Optics & Laser Technology 88 (2017) 184–187

Contents lists available at ScienceDirect

Optics & Laser Technology

journal homepage: www.elsevier.com/locate/optlastec

Full length article

Generation of a femtosecond vacuum ultraviolet optical pulse by four-wave Raman mixing

Duong Vu*, Trong Nghia Nguyen, Totaro Imasaka

Division of International Strategy, Center of Future Chemistry, Kyushu University, Japan

ARTICLE INFO

Keywords:
Nonlinear wave mixing
Nonlinear optics
Four-wave mixing
Raman effect
UV
EUV
X-ray lasers

1. Introduction

An ultrashort optical pulse emitting in the deep ultraviolet (DUV) and VUV regions has attracted considerable attention due to the possibility of its numerous applications in spectroscopic and spectrometric studies. A conventional technique, in which a birefringent crystal is used, can be useful but has several limitations with respect to transparency in the VUV region and a large dispersion which induces phase mismatching. For example, a nonlinear crystal of potassium fluoroboratoberyllate has been utilized to generate an optical pulse with a wavelength below 200 nm, but the shortest pulse duration is limited to a few hundred femtoseconds [1,2]. Four-wave Raman mixing has been proven to be useful for the generation of an optical pulse in the VUV region [3,4]. Four-wave difference-frequency mixing (FWDM), which is based on the third-order nonlinear process, has been successfully used for the generation of sub 50 fs pulses at 160 nm, but the energy ratio between the VUV beam and the probe beam (200 μJ, 267 nm) is about 0.1% [5]. The efficiency of FWM can be enhanced by using a gas-filled capillary, which results in the generation of broadly tunable emission lines in a variety of spectral ranges [6–9]. A method for generating numerous Raman sidebands through FWRM has several advantages, in that the emission lines can be generated in the entire ultraviolet-visible (UV–vis) region [10] and can be synthesized to sub-femtosecond pulse trains [11,12].

In this study, the mechanism responsible for three-color FWRM was investigated, for the first time, using a 200 nm probe beam. Under optimal conditions, the conversion efficiency for generating an anti-Stokes beam at 187 nm approaches 16%, with an estimated pulse energy of 1.6 μJ. By changing the time delay of the probe pulse from the two-color pump pulse, we were able to verify that the coherent motion (vibration) of hydrogen molecules revives after a time delay of about 6 ps. At this time delay, the probe beam interacts most efficiently with the coherent vibration of molecules without competition from other nonlinear phenomena such as cross-phase modulation (XPM). The advantage of the present technique based on three-color FWRM is discussed in detail in this report.

2. Experimental

The apparatus used in the experiment is shown in Fig. 1. Two pump beams and a probe beam were generated using a Ti: sapphire laser (Legend Elite, Coherent Inc.). The fundamental beam (800 nm, 35 fs, 3.6 W, 1 kHz) was divided into two parts using a beam splitter. A part of the fundamental beam (30%) was used as the first pump beam in the three-color FWRM experiments. The remaining part (70%) was used as a pump beam for an optical parametric oscillator/amplifier (OPA – OPerA Solo, Coherent Inc.) to produce the second pump beam (1200 nm). The separation between the first and the second pump beams was adjusted so as to coincide with the vibrational Raman shift frequency of molecular hydrogen (4155 cm^{-1}). The output of the fundamental beam (1.6 W) from the OPA was used to generate the fourth harmonic emission (200 nm, 10 μJ). The pulse width of the third harmonic emission was measured using a self-diffraction frequency-resolved optical gating (SD FROG) system, suggesting a pulse

* Corresponding author.
E-mail address: duongvu@iop.vast.vn (D. Vu).

http://dx.doi.org/10.1016/j.optlastec.2016.09.010
Received 3 June 2016; Received in revised form 2 August 2016; Accepted 8 September 2016

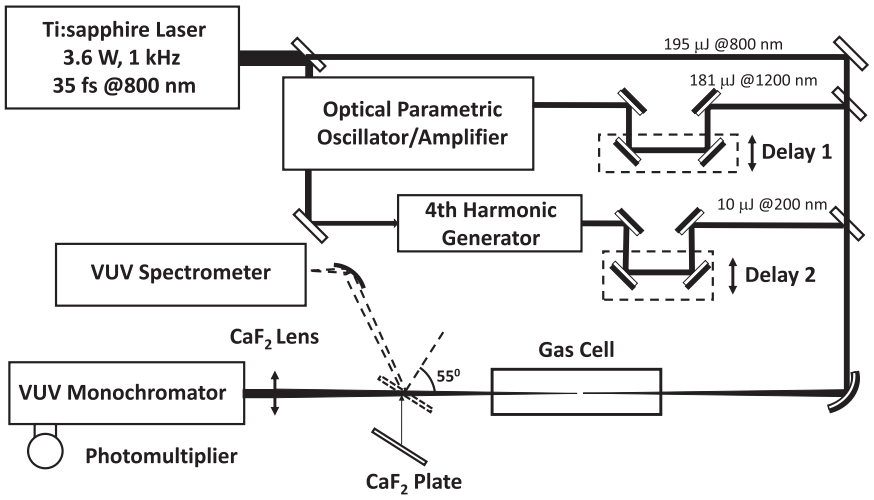

Fig. 1. Experimental setup used for the femtosecond pulse generation in the VUV. Three-color beams are delivered from a Ti:Sapphire laser. The temporal delays among these pulses can be adjusted precisely by two delay stages. After being temporally and spatially overlapped, these beams were focused into the hydrogen gas cell. The FWRM signal was measured using a VUV spectrometer or a VUV monochromator equipped with a photomultiplier.

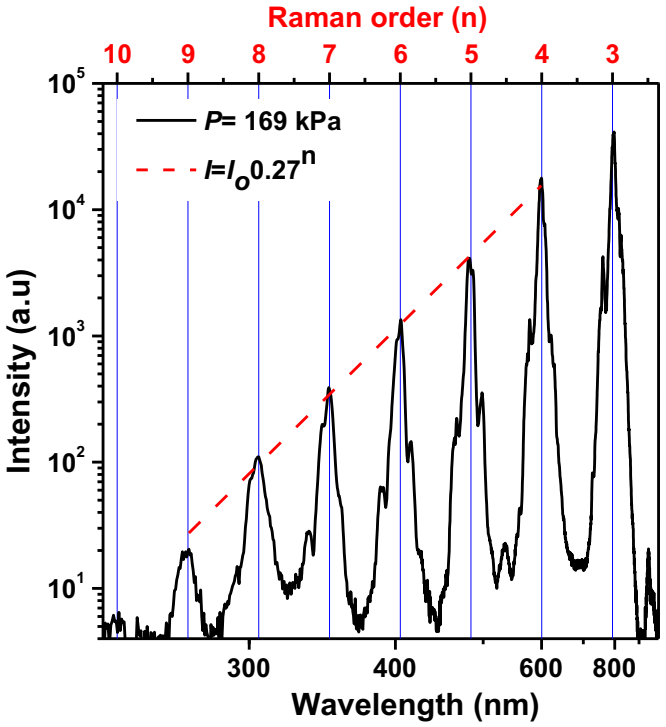

Fig. 2. UV–vis spectrum obtained when the two pump pulses (800 nm and 1200 nm) are temporally and spatially overlapped with each other. The peak intensities were fit to a straight line in a logarithmic plot (red line).

width of 60 fs. Due to a large group velocity dispersion and the small acceptance angle of the β-barium borate (BBO) crystal at shorter wavelengths, the pulse width of the fourth harmonic emission would be predicted to be slightly larger than the value of the third harmonic emission. The laser energies of the first and the second pump beams were 195 μJ and 181 μJ, respectively. After being spatially overlapped with each other, a three-color beam was focused on a Raman cell, 1 m in length, using a concave mirror with a focal length of 50 cm. Two CaF_2 plates with thicknesses of 0.5 mm were used as optical windows at both sides of the Raman cell. The relative temporal delays among these three pulses were adjusted precisely using two delay stages. The first delay stage was used to adjust the time delay between the two pump pulses. The temporal overlap of the pulses was confirmed by observing the generation of cascade Raman sidebands using a spectrometer (USB Maya-pro 2000, Ocean Optics). When the two pump pulses were perfectly overlapped, i.e., zero-delay, multicolor Raman sidebands appeared efficiently. The second delay stage was used to adjust the temporal delay of the probe pulse against the two-color pump pulse. The hydrogen gas pressure was monitored using a pressure meter (AP44, Keyence). The output beam was focused using a CaF_2 lens or an aluminum concave mirror with the same focal length of 10 cm onto a slit of a spectrometer (Custom USB Maya-pro 2000, Ocean Optics) or a VUV monochromator (VM502, Acton Research Corporation) equipped with a photomultiplier (R8487, Hamamatsu Photonics), respectively. During the acquisition of the spectral data, oxygen inside the instruments was removed by flowing nitrogen gas through the system at a flow rate of 4 mL/min. A CaF_2 plate was used to reflect the beam after the Raman cell more efficiently at shorter wavelengths. The incident angle was set at 55° in order to reduce the intensity of the visible-infrared pump beams.

3. Results and discussion

The temporal and spatial overlaps between two pump pulses were optimized by maximizing the conversion efficiency from a specific Raman sideband to the next higher order one. The spectrum of the beam after the Raman cell is shown in Fig. 2. The CaF_2 plate (see Fig. 1) was removed in this experiment. The peak intensities in a logarithmic plot were fit to a straight line. The slope of the line suggests that the conversion efficiency from a Raman sideband to the next higher order one is 27%. This represents the highest value reported to date without using a gas-filled capillary.

Since the intensity of the Raman sideband decreases exponentially at high orders in two-color FWRM, it would be expected that the pulse energy for the Raman sidebands in the VUV region using a pair of pump beams emitting at shorter wavelengths would be increased. However, the advantage of three-color FWRM is apparent, since the intensity of the Raman sideband can be increased drastically in the UV region by introducing the UV probe beam. It is reasonable to expect that a probe beam emitting at 200 nm could be frequency-modulated by the coherent molecular vibrations of hydrogen to generate several Raman sidebands at the same conversion efficiency. The temporal and spatial overlapping of the probe and two-color pump beams can be optimized by observing the Raman sideband at 185 nm.

Several spectra were measured at different time delays of the probe pulse from the two-color pump pulse and the results are shown in Fig. 3. No signal was observed at 185 nm, when the probe beam propagates in advance of the pump beams. When the three pulses are