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Raman Shifting a Tunable ArF Excimer Laser to Wavelengths of 190 to 240 nm With a Forced Convection Raman Cell

R. Jeffrey Balla and G. C. Herring
Langley Research Center, Hampton, Virginia

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Abstract

Tunable radiation, at ultraviolet wavelengths, is produced by Raman shifting a modified 285-mJ ArF excimer laser. Multiple Stokes outputs are observed in H₂, CH₄, D₂, N₂, SF₆, and CF₄ (20, 22, 53, 21, 2.1, and 0.35 percent, respectively). Numbers in parentheses are the first Stokes energy conversion efficiencies. We can access 70 percent of the frequency range 42000–52000 cm⁻¹ (190–240 nm) with Stokes energies that vary from 0.2 µJ to 58 mJ inside the Raman cell. By using 110 mJ of pump energy and D₂, the tunable first Stokes energy varies over the 29–58 mJ range as the wavelength is tuned over the 204–206 nm range. Dependence on input energy, gas pressure, He mixture fraction, and circulation of the gas in the forced convection Raman cell is discussed. Stokes conversion is also discussed for laser repetition rates from 1 to 100 Hz. An empirical equation is given to determine whether forced convection can improve outputs for a given repetition rate.

1. Introduction

Molecules such as H₂, O₂, N₂, NO, N₂O, H₂O, CO, and CO₂ have spectroscopic transitions in the region from 190 to 240 nm (ref. 1). These molecules are important for many processes, which include combustion chemistry, atmospheric chemistry, and high-speed wind tunnel testing. For example, we are interested in developing diagnostics for O₂ because many wind tunnels use air, and N₂ is more difficult to detect than O₂.

A variety (refs. 2 to 5) of methods for the generation of laser beams in the far ultraviolet (UV) at 200 to 250 nm and vacuum UV (VUV) at ≤200 nm have been pursued. Stimulated Raman scattering (SRS) from an ArF laser is one (refs. 6 to 8) of many approaches. To avoid the disadvantages of multiple lasers, a single laser solution is desirable. Thus, we studied SRS in a forced convection Raman cell (FCRC), which is pumped by a tunable 285-mJ ArF laser. The laser was modified to produce 220 times the spectral brightness of the original commercially available system. Additional excimer-based studies of SRS in H₂ isotopes are in reference 9. Most previous ArF studies used bandwidths ≥1 cm⁻¹ and pump energies <100 mJ in static Raman cells. A localized gas flow to replenish gas near the focal volume was previously used in reference 10.

The approach in this paper is oriented towards Stokes production and has several advantages. First, it offers a passive and single-laser method for generating tunable coherent radiation at frequencies outside the laser gain profile. Second, because the Raman gain coefficient increases linearly with increasing pump laser frequency, ArF pumping is enhanced compared with lower frequency sources. Third, ArF pulse energy is high compared with many other laser sources. Last, in specific conditions in some gases, SRS perturbs the gas medium and reduces conversion efficiency on successive pulses. To maintain conversion, the gas medium along the entire length of interaction with the laser beam was replenished between pulses with a commercially available FCRC. The FCRC uses an internal fan to circulate continuously the gas medium transverse to the laser beam propagation direction. This circulation allows improvement of the SRS conversion at higher repetition rates. Short test times (typically 1 to 60 s) in high-speed wind tunnels combined with high run-time costs often dictate a high-repetition-rate laser. This approach also has disadvantages. ArF laser tunability is limited to approximately 300 cm⁻¹. Hence, multiple Stokes shifts in several gases will be required to access a wide frequency range. Generally, excimer-based Raman beam spatial profiles are poorer than YAG-based profiles. Also, absorption of ArF radiation is a problem in many gases.

In this work, the conditions producing optimal SRS are determined empirically by studying conversion efficiency versus relevant parameters for stimulated vibrational Raman scattering (SVRS) and stimulated pure rotational Raman scattering (SRRS) in H₂, D₂, CH₄, N₂, SF₆, and CF₄ at room temperature. The largest tunable Stokes pulse energy of 58 mJ (before exiting the cell), obtained near 205 nm, is
larger than that achieved in earlier studies (refs. 7 and 11). SRS conversion efficiency versus laser repetition rate is measured from 1 to 100 Hz. Results are obtained with and without the FCRC and include an empirical rule to determine when forced convection will be helpful.

2. Stimulated Raman Scattering

When a plane wave of intensity $I_p$ and frequency $\nu_p$ (pump wave) traverses a Raman active medium parallel to the x-axis, a fraction of the intensity at $\nu_p$ is converted to a copropagating wave at frequency $\nu_s$ (Stokes wave) through the Raman scattering process. The conversion of pump wave to Stokes wave is described by (ref. 12)

$$I_o = I_i \exp(g l_p L)$$

where $I_i$ is the initial Stokes intensity (J/sec-m$^2$) at $x = 0$ and $I_o$ is the amplified Stokes intensity at $x = L$ after traversing a distance $L$. If the product $g l_p L$ is $<1$, then equation (1) reduces to

$$I_o - I_i = I_i g l_p L$$

In this limit, the generated Stokes intensity, $I_o - I_i$, is linearly dependent on $I_i$ and is determined from the product of two different intensities, $I_i$ and $I_p$. The plane-wave, steady-state Raman gain coefficient $g$ (m/W) (ref. 9) is

$$g = \frac{2\lambda_s^2}{\hbar \nu_s \pi} \frac{d\sigma}{d\nu}$$

where $\lambda_s$ is the Stokes wavelength, $\Delta\nu$ is the full width at half maximum (FWHM) Raman bandwidth, $d\sigma/d\Omega$ is the differential Raman cross section for the chosen Raman transition, polarization, and scattering direction. The population difference between the initial and the final levels is $\Delta N$. Usually $\nu$ and $J$ denote vibrational and rotational quantum numbers. Since $d\sigma/d\Omega$ varies as $\nu^4$, $g$ varies as $\nu$; hence $g$ is enhanced for the ArF laser compared with all lower frequency light sources.

If a second wave at the Stokes frequency $\nu_s$ is also injected with the pump wave such that it copropagates with the pump, then this second wave will act as the seed $I_i$ for the conversion process. In Raman shifting, we are interested in one-laser configurations where only the pump wave at $\nu_p$ initially enters the Raman medium. Then the initial Stokes intensity $I_i$ originates from spontaneous Raman scattering that occurs in a small solid angle centered on the forward direction. Equation (1) can still be used to calculate the conversion to the Stokes frequency if the strength of the spontaneous Raman scattering can be estimated.

Equations (1) to (3) describe conversion for an ideal plane wave, but are not adequate for accurate calculations in the more practical case of a focused Gaussian-shaped laser beam. For Gaussian beams, one can integrate over the focal region to get (refs. 9 and 12)

$$P_o = P_i \exp\left[\frac{4g P_p \arctan(L/b)}{\lambda_p + \lambda_s}\right]$$

where $b$ is the confocal parameter, $\lambda_p$ is the wavelength of the pump beam, $P_i$ and $P_o$ are the powers (J/s) of the Stokes beam at $x = 0$ and $x = L$, respectively, and $P_p$ is the pump beam power.

Equations (1) and (4) illustrate how the generation of Stokes light is nonlinearly dependent on $I_i$ or $P_p$. This Raman conversion to Stokes energy also exhibits a threshold behavior. As the pump intensity is increased from very small values, one observes relatively little conversion to the Stokes wavelength. Near the threshold intensity, $I_{th} = I_{th}$, the conversion dramatically increases as $I_p$ increases only slightly. Finally, the fraction of light converted becomes roughly constant as $I_p$ is increased further. Most of the conversion occurs over a small range of pump intensities near $I_{th}$.

For a focused Gaussian beam in a Raman medium much longer than the confocal parameter, the pump beam power $P_{th}$ needed to achieve threshold for the Raman process is (refs. 9 and 13)

$$P_{th} = \frac{25 \lambda_p}{\pi g}$$

Threshold is defined as the pump power necessary to obtain $P_o/P_i = \exp(25)$ in equation (4). This empirical definition is based on a variety of experimental observations. The ratio $P_o/P_i$ represents the total integrated gain through the focal region. This gain is sometimes expressed in nepers (Np). One Np is defined as a gain of $\exp(1)$, thus $\exp(25) = e^{25} = 25$ Np.
3. Experiment

3.1. ArF Excimer Laser Configuration

A schematic diagram of the experiment is shown in figure 1. The pump laser consists of a modified Lambda Physik model LPX250T pulsed, dual-discharge tube tunable ArF excimer laser. A detailed description of the characteristics and modifications of this design has been presented in reference 14. This design has a bandwidth of 0.33 cm\(^{-1}\), a FWHM pulse of 15 ns, peak locking efficiency of 98 percent, and tunability from 51570 to 51870 cm\(^{-1}\) with >30-percent locking efficiency. The output beam is 8 mm high by 10 mm wide, diverges nine times the diffraction limit of a 10-mm-diameter Gaussian beam, and has a horizontal-to-vertical polarization ratio of 35:1. Overall, spectral brightness is increased 2 orders of magnitude when compared with the standard unstable-resonator amplifier design. With new discharge tube windows, the output is 285 mJ. As the MgF\(_2\) windows absorb water and become dirty, the output can significantly decrease.

All optics (outside the laser) in figure 1 are uncoated Heraeus Amsersil Suprasil quartz with focal lengths specified for 193-nm radiation. Those between the laser output and center of the FCRC transmit 70 percent, giving a pump energy of 200 mJ inside the cell if the laser produces 285 mJ. The beamsplitter before the FCRC diverted a fraction of the backward stimulated Brillouin scattering (SBS) and backward SRS. These were measured following dispersion by a prism. The optics on the output side of the FCRC also transmitted about 70 percent of the output Stokes wavelengths. Thus usable pulse energies after the final dispersing prism are about 70 percent of what is generated inside the cell.

3.2. Forced Convection Raman Cell

The FCRC consists of a closed Raman cell with an internal fan to circulate continuously the gas medium transverse to the laser beam propagation direction. It removes or dilutes the gas medium that has been perturbed by SRS on prior laser pulses. This device is available commercially from Light Age, Inc. Using the manufacturer’s lens or window thickness of 0.95 cm, the maximum recommended operating density was 157 amagats (2500 psi at 296 K). Densities are specified in amagats, where 1 amagat is the density of an ideal gas at standard temperature and density (273.15 K and 1 atm). The FCRC contains an internal variable-speed fan with four blades. Each blade extends outward 0.95 cm from the main shaft. The shaft and blades run parallel to the laser beam, span the length of the FCRC, and circulate the gas transverse to the laser beam. A 0–24 V DC (direct-current) motor drives the fan. Fan tip rotation was measured at atmospheric density and was found to be linear with applied voltage. The maximum of 24 V corresponds to 100 rps and a fan tip speed of 600 cm/s, but only at 1 atmosphere.

All gases were research grade purity except D\(_2\) (99.7+ percent), CF\(_4\) (99.97 percent), and SF\(_6\) (99.999 percent). The gases in the following table with the manufacturer’s stated minimum purities were used:

<table>
<thead>
<tr>
<th>Gas</th>
<th>Minimum purity, percent</th>
</tr>
</thead>
<tbody>
<tr>
<td>H(_2)</td>
<td>99.9999</td>
</tr>
<tr>
<td>D(_2)</td>
<td>99.7+</td>
</tr>
<tr>
<td>CH(_4)</td>
<td>99.995</td>
</tr>
<tr>
<td>CF(_4)</td>
<td>99.97</td>
</tr>
<tr>
<td>SF(_6)</td>
<td>99.999</td>
</tr>
<tr>
<td>N(_2)</td>
<td>99.9995</td>
</tr>
<tr>
<td>He</td>
<td>99.9999</td>
</tr>
</tbody>
</table>

Several cell geometries were used to vary confocal parameter. The first, in figure 1, uses a lens (2 in. diameter by 65 cm focal length) to reduce the beam diameter before entering the FCRC input window (12 mm diameter). This eases alignment difficulties and focuses more energy into the cell than an integrated (i.e., a lens also acting as the Raman cell window) optic. A second geometry used a 1-m long cell with integrated optics (50-cm focal length). A third geometry combined 0.5 and 1.0 m FCRC’s to produce a single cell (with two internal fans) with a length of 1.5 m and integrated optics (80-cm focal length). The FCRC window-seal design required densities above 1.2 amagats to maintain gas seal integrity.

3.3. Optimization of SRS

Output Raman energy was studied as a function of several parameters:
Energy measurements were made of the incident pump, transmitted pump $Q_0$, first SVRS Stokes order $Q_1$, and higher Stokes orders $Q_n$. Surrounding each $Q_n$, a series of equally spaced SRRS transitions on both the high and low frequency side was observed. SRRS transitions on the lower frequency side are designated $Q_{n+1}$, $Q_{n+2}$, $Q_{n+3}$, ..., whereas those on the higher frequency side are designated $Q_{n-1}$, $Q_{n-2}$, $Q_{n-3}$, .... With this notation, the first Stokes pure rotational shift is $Q_{0+1}$, the second Stokes pure vibrational shift is $Q_2$, and the third anti-Stokes rotational shift off the first Stokes vibrational shift is denoted by $Q_{1-3}$. Multiple detectors were used to verify detector calibrations. These detectors included both pyroelectric (Molectron J4-09, Laser Precision model RJP-735-RO, and Coherent model LMP10) and calorimeter energy meters (Coherent model LM100E and Scientech Model 380103). All detectors agreed within 10 percent over the appropriate energy ranges. Single pulse spatial profiles of selected beams were captured with a CCD camera.

Typically, 50 to 90 percent energy conversion was observed into the sum of all Stokes transitions. Anti-Stokes production occurs at VUV frequencies. No attempt was made to measure vibrational anti-Stokes yields, where the room air and optics strongly absorb radiation below 190 nm. The few anti-Stokes transitions observed were pure rotational shifts with wavelengths above 190 nm.

4. Results

When a high-intensity laser is focused into a gas, SRS and four-wave mixing (FWM) occur (ref. 15) simultaneously. The first is cascade SRS where $Q_n$ becomes intense enough to generate $Q_{n+1}$. SRS exhibits a threshold behavior; that is, at a specific input $Q_n$, intensity, the conversion to Stokes light $Q_{n+1}$ dramatically increases in a nonlinear manner. The second process is FWM using four consecutive $Q_n$ orders. FWM processes have no threshold but do require phase matching. Cascade SRS tends to dominate the production of low order ($n = 1,2$) $Q_n$ and FWM tends to dominate the production of higher orders ($n \geq 3$) (ref. 7).

To maximize the energy in $Q_1$ or $Q_2$, FWM should be minimized. FWM can be reduced by two methods (ref. 16). Large densities will prohibit phase matching in the forward direction, whereas long focal lengths eliminate off-axis propagation angles that satisfy the phase-matching condition in the near-forward direction.

The pump-beam focusing was primarily constrained by the FCRC length. A focal length too short requires a collimating lens internal to the cell, and one too long may burn the cell windows. We measured conversion in $H_2$ and $N_2$ for 50- and 65-cm focal lengths at 193 nm. We used pump energies above 50 mJ because this study seeks maximum output energies, not high conversion efficiency. Below 10 amagats, $Q_1$ and SRRS transitions are larger with the 65-cm focal length. Above 10 amagats, $Q_2$ generally dominates the output spectrum. Conversions for $Q_1 - Q_3$ vary less than 25 percent for the two focal lengths. Unless stated otherwise, a 65-cm focal length was used for this work; the few times that a 50-cm lens was used are clearly stated. Optimum energy production into any desired order is achieved by varying density.

The following conditions apply to all results unless stated otherwise. Absolute energy measurements were made of the incident pump energy, the residual pump energy $Q_0$, and SRS energy in all observed Stokes orders. All conversion efficiencies and all pulse energies for the transmitted pump $Q_0$ and Stokes beams are values inside the FCRC after correction for transmission losses of 30 percent exiting the cell. Since the input optics of the Raman cell also attenuate the pump energy by 30 percent, pump energy values listed on the x-axis of the figures in this report have been reduced by 30 percent to give energy values inside the Raman cell. Conversion efficiencies are listed as percent energy conversions inside the cell. All experiments were performed at 296 K. Those that varied pump energy, confocal parameter, He mixture...
fraction, or gas density are performed at 10 Hz with a 1-m-long FCRC operating at 24 V on the fan motor. All experiments, except the few times that the laser was scanned, are performed with the laser tuned to 51714 cm⁻¹.

4.1. H₂

The vibrational Raman shift of H₂ is 4155.3 cm⁻¹ (ref. 17). Stokes Q-branch conversion versus H₂ density is shown in figure 2 using 83 mJ of pump. Although not shown in the figure, the Stokes outputs remain constant at the 60-amagat values out to 100 amagats. Attempts were made to optimize Q₁ at pump energies above 100 mJ by adding 50-amagat helium or argon (refs. 16 and 18) to 50-amagat H₂. Energy conversions Q₁ – Q₃ were unaffected by these additions. Addition of 6-amagat N₂ to 50-amagat H₂ increased Q₁ conversion by 20 percent at the expense of Q₂ – Q₃. Energy conversion Q₁ – Q₃ versus pump energy is shown in figure 3 for 20-amagat pure H₂. Experiments performed with the FCRC fan on and off show no measurable effect on any Q₁ – Q₃ over a range of 10- to 30-Hz repetition rates.

At H₂ densities <6 amagats, a series of transitions were observed surrounding each Qₙ transition, and this is shown in figure 4. To determine if these Raman orders are pure rotational (ΔJ = 2) or O and S branch rotational-vibrational transitions, we accurately measured the wavelengths. For any Qₙ, the frequency difference between Q₀ and Q₀,1, Q₁ and Q₁,1, Q₂ and Q₂,1 was measured to be 587.0 ± 0.3 cm⁻¹, where the uncertainty of ±0.3 cm⁻¹ is estimated from the variation of repeated measurements. This frequency difference corresponds to ΔJ = 2, for J = 1 to 3. The transition is known to be 587.0 cm⁻¹ (ref. 19). Thus these transitions correspond to pure rotational Raman shifting. They are referred to as SRRS transitions and labeled Qₙ±ₘₚ, where n refers to the vibrational order and ±ₘ refers to the rotational order.

Conversions versus pump energy for SRRS transitions are shown in figure 5. Conversions versus H₂ density are shown in figure 6. Above 6 amagats, only Qₙ transitions are observed, whereas the SRRS orders disappear. Note that >90 percent of the pump energy is converted into forward Stokes orders under some conditions.

4.2. CH₄

The vibrational Raman shift corresponding to the v₁ transition is 2916.5 cm⁻¹ (refs. 20 and 21). SRS in static CH₄ with high UV intensities generally results in multiphoton decomposition or breakdown (ref. 22). We minimized this problem by flowing the CH₄. Figure 7 shows CH₄ SRS in a closed FCRC, which produced a rapid temporal decay in conversion. The transmitted pump laser energy decreased 30 percent during the experiment. Results identical to figure 7 were obtained four successive times by refilling the FCRC. Results were independent of fan on or off. Removal and examination of the FCRC windows following all four runs indicated they were unaffected by the window-soot problem sometimes encountered (ref. 23). Results suggest a process creating photochemical decomposition whose products absorb ArF radiation. To overcome this problem, the laser entrance end of the FCRC was opened to the CH₄ cylinder while a mass flow controller was attached to the other end. The results obtained are shown in figure 7 with a flow of 6 SLM (standard liters per minute) of CH₄. All results in pure CH₄ and CH₄-He mixtures were acquired with this flowing Raman medium. After 2 to 3 days of work, a round black spot coincident with the pump beam appeared on the FCRC input window. The transmitted pump beam energy decreased by 30 percent. The window was readily cleaned with methanol.

Above 7 amagats, Q₁ and Q₂ conversions were the same for the 65- and 50-cm lenses; therefore, the 65-cm lens was used to acquire all further CH₄ data. Conversions versus CH₄ density are shown in figure 8 and are independent of fan on compared with fan off at ≤10 Hz. As with H₂, Q₂ exceeds Q₁ as density increases.

Helium addition (ref. 24) can vary the competition between backward SBS and SRS in a CH₄-He mixture and forward SRS. Results of backward and forward SRS versus He density are shown in figure 9. The backward SBS signal could not be distinguished from scattered pump laser in the case of CH₄. The maximum backward Q₁ signal, measured to be 1 μJ, was swamped by back-scattered pump. To remove the latter, backward Q₁ was observed by using a photomultiplier tube (PMT) after passage through a 1/4-m monochromator. Relative values obtained versus He
were normalized to the 1-μJ signal levels, then corrected for transmission losses to calculate conversions inside the FCRC. This method provides an order-of-magnitude estimate of backward $Q_1$. Forward $Q_1$ increases slightly at the expense of forward $Q_2$ and backward $Q_1$, as the He partial density is increased.

Conversion versus pump energy is shown in figure 10; $Q_2$ exceeds $Q_1$ at 50 mJ and both exceed $Q_0$. As with $H_2$, the FCRC has no measurable effect on conversion under these conditions.

4.3. $D_2$

The vibrational Raman shift of $D_2$ is 2987.3 cm$^{-1}$ (ref. 25). No experiments were performed to study conversion in $D_2$ versus focal length. Results similar to $H_2$ are expected. $Q$-branch conversion versus $D_2$ density is shown in figure 11. Above 4 amagats, Stokes lines greater than $Q_3$ decrease with increasing $D_2$ density indicating FWM effects. Like $H_2$, $Q_1 - Q_3$ conversion is independent of density above 5 amagats. Energy conversion $Q_1 - Q_4$ at 26-amagat $D_2$ versus pump energy is shown in figure 12; $Q_2$ exceeds $Q_1$ above 65 mJ, but unlike $H_2$ and CH$_4$, neither exceeds $Q_0$.

At pump energies above 50 mJ and densities from 1 to 10 amagats, rotational transitions surrounding the expected $Q_n$ vibrational transitions were observed in $D_2$ similar to those observed in $H_2$. The frequency spacing is measured as 408 ± 11 (±1σ) cm$^{-1}$. This spacing corresponds to transitions between $J = 2$ and 4. The rotational Raman shift corresponding to the $S(2)$ transition is 414.6 cm$^{-1}$ (ref. 25). By analogy with $H_2$, these transitions are the result of cascade pure SRRS. Conversion versus pump energy is shown in figure 13 for transitions near $Q_0$ and $Q_1$. Conversion versus $D_2$ density is shown in figure 14. Unlike $H_2$, $D_2$ transitions require higher pump energies and generate fewer SRRS transitions. Like $H_2$, they have similar conversions and occur over similar density ranges. At 1.2 amagats, only transitions surrounding $Q_0$ and $Q_1$ were observed, with 53 percent conversion into $Q_1$. Small density changes in the range of 1 to 5 amagats change significantly the conversions observed in various orders. As with $H_2$ and CH$_4$, the FCRC has no measurable effect on conversion.

As in $H_2$, results show an optimal density for conversion into a given transition. Also, as in $H_2$, low densities (1 to 3 amagats) optimize pure rotational transitions and $Q_1$. Moderate densities (3 to 5 amagats) optimize Stokes above $Q_3$. High densities (>5 amagats) optimize $Q_2$ and $Q_3$. Note that the intensity of our pump laser is large enough that we easily observe SRRS for $H_2$ and $D_2$ at 296 K.

4.4. $N_2$

The vibrational Raman shift is 2329.9 cm$^{-1}$ (ref. 26). The FCRC affects SRS significantly in $N_2$. Results showing conversion versus $N_2$ density with the FCRC fan on and off are presented in figure 15. The fan-off condition versus time caused the location of the laser beam on the FCRC output window to fluctuate from pulse to pulse. The beam also increases in size and overfills the FCRC output window because of thermal self-actions in the static gas as discussed in sections 4.8 and 5.1. In this case, accurate conversions could not be measured. The measured output energies with the fan off (and the cell clipping the beam) are shown for comparison. At 89 amagats, the fan-on condition improves measured output energy by a factor of approximately 300. At intensities less than 1 percent of $Q_1$, $Q_2$ was observed intermittently. Most $N_2$ data were acquired with a fan voltage of 24 V. Reducing the fan voltage from 24 V to 10 V could decrease conversion by 25 percent.

Conversions at 95-amagat $N_2$ were measured versus time for fan-on and fan-off conditions. For fan off, temporal decays similar to those of CH$_4$ were obtained. With the fan off at Time = 0, a 5-shot average at 10 Hz produced 8 percent conversion. After 30 s at 10 Hz with fan off, conversion decreased by a factor of 20. This decay was reproduced after running the fan for a minute or waiting 15 min with the fan off. Successive laser pulses build up the vibrational excitation but do not destroy the $N_2$ medium.

Previous work (ref. 27) used relatively long focal lengths for Raman shifting in $O_2$. For $N_2$, similar to $O_2$, we first determined that the 65-cm lens gave 25 percent more conversion than the 50-cm lens. All other $N_2$ results were acquired by using the 65-cm lens and the 1.5-m FCRC. Results in pure $N_2$ and two $N_2$-He mixtures are shown in figure 16. $N_2$ partial densities are listed on the abscissa. To ensure uniform composition, the FCRC circulated each mixture for at least 4 hr and the mixture stood overnight. Next, a
series of measurements were obtained by decreasing the total pressure.

Results of backward SRS and SBS versus added He density are shown in figure 17. In pure N₂, backward and forward SRS conversions are comparable. He addition reduces backward SRS and SBS while increasing forward SRS.

Energy conversion $Q_1$ was measured versus pump energy (18 to 100 mJ) for a 7:1 N₂-He mixture (Total density = 126 amagats). Unlike H₂, CH₄, and D₂, $Q_1$ conversion in N₂ remains approximately constant. Energy conversion $Q_1$ decreases by only 20 percent as the pump varies from 40 to 18 mJ. Above pump energies of 40 mJ, $Q_1$ conversion was constant.

4.5. SF₆ and CF₄

The prominent $v_1$ SF₆ vibrational Raman transition is shifted 774.5 cm⁻¹ (refs. 21 and 28). Conversions in SF₆ are low and difficult to obtain. Conversions of both $Q_1$ and $Q_2$ could not be maintained above 4 Hz. Most experiments were performed at 3 Hz. None were performed as a function of focal length. Conversion in pure SF₆ was optimal at pressures near the SF₆ condensation point (∼20 amagats in the nonshaded part of fig. 18) and when fan speeds were minimal. The fan motor was powered with 8 V. Helium addition produced results similar to CH₄ and N₂ as shown in the shaded part of figure 18. At the curve peak in the shaded part of figure 18, $Q_3$, $Q_4$, and $Q_{-1}$ were observed, all three with conversions of about 0.003 percent. No backward SRS or SBS experiments were performed.

Conversions in the optimal SF₆-He mixture in the shaded part of figure 18 were independent of pump energy from 15 to 60 mJ. With the FCRC fan off and using the optimum SF₆-He mixture, results similar to CH₄ and N₂ were observed. Conversions decreased by 1 order of magnitude after 10 min at 1 Hz in a closed cell with the fan off.

The vibrational Raman shift in CF₄ of the $v_1$ transition is 909.1 cm⁻¹ (refs. 21 and 29). Results are similar to SF₆. Conversions in pure CF₄ and CF₄-He mixtures are presented in table 1. Helium addition produced results similar to CH₄, N₂, and SF₆. Conversions $Q_3$, $Q_4$, and $Q_{-1}$ were generated as rings, which suggests they were due to FWM. Like SF₆, conversion was optimal when fan speeds were minimal (8 V) and could not be maintained above 4 Hz. No backward SRS, SBS, or further studies were performed due to the low conversions.

Low conversions in these two gases have several reasons. First, the Raman gain coefficients are small. Second, competition with backward SRS and SBS (ref. 30) reduces the forward Raman. Third, without a FCRC there are severe prior-pulse effects. Using a FCRC, along with He addition, reduces the magnitude of this problem. Fourth, SRS is observed only near the liquefaction point; this combined with high fan speeds created refractive index gradients, which were observed visually in the FCRC. Their intensity increased with fan speed, which precluded the use of relatively large fan speeds for these two gases. However, some finite fan speed is necessary to get any increase in the heat dissipation. These two ideas are consistent with our observation of optimal conversion at one particular relatively low fan speed.

4.6. Spatial Profiles of Raman Shifted Beams

Figure 19(a) shows 25 pulse averages for the first four Stokes orders after Raman shifting in pure CH₄. These images were captured on a video camera. Figure 19(b) shows four different single pulse images for the first Stokes, and figure 19(c) shows four single pulse profiles for the fourth Stokes. These pure CH₄ results are typical of most of our spatial profiles.

Raman shifting in H₂, D₂, and CH₄-He mixtures generally gave better spatial profiles than those of pure CH₄ in figure 19. Large gain coefficients (i.e., well above threshold) and efficient thermal conduction for these cases help to give better spatial profiles. By better, we mean more circularly symmetric than those of figures 19(b) and 19(c). Conversely, Raman shifting in CF₄, SF₆, and N₂ (all three mixed with He) consistently gave poorer spatial profiles (i.e., unsymmetric patterns with random hot spots) than those of figure 19. These poorer spatial profiles occur because of the relatively small gain coefficients (i.e., closer to threshold) and poorer thermal conductivity. Pure N₂ profiles were the worst, showing filamentary-type intensity distributions (ref. 31) that are characteristic of self-focusing or self-trapping.
4.7. Accessible Frequency Range

Table 2 lists the accessible frequency range for each Raman transition of each gas, along with the maximum conversion efficiencies obtained at the center of each range. Combined, they cover approximately 70 percent of the frequency range from 42,000 to 52,000 cm\(^{-1}\). The endpoints of the tuning ranges are defined to be where the locking efficiency of the laser drops to 30 percent. At both endpoints, the Stokes output pulse energies typically fall to about one half maximum because the laser energy and the locking efficiency both decrease as it is tuned off of the center frequency. We observe only minor variation in conversion efficiency above 40 mJ of input pump energy in most gases, and keeping the laser energy above 150 mJ is very time-consuming. Thus the results in table 2 were acquired over a wide range (40–170 mJ inside the cell) of pump energies. The Stokes energies corresponding to the conversion efficiencies in table 2 vary from 0.2 µJ to 58 mJ, depending on the input pump energy, gas, and Stokes transition. One result, using 110 mJ of pump energy and D\(_2\), is a conversion efficiency of 53 percent and a tunable Stokes energy that varies over the range of 29 to 58 mJ as the first Stokes wavelength is tuned over the range of 204 to 206 nm. We anticipate increasing these Stokes energies by about a factor of 2 by using the full pump energy and MgF optics in the cell.

4.8. Laser Pulse Repetition Rate

Stokes conversion, input pump energy, and residual pump energy versus laser repetition rate are shown for H\(_2\), CH\(_4\), D\(_2\), N\(_2\), and SF\(_6\) in figures 20 through 25. Figure 21 differs from figure 20 by the pump energy that is used. For the first Stokes of H\(_2\) and D\(_2\), forced convection does not make any difference, even at a pulse rate of 100 Hz. Higher Stokes orders are helped by the circulation of the gas. In CH\(_4\), in spite of the pressure-induced flow from the CH\(_4\) bottle, the fan-induced motion helps Raman conversion at >10 Hz. For N\(_2\), SF\(_6\), and CF\(_4\), using forced convection (i.e., fan on) improves the conversion at higher repetition rates compared with fan off. At rates as low as 1 Hz, for these 3 gases, circulation of the gas is necessary to get any consistent output.

In many gases, we observe a 10- to 20-percent increase in \(Q_1\) and \(Q_2\) conversion as the repetition rate varies from 10 to 100 Hz. This increase occurs because the ArF laser cannot maintain pulse energies above 50 Hz. The input pump energies are plotted with the right-hand axes. The decreasing pump energy eventually drops below threshold for the higher orders; this makes more pump energy available for conversion into the lower orders, which are still above threshold. This results in the increase, versus repetition rate, of the low orders in figures 20 to 25.

Without exchanging the gas between laser shots, N\(_2\) SRS is limited to very low (<1 Hz) repetition rates, where mass and thermal diffusion can maintain equilibrium. Pure N\(_2\) has a relatively long vibrational lifetime for \(v = 1\). At 298 K, reference 32 quotes a lower limit of >1 atm-s. We estimate \(3 \times 10^3\) atm-s, which is based on high temperature data (ref. 33). The N\(_2\) lifetime at 157 amagats (170 atm) is estimated to be 18 s. Successive pulses at 10 Hz would create significant population in \(v = 1\) and reduce conversion.

Helium (ref. 32) was added to N\(_2\) to increase the vibrational relaxation rate. However, in static conditions (i.e., no fan), good conversion was still limited to low repetition rates. High conversion for the first 5 to 10 pulses suggests that gas breakdown is not the problem. But observations of fluctuations in beam size and location on the output FCRC window indicate that thermal “lensing” is a problem. Using the fan did eliminate these effects and improve the output beam profile and reduce the motion. Figure 24 shows that the fan allows reasonable conversion at repetition rates up to 10 Hz.

4.9. Raman Threshold Power Measurements

Our best threshold measurement is in H\(_2\) at 4 amagats. Using 1.5 mJ of pump, 18 percent conversion into \(Q_1\) is observed without any higher Stokes orders. Decreasing the pump energy to 0.75 mJ produced no \(Q_1\). Thus we estimate an experimental threshold power of 1 mJ/15 ns = 70 ± 30 kW for our apparatus. This estimate is about 8 times larger than the calculated value of 8 kW from equation (5).

We also measured threshold for N\(_2\), although less reliably than for H\(_2\). For 146 amagats of pure N\(_2\), we see minimal Stokes production with 12 mJ of pump. Visual inspection of the beam suggested that we were close to threshold; however, we did not decrease the
power to watch the Raman conversion disappear because we were not interested in threshold powers at the time. Thus \( P_{th} \leq 12 \text{ mJ/15 ns} = 0.8 \text{ MW} \). The calculation with equation (5) is 95 kW. As in the measurement of \( \text{H}_2 \), the \( \text{N}_2 \) upper limit is \( \approx 8 \) to 9 times larger than expected for an ideal focused Gaussian beam.

This disagreement is reasonable, since our pump beam is \( \approx 9 \) times worse than the diffraction limit (section 3.1). Hence we expect that the pump beam will not focus as tight as possible and that the threshold will be raised above the expected value of \( \varepsilon^{25} = 25 \text{ Np} \) for near diffraction limited beams. Since we observe threshold at \( 9 = \varepsilon^2 \) times the value predicted by equation (5), we are observing threshold at about \( \varepsilon^2 \varepsilon^{25} = 27 \text{ Np} \) for both \( \text{H}_2 \) and \( \text{N}_2 \).

Similar to \( \text{N}_2 \), we also obtained an upper limit of threshold for \( \text{SF}_6 \). For a 1:1 mixture of \( \text{SF}_6-\text{He} \) at a total density of 29 amagats, \( P_{th} \) is less than or equal to 0.47 MW for \( \text{SF}_6 \).

5. Discussion

5.1. Limitations for Raman Conversion

The SRS process perturbs a gas medium in a variety of ways (refs. 34 and 35) that can reduce conversion efficiency. A few of these processes are discussed. First, for each Stokes photon produced, a molecule is excited to \( \nu = 1 \). But conversion efficiency depends on the population difference. If these excited molecules do not return to \( \nu = 0 \) between successive pulses, conversion is reduced. Vibrational relaxation lifetimes are listed in table 3. (Data in table 3 are from refs. 36 to 42, 33, and 32.) Even at 100 Hz, relaxation between laser pulses for all gases is complete except in \( \text{N}_2 \). Second, regardless of the mechanism or rate by which the perturbed medium equilibrates, the final result is energy transfer into translational motion. Hence, significant heat can be deposited in the interaction volume and perturb the propagation of subsequent pulses through the medium. Third, optically induced ionization can destroy the Raman medium. Finally, large laser intensities can nonlinearly perturb the index of refraction to change the beam propagation on the nanosecond time scale of a single pulse. These previous processes are known as saturation, thermal lensing, optical breakdown, and self-focusing, respectively.

These processes can degrade the spatial beam quality, create large fluctuations in the spatial location of the output Raman beam, and prevent the beam from focusing properly. In general, they also reduce conversion in the SRS process. They can occur during a single pulse (transient self-actions) or be the result of a prior pulse or series of pulses (thermal self-actions). The influence of thermally related perturbations is governed by the removal of heat through natural convection and diffusion. Previous researchers (e.g., ref. 24) added He to the gas medium to increase the thermal conductivity.

For \( \text{N}_2 \), Stokes output beam spatial profiles that are filamentary in nature are observed. This observation suggests that self-focusing is occurring and that the input beam is not uniform. Addition of He to \( \text{N}_2 \) reduces, but does not eliminate, this beam degradation.

To check for laser-induced breakdown of the gas medium, a second cell (one with a side window but unable to take high pressures) was filled with 1 atm of \( \text{N}_2 \). With 200 mJ inside the cell, we observed breakdown with a 50-cm focal length lens and no breakdown with 57- and 65-cm lenses. Filling the cell with 1 atm of \( \text{H}_2 \) again showed no sign of breakdown with the 65-cm lens. Almost all the data presented here were obtained with the 65-cm lens. These tests do not prove that we are avoiding breakdown because the breakdown threshold will be lowered (relative to 1 atm) at the high pressures used for Raman shifting. Unfortunately, there are no side windows on the FCRC to use for visual confirmation of breakdown.

5.2. Increasing the Repetition Rate With a FCRC

In this section, an empirical method is presented for determining repetition rates for which our FCRC will be effective in improving Raman beam output energies or spatial profiles. One method for eliminating prior-pulse effects is to exchange the medium after each laser pulse (not only at the focal volume, but along the entire length of the laser beam). This was demonstrated (ref. 43) with a cw (continuous-wave) laser beam inside a wind tunnel. That work showed
that prior-pulse thermal effects were minimized at velocities near 250 cm/s, and these velocities had negligible effects on beam profile.

The FCRC achieves similar improvements except in a closed system. The nonequilibrium medium is removed or diluted in the surrounding medium, and this provided time to equilibrate. The FCRC will be effective for nonequilibrium whenever the perturbed medium cannot equilibrate between pulses. If vibrational relaxation is assumed complete and no optical breakdown, the problem reduces to heat dissipation. One method for estimating when the FCRC will be effective for heat dissipation is to compare the thermal diffusion length \( L \) (ref. 15) with the focused beam diameter \( D \). The diffusion length depends on the pulse repetition rate \( f \) as

\[
L(f) = \left( \frac{8k}{fpC_p} \right)^{1/2}
\]

(6)

The density is \( \rho \), the specific heat capacity is \( C_p \), from reference 21, and the thermal conductivity is \( k \), from reference 44 for all gases except SF\(_6\) and CF\(_4\) and from reference 45 for SF\(_6\) and CF\(_4\). For comparison of gases, table 4 shows values of diffusion length \( L(10) \) calculated for our Raman cell conditions and a pulse rate of 10 Hz. Table 4 suggests that conversion can be maintained at higher repetition rates for the sequence SF\(_6\) < N\(_2\) < CH\(_4\) < D\(_2\) < H\(_2\). Results in figures 20 to 25 are consistent with this suggestion.

Prior-pulse effects should be small when \( L \) becomes several times \( D \). To test this idea, calculate \( L(f_{1/2}) \) at the repetition rate \( f_{1/2} \), which is the frequency where the Stokes output has decreased to 50 percent of the low repetition rate value. Table 5 presents the ratio \( L(f_{1/2})/D \) for five cases. In each case, we use the lowest Stokes transition that is observed to weaken with increasing frequency. For all transitions listed in table 5, the conversion efficiency is \( \geq 10 \) percent (except the backward O\(_2\)-He mixture case which is 7 percent). This criterion implies that the gain in these various transitions is at similar relatively large levels above threshold. Thus significant heating is taking place in each case. The data for H\(_2\), CH\(_4\), and D\(_2\) come from figures 21 to 23. The O\(_2\)-He mixture data for forward and backward scattering are estimated from reference 46. The thermal conductivity for the O\(_2\)-He mixture is estimated as \( \approx 0.06 \) J/m-s-K from reference 47. We believe that the two data points for the two O\(_2\)-He mixture cases are less reliable than the pure gas results, because of our crude calculation of the thermal conductivity and the limited data points for conversion versus frequency in reference 47.

The ratio \( L(f_{1/2})/D \) is about constant at \( 4 \pm 1 \) for all entries in table 5. The uncertainty is an estimate simply based on the spread of values in table 5. This implies that \( L(f_{1/2})/D = 4 \) can be used as a rule of thumb to estimate repetition rates where this FCRC will become effective in improving conversion and beam quality. If \( L/D < 4 \), then the internal fan will help. If \( L/D > 4 \), natural diffusion and convection will suffice. Our observed value of \( 4 \pm 1 \) for \( L/D \) in gases is similar to the value of 2.7 observed (ref. 15) in liquid nitrogen.

6. Concluding Remarks

We have studied the production of high-pulse-energy tunable laser radiation at far ultraviolet frequencies. This is achieved by combining a high-spectral-brightness tunable 285-mJ ArF excimer laser, stimulated Raman scattering, and a forced convection Raman cell. Multiple stimulated Raman transitions in H\(_2\), CH\(_4\), D\(_2\), N\(_2\), SF\(_6\), and CF\(_4\) are observed. We can access 70 percent of the frequency range from 42000 to 52000 cm\(^{-1}\) with a Raman beam energy of \( \approx 60 \) mJ in the best case of D\(_2\) (first Stokes energy conversion efficiency of 53 percent). Addition of He to the Raman gas greatly improves the conversion in N\(_2\), SF\(_6\), and CF\(_4\). Threshold intensities for vibrational Raman conversion are found to be \( \approx 8 \) times larger than calculations for ideal Gaussian beams. Pure rotational Raman conversion in H\(_2\) and D\(_2\) is obtained at room temperature.

The vibrational Raman conversion efficiency versus laser repetition rate was measured. Energy conversions of \( Q_1 \) and \( Q_2 \) in H\(_2\) and \( Q_1 \) in D\(_2\) do not decrease from 1 to 100 Hz in a static cell. But, conversions in higher orders of H\(_2\) and D\(_2\) and \( Q_1 \) of lower-gain gases (N\(_2\), CH\(_4\), and SF\(_6\)) decrease as repetition rate increases due to prior-pulse effects (e.g., heating) on the Raman medium. We show the effectiveness of forced convection to improve Raman conversion at higher repetition rates by circulating the gas inside the Raman cell to decrease these effects. At room temperature, if the thermal diffusion length is less than
=4 times the focused beam diameter, the FCRC will be useful in increasing conversion efficiencies at higher pulse rates. Using this FCRC, maximum conversion efficiency can be maintained up to 100 Hz in Q1 of CH4, 50 Hz in Q2 of D2, 20 Hz in Q2 of CH4, 10 Hz in Q1 of N2-He, and 5 Hz in Q1 and Q2 of SF6-He.

7. References


Table 1. Vibrational Branch Stokes Energy Conversion in CF$_4$
[Data acquired using 40 mJ of pump laser energy and a 5-Hz repetition rate]

<table>
<thead>
<tr>
<th>Density, amagats</th>
<th>Energy generated, $\mu$J</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$Q_1$</td>
</tr>
<tr>
<td>CF$_4$ 4.1</td>
<td>0</td>
</tr>
<tr>
<td>16.6</td>
<td>0</td>
</tr>
<tr>
<td>18.5</td>
<td>0</td>
</tr>
<tr>
<td>18.5 6.3</td>
<td>142</td>
</tr>
<tr>
<td>18.5 12.6</td>
<td>140</td>
</tr>
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</table>
Table 2. Accessible Frequency Ranges and Maximum Energy Conversions Obtainable by SRS Using a FCRC and Tunable ArF Excimer

Conversions are obtained at laser frequency of 51714 cm\(^{-1}\) and conditions that optimize conversion for each particular transition.

<table>
<thead>
<tr>
<th>Gas</th>
<th>Transition</th>
<th>Frequency range, cm(^{-1})</th>
<th>Energy conversion, percent</th>
</tr>
</thead>
<tbody>
<tr>
<td>H(_2)</td>
<td>Q(_{0,1})</td>
<td>52 460–52 160</td>
<td>0.35</td>
</tr>
<tr>
<td>D(_2)</td>
<td>Q(_{0,1})</td>
<td>52 285–51 985</td>
<td>0.75</td>
</tr>
<tr>
<td>D(_2)</td>
<td>Q(_{0,11})</td>
<td>51 455–51 155</td>
<td>2.5</td>
</tr>
<tr>
<td>H(_2)</td>
<td>Q(_{0,11})</td>
<td>51 280–50 980</td>
<td>1.3</td>
</tr>
<tr>
<td>SF(_6)</td>
<td>Q(_1)</td>
<td>51 095–50 795</td>
<td>2.1</td>
</tr>
<tr>
<td>D(_2)</td>
<td>Q(_{0,12})</td>
<td>51 040–50 740</td>
<td>0.002</td>
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<td>CF(_4)</td>
<td>Q(_1)</td>
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</tr>
<tr>
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<td>Q(_{0,12})</td>
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<td>0.053</td>
</tr>
<tr>
<td>SF(_6)</td>
<td>Q(_2)</td>
<td>50 320–50 020</td>
<td>0.26</td>
</tr>
<tr>
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<td>0.0033</td>
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<td>21</td>
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<td>22</td>
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<tr>
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<td>Q(_1)</td>
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<td>Q(_{2,2})</td>
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<tr>
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<td>Q(_2)</td>
<td>46 040–45 740</td>
<td>28</td>
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<td>Q(_{1,13})</td>
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<tr>
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<td>45 480–45 180</td>
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<td>Q(_2)</td>
<td>43 560–43 260</td>
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<td>Q(_{3,1})</td>
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<td>43 120–42 820</td>
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<td>D(_2)</td>
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<td>13</td>
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<td>Q(_{2,12})</td>
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<td>Q(_{3,1})</td>
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<td>Q(_4)</td>
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Table 3. Vibrational Relaxation Data for $v = 1$

<table>
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<th>Molecule pair</th>
<th>$p \tau$, atm-s</th>
<th>Reference</th>
<th>Temperature, K</th>
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<tr>
<td>$H_2$ $H_2$</td>
<td>$1060 \pm 100 \times 10^{-6}$</td>
<td>36</td>
<td>300</td>
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<td>$H_2$ $He$</td>
<td>$385 \times 10^{-6}$</td>
<td>37</td>
<td>295</td>
</tr>
<tr>
<td>$H_2$ $He$</td>
<td>$2280 \times 10^{-6}$</td>
<td>38</td>
<td>298</td>
</tr>
<tr>
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<td>295</td>
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<tr>
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<td>39</td>
<td>295</td>
</tr>
<tr>
<td>$D_2$ $D_2$</td>
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<td>40</td>
<td>296</td>
</tr>
<tr>
<td>$D_2$ $He$</td>
<td>$6 \times 10^{-3}$</td>
<td>41</td>
<td>296</td>
</tr>
<tr>
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<td>$^{a}33$</td>
<td>300</td>
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<td>$6.9 \pm 0.3 \times 10^{-3}$</td>
<td>32</td>
<td>298</td>
</tr>
<tr>
<td>$N_2$ $He$</td>
<td>$1.5 \times 10^{-2}$</td>
<td>$^{a}33$</td>
<td>298</td>
</tr>
<tr>
<td>$SF_6$ $SF_6$</td>
<td>$&gt;2 \times 10^{-8}$</td>
<td>42</td>
<td>300</td>
</tr>
<tr>
<td>$CF_4$ $CF_4$</td>
<td>No data available</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^{a}$Extrapolated to $\approx 300$ K from high temperature measurements.

Table 4. 10-Hz Thermal Diffusion Lengths $L(10)$

<table>
<thead>
<tr>
<th>Gas</th>
<th>$C_p$, J/mol-K</th>
<th>$k$, J/m-s-K</th>
<th>$\rho$, amagats</th>
<th>$L(10)$, $\mu$m</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>20.8</td>
<td>0.15</td>
<td>20</td>
<td>2500</td>
</tr>
<tr>
<td>$H_2$</td>
<td>28.8</td>
<td>0.182</td>
<td>20</td>
<td>2400</td>
</tr>
<tr>
<td>$D_2$</td>
<td>29.2</td>
<td>0.14</td>
<td>26</td>
<td>1800</td>
</tr>
<tr>
<td>$CH_4$</td>
<td>35.6</td>
<td>0.034</td>
<td>7</td>
<td>1600</td>
</tr>
<tr>
<td>$N_2$</td>
<td>29.1</td>
<td>0.026</td>
<td>40</td>
<td>600</td>
</tr>
<tr>
<td>$CF_4$</td>
<td>61</td>
<td>0.016</td>
<td>18.5</td>
<td>500</td>
</tr>
<tr>
<td>$SF_6$</td>
<td>97</td>
<td>0.015</td>
<td>18</td>
<td>400</td>
</tr>
</tbody>
</table>

Table 5. Half-Stokes-Power Thermal Diffusion Lengths $L(f_{1/2})$

[For $O_2$-$He$, $D = 300 \mu$m; for all others, $D = 167 \mu$m]

<table>
<thead>
<tr>
<th>Gas</th>
<th>Stokes order</th>
<th>$f_{1/2}$, Hz</th>
<th>$\rho$, amagats</th>
<th>$L(f_{1/2})$, $\mu$m</th>
<th>$L(f_{1/2})/D$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$H_2$</td>
<td>3</td>
<td>79</td>
<td>20</td>
<td>2400</td>
<td>5.1</td>
</tr>
<tr>
<td>$D_2$</td>
<td>2</td>
<td>64</td>
<td>26</td>
<td>1800</td>
<td>4.3</td>
</tr>
<tr>
<td>$CH_4$</td>
<td>1</td>
<td>42</td>
<td>7</td>
<td>1600</td>
<td>4.6</td>
</tr>
<tr>
<td>$O_2$-$He$ (8:9)</td>
<td>1 (back)</td>
<td>$=3$</td>
<td>65</td>
<td>1484</td>
<td>4.9</td>
</tr>
<tr>
<td>$O_2$-$He$ (2:3)</td>
<td>1 (forward)</td>
<td>$=8$</td>
<td>63</td>
<td>923</td>
<td>3.1</td>
</tr>
</tbody>
</table>
Figure 1. Schematic diagram of apparatus for Raman shifting using tunable ArF laser and forced convection Raman cell.
Figure 2. Conversion in H$_2$ at pump energy ($E_{\text{pump}}$) of 83 mJ versus density.
Figure 3. Conversion using 20-amagat H$_2$ versus input pump energy.
Figure 4. Summary of SRS transitions and conversions observed in 1.55-amagat H$_2$ with pump energy of 101 mJ.
Figure 5. SRS pure rotational transitions and conversions observed in 1.55-amagat H$_2$ versus pump energy.
Figure 5. Concluded.
Figure 6. SRS pure rotational transitions and conversions observed versus H$_2$ density using pump laser energy of 107 mJ.
Figure 6. Continued
Figure 6. Continued.
Figure 6. Concluded.
Figure 7. Conversion in 2.5-amagat CH$_4$ versus time at pump of energy 45 mJ and repetition rate of 5 Hz. The FCRC fan is on for both data sets.
Figure 8. Conversion versus CH$_4$ density using pump energy of 45 mJ.
Figure 9. Forward and backward Stokes conversion in 19.7-amagat CH$_4$ versus helium partial density using pump energy of 42 mJ.
Figure 10. Conversion in 19.7-amagat pure CH\textsubscript{4} versus pump energy.
Figure 11. Conversion in D\textsubscript{2} versus density for pump energy of 53 mJ.
Figure 12. Conversion in 26-amagat D$_2$ versus pump energy.
Figure 13. Conversions of transitions observed in 1.55-amagat D$_2$ versus pump energy.
Figure 14. Conversions of transitions observed in D$_2$ versus density using pump energy of 110 mJ.
Figure 14. Continued.

(b) $Q_1$. 

Energy conversion efficiency, percent

$D_2$ density, amagats
Figure 14. Continued.

(c) $Q_2$. 

Figure 14. Continued.
Figure 14. Concluded.

(d) $Q_3$, $Q_4$, and $Q_5$. 

(d) $Q_3$, $Q_4$, and $Q_5$. 

Figure 14. Concluded.
Figure 15. Conversion in pure N$_2$ versus density using pump energy of 45 mJ at 5 Hz.
Figure 16. Forward conversion $Q_1$ in pure $N_2$ and $N_2$-He mixtures versus $N_2$ and using pump energy of 53 mJ at 5 Hz.
Figure 17. Backward SRS Stokes $Q_1$ and backward SBS conversion efficiencies in pure N$_2$ and a 3:1 N$_2$-He mixture for pump laser energy of 82 mJ at 5 Hz.
Figure 18. Conversion in SF$_6$ versus SF$_6$ density and He density using input pump energy of 59 mJ at 3 Hz with fan on. Unshaded part of figure shows conversion in pure SF$_6$ versus density and shaded part shows conversion versus partial He density at fixed SF$_6$ density of 20 amagats.
(a) 25-pulse averages of $Q_1$ through $Q_4$.

Figure 19. Typical CH$_4$ pulse intensity profiles.
(b) Four single pulse examples of $Q_1$.

Figure 19. Continued.
(c) Four single pulse examples of $Q_4$ doughnut profile.

Figure 19. Concluded.
Figure 20. Conversion in 20-amagat H₂ versus repetition rate. Input pump energy (E_{pump}) is read from right-hand scale.
Figure 21. Conversion in 20-amagat H$_2$. 

Energy conversion efficiency, percent

Laser repetition rate, Hz

Q$_1$  Q$_0$  Q$_3$  Q$_2$  E$_{\text{pump}}$

Fan off  Fan on
Figure 22. Conversion in 7.2-amagat CH\textsubscript{4} versus repetition rate. To minimize photochemical decomposition, the CH\textsubscript{4} flow rate is 6 SLM.
Figure 23. Conversion in 26-amagat D₂ versus repetition rate.
Figure 24. Conversion in 3:1 N₂-He mixture (total density is 126 amagats) versus repetition rate. Input pump energy, 71 mJ; $Q_0$ constant over range of 1 to 30 Hz.
Figure 25. Conversion in mixture of 18-amagat SF₆ and 12-amagat He versus repetition rate. Fan on; input pump energy, 59 mJ; \( Q_0 \) constant over range of 1 to 20 Hz.
**Title:** Raman Shifting a Tunable ArF Excimer Laser to Wavelengths of 190 to 240 nm With a Forced Convection Raman Cell

**Authors:** R. Jeffrey Balla and G. C. Herring

**Performing Organization:** NASA Langley Research Center

**Funding Numbers:** WU 522-31-61-01

**Performing Organization Report Number:** L-17909

**Sponsoring/Monitoring Agency:** National Aeronautics and Space Administration

**Sponsoring/Monitoring Agency Report Number:** NASA/TM-2000-209710

**Abstract:**

Tunable radiation, at ultraviolet wavelengths, is produced by Raman shifting a modified 285-mJ ArF excimer laser. Multiple Stokes outputs are observed in H₂, CH₄, D₂, N₂, SF₆, and CF₄ (20, 22, 53, 21, 2.1, and 0.35 percent, respectively). Numbers in parentheses are the first Stokes energy conversion efficiencies. We can access 70 percent of the frequency range 42,000–52,000 cm⁻¹ (190–240 nm) with Stokes energies that vary from 0.2 μJ to 58 mJ inside the Raman cell. By using 110 mJ of pump energy and D₂, the tunable first Stokes energy varies over the 29–58 mJ range as the wavelength is tuned over the 204–206 nm range. Dependence on input energy, gas pressure, He mixture fraction, and circulation of the gas in the forced convection Raman cell is discussed; Stokes conversion is also discussed for laser repetition rates from 1 to 100 Hz. An empirical equation is given to determine whether forced convection can improve outputs for a given repetition rate.