Intro to SR & FEL spectroscopy; interaction between radiation & matter (2)

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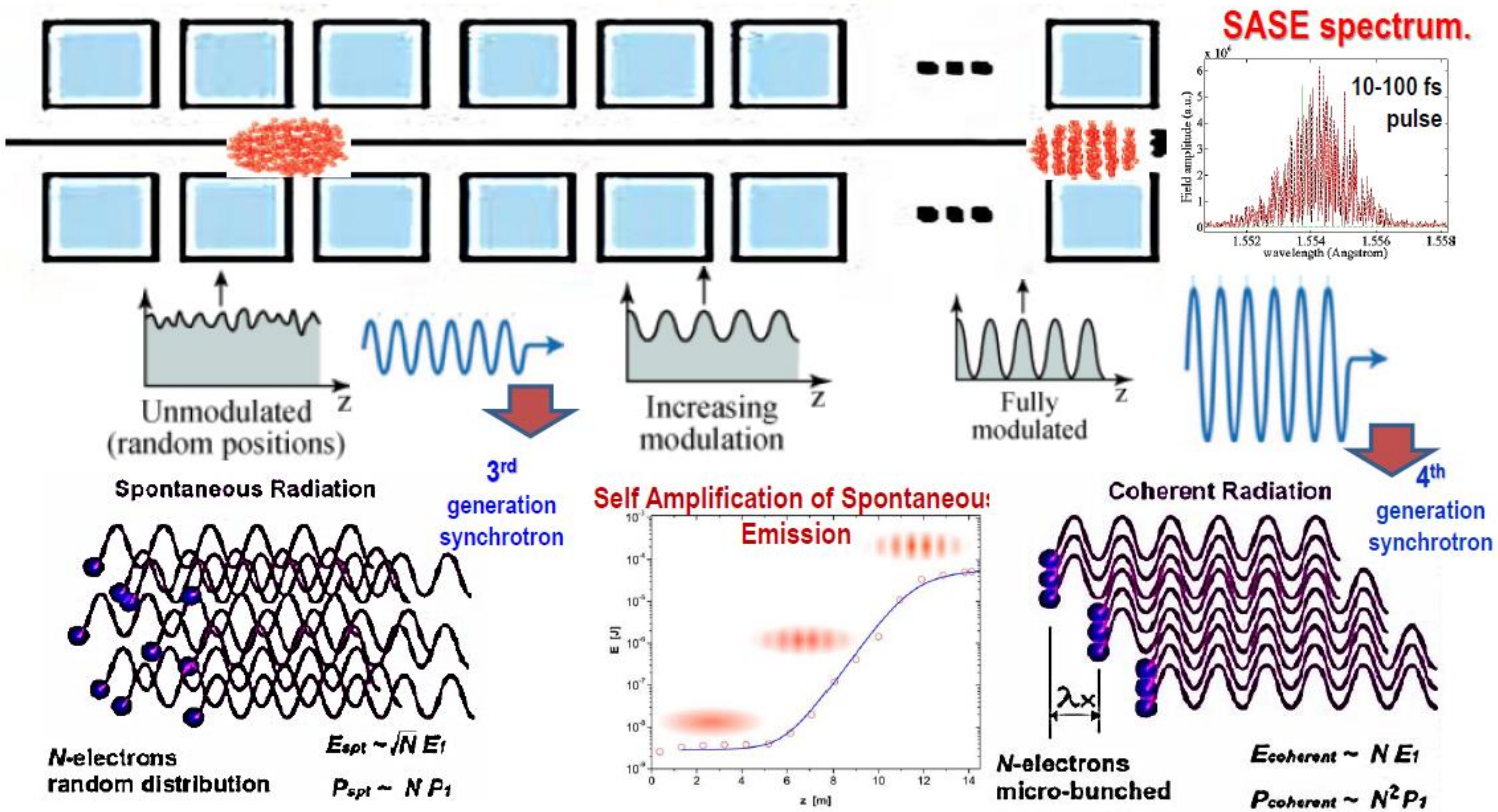
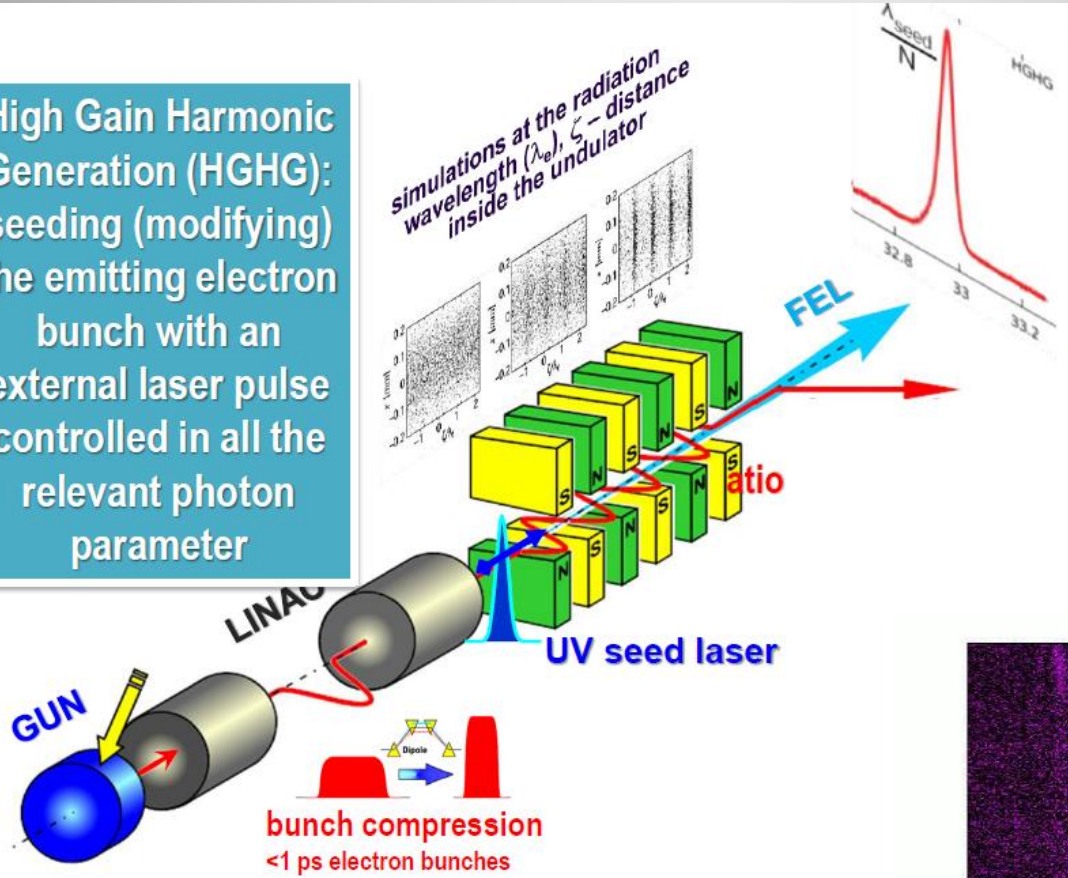
With a very long undulator the radiated fields become stronger and lead to microbunching, i.e. transform the random positions and motions of electrons into correlated waves of electrons, emitting radiation in phase.

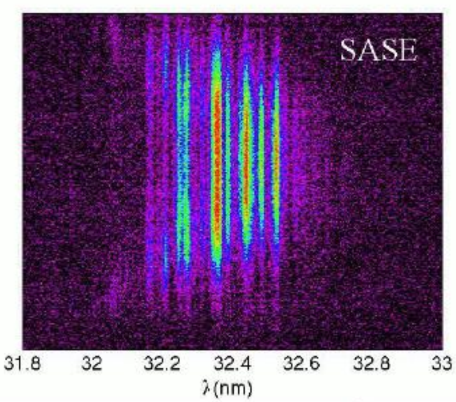


High Gain Harmonic Generation (HGHG): seeding (modifying) the emitting electron bunch with an external laser pulse controlled in all the relevant photon parameter



"SASE" FEL – several separate "waves" of electrons with uncorrelated phase. Less peak power, broader spectrum.

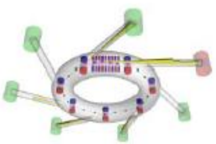
The properties of the FEL radiation are entangled with those of the seed laser. Defined energy-time profile.

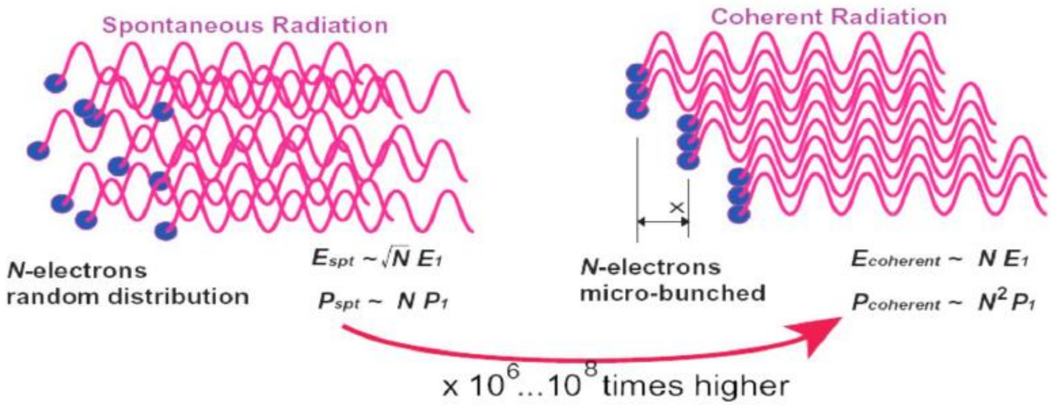


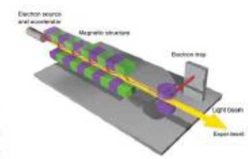
Synchrotron vs FEL Radiation

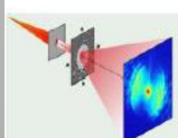
long pulses (sub-ns) max ~10^8 photons/pulse

Fs pulses > 10^{11} photons/pulse

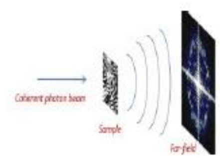






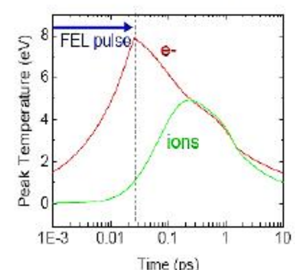


Synchrotron radiation:
pinhole and monochromators for spatial and spectral filtering, but at the expense of intensity!



FEL (FLASH, LSLS, SACLA, FERMI):
natural space coherence: each electron - spontaneous emission that overlap each other in phase

Ultra-short (fs) and ultra-bright coherent FEL pulses allow imaging with single pulse before the radiation damage manifests itself !



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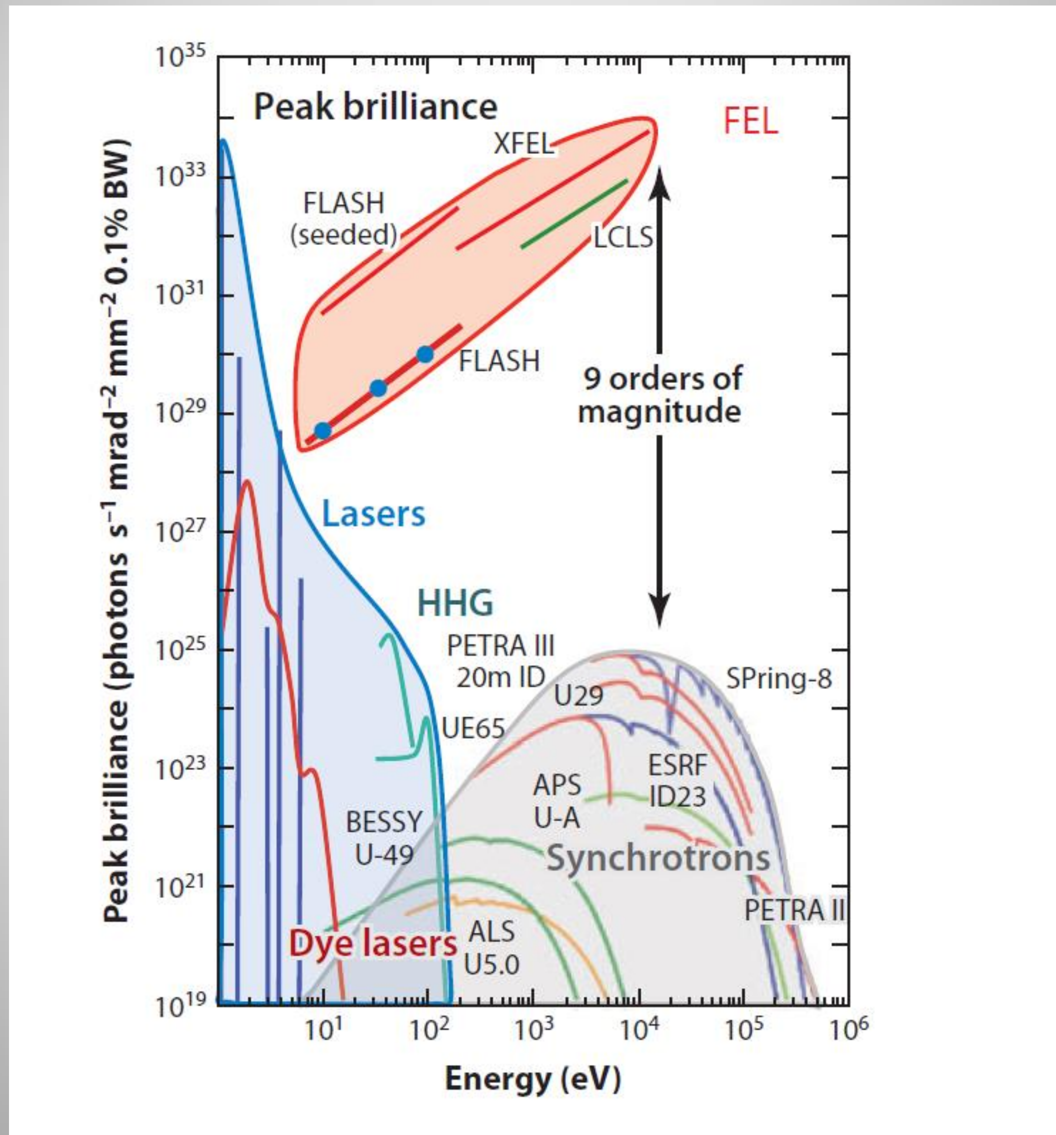


	LCLS	LCLS II	Eu-XFEL	SACLA	FLASH	FLASH II	FERMI	SwissFEL	PAL XFEL	Shanghai XFEL
Shortest wavelength	1.5 Å	1 Å	0.5 Å	1 Å	40 Å	40 Å	40 Å	1 Å	1 (0.6) Å	1 Å
Undulator type hard X-ray.	Fixed gap	Variable gap	Variable gap	In-vacuum Var. gap	n.a.	n.a.	n.a.	In-vacuum var. gap	Variable gap	Variable gap
Undulator type soft X-ray.	n.a.	Variable gap	Variable gap	n.a.	Fixed gap	Variable gap	Apple II	Apple II	Apple II	?
Injector	S-band RF gun	S-band RF gun	L-band RF gun	Pulsed Diode	L-band RF gun	L-band RF gun	S-band RF gun	S-band RF gun	S-band RF gun	S-band RF gun
Cathode	Cu	Cu	Cs_2Te	CeB_6 (thermionic)	Cs_2Te	Cs_2Te	Cu	Cu	Cu	Cu
Main linac technology	n.c. Pulsed	n.c. pulsed	s.c. pulsed	n.c. pulsed	s.c. pulsed	s.c. pulsed	n.c. pulsed	n.c. pulsed	n.c. pulsed	n.c. pulsed
RF frequency	S-band	S-band	L-band	C-band	L-band	L-band	S-band	C-band	S-band	C-band
RF Rep. rate	120 Hz	120 Hz	10 Hz	60 Hz	10 Hz	10 Hz	10-50 Hz	100 Hz	120 Hz	60 Hz
FEL pulses/RF pulse	1	1	2700	1	2700	2700	1	2	1	1
max. bunch charge	0.25 nC	0.25 nC	1 nC	0.2 nC	1 nC	1 nC	0.5 nC	0.2 nC	0.2 nC	0.2 nC
max. electron energy	13.6 GeV	14 GeV	17.5 GeV	8 GeV	1.2 GeV	1.2 GeV	1.5 GeV	5.8 GeV	10 GeV	6.4 GeV
No. RF stations	81	81	29	69	5	5	15	34	49	?
Approx. facility length	1.7km	1.7km	3.4 km	0.8km	0.32 km	0.32 km	0.5 km	0.7km	1.1 km	0.6 km
Start operation	2009	2017	2015	2011	2005	2013	2010	2016	2015	2019

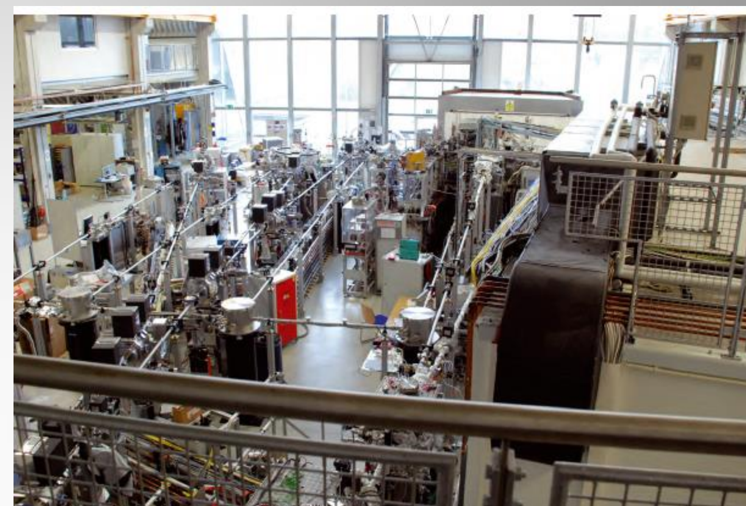
The Physics of Free Electron Lasers
By *Evgeny Saldin, Evgeny A. Schneidmiller, M.V. Yurkov*
Springer, 2000

Classical Theory of Free-Electron Lasers
By *Eric B. Szarmes*
Morgan & Claypool Publishers, 2014

Insertion Devices for Synchrotron Radiation and Free Electron Laser
By *F. Ciocci , G. Dattoli, A. Torre, A. Renieri*
World Scientific, 2000

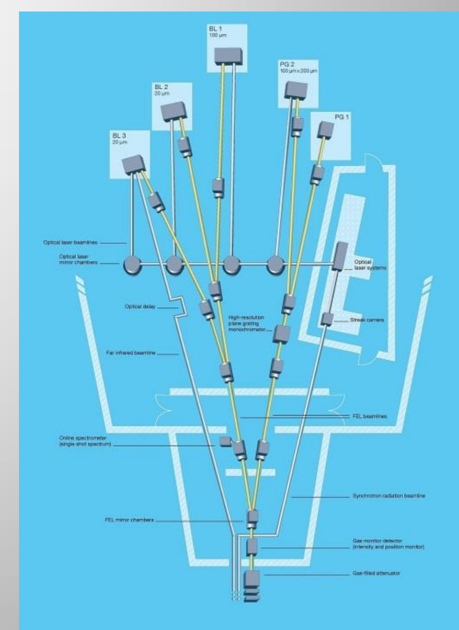
FLASH

DESY, Hamburg, Germany

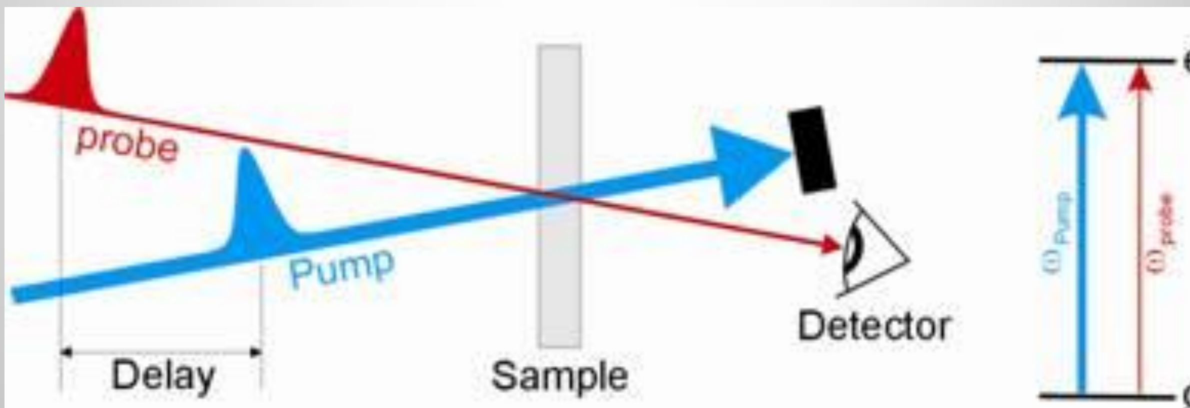


FLASH Parameters

XUV - Photons		
Number of Bunches per second+	1 - 8000 (delivered up to 5000 to users)	
Pulse Repetition Rates (within pulse train)+	40, 50, 100, 200, 250, 500, 1000	kHz
Wavelength	4.2 - 45	nm
Photon Energy	28 - 295	eV
Pulse Duration (FWHM)	30 - 300	fs
Average Pulse Energy (single bunch)*	1 - 500	µJ
Average Pulse Energy (pulse trains)*	1 - 200	µJ



Pump-probe experiments



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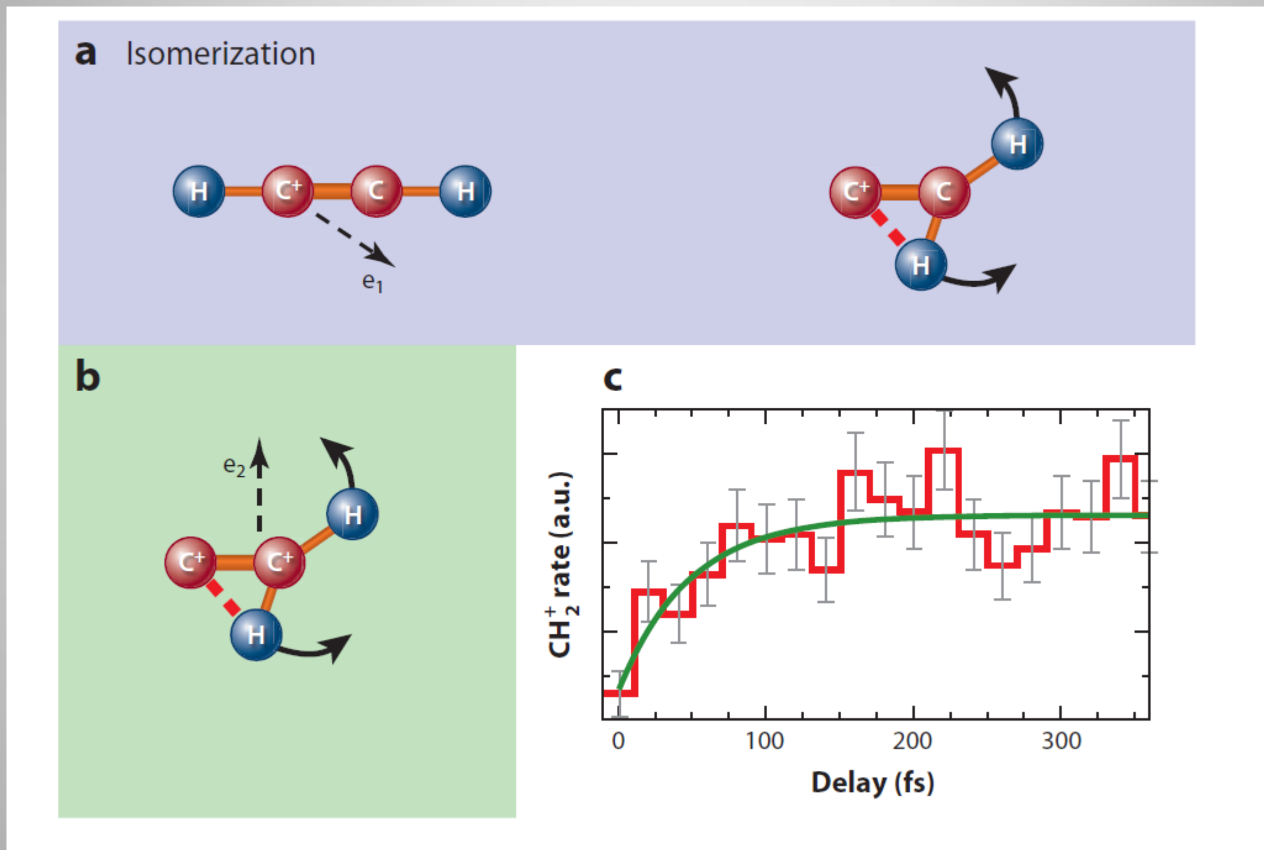

Y. H. Jiang et al., PRL 105, 263002 (2010)

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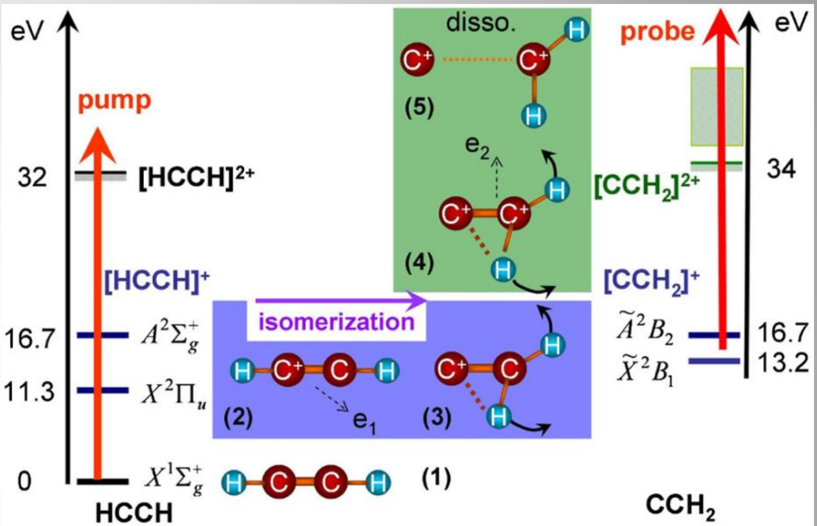
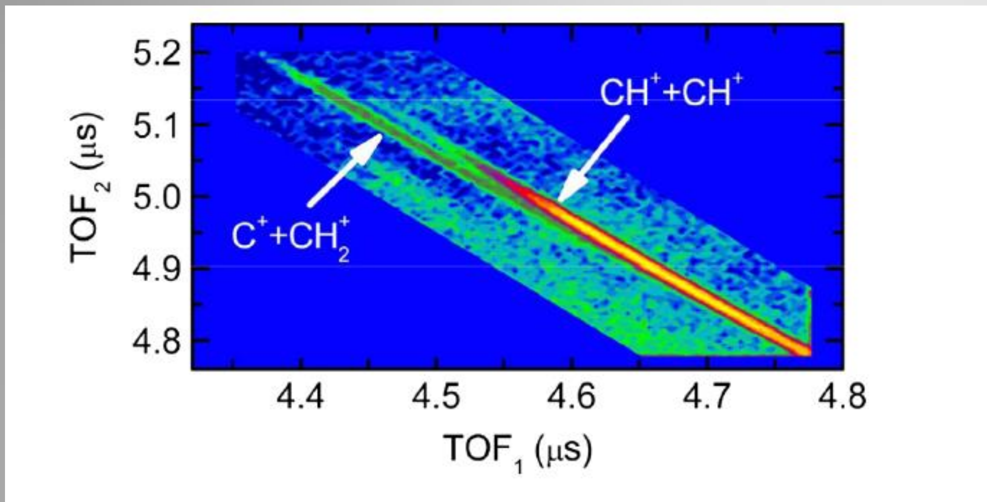
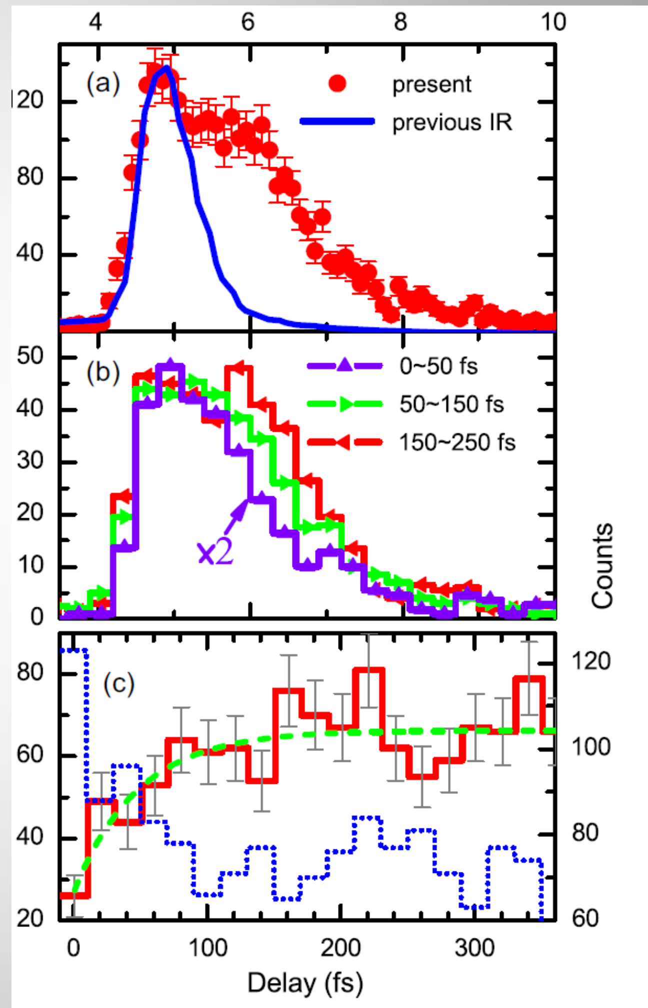


$$\mathrm{HCCH}(X^1\Sigma_g^+) + h\upsilon(\mathrm{pump})$$
$$\rightarrow [\mathrm{HCCH}]^+(X^2\Pi_u, A^2\Sigma_g) + e_1$$
$$\xrightarrow{\mathrm{migra}} [\mathrm{H_2CC}]^+(\tilde{X}^2B_1, \tilde{A}^2B_2) + e_1, \qquad (1)$$

and

$$[\mathrm{H_2CC}]^+(\tilde{X}^2B_1, \tilde{A}^2B_2) + h\upsilon(\mathrm{probe}) \rightarrow [\mathrm{H_2CC}]^{2+} + e_2$$
$$\xrightarrow{\mathrm{diss}} \mathrm{CH_2^+} + \mathrm{C^+} + e_2, \qquad (2)$$





Mean isomerization time:

52 ± 15 fs

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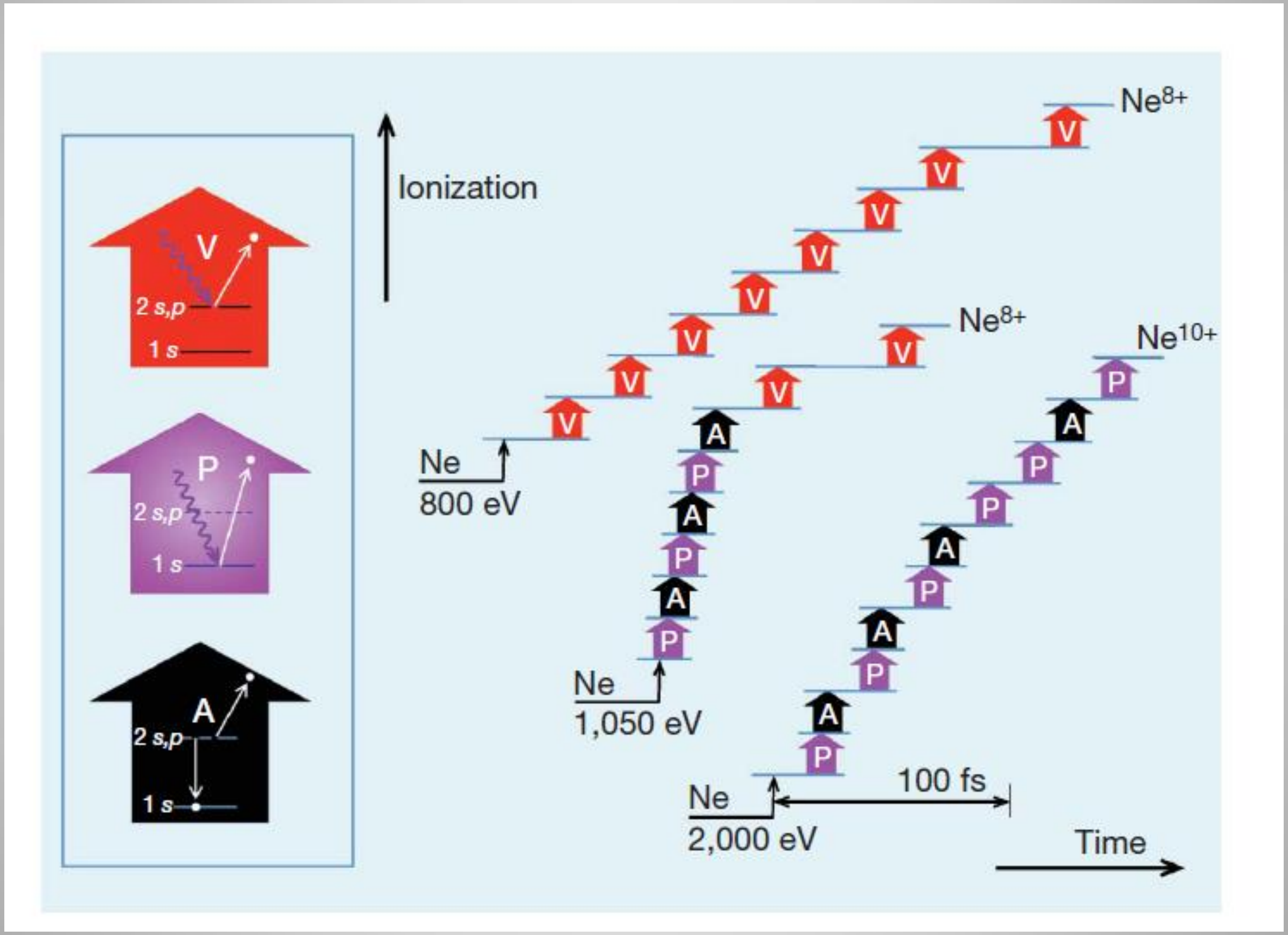
Photon Beam Parameters	Symbol	hard x-rays	soft x-rays	short pulse soft	short pulse hard	unit
Fundamental wavelength	λ_r	6.2-1.3	43.7-6.2	43.7-6.2	6.2-1.3	Å
Photon energy	$\hbar\omega$	2000-9600	285-2000	285-2000	2000-9600	eV
Final linac e^- energy	γmc^2	6.7-14.7	2.5-6.7	2.5-6.7	6.7-14.7	GeV
FEL 3-D gain length	L_G	3.3	1.5	~1.5	~3.3	m
Photons per pulse	N_γ	2	20	0.5	0.2	10^{12}
Peak brightness	B_{pk}	20	0.3	?	?	10^{32} §
Average brightness (120 Hz)	$\langle B \rangle$	160	8	?	?	10^{20} §
SASE bandwidth (fwhm)	$\Delta\omega/\omega$	~0.2-0.5	~0.2-1.0	?	?	%
Final pulse duration (fwhm)	$\Delta\tau_f$	50-250	70-400	<10	<10	fs

LCLS

SLAC, Stanford, CA, USA



L.Young et al., Nature 466, 56 (2010)

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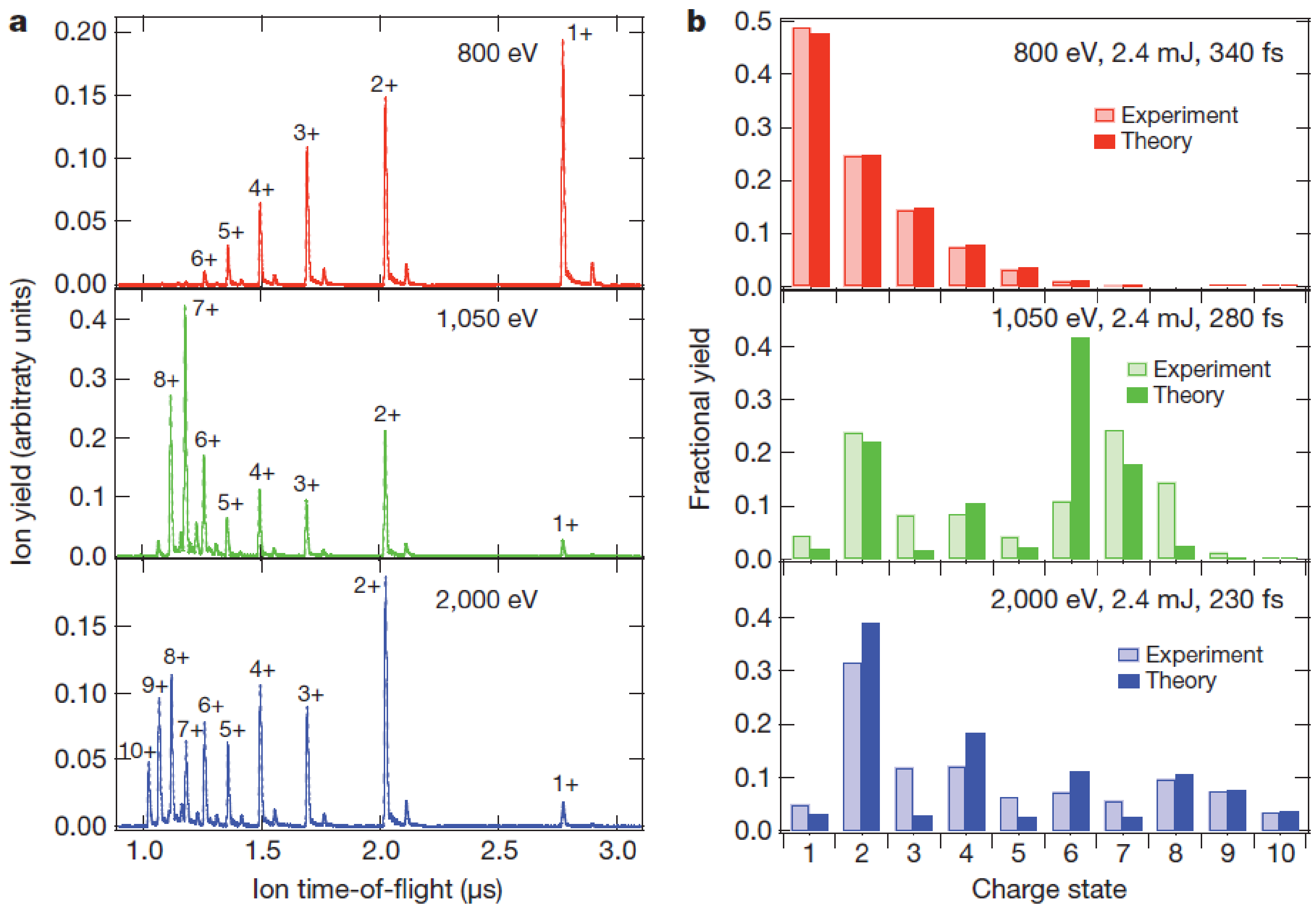

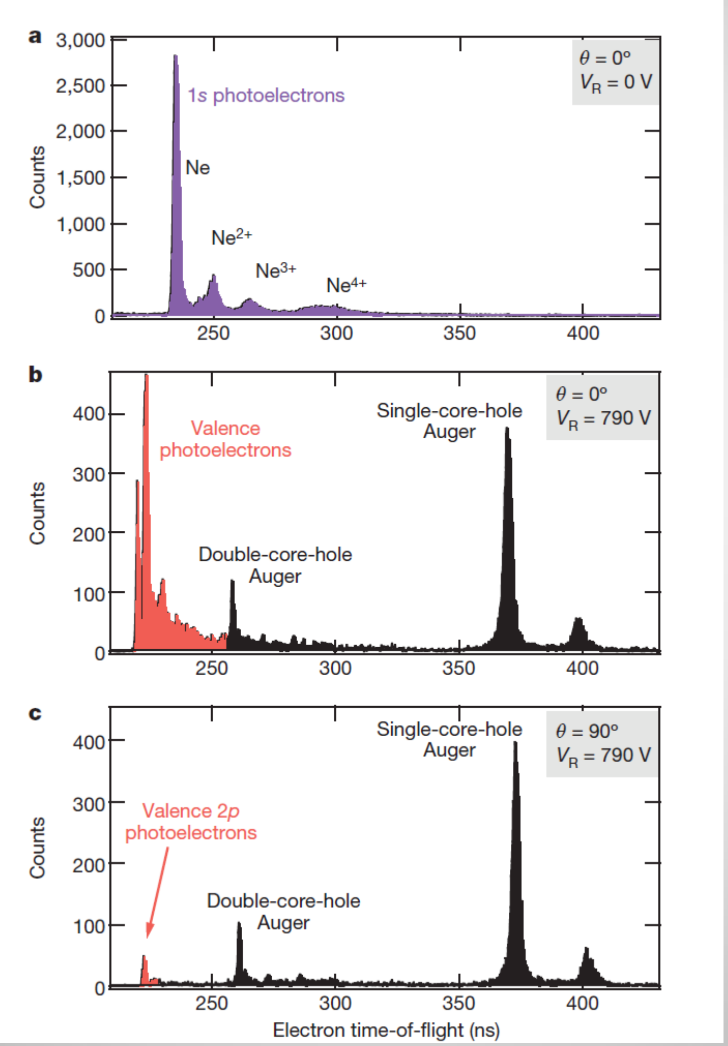

a
3,000
2,500
2,000
1,500
1,000
500
0
Counts
1s photoelectrons
Ne
Ne2+
Ne3+
Ne4+
θ = 0°
VR = 0 V
250
300
350
400
b
400
300
200
100
0
Counts
Valence
photoelectrons
Double-core-hole
Auger
Single-core-hole
Auger
θ = 0°
VR = 790 V
250
300
350
400
c
400
300
200
100
0
Counts
Valence 2p
photoelectrons
Double-core-hole
Auger
Single-core-hole
Auger
θ = 90°
VR = 790 V
250
300
350
400
Electron time-of-flight (ns)

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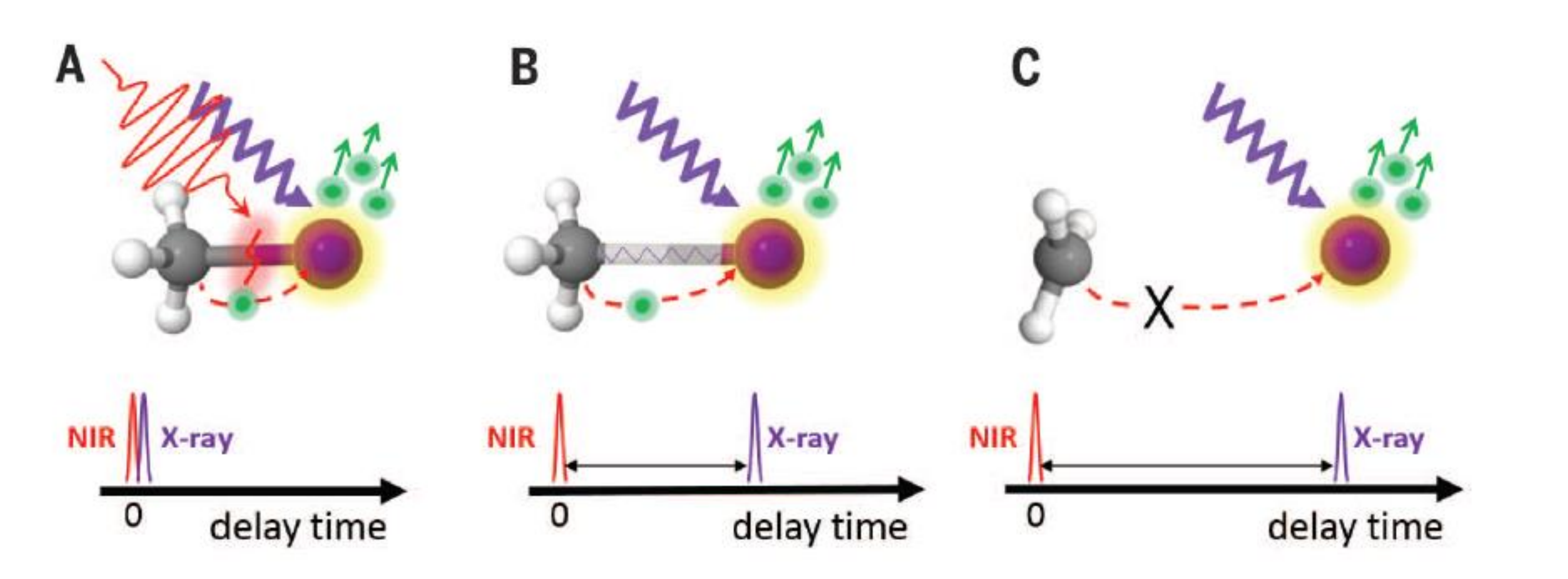

B.Erk et al., Science 345, 288 (2014)

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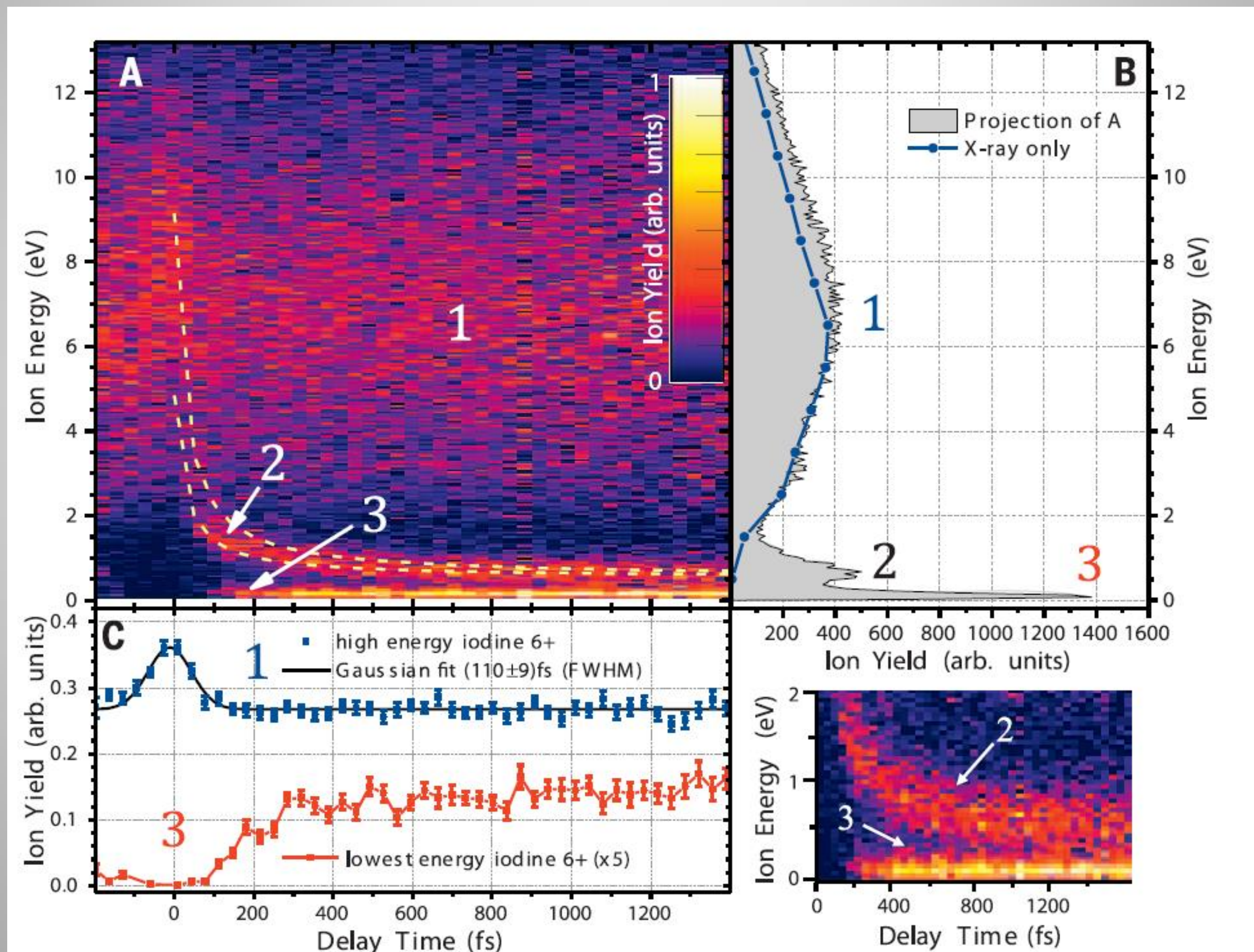

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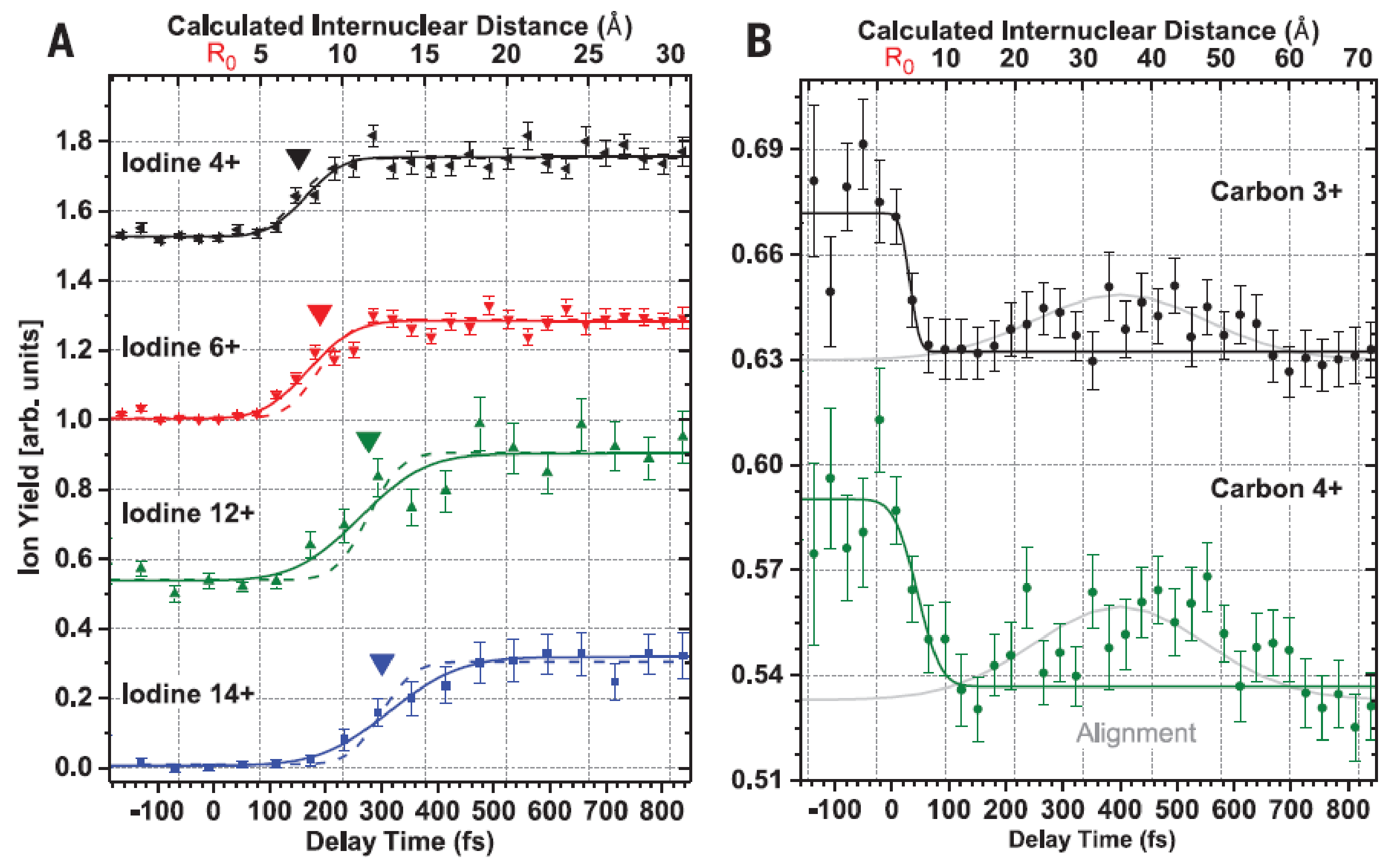
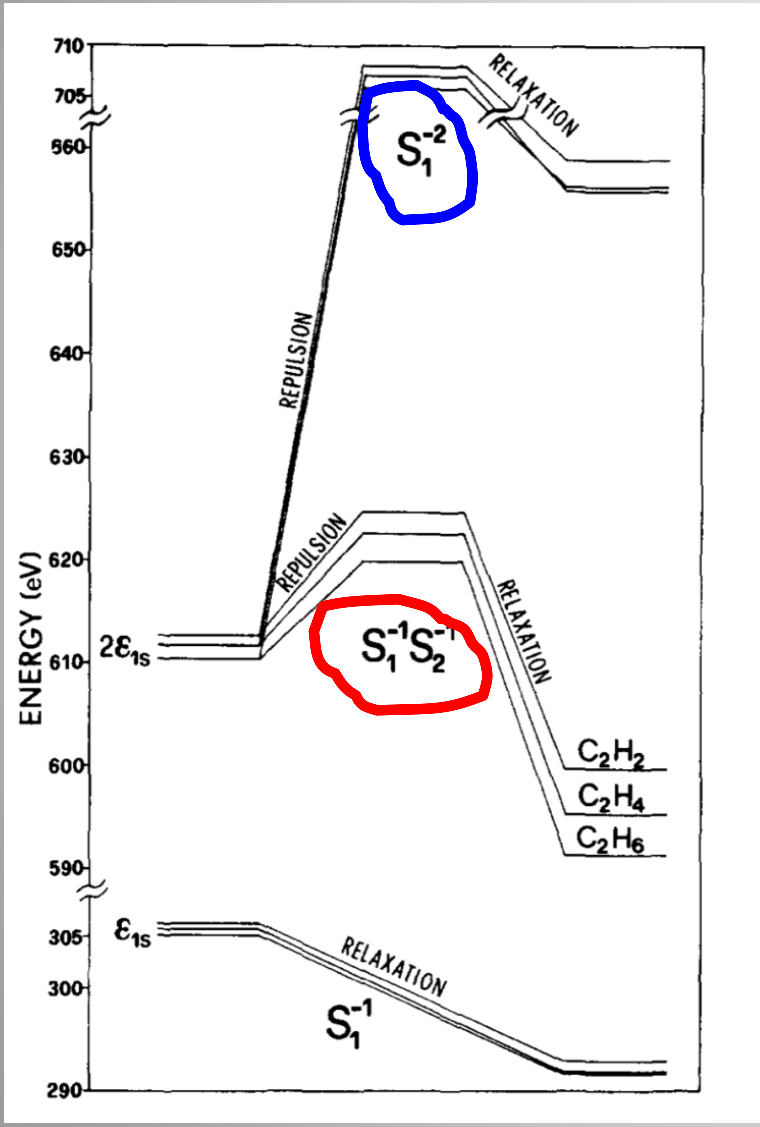




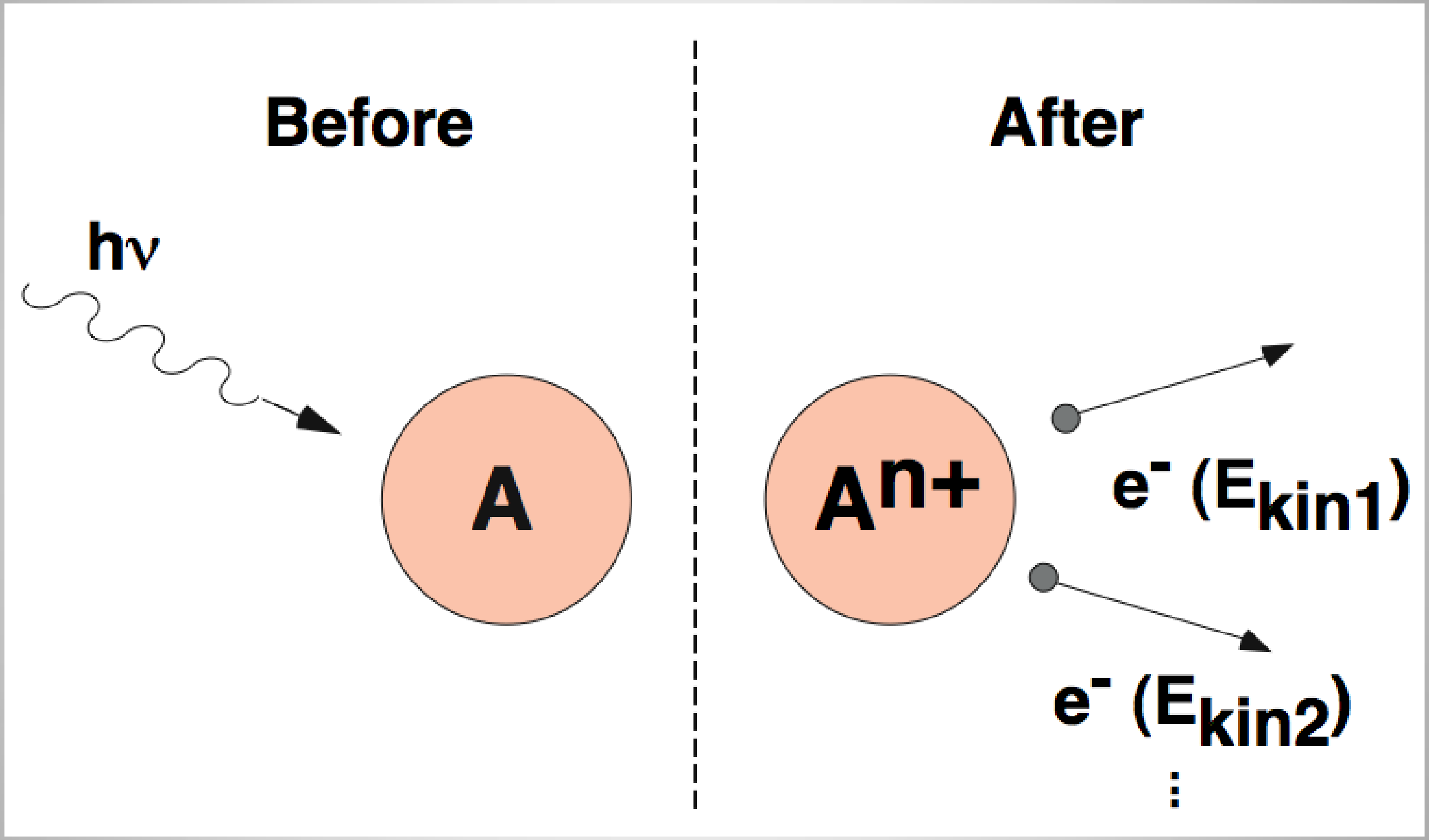
DCH Single-Site (SS)

DCH Two-Site (TS)

Substantially enhanced chemical sensitivity

L.S.Cederbaum et al, J.Chem.Phys. 85, (1986) 6513

Single Photon – Multiple Ionisation

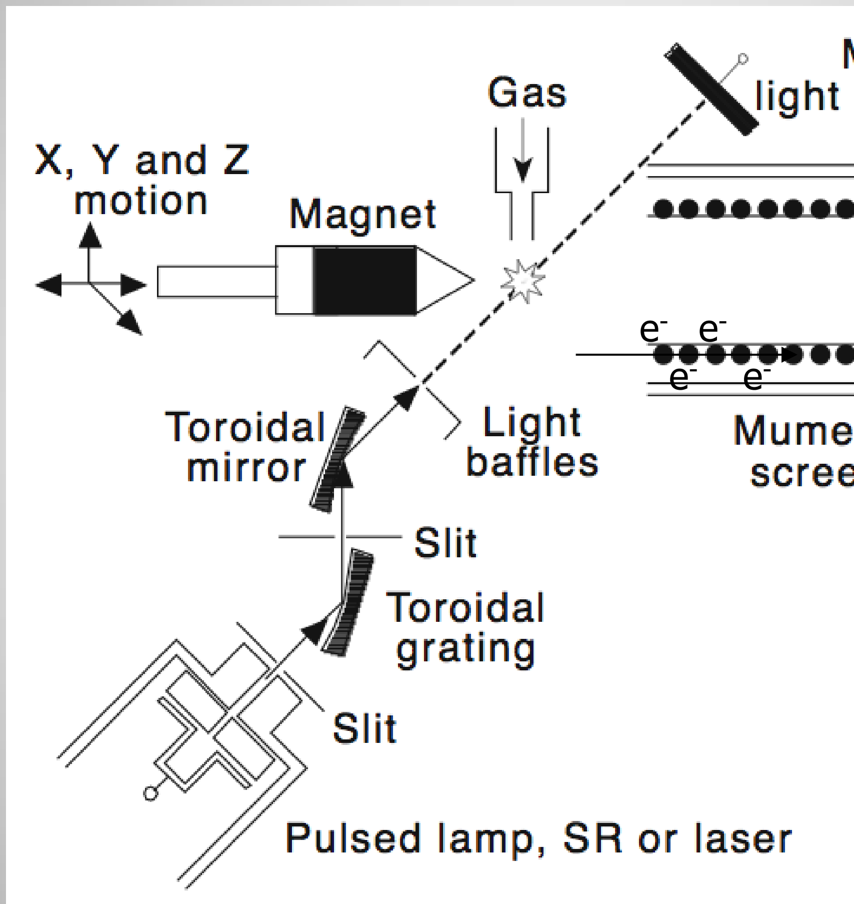


$$E_{B1} + E_{B2} \ (+ \ldots) = h\nu - E_{kin1} - E_{kin2} \ (- \ldots)$$

Double (multiple) ionisation energy (DIP, …)

Time-Of-Flight Magnetic Bottle Multi-Electron Spectrometer

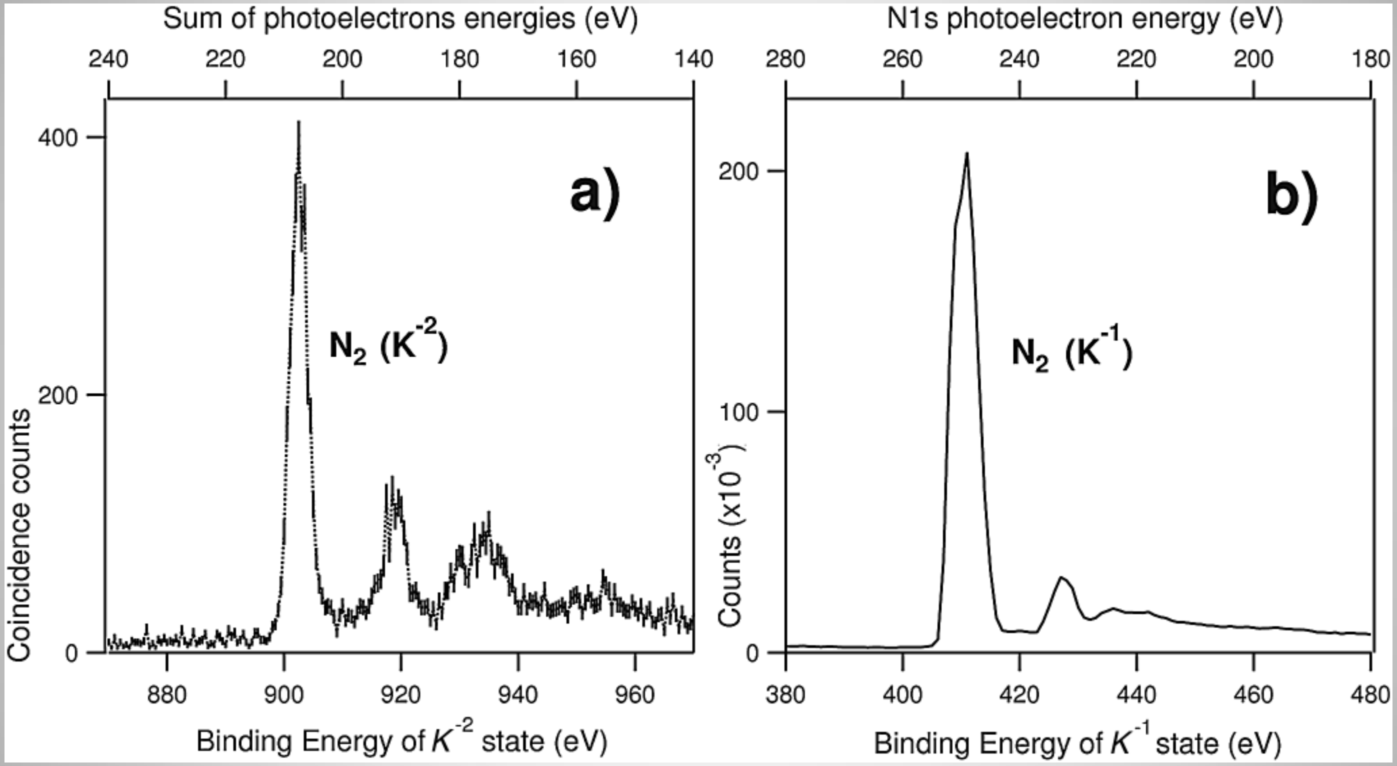
"4π" correlation device



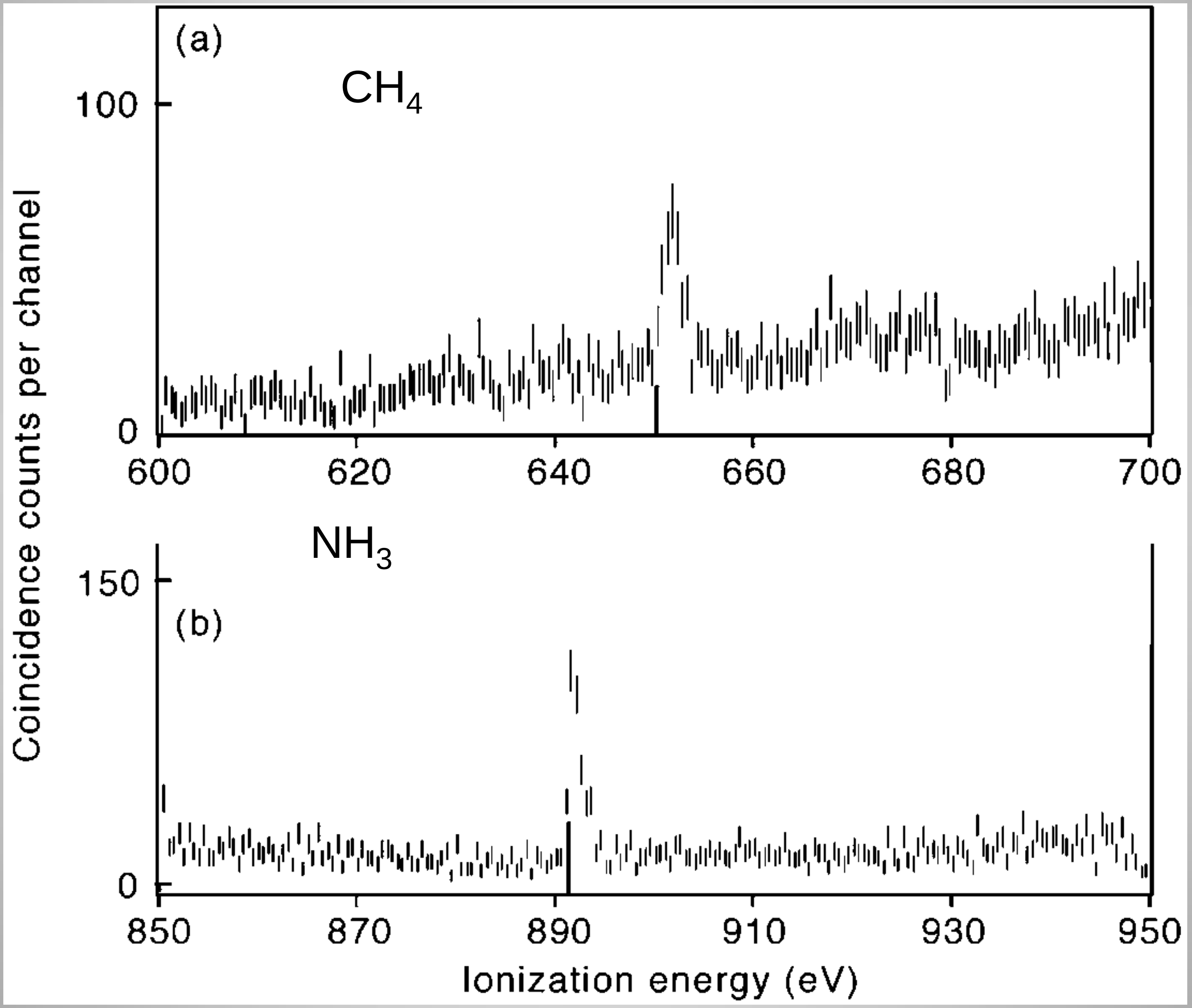
$$t = \frac{D}{\sqrt{E_{Kin}}}$$

Pulsed light source: He-lamp, SR, fs-laser system, FEL

J.H.D. Eland *et al.*, Phys. Rev. Lett. **90**, 053003 (2003)

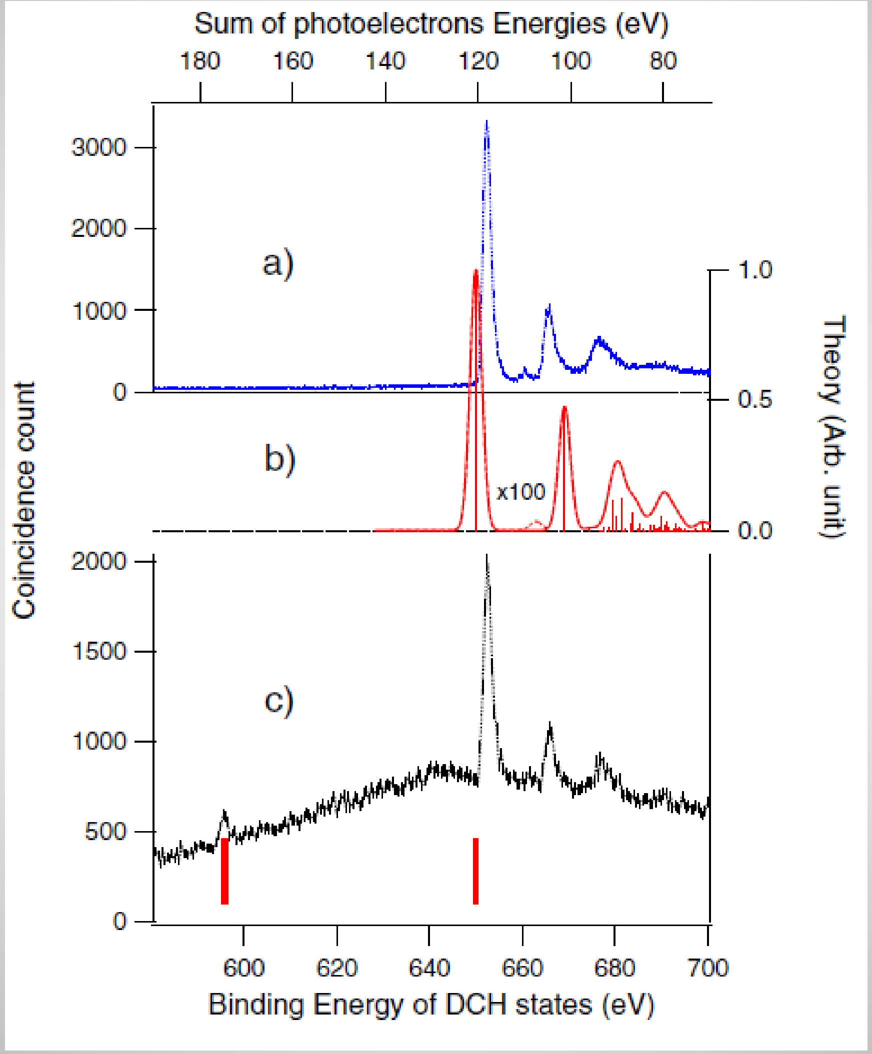


P.Lablanquie, F.Penent,J.Palaudoux, L.Andric, P.Selles, S.Carniato, K.Buĉar, M.Žitnik, M. Huttula, J.H.D.Eland, E.Shigemasa, K.Soejima, Y.Hikosaka, I.H.Suzuki, M.Nakano and K.Ito, PRL 106, 063003 (2011)



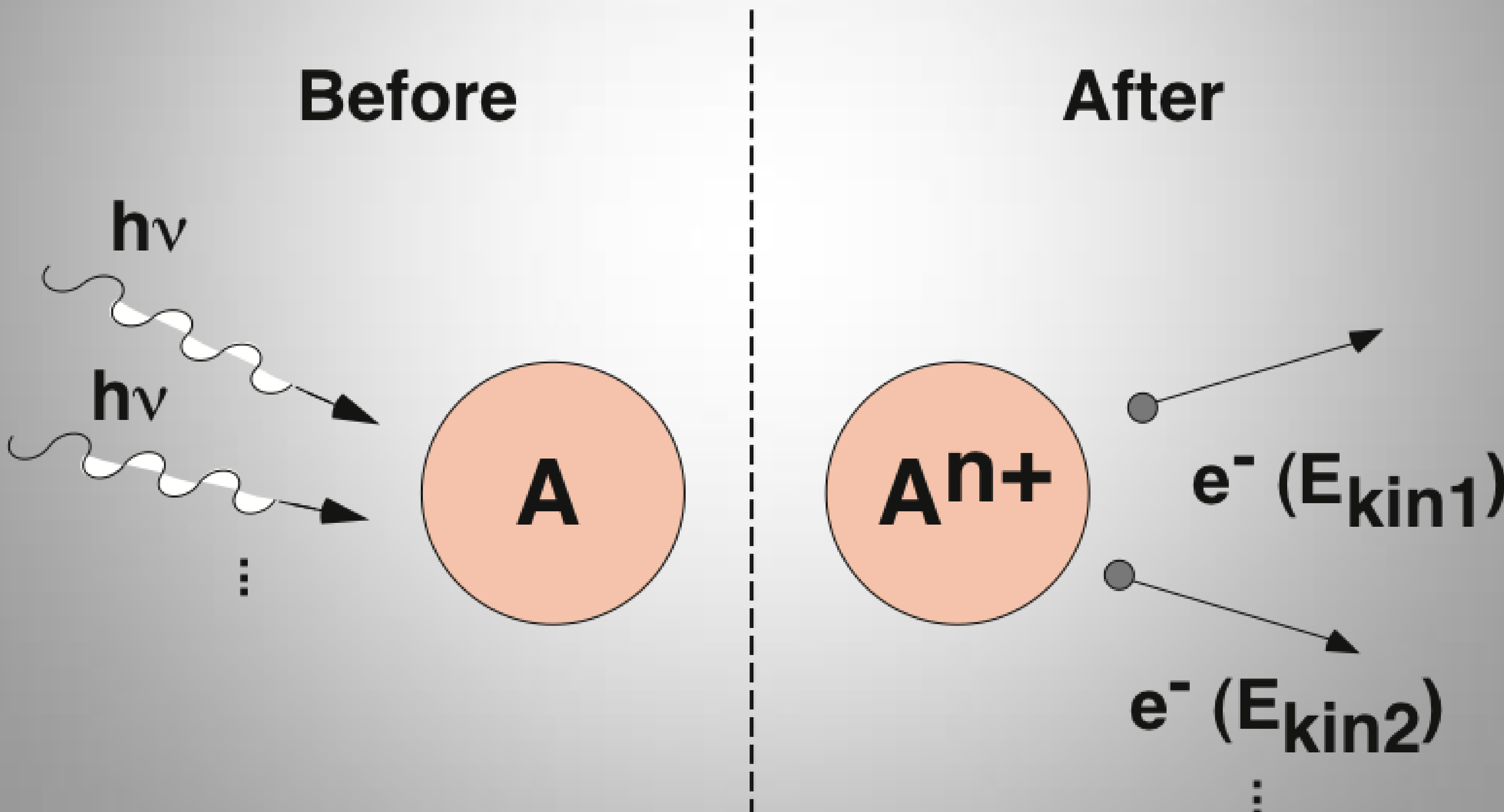
J.H.D.Eland, M.Tashiro, P.Linusson, M.Ehara, K.Ueda and R. Feifel, PRL 105, 213005 (2010)

C_2H_2



P. Lablanquie,T. P. Grozdanov, M. Žitnik, S. Carniato, P. Selles, L. Andric, J. Palaudoux, F. Penent, H. Iwayama, E. Shigemasa, Y. Hikosaka, K. Soejima, M. Nakano, I. H. Suzuki and K. Ito, PRL 107, 193004 (011)

Few Photon – Multiple Ionisation



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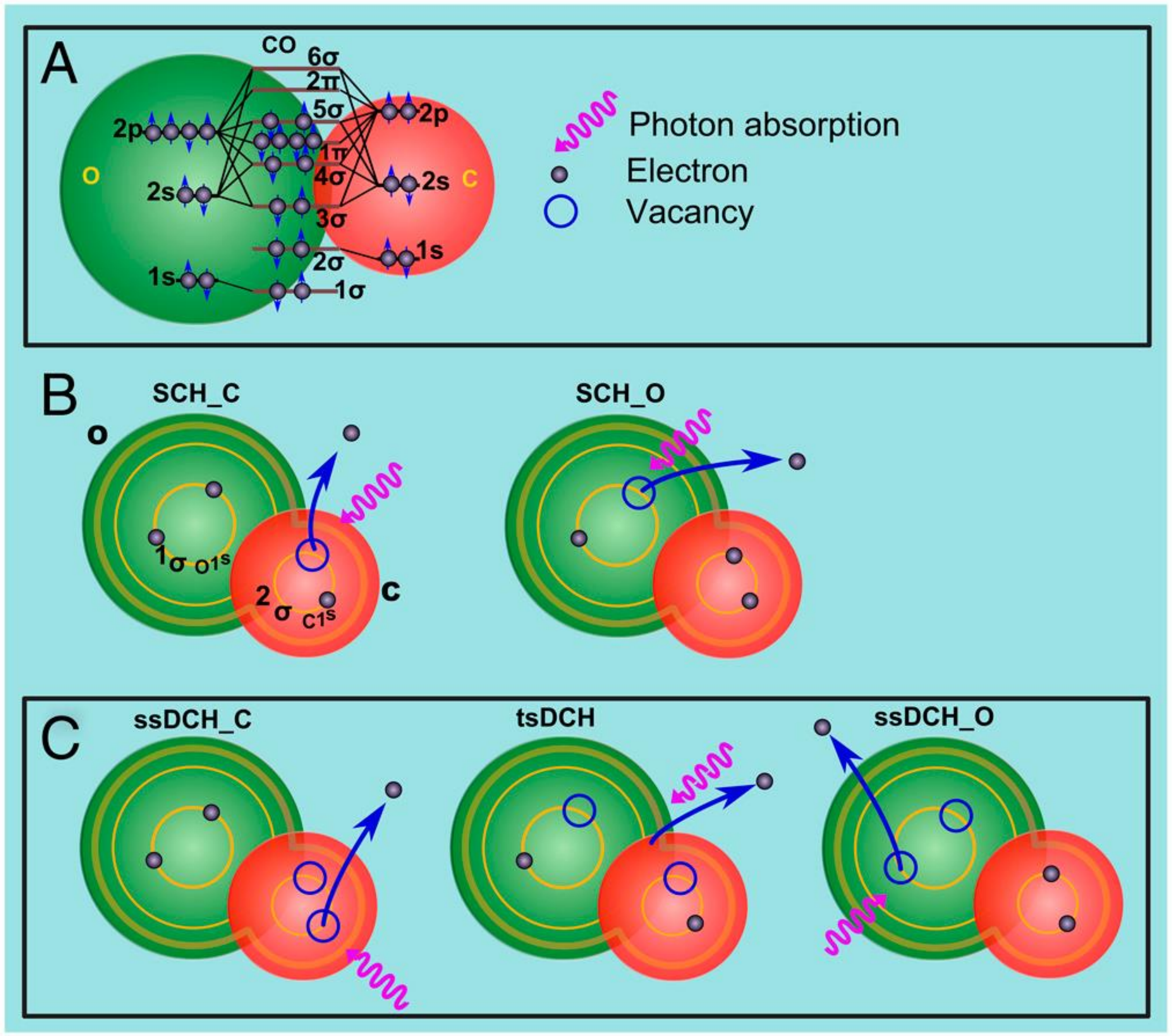
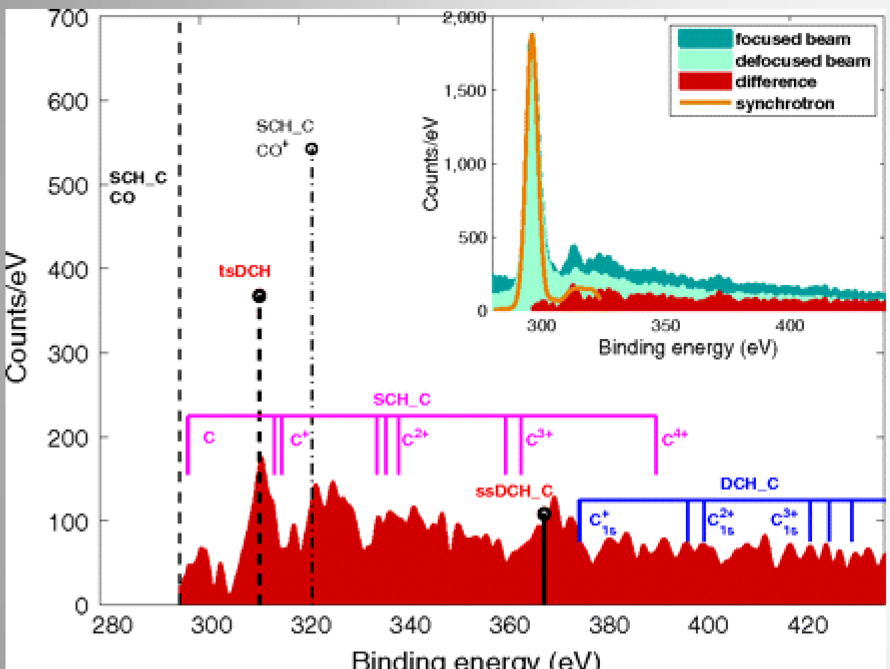
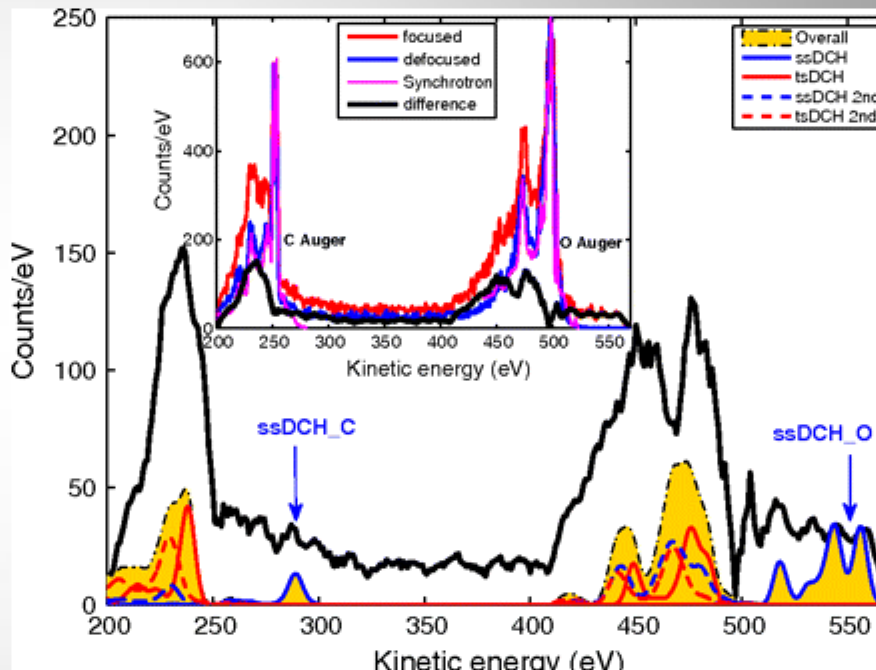


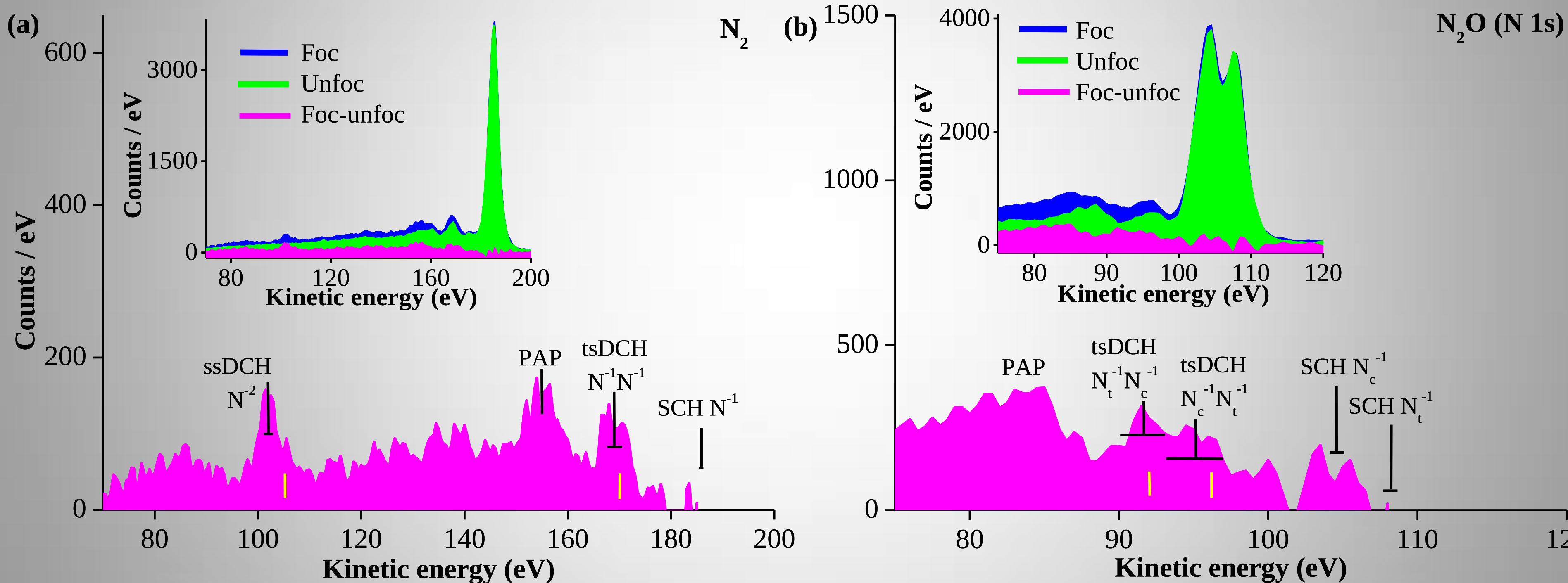
DCH measurements: CO





N. Berrah, ……M.N.Piancastelli *et al.*, PNAS 108, 16912 (2011)

Challenge remains:
TS-DCH → CVV/VVVV Auger
and Auger from other channels overlap



- P. Salén, P. van der Meulen, H.T. Schmidt, R.D. Thomas, M. Larsson, R. Feifel, M.N. Piancastelli, L.Fang, B. Murphy, T. Osipov, N. Berrah, E. Kukk, K. Ueda, J.D. Bozek, C. Bostedt, S. Wada, R. Richter, V. Feyer and K.C. Prince, PRL 108, 153003 (2012)

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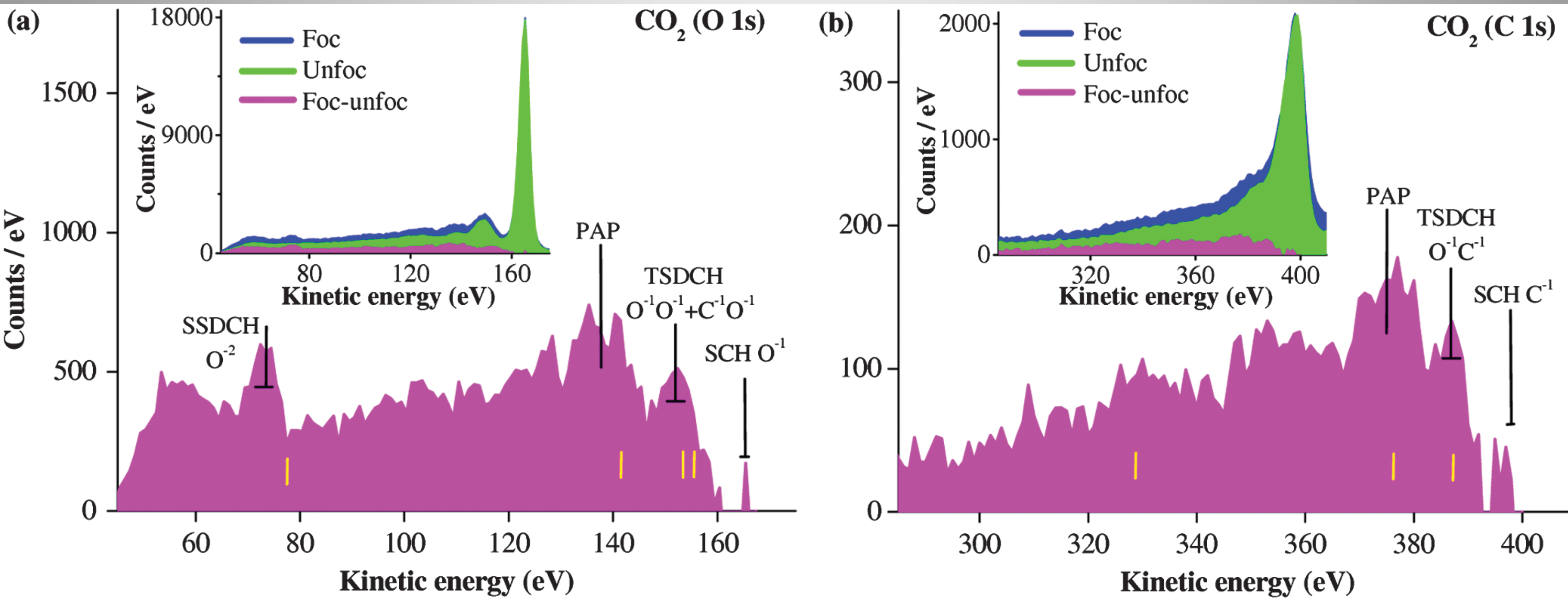

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High performances of the GALAXIES beam line:

high flux and high resolution

Single-channel measurements

Immediate identification of states of the type:

$$(a) \quad \mathrm{Ar}\ 1s^{-1}2p^{-1}np(^2S_{1/2})\epsilon p$$
$$(b) \quad \mathrm{Ar}\ 1s^{-1}2p^{-1}ns(^2P_{1/2,3/2})\epsilon s, \epsilon d.$$

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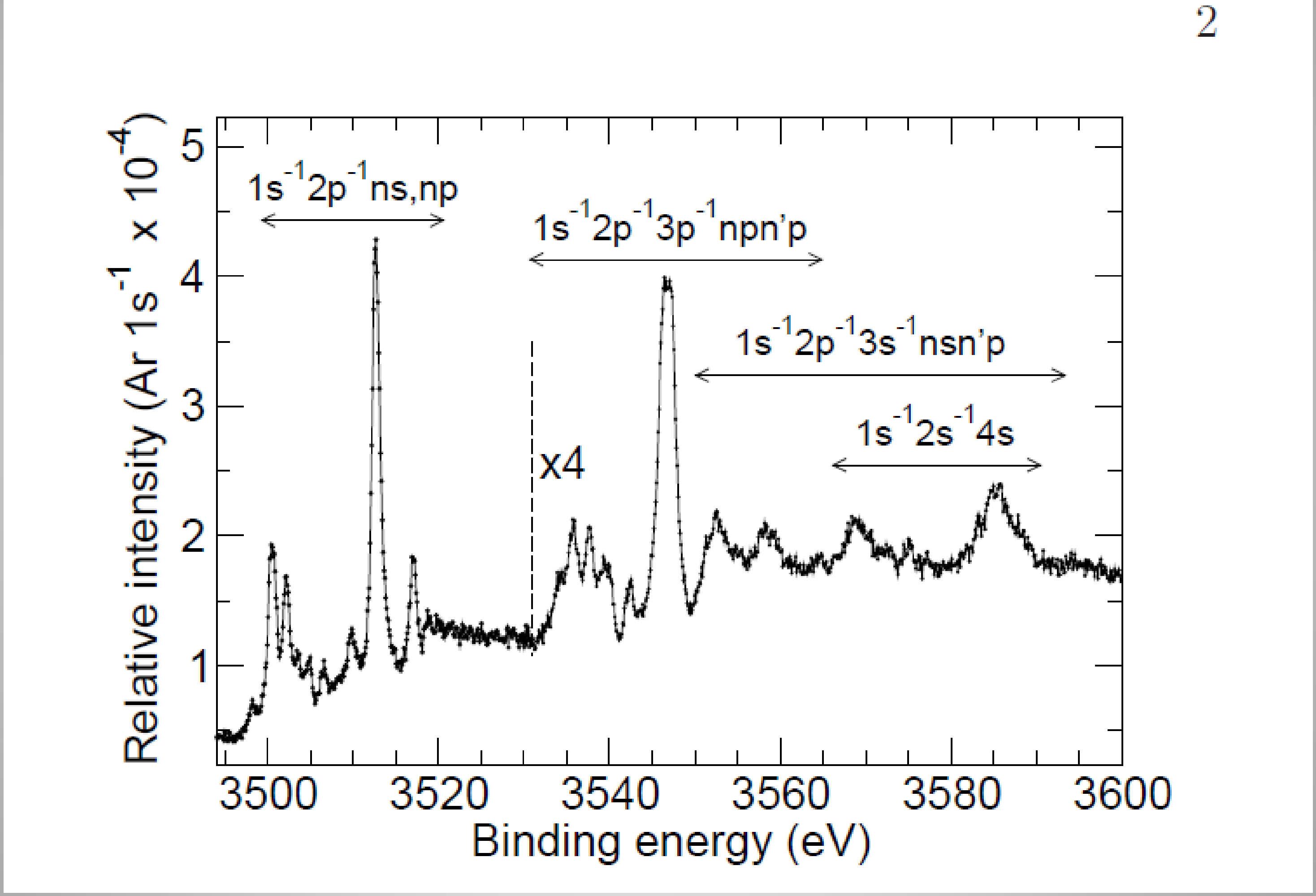
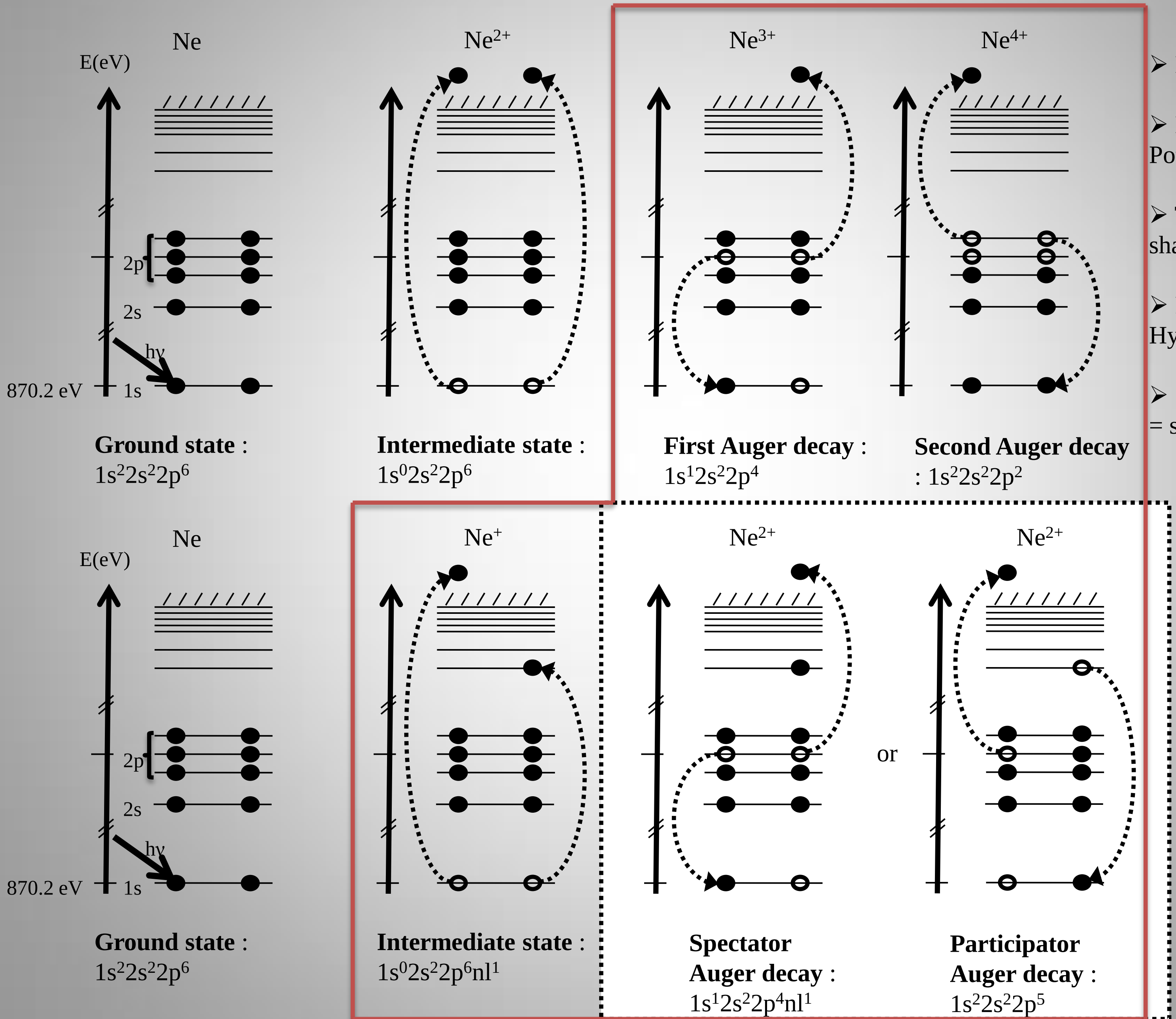


R.Püttner, G. Goldsztejn, D. Céolin, J.-P.Rueff, T. Moreno, R. K. Kushawaha, T. Marchenko, R. Guillemin, L.Journel, D. W. Lindle, M.N. Piancastelli and M. Simon, Phys. Rev. Lett. 114, 093001 (2015)

Double core hole (DCH) in Neon

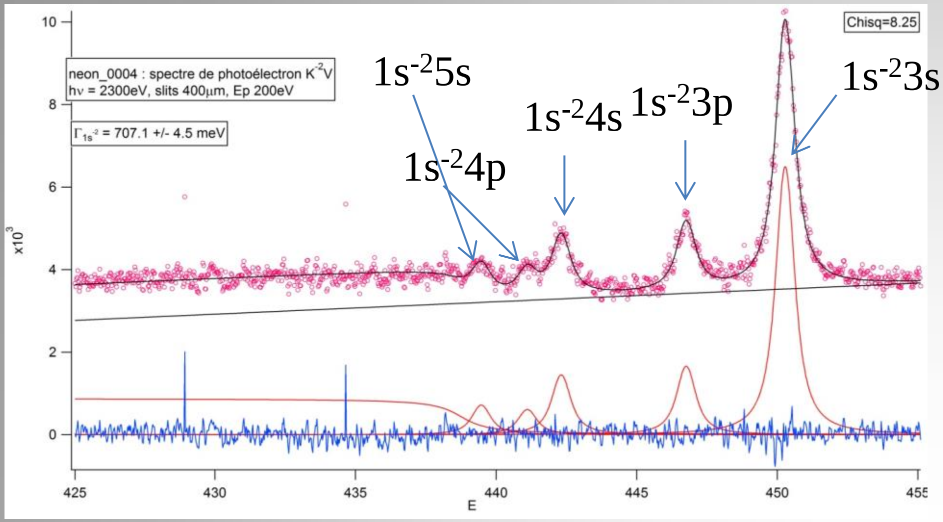


- K^{-2} in Neon.
- Double Ionization Potential = 1863 eV.
- The two photoelectrons share the energy
- First Auger electron = Hypersatellite.
- Second Auger electron = satellite.

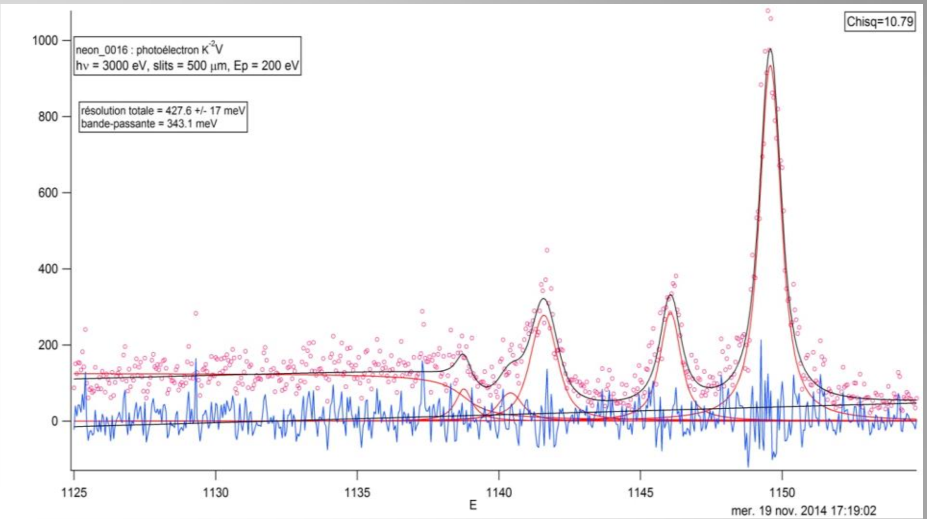
- $K^{-2}V$ in Neon.
- Photoelectrons well separated and easy to identify.
- Different type of decays at different energies.

Photon energy dependence of DCH

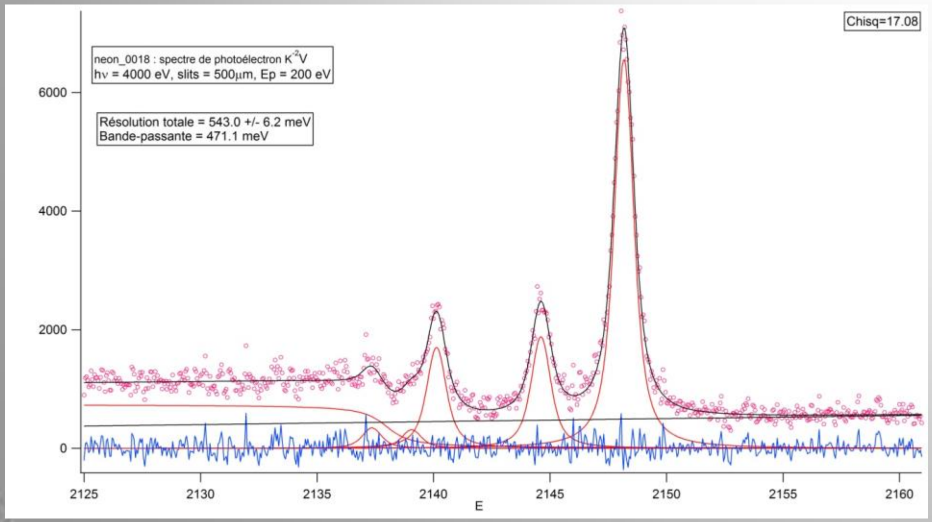
hν = 2300 eV



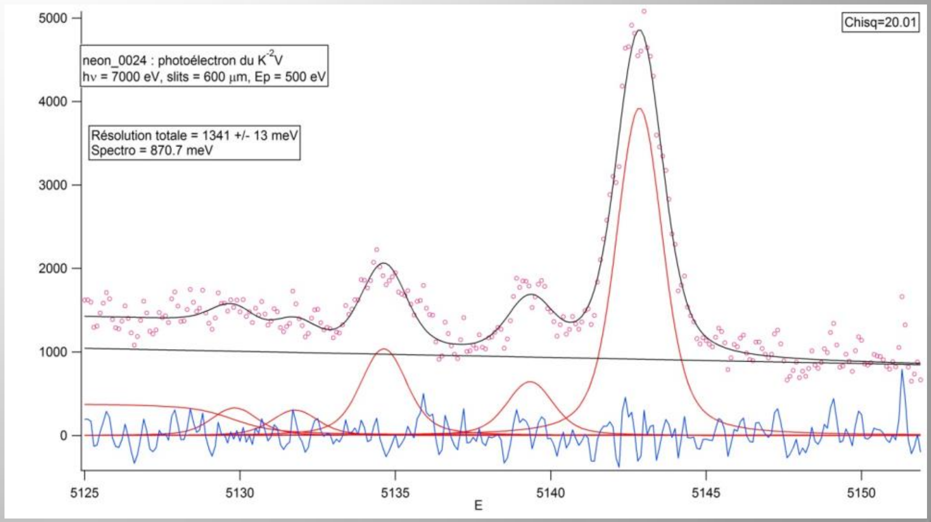
hν = 3000 eV



hν = 4000 eV



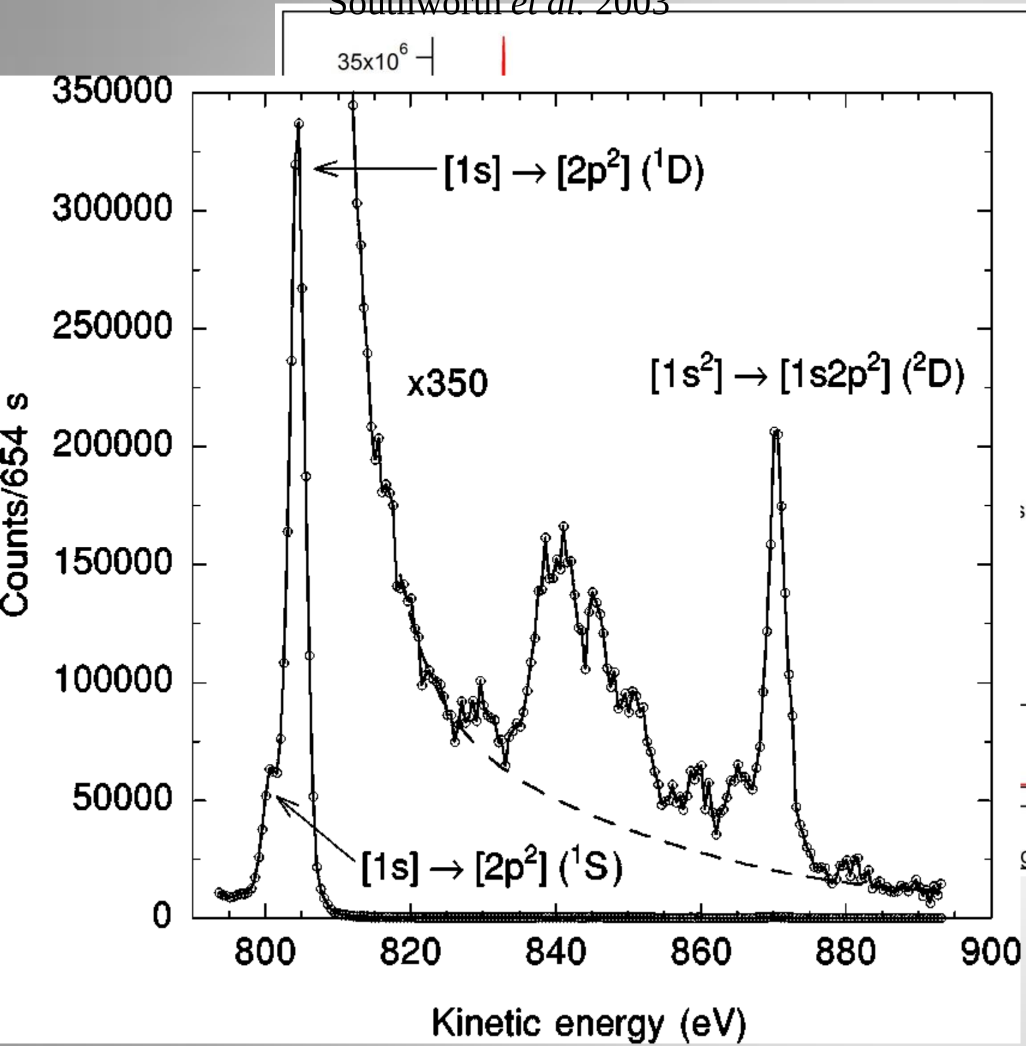
hν = 7000 eV

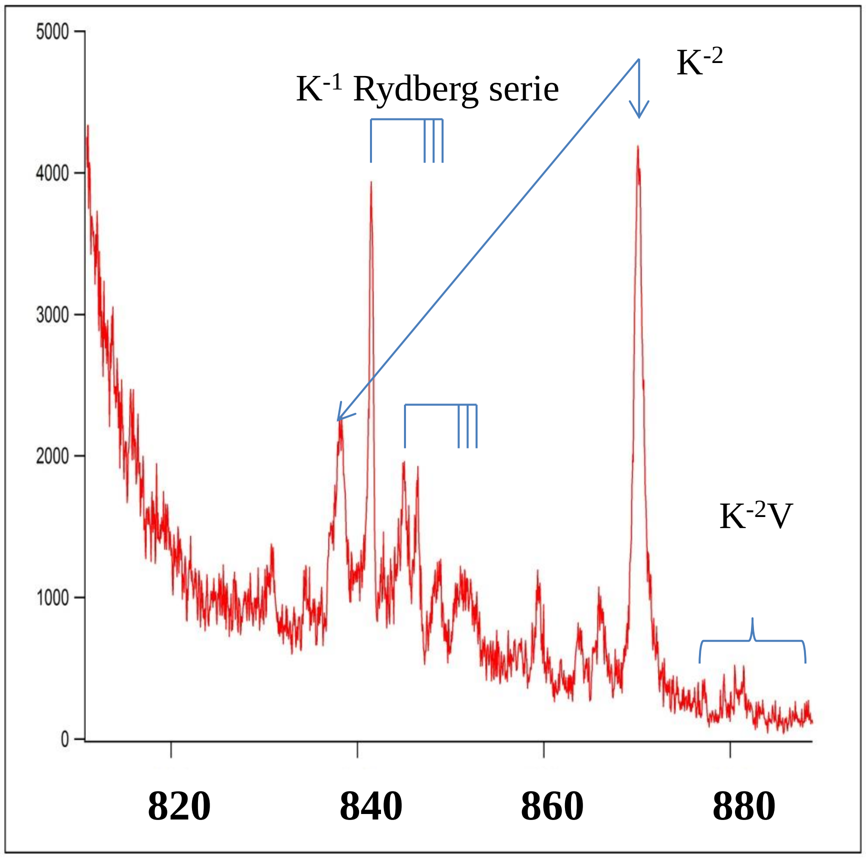


The hypersatellite spectra

Southworth *et al.* 2003

HAXPES 2014





- ➢ K^{-1} ≈ 1000 times more intense than K^{-2}
- ➢ Thanks to much better resolution we were able to identify K^{-1} satellites

Southworth *et al.* PRA 67, 062712 (2003)
Svensson *et al.* J. Electron Spectrosc. Relat. Phenom. 47, 327 (1988)

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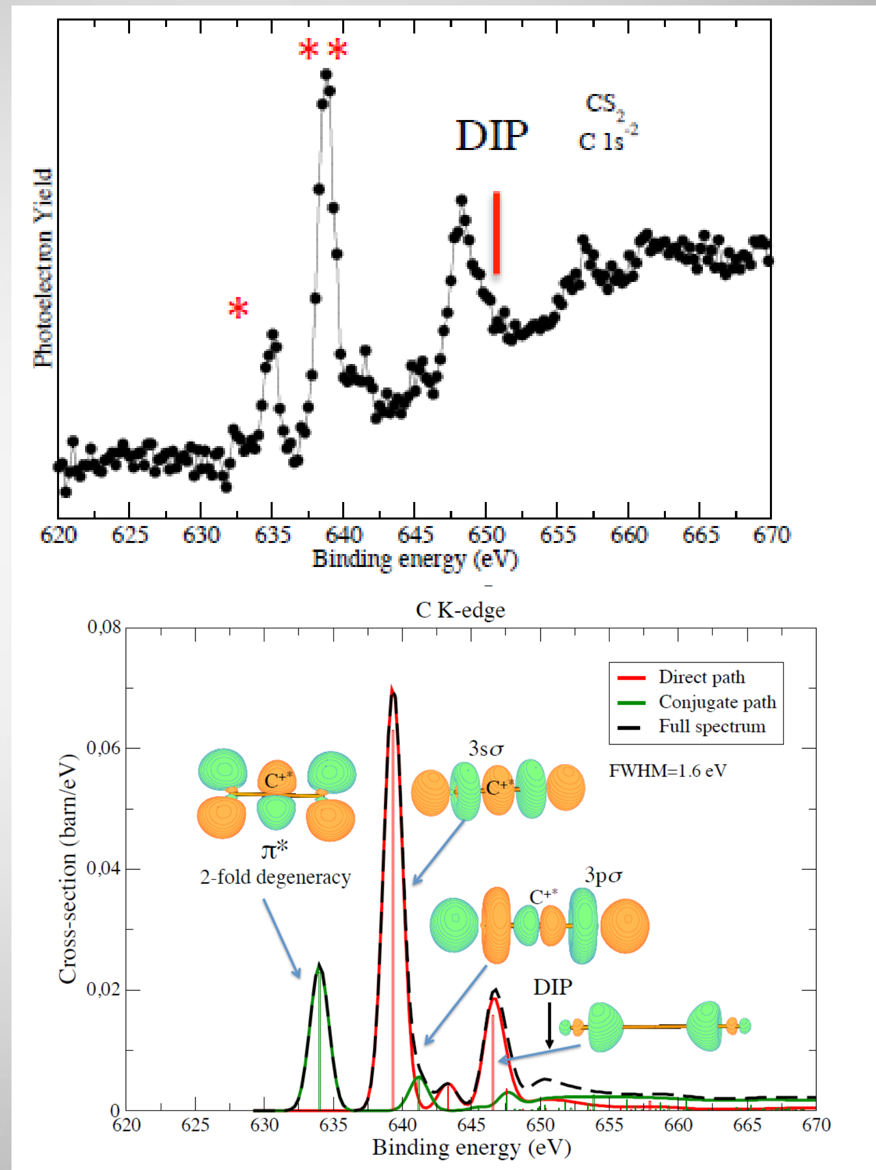
$\underline{C}S_2$



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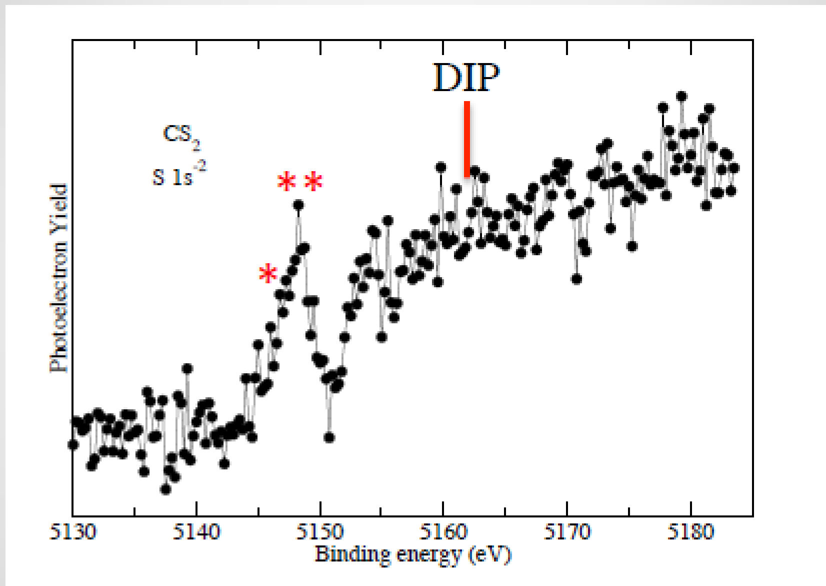
CS_2



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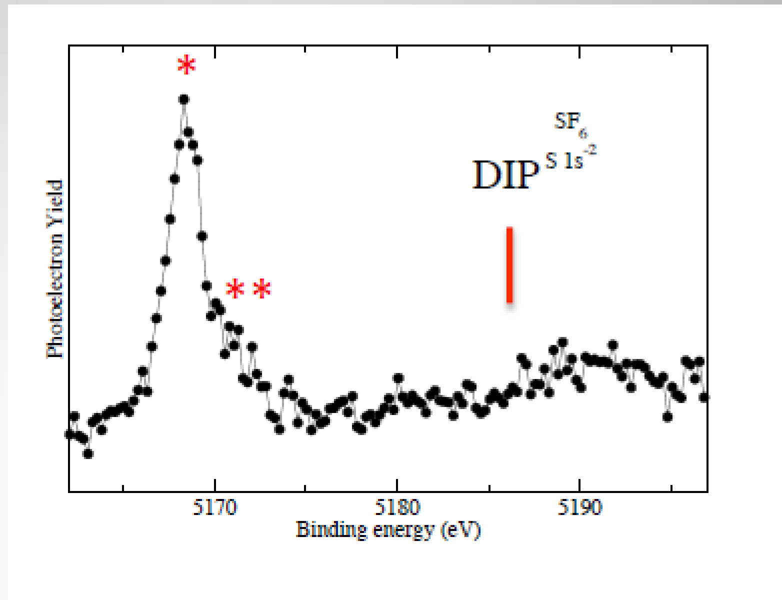
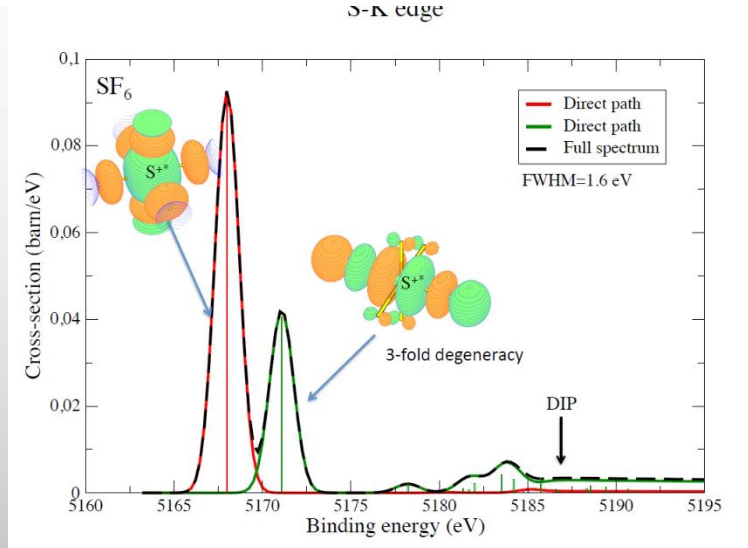
SF_6





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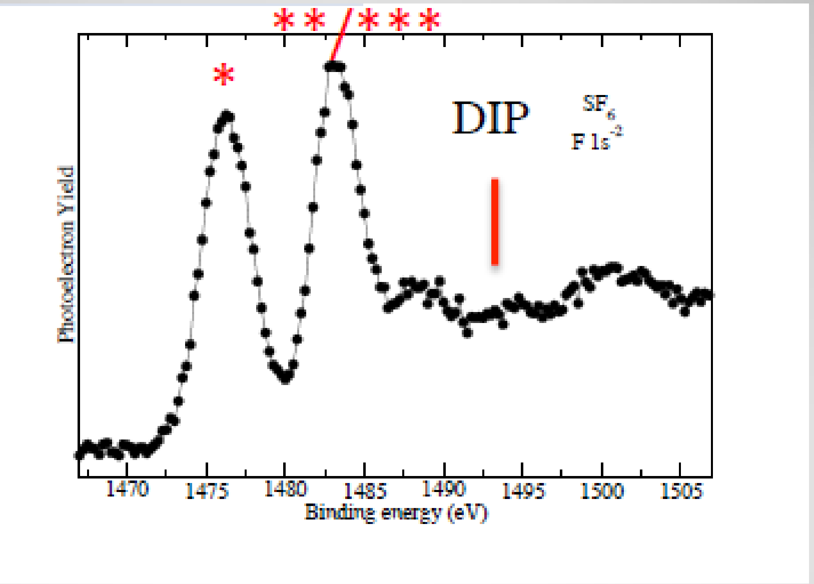
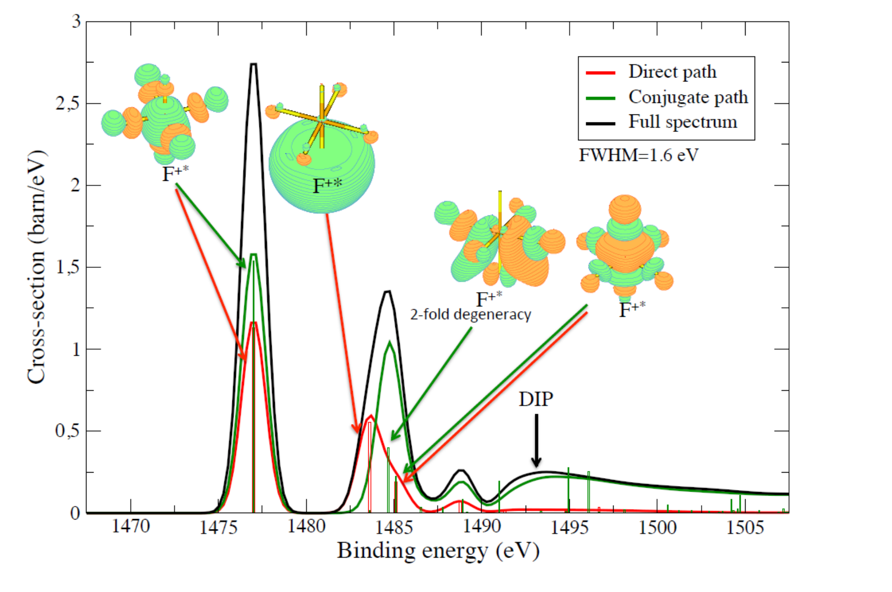
SF$_6$

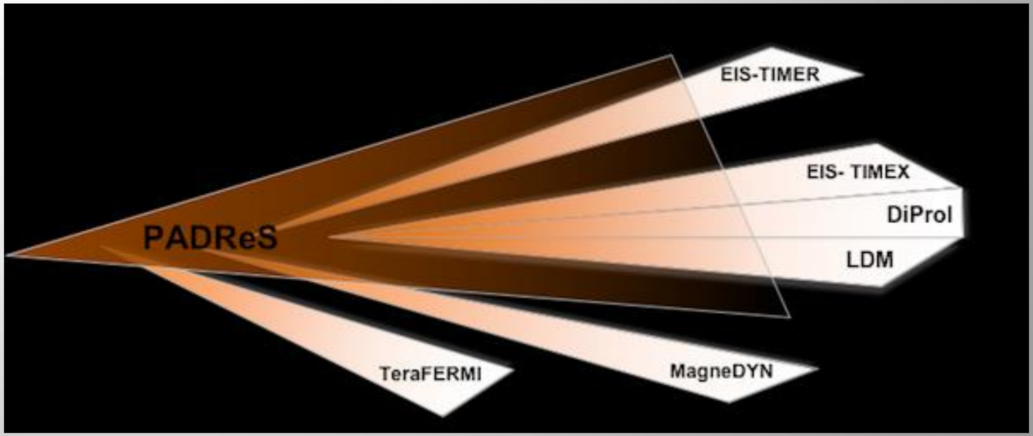




FERMI

Elettra, Trieste, Italy

Aerial view



Beam transport and beamlines

FERMI

First seeded FEL

	λ seed	261.60	(±0.5) nm		4.7394	eV
harmonic stage1↓	1st stage wl (nm)	2nd stage wl (nm)				
4	65.40	21.80	16.35	13.08	10.90	9.34
5	52.32	17.44	13.08	10.46	8.72	7.47
6	43.60	14.53	10.90	8.72	7.27	6.23
7	37.37	12.46	9.34	7.47	6.23	5.34
8	32.70	10.90	8.18	6.54	5.45	4.67
9	29.07	9.69	7.27	5.81	4.84	4.15
10	26.16	8.72	6.54	5.23	4.36	
11	23.78	7.93	5.95	4.76		
12	21.80	7.27	5.45	4.36		
13	20.12	6.71	5.03	4.02		
harmonic stage 2→		3	4	5	6	7
harmonic stage1↓	1st stage En. (eV)	2nd stage Energy (eV)				
4	18.96	56.87	75.83	94.79	113.75	132.70
5	23.70	71.09	94.79	118.49	142.18	165.88
6	28.44	85.31	113.75	142.18	170.62	199.06
7	33.18	99.53	132.70	165.88	199.06	232.23
8	37.92	113.75	151.66	189.58	227.49	265.41
9	42.66	127.97	170.62	213.28	255.93	298.59
10	47.39	142.18	189.58	236.97	284.37	
11	52.13	156.40	208.54	260.67		
12	56.87	170.62	227.49	284.37		
13	61.61	184.84	246.45	308.06		

Variable polarization

Negligible photon energy jitter

Negligible time jitter

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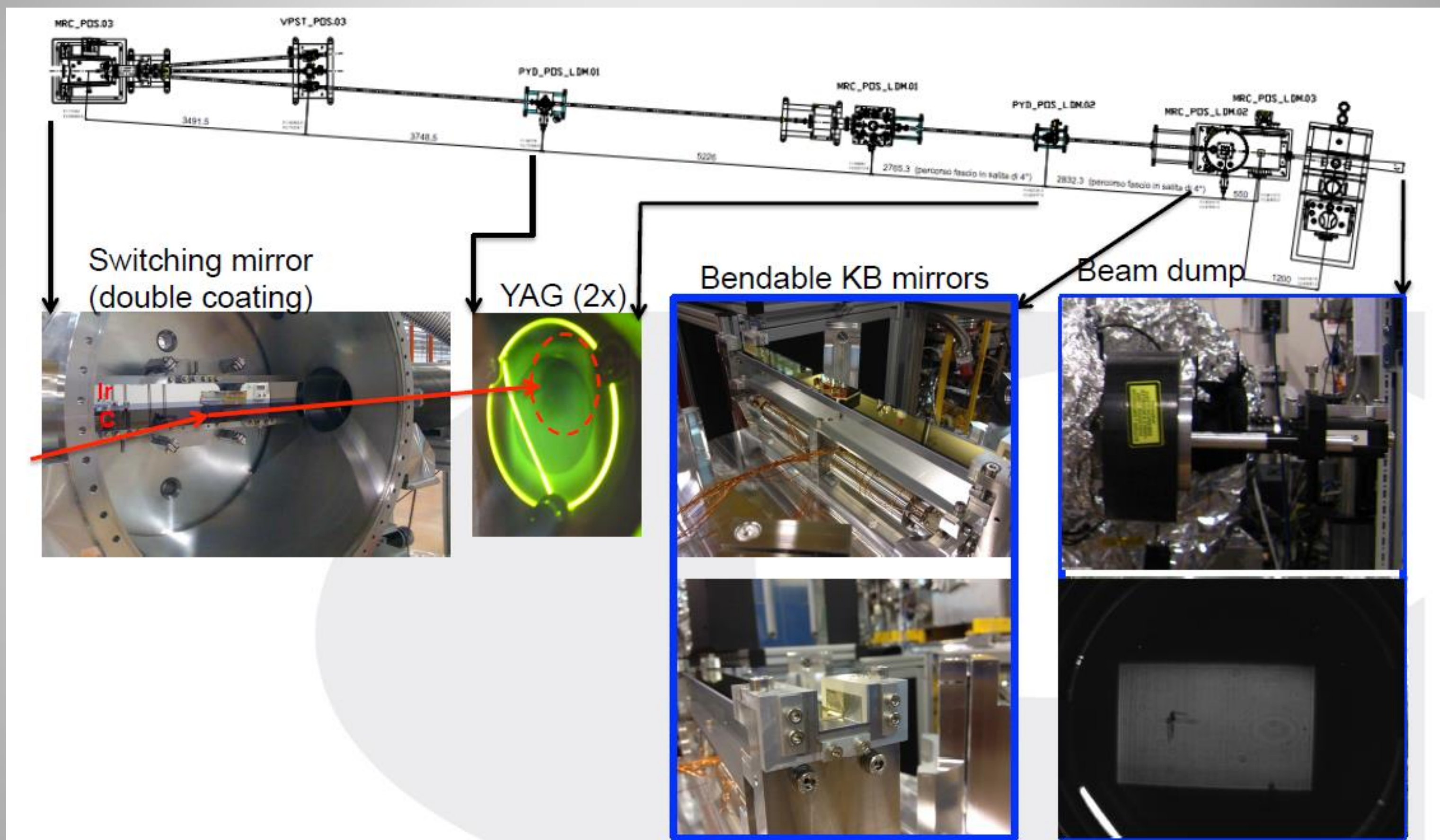
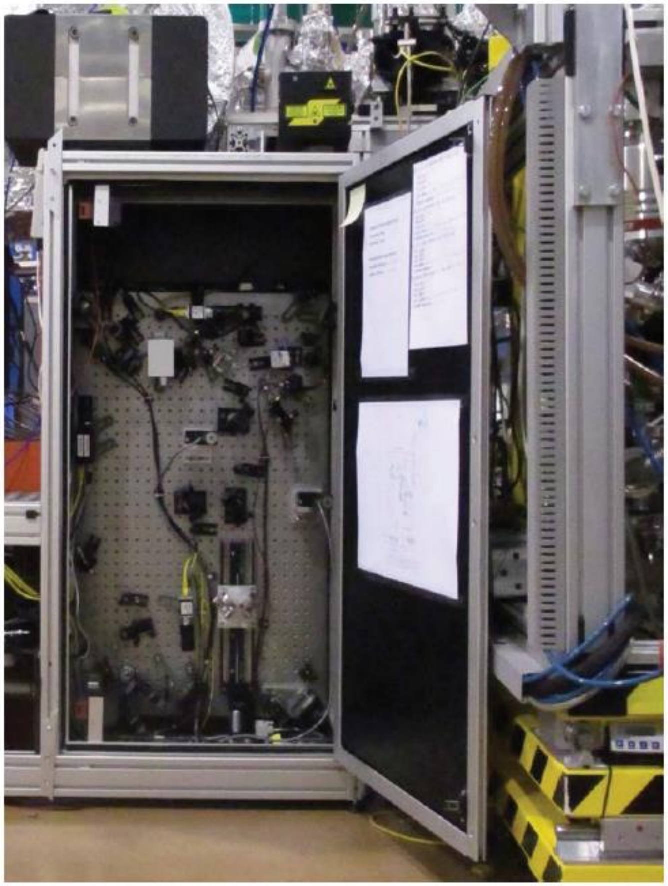


Low Density Matter (LDM)



Ti:sapphire (same used to form the FEL pulse)

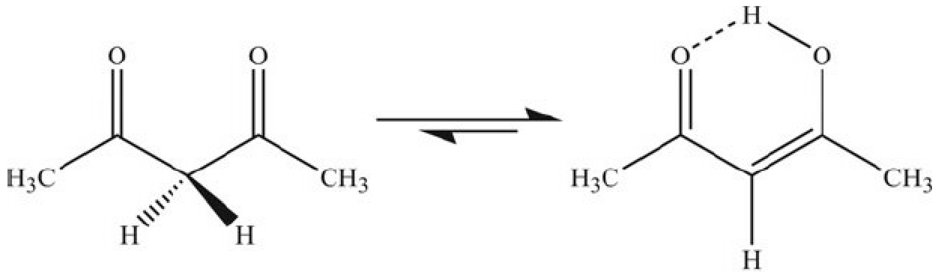
λ=780 nm

Energy/pulse: 1 mJ
Pulse width 140 fs
(autocorrelation)

USL-LDM Controls	
Attenuation (energy meter avaialble)	0 – 100 %
Polarization	Full control
Delay resolution	2.6 fs
Beam Position resolution	10 μrad (upgrade: piezo tip-tilt)

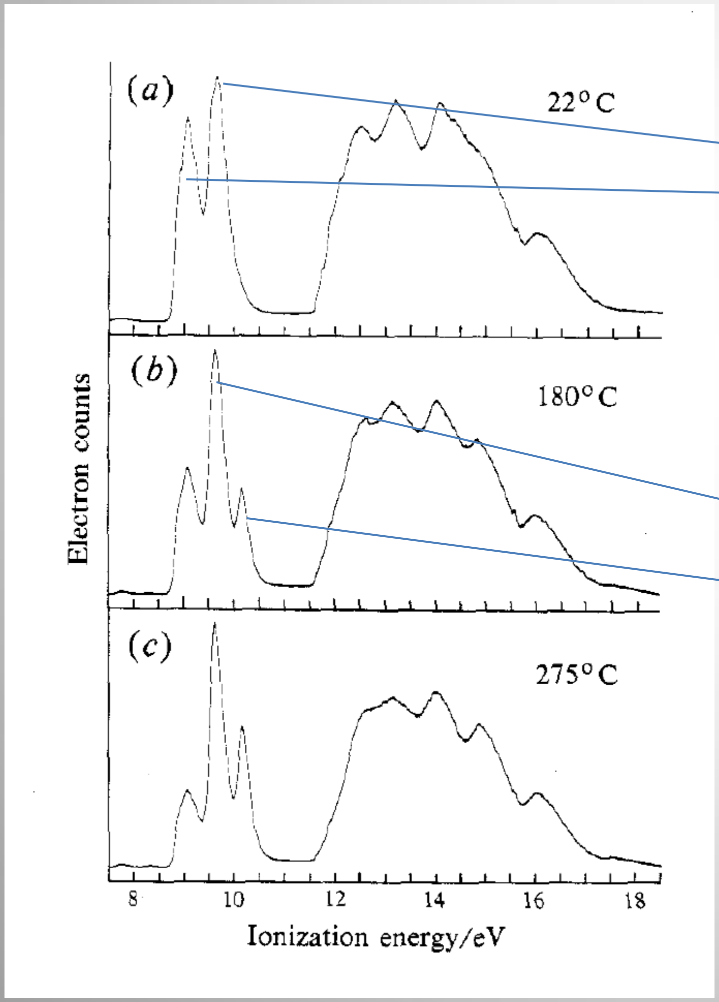
Probing keto-enol tautomerism

in acetylacetone



Keto   Enol

Valence Photoelectron spectra are characteristic of the forms (Hush *et al.* Aust. J.Chem. **40,** (1987) 559)



Enol form

Keto form

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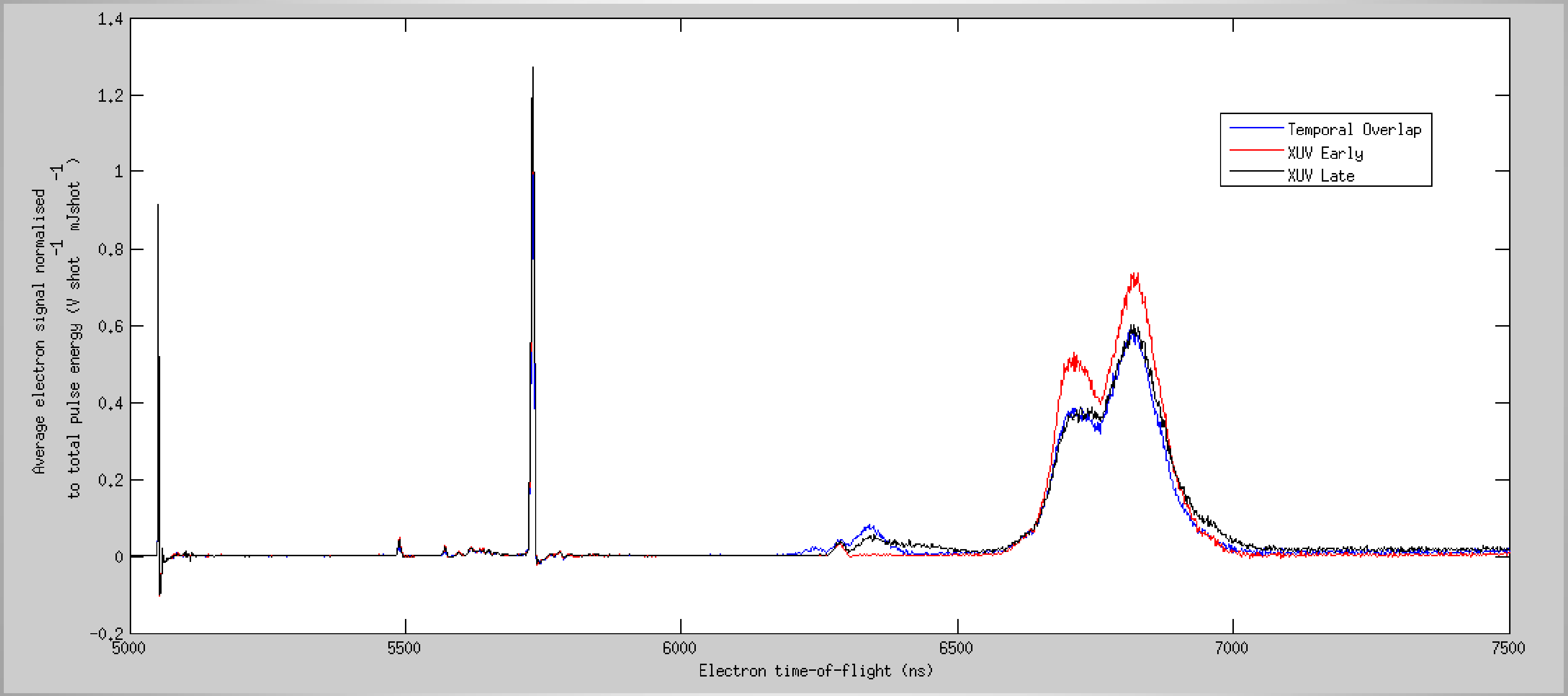

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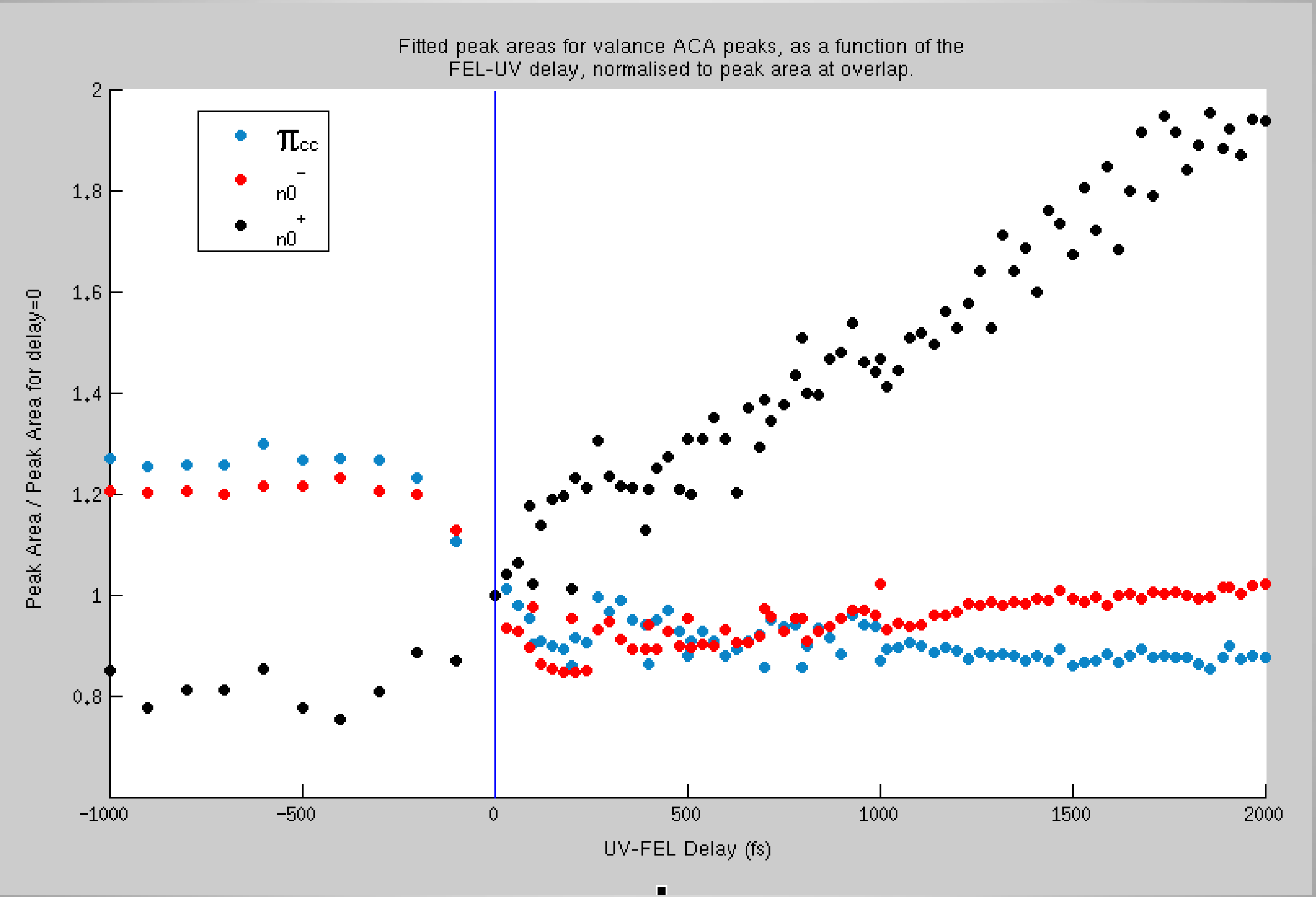
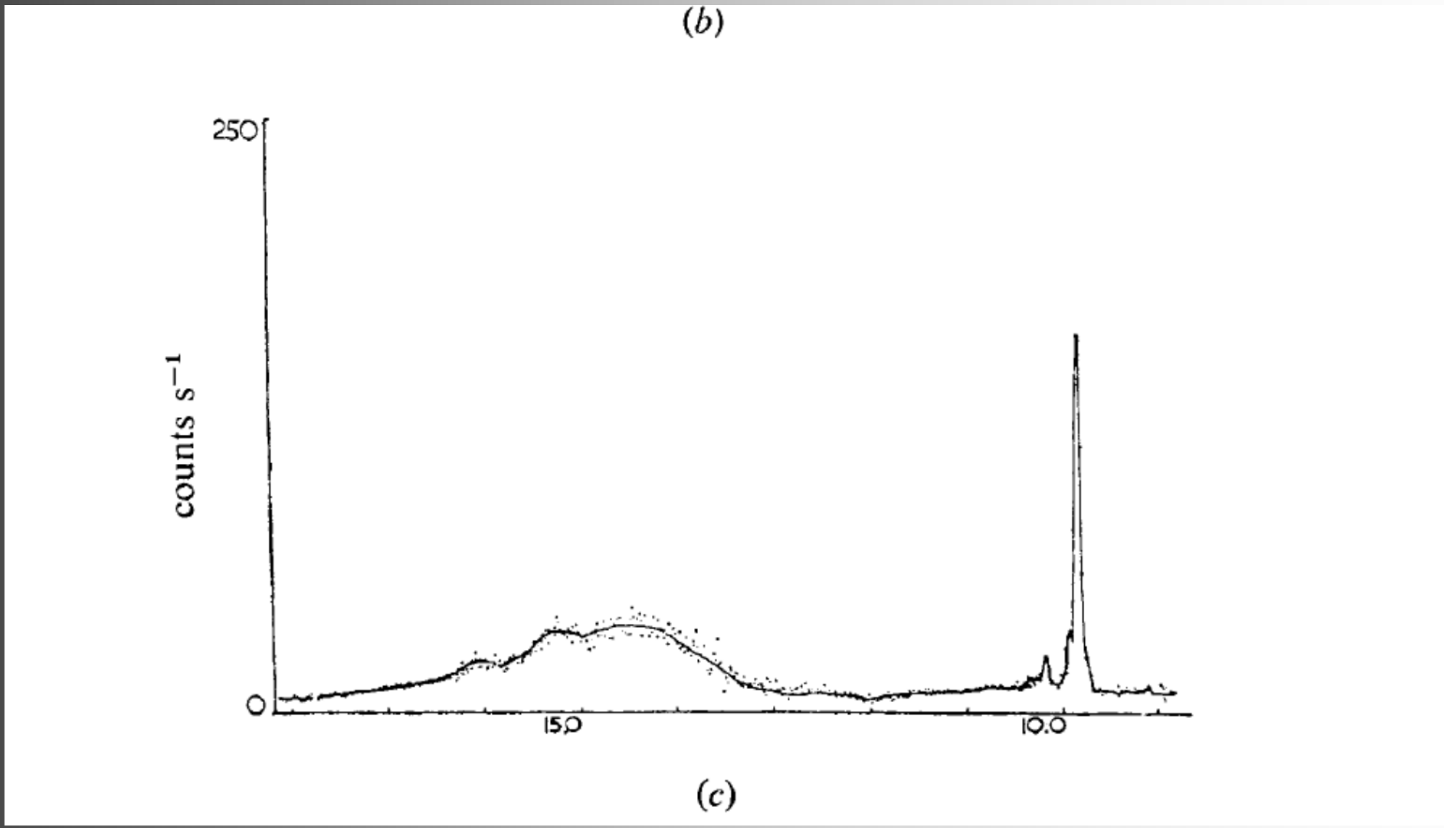


CH_3



J. Dyke, N. Jonathan, E. Lee and A. Morris
J. Chem. Soc., Faraday Trans. 2 72, (1976) 1385

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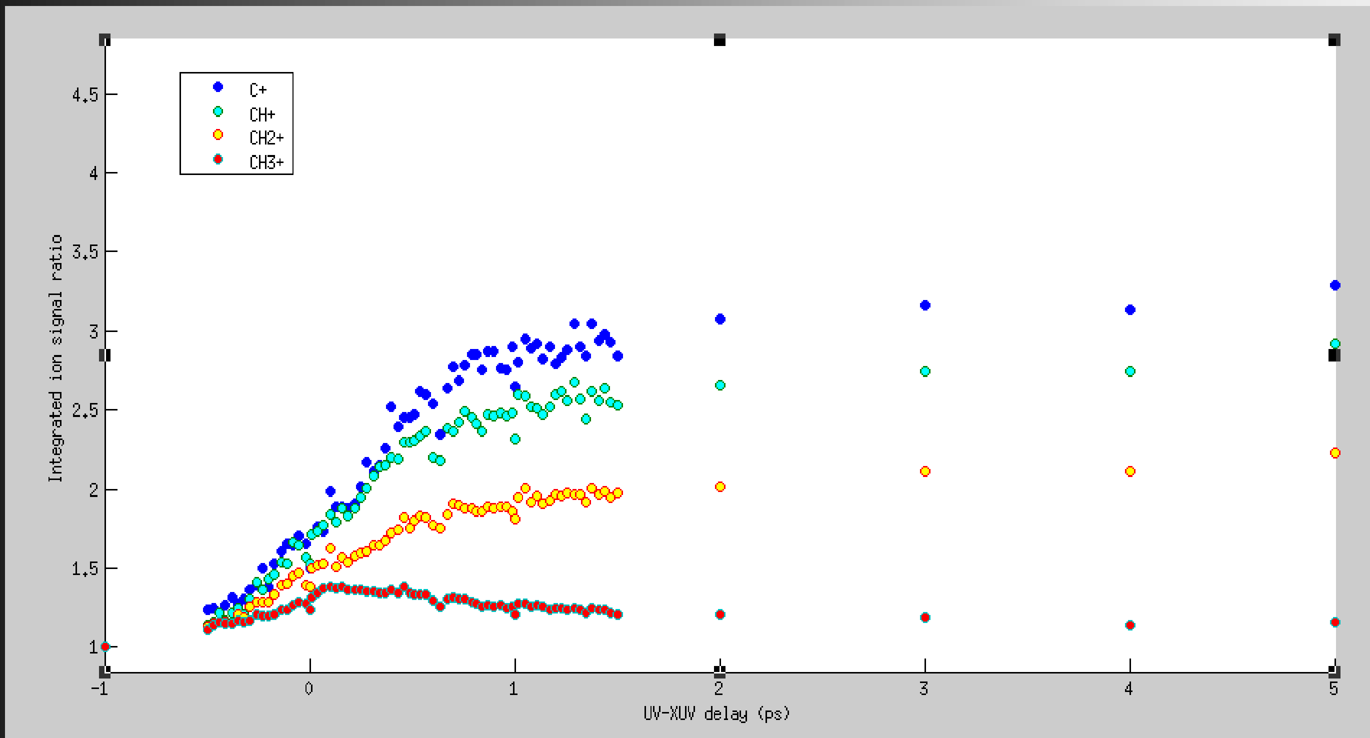

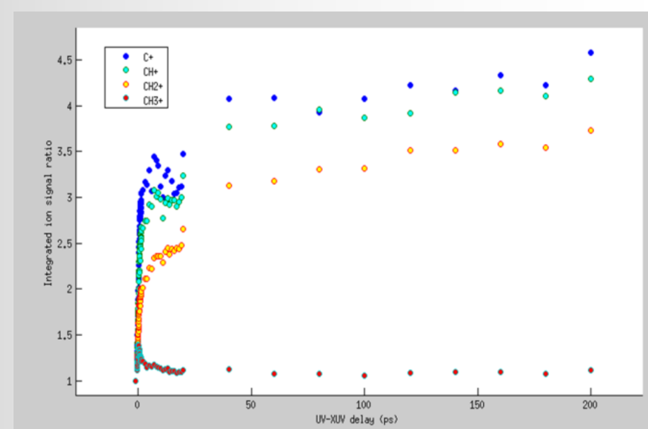

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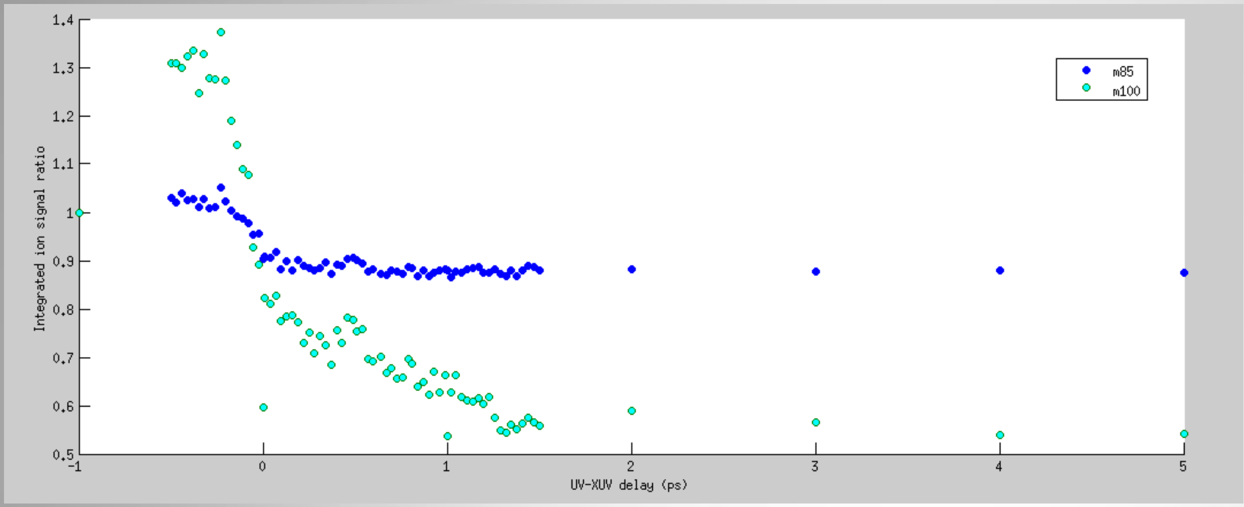

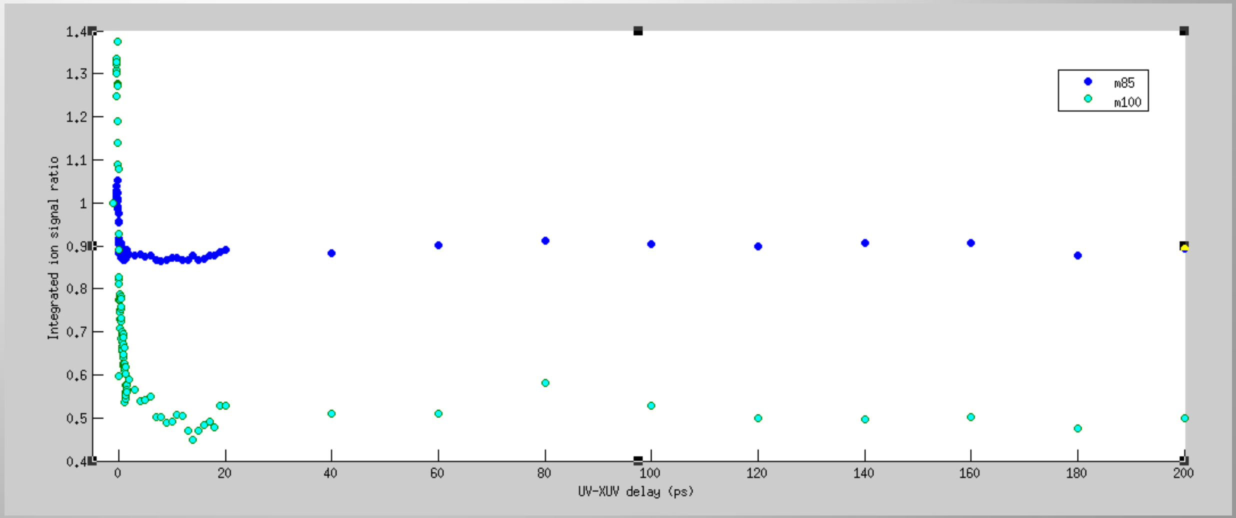

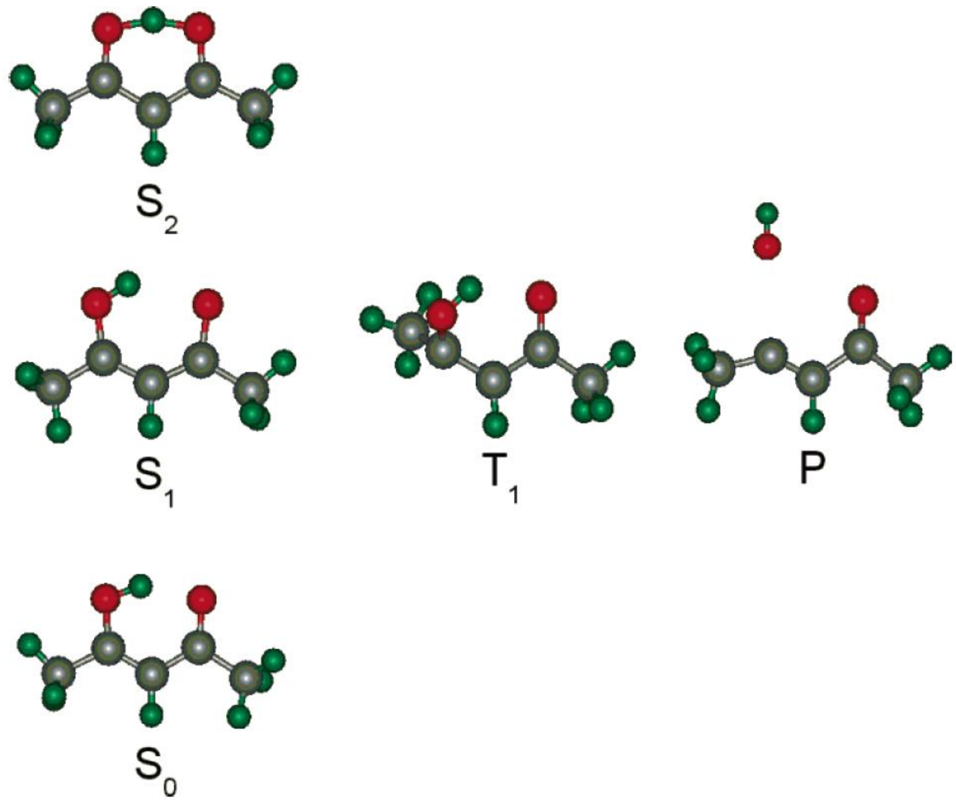

6650 *J. Phys. Chem. A* **2004,** *108,* 6650−6655

Ultrafast Electron Diffraction: Structural Dynamics of the Elimination Reaction of Acetylacetone

Shoujun Xu, Sang Tae Park, Jonathan S. Feenstra, Ramesh Srinivasan, and Ahmed H. Zewail*

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