Novel ultrafast structured EUV/x-ray sources from nonlinear optics

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Abstract. Coherent extreme-ultraviolet (EUV)/x-ray laser sources, structured in their temporal/spectral, spatial and angular momentum properties are emerging as unique tools to probe the nanoworld. One of the key ingredients for the emergence of such sources is the extraordinary coherence in the up-conversion of infrared laser sources through the highly nonlinear process of high-order harmonic generation. In this contribution we will review the advances during the last decade that led to the generation of structured EUV/x-ray sources, such as circularly polarized attosecond pulses, harmonic vortices with time-varying orbital angular momentum, ultrafast vector and vector/vortex beams, tunable high-order harmonic combs or attosecond pulse trains with time-dependent polarization states. The use of such sources is being already applied to the investigation of chiral matter or magnetic materials. In the latter case, structured ultrafast sources are very promising to achieve a complete understanding of the electronic and spin interactions that govern sub-femtosecond magnetization dynamics.

The development of structured ultrafast laser sources is a key ingredient to advance our knowledge about the fundamental dynamics of electronic and spin processes in matter. In particular, it has been widely recognized the relevance of ultrafast sources structured in their spin angular momentum (SAM, associated to the polarization of light) and orbital angular momentum (OAM, associated with the transverse phase profile, or vorticity of a light beam) to study chiral systems and magnetic materials in their fundamental temporal and spatial scales. In that scenario, structured coherent extreme-ultraviolet (EUV)/soft x-ray pulses are emerging thanks to the highly nonlinear process of high harmonic generation (HHG).

HHG is among the most extreme nonlinear up-conversion processes to date, and it can be semiclassically understood through the so-called three-step model. An intense, infrared laser field is focused into an atomic or molecular target, and an electronic wavepacket is detached from its parent ion through tunnel ionization. Afterwards, the electronic wavepacket acquires kinetic energy from the laser field, and due to the oscillatory behavior of the later one, it recollides with its parent atom, releasing the kinetic energy as high frequency radiation. The emitted radiation spectrum is composed of harmonics of the infrared driving field, that can extend towards the soft x-rays [1], being temporally emitted as attosecond pulses [2]. In the last decade, HHG in solid systems is being also investigated, where an analogy with the three-step model can be found. In HHG, some of the driving laser beam properties are imprinted on the dynamics of the radiating electron, and, in turn, on the emitted EUV/soft X-ray light, making it a very versatile process to engineer the temporal, spectral and spatial properties of high-harmonic and attosecond pulses.

In this contribution we review our recent work in the generation of structured laser pulses at the attosecond scale. Several works that revealed the mapping properties of SAM and OAM in the HHG process were crucial. First, circularly polarized attosecond pulses were obtained by using non-trivial geometries of the driving field [3-5]. Second, high harmonic vortices with high topological charge can be obtained through OAM conservation [6-8]. These two achievements over the SAM and OAM of high-order harmonic pulses, that resulted from several works from of many research groups, paved the way towards more sophisticated structured high harmonic and attosecond pulses: high-harmonic pulses with both OAM and SAM [9], high-harmonic vortices with time-dependent OAM or self-torque [10], attosecond pulses with time-dependent polarization states [11], or EUV vector and vector/vortex beams with spatially varying polarization states [12,13], among others.

Very recently, the proper use of OAM driving fields has allowed to achieve precise coherent control over the frequency content of high-harmonic pulses—a key property to perform high-harmonic spectroscopy. In particular, by harnessing OAM conservation in HHG, we can generate a transverse necklace-shaped spatial phased array of harmonic emitters that allow us to tune the line spacing and divergence of the emitted harmonic combs [14] (see Fig. 1). Our theoretical and experimental results

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show that the resulting on-axis HHG emission is composed of harmonics whose frequency separation can be controlled through the OAM of the driving field, extending towards the soft X-rays. In addition, such harmonic radiation presents extremely low divergence, well below that obtained when using Gaussian driving beams, which further decreases with the harmonic order. This work provides a new degree of freedom for the design of harmonic combs—particularly in the soft X-ray regime, where very limited options are available.

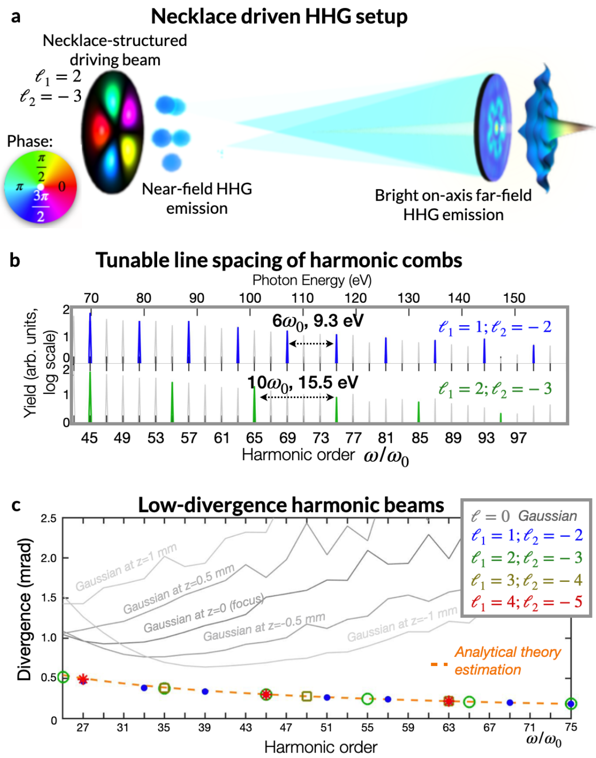

Fig. 1. a) Generation scheme of phased-necklace HHG, where the driving field is composed of two collinear linearly polarized vortex beams with opposite—and different—OAM content (ℓ_1, ℓ_2) and same frequency (ω_0). b) Tunability of the on-axis HHG spectra in He driven by a 800 nm field. The line spacing can be tuned through the OAM content of the driving fields. Panel c) shows the extremely low divergence of the on-axis harmonic beams. Remarkably, the divergence decreases with frequency [14].

Another way to structure ultrafast light pulses is to use a structured material. For example, HHG in an anisotropic material [15], such as graphene, possess the potential to structure the intensity and polarization state of light beams. Another recent proposal consists in the interaction of ultrafast azimuthally polarized beams with metals, where a large current oscillates at the laser frequency, generating ultrafast, intense longitudinal magnetic fields isolated from the electric fields [16] (see Fig. 2). Such structured sources offer an appealing alternative to study sub-femtosecond magnetization dynamics, where a complete understanding of the electronic and spin interactions remains unexplored.

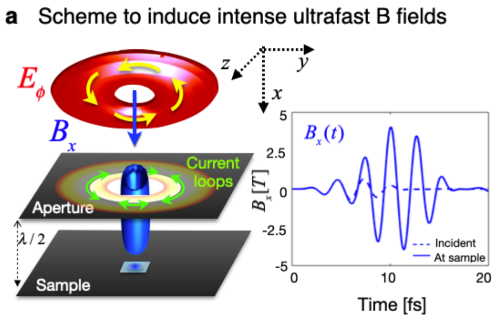

Fig. 2. Enhanced ultrafast magnetic field (blue) at a metal sample irradiated by an infrared femtosecond azimuthally polarized beam [16].

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