Ultrafast dynamics in CO_2 studied by XUV-pump – NIR-probe experiments

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Synopsis We present the results of an XUV-pump – NIR-probe experiment studying ultrafast dynamics of photo-ionized CO_2. The multi-harmonics XUV pulse, which contains the 11th to 39th harmonics of a 790 nm near-infrared (NIR) laser, is used to excite molecular wave packets in CO_2^+, which are then probed by a near-infrared pulse that induces dissociation. The yields and angle-resolved kinetic energy distributions of all charged fragments are measured as a function of the XUV-NIR delay using a velocity map imaging setup.

We have developed a versatile experimental setup for XUV-pump – NIR-probe experiments using a 10 kHz high-harmonic generation (HHG) source and two different charged-particle momentum imaging spectrometers. The HHG source, based on a commercial KMLabs eXtreme Ultraviolet Ultrafast Source (XUUS), is capable of delivering XUV radiation of less than 30 fs pulse duration in the photon energy range of ~17 eV to 100 eV. It can be coupled either to a conventional velocity map imaging (VMI) setup with an atomic, molecular, or nanoparticle target; or to a double–sided VMI spectrometer equipped with two delay-line detectors for electron-ion and ion-ion coincidence measurements.

We present an overview of the setup and results of first pump-probe experiments, including studies of the ultrafast dynamics of photo-ionized CO_2 molecules. The pump-probe studies are performed using a combination of broadband XUV pulses and near-infrared (NIR) pulses, both of ~25 fs duration. The XUV pulse containing the 11th to 39th harmonics of a 790 nm NIR laser is used to excite molecular wave packets in CO_2^+, which are then probed by near-infrared-induced dissociation.

We use a velocity map imaging setup to measure the yields and angle-resolved kinetic energy distributions of all charged fragments as a function of the XUV-NIR delay. The results, in particular, the delay dependence of O^+ and CO^+ ion production for parallel and perpendicular NIR and XUV polarizations, are compared to the data reported by Timmers *et al.* [1] for a shorter harmonics train, and to our earlier data obtained using narrow-band isolated harmonics (11th or 13th) as a pump.

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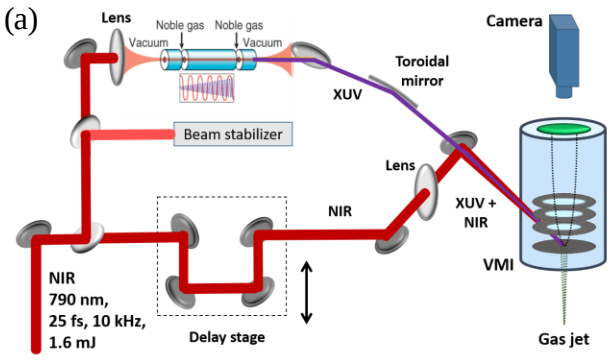

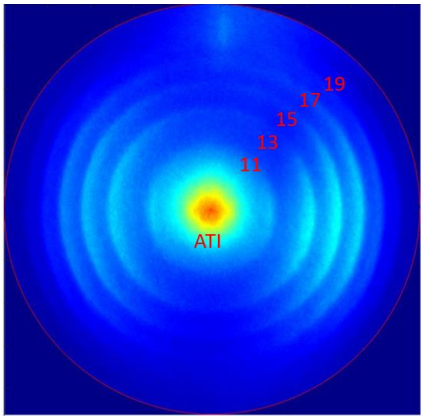

Figure 1. (a) Schematic of the XUV-pump – NIR-probe setup with the VMI spectrometer. (b) VMI image of photoelectrons produced by the ionization of argon atoms with a combination of XUV and NIR fields. The low-energy electrons at the center of the image are from above-threshold ionization (ATI), while the circular structures are the photoelectrons produced by the XUV, with the corresponding harmonic order indicated in red.

References

[1] H. Timmers *et al.* 2014 *Phys. Rev. Lett.* **113** 113003

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