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Two-Dimensional Control of Electron Localization in H_2 Dissociation with Elliptically Polarized Few-Cycle Pulses

Sarayoo Kangaparambil[1,*], *Václav* Hanus[1], *Seyedreza* Larimian[1], *Xinhua* Xie[1], *Markus* Schöffler[2], *Gerhard* Paulus[3], *Andrius* Baltuska[1] and *Markus* Kitzler[1]

[1]Photonics Institute, Technische Universität Wien, Gusshausstrasse 27, 1040 Wien, Austria
[2]Institut für Kernphysik, J.W. Goethe-Universität, Max-von-Laue-Strasse 1, 60438 Frankfurt am Main, Germany
[3]Friedrich-Schiller-Universität Jena, Max-Wien-Platz 1, 07743 Jena, Germany

Abstract. We demonstrate two-dimensional control over the charge-localization in H_2 dissociation using elliptically polarized laser pulses. The influences of the CEP and the laser phase at the instant of ionization are investigated.

1 Introduction

The possibility of gaining control over charge-localization processes with strong laser fields has been discussed as a possibility to control chemical reactions on the ultrafast time scale [1-3]. Control over directional bond-breaking of the hydrogen molecule has been demonstrated in one dimension using the CEP of linearly polarized few-cycle laser pulses [1-3], or the relative phase between two colors in multicycle tailored laser fields [4]. Recently, bond-breaking control was extended to two spatial dimensions by employing orthogonally polarized and circularly polarized multicycle two-color laser fields. In all of these experiments [1-5], asymmetric bond-breaking in the lab-frame is achieved by employing laser pulses that exhibit an asymmetric electric field in this frame of reference. It was, however, also demonstrated that due to the fast nuclear motion of H_2 there exists an intrinsic asymmetry that merely depends on the initial values of the dissociation process. This asymmetry manifests itself and can be revealed using angular streaking [6].

Here we investigate for the first time two-dimensional control of charge-localization during dissociation of the H_2 molecule at the few-cycle limit. We employ elliptically polarized fields and demonstrate two-dimensional directional dissociation using the CEP as the control parameter. We furthermore extend the investigation of the role of the laser phase in the dissociation process, as initially revealed in Ref. [6], to the few-cycle range and discuss the connection between this molecular-frame asymmetry with the observed lab-frame asymmetry controlled by the CEP.In our experiments, we studied dissociation of H_2, the

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[*] Corresponding author: sarayoo.kangaparambil@tuwien.ac.at

prototypical model system used in directional bond breaking experiments [1-6]. We used near-circularly polarized laser pulses with a duration of ~5 fs, with a spectrum centered around 760 nm. The laser beam was focused on a cold H_2 beam with a pulse peak intensity of 0.8 PWcm^{-2}.The charged particles produced during the interaction of a laser pulse with a H_2 molecule were detected by time and position sensitive detectors. By this, their three dimensional momentum vectors were determined. CEP tagging of the pulses was performed with a phase meter.

2 Dissociation of H_2^+

Upon interaction with a strong laser field, the H_2 molecule may become singly ionized and undergo dissociation. During the dissociation, the electric field can drive the remaining bound electron back and forth between the two nuclei until, at large inter-nuclear distances, the electron localizes on one of them. The underlying physical process in the directional dissociation of the hydrogen molecule is understood by the coupling between the $1s\sigma_g$ and $2p\sigma_u$ states [7]. The molecular ion, vibrating on the $1s\sigma_g$ state, can be excited to the $2p\sigma_u$ state in multiple ways via the absorption of an odd number of photons. This leads to different dissociation pathways such as bond softening (BS), above-threshold dissociation (ATD) and three-photon dissociation (3PD). The interference of these dissociation pathways results in electron localization. The relative phase between two interfering pathways is the key parameter for controlling the directionality of the bond-breaking process and can be manipulated with the CEP [1-3] and the laser ionization-phase at the ionization instant [6].

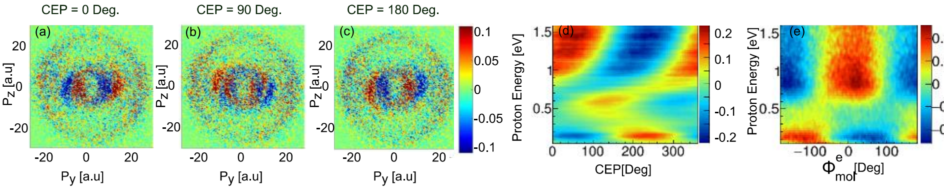

Fig. 1. (a)-(c) 2D asymmetry patterns of proton yield in the polarization plane for three different values of the CEP. (d) CEP dependent asymmetry observed in the lab frame as a function of proton energy. (e) Asymmetry in proton ejection as a function of the electron emission angle (ϕ^{e}_{mol}).

3 Controlling the dissociation process in the lab – and molecular frames

Control over the dissociation process, i.e., the determination whether the proton is emitted along a certain direction in the lab-frame of reference, implies gaining control over the charge localization dynamics. Exploiting the fact that by the CEP we are able to determine the evolution of the laser field of our few-cycle pulses in two-dimensions, we demonstrate control over the localization of the electron on one of the two nuclei, as visualized by the rotations of the asymmetry patterns in the polarization plane [Figs. 1(a)-(c)]. A more intuitive representation of the achieved control is obtained by plotting the asymmetry of the proton yield along the major axis of the polarization ellipse as a function of the CEP and the proton energy, see Fig 1(d).

It has been shown recently using much longer pulses, that the charge-localization process can be intrinsically linked to the laser-phase at the ionization instant [7]. To see the effect of this in our experiment, we plot the left-right asymmetry of the proton yield as a function of proton energy and the electron emission angle in the molecular frame of reference (ϕ^{e}_{mol}),

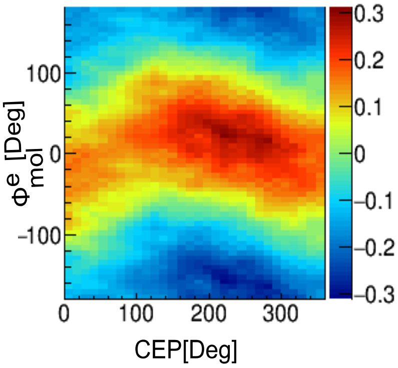

Fig. 2 Asymmetry in proton emission as a function of the electron emission angle ϕ_{mol}^{e} and CEP for proton energies 1-1.2eV.

see Fig. 1(e). Because in elliptically polarized fields the electron's emission angle is uniquely linked to the ionization time, the angle ϕ_{mol}^{e} is a direct measure of the laser ionization phase. The asymmetry map is markedly different from that measured with more narrow-band multi-cycle laser pulses [7].In both Fig. 1(d) and 1(e) one can identify two different proton energy regions (0.1-0.6 and 0.7-1.7 eV) that exhibit opposite dependence of the asymmetry on the CEP (lab frame) and the electron emission angle (molecular frame), respectively. The largest modulation of the asymmetry (~16%) observed between 0.7 to 1.7 eV is attributed to interference of the ATD and 3PD pathways. The smaller asymmetry-modulation (~5%) in the energy range 0.1-0.6 eV comes from the interference of the BS and ATD pathways.

Fig. 1 demonstrates the sensitivity of the charge localization process to both the ionization phase and the CEP, both of which influence the phase of the vibrational wavepacket accumulated during dissociation. It is this phase that determines the pathway interference during dissociation and thus is responsible for the observed asymmetry. To investigate the connection between the two parameters that determine the charge localization, i.e., the CEP and ϕ_{mol}^{e}, we plot in Fig. 2 the proton asymmetry along the major polarization ellipse as a function of these two parameters. It can be seen that for varying ϕ_{mol}^{e} the CEP needs to be adjusted to maintain a certain asymmetry. This means that both phases, the field's phase (CEP) and ionization phase (determined by ϕ_{mol}^{e}) are not independent of each other. We are working on numerical results that reproduce and explain the connection of the two parameters.

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