

Abstract Submitted
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Direct femtosecond laser excitation of the 2p state of H by a resonant 8-photon transition in H_2^+ . GEORGE GIBSON, LI FANG, BRAD MOSER, University of Connecticut — We observe Lyman- α radiation produced by the direct excitation of thermal H_2 molecules by 25-fs 800-nm laser pulses. The excitation proceeds in 4 steps. First the laser pulse ionizes the H_2 molecule. The H_2^+ molecular ion then begins to dissociate through bond softening. The expanding H_2^+ ion enters a region of strong multiphoton coupling with the laser field in which resonant excitation from the strongly coupled $1s\sigma_g$, $2p\sigma_u$ states to the $2s\sigma_g$, $3p\sigma_u$ states can occur. The population in the $2s\sigma_g$ and $3p\sigma_u$ states finally dissociate into H atoms in the 2s and 2p states. This scenario is supported by several independent experiments. 1) The Lyman- α fluorescence has a linear dependence on pressure, which is consistent with direct, but not plasma, excitation. 2) A weak probe pulse delayed by 500 fs can quench the radiation, as the H(2p) state is easily ionized. This shows that the excited state is populated within 500 fs of the first pulse, again ruling out plasma excitation. 3) The radiation is seen with circularly polarized light, ruling out excitation through rescattering. 4) Ion time-of-flight coincidence measurements show a new $H^+ + H^+$ channel when a weak probe pulse is applied, as would be expected from the quenching experiment. All of these results are fully consistent with a new theory of high-order strong-field coupling in diatomic molecules.

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