DESCRIPTION FOR THE GENERAL PUBLIC

Linear X-ray spectroscopy, such as absorption or emission spectroscopies, relies on weak-limit interaction of X-rays with matter. In such a regime, the strength of X-ray signals is correlated in a linear way and the measurement is performed in a one-photon-in and one-photon-out manner. However, for strong enough X-ray fields, the nonlinear regime may be accessed where the X-ray signals are not correlated anymore by linear dependence and the spectroscopic measurements are based on multi-photons-in multi-photons-out processes. Such a nonlinear regime, since many years available at optical frequencies[1-4], at X-ray wavelengths was remaining as unexplored area due to the lack of strong enough X-ray sources. Ability to access nonlinear light-matter interactions at X-ray wavelengths become possible only recently thanks to the development of X-ray Free Electron Lasers (XFELs)[5-6]. In contrast to the optical laser wavelengths, the photon-atom interaction at hard X-ray energies involves bound core-electrons and leads to the intermediate electronic states with sub-femtosecond lifetimes[7-10]. The femtosecond X-ray lasers allow thus to access an uninvestigated area of physics and to probe the physical mechanisms that drives to the nonlinear interaction of X-rays with matter[11-13].

In application at hard X-ray energies, the two-photon absorption (TPA) mechanism will allow accessing different excitation states of matter thanks to the nature of electronic transitions. Comparing to X-ray linear regime with one photon absorption (OPA) process which is determined by dipole-allowed transitions, the TPA process requests changing the electron quantum number by +/-2 or 0, allowing thus to access a quadrupole or forbidden excitations. This may lead to creation of new and unexplored states of matter. However to this point, the real application of TPA at XFELs is still questionable. Because of low cross-sections, the TPA process is competing with other first- and second-order photon-atom interactions induced in the sample during the course of ultra-short X-ray pulse.

The project aims at a combination of X-ray spectroscopy and theory to study TPA mechanisms in the hard X-ray regime. The experimental data from XFEL experiments will be used to determine the cross-sections for TPA processes in different materials as well as different material structures (like molecules or nanoparticles). We intend to probe the dependence on excitation energy of TPA signal trying to identify the regimes at which this process become dominant. Moreover, thanks to available high X-ray fluence at LCLS XFEL, we intend to disentangle two possible paths of TPA mechanism: sequential and simultaneous ionization. While at low X-ray fluence the sequential absorption of two X-rays dominates in TPA process, the higher X-ray fluence may allow for detection of simultaneous two-photon absorption. Establishing relative cross-sections for sequential and simultaneous two-photon absorption will allow for direct tests of quantum theorem describing interaction of X-ray radiation with bound-electrons.

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