High-order Raman sidebands generated from the near-infrared to ultraviolet region by four-wave Raman mixing of hydrogen using an ultrashort two-color pump beam

Osamu Shitamichi[1,*] and Totaro Imasaka[1,2]

[1]*Department of Applied Chemistry, Graduate School of Engineering, Kyushu University, Motooka, Nishi-ku, Fukuoka, 819-0395, Japan*

[2]*Division of Translational Research, Center for Future Chemistry, Kyushu University, Motooka, Nishi-ku, Fukuoka, 819-0395, Japan*

**o-shitamichi@cstf.kyushu-u.ac.jp*

Abstract: A two-color pump beam consisting of a fundamental beam of a Ti:sapphire laser (35 fs, 802 nm) and a signal beam generated by optical parametric amplification (55 fs, 1203 nm) was utilized to generate multiple Raman sidebands by vibrational four-wave Raman mixing. The second harmonic emission (401 nm) was further employed as a seed beam for enhancing efficiency. Numerous sidebands emitting at 602, 481, 344, 301, 267, 241, 219, 200, and 185 nm were observed by irradiating the beam onto a screen coated with sodium salicylate. The spectral band width of these emission lines was capable of generating 0.9-fs optical pulses by Fourier synthesis.

OCIS codes: (190.4380) Nonlinear optics, four-wave mixing; (290.5910) Scattering, stimulated Raman; (320.7110) Ultrafast nonlinear optics.

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1. Introduction

An ultrashort optical pulse can be useful for studies of ultrafast phenomena. To generate an ultrashort optical pulse approaching 1 fs, it is necessary to expand the spectral domain from the near-infrared to the ultraviolet region. Several methods for expanding the spectral region, based on nonlinear optical effects, have been reported to date, i.e., the generation of a supercontinuum, optical parametric amplification (OPA), optical parametric chirp-pulse amplification (OPCPA), and four-wave mixing (FWM) and four-wave Raman mixing (FWRM) in solid media as well as in gases.

In the past few decades, the generation of numerous rotational Raman sidebands utilizing a two-color pump beam spaced by a Raman shift frequency (587 cm^{-1}) has been reported [1,2]. This wide spectral domain can be used in the generation of an ultrashort optical pulse [3]. Impulsive stimulated Raman scattering induced by molecular phase modulation based on a pump-probe technique has been used to generate a 3.8-fs optical pulse [4–7]. The pump-probe technique has been employed for the generation of an 11-fs optical pulse in the transient regime [8–11]. In addition, the vibrational four-wave Raman mixing is more useful for expanding the spectral domain for the generation of a shorter optical pulse, because of a larger Raman shift frequency (4155 cm^{-1}). An optical pulse shorter than the molecular vibration (8 fs) should be used in the impulsive technique. On the contrary, a pair of longer pump pulses can be used for the transient Raman technique. In fact, numerous sidebands were generated using a femtosecond two-color pump beam (200 fs, 800 and 600 nm) focused on a hollow core fiber filled with hydrogen [12,13]. This approach of using a two-color pump beam (780 and 830 nm) was applied to SF_6 in the hollow core fiber for generating high-order vibrational sidebands [14,15]. On the other hand, a train of 1.6-fs optical pulses in a nanosecond envelope was generated by using a two-color nanosecond pump beam [16–19].

In this study, we report on FWRM in molecular hydrogen to generate multiple vibrational lines in the near-infrared to the deep-ultraviolet region in the transient regime. The fundamental beam of a Ti:sapphire laser (802 nm, 3ω) and a signal beam generated by OPA (1203 nm, 2ω) were employed as pump beams; the frequencies of these beams were adjusted to multiples of the Raman shift frequency (ω = 4155 cm^{-1}). A series of sidebands spaced by ω were generated by vibrational FWRM. When the second harmonic emission of the Ti:sapphire laser (6ω) was introduced as a seed beam, the frequency domain was expanded into the spectral region of 2 - 13ω. The use of the near-infrared emissions (2 and 3ω) prevents the generation of cascaded stimulated Raman emission (SRS) toward lower frequencies, since the efficiency is reciprocally proportional to the wavelength of the laser emission. As a result, it improves the efficiency of generating anti-Stokes emissions through FWRM toward higher frequencies.

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2. Experimental setup

Experimental apparatus used in this study is shown in Fig. 1. The signal beam, generated in an OPA system (1203 nm, OPerA, Coherent Inc.) pumped by a Ti:sapphire laser (802 nm, 35 fs, 1 kHz, 4 mJ), was employed to generate one of the pump beams (2ω). Gold-coated mirrors were used for beam reflections. The beam was passed through a half-wave plate, in order to produce a *p*-polarized beam. The remaining part of the fundamental beam in the OPA system was utilized as a second pump beam. These two pump beams were coaxially aligned with each other using a dielectric beam splitter. The timing between the pulses was adjusted by using a delay line controlled by a translational stage (New Focus 9067-COM-E, Newport Inc.) equipped with a picomotor actuator (New Focus 8302, Newport Inc.). An aluminum-coated off-axis parabolic mirror was used to focus the two-color beam into a Raman cell filled with a hydrogen gas to generate anti-Stokes Raman emissions by FWRM. In order to improve the efficiencies of the system, the second harmonic emission (6ω) of the Ti:sapphire laser (3ω) was generated by passing the beam through a β-BaB_2O_4 crystal (BBO). For dispersion compensation, this beam (6ω) was reflected by 2 bounces with a pair of chirp mirrors (-50 ± 10 fs^2 per bounce) since the beam was chirped slightly positively.

The spectrum of the Raman emission was measured using a spectrometer (HR4000, Ocean Optics Inc.). The spectral response of the monochromator was calibrated against the parameters obtained using a deuterium-halogen lamp (DH-2000 CAL, Ocean Optics Inc.). The data of spectrum vs. phase was measured by means of self-diffracting frequency-resolved optical gating (SD-FROG) [20]. Two types of monochromators were utilized; HR4000 for the measurement of 3ω and AvaSpec-2048-SPU (Avantes Inc.) for 6ω.

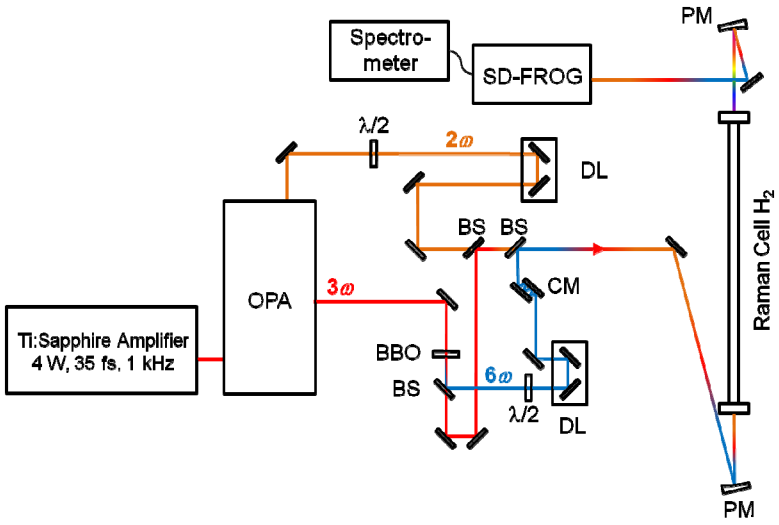

Fig. 1. Experimental apparatus: BBO, β-BBO crystal; DL, delay line; λ/2, half-wave plate; CM, chirp mirror; BS, dichroic beam splitter; PM, off-axis parabolic mirror.

3. Results and discussion

3.1. Four-wave Raman mixing induced by 2ω and 3ω

Six sidebands ranging from 4ω to 9ω were generated using a two-color pump beam polarized in parallel with each other, as shown in Fig. 2. No sideband was, however, generated, when only 3ω was introduced into the hydrogen gas. Direct FWRM induced by a two-color pump beam would be more efficient than FWRM induced by two-color beams, one of which is generated by SRS. In a previous study, a similar result was obtained using a two-color pump beam (800 nm, 70 fs, 500 μJ; 600 nm, 250 ± 50 fs, 60 μJ) and a hollow-core capillary (170

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μm i.d., 50 cm long) filled with a hydrogen gas (1.7 atm) [12,13]. In our study, the pulse widths of the pump beams were shorter and the energies were larger (55 fs, 560 μJ for 2ω; 35 fs, 650 μJ for 3ω). The spectrum appeared to be broadened, suggesting efficient self phase modulation (SPM), which can be attributed to the high peak powers of the pump beams.

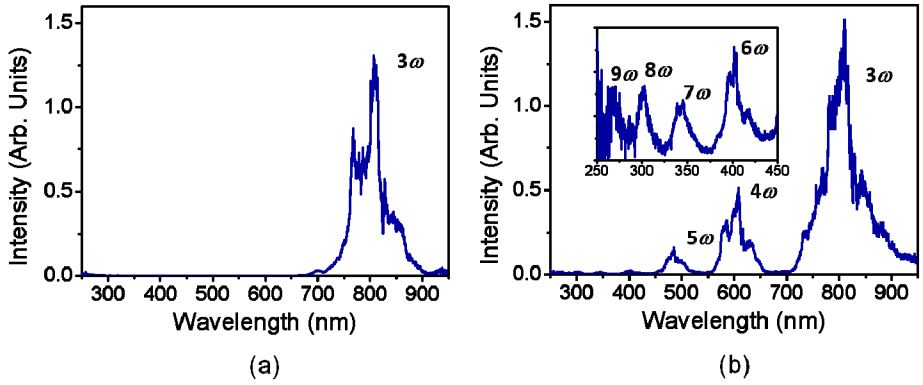

Fig. 2. Raman spectra obtained after the Raman cell when (a) 3ω and (b) 3ω and 2ω were focused into a hydrogen gas pressurized at 1.4 atm.

3.2. Enhancement of high-order sidebands using the second harmonic beam

The frequency of the second harmonic emission corresponds to the sixth order of Raman emission (6ω), and the second harmonic beam was then focused into the Raman cell to enhance the efficiency in the generation of the high-order sidebands. In the experiment, the Raman emission was separated by a prism and was projected onto a white screen (paper) coated with sodium salicylate; 4ω and 5ω were attenuated using a black screen (paper). The obtained spectrograph is shown in Fig. 3; the pulse energy was 430 μJ and 60 μJ for the fundamental beam (3ω) and the second harmonic beam (6ω) before the Raman cell, respectively. High-order Raman sidebands up to 13ω could be observed by the naked eye. Higher-order sidebands above 14ω (172 nm) would not be observed because the emission is absorbed by the surrounding air and is beyond the range of the measurement using a prism made of fused silica.

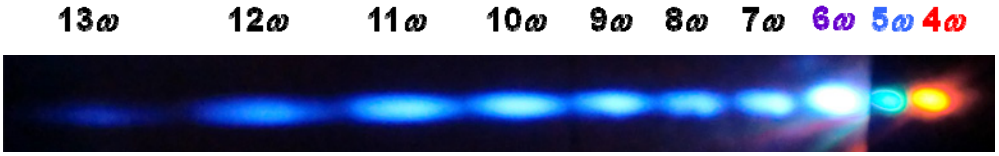

Fig. 3. Spectrograph of Raman sidebands.

When a spectrometer was used, only 7 sidebands were measured due to lower sensitivity in the deep-ultraviolet region, as shown in Fig. 4. The number of sidebands increased when the second harmonic beam was introduced. The relative intensity of the sideband was 1 (3ω), 0.2 (4ω), 0.1 (5ω), 0.01 (6ω), 0.008 (7ω), 0.006 (8ω), and 0.004 (9ω) in the absence of an SHG beam. On the other hand, the value was 1 (3ω), 0.28 (4ω), 0.12 (5ω), 0.21 (6ω), 0.03 (7ω), 0.021 (8ω), 0.016 (9ω), 0.011 (10ω) by introducing an SHG beam. This result suggests that the seed pulse assists in the generation of high-order anti-Stokes emissions. The full width at half maximum (FWHM) of the sideband tends to decrease with an increase in the order of the sideband; 866 cm^{-1} (3ω), 662 cm^{-1} (4ω), 570 cm^{-1} (5ω), 371 cm^{-1} (6ω), 487 cm^{-1} (7ω), 438 cm^{-1} (8ω), 480 cm^{-1} (9ω), 310 cm^{-1} (10ω). To explain this, it would be necessary to investigate the chirps for the pump beams which are strongly affected by SPM.

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To further enhance high-order sidebands, the third harmonic beam of the Ti:sapphire laser, corresponding to 9ω, would need to be introduced as a second seed beam. It should be noted that a larger number of sidebands can be observed when the seed beam at 6ω is introduced, even though the pulse energy of the pump beam at 3ω decreased from 650 to 430 μJ (cf. Figure 2(b) and Fig. 4(b)). Thus, additional seed beams would be useful for the enhancement of the sidebands, although the system would become more complicated.

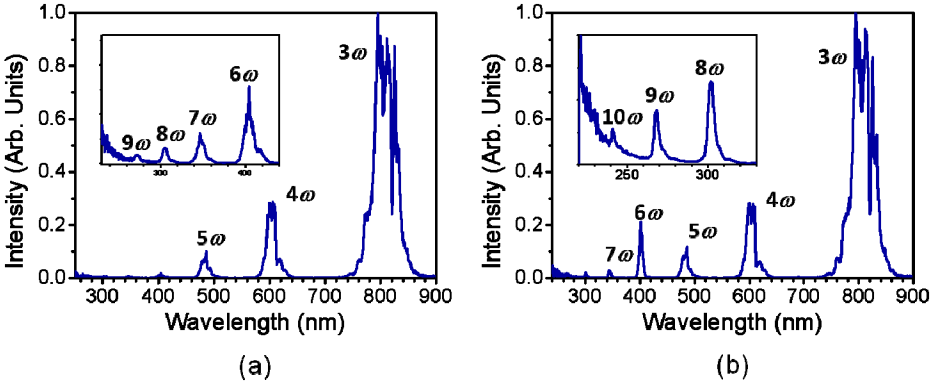

Fig. 4. Raman spectra measured after the Raman cell. (a) Two-color pump beam without a seed beam emitting at 6ω. (b) Two-color pump beam with a seed beam emitting at 6ω.

3.3. Self phase modulation of the pump and seed beams

As recognized from the uncertainty principle, a broad spectral band width is necessary for generating a short optical pulse. In addition, expanding the spectral domain for individual sidebands for the synthesis of a monocycle optical pulse is also highly desirable. One of the approaches is the use of a nonlinear optical effect. In this study, the spectral band width was expanded by SPM in gaseous hydrogen. In order to confirm this, a FROG trace of the pump pulse (3ω) was obtained at different hydrogen pressures, and the results ranging from 0 to 1 atm are shown in Fig. 5. The pump pulse was negatively chirped in front of the Raman cell in this study, probably due to a slight misalignment of the compressor in the Ti:sapphire amplifier.

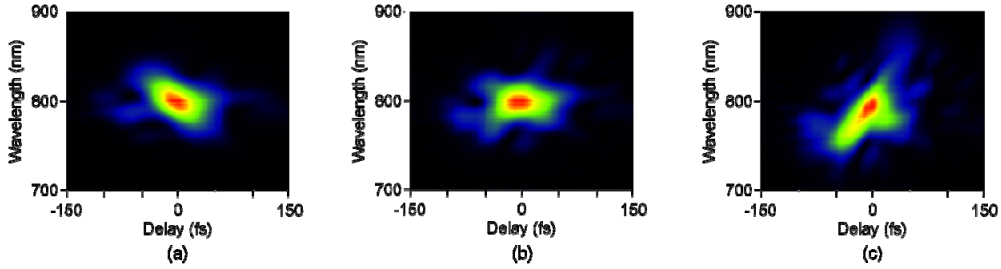

Fig. 5. FROG traces observed for a pump beam measured at different hydrogen pressures. (a) 0 atm (b) 0.5 atm (c) 1 atm.

When the pressure was increased to 0.5 atm, the spectral domain became slightly narrower and the temporal width was then slightly expanded by adding a positive chirp by SPM. The profile of the trace extended diagonally from the lower left to the upper right at 1 atm. This tendency was more significant at high pressures. Accordingly, the spectral domain can be expanded using SPM, and the resulting nonlinear optical effect would be useful for suppressing the pre- and post-pulses to generate a single optical pulse.

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The profile of the seed pulse (6ω) focused into gaseous hydrogen was measured using a SD-FROG system. The trace was diagonally stretched more significantly, as shown in Fig. 6; the spectrum and the pulse width were more efficiently stretched, probably due to the shorter wavelength, which resulted in a better focusing capability of the second harmonic beam.

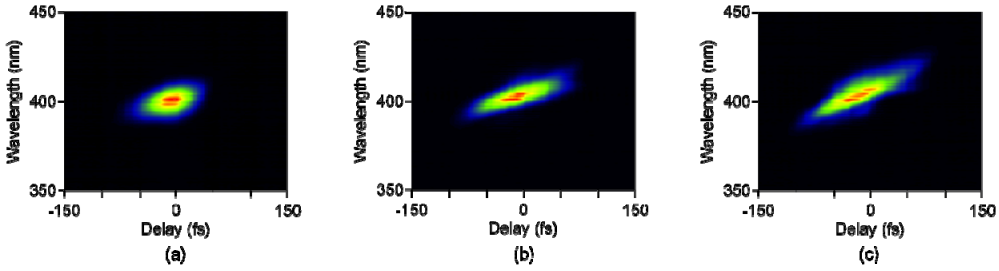

Fig. 6. FROG traces observed for a seed beam measured at different hydrogen pressures. (a) 0 atm (b) 0.5 atm (c) 1 atm.

3.4. Cross phase modulation of the pump and seed beams

In addition to SPM, cross phase modulation (XPM) should also be investigated, since two intense pump beams (2ω and 3ω) were used in this study. After generating numerous sidebands by introducing a seed beam (6ω), only the emission at 3ω was separated using a dielectric beam splitter and the FROG trace was measured at around 800 nm. The results obtained for the FROG trace are shown in Fig. 7. The pulse was appreciably chirped even at 0.1 atm. This is apparently due to XPM induced by the second pump beam (2ω). This suggests that XPM can be useful for expanding the spectral domain at lower hydrogen pressures, which is more favorable for phase matching to generate high-order Raman sidebands. The temporal pulse width, measured at 0.1 and 0.4 atm, was found to be 60 and 70 fs, respectively. The FWHM of the spectral band width was 29 and 28 nm, respectively. The FROG trace obtained at higher pressure consists of more complicated structures, which is probably due to the efficient generation of high-order sidebands and a significant loss of the portion of the pump beam (3ω) with better spectral quality.

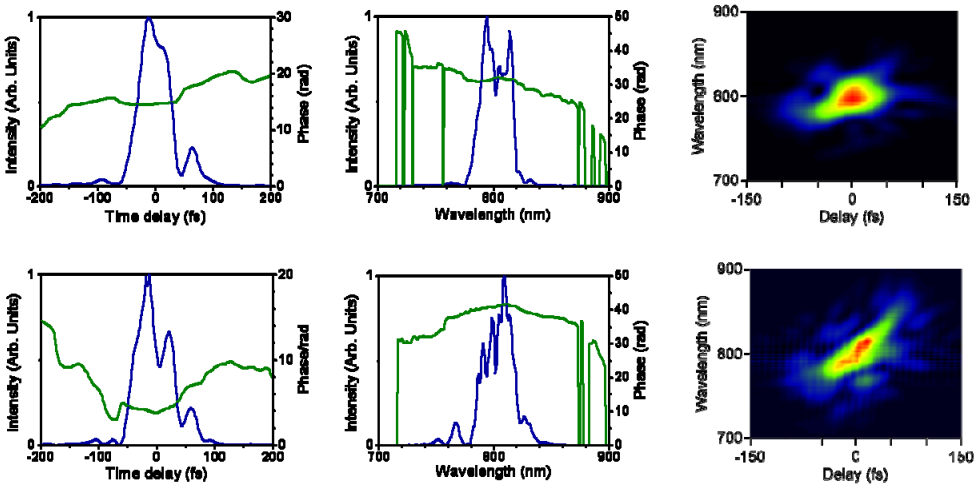

Fig. 7. FROG traced for the pump beam (3ω) when the two pump beams (3ω and 2ω) were introduced into hydrogen gas with a seed beam (6ω) to generate Raman sidebands. The hydrogen pressure was adjusted at 0.1 atm (upper trace) and 0.4 atm (lower trace).

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3.5. Pulse width

Several methods have been proposed for characterizing a laser pulse with an extremely-broad band width expanding from the near-infrared to the deep-ultraviolet region [20–23]. The pulse width that can be achieved using the high-order Raman sidebands was estimated by assuming a Gaussian profile for the intensity distribution of the emission lines observed in Fig. 4(b). The results suggest that the generation of 0.9-fs optical pulses is possible. The results reported herein suggest that the sidebands generated by FWRM using the two-color pump beam in a transition regime would have the potential for use in generating a sub-femtosecond pulse in the optical domain.

Conclusion

In this study, we employed a seed beam for the enhancement of FWRM induced by a two-color pump beam. High-order sidebands (2ω - 13ω) were generated in the region extending from the near-infrared to the deep-ultraviolet at multiples of the vibrational Raman shift frequency of hydrogen. Accordingly, the pulse width estimated from the spectral domain measured in this study approaches 0.9 fs. The findings suggest that SPM and XPM are useful for expansion of the spectral lines to generate a single optical pulse. In theory, there is no loss of energy in the FWRM process, simply reducing the pulse width of the femtosecond laser. As a result, it is possible to generate a high-peak-power laser, which would have numerous applications in science and technology.

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