Controlling Asymmetric Coulomb Explosion Using 2-Color Fields

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Synopsis Intense, asymmetric, two-color laser fields have been used to control directional Coulomb explosion in several small molecules. A strong asymmetry in the forward/backward fragment directionality from asymmetric dissociation channels is detected as a function of the relative phase between the 800 nm and 400 nm pulses. Manipulation of the optical phase enables control over native and/or induced electron localization in the strong optical field. The robust effect might be utilized as an effective phase-meter for few-cycle laser pulses.

Recently, the ability to specifically tailor the electric field of an ultrashort laser pulse through carrier-envelope phase (CEP) stabilization has become available. Application of such asymmetric CEP stabilized pulses to D_2 molecules has illustrated control of electron localization between nuclear centers during the dissociation process [1]. Asymmetric electric fields are also readily created by combining a laser field at fundamental frequency ω with its second harmonic at 2ω; variation of the relative phase between the two components tailors the field asymmetry [2].

In our experiments, laser pulses of 35 fsec duration at 800 nm are frequency doubled in a BBO crystal, resulting in a two-color laser field with total intensities of $10^{13} - 10^{14}$ W cm^{-2} and field asymmetries $0.2 \leq (|E_f/E_b| - 1) \leq 0.9$. The relative phase between the red and blue components is continuously varied as the pulses are applied to the dissociation of N_2, O_2, CO, HBr, and CO_2. Coulomb explosion occurs along the laser polarization, which is parallel to the axis of a single-stage time of flight (TOF) mass spectrometer. In the symmetric molecules, a distinct phase-dependent asymmetry in the forward/backward fragment directionality is observed for asymmetric dissociation channels, i.e. $N_2^{+(p+q)} \rightarrow N^{+p} + N^{+q}$, $p \neq q$, for $(p+q) \geq 3$. Charge symmetric channels in hetero-nuclear molecules, e.g. $CO^{+4} \rightarrow C^{+2} + O^{+2}$, also exhibit this asymmetry.

The target molecules were selected to explore the role of orbital structure in the asymmetric dissociation. Oxygen, which has a π-shaped highest occupied molecular orbital (HOMO), was found to exhibit an asymmetry parameter as large as that for N_2, which has a σ-shaped HOMO. Carbon dioxide, also possessing a π-shaped HOMO, exhibits asymmetries in several dissociation channels, indicating that HOMO structure is not a barrier to electron localization.

Repeating the N_2 and CO experiments with an elliptically polarized field also resulted in large phase-dependent fragment directionalities. This suggests that electron recollision is not the dominant mechanism for the observed asymmetric dissociation.

By adding a third, time-delayed probe pulse, the possibility of laser-induced molecular orientation in CO and HBr is explored. No field-free orientation at the characteristic revival times was observed, indicating that transient molecular orientation does not occur. Instead, our results are consistent with the notion of charge localization and enhanced ionization in the asymmetric field.

Analogous fragmentation directionality induced by CEP-stabilized pulses [3] could be used to measure the carrier-envelope phase.

References

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