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Ultrafast Talbot Spectroscopy

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High-order harmonic generation (HHG) is a well-established technique to generate attosecond x-ray coherent radiation and, also, an extraordinary spectroscopic tool to probe ultrafast dynamics in the matter. The highly nonlinear HHG mechanism has been thoroughly studied in atomic and molecular gases while, lately, there is an increasing interest in driving it in solid targets. In this latter case, HHG has been used to reveal, for instance, the electronic band structure of crystals. In atomic and molecular targets, HHG can be easily understood by means of the so-called *three-step model*. According to this interpretation, when an intense laser field interacts with an atomic or molecular target, electronic wavepackets are liberated through tunnel ionization. Then, they are accelerated by the laser field and, finally, after the reversal of the field, they are driven back to rescatter with the parent ion. The high-frequency radiation, resulting from the oscillating dipole induced in this final step, is emitted in the form of several high-order harmonics of similar intensity, giving rise to a spectral *plateau*. Although in solids the driving laser field polarization is typically considered perpendicular to the target's surface, it has been shown that HHG from two-dimensional solids at grazing incidence is also possible and that stems from a mechanism very similar to the atomic three-step model[1].

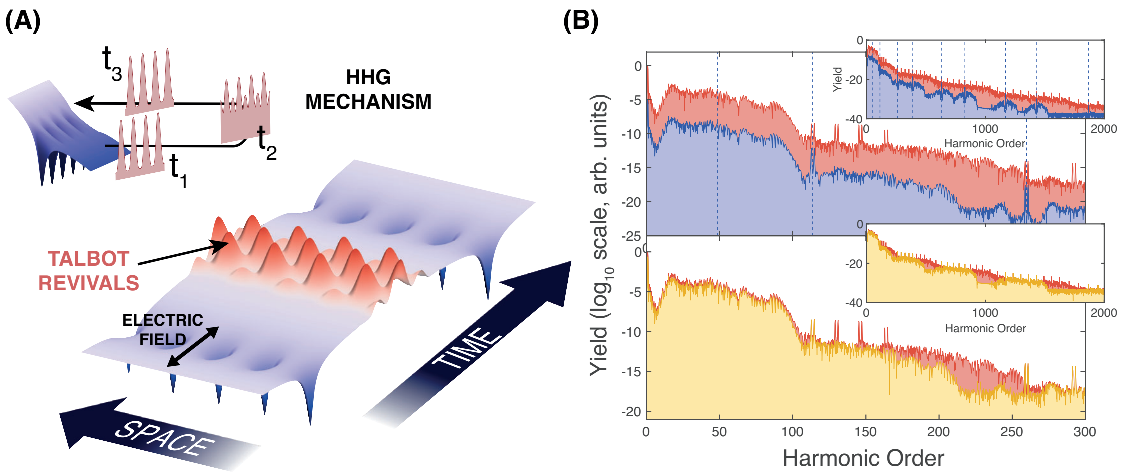

Figure 1: (A) Interaction geometry and HHG process in a low-dimensional solid under grazing incidence. (B) HHG spectra for a single Bloch state (blue), ten Bloch states equally distributed along the Brillouin zone (red) and five of those ten states with lower energy (first band partially filled).

One of the most relevant properties of HHG is that the coherence of the electronic wavefunction is preserved during the process. In this work, we demonstrate that HHG from 2D solids at grazing incidence is an excellent scenario to explore common phenomena in coherent optics[2], such as

Talbot imaging, in matter waves at the ultrafast time scale. The possibility of exploring subfemtosecond subnanometer coherent matter-wave phenomena has, to the best of our knowledge, not been addressed yet. As a main result, we show that the harmonic spectra enclose Talbot interferometric signals. Therefore, our proposal of *ultrafast Talbot spectroscopy* emerges as a natural procedure to access interferometric measurement in the unprecedented subfemtosecond/subnanometer scales[3].

Talbot effect, in coherent optics, refers to the formation of a series of self-images at periodic distances that follows on from the near-field diffraction of light by a periodic grating. In this work, to study the analogous matter-wave phenomenon in the ultrafast scale, we consider a few-cycle mid-infrared laser pulse interacting with a one-dimensional 2.1 Å-lattice-parameter crystal at grazing incidence. Figure 1(A) schematizes the interaction geometry and the HHG process. In this scenario, the periodic electronic wavepackets in the crystal potential (blue) are tunnel-ionized to the continuum, a situation that is analogue to the diffraction of light by a grating. The released electronic wavepackets (red), under the influence of the laser field, experience a series of ultrafast Talbot revivals before being driven back to recollide with the crystal. During this rescattering, the Talbot modulations in the ionized electronic wavefunction leave an unequivocal signal in the HHG spectrum. To some extent, the crystal potential at rescattering plays the role of the second optical grating, of a Talbot-Lau interferometer.

Figure 1(B) shows the HHG spectrum arising from the numerical resolution of the two-dimensional time-dependent Schrödinger equation (TDSE). The blue filled curve shows the spectrum corresponding to an electron that is initially occupying a single Bloch state. The Talbot revivals of the ionized electron lead to the appearance of a series of high frequencies, well above the cut-off. These frequencies emerge as a sequence of *plateaus* with decreasing intensity which are, indeed, replicas of the first *plateau*. The red-filled plot corresponds to the HHG spectra resulting from the total contribution of a set of ten Bloch states equally distributed along the Brillouin zone, showing a variety of replicas. We have developed a strong-field approximation description of the process, that allows us to identify the Talbot frequencies (blue dashed lines in Fig. 1(B)). As proof of the practical application of Talbot spectroscopy, we present at the bottom of Figure 1(B) a comparison between the spectrum when the valence band is totally filled with that of a partially filled band (yellow-filled), demonstrating that the spectral signal is sensitive to the band occupation.

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