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Controlling high harmonic generation in diatomic molecules by phase shaped laser pulses

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Synopsis In this work we explore the high-order harmonic generation in the H_2^+ molecule under the action of phase shaped pulses. To this end, quantum mechanical wavepacket calculations are performed within a collinear model of H_2^+. We analyze the effect of a chirp to the plateau extension of the HHG spectrum, observing significant differences with respect to the sign of the chirp parameter. This result is found to originate from a subtle electron recollision dynamics within the chirped laser pulses, as substantiated by classsical trajectory calculations. Extension of this work focuses on including the nuclear motion of H_2^+.

The process of high harmonic generation has proven to be an effective source of extreme utraviolet and soft x-ray light with a crucial application to generate attosecond light pulses, which can be used in time-resolved pump-probe experiments. Coherent control over the HHG spectrum has been studied by many authors, commonly in atoms, for various purposes such as the selective enhancement of harmonics or the temporal shape harmonic emission.

The aim of the present work is to analyze the effect of modifying the spectral phase of the IR laser onto the generation of high harmonics. For that purpose, we first analyze the dependence of the cutoff energy with respect to the chirp parameter. In addition to an extension of the plateau region for chirped pulses, we have found significant differences with respect to the sign of the chirp parameter depending also of the peak intensity of the laser pulse which determines the tunnel rate.

For the usual laser intensities, negative (down) chirp rates are found to be more efficient than positive (up) chirps. Nevertheless, when using strong pulses, the efficiency of HHG is reversed. An increase of the ionization yield penalizes down-chirped pulses due to significant groundstate depletion. Since the up/down chirped pulses have the same spectrum, the observed asymmetry cannot be attributed to purely spectral arguments, using the three-step model, but is the result of a complex electron recombination dynamics. Quantum results are substantiated by classical trajectory calculations.

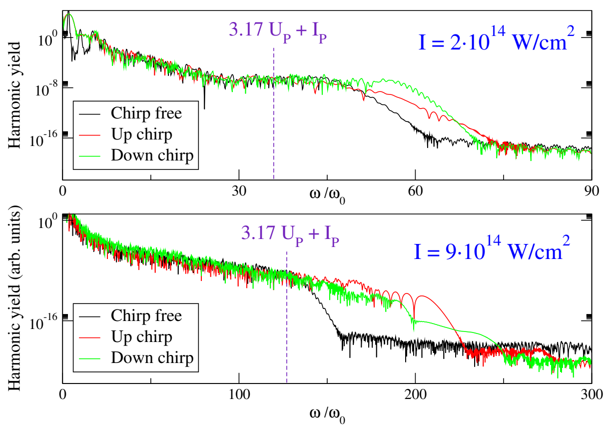

Figure 1. Chirp effects on harmonic generation. The harmonic yield is greatly enhanced near the cutoff frequency, especially for the down-chirped pulse (green) when normal intensities are applied. In the high-intensity regime, up chirps (red) become more efficient to extend the cutoff.

Founded on the obtained results, the effect on nuclear motion for those pulses is analyzed by 2-dimensional wavepacket calculations. Indeed, recent calculations have shown the importance of correlated electron-nuclear dynamics in the high-order harmonic generation process. [1]

References

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