Quantum Path Interferences in High-Harmonic Generation: Ionization Effects and Spatial Structure

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Abstract: We investigate the influence of microscopic and macroscopic ionization effects on the intensity-dependent quantum-path interferences in high-harmonic generation. The resulting interference structures were analyzed in different gases and in spatially resolved harmonic spectra.

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1. Introduction

Theoretical models describing high-order harmonic generation (HHG) predict multiple electron trajectories leading to the emission of given spectral components in the plateau region [1]. Significant contributions from more than one trajectory may be observed for proper macroscopic conditions. Since the phase of the emission depends on the trajectory and the laser peak intensity, interference will occur in spectral regions with contributions from multiple trajectories. The resulting yield modulations in the HHG emission were recently experimentally observed for the first time [2]. The measured quantum path interference (QPI) data demonstrates that laser intensity can be used to control the electron excursion time on each trajectory with an accuracy of tens of attoseconds. Here, we report the investigation of the influence of the important ionization effects on the observed interference structures. The experimental analysis with HHG media with different ionization potential is supported by theoretical results. Spatially resolved harmonic spectra provide a new view of QPI and the ionization processes intrinsic to HHG.

Measuring the intensity-dependent QPI in three gases with different ionization potential and comparison with theory allows us to pinpoint the impact of microscopic and macroscopic ionization effects on the observed interference structures. While the microscopic effect of ground-state depletion leads to an asymmetry in the harmonic spectra due to the suppressed emission on the falling edge of the laser pulse, the macroscopic effect of free-electron dispersion leads to distortions of the fringe pattern. The latter may result in apparent merging or break-up of interference fringes. Spatially resolved QPI spectra provide a deeper insight into the dynamics of these processes, which are of fundamental importance for HHG.

2. Experiment and Results

Our experiments are based on a Ti:sapphire laser system delivering 30 fs pulses at a repetition rate of 1 kHz with maximum pulse energy of 3 mJ. The laser beam enters a vacuum chamber and is focused by a spherical mirror (ROC = 500 mm for the xenon and argon target, ROC = 250 mm for the neon target) into a pulsed gas jet. The jet is movable along the laser propagation direction to control which trajectory is involved in the high-order harmonic generation process: with the jet positioned after the laser focus, we can phase match the short and long trajectories simultaneously. A variable attenuator is placed in the IR beam for fine control of the laser peak intensity in the jet. Far-field spatial filtering with 6 mrad acceptance was used before the XUV spectrometer to select different transverse areas (from on- to off-axis positions). This enables us to select the trajectory contributions sent to the XUV spectrometer and to control the contrast of the interference pattern.

As the HHG media, we use xenon, argon, and neon. The presence of QPI in the data for all three gas species demonstrates that the experimental conditions for their observation as defined in Ref. [2] are universally applicable. The comparison of measurements taken in the different gases allows us to study the behavior of QPI with laser intensities above, around and below the ionization threshold. Depending on this regime, the plateau harmonics exhibit intensity-dependent features such as saturation of the harmonic yield, spectral asymmetry and blue shift, trajectory dependent spectral broadening, and fringe pattern distortions due to free electron dispersion (see Fig. 1). The measurements are well supported through simulations.

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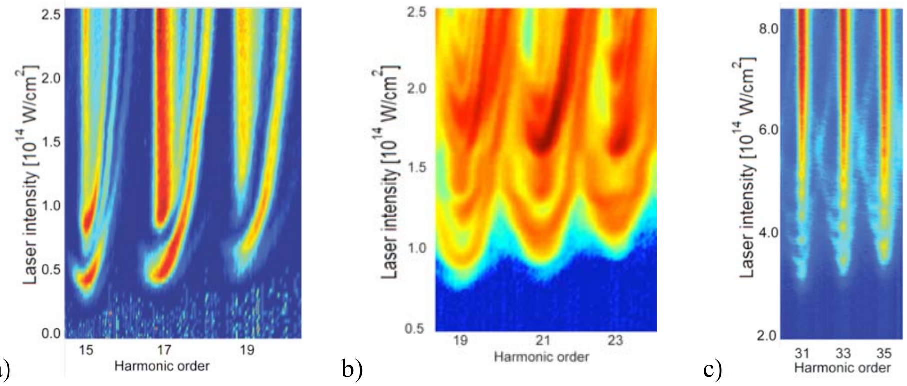

Fig. 1. Three plateau harmonics for each of the gases investigated. Xenon data is depicted in a), argon data in b), and neon data in c). In xenon, the harmonic emission is strongly asymmetric and only a single QPI period can be observed. The spectral asymmetry gets less, while more QPI modulation periods are observed for the target media with higher ionization potential. The dispersion from free electrons distorts the fringe pattern leading to apparent fringe splitting in the fine structure (best seen in argon).

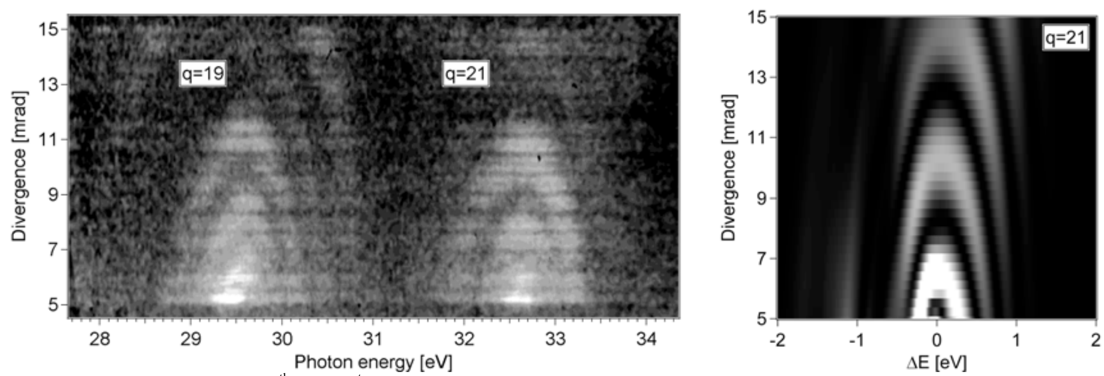

Fig. 2. Spatially resolved 19th and 21st harmonic in argon in the presence of significant contributions from the short and the long trajectory and at a fixed peak intensity of ~$3{\cdot}10^{14}$ W/cm^2 (left graph). Right graph: Corresponding simulations from a theoretical model based on the strong-field approximation and including propagation effects.

The spatially resolved spectrum shown in Fig. 2 is recorded at a fixed peak intensity value of ~$3{\cdot}10^{14}$ W/cm^2 for phase matching conditions resulting in comparable contributions from the short and long trajectories. For this measurement, the far field spatial filter is removed and an imaging detector is used compared to the setup described above. The observed spatial fringe pattern is in excellent agreement with theoretical calculations based on the strong-field approximation with subsequent propagation. The intrinsic time-to-frequency mapping in the harmonic emission allows us to track ionization dynamics and its influence on the QPI with time in the spatially resolved fringe pattern. We observe a harmonic yield asymmetry propagating from lower to higher harmonic frequencies with increasing peak intensities of the laser above barrier suppression. This is consistent with the onset of ground state depletion moving to earlier times within the driving pulse.

3. Conclusions and Outlook

In conclusion, we found general experimental conditions that enable us to study quantum path interferences in various HHG media. The comparison between the different media and with theoretical considerations allows identifying the impact of ionization related microscopic and macroscopic effects on the QPI. Spatially resolved QPI permit us to track the ionization dynamics occurring within the infrared pulse driving the HHG process. In the future, our studies can be extended to molecular targets while the phase information contained in the QPI may be used as an interferometrically sensitive probe.

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