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Stimulated Raman scattering and four-wave Raman mixing seeded by a supercontinuum generated in dibromomethane using picosecond and femtosecond laser sources

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Abstract

Stimulated Raman emission from liquid dibromomethane (vibrational Raman shift frequency, 588 cm^{-1}) is introduced into hydrogen gas (rotational Raman shift frequency, 587 cm^{-1}) as a seed beam, in order to generate numerous rotational lines by four-wave Raman mixing. Unexpectedly, a supercontinuum, which is generated by self-phase modulation in dibromomethane, acted as a seed beam to exclusively generate vibrational lines; the rotational lines are generated only when the supercontinuum is minimal. The former is explained by a competition between the high-gain vibrational and low-gain rotational Raman effects when strongly seeded by a supercontinuum. The latter is explained by stimulated Raman gain under the seed effect exclusively to the first-Stokes rotational line.

Keywords: Raman scattering; Dibromomethane; Hydrogen gas

1. Introduction

Stimulated Raman scattering (SRS) and four-wave Raman mixing (FWRM) are frequently employed for frequency conversion of a tunable laser, since angle phase matching is not required, making the frequency conversion system a simple one. The frequency domain can be extended to the wavelength region from the vacuum ultraviolet to the mid infrared. Recently, FWRM has been used for the generation of numerous Stokes and anti-Stokes Raman lines [1]. For example, a two-color pump beam, whose frequency is separated by 587 cm^{-1}, is introduced into hydrogen to simultaneously generate more than 40 Raman lines [2–4]. This approach is useful for enhancing the rotational lines, since this frequency separation corresponds to a resonant frequency for a rotational transition from $J=1$ to $J=3$ for hydrogen. This multi-color laser has great potential for use in illumination work and can be displayed via combination with diffractive optics such as multiple transmission gratings [5].

A mode-locking technique has been utilized for the generation of ultrashort optical pulses. In 1987, a 6-fs optical pulse was generated in conjunction with a pulse compression technique [6,7]. Although this accomplishment was shortened to 5 fs in 1997 [8,9], there are two major difficulties for further shortening of the laser pulsewidth: (1) a laser material with a wider frequency domain is required, but has practical limitations; (2) phase distortion including group velocity dispersion (GVD) in optical components requires compensation, but this is also difficult, because of its very wide frequency domain. For example, it may be possible to generate a supercontinuum (white light continuum) by self-phase modulation (SPM) in an optical fiber [6], and the GVD could, in part, be compensated using grating pairs and prism pairs. The spectral bandwidth of the supercontinuum is, however, not sufficiently broad to form a subfemtosecond optical pulse. In addition, it is difficult to compensate positive high-order nonlinear dispersions completely, by employing solid op-

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tics with negative dispersion because of the different natures of their nonlinearities.

As described previously, FWRM produces numerous, equally spaced, rotational lines. While this behavior is similar to the transverse modes of the modelocked laser, this approach has an extremely wide spectral region, e.g., from the far-ultraviolet to the near-infrared, with a considerably flat intensity profile. It is emphasized that a gas medium, in which the GVD is negligible or is small and easily compensated, is required for this type of experiment. These characteristics are preferred for the generation of ultrashort laser pulses. This multifrequency laser consisting of many high-order rotational lines has the potential for use in the generation of ultrashort optical pulses by Fourier synthesis, i.e., modelocking, of the emission lines [10,11].

In a previous study, we reported the generation and enhancement of the rotational lines by introduction of a seed beam, which is generated by vibrational SRS in liquid dibromomethane (DBM) using a nanosecond laser pulse [12]. The vibrational Raman shift frequency of DBM is 588 cm^{-1}, which is very close to a rotational Raman shift frequency of 587 cm^{-1} for hydrogen. In this report, we propose the use of this approach in the femtosecond region, because of its simplicity, for the generation of numerous rotational lines.

In this study, the generation and enhancement of the rotational emission is investigated by passing the picosecond and femtosecond beam through a DBM cell before the Raman cell filled with hydrogen. Unexpectedly, a supercontinuum generated by SPM in DBM was found to play an important role in the effect of seeding to generate vibrational lines, and the rotational lines are substantially suppressed. The explanation of this effect is given and an approach, designed to enhance the rotational lines is also discussed in this paper.

2. Experimental

A block diagram of the experimental apparatus is shown in Fig. 1. A modelocked Ti:sapphire laser (Spectra-Physics, Tsunami, 82 MHz, 100 fs, 500 mW, 800 nm) is pumped by an argon ion laser (Spectra-Physics, Beamlok 2060-7S, 6 W). The pulsewidth is extended to 200 ps by a pulse stretcher (Positive Light, TSC), in order to avoid optical damage in the amplifier. The output beam is introduced into a regenerative amplifier (Spectra-Physics, TSA), which is pumped by a second harmonic emission of a Nd:YAG laser (Spectra-Physics, GCR-6, 10 Hz, 6 ns, 600 mJ, 532 nm). The output pulse is compressed by a pulse compressor (Spectra-Physics), and the pulse energy of the laser was 8 mJ. In this experiment, the pulsewidth was adjusted within a range from 10 ps (it was not possible to exactly measure this value with the autocorrelator used in the experiment, due to over range of the specification) to 310 fs by changing the position of the mirrors in the pulse compressor. By this procedure, the pulsewidth can be adjusted without any change in pulse energy, wavelength, linewidth, and polarization. The pulsewidth was measured by a single shot autocorrelator (Positive Light, SSA). The measured spectral linewidth was 12 nm. These results suggest that the pump pulse is nearly transform limited. The unfocused pump beam propagates through a DBM cell (dimension, $2 \times 4 \times 3.7$ cm; path length, 2 or 4 cm). The pulse energy of the transmitted beam from the cell decreased to 6 mJ, mainly due to reflection losses at the cell walls. The pump beam was linearly polarized, and the beam pattern was elliptical (1×0.5 cm). The laser beam is focused by a lens with a focal length of 1 m into a 1-m Raman cell, which is pressurized to 10 atm with hydrogen (Iwatani, 99.9%). The spectrum is measured by a multichannel spectrometer (Ocean Optics, S-1000, 300 lines/mm, 300–1100 nm). The intensities of the emission lines are reduced by a combination of neutral density filters and a frosted glass placed in front of the optical fiber. A frosted glass is used, in order to avoid position sensitivity in the spectral measurements. The spectral response of the multichannel spectrometer changes within a factor of 10 in the wavelength region from 400 to 900 nm. The pulse energy of the laser was measured by a power meter (Melles Griot, 13PEM001).

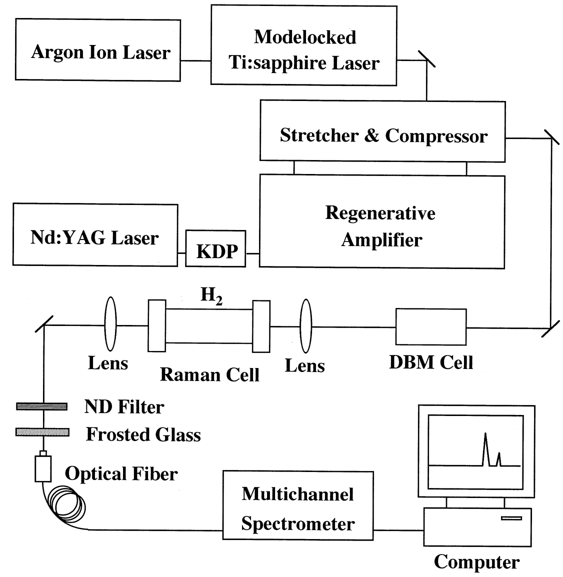

Fig. 1. Experimental setup. DBM: dibromomethane.

3. Results and discussion

Initially, the pump beam was focused with a lens having a focal length of 10 cm into a 2-cm DBM cell, in order to increase the beam intensity for the efficient generation of a seed beam for FWRM in hydrogen. The spectra of the laser beam, measured after the hydrogen cell at different pulsewidths, are shown in Fig. 2. When the pulsewidth of the pump laser is adjusted to 1.3–10 ps, the first Stokes rotational line emitting at 839 nm and the first anti-Stokes rotational line at 764 nm are clearly observed in the vicinity of the fundamental line. These emission lines were found to originate from SRS and FWRM in DBM, since no change was observed in the spectrum, even when the hydrogen cell was removed. By shortening the pulsewidth, the intensities of the rotational lines are reduced and the spectral line of the fundamental is broadened by SPM, which reduces the gains of SRS, stimulated Raman amplification (SRA), and FWRM in DBM as well as in hydrogen. No Raman emission is observable below 730 fs. In these experiments, the laser beam from the DBM cell consisted of irregular beam patterns and was unfocusable due to filament formation, resulting from self-focusing (SF) and breakdown of the DBM, which makes its use as a seed beam for the generation of rotational emission by FWRM in the succeeding hydrogen cell difficult.

In order to avoid this undesirable effect, the pump beam was unfocused in the DBM cell. In this case, the beam pattern observed after the DBM cell consisted of an intense transmitted beam and a weak supercontinuum with a large divergence angle, probably caused by SPM coupled with SF. The latter is probably formed by a limited number of filaments which occurred from hot spots which exist in the pump beam. The vibrational line, which emits at 588 cm^{-1} from the fundamental line, was not observed after the DBM cell by using the present detection system. This is mainly due to a low conversion efficiency, which is the result of the low intensity of the unfocused pump beam.

The spectrum of the beam after the hydrogen cell was measured with and without the DBM cell. As shown in Fig. 3(a), the first Stokes rotational line, in addition to the weak anti-Stokes line, is observed in the spectrum. This is explained by SRA of the first rotational line, which is assisted by a supercontinuum generated by SPM with optical components or hydrogen itself, or which could occur as the result of a slight ellipticity of the pump beam polarization, since rotational Raman scattering does not occur for a linearly polarized beam. No vibrational line is

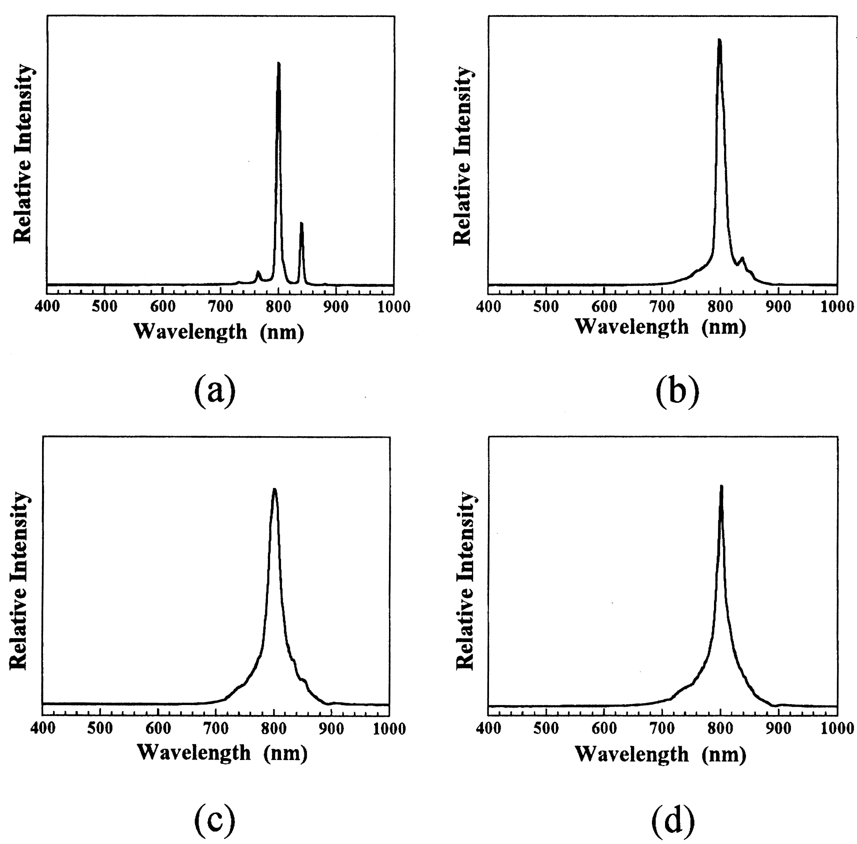

Fig. 2. Spectra of laser emissions from the hydrogen cell. A pump beam is focused into a DBM cell. The pulsewidth of the pump laser is (a) 1.3–10 ps (b) 860 fs (c) 730 fs (d) 310 fs. The pulse energy is reduced to 3 mJ after the DBM cell, in order to avoid breakdown in the cell and at the cell windows.

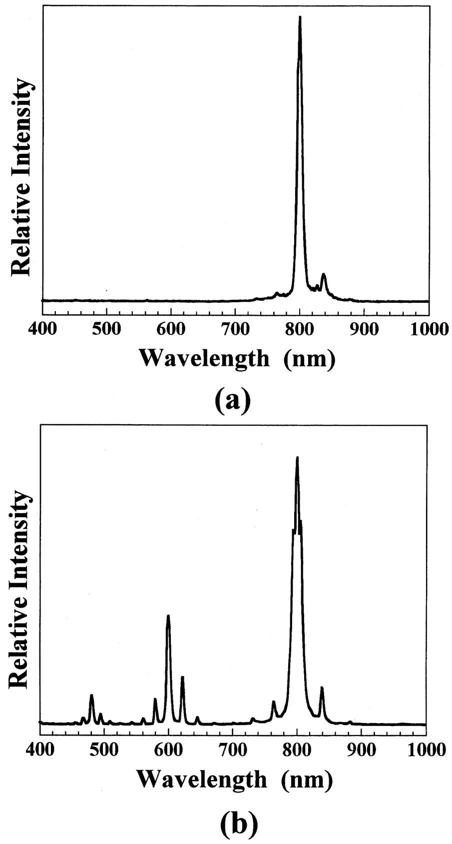

Fig. 3. Spectra of laser emissions from the hydrogen cell. The pulsewidth of the pump beam is adjusted to 1.1 ps. The pump beam is focused into a hydrogen cell after passing the beam through (a) no DBM (b) 2-cm DBM cell.

observed, probably due to a high threshold of SRS; the seed effect of a supercontinuum for the SRA of the vibrational line is negligibly small, due to a large Raman shift frequency (4155 cm^{-1}). As shown in Fig. 3(b), when the DBM cell is inserted into the beam path, numerous rotational, vibrational, and vibrationally shifted rotational lines are observed. It is interesting to note that the vibrational lines, rather than the rotational lines, are more pronounced as the result of passing the beam through the DBM. Initially, we expected an enhancement in the rotational lines by FWRM, since it is assisted by introduction of a seed beam separated by 588 cm^{-1}, which coincides with a rotational Raman shift frequency of 587 cm^{-1} and within the experimental error. In fact, pure rotational lines appeared in the vicinity of the fundamental line, which are enhanced to some extent. The vibrational and vibrationally shifted rotational lines, which previously could not be seen in Fig. 3(a), are, however, more efficiently generated. In Fig. 4, the spectra observed using DBM cells having different path lengths are shown. Remarkably, the rotational lines are nearly completely suppressed by increasing the cell length from 2 to 4 cm. The gains of SRS and SRA increase with increasing path length, and therefore the rotational lines would be expected to be enhanced more efficiently. This is in contradiction with the experimental result. This can be explained by the seed effect for the SRA of a vibrational line, which is assisted by a supercontinuum which occurred in the DBM.

When the saturation of the laser intensity is negligible, the line-broadening by SPM is proportional to the length of the interaction with a medium [13]. Thus, it is predicted that a longer DBM cell produces a stronger supercontinuum by SPM. In fact, such a supercontinuum is barely observed in the experiment using an unfocused beam. The seed beam, whose energy is in the order of 10^{-12} J, has been reported to be effective in FWRM [14]. Thus the supercontinuum extending to 4155 cm^{-1} from the fundamental line may enhance the first Stokes vibrational line at 1198 nm by SRA, although this emission line could not be measured in this study due to a low sensitivity of the detector in this region. Once the first Stokes vibrational

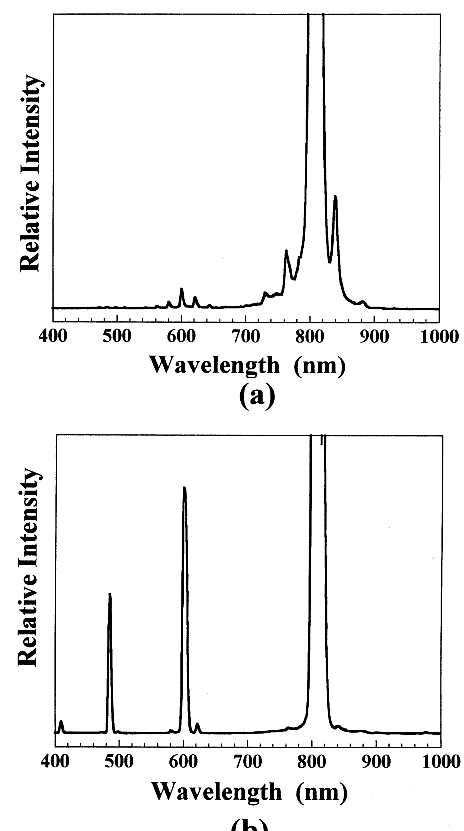

Fig. 4. Spectra of laser emissions from the hydrogen cell. The pulsewidth of the pump beam is adjusted to 1.1 ps. The pump beam is passed through (a) 2-cm and (b) 4-cm DBM cell. In the figures, the fundamental line emitting at 800 nm is scaled out, in order to observe the week vibrational and rotational lines.

line is generated, the FWRM associated with the fundamental and the first Stokes lines successively produce the first ($\lambda = 600$ nm) and second ($\lambda = 481$ nm) anti-Stokes vibrational lines. Assuming that the gains for the vibrational line occurring from SRA and FWRM are much larger than those of rotational lines, the generation of the seed beam (supercontinuum) may strongly enhance the vibrational line. The vibrational and rotational lines appear competitively, and, as a result, the rotational lines are suppressed. The present results suggest that the seed effect should be effective only to the first Stokes rotational line emitting at 587 cm^{-1} from the fundamental line and should be negligible for the first Stokes vibrational line at 4155 cm^{-1}.

The seed effect can be enhanced by line-broadening of the fundamental laser through SPM. The line-broadening is also proportional to the intensity of the laser [13]. When the pulsewidth is reduced to 700 fs, the supercontinuum was further enhanced. In this case, the first Stokes and anti-Stokes rotational lines were appreciable but very weak, and the pump energy was more efficiently transferred to SPM than to SRS, SRA, and FWRM. In addition, irregular beam patterns were observed, which may arise from SF. Thus SPM seriously suppresses the gains of the nonlinear processes related to Raman scattering in the femtosecond regime, as has been reported [15].

A supercontinuum plays an important role as a seed beam for the generation of Raman emission through SRA and the succeeding FWRM, as long as the beam focusing capability is properly retained. In the present study, H_2O and CH_3OH, common laboratory solvents, were used as media for the generation of the supercontinuum. No white light could be visually observed, which is in contrast to the case of DBM. The spectra obtained for the beam after the hydrogen cell are shown in Fig. 5. Only weak lines originating from the first Stokes and anti-Stokes emissions are observed for H_2O, indicating that the seed effect is much smaller than the case of DBM. When CH_3OH is used as a medium, no Raman emission is observed. This is probably due to a low efficiency in the generation of a supercontinuum, which results in lower efficiencies in SRA and FWRM in hydrogen. The supercontinuum is most efficiently generated with DBM among these media, which may be attributed to the large value of the third-order nonlinear susceptibility, $\chi^{(3)}$, for DBM. This is supported by the fact that Raman emission, which is more efficiently generated in a medium with a larger $\chi^{(3)}$ value, is most efficiently generated in DBM. In addition, the quality of the beam transmitted from H_2O or CH_3OH had seriously deteriorated, probably the result of heat and the convection which occurred during the experiment. This undesirable effect was more serious when a longer cell (4 cm) was used in the experiment. It is known that low quality of the pump beam decreases the conversion efficiencies of SRA and FWRM, and, as a result, these media are inadequate for the efficient generation of numerous Raman lines.

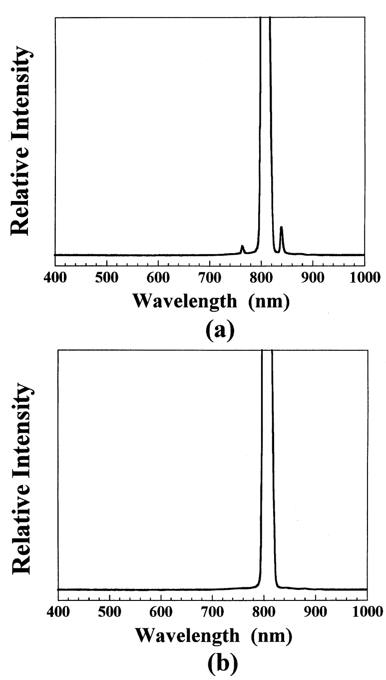

Fig. 5. Spectra of laser emissions from the hydrogen cell. A medium of (a) H_2O and (b) CH_3OH is used for the generation of a supercontinuum. The pulsewidth of the pump beam is adjusted to 1.1 ps. The cell length is 2 cm.

4. Conclusion

In the picosecond and especially in the femtosecond regime, the vibrational Raman emission arising from DBM gives a minimum or negligible effect, and, in contrast, the supercontinuum generated by SPM from DBM plays an important role as a seed beam for the generation of Raman lines by SRA and FWRM. When a supercontinuum is small, high-order rotational lines are generated by FWRM after SRA of the first Stokes rotational line. Under this condition, no vibrational lines are generated, since the gain of SRS is too low to generate the first Stokes vibrational line. When the supercontinuum is further increased, it also acts as a seed beam for the generation of the first Stokes vibrational line through SRA and assists FWRM for the generation of high-order Stokes vibrational and vibrationally shifted rotational lines. Due to the high gain for vibrational lines, the pump energy flows exclusively to the vibrational lines, and, as a result, the rotational and vibrationally shifted rotational lines are completely suppressed in such an extreme case. In order to exclusively generate high-order rotational lines, it is necessary to introduce a seed beam emitting only at the wavelength of the first Stokes rotational line and to suppress the generation of a supercontinuum at the wavelength of the first Stokes vibrational line. A possible approach for this is to optimize the conditions, in order to avoid the excess generation of a supercontinuum by SPM. The other approach might be the generation of a strong SPM for use as a seed beam to assist SRA and FWRM in generating rotational lines and the removal of unwanted emission by a filter at the first Stokes vibrational line. Liquid DBM is one of the best media for the generation of a supercontinuum, because of its ease of handling, its large value of $\chi^{(3)}$, and its favorable thermo-optical characteristics.

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