Two-source double-slit interference in angle-resolved high-energy above-threshold ionization spectra of diatoms

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Synopsis Experimental data are reported and compared with molecular strong-field-approximation calculations that display an interference where four electronic quantum orbits conspire. For random orientation of the molecule, footprints of the destructive interference survive for O_2 , but not for N_2 .

Owing to the two centers of a diatom, the typical high-order above-threshold ionization quantum orbit of a freed electron is replaced by four such orbits, depending upon at which center the electron is ionized and rescatters. The coherent superposition of the contributions of these four orbits in the ionization amplitude gives rise to a particular destructive interference, which defines a curve in the angle-energy plane of the ionized electron along which the electron yield is almost zero. These curves, which depend on the orientation of the molecule, are almost identical for the two molecules N_2 and O_2 , in contrast to the usual two-center destructive interference, which depends on the symmetry of the molecular ground state (σ or π).

For randomly oriented molecules, this interference pattern is washed out. However, for orientation of the molecule with respect to the field intermediate between parallel and perpendicular, the yield is suppressed for emission in the field-polarization direction. For O₂, due to its π symmetry there is little emission for parallel and perpendicular orientation, and the suppression survives in the averaged yield (see Fig. 1). For N₂ emission from perpendicular alignment is dominant, and the suppression cannot be seen (not shown).

We present calculations based on the molecular strong-field approximation (MSFA) extended to include rescattering [1, 2] and compare them with experimental data obtained with a setup used previously [3]. The agreement is good with respect to the essential features of the phenomenon, including the intensity dependence.



Fig. 1. Angle-resolved electron energy spectra of randomly oriented O_2 as a function of the angle θ_e between electron emission and field polarization. Upper panel: experimental data for a 100-fs 800-nm laser pulse with estimated peak intensity of 7×10^{13} Wcm⁻². Lower panel: calculation via the MSFA, averaged over the molecular orientation.

References

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