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Strong-field dissociation dynamics of molecular dications

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Synopsis We focus on the dissociation of metastable molecular dications induced by intense, ultrafast laser pulses. In particular, we demonstrate the dominant role of commonly-neglected permanent-dipole transitions and drive dissociation via a pump-dump-like mechanism within a single laser pulse.

Many doubly-charged molecules have ground electronic states with lifetimes spanning from picoseconds to seconds [1], thus enabling unique studies. We focus on dissociation of several-keV beams induced by intense, ultrafast laser pulses. Specifically, we demonstrate that permanent-dipole (PD) transitions can play a key role and that the structure of the metastable electronic ground state can delay stimulated emission, leading to pump-dump-like transitions.

In NO2+, dissociation parallel to the laser polarization is prominent at high intensities, as shown in Fig. 1(a). Contrary to common intuition dictating that electronic transitions always prevail, we find that permanent-dipole transitions are extremely important in this system. We demonstrate this with time-dependent Schrödinger equation (TDSE) calculations [2], shown in Fig. 1(b), together with the most likely number of photons involved in the transition, derived from fitting the measured angular distribution in Fig. 1(a) [3]. Specifically, we find that the dominant pathways leading to the aligned feature at high kinetic energy release (KER≥8.9 eV) are multiphoton permanent-dipole-driven vibrational excitations on the electronic ground state leading to its continuum.

Figure 1. (a) Density plot of NO2+ → N+ + O+ dissociation as a function of KER and cosθ for 5×1015 W/cm² laser intensity. (b) Calculated KER spectra for transitions starting from X 3Π → A 3Σ− transitions. (c) Measured KER spectrum for CS2+ → C+ + S+.

Figure 2. (a) Potential energy curves for CS2+ → C+ + S+. (b) Potential energy curves for CS2+ [4] with dissociation pathways for the one-photon (1ω) and pump-dump (2ω) transitions.

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