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Zero photon dissociation of CS^{2+} in intense ultrashort laser pulses¹ TRAVIS SEVERT, K.J. BETSCH, M. ZOHRABI, U. ABLIKIM, BETHANY JOCHIM, K.D. CARNES, B.D. ESRY, I. BEN-ITZHAK, J.R. Macdonald Laboratory, Physics Department, Kansas State University, Manhattan, KS 66506, USA — We measured the dissociation of a CS^{2+} molecular ion beam in intense laser pulses (<50 fs, $<10^{15}$ W/cm²), focusing on the zero photon dissociation (ZPD) and above threshold dissociation (ATD) mechanisms. The ZPD mechanism leads to dissociation with the net absorption of zero photons in a strong field. The present work extends the idea of ZPD to more complex molecules than the H_{2}^{+} discussed in literature. Preliminary data suggests that ZPD is larger than ATD for $CS^{2+} \rightarrow C^+ + S^+$. We speculate that a pump-dump process occurs whereby the vibrational wavepacket in the electronic ground state of CS^{2+} is pumped into the electronic first excited state's continuum by a single photon during the laser pulse. Once this continuum vibrational wavepacket passes the potential barrier in the ground electronic potential, the emission of a second photon is stimulated by the same laser pulse, most likely when the wavepacket moves through the internuclear distance where the two electronic states are in resonance with the driving field. A comparison is made to ZPD and ATD in the isovalent CO^{2+} species. Curiously, ATD is the favored mechanism in CO^{2+} . The underlying molecular structure and dynamics determining this preference will be discussed.

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