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Partial covariance mapping techniques at FELs[1]

LESZEK FRASINSKI, Imperial College London

The development of free-electron lasers (FELs) is driven by the desire to access the structure and chemical dynamics of biomolecules with atomic resolution. Short, intense FEL pulses have the potential to record x-ray diffraction images before the molecular structure is destroyed by radiation damage [1]. However, even during the shortest, few-femtosecond pulses currently available, there are some significant changes induced by massive ionisation and onset of Coulomb explosion. To interpret the diffraction images it is vital to gain insight into the electronic and nuclear dynamics during multiple core and valence ionisations that compete with Auger cascades. This paper focuses on a technique that is capable to probe these processes. The covariance mapping technique [2] is well suited to the high intensity and low repetition rate of FEL pulses. While the multitude of charges ejected at each pulse overwhelm conventional coincidence methods [3], an improved technique of *partial* covariance mapping can cope with hundreds of photoelectrons [4] or photoions [5] detected at each FEL shot. The technique, however, often reveals spurious, uninteresting correlations that spoil the maps. This work will discuss the strengths and limitations of various forms of covariance mapping techniques. Quantitative information extracted from the maps will be linked to theoretical modelling of ionisation and fragmentation paths. Special attention will be given to critical experimental parameters, such as counting rate, FEL intensity fluctuations, vacuum impurities or detector efficiency and nonlinearities. Methods of assessing and optimising signal-to-noise ratio will be described. Emphasis will be put on possible future developments such as multidimensional covariance mapping, compensation for various experimental instabilities and improvements in the detector response.

[1] R. Neutze, R. Wouts, D. van der Spoel, E. Weckert and J. Hajdu, *Nature (London)*, 2000, **406**, 752.
[2] L. J. Frasinski, K. Codling and P. A. Hatherly, *Science*, 1989, **246**, 1029.
[3] J. H. D. Eland, *Adv. Chem. Phys.*, 2009, **141**, 103.
[4] L. J. Frasinski, *et al.*, *Phys. Rev. Lett.*, 2013, **111**, 073002.
[5] O. Kornilov, *et al.*, *J. Phys. B: At. Mol. Opt. Phys.*, 2013, **46**, 164028.

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