Probing Xenon Electronic Structure by Two-Color Driven High-Order Harmonic Generation

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Abstract We studied the two-color HHG emission from xenon in the giant resonance spectral region. We found a substantial departure from the behavior expected for the single-active-electron picture which could be ascribed to electron correlation effects.

1 Introduction

High-order harmonic generation (HHG) is a sensitive probe of atomic and molecular structures. Recently this research field greatly benefited from the exploitation of mid-IR driving pulses that allowed the extension of the harmonic emission to higher photon energies, giving access to several phenomena previously unexplored, such as the giant resonance in xenon [1]. This enhancement in the harmonic generation yield around 100 eV has been interpreted in terms of the electronic structure of xenon, suggesting the key role of single [2] and multi-electron [1] contribution to the

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harmonic generation process. The presence of phase-matching effects could play an important role as well, making the interpretation even more complicated [3].

In order to provide further insight into this interesting phenomenon, we exploited HHG by a two-color field, combining this powerful experimental approach with a mid-IR driving source. Harmonic spectroscopy based on two-color driving pulses has already been successfully applied to the study of electronic structure and attosecond dynamics in atoms and molecules [4, 5]. In this work we provide the evidence of a deviation of the xenon response with respect to the expected atomic behavior which could be attributed to electron correlation effects as introduced by Shiner et al. [1] and recently theoretically confirmed by Pabst and Santra [6].

2 The Giant Resonance in Xenon

The giant resonance in xenon [1, 6] involves electron correlation effects. In particular, in the first step of the HHG process (see Fig. 1), the electron is mainly ionized by tunneling from the valence 5p orbital. Under the influence of the external laser field, the electron is driven back to the parent ion. In the recombination step, the electron may either recombine with the 5p hole in the parent ion, or it could exchange energy with the ion promoting an inner shell electron from the 4d shell to the valence shell via Coulomb interaction (dotted arrow in Fig. 1). In this latter case the electron will then recombine with the 4d hole. This inter-channel coupling gives rise to the enhancement in the harmonic yield at high photon energy.

3 Experimental Results

We exploited an optical parametric amplifier (OPA) pumped by an amplified Ti: sapphire laser system (60 fs, 20 mJ, 800 nm). The OPA is based on difference frequency generation and provides driving pulses with 1,500 nm central wavelength, pulse duration of 20 fs and pulse energy 1.2 mJ. We used a BBO crystal followed by

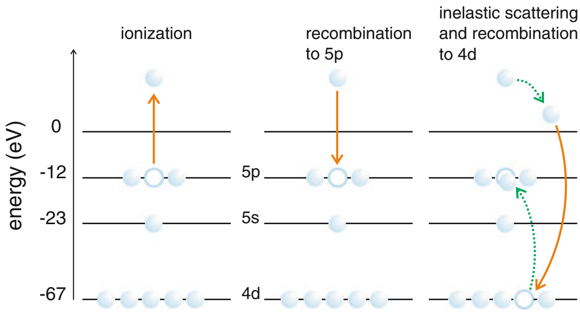

Fig. 1 Sketch of the ionization and recombination steps of HHG (spin states are not considered)

a calcite plate for generating a two color driving field with two components with perpendicular polarization. We changed the relative phase of the two driving field components with a pair of wedges. High order harmonics were generated by focusing the two-color driving field on a pulsed gas jet. Harmonics were detected by means of a flat field XUV spectrometer coupled to an MCP and a CCD detector.

Figure 2a shows a sequence of harmonic spectra generated in xenon for different values of the relative phase between the two components of the driving field as a 2D colormap. By changing this relative phase, the harmonic yield oscillates giving rise to the spectral modulation clearly visible in the figure. The spectra at high photon energy show for some value of the relative phase a feature which can be related to the giant resonance [1]. It is worth noting that the measurement reported in Fig. 2a has been performed with a very low xenon pressure in order to avoid clusterization and to minimize the role of phase-matching in the HHG process [3]. The phase of the spectral oscillation has been retrieved by Fourier transforming the sequence of harmonic spectra for each photon energy and by selecting the component at the oscillation frequency. The retrieved phase is shown as a solid black line on top of the 2D colormap in Fig. 2a. For the sake of comparison we show the same results for a measurement performed in krypton in Fig. 2b. In atoms, in the framework of the single-active-electron picture, one expects to see a smooth monotonic increase in the phase up to the cutoff [5], as the one we retrieved in krypton. In the framework of the single-active electron picture this behavior is intimately connected to the evolution of the electron trajectories related to the harmonic emission. In xenon we observed a very different behavior: a clear non-monotonic change in the slope of the phase corresponding to the spectral region of the giant resonance. Such finding is compatible with the occurrence of multielectron dynamics in HHG. Indeed, two-color HHG has been suggested as a sensitive approach for probing collective effects on an attosecond time scale [5].

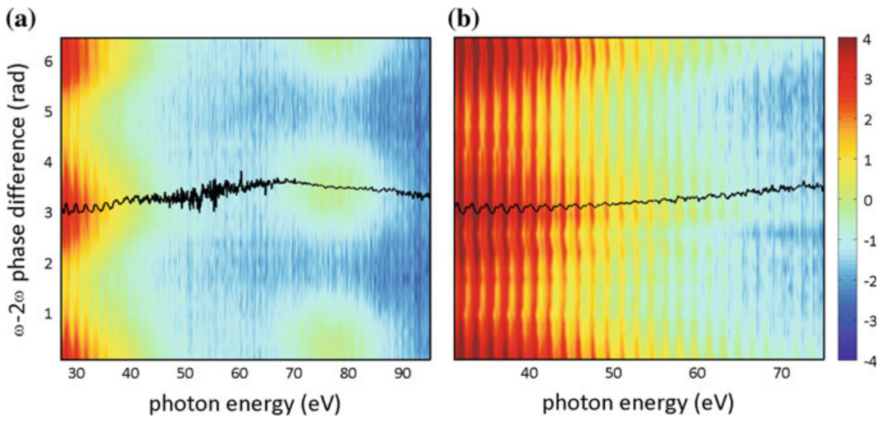

Fig. 2 Scan of harmonic spectra acquired in xenon (**a**) and krypton (**b**) as a function of photon energy and two-color phase difference (log scale). The *black line* corresponds to the phase of the spectral oscillation retrieved by Fourier transforming the sequence of harmonic spectra for each photon energy and by selecting the component at the oscillation frequency

4 Conclusions

In conclusion we investigated the xenon response by two-color HHG spectroscopy. We found a substantial departure from the expected behavior in atoms at high photon energy. This finding might support the evidence of correlation effects occurring in xenon and paves the way to the study of these dynamics on the attosecond time-scale.

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