We investigate femtosecond stimulated Raman scattering (SRS) in H\textsubscript{2}, D\textsubscript{2}, HD, and CH\textsubscript{4} excited with 300-fs-duration pulses at 390-nm wavelength and with as much as 0.1 mJ of energy. We show that the SRS-generation threshold and conversion efficiency are due to the transient nature of SRS in femtosecond regime. We determine optimal conditions for efficient generation in the broad spectral range 289–797 nm and show how self-phase modulation and white-light generation limit the ultimate conversion efficiency. © 1998 Optical Society of America [S0740-3224(98)00912-6]

1. INTRODUCTION
In recent years there has been a rapid development of lasers that generate high-intensity ultrashort pulses. This development has prompted renewed interest in techniques of nonlinear frequency conversion of ultrashort pulses. One of the most attractive ways to convert laser light from one wavelength into a broad range of wavelengths is to use stimulated Raman scattering (SRS). Besides providing for high conversion efficiency, a special property of SRS is that one can use it to obtain coherent light simultaneously at several wavelengths. One can manipulate the exact values of these multiple Raman wavelengths and their relative intensities to a large degree by varying the active medium and by choosing from among several rotational and vibrational types of interaction.

Among known Raman-active media, the most suitable for frequency conversion of intense femtosecond pulses appear to be pressurized molecular gases such as hydrogen, deuterium, and methane\textsuperscript{1}. These gases possess several favorable properties, including large values of rotational and vibrational Stokes frequency shifts, high degrees of optical homogeneity, and relatively low dispersion of group velocity for ultrashort pulses. In addition, these gases are photochemically stable, they are not chemically aggressive, and one can vary their optical and spectral parameters by changing the pressure.

Previously hydrogen gas was used for generation of nanosecond-duration bandwidth-limited pulses with conversion efficiencies as large as 50%\textsuperscript{1–3}. In the femtosecond domain, the SRS experiments reported so far concern mostly generation of selected Raman components in H\textsubscript{2} and CH\textsubscript{4}\textsuperscript{4–6}. Recently\textsuperscript{7} we performed experiments in which we investigated generation of vibrational, rotational, and rotational–vibrational Raman components in pressurized H\textsubscript{2} by using as the pump source 200-fs-duration 390-nm-wavelength pulses from a frequency-doubled commercial regenerative amplified Ti:sapphire laser system. With this pump source it is feasible to obtain ultrashort SRS pulses in many different Stokes and anti-Stokes spectral components. By tuning the wavelength of the frequency-doubled Ti:sapphire laser in the interval 360–425 nm one can potentially cover the whole range from 280 to 630 nm.

In this paper we investigate several gases to achieve maximum broad-wavelength coverage for SRS generation without the need to change the wavelength of the pump source. We perform, for the first time to our knowledge, a comprehensive study of femtosecond SRS in the gases H\textsubscript{2}, D\textsubscript{2}, CH\textsubscript{4}, and HD. Our objective is to characterize spectral and temporal properties of various Raman components as a function of gas pressure and excitation intensity.

First we give a summary of the theoretical description of Raman gain that is relevant to a strongly transient regime. Then we discuss experimental results of measurements with femtosecond excitation pulses of SRS in several gases.

2. EVALUATION OF TRANSIENT SIMULATED RAMAN SCATTERING GAIN COEFFICIENT FOR SEVERAL GASES
Here we provide the theoretical background for understanding SRS threshold and amplification in the transient regime. Strongly transient SRS occurs when the
duration of the pump pulse $\tau_p$ is much shorter than the vibrational and rotational dephasing time $T_2$ of the gas molecules, $\tau_p \ll T_2$. In this case the nonlinear polarization of the medium does not reach steady state within the duration of the excitation pulses.\textsuperscript{8} With ultrashort-pulse excitation, phonon decay and dephasing during the pump pulse can be neglected. With the assumption of low depletions of the energy of the pump pulse, the intensity of Stokes beam at the output of the medium can be expressed as\textsuperscript{8,9}

$$I_s \sim I_0 \exp \left[ \frac{8g}{T_2} \int_0^L I_p(z, t) \, dz \, dt \right]^{1/2},$$

(1)

where $I_p$ is the intensity of the pump pulse, $I_0$ is the initial Stokes intensity, and $g$ is the steady-state plane-wave gain coefficient. The integration of the pump intensity is performed here over the length of interaction, $z = 0, L$, and over the duration of the excitation pulse, $t = 0, \tau_p$. For a diffraction-limited Gaussian beam\textsuperscript{10}

$$I_s \sim I_0 \exp \left[ \frac{16\pi g W_p^2}{T_2 \lambda_p} \right]^{1/2},$$

(2)

where the pump energy is

$$W_p = \int_{-\infty}^{\infty} P(t) \, dt$$

(3)

and $P(t)$ is the instantaneous pump power.

Note that for a focused pump beam the steady-state regime SRS threshold depends on the pump power,\textsuperscript{11} whereas the transient regime SRS threshold depends on the pump energy. If the steady-state gain $g$ and the molecular dephasing time $T_2$ are both known, we can use relation (2) to evaluate the gain for the transient regime. To find the generation threshold we use the same approach as for the steady-state regime, which consists in the assumption that the required gain to reach the SRS generation threshold by amplification of vacuum field fluctuations is $\sim e^{23,12}$. The threshold energy for transient SRS then becomes

$$W_{th} = \frac{10.5T_2\lambda_p}{g}.$$  

(4)

However, using Eq. (4) requires that we know the values of parameters $g$ and $T_2$ as a function of gas pressure. Unfortunately, reliable numerical values are not available for all molecules studied (H$_2$, HD, D$_2$, and CH$_4$) or for all pressures ($1 \times 10^5-4.5 \times 10^6$ Pa) and excitation wavelengths (390 nm) used in our experiments. To compensate for this deficiency we use a different approach in which we calculate $g$ and $T_2$ from other known molecular parameters.\textsuperscript{13-19} The results of the calculations are presented in Table 1.

As the last step, we evaluate the group-velocity mismatch (GVM) $\Delta \nu$ between the pump wave and the Stokes wave:

$$\Delta \nu = \frac{1}{u_p} - \frac{1}{u_s} = \frac{1}{c} \left( \frac{n_p - n_s - \lambda_p}{\lambda_p} \frac{dn_p}{d\lambda_p} - \frac{dn_s}{d\lambda_s} \right),$$

(5)

where $u_p$ and $u_s$ are the group velocities for the pump and the Stokes wavelengths, respectively, and $n_p - n_s$ is the corresponding difference between the linear refractive indices of the pump and the Stokes waves. For our gases, at normal conditions ($T = 300$ K, $p = 10^6$ Pa) Cauchy’s dispersion formula yields\textsuperscript{20}:

$$n - 1 = A_1 \left( 1 + \frac{B_1}{\lambda^2} \right),$$

(6)

where $A_1$ and $B_1$ are constants. By substituting Eq. (6) into Eq. (5) and by accounting for the Lorentz–Lorenz formula $(n - 1 \propto \rho)$, we finally obtain

$$\Delta \nu = \frac{pA_1B_1}{c} \left( \frac{1}{\lambda_p^2} - \frac{1}{\lambda_s^2} \right) = \frac{pA_1B_1}{c} \left( 2\nu_{ph} - \nu_{ph}^2 \right),$$

(7)

where we have introduced the Raman frequency shift $\nu_{ph}$. The value of coefficient $A_1B_1$ can be found from the literature, at least for some of our gases: H$_2$, $1.02 \times 10^{-14}$; D$_2$, $1.13 \times 10^{-14}$; and CH$_4$, $6.03 \times 10^{-14}$.\textsuperscript{21}

For HD, however, no literature value was found. In the region of normal dispersion the GVM depends both on the pump wavelength and on the frequency shift. In particular, it follows from Eq. (7) that the GVM between the first Stokes and the first anti-Stokes is approximately double that between the pump and the first Stokes. This circumstance is important for generation of higher Stokes and anti-Stokes components.

Table 1 summarizes the values of steady-state gain coefficient and the molecular dephasing time, which we then use to evaluate the SRS generation threshold for four gases for excitation wavelength $\lambda_p = 390$ nm and pressure $p = 3 \times 10^6$ Pa. The table also gives calculated values of GVM for purely vibrational SRS.

By analyzing the data presented in Table 1 we can draw certain conclusions about the character of transient SRS compared with that of the well-known stationary regime. First, in the stationary regime the generation

<table>
<thead>
<tr>
<th>Gas</th>
<th>Vibrational/ Rotational Stokes Shift $\Delta \nu$ (cm$^{-1}$)</th>
<th>Steady-State Gain Coefficient $g$ (cm/GW)</th>
<th>Dephasing Time $T_2$ (ps)</th>
<th>GVM (fs/cm)</th>
<th>Experimental SRS Threshold ($\mu\text{m}$)</th>
<th>Theoretical SRS Threshold ($\mu\text{m}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H$_2$</td>
<td>4.155/587</td>
<td>4.72</td>
<td>203</td>
<td>2</td>
<td>27</td>
<td>20</td>
</tr>
<tr>
<td>HD</td>
<td>3631</td>
<td>0.13</td>
<td>15</td>
<td>–</td>
<td>65</td>
<td>50</td>
</tr>
<tr>
<td>D$_2$</td>
<td>2997/414</td>
<td>0.79</td>
<td>88</td>
<td>1.6</td>
<td>32</td>
<td>23</td>
</tr>
<tr>
<td>CH$_4$</td>
<td>2917</td>
<td>1.2</td>
<td>16</td>
<td>8.7</td>
<td>12</td>
<td>6</td>
</tr>
</tbody>
</table>
threshold differs from one gas to another by as much as a factor of 40, with $\text{H}_2$ having the lowest value. In the transient regime, however, the lowest threshold energy is in $\text{CH}_4$, whereas all the gases listed in Table 1 have rather similar threshold values. In particular, in the stationary regime the ratio between the threshold in $\text{H}_2$ and $\text{D}_2$ possesses narrow SRS lines and exhibit a strong dependence on pressure. In the stationary regime the SRS threshold is practically independent of the gas pressure in the range $3 \times 10^6$–$5 \times 10^6$ Pa, whereas in the transient regime we can expect that the threshold will increase as the gas pressure is decreased. This circumstance can create certain difficulties in using rotational Raman components, which in the case of the stationary regime are known to be most efficiently excited at pressures of $10^5$–$10^6$ Pa.\textsuperscript{22}

### 3. EXPERIMENTAL RESULTS AND DISCUSSION

A schematic of our experimental setup is shown in Fig. 1. We used a commercial femtosecond Ti:sapphire regenerative amplifier system\textsuperscript{23} to generate 200-fs-duration pulses at the 780-nm wavelength and a pulse energy of 0.75 mJ at a 1-kHz repetition rate. Frequency doubling was performed in a 2.5-mm-thick KDP crystal. The energy of the pulses at 390 nm was 0.3 mJ, and the pulse duration was 300 fs. The gas cell was a stainless-steel cylinder with 25-mm-diameter quartz windows of two different lengths, 30 and 100 cm. The gas pressure varied in the range $10^6$–$9 \times 10^6$ Pa.

We varied the pump intensity by attenuating the pulses with neutral-density filters. We also changed the focusing conditions in the Raman cell by using spherical lenses with focal lengths $f = 25$, 50, 100, 200 cm and a telescope with variable beam-compression factors of $2 \times$, $3 \times$, and $4 \times$.

Although focusing conditions do not enter directly into Eq. (5), which describes the amplification process, we observed that focusing had a profound effect on the efficiency of SRS, basically because of the self-phase modulation and the white-light-continuum generation that occur at high peak intensities. In particular, we observed that for strong focusing with a short focal-length lens the white-light-continuum generation developed before the SRS threshold was reached. Without focusing, however, at our excitation intensities of $\sim 10$ GW cm$^{-2}$ the SRS generation threshold could not be reached. Thus we used weak focusing, so the threshold for SRS was reached before generation of the white-light continuum. Empirically, we achieved the best conversion by using a 100-cm-long Raman cell and by focusing the pump beam in the center of the cell with an $f = 100$ cm lens or by using a $4 \times$ focusing telescope.

To vary the pressure in $\text{H}_2$, $\text{D}_2$, and $\text{CH}_4$ we used the range $10^6$–$7 \times 10^6$ Pa. In the case of HD the highest pressure was limited to $2.5 \times 10^6$ Pa, as recommended by the suppliers of the gas to avoid chemical decomposition of the molecules.

In the first experiment we investigated the dependence of the conversion efficiency into the first Stokes as a function of pressure. Inasmuch as both thresholds, for SRS as well as for white-light generation, increase with decreasing pressure, we are particularly interested in achieving efficient SRS without competition from the white-light-generation process. Figure 2 shows the measured dependence in four gases when the energy of the pump pulses was held constant at $W_p = 0.1$ mJ. At this pump power there is a limited range of pressure values, where the SRS generation starts before the white-light generation. In this range the maximum efficiency is 6–16%. However, the absolute maximum efficiency of 12–18% is reached at higher pressures when the white light is already present. This result is in agreement with our previous results for $\text{H}_2$,\textsuperscript{7} for which the efficiency of 15% was achieved when the energy of the pump pulses was 0.4–0.6 mJ and the duration of the pump pulses was 300 fs.

From this measurement we conclude that nonlinear index of refraction $n_2$ of the Raman medium, which was ne-
glected in the simple description given above, gives rise to significant competing nonlinear effects. The fact that the white-light-generation threshold is similar for H\textsubscript{2} and D\textsubscript{2} at \(\sim 4.5 \times 10^6\) Pa indicates that \(n_2\) has similar magnitudes in both media. In comparison, the white-light generation in CH\textsubscript{4} begins at a pressure of \(3 \times 10^6\) Pa.

For H\textsubscript{2} and D\textsubscript{2} the conversion efficiency increased almost linearly with the increase of pressure, whereas in CH\textsubscript{4} we observed a distinct maximum in the range of \(3 \times 10^6\) Pa beyond which the efficiency started to decrease. A possible explanation for such different behavior can perhaps be found in the dispersion of group velocities, which in CH\textsubscript{4} is \(\sim 8.6\) fs cm\textsuperscript{-1}. In a 100-cm-long cell, this value corresponds to a maximum time mismatch between the pump and the SRS pulse of \(\sim 800\) fs. Even if we consider that the actual interaction length in the cell is much less than 100 cm, it appears to be sufficiently large to influence the efficiency of conversion. Another possible explanation for the bend-over in the CH\textsubscript{4} curve is that it is due to the lower white-light-generation threshold. It may turn out that stronger self-phase modulation in this gas impedes buildup of a SRS wave.

The result discussed above allows us to determine optimal pressure, which for given types of molecules is \(4.5 \times 10^6\) Pa for H\textsubscript{2} and D\textsubscript{2}, \(3 \times 10^6\) Pa for CH\textsubscript{4}, and \(2.6 \times 10^6\) Pa for HD. Figure 3 shows how the efficiency varies with the energy of the pump pulses, once the optimal pressure is selected. In the case of H\textsubscript{2}, D\textsubscript{2}, and CH\textsubscript{4} the curves exhibit characteristic saturation behavior, showing more- and less-pronounced regions with fast and slow dependence, respectively, on the pump energy. Similar dependencies were observed earlier\textsuperscript{6}; the occurrence of slow-growth regions was associated with nonlinear effects that were due to self-phase modulation of the pump pulses. The absence of a flattening out of the curve obtained in HD can be explained by a higher threshold of SRS generation, which would limit the conversion efficiency in the given pump pulse energy range.

![Fig. 3. Dependence of energy of the first vibrational Stokes pulse on the pump pulse energy for four gases. Gas pressure: in CH\textsubscript{4}, \(3 \times 10^6\) Pa; in H\textsubscript{2}, \(4.5 \times 10^6\) Pa; in D\textsubscript{2}, \(4.5 \times 10^6\) Pa; in HD, \(2.6 \times 10^6\) Pa.](image)

Figure 4 shows in more detail the dependence on pressure of the threshold of the first vibrational Stokes generation. In agreement with the results shown in Fig. 3, the lowest threshold was observed in CH\textsubscript{4}, and the threshold in H\textsubscript{2} was slightly lower than in D\textsubscript{2}. If pressure \(p\) is increased, the threshold will decrease for all gases, whereas for H\textsubscript{2}, D\textsubscript{2}, and HD this dependence is roughly proportional to \(1/p\). This dependence is due mostly to a linear decrease of the molecular coherence time, \(T_2\), with increasing pressure.

We can follow these dependences even better if we compare the theoretical and experimental threshold values summarized in Table 1. We note that the low threshold observed in CH\textsubscript{4} correlates very well with the theoretical prediction. In general, our experimental values are in good agreement with the theoretically estimated values, even if the present theory does not account for any of the accompanying nonlinear effects.

When the pump energy exceeded the threshold by a factor of 2–3 we observed emission at wavelengths corresponding to higher Stokes and anti-Stokes components. In H\textsubscript{2} at pressures up to \(4 \times 10^6\) Pa the maximum efficiency of higher Stokes components was a few percent. At the output of the Raman cell the corresponding beams were propagating in a cone centered about the direction of the pump beam. This special beam shape was caused by wave matching between the pump beam and the scattered beams.\textsuperscript{22} However, when the pressure of H\textsubscript{2} was increased to \(7 \times 10^6\) Pa the energy of the second Stokes component increased to as much as 12% of the pump energy, and the beam was propagating in the same direction as the pump. The third Stokes component as well as the first and the second anti-Stokes components was generated in a cone with an efficiency of 1–3%.

![Fig. 4. Dependence of SRS-generation threshold energy on gas pressure.](image)

Table 2 summarizes several Stokes and anti-Stokes wavelengths that are due to vibrational, rotational, and vibrational–rotational components in the gases investigated. To excite rotational and vibrational-rotational components we used circularly polarized pulses.\textsuperscript{24} The most efficient generation of rotational Stokes and anti-
Stokes components was observed in H\textsubscript{2} (Fig. 5), whereas in D\textsubscript{2} we observed only the Stokes components of the rotational spectrum. In CH\textsubscript{4} and HD no rotational components were observed. For CH\textsubscript{4} the rotational lines were absent because rotational scattering is forbidden by the symmetry of the molecule. For HD the rotational scattering is allowed. However, the corresponding spectral lines were absent because the amplification factor was insufficient.\textsuperscript{25} Note that the various frequency shifts shown in Table 2 cover a broad interval from a few to several hundred inverse centimeters.

Figure 5 shows the spectrum of Stokes components in H\textsubscript{2}, D\textsubscript{2}, and CH\textsubscript{4} excited with linearly and circularly polarized light. With the linearly polarized excitation we observed as many as three higher-order vibrational Stokes components, shifted by multiples of fundamental frequencies $\Delta \nu = 4155$ cm\textsuperscript{-1} (H\textsubscript{2}), $\Delta \nu = 2997$ cm\textsuperscript{-1} (D\textsubscript{2}), and $\Delta \nu = 2917$ cm\textsuperscript{-1} (CH\textsubscript{4}). When the polarization of the pump was changed to circular, we observed rotational Stokes and anti-Stokes satellite components shifted by $\Delta \nu = 587$ cm\textsuperscript{-1} (H\textsubscript{2}) and $\Delta \nu = 414$ cm\textsuperscript{-1} (D\textsubscript{2}) with respect to the vibrational Stokes lines. For H\textsubscript{2} the intensity of the rotational satellites was comparable with that of the vibrational SRS lines. Properties of additional Stokes components in H\textsubscript{2} are discussed in Refs. 7 and 26.

As the last measurement we obtained the temporal intensity profile of the first Stokes component in three gases. We accomplished this by using nonlinear frequency-doubling autocorrelation in a 1-mm-thick BBO crystal (type 1 phase matching). Figure 6 presents the autocorrelation functions of a SRS pulse for H\textsubscript{2}, D\textsubscript{2}, and CH\textsubscript{4}. For reference, the autocorrelation of the pump pulse, measured at the input to the SRS cell, is given. The energy of the pulses was 50 \(\mu\)J for H\textsubscript{2} and D\textsubscript{2} and 30 \(\mu\)J for CH\textsubscript{4}. The spectrum of the first Stokes pulse in D\textsubscript{2} is shown in Fig. 5(c).

We observed that the duration of the SRS pulse experienced appreciable shortening with respect to the duration of the pump pulses. This effect was most pronounced in H\textsubscript{2} and was less obvious in CH\textsubscript{4}. The explanation for this phenomenon may involve the fact that in the transient regime the SRS pulse is formed on the trailing edge of the pump pulse.\textsuperscript{27} We are preparing a detailed analysis of the femtosecond SRS pulse forms, including the effects of GVM.

We mentioned above that competing nonlinear processes such as white-light-continuum generation appear to reduce the conversion efficiency, especially with increasing pump power. Nonlinear self-phase modulation manifests itself also as spectral broadening of the Raman lines, especially those closer to the excitation wavelength. Figure 7 illustrates this observation; for this figure we used a grating monochromator to monitor the spectral intensity profile of the radiation at the output of the cell. First we measured the spectrum of the transmitted pump (Rayleigh scattering) at several powers. Below the SRS threshold the spectrum is identical with that at the input to the cell. Above the threshold we first observe a gradual broadening of the spectral line shape to a factor of 3 to 4 (measured at half-maximum). After that the

<table>
<thead>
<tr>
<th>Gas</th>
<th>AS2–AS1</th>
<th>AS2–S1</th>
<th>AS1–AS1</th>
<th>AS1–S1</th>
<th>O–AS1</th>
<th>O–S1</th>
<th>S1–AS1</th>
<th>S1–S1</th>
<th>S2–AS1</th>
<th>S2–S1</th>
<th>S3–AS1</th>
<th>S3–S1</th>
</tr>
</thead>
<tbody>
<tr>
<td>H\textsubscript{2}</td>
<td>289</td>
<td>294</td>
<td>300</td>
<td>330</td>
<td>336</td>
<td>342</td>
<td>381</td>
<td>399</td>
<td>453</td>
<td>465</td>
<td>478</td>
<td>558</td>
</tr>
<tr>
<td>HD</td>
<td>342</td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>D\textsubscript{2}</td>
<td>344</td>
<td>349</td>
<td>354</td>
<td>396</td>
<td>384</td>
<td>441</td>
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<tr>
<td>CH\textsubscript{4}</td>
<td>350</td>
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</tr>
</tbody>
</table>

\textsuperscript{a} AS1, AS2, and second anti-Stokes, respectively; S1, S2, S3, first, second, and third Stokes, respectively; AS2–AS1, combination of first rotational anti-Stokes with second vibrational anti-Stokes.
most intense part of the spectrum suddenly goes back to
the initial narrow width, whereas on the Stokes side of
the spectrum there appears a broad structureless back-
ground that is due to white-light generation. The aver-
age intensity and spectral extent of the broad spectral
component increase with increasing the pump power,
gradually filling in the gap between the pump and the
first Stokes, and extend further toward higher Stokes
components.

Different molecules showed different spectral behav-
iors as functions of pump intensity. In H_2 the Stokes
lines broadened simultaneously with the broadening of
the pump beam spectrum. At intensities above the
white-light-generation threshold the Stokes spectrum had
a similar broad wing, which was dropped off toward
longer wavelengths. In CH_4, however, a much different
situation was observed. In particular, the broadening of
the Stokes lines started at lower pump intensities, even
before the broadening in the pump beam itself occurred.
In some instances we were able to observe that the width
of the Stokes lines was broader than that of the pump by
a factor of 3 or 4, approaching 400 cm^{-1}. Again, when
the power of the pump was increased further, the Stokes
lines collapsed to 100–150 cm^{-1}, and the intensity of
broad background intensity dropped as well.

Considerable differences were observed also in the
character of the broadening of the Rayleigh spectrum at
high excitation power. In H_2 the spectrum broadened in
the direction of longer wavelengths, whereas in CH_4 the
broadening was mostly in the anti-Stokes region. We
can attribute these peculiarities to possible different
wavelength dependences of the gain factor and different
values of the nonlinear index of refraction in H_2 and CH_4.
The exact mechanisms of these processes will be the sub-
ject of our further investigation.

4. CONCLUSION

To summarize, we have investigated in detail the effi-
ciency and the spectral–temporal characteristics of SRS
in the femtosecond time domain for gases such as H_2, D_2,
HD, and CH_4. Experimental conditions for efficient con-
version of the wavelength of energetic femtosecond pulses
with a possibility of choosing from among many vibra-
tional and rotational Raman shift frequencies were deter-
dined. A 18% conversion efficiency to the first Stokes
component in H_2 was achieved, despite the fact that we
were dealing with a strongly transient regime. In addi-
tion, a theoretical analysis of the transient amplification
factor and the SRS-generation threshold was presented
for H_2, D_2, HD, and CH_4 and was in good accord with our
experimental results.

Despite evidence of self-phase modulation and white-
light-continuum generation at high intensities, it is pos-
sible by taking into account the properties of the various
gases to apply this technique to efficient frequency con-
version of commercial amplified Ti:sapphire lasers in a
fairly broad spectral range, extending from the near UV
to the near IR. Efficient generation of first, second, and
third Stokes and anti-Stokes lines demonstrated possibil-
ity of achieving femtosecond-pulse generation in the spec-
tral range 280–797 nm simultaneously in as many as 10
lines. The principal advantage of this technique consists in its relatively simple operation principle and high efficiency, especially when it is compared with parametric amplification in nonlinear crystals.

By analyzing the spectral characteristics of the Raman and the Rayleigh components of the signal we found evidence that self-phase modulation and white-light generation, in fact, limit the ultimate conversion efficiency. These nonlinear processes, which occur at high excitation intensity, broaden the spectrum and distort the phase front of the pump pulse and thus act to deplete the intensity of the pump.

Further investigations are needed to clarify the role of competing nonlinear processes, which tend to broaden the spectrum of the SRS signal and to be responsible for limiting efficiency because of distortion of the phase front of the pump pulses.

ACKNOWLEDGMENTS
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