Ultrafast Molecular Imaging With Laser Driven Electron Diffraction

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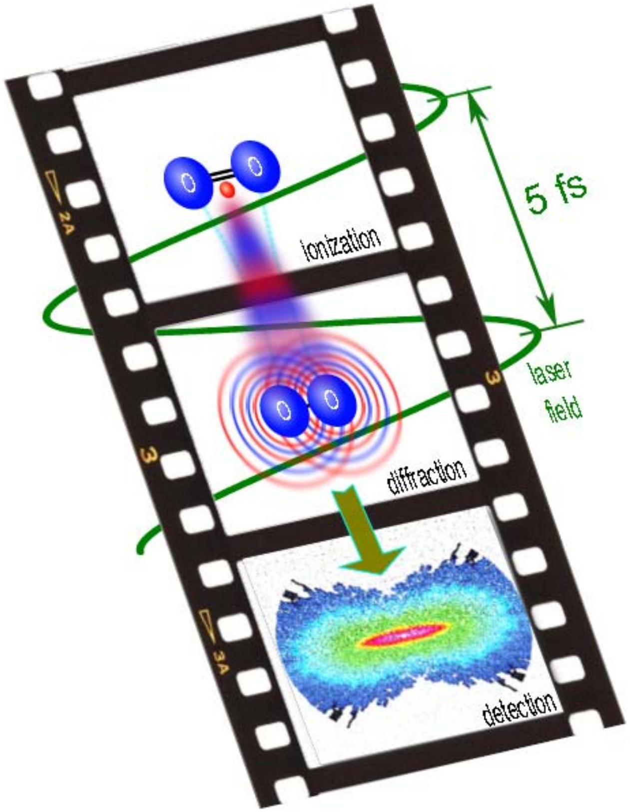
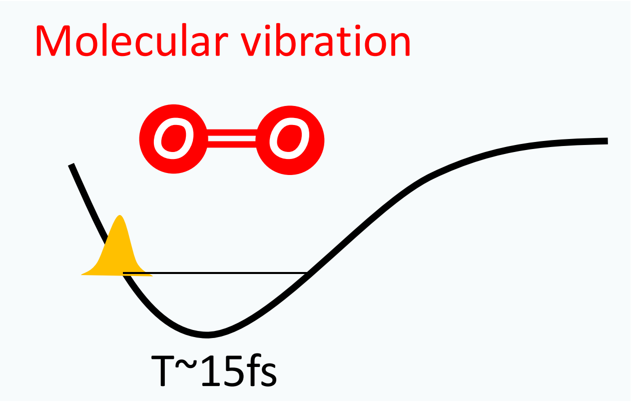
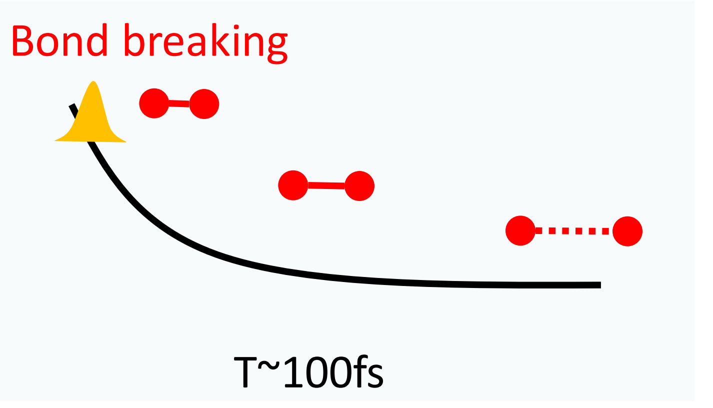


Nature **483**, 194–197 (2012)

COLDBEAMS Workshop – Nîmes, France
October 3rd, 2012

Motivation: molecular imaging





Needed: sub-Angstrom and femtosecond spatio-temporal resolutions

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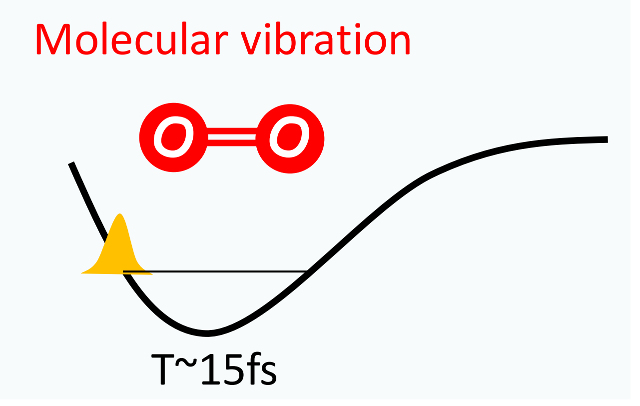
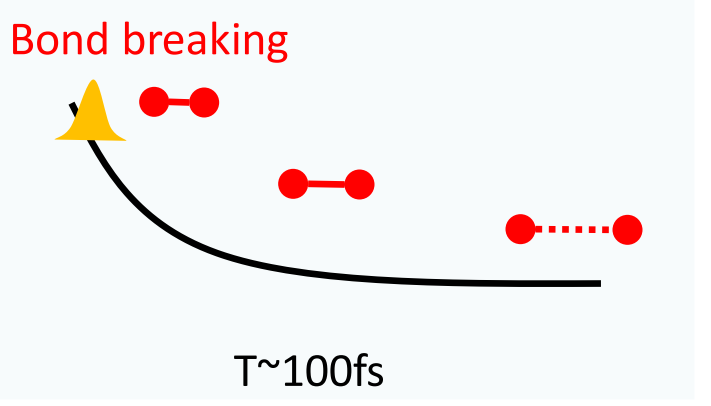
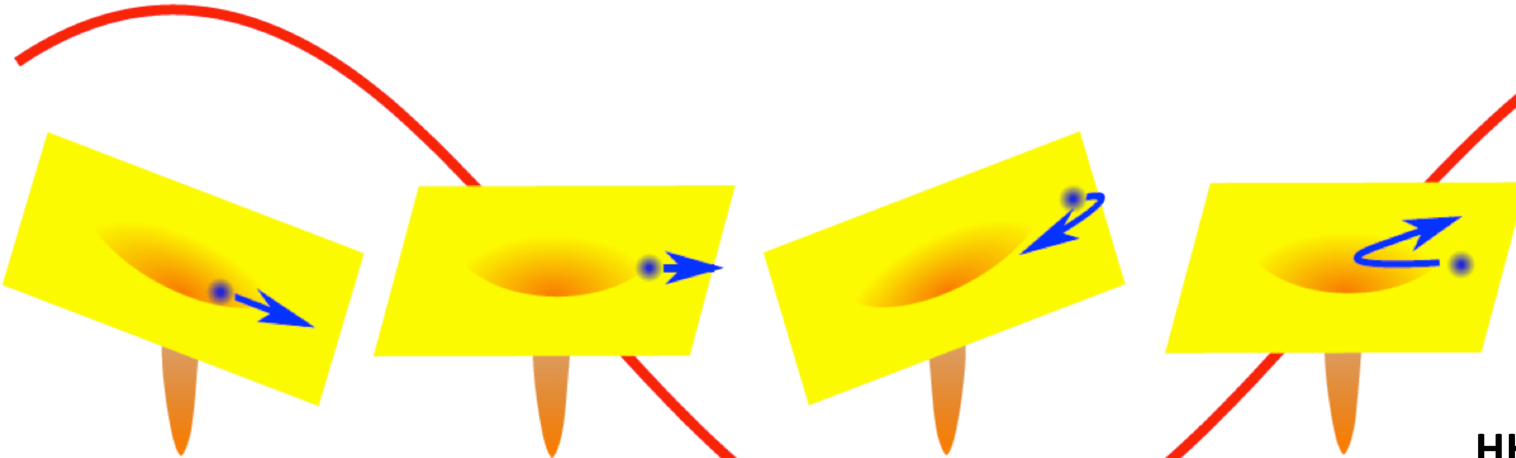




Needed: sub-Angstrom and femtosecond spatio-temporal resolutions

Outline:
- **brief introduction to conventional gas phase electron diffraction**
  - **independent atom model**
- **laser-induced electron diffraction**
  - **atomic results**
  - **molecular results**
    - first direct observation of molecular relaxation
- **outlook**

Traditional imaging methods



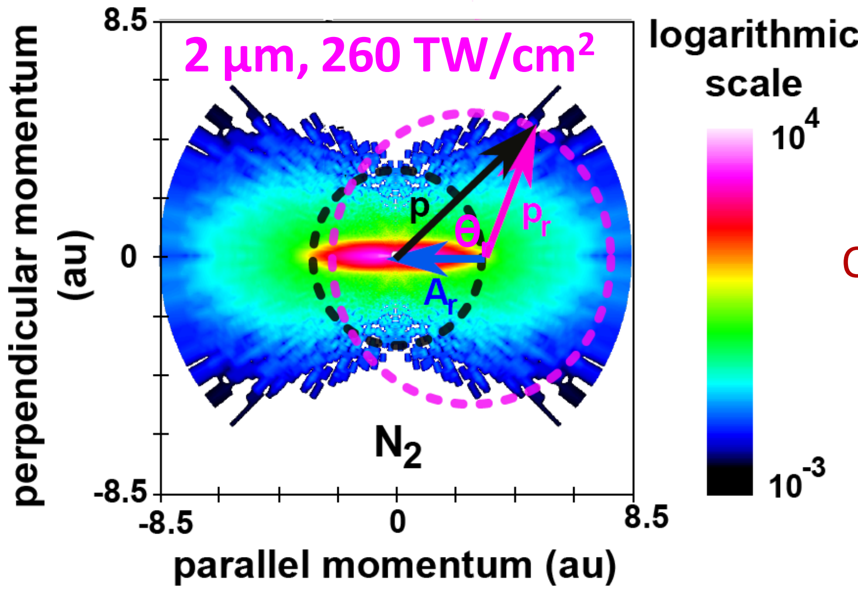
HHG
Inelastic Scattering
Elastic Scattering

Alternative path (NRC, BNL, Imperial College, Texas A&M, MPI, etc.)

Laser-Induced Electron Diffraction (LIED) → Based on rescattered electrons generated by intense fs pulses

1. spatial resolution
2. Understanding and decoupling the effect of the laser field

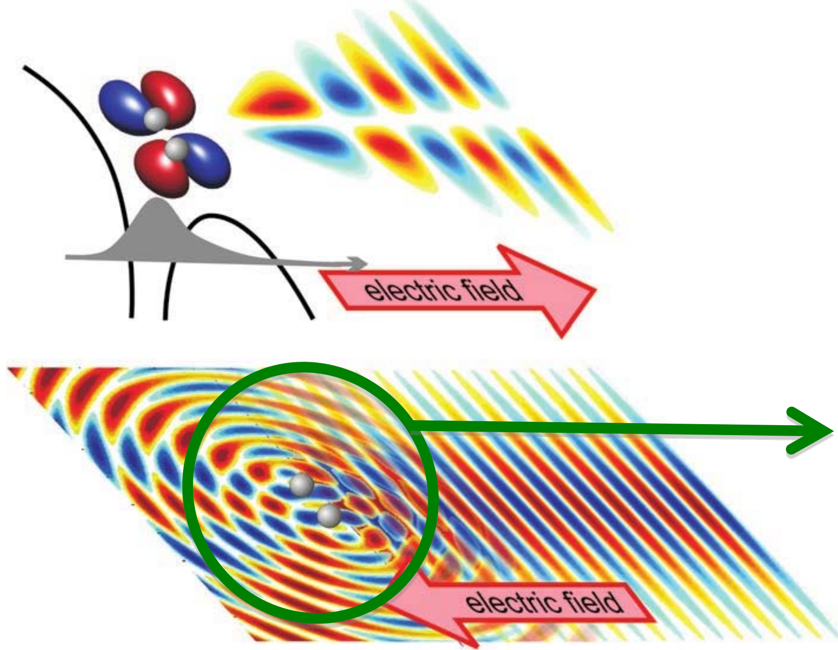
Laser-Driven Electron Diffraction (LIED)

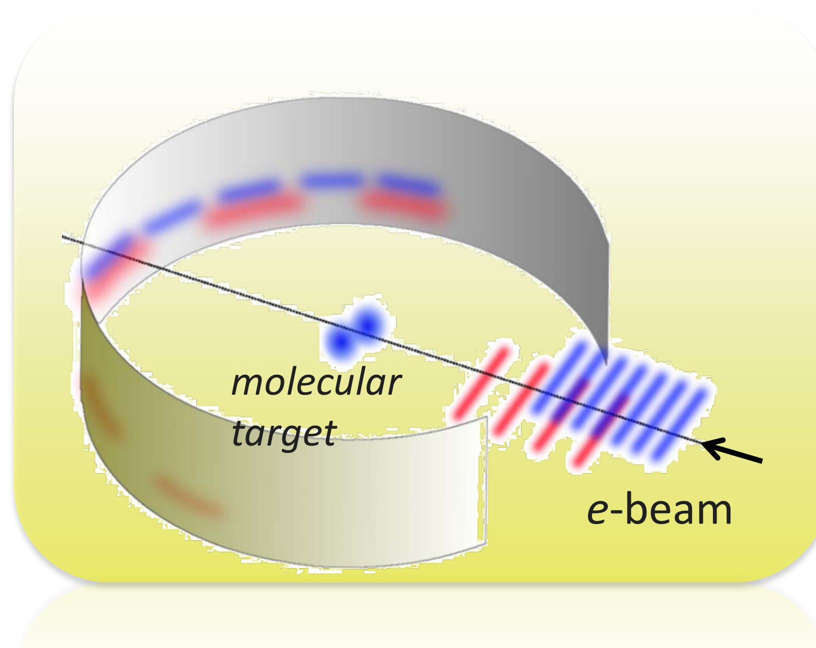


Quantitative Rescattering Theory (QRS)

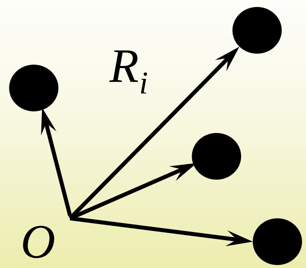
Chen *et al.*, PRA **79**, 033409 (2009)

Meckel *et al.*, Science **320**, 1478 (2008)





Independent atom model (IAM)



Molecular scattering amplitude:

$$f_{tot}(\theta,\varphi) = \sum_i e^{i\vec{q}\cdot\vec{R}_i} f_i(\theta)$$

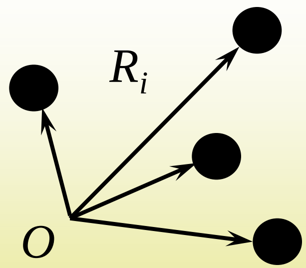
Molecular DCS:

$$\sigma_{tot}(\theta) = \sum_i \sigma_i(\theta) + \sum_{i\neq j} e^{i\vec{q}\cdot\vec{R}_{ij}} f_i(\theta) f_j^*(\theta)$$

atomic term — molecular (interference) term

- relevant quantity: - momentum transfer $q=2p_r\sin(\theta/2)$

Independent atom model (IAM)



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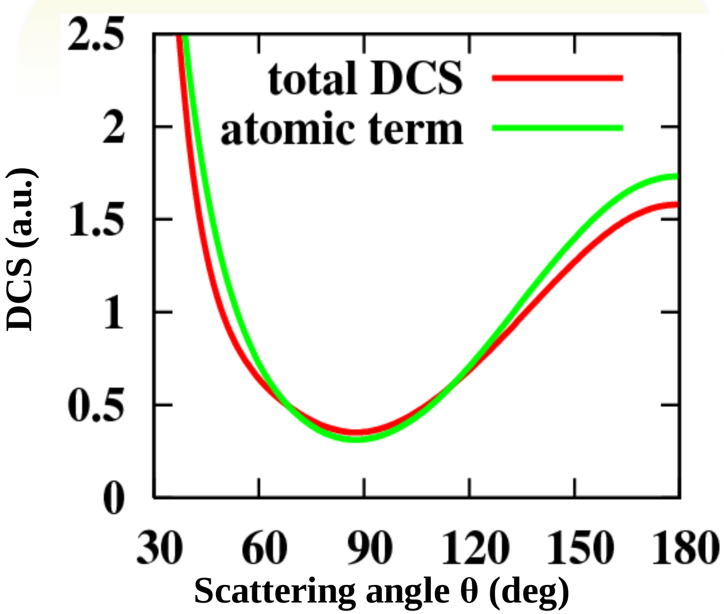


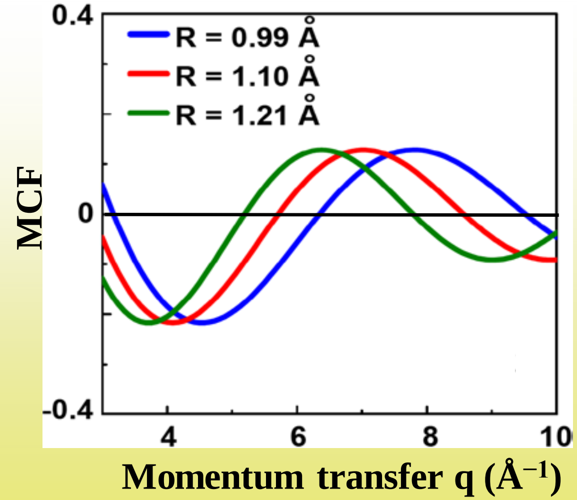
Molecular contrast factor (MCF):

$$\gamma = \frac{\text{Molecular term}}{\text{Atomic term}}$$

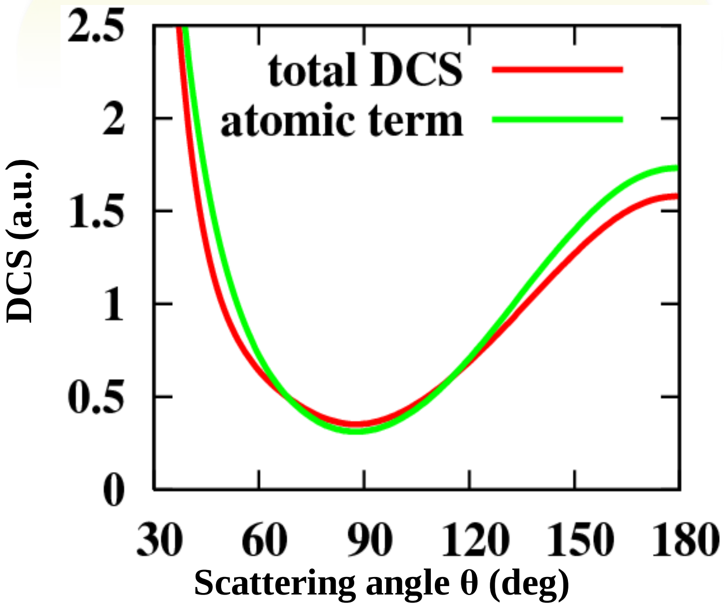
Independent atom model (IAM)

Randomly oriented sample

$$\gamma = \frac{1}{I_A} \sum_{i \neq j} f_i f_j^* \frac{\sin(q R_{ij})}{q R_{ij}}$$



Xu *et. al.*, *Phys. Rev. A* **82**, 023814 (2010)



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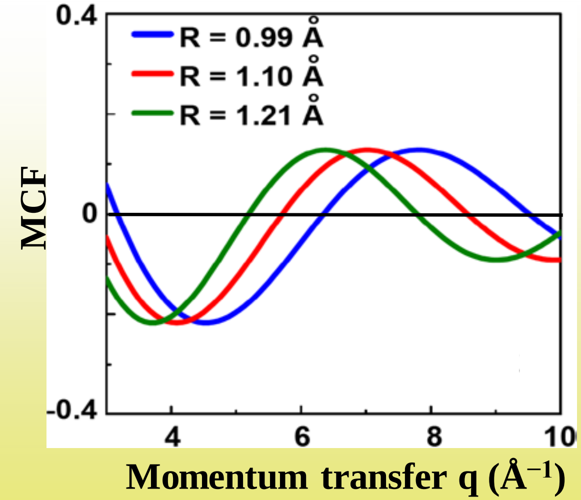
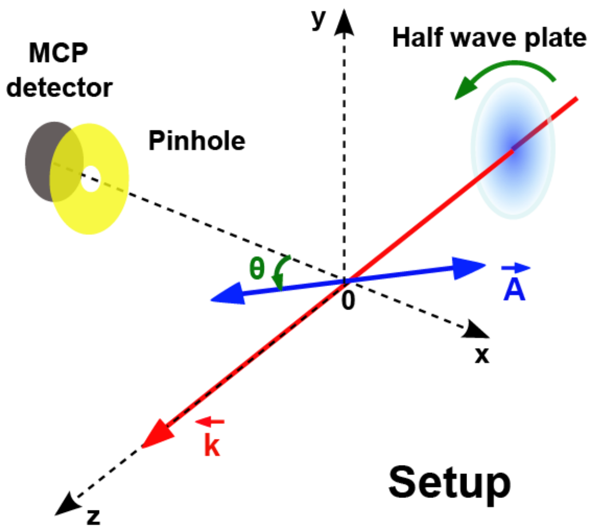


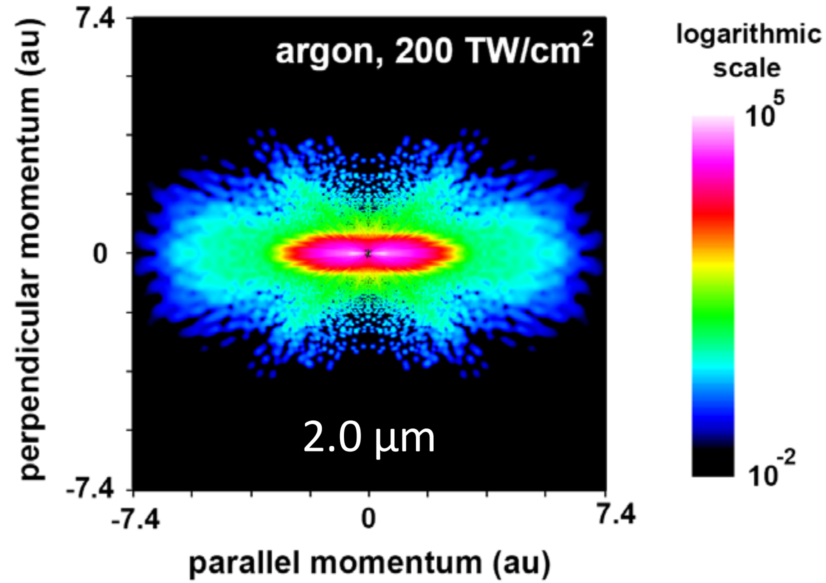
Xu *et. al.*, *Phys. Rev. A* **82**, 023814 (2010)

- momentum transfer q determines spatial resolution
- q > 5 $Å^{-1}$ are needed even for simple molecules

$q=2p_r\sin(\theta/2)$ → Recollision electrons with KE > 100 eV are needed → KE ~ $I^*\lambda^2$ → mid-IR laser sources are required

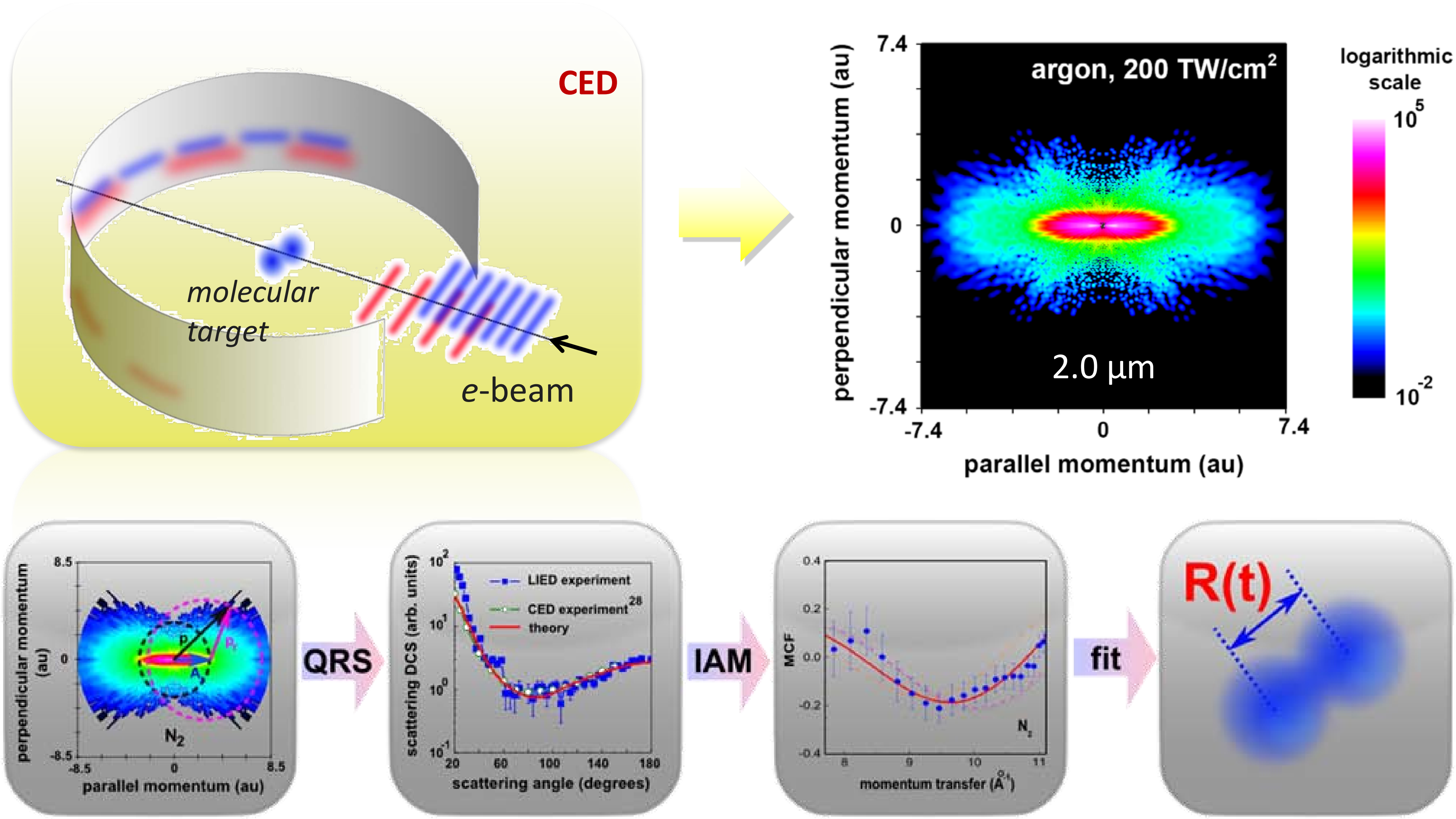
Laser-Induced Electron Diffraction (LIED)





- **long wavelength:** rescattering electron energy > 100 eV (hard collisions)
- **backscattering:** spatial resolution satisfied if $\theta \rightarrow 180°$
- invoke same analysis as conventional e-beam diffraction
- ***Challenge: extracting the DCS from the recorded angular distributions***

Laser-Induced Electron Diffraction (LIED)



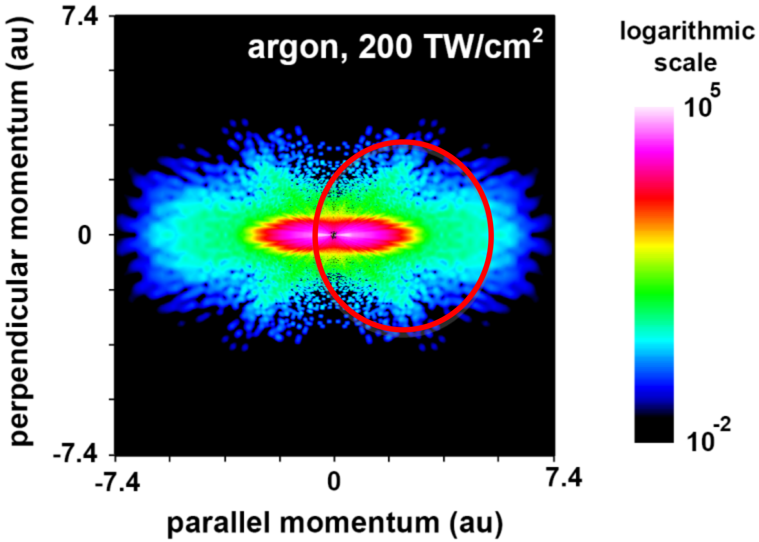
photoelectron angular distribution

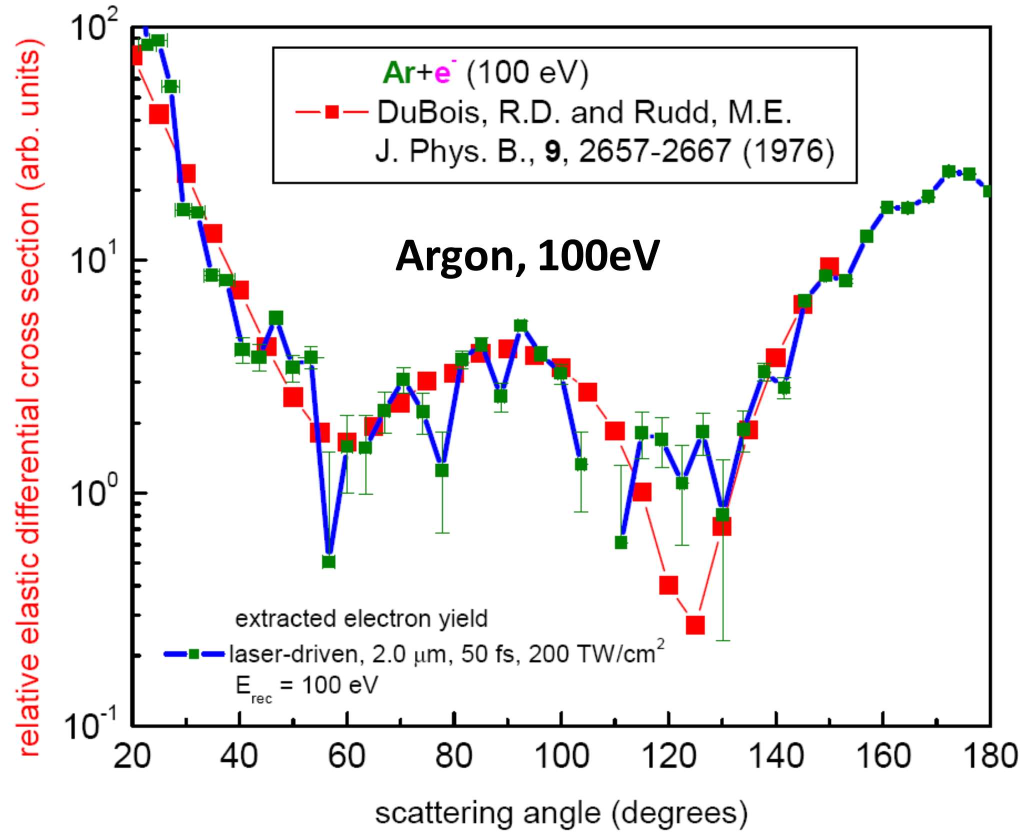
elastic DCS

molecular contrast factor

structural information

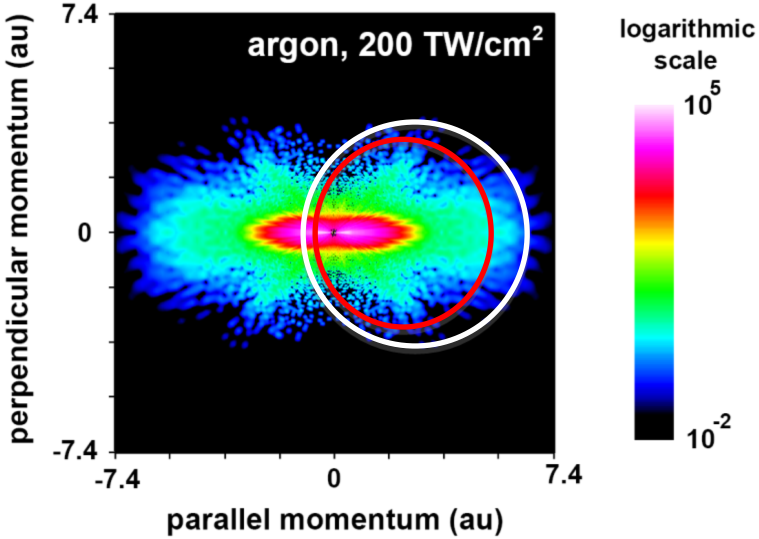
Laser-Induced Electron Diffraction (LIED) – Atomic Response

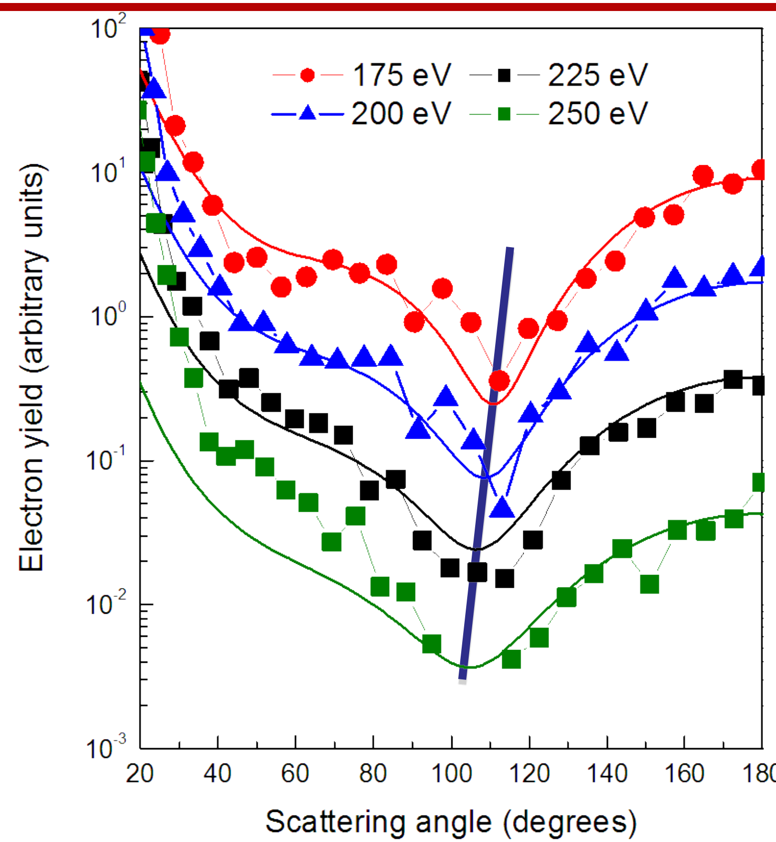


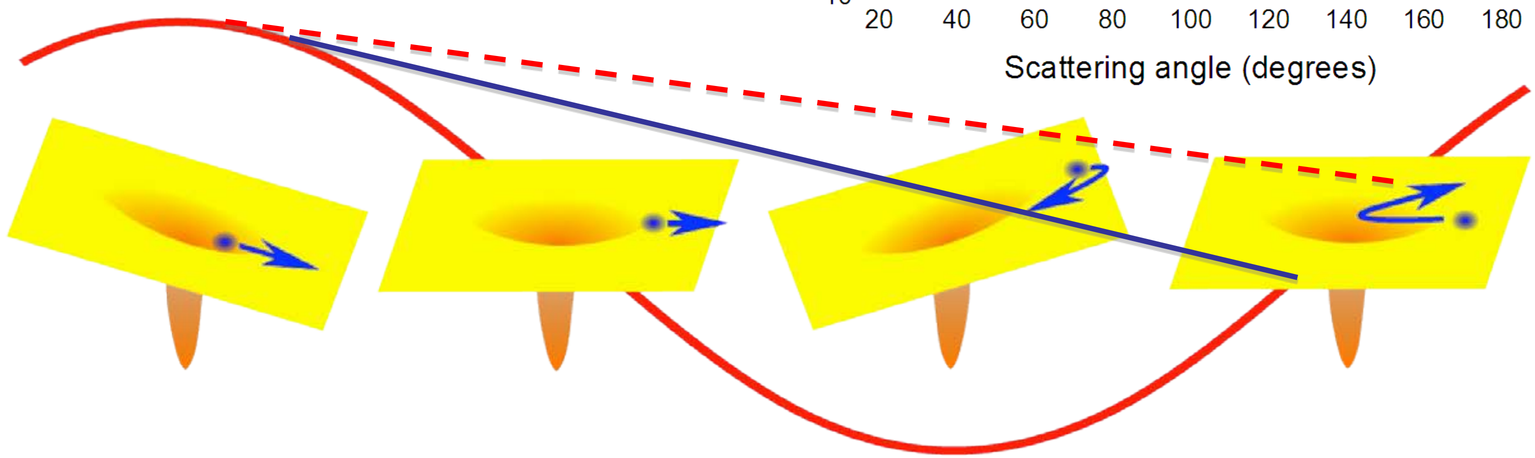


- accurate positions for diffraction minima and maxima
- two orders of dynamic range
- LIED DCS identical with field-free CED result – proves the validity of QRS
- ionic (LIED) and neutral (CED) DCSs are identical – short range interaction – justify IAM

Laser-Induced Electron Diffraction (LIED) – Atomic Response

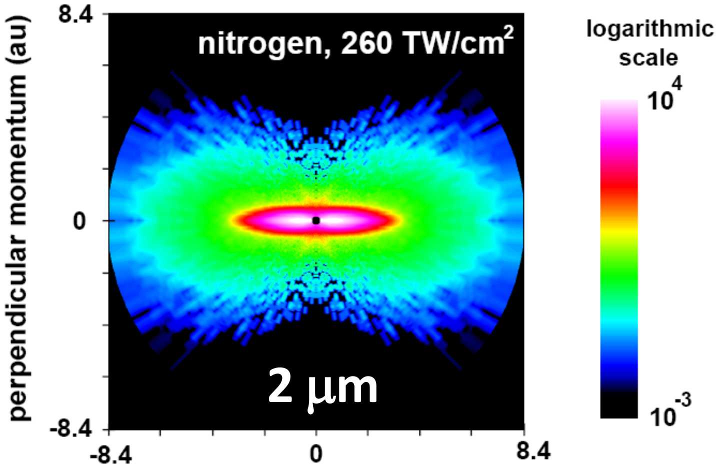


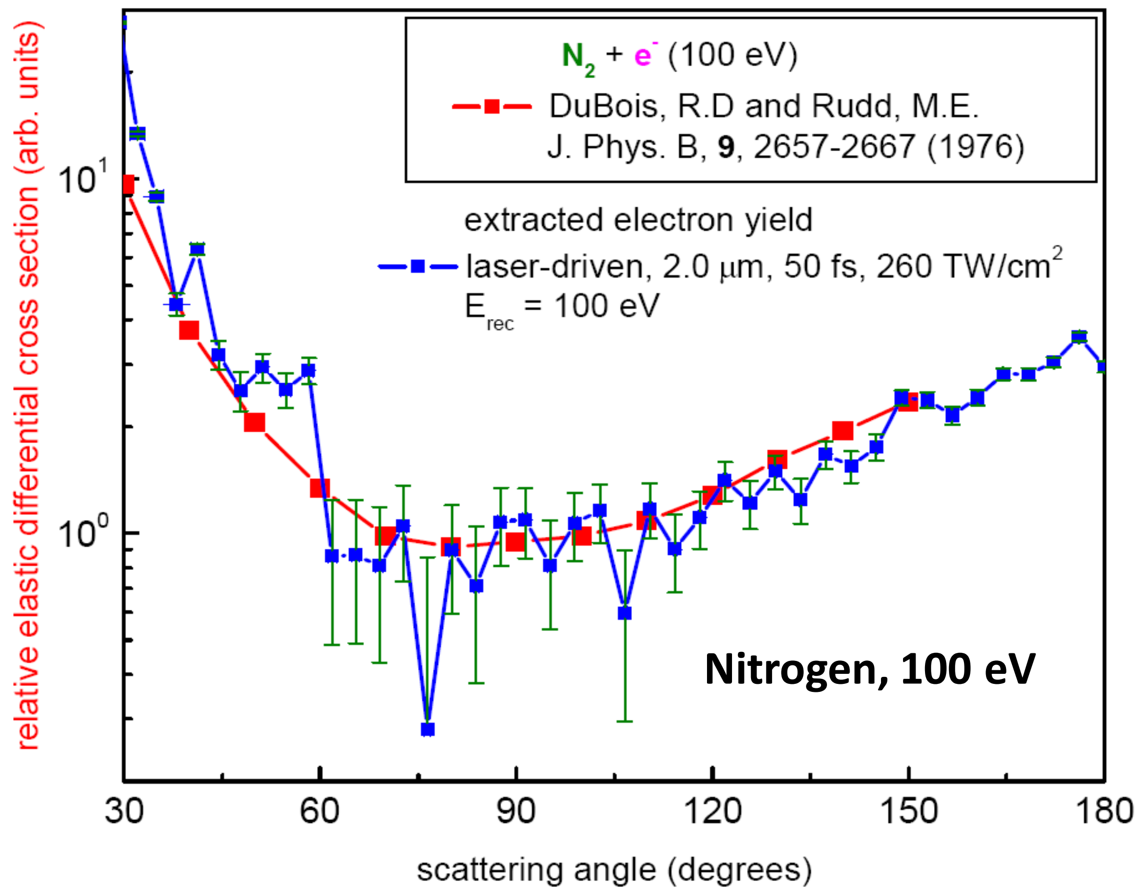




Suggests temporal resolutions below 100 as

Laser-Induced Electron Diffraction (LIED) – Molecular Response





- DCS in very good agreement with conventional method

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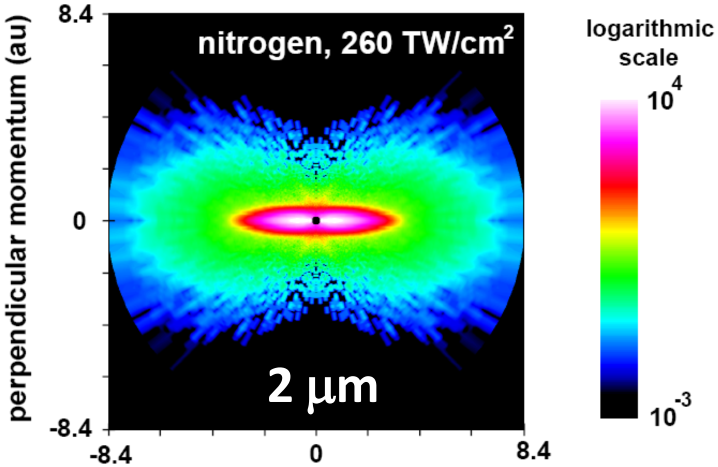
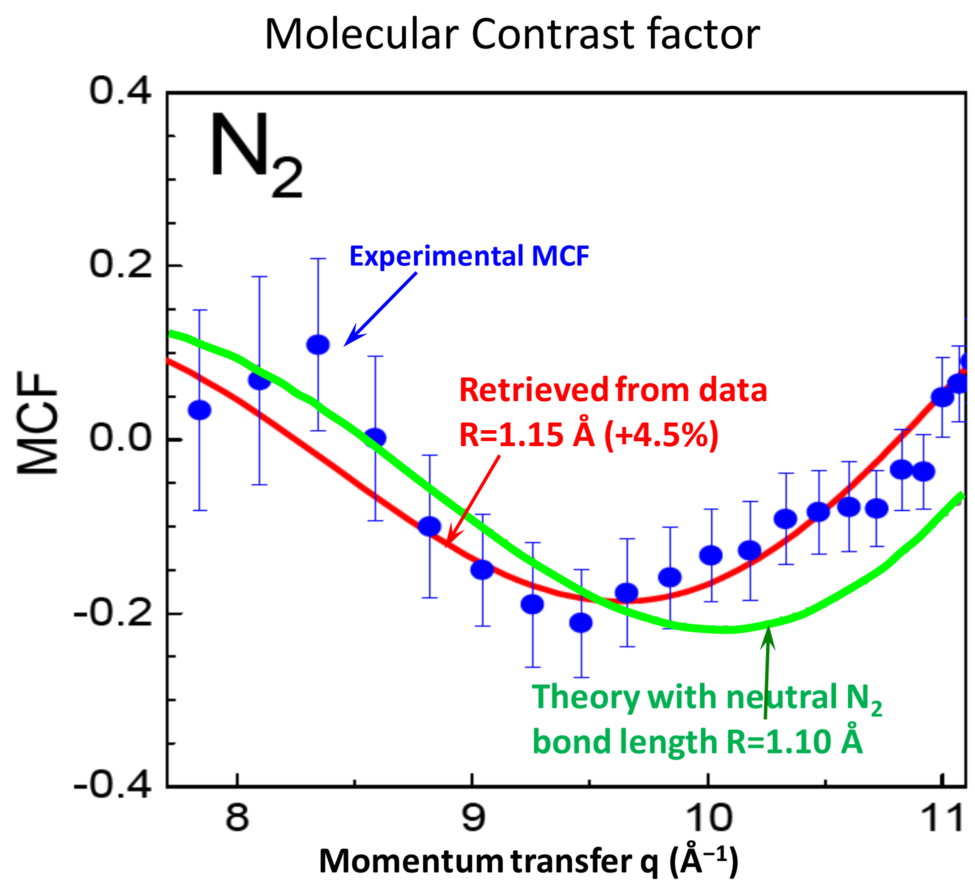
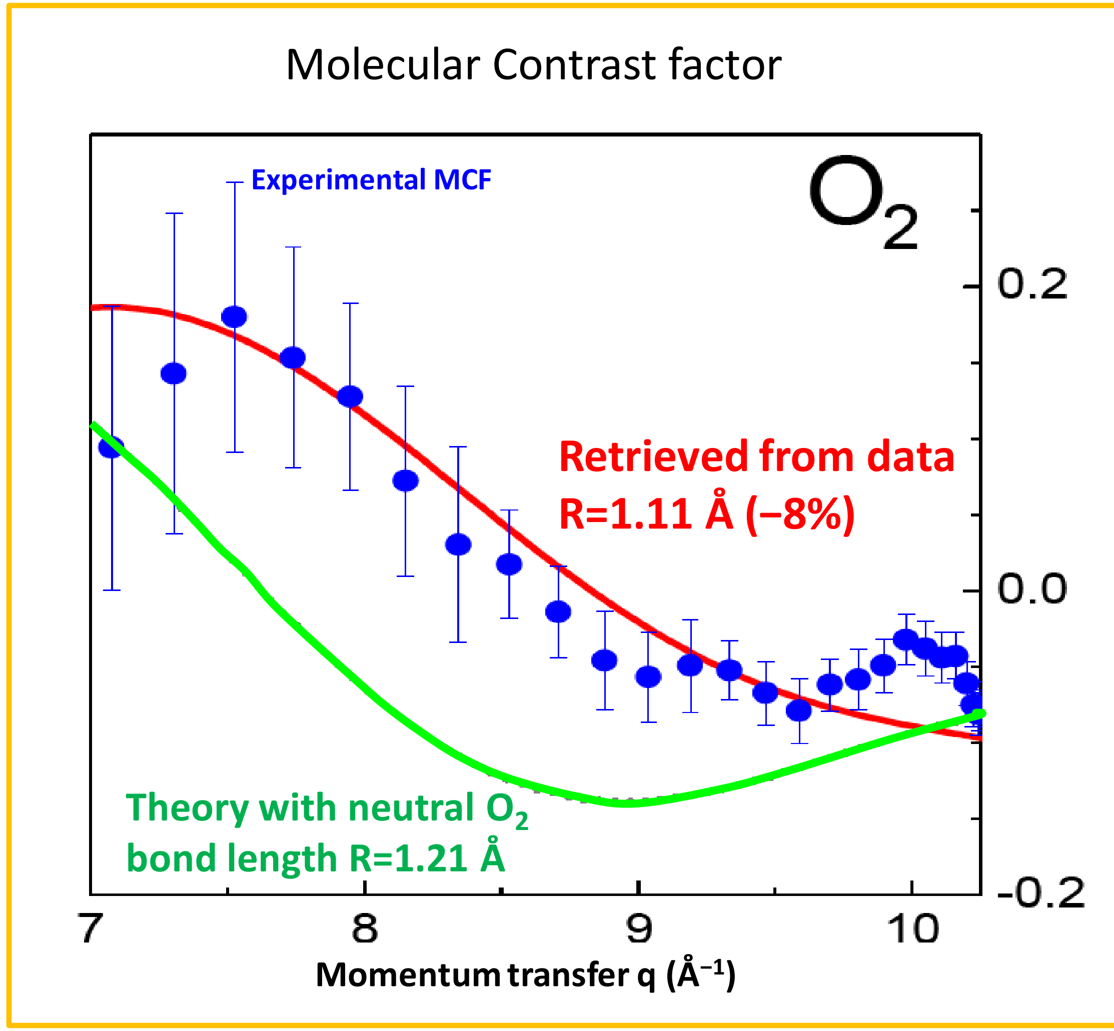




- **DCS in very good agreement with conventional method**

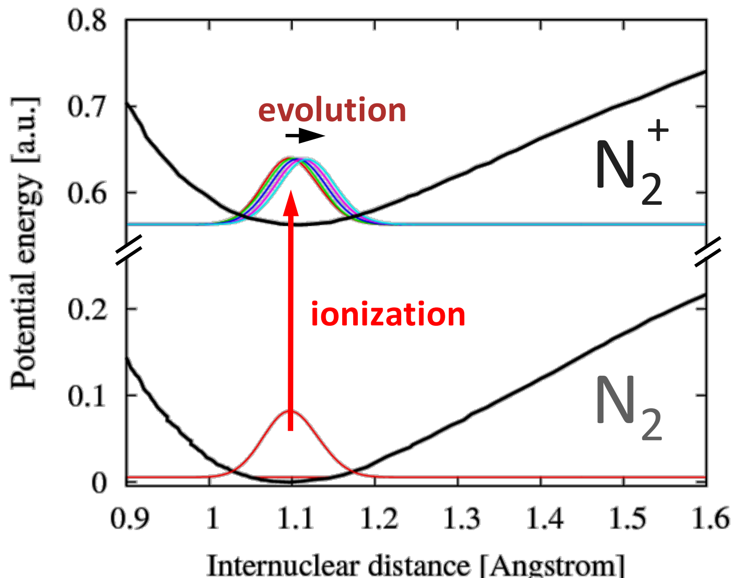
Laser-Induced Electron Diffraction (LIED) – Molecular Response

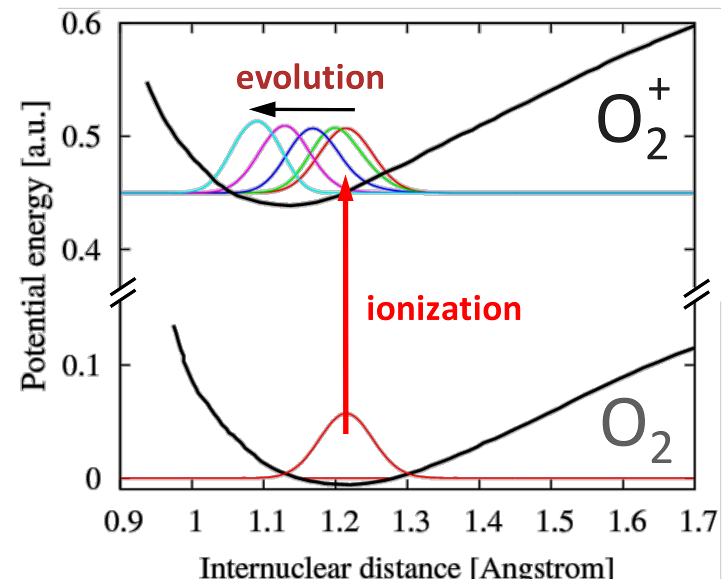
perpendicular momentum (au)

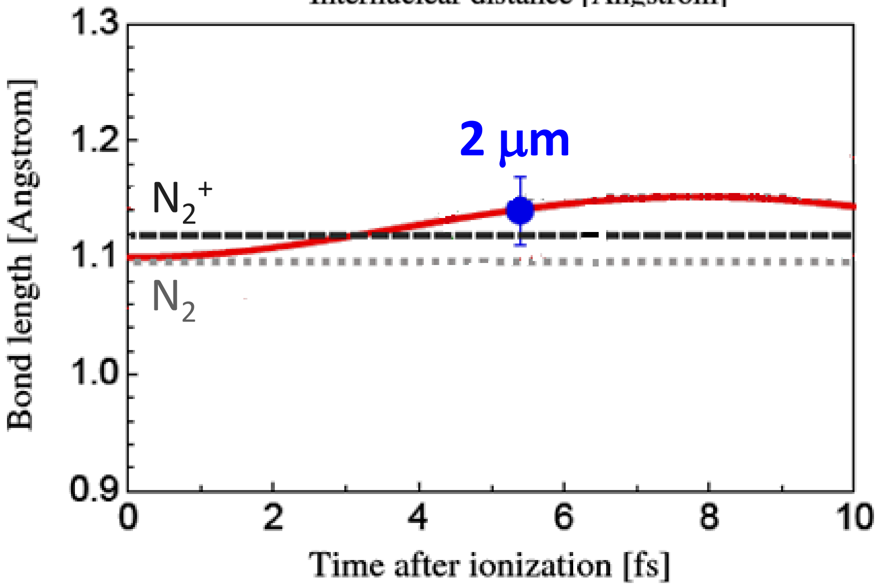


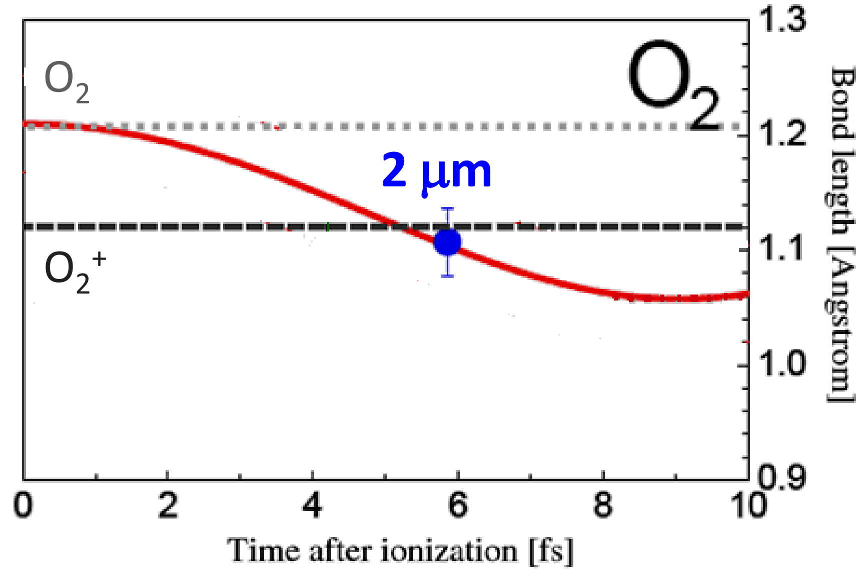
- Why is the ionic O-O distance shorter than the neutral O-O?

Laser-Induced Electron Diffraction (LIED) – Molecular Response





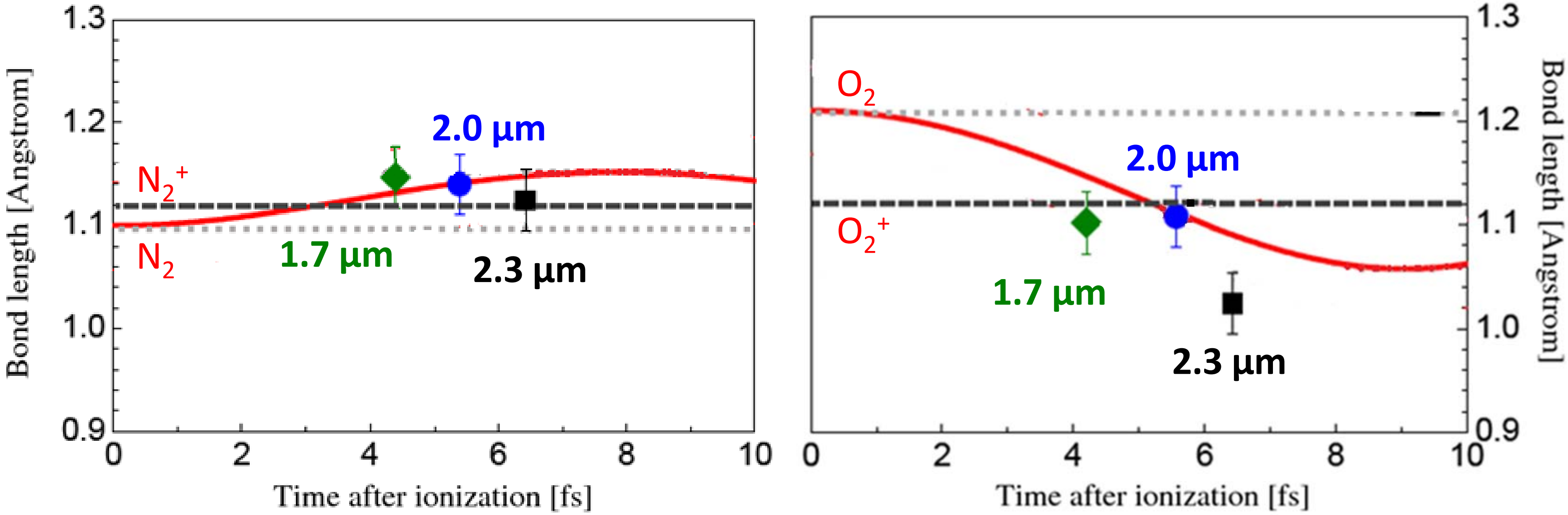




- **Pump-probe: ionization = pump, rescattering = probe**

Laser-Induced Electron Diffraction (LIED) – Molecular Response

- By changing the driving field wavelength we control the recollision (imaging) time!



- Alternative methods: - bichromatic fields
  - ellipticity control

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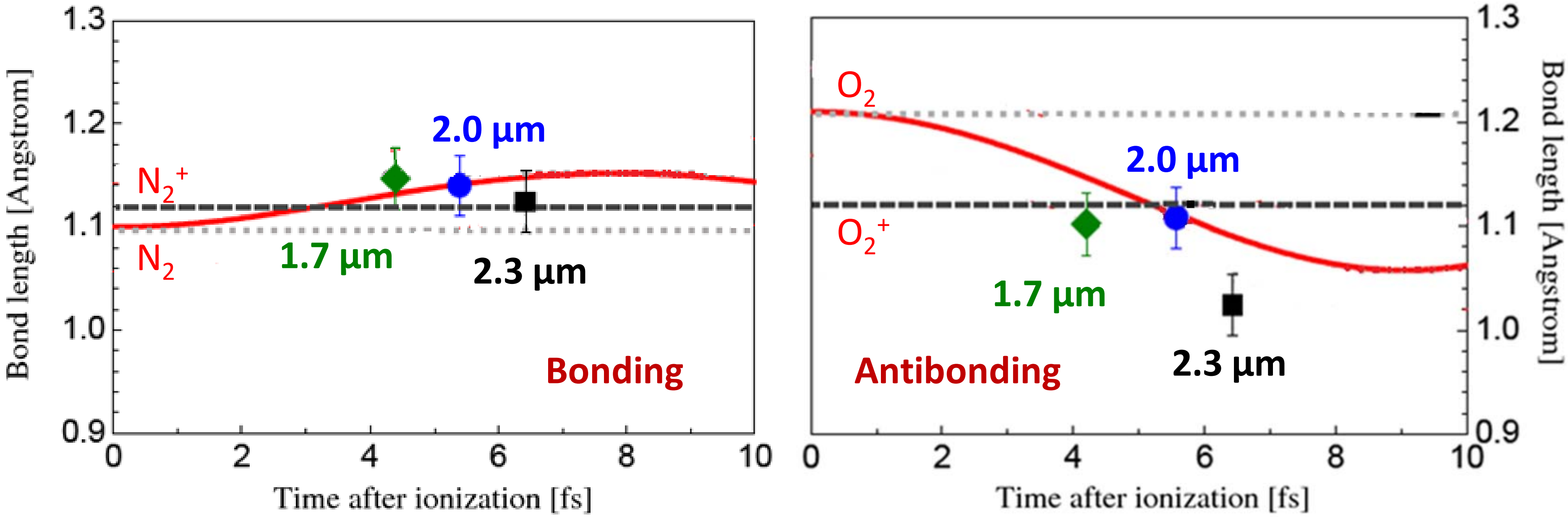
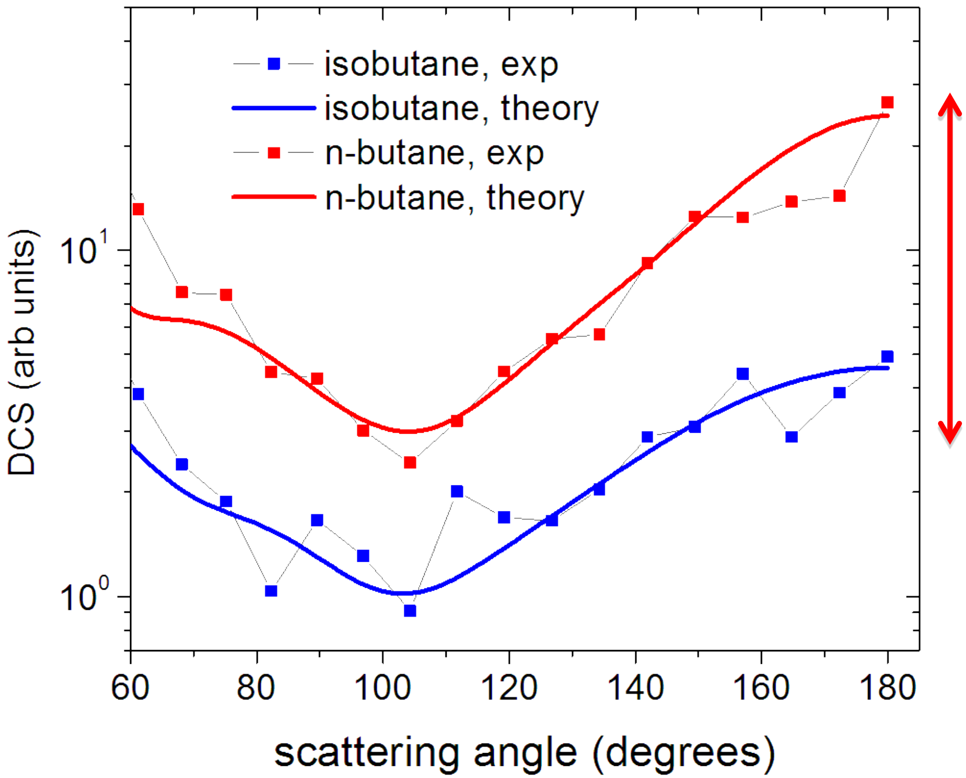


- **Alternative methods: - bichromatic fields**
  - ellipticity control

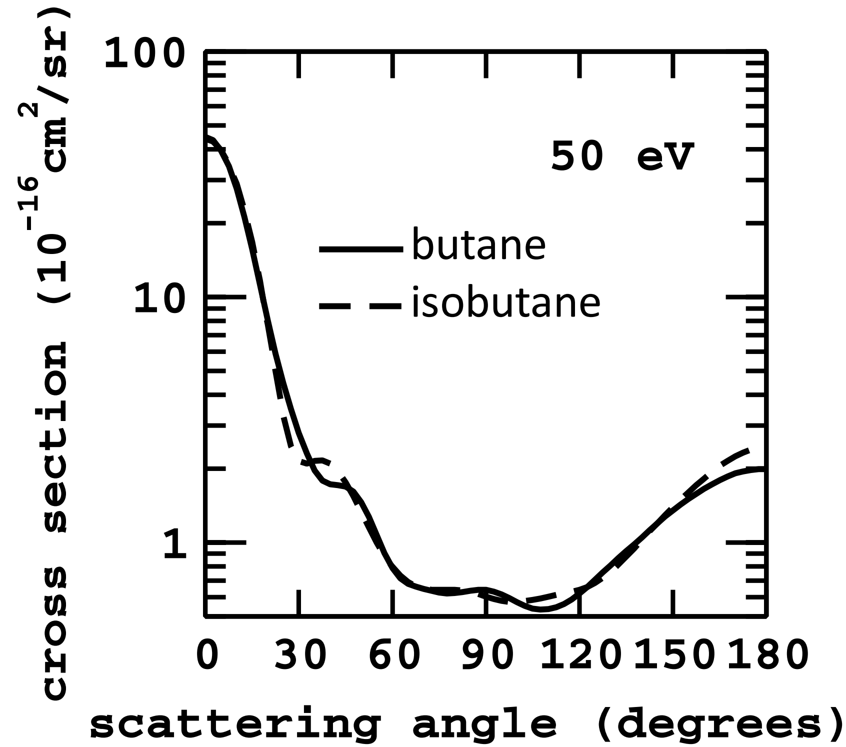
- **LIED directly sees the type of molecular orbital**

Laser-Induced Electron Diffraction (LIED) – More Complex Molecules

50 eV w/ 3.6 μm, 100 fs pulses, 30 TW/cm^2



Lopes *et al.*, J Phys B, **37**, 997 (2004)



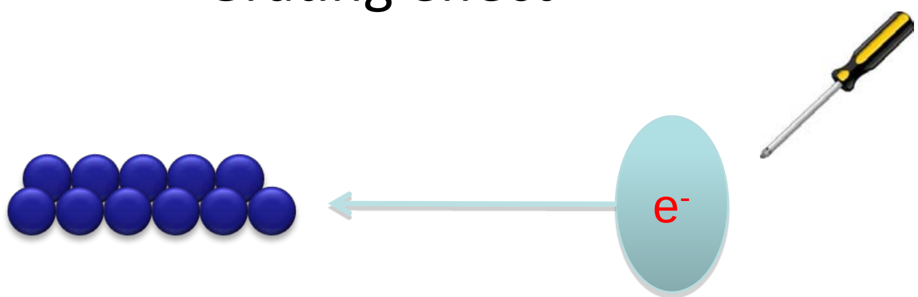
Q: Why is the contrast better in butane?

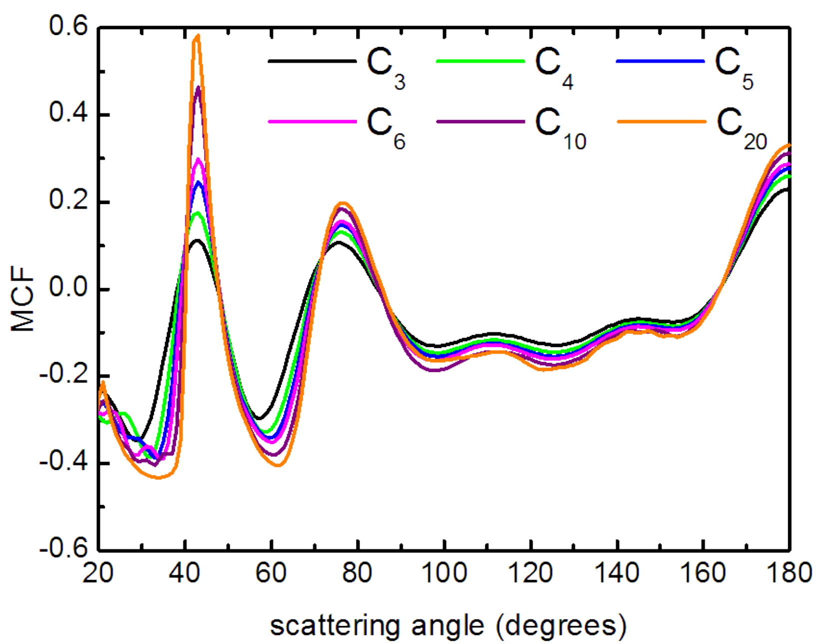
A: Tunneling ionization strongly favors certain molecular orientations.

We can record DCS for aligned samples for free!

Laser-Induced Electron Diffraction (LIED) – More Complex Molecules

Grating effect





Broadband attosecond electron wave packet studies

Laser-Induced Electron Diffraction (LIED) – More Complex Molecules

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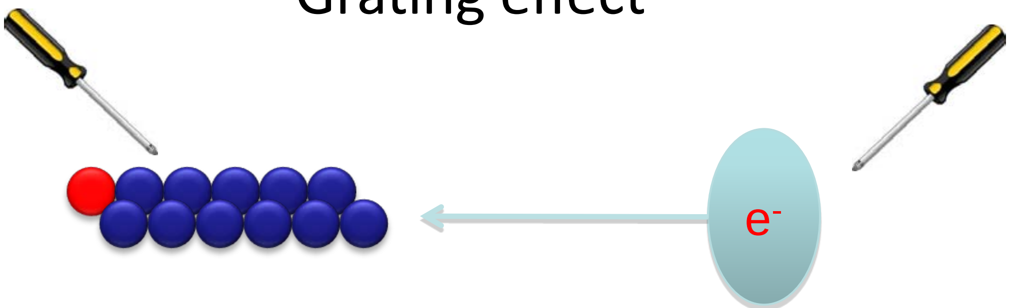
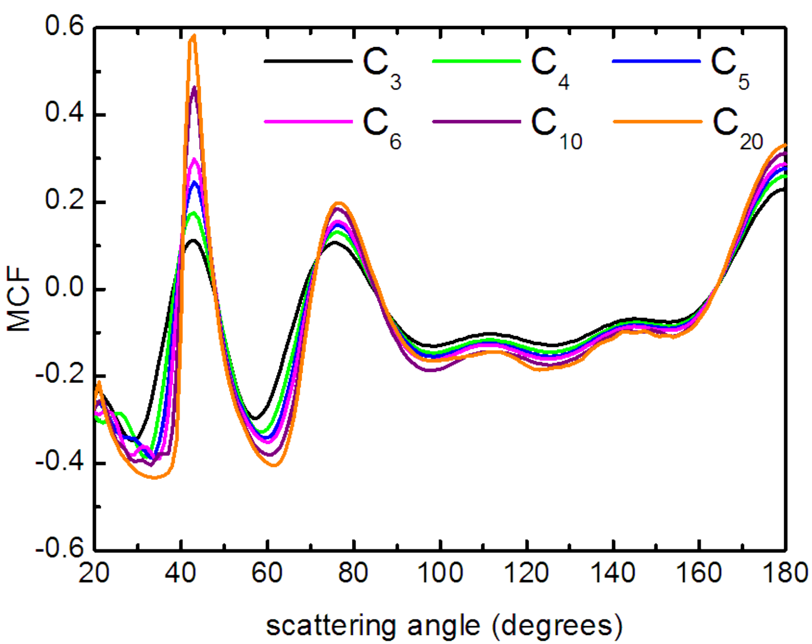


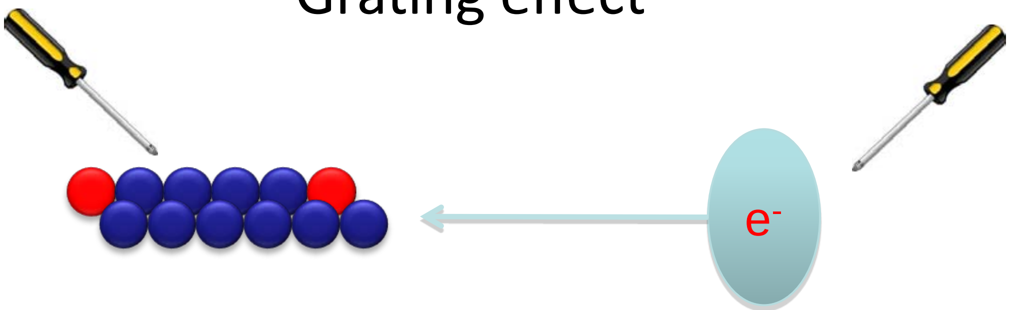


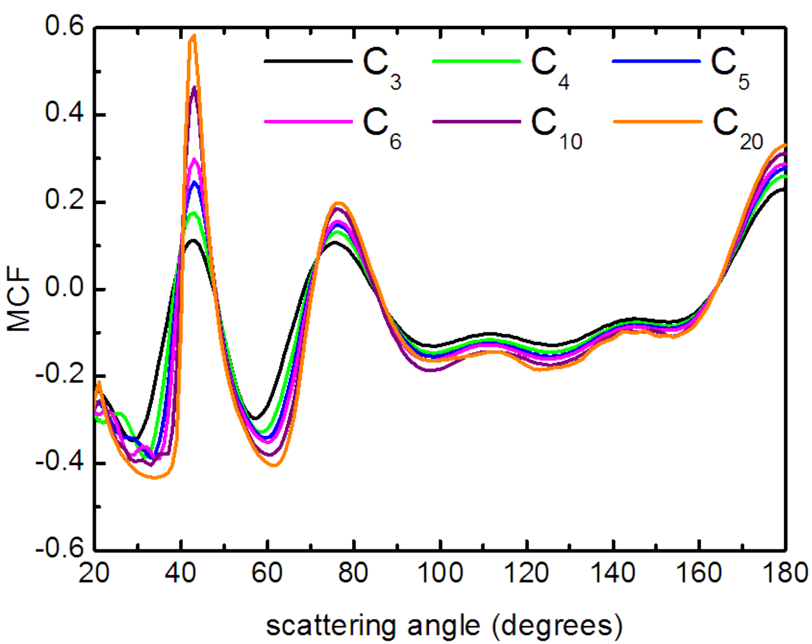
Broadband attosecond electron wave packet studies

Molecular tunneling studies

Laser-Induced Electron Diffraction (LIED) – More Complex Molecules

Grating effect





Broadband attosecond electron wave packet studies

Molecular tunneling studies

Laser-Induced Electron Diffraction (LIED) – Conclusions

- We can achieve spatio-temporal resolutions good enough to follow ultrafast molecular transformations
- Single atom/molecule method
- Applicable to complex molecules

Laser-Induced Electron Diffraction (LIED) – What we need next

- Faster acquisition times for true pump-probe experiments

→ Velocity map imaging detector

- Long wavelength (3-20 μm), few cycle, high repetition rate drivers

→ New optical parametric amplifiers (KTA, AGS – based)

Thank you!

<u>Cosmin Blaga</u>, Junliang Xu, Anthony DiChiara, Kaikai Zhang, Urszula Szafruga, **Pierre Agostini, Louis F. DiMauro**

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