Low-density matter with synchrotrons and time-resolved experiments with FELs

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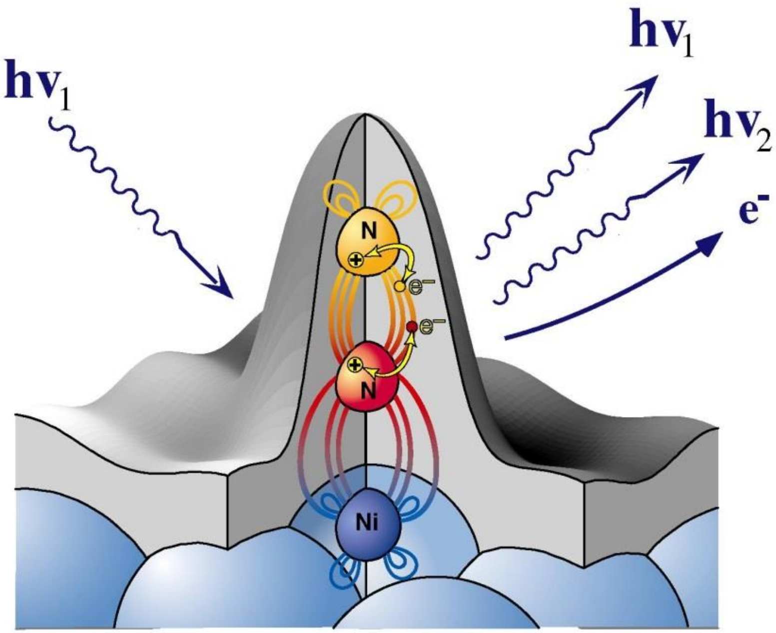
Photon Interaction

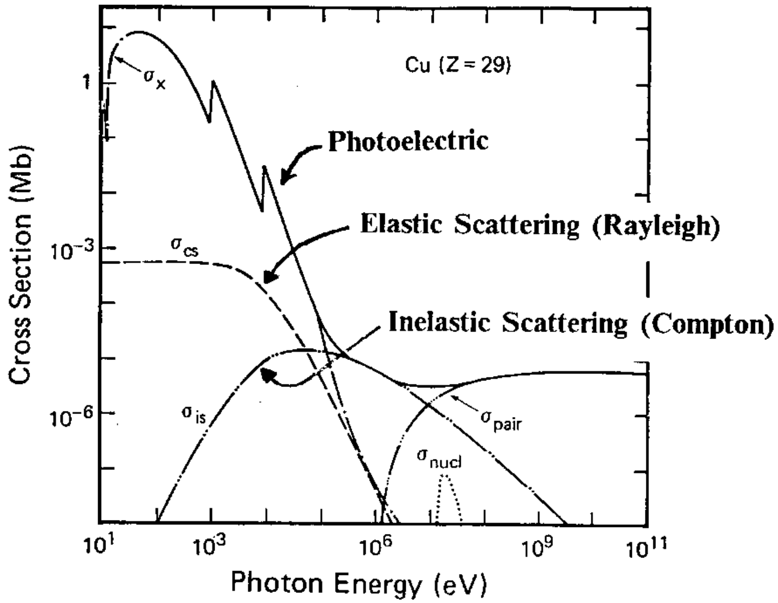
Incident photon interacts with electrons



- Photon is
  - Adsorbed
  - Elastically Scattered
  - Inelastically Scattered

- Electron is
  - Emitted
  - Excited
  - De-excited

Cross Sections

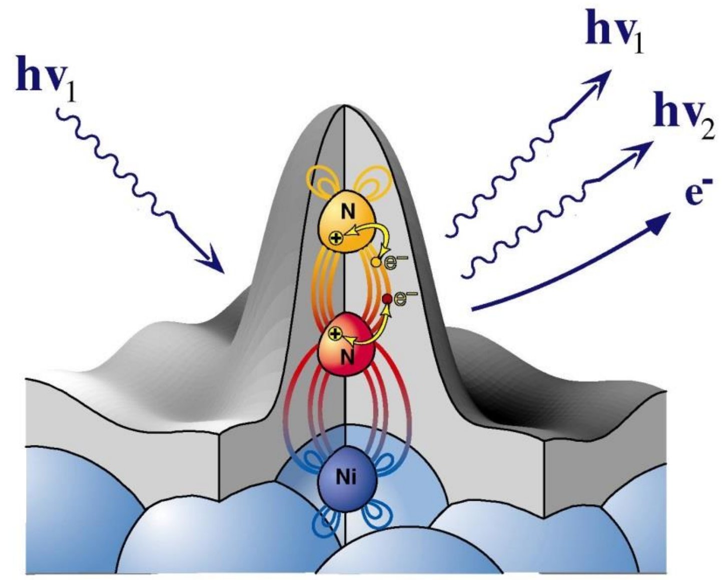


Below 100 keV

Photoelectric and elastic cross section dominate

Spectroscopy-Scattering

Detected Particles



EMITTED PARTICLE

- *Elastic Scattering* X-Diffraction
- *Inelastic Scattering* X-ray Emission Spectroscopy
- *Electron Emission* Photoelectron Spectroscopy

NO EMITTED PARTICLE

- *Photon Adsorbed* X-ray Absorption Spectroscopy

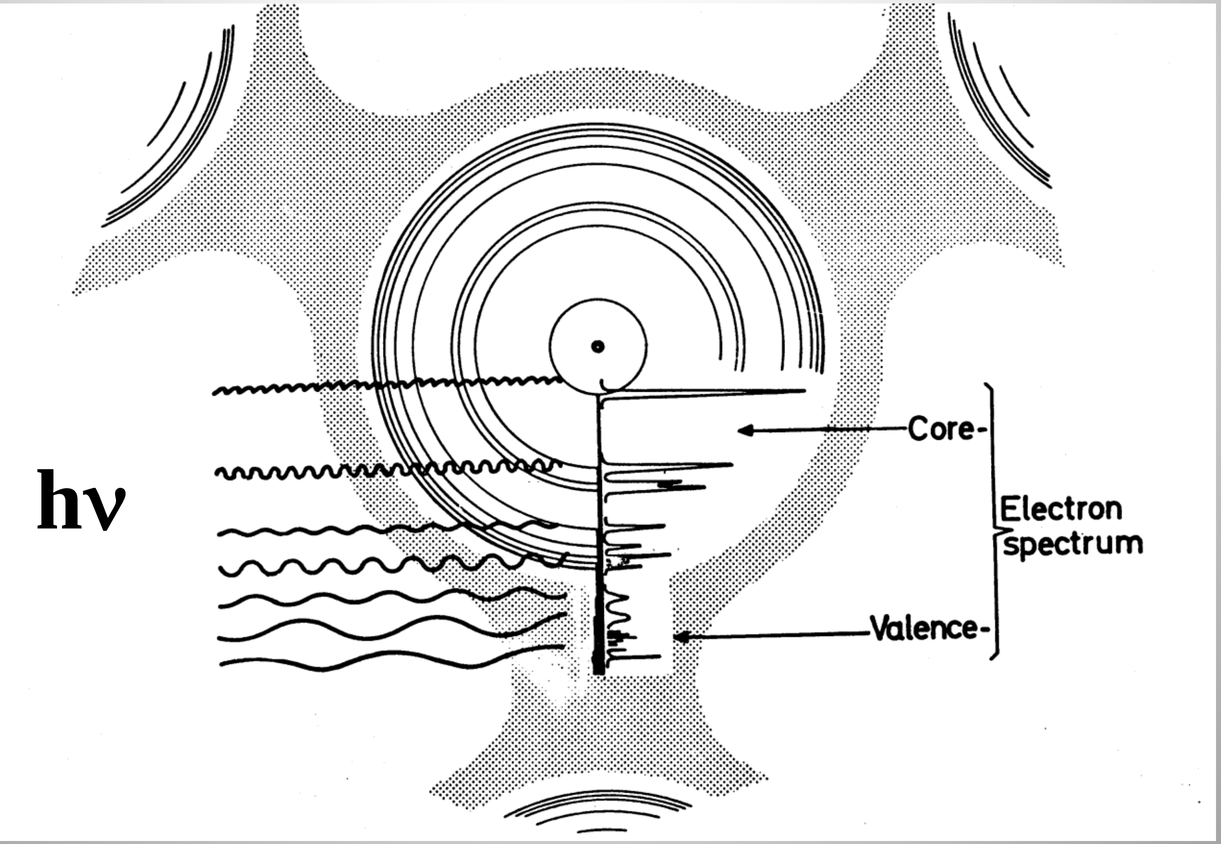
Spectroscopy

Valence electrons → Chemical Bonding

Core electrons → Non interacting

Ionization → Photoelectron Spectroscopy

hν



Methods

- X-ray Diffraction

- Photoelectron Spectroscopy (PES)

Core level electron spectroscopy

Valence band photoemission

Resonant photoemission

- X-ray Absorption Spectroscopy (XAS)

Near Edge X-ray Absorption Spectroscopy (NEXAFS)

Extended X-ray Absorption Fine Structure (EXAFS)

- X-ray Emission Spectroscopy (XES)

Resonant Inelastic X-ray Scattering (RIXS)

Selected examples:

Resonant photoemission

Ultrafast dynamics

Young’s double slit-type interference

Doppler effects

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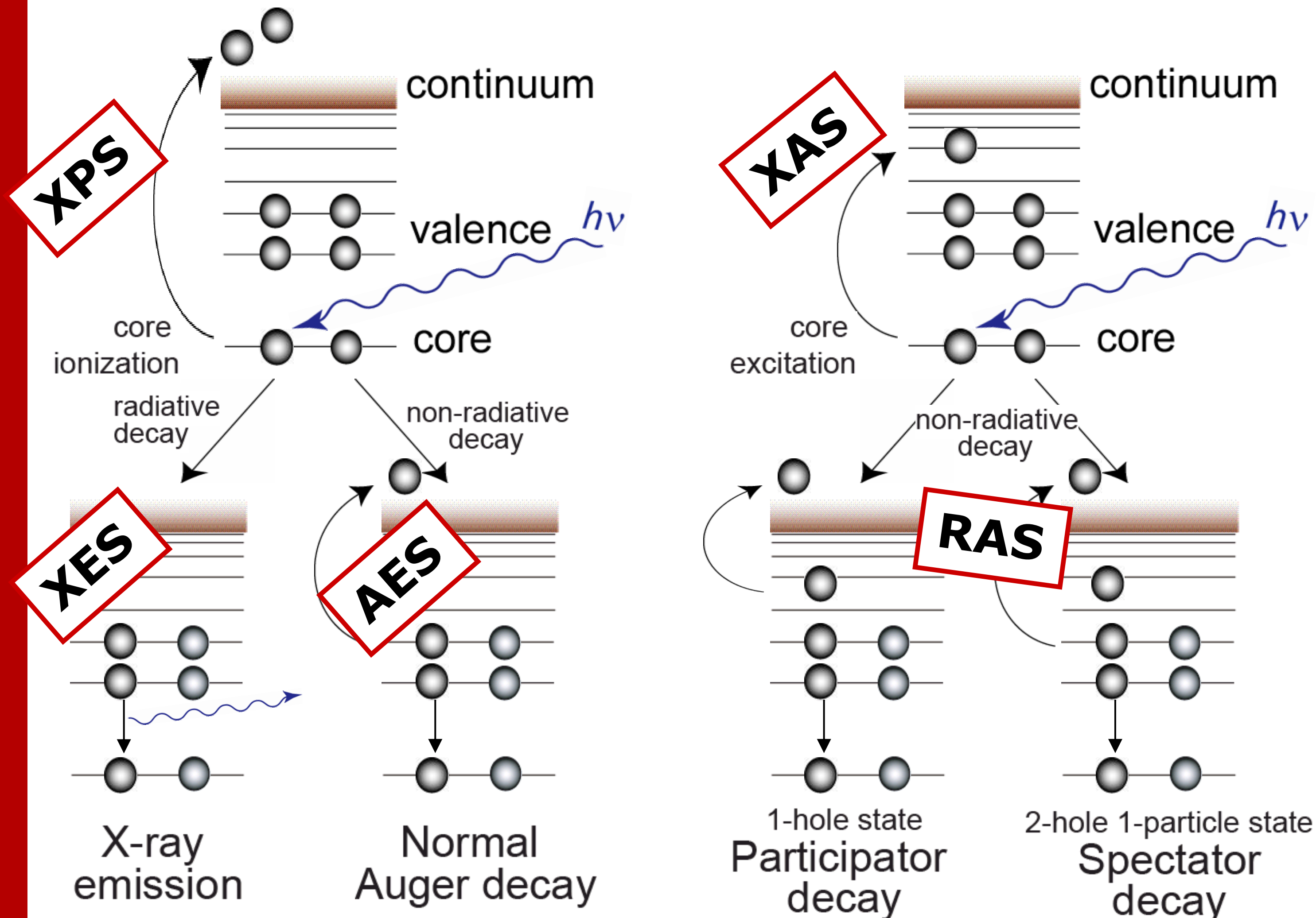
Electron Spectroscopy



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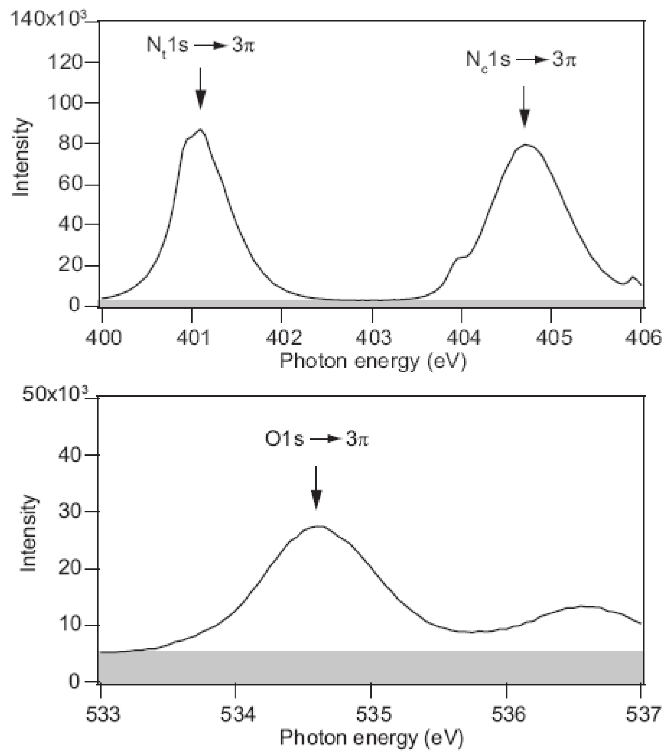
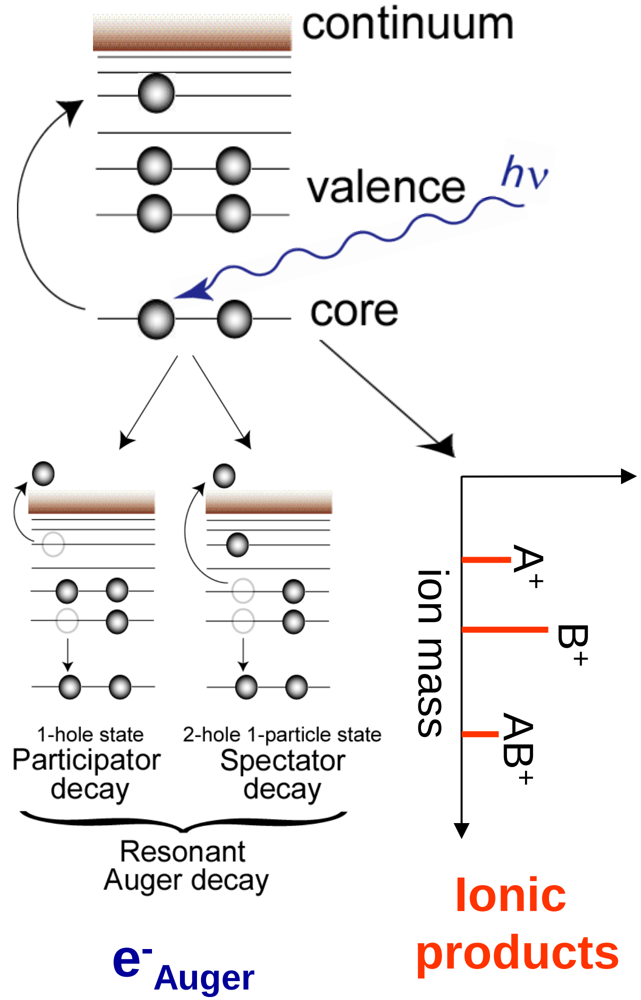
X-ray Absorption Spectroscopy of N_2O

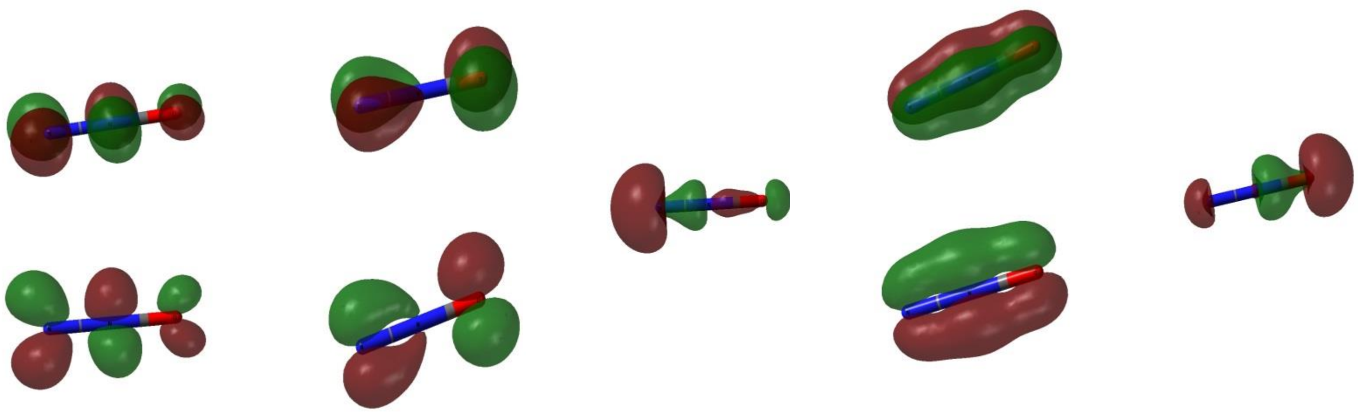
N≡N=O





Decay Processes in Core-Excited N_2O

$$1\sigma^2 2\sigma^2 3\sigma^2 4\sigma^2 5\sigma^2 6\sigma^2 1\pi^4 7\sigma^2 2\pi^4 3\pi \quad {}^1\Sigma^+$$



M N Piancastelli et al., J.Phys.B: At.Mol.Opt.Phys. 40, 3357(2007)

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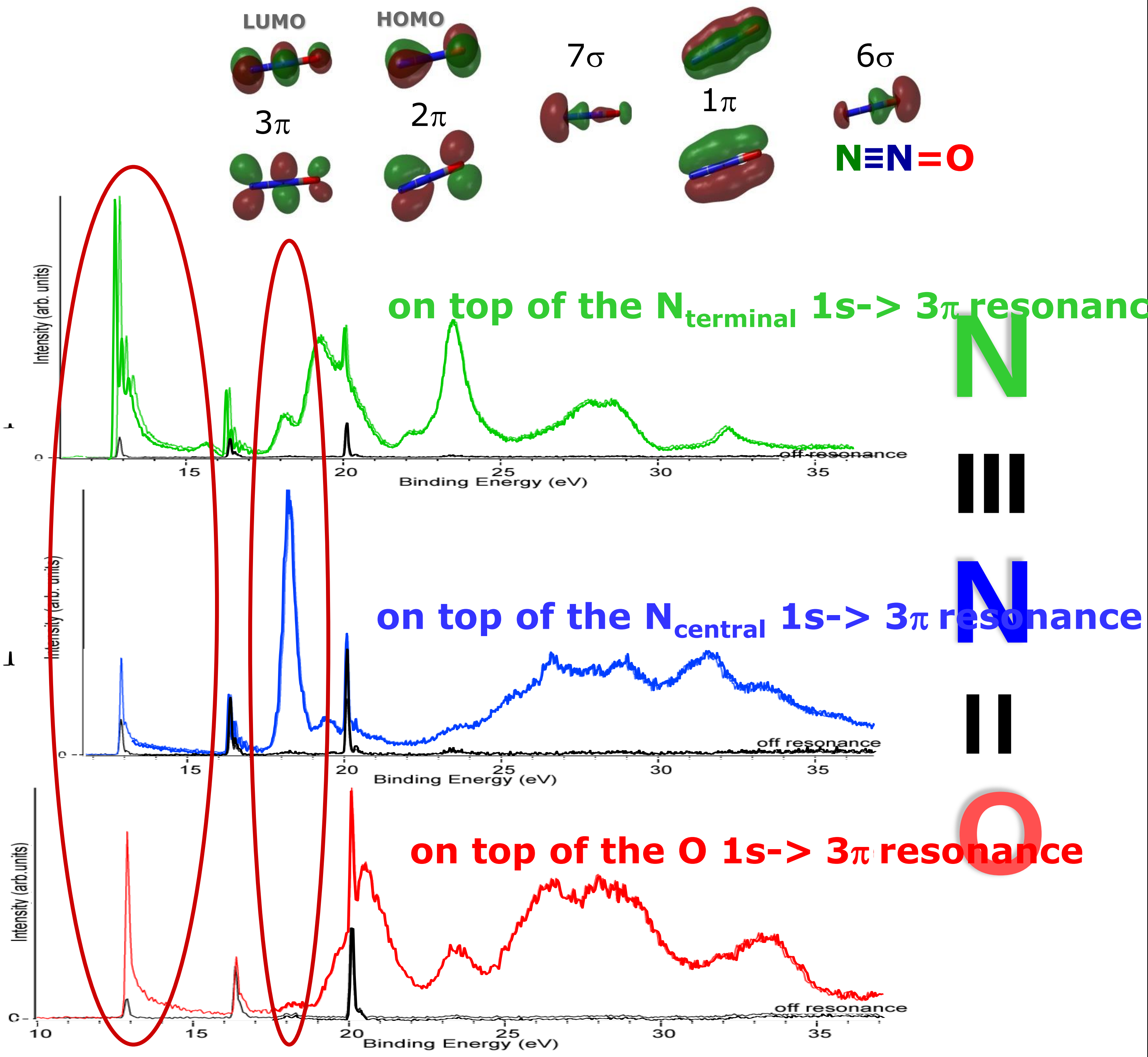

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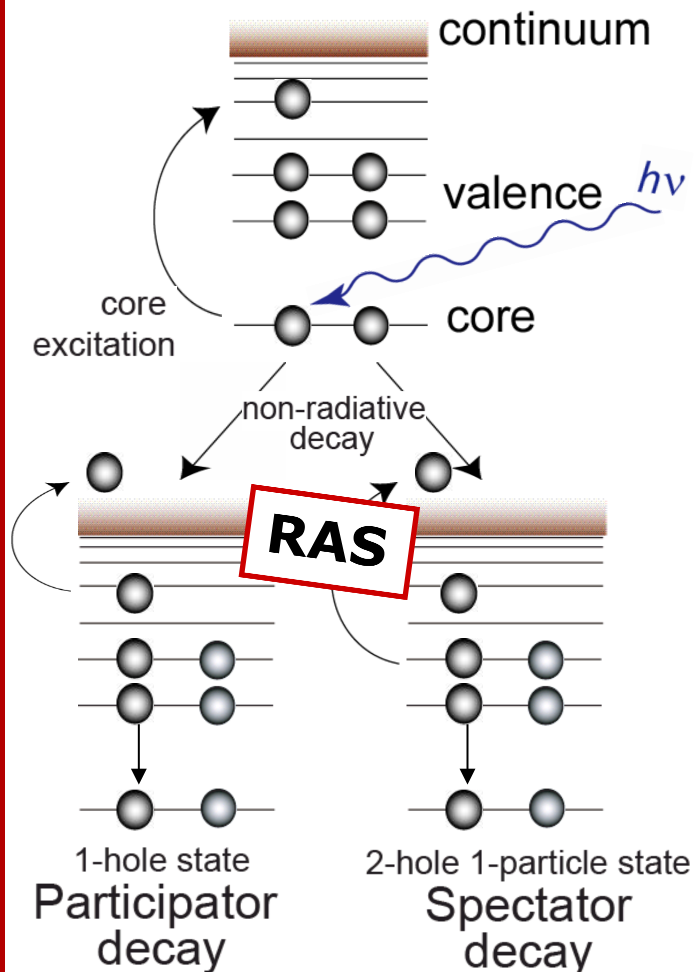
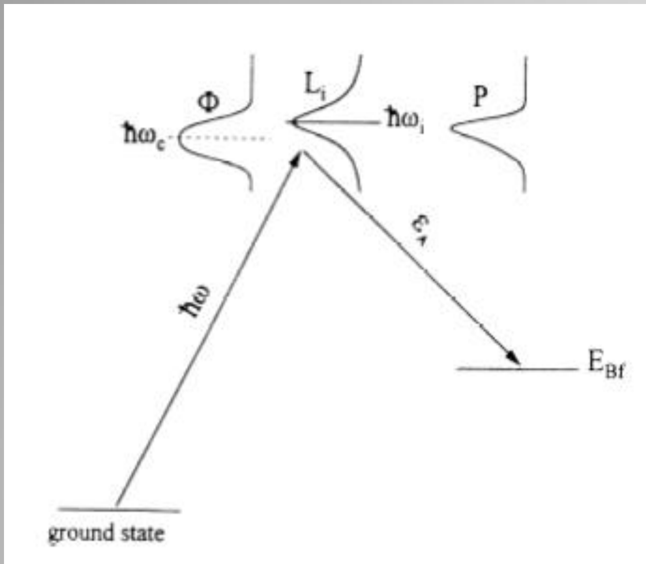
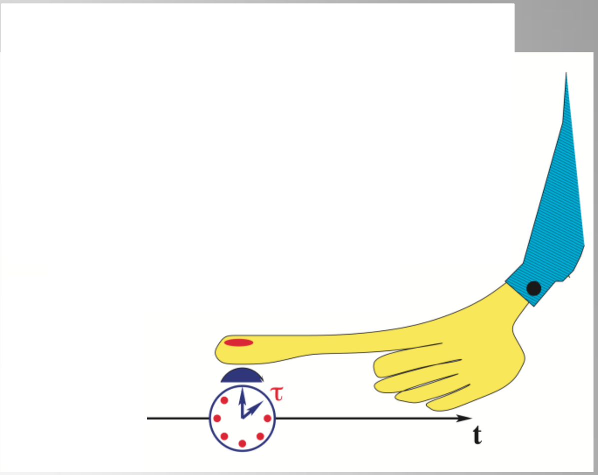
Nuclear dynamics of core-excited systems



Possible mechanisms of nuclear dynamics:

- ultrafast dissociation
- geometry change
  - e.g. bending, twisting
- conformational changes



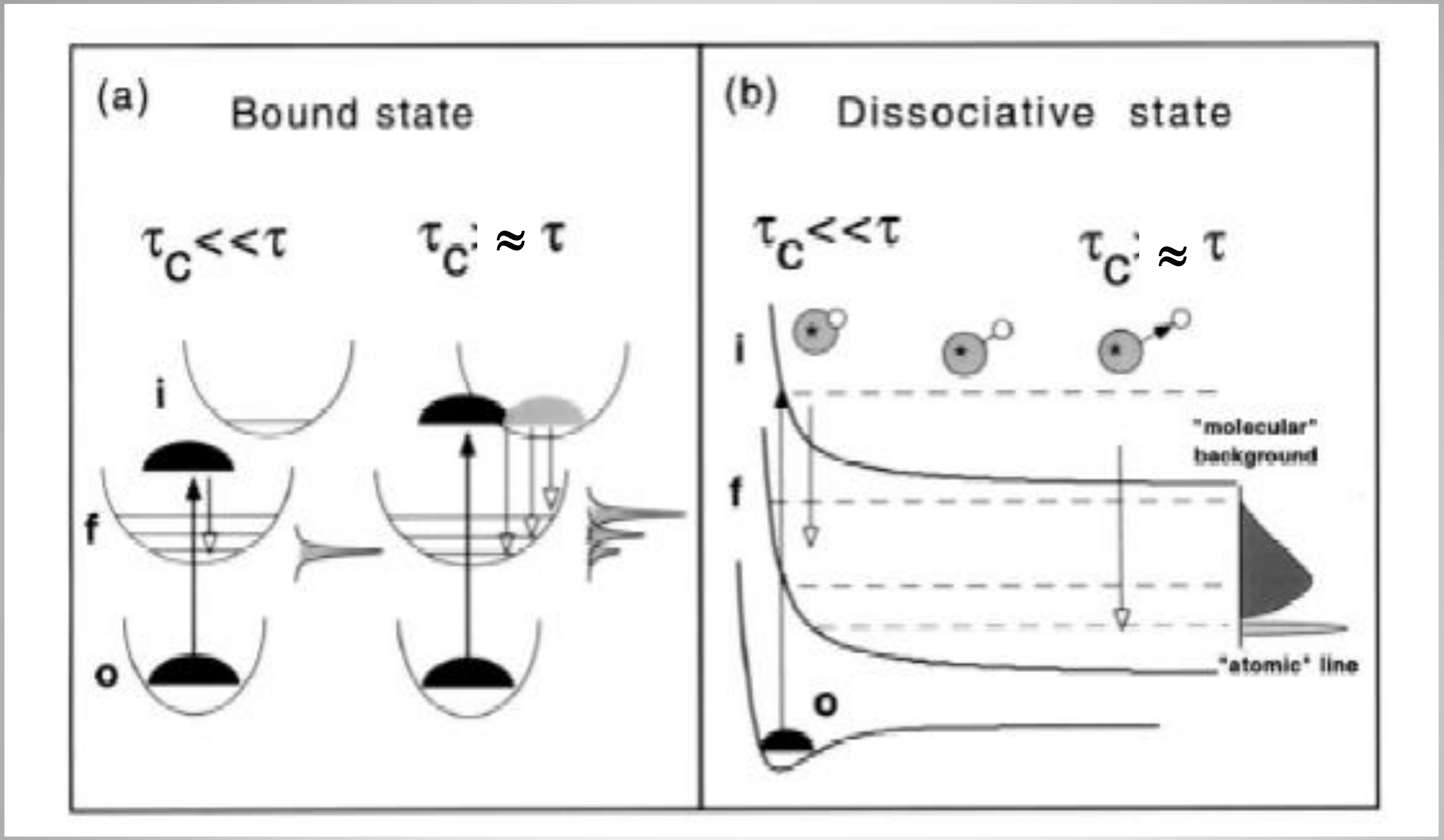


Auger resonant Raman conditions:

Photon bandwidth much narrower than the natural lifetime width of the intermediate state

Core-hole clock

$$(\Delta T)\ (\Delta E) \geq \hbar/2$$



duration time

$$\tau_c = \frac{1}{\sqrt{\Gamma^2 + \Omega^2}}$$

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Nuclear Dynamics of core-excited systems

Ultrafast dissociation

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Atomic Autoionization Following Very Fast Dissociation of Core-Excited HBr

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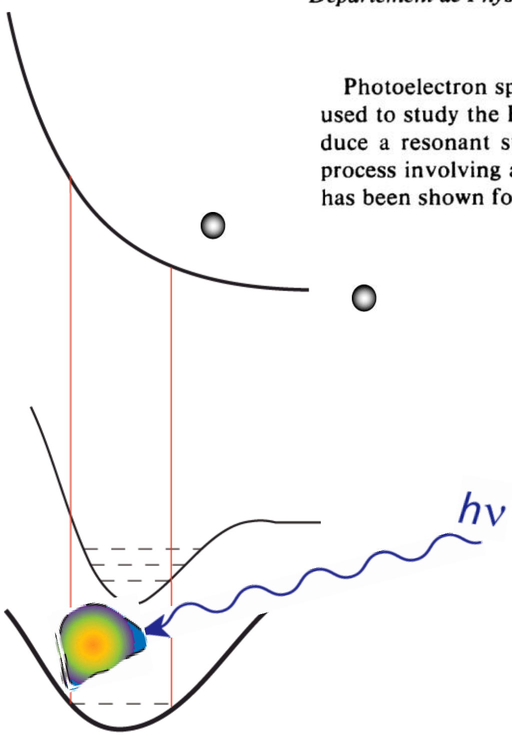
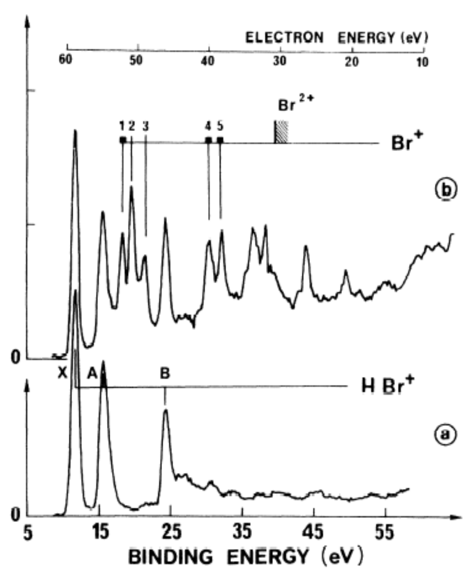
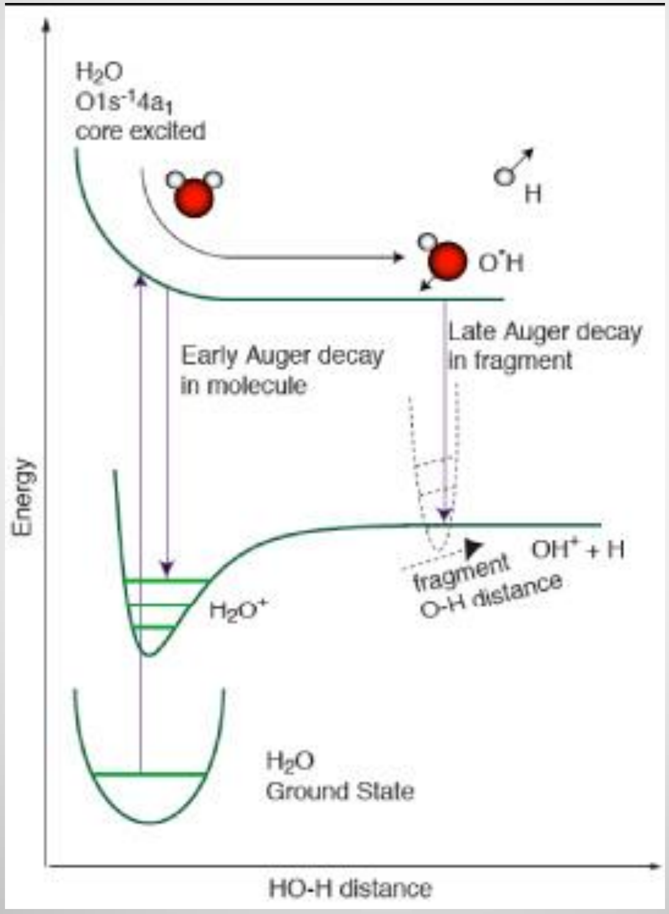
(Received 28 February 1986)

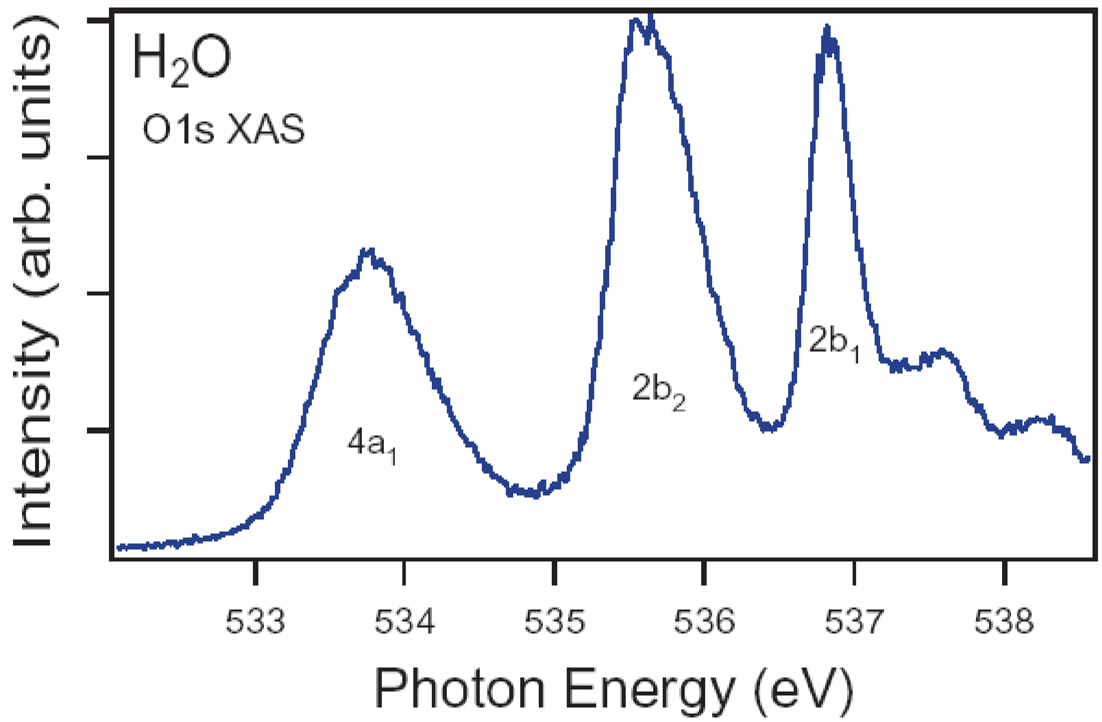
Photoelectron spectroscopy excited by monochromatic synchrotron radiation (68–80 eV range) is used to study the Br 3*d* excitation in HBr. The transition to an antibonding orbital is shown to produce a resonant state whose repulsive nature has been observed directly. A two-step relaxation process involving a fast neutral dissociation followed by the autoionization of the excited fragment has been shown for the first time.



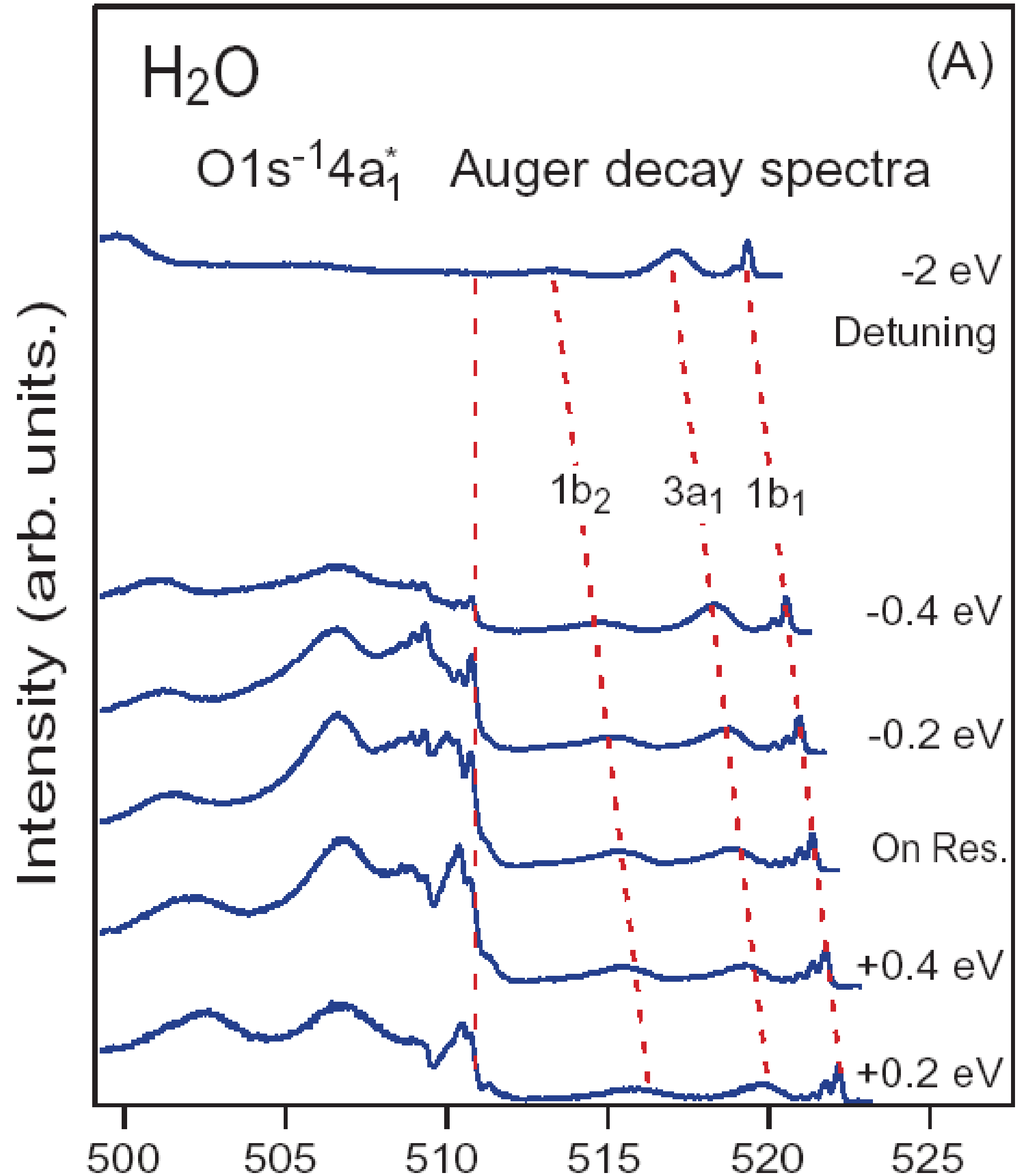


I.Hjelte, M.N.Piancastelli, R.F.Fink, O.Björneholm, M.Bässler, R.Feifel, A.Giertz, H.Wang, K.Wiesner, A.Ausmees, C.Miron, S.L.Sorensen and S.Svensson, Chem.Phys.Lett. 334, (2001) 151

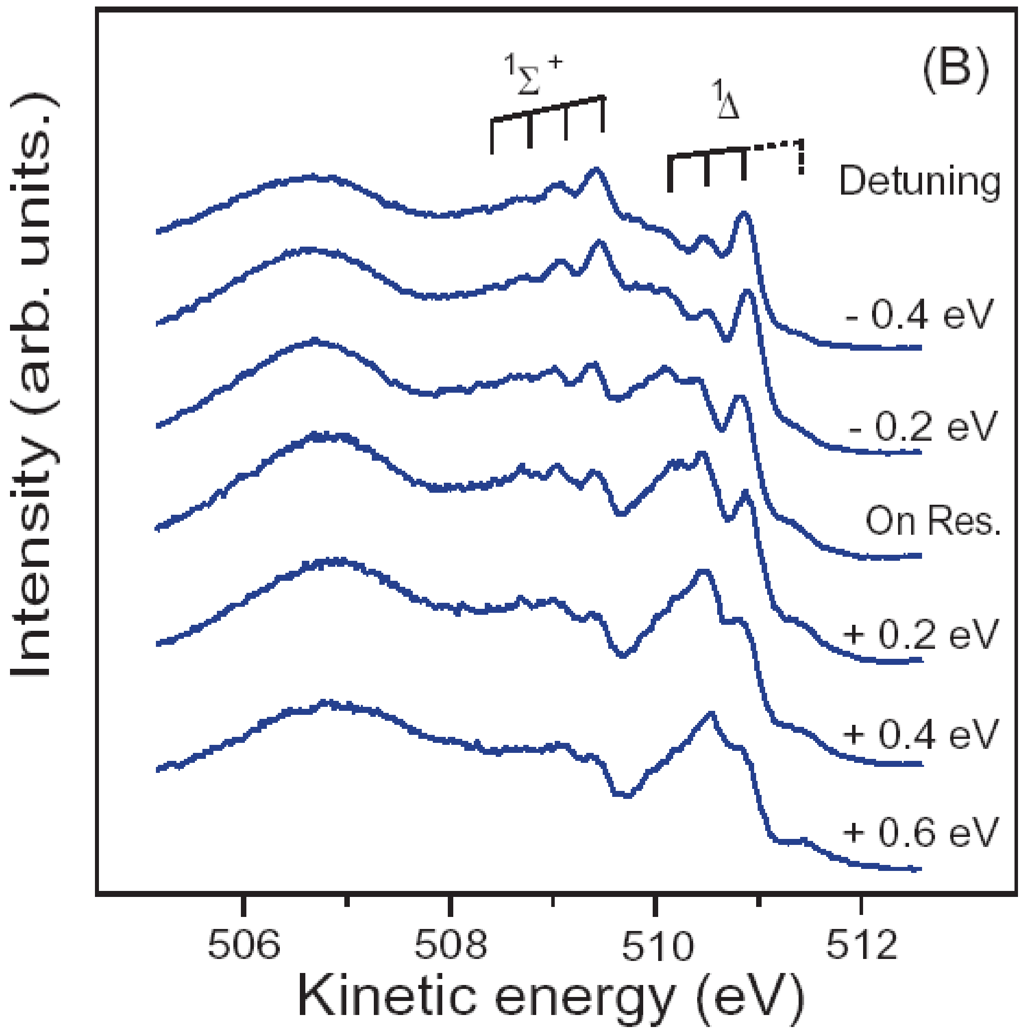




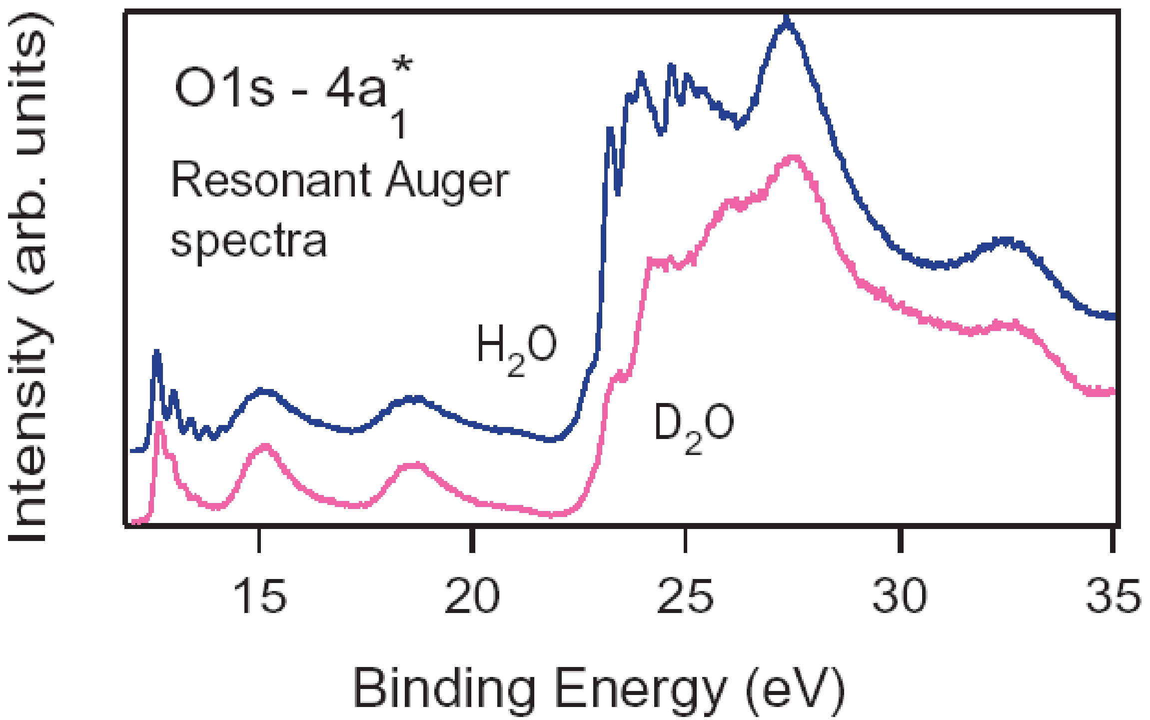
H_2O
O1s XAS
$4a_1$
$2b_2$
$2b_1$
Intensity (arb. units)
533
534
535
536
537
538
Photon Energy (eV)



H2O
(A)
O1s-14a1* Auger decay spectra
-2 eV
Detuning
1b2
3a1
1b1
-0.4 eV
-0.2 eV
On Res.
+0.4 eV
+0.2 eV
Intensity (arb. units.)
500
505
510
515
520
525

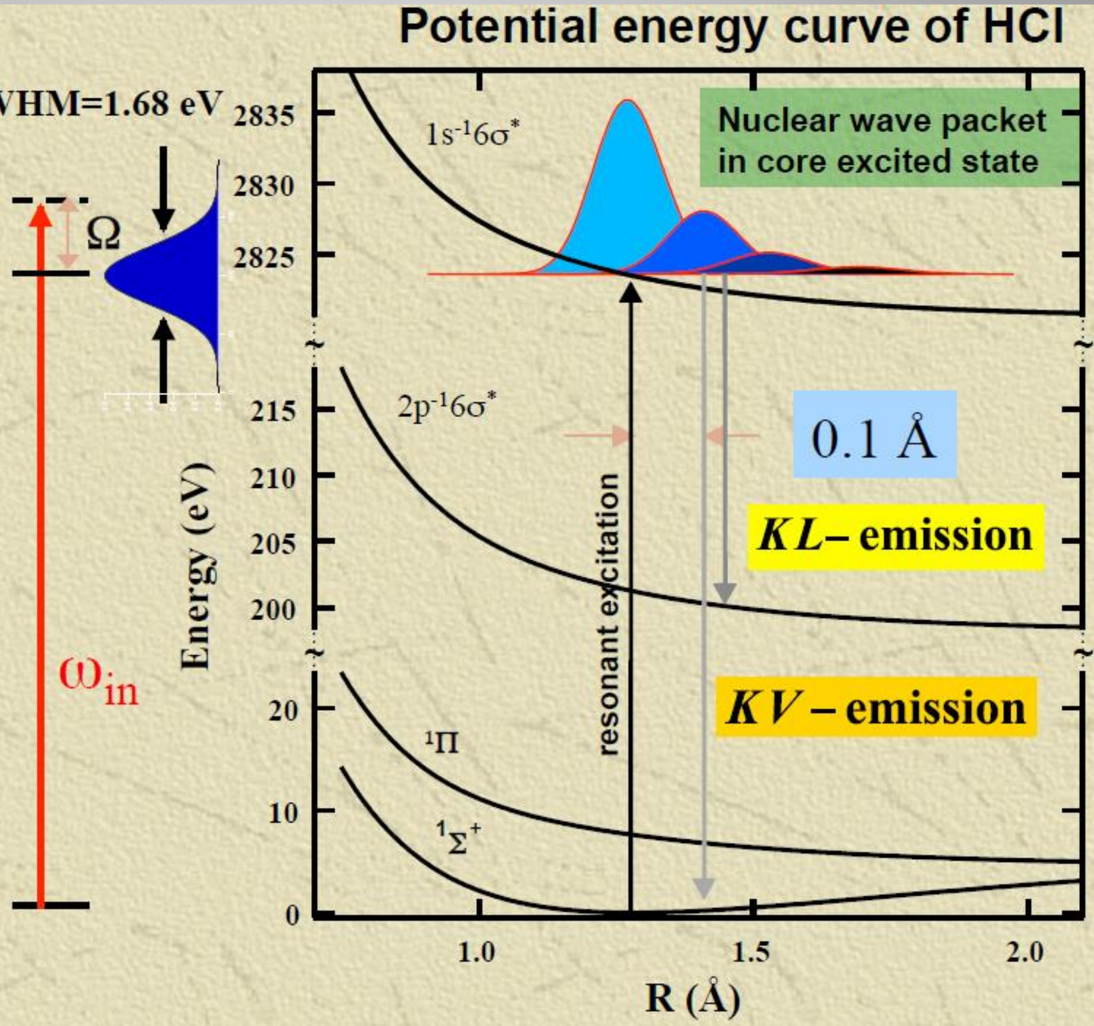


(B)
$^1\Sigma^+$
$^1\Delta$
Detuning
- 0.4 eV
- 0.2 eV
On Res.
+ 0.2 eV
+ 0.4 eV
+ 0.6 eV
Intensity (arb. units.)
506
508
510
512
Kinetic energy (eV)

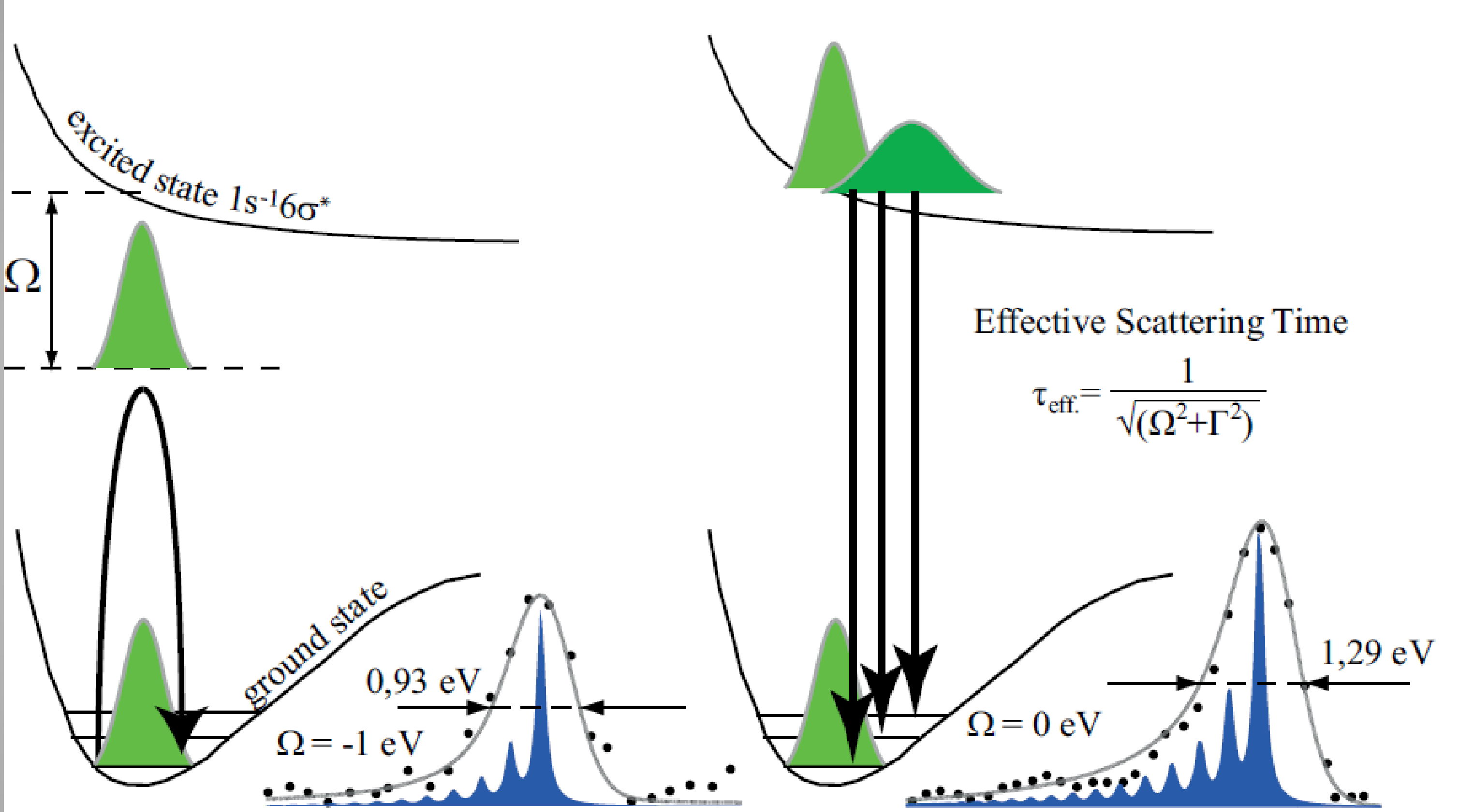


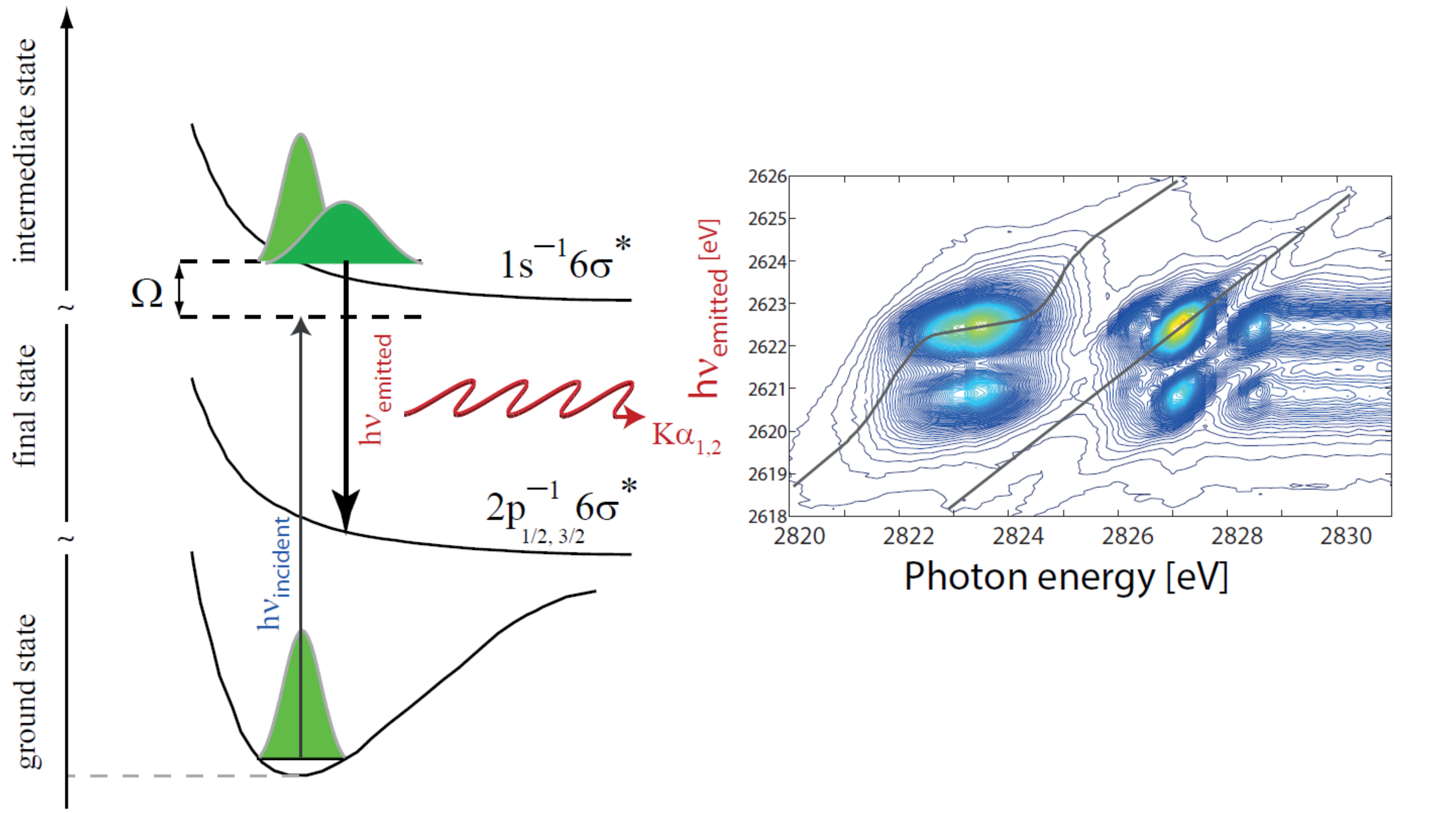
O1s - 4a$_1^*$
Resonant Auger spectra
H_2O
D_2O
Intensity (arb. units)
15
20
25
30
35
Binding Energy (eV)

- lifetime(~1 fs)
- Fluorescence yield
- Cl K edge HCl, CH_3Cl, CF_3Cl….
- "Core-hole clock experiment" using RIXS
- Nuclear dynamics



M. Simon, L. Journel, R. Guillemin, W. C. Stolte, I. Minkov, F. Gel'mukhanov, P. Salek, H. Ågren, S. Carniato, R. Taïeb, A. C. Hudson, and D. W. Lindle, **Phys. Rev. A** 73 (2006) 020706

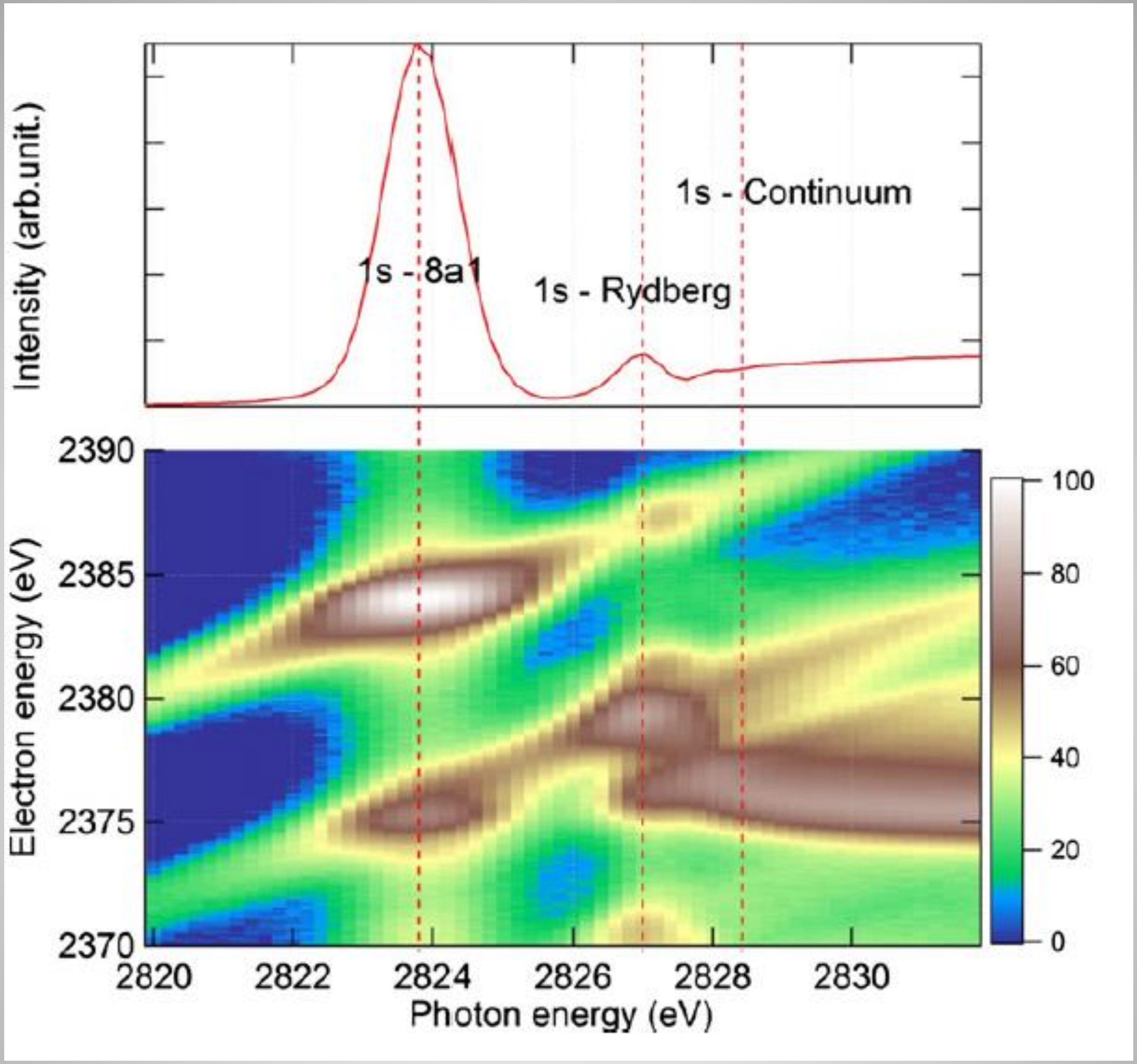

excited state 1s-16σ*
Ω
ground state
0,93 eV
Ω = -1 eV
Effective Scattering Time
τeff.= 1/√(Ω2+Γ2)
1,29 eV
Ω = 0 eV



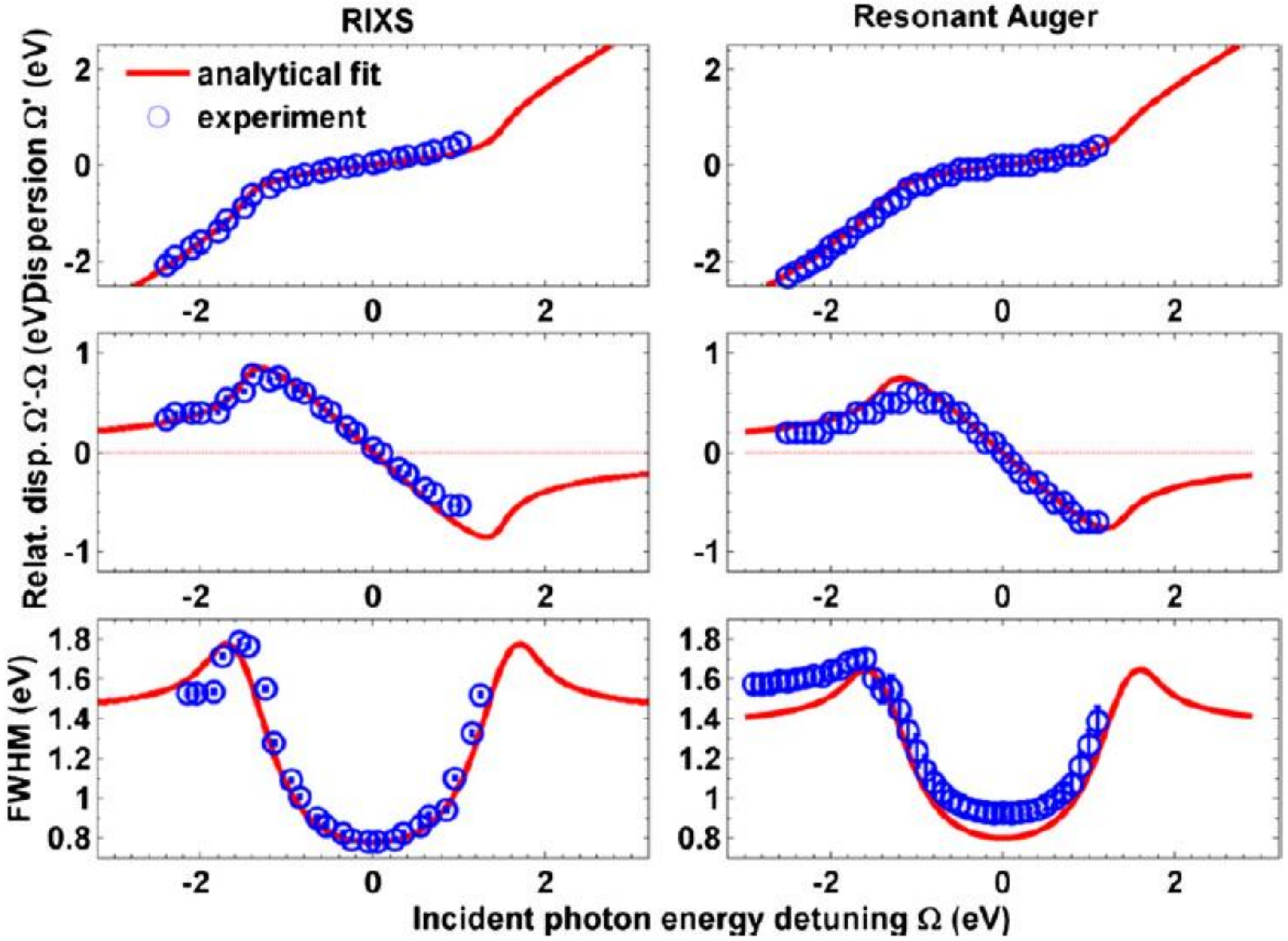
intermediate state
final state
ground state
Ω
$1s^{-1}6\sigma^*$
$h\nu_{emitted}$
$K\alpha_{1,2}$
$2p^{-1}_{1/2,\,3/2}\ 6\sigma^*$
$h\nu_{incident}$
$h\nu_{emitted}$ [eV]
2626
2625
2624
2623
2622
2621
2620
2619
2618
2820
2822
2824
2826
2828
2830
Photon energy [eV]

CH3Cl

Cl K-edge



M. N. Piancastelli, G. Goldsztejn, T. Marchenko, R. Guillemin, R. K. Kushawaha, L. Journel, S. Carniato, J.-P. Rueff, D. Céolin and M. Simon, J.Phys.B: At.Mol.Opt.Phys. 47, (2014) 124031

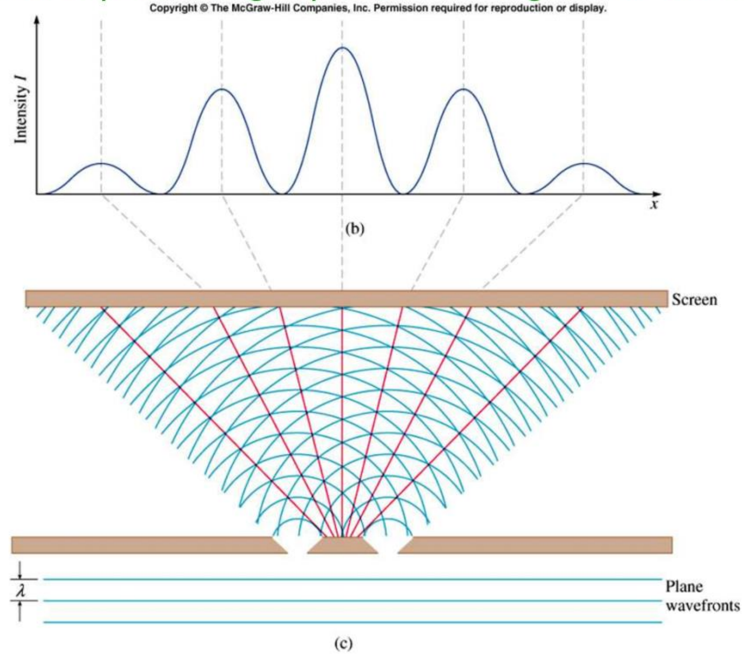


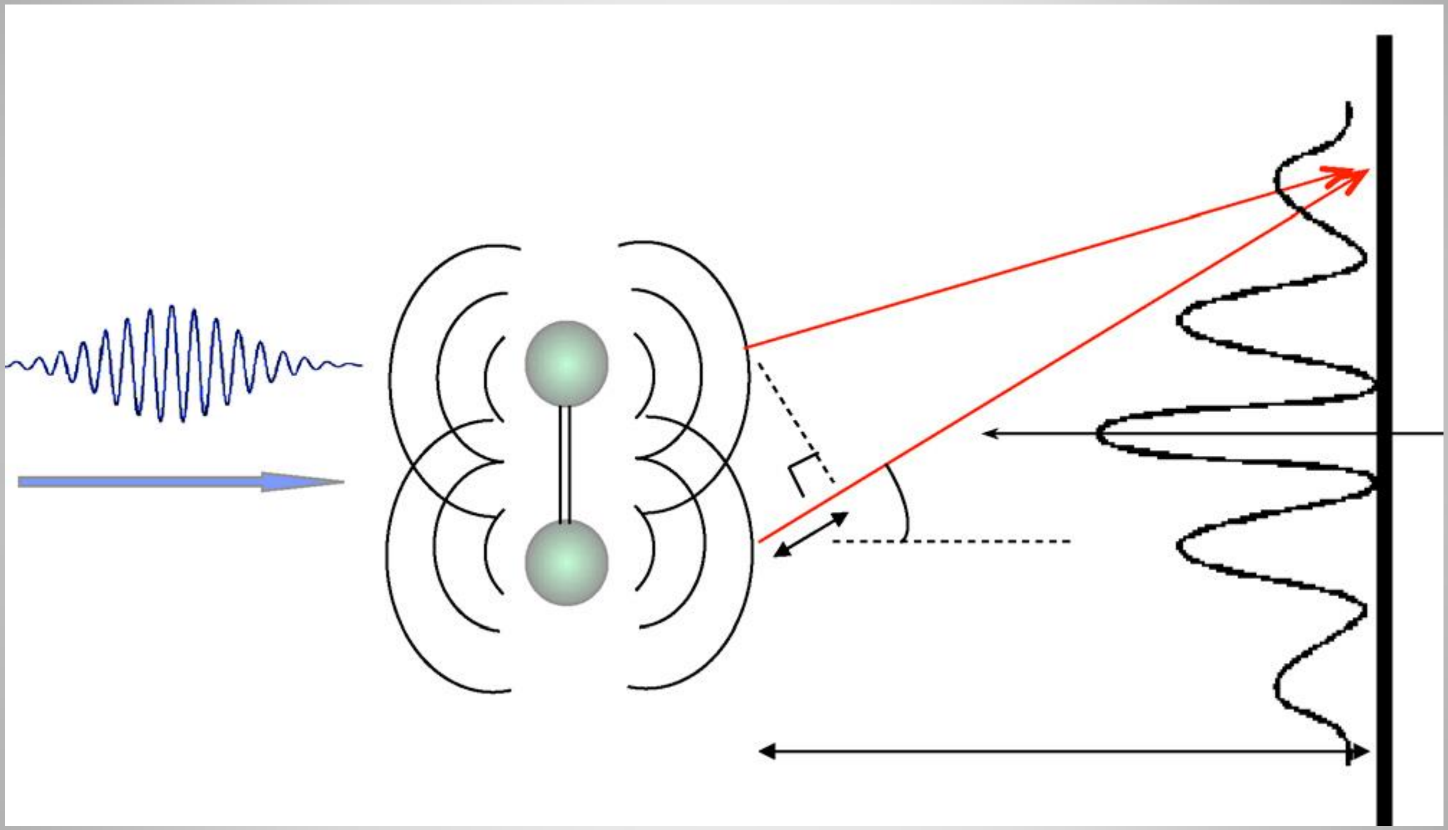
RIXS
Resonant Auger
analytical fit
experiment
Dispersion Ω' (eV)
Relat. disp. Ω'-Ω (eV)
FWHM (eV)
Incident photon energy detuning Ω (eV)

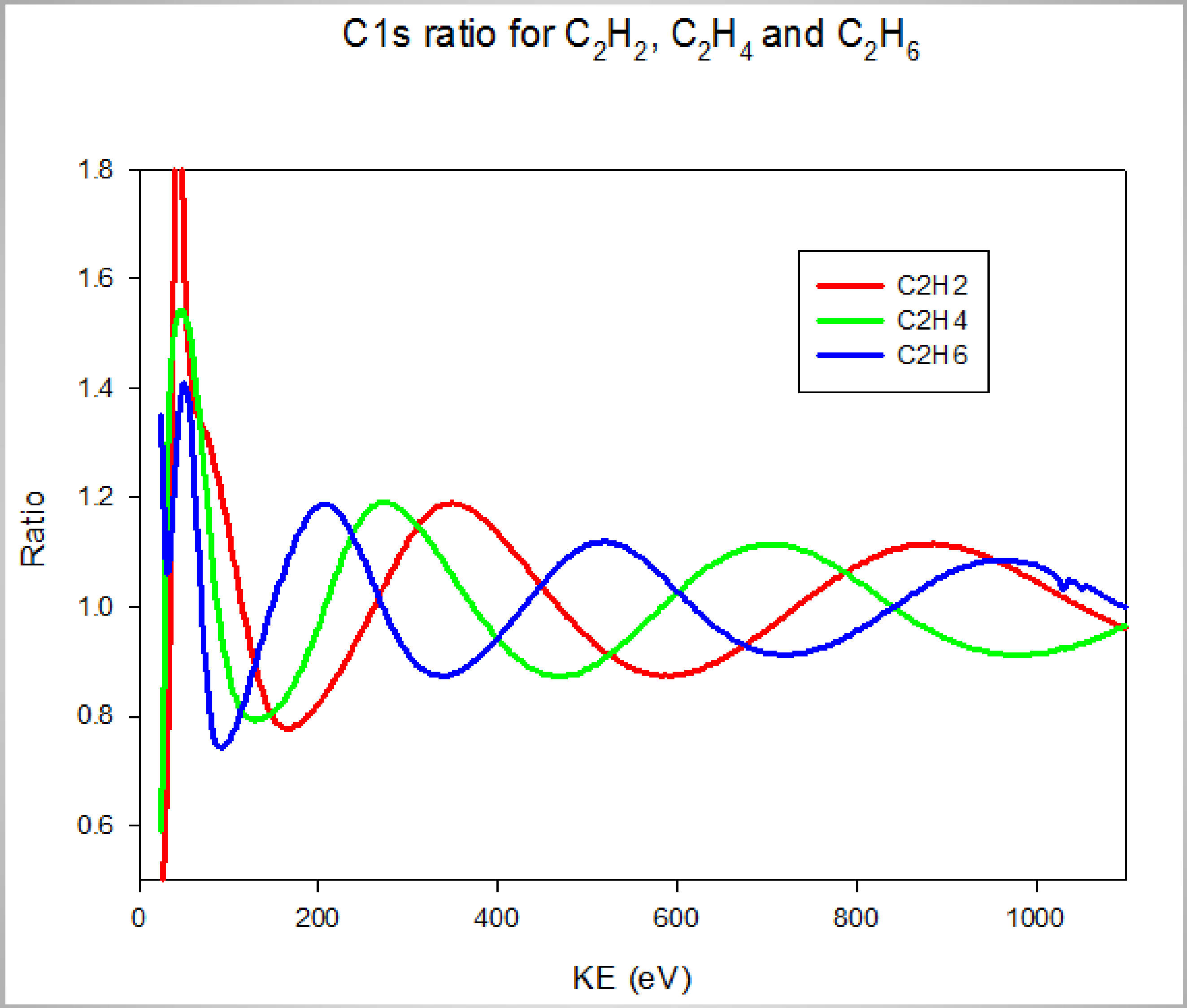
Young´s double slit interference in photoemission

Young's Double Slit Experiment

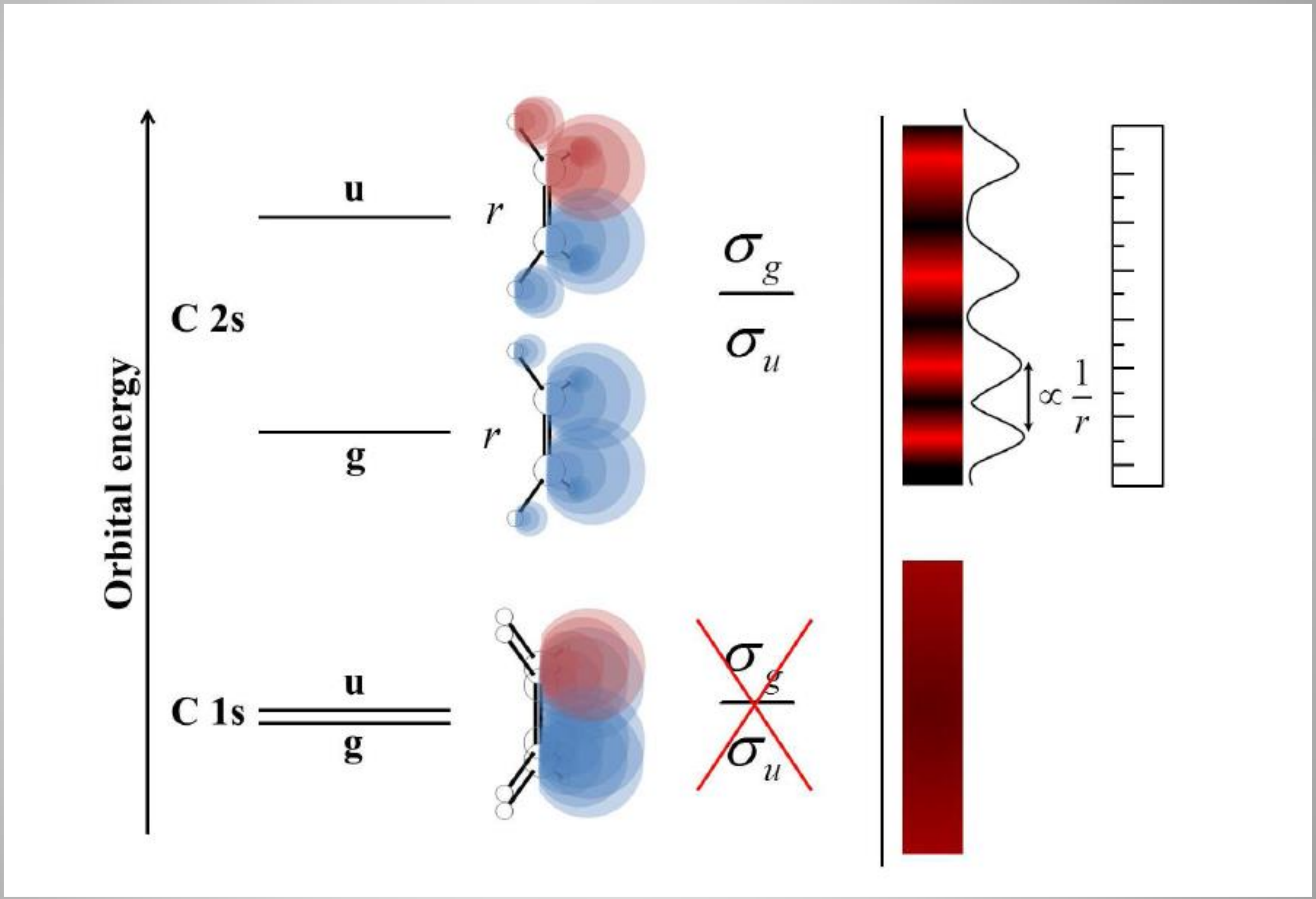
Light passing through two parallel slits
will interfere, producing a pattern of bright and dark fringes.



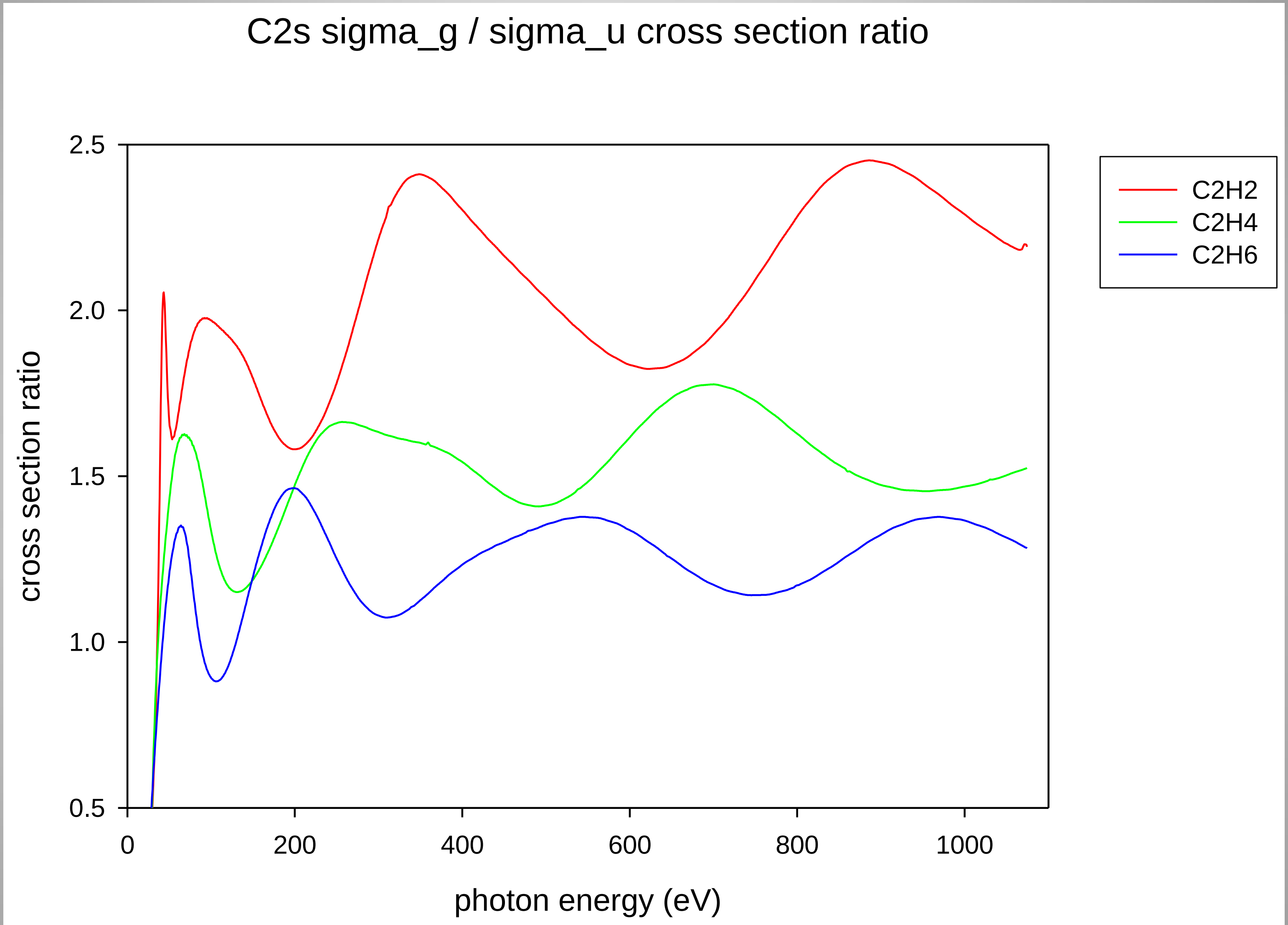


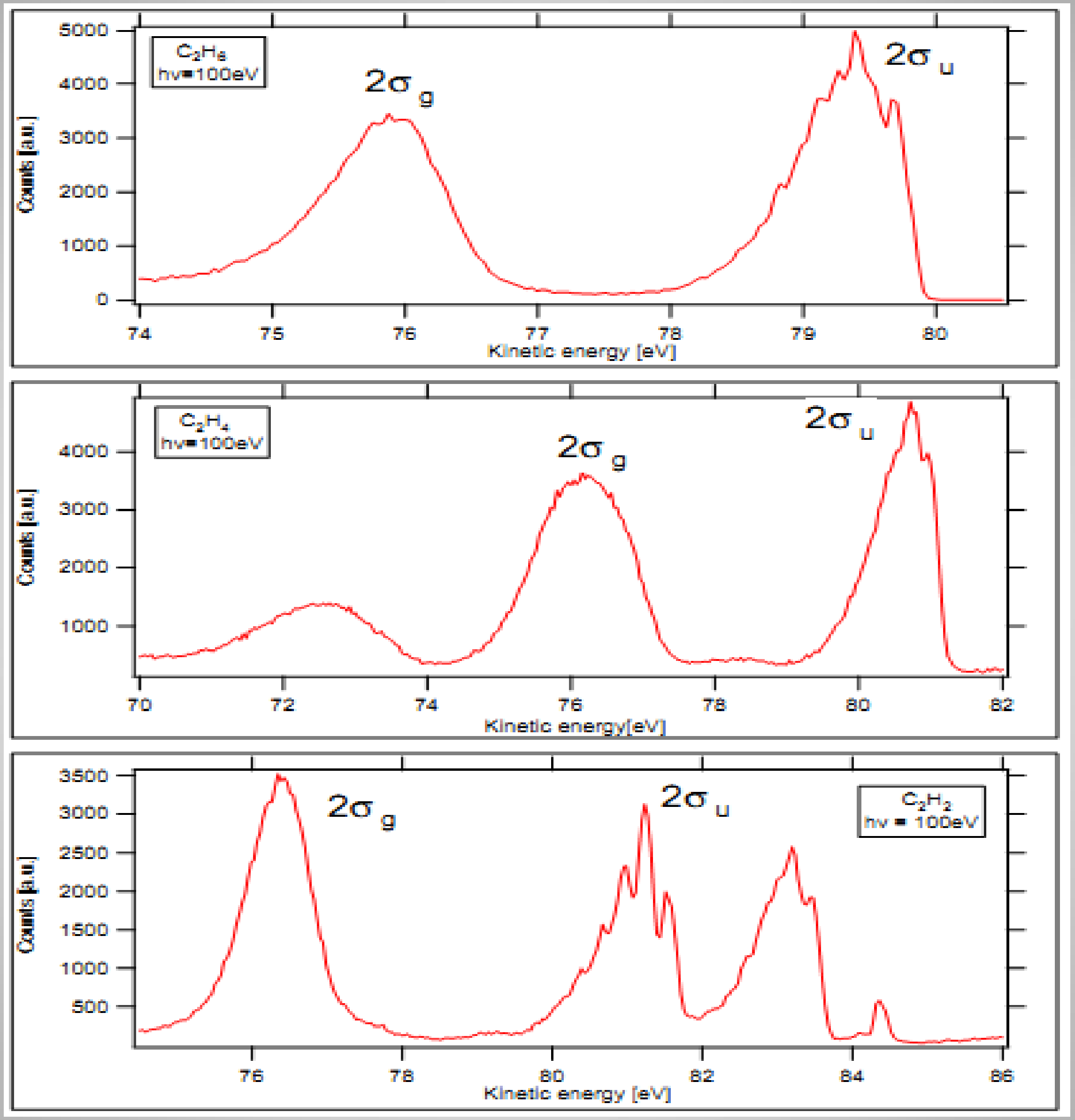


Calculations by P.Decleva

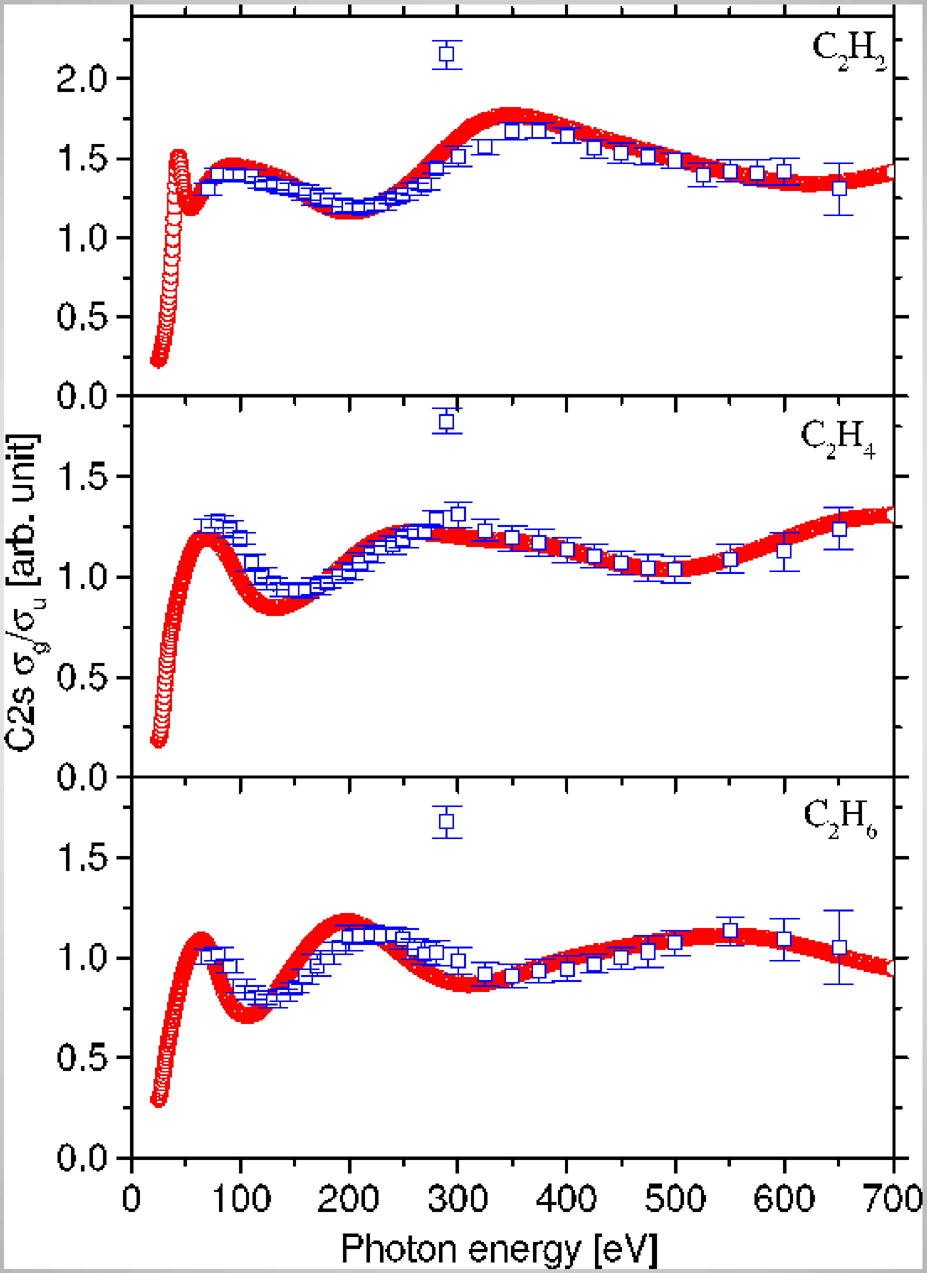


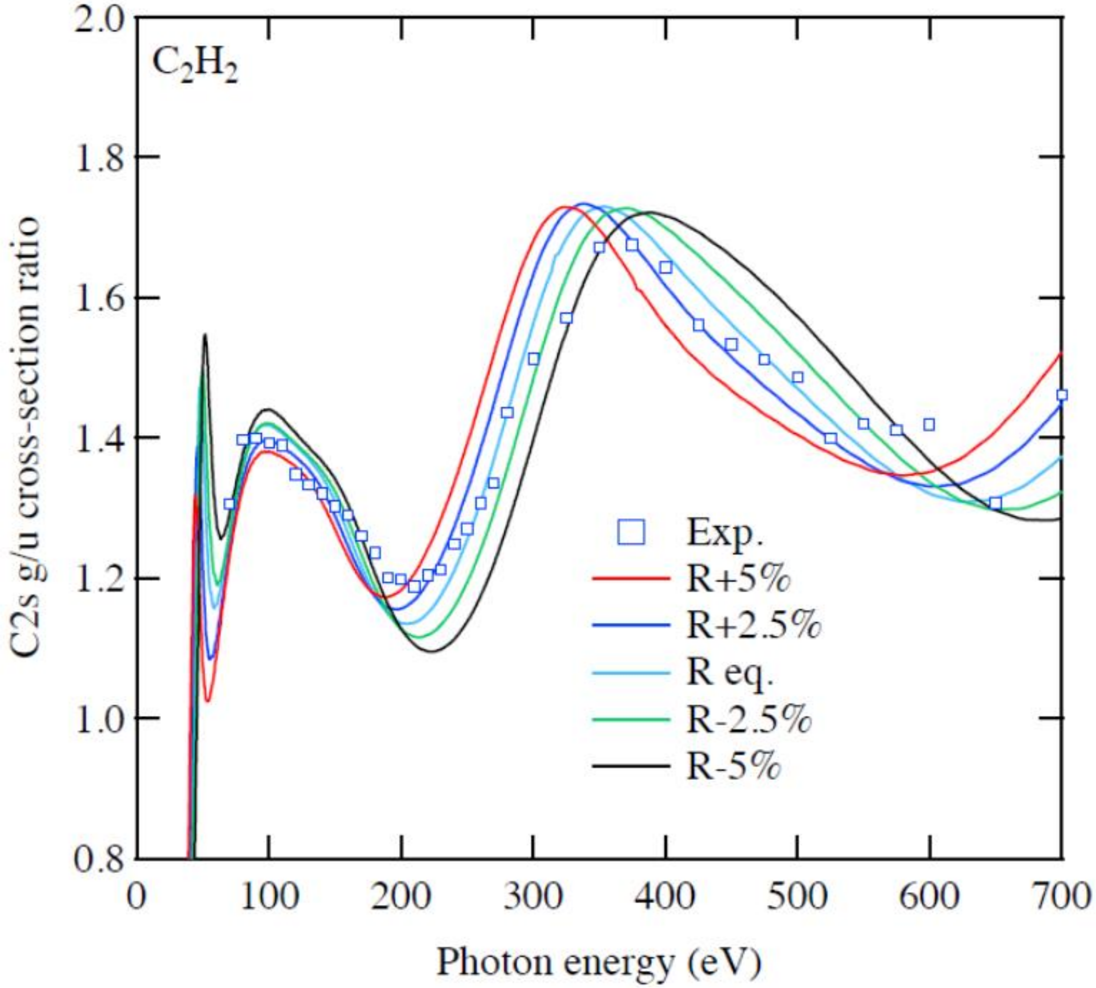
R.K. Kushawaha, M. Patanen, R. Guillemin, L. Journel, C.Miron, M. Simon, M.N.Piancastelli and P. Decleva, PNAS 110, (2013) 15201


C2s sigma_g / sigma_u cross section ratio
cross section ratio
2.5
2.0
1.5
1.0
0.5
0
200
400
600
800
1000
photon energy (eV)
C2H2
C2H4
C2H6



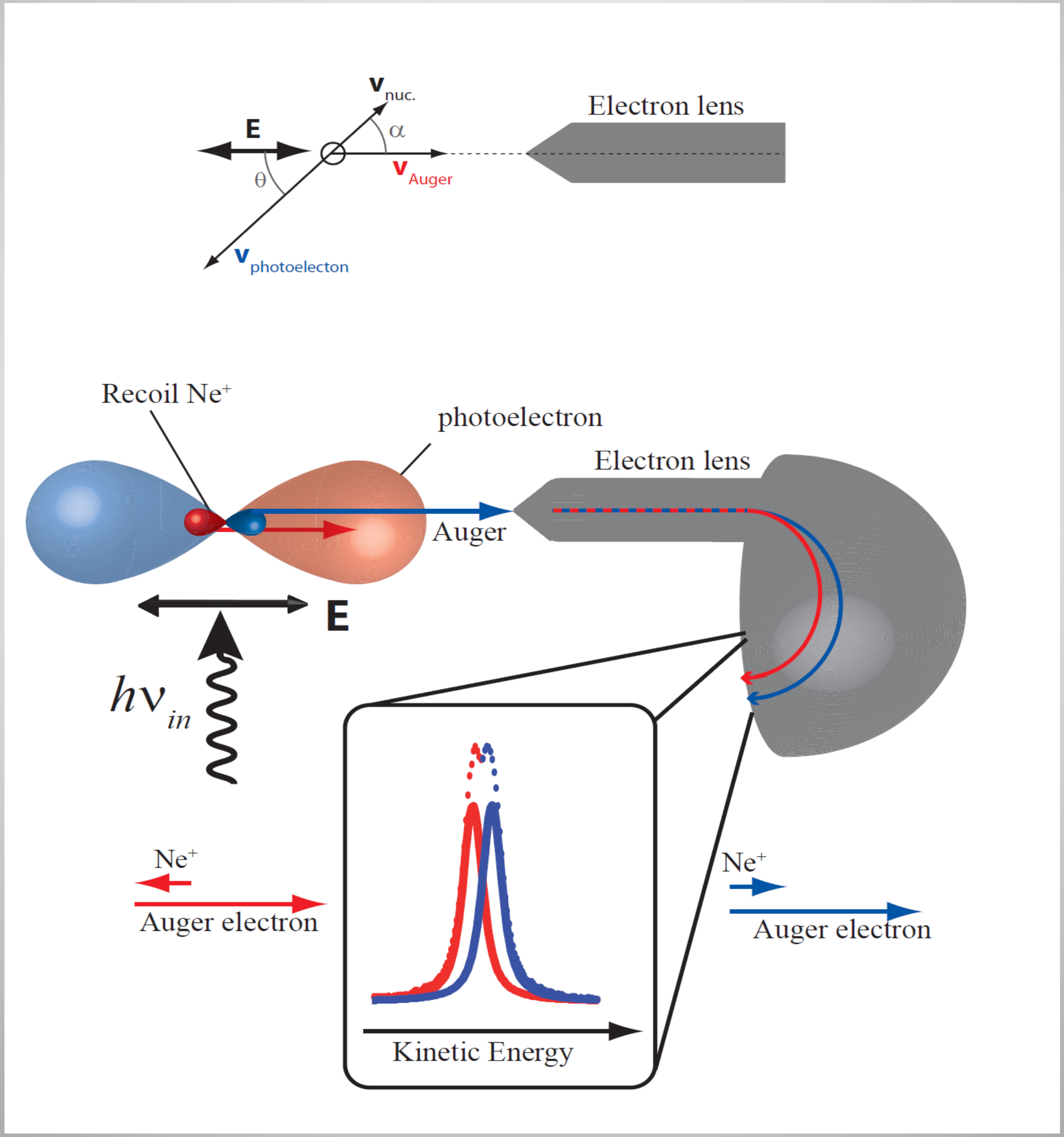
C2H6
hv=100eV
2σ g
2σ u
Counts [a.u.]
Kinetic energy [eV]
C2H4
hv=100eV
2σ g
2σ u
Counts [a.u.]
Kinetic energy[eV]
C2H2
hv = 100eV
2σ g
2σ u
Counts [a.u.]
Kinetic energy [eV]


C$_2$H$_2$
C$_2$H$_4$
C$_2$H$_6$
C2s σg/σu [arb. unit]
Photon energy [eV]
0.0
0.5
1.0
1.5
2.0
0
100
200
300
400
500
600
700



C2s g/u cross-section ratio
2.0
1.8
1.6
1.4
1.2
1.0
0.8
C_2H_2
Exp.
R+5%
R+2.5%
R eq.
R-2.5%
R-5%
0
100
200
300
400
500
600
700
Photon energy (eV)

Atomic Auger Doppler effects upon emission of fast photoelectrons



M.Simon, R.Püttner, T.Marchenko, R.Guillemin, R.K. Kushawaha, L.Journel, G.Goldsztejn, M.N.Piancastelli, J.M. Ablett, J.-P.Rueff and D.Céolin, Nature Comm. 5, 4069 (2014)