Potential of X-ray spectroscopy methods with pulsed sources

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The stochastic nature of self-amplified spontaneous emission (SASE) beams available at 4th generation synchrotron sources can be used as an advantage to map simultaneously the unoccupied and occupied electronic states of a scattering atom [1]. The spectral distribution of the SASE pulses incident on the sample and the corresponding X-ray emission spectra for a series of X-ray Free Electron Laser (XEFL) pulses allows unveiling the dependence of the X-ray emission spectra on the energy of the incident X-ray photons. As a consequence, nonlinear X-ray processes in the vicinity of an ionization threshold can be investigated, allowing the expansion of the scope of two-dimensional spectroscopy techniques available at XFELs beyond the applications of resonant X-ray emission spectroscopy at synchrotron radiation facilities. The discrimination of intensity-induced X-ray transparency towards opacity due to a few-eV variation of the incident photon energy will be discussed and attributed to the sequential ionization and excitation of atomic states with femtosecond lifetimes. The presented methodology for measuring the unoccupied and occupied electronic states of atoms in a single experiment allows the use of the entire properties of XFELs, in particular effectively exploiting the most intense and the shortest pulse duration available to ultimately realize nonlinear studies on the electronic structure of a system. The ability to reconstruct RXES maps using XFEL SASE pulses allows exploratory experiments and positioning spectroscopy at XFELs as a driving force towards a fundamental understanding of the interaction between matter and intense X-rays. Finally, limits on the minimal number of X-ray pulses requested for 2-dimensional X-ray spectroscopy are estimated [2] and supported with experimental evaluation.

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References

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2. W. Błachucki et al., Appl. Sci., 11, (2021) 8775.