Construction of DUV and VUV light source for ultrafast photoelectron spectroscopy

Huan Shen[1], Peng Zuo[1], Shunsuke Adachi[1, 2] and Toshinori Suzuki[1,2]

[1] *Molecules Reaction Dynamics Research Team, RIKEN ASI,* [2] *Kyoto University*

Introduction: An ultrashort laser pulse in deep UV (DUV) and vacuum UV (VUV) regions is highly useful for fundamental studies of molecular reaction dynamics in gas and liquid phases [1]. In our previous studies, we generated DUV and VUV pulses simultaneously by four-wave mixing in a single filamentation cell [2]. This multi-color filamentation light source is highly useful for ultrafast photoelectron spectroscopy that requires both the pump and probe laser pulses in the DUV and VUV region. The pulse duration is typically sub-30 fs. However, since the wavelength thus generated is fixed, samples are limited to those whose absorption spectra overlap with the spectrum of DUV or VUV pulse. In the present study, we construct a tunable DUV light source, based on a non-collinear parametric amplification (NOPA), to enable resonant one-photon excitation of samples, and we integrate it with the filamentation VUV light source.

Experiment: Figure 1 shows a schematic diagram of our experimental setup. A cryogenically cooled Ti: sapphire amplifier delivers 80-fs, 2.8-mJ and 792-nm at 1 kHz repetition rate. Around 4% (~100 μJ) of the fundamental beam reflected by a glass plate is used for DUV generation, while the main (~2.7 mJ) fundamental beam is used for VUV generation. In the DUV branch, a small portion of the fundamental beam is separated by a beam splitter and focused onto a 3-mm-thick sapphire plate to generate a white light continuum (WLC). The remaining fundamental beam is used to generate the second harmonic (SH). A minor (5 μJ) portion of the SH beam was reflected by a beam splitter and sent to a DUV frequency-conversion system, while the transmitted portion (43 μJ) is used to pump the NOPA.

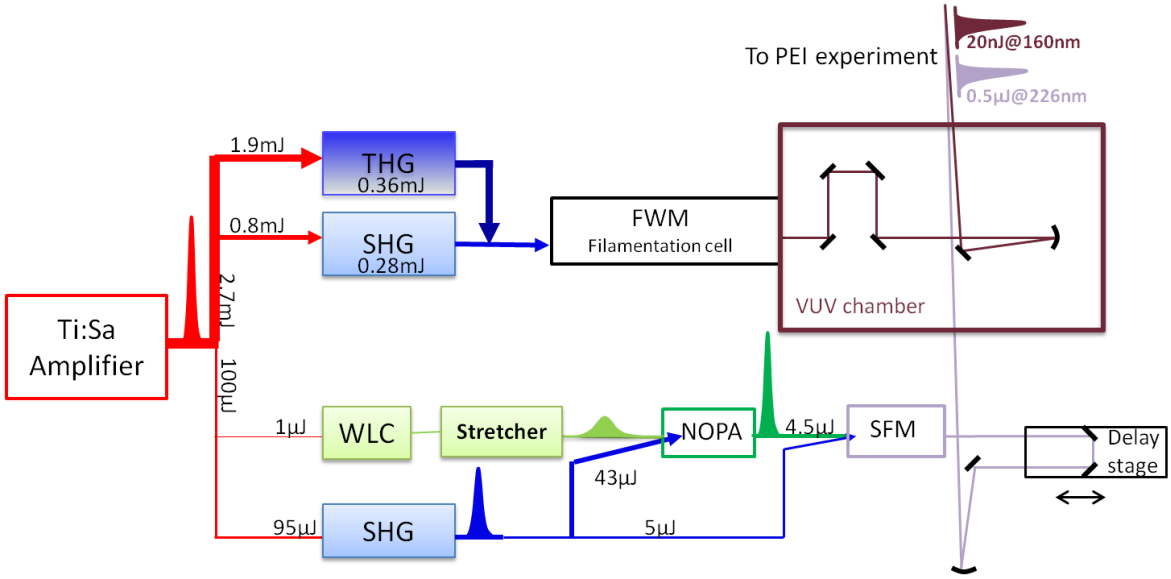

Figure 1: The schematic experimental setup for the DUV and VUV generation.

The white light is recollimated and sent to a stretcher. The stretcher consists of a pair of fused-silica prisms separated by 55 cm, and it provides a negative chirp to the seed pulse. The chirp corresponds to a group delay of 200-fs for wavelength between 510 and 530 nm. Because of the stretching, only a portion of the NOPA output temporally overlaps with the pump pulse, allowing the bandwidth and the center wavelength to be selected by fine-tuning of the chirp and the delay of the seed pulse, respectively.

The pump and seed pulses are focused onto a 2-mm-thick type-I BBO crystal. The non-collinear and phase-matching angles for the NOPA were set to 3.3° (internal) and 30.2°, respectively. The seed pulse energy of 1 nJ was amplified to 4.5 μJ, which corresponds to a parametric gain of 4.5×10^3. The NOPA spectrum shown in figure 2(a) supports a Fourier-transform-limited pulse duration of 40 fs. The NOPA output is used to generate DUV (226-nm) by sum-frequency-mixing (SFM) with 400-nm beam. The generated DUV pulse energy was 0.5 μJ at 226 nm, which corresponds to conversion efficiency from the NOPA output to DUV of up to 15%. The spectrum of 226 nm is shown in figure 2(b) and the pulse duration is expected to be 50 fs. In the present setup, the tuning range of the NOPA output is of 500–550 nm, consequently, the generated DUV can be tuned from 222 to 232 nm.

In the VUV branch, the fundamental beam is split into two and used for SH and third harmonic (TH) generation. The generated SH and TH pulses are gently focused by concave mirrors independently, combined through a dichotic mirror, and delivered into an Ar gas cell for filamentation. The ultrashort 198- and 158-nm pulses are generated simultaneously by cascaded four-wave mixing of SH and TH in the filamentation cell [2]. The pulse duration of 158-nm is expected to be ~50 fs after pass through the rear window of the cell. Both DUV and VUV pulses are introduced into our photoelectron imaging apparatus.

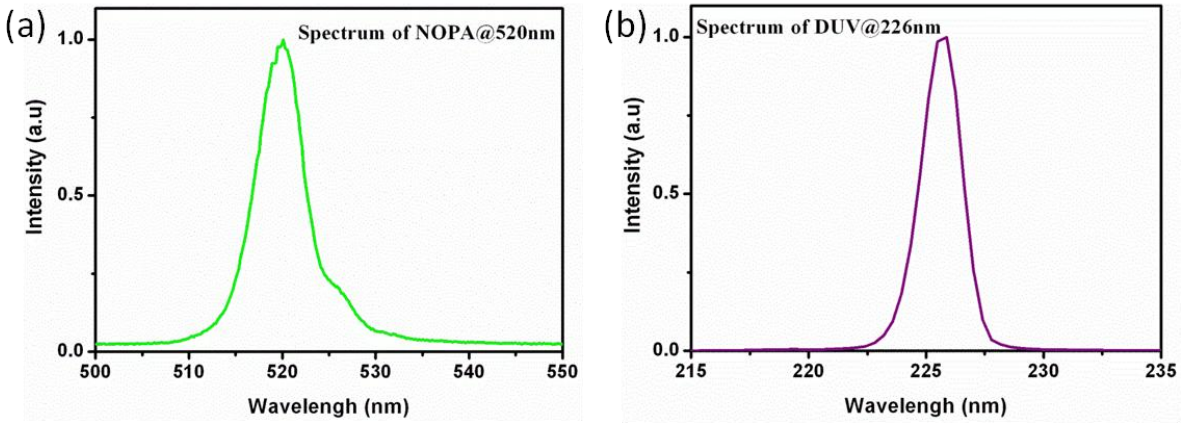

Figure 2: (a) The spectrum of NOPA at 520nm. (b) The spectrum of DUV at 226nm.

Since we started construction of NOPA in April 2012, we have not obtained spectroscopic results yet. The system, however, is almost ready for ultrafast photoelectron imaging.

References:

[1] Toshinori Suzuki, International Reviews in Physical Chemistry, 31, 265-318 (2012).

[2] Peng Zuo, Takao Fuji, Takuya Horio, Shunsuke Adachi, Toshinori Suzuki, Appl. Phys. B, in press (2012)