Intramolecular interference effects in photoelectron momentum distributions arising due to strong-field molecular ionization

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Synopsis

The strong-field phenomenon of multiphoton above-threshold ionization (ATI) in laser-irradiated molecular dimers (H₂, N₂ and O₂) is addressed theoretically within the velocity-gauge formulation of strong-field approximation under conditions of experiments with a laser field of mid-infrared wavelength (λ ≥ 800 nm). Our calculation results demonstrate clear signatures of intramolecular interference manifested as pronounced and well-established interference minima arising in photoelectron momentum distributions whose form and structure proved to be sensitive to internuclear separation R₀ and spatial orientation of molecular axis.

We report about the results of our theoretical study of strong-field (multiphoton) above-threshold ionization (ATI) in laser-irradiated molecular diatomic species (H₂, N₂ and O₂) under conditions of experiments [1, 2] observing a characteristic (the so-called two-hump) structure in longitudinal photoelectron momentum distributions (PMD), which becomes especially prominent using mid-infrared laser wavelengths (λ ≥ 800 nm). The problem is addressed within the velocity-gauge (VG) formulation of molecular strong-field approximation (SFA) [3]. In particular, only the so-called direct ATI process is supposed as quite a sufficient underlying physical mechanism solely contributing to the phenomenon under study. Moreover, the density-functional theory (DFT) is also essentially exploited here for numerical composition of initial (laser-free) atomic and/or molecular state using the routines of GAUSSIAN-03 code [4].

Our present DFT-SFA based calculation results suggest the two-hump form of longitudinal atomic/molecular PMDs similar to those observed in experiments for atomic ionization in the tunneling regime (the value of the Keldysh parameter γ ≤ 1). In particular, for the photoelectron emission parallel to the laser field polarization, the calculated molecular PMDs (e.g. for ionization of N₂ see Figure 1) also represent a symmetric sequence of discrete photoelectron peaks in the parallel-momentum distributions with a prominent central minimum between the two symmetrically located and well-extended “horn-like” side-lobes (humps). However, unlike the atomic case, Figure 1 remarkably demonstrates a substantially different form of calculated molecular PMDs (as an envelope of peaks) which is well seen to suggest a presence of two distinct and pronounced minima - each located within a respective (either of two symmetric) hump. In addition, the form and location of these minima proved to be well sensitive to spatial orientation of molecular axis relative to the incident laser field polarization (though quite insensitive to the laser wavelength λ and peak intensity I). The latter feature allows to identify the origin of minima and interpret them as arising due to intramolecular interference of multiphoton transition amplitudes corresponding to the photoelectron emission from either of two identical atomic centers of molecular dimer under ionization.

Figure 1. PMDs calculated for ionization of Ar atom and N₂ molecule aligned along the polarization of incident laser field of fixed wavelength λ and peak intensity I (as indicated in the picture inset).

References


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