# Noise characteristics of single shot broadband Raman-resonant CARS with single- and multimode lasers

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*Published in:*
Applied Optics

*DOI:*
10.1364/AO.26.001068

1987

# Noise characteristics of single shot broadband Raman-resonant CARS with single- and multimode lasers

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A simple model is presented as an aid in understanding, first, the relative noise performance and, second, the noise reduction achievable by referencing, in different experimental approaches to single shot broadband coherent anti-Stokes Raman scattering (CARS). Qualitative agreement is obtained with previous experimental investigations of CARS noise. The broadband dye laser radiation is described as the sum of independent modes with random phases. The dye laser contribution to the CARS noise is then approximately inversely proportional to the square root of the number of dye laser modes generating the detected signal. A fundamental idea is that in Raman resonant spectra only the number of Stokes modes actually participating in driving the Raman resonance should be counted. This means, e.g., that for narrow Raman resonances, as in an atmospheric flame, the noise generated by the dye laser will be higher for a single-mode pump laser than for a multimode pump laser with the experimental CARS configuration normally employed. The implications of the model for the dual broadband type CARS techniques are also discussed.

## I. Introduction

The possibility of using coherent anti-Stokes Raman scattering (CARS) for nonintrusive temperature and/or species concentration determination[1,2] has generated increasing interest from groups in the combustion field to develop the technique into a reliable instrument for accurate measurements in combustion processes.[3–8] The most common CARS setup utilizes a Nd:YAG system where the CARS beam at the anti-Stokes frequency $\omega_{as}$ is generated at $\omega_{as} = 2\omega_p - \omega_s$. Here $\omega_p$ is the frequency-doubled output from the YAG laser and $\omega_s$ is the output from a broadband dye laser (typical bandwidth about one hundred wavenumbers). The generated anti-Stokes beam is dispersed by a spectrograph and detected with an optical multichannel analyzer (OMA). A complete anti-Stokes spectrum of the investigated molecule is thus obtained in a single laser shot.[9] Within the CARS community there has been a controversy whether a single-mode or a multimode YAG laser is preferred for minimizing the noise in CARS spectra. Complications arising when using a multimode YAG laser have been pointed out.[10–12,5] These have predominantly been related to rapid fluctuations in the pump laser intensity arising from mode beating. Snelling *et al.*[13] have also presented non-Raman-resonant CARS spectra where a single-mode pump laser gave a better signal-to-noise ratio (S/N) than a multimode pump laser. On the other hand Greenhalgh and Whittley[14] have reported non-Raman-resonant noise figures for a multimode pump laser which are comparable with the single-mode figures of Snelling and co-workers. Hall and Greenhalgh[15] claimed that the single- and multimode pump noise figures should be about the same for the nonresonant case if all intermode beating terms are averaged out by long pulse duration.

Snelling *et al.* have recently extended their investigations to include resonant flame spectra.[16] (The term resonant here and throughout the rest of the paper stands for Raman resonant, not electronically resonant, spectra. Similarly the term nonresonant refers to spectra without either Raman resonance or electronic resonance.) While the nonresonant spectra, in agreement with their previous investigation, showed a better S/N with the single-mode laser, the multimode laser gave the lowest noise in the resonant spectra. In light of previous publications on the issue of single- vs multimode pump lasers this result was somewhat surprising. The results of Snelling *et al.*[16] are reprinted in Table I to facilitate the discussion. Shot noise and dark current noise have been subtracted as per Ref. 16.

In this paper we first show that the strongly increased noise figures of Snelling *et al.*, when going from the nonresonant to the resonant case using a single-mode laser, can be approximately reconstructed by considering only one single source of noise. Namely, that arising from the finite number of independent dye-laser modes actually contributing to the CARS signal. (A more elaborate theoretical model would, e.g., have to consider the role of pulse-to-pulse fluctuations in pump mode intensities and/or possible nonaveraged intermode beat terms.[15,17] Such a treatment would need to be more mathematical. Preliminary considerations indicate that the higher nonresonant

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Received 4 October 1986.
0003-6935/87/061068-06$02.00/0.

Table I. CARS Noise Obtained for Different Experimental Situations by Snelling

| | Pump laser (%) Single-mode | Multimode |
|---|---|---|
| Nonresonant spectra | 6.6 | 8.4 |
| Resonant spectra $N_2$ at $T$ = 1580 K | 22.0 | 15.6 |

figures for the multimode laser could be explained with such an approach.[17] In the next section it is argued that an upper limit on the noise reduction obtainable by referencing can be determined by considering the dye laser modes contributing to the signal and reference spectra, respectively. Finally, dual broadband vibrational[18,19] and rotational[20,21] CARS are discussed, since our model implies that these techniques would be particularly insensitive to dye-laser mode noise. Recent measurements support this conclusion.[21]

Throughout the paper we assume that both the phases and the amplitudes of the various dye laser modes can be regarded as independent of each other. This assumption is expected to be valid for a pulsed multimode dye laser operating high above threshold.[22] For such a laser spatial hole burning and mode competition are negligible. Greenhalgh and Whittley have analyzed in detail the mode noise in a broadband dye laser[14] and their results were also consistent with the assumption of independent modes. The probability distribution for the mode intensities is assumed to be exponential.[14] We define intensities and standard deviations as those registered by a single pixel in the OMA diode array. If these signals are normalized by dividing the signal recorded in each pixel with the integrated CARS intensity from all pixels, as was done in Refs. 13 and 16, this normalized pixel intensity will not fluctuate due to variations in the total intensity of the pump or Stokes beams. However, there will be shot-to-shot fluctuations due to, e.g., variations in the intensity of the particular Stokes modes contributing to the signal in a given pixel. Following Ref. 14, noise is defined as the standard deviation of the pixel intensity, normalized as above, divided by the average normalized intensity recorded by that pixel. The number of Stokes modes contributing to the signal in a specific pixel is determined by the convolution of the pump laser bandwidth and the detection system slit function. For simplicity we assume that the width of the slit function $W$ is much larger than the pump laser bandwidth $\Gamma_p$. For the values in Ref. 16, $W \approx 2\ \text{cm}^{-1}$ and $\Gamma_p \approx 0.1\ \text{cm}^{-1}$, this is a good approximation. Line shapes and slit widths will be assumed to be rectangular. It is, at the expense of a need for more elaborate mathematics, straightforward to include the correct profiles or different relationships between the various widths in the treatment below. The conclusions drawn from the treatment outlined in this paper do not depend on the exact shapes of laser or line profiles. The numerical calculations presented here are not to be regarded as exact, however; instead their aim is to show trends, demonstrate orders of magnitude, and clarify the discussion.

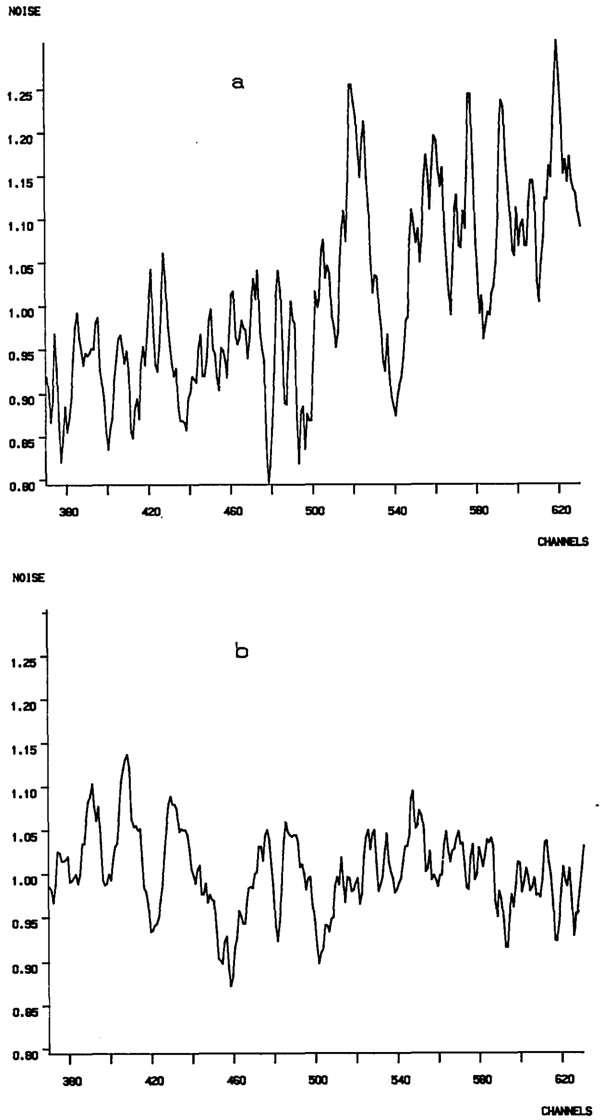


Fig. 1. Schematic view of the CARS process employing (a) a single-mode pump laser and (b) a multimode pump laser. The figure is a visualization of how the multimode pump laser enables a larger number of Stokes laser modes to participate in the excitation of the Raman resonance. Vertical scale is energy, vertical arrows and piles represent energies of the photons used in each step in the CARS process. One Raman resonance is indicated. Horizontal lines represent laser modes. These lines have different lengths symbolizing the randomness in intensity of the individual modes.

## II. Influence of Stokes Laser Noise on Resonant CARS

In Fig. 1 the CARS process is schematically pictured for (a) a single-mode pump laser and (b) a multimode pump laser. The vertical axis corresponds to energy and the vertical arrows represent the energy of the photons contributing to the resonant CARS signal. The horizontal lines represent laser modes. One Raman resonance has been indicated. The figure visualizes how the multimode laser enables a larger number of Stokes modes to participate in the excitation of the Raman resonance. This is the case since Stokes modes that would fall just outside the Raman resonance if combined by the single mode from a single-mode pump laser are shifted in under the Raman line by combining with modes in the wings of the multimode pump profile. Let $\Gamma_r$, $\Gamma_{(r+p)}$, $\Omega_p$ and $\Omega_s$ be the FWHM of the Raman line, the FWHM of the profile corresponding to the convolution of the Raman line and the pump laser profile, the mode separation in the pump laser, and the mode separation in the Stokes laser, respectively. Only the lowest-order transverse modes ($TEM_{00}$) are considered here. The higher-order modes do not focus as tightly as the lower-order modes and are less effective in generating the CARS signals.[14,16,23] There will be $\Gamma_r/\Omega_s$ and $\Gamma_{(r+p)}/\Omega_s$ Stokes modes contributing to the Raman resonant signal with a single-mode pump laser and a multimode pump laser, respectively. Provided that the Raman linewidth is larger than the mode separation in the dye laser, it is also clear that all pump modes contribute to the Raman resonant signal for the multimode case. If the pump mode separation is larger than the Raman linewidth all the pump modes will still participate in the excitation. In this case the number of independent Stokes modes contributing would be $N_p(\Gamma_r/\Omega_s)$, where $N_p$ is the number of pump laser modes.

Greenhalgh and Whittley[14] demonstrated that the noise in the spectral profile of a multimode laser and in a nonresonant CARS spectra generated with such a laser, depended on the number of dye laser modes detected by each pixel. For independent modes we have[14]

$$\langle I_T\rangle = \sum_{k=1}^{l} \langle I_k\rangle,$$

$$\sigma_k \equiv (\langle I_k^2\rangle - \langle I_k\rangle^2)^{1/2} = \langle I_k\rangle,$$

$$\sigma_T = \left(\sum_{k=1}^{l} \sigma_k^2\right)^{1/2};$$

$\langle\ \rangle$ indicates an ensemble average, $I_k$ denotes the intensity of mode $k$, $I_T$ is the registered intensity when $l$ modes are simultaneously observed, $\sigma_k$ is the standard deviation in the intensity of mode $k$, and $\sigma_T$ is the standard deviation in the intensity $I_T$. If a nonexponential probability distribution is assumed for the mode intensities instead the expectation values in the second equation above need to be changed. The subsequent treatment is then still valid. The noise when $l$ modes are observed simultaneously is given by Eq. (1):

$$\frac{\sigma_T}{\langle I_T\rangle} = \frac{\sqrt{\sum_{k=1}^{l} \langle I_k\rangle^2}}{\sum_{k=1}^{l} \langle I_k\rangle} = \frac{1}{\sqrt{l}}. \tag{1}$$

Consequently, the observed noise is inversely proportional to the square root of the number of modes contributing to the signal. (A more general form for the right-hand side would be $\gamma/\sqrt{l}$, where $\gamma$ is a line shape dependent numerical factor. Here, for rectangular profiles, $\gamma$ equals one.) To calculate the noise in a given pixel for a resonant or nonresonant CARS spectrum the number of independent Stokes modes contributing to the signal must therefore be estimated. In the nonresonant case $W/\Omega_s$ modes are seen. As the nonresonant part of the third-order susceptibility essentially is constant all the modes will have equal weight. In a resonant spectrum the recorded signal essentially only arises from those mode combinations which actually excite the Raman resonance. Other, nonresonant, modes will only give a small contribution to the signal (majority species assumed). Let $\Delta$ denote an effective average line spacing between the Raman resonances within the observed spectral interval. There will be (for $\Delta > \Gamma_r, \Gamma_{(r+p)}$)

$$\frac{W}{\Delta}\cdot\frac{\Gamma_r}{\Omega_s}$$

modes contributing to the signal for the single-mode case and, for the multimode case,

$$\frac{W}{\Delta}\cdot\frac{\Gamma_{(r+p)}}{\Omega_s}.$$

In comparison with a nonresonant spectrum the number of participating Stokes laser modes in a resonant spectrum then decreases from $W/\Omega_s$ to $(W\cdot\Gamma_r)/(\Delta\cdot\Omega_s)$ (single-mode case) and $(W\cdot\Gamma_{(r+p)})/(\Delta\cdot\Omega_s)$ (multimode case). The increase in noise according to Eq. (1) is then

$$\sqrt{\frac{\Delta}{\Gamma_r}} \quad \text{single-mode laser,}$$

$$\sqrt{\frac{\Delta}{\Gamma_{(r+p)}}} \quad \text{multimode laser.}$$

If $\Delta$ is not known one may instead study the ratio of the increase in noise when the measurement is changed from the nonresonant to the resonant case:

$$\frac{\text{noise}_{NR,SM}}{\text{noise}_{NR,MM}}\cdot\sqrt{\frac{\frac{\Delta}{\Gamma_r}}{\frac{\Delta}{\Gamma_{(r+p)}}}} = \frac{\text{noise}_{NR,SM}}{\text{noise}_{NR,MM}}\cdot\sqrt{\frac{\Gamma_{(r+p)}}{\Gamma_r}} = \frac{\text{noise}_{R,SM}}{\text{noise}_{R,MM}}.$$

The indices $R$, $NR$, $SM$, and $MM$ stand for resonant, nonresonant, single-mode, and multimode, respectively.

For a flame spectrum at 1580 K, $\Gamma_r \approx 0.04\ cm^{-1}$. Snelling *et al.* measured their pump laser bandwidth to $\Gamma_p \approx 0.1\ cm^{-1}$. Using the approximate formula[24]

$$W_V \approx \frac{W_L}{2} + \sqrt{\left(\frac{W_L}{2}\right)^2 + W_G^2},$$

where $W_V$ is the FWHM of a Voigt profile resulting from the convolution of a Lorentzian and a Gaussian profile with FWHM of $W_L$ and $W_G$, respectively, the convoluted pump laser and Raman width are calculated to

$$\Gamma_{(r+p)} \approx \frac{\Gamma_r}{2} + \sqrt{\left(\frac{\Gamma_r}{2}\right)^2 + \Gamma_p^2} \approx 0.12\ \text{cm}^{-1}.$$

The values in Table I together with the values on $\Gamma_r$ and $\Gamma_{(r+p)}$ inserted on the left-hand side give

$$\frac{6.4}{8.6}\sqrt{\frac{0.12}{0.04}} \approx 1.4,$$

and for the right-hand side one obtains

$$\frac{22}{15.6} \approx 1.4.$$

As further numerical examples we may estimate the noise for the resonant and nonresonant single-mode cases. Let $W = 2\ \text{cm}^{-1}$, $\Gamma_r = 0.04\ \text{cm}^{-1}$, $\Omega_s = 0.009\ \text{cm}^{-1}$.[13] The noise will be $(\Omega_s/W)^{1/2} \approx 7\%$, nonresonant case, and $[(\Omega_s\Delta)/(W\Gamma_r)]^{1/2} \approx 30 \cdot \Delta^{1/2}\%$, resonant case, where any value between 0.1 and 2 $\text{cm}^{-1}$ may be reasonable for $\Delta$ in a $N_2$ $Q$-branch spectrum. In general attempting to narrow the bandwidth of the pump laser by an intracavity etalon will only result in increased single pulse CARS noise; the best performance should result when the pump bandwidth is as large as is consistent with acceptable resolution. We also note that the observation in Ref. 16 of reduced CARS noise when introducing aberration effects in the Stokes beam focusing optics (thus enabling also higher-order transverse modes in the Stokes laser to participate in the CARS process) is consistent with the model.

Although the model is partly successful it is clear that several details concerning noise in CARS spectra are still unclear and the issue seems to need further investigations.

## III. Referencing

In principle, variations in the CARS signal due to shot-to-shot fluctuations in the dye laser spectral profile can be eliminated by recording an anti-Stokes spectrum from an element with a constant nonresonant susceptibility (a reference spectrum) simultaneously with the resonant CARS spectrum. The reference spectrum then mirrors the dye laser profile in that particular laser shot. The ratio between the resonant spectrum and the reference spectrum should be insensitive to shot-to-shot variations in the dye laser spectral profile. It is well known that it is very difficult to completely eliminate noise due to shot-to-shot fluctuations in the dye laser spectral profile using this technique.[4,5,25]

The extent to which the spectral fluctuations can be reduced critically depends on the experimental situation in a way closely related to the discussion in the previous section. In Fig. 2 a broad slit function, a narrow isolated Raman line, and a number of Stokes laser modes are shown. The reference spectrum intensity in a given pixel gives the average intensity of the $W/\Omega_s$ dye laser modes generating the signal recorded by that particular pixel. However, the intensity in the resonant spectrum is determined by the intensity of the $\Gamma_r/\Omega_s$ (single-mode case) or $\Gamma_{(r+p)}/\Omega_s$ (multimode case) modes exciting the Raman resonance. If $W \gg \Gamma_r$ [or alternatively $W \gg \Gamma_{(r+p)}$], our information about the intensity of the particular modes exciting the Raman resonance will only be marginally improved. On the average the noise will decrease a factor of $(1 - \eta)^{1/2}$ where $\eta$ is the ratio between the number of modes contributing to the resonant spectrum and the number of modes contributing to the nonresonant spectrum. The best improvement of the S/N with referencing is obtained when the slit function is narrower than the Raman resonance. This condition is easiest to fulfill at high pressure because the resonance lines will then overlap. According to our model referencing may be easier to perform using a broadband pump laser. Preferably the spectral width of the pump laser should be of about the same size as the line spacing (or alternatively of the same width as the slit function in the case of an isolated line). There may be other possibilities like introducing a frequency chirp such that the Stokes laser modes are scanned over the resonance.

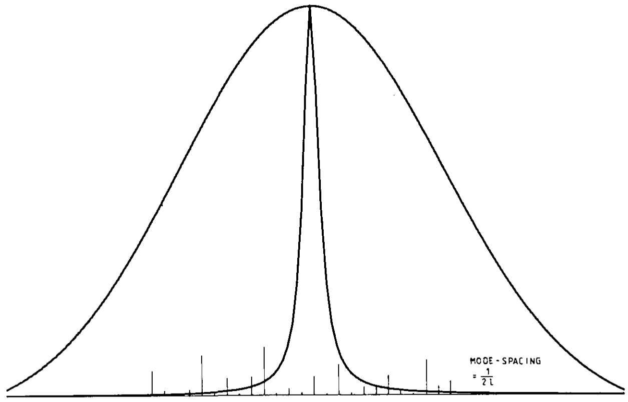


Fig. 2. Dye laser modes, a narrow isolated Lorentzian Raman resonance, and a wider Gaussian slit function are shown. $L$ is the (optical) length of the dye laser cavity. The intensity in the nonresonant spectra will be determined by the total intensity in the modes selected by the detection slit function. The intensity in the resonant spectra, however, is determined only by the total intensity in the smaller subgroup selected by the Raman resonance.

While the arguments above imply that successful referencing is most easily obtained with a multimode pump laser, there may also be disadvantages. It has been pointed out that the reference spectra may be sensitive to the precise time overlap of the pump laser and dye laser beams.[5] In particular this might be the case if the duration of the anti-Stokes pulse is small compared to the intermode beat periods. The sensitivity of reference spectra to the precise time overlap can possibly be investigated by simultaneously recording two reference spectra and looking at the cross correlation between the spectra as one of the beams in one of the reference cells is delayed.

## IV. Discussion and Conclusions

The model presented here indicates that the new techniques in which the molecular Raman resonance is excited with one[20,21,26] or two[18,19] broadband dye lasers should be particularly noise free as every transition is

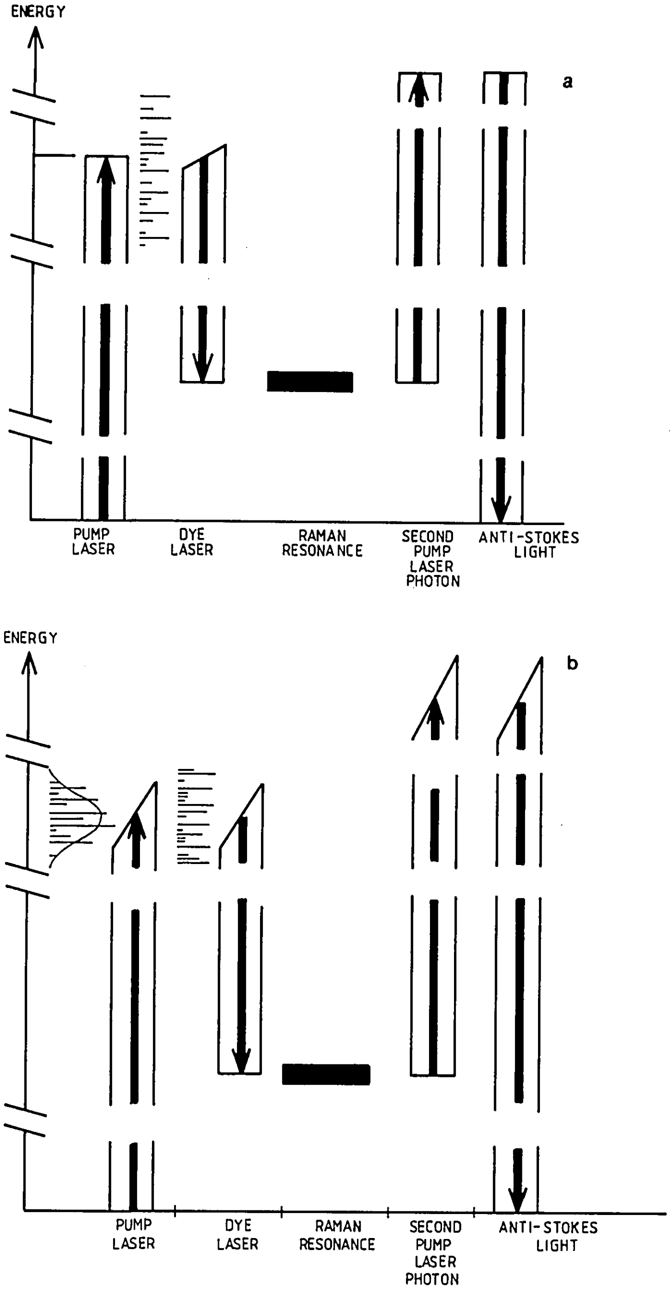


Fig. 3. Noise, as defined in the text, in two nonresonant CARS spectra recorded using (a) normal vibrational CARS and (b) dual broadband rotational CARS is shown. In a spectra with no noise all points would have intensity 1.0. The noise in the two spectra is 9.3% and 4.5%, respectively. The slope in the upper spectra arises from shot-to-shot fluctuations in the position of the dye laser spectral peak. The dispersion is 0.6 Å/channel.

excited by a very large number of modes. Indeed, this seems to be the case. In Fig. 3 the noise in a nonresonant spectrum recorded with the normal CARS configuration (a) and the (rotational) dual broadband technique (b) are shown. Twenty Stokes laser spectra and twenty spectra for each of the two cases above were recorded using the dye rhodamine 640. The noise was evaluated to 4.9% for the Stokes laser spectra, 9.6% for the normal CARS configuration, and 4.7% for the dual broadband configuration. The noise in the two spectra in Fig. 3 was (a) 9.3% and (b) 4.5%. That the noise in the dual broadband case is not higher in the CARS spectra than in the Stokes laser spectra is in contrast to what is obtained using the normal CARS configuration. The present investigation as well as previous ones have shown that for the normal configuration the noise is higher in the CARS spectra than in the Stokes laser spectra regardless of whether it is a single-mode[13,16] or a multimode[13,14,16,21] pump laser that is used. The spectrum in Fig. 3(a) sits on a slope because the position of the dye spectral peak fluctuates from shot to shot. This type of fluctuation has no effect on the rotational dual broadband spectra, on the other hand these are more sensitive to fluctuations in the dye laser bandwidth. It is also likely that referencing should work well with this type of technique.

Furthermore, the dual broadband techniques have calculational advantages. For these techniques the intensity convolution integral needed for theoretical spectra synthesis has a particularly simple form.[26] This is so for one thing because all sources are statistically independent. The issue of cross-coherence effects[10,27] does not arise, and there need be no concern that non-Gaussian second harmonic pump statistics[28–31] will invalidate a convolution integral based on Gaussian statistics for the laser sources.

Summarizing: a simple, semiquantitative model that may be useful as an aid in understanding the relative noise performance of various broadband CARS techniques has been presented. The model states that the CARS noise is approximately equal to the inverse square root of the number of modes from the Stokes laser generating the signal recorded by a single pixel. This means that compared with using a single-mode pump laser for generating resonant CARS spectra the use of a multimode pump laser will enable a higher number of Stokes laser modes to excite the Raman resonance. This property of the multimode pump laser will tend to reduce the noise. However, more definite statements can probably not be made as issues like pump mode intensity fluctuations, possible intermode beats, or questions whether the dye pumping process causes the dye laser intensity to follow fluctuations in the pump laser intensity still may need further investigation. [Clearly the conclusion above concerning the relative noise performance of multimode and single-mode lasers applies only when pump laser light is used for exciting the molecular Raman resonance. The situation where the pump laser light only is scattered off the vibrating molecule (i.e., providing only what is denoted the second pump laser photon in Fig. 1) is not treated here. Further, these conclusions might also change for high pressure applications with broad Raman resonances.]

The model also implies that to have optimum performance in referencing it must be assured that the same Stokes modes contribute to the signal in a given diode detector element in the reference and in the resonant spectra. This is more easily achieved at high pressure, with a narrow slit function and/or with a broadband pump laser.

Finally, we note that the present analysis agrees with the observation that the noise in CARS spectra seems lower for the dual broadband type techniques than for the ordinary CARS configurations. The definitive answer to this question will have to await the demonstration of single pulse capability with the former technique. After submission of this paper it came to our knowledge that an analysis similar to the one presented in this paper has been proposed by Snelling *et al.*[32]

The authors wish to thank D. A. Greenhalgh, L. G. Rosengren, S. Svanberg, H. Edner, and H. Bergström for valuable discussions. The financial support by Sydkraft AB, the Swedish National Board for Technical Development (STU), AB Volvo, and the Swedish National Energy Administration (STEV) is gratefully acknowledged. R. J. Hall wishes to acknowledge the hospitality of A. Leipertz and Ruhr-Universitat Bochum and the financial support of the Volkswagen Foundation.

## References

1. S. A. J. Druet and J.-P. E. Taran, "CARS Spectroscopy," Prog. Quantum. Electron. **7**, 1 (1981).
2. R. J. Hall and A. C. Eckbreth, "Coherent Anti-Stokes Raman Spectroscopy (CARS) Application to Combustion Diagnostics," Laser Appl. **5**, 213 (1984).
3. R. L. Farrow, R. P. Lucht, G. L. Clark, and R. E. Palmer, "Species Concentration Measurements Using CARS with Nonresonant Susceptibility Normalization," Appl. Opt. **24**, 2241 (1985).
4. M. Pealat, P. Bouchardy, M. Lefebvre, and J.-P. Taran, "Precision of Multiplex CARS Temperature Measurements," Appl. Opt. **24**, 1012 (1985).
5. A. C. Eckbreth, G. M. Dobbs, J. H. Stufflebeam, and P. A. Tellex, "CARS Temperature and Species Measurements in Augmented Jet Engine Exhausts," Appl. Opt. **23**, 1328 (1984).
6. L. P. Goss, D. D. Trump, B. G. MacDonald, and G. L. Switzer, "10-Hz Coherent Anti-Stokes Raman Spectroscopy Apparatus for Turbulent Combustion Studies," Rev. Sci. Instrum. **54**, 563 (1983).
7. M. Alden and S. Wallin, "CARS Experiments in a Full-Scale (10 × 10 m) Industrial Coal Furnace," Appl. Opt. **24**, 3434 (1985).
8. E. J. Beiting, "Multiplex CARS Temperature Measurements in a Coal-Fired MHD Environment," Appl. Opt. **25**, 1684 (1986).
9. W. B. Roh, P. W. Schreiber, and J. P. E. Taran, "Single-Pulse Coherent Anti-Stokes Raman Scattering," Appl. Phys. Lett. **29**, 174 (1976).
10. R. E. Teets, "Accurate Convolutions of Coherent Anti-Stokes Raman Spectra," Opt. Lett. **9**, 226 (1984).
11. L. A. Rahn, R. L. Farrow, and R. P. Lucht, "Effects of Laser Field Statistics on Coherent Anti-Stokes Raman Spectroscopy Intensities," Opt. Lett. **9**, 223 (1984).
12. R. L. Farrow and L. A. Rahn, "Interpreting Coherent Anti-Stokes Raman Spectra Measured with Multimode Nd:YAG Pump Lasers," J. Opt. Soc. Am. B **2**, 903 (1985).
13. D. R. Snelling, R. A. Sawchuk, and R. E. Mueller, "Single Pulse CARS Noise: a Comparison Between Single-Mode and Multimode Pump Lasers," Appl. Opt. **24**, 2771 (1985).
14. D. A. Greenhalgh and S. T. Whittley, "Mode Noise in Broadband CARS Spectroscopy," Appl. Opt. **24**, 907 (1985).
15. R. J. Hall and D. A. Greenhalgh, "Noise Properties of Single-Pulse Coherent Anti-Stokes Raman Spectroscopy with Multimode Pump Sources," J. Opt. Soc. Am. B **3**, 1637 (1986).
16. D. R. Snelling, G. I. Smallwood, R. A. Sawchuk, and T. Parameswaran, "Nonlinearity and Single Shot Noise Problems in CARS Spectroscopy," AGARD Conf. Proc. **00**, 000 (198x), to be published.
17. S. Kröll, "The influence of mode beating and pump mode fluctuations on noise in broadband CARS," in progress.
18. A. C. Eckbreth and T. J. Anderson, "Dual Broadband CARS for Simultaneous, Multiple Species Measurements," Appl. Opt. **24**, 2731 (1985).
19. A. C. Eckbreth and T. J. Anderson, "Dual broadband USED CARS," Appl. Opt. **25**, 1534 (1986).
20. A. C. Eckbreth and T. J. Anderson, "Simultaneous Rotational Coherent Anti-Stokes Raman Spectroscopy and Coherent Stokes Raman Spectroscopy with Arbitrary Pump–Stokes Spectral Separation," Opt. Lett. **11**, 496 (1986).
21. M. Aldén, P.-E. Bengtsson and H. Edner, "Rotational CARS Generation Through a Multiple Four-Color Interaction," Appl. Opt. **25**, 4493 (1986).
22. A. V. Masalov, "Spectral and Temporal Fluctuations of Broadband Laser Radiation," Prog. Opt. **22**, 145 (1985).
23. R. E. Teets, "CARS Signals: Phase Matching, Transverse Modes, and Optical Damage Effects," Appl. Opt. **25**, 855 (1986).
24. E. E. Whiting, "An Empirical Approximation to the Voigt Profile," J. Quant. Spectrosc. Radiat. Transfer **8**, 1379 (1968).
25. M. Aldén, K. Fredriksson, and S. Wallin, "Application of a Two-Color Dye Laser in CARS Experiments for Fast Determinations of Temperatures," Appl. Opt. **23**, 2053 (1984).
26. M. A. Yuratich, "Effects of Laser Linewidth on Coherent Anti-Stokes Raman Spectroscopy," Mol. Phys. **38**, 625 (1979).
27. H. Kataoka, S. Maeda, and C. Hirose, "Effects of Laser Linewidth on the Coherent Anti-Stokes Raman Spectroscopy Spectral Profile," Appl. Spectrosc. **36**, 565 (1982).
28. J. H. Churnside, "Second Harmonic Generation Using Partially Coherent Light," Opt. Commun. **51**, 207 (1984).
29. R. J. Hall, "The Statistical Behaviour of Nonresonant CARS Intensities," Opt. Commun. **56**, 127 (1985).
30. R. J. Hall, "Theoretical Analysis of Non-Thermal Pump Effects in Broadband CARS Spectroscopy," Opt. Quantum Electron. **18**, 319 (1986).
31. G. S. Agarwal and R. L. Farrow, "Theoretical Modeling of Two-Color Coherent Anti-Stokes Raman Spectroscopy Spectra Measured with a Frequency-Doubled, Multimode Pump Laser," J. Opt. Soc. Am. B **3**, 1596 (1986).
32. D. R. Snelling, G. J. Smallwood, R. A. Sawchuk, and T. Parameswaran, "Precision of Multiplex CARS Temperatures Using Both Single-Mode and Multimode Pump Lasers," Appl. Opt. **26**, 99 (1987).