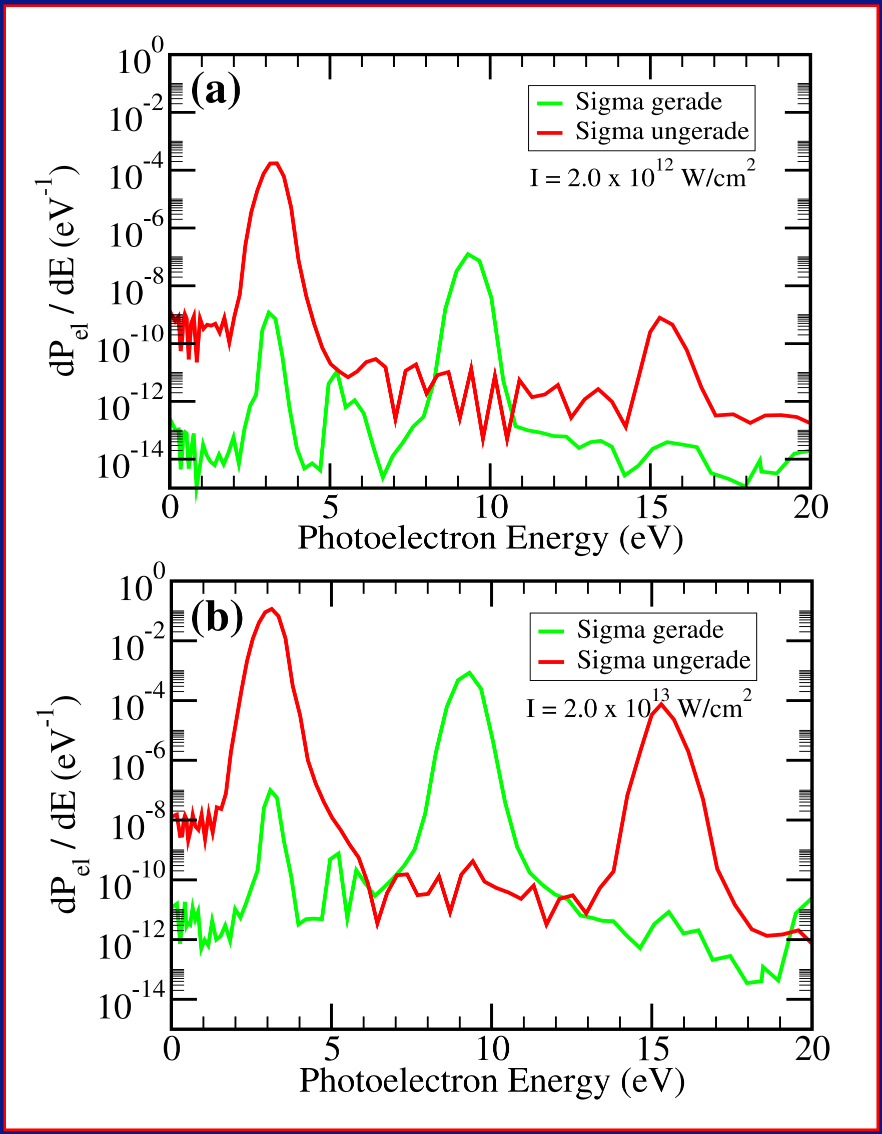


Photoelectron spectra for H_2



LOPT prediction:

only first Σ_u peak exists (3 photon process)!

Additional peaks:

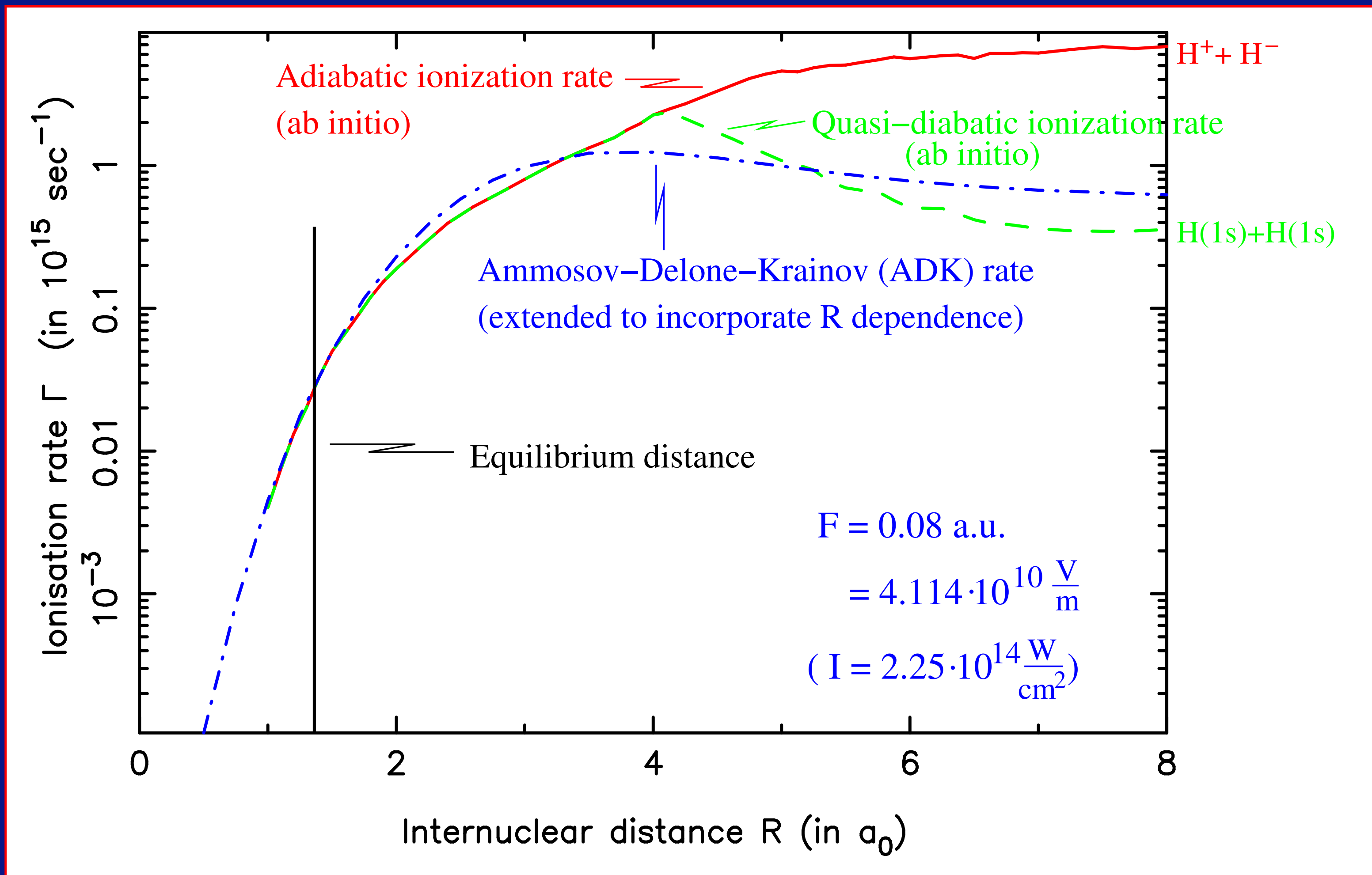
above-threshold ionization (ATI)

H_2 at $R = 1.40\,a_0$

Photon energy: 6.2 eV

Pulse length: 25 cycles ≈ 16.5 fs

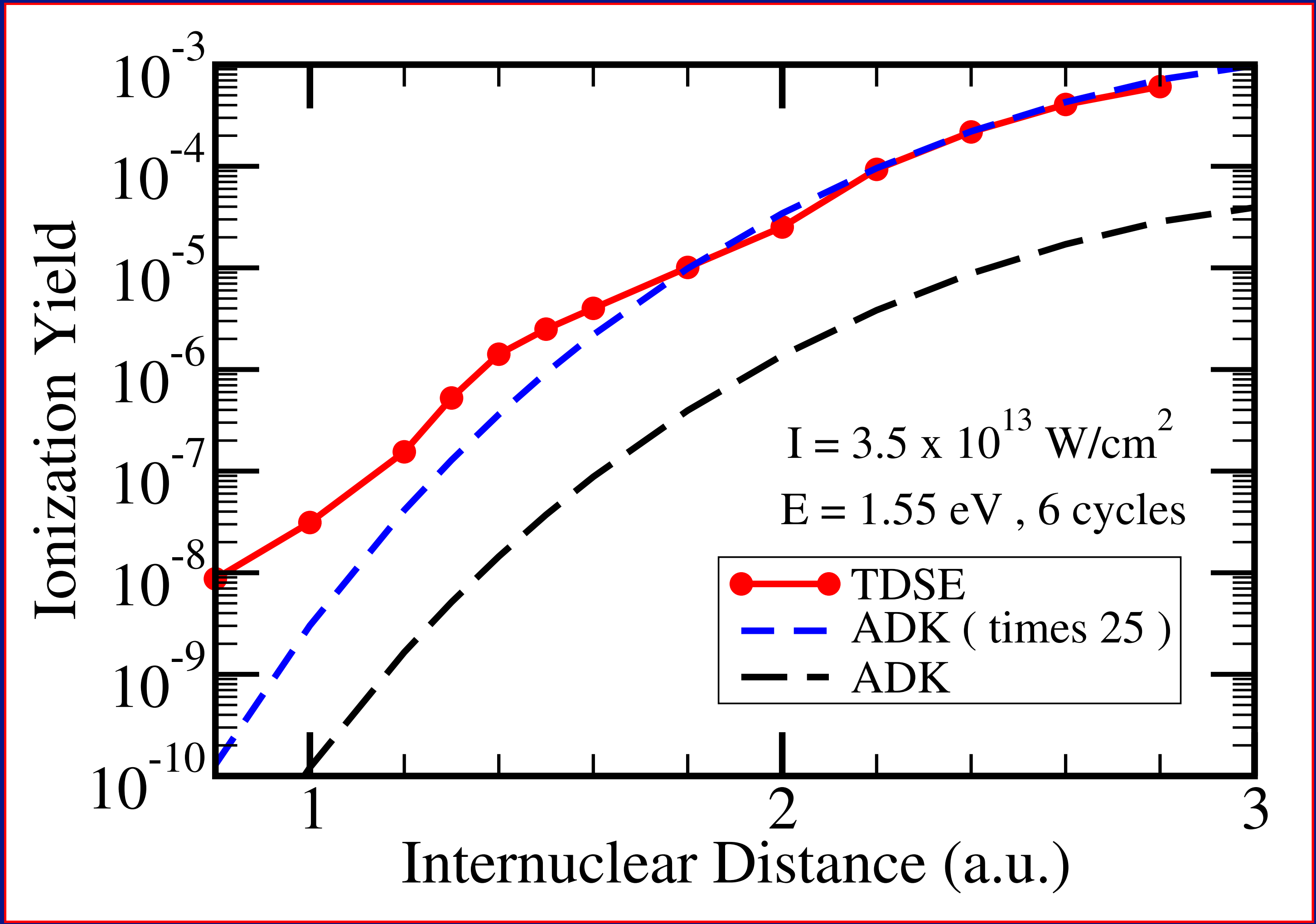
R-dependent ab initio dc ionization rate for H_2



Ab initio calculation (dc field) and experiment confirms: No FC distribution for H_2.

[A. Saenz, *Phys. Rev. A* **61**, 051402 (R) (2000); *Phys. Rev. A* **66**, 063408 (2002).]

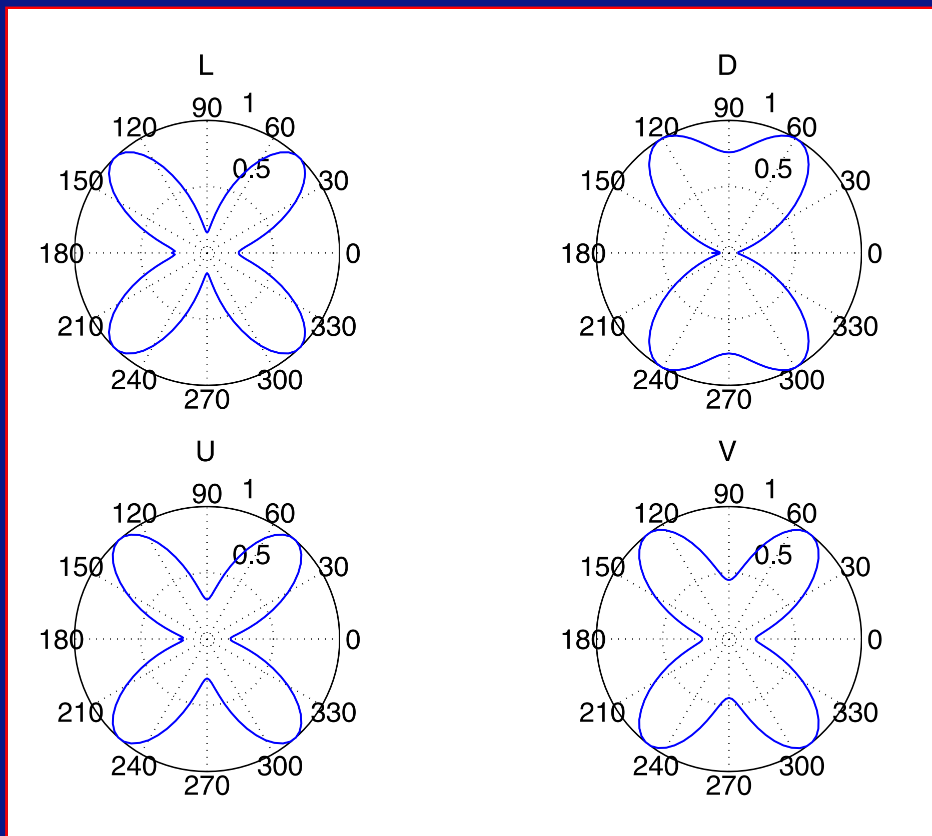
Validity of quasi-static approximation for H_2



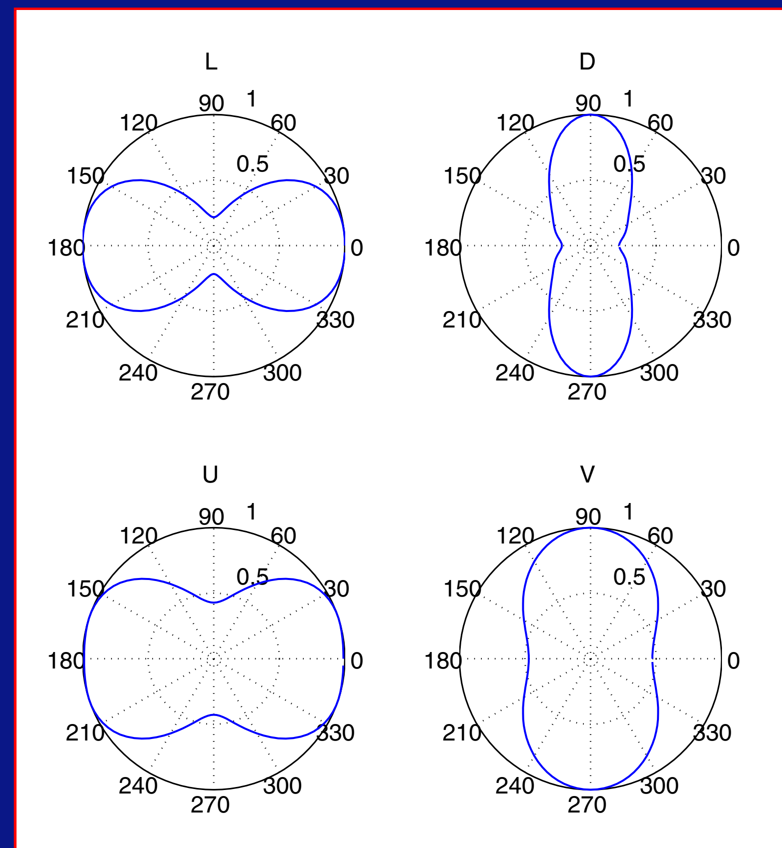
Full dimensional solution of TDSE: M. Awasthi, Y. V. Vanne, A. Saenz, *J. Phys. B* **38**, 3973 (2005) [method];
M. Awasthi and A. Saenz, *J. Phys. B:* **39**, S389 (2006) [R dependence].

Strong-field approximation (SFA) for oxygen and nitrogen

Oxygen O_2

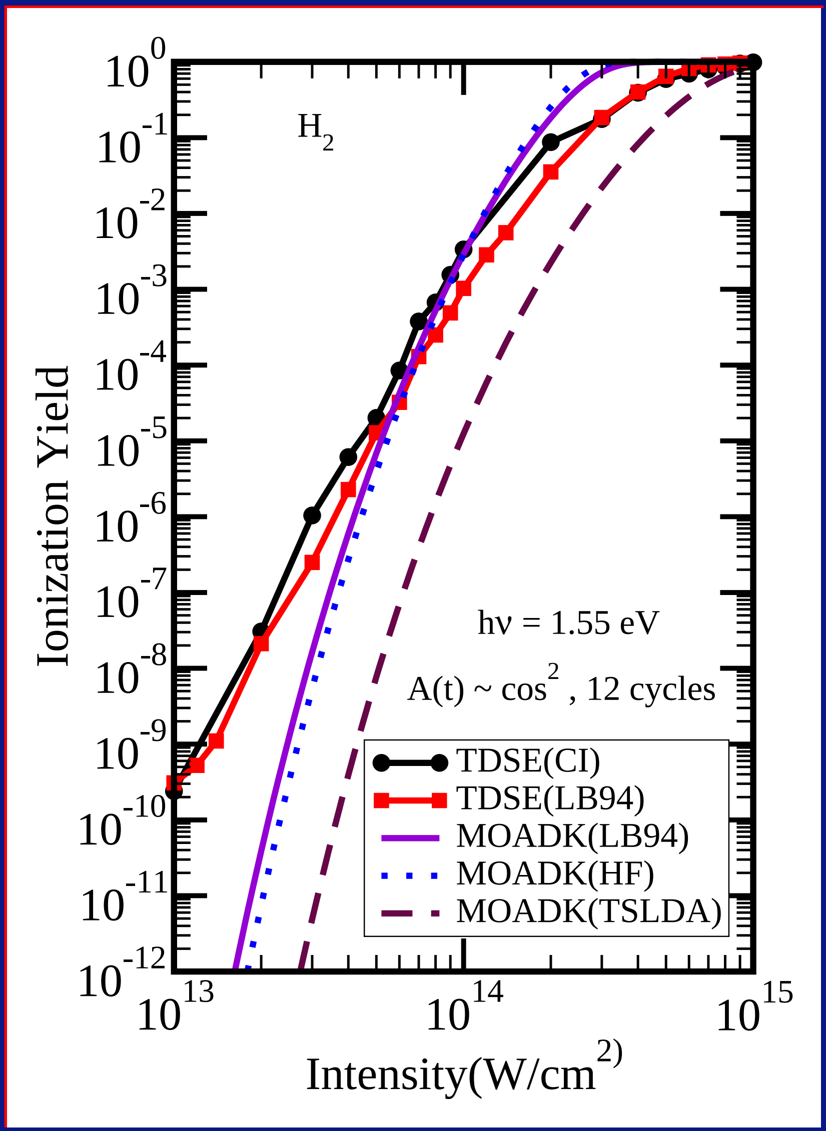


Nitrogen N_2



(from D. Milosevic, Phys. Rev. A **74**, 063404 (2006))

Validity of MO-ADK and MO-SFA-V/L for H_2



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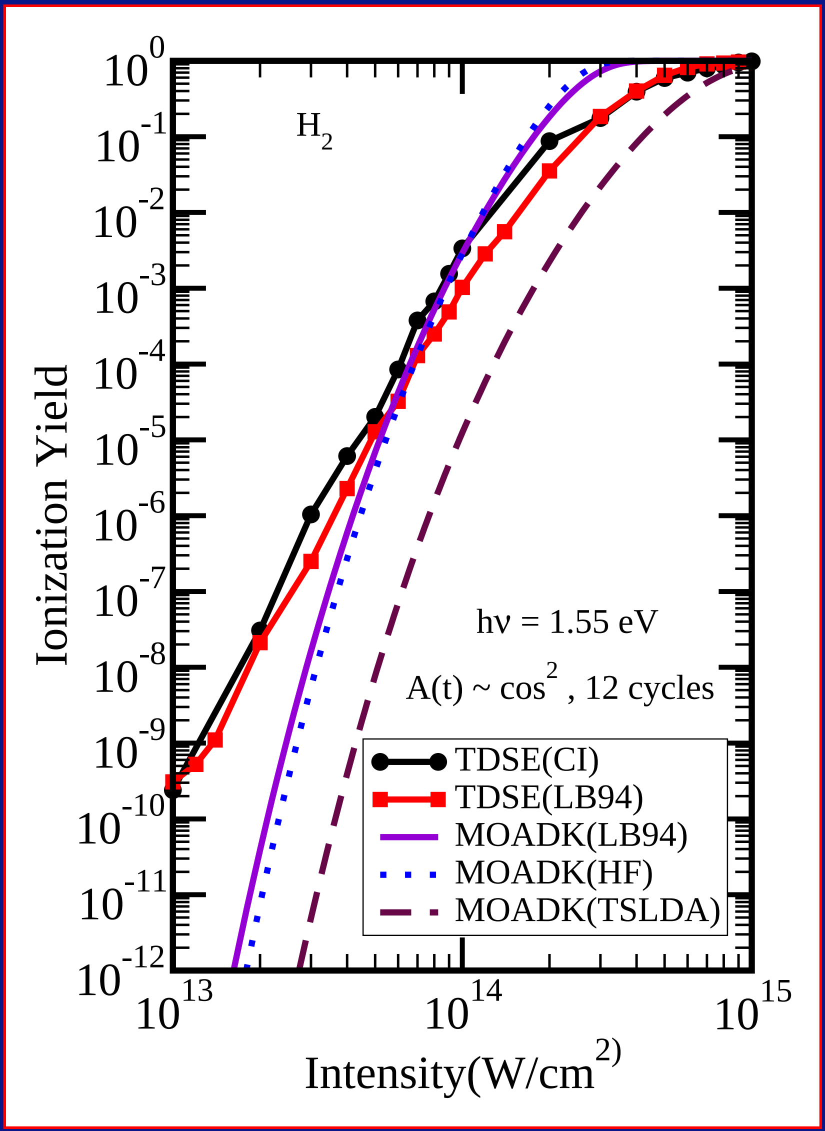
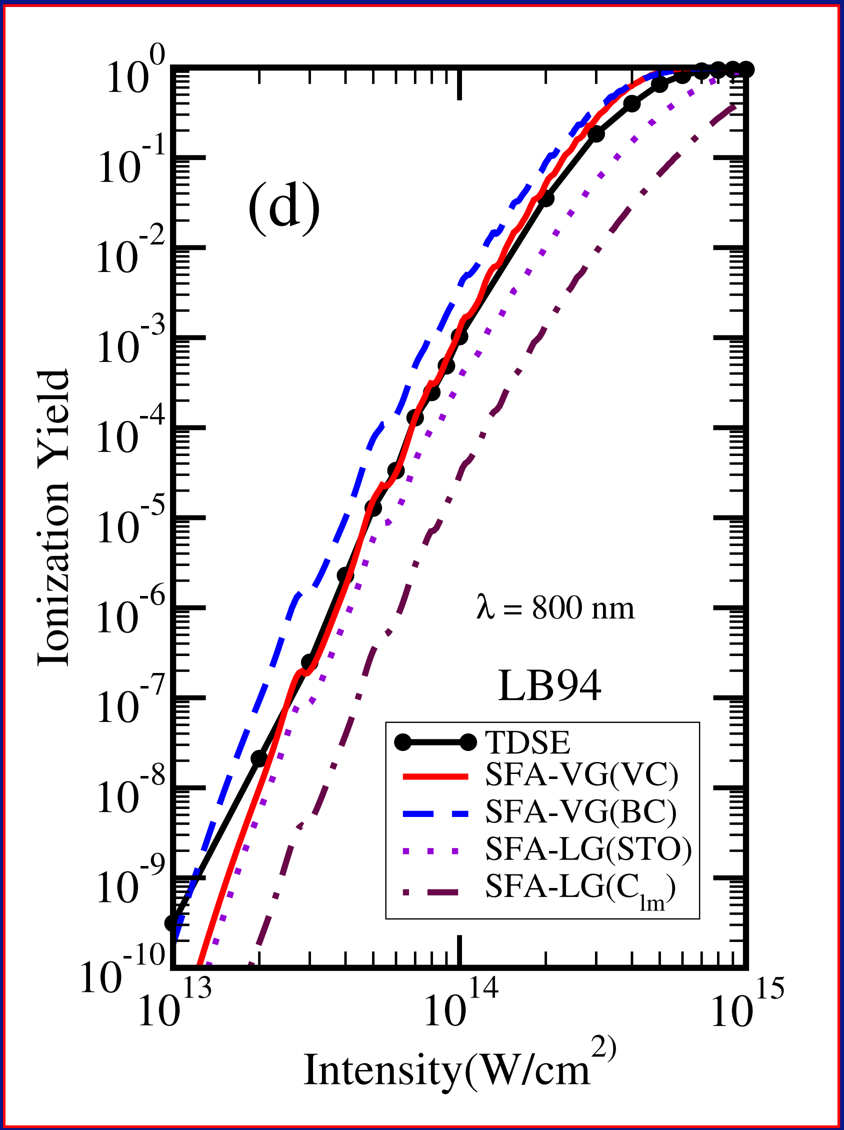




Floquet versus SFA and TDSE

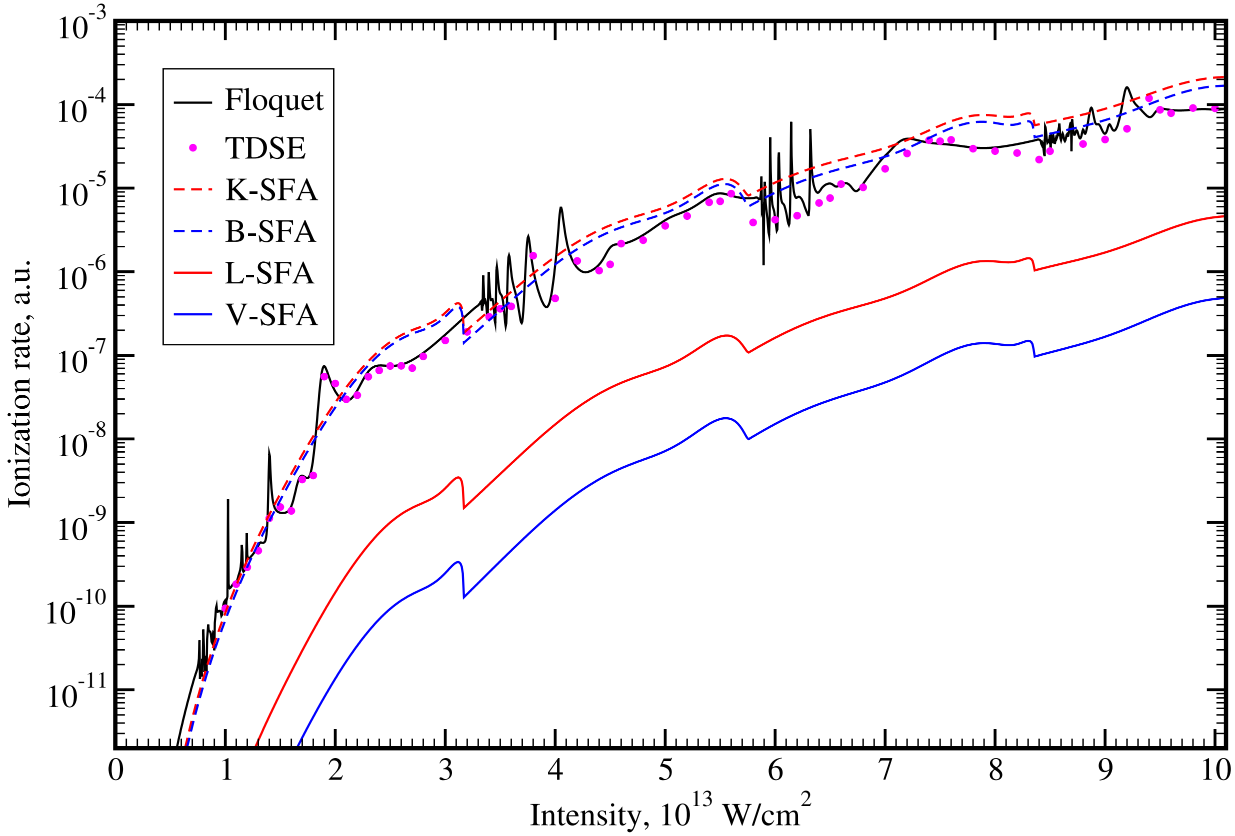
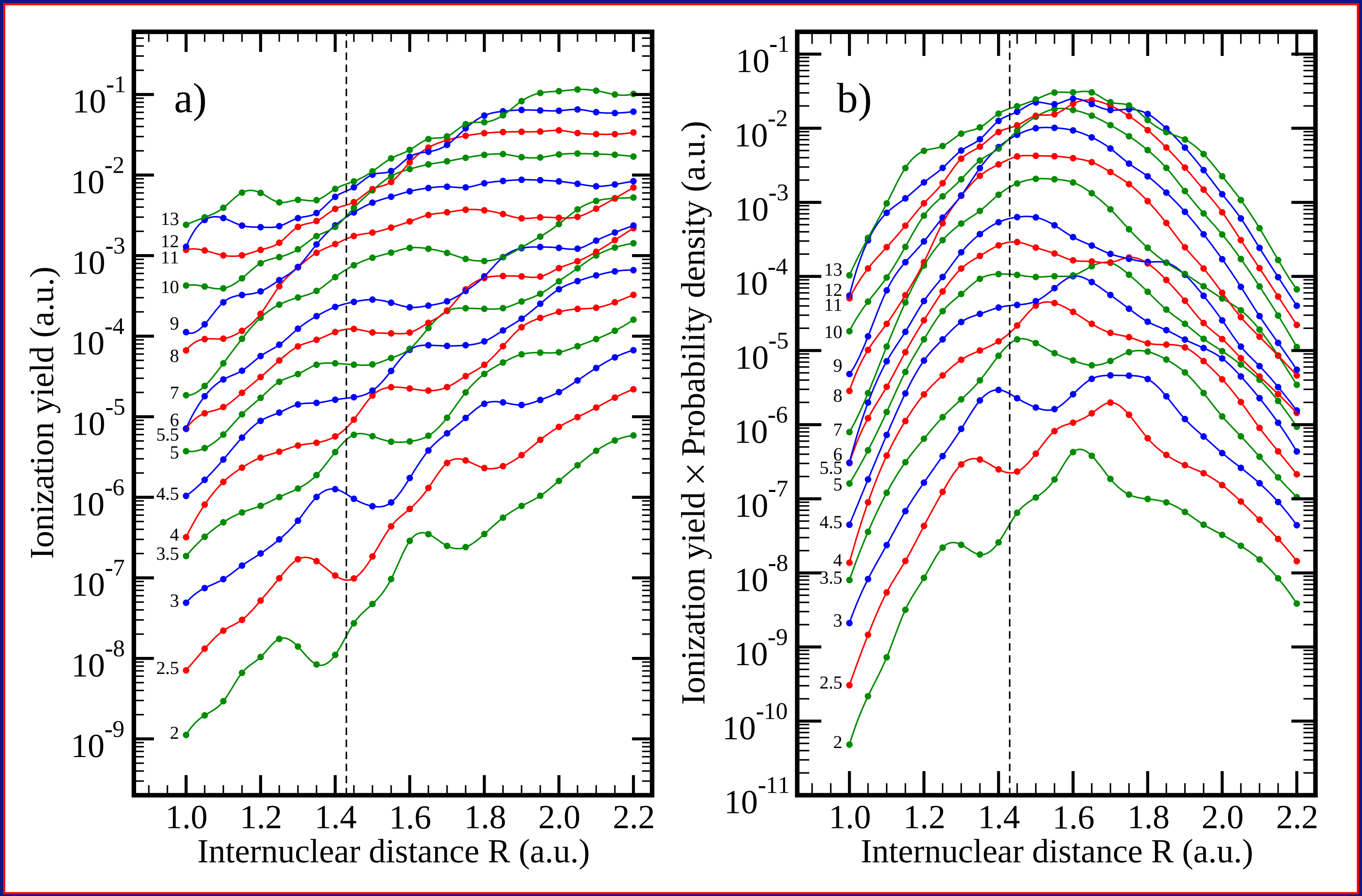


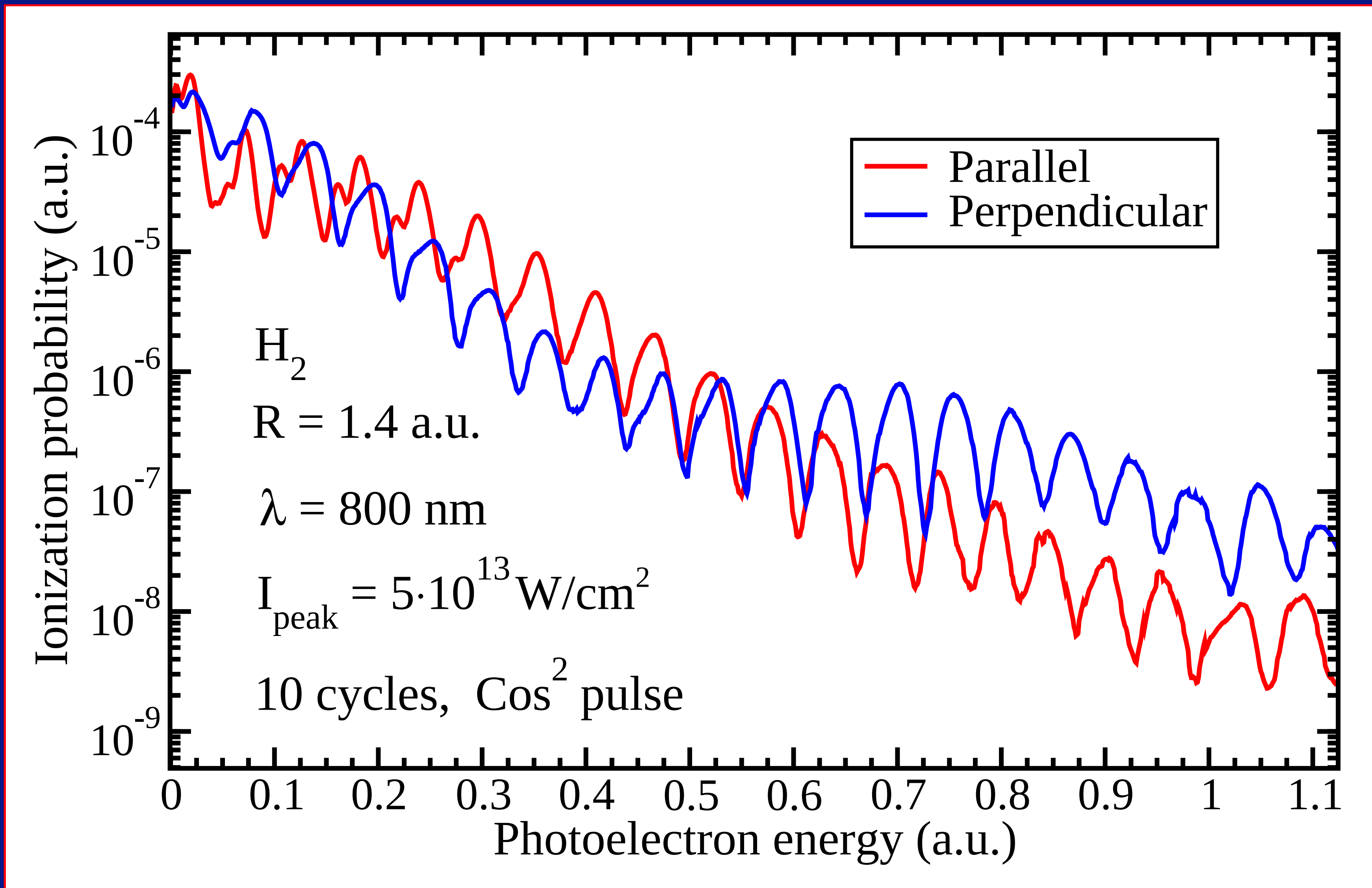
Figure 6.10.: Ionization rates of an H atom for an 800 nm laser field vs. laser intensity. Floquet results are compared with ionization rates yielded by the TDSE method, L-gauge SFA (L-SFA), L-gauge SFA with Krainov's Coulomb correction (K-SFA), V-gauge SFA (V-SFA), and V-gauge SFA with the Coulomb correction of A. Becker *et al.* (B-SFA).

Internuclear-distance dependent ion yields of H_2 (800 nm, perp.)

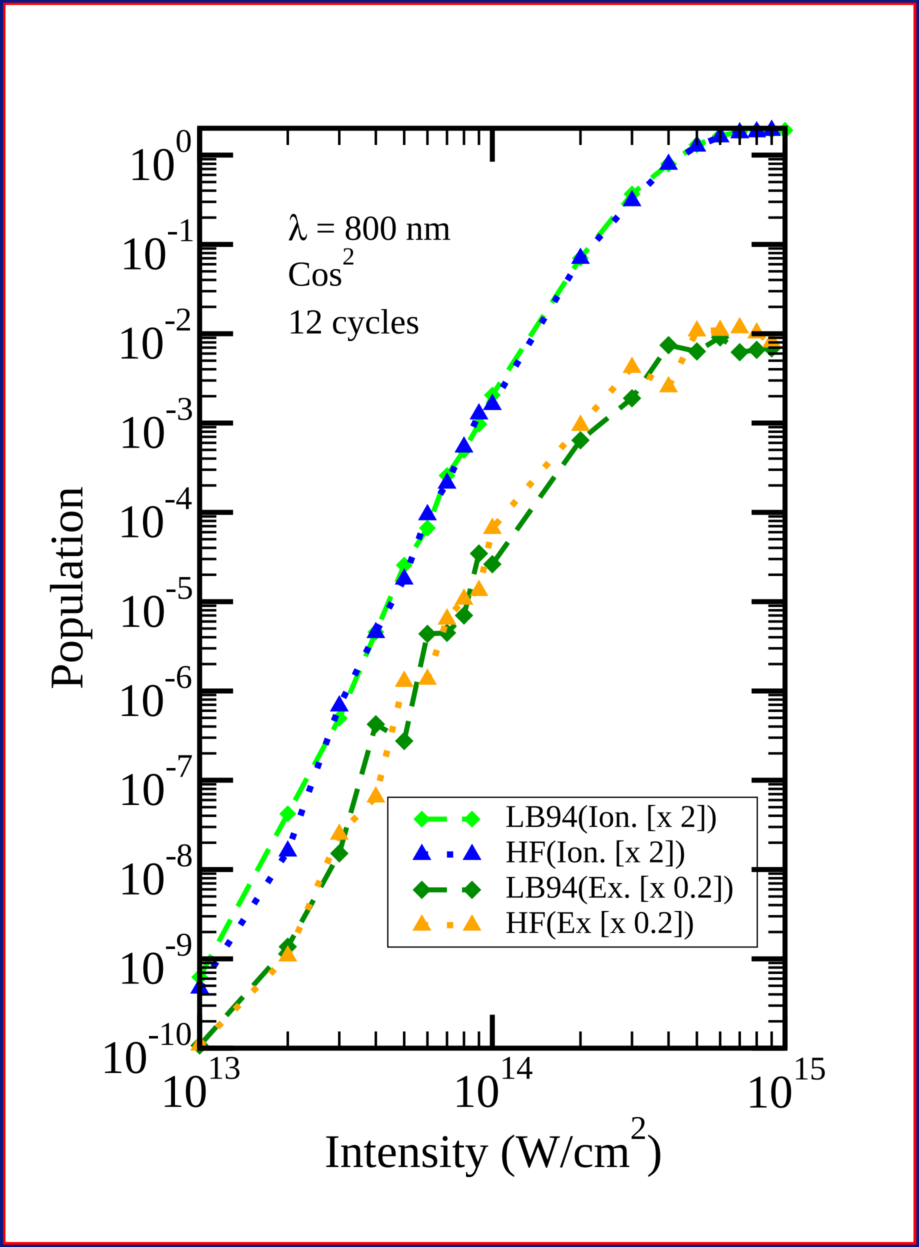


[for method see: Y.V. Vanne and A. Saenz, J. Modern Optics **55**, 2655 (2008); Phys. Rev. A **80**, 053422 (2009)]

Energy-resolved electron spectra (ATI)

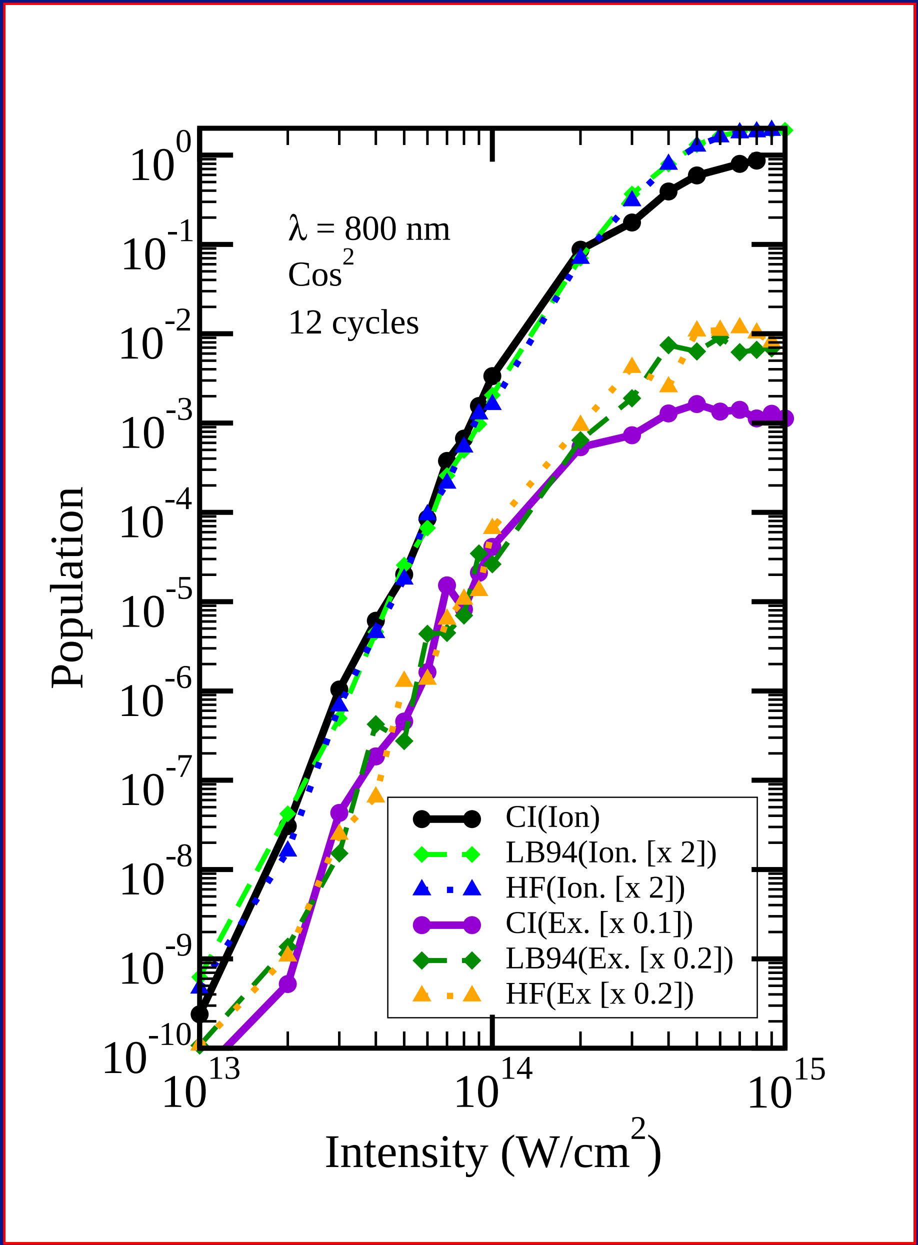


H_2: Hartree-Fock vs. DFT core (excitation)



M. Awasthi et al.
PRA **77**, 063403 (2008)

Validity of the SAE approximation for H_2



M. Awasthi et al.
PRA **77**, 063403 (2008)