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Explicit control of spectral distribution in impulsively stimulated rotational Raman scattering

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ABSTRACT We demonstrate control of the spectral distribution of Raman sidebands in impulsively stimulated rotational Raman scattering from hydrogen gas by 70 fs infrared pump and 60 fs blue probe pulses. The results show that the energy flow direction of the probe pulse can be confined to either the Stokes side or the anti-Stokes side by changing the polarization of the probe pulse. It is found that the combination of a circularly polarized pump pulse and a linearly polarized probe pulse is the most efficient method for generating multiple Raman components.

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1 Introduction

Laser frequency conversion in the femtosecond regime is often required in fundamental and applied sciences. Stimulated Raman scattering (SRS) is one of the most useful techniques for laser-frequency conversion, for the following main reasons: the experimental setup is simple; the spatial quality of the output beam is high; the conversion efficiency from the pump pulse to the output laser is high; and it is possible to obtain various frequencies of a laser simultaneously. Although there have been some reports on the generation of multiple Raman radiation in the picosecond regime [1, 2], major difficulties arise in the femtosecond region because of the appearance of other nonlinear optical phenomena such as self-phase modulation (SPM), self-focusing, and harmonic generation. These phenomena compete with SRS and suppress the Raman gain. Thus far, the generation of more than 40 pure rotational Raman lines from H_2 has been carried out by pumping with an 800 fs 800 nm pulse [3]. In a shorter temporal region, vibrationally shifted rotational lines have been reported with pump pulses of a 200 fs 400 nm [4] and a 150 fs 248 nm laser [5]. However, the Raman lines suffer severe spectral broadening by SPM, resulting in low coherence of the Raman radiation.

Wittmann et al. demonstrated low-order rotational Raman scattering from H_2 with a 200 fs 400 nm pump laser using an impulsive excitation method [6]. This technique was originally investigated by Ruhman et al. for the observation of molecular coherent vibration [7]. In this method, the sample molecule is excited a priori by an intense pulse with a pulse width much shorter than the molecular vibrational period, so as to impulsively induce Raman polarization before the occurrence of SRS.

In this study, we report the efficient multiple generation of stimulated rotational Raman radiation from H_2, as a result of optimizing the polarization of a 60 fs probe pulse and a 70 fs pump pulse. According to our measurements, the intensity distribution of the Raman sidebands is strongly influenced by the polarization of the pump and probe pulses. We demonstrate, for the first time, the explicit control of the energy-transfer direction from the probe pulse to either the Stokes or anti-Stokes sidebands by changing the polarization of the probe laser. In particular, the selective generation of anti-Stokes radiation without generation of Stokes radiation is demonstrated. The results presented in this paper show the optimal conditions for achieving efficient SRS in the sub-100 fs region.

2 Experiment

Figure 1 shows the experimental apparatus. We use a chirped-pulse-amplified Ti:sapphire femtosecond laser system which provides ~ 785 nm, 70 fs, and 1 mJ pulses at 10 Hz. The infrared pulses are divided by a broadband 50% beam splitter for s-polarization at a 45° angle of incidence (CVI: FABS-800-45S-PW-1012-UV). One pulse is used as a pump pulse and the other pulse is transformed to the second harmonic as a probe pulse by passing it through a 100 μm thick BBO crystal. The center wavelength of the probe pulse is 389 nm and the pulse energy is 3 μJ. The delay of the probe pulse from the pump pulse is 570 fs. The pulse width of the second harmonic is estimated to be 60 fs from the Fourier transform of its spectrum, taking account of the dispersion through the transparent media. Polarizations of the pump and probe pulses are changed by rotating individual quarter-wave plates (CVI: QWPO-780-04-4-R15, QWPO-400-04-4-R15) to achieve efficient molecular rotational excitation. The two beams are focused by FL1 ($f = 50$ cm) and FL2 ($f = 80$ cm), respectively, and overlapped by a dichroic mirror (SIGMA KOKI: DIM-50S-RED) which reflects more than 90% at

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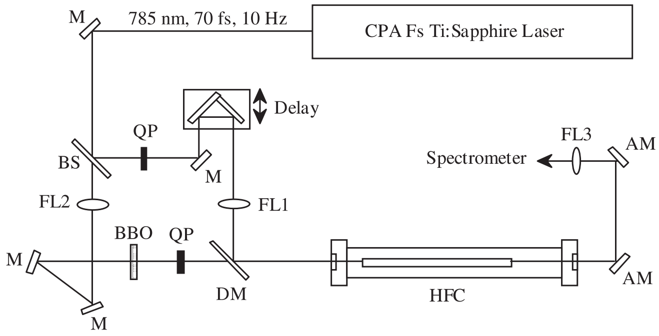

FIGURE 1 Experimental apparatus for stimulated rotational Raman scattering by two-beam pumping. M, dielectric multilayer mirror; BS, beam splitter; QP, quarter-wave plate; PS, periscope; OD, optical delay; FL, focusing lens; BBO, β-barium borate crystal; DM, dichroic mirror; HFC, hollow fiber cell; AM, aluminum mirror

800 nm and is about 80% transparent at 400 nm. Then the beams are injected into the entrance of a 75 cm long hollow waveguide (inner diameter = 126 μm) which is set in a Raman cell filled with H_2 gas at a pressure of 2 atm. The thickness of the fused silica windows for the entrance and exit of the Raman cell is 1 mm. The temporal delay between the prepump pulse and the pump pulse is optimized to achieve the highest conversion efficiency of Raman sidebands. The output spectrum from the exit of the Raman cell is detected on a shot-to-shot basis by a spectrometer (Ocean Optics: USB2000).

3 Results

Figure 2 shows the spectra of the output laser from the Raman cell. When the probe pulse, as well as the pump pulse, is circularly polarized clockwise, only Stokes components from the first to the third lines are obtained, and no signals are detected in the anti-Stokes side, as shown in Fig. 2a. On the other hand, when the probe pulse is circularly polarized counter-clockwise, only the first anti-Stokes line is observed with a weak irregular spectral structure caused by SPM in the Stokes side, as in Fig. 2c. Normally, anti-Stokes generation always coincides with Stokes generation to satisfy the coupling condition of four-wave Raman mixing (FWRM). In our study, however, no Stokes structures can be observed. According to our measurement in the case shown in Fig. 2a and c, it was found that the first Stokes and first anti-Stokes have circular polarization with a rotational direction opposite to the probe pulse, respectively. These findings indicate that the direction of energy flow from the probe pulse to the Stokes or anti-Stokes components is determined by the conservation law of angular momentum. In the process of impulsively stimulated Raman scattering, the pump pulse induces Raman coherence on a Raman-active molecule before a probe pulse interacts with the molecule, which results in the high efficiency of stimulated Raman scattering induced by such a low-energy probe pulse in the femtosecond regime. The idea of this selection rule is represented in Fig. 3. In impulsively stimulated Raman scattering, the pump pulse should be shorter than the period of the motion of the Raman-active molecule. This means that the pump pulse should have a spectral width that is sufficiently broader than the Raman shift frequency. Therefore, within the spectrum of the pump pulse, any two spectral components separated at the Raman shift frequency can efficiently induce Raman coherence on the molecule. Generally, a perfectly circularly polarized pump pulse (all spectral components have an identical circular polarization) cannot enhance the generation of the efficient rotational Raman coherence due to the limitation of the selection rule. In our experiment, it could be enhanced due to the imperfection of the circular polarization with respect to the pump pulse used. The quarter-wave plate and dichroic mirror employed here cannot realize perfect circular polarization of all the spectral components over the entire spectrum. As shown in Fig. 3, the longer and shorter wavelength components have slightly different ellipticities of the polarization. In such a case, corresponding rotational Raman coherence is produced on the molecule efficiently. After that, a clockwise/counter-clockwise circularly polarized probe pulse scatters the counter-clockwise/clockwise Stokes or anti-Stokes components, respectively, conserving the angular momentum of the system. In the Stokes process, the rotational direction of the Raman coherence should be the same as that of Stokes polarization. On the other hand, in the anti-Stokes process, it should be the same as that of

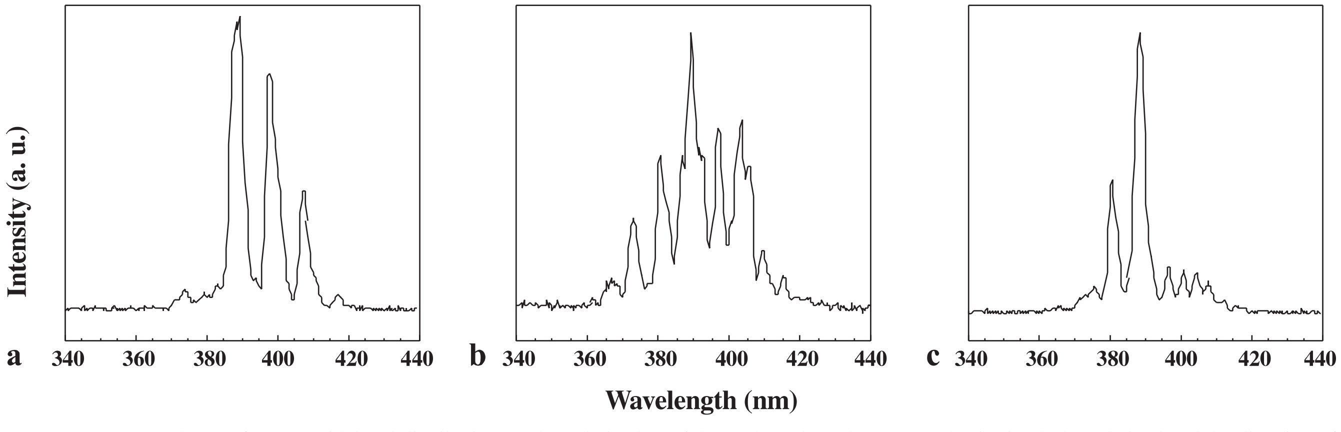

FIGURE 2 Dependence of Raman-sideband distribution on the polarization of the probe pulse. The pump pulse is circularly polarized and the direction of the polarization rotation is clockwise. The probe pulse has **a** clockwise circular, **b** linear, and **c** counterclockwise circular polarizations

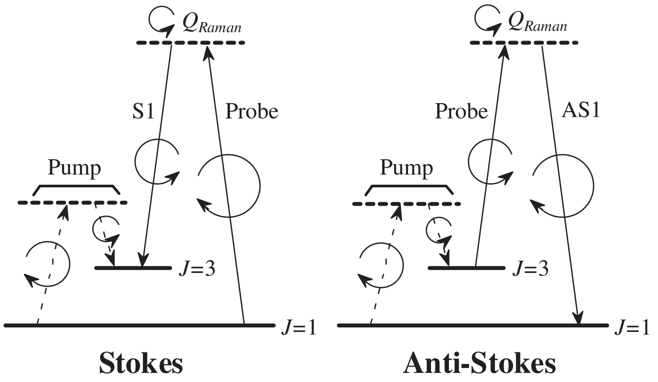

FIGURE 3 Diagram of the generation process of Stokes and anti-Stokes radiation. *Dotted arrows* are pump pulse and the corresponding first Stokes. *Solid arrows* are probe pulse and the corresponding Stokes and anti-Stokes. *Curved arrows* indicate the rotational directions of the respective circular polarizations

probe pulse polarization. This is why the clockwise circularly polarized probe pulse is preferred for Stokes generation and the counter-clockwise circularly polarized is preferred for anti-Stokes generation. In the presence of high-order Raman components, the situation is more complicated because of the effect of parametric coupling among the Raman components. When the polarization of the probe pulse is linear, Stokes and anti-Stokes lines are generated almost symmetrically, as shown in Fig. 2b, thus the linear polarization can be regarded as the synthesis of the clockwise and counter-clockwise circular polarizations. The frequencies of the Stokes lines are slightly blue-shifted due to the frequency chirp of Raman radiation in the highly transient regime [8]. When the pump pulse was linearly polarized, no Raman components could be observed, regardless of the polarization of the probe pulse, since the linearly polarized pump pulse could not induce a Raman polarization sufficiently for SRS or FWRM. It is found that the polarization of the excited molecule is a critical factor in the generation of Raman sidebands by this method.

Based on the result that the use of a circularly polarized pump pulse and a linearly polarized probe pulse is the best combination to obtain Raman components, we investigated the dependence of the intensities of Raman components on the H_2 pressure, the results of which are shown in Fig. 4. The polarization of the pump and probe pulses is the same as in the case shown in Fig. 2b. The H_2 pressure is changed from 0 to 2 atm. According to this result, stimulated rotational Raman scattering (i.e. generation of the first Stokes and first anti-Stokes line) starts at 0.4 atm, and an obvious depletion of the probe pulse energy begins at 0.6 atm. In this measurement, Raman sidebands from the third Stokes line to the fourth anti-Stokes line are observed at pressures above 1.2 atm. The intensity of the first anti-Stokes line exceeds that of the probe pulse at H_2 pressures higher than 1.0 atm. When the pressure is more than 1.6 atm, the intensity increase of the Raman components saturates, and no significant improvement can be observed. The total energy of the output beam is 3 μJ, i.e. 60% transmittance through a hollow fiber. The intrinsic efficiency of the Raman sidebands compared to the transmitted pump pulse is 84% in the output from the Raman cell.

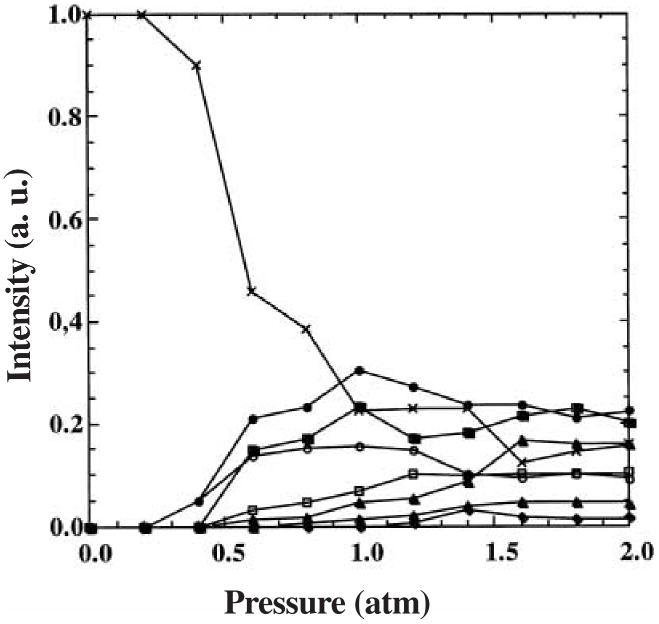

FIGURE 4 Dependence of the intensities of Raman components on the hydrogen pressure. *Crosses*, probe pulse; *open circles*, first Stokes; *open squares*, second Stokes; *open triangles*, third Stokes; *filled circles*, first anti-Stokes; *filled squares*, second anti-Stokes; *filled triangles*, 3rd anti-Stokes; *filled diamonds*, 4th anti-Stokes

4 Conclusion

In this study, we report the dependence of the intensity distribution of the rotational Raman sidebands from H_2 on the polarization of the pump and probe pulses. We confined the direction of the energy flow of the probe pulse to either the Stokes side or the anti-Stokes side to change the polarization of the probe pulse. In particular, selective anti-Stokes generation can be achieved without the occurrence of any Stokes emissions. It was found that combination of the circularly polarized pump pulse and the linearly polarized probe pulse is optimal for realizing the most efficient stimulated rotational Raman scattering. Our optimization of the experimental conditions allowed the generation of multiple Raman components in the spectral range of 360–440 nm. We can easily obtain the continuous frequency sequence in the spectral regime when the fundamental wavelength is scanned for 10 nm. We achieved an intrinsic efficiency of more than 80% from the probe pulse to the Raman sidebands in this study, i.e. the multiple-wavelength laser radiation obtained here is a promising laser source for the sub-100 fs ultrashort-time region.

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