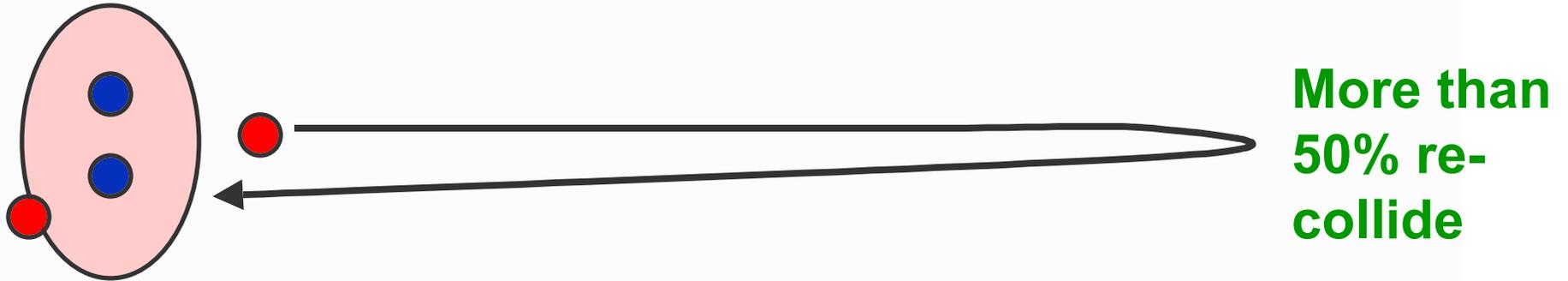


X-rays vs electrons

- 4th generation light source 10^6 photons / Å
single molecule imaging
- X-rays from conventional lasers will never be able to compete
- But lasers can generate <1 Å **electrons**
conveniently
with high current density ($\sim 10^{-7}$ to 10^{-8} X-ray density)
short duration
- For diffraction, electrons are much more effective than photons.

The key Idea



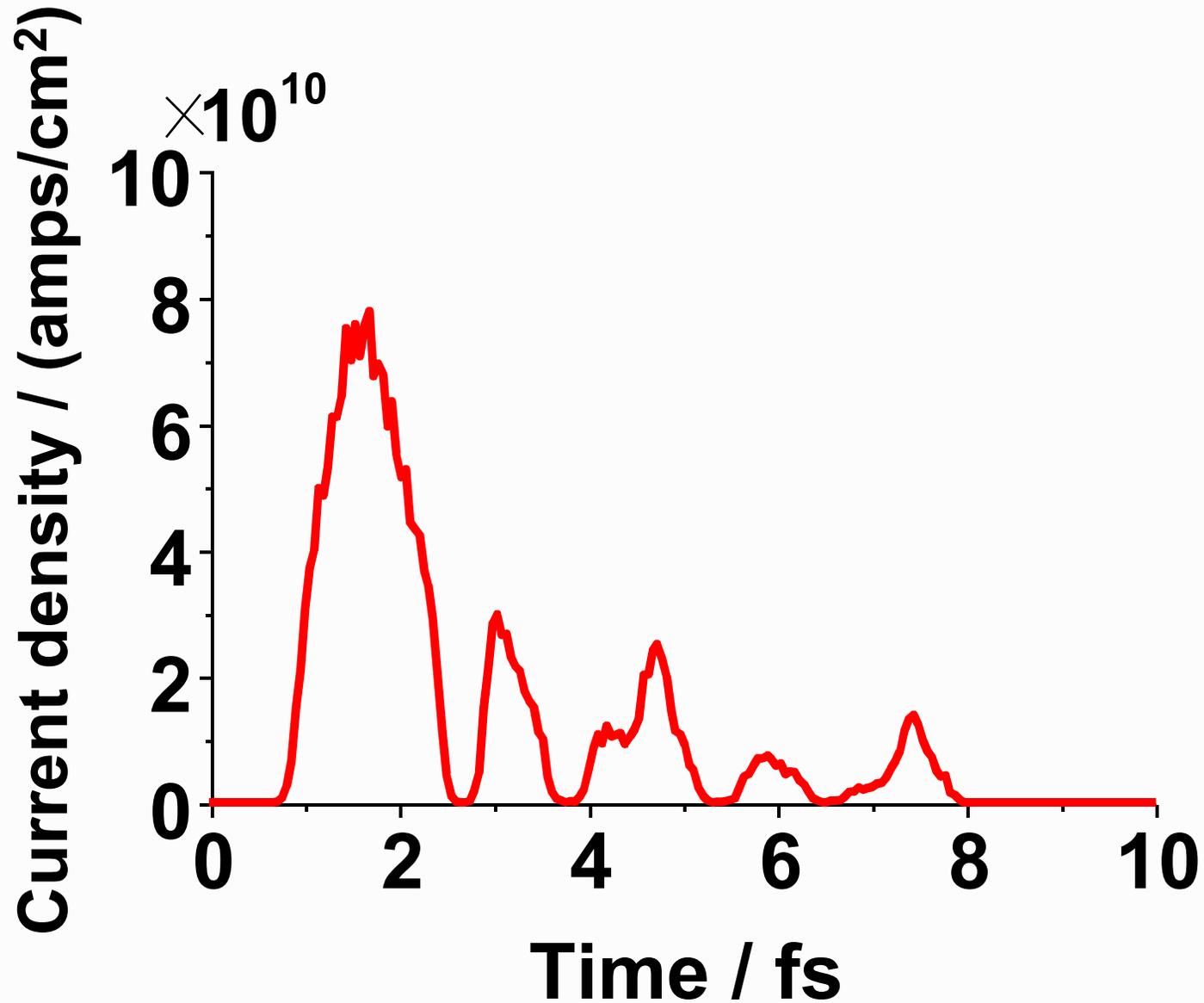
$J \sim 10^{11}$ Amps/cm²; $\tau \sim 1$ fs;

Energy ~ 100 -1000 eV; $\lambda \sim 1.2$ Å - 0.4 Å

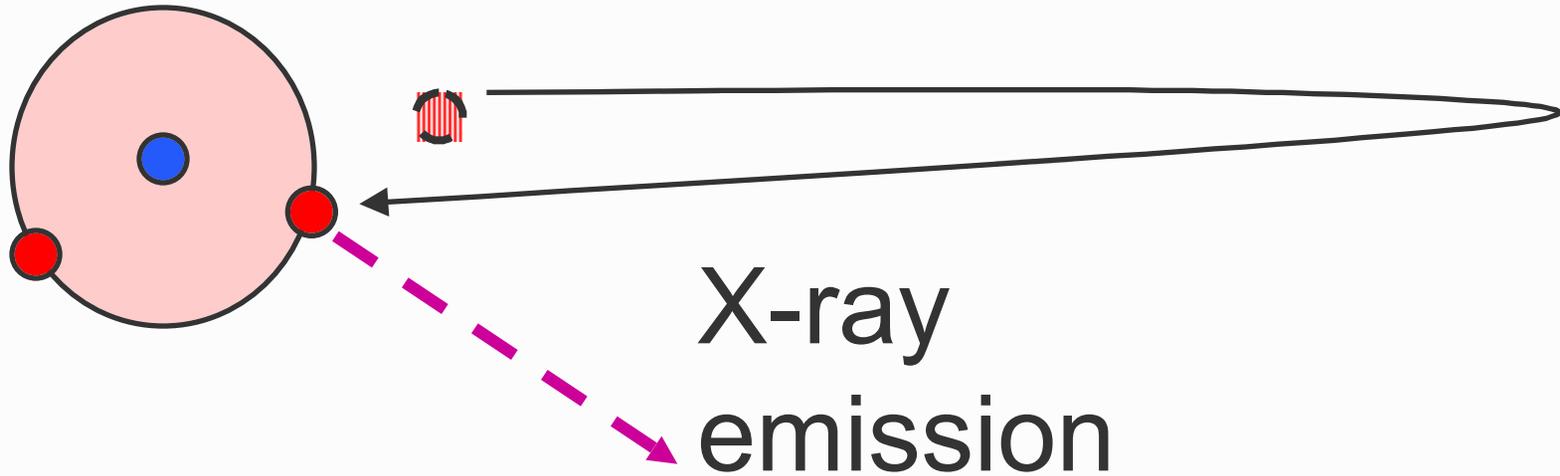
such large current densities are unavailable from *any other source*

To get the same wavelength in X-rays requires >10keV photons

Calculated |Current Density|



Attosecond Optical Pulses

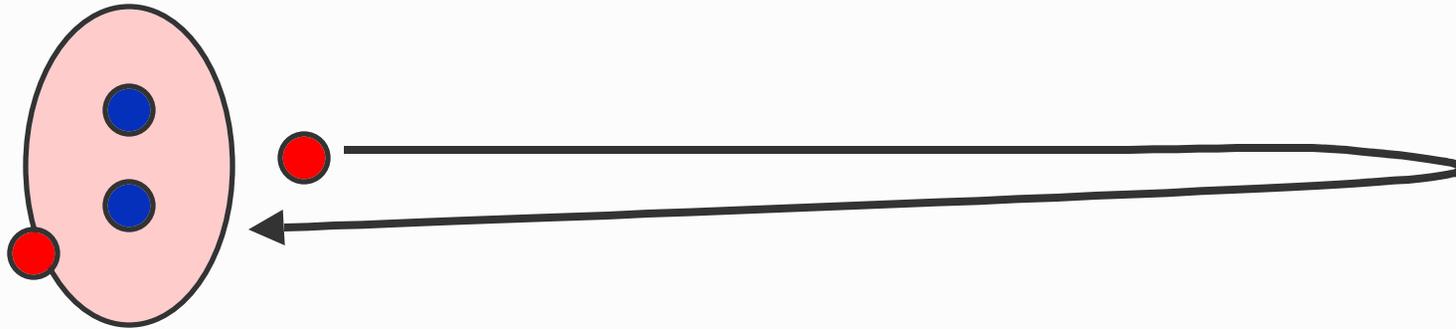


Attosecond pulses are

- produced inefficiently
- focused poorly
- interact weakly

Perhaps the electrons are as important ----- or more important ----- than optical pulses they produce

Outline



More than
50% re-
collide

- Measuring: $J=q/\text{Area}/\text{time}$

the area ---- $\sim 50 \text{ \AA}^2$

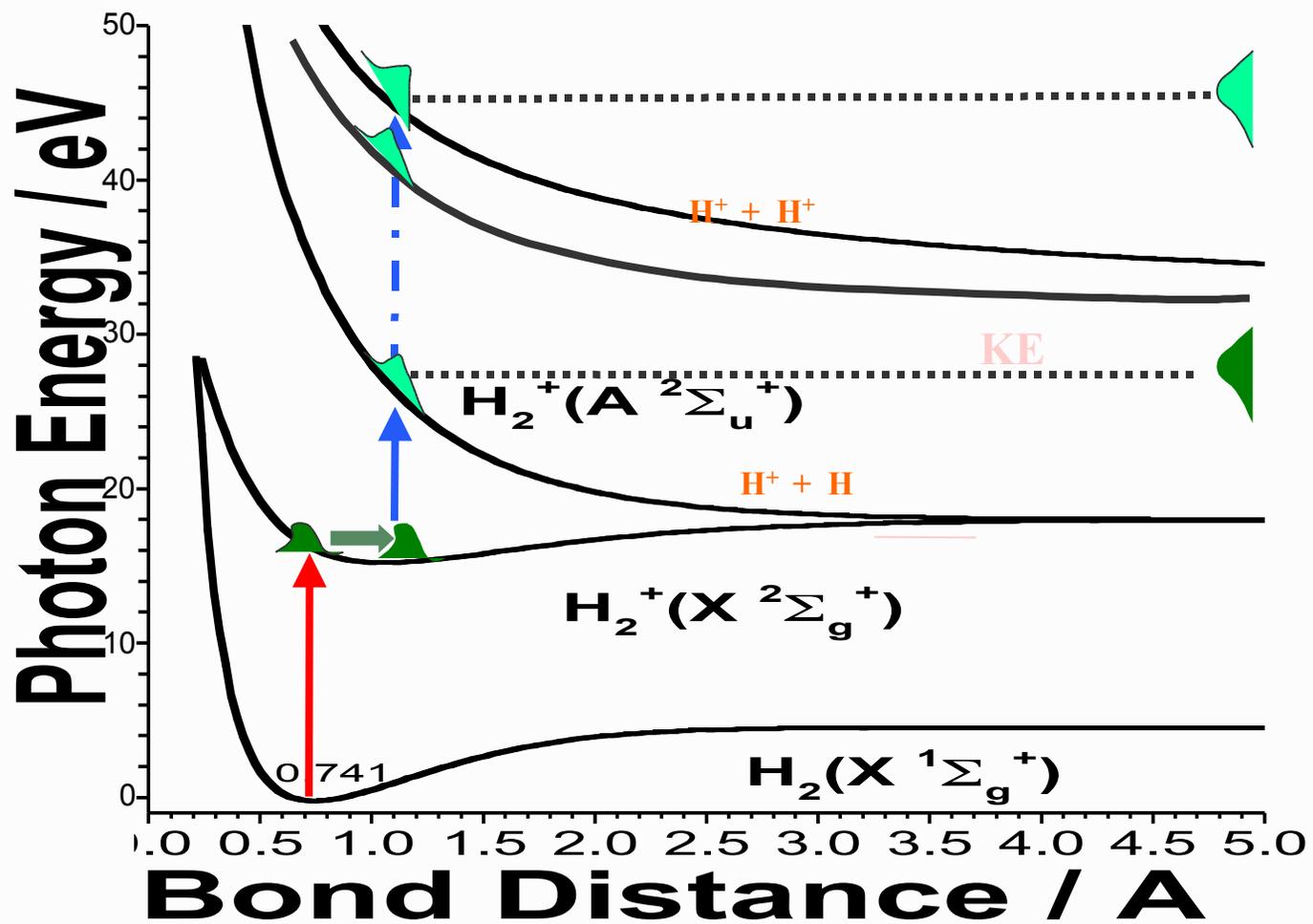
the time ---- $\sim 1 \text{ fs}$

- Consequences:

short pulses are not essential for fast measurements

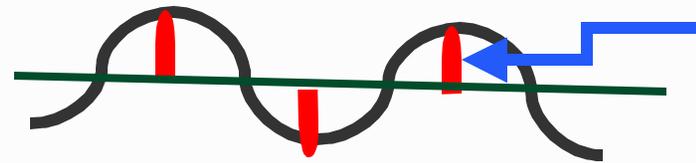
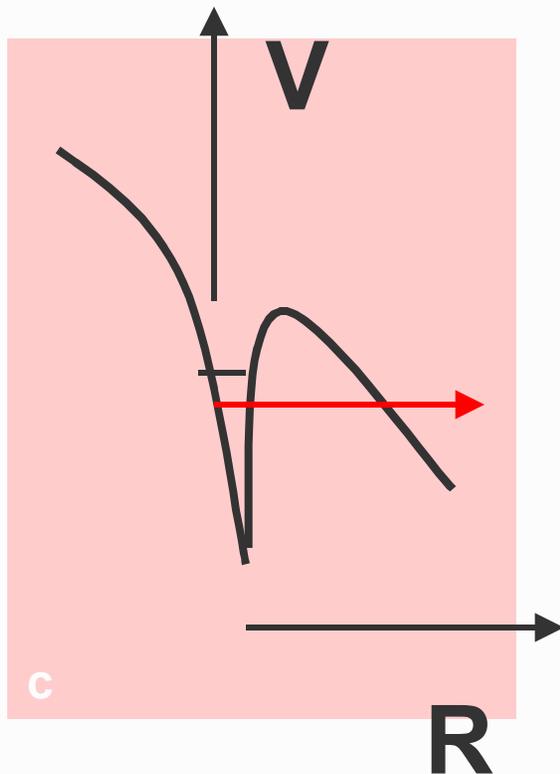
electron diffraction from parent molecule

Measuring time: Molecular clock



Strong Field Ionization

$$V(x,t) = V(x) - xE \cos \omega t$$

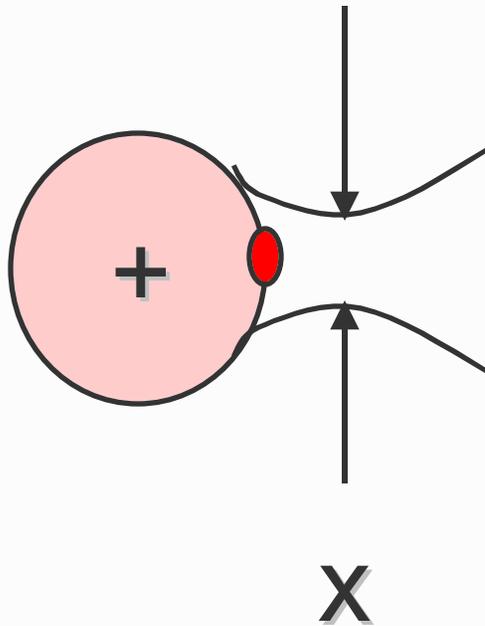


**Ionization
occurs in
bursts**

$$\omega(t) = 4\omega_0 \frac{E_i}{E_h}^{5/2} \frac{E_a}{E(t)} \exp \left[-\frac{2}{3} \frac{E_i}{E_h} \frac{E_a}{E(t)} \right]$$

**The ionization rate reaches about 10^{13}
at E about $1/20 E_a$**

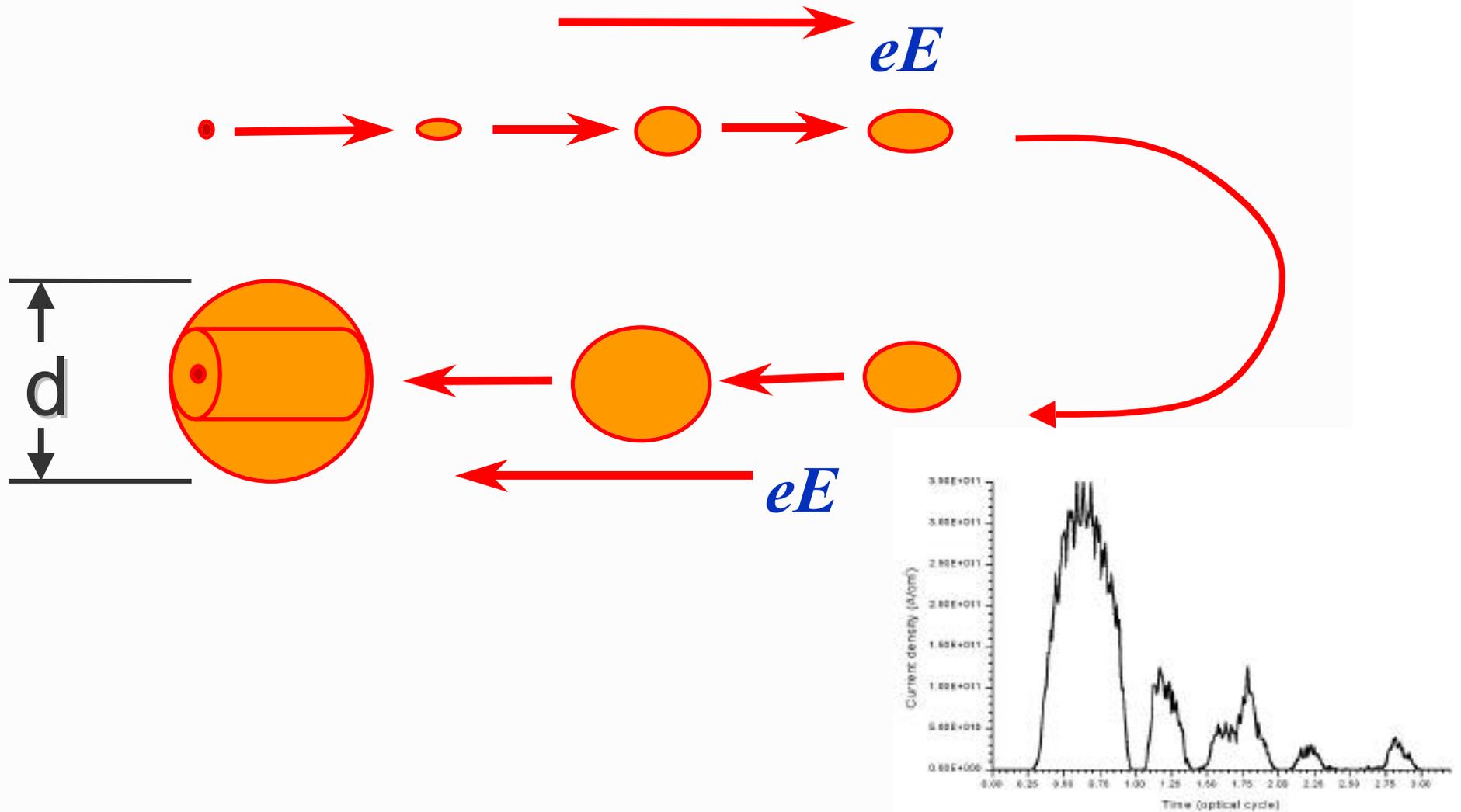
The initial conditions for the electron



$$p \sim h / x$$

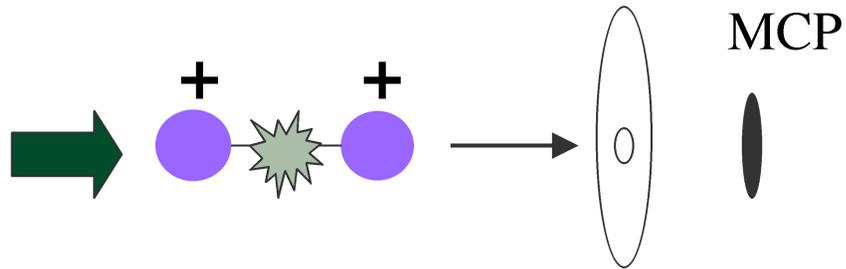
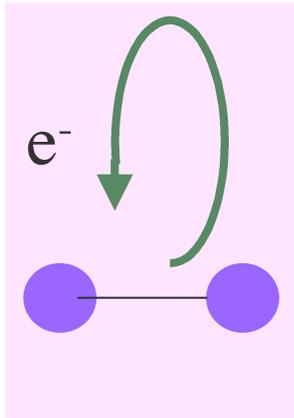
Classical physics mostly determines $V_{//}$

Electron motion

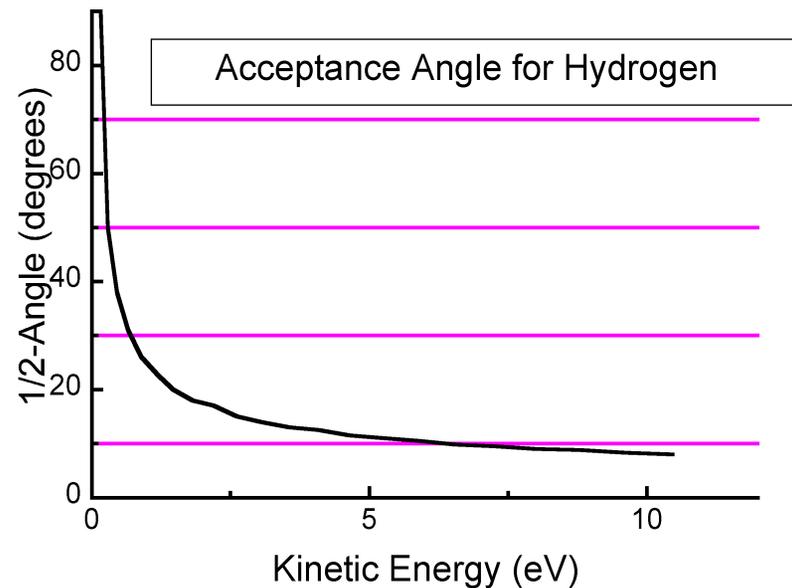
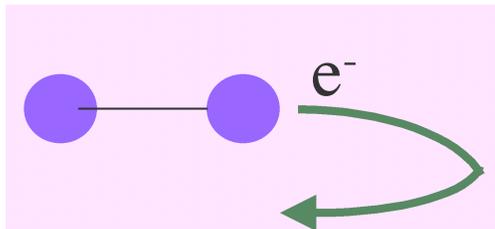


Measuring sub-cycle electron dynamics

Perpendicular

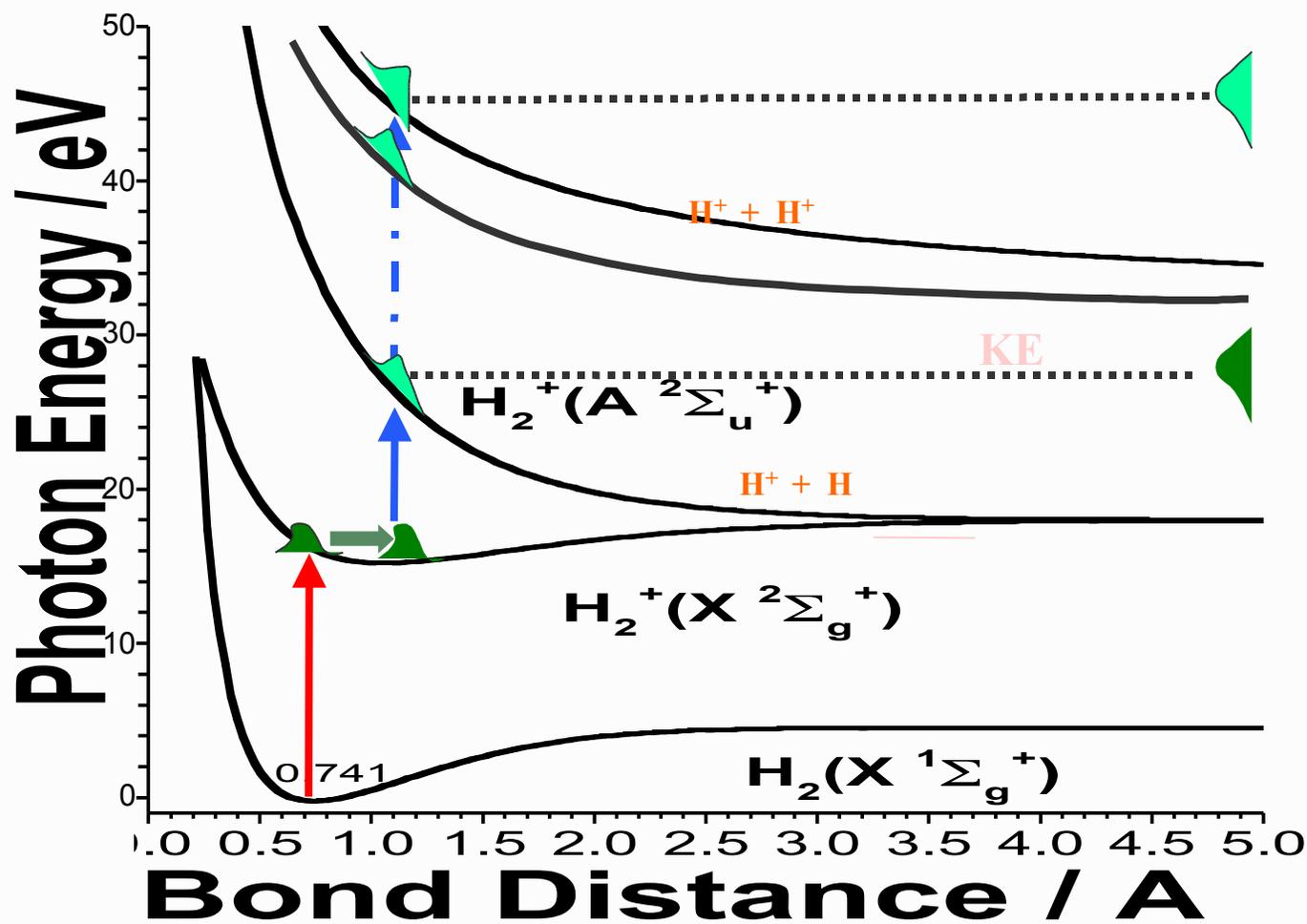


Parallel



The acceptance angle is about 8° for 10 eV protons

In H_2 an electron wavepacket is always formed with a Vibrational Wavepacket

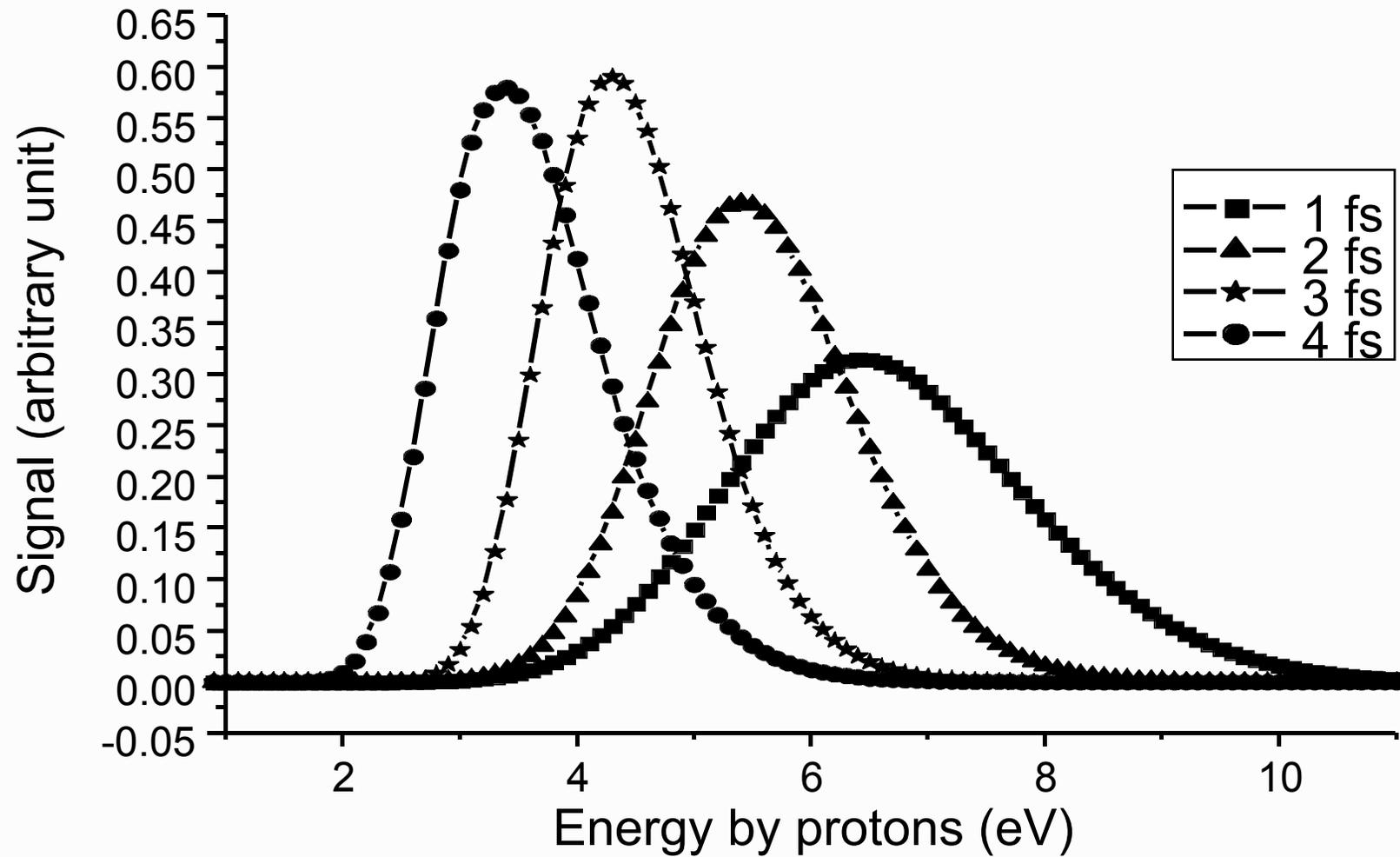


Potential Energy surface of H_2

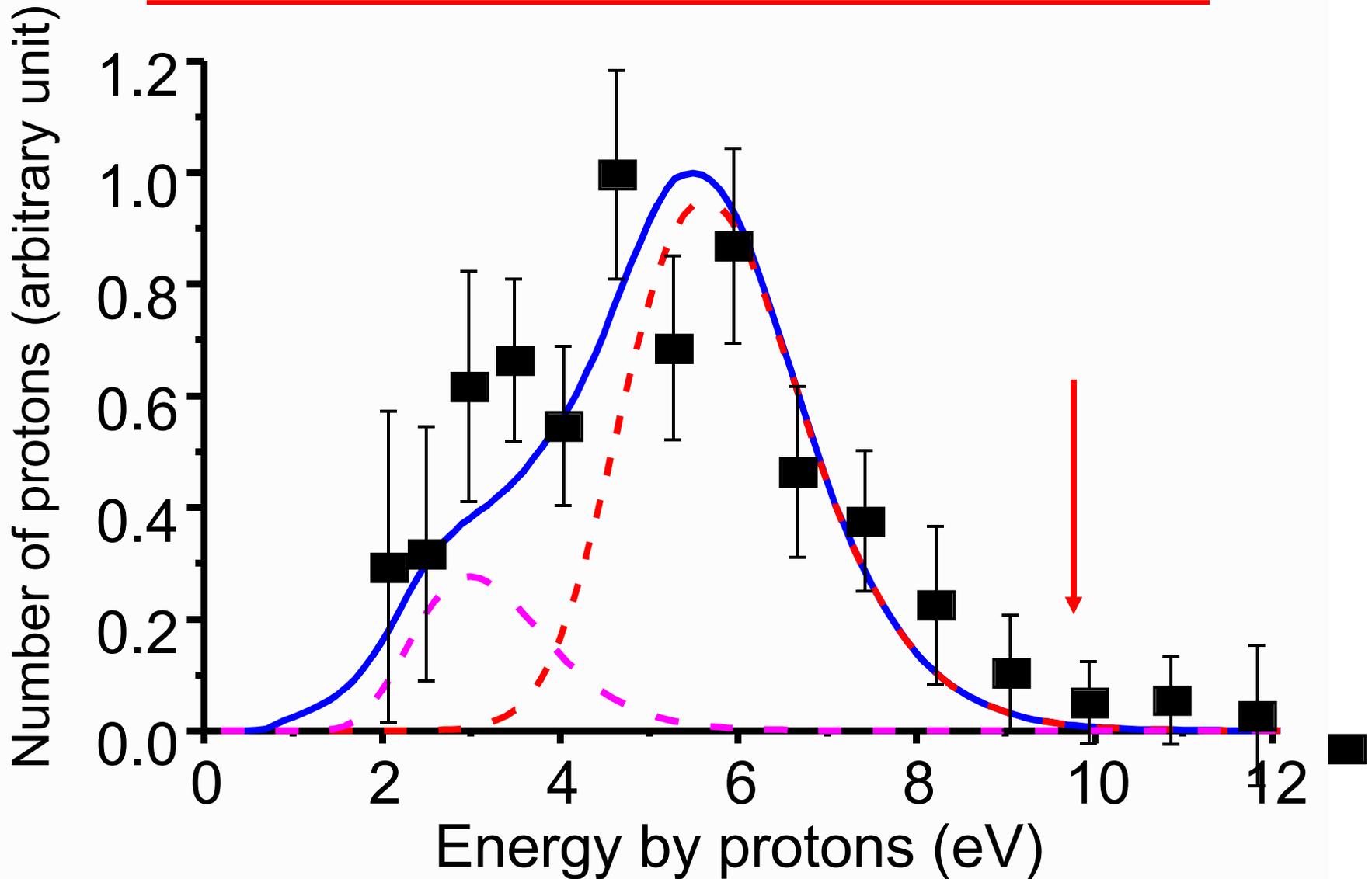
Wave packet motion

- ☞ Ionization forms a Σ_g wave packet
- ☞ *slightly* distorted from the ground state wave function of H_2 --- due to the radial dependence of the ionization potential
- ☞ The wave packet evolves until the re-collision electron excites H_2^+ according to published cross-sections
- ☞ *We predict the kinetic energy of the fragments, adding each re-collision incoherently*

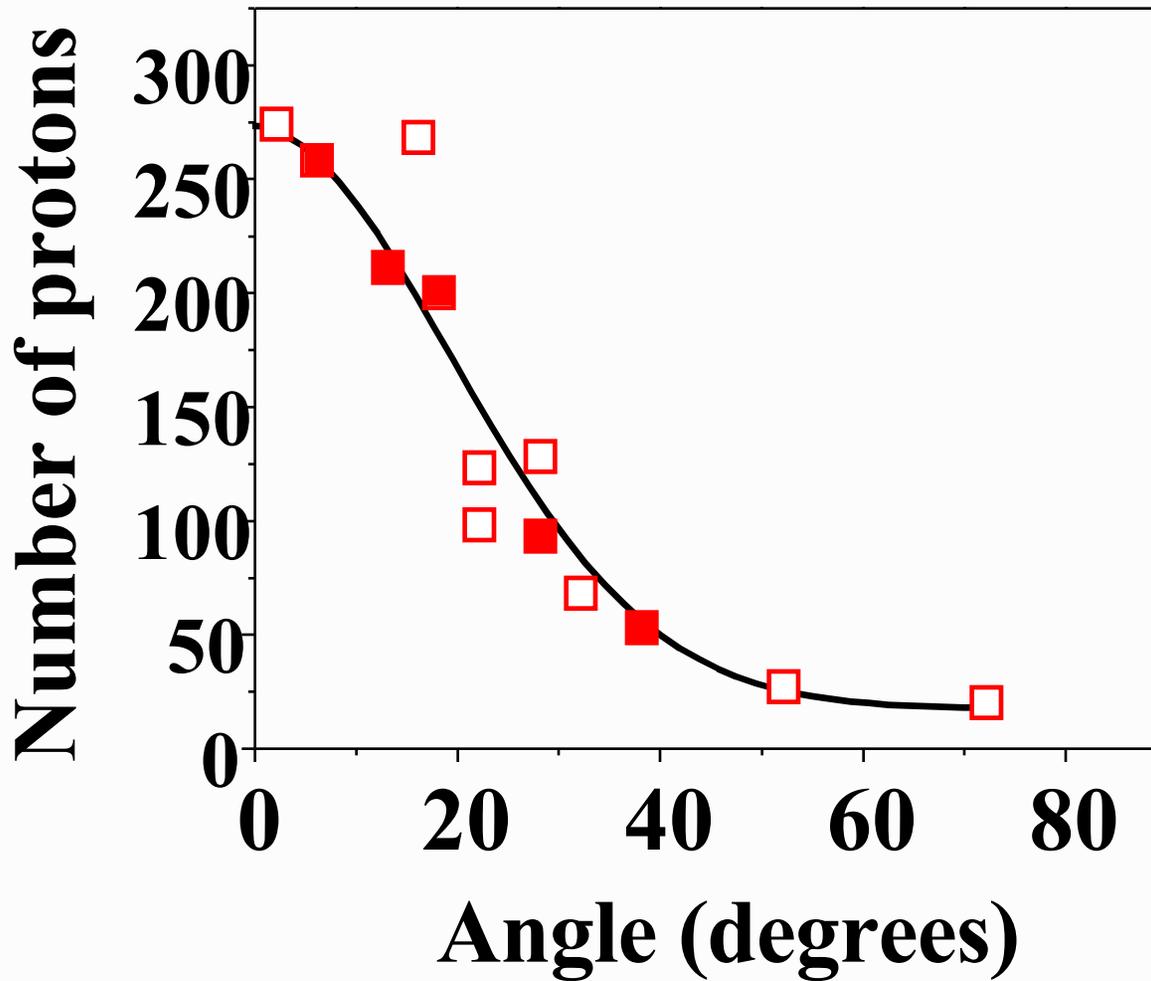
Time Resolution of Molecular Clock



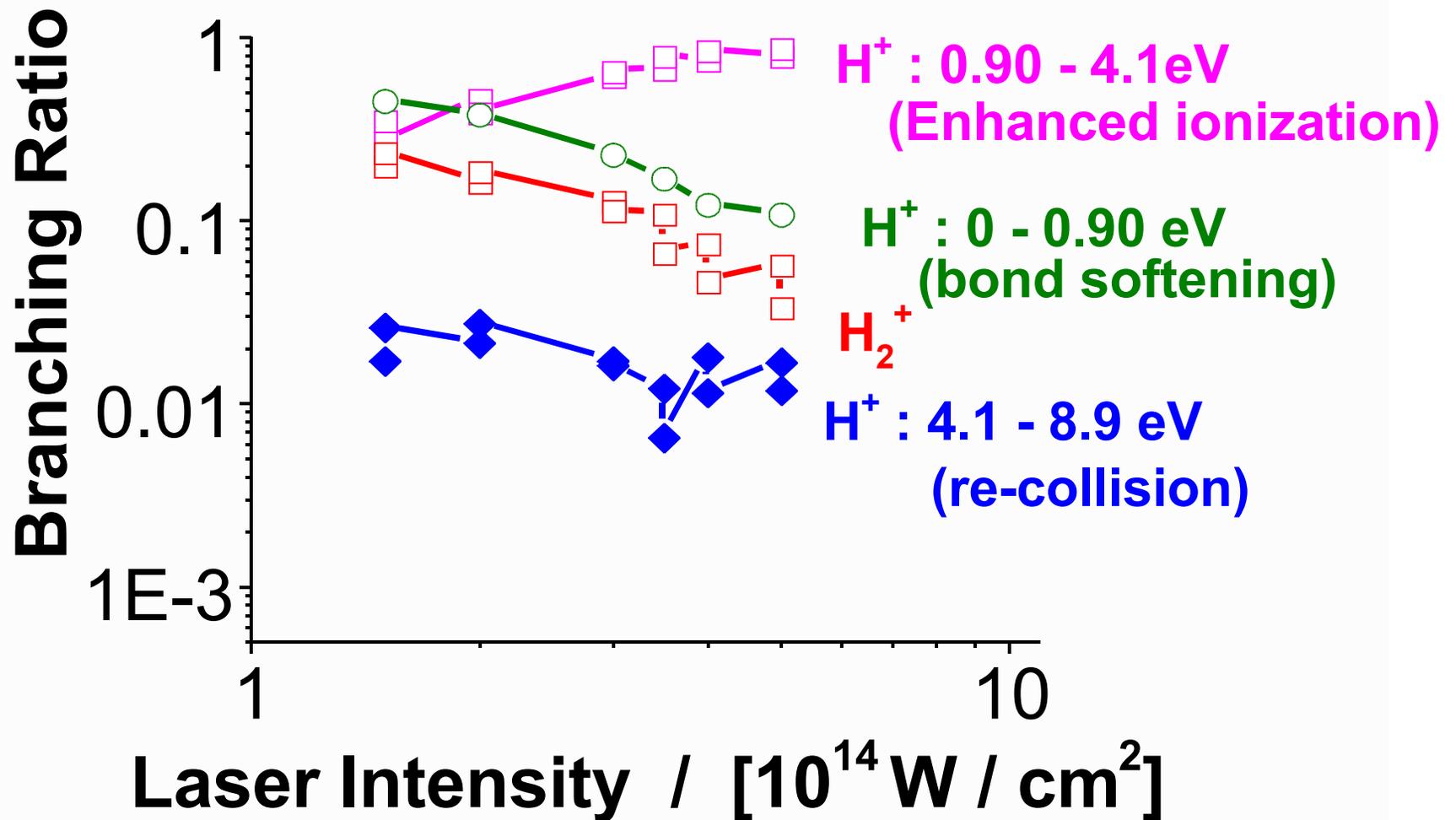
$<10^{-4}$ probability of instantaneous double ionization



Alignment dependence



Confirming the magnitude of the current



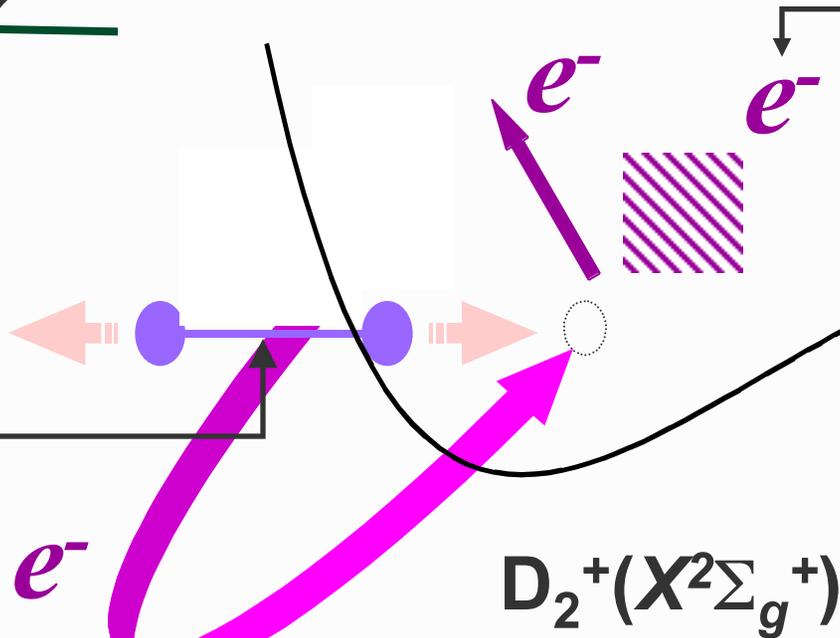
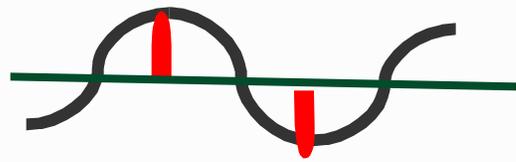
Attosecond electrons

- ☛ **Attosecond** electron bunches will gain equal footing with attosecond optical pulses in attosecond science -- fastest vibrational wavepacket motion ever observed.
- ☛ **Femtosecond** science has *no convenient short wavelength source for either pumping or probing*. Re-collision electrons can play that role.
 - Elastic scattering ---- Electron diffraction
 - Inelastic scattering
- ☛ **Single molecule imaging** appears to be an important enough goal to justify the 4th generation light source. Re-collision electrons may have a role by allowing *few molecule, gas phase imaging*.

Measuring D_2^+ vibrational wavepackets with sub-femtosecond precision

- ☞ For the past 30 years each advance in fast measurement required shorter pulses.
- ☞ **Exploiting correlation is an alternate approach --- *the evolution of the correlated partners after their formation largely determines the resolution***
- ☞ ***sub-femtosecond resolution over the first 5 fs following the formation of a D_2^+ vibrational wavepacket --- using a 50-100 fs pulses***

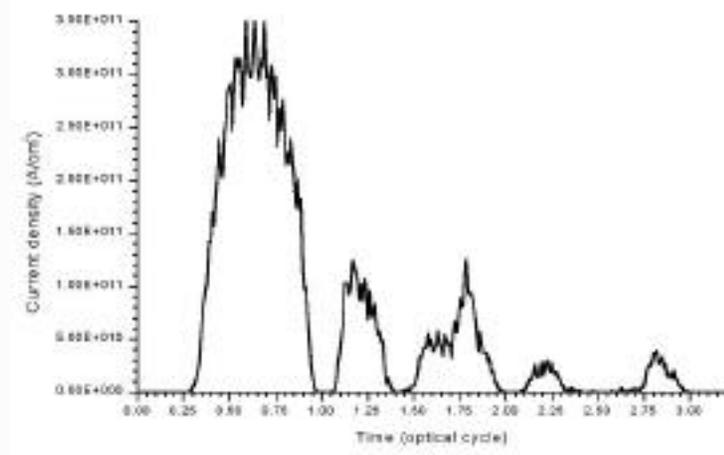
Using correlated wavepackets for dynamics measurements



Pump step

delay

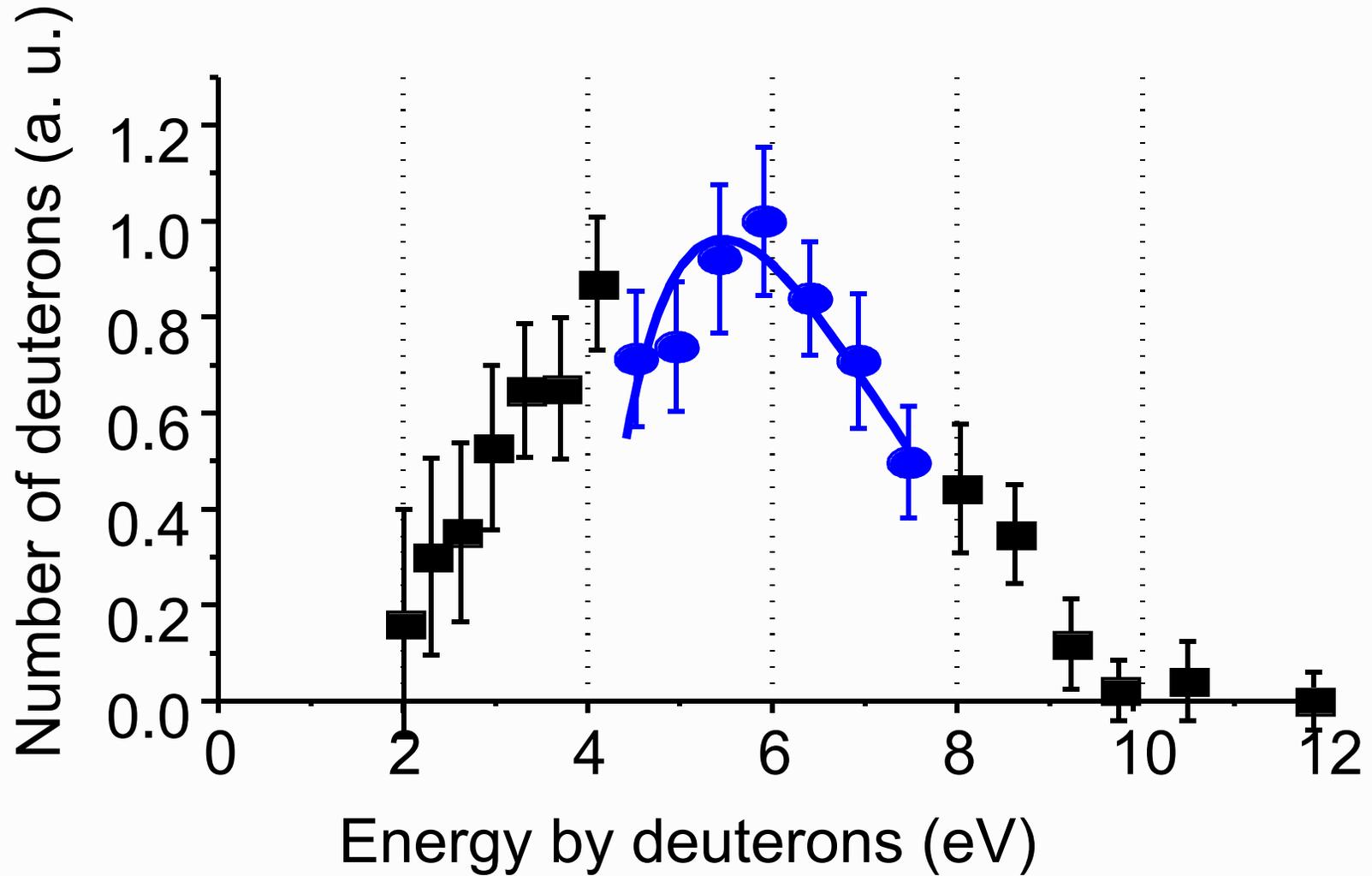
Probe step



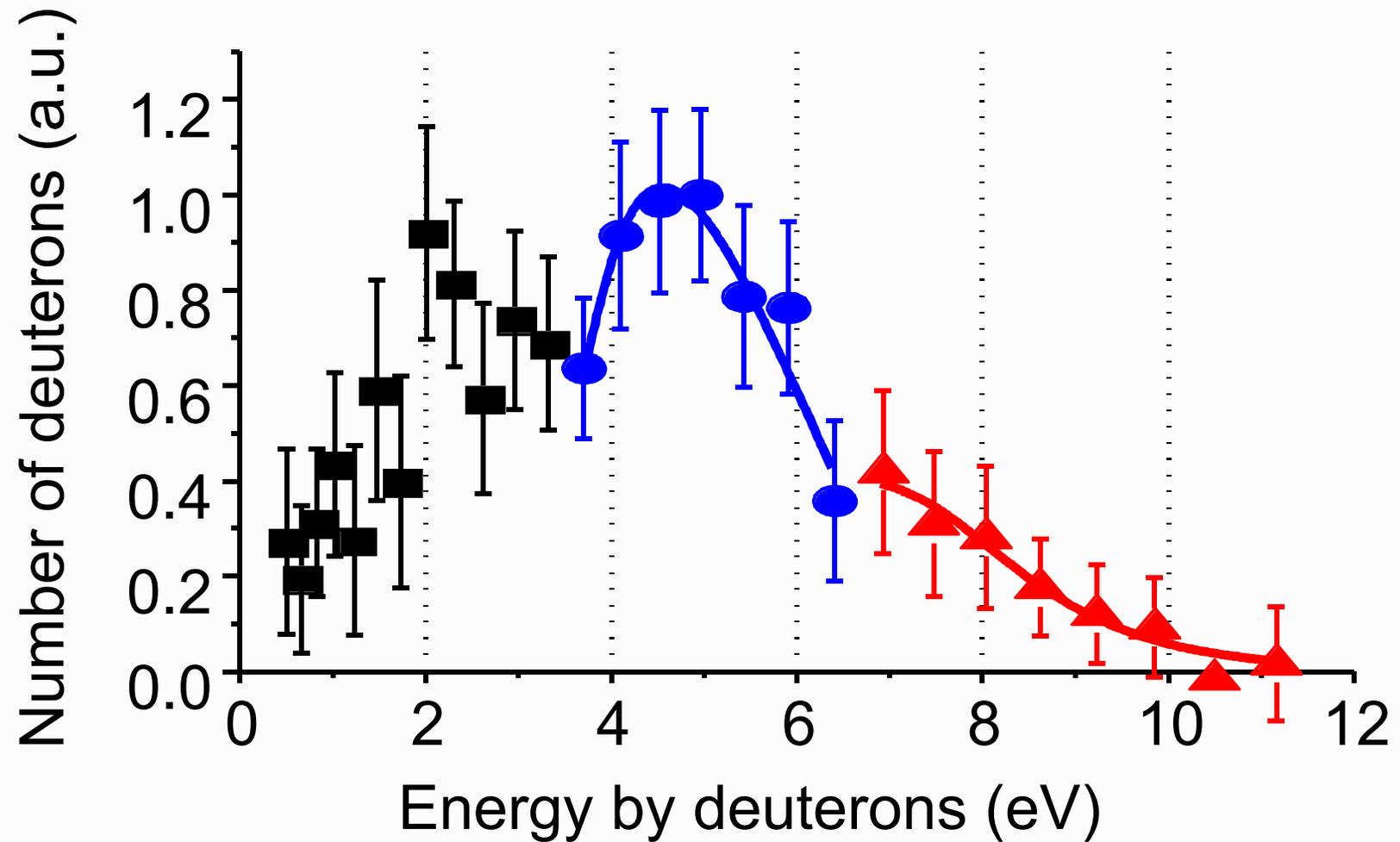
Characteristics of electrons

- Broad distribution of electron energies
- Peaked near $E \sim 3.17 (q^2 E^2 / m^2)$
(at $2 \mu\text{m}$ and 10^{14} W/cm^2 $E \sim 120 \text{ eV}$)
(at $0.8 \mu\text{m}$ and 10^{14} W/cm^2 $E \sim 30 \text{ eV}$)
- Diameter of the returning wavefunction increases linearly with time.

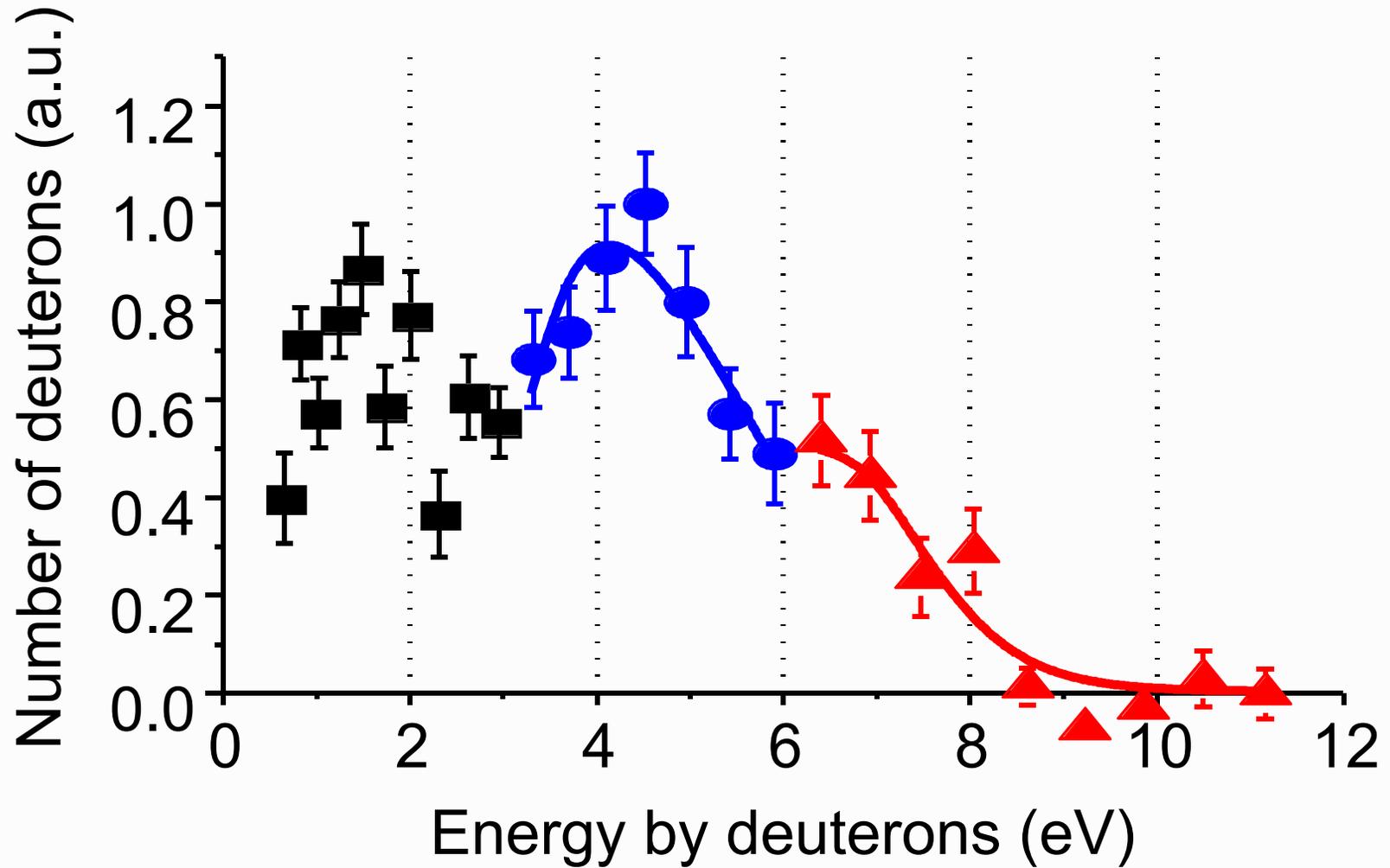
800 nm



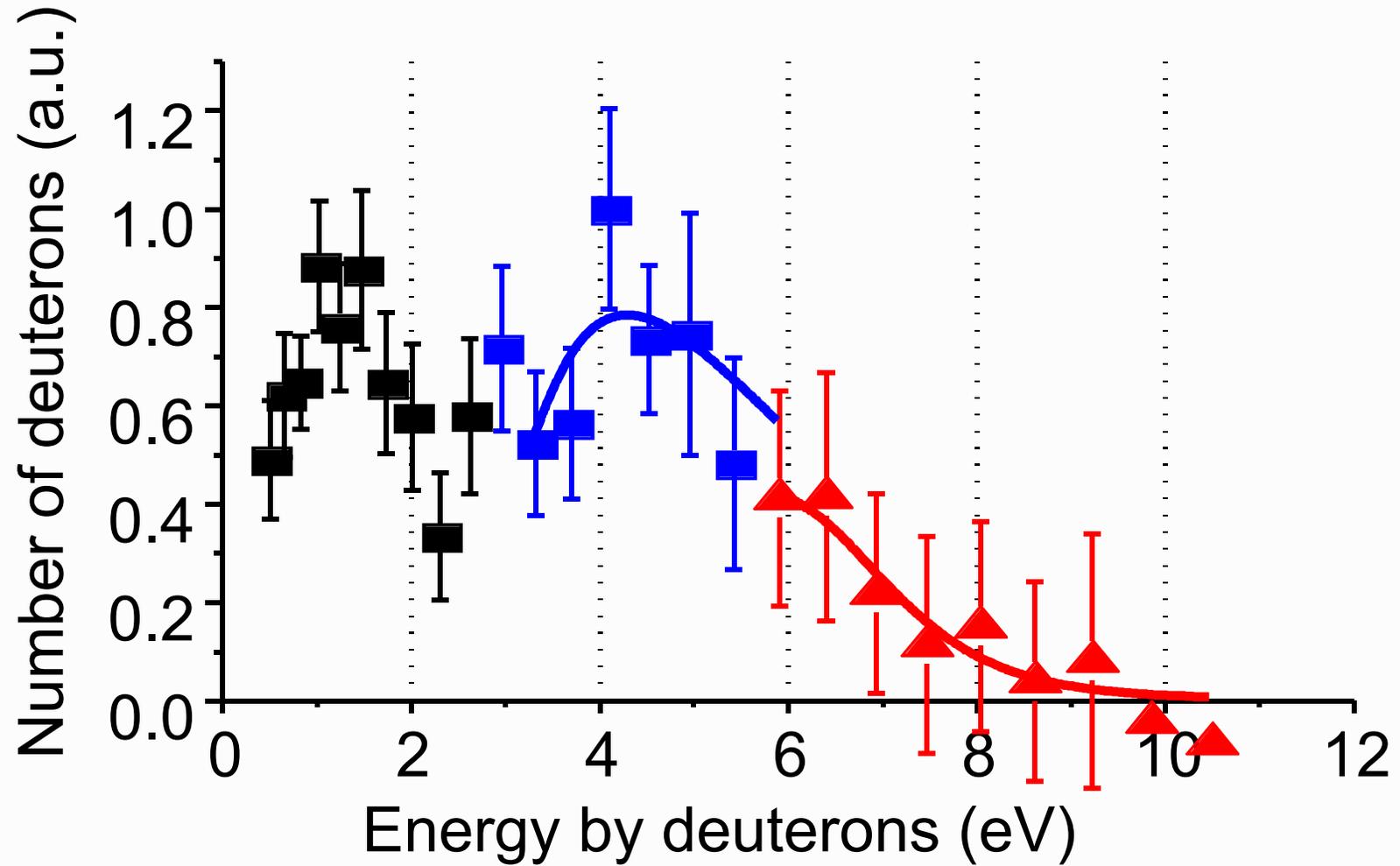
1200 nm



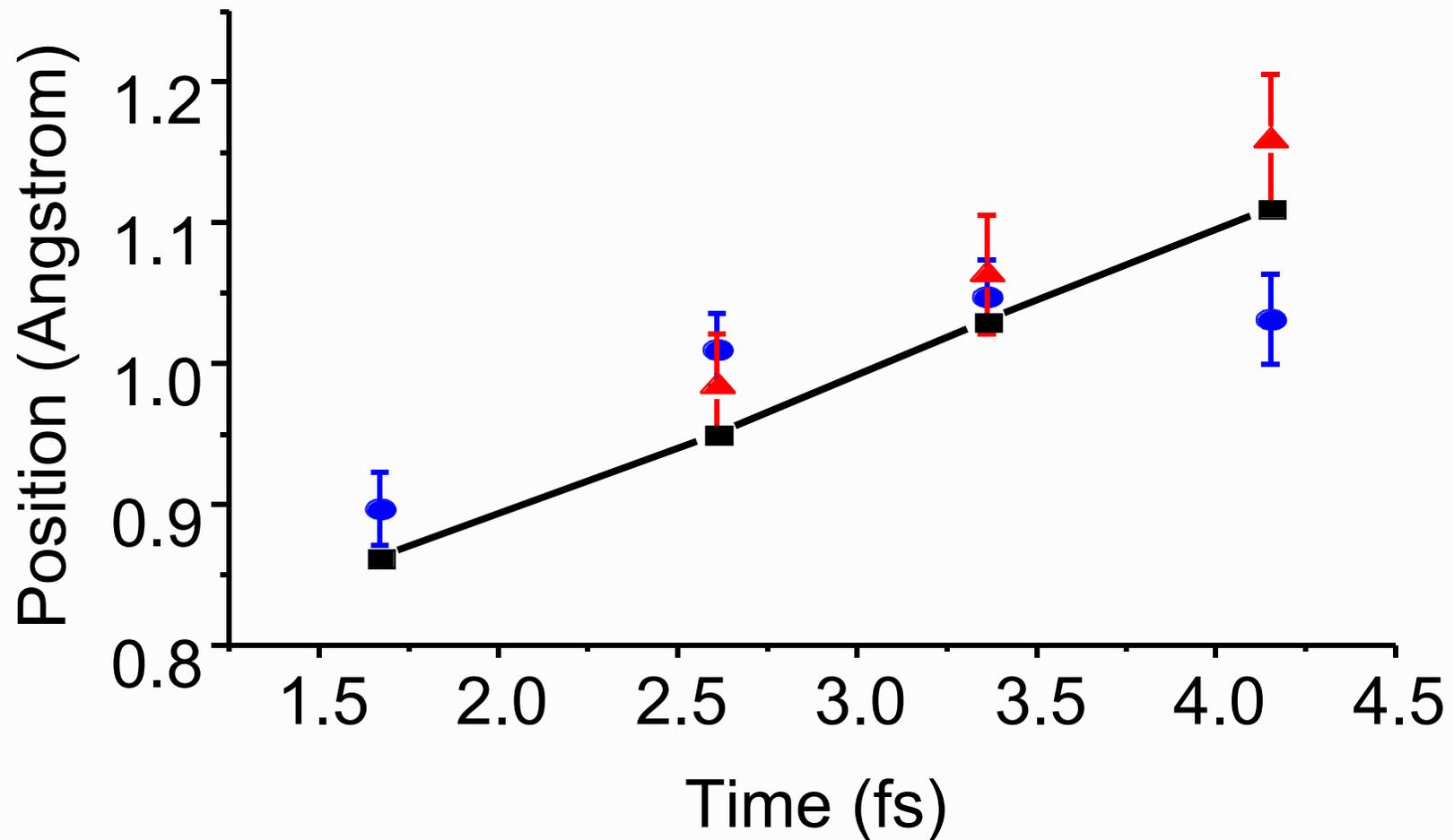
1530 nm



1850 nm



The first few femtoseconds for D_2^+



Specialized problem?

- Strong fields control electrons

In molecules:

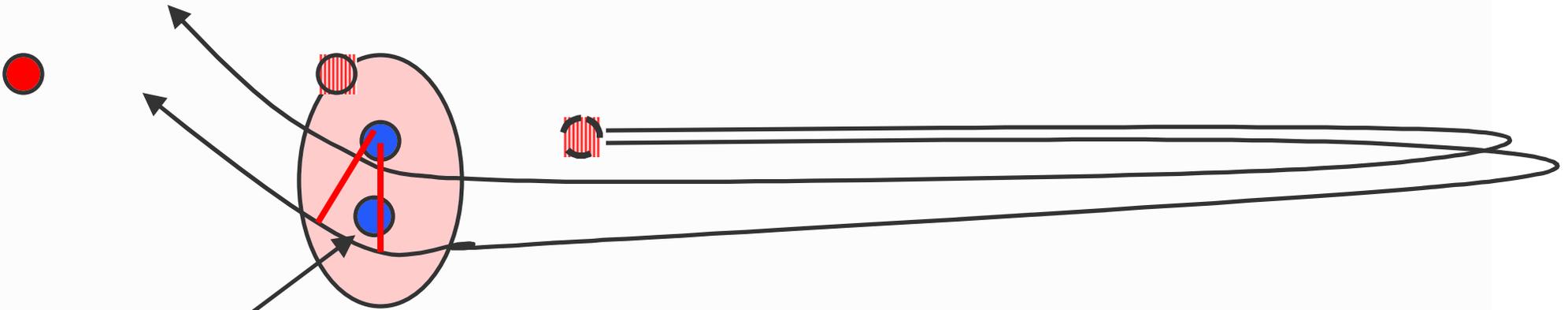
- Harmonic emission
- Elastic scattering
- Inelastic scattering

In electron correlation dynamics:

- Energy and direction of correlated electrons

- Superstrong fields control protons

Elastic scattering --- Molecular structure: The most important implication



$$n\lambda = d \sin(\theta)$$

Elastic scattering --- electron diffraction

In any electron diffraction experiment only the momentum transverse to the electron direction is measured.

Our proposal for self-diffraction

- Use strong field molecular optics to align the molecule
- Use a pump pulse to excite a process of interest if needed
- Use strong ir beam to (multiply) ionize molecule and give electron energy needed for self-diffraction

Recollision electrons can explore
feasibility and procedures needed for
femtosecond X-rays

The Femtogroup (2001)

Scientists: *Paul Corkum, David Villeneuve,* Albert Stolow, *Misha Ivanov* and David Rayner

Technologists: Bert Avery, David Joines, John Parsons and Shutao Li

Postdoctoral Fellows: Ravi Bhardwaj, *Igor Litvinyuk,* Jiro Itatani, Fabien Quere, *Hiromichi Niikura, Rachel Hasbani,* Thomas Schultz, Jonathan Underwood, Eugena Shapiro

Visitors: Gennady Yudin

Ph. D. Students: *Pat Dooley* and Kevin Lee (McMaster), *Francois Legare (Sherbrooke),* Anthony Lee and Ben Sussman (Queens), Marc Smits (Holland), Michael Spanner (Waterloo)