Electron localization following attosecond molecular photionization

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Synopsis Observation and control of electron dynamics on attosecond timescales are the main driving forces behind the recent emergence of attosecond science. In this experiment an isolated attosecond laser pulse ionizes D_2. and part of the ionized molecules dissociate. Interaction of these dissociating wave packets with a moderately strong IR field localizes the electron on one of the D^+ ions. By varying the delay between the XUV pulse and the few-cycle IR pulse, the localization is controlled with attosecond resolution. A detailed comparison between the experiment and a full-dimensional model is presented.

Using attosecond laser pulses, experiments can be conceived where electron dynamics is initiated on attosecond timescales. Subsequently, the electrons are allowed to evolve and are probed (again, on an attosecond timescale), providing new insights into the fundamentals of photoexcitation, as well as the role of electron-electron correlations and electron-nuclear energy transfer on these ultrafast timescales. A technical breakthrough that has been very influential and that has greatly improved the ability to generate attosecond laser pulses in a controlled manner, was the development of amplified femtosecond laser systems with a controlled carrier-envelope-phase (CEP) in 2003 [1]. This enabled the controlled generation of isolated attosecond laser pulses with a pulse duration down to the present record of 80 attoseconds [2]. In addition, CEP-stable pulses have been used to control atomic ionization and the localization of electrons in D_2^+ molecules [3]. In this experiment an isolated attosecond laser pulse ionizes D_2, and part of the ionized molecules dissociate. Isolated attosecond pulses are produced by means of high-harmonic generation in Krypton using the polarization gating technique [4]. Due to the wide photon energy spectrum (20-40 eV) associated to the attosecond pulse, a large manifold of continuum as well as autoionizing states are populated. Several interfering pathways are then possible, direct dissociative and non dissociative ionization as well as autoionization from the Q_n doubly excited states.Interaction of these dissociating wave packets with a moderately strong IR field localizes the electron on the upper or lower D^+ ion. By varying the delay between the XUV pulse and the few-cycle IR pulse the localization is controlled with attosecond resolution. To include all possible ionization pathways a full dimensional model is developed for the interaction of the molecule with the XUV-IR pulse sequence. We will present a detailed comparison between this theory and the experimental results. It will be also shown that the essence of the electron localization can be understood in terms of a few discrete transitions between the quasi-static states of the dissociating molecule.

References

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