

Vibrationally cold CO²⁺ probed by intense femtosecond laser pulses

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Synopsis Using a novel approach, we produce a vibrationally cold CO²⁺ beam for study in an intense ultrashort laser field. We observe perpendicular dissociation of the simple two-level CO²⁺ $v=0$ ion and above-threshold dissociation peaks spaced by the photon energy.

H₂⁺ seems the ideal candidate to study ultrashort intense laser-molecule interactions since its simple one-electron structure allows it to be treated accurately by theory. This is aided by the fact that at low intensity ($<5 \times 10^{13}$ W/cm²) it may be approximated as a two electronic state system. However, as recently pointed out by Posthumus *et al.* [1], vibrational excitation of H₂⁺, with the population often unknown, can severely complicate its dynamics in an intense laser field. This makes experimental observation, and study, of important laser-induced phenomena such as bond-softening and above-threshold dissociation (ATD) more difficult and confusing.

Ion traps are currently used to vibrationally cool H₂⁺ [2]. Alternatively, we present novel measurements of intense field dissociation of vibrationally cold CO²⁺, which is in many respects a similar target to cold H₂⁺. Our method involves production of CO²⁺ ions, by electron-impact in an ion source, that predissociate during their travel ($\sim 20 \mu\text{s}$ transport time) to the laser interaction region — leaving a pure target of $v=0$ ground state CO²⁺ molecules. Laser-induced fragmentation of CO²⁺ is measured by 3D momentum imaging of the C⁺ and O⁺ fragments detected in coincidence [3].

Specifically, we demonstrate that cold CO²⁺, like H₂⁺, can be considered as a two-state system at low intensity (involving only its lowest triplet states). This allows some of the important theoretical foundations developed for H₂⁺ to be applied and tested on this more complex multielectron system. Work along these lines will be presented in this poster. Moreover, as the CO²⁺ initial state is well-defined, we observe sharp peaks in the kinetic energy release from dissociation [Fig. 1(a)], attributable to one- and two-photon processes, measured for both 790 nm and 395 nm

wavelengths. Interestingly, unlike H₂⁺ where dissociation occurs for molecules predominantly aligned to the laser polarization [Fig. 1(c)], CO²⁺ displays the opposite behavior, showing a preference to be aligned perpendicular to the laser polarization [Fig. 1(b)] due to the nature of the dominant dissociative transition.

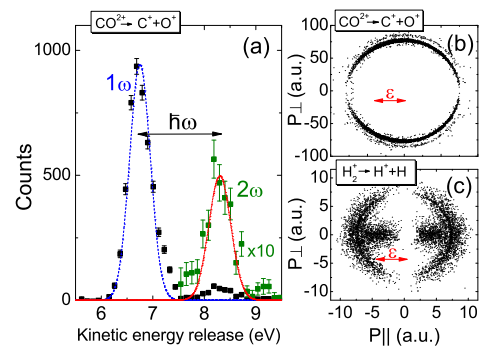


Fig. 1. (a) Kinetic energy release and (b,c) angular distributions for dissociation of CO²⁺ and H₂⁺ at 2×10^{15} W/cm². The angle θ is between the molecular dissociation axis and the laser polarization. P_∥ and P_⊥ denote momentum parallel and perpendicular to the laser polarization, respectively.

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References

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