X-ray spectroscopy of nonlinear interactions of X-rays with matter

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X-rays have long been used to explore the electronic and structural properties of all forms of matter, using sources as varied as X-ray tubes to accelerator-based storage rings. X-ray methods have evolved over decades to become specialized tools for a broad range of investigations, with techniques ranging from X-ray scattering through X-ray spectroscopy to X-ray tomography. In general these methods all rely on X-ray measurements that depend linearly on the number of incident X-ray photons. With the advent of X-ray free electron lasers (XFELs), the ability to reach extremely high photon numbers in ultrashort pulse durations has resulted in a paradigm shift in our ability to observe nonlinear X-ray signals. This enormous increase in peak power (pulse energy/pulse duration) has been a double-edged sword, with new and exciting techniques being developed but at the same time well-established techniques proving unreliable [1-3]. This requires a fundamental change in our approach to X-ray science at FELs, since this nonlinear regime is a largely unexplored area, making it hard to predict not only when to expect nonlinear contributions to a measurement, but also to understand the very nature of this response [4-7].

We will report on application of X-ray spectroscopy methods in a regime where the penetrating properties of matter become inverted. The deliberate use of stochastic X-ray pulses in resonant X-ray emission spectroscopy is demonstrated to allow for simultaneous mapping of the electron and hole states of atoms. The discrimination of intensity-induced X-ray transparency towards opacity due to a few-eV variation of the incident photon energy around ionization threshold will be discussed and attributed to the sequential ionization and excitation of atomic states with femtosecond lifetimes. Finally, interaction of electrons generated in solvent with metal-centre molecule will be discussed and evaluated by means of X-ray emission spectroscopy at above edge ionization energies.

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