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Ultrafast photoelectron imaging using VUV and tunable DUV pulses

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Time-resolved photoelectron imaging (TRPEI) enables direct access to ultrafast electronic dynamics in a molecule. We have been performing TRPEI using sub-20 fs deep ultraviolet (DUV) and vacuum ultraviolet (VUV) pulses generated in filamentation four-wave mixing to observe non-adiabatic dynamics in photoexcited molecules. So far, we have employed cascaded four-wave mixing of either the fundamental (w) and the second harmonic (2ω) in Ne [1] or of 2ω and 3ω [2] in Ar to generate DUV and VUV simultaneously in a single gas cell. The limitation of these methods is that the wavelength of the DUV is not tunable. Tunable DUV will be highly useful for exploring wavelength dependence of photophysical and photochemical processes. Therefore, we introduce here a non-collinear parametric amplifier (NOPA) to attain tunable DUV to excite a target molecule to different states more easily. This modification also obviates the need of a cascaded mixing scheme and enables the use of a more straightforward four-wave mixing scheme of $3\omega + 3\omega - \omega = 5\omega$ to generate a 5ω (159 nm) pulse.

The experimental layout is displayed in Figure 1. A cryogenically-cooled Ti:Sa regenerative amplifier (KMLabs, Wyvern) delivers 80 fs pulses at 1 kHz with a pulse energy of 2.7 mJ. The center wavelength of 795 nm was used to generate 159 nm and 260 nm radiation from filamentation and NOPA, respectively. The output of the Ti:Sa laser is split into two, with 2.6 mJ delivered for generating filamentation and 100 μJ used to pump the NOPA. The output of the Ti:Sa laser was further split in a 30:70 ratio, with the higher fraction used to generate the third harmonic (TH). The fundamental frequency (FF) (700 μJ) and TH (340 μJ) beams were gently focused (r = -2000 mm) into a krypton gas cell (L ~ 1.5 m) in a collinear geometry. When the beams are spatially and temporally overlapped, a bright filament appears and the 5ω beam can be observed on fluorescent glass (Lumilass G9) or a Ce:YAG plate after separation from the FF and TH, either using dichroic mirrors or a VUV compatible prism. The conversion efficiency and spatial mode of the 5ω beam is strongly dependent on the pressure of the krypton gas, with ca. 10 Torr krypton giving the best performance for both. At this gas pressure, up to 600 nJ of the fifth harmonic can be produced, corresponding to a conversion efficiency of 1.8×10^{-3}, and the beam mode is best described as a "ring." The 5ω beam then enters a vacuum chamber that houses optics used to filter out the generating harmonics before being delivered into our photoelectron imaging instrument.

Our NOPA system uses a 90 μJ input pulse to generate tunable DUV radiation by performing second harmonic generation of the visible output. The 795 nm input beam is split into two portions, with a small amount (> 1 μJ) used to generate the white light continuum seed, and the major portion used to generate the 400 nm pump beam (40 μJ) by performing SHG. The pump and seed beams are focused onto a

type-I beta-barium borate crystal (BBO, 30.2°), which generates ca. 4.5 μJ in the range of 500-540 nm. The tuning range can be expanded by using broadband optics inside the NOPA. The output is directed into another BBO crystal (44.3°) to produce the second harmonic, in this case 260 nm, with a pulse energy of around 450 nJ), which is then delivered to our photoelectron imaging apparatus.

The PEI apparatus is of a standard design, which we employed previously for TRPEI using a VUV free electron laser SCSS (SPring-8 SASE source) [3]. The pulse energies of several hundred nanojoules for both our 260 nm DUV and 159 nm VUV beams have enabled us to observe photoelectron images of molecules. An example of a one-color photoelectron image of toluene using ionization at 260 nm is shown in Figure 2. We will present preliminary pump-probe results at the conference.

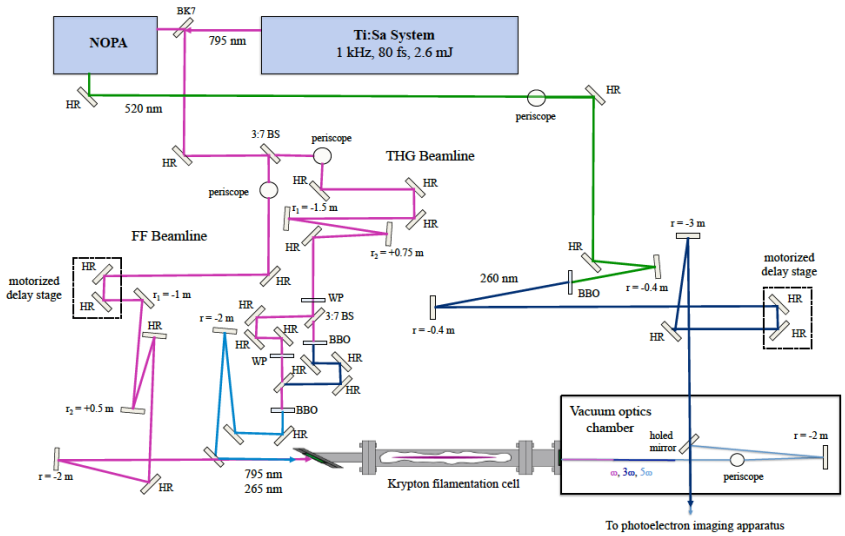

Figure 1: Schematic of the experimental layout showing the combination of tunable DUV (NOPA) and VUV (filamentation) generation for photoelectron imaging.

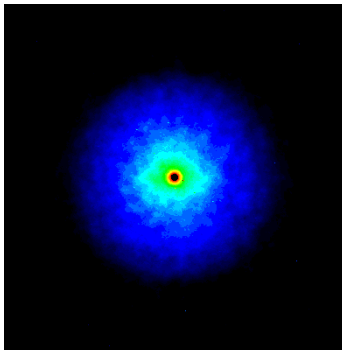

Figure 2: Photoelectron image obtained from toluene ionization (1+1) at 260 nm.

References

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