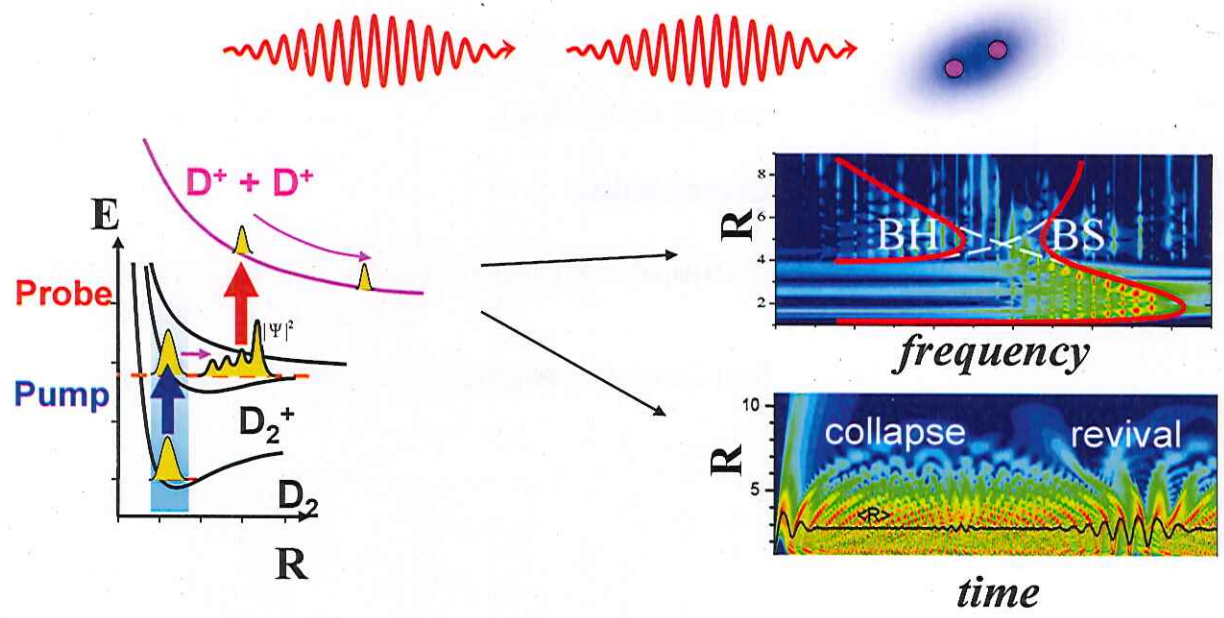


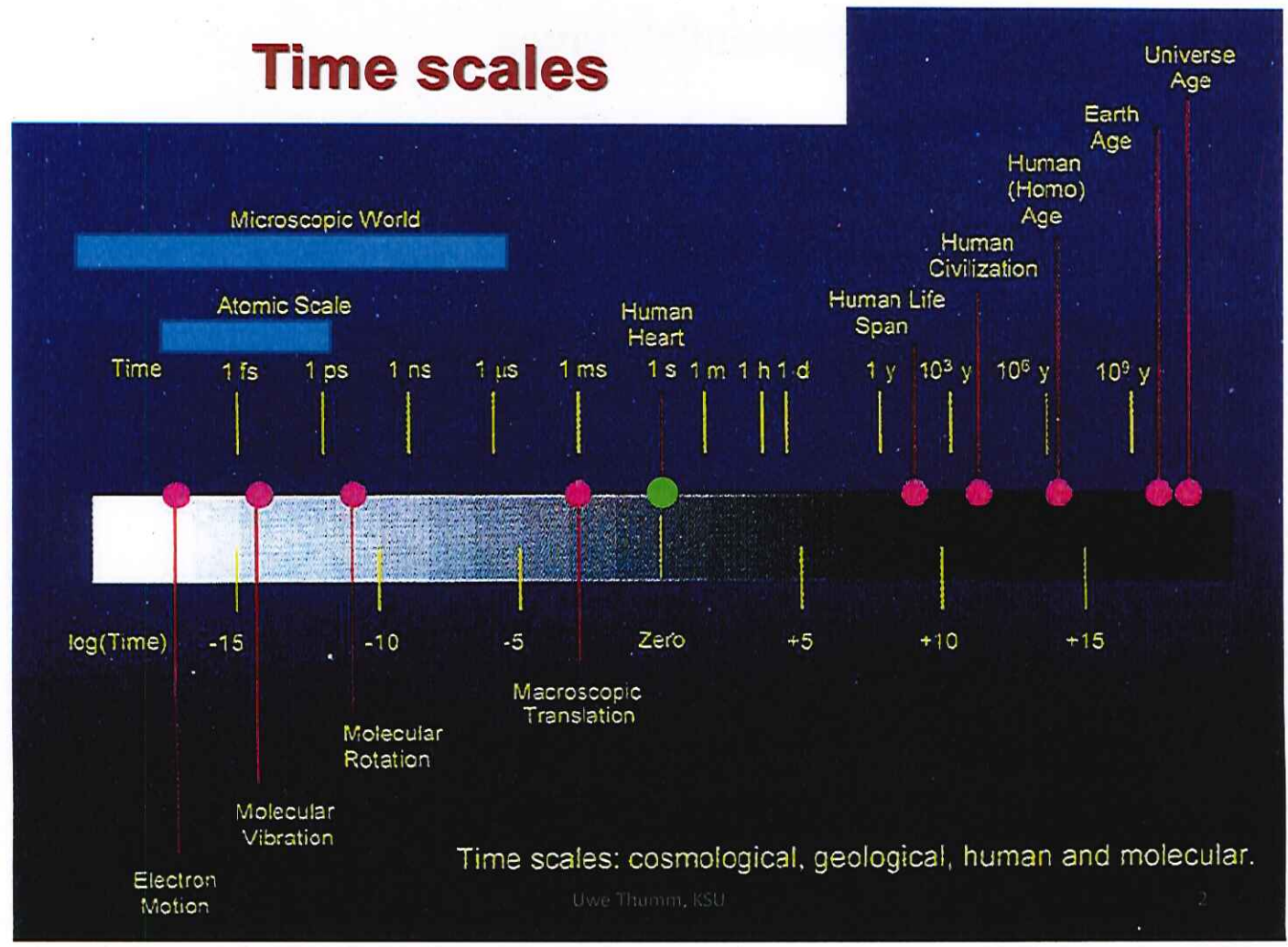
# Topics in atto- and femtosecond science Lecture 13

## Probing and controlling the dissociative and bound motion in diatomic molecules with *ultrashort laser and XUV pulses*



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## Time scales



Time scales: cosmological, geological, human and molecular.

# Basic processes

**H<sub>2</sub> / D<sub>2</sub>**  
**Introduction**  
**Numerical model**  
**Single-pulse results**

- dissociation / ionization

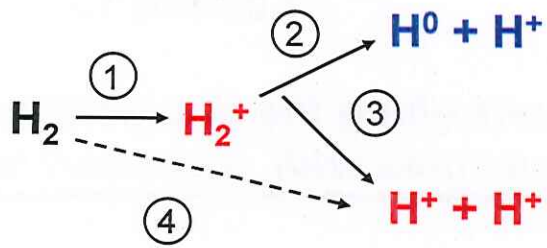
**Pump-probe results**

- time - space imaging
- dephasing & revivals
- pump-control-probe
- shaping wave packets

**Quantum-beat analysis**

- frequency – space imaging
- potential & wf reconstruction

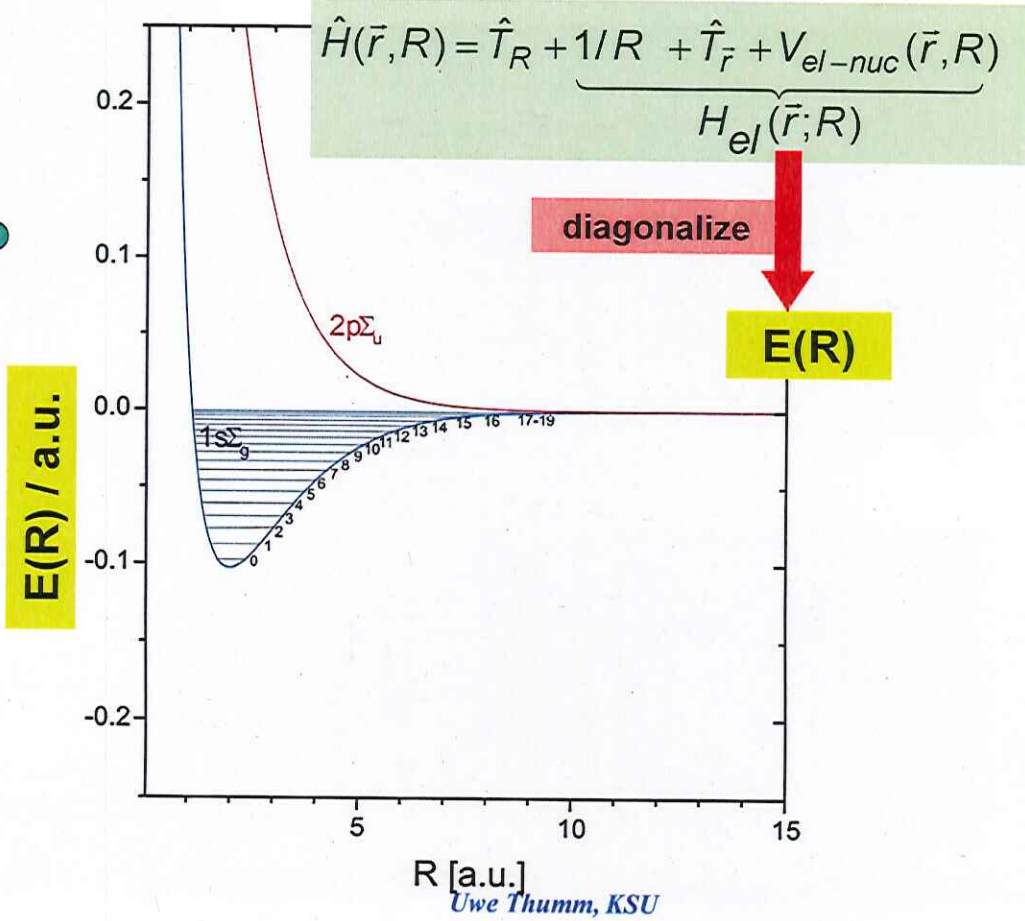
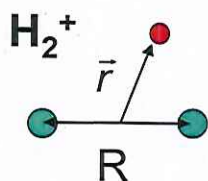
**N<sub>2</sub>, O<sub>2</sub>, CO, Ar<sub>2</sub>, ...**  
**dissociation pathways**  
**XUV pump – XUV probe**



- ① single ionization
- ② dissociation
- ③ Coulomb explosion (CREI = charge resonance enhanced ionization)
- ④ fast Coulomb explosion (double ionization)

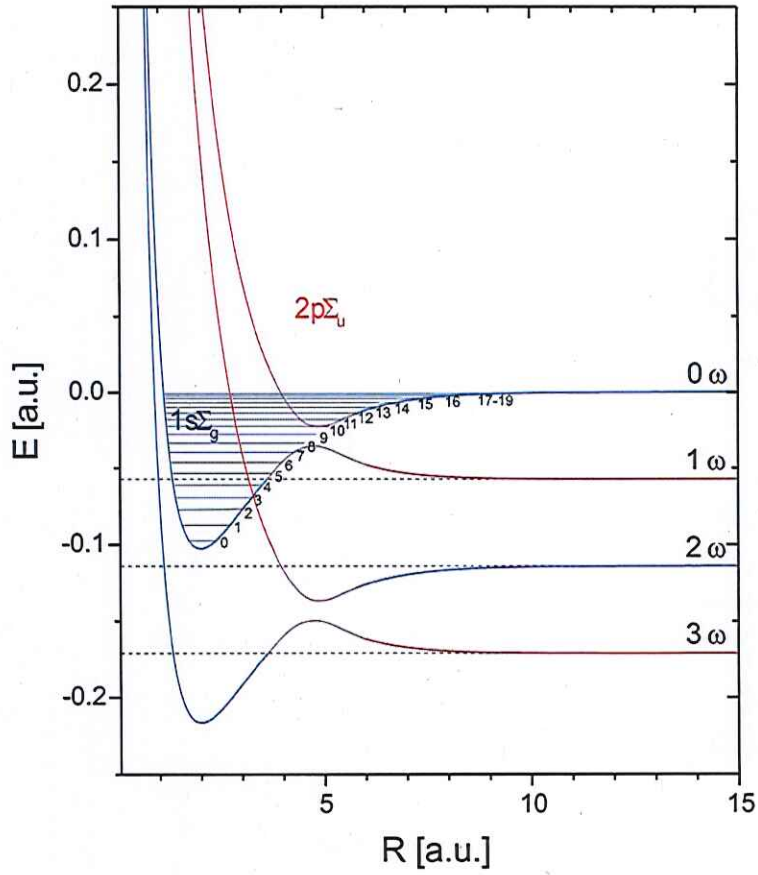
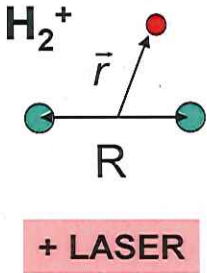
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## “Adiabatic” molecular potential curves



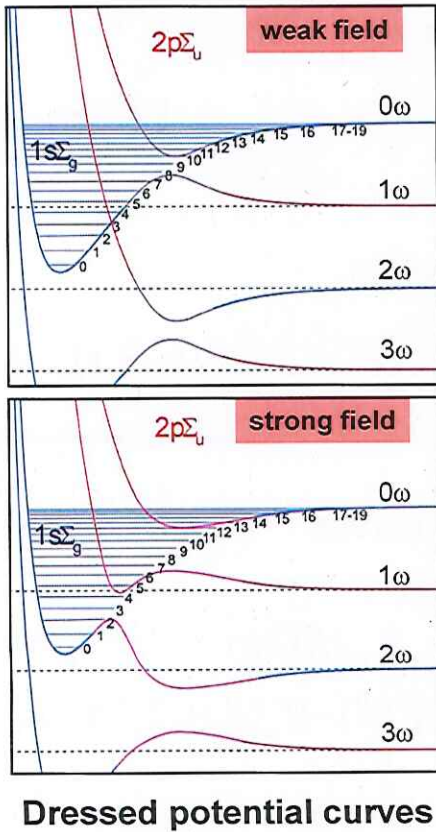
# "Light-dressed" molecular potential curve

(Floquet picture)

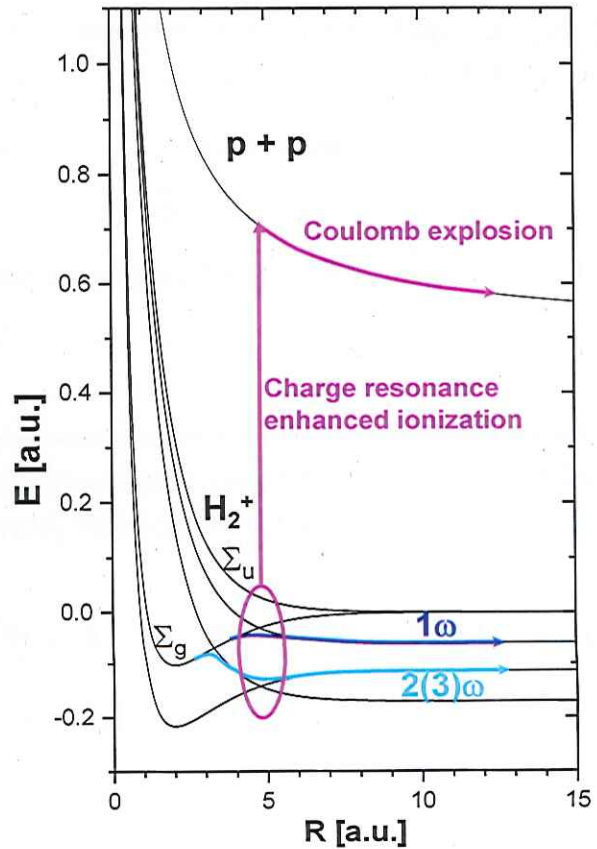


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# Dissociation and ionization paths



Dressed potential curves



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**H<sub>2</sub> / D<sub>2</sub>**

**Introduction**

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**Single-pulse results**

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- time - space imaging
- dephasing & revivals
- pump-control-probe
- shaping wave packets

**Quantum-beat analysis**

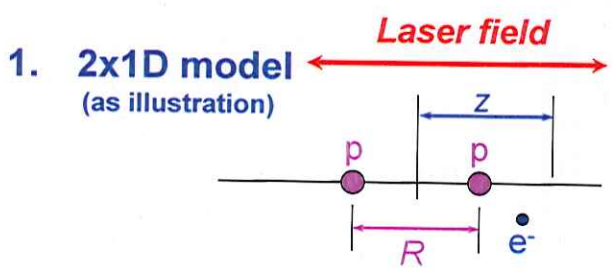
- frequency - space imaging
- potential & wf reconstruction

**N<sub>2</sub>, O<sub>2</sub>, CO, Ar<sub>2</sub>, .....**

**dissociation pathways**

**XUV pump - XUV probe**

**NUMERICAL METHODS**



$$\hat{H} = \hat{T}_R + 1/R + \hat{T}_z + \underbrace{V_{sce}}_{e\text{-nucleus}} + \underbrace{\mathbf{E}(t) \cos(\omega t) \cdot \mathbf{z}}_{laser}$$

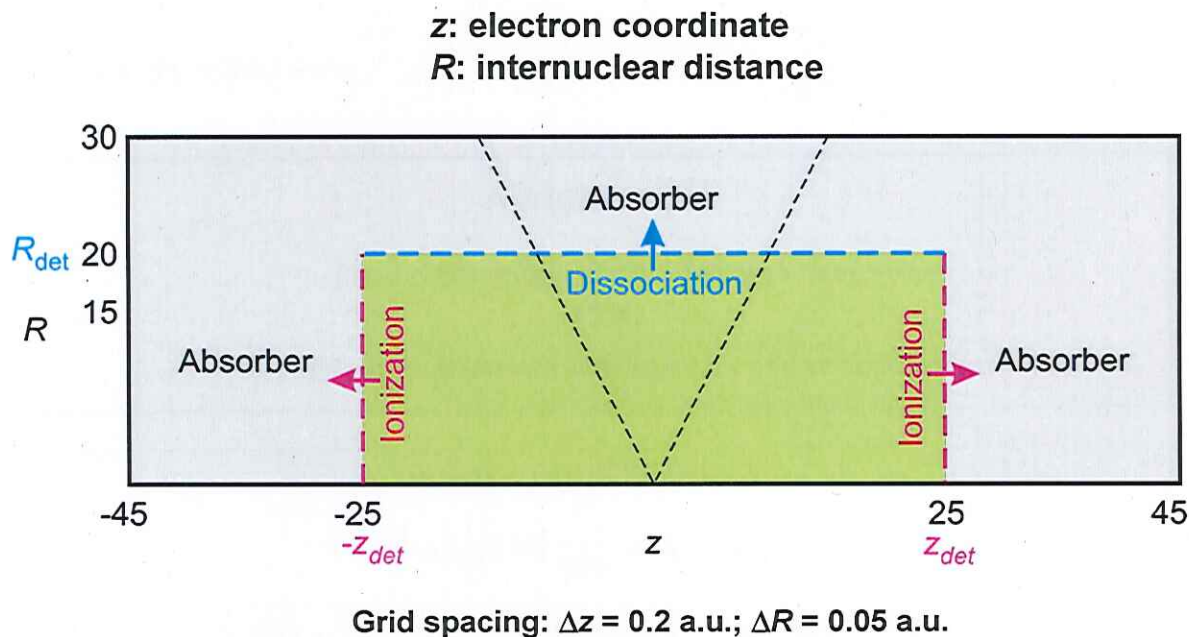
2D Crank - Nicholson split-operator propagation

$$\Psi(z, R, t + \Delta t) = e^{-i\hat{T}_R \Delta t/2} \cdot e^{-i(\hat{T}_z + 1/R + V_{sce} + V_{laser}) \Delta t} \cdot e^{-i\hat{T}_R \Delta t/2} \Psi(z, R, t) + O(\Delta t^3)$$

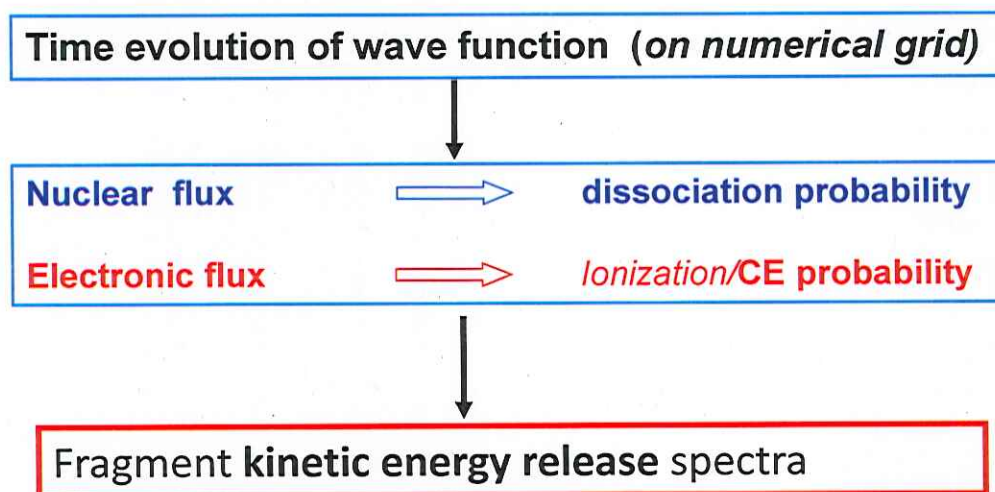
**2. vector Crank - Nicholson technique**  
propagation on coupled adiabatic potential curves

$$i \frac{d}{dt} \begin{pmatrix} \Psi_g(R, t) \\ \Psi_u(R, t) \end{pmatrix} = \begin{pmatrix} \hat{T}_R + V_g(R) - i\Gamma_g(R, t)/2 & D_{g-u}(R)E(t) \\ D_{g-u}(R)E(t) & \hat{T}_R + V_u(R) - i\Gamma_u(R, t)/2 \end{pmatrix} \begin{pmatrix} \Psi_g(R, t) \\ \Psi_u(R, t) \end{pmatrix}$$

# Num. grid for wave packet propagation "virtual detector" method



## "virtual detector" method



# “virtual detector”: data analysis

$$\Psi(z, R, t) = A(z, R, t) e^{i \cdot \phi(z, R, t)}$$

$$j_R(z, R, t) = \frac{\rho}{\mu} \frac{\partial}{\partial R} \phi(z, R, t) = \rho \cdot v_R, \quad \rho = |A(z, R, t)|^2$$

### Dissociation

$$p_R^{(D)}(z, t) = \frac{\partial}{\partial R} \phi(z, R_{\text{det}}, t)$$

Integration over z  $\Rightarrow$  fragment momentum distribution

### Coulomb explosion

$$p_R(z_{\text{det}}, t) = \frac{\partial}{\partial R} \phi(z_{\text{det}}, R, t)$$

$$p_R^{(CE)}(R, t) = \sqrt{p_R^2(z_{\text{det}}, R, t) + 2\mu/R}$$

Integration over R  $\Rightarrow$  fragment momentum distribution

## H<sub>2</sub> / D<sub>2</sub>

### Introduction

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### Single-pulse results

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### Pump-probe results

- time - space imaging
- dephasing & revivals
- pump-control-probe
- shaping wave packets

### Quantum-beat analysis

- frequency – space imaging
- potential & wf reconstruction

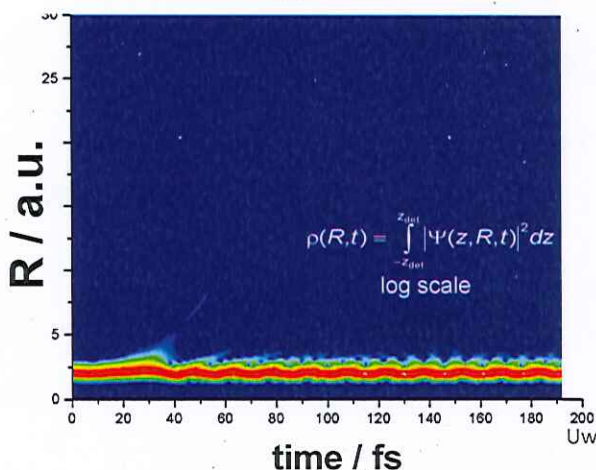
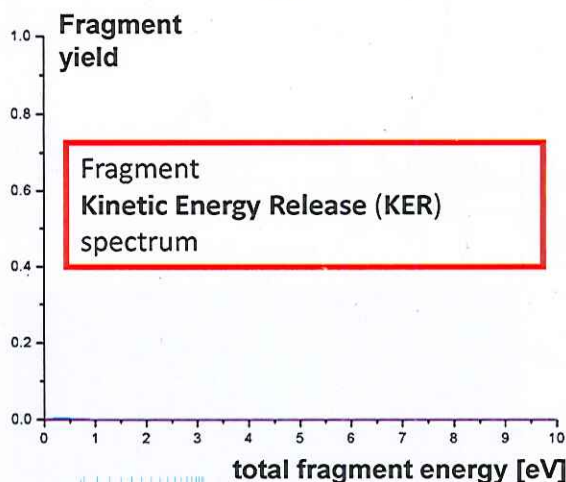
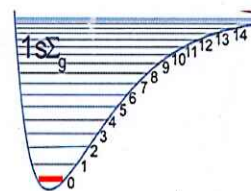
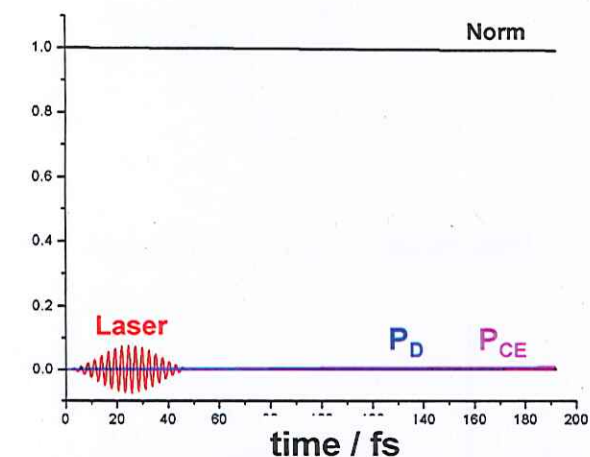
## N<sub>2</sub>, O<sub>2</sub>, CO, Ar<sub>2</sub>, ....

### dissociation pathways

### XUV pump – XUV probe

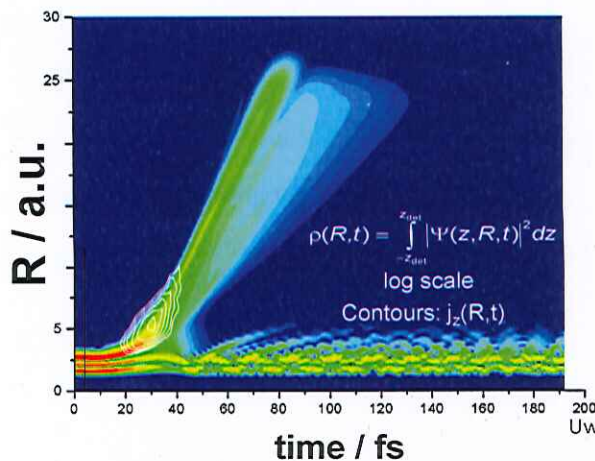
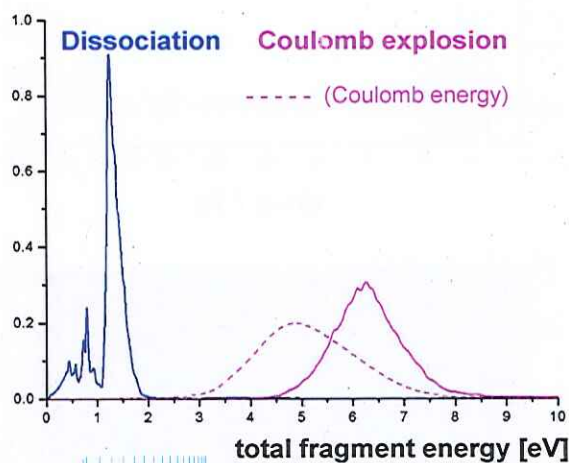
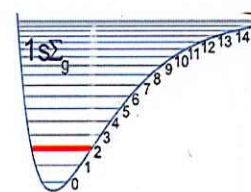
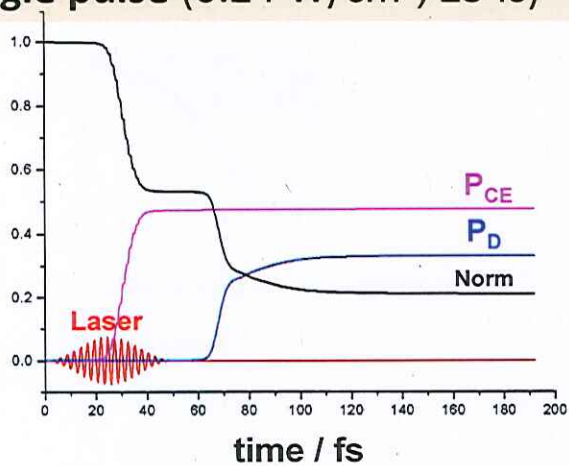
Single pulse (0.2 PW/cm<sup>2</sup>, 25 fs)

Initial state: H<sub>2</sub><sup>+</sup>(v = 0)



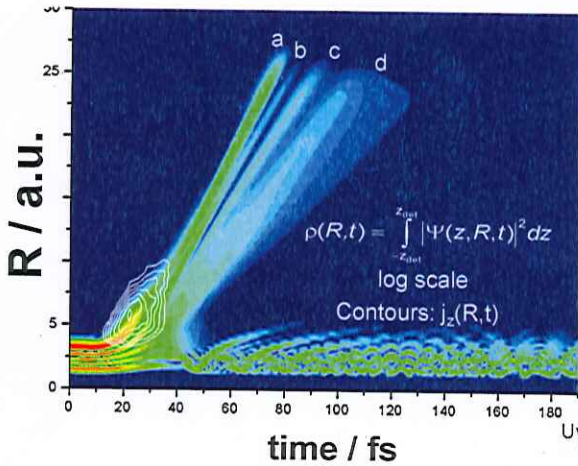
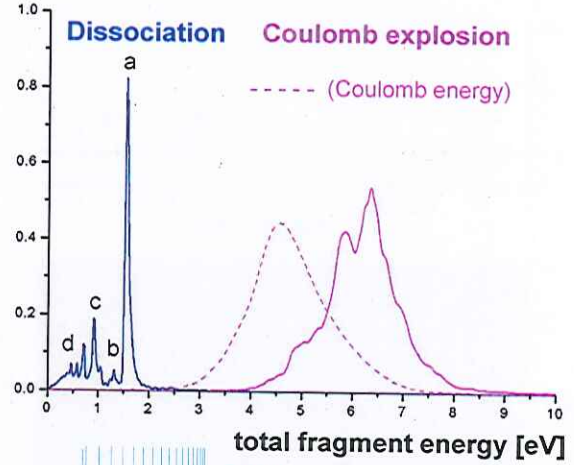
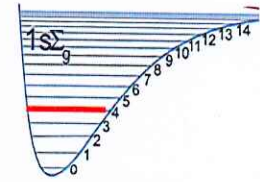
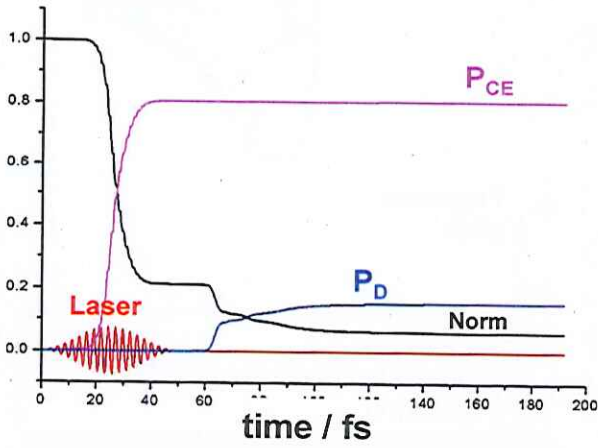
Single pulse (0.2 PW/cm<sup>2</sup>, 25 fs)

Initial state: H<sub>2</sub><sup>+</sup>(v = 2)



### Single pulse (0.2 PW/cm<sup>2</sup>, 25 fs)

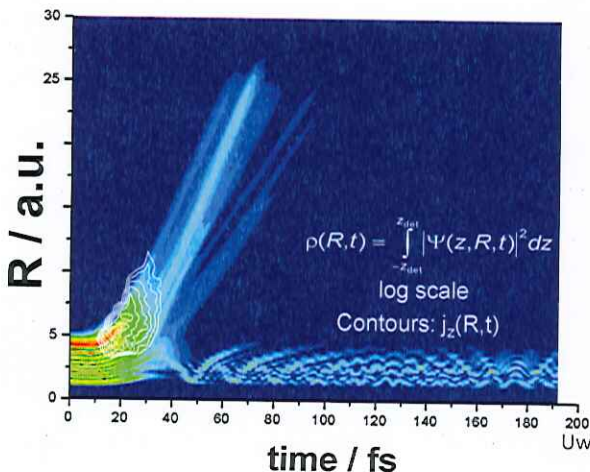
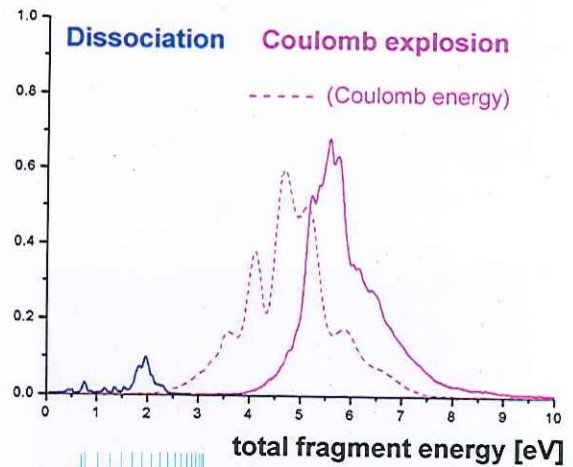
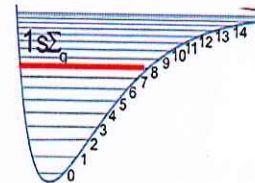
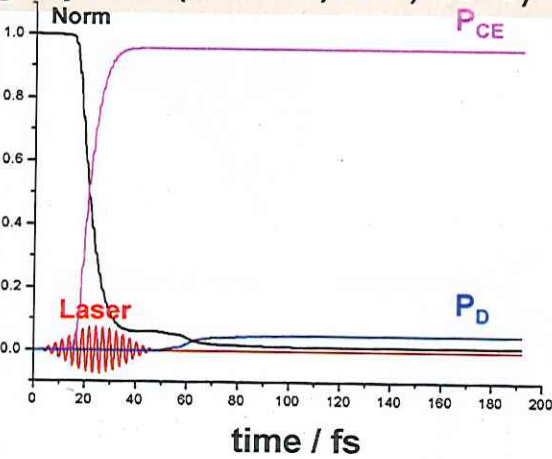
Initial state: H<sub>2</sub><sup>+</sup>(v = 4)



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### Single pulse (0.2 PW/cm<sup>2</sup>, 25 fs)

Initial state: H<sub>2</sub><sup>+</sup>(v = 8)

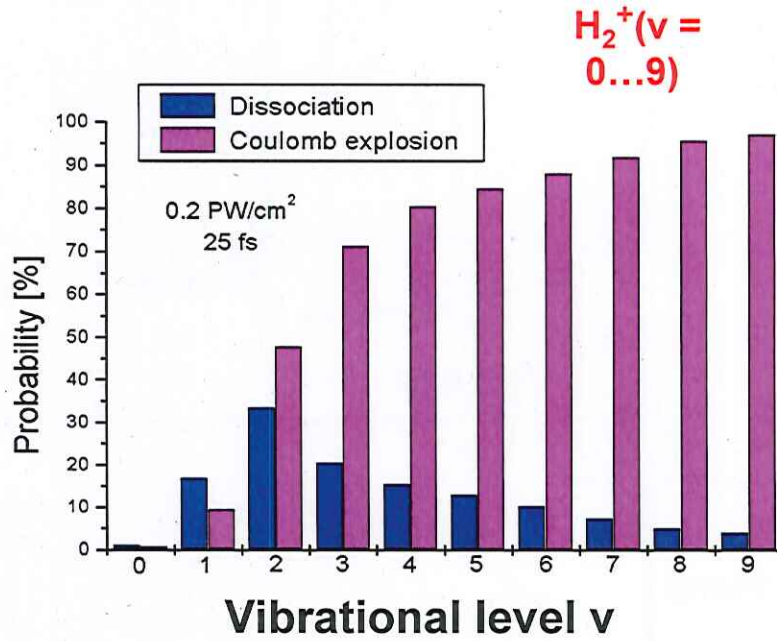


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# Branching ratio : Dissociation vs. Coulomb explosion

Single pulse (0.2 PW/cm<sup>2</sup>, 25 fs)



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## H<sub>2</sub> / D<sub>2</sub>

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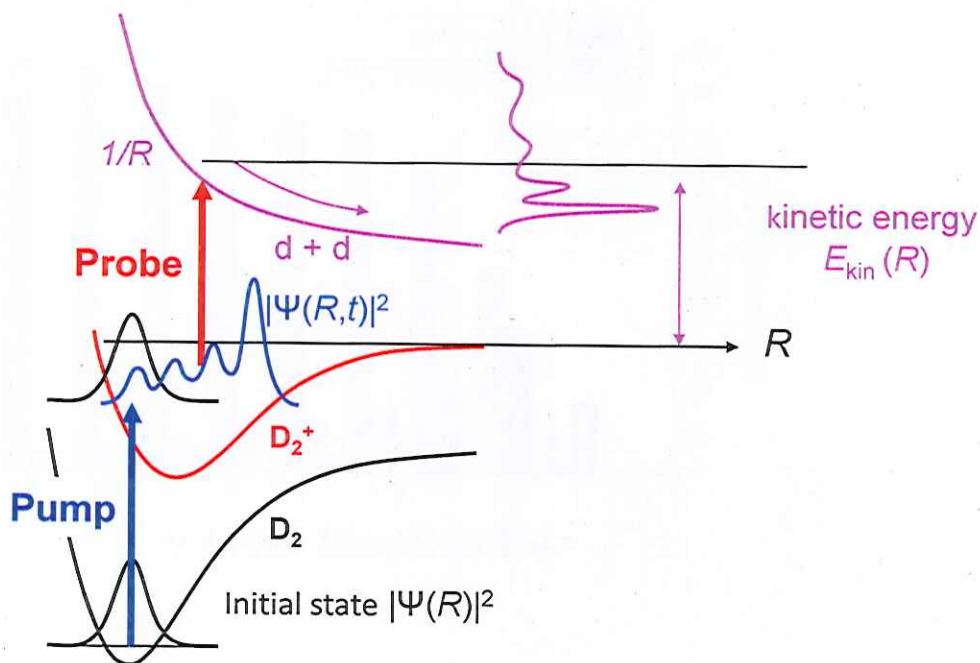
### Quantum-beat analysis

- frequency - space imaging
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## N<sub>2</sub>, O<sub>2</sub>, CO, Ar<sub>2</sub>, ...

### dissociation pathways

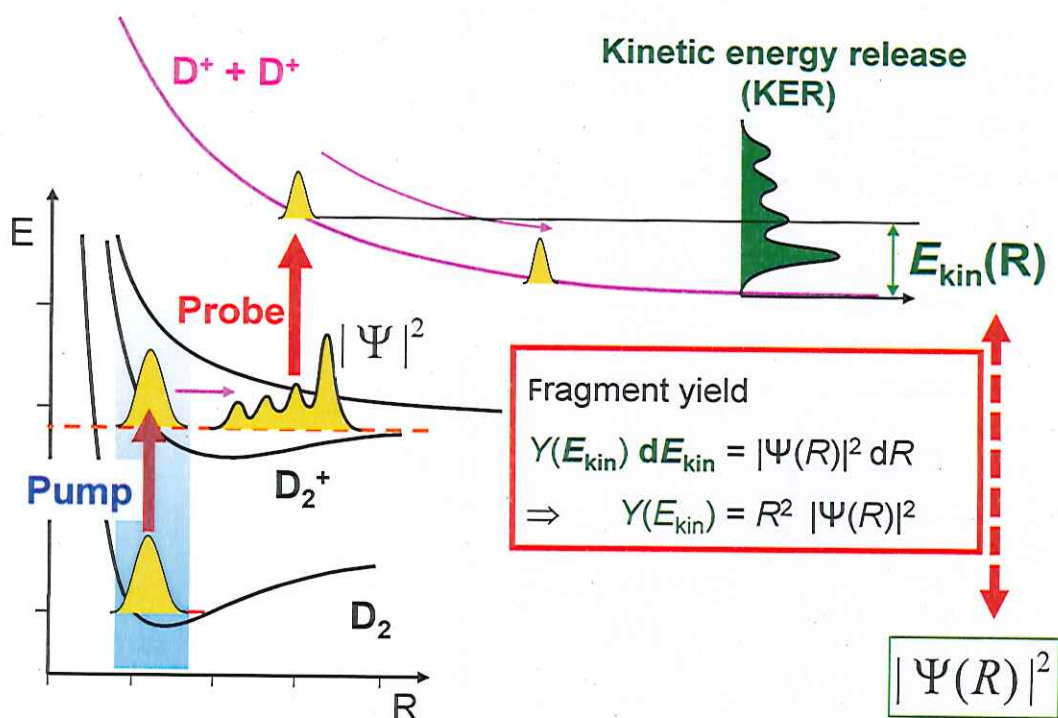
### XUV pump - XUV probe



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Coulomb-explosion imaging of dissociating & bound nuclear motion



Feuerstein, Thumm, PRA 67, 063408 ('03)

Thumm et al., PRA 77, 065401 ('08)

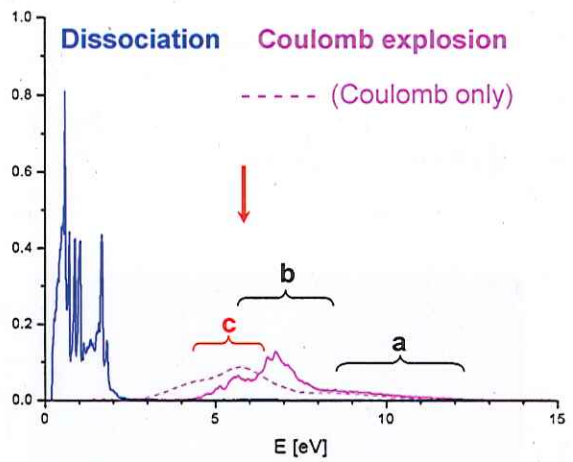
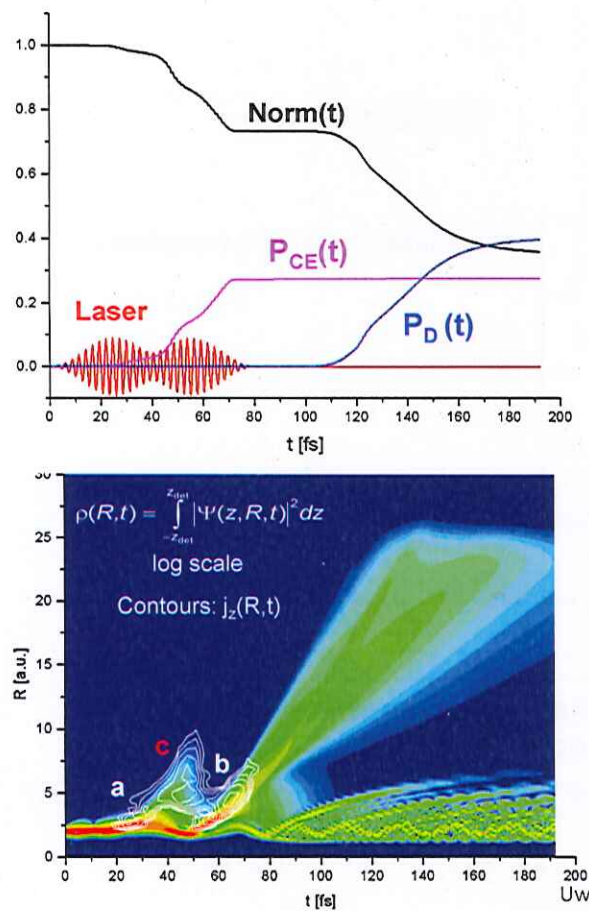
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# Pump-probe: D<sub>2</sub>, 0.3 PW/cm<sup>2</sup>, 2 x 25 fs

Initial state: D<sub>2</sub><sup>+</sup>(v = 0)

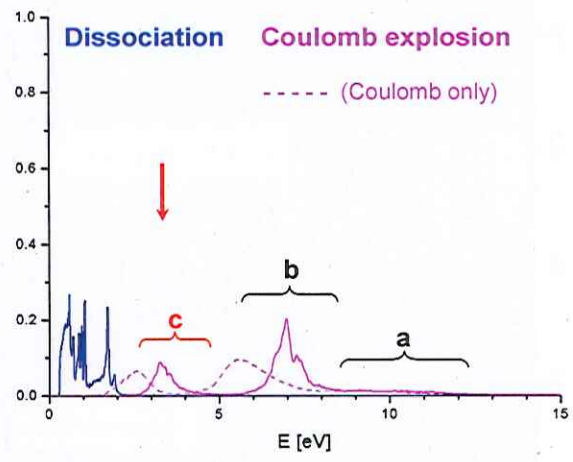
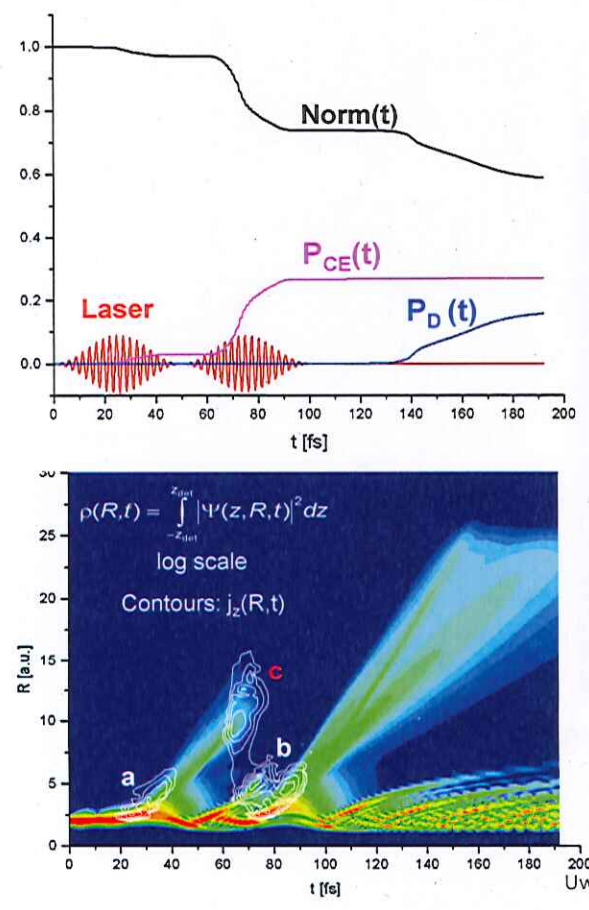
delay 30 fs



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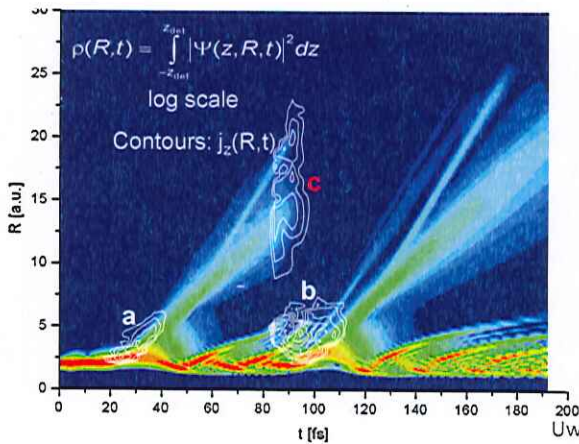
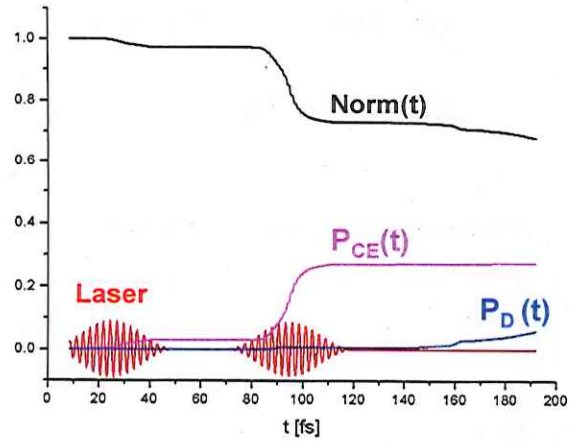
# Pump-probe: D<sub>2</sub>, 0.3 PW/cm<sup>2</sup>, 2 x 25 fs

delay 50 fs



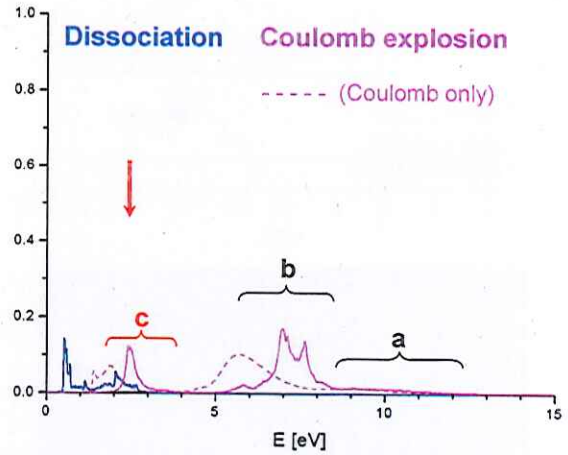
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**Pump-probe: D<sub>2</sub>, 0.3 PW/cm<sup>2</sup>, 2 x 25 fs**



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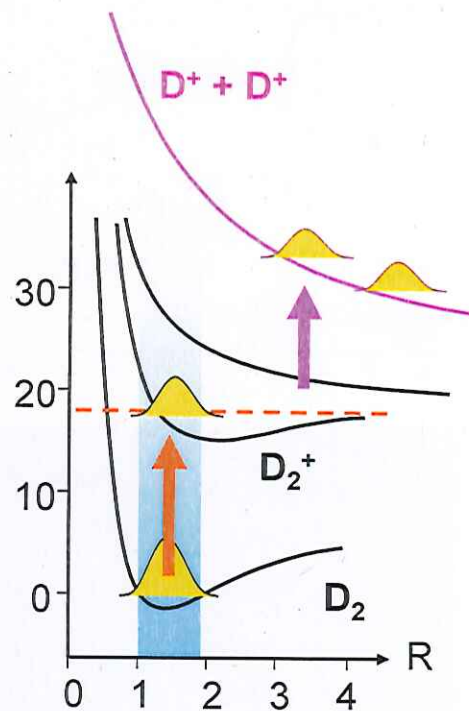
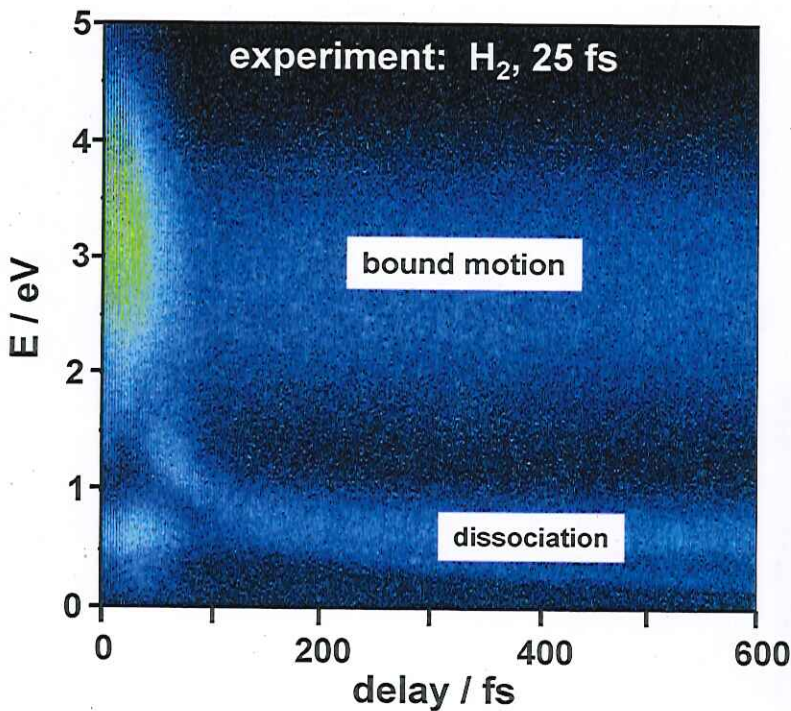
**delay 70 fs**



**Qual. agreement with experiment:**  
 (D<sub>2</sub>, 0.1 PW/cm<sup>2</sup>, 2 x 80 fs, 0 - 300 fs delay)  
 Trump, Rottke, Sandner, PRA **59**, 2858 (1999)

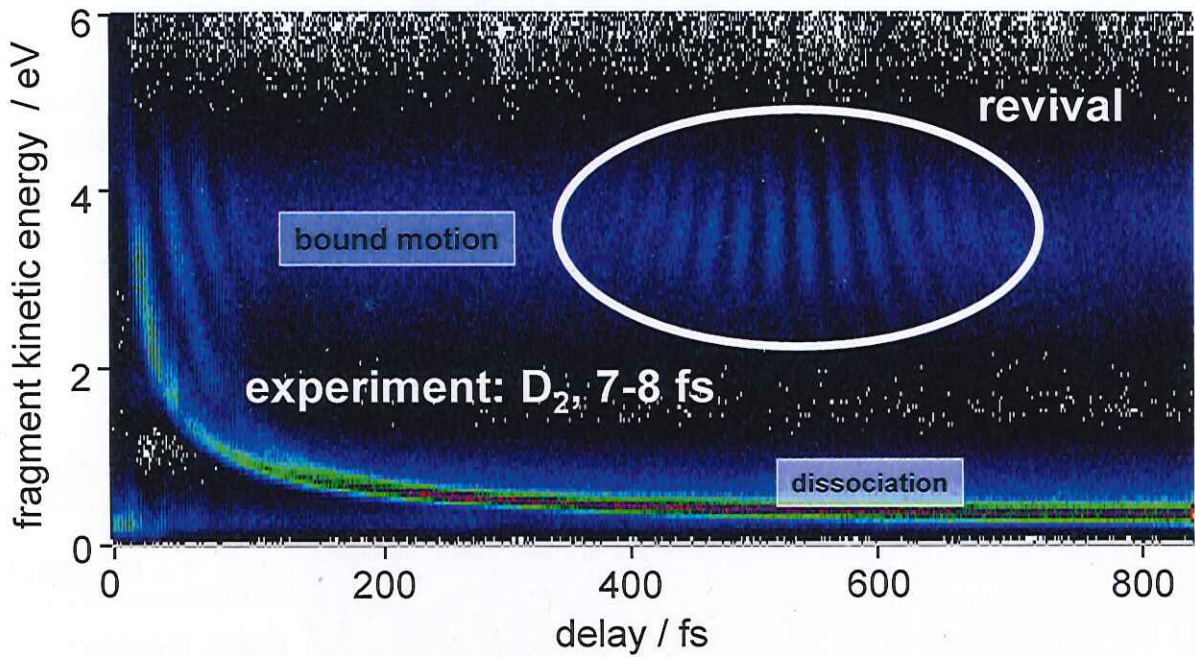
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**Pump-probe experiment: H<sub>2</sub> + 25 fs IR pulses**



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**“Revival”** of the vibrational wave packet

Ergler, Moshhammer, Ullrich, et al. PRL 97, 193001 (2006)

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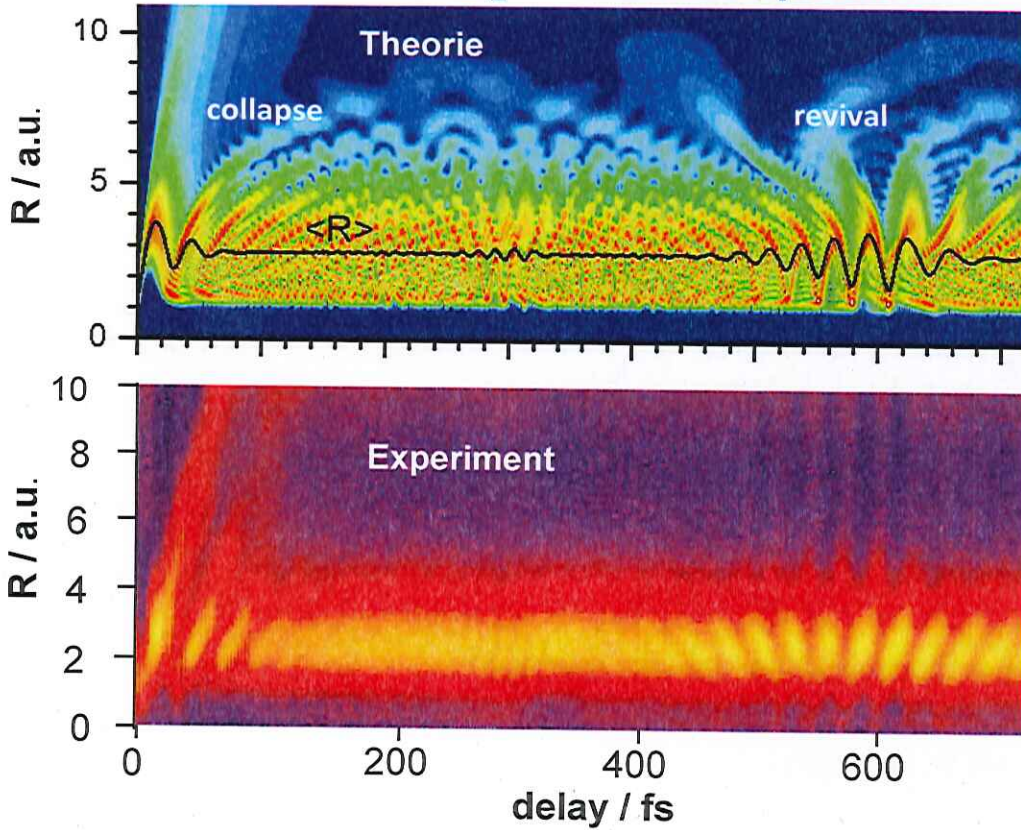
**Pump-probe experiment:  $D_2$  + fs pulses**

	period
Grandfather' clock:	1s
Cs atomic clock:	$10^{-10}$ s
<b>Molecular clock:</b>	<b><math>2 \times 10^{-14}</math> s</b>



# Time-resolved nuclear motion in $D_2^+$

## $D_2 + 7-8$ fs IR pulses



**Theory:**  
Feuerstein & Thumm  
PRA 67, 063408 (2003)

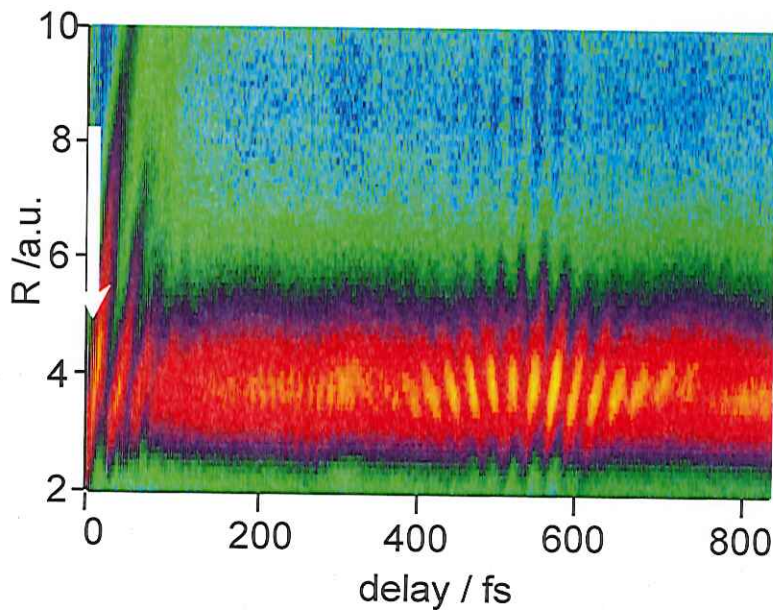
**Experiment:**  
Fragment yield:  
 $Y(E_{kin}) = R^2 |\phi(R)|^2$

Ergler, Moshhammer,  
Ullrich, et al.  
PRL 97, 193001 (2006)

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## The sound of $D_2^+$



Nuclear wave packet:

$$\Psi(R,t) = \sum_{\nu} a_{\nu} \chi_{\nu}(R) e^{-i\omega_{\nu}t}$$

↓ 23 fs ↔ 1 s  
 $\omega_0 = 440$  Hz

Audio:

$$f(t) = \left| \sum_{\nu} a_{\nu} \sin(\omega_{\nu} + \omega_0)t \right|^2$$



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