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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 NO^{2+}, 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.

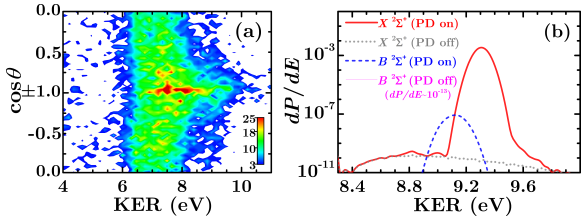

Figure 1. (a) Density plot of $NO^{2+} \rightarrow N^+ + O^+$ dissociation as a function of KER and $\cos\theta$ for 5×10^{15} W/cm^2 laser intensity. (b) Calculated KER spectra for transitions starting from $X\,^2\Sigma^+(v{=}3)$ and dissociating on the indicated state.

In CS^{2+}, photodissociation via $X\,^3\Pi \rightarrow A\,^3\Sigma^-$ transitions yield the main KER peak, marked as 1ω in Fig. 2(a). In addition, a more intriguing peak, marked 0ω, appears at about one-photon energy lower KER. This 0ω peak can be understood as a pump-dump process driven by one laser pulse, where the same $X\,^3\Pi \rightarrow A\,^3\Sigma^-$ excitation ("pump") is followed by delayed stimulated emission ("dump"). This delay, on the order of a few tens of fs, is due to the potential barrier of the electronic ground state, which makes stimulated emission energetically forbidden until the dication has stretched far enough, as indicated in Fig. 2(b). Once the Condon point is reached, stimulated emission is most likely, and the pump-dump-like process results in $C^+ + S^+$ fragments with a KER value reflecting no net energy exchanged with the laser field. Finally, we also observe this "pump-dump-like" mechanism in $CO^{2+}(v{=}0)$.

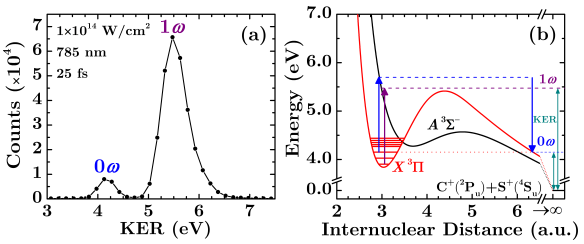

Figure 2. (a) Measured KER spectrum for $CS^{2+} \rightarrow C^+ + S^+$. (b) Potential energy curves for CS^{2+} [4] with dissociation pathways for the one-photon (1ω) and pump-dump (0ω) transitions.

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