

Directional control of dissociative ionization by a two-colour laser field

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Abstract. Using asymmetric two-color laser fields composed of 1800 and 900nm, we have simultaneously controlled four well identified fragmentation channels in dissociative ionization of the hydrogen molecule, resulting in enhanced electron-localization sensitivities of up to 65%.

1 Control principle

The interaction of an asymmetric laser field with a molecular system can lead to efficient control in the directionality of its dissociative ionization. Whether an electron remains on one side or the other of a dissociating molecule depends on the composition of this electric field; due to the longer period of one optical cycle (6.0fs at 1800nm versus 2.7fs at 800nm), the mid-infrared (MIR) spectral range enables a more efficient subcycle control of slower nuclear dynamics compared to 800nm, which are the preponderant dynamics in large molecular systems. Also for the simplest of all molecules, the hydrogen cation, one can achieve excellent timing for the coupling of the electronic states involved, σ_g and σ_u (see Fig. 1a), since half a vibrational period matches the optical cycle. This leads to very efficient control of electron localization. At the same time, MIR photons comprise a smaller energy (0.69eV @ 1800nm vs. 1.55eV @ 800nm), which opens up additional interference pathways when overcoming the energy gap of relevant electronic states, due to more photon-combination possibilities.

This variety further enriches the “play on the control-piano” of dissociation channels.

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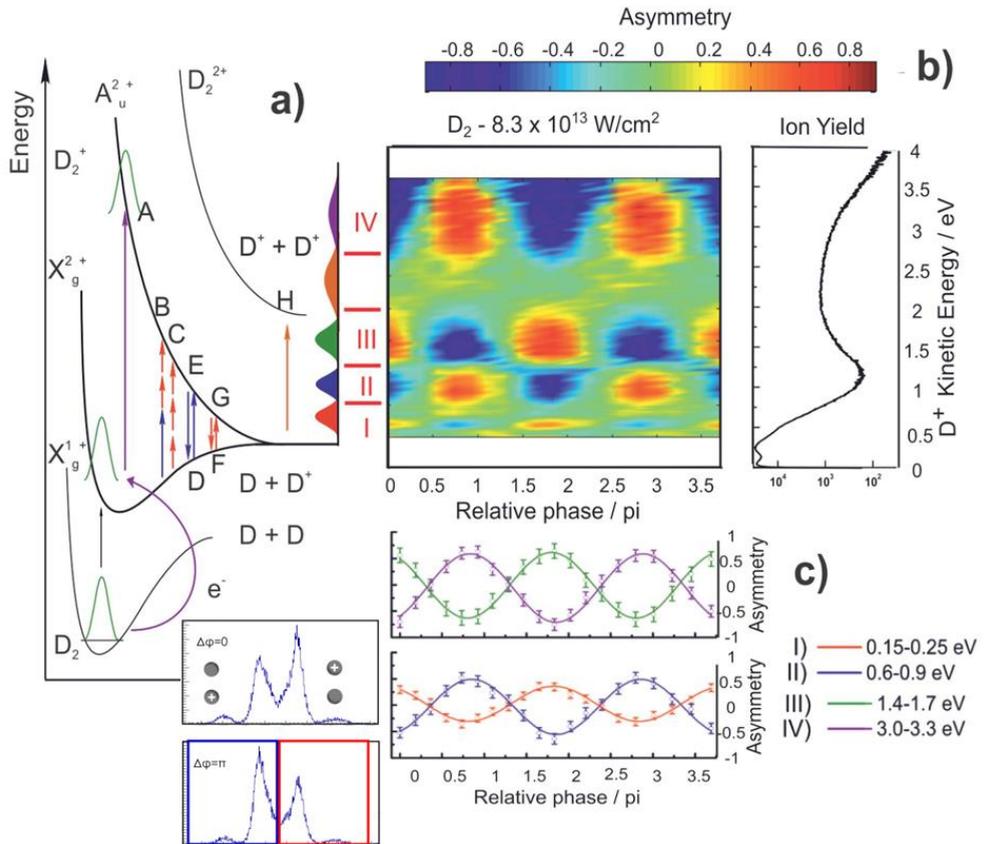


Fig. 1. Principle of D_2^+ dissociation control experiment with control channels at different kinetic energy (KE) regions, as indicated with roman numbers. (a) Scheme of the reflection principle associated to the formation of D^+ ions from bond softening (red KE region), above-threshold dissociation (blue KE region), three-photon dissociation (green KE region), charge-resonance-enhanced ionization (orange KE region, no control) and recollision excitation (purple KE region). Capital letters correspond to different pathways. Blue arrows indicate absorption of 2ω photons, red arrows of ω photons. The two insets show the occurring asymmetry depending on the relative phase φ in the time-of-flight spectrum. The left two peaks correspond to D^+ fragments pointing towards the detector, the right two peaks correspond to D^+ fragments pointing away from it (b) left panel: experimental calculated asymmetries for the detected D^+ fragments at an intensity of $8.3 \times 10^{13} \text{ Wcm}^{-2}$ and right panel: corresponding logarithmic energy distribution. (c) Averaged asymmetries for the specified KE cuts.

2 Results

For the experiments reported here, MIR two-colour (1800 and 900nm) laser pulses of 50fs duration were used to coherently control the dissociative ionization of H_2 and D_2 molecules [1,2]. In the two-colour technique, which is experimentally significantly easier to implement compared to CEP stabilized few-cycle pulses, an asymmetry over such a relatively long duration can be achieved, since the electric field can basically be tailored on any chosen timescale. This is because in the case of CEP stabilization the time-averaged electric field goes to zero with increasing

duration, in contrary to two- colour fields where an asymmetry is imprinted on every single cycle and thus remains independent on the pulse duration.

With this two-colour approach we have obtained control over four distinct dissociation pathways, occurring from different areas of kinetic energy (KE), as indicated in Fig.1, utilizing the normalized asymmetry parameter $A(\varphi, E) = (N_{up}(\varphi, E) - N_{down}(\varphi, E))/(N_{up}(\varphi, E) + N_{down}(\varphi, E))$:

- (I) Bond Softening channel (red and blue region of kinetic energy - KE, indicated in Fig. 1a): Absorption of one ω or one 2ω photon (pathways G and E, respectively) can lead to dissociation of the molecule via the σ_u cationic state (net one photon).
- (II) Above threshold dissociation (ATD, blue KE range): E.g. three photon absorption (C) followed by stimulated emission of one photon (F) may lead to dissociation of the molecule via the σ_g cationic state (net two photons).
- (III) Three-photon dissociation (green KE range): E.g. three photon absorption (C) (net three photons).
- (IV) Recollision excitation (RE, purple KE region): While channels (I-III) arise from radiative coupling of the molecule with the laser field, channel (IV) results from an inelastic recollision of the ionized electron with its remaining molecular ion. This leads to an excitation of the molecule to the σ_u dissociative state (A). Because the dissociative state is populated rather early, the generated ions possess a high KE.

The actual control effect occurs from an overlap of involved nuclear wavepackets in space and time on the potential energy surfaces - in general the case when their kinetic energy distribution is similar. This leads to tuneable interferences between dissociation pathways, each populating electronic states of different parity via the concurrent net absorption of n and $n+1$ photon(s) in the presence of a strong laser field. As an example, because BS (pathway E) and ATD (pathways C+F) both lead to the same final KE but with wavepackets propagating on different electronic states (σ_g and σ_u), the resulting interference allows us to coherently control the dissociation channel of KE region II. Hereby enhanced electron-localization sensitivities of up to 65% are achieved.

The experimental results are well reproduced by a theoretical model solving the time-dependent Schrödinger equation for the molecular ion, identifying the involved dissociation mechanisms. Furthermore, the comparison between theory and experiment allowed us to highlight the importance of the initial vibrational distribution on electron localization. We have found that in our case fewer vibrational states are being involved, compared to a Franck Condon type distribution.

References

1. V. Wanie, H. Ibrahim, S. Beaulieu, N. Thiré, B. E. Schmidt, Y. Deng, A. S. Alnaser, I. V. Litvinyuk, X.-M. Tong, F. Légaré, J. Phys. B: At. Mol. Opt. Phys. **49**, 025601 (2016)
2. H. Ibrahim, C. Lefebvre, A. D. Bandrauk, A. Staudte, F. Légaré, J. Phys. B: At. Mol. Opt. Phys., **51**, 042002 (2018)