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Covariance mapping of molecular fragmentation

from simple to complex systems

Leszek Frasiński

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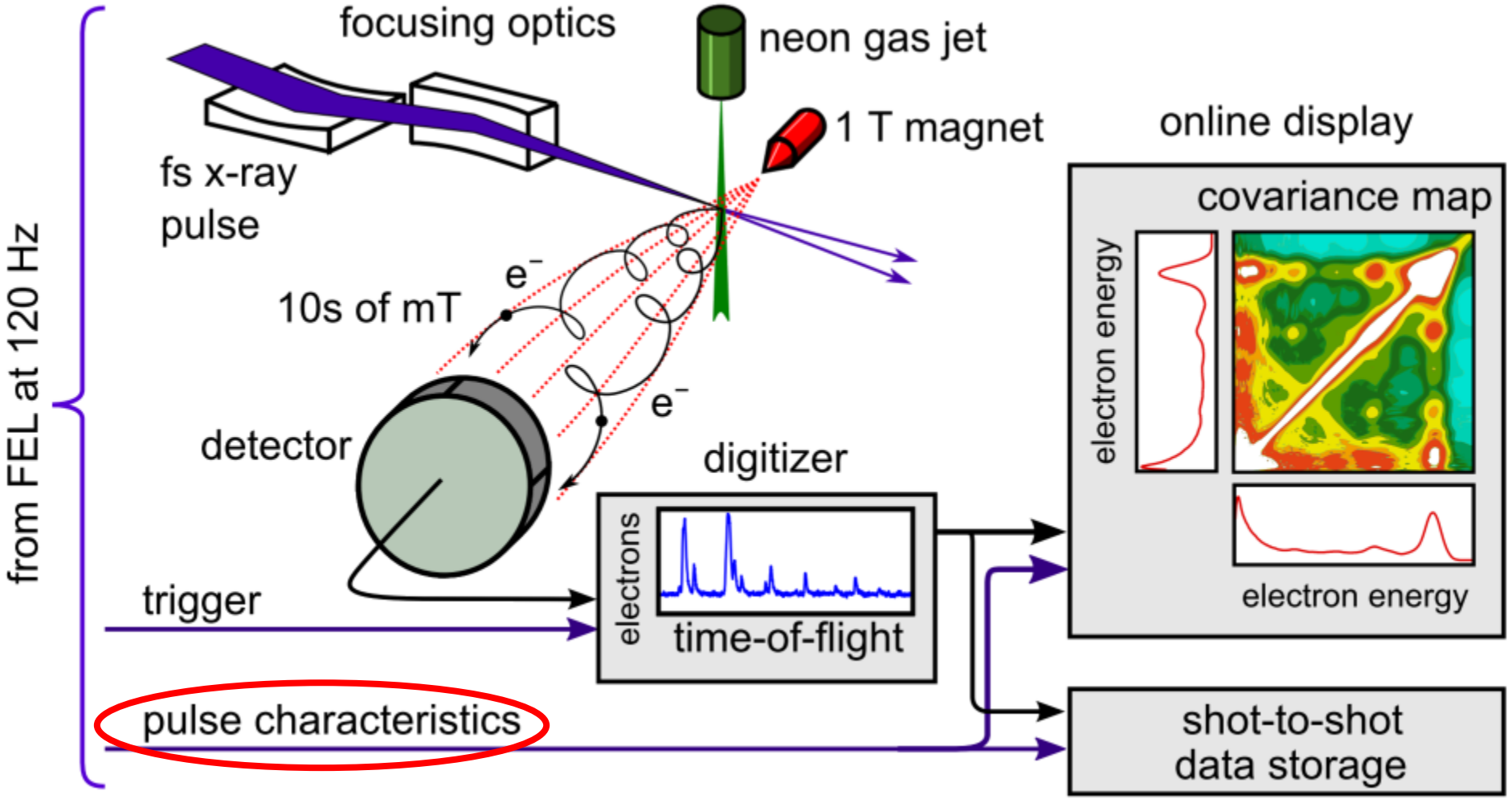
Neon photoelectron covariance mapping at LCLS



L J Frasinski *et al*, *Phys. Rev. Lett.* **111** 073002 (2013)

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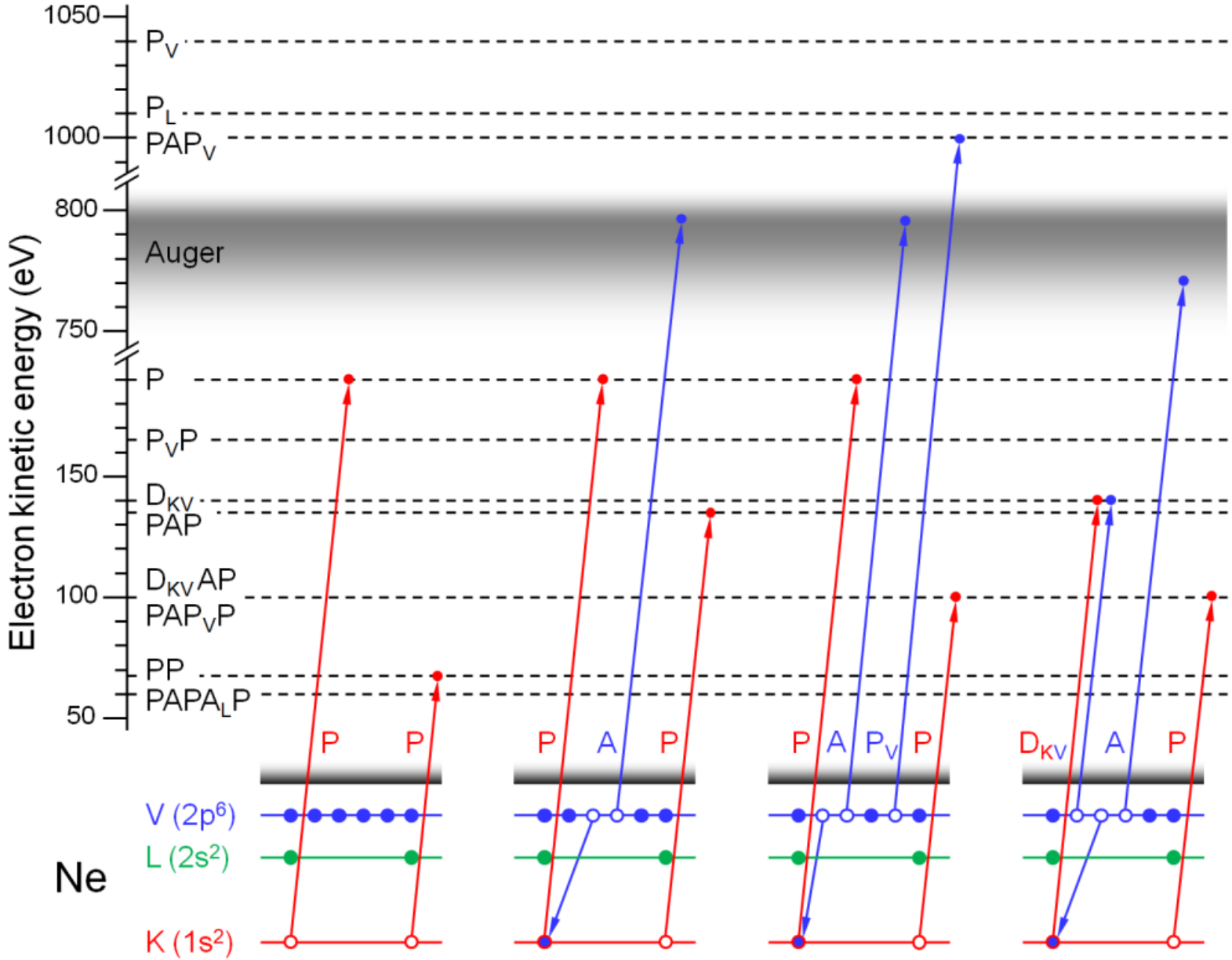
Examples of ionization processes in neon



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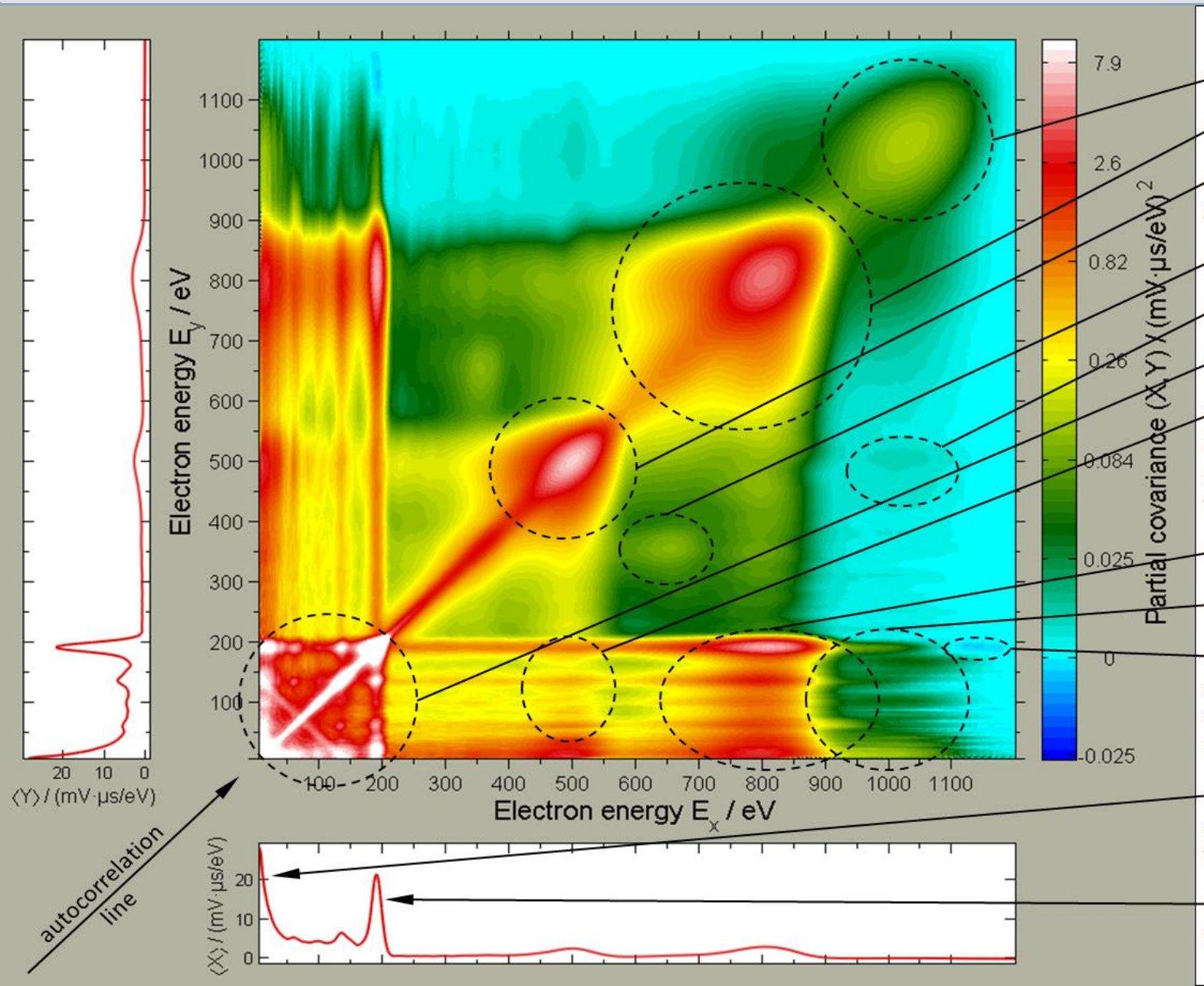
Partial covariance map of Ne at 1062 eV



Origin of electrons:

- Ne valence-valence
- Ne Auger-Auger
- H_2O core-core, core-Auger and Auger-Auger
- N_2 core-Auger
- H_2O core-valence
- Ne core-core
- spurious Ne-H_2O correlations due to common sample density fluctuations
- Ne core-Auger
- Ne core-valence
- artefact from photon energy jitter compensation, which distorts Auger peaks
- double Auger and secondary electrons from electrode surfaces
- Ne main (core) photoline

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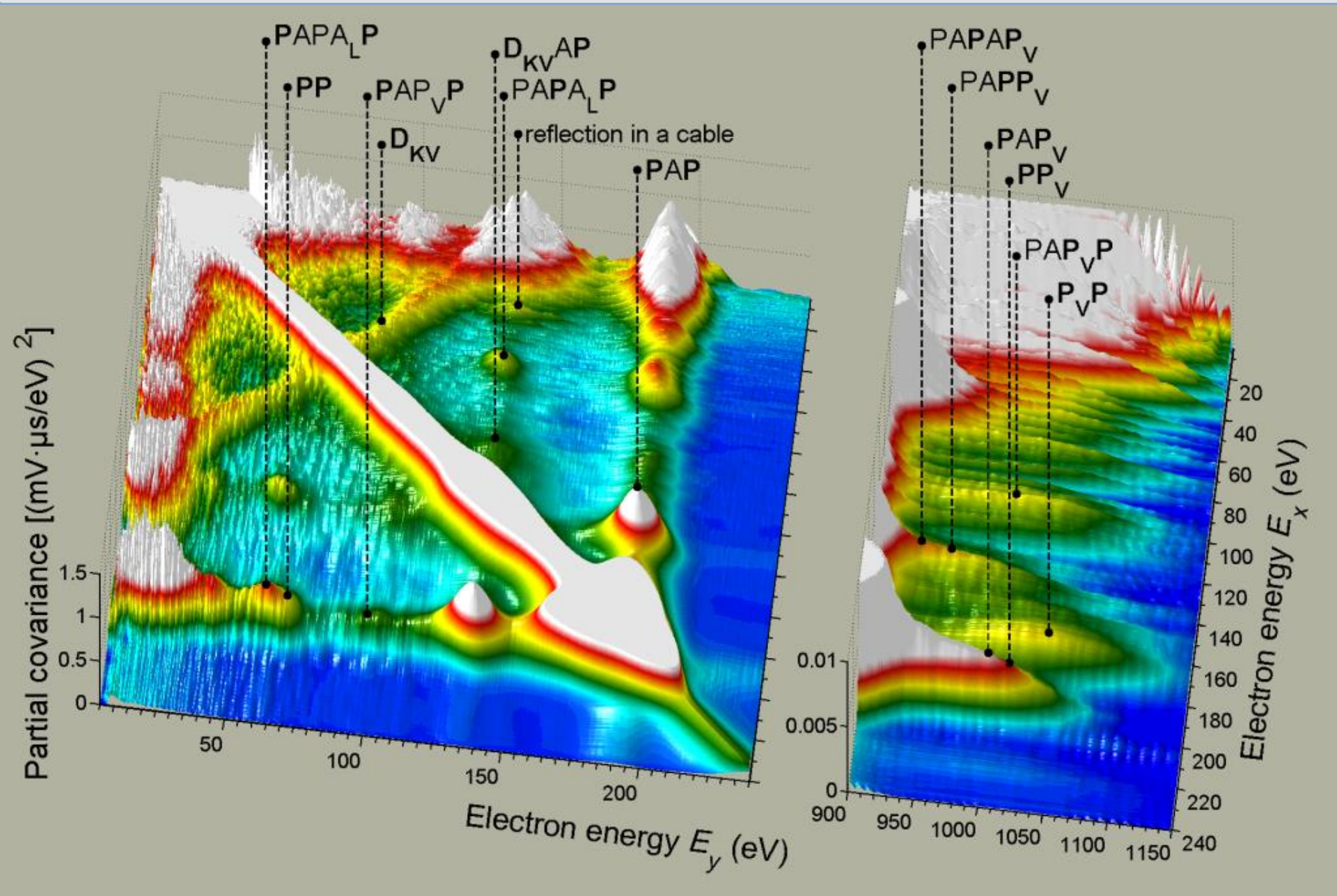
Identification of ionization processes in neon



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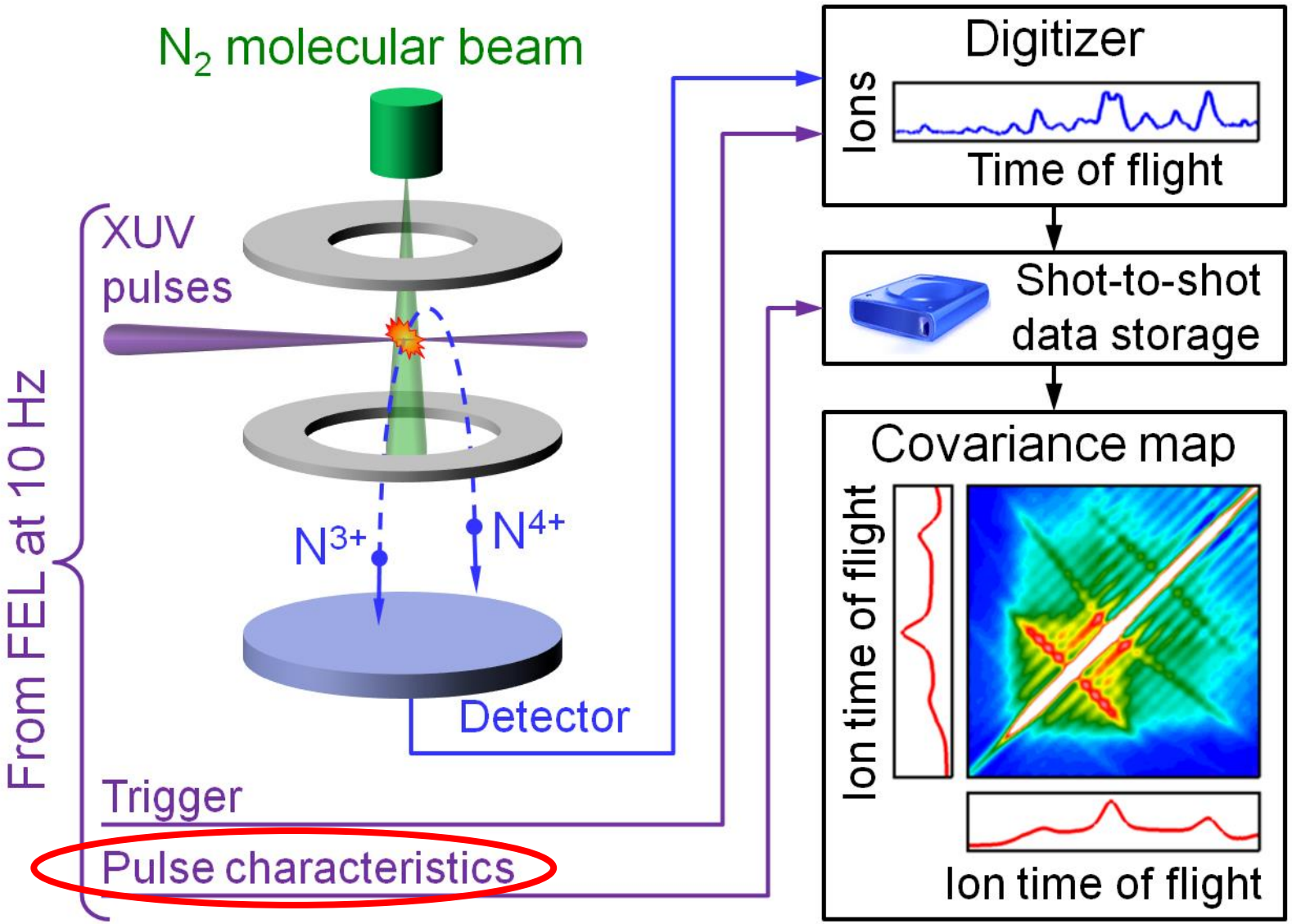
Diatomics: Coulomb explosion of N_2 and I_2 at FLASH



O Kornilov *et al*, *J. Phys. B* **46** 164028 (2013)

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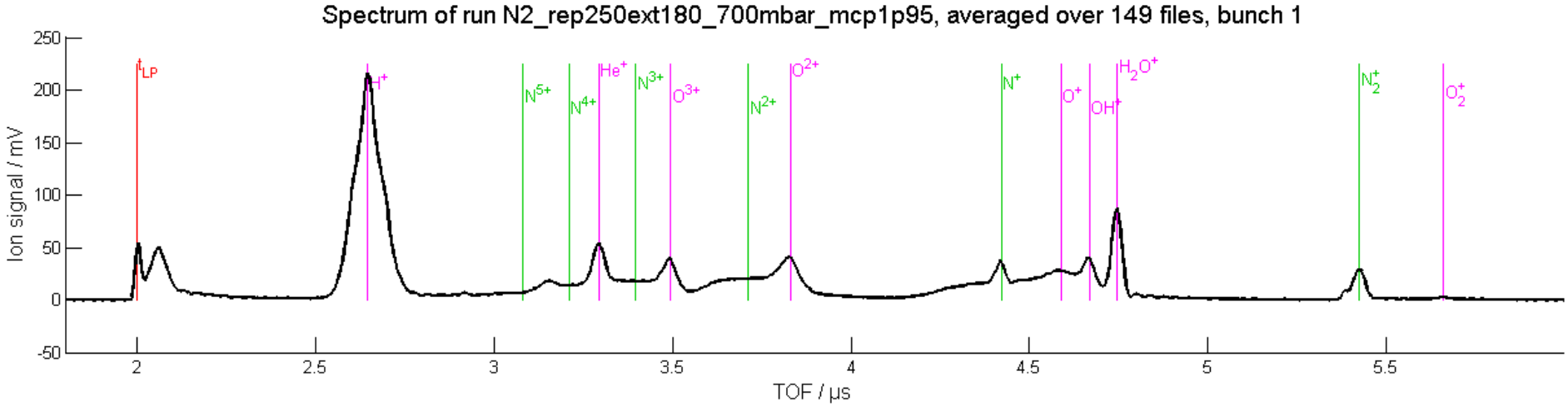
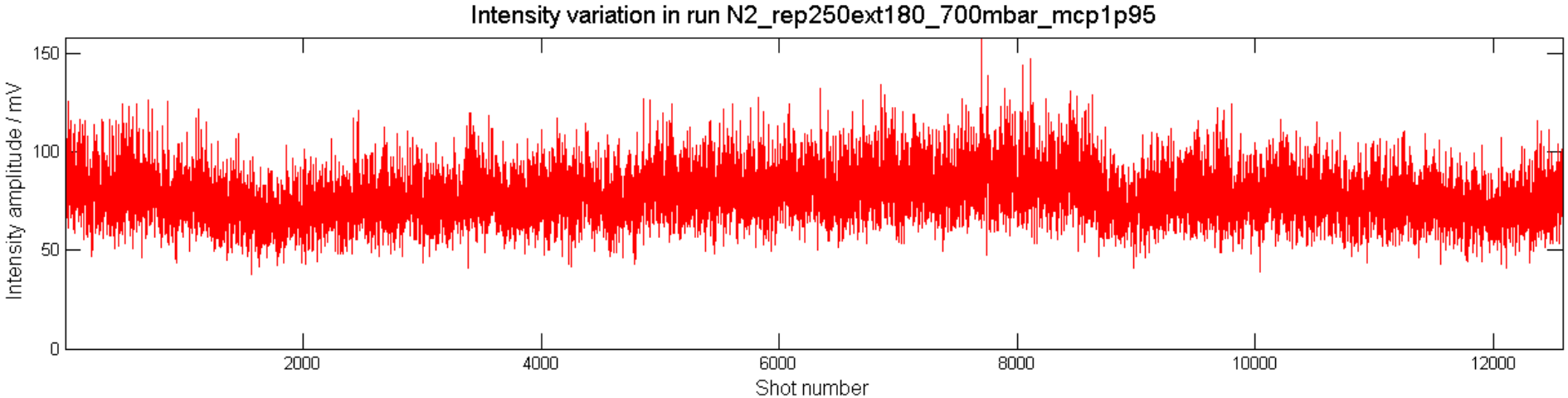
N_2 spectrum and intensity variation

Nice conventional, 1D spectra:



But for covariance mapping there are problems due to intensity fluctuations:



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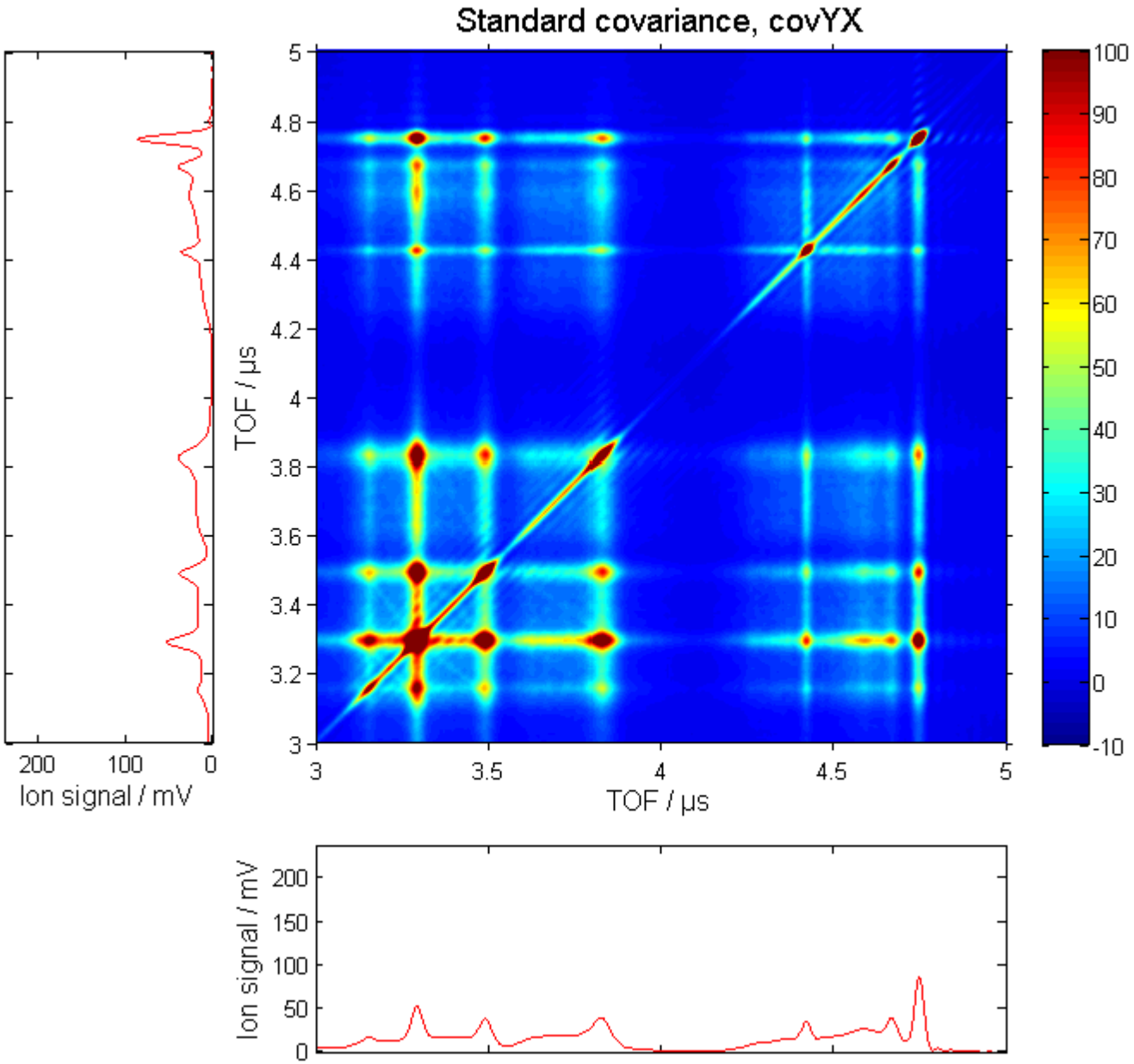
Simple covariance map of N_2



Every ion peak is correlated with every other peak via intensity fluctuations.

This is because when intensity increases then the production of every ion increases.

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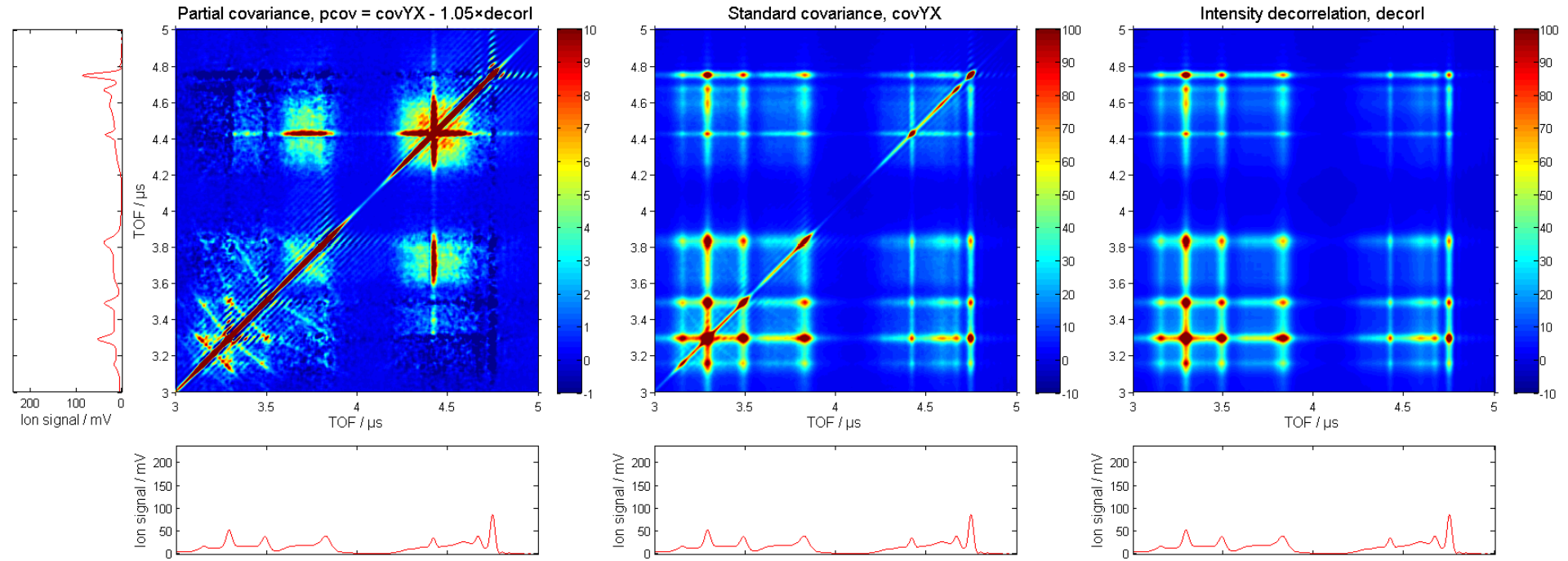
Solution: partial covariance mapping

("partial" = the part not induced by intensity fluctuations)

From W J Krzanowski "Principles of Multivariate Analysis", Clarendon Press, Oxford 1990, page 428:

$$\mathrm{pcov}(\mathbf{Y},\mathbf{X}) = \langle \mathbf{Y}\mathbf{X} \rangle - \langle \mathbf{Y} \rangle \langle \mathbf{X} \rangle - \frac{\left(\langle I\,\mathbf{Y} \rangle - \langle I \rangle \langle \mathbf{Y} \rangle\right)\left(\langle I\,\mathbf{X} \rangle - \langle I \rangle \langle \mathbf{X} \rangle\right)}{\langle I^2 \rangle - \langle I \rangle^2}$$

where $I = I_n$ – laser intensity at shot n



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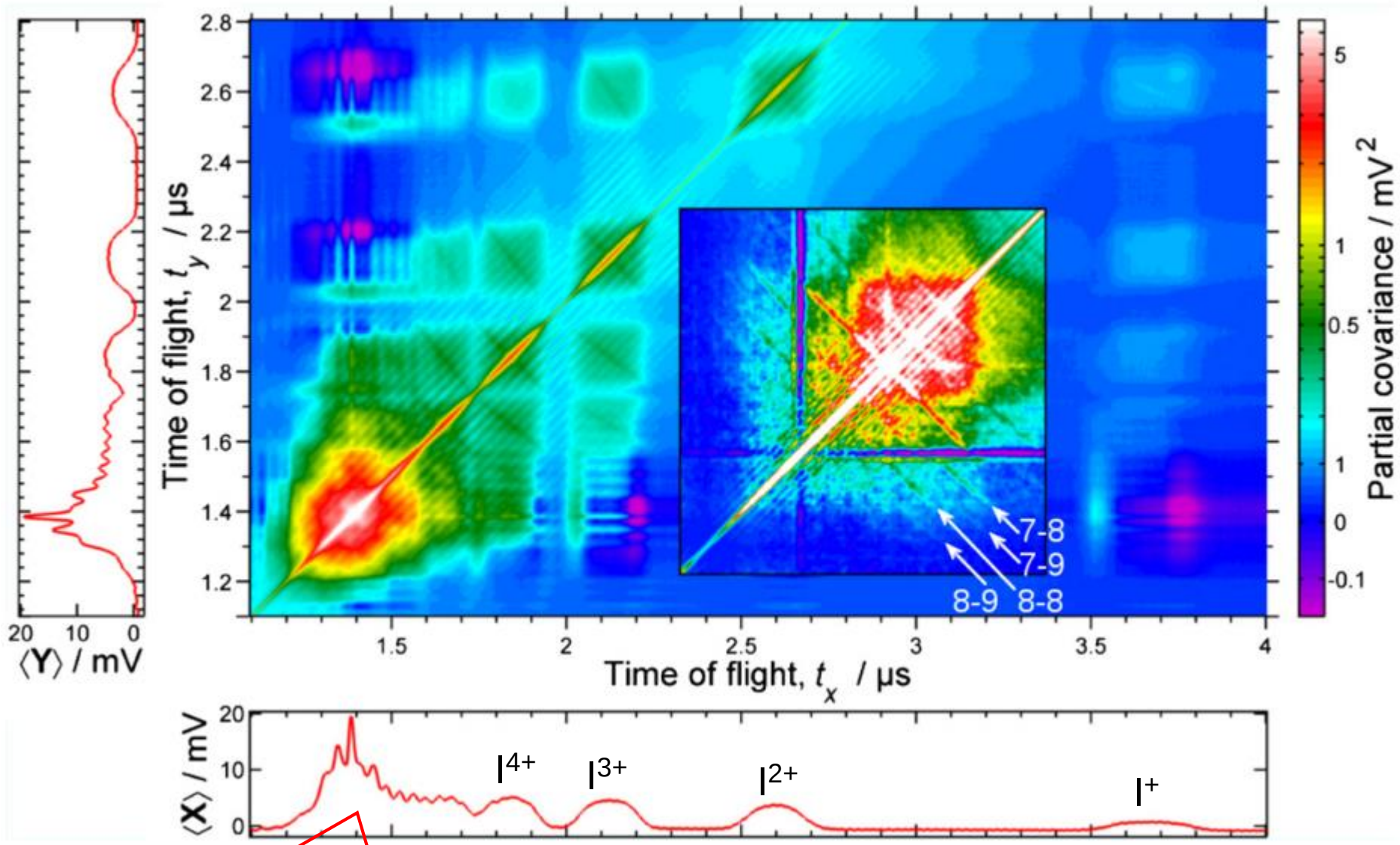
Coulomb explosion of I_2



$q \geq 5$ unresolved here

Coulomb explosion of diatomic molecules in intense XUV fields mapped by partial covariance O. Kornilov *et al, J. Phys. B* **46** 164028 (13 August 2013)

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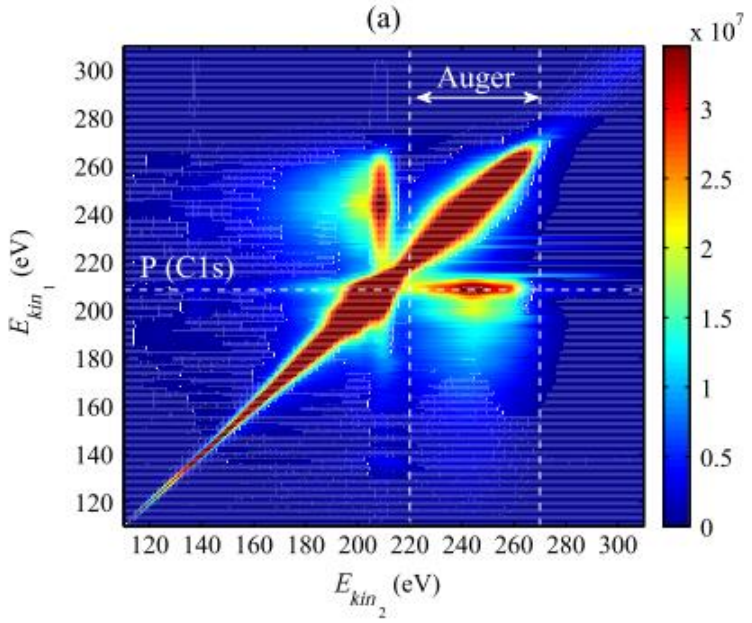
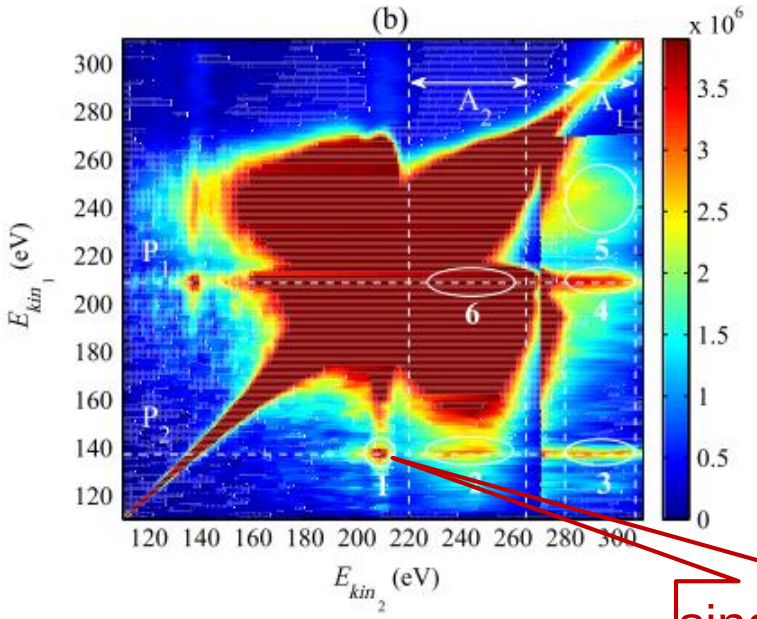
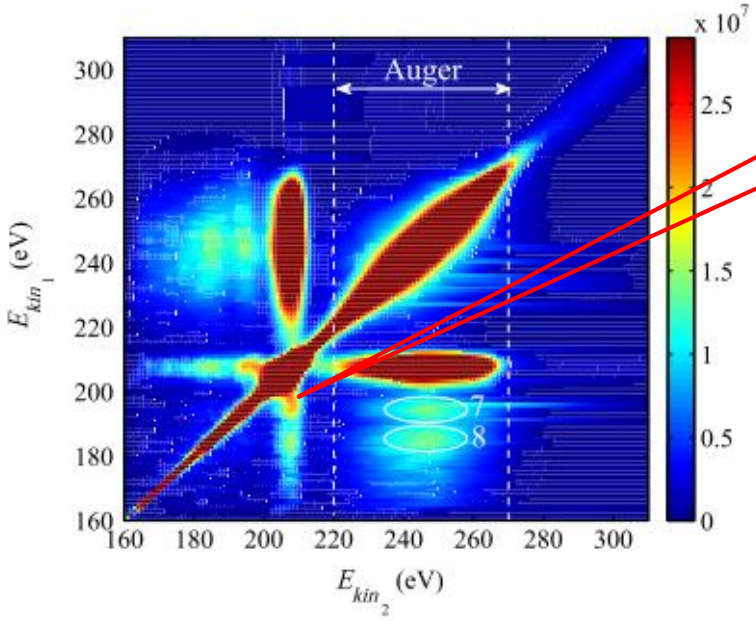
Polyatomics: double core holes in C_2H_2 and C_2H_6





single-site DCH



two-site DCH

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PAPER

Covariance mapping of two-photon double core hole states in C_2H_2 and C_2H_6 produced by an x-ray free electron laser

M Mucke[1], V Zhaunerchyk[1,2,3], L J Frasinski[4], R J Squibb[1,2,4], M Siano[4], J H D Eland[1,5], P Linusson[3], P Salén[3], P v d Meulen[3], R D Thomas[3], M Larsson[3], L Foucar[6,7], J Ullrich[8,9], K Motomura[10], S Mondal[10], K Ueda[10], T Osipov[11], L Fang[11,12], B F Murphy[11], N Berrah[11,13], C Bostedt[14], J D Bozek[14,15], S Schorb[14], M Messerschmidt[14,16], J M Glownia[14], J P Cryan[14], R N Coffee[14], O Takahashi[17], S Wada[18], M N Piancastelli[1,19], R Richter[20], K C Prince[20] and R Feifel[1,2,21]

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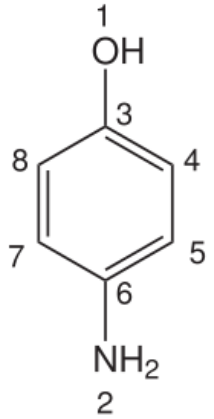
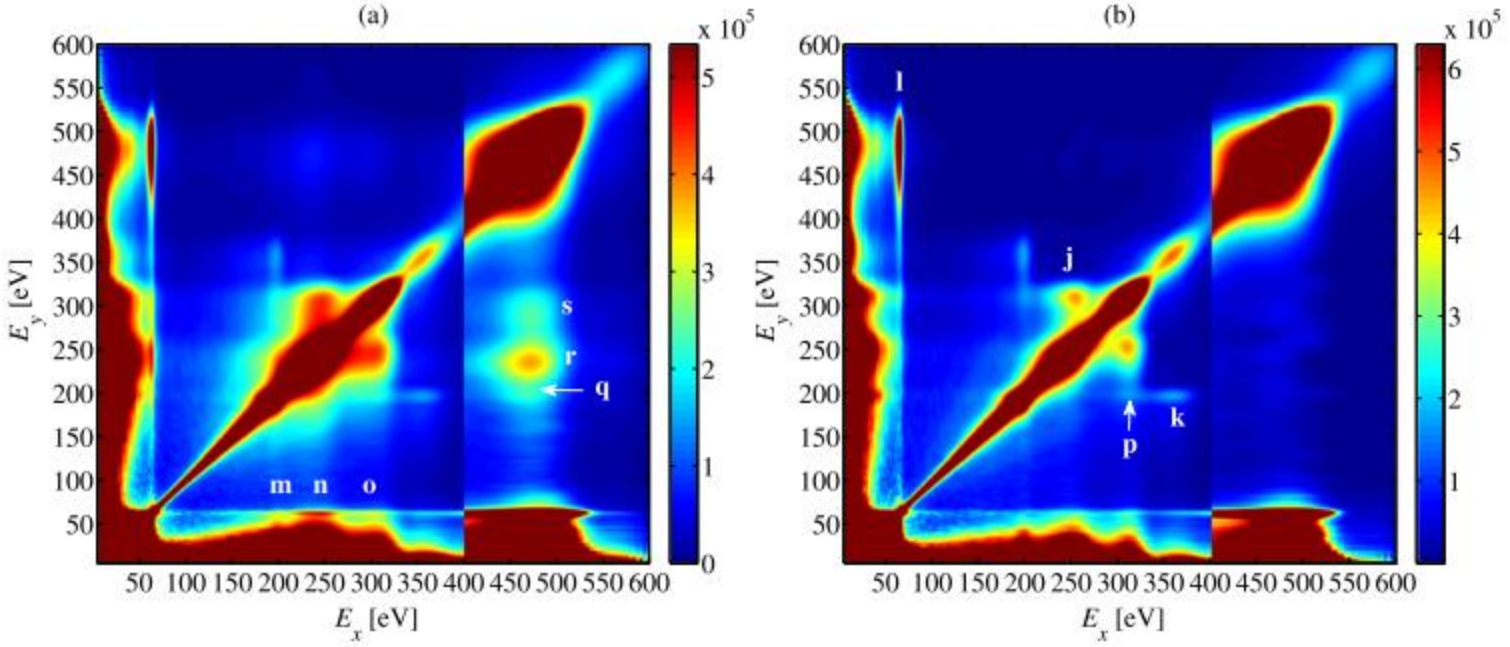
Aminophenol studies at LCLS

IOP Publishing
Journal of Physics B: Atomic, Molecular and Optical Physics
J. Phys. B: At. Mol. Opt. Phys. **48** (2015) 244003 (9pp)
doi:10.1088/0953-4075/48/24/244003

Disentangling formation of multiple-core holes in aminophenol molecules exposed to bright X-FEL radiation



V Zhaunerchyk[1,2], M Kamińska[2,3,4], M Mucke[2], R J Squibb[1,2,5], J H D Eland[1,2,6], M N Piancastelli[2,7], L J Frasinski[5], J Grilj[8], M Koch[8,9], B K McFarland[8], E Sistrunk[8], M Gühr[8], R N Coffee[8], C Bostedt[8], J D Bozek[8], P Salén[3], P v d Meulen[3], P Linusson[3], R D Thomas[3], M Larsson[3], L Foucar[10], J Ullrich[11,12], K Motomura[13], S Mondal[13], K Ueda[13], R Richter[14], K C Prince[14], O Takahashi[14], T Osipov[16], L Fang[16], B F Murphy[16], N Berrah[16,17] and R Feifel[1,2]



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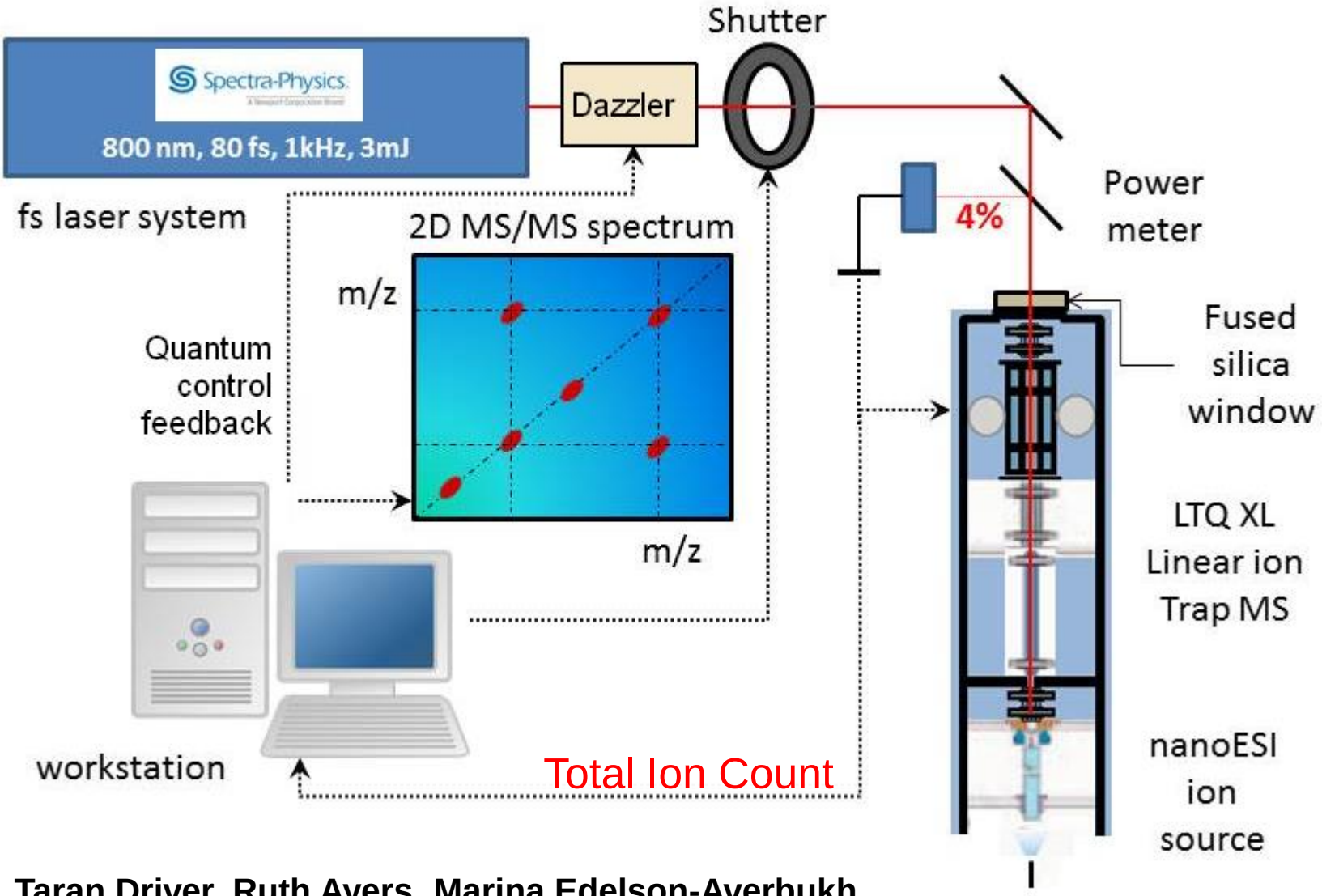
Mass spectrometry of peptides



Taran Driver, Ruth Ayers, Marina Edelson-Averbukh

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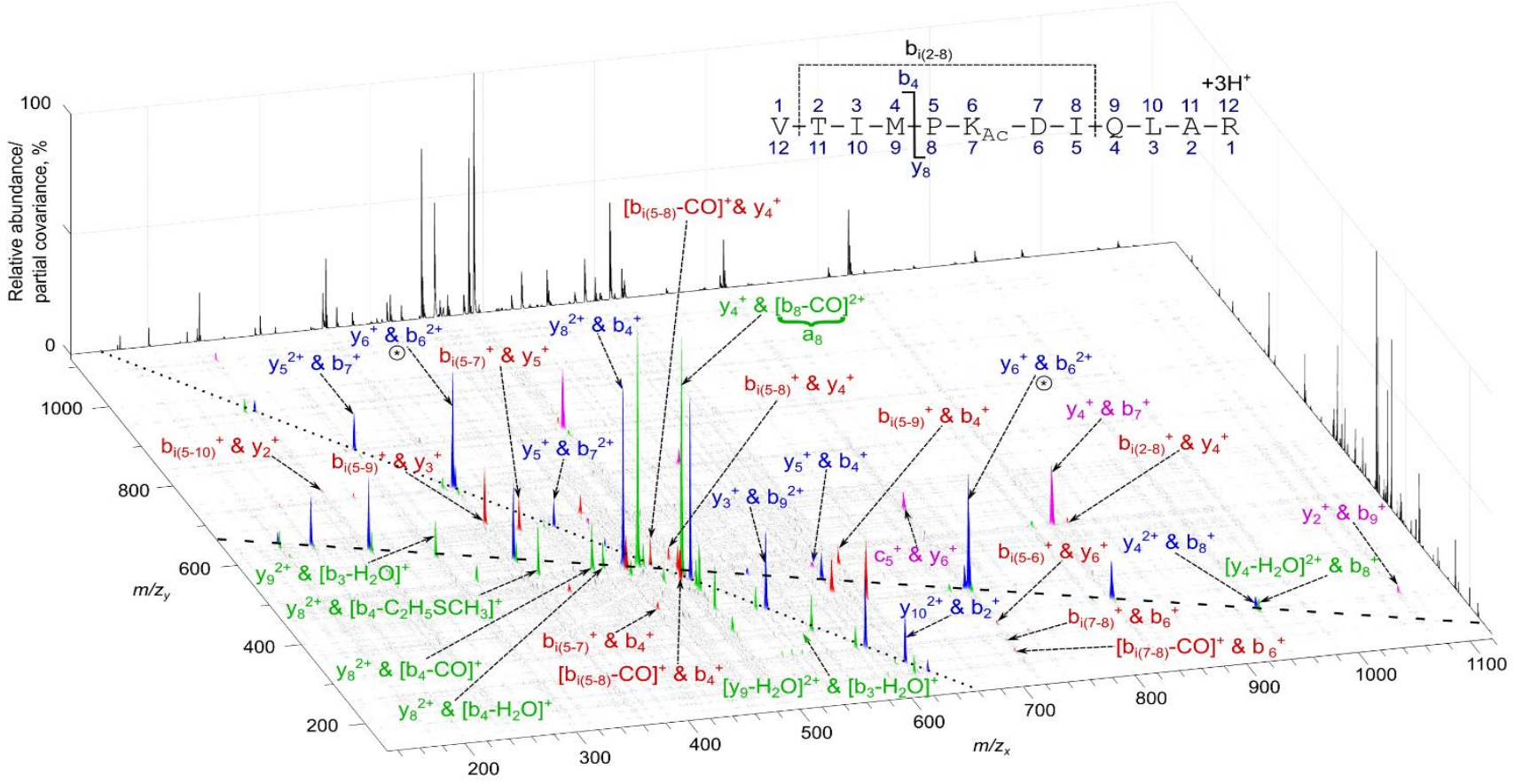
Works beautifully even without the laser!

Two-dimensional mass spectrometry of large molecules based on fragment correlations

Taran Driver[1], Bridgette Cooper[1], Ruth Ayers[1], Rüdiger Pipkorn[2], Serguei Patchkovskii[3], Vitali Averbukh[1], David R. Klug[4], Jon P. Marangos[1], Leszek J. Frasinski[1], and Marina Edelson-Averbukh[1*]

submitted to PRX



International patent **WO 2018/051120**