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Measurement of the attosecond emission from aligned molecules

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Recently, a number of papers have demonstrated the interest of high-order harmonic generation (HHG) from molecules aligned with respect to the laser polarization. Itatani et al. (*Nature* **432**, 867 (2004)) have shown that a precise characterization of the harmonic emission allows performing a tomographic reconstruction of the molecular orbitals that radiate. Kanai et al. (*Nature* **435**, 470 (2005)) have evidenced quantum interferences in the recombination process of HHG that are directly related to the molecular structure. In all of these papers, only the HHG intensity was measured. The relative harmonic phase, though more difficult to measure, contains important information on the interference process, and is needed for an ab initio tomographic reconstruction. Finally, while the attosecond emission from atoms has been thoroughly studied, in particular by our group (Mairesse et al., *Science* **302**, 1540 (2003)), it has not been investigated in molecules.

In a first experiment (Wabnitz et al., *EPJD (2006)*), we measured the amplitude and relative phase of harmonics radiated by ***un-aligned*** nitrogen molecules. Small but reproducible deviations from the phase of harmonics generated in argon (same ionization potential as nitrogen) were measured for low orders.

In a recent experiment, we have measured, up to high order, the harmonic amplitude and relative phase for ***aligned*** molecules (N_2 and CO_2). In order to align the molecules, we used the so-called nonadiabatic technique: a rotational wavepacket is created by a strong enough and short aligning pulse, so that a field-free alignment is obtained at the revival (a few ps after the aligning pulse). The measurement of phase locking between neighboring harmonics was performed through the photoionization of a target gas by the harmonic beam in presence of a sufficiently intense "dressing" laser beam (RABITT technique).

The harmonic phase measured when the CO_2 molecules are aligned parallel to the generating laser polarization (at the revival of the rotational wavepacket, 22 ps after the aligning laser pulse) are significantly different from the Krypton case (which has a similar ionization potential). Up to harmonic 21, their behavior is similar, but at harmonic 23 we observe a phase jump spread over 3 harmonic orders. The value of the total phase shift is around 2 radians, which is close to the phase jump of π that is predicted when destructive interference occurs in the recombination process. The position of the jump coincides with the dip in the harmonic spectrum measured at harmonic 23 by Kanai et al. (*Nature* **435**, 470 (2005)). This behavior disappears when the CO_2 molecules are aligned at 90°, which is consistent with the two-center interference model.

The reconstructed temporal profile of the attosecond pulse trains emitted by aligned CO_2 molecules are strongly affected by this phase jump that leads to a splitting of the pulses.

We also investigated the case of aligned nitrogen molecules. We observed a phase jump at harmonic 25, but this jump is independent of the angle of alignment. The origin of this interference would thus be different from that observed in CO_2.

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