

Polarization of multiple rotational Raman sidebands from hydrogen gas by delayed four-wave Raman mixing in the femtosecond regime

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We demonstrate delayed four-wave Raman mixing in hydrogen gas and discuss the polarization of multiple rotational Raman radiation in the sub-100-fs regime. The mechanism of sideband generation through the interaction between a probe pulse and coherence of molecular motions induced by a pump pulse in hydrogen is revealed. One can artificially control the ellipticity of the polarization of the rotational Raman sidebands by changing the pump pulse polarization. © 2002 Optical Society of America

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Recently, ultrashort optical pulses in the visible region have come to be strongly desired as probe light sources, taking into account applications such as spectroscopy. Generation of 5-fs pulses from the visible to near-infrared region by use of a supercontinuum produced by self-phase modulation in a nonlinear medium¹ and by an optical parametric generator² have been reported. These pulses have been applied for ultrafast spectroscopy.³ However, stimulated Raman scattering (SRS) has been proposed as another suitable method of broadening the spectral width of laser pulses for ultrashort-pulse formation.^{4–8} One of the advantages of SRS is its high conversion efficiency of $>10^{-1}$. In addition, the phase dispersion is, in principle, quite small in the wide spectral region compared with that of the supercontinuum from self-phase modulation. By use of nanosecond pump lasers, ultrashort-pulse-train formation has been successfully demonstrated by compensation for the phase dispersion of Raman components.⁹ In the femtosecond regime, an impulsive SRS technique has been proposed^{10,11} and experimentally demonstrated.^{12,13} Zhavoronkov and Korn have reported vibrational SRS from SF₆ by use of this technique and demonstrated a 3.8-fs pulse.¹⁴ Molecular hydrogen is also an attractive Raman medium. Rotational SRS from H₂ provides longer time intervals of pulse trains that depend on the Raman frequency shift, 57 fs from *ortho*-H₂ and 94 fs from *para*-H₂. This makes it easy to get single-pulse formation by Fourier synthesis of the Raman components. In addition, H₂ gas is transparent in the shorter-wavelength region, which is important for the spectral broadening to lead to short-pulse generation. These subjects have been analyzed numerically.^{15,16}

Here we propose a unique technique, delayed four-wave Raman mixing (DFWRM). In this technique, four-wave Raman mixing as well as impulsive SRS is separated into two processes, pumping and scattering. The point is that it is not necessary to prepare such an extremely short pulse, which has a broad spectral width including fundamental and Stokes spectral components. At first, a pump pulse

propagates through the medium and generates the first-Stokes radiation, which grows from spontaneous scattering, together with the coherent rotational motion of the molecule induced by those two pulses. After some delay during the coherence lifetime of the molecular motions, a probe pulse comes in contact with the molecule and is scattered, generating the fourth component, i.e., the Raman sideband.

Taking into account applications of the rotational Raman sidebands as light sources, we note that information about the polarization of the high-order rotational Raman components is essential for applications. This polarization has attracted much physical interest with respect to its complicated mechanism, because the angular momentum conservation law strictly influences rotational SRS differently from vibrational SRS. In this study, to explain this influence, we discuss angular momentum conservation among the rotational motion of the molecules, incident pulses, and scattered sideband pulses. We present measurements of the polarization of multiple rotational Raman sidebands and investigate the measurements' dependence on the Raman coherence in the as-prepared H₂ molecules, which gives us an overall view of the mechanism of high-order rotational Stokes–anti-Stokes sideband generation in DFWRM.

Figure 1 shows the experimental setup. Details of our laser system are described in Ref. 17. A pump pulse (wavelength 785 nm, pulse width 70 fs, energy 300 μ J, laser repetition rate 10 Hz) and probe pulse

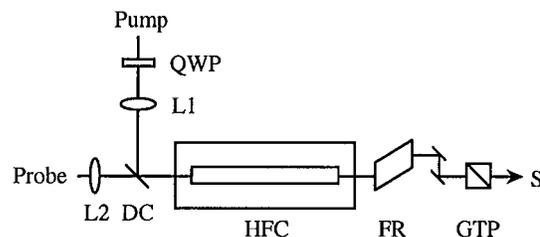


Fig. 1. Experimental setup: QWP, quarter-wave plate; DC, dichroic mirror; HFC, hollow fiber cell; FR, Fresnel rhomb quarter-wave retarder; GTP, Glan–Taylor prism; S, spectrometer.

(393 nm, 60 fs, 5 μ J, 10 Hz) were overlapped by a dichroic mirror (Sigma Koki, DIM-50S-RED), which reflected more than 90% at 800 nm and was \sim 80% transparent at 400 nm. The delay of the probe pulse from the pump pulse was set at 570 fs. Polarization of the pump pulse was changed between linear and circular by rotation of a quarter-wave plate (CVI, QWPO-780-04-4-R15), and we linearly polarized the probe pulse to obtain multiple Stokes and anti-Stokes sidebands.¹⁷ The two beams were focused by focal lenses L1 and L2 ($f = 50$ cm and $f = 80$ cm, respectively) to the entrance of a 75-cm-long hollow waveguide (inner diameter, 126 μ m), which was set in a Raman cell filled with H₂ gas at a pressure of 2 atm. In this experiment, *n*-H₂, which consists of 75% of *ortho*-H₂ and 25% of *para*-H₂, is used. Under this experimental condition, one can see the dominant contribution of *ortho*-H₂ to the frequency shift of the probe pulse.^{12,14} The thickness of the fused-silica windows at the entrance and exit of the Raman cell was 1 mm. To measure the rotational directions and ellipticities of the output pulse polarization, we used a pair of a Fresnel rhomb quarter-wave retarders (Sigma Koki, FRB-1515-4) and an analyzer (Glan-Taylor prism). The output spectra from the exit of the Raman cell were detected on a shot-to-shot basis by a multichannel spectrometer (Ocean Optics, USB2000).

Figure 2 shows the dependence of the spectrum of high-order Raman sidebands on ψ , the ellipticity of the pump pulse polarization. When $\psi = 0.0$, i.e., when the pump pulse is linearly polarized, rotational coherence of the molecule is scarcely induced because of parametric gain suppression by Stokes-anti-Stokes coupling, as shown in Fig. 2(a). When the polarization of the pump pulse has some ellipticity, high-order sidebands are apparent, as shown in Fig. 2(b). Rotational Raman components from the third Stokes to the third anti-Stokes were observed. However, when the pump pulse is circularly polarized with $\psi = 1.0$, SRS ceases up to the first Stokes and first anti-Stokes radiation, as shown in Fig. 2(c). Even when ψ is only 0.003, multiple Raman sidebands such as those in Fig. 2(b) can be clearly seen, as is the case with $\psi = 0.8$. In fact, the intensity of the first Stokes from a pump pulse with $\psi = 0.003$ is relatively lower than that from a pump pulse with $\psi = 0.8$. Nevertheless, a sufficient amount of molecular coherence can be excited for realization of distinct multiple Raman sidebands even in the case of $\psi = 0.003$.

We measured the ellipticities of Raman sideband polarization. In the cases of $\psi = 0.003$ and $\psi = 0.8$, significantly specific results were found, as shown in Fig. 3. When $\psi = 0.8$, one can apparently see the tendency that the even-order components are close to linearly polarized (ellipticity, \approx 0.1) and the odd-order components have relatively near-circular polarization (ellipticity, \approx 0.7). However, when $\psi = 0.003$, the ellipticities of both even- and odd-order components are low (ellipticity, \approx 0.3). These phenomena can be interpreted from the diagram in Fig. 4, which is defined by the well-known angular momentum conservation law. In DFWRM, the angular momentum conservation

law is described as below. For the Stokes-generation process,

$$m_{P,+} - m_{S,-} = m_{j,+} - m_{j-1,-}, \quad (1)$$

$$m_{P,-} - m_{S,+} = m_{j,-} - m_{j-1,+}. \quad (2)$$

However, for the anti-Stokes,

$$m_{P,+} - m_{S,-} = m_{j,-} - m_{j+1,+}, \quad (3)$$

$$m_{P,-} - m_{S,+} = m_{j,+} - m_{j+1,-}. \quad (4)$$

Here m is the angular momentum, P and S are the pump pulse and the first Stokes from the pump pulse, respectively, $+$ and $-$ are the right- and left-circular polarization, respectively, and j is the order of Raman radiation from the probe pulse. According to Eqs. (1)–(4), for simultaneous multiple Stokes and anti-Stokes generation, both the $+$ and $-$ components of the angular momenta should be included in both the probe and the pump pulses. In this study we linearly polarized the probe pulse to obtain the largest number of sideband components; i.e., the right- and left-circular polarized components were included

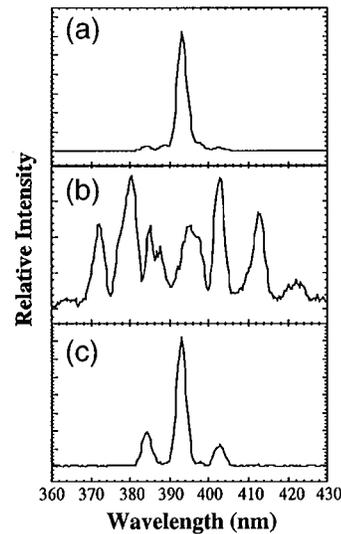


Fig. 2. Spectra of high-order Raman sidebands: (a) $\psi = 0.0$, (b) $\psi = 0.003$, (c) $\psi = 1.0$.

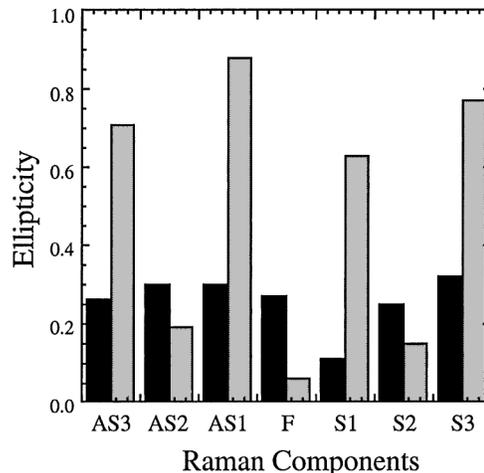


Fig. 3. Ellipticities of Raman sideband polarizations. Black bars, $\psi = 0.003$; gray bars $\psi = 0.8$.

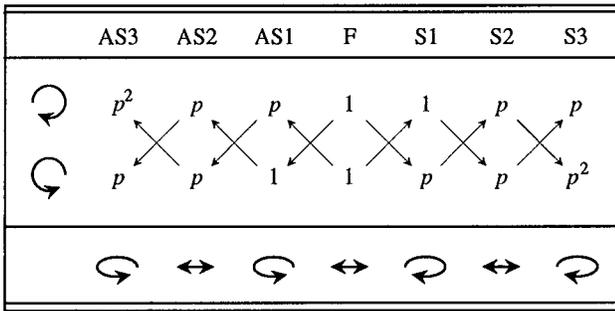


Fig. 4. Final states of the polarization of the multiple rotational Raman components predicted by theory. Right- and left-circular components are included in the molecular rotations at the rate of $1:p$ ($0 \leq p \leq 1$). If a pump pulse is linearly polarized, $p = 1$. If a pump pulse is circularly polarized, $p = 0$. The shapes of the arrows at the bottom indicate the polarization of the final output Raman components.

equally. However, if the pump pulse polarization has some ellipticity, it can provide + and - angular momenta in the molecules. If the respective numbers of Raman-active molecules that have + and - angular momenta are at the rate of $1:p$ ($0 \leq p \leq 1$), the rate of the amounts of right- and left-circular polarization of the output Raman components should be approximately $1:p$ for the first Stokes and $p:1$ for the first anti-Stokes. The final states of the output polarization of Raman radiation result from the synthesis of their right- and left-circular polarized components. This diagram shows that the even-order components have linear polarization; however, the odd-order components are elliptically polarized. According to Fig. 3, if the ellipticity of odd-order components is ~ 0.7 , $p \approx 0.1$, and for an ellipticity of ~ 0.3 , $p \approx 0.3$. Throughout the experiments, the fluctuation of these ellipticities measured with respect to the Raman sidebands was almost negligible. This leads to the important conclusion that all the processes in Eqs. (1)–(4) proceed in phase.

In this study we have investigated the polarizations of rotational Raman sidebands by use of DFWRM and given a clear explanation for the mechanism of multiple rotational Raman sideband generation. The polarizations of the sidebands are dependent on the ellipticity of the pump pulse, i.e., the rate of the rotational directions prepared in the molecules. It was found that one can artificially control the ellipticity of the polarization of the Raman sidebands by changing the pump pulse polarization. We demonstrated the generation of multiple Raman sidebands that have quasi-linear polarization by

the use of almost-linearly polarized pump pulses. Such quasi-linearly polarized multiple radiation can be a powerful source not only for pulse compression by Fourier synthesis but also for other spectroscopic applications in the femtosecond regime. In addition, we presented interesting characteristics of polarization with different ellipticities between odd- and even-order Raman components when the ellipticity of the pump pulse polarization was relatively large. Odd-order components had polarization with large ellipticity. The present results can ensure the possibility of multiple rotational Raman sidebands for a variety of scientific and industrial applications in the ultrashort time regime.

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