

High-order harmonic generation from ultrafast matter Talbot effect

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Abstract. High-order harmonic spectroscopy is a robust method for probing electron dynamics under the influence of a driving field, capturing phenomena as brief as attoseconds. It relies on the extreme non-linear process of high-harmonic generation (HHG), where intense laser pulses are directed at a material, causing it to emit high-energy photons in harmonics of the laser frequency. In this contribution we explore the possibility to generate high-order harmonics from low-dimensional crystalline solids driven under grazing incidence. We demonstrate that, in this unconventional geometry, the electron wavefunction is ejected from the solid and, subsequently, redirected to it to generate harmonics. Most appealingly, we show that the crystal's periodicity imprinted in the electron's wavefunction introduces a revival dynamics closely connected with the matter temporal Talbot effect. These Talbot oscillations are ultrafast ($<$ femtosecond) and leave a distinct signature in the high-frequency harmonic spectrum, in the form of structures extending beyond the main spectral cutoff.

1 Introduction

The utilization of ultrashort laser sources, operating at femtosecond and attosecond scales, presents unparalleled capabilities for manipulating elemental matter constituents at their natural dimensions. A significant aspect of this manipulation lies in controlled interaction geometries that enable precise management of electron dynamics while preserving wavefunction coherence. High-order harmonic generation (HHG) exemplifies this control, where high-frequency emission arises from the coherent superposition of bound and ionized electron wavefunctions [1]. This coherent ultrafast domain offers a promising platform for extending conventional matter-wave coherent optics to subnanometer and subfemtosecond scales, a domain ripe for exploration.

Coherent optics, rooted in phase-preserving light manipulation, underpins various applications across metrology, imaging, and information processing. Notably, the translation of coherent optical phenomena to matter waves has sparked considerable interest, with Talbot self-imaging serving as a practical example. Talbot revivals, stemming from near-field diffraction, manifest as repeated images of an initial periodic

pattern, with implications across diverse fields from image processing to quantum computing.

Recently we have proposed translating matter Talbot interferometry to unprecedented spatial nanometer and temporal subfemtosecond scales, leveraging HHG from periodic crystals as a means to induce electron Talbot effects and realize ultrafast nanometric Talbot-Lau interferometers [2]. This convergence of Talbot interferometry and high-harmonic spectroscopy heralds a new era of ultrafast techniques, offering insights into the coherent dynamics of electronic wavefunctions within their natural spatial and temporal domains. These techniques, capable of generating extreme-ultraviolet and soft x-ray coherent radiation, provide a deeper understanding of electron dynamics in both atomic/molecular gases and crystalline solids, opening avenues for studying band structures, interaction dynamics, and electron coherence with unprecedented precision.

2 Mechanism.

We examine the HHG setup depicted in Figure 1a, where a mid-infrared laser pulse interacts with a periodic structure at grazing incidence. The driving field detaches the electrons from the crystal (t_1), whose

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wavefunction contains transverse modulations due to the crystal's periodicity. The electron evolves detached from the crystal, while accelerated by the driving field and redirected towards the crystal target (t_2). During the excursion, the transverse periodicity of the wavefunction will produce Talbot revivals. At rescattering, the electron will radiate harmonics with an efficiency modulated by the instantaneous Talbot structure (t_3).

The crystal target is described as a periodic chain of ions, with a period of $d = 2.1 \text{ \AA}$ (4 a.u.), chosen to replicate in one dimension the talbot dynamics of the hexagonal lattice forming graphene. Other parameters of our model potential are set to reproduce a work function of 5 eV, akin to graphene. Figure 1b outlines the rescattering process over time, highlighting the temporal Talbot revivals and the resemblance of the rescattering HHG sequence to a Talbot-Lau interferometer configuration. After rescattering, these ultrafast Talbot revivals imprint a distinct signal in the HHG spectrum.

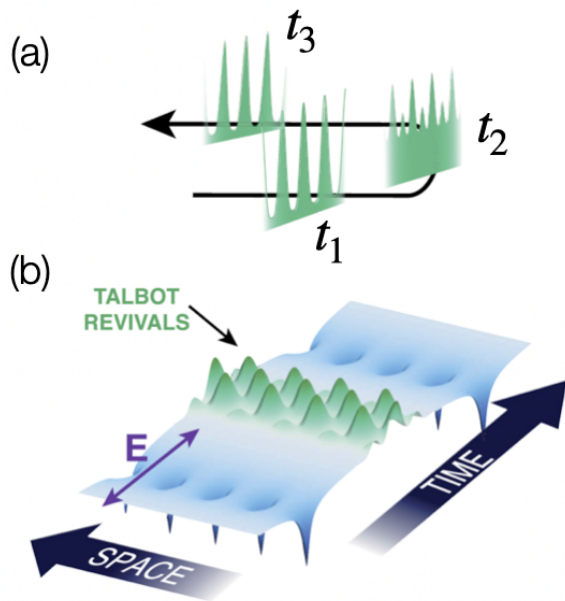


Fig. 1. (a) Scheme of the interaction: a laser is aimed with polarization perpendicular to the crystal surface, detaching an electron (t_1) whose wavefunction is modulated transversely by the crystal potential. The wavepacket is accelerated (t_2) and redirected back to the crystal (t_3), where it generates harmonics. (b) space time evolution of the electron wavefunction, showing the emergence of Talbot revivals.

3 Results

In Figure 2, we observe the harmonic emission generated by an electron under the influence of a $3 \mu\text{m}$,

10^{13} Wcm^{-2} pulse lasting 5 cycles (18 fs FWHM). The blue curve showcases the contribution of a single Bloch state at $k = 0.2g$, whereas the purple curve illustrates the collective contribution of ten states evenly dispersed throughout the Brillouin Zone (BZ). The blue dashed lines highlight the energies of the harmonic components induced by Talbot modulation. These harmonic spectra extend towards photon energies of 0.8 keV, presenting themselves as a series of plateaus with gradually declining efficiency. Interestingly, each plateau within this sequence undergoes a distinct upward shift towards higher energies, owing to discrete energy displacements linked with the periodicity of the Talbot revivals.

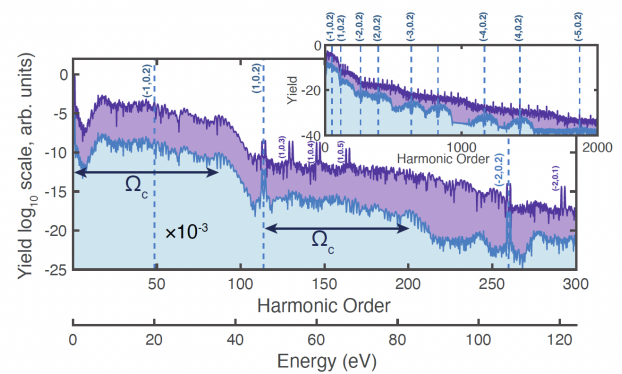


Fig. 1. High-harmonic spectrum for a $3 \mu\text{m}$, 10^{13} Wcm^{-2} driving pulse, lasting 5 cycles (18 fs FWHM). In this visualization, the blue curve delineates the contribution of a single Bloch state at $k = 0.2g$, while the purple curve illustrates the cumulative contribution of ten states evenly dispersed along the Brillouin Zone (BZ). The blue dashed lines denote the energies associated with the Talbot revivals.

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