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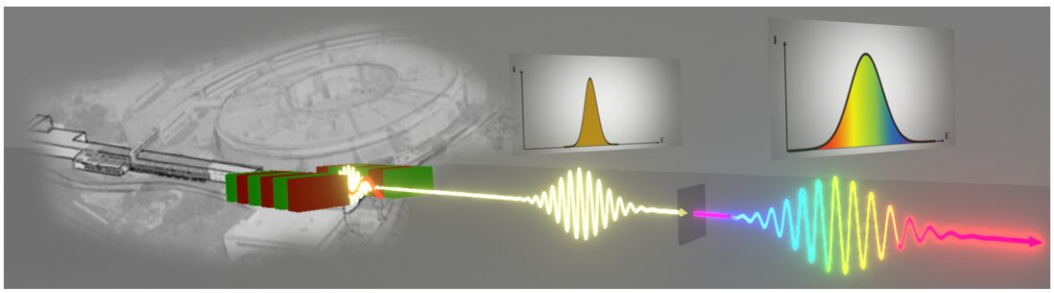
Non-linear optics meets X-rays

May 13 2021



An illustrative scenario of the observed effect is shown. The pulses with a subpicosecond duration pulsed in the soft X-ray generated from FERMI@elettra interacts with a submicrometric metallic foil. The non-linear optical interaction generates a modification of chromatic components in the pulses. Credit: Carino Ferrante, Emiliano Principi, Andrea Marini, Giovanni Batignani, Giuseppe Fumero, Alessandra Virga, Laura Foglia, Riccardo Mincigrucci, Alberto Simoncig, Carlo Spezzani, Claudio Masciovecchio, Tullio Scopigno.

The recent advent of femtosecond X-ray sources offers unprecedented opportunities for structural and dynamical studies. It requires, however, manipulating spectral properties, as commonly done by non-linear optics at visible/infrared wavelengths. Here we show the first evidence for Self-Phase Modulation, a key non-linear effect in ultrafast laser science, in soft X-Rays. Building on such an effect, we demonstrate how to tune spectral properties in this wavelength region critical for core electron pump-probe spectroscopy and nanoimaging.

The relevance for radiology applications is probably the most well-known advantage of X-ray beams (keV energies) with respect to visible radiation (eV energies) and can be traced back to their superior penetration depth. On a more fundamental point, however, the relevance of this photon energy range relies on the capability of probing inner shell electrons (as they have comparable binding energies) and mapping molecular structures on the atomic-scale (as typical interatomic spacings are comparable to X-ray wavelengths). Building on such capabilities, efforts have been devoted by the scientific community to develop X-ray sub-picosecond sources able to access matter properties with a time resolution sufficient to access elemental molecular motions. Free electron lasers (FEL), nowadays available at several large-scale facilities around the world, represent a prime candidate to generate femtosecond X-ray pulses with high brilliance. One of the main challenges to exploit the enormous potential of FEL sources is developing methods for tuning the spectral and temporal beam properties, a task which is customarily achieved at visible wavelengths resorting to non-linear optics.

In a new paper published in *Light Science & Applications,* a team of scientists from the Italian Institute of Technology, University of L'Aquila, FERMI Trieste and "Sapienza" University of Rome have shown the first evidence of self-phase modulation (SPM) in the soft X-ray regime. The experiment, performed in the facility FERMI@elettra of Trieste, consists in the observation of spectral modulation after the interaction of focused FEL beams with a very thin metallic foil (100-300 nm).

"Our experiment demonstrates a new control knob for spectral shaping of FEL pulses. Blue to red shift accompanied by bandwidth increase can be obtained by moving the input wavelength across the material's absorption edge," prof. Tullio Scopigno explains.

The atomic absorption edges in the X-ray region feature sharp discontinuities: an optical transparent material can absorb light modifying the photon energy by less than 1%, correspondingly generating specific core electron excitations.

"This first observation of SPM effects in the soft X-Ray regime allows to unveil specific atomic properties on the subpicosecond time scale. In particular, the interplay with a light-induced out-of-equilibrium electron plasma generated on the femtosecond timescale in thin metallic foils," concludes Dr. Carino Ferrante.

Below the absorption edge, the observed SPM is induced by the Kerr effect, i.e. by a modification of the non-linear refractive index mimicking the pulse intensity profile, which ultimately results in spectral broadening, accompanied by a redshift due to valence electrons heating. In striking difference, above edge, the highly excited core photoelectrons generated by the pulse leading edge form a transient hot dense ionized plasma, responsible for a sharp decrease of the refractive index. Consequently, the pulse trailing edge is accelerated giving rise to an asymmetric temporal compression which, in turn, results in a blueshift.

The results provide a proof of concept for spectral shaping of soft X-ray pulses, a key milestone towards the development of new protocols for femtosecond core electrons spectroscopies.

More information: Carino Ferrante et al, Non-linear self-driven spectral tuning of Extreme Ultraviolet Femtosecond Pulses in monoatomic materials, *Light: Science & Applications* (2021). DOI:

10.1038/s41377-021-00531-8

Provided by Chinese Academy of Sciences