Fourier-limited attosecond pulse generation with magnetically pumped high-order harmonic generation

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Abstract. After more than two decades of attosecond physics, the generation and control of the shortest laser pulses available remains as a complex task. One of the main limitations of reducing the temporal duration of attosecond pulses emitted from high-order harmonic generation (HHG) is the *attochirp*. In this contribution, we demonstrate that HHG assisted by strong fast oscillating magnetic fields enables the generation of Fourier-limited attosecond pulses in the water window. In short, the magnetic field generates a nanowire-like structure, which transversally confines the electronic wavefunction in the HHG process. We demonstrate that the resulting HHG spectrum extends well beyond the semiclassical cutoff frequency, and most interestingly, it is emitted in the form of few-cycle, Fourier-limited, attosecond pulses.

1 Introduction

High-order harmonic generation (HHG) is an extreme non-linear process resulting from the non-perturbative interaction between matter and intense laser fields. In the standard picture, using a gas target and a linearly polarized laser, the harmonic generation stems from the recollision of previously ionized electrons with their parent ions. During its excursion along the continuum, the electron gains kinetic energy, that is subsequently radiated in the form of high-order harmonics of the incident field. In this simple approach, the detached electron behaves as a free particle following classical trajectories along the longitudinal direction, from the ionization time until the recollision [1]. The electron's excursion along the continuum is essential to build the main properties of HHG, and several approaches have been studied to control the electron trajectories in order to modify the HHG process. For example, the use of multicolor drivers, complex polarization schemes, or external magnetic fields [2]. In the latter case, however, only static magnetic fields have been considered, to the best of our knowledge, as in principle strong oscillating magnetic fields are only present in electromagnetic waves. In this latter case, the force exerted by an electric field is orders of magnitude over the magnetic field, therefore hindering any effect of

On the one hand, one of the major interests of HHG is its ability to generate attosecond pulses. Attosecond pulse generation stands as one of the most robust tools to probe ultrafast processes at molecular and atomic scales. The extension of pump-probe experiments to these timescales allows for time-resolved observation of electronic dynamics, for example, in chemical reactions. Moreover, they are considered the main tool to understand, and

ultimately control, charge migration in molecular systems, with huge implications for understanding the alteration of biological macromolecules, the development of more efficient photovoltaic systems, or more selective catalysts [3,4]. HHG stands as the standard tool for attosecond pulse generation thanks to the broad spectra generated towards the XUV or soft X-rays. Counterintuitively, the shortest duration of an HHG attosecond pulse is not limited by its bandwidth [5], but by its intrinsic chirp, also known as attochirp [6]. By using metallic foils, the attochirp can be partially compensated, reaching the record established at 43 as [7].

On the other hand, recently there has been an increasing interest and a huge development of structured light beams [8], opening new perspectives on light-matter interactions. For example, it has been shown how azimuthally polarized vector beams exhibit an isolated Tesla-scale femtosecond magnetic field on-axis oscillating at optical frequencies, paving the way to pure magnetic interaction with matter [9]. Isolated fast oscillating magnetic fields are of great interest in fields such as ultrafast magnetism or spintronics [10], but also in ultrafast and non-linear optics.

In this contribution, we perform theoretical simulations demonstrating that HHG assisted by a linearly-polarized magnetic field in coincidence with a circularly-polarized driver laser results in the generation of few-cycle, near Fourier limited, attosecond pulses

2 Model

We solve numerically the three-dimensional, timedependent Schrödinger equation for the hydrogen atom. The Hamiltonian, using atomic units, for the electron under the action of both fields reads:

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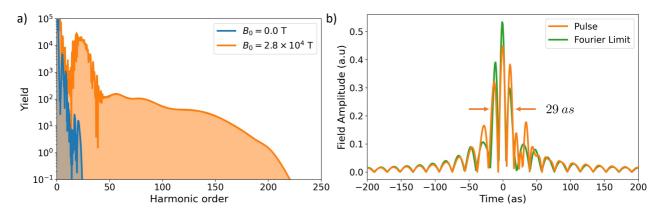


Fig. 1. (a) HHG spectra resulting from a circularly polarized driver laser without magnetic field (blue line) and with the magnetic field linearly-polarized. (b) Attosecond pulse arising from the spectra with the magnetic field in panel a), filtering above the 70th harmonic order (blue line). The measured FWHM pulse duration is approximately 29 as. The Fourier-limited pulse, plotted in green, shows a FWHM around 27 as.

$$\hat{H} = \frac{1}{2} \left[\boldsymbol{p} - \frac{1}{c} \boldsymbol{A}_{E} \right]^{2} + \frac{1}{2c} B(t) \hat{L}_{z} + \frac{1}{8c^{2}} B^{2}(t) (x^{2} + y^{2}) - V(\boldsymbol{r})$$

The magnetic field *B* creates a time-dependent harmonic oscillator potential in the x-y plane, confining the detached photoelectron and its excursion along the continuum, inducing a restructuration of the continuum energy levels in the same fashion as quantum wire. Moreover, note that one of the components of the circularly-polarized electric field will exert a transverse force over the photoelectron inducing transitions to higher states inside the magnetic harmonic oscillator. These highly excited states will contribute to the HHG emission.

3 Results

We consider a four-period circularly polarized electric field at 800 nm with a peak intensity of 1.6×10¹⁴ W/cm2 and a linearly polarized oscillating magnetic field. While in the standard scheme, without a magnetic field, the harmonic yield drops immediately using drivers with circular polarization, we show the generation of broad supercontinuum by adding an external magnetic field with amplitudes around 2.8×10⁴ T, oscillating at 1600 nm (Fig. 1 a). This emission, extending over 200 harmonic orders (110 eV), results from the excitation of the quantum wire transverse states by the circularly-polarized electric field. These highly excited states contribute to the harmonic emission explaining the broad spectrum observed. In fact, we have seen that the electron in the continuum is a semiclassical wave packet, following a well-defined trajectory under electric field driving. The emission yield is maximized when the collision with the parent ion occurs simultaneously in the longitudinal and the transverse directions, highly dependent on the magnetic field wavelength, phase, and amplitude.

By filtering the spectrum above the 70th harmonic order, we see how a 29 as FWHM pulse can be generated nearly Fourier-limited (Fig. 1 b). These attosecond pulses are centered at 6.7 nm (27 as period), meaning they are

almost single cycle. To our knowledge, this is the first proposal to generate chirp-free HHG attosecond pulses. Thus, in this scenario, attosecond pulse generation is not limited by the use of negative GDD metallic foils, difficult to use at these wavelengths, and could be extended for broader spectra. More interestingly, by using longer wavelengths [5], we foresee that broader chirp-free spectra could be obtained, with the potential to reach fewcycle, few attosecond pulses.

We note that such isolated linearly-polarized magnetic fields could be obtained from structured Petawatt laser beams, with laser intensities around 10¹⁸ W/cm². This work opens the route to new and exotic HHG scenarios in which state-of-the-art Petawatt laser systems may play a determinant role, allowing the generation of nearly Fourier-limited attosecond pulses.

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