High-order harmonic generation from field-distorted orbitals

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A recently proposed extension of the Lewenstein model [1] showed that the energy shift induced by the field driving a molecular target during high-order harmonic generation (HHG) plays a crucial role. For example, this theory accounting for the Stark effect predicts a shift of the two-center interference minimum in the HHG spectrum from polar molecules with respect to previous predictions neglecting the field-induced energy shift [2].

Here, we take into account field-distortion of molecular orbitals by the driving laser field as a further development of the theory. The effects of the orbital distortion on the HHG spectra reveal that structural features in the spectrum strongly depend on the polarizability of the target. A study of HHG from N_2 (high polarizability) suggests that the absence of the two-center interference minimum in the HHG spectra, evidenced by many experiments (see e.g. [3]), may be explained by orbital distortion (see Fig. 1). Moreover, we propose that the minimum may be observed experimentally from the long trajectory part of the spectrum, by carefully adjustment of the molecular orientation and the field parameters. For further validation of our theory, we study CO_2 (low polarizability). For CO_2 experiments proved the survival of the minimum in the HHG spectrum. Our results agree well with experiments (see e.g. [4]).

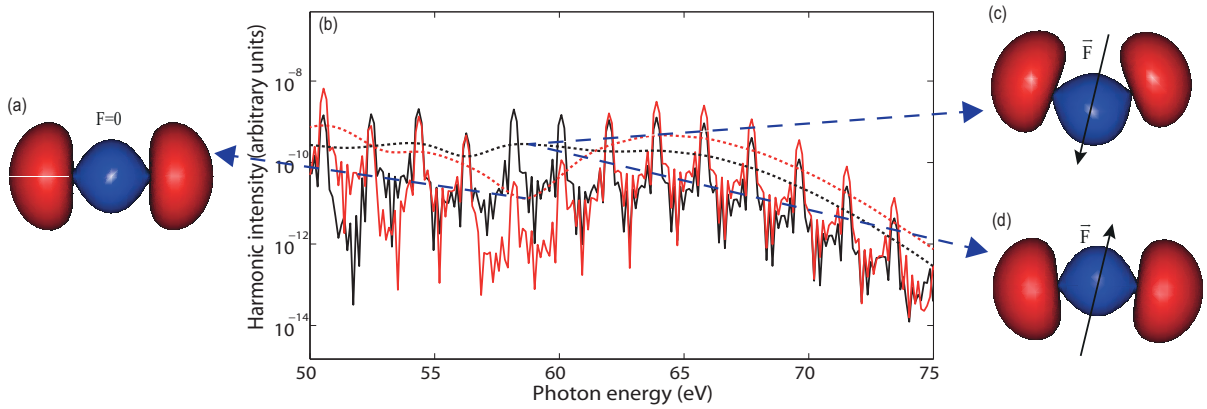

Figure 1. HHG spectra for N_2 from field-free and field distorted-orbitals. (a) Field-free, undistorted orbital. (b) HHG spectra for field-free (solid red), and field-distorted (solid black) orbital. The smoothed spectra (dashed lines) are obtained by averaging over four harmonics. (c) Field-distorted orbital for a recombination time when 58 eV photons are emitted from the short trajectory electrons (F=0.04 a.u.). (d) Field-distorted orbital for a recombination time when 58 eV photons are emitted from the long trajectory electrons (F=0.01 a.u.). In (c) and (d) full (black) arrows indicate the field direction. The (blue dashed) arrows from (b) to (a), (c), and (d) indicate the shape of the orbitals for which the given part of the spectrum is created.

References:

[1] A. Etches and L. B. Madsen, J. Phys. B **43**, 155602 (2010).
[2] A. Etches, M. B. Gaarde, and L. B. Madsen, Phys. Rev. A **84**, 023418 (2011).
[3] J. B. Bertrand *et al.*, Phys. Rev. Lett. 109, 143001 (2012).
[4] C. Vozzi *et. al.*, Nature Physics 7, 822 (2011).