Control of H_2 dissociative ionization in the non-linear regime using EUV femtosecond pulses @FERMI

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Michael Meyer
Thomas Baumann
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Oksana Plekan
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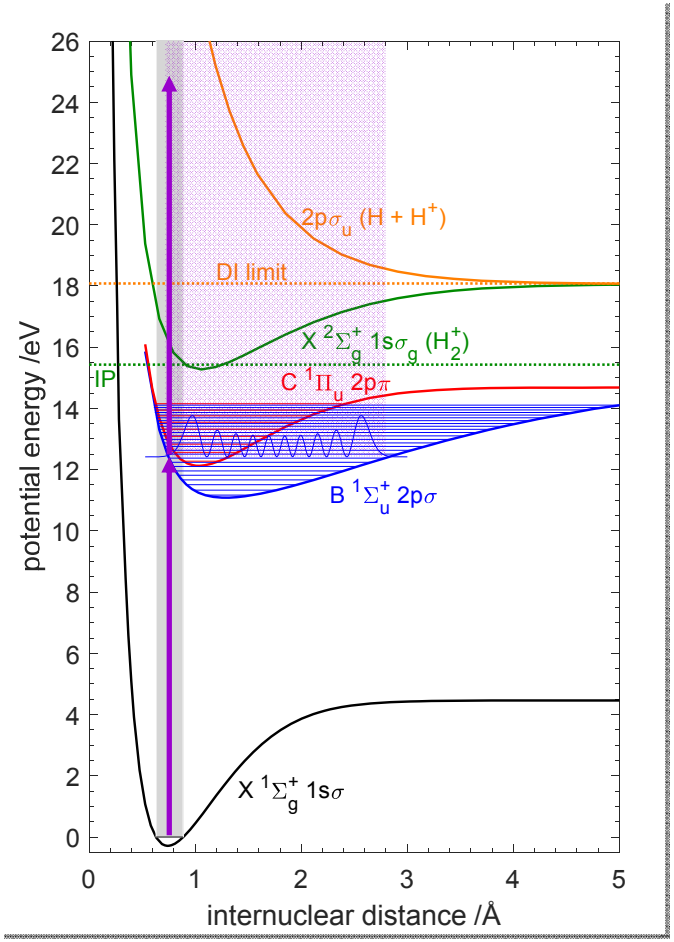
David Gauthier

Elettra Sincrotrone
Kevin Prince
Robert Richter
Paola Finetti

Henri Bachau

Fernando Martín
Alicia Palacios
Roger Bello

OBJECTIVE



- brilliance for a non-linear 2-photon process
- short pulses
- tunable in the EUV photon energy range

>> FERMI FEL

first user experiments at wavelengths around 100 nm

TWO-PHOTON IONIZATION OF H_2: THEORY

PRL 96, 143001 (2006) PHYSICAL REVIEW LETTERS week ending 14 APRIL 2006

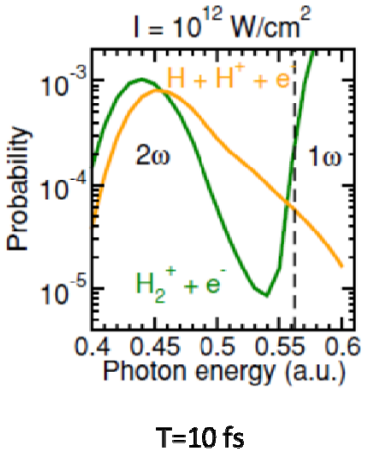
Enhancement and Control of H_2 Dissociative Ionization by Femtosecond VUV Laser Pulses

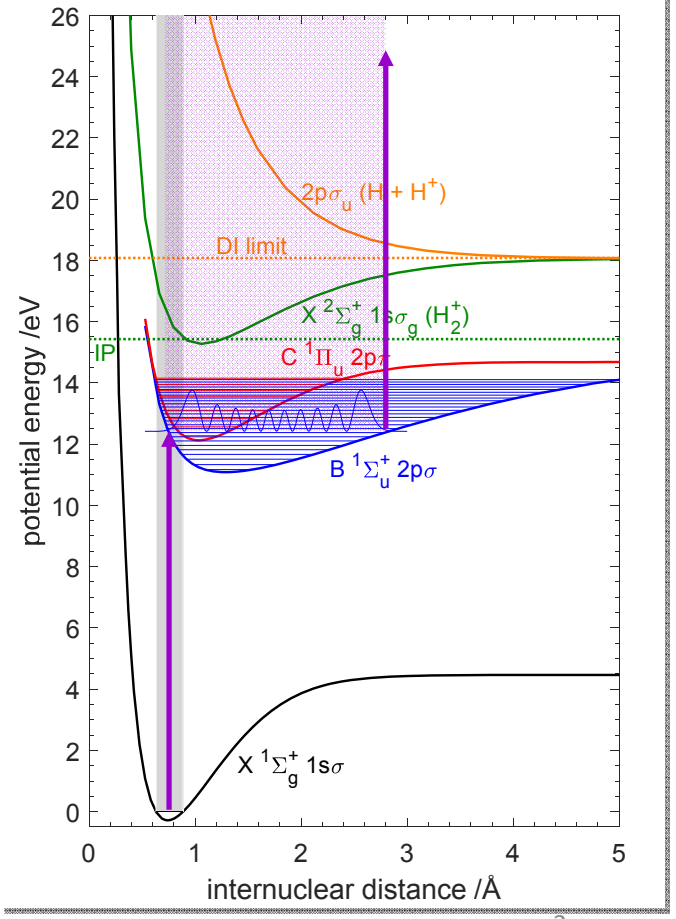
A. Palacios,[1] H. Bachau,[2] and F. Martín[1]

IOP Publishing Journal of Physics B: Atomic, Molecular and Optical Physics
J. Phys. B: At. Mol. Opt. Phys. 43 (2010) 015204 (8pp) doi:10.1088/0953-4075/43/1/015204

The role of autoionizing states in two-photon dissociative ionization of H_2 by xuv ultrashort laser pulses

J F Pérez-Torres[1], J L Sanz-Vicario[2], H Bachau[3] and F Martín[1]

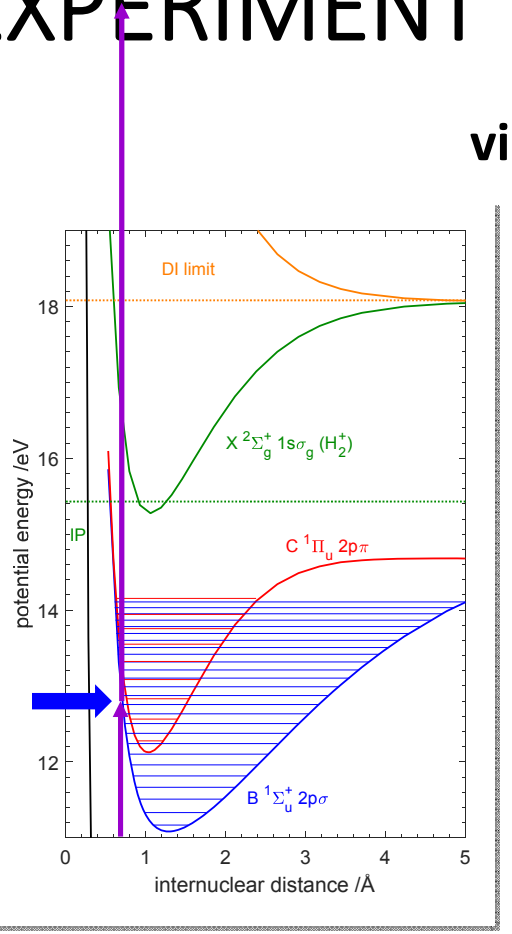




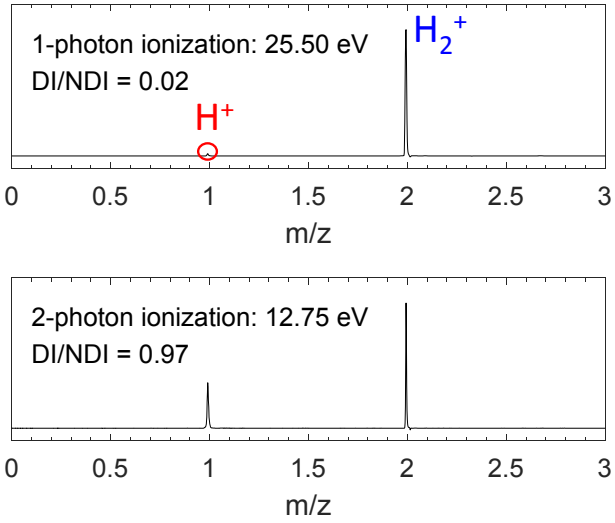
- **non-linear** (1+1) resonance enhanced **2 photon ionization**
- study the role of electronic and nuclear degrees of freedom
  - ➢ pulses with 1-10 fs durations
  - ➢ **dissociative ionization** (DI) becomes **dominant** and can be **controlled by** the **pulse length**, **laser intensity**, and **photon energy**

EXPERIMENT @FERMI

vibrationally resolved 1+1 REMPI



FEL pulses:
-12.3 - 14.8 eV (101-84 nm)
-t ≈ 50-100 fs
-10 Hz repetition rate

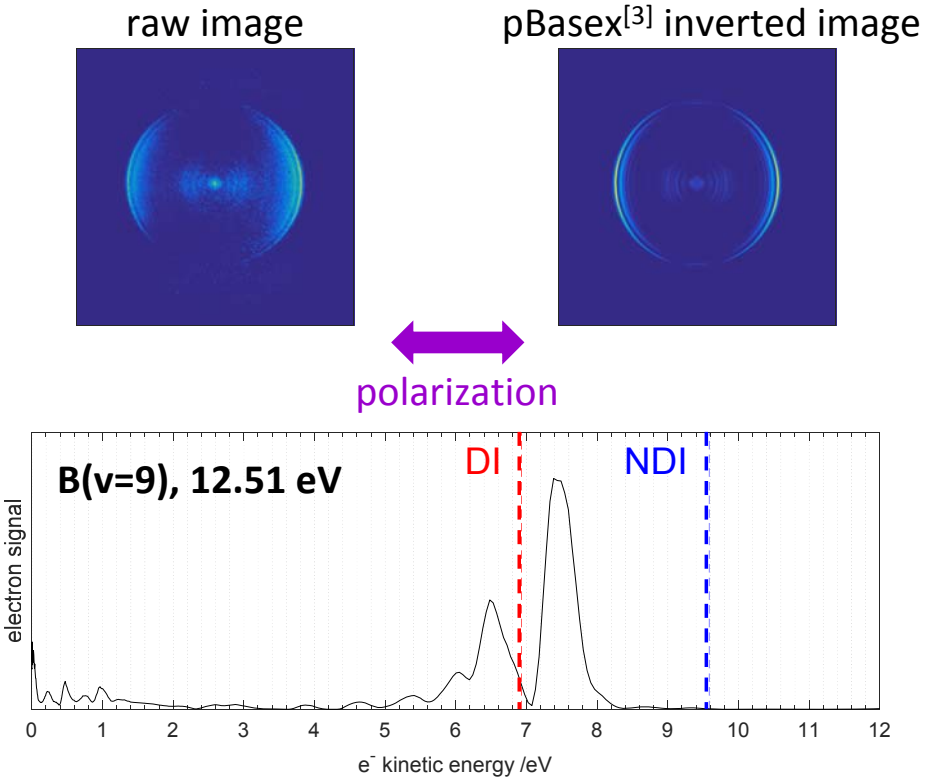


FERMI LDM beamline:[1,2]
- **TOF-Mass Spectrometry**
- **Photoelectron velocity map imaging (VMI)**
- **Ion VMI**

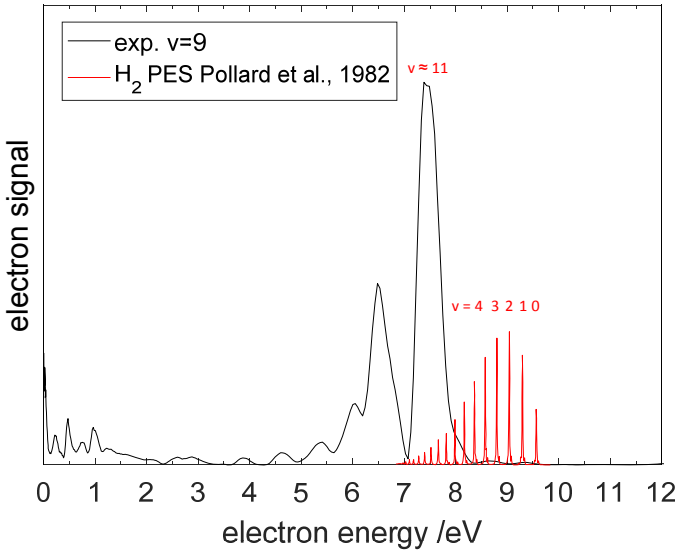
➢ very strong enhancement of dissociative ionization

[1] O'Keeffe et al., Nucl. Instrum. Methods Phys. Res. B **284** (2012) 69-73.
[2] Lyamayev et al., J. Phys. B: At. Mol. Opt. Phys. **46** (2013) 164007.

ELECTRON VMI FOR A Σ INTERMEDIATE STATE



comparison with one-photon PES



- Clear separation of electrons correlated to DI or NDI
- DI/NDI ratio can be determined: DI/NDI = 0.66
- Excitation of H_2^+ vib. levels outside the one-photon FC region >> indication for **nuclear dynamics**

[3] Garcia et al., Rev. Sci. Instrum. **75**, 4989 (2004).

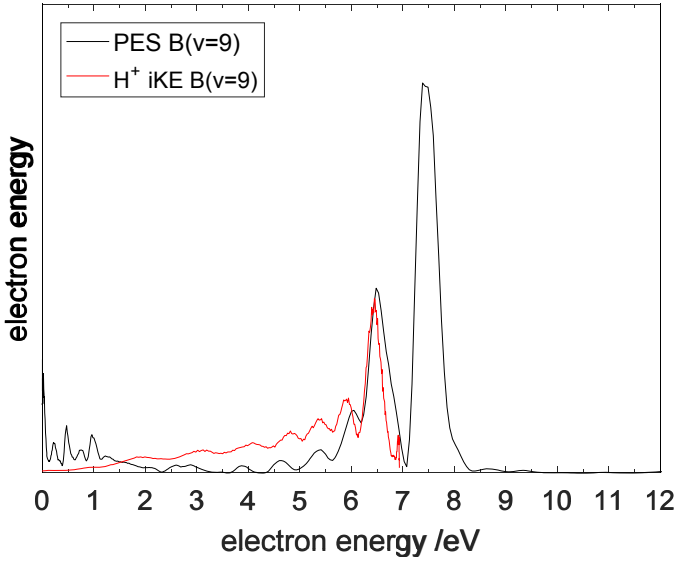
ION VMI FOR A Σ INTERMEDIATE STATE

Electrons correlated to dissociative ionization

➢Energy conservation for DI:

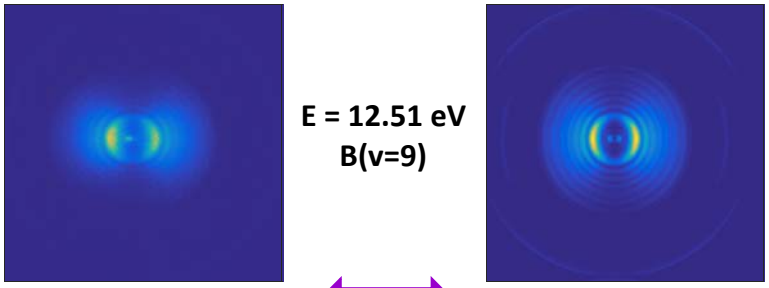
$$2 \cdot h\nu = E_e + 2 \cdot E_i + E_{diss}$$

comparison of electron and ion spectrum



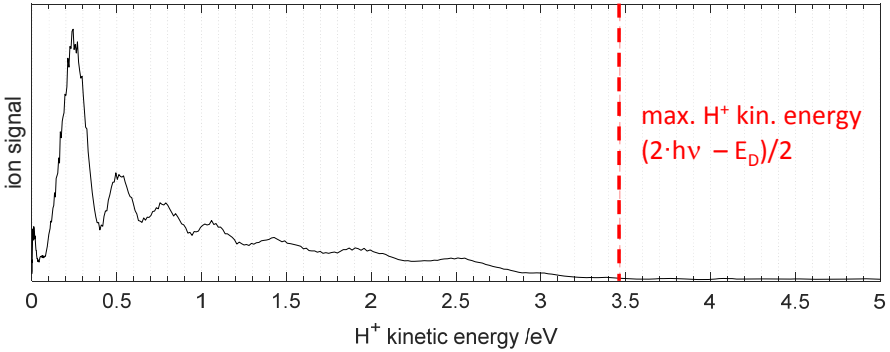
raw image

pBasex inverted image



E = 12.51 eV
B(v=9)

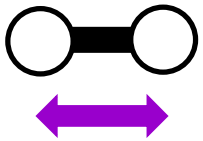
polarization

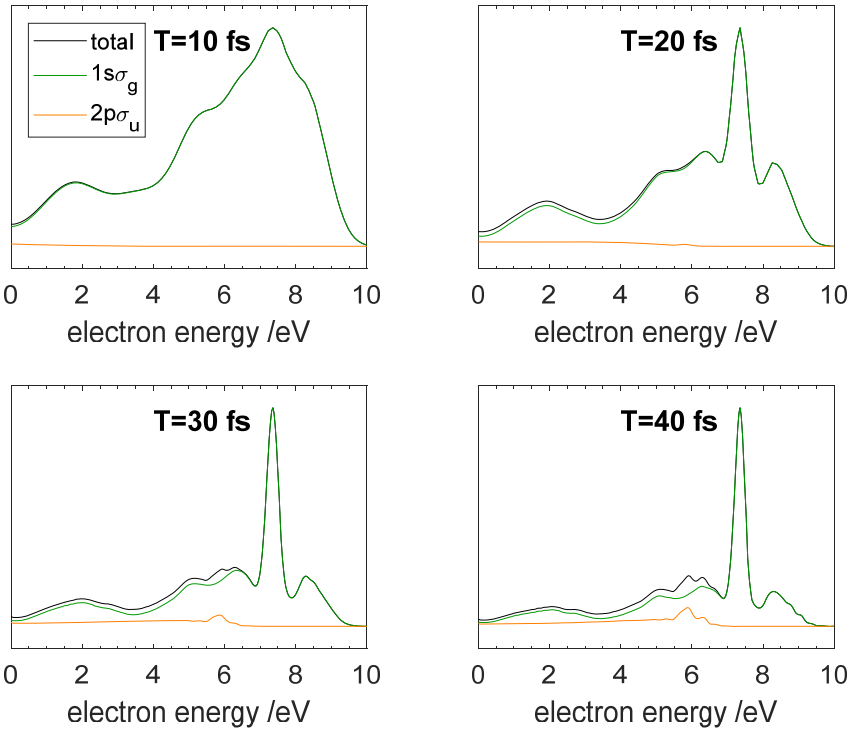


COMPUTED ELECTRON SPECTRA

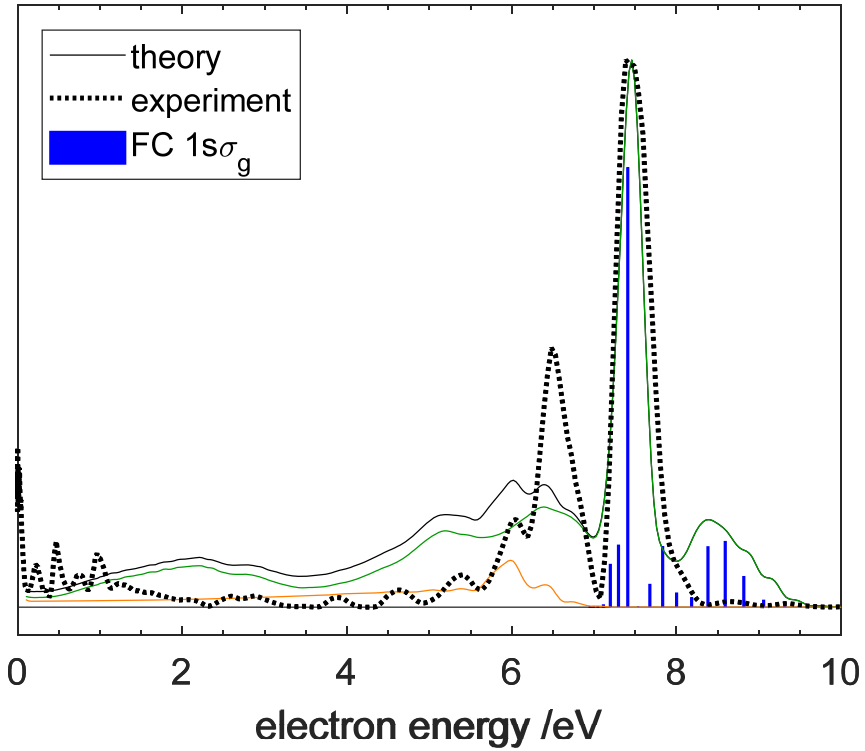
Work in progress by R. Bello, A. Palacios, F. Martín, and H. Bachau :

- time-dependent 2nd order **perturbation theory**
- inclusion of **autoionizing states** and all couplings between direct ionization and autoionization
- restricted to **parallel orientation** only (for the moment)





EXPERIMENT VS. THEORY



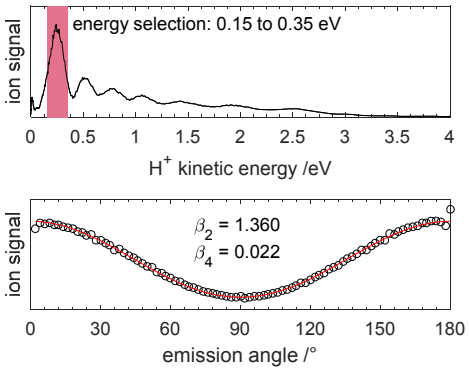
Electrons correlated to non-dissociative ionization

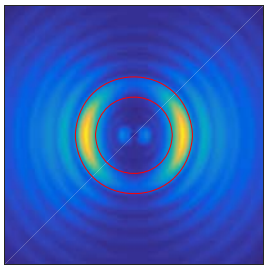
- good agreement with theory
- Franck-Condon overlap of $H_2(B^1\Sigma_u^+)$ vibrational wavefunction with $H_2^+(X^2\Sigma_g^+)$ including dipoles

Electrons correlated to dissociative ionization?

- We're working on it…

ION ANGULAR DISTRIBUTIONS





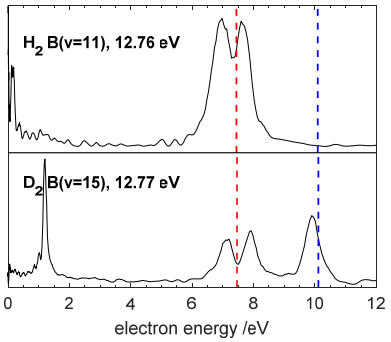
$$H_2\ X(^1\Sigma_g^+) \xrightarrow{h\nu} H_2^*\ B(^1\Sigma_u^+, v=9) \xrightarrow{h\nu} H + H^+ + e^-$$

$$I(\theta) = 1 + \boldsymbol{\beta_2}\ P_2(\cos\theta) + \boldsymbol{\beta_4}\ P_4(cos\theta)$$

expected:

- $\beta = 2$ for excitation step
- $\beta \in [-1\ 2]$ for ionization step

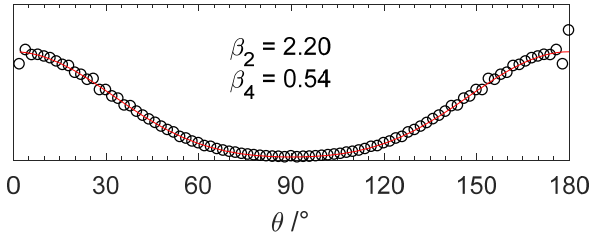
D_2



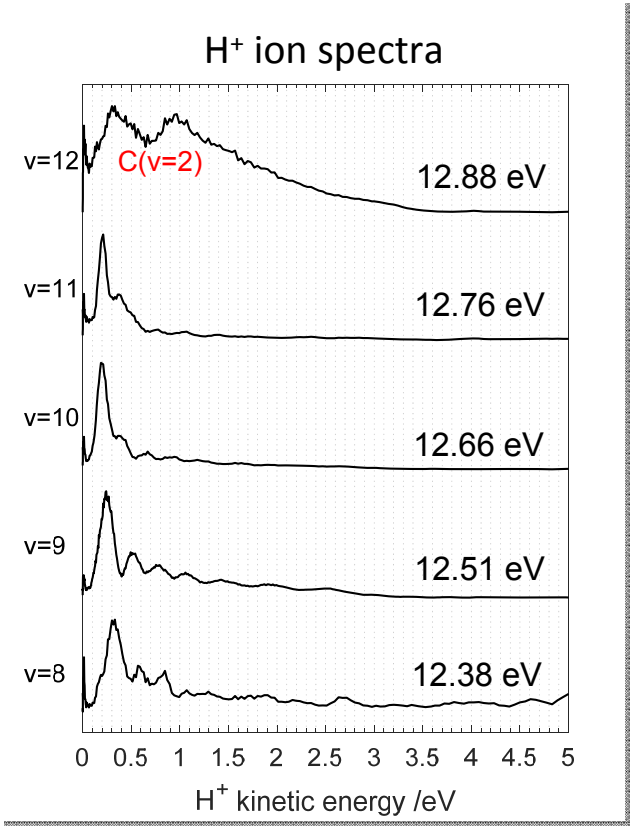
Electron spectrum

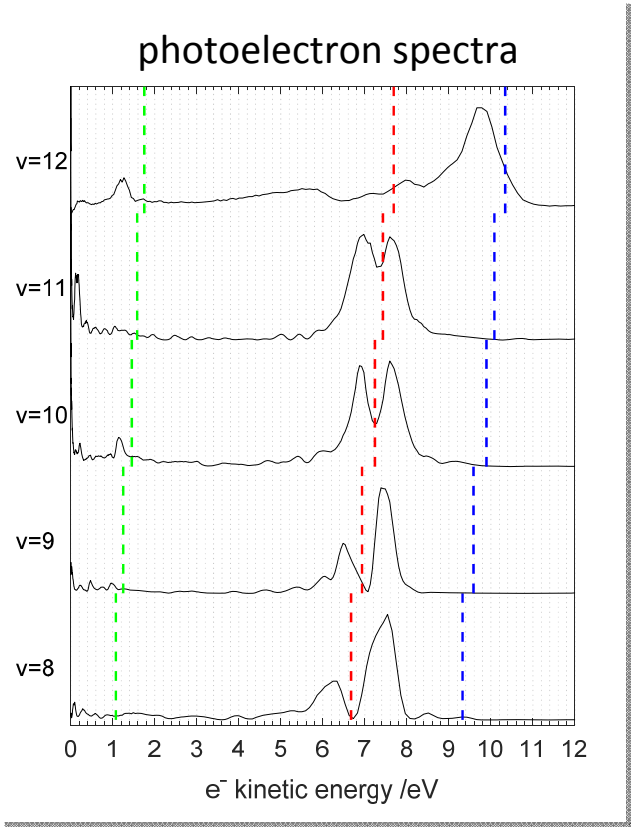
➢ poster

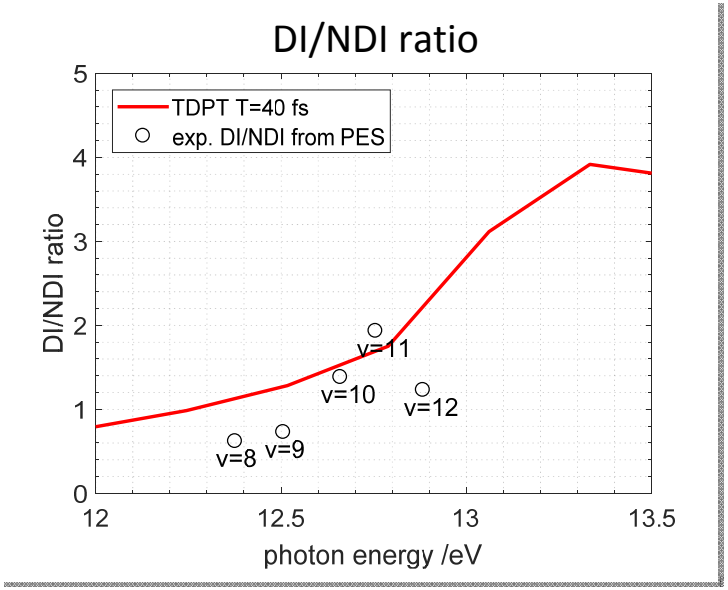
D^+ angular distribution



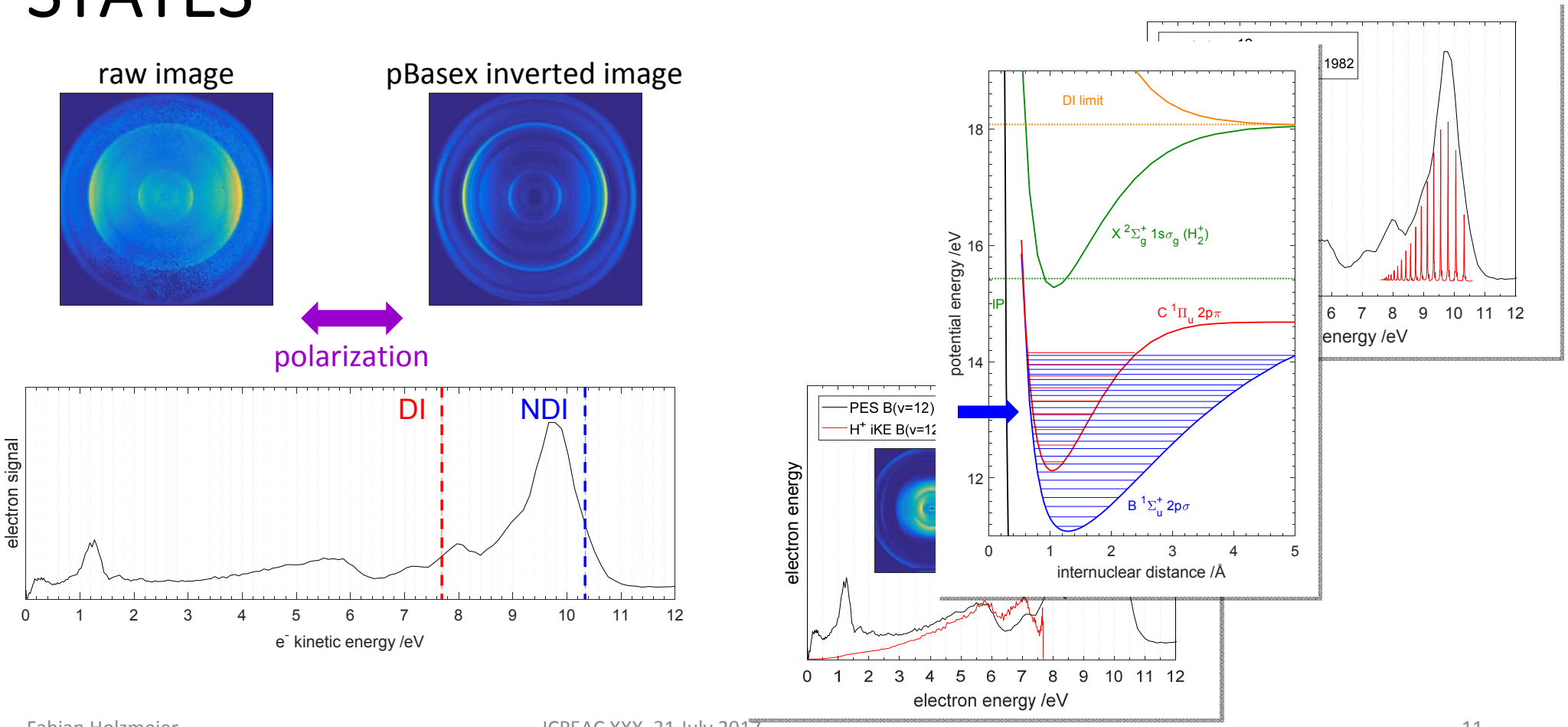
OVERVIEW OF SPECTRA







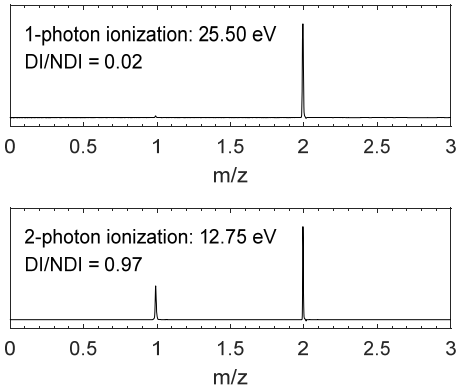
EXCITATION OF Σ AND Π INTERMEDIATE STATES



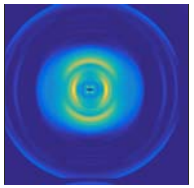
SUMMARY

Probe effect of nuclear dynamics in the outcome of 2-photon ionization of H_2 with femtosecond EUV FEL pulses

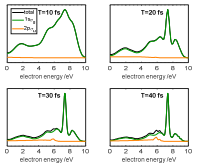
- Significant enhancement of dissociative ionization



- Dissociative and non-dissociative photoionization depend strongly on the photon energy and the symmetry of the intermediate state(s) >> **control**

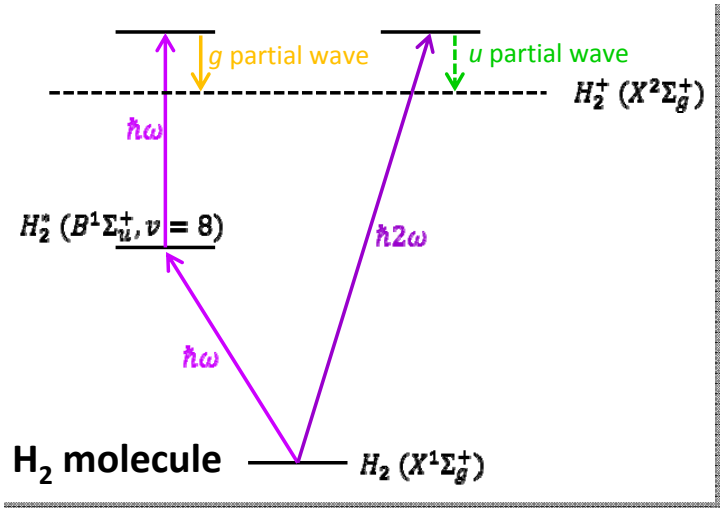


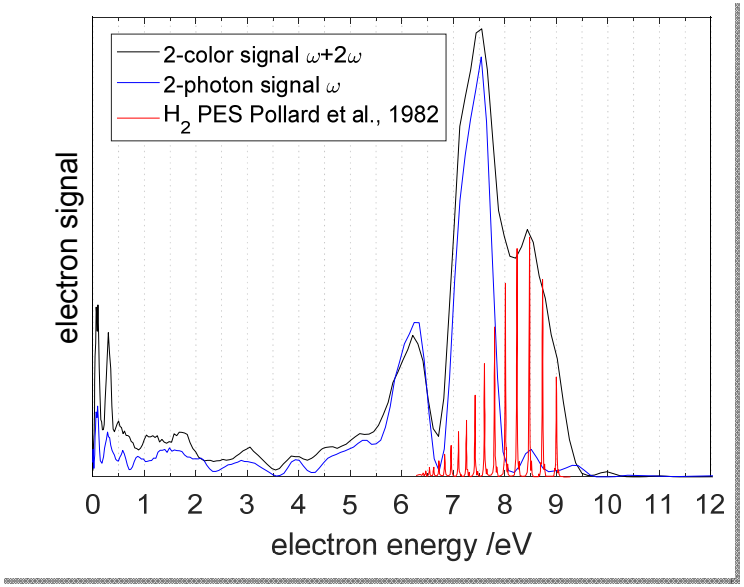
- 2nd order time-dependent perturbation theory to complement the experimental data (UA Madrid)
- Parallel (Σ) and perpendicular (Π) transitions soon

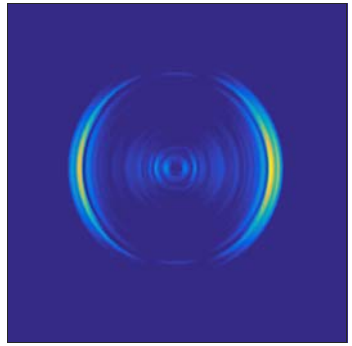


TOWARDS COHERENT CONTROL IN MOLECULES

In analogy to K. C. Prince et al., Nat. Phot. **10** (2016) 176-179







THANK YOU!

Funding for Fermi experiment and ICPEAC:

SOLEIL SYNCHROTRON

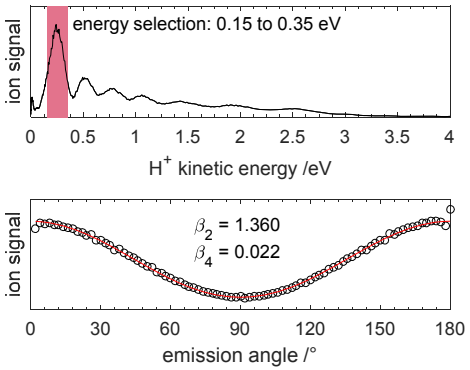
ATTO Lab

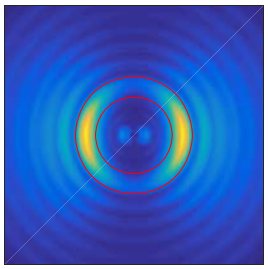
PALM
Laboratoire d'Excellence
Physique : Atomes Lumière Matière

INP Institut de physique

MARIE CURIE ACTIONS

ION ANGULAR DISTRIBUTIONS





$$H_2\, X(^1\Sigma_g^+) \xrightarrow{h\nu} H_2^*\, B(^1\Sigma_u^+, v=9) \xrightarrow{h\nu} H + H^+ + e^-$$

$$I(\theta) = 1 + \boldsymbol{\beta_2}\, P_2(\cos\theta) + \boldsymbol{\beta_4}\, P_4(cos\theta)$$

expected:

- $\beta = 2$ for excitation step
- $\beta \in [-1\ 2]$ for ionization step

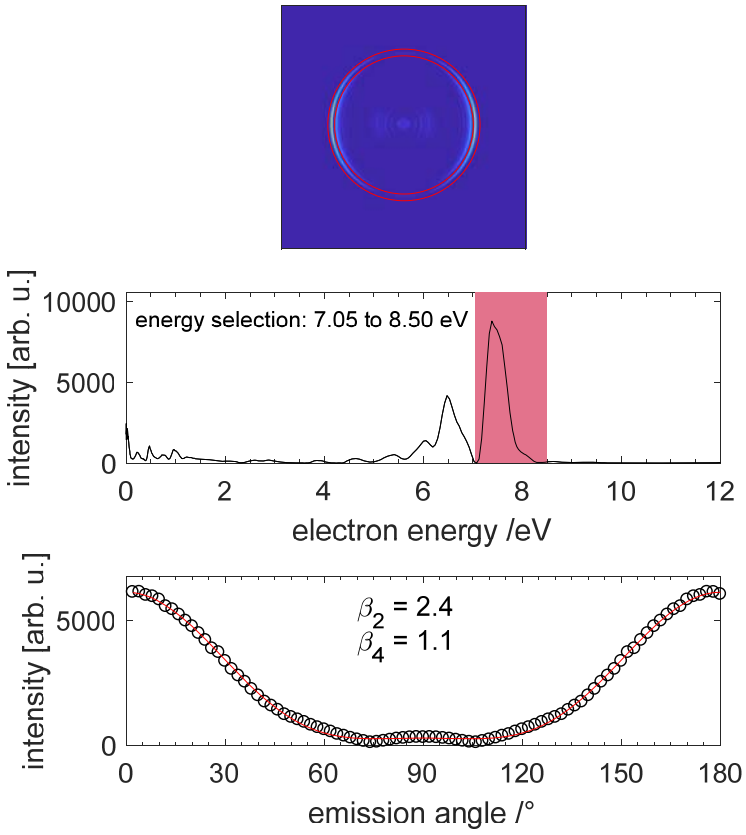
rotation?

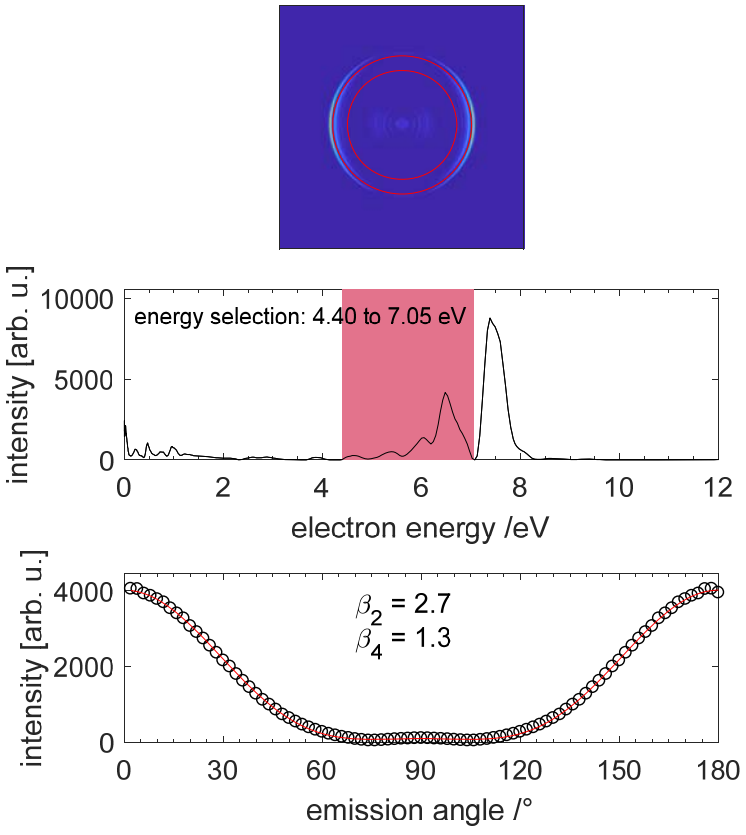
Rotational periods from exp. rotational constants[5] for v=9 (rigid rotor model):

J	T_{rot} /fs
1	870
2 (+10 meV)	502
3 (+20 meV)	355
4 (+33 meV)	275
5 (+49 meV)	225

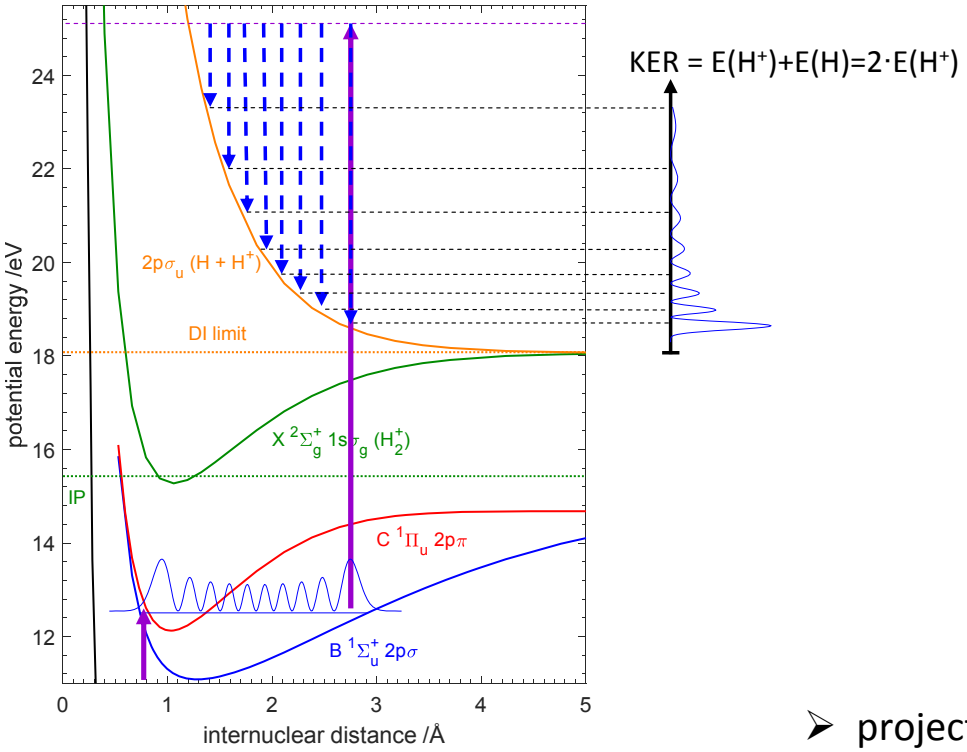
[5] Braig and Connerade, J. Phys. B: At. Mol. Opt. Phys. **18**, L809 (1984).

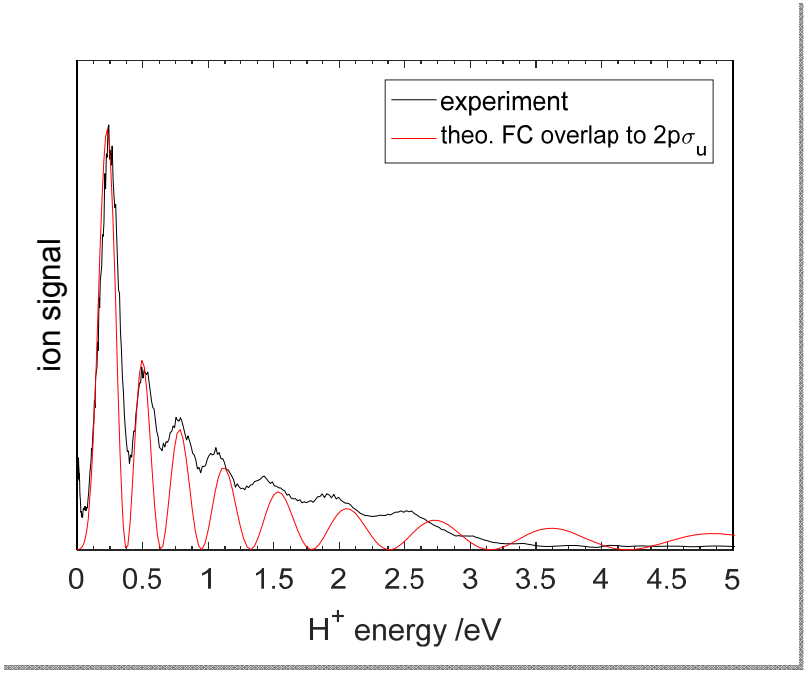
ELECTRON ANGULAR DISTRIBUTIONS





STRUCTURE IN ION SPECTRUM



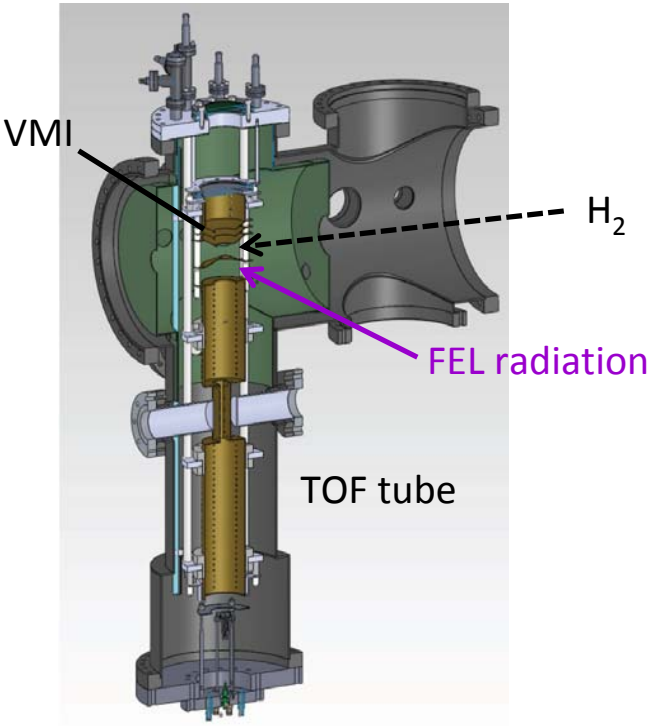


- projection of nuclear probability function onto $2p\sigma_u$ continuum state

EXPERIMENT @FERMI: LDM BEAMLINE

- 2-photon ionization at photon energies between 12.3 and 14.75 eV (101 – 84 nm)
- Pulse duration $\tau \approx 100$ fs, 10 Hz repetition rate
- pulsed supersonic molecular beam of H_2
- resonant excitation of selected vibrational levels in the B, C, and B' intermediate states of H_2 and ionization with a second photon
- VMI and TOF spectrometer[2,3]

  - **TOF mass spectra**
  - **Electron velocity map images**
  - **H^+ ion velocity map images**

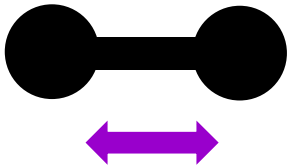


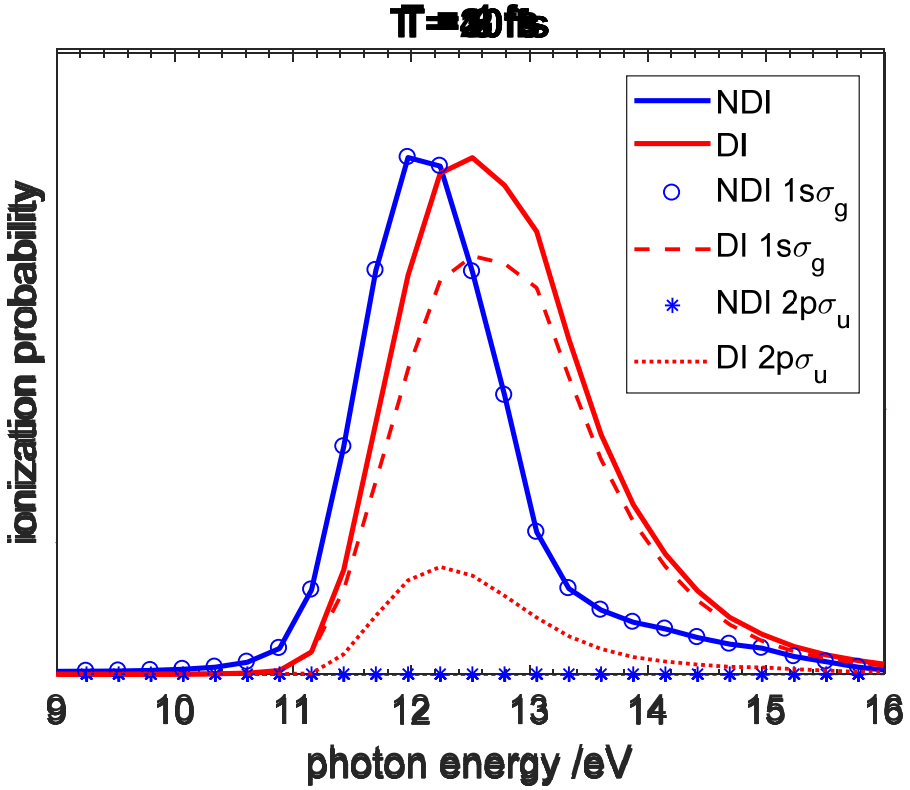
[2] O'Keeffe et al., Nucl. Instrum. Methods Phys. Res. B **284** (2012) 69-73.
[3] Lyamayev et al., J. Phys. B: At. Mol. Opt. Phys. **46** (2013) 164007.

DI FOR DIFFERENT PULSE DURATIONS

time-dependent 2nd order perturbation theory computations for two-photon ionization via the B $^1\Sigma_u^+$ intermediate state: first results

R. Bello, A. Palacios, F. Martín (**UA Madrid**)





>> ionization into the repulsive 2p σ_u continuum state becomes more important for long pulse durations

COHERENT CONTROL IN ATOMS WITH AN FEL

LETTERS
PUBLISHED ONLINE: 22 FEBRUARY 2016 | DOI: 10.1038/NPHOTON.2016.13
nature photonics

Coherent control with a short-wavelength free-electron laser

K. C. Prince[1,2,3]*, E. Allaria[1], C. Callegari[1], R. Cucini[1], G. De Ninno[1,4], S. Di Mitri[1], B. Diviacco[1], E. Ferrari[1], P. Finetti[1], D. Gauthier[1], L. Giannessi[1,5], N. Mahne[1], G. Penco[1], O. Plekan[1], L. Raimondi[1], P. Rebernik[1], E. Roussel[1], C. Svetina[1,6], M. Trovò[1], M. Zangrando[1,3], M. Negro[7], P. Carpeggiani[7], M. Reduzzi[7], G. Sansone[7]*, A. N. Grum-Grzhimailo[8], E. V. Gryzlova[8], S. I. Strakhova[8], K. Bartschat[9], N. Douguet[9], J. Venzke[9], D. Iablonskyi[10], Y. Kumagai[10], T. Takanashi[10], K. Ueda[10]*, A. Fischer[11], M. Coreno[12], F. Stienkemeier[13], Y. Ovcharenko[14], T. Mazza[15] and M. Meyer[15]

Two-color (ω and 2ω) pulses from the FERMI seeded-FEL are longitudinally coherent and can be used for coherent control experiments with ultrahigh time-resolution (3 as)