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The importance of Rydberg orbitals in dissociative ionization of small hydrocarbon molecules in intense few-cycle laser pulses<sup>1</sup> E. WELLS, A. VOZNYUK, J.B. MAHOWALD, D.G. SCHMITZ, T.G. BURWITZ, Augustana College, B. JOCHIM, M. ZOHRABI, K.J. BETSCH, T. SEVERT, B. BERRY, N.G. KLING, U. ABLIKIM, K.D. CARNES, I. BEN-ITZHAK, J.R. Macdonald Laboratory, Kansas State University, R. SIEMERING, M.F. KLING, R. DE VIVIE-RIEDLE, Ludwig-Maximilians-University Munich — Much of our intuition about strong-field processes is built upon studies of diatomic molecules, which have relatively well separated electronic states. In polyatomic molecules, however, the electronic states are closer together, leading to more complex interactions. A combined experimental and theoretical investigation of strong-field ionization followed by hydrogen elimination in the hydrocarbon series  $C_2D_2$ ,  $C_2D_4$ , and  $C_2D_6$  reveals that the photofragment angular distributions can only be understood when ionization from Rydberg orbitals is considered. These commonly neglected Rydberg orbitals are readily populated for some orientations of the molecule relative to the laser polarization, leading to dissociation patterns and an intensity dependence consistent with significant Rydberg orbital influence. Our results suggest that Rydberg states should be routinely considered when studying polyatomic molecules in intense laser fields.

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