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Editorial

Special issue on experimental and theoretical extreme ultraviolet to x-ray non-linear methods

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The advent of the laser in the early 1960s represented a revolution in Optics and Spectroscopy, as it also enabled the birth of non-linear (NL) optics thanks to the intense and coherent nature of the laser beams. The rapid development of a high diversity of NL optical methods had a deep impact in science that was recognized by the 1981 and 2018 Physics and the 1999 Chemistry Nobel Prizes. These developments concerned the optical wavelength-domain (ultraviolet, visible, infrared and terahertz), and led to the birth of photonics and optoelectronics, along with a myriad of new analytical and imaging techniques [1]. NL optics and spectroscopy have also deeply advanced our understanding of fundamental processes in atomic, molecular and condensed matter systems, but also of surface and interface physics with striking applications to technology. This was particularly felt after the birth of femtosecond (fs) spectroscopy in the second half of the 1980s and was later extended into the attosecond regime highlighted by the Physics Nobel prize 2023 [2].

In parallel to these developments, there was the awareness that optical techniques view the dynamics through their projection on the dipole moment that is typically generated by valence electron polarizations, which means that they are subject to selection rules, involve delocalized and often unspecific states, and therefore provide an indirect and partial picture of the dynamics. A more direct and complete observation of nuclear motions in molecules, liquids, solid materials and biological systems is achieved through the combination of the high temporal resolution of femtosecond laser spectroscopy, with the atomic scale spatial resolution of structural probes such as x-ray or electron diffraction and x-ray spectroscopy, which look directly at the motions of charges localized around atomic nuclei [3]. This lead to a major development of time-resolved x-ray techniques starting from the early 2000s, which experienced a major leap forward with the advent of x-ray free electron lasers (XFELs) [4].

With their high peak power XFELs have also enabled x-ray NL optics and spectroscopy, just like the optical laser did over 60 years ago. Indeed, starting from the launch of the XFELs around 2010, and more recently, with the advent of table-top sources of extreme ultraviolet (EUV) to soft x-ray sources based on the principle of high harmonic generation (HHG) [5], we have witnessed several breakthrough experiments demonstrating x-ray NL phenomena, such as multiphoton absorption, saturated absorption, stimulated x-ray Raman and emission, second harmonic generation, sum-frequency generation and four-wave mixing [6]. Following these breakthrough experiments, still much needs to be

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done to consolidate the methods, firmly establish their observables and widen their scope in fundamental science and in applications. In this respect, an open, international network, WavemiX, was established in 2020 in order to consolidate and share the know-how of such complex experiment and foster the development of novel theoretical methods and approaches. The network also contributes a large share to the more recent COST action NEXT: 'An international network for NL EUV to hard x-ray techniques' (CA22148, https://ca-next.eu/) with a similar focus. This COST action organizes regular workshops, contributes to training schools and largely aims at fostering the exchange and education around the novel methodologies enabled through x-ray NL processes.

The contents of this special issue represent a snapshot of recent developments in the extremely fast-moving area of NL Science at high photon energies. It features a collection of articles highlighting techniques, methods, and scientific advancements—both experimental and theoretical—in the fields of atomic and molecular physics, chemistry, and materials science, enabled by the use of ultrafast nonlinear x-ray tools, as well as some technical developments. It contains articles showing how the ultrashort x-ray pulses can be used to monitor the evolution of electronic and structural dynamics in solid materials [7–10] or can be used to trigger changes therein [11]. This issue presents several applications of NL x-ray techniques, with a particular focus on four-wave mixing methods, especially transient grating (TG) spectroscopy from the EUV to the Hard x-ray range [8–10, 12]. This approach stands out as the most widely used due to its ability and potential, illustrated here, to probe transport phenomena and diffusion at nano- and mesoscopic scales [8–10], and dynamics of chiral systems [12]. TG spectroscopy relies on the generation of at least two identical EUV/x-ray pulses and an optical probe [8], or three ultrashort EUV/x-ray pulses of which two are identical [10]. The delivery of multiple and multicolor x-ray pulses is, as a consequence, a central issue, which is discussed in [9], and is applicable to the other major sources of EUV pulses, such as HHG sources [13]. Moreover, second-order processes, which are extremely important for surface and interface science, are also addressed in this special issue, showcasing novel approaches to performing such experiments at high photon energies [14]. Finally, the issue showcases the investigation of electronic and ionization dynamics in Neon triggered by NL EUV/x-ray processes [15–17].

Undoubtedly, the advent of XFELs and HHG sources is heralding the birth of NL core-level spectroscopies, with the advantage of element-selectivity, atomic-scale structural resolution and large momentum transfers to the system, offering unique opportunities to interrogate the fundamental processes in matter. The guest editors share the confident belief that the works presented in this special issue capture a snapshot of the present state of the art of this dynamic area of science and, that readers will be invigorated in pursuing future research by the contributions selected herein. The guest editors are grateful to all contributors and referees, and warmly thank Dillon Eastoe for his support and his guidance.

The Guest Editors: Martin Beye, Majed Chergui, Claudio Masciovecchio, Cristian Svetina

Data availability statement

No new data were created or analysed in this study.

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