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Energy-resolved photoion angular distributions from ion-pair formation in O_2 following 2-photon absorption of a 9.3 eV femtosecond pulse KIRK LARSEN, Univ of California - Berkeley, ROBERT LUCCHESE, DANIEL SLAUGHTER, THORSTEN WEBER, Lawrence Berkeley National Laboratory — We present a combined experimental and theoretical study on the photodissociation dynamics of ion-pair formation in O_2 following resonant 2-photon absorption of a 35 femtosecond 9.3 eV pulse produced via 400 nm driven high harmonic generation, where the resulting O^+ ions are detected using a 3-D momentum imaging spectrometer. Ion-pair formation states of ${}^{3}\Sigma_{g}$ and ${}^{3}\Pi_{g}$ symmetry are accessed through predissociation of continuum molecular Rydberg states that are resonantly populated via a mixture of both $\|\cdot\|$ and $\|\cdot \perp 2$ -photon transitions, where this mixture varies with the kinetic energy release (KER) of the dissociating ion-pair. The variation in the mixture's composition is captured by the KER dependent photoion angular distribution, which helps clarify the underlying 2-photon absorption dynamics involved in the ion-pair formation mechanism and indicates that the propensity towards undergoing a $\|-\|$ or $\|-\bot$ transition varies with the molecular structure.

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