International workshop on quantum many-body problems,
in particle, nuclear, and atomic physics
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EXTRACTING DYNAMIC STRUCTURAL INFORMATION FROM LASER-INDUCED ELECTRON DIFFRACTION SPECTRA

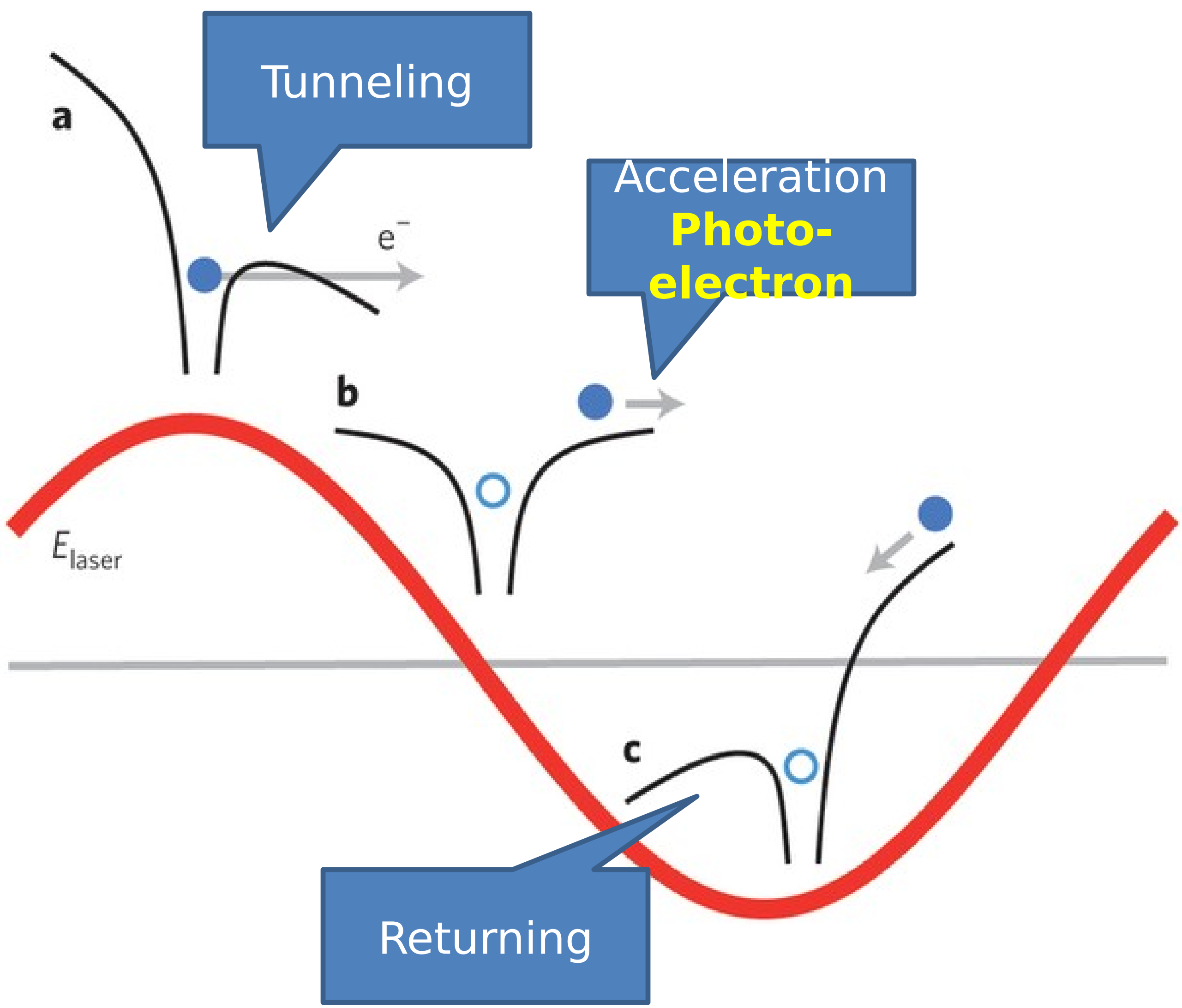
Vu Tran Dinh Duy,
Ho Chi Minh City University of Education

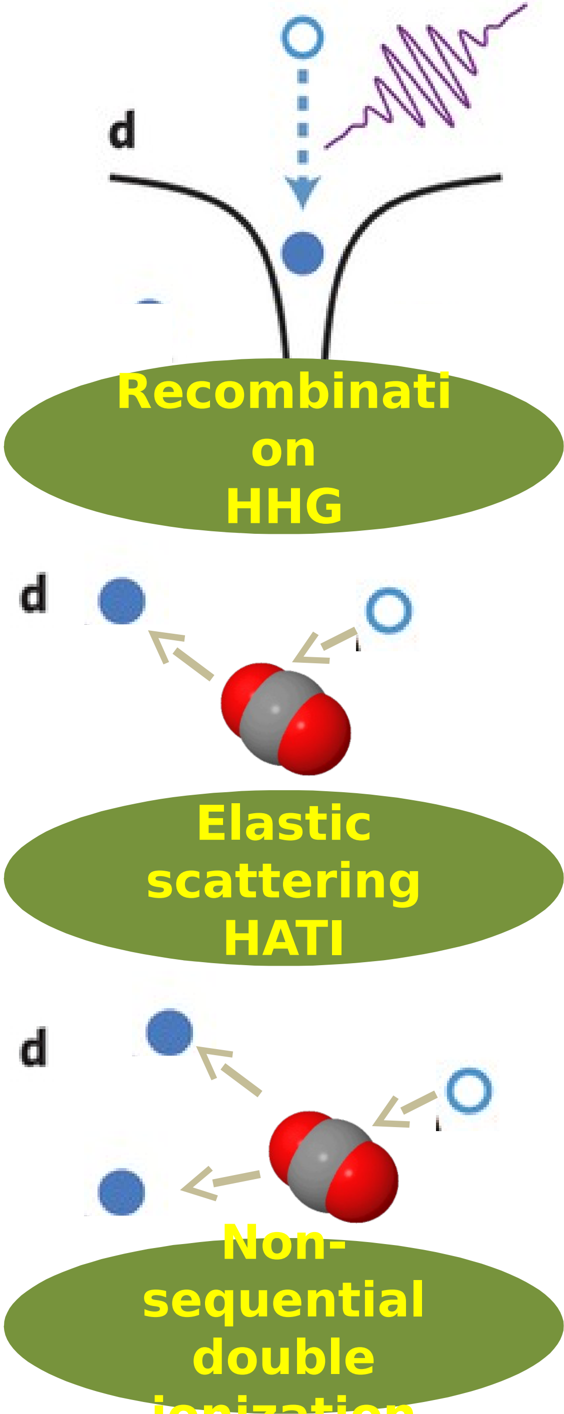
ĐẠI HỌC SP TP. HỒ CHÍ MINH

Atomic, Molecular & Optical physics group at HCMUE

Photo

Matter – strong field laser interaction





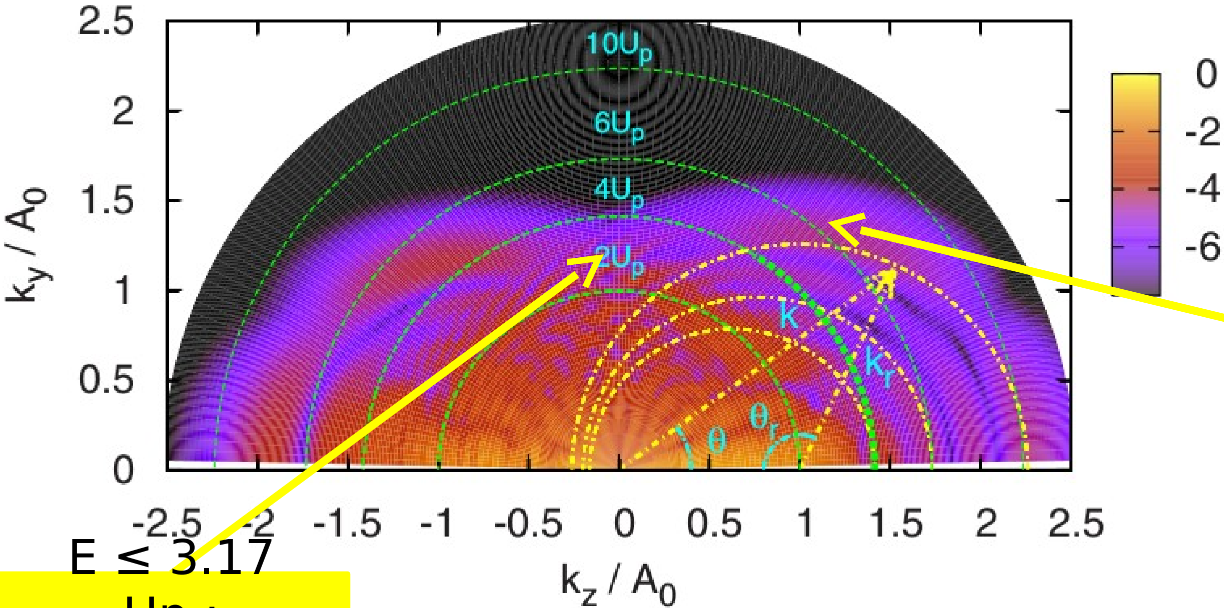
Why are these phenomena important?

- Contain target's signature
- Interaction takes place in atto – femto second scale -> information with high temporal resolution

Suitable tool to study ultra-fast atomic or molecular physics

High-energy above threshold ionization

Zhangjin Chen, Anh-Thu Le, Toru Morishita, and C. D. Lin, Phys. Rev. A, **79**, 033409 (2009)



3.17 Up ≤ E ≤ 10 Up : exclusively rescattered electron

E ≤ 3.17 Up : dominated by direct electron

Semi circle: collection of rescattered electron with incoming momentum k_r

$$k_z = k\cos\theta = \pm A(t_r) \mp k_r \cos\theta_r$$

$$k_y = k\sin\theta = k_r \sin\theta_r$$

Quantitative rescattering theory

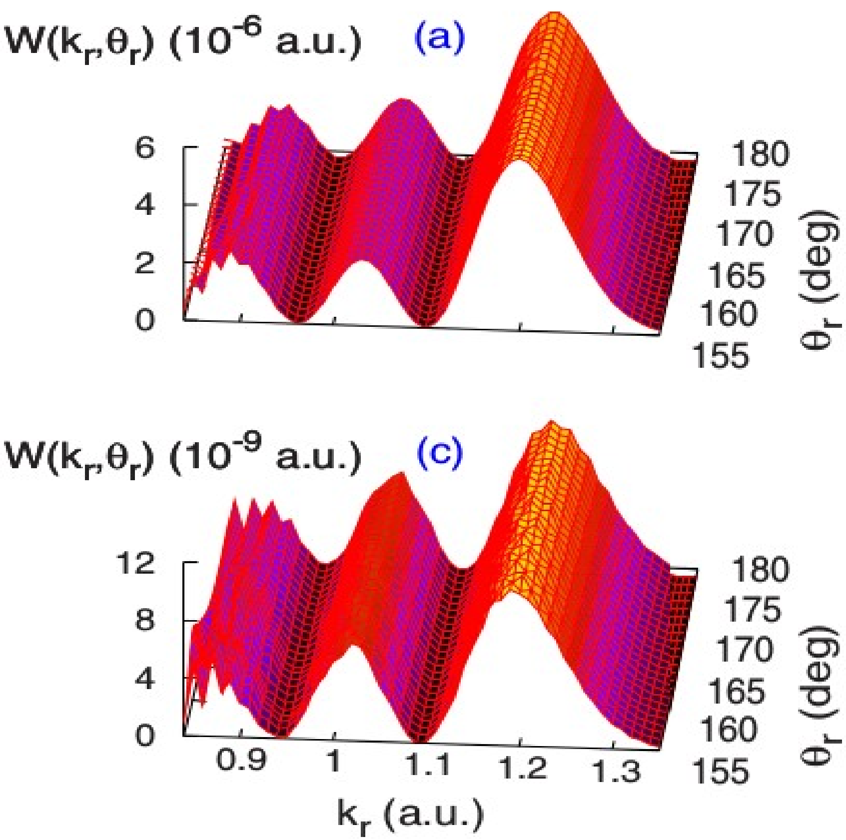
Zhangjin Chen, Anh-Thu Le, Toru Morishita, and C. D. Lin, Phys. Rev. A, **79**, 033409 (2009)

$$D(k,\theta) = W(k_r,\theta_r)\sigma(k_r,\theta_r)$$

Rescattered electron distribution

Distribution of returning electron wave

Laser-free differential cross section

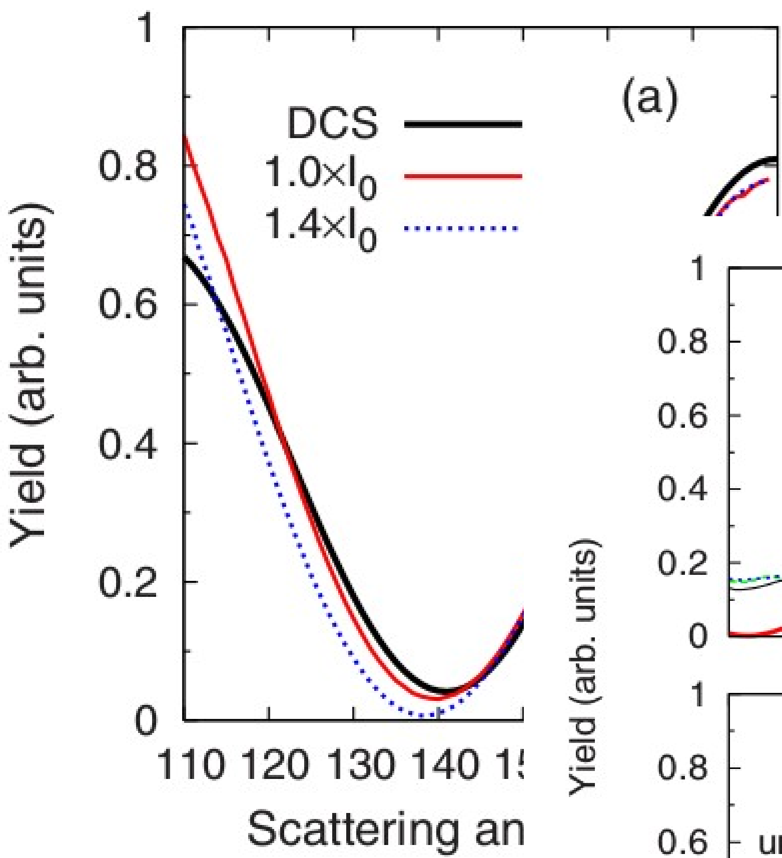


Returning electron wave is weakly dependent of scattering angle

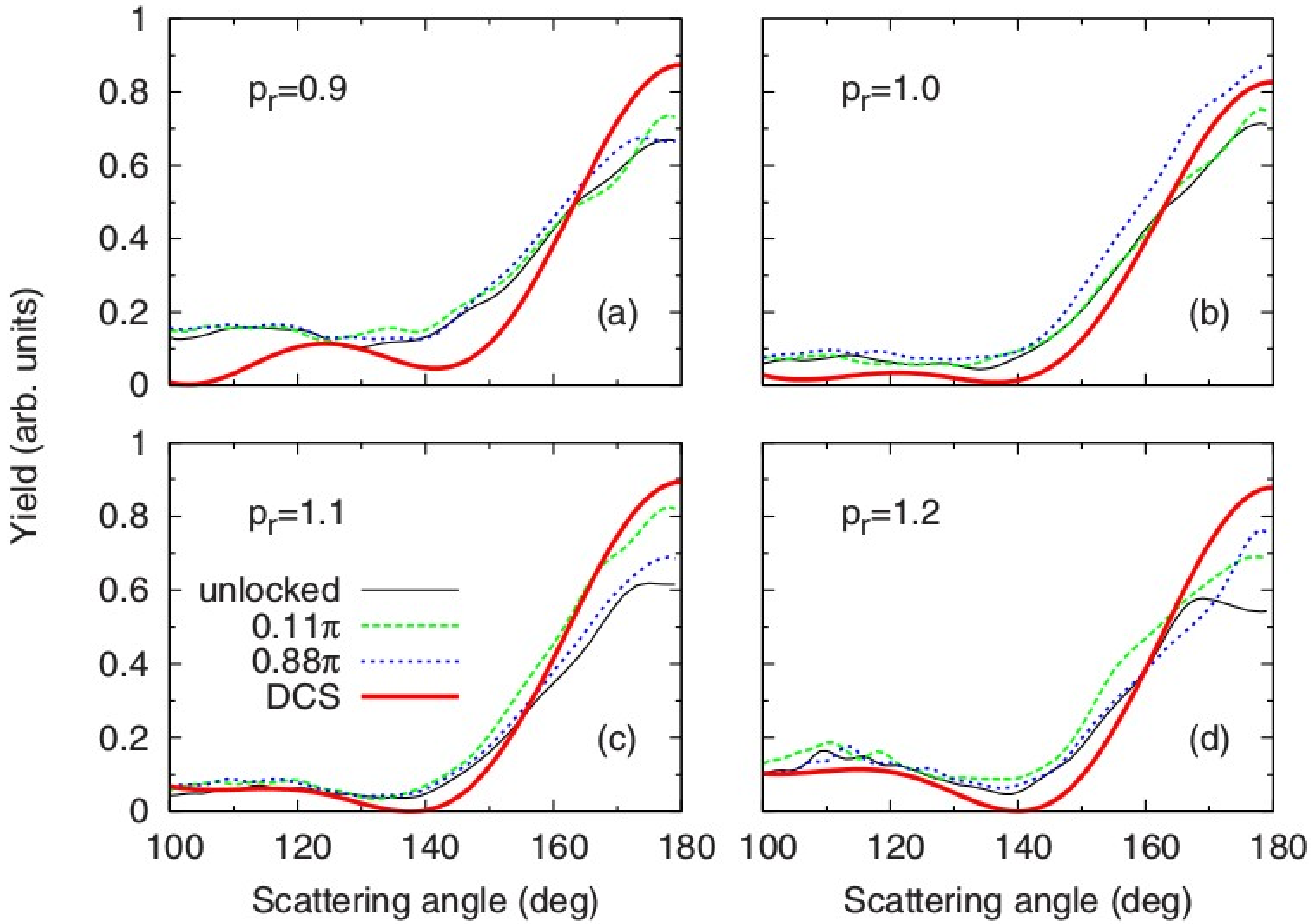
$$\rightarrow D(k,\theta) \propto \sigma(k_r,\theta_r)$$

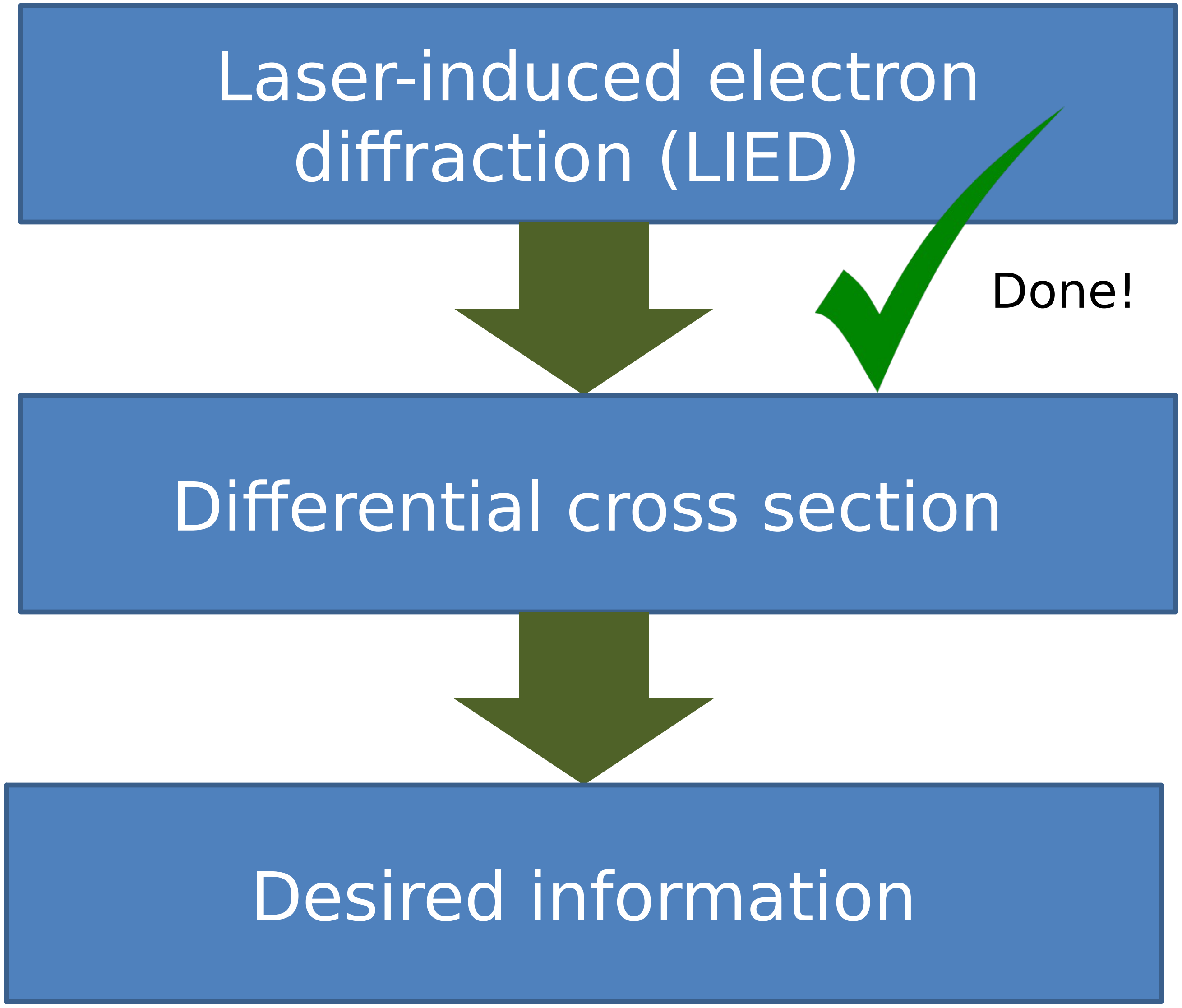
Quantitative rescattering theory

Zhangjin Chen, Anh-Thu Le, Toru Morishita, and C. D. Lin, Phys. Rev. A, **79**, 033409 (2009)



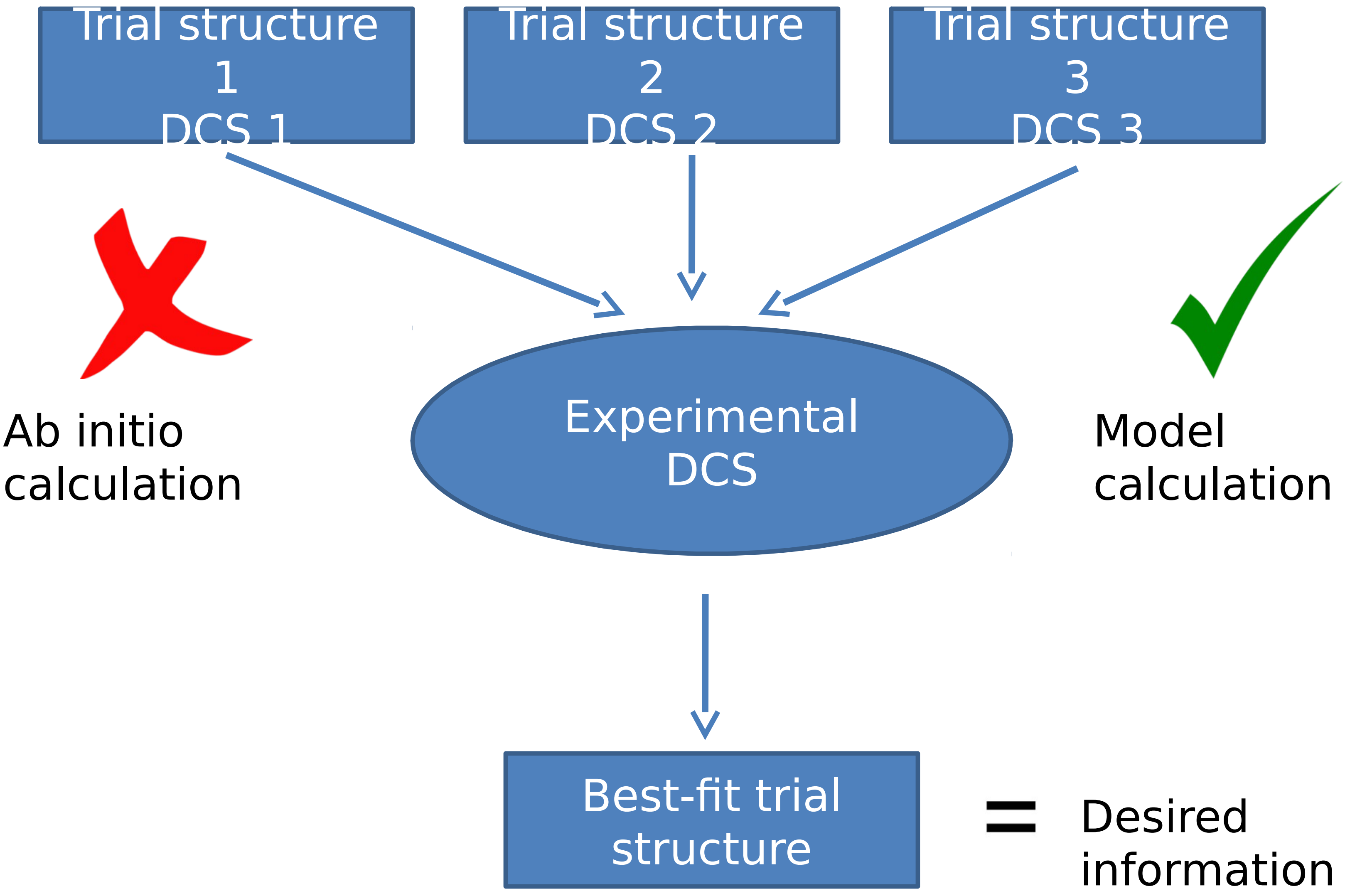
Extracted cross section is fairly independent of laser parameters




Laser-induced electron diffraction (LIED)
Done!
Differential cross section
Desired information

Retrieve structure from cross section

Cosmin I. Blaga et al, Nature **483** (2012)



Retrieve structure from cross section

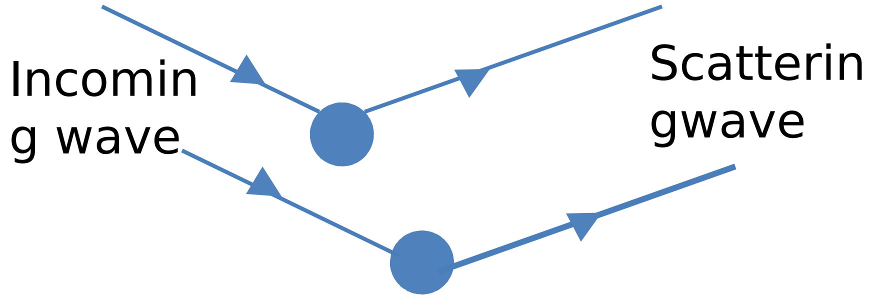
Cosmin I. Blaga et al, Nature **483** (2012)

- Independent Atoms Model (IAM) to generate theoretical DCS
- Molecule = collection of free atoms, omitting multiple scattering

$$F = \sum f_i \exp(i\mathbf{q}.\mathbf{R}_i)$$

Scattering amplitude of free component atoms

Spatial phase



$$I(\theta) = \langle |F|^2 \rangle = \sum |f_i(\theta)|^2 + \sum_{i \neq j} f_i(\theta) f_j^*(\theta) \frac{\sin qR_{ij}}{qR_{ij}}$$

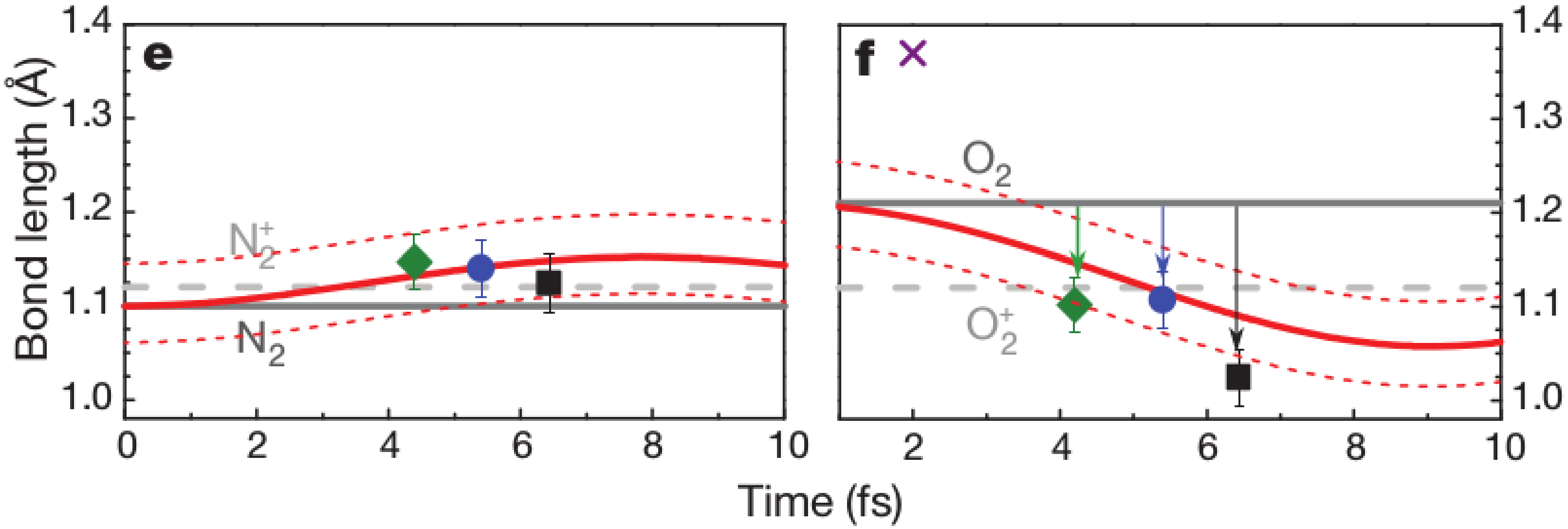
Molecular structure

IA: background term

Molecular Interference Factor (MIT)

Retrieve structure from cross section

Cosmin I. Blaga et al, Nature **483** (2012)



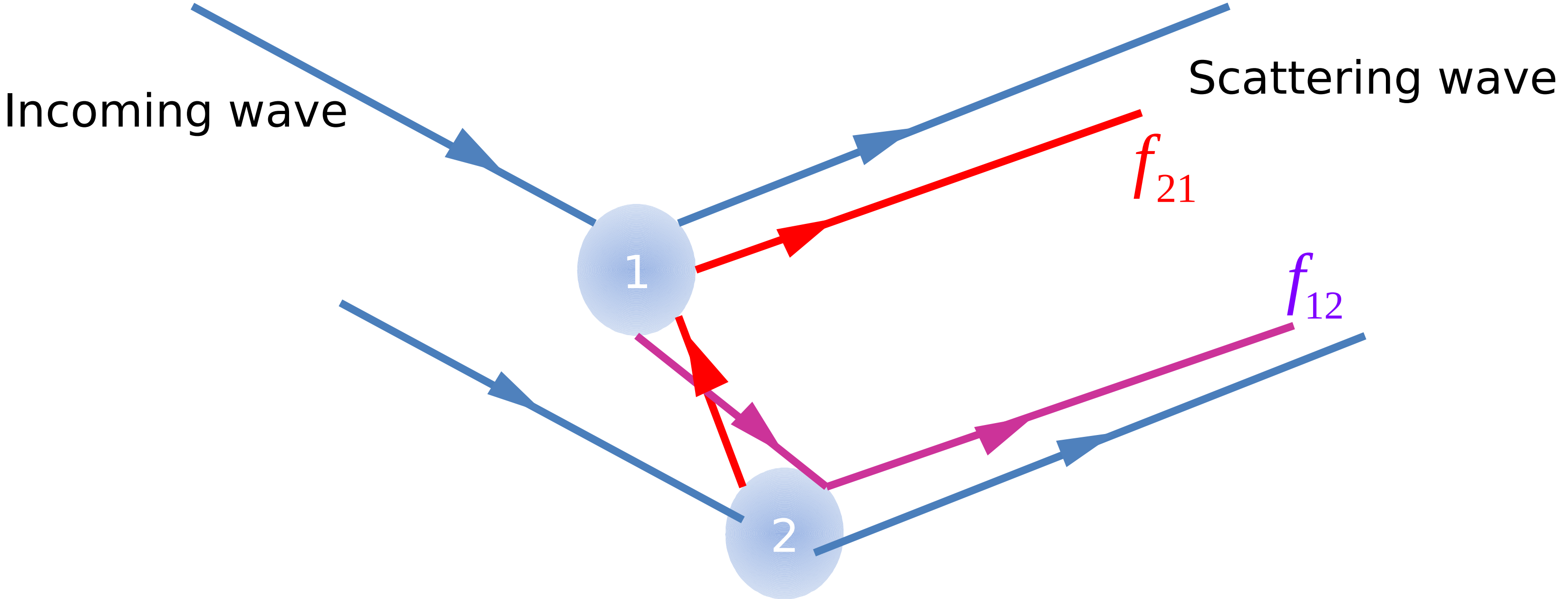
- Each point at different time is obtained by changing laser parameters (long wavelength -> long time)
- Problems

- IAM does not describe collision at low impact energy
- Experiments are confined to long wavelength laser (mid-infrared laser)
  → cannot probe at short time after ionization

Our solution: multiple scattering

Hayashi Shigeo and Kuchitsu Kozo, J. Phys. Soc. Jpn 41, 5, (1976)

- Multiple scattering (MS) theory: Including higher-orders scattering inside molecule analytically



Multiple scattering theory

- Up to 2nd order scattering

Incoming wave

$$F = -\frac{1}{4\pi}\langle \phi_{\mathbf{k}'} | T | \phi_{\mathbf{k}} \rangle$$

Outgoing wave

$$-\frac{1}{4\pi}\langle \phi_{\mathbf{k}'} | T_j | \phi_{\mathbf{k}} \rangle = e^{i(\mathbf{k}-\mathbf{k}').\mathbf{R}_j} f_j.$$

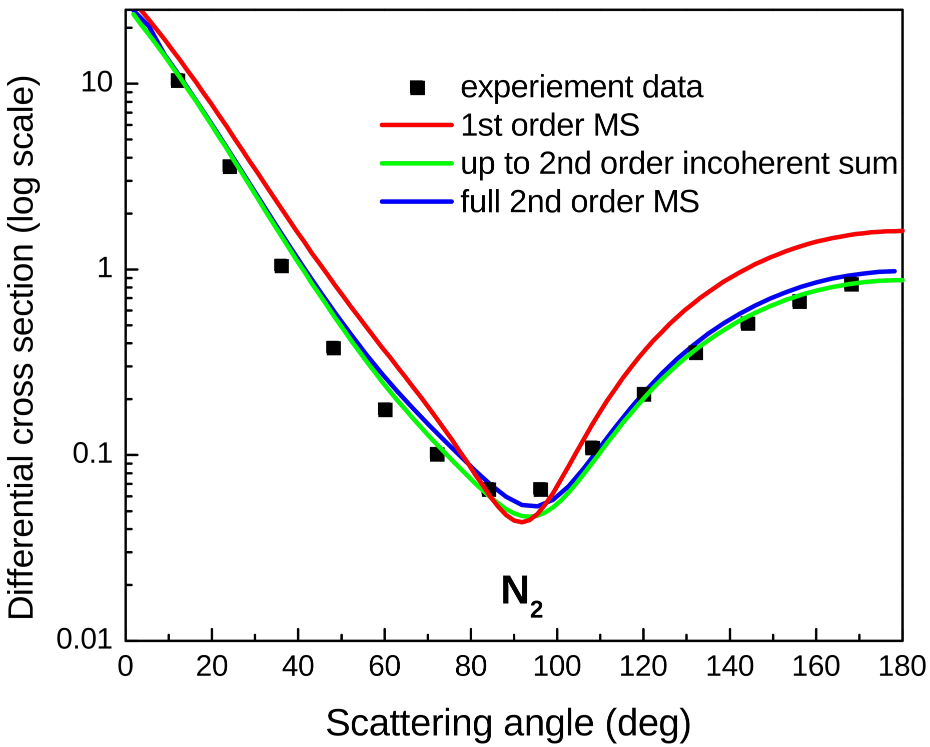
$$T = T_j + T_t + T_j G T_t + T_j G T_t$$

$$-\frac{1}{4\pi}\langle \phi_{\mathbf{k}'} | T_j G T_t | \phi_{\mathbf{k}} \rangle$$

$$= \frac{2\pi i}{k} e^{i\left(\mathbf{k}.\mathbf{R}_j - \mathbf{k}'.\mathbf{R}_t\right)} \sum_{l_1,l_2,l_3} i^{l_3} A_{l_1,j} A_{l_2,t} J_{l_3}(kR_{jt})(2l_1+1)\left[(2l_2+1)(2l_3+1)\right]^{1/2}$$

$$\times \begin{pmatrix} l_1 & l_2 & l_3 \\ 0 & 0 & 0 \end{pmatrix} \sum_{m_3} Y_{l_2}^{-m_3*}(\hat{\mathbf{k}}', \hat{\mathbf{k}}) Y_{l_3}^{m_3*}(\hat{\mathbf{R}}_{tj}, \hat{\mathbf{k}}) \begin{pmatrix} l_1 & l_2 & l_3 \\ 0 & -m_3 & m_3 \end{pmatrix}$$

$A_l = e^{i\delta_l}\sin\delta_l$ for δ_l is the phase shift

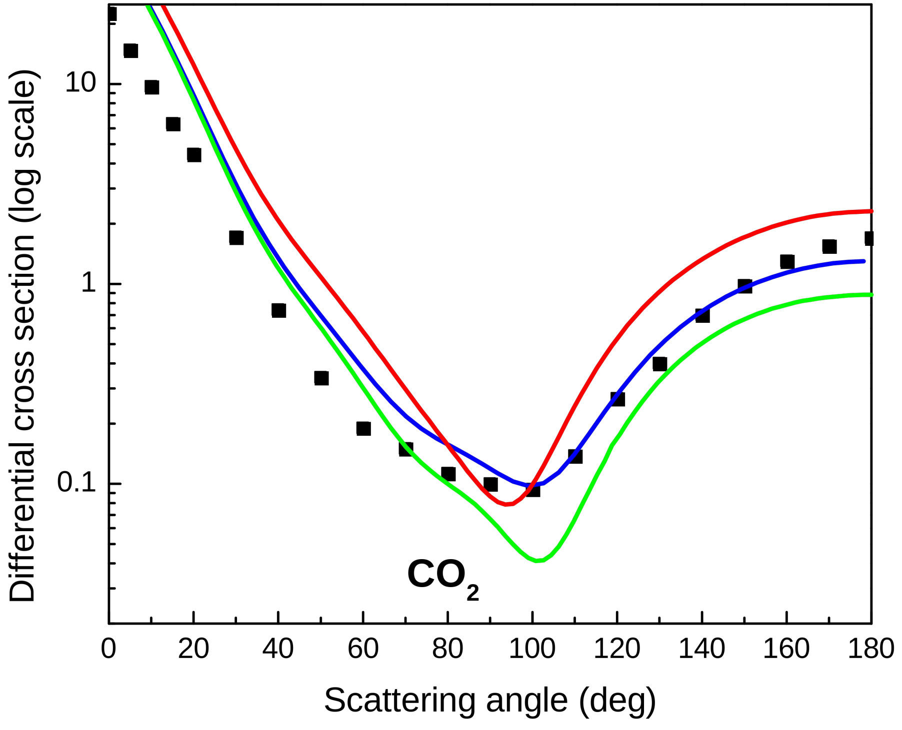


Experimental DCS:

T W Shyn, and G R Carignan, Phys. Rev. A **22**, 3 (1980)

T W Shyn, W E Sharp, and G R Carignan, Phys. Rev. A **17**, 6 (1978)

New model MS produces better differential cross section than IAM



Test on gas phase electron diffraction

Collision energy	By IAM	By MS
20	-	1.293 (7.1%)
30	1.697 (41%)	1.201 (0.5%)
40	1.604 (33%)	1.165 (3.5%)
50	1.372 (14%)	1.193 (1.2%)

-: error > 50% or not single local minimum inside search zone

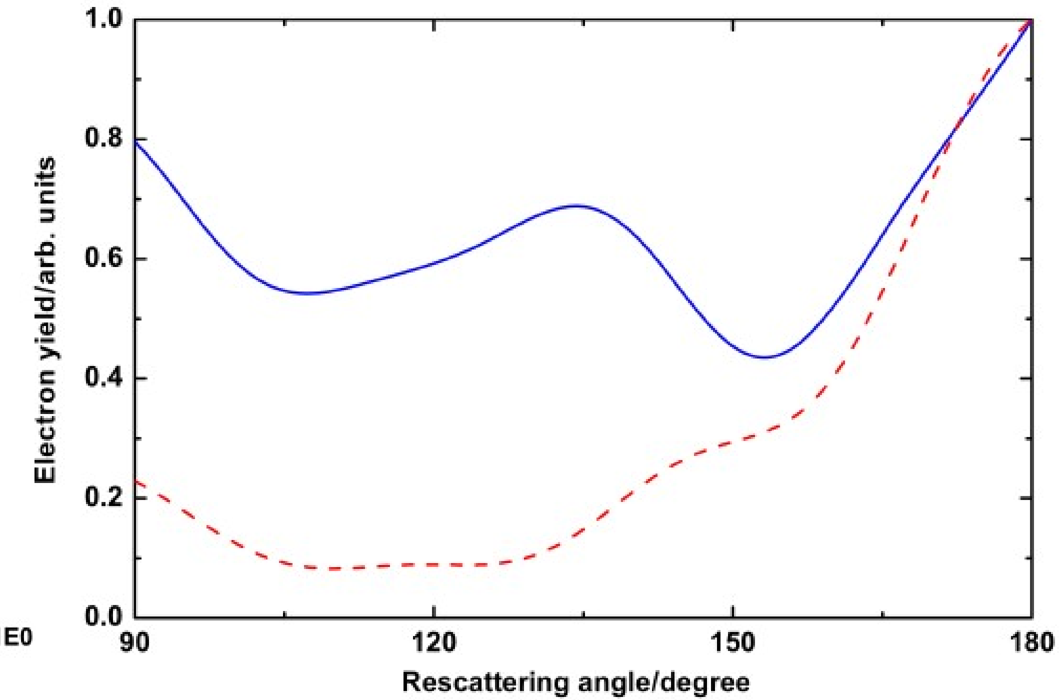
Collision energy	By IAM	By MS
20	-	1.174 (7.0%)
30	-	1.042 (5.1%)
40	-	1.157 (5.4%)

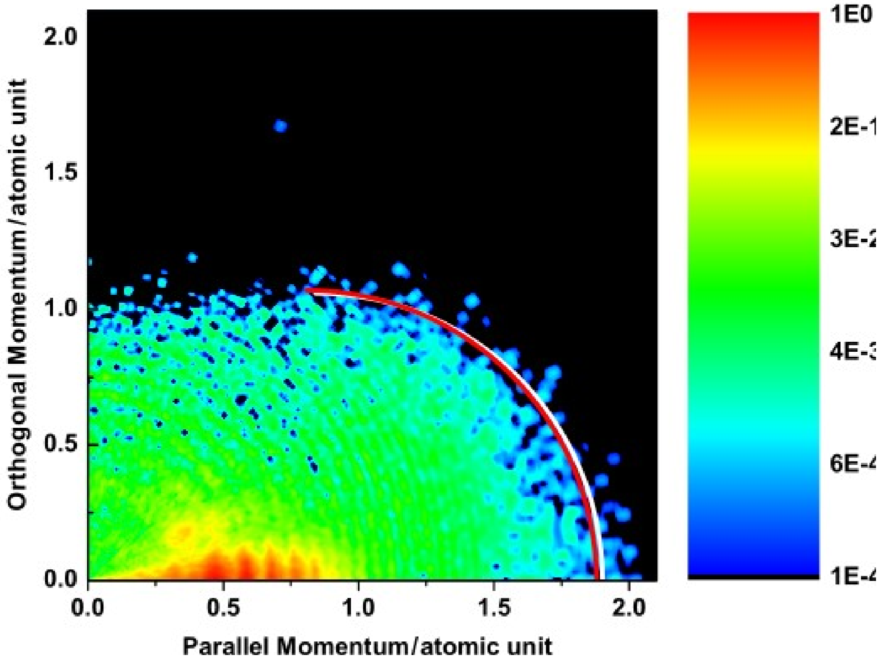
Structural retrieval from LIED data

Differential cross section of CO_2

15 eV

30 eV





C Cornaggia, J. Phys. B: At. Mol. Opt. Phys. 42 (2009)

$$\lambda = 795 \text{ nm}, I = 0.83 \times 10^{14} \text{ W/cm}^2$$

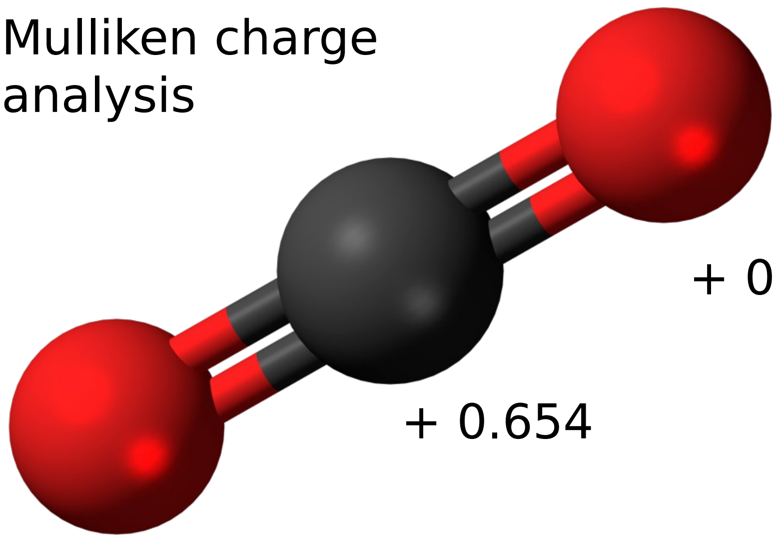
$$\rightarrow E \approx 15 \text{ eV}$$

Structural retrieval from LIED data

- Compared to gas phase electron diffraction, LIED differs in two points

- the collision is e – ion -> require long-ranged Coulomb potential
- Incoming electron flux depends on molecule orientation through ionization r
average

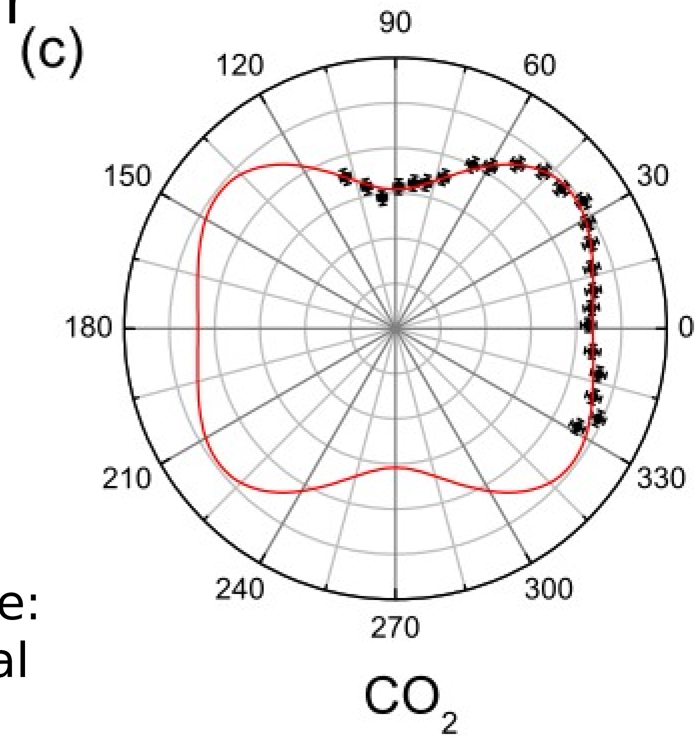
Mulliken charge analysis



+ 0.173

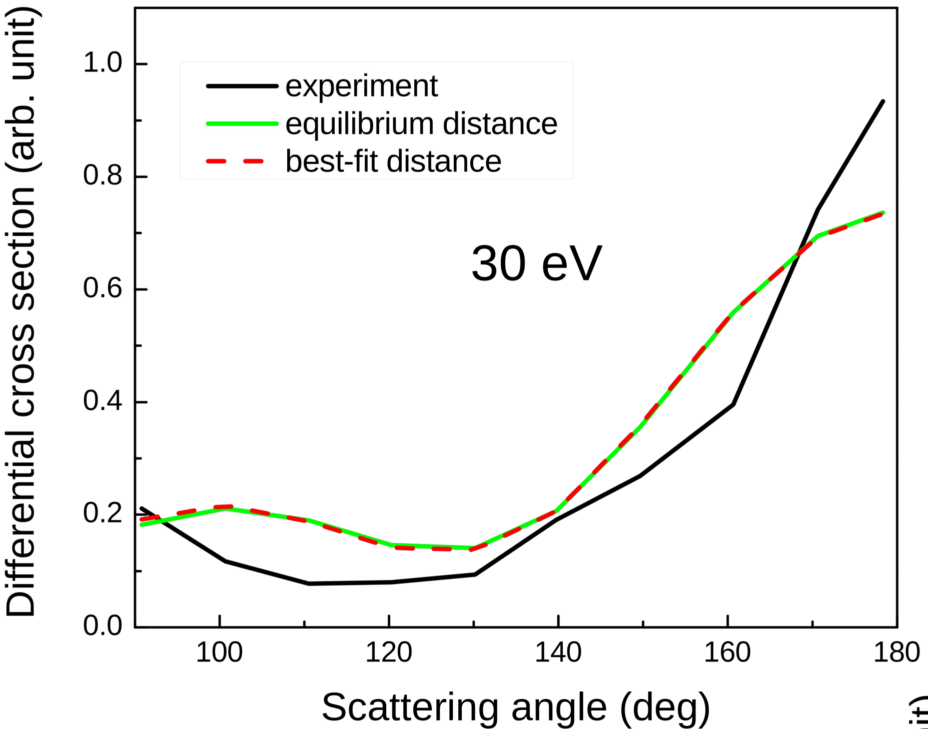
+ 0.654

+ 0.173



Ionization rate:
- Experimental data
- Calculate by MO-ADK

Domagoj Pavičić et al, Phys. Rev. Lett 98,

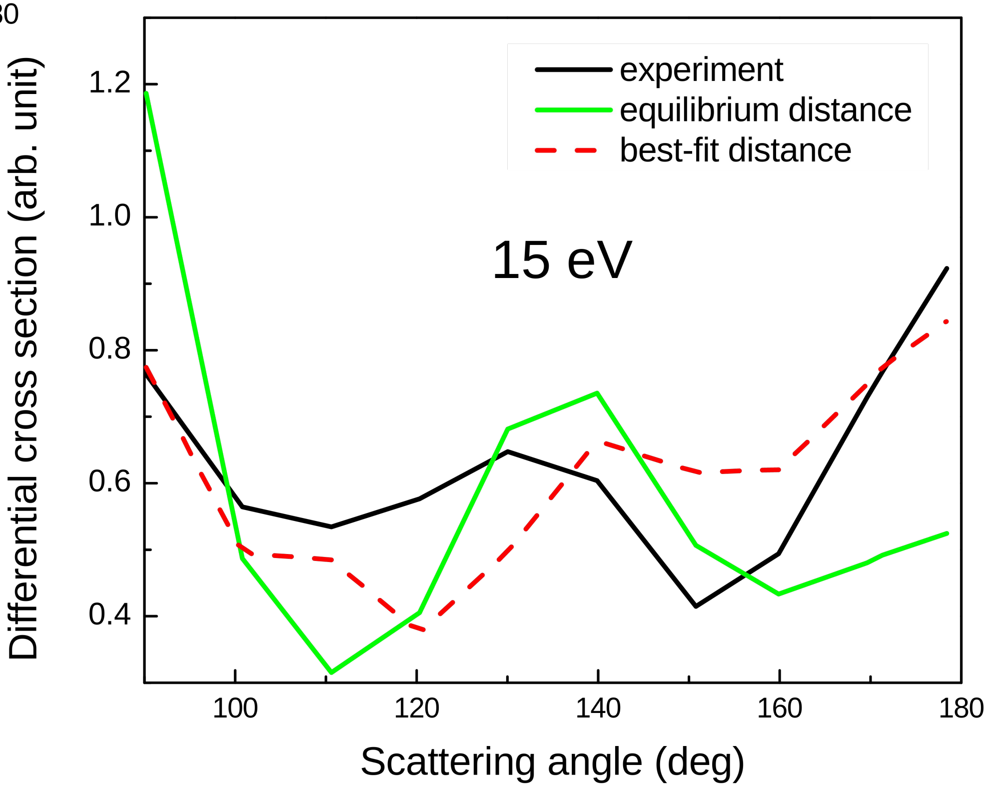


Collision energy = 15eV

$$\varepsilon = \frac{|R_{best-fit} - R_0|}{R_0} = 16\%$$

Collision energy = 30eV

$$\varepsilon = \frac{|R_{best-fit} - R_0|}{R_0} = 1.9\%$$



THANK YOU FOR YOUR ATTENTION