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Kröll, Stefan; Sandell, D

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Influence of laser-mode statistics on noise in nonlinear-optical processes—application to single-shot broadband coherent anti-Stokes Raman scattering thermometry

Stefan Kröll

Department of Atomic Physics, Combustion Centre, Lund Institute of Technology, Box 118, S-221 00 Lund, Sweden

Dennis Sandell

Department of Mathematical Statistics, Lund Institute of Technology, Box 118, S-221 00 Lund, Sweden

A quantitative approach for calculating the noise due to the stochastic nature of multimode laser radiation in nonlinear-optical processes is presented. The model is applicable when it is appropriate to describe the nonlinear interaction with a perturbation expansion in the incoming field, and it is derived under the assumption of independent individual laser-mode intensities and phases. It is possible to separate noise contributions from mode-amplitude and -phase fluctuations, respectively, and also to identify the noise contribution from each laser source. For coherent anti-Stokes Raman scattering (CARS) thermometry, the model shows that with a single-mode pump laser the stochastic phases in the dye laser do not generate noise in the conventional approach and that amplitude fluctuations in the dye laser(s) do not (significantly) generate noise in the dual-broadband approaches. Thus, in the dual-broadband approaches, the spectral noise in the Stokes beam is not a lower limit for the noise in the CARS beam. The model seems to overestimate the noise due to phase fluctuations and to underestimate the noise due to amplitude fluctuations.

1. INTRODUCTION

There has been great interest lately in various ways in which laser statistics influence coherent anti-Stokes Raman scattering (CARS) spectra.[1-10] As CARS is used for temperature and concentration measurements,[11] it is important to characterize the CARS process accurately. It is generally assumed that the statistical fluctuations of the laser fields impose a fundamental limitation on the accuracy that can be obtained in CARS thermometry [e.g., see Refs. 2 and 12-14]. There has been a lively debate on whether the best accuracy in CARS thermometry is obtained with a multimode or a single-mode pump laser.[4,6,8-10,13] Recently new approaches to CARS thermometry have been demonstrated.[15-17] In one of these approaches (rotational dual-broadband CARS) the noise obtained in non-Raman resonant spectra was a factor of 2 lower than with a conventional CARS setup.[17] Currently the optimum configuration of a CARS thermometry setup is an open question. Comparisons of the experimental characteristics of CARS spectra recorded using a multimode or a single-mode pump laser have been performed for the conventional CARS setup. These have demonstrated lower noise for non-Raman-resonant spectra recorded with a single-mode YAG laser than those recorded with a multimode YAG laser but significantly higher noise for a single-mode than for a multimode YAG laser when Raman-resonant spectra were recorded.[4,9,14] A qualitative explanation of the comparatively high noise level in the Raman-resonant single-mode spectra has been proposed.[9,10] However, the model used there failed to explain why a single-mode laser yielded lower noise than a multimode laser in non-Raman-reso-

nant spectra. We have thus developed a quantitative model that can be used to estimate the noise in a general nonlinear-optical process, provided that the induced polarization can be expressed by using a perturbation expansion in the electric fields and that the generated radiation has negligible intensity in comparison with the intensity of the incoming fields. This model has been used to calculate the noise in different experimental approaches to broadband CARS.

The CARS process is schematically depicted in Fig. 1. The most common experimental setup for CARS thermometry utilizes a Nd:YAG-pumped dye-laser system, in which the photons at frequencies ω_a and ω_c are taken from the frequency-doubled YAG laser light and the photon at frequency ω_b is provided by the dye laser. In broadband CARS thermometry the dye laser is operated in broadband mode (FWHM >100 cm^{-1}). In this way the population differences in all rotational–vibrational transitions are probed simultaneously. The generated anti-Stokes beam, with the frequency ω_{as}, is dispersed in a spectrograph and imaged onto a diode array. In this way the anti-Stokes spectrum of the investigated molecule is obtained in one single laser shot.[18]

Considerable effort has been put into theoretical modeling of CARS spectra. The calculation of theoretical spectra involves integration over all laser frequencies.[19-23] Clearly this is much simplified if a single-mode pump laser is used, as the laser linewidth is then considerably smaller than the Raman linewidth and the laser radiation can simply be described by a delta function in frequency space. For the multimode YAG laser, an accurate calculation turns out to be quite complicated.[5-7] CARS spectra recorded with the
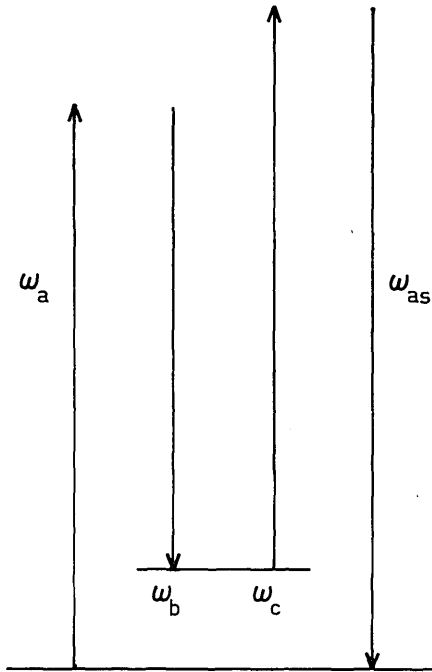
Fig. 1. Conceptual picture of the CARS process. ω_a, ω_b, and ω_c represent the frequencies of the incoming photons, and ω_{as} represents the frequency of the CARS photon. A Raman resonance is indicated.

conventional setup are generated by a second-order process in the frequency-doubled YAG laser light. Thus, to calculate such a CARS spectrum, the autocorrelation function of the frequency-doubled YAG radiation must be known. (This is the case in principle; however, for temperature determinations, details of the laser statistics may not in practice necessarily affect the result to any significant degree. Compare, e.g., Ref. 7.) The more complicated calculation is an argument against using a multimode YAG laser in CARS thermometry, in particular as the autocorrelation function appears to be a characteristic of each individual laser[6,7] and also varies with the operating conditions.[3,24] Thus, although yielding higher noise in Raman resonant spectra, the single-mode laser is more attractive from a modeling point of view.

At this point the new experimental approaches to CARS seem to have attractive properties. In these, only one photon from the YAG laser is used for the generation of each anti-Stokes photon, thus eliminating a major computational difficulty. In these so-called dual-broadband techniques, two broadband dye lasers drive the vibrational Raman resonance,[15] or, in the case of rotational CARS, both photons driving the Raman resonance may actually be provided by one single broadband dye laser.[16,17] The YAG laser beam is then scattered off the vibrating molecules. The three experimental approaches, conventional CARS, dual-broadband CARS, and rotational dual-broadband CARS, henceforth abbreviated CC, DBC, and RDBC, respectively, are conceptually described in Fig. 1 and Table 1.

As usual, in the case of broadband CARS,[2,4,8,9] noise is here defined as $\sigma(I)/\langle I \rangle$, where $\sigma(I)$ is the shot-to-shot standard deviation of the intensity I, the anti-Stokes intensity registered by a diode, and $\langle I \rangle$ is the average intensity registered

by the diode. The weak-signal limit, where, e.g., shot noise, Johnson noise, and incoherent background must be taken into account, was previously analyzed for several different four-wave-mixing processes (Ref. 25 and Ref. 26, p. 156). The present analysis is primarily valid for the high-signal-intensity limit, where the noise level is primarily determined by the shot-to-shot fluctuations in the laser-mode phases and intensities. A main feature of the present model is that the noise contributions from the stochastic laser-mode phases and the stochastic laser-mode intensities can be calculated separately and that the noise contribution from each laser source is obtained separately. A simplified physical picture of the different sources of noise, as defined in this paper, can be obtained by considering two electric-field modes, E_a and E_b, with amplitudes a and b and frequencies ω_a and ω_b and phases θ_a and θ_b, where

$$E_a = a \exp[i(\omega_a t + \theta_a)],$$

$$E_b = b \exp[i(\omega_b t + \theta_b)],$$

and t is time.

The intensity (I_0) from such a field would be

$$I_0 = |E_a + E_b|^2 = a^2 + b^2 + 2ab \cos[(\omega_a - \omega_b)t + \theta_a - \theta_b].$$

The noise due to shot-to-shot fluctuations in the term a^2 (b^2) is considered to arise from mode-intensity fluctuations in mode a (b), and the noise due to shot-to-shot fluctuations in the cross term

$$2ab \cos[(\omega_a - \omega_b)t + \theta_a - \theta_b]$$

is considered to arise from phase fluctuations. It may be noted that in contrast to the two first terms (a^2, b^2) the average intensity contribution from several shots is zero for the cross term. Noise due to phase fluctuations is often referred to as mode beating. We have used a notation in which we call the noise from the cross term mode beating if $\omega_a \neq \omega_b$ and phase incoherence if $\omega_a = \omega_b$. The multimode laser fields are modeled as a sum of a large number of modes with independent phases and amplitudes. This is a commonly used representation for the radiation from a broadband laser.[27] The modes in the frequency-doubled YAG laser radiation are, however, not strictly independent because partial mode locking occurs in the frequency-doubling process.[1,6] Also, a number of publications have appeared lately that indicate that this representation may not be correct for a broadband pulsed dye laser; at least this seems to be the case if the number of modes is small ($\sim$10).[28,29] To keep the problem tractable, independent phases and amplitudes have still been assumed for all fields. Spatial effects in the mode structure[2,6,30] and the time dependence of both phases and amplitudes have also been neglected. Given the assumptions above, a general expression for the noise in a four-wave-mixing process is given in Section 2. In Section 3

Table 1. Laser Types Used for Each of the Steps in the CARS Process Depicted in Fig. 1

	CC	DCB	RDBC
Pump laser (narrow band)	ω_a, ω_c	ω_c	ω_c
First broadband dye laser	ω_b	ω_a	ω_a, ω_b
Second broadband dye laser	—	ω_b	—

we describe how we have applied the approach outlined in Section 2 to calculate the noise in non-Raman resonant CARS and to rotational Raman-resonant CARS for the case in which the spectral width of the pump laser is much broader than the Raman resonance. A discussion of some crucial details in the evaluation of the expressions for the noise and a physical interpretation of the different mechanisms that give rise to the noise are given in Section 4. In Section 5 a discussion is given, and in Section 6 a summary of the results is presented. All explicit calculations and proofs are collected in Appendixes A–E. In Appendix A, we show how, by starting with light fields with stochastic-mode amplitudes and phases, it is possible to obtain a deterministic expression for the mean and variance for a signal generated in a four-wave-mixing process by using three independent light sources. In Appendixes B and C examples of the explicit calculations of the noise and some mathematical details are given. In Appendix D expressions that can be used to calculate the noise in a more general n-wave-mixing process are given, and, finally, in Appendix E analytical expressions are given for the noise in various approaches to CARS as calculated by the presented model.

2. MODEL FOR CALCULATING NOISE

The description of the nonlinear interaction is similar to the ones in Refs. 6 and 31. The laser fields are described as a summation over an infinite number of independent modes:

$$E_a = \sum_k a_k \exp\left\{-\frac{1}{2}\left(\frac{k\Omega_a}{\Gamma_a}\right)^2 + i[(\omega_a^0 + k\Omega_a)t + \theta_k]\right\} + \text{c.c.},$$

$$E_b = \sum_r b_r \exp\left\{-\frac{1}{2}\left(\frac{r\Omega_b}{\Gamma_b}\right)^2 + i[(\omega_b^0 + r\Omega_b)t + \phi_r]\right\} + \text{c.c.},$$

$$E_c = \sum_m c_m \exp\left\{-\frac{1}{2}\left(\frac{m\Omega_c}{\Gamma_c}\right)^2 + i[(\omega_c^0 + m\Omega_c)t + \psi_m]\right\} + \text{c.c.}$$

Many symbols used in this paper are defined in Table 2. Here ω_a^0, ω_b^0, and ω_c^0 are the center frequencies of the multi-

Table 2. Definition of Symbols

Symbol	Definition
Γ_i	Bandwidth of laser i
Γ_r	Linewidth of Raman resonance
W	Spectral width of detection system's instrument function
Γ	$(\Gamma_a^2 + \Gamma_b^2 + \Gamma_c^2 + W^2)^{1/2}$, where a, b, and c may or may not be different
Ω_i	Mode spacing of laser i
ω_i	Frequency of laser i
ω_{as}	Anti-Stokes frequency
Δ_α	Frequency difference between the center frequency transmitted through the detection system to diode α and the frequency where a nonresonant CARS signal would have its maximum
T	Duration of anti-Stokes pulse
t	Time
I	Intensity detected by one diode in the diode array
$\langle x \rangle$	Expected value of x
$\sigma(x)$	Standard deviation of x
$\sigma(I)/\langle I \rangle$	noise

mode fields and c.c. stands for a complex conjugate. Ω_a, Ω_b, and Ω_c and Γ_a, Γ_b, and Γ_c are mode spacings and linewidths, respectively, of the laser sources. The electric-field mode amplitudes a, b, and c and phases ϕ, θ, and ψ are assumed to be stochastic variables varying slowly in time. This time dependence will be neglected henceforth. a_k^2, b_k^2, and c_k^2 belong to the distributions $\Gamma(1, i_a)$, $\Gamma(1, i_b)$, and $\Gamma(1, i_c)$, respectively (see Appendix A), and the phase factors are all uniformly distributed in the interval $(0, 2\pi)$. The phase factors have been chosen such that the mode amplitudes are real. The lower-order spatial modes focus more tightly and are thus the ones mainly contributing to the generation of the CARS signal.[2] The lasers are assumed to operate in their lowest-order spatial mode, and differences in beam overlap are neglected. Thus the spatial integration may be replaced by multiplication by a constant.[6]

In the weak-interaction limit the nonlinear polarization $[P^{(3)}]$ for a general four-wave-mixing process can be written as (Ref. 26, p. 46)

$$P^{(3)} \sim \chi^{(3)} E_a E_b E_c, \tag{1}$$

where $\chi^{(3)}$ is the third-order nonlinear susceptibility. In the weak-signal limit the generated electric field is proportional to the induced polarization. In particular, the field at frequencies near

$$\omega_{as}^0 = \omega_a^0 - \omega_b^0 + \omega_c^0$$

[which would correspond to the anti-Stokes field (E_{as}) in CARS] is given by

$$E_{as} \sim \sum_{krm} E_{as}(\omega_{krm}) = \sum_k \sum_r \sum_m \chi_{krm}^{(3)} a_k b_r c_m$$
$$\times \exp\left\{-\frac{1}{2}\left[\left(\frac{k\Omega_a}{\Gamma_a}\right)^2 + \left(\frac{r\Omega_b}{\Gamma_b}\right)^2 + \left(\frac{m\Omega_c}{\Gamma_c}\right)^2\right]\right\}$$
$$\times \exp\{i[(\omega_a^0 - \omega_b^0 + \omega_c^0 + k\Omega_a - r\Omega_b + m\Omega_c)t$$
$$+ \theta_k - \phi_r + \psi_m]\} + \text{c.c.},$$

where ω_{krm} is the frequency of the generated field for the particular mode combination k, r, and m, and $\chi_{krm}^{(3)}$ may be frequency dependent. For the generated intensity (I) at, e.g., frequency $\omega_{as} = \omega_a - \omega_b + \omega_c$, we may write

$$I = \sum_{\substack{klr \\ smn}} a_k a_l b_r b_s c_m c_n \exp[i(\theta_k - \theta_l - \phi_r + \phi_s + \psi_m - \psi_n)]$$
$$\times H(k, l, r, s, m, n).$$

H is a deterministic function containing, e.g., the third-order nonlinear susceptibility, the response of the detection system (to be briefly discussed in Section 3), and the nonstochastic parts of the laser fields. The expected value $E(I)$ and the variance $V(I)$ for the intensity above (assuming three independent laser sources) can be written in the simple form (see Appendix A)

$$E(I) = i_a i_b i_c \sum_{krm} H(k, k, r, r, m, m), \tag{2a}$$

$$V(I) = (i_a i_b i_c)^2 \sum_{\substack{klr \\ smn}} [H(klrrmm)H(klssnn)$$

$$+ H(kkrsmm)H(llrsnn) + H(kkrrmn)H(llssmn)$$

$$+ H(kkrsmn)H(llrsmn) + H(klrrmn)H(klssmn)$$

$$+ H(klrsmm)H(klrsnn) + H(klrsmn)H(klrsmn)].$$

$$(2b)$$

(The commas between indices have been omitted.) Provided that approximation (1) is valid and the generated field is

weak, the variance above is a general analytical expression for the fluctuations in a signal from a four-wave-mixing process, where all the process-dependent parts are contained in the deterministic function H. In particular, it may be noted that the expression is valid for all frequency combinations $\omega_4 = \pm\omega_a \pm \omega_b \pm \omega_c$. In Appendix D we give more-general expressions, which can be used to calculate the noise in a nonlinear process in which an arbitrary number of photons are taken from each laser source. Consequently, these expressions are applicable to the CC and RDBC cases. With N different lasers and if we take f_k photons from laser k, the variance will contain

$$\prod_{k=1}^{N} (2f_k)! - \prod_{k=1}^{N} (f_k!)^2$$

terms, and the expected value will contain

$$\prod_{k=1}^{N} f_k!$$

terms. Thus, unless the function H has some symmetries in the permutation of the indices (as is the case in, e.g., nonresonant CC, where the number of terms in the variance is reduced from 44 to 5 by symmetry), the large number of terms begins to make the explicit evaluation rather cumbersome when more than one photon is taken from a laser source.

3. APPLICATION TO COHERENT ANTI-STOKES RAMAN SCATTERING

In this section we describe how the model in Section 2 can be applied to calculate the noise in non-Raman-resonant and Raman-resonant broadband CARS. The signal in broadband CARS thermometry is normally dispersed by a spectrograph and registered by a diode array. It is assumed that each diode in the detector array sees the detection system as a Gaussian with a width W. The electric field reaching a diode in the diode array is then given by

$$\sum_{krm} E_{as}(\omega_{krm})S_\alpha(\omega_{krm}),$$

where $[S_\alpha(\omega)]^2$ is the detection system's instrument function for diode α, chosen such that $S_\alpha(\omega) = \exp[-(\omega_\alpha - \omega)^2/(2W^2)]$, and ω_α is the center frequency impinging upon diode α. The nonlinear third-order susceptibility $\chi^{(3)}$ is a real constant in the non-Raman-resonant case. The anti-Stokes intensity registered by the detector is then proportional to

$$\sum_{\substack{krm \\ lsn}} \frac{1}{T} \int_{-T/2}^{T/2} E_{as}(\omega_{krm})S_\alpha(\omega_{krm})\overline{E_{as}}(\omega_{lsn})\overline{S_\alpha}(\omega_{lsn})dt = I(\Delta_\alpha),$$

yielding

$$H_{\Delta_\alpha}(k, l, r, s, m, n) = \chi_{krm}^{\overline{(2)}}\chi_{lsn}^{(37)} \exp\left\{-\frac{1}{2}\left[\frac{(k^2 + l^2)\Omega_a{}^2}{\Gamma_a{}^2} + \frac{(r^2 + s^2)\Omega_b{}^2}{\Gamma_b{}^2} + \frac{(m^2 + n^2)\Omega_c{}^2}{\Gamma_c{}^2}\right]\right\}$$

$$\times \exp\left\{-\frac{1}{2}\left[\frac{(k\Omega_a - r\Omega_b + m\Omega_c - \Delta_\alpha)^2 + (l\Omega_a - s\Omega_b + n\Omega_c - \Delta_\alpha)^2}{W^2}\right]\right\}\text{sinc}\left(\frac{\Delta\omega T}{2}\right), \quad (3)$$

where

$$\text{sinc}(x) = \frac{\sin(x)}{x},$$

$$\Delta\omega = (k - l)\Omega_a - (r - s)\Omega_b + (m - n)\Omega_c,$$

and

$$\Delta_\alpha = \omega_\alpha - \omega_a{}^0 + \omega_b{}^0 - \omega_c{}^0.$$

Consider a single diode in the diode-array detector. Assume that the intensity (I) in this diode is normalized by dividing by the total intensity recorded by all diodes. This is normally done in order to eliminate the overall intensity fluctuations in the anti-Stokes pulse such that only the spectral fluctuations in the laser pulses contribute to the noise.[4,9] The normalized intensity (I_N) in an arbitrary diode α may then be written as

$$I_N(\Delta_\alpha) = \frac{I(\Delta_\alpha)}{\sum_\beta I(\Delta_\beta)},$$

where the summation over β is made over all diodes in the diode array. The mean and variance for I_N can be estimated by using the first-order Gaussian approximation[32] for the ratio between two random variables. For broadband CARS the following conditions will generally be valid ($x = a, b, c$):

$$\Omega_x \ll \min(W, \Gamma_x), \quad (4a)$$

$$T\Omega_x \gg \pi/2. \quad (4b)$$

The conditions in expressions (4) imply that the summations over mode combinations necessary to calculate the intensity, in most cases, may be replaced by integrations without significant loss in accuracy. The exceptions when this is not the case are discussed in Section 4. The evaluation of $E(I)$ and $V(I)$ for the three-laser case is outlined in Appendix B. Explicit expressions for the noise calculated for non-Raman-resonant CARS using CC, DBC, and RDBC are given in Appendix E. In the calculations for single-mode pump laser cases the single-mode fields are assumed to be deterministic and are represented by delta functions in frequency space. For Raman-resonant CARS the calcula-

tion is restricted to the special case of a single Raman line centered at the transmission maximum of the detection system. With the exception of the RDBC case, the laser profiles are assumed to be tuned to give maximum intensity at the corresponding anti-Stokes wavelength. Thus, for the Raman-resonant case,

$$H(k, l, r, s, m, n) = \exp\left\{-\frac{1}{2}\left[\frac{(k^2 + l^2)\Omega_a^2}{\Gamma_a^2} + \frac{(r^2 + s^2)\Omega_b^2}{\Gamma_b^2} + \frac{(m^2 + n^2)\Omega_c^2}{\Gamma_c^2}\right]\right\}$$
$$\times \exp\left\{-\frac{1}{2}\left[\frac{(k\Omega_a - r\Omega_b + m\Omega_c - \Delta_\alpha)^2 + (l\Omega_a - s\Omega_b + n\Omega_c - \Delta_\alpha)^2}{W^2}\right]\right\} \quad (5)$$
$$\times \frac{\text{sinc}\left\{\frac{T}{2}[(k - l)\Omega_a - (r - s)\Omega_b + (m - n)\Omega_c]\right\}}{(k\Omega_a - r\Omega_b - \Delta_\alpha + i\Gamma_r)(l\Omega_a - s\Omega_b - \Delta_\alpha - i\Gamma_r)},$$

where $\Delta_\alpha = 0$ for CC and DBC. This approach is reasonably correct for rotational CARS, where the spacings between the Raman resonances are normally larger than the width of the instrument function. A vibrational CARS spectrum corresponds to an intermediate case, and the non-Raman-resonant situation corresponds to a case in which all anti-Stokes modes have nearly the same weight. The explicit evaluation of the Raman-resonant case, as above, is performed for

$$\Gamma_a, \Gamma_b, \Gamma_c, W \gg \Gamma_r \gg 1/T,$$

where Γ_r is the width of the Raman resonance. These conditions will generally be fulfilled for the YAG laser pumped system operating without an étalon in the YAG laser, but they are normally not fulfilled if an étalon is inserted into the YAG laser in order to narrow its bandwidth. In the second part of Appendix B the calculation of the noise for the Raman-resonant cases is briefly described, and the expressions for the noise are given in Appendix E.

4. ANALYSIS

One of the main differences between this treatment and the previous treatments of noise in broadband CARS[2,8,9,14] is that a quantitative estimate is made of the effect that random phases of the individual modes in the multimode lasers will have on the noise. The noise depends critically on how the sinc function in Eq. (3) is treated in the summation over all modes. This point is thus examined in some detail.

A necessary requirement for replacing the summations in Eqs. (2) by integrations is that there are not too few combinations of k, l, r, s, m, n yielding a value of $|\Delta\omega T| < \pi$.

As an example we have, for CC,

$$\Delta\omega = (k - l + m - n)\Omega_a - (r - s)\Omega_b = P\Omega_a + S\Omega_b,$$

where $P \equiv k - l + m - n$ and $S \equiv s - r$ are integers. $\Delta\omega$ is the beat frequency between the anti-Stokes modes $k\Omega_a - r\Omega_b + m\Omega_a$ and $l\Omega_a - s\Omega_b + n\Omega_a$. It is clear that for every P there exists an $S = S_{\text{MIN}}(P)$ yielding a minimum value of $\Delta\omega$ ($\Delta\omega_{\text{MIN}}$) such that $\Delta\omega_{\text{MIN}} = |P\Omega_a + S_{\text{MIN}}(P)\Omega_b| < \Omega_b/2$. With N pump modes these can be combined to a P in N^4 ways, and there will be $\sim 4N$ different values for P. For $T = 10^{-8}$ sec and $\Omega_b = 0.01$ cm^{-1}, $|\Delta\omega_{\text{MIN}}T| \approx 3\pi$. $\Delta\omega_{\text{MIN}}$ may be expected to be uniformly distributed in the interval $(-3\pi/T, 3\pi/T)$ as

P runs through its $4N$ different values. Thus there will be approximately $N^4/3$ pump-mode combinations yielding a value of $\Delta\omega T/2$ in the interval $(-\pi/2, \pi/2)$, and there will be roughly $4N/3$ different values of $\Delta\omega_{\text{MIN}}T$ in this interval. Computer simulations show that even for such small values of Γ_a as $\Gamma_a/\Omega_a = 10$, the error in approximating the sum by an integral is less than 1%. With a single-mode pump laser, however, $\Delta\omega_{\text{MIN}} = \Omega_b$ for $r \neq s$, and there are now no mode-beating terms with a value of $\Delta\omega T$ in the interval $(-6\pi, 6\pi)$. Thus terms for which $r \neq s$ will now only give a negligible contribution, and the sinc function is best replaced by a delta function $\delta(r - s)$. To visualize how the different noise terms appear, we examine the expression for the noise in non-Raman-resonant DBC [rewritten from Eq. (E3) below]:

$$\frac{\sigma(I)}{\langle I\rangle} = \frac{1}{(2\pi)^{1/4}}\left\{\sum_{x=a,b,c}\frac{\Omega_x\Gamma_x}{\Gamma}\frac{1}{[(\Gamma^2 - \Gamma_x^2]^{1/2}}\right.$$
$$\left.+ \frac{2\pi}{T}\sum_{x=a,b,c}\left[\frac{1}{W^2 + \Gamma_x^2} - \frac{1}{\Gamma^2}\right]^{1/2} + \frac{2\pi}{WT}\right\}^{1/2}. \quad (6)$$

Now, again consider Eq. (3):

$$H(k, l, r, s, m, n) = \exp\left\{-\frac{1}{2}\left[\frac{(k^2 + l^2)\Omega_a^2}{\Gamma_a^2} + \frac{(r^2 + s^2)\Omega_b^2}{\Gamma_b^2}\right.\right.$$
$$+ \frac{(m^2 + n^2)\Omega_c^2}{\Gamma_c^2}$$
$$\left.\left.+ \frac{(k\Omega_a - r\Omega_b + m\Omega_c - \Delta_\alpha)^2 + (l\Omega_a - s\Omega_b - n\Omega_c - \Delta_\alpha)^2}{W^2}\right]\right\}$$
$$\times \text{sinc}\left\{\frac{T}{2}[(k - l)\Omega_a - (r - s)\Omega_b + (m - n)\Omega_c]\right\},$$

where the indices have been grouped in pairs. If one and only one of the three pairs is different, as in the first three terms in Eq. (2b) [corresponding to the first three terms, respectively, in Eq. (6)], the sinc function is approximated by a delta function, as discussed above. This is explicitly done in Appendix B by setting $\sum H(k, l, r, r, m, m)H(k, l, s, s, n, n) \approx \int H(k, k, r, r, m, m)H(k, k, s, s, n, n)$ for the first term in Eq. (2b). If the first-order Gaussian approximation is used to calculate the variance for a function F, which is a function of the independent stochastic variables x_i, $i = 1, 2, 3, \ldots$, we have

$$V(F) = \sum_i V(x_i)\left[\left(\frac{\partial F}{\partial x_i}\right)^2\right]_E,$$

where the index E indicates that the derivative is to be evaluated at the expected values of x_i. The first term in Eq. (6) is (apart from a numerical factor) equal to

$$\sum_k V(a_k)\left[\left(\frac{\partial I_N}{\partial a_k}\right)^2\right]_E$$

in the Gaussian approximation, where a_k are the stochastic-mode amplitudes in laser a and I_N is the normalized diode intensity. Thus the term above can be interpreted as arising from amplitude fluctuations in the modes from laser a. As can be seen from Eq. (6), each of the first three terms is inversely proportional to the square root of the number of modes from one of the lasers contributing to the signal in that particular diode. Thus, for all three lasers, there is one noise contribution due to stochastic-mode-intensity (or -amplitude) variations. In non-Raman-resonant CARS the signal contribution from mode-intensity fluctuations is the same for all diodes in the diode array for a laser x with

$$\Gamma_x \ll (\Gamma_a^2 + \Gamma_b^2 + \Gamma_c^2 + W^2)^{1/2}$$

(e.g., a multimode pump laser in CC). For such a case, the normalization employed in Section 3 eliminates this noise contribution. In Eq. (6) this is manifested through the function F_Δ, which arises from the normalization [see Eq. (B8) below].

If the two components in one, and only one, of the three pairs of indices in H above are equal, the next three terms in Eq. (6) are obtained. If none of the pairs has two indices that are equal, then the last term is obtained [compare Eq. (2b)]. These last four terms correspond to mode beating between anti-Stokes modes. The first three of these terms correspond to beating between anti-Stokes modes where, for one, and only one, of the lasers, photons from the same mode have been used to create the anti-Stokes photons. In the last four terms in Eq. (6) none of the lasers contributes a photon from the same mode to the different pairs of anti-Stokes components beating against each other.

A possibly more physical picture of how the mode-beating terms contribute to the noise is to consider anti-Stokes electric-field modes with random phases and a mode separation $\Delta\omega$ impinging upon a diode after passing the detection system with an instrument function with a spectral width W. The intensity registered by that diode is proportional to the square of the electric field averaged over the anti-Stokes laser pulse duration T. In Fig. 2 an instrument function with a width W and two electric-field modes, separated by an interval $\Delta\omega$ in frequency, is shown. The intensity generated by these two modes contains a term oscillating with a frequency $\Delta\omega$. If $\Delta\omega T \gg \pi$, this term will average to zero if integrated over the laser pulse, but if $\Delta\omega < \pi/T$, there will generally be a net contribution to the intensity registered by the diode from the beat signal. By considering all possible mode combinations, we can infer that the number of phase-incoherent terms fulfilling the condition $\Delta\omega T < \pi$ is proportional to W/T. Because of the random phases, the average value of their contribution is zero with a standard deviation of $(W/T)^{1/2}$. The number of mode combinations contributing to the recorded intensity is proportional to W. Thus this noise contribution is proportional to $1/(WT)^{1/2}$, which corresponds to the last term in Eq. (6). The three terms before this last one can be motivated by similar arguments.
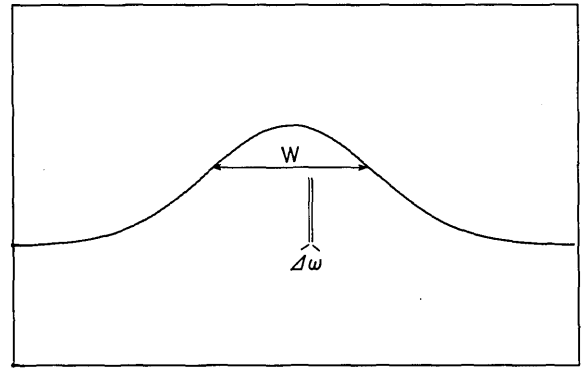

Fig. 2. A conceptual picture of an instrument function with the spectral width W and two anti-Stokes modes with a frequency separation $\Delta\omega$ transmitted through the detection window. The two modes will give rise to a beat signal at frequency $\Delta\omega$ in the detector. The horizontal scale is frequency, and the vertical scale is intensity (arbitrary units).

As a special case, consider now the situation when two photons (e.g., a and c) are taken from the same laser. The sinc^2 function in the term where $k \neq l$ and $r = s$ but $m \neq n$ should here again be replaced by $\delta(k - l - m + n)$ in the calculation. The influence of such a term is discussed in Ref. 8 for the case $\Gamma_a = \Gamma_c \ll \Gamma$. For such a case all diodes in the diode array would experience the same contribution from this term, and this noise contribution is, as pointed out in Ref. 8, eliminated by the type of normalization that has been employed in Section 3 before calculating the noise. If, however, the bandwidth of the laser generating this incoherent term is large, as in RDBC, this contribution will be different for different diodes and can no longer be eliminated by normalization. This term is the main contribution to the noise in RDBC. One way to understand this term is to consider the mode-beating case first and then to assume that the two modes have the same frequency. With two anti-Stokes modes ω'_{as} and ω''_{as}, with phases ϕ' and ϕ'', the registered intensity $\int E_{as}\overline{E}_{as}dt$ will contain terms of the type $\int_0^T \exp\{i[(\omega'_{as} - \omega''_{as})t + \phi' - \phi'']\}dt$. Assume that a mode combination has been found for which ω'_{as} exactly equals ω''_{as}. The recorded signal will then be proportional to $\cos(\phi' - \phi'')$. The size of this contribution is thus determined by the relative phases of the modes, and, although the mode beating is now absent, there will still be a phase-incoherent noise contribution from these modes. Furthermore, this contribution will not be decreased by an averaging over the laser pulse as a mode-beating term is. Thus this type of phase-incoherent contribution may be expected to be particularly strong. If two photons are taken from the same (multi-mode) laser, there will clearly be a large number of mode-combination pairs that have exactly the same frequency. An equivalent term arising in sum-frequency generation is briefly discussed in Ref. 31.

5. DISCUSSION

In deriving the expression for the noise in a nonlinear-optical process the electric fields have been modeled as a sum of modes with stochastic and independent amplitudes and phases. On the basis of this approach, it is possible to separate the noise contribution into two types, one due to the random amplitudes and one due to the random phases.

Table 3. Values Used When Calculating the Noise in Tables 4 and 5[a]

Parameter	CC	DBC	RDBC
Γ_a	0.5	100	100
Γ_b	100	100	$=\Gamma_a$
Γ_c	$=\Gamma_a$	0.5	0.5
Common for All Three Experimental Approaches			
$\Omega_a = \Omega_b = \Omega_c \approx 0.01$			
$W = 1.0$			
$T = 8.3$ nsec			
$\Gamma_r = 0.03$			

[a] Units are inverse centimeters if not otherwise stated. All Ω's, Γ's, and W must be multiplied by a factor of $2\pi c$, where c is the velocity of light ($=3 \times 10^{10}$ cm/sec). To convert to FWHM's the Γ's and W need to be multiplied by a factor of $2(\ln 2)^{1/2}$. All quantities are defined in Table 2.

Table 4. Calculated Noise for Nonresonant CARS

Parameter	Percentage Noise[a]					
	CC		DBC		RDBC	
	SM	MM	SM	MM	SM	MM
Mode-amplitude fluctuations	6	6	<1	<1	<1	<1
Mode beating	—	8	4	5	—	4
Phase-incoherence non mode beating	—	≈0	—	—	6	6
Total (added in quadrature)	6	10	4	5	6	7

[a] SM, single-mode pump laser; MM, multimode pump laser.

Table 5. Calculated Noise in a Spectrum Where a Single Raman Resonance Contributes to the Signal in Each Diode

Parameter	Percentage Noise[a]					
	CC		DBC		RDBC	
	SM	MM	SM	MM	SM	MM
Mode-amplitude fluctuations	23	11	<1	3	<1	4
Mode beating	—	20	15	16	—	3
Phase-incoherence non mode beating	—	≈0	—	—	23	23
Total (added in quadrature)	23	23	15	16	23	24

[a] SM, single-mode pump laser; MM, multimode pump laser.

The incoherent contribution due to the random phases may, as was described in Section 4, be separated into a mode-beating and a non-mode-beating term. For all three techniques, CC, DBC, and RDBC, the noise in non-Raman-resonant and Raman-resonant (for the particular situation described in Section 3) CARS is calculated for both a multimode and a single-mode pump laser by using the data given in Table 3. The results of these calculations are given in Tables 4 and 5.

The two types of noise have quite different characteristics. The random amplitudes cause variations in the coherent intensity contribution to the nonlinear signal. These variations can be reduced by letting as many modes as possible participate in the generation of the signal. This is effectively what is happening in DBC and RDBC, where the mode-amplitude fluctuation part is consequently negligible. An effect of this type was anticipated in the papers originally introducing these approaches to CARS.[15,17]

The noise from phase fluctuations is, in contrast to the amplitude noise, incoherent. Contrary to the case for the coherent noise, the incoherent noise is decreased when as few incoherent terms as possible contribute to the signal. Thus reducing the coherent noise seems (at least at first sight) incompatible with reducing the incoherent noise and vice versa. The incoherent-noise contribution arises as soon as more than one photon in the wave-mixing process comes from a multimode source. (If $T\Omega_x < \pi$ the incoherent-signal contribution may also be significant with only one photon from a multimode laser.) It may be clarifying to study a particular case in more detail. The main noise term in Raman-resonant CARS with a single-mode pump laser is

$$(\Omega_b/2\pi\Gamma_r)^{1/2} \qquad (7a)$$

for CC and RDBC and

$$1/(\Gamma_r T)^{1/2} \qquad (7b)$$

for DBC. In CC the noise comes from mode-amplitude variations. The dye-laser mode spacing may then be decreased in order to increase the number of modes participating in driving the Raman resonance and thus reducing the noise. This will be effective until $\Omega_x T$ is of the order of π. Two adjacent dye-laser modes are then so close in frequency that mode beating between these two modes will be too slow to be averaged to zero during the anti-Stokes pulse, and a phase-incoherent contribution will start to appear for the CC single-mode case. In a more mathematical approach the fact that $\Omega_x T$ is of the order of π means that the sinc^2 function in Eq. (3) can no longer be replaced by a delta function before the integrations are carried out, as was described in Section 4. We can actually show that in the limit of zero mode spacing, the noise in CC (single mode) will decrease to $1/(TT_r)^{1/2}$, the noise observed in DBC. As can be seen from Table 4, the noise in the non-resonant CC single-mode case is determined by mode-amplitude fluctuations in the dye laser only. This is the spectral noise in the dye laser. In the case of DBC the total noise is indeed below this value. Essentially, the noise in DBC is given by expression (7b) with Γ_r exchanged for W. This can also be inferred from Eq. (E4 below) and Table 3.

At this point, it is interesting to compare the calculated results with experimental data. In CC (single-mode) we essentially need to take only the amplitude noise into account. Snelling et al.[9,14] have compared the noise in experimental vibrational CARS spectra with theoretical calculations of mode-amplitude noise for this experimental configuration. A factor-of-1.4 higher noise than predicted by the theoretical calculations is observed in the experimental spectra for the non-Raman-resonant, as well as the Raman-resonant, case.[14] On the other hand, in a comparison with the experimental noise measured by Aldén et al.[17] for non-Raman-resonant RDBC (multimode), the theoretical model presented here overestimates the noise by a factor of 1.8. From expressions (7) it is seen that the analytical expressions for the main noise term are the same in CC and RDBC, and this is, as a matter of fact, also the case for non-Raman-resonant CARS. A more extensive investigation of the noise- and temperature-accuracy properties of rotational CARS, which may be compared with the present model, is in progress.[33] In practice, CC and RDBC may not give the same noise at all. The theoretical expressions in Eq. (2) and in Eqs. (D1) and (D2) below are exact under the conditions given. However, several approximations have been made in

order to arrive at them. Transverse modes have been neglected, amplitudes and phases have been assumed not to change during the laser pulse, and all mode amplitudes and phases have been assumed to be independent. Deviations from these conditions may affect, e.g., CC and RDBC differently. In particular, we may expect that the CC noise would be reduced for a laser experiencing amplitude locking and that the RDBC noise would be reduced for a laser experiencing phase locking. Spontaneous processes of these types have been observed in some multimode dye lasers.[28,29,34]

It is thus questionable whether the model can be used for quantitative predictions of noise. It may, however, be helpful for qualitative interpretations and, in particular, it shows that if we assume multimode sources with independent modes the noise should be larger in non-Raman-resonant CARS when the CC multimode approach is used than with the CC single-mode approach, as has been shown experimentally.[4] The mode-beating term contributing in this case will also contribute to Raman-resonant spectra and may thus be one of the sources of the noise observed by Snelling et al.[14] in addition to the mode-amplitude noise in their multimode spectra. They have noted that (for their system) the Stokes and pump laser intensities appear to be correlated in time (on a nanosecond time scale),[4,14] and they point out that this may be the reason for the increased noise when a multimode pump laser is used. As can be seen from Table 5, the model fails to predict the reduction of noise that is obtained in Raman-resonant spectra when a multimode pump laser is used. This would be consistent with the fact that the model underestimates the noise due to mode-amplitude fluctuations and overestimates the noise due to phase fluctuations. It is unclear exactly how the statistics of laser radiation should deviate from what has been assumed in order to reproduce this behavior.

From the analysis presented here and the comparisons with experimental measurements, it seems that to design the optimum experimental approach it would be helpful to know more about the characteristics of the statistics of the laser radiation. One may also attempt to manipulate the laser radiation such that it suits the experimental approach chosen. An approach to achieve phase locking has recently been demonstrated by Li et al.[35]

A completely different approach to reduce the noise in CARS measurements would be to use accurate referencing. This has long been considered a difficult task. As discussed in Ref. 10, it is important to ascertain that the same modes contribute to the reference spectra and the signal for successful referencing. This seems to be a necessary condition. At present, it is, however, unclear to what extent it is a sufficient condition.

6. SUMMARY

An approach for calculating the noise in a general four-wave-mixing process, in which the generated field is weak and a perturbation expansion in the incoming fields is sufficient for calculating the induced polarization, has been presented. Mode amplitudes and phases of the electromagnetic fields have been assumed to be uncorrelated, and their time dependence during the laser pulse has been neglected. The model is strictly correct only for uncorrelated and uniformly distributed laser-mode phases. This model has been used to

evaluate explicitly the noise in different experimental approaches to broadband CARS, yielding the following information on noise in broadband CARS as it is used in thermometry.

Generally two principal sources of noise can be identified. From each multimode source there is a term, inversely proportional to the square root of the number of modes, that contributes to the CARS signal. Normalizing the intensity registered by a single diode to the total intensity registered by the diode array, as is generally done in CARS thermometry, will tend to eliminate this noise contribution for a laser with a bandwidth that is much smaller than the convoluted bandwidth of all laser sources and the instrument function. The reason for this is that this contribution will be the same for all diodes and that the different types of noise contribution are statistically independent. The other main source of noise arises from the phase incoherence in the multimode lasers, which adds noise primarily through mode beating between the generated anti-Stokes modes. If the different mode combinations are frequency degenerate, noise will still be added because the phases for different mode combinations will be different. The analysis shows that it is essentially only the stochastic laser-mode phases (and not the stochastic-mode amplitudes) that cause the noise in the dual-broadband techniques, and, conversely, it is only the stochastic-mode amplitudes (i.e., the spectral noise in the dye laser) that cause the noise in the conventional CARS single-mode pump laser case. Consequently, the dye-laser spectral noise is not strictly a limiting factor in the temperature accuracy in the dual-broadband approaches.

The model predicts that the noise in nonresonant broadband CARS is higher with a multimode YAG laser than with a single-mode YAG laser, as a multimode YAG laser will contribute to the noise through phase incoherence. This result is supported by existing experimental data.[4,9]

In general, however, the model seems to underestimate the noise due to amplitude fluctuations and to overestimate the noise due to phase incoherence. As the exact mathematical and statistical description of the laser radiation is unknown, the discrepancy may, in principle, arise from an incorrect representation of the laser fields. A conclusion that may be drawn from the calculations is that it may be possible to reduce the noise and thus to improve the accuracy in single-shot broadband CARS by using amplitude- and/or phase-locked lasers.

APPENDIX A

Consider two complex-valued random variables

$$I_j = \sum_{klrsmn} a_k a_l b_r b_s c_m \dot{c}_n \exp\{i[(\theta_k - \theta_l) - (\phi_r - \phi_s)$$
$$+ (\psi_m - \psi_n)]\} H_j(k, l, r, s, m, n), \qquad j = 1, 2,$$

where the summation is performed over all integers, including infinity. The functions H_j are deterministic but otherwise arbitrary, $j = 1, 2$. $\{x_k^2\}$ is a sequence of independent, exponentially distributed random variables with mean $i_x(x = a, b, c)$, and $\{y_k\}$ is a sequence of independent random variables that are uniformly distributed over the interval

$(0, 2\pi), y = \theta, \phi, \psi$. Furthermore, these six sequences of real random variables are independent of one another.

We will calculate here the expected value $E(I_1)$ and the covariance $C(I_1, I_2)$ between the random variables I_1 and I_2. At the end of this appendix we will choose $H_1 = H_2 = H$, and hence $I_1 = I_2 = I$, and we will determine $E(I)$ and $V(I) = C(I, I)$ as a special case.

For a complex-valued random variable $Z = Z_1 + iZ_2$, where Z_1 and Z_2 are random real variables, the mean is $E(Z) = E(Z_1) + iE(Z_2)$. The covariance between two complex-valued random variables Z and W is

$$C(Z, W) = C\{[Z - E(Z)][\overline{W - E(W)}]\}$$
$$= E(Z\overline{W}) - E(Z)\overline{E(W)}.$$

Suppose that X^2 is exponentially distributed with a mean m, i.e., that $X^2 \in \Gamma(1, m)$. Then, X^2 has the density $f(z) = (1/m)\exp(-z/m)$ for $z > 0$, and hence

$$E(X^2) = m, \tag{A1}$$

$$E(X^4) = 2m^2. \tag{A2}$$

If Y is uniformly distributed over the interval $(0, 2\pi)$ we have, for an integer k,

$$E(e^{ikY}) = 1 \qquad \text{if } k = 0, \text{ and } 0 \text{ otherwise.} \tag{A3}$$

Mean
Because the six sequences are independent, we have

$$E(I_1) = \sum_{\substack{klr \\ smn}} E(a_k a_l) E(b_r b_s) E(c_m c_n) E\{\exp[i(\theta_k - \theta_l)]\}$$

$$\times E\{\exp[i(\phi_s - \phi_r)]\} E\{\exp[i(\psi_m - \psi_n)]\}$$

$$\times H_1(k, l, r, s, m, n).$$

Consider the expected value $E\{\exp[i(\theta_k - \theta_l)]\}$. If $k \neq l$, θ_k and θ_l are independent, and if we use (A3), we have

$$E\{\exp[i(\theta_k - \theta_l)]\} = E[\exp(i\theta_k)]E[e(-i\theta_l)] = 0.$$

But if $k = l$, we have $E\{\exp[i(\theta_k - \theta_l)]\} = 1$.

The same argument for ϕ and ψ gives

$$E(I_1) = \sum_{krm} E(a_k^2) E(b_r^2) E(c_m^2) H_1(k, k, r, r, m, m).$$

Finally, from Eq. (A1), we have

$$E(I_1) = (i_a i_b i_c) \sum_{krm} H_1(k, k, r, r, m, m). \tag{A4}$$

Covariance
First we write

$$I_j = \sum_{n=1}^{8} S_j(n), \qquad j = 1, 2,$$

where

$$S_j(1) = \sum_{\substack{klrsmn \\ k \neq l, r \neq s, m \neq n}} a_k a_l b_r b_s c_m c_n \exp\{i[(\theta_k - \theta_l) + (\phi_s - \phi_r)$$
$$+ (\psi_m - \psi_n)]\} H_j(k, l, r, s, m, n),$$

$$S_j(2) = \sum_{\substack{klrsm \\ k \neq l, r \neq s}} a_k a_l b_r b_s c_m^2 \exp\{i[(\theta_k - \theta_l) + (\phi_s - \phi_r)]\}$$
$$\times H_j(k, l, r, s, m, m),$$

$$S_j(3) = \sum_{\substack{klrmn \\ k \neq l, m \neq n}} a_k a_l b_r^2 c_m c_n \exp\{i[(\theta_k - \theta_l) + (\psi_m - \psi_n)]\}$$
$$\times H_j(k, l, r, r, m, n),$$

$$S_j(4) = \sum_{\substack{krsmn \\ r \neq s, m \neq n}} a_k^2 b_r b_s c_m c_n \exp\{i[(\phi_s - \phi_r) + (\psi_m - \psi_n)]\}$$
$$\times H_j(k, k, r, s, m, n),$$

$$S_j(5) = \sum_{\substack{klrm \\ k \neq l}} a_k a_l b_r^2 c_m^2 \exp[i(\theta_k - \theta_l)] H_j(k, l, r, r, m, m),$$

$$S_j(6) = \sum_{\substack{krsm \\ r \neq s}} a_k^2 b_r b_s c_m^2 \exp[i(\phi_s - \phi_r)] H_j(k, k, r, s, m, m),$$

$$S_j(7) = \sum_{\substack{krmn \\ m \neq n}} a_k^2 b_r^2 c_m c_n \exp[i(\psi_m - \psi_n)] H_j(k, k, r, r, m, n),$$

$$S_j(8) = \sum_{krm} a_k^2 b_r^2 c_m^2 H_j(k, k, r, r, m, m), \qquad j = 1, 2.$$

This partition of I_j is convenient because we can show that $C[S_1(u), S_2(v)] = 0$ when $u \neq v$, and hence

$$C(I_1, I_2) = \sum_{u=1}^{8} C[S_1(u), S_2(u)]. \tag{A5}$$

Therefore it only remains to calculate the covariances

$$C[S_1(u), S_2(u)], u = 1, \ldots 8:$$

$$C[S_1(1), S_2(1)]$$

$$= \sum_{\substack{klrsmnopqtuv \\ k \neq l, r \neq s, m \neq n \\ o \neq p, q \neq t, u \neq v}} H_1(k, l, r, s, m, n) \overline{H_2(o, p, q, t, u, v)} C_1,$$

where

$$C_1 = C(a_k a_l b_r b_s c_m c_n \exp\{i[(\theta_k - \theta_l) + (\phi_s - \phi_r) + (\psi_m - \psi_n)]\},$$
$$a_o a_p b_q b_t c_u c_v \exp\{i[(\theta_o - \theta_p) + (\phi_t - \phi_q) + (\psi_u - \psi_v)]\}).$$

Because

$$E(a_k a_l b_r b_s c_m c_n \exp\{i[(\theta_k - \theta_l) + (\phi_s - \phi_r) + (\psi_m - \psi_n)]\}) = 0$$

and because the sequences are independent, we have

$$C_1 = E(a_k a_l a_o a_p) E(b_r b_s b_q b_t) E(c_m c_n c_u c_v)$$

$$\times E\{\exp[i(\theta_k - \theta_l + \theta_p - \theta_o)]\} E\{\exp[i(\phi_s - \phi_r + \phi_q - \phi_t)]\}$$

$$\times E\{\exp[i(\psi_m - \psi_n + \psi_u - \psi_v)]\}.$$

Now consider the mean $E\{\exp[i(\theta_k - \theta_l + \theta_p - \theta_o)]\}$. Clearly, if one index differs from the other three, say k, then θ_k is independent of θ_l, θ_p, and θ_o, and hence the mean is zero. Furthermore, it is obvious that this mean differs from zero only when $\theta_k - \theta_l + \theta_p - \theta_o = 0$, which, because $k \neq l$ and $p \neq o$, happens only when $k = o$ and $l = p$. A similar argument holds for ϕ and ψ, so when $k = o, l = p, r = q, s = t$, $m = u$, and $n = v$, we have $C_1 = E(a_k^2 a_l^2) E(b_r^2 b_s^2) E(c_m^2 c_n^2)$, and $C_1 = 0$ otherwise.

Because $k \neq l, r \neq s$, and $m \neq n$, by independence and by using Eq. (A1), we then have

$$C_1 = (i_a i_b i_c)^2.$$

Hence we have

$$C[S_1(1), S_2(1)]$$

$$= (i_a i_b i_c)^2 \sum_{\substack{klrsmn \\ k \neq l, r \neq s, m \neq n}} H_1(k, l, r, s, m, n) \overline{H_2(k, l, r, s, m, n)},$$

$$C[S_1(2), S_2(2)]$$

$$= \sum_{\substack{klrsmopqtn \\ k \neq l, r \neq s, o \neq p, q \neq t}} H_1(k, l, r, s, m, m) \overline{H_2(o, p, q, t, n, n)} C_2,$$

$$(A6)$$

where

$$C_2 = C(a_k a_l b_r b_s c_m^2 \exp\{i[(\theta_k - \theta_l) + (\phi_s - \phi_r)]\},$$

$$a_o a_p b_q b_t c_n^2 \exp\{i[(\theta_o - \theta_p) + (\phi_t - \phi_q)]\}).$$

By using arguments similar to those for the calculation of C_1, we have

$$C_2 = E(a_k a_l a_o a_p) E(b_r b_s b_q b_t) E(c_m^2 c_n^2)$$

$$\times E\{\exp[i(\theta_k - \theta_l + \theta_p - \theta_o)]\}$$

$$\times E\{\exp[i(\phi_s - \phi_r + \phi_q - \phi_t)]\}$$

$$= E(a_k^2 a_l^2) E(b_r^2 b_s^2) E(c_m^2 c_n^2)$$

$$= (i_a i_b)^2 E(c_m^2 c_n^2),$$

when $k = o, l = p, s = t, r = q$, and $C_2 = 0$ otherwise.

We have to separate the cases where $m = n$ or $m \neq n$. If $m = n$, from Eq. (A2),

$$C_2 = (i_a i_b)^2 E(c_m^4) = 2(i_a i_b i_c)^2.$$

If $m \neq n$, c_m^2 and c_n^2 are independent, and hence

$$C_2 = (i_a i_b)^2 E(c_m^2) E(c_n^2) = (i_a i_b i_c)^2.$$

We therefore have

$$C[S_1(2), S_2(2)] = (i_a i_b i_c)^2 \left[\sum_{\substack{klrsmn \\ k \neq l, r \neq s, m \neq n}} H_1(k, l, r, s, m, m) \right.$$

$$\times \overline{H_2(k, l, r, s, n, n)}$$

$$+ 2 \sum_{\substack{klrsm \\ k \neq l, r \neq s}} H_1(k, l, r, s, m, m)$$

$$\left. \times \overline{H_2(k, l, r, s, m, m)} \right].$$

$$(A7)$$

By symmetry we must have

$$C[S_1(3), S_2(3)]$$

$$= (i_a i_b i_c)^2 \left[\sum_{\substack{klrsmn \\ k \neq l, r \neq s, m \neq n}} H_1(k, l, r, r, m, n) \overline{H_2(k, l, s, s, m, n)} \right.$$

$$\left. + 2 \sum_{\substack{klrmn \\ k \neq l, m \neq n}} H_1(k, l, r, r, m, n) \overline{H_2(k, l, r, r, m, n)} \right]. \quad (A8)$$

$$C[S_1(4), S_2(4)]$$

$$= (i_a i_b i_c)^2 \left[\sum_{\substack{klrsmn \\ k \neq l, r \neq s, m \neq n}} H_1(k, k, r, s, m, n) \overline{H_2(l, l, r, s, m, n)} \right.$$

$$\left. + 2 \sum_{\substack{krsmn \\ r \neq s, m \neq n}} H_1(k, k, r, s, m, n) \overline{H_2(k, k, r, s, m, n)} \right].$$

$$(A9)$$

$$C[S_1(5), S_2(5)]$$

$$= \sum_{\substack{klrmopsn \\ k \neq l, o \neq p}} H_1(k, l, r, r, m, m) \overline{H_2(o, p, s, s, n, n)} C_3,$$

where

$$C_3 = C\{a_k a_l b_r^2 c_m^2 \exp[i(\theta_k - \theta_l)], a_o a_p b_s^2 c_n^2 \exp[i(\theta_o - \theta_p)]\}.$$

By the same argument as before we have

$$C_3 = i_a^2 E(b_r^2 b_s^2) E(c_m^2 c_n^2)$$

$$\text{if } k = o \text{ and } l = p, \text{ and } 0 \text{ otherwise.}$$

We now have to distinguish the four cases

$$r = s, m = n \rightarrow C_3 = 4(i_a i_b i_c)^2,$$

$$r = s, m \neq n \rightarrow C_3 = 2(i_a i_b i_c)^2,$$

$$r \neq s, m = n \rightarrow C_3 = 2(i_a i_b i_c)^2,$$

$$r \neq s, m \neq n \rightarrow C_3 = (i_a i_b i_c)^2$$

because of independence and Eqs. (A1) and (A2). Hence we have

$C[S_1(5), S_2(5)]$

$$= (i_a i_b i_c)^2 \left[\sum_{\substack{klrsmn \\ k \neq l, r \neq s, m \neq n}} H_1(k, l, r, r, m, m)\overline{H_2(k, l, s, s, n, n)} \right.$$

$$+ 2 \sum_{\substack{klrsm \\ k \neq l, r \neq s}} H_1(k, l, r, r, m, m)\overline{H_2(k, l, s, s, m, m)}$$

$$+ 2 \sum_{\substack{klrmn \\ k \neq l, m \neq n}} H_1(k, l, r, r, m, m)\overline{H_2(k, l, r, r, n, n)}$$

$$+ 4 \sum_{\substack{klrm \\ k \neq l}} H_1(k, l, r, r, m, m)\overline{H_2(k, l, r, r, m, m)} \left. \right] . \quad (A10)$$

As in the previous case, by symmetry we have

$C[S_1(6), S_2(6)]$

$$= (i_a i_b i_c)^2 \left[\sum_{\substack{klrsmn \\ k \neq l, r \neq s, m \neq n}} H_1(k, k, r, s, m, m)\overline{H_2(l, l, r, s, n, n)} \right.$$

$$+ 2 \sum_{\substack{klrsm \\ k \neq l, r \neq s}} H_1(k, k, r, s, m, m)\overline{H_2(l, l, r, s, m, m)}$$

$$+ 2 \sum_{\substack{krsmn \\ r \neq s, m \neq n}} H_1(k, k, r, s, m, m)\overline{H_2(k, k, r, s, n, n)}$$

$$+ 4 \sum_{\substack{krsm \\ r \neq s}} H_1(k, k, r, s, m, m)\overline{H_2(k, k, r, s, m, m)} \left. \right] , \quad (A11)$$

$C[S_1(7), S_2(7)]$

$$= (i_a i_b i_c)^2 \left[\sum_{\substack{klrsmn \\ k \neq l, r \neq s, m \neq n}} H_1(k, k, r, r, m, n)\overline{H_2(l, l, s, s, m, n)} \right.$$

$$+ 2 \sum_{\substack{klrmn \\ k \neq l, m \neq n}} H_1(k, k, r, r, m, n)\overline{H_2(l, l, r, r, m, n)}$$

$$+ 2 \sum_{\substack{krsmn \\ r \neq s, m \neq n}} H_1(k, k, r, r, m, n)\overline{H_2(k, k, s, s, m, n)}$$

$$+ 4 \sum_{\substack{krmn \\ m \neq n}} H_1(k, k, r, r, m, n)\overline{H_2(k, k, r, r, m, n)} \left. \right] . \quad (A12)$$

Finally, we calculate

$$C[S_1(8), S_2(8)] = \sum_{klrsmn} H_1(k, k, r, r, m, m)\overline{H_2(l, l, s, s, n, n)} C_4,$$

where

$$C_4 = C(a_k{}^2 b_r{}^2 c_m{}^2, a_l{}^2 b_s{}^2 c_n{}^2)$$

$$= E(a_k{}^2 a_l{}^2) E(b_r{}^2 b_s{}^2) E(c_m{}^2 c_n{}^2) - (i_a i_b i_c)^2.$$

As for the previous cases, we separate the covariance into the (eight) appropriate cases, which finally yields

$C[S_1(8), S_2(8)]$

$$= (i_a i_b i_c)^2 \left[7 \sum_{krm} H_1(k, k, r, r, m, m)\overline{H_2(k, k, r, r, m, m)} \right.$$

$$+ 3 \sum_{\substack{krmn \\ m \neq n}} H_1(k, k, r, r, m, m)\overline{H_2(k, k, r, r, n, n)}$$

$$+ 3 \sum_{\substack{krsm \\ r \neq s}} H_1(k, k, r, r, m, m)\overline{H_2(k, k, s, s, m, m)}$$

$$+ 3 \sum_{\substack{klrm \\ k \neq l}} H_1(k, k, r, r, m, m)\overline{H_2(l, l, r, r, m, m)}$$

$$+ \sum_{\substack{krsmn \\ r \neq s, m \neq n}} H_1(k, k, r, r, m, m)\overline{H_2(k, k, s, s, n, n)}$$

$$+ \sum_{\substack{klrmn \\ k \neq l, m \neq n}} H_1(k, k, r, r, m, m)\overline{H_2(l, l, r, r, n, n)}$$

$$+ \sum_{\substack{klrsm \\ k \neq l, r \neq s}} H_1(k, k, r, r, m, m)\overline{H_2(l, l, s, s, m, m)} \left. \right] . \quad (A13)$$

Thus we have computed $C[S_1(u), S_2(u)]$, $u = 1 \ldots 8$, and therefore, by using Eq. (A5), we have found $C(I_1, I_2)$ to be the sum of $1 + 3(2) + 3(4) + 7 = 26$ different sums. Fortunately, when we simplify the expression, it turns out that most terms vanish. Because these tedious calculations are of little interest, we will just give the final result without any details:

$$C(I_1, I_2) = (i_a i_b i_c)^2 \left[\sum H_1(k, l, r, s, m, n)\overline{H_2(k, l, r, s, m, n)} \right.$$

$$+ \sum H_1(k, l, r, s, m, m)\overline{H_2(k, l, r, s, n, n)}$$

$$+ \sum H_1(k, l, r, r, m, n)\overline{H_2(k, l, s, s, m, n)}$$

$$+ \sum H_1(k, k, r, s, m, n)\overline{H_2(l, l, r, s, m, n)}$$

$$+ \sum H_1(k, l, r, r, m, m)\overline{H_2(k, l, s, s, n, n)}$$

$$+ \sum H_1(k, k, r, s, m, m)\overline{H_2(l, l, r, s, n, n)}$$

$$+ \sum H_1(k, k, r, r, m, n)\overline{H_2(l, l, s, s, m, n)} \left. \right] ,$$

$$(A14)$$

where each summation is performed over all occurring indices. If we now choose $H_1 = H_2 = H$ and if we use Eqs. (A4) and (A14), we have [as in Eq. (2)]

$$E(I) = (i_a i_b i_c) \sum_{krm} H(k, k, r, r, m, m),$$

$$\begin{aligned}
V(I) = (i_a i_b i_c)^2 \sum_{klrsmn} & [|H(k, l, r, s, m, n)|^2 \\
& + H(k, l, r, s, m, m)\overline{H(k, l, r, s, n, n)} \\
& + H(k, l, r, r, m, n)\overline{H(k, l, s, s, m, n)} \\
& + H(k, k, r, s, m, n)\overline{H(l, l, r, s, m, n)} \\
& + H(k, l, r, r, m, n)\overline{H(k, l, s, s, n, n)} \\
& + H(k, k, r, s, m, m)\overline{H(l, l, r, s, n, n)} \\
& + H(k, k, r, r, m, n)\overline{H(l, l, s, s, m, n)}].
\end{aligned}$$

APPENDIX B

In this appendix we will evaluate $E(I)$ and $V(I)$, which are given at the end of Appendix A. We will thus be able to obtain a corresponding expression for the noise $[V(I)]^{1/2}/E(I)$. At the end of this appendix we give an expression for the noise in the normalized signal $I_N(\Delta_\alpha)$.

To simplify forthcoming calculations, we begin with a slightly more general expression. Let

$$\begin{aligned}
I(\Delta) = \sum_{klrsmn} & a_k a_l b_r b_s c_m c_n \exp\{i[(\theta_k - \theta_l) - (\phi_r - \phi_s) \\
& + (\psi_m - \psi_n)]\}H_\Delta(k, l, r, s, m, n),
\end{aligned} \quad (B1)$$

where

$$\begin{aligned}
H_\Delta(k, l, r, s, m, n) = \exp\Bigg\{ & -\frac{1}{2}\Bigg[\frac{\Omega_a{}^2}{\Gamma_a{}^2}(k^2 + l^2) \\
& + \frac{\Omega_b{}^2}{\Gamma_b{}^2}(r^2 + s^2) + \frac{\Omega_c{}^2}{\Gamma_c{}^2}(m^2 + n^2)\Bigg]\Bigg\} \\
\times \exp\Bigg\{ & -\frac{1}{2W^2}[(\Omega_a k - \Omega_b r + \Omega_c m - \Delta)^2 \\
& + (\Omega_a l - \Omega_b s + \Omega_c n - \Delta)^2]\Bigg\} \\
\times \operatorname{sinc}\Bigg\{ & \frac{T}{2}[\Omega_a(k - l) - \Omega_b(r - s) + \Omega_c(m - n)]\Bigg\}.
\end{aligned}$$

After we have evaluated the mean $E[I(\Delta)]$ and the covariance $C[I(\Delta_\alpha), I(\Delta_\beta)]$, $E(I)$ and $V(I)$ above will easily follow as a special case, letting $\Delta = \Delta_\alpha = \Delta_\beta = 0$.

To be able to replace summations with integrations, we assume throughout this appendix that

$$\Omega_x \ll \min(W, \Gamma_x), \qquad x = a, b, c.$$

The mean $E[I(\Delta)]$ is given by Eq. (A4). Hence we should evaluate the following integral:

$$\begin{aligned}
\sum_{krm} H_\Delta(k, k, r, r, m, m) \approx \int_{R^3} & \Bigg\{\exp\Bigg[-\Bigg(\frac{\Omega_a{}^2}{\Gamma_a{}^2}k^2 + \frac{\Omega_b{}^2}{\Gamma_b{}^2}r^2 \\
& + \frac{\Omega_c{}^2}{\Gamma_c{}^2}m^2\Bigg)\Bigg]\exp\Bigg[-\frac{1}{W^2}(k\Omega_a - r\Omega_b + m\Omega_c - \Delta)^2\Bigg]\Bigg\} dm\,dr\,dk.
\end{aligned}$$

After suitable variable substitutions and repeated use of the identity

$$\int_R \exp(-px^2 \pm qx)dx = \exp\{q^2/(4p)\}(\pi/p)^{1/2},$$

we obtain

$$E[I(\Delta)] \approx (i_a i_b i_c)\pi(\pi)^{1/2}\left(\frac{\Gamma_a \Gamma_b \Gamma_c}{\Omega_a \Omega_b \Omega_c}\right)\frac{W}{\Gamma}\exp(-\Delta^2/\Gamma^2), \quad (B2)$$

where $\Gamma^2 = W^2 + \Gamma_a{}^2 + \Gamma_a{}^2 + \Gamma_c{}^2$.

Define a constant C_Δ by

$$C_\Delta = \pi(\pi)^{1/2}\left(\frac{\Gamma_a \Gamma_b \Gamma_c}{\Omega_a \Omega_b \Omega_c}\right)\frac{W}{\Gamma}\exp(-\Delta^2/\Gamma^2). \quad (B3)$$

We will now evaluate the covariance $C[I(\Delta_\alpha), I(\Delta_\beta)]$ by using Eq. (A14) with $H_1 = H_{\Delta_\alpha}$ and $H_2 = H_{\Delta_\beta}$. We first note that there are three unique sums; the remaining four are given by obvious symmetry. Although the evaluation of these three unique sums is somewhat more difficult than for the mean, we shall only mention some difficulties and not give the explicit calculations.

First, we observe that for the last three sums in Eq. (A14) we have to replace the sinc function in $H_{\Delta_\alpha}H_{\Delta_\beta}$ with a delta function in order to replace the summation with integration. The reason for this substitution is discussed in Section 4. Hence the integration in these three cases only goes over five variables.

Second, the remaining four integrals cannot be solved exactly because the sinc function does not vanish, as in the case of the mean, nor can they be replaced with a delta function, as above. Hence the result in these cases will depend on an integral $f(a)$, defined for $a > 0$ by

$$f(a) = \int_R \exp(-ax^2)\operatorname{sinc}^2(x)dx. \quad (B4)$$

Using this function, we obtain the following expression for the covariance:

$$C[I(\Delta_\alpha), I(\Delta_\beta)] \approx (i_a i_b i_c)^2 C_{\Delta_\alpha} C_{\Delta_\beta} \frac{1}{(2\pi)^{1/2}}$$

$$\times \left\{ \frac{2}{TW}\left(1 - \frac{W^2}{\Gamma^2}\right)^{-1/2} f\left[\frac{2\Gamma^2}{T^2 W^2 (\Gamma^2 - W^2)}\right] \right.$$

$$\times \exp\left[-\frac{1}{2}(\Delta_\alpha - \Delta_\beta)^2 \frac{\Gamma^2 - W^2}{\Gamma^2 W^2}\right]$$

$$+ \sum_{x=a,b,c}\left(\frac{\Omega_x}{\Gamma_x}\left(1 - \frac{\Gamma_x^2}{\Gamma^2}\right)^{-1/2}\right.$$

$$\times \exp\left[-\frac{1}{2}(\Delta_\alpha - \Delta_\beta)^2 \frac{\Gamma_x^2}{\Gamma^2(\Gamma^2 - \Gamma_x^2)}\right]$$

$$+ \frac{2}{T}\left\{\left[(W^2 + \Gamma_x^2)\left(1 - \frac{W^2 + \Gamma_x^2}{\Gamma^2}\right)\right]^{-1/2}\right.$$

$$\times f\left[\frac{2(\Gamma^2 - \Gamma_x^2)}{T^2 W^2 (\Gamma^2 - W^2 - \Gamma_x^2)}\right]$$

$$\left.\left.\left.\times \exp\left[-\frac{1}{2}(\Delta_\alpha - \Delta_\beta)^2 \frac{\Gamma^2 - W^2 - \Gamma_x^2}{\Gamma^2(W^2 + \Gamma_x^2)}\right]\right\}\right)\right\}, \quad (B5)$$

where the constant C_Δ is defined in Eq. (B3).

If we let $\Delta = 0$ in Eq. (B2) and $\Delta_\alpha = \Delta_\beta = 0$ in Eq. (B5) we find $E(I)$ and $V(I)$. Finally, we give an expression for the noise in the signal:

$$\frac{\sigma(I)}{\langle I \rangle} = \left[\frac{V(I)}{E(I)^2}\right]^{1/2}$$

$$\approx (2\pi)^{-1/4}\left[\sum_{x=a,b,c}\left[\frac{\Omega_x}{\Gamma_x}\left(1 - \frac{\Gamma_x^2}{\Gamma^2}\right)^{-1/2}\right]\right.$$

$$+ \frac{2}{T}\sum_{x=a,b,c}\left(f\left\{\frac{2}{W^2 T^2}\left[1 + \frac{W^2}{\Gamma^2 - (\Gamma_x^2 + W^2)}\right]\right\}\right.$$

$$\left.\times \left\{(W^2 + \Gamma_x^2)\left[1 - \frac{(W^2 + \Gamma_x^2)}{\Gamma^2}\right]\right\}^{-1/2}\right)$$

$$\left.+ \frac{2}{TW}\left(1 - \frac{W^2}{\Gamma^2}\right)^{-1/2} f\left[\frac{2\Gamma^2}{T^2 W^2 (\Gamma^2 - W^2)}\right]\right]^{1/2}. \quad (B6)$$

We will now evaluate the noise in the normalized signal

$$I_N(\Delta_\alpha) = I(\Delta_\alpha)\bigg/\sum_\beta I(\Delta_\beta),$$

where $I(\Delta)$ is given by Eq. (B1).

Using the first-order Gaussian approximation[32] for the quotient X/Y of two complex-valued random variables, we have

$$E\left(\frac{X}{Y}\right) \approx \frac{E(X)}{E(Y)},$$

$$V\left(\frac{X}{Y}\right) \approx \frac{V(X)}{E(Y)^2} + V(Y)\frac{E(X)^2}{E(Y)^4} - 2\,\mathrm{Re}[C(X,Y)]\frac{E(X)}{E(Y)^3},$$

and hence

$$\frac{V(X/Y)}{E(X/Y)^2} \approx \frac{V(X)}{E(X)^2} + \frac{V(Y)}{E(Y)^2} - \frac{2\,\mathrm{Re}[C(X,Y)]}{E(X)E(Y)}. \quad (B7)$$

Let $X = I(\Delta_\alpha)$ and $Y = \sum_\beta I(\Delta_\beta)$. Now

$$E(X) = E[I(\Delta_\alpha)],$$

$$E(Y) = E\left[\sum_\beta I(\Delta_\beta)\right] = \sum_\beta E[I(\Delta_\beta)],$$

$$V(X) = V[I(\Delta_\alpha)] = C[I(\Delta_\alpha), I(\Delta_\alpha)],$$

$$C(X,Y) = C\left[I(\Delta_\alpha), \sum_\beta I(\Delta_\beta)\right] = \sum_\beta C[I(\Delta_\alpha), I(\Delta_\beta)],$$

$$V(Y) = V\left[\sum_\beta I(\Delta_\beta)\right] = C\left[\sum_\beta I(\Delta_\beta), \sum_\alpha I(\Delta_\alpha)\right]$$

$$= \sum_{\alpha\beta} C[I(\Delta_\alpha), I(\Delta_\beta)].$$

For suitable choice of Δ, Δ_α, and Δ_β in expression (B2) and Eq. (B5), these moments can be evaluated. Hence, using Eq. (B7), we are able to get an expression for the noise in the normalized signal. If we introduce a function $F_{\Delta_\alpha}(x)$ by

$$F_{\Delta_\alpha}(x) = 1 - 2\frac{\displaystyle\sum_\beta \exp\left[-\frac{\Delta_\beta^2}{\Gamma^2} - \frac{1}{\Gamma^2}(\Delta_\beta - \Delta_\alpha)^2 \frac{x}{2(1-x)}\right]}{\displaystyle\sum_\beta \exp(-\Delta_\beta^2/\Gamma^2)}$$

$$+ \frac{\displaystyle\sum_{\beta\gamma} \exp\left[-\frac{1}{\Gamma^2}(\Delta_\beta^2 + \Delta_\gamma^2) - \frac{1}{\Gamma^2}(\Delta_\beta - \Delta_\gamma)^2 \frac{x}{2(1-x)}\right]}{\left[\displaystyle\sum_\beta \exp(-\Delta_\beta^2/\Gamma^2)\right]^2}, \quad (B8)$$

we have, after some rearranging,

$$\frac{V[I_N(\Delta_\alpha)]}{E[I_N(\Delta_\alpha)]^2} \approx \frac{1}{(2\pi)^{1/2}}\left(\frac{2}{TW}\left(1 - \frac{W^2}{\Gamma^2}\right)^{-1/2} f\left[\frac{2\Gamma^2}{T^2 W^2 (\Gamma^2 - W^2)}\right]\right.$$

$$\times F_{\Delta_\alpha}\left(\frac{W^2}{\Gamma^2}\right) + \frac{2}{T}\sum_{a,b,c}\left\{\left[(W^2 + \Gamma_x^2)\left(1 - \frac{W^2 + \Gamma_x^2}{\Gamma^2}\right)\right]^{-1/2}\right.$$

$$\left.\times f\left[\frac{2(\Gamma^2 - \Gamma_x^2)}{T^2 W^2 (\Gamma^2 - W^2 - \Gamma_x^2)}\right]F_{\Delta_\alpha}\left(1 - \frac{W^2 + \Gamma_x^2}{\Gamma^2}\right)\right\}$$

$$\left.+ \sum_{x=a,b,c}\left[\frac{\Omega_x}{\Gamma_x}\left(1 - \frac{\Gamma_x^2}{\Gamma^2}\right)^{-1/2}F_{\Delta_\alpha}\left(\frac{\Gamma_x^2}{\Gamma^2}\right)\right]\right). \quad (B9)$$

APPENDIX C

In this appendix we will express the function $f(a)$, defined by Eq. (B4), as a composition of simpler functions and

$$\Phi(x) = \frac{1}{(2\pi)^{1/2}}\int_{-\infty}^x \exp(-y^2/2)dy.$$

$\Phi(x)$ is the distribution function for a standard normally distributed random variable.

We will also find approximations for $f(a)$ when $a > 0$ is small and will estimate $F_\Delta(x)$, defined in Eq. (B8), when x is close to zero or unity. We have

$$f(a) - f(0) = \int_0^a f'(y) dy,$$

$$f(0) = \int_R \mathrm{sinc}^2(x) dx = \pi,$$

$$f'(y) = -\int_R \sin^2(x) \exp(-yx^2) dx$$

$$= -\frac{1}{2} (\pi/y)^{1/2} [1 - \exp(-1/y)].$$

Furthermore,

$$\int_0^a f'(y) dy = \frac{(\pi)^{1/2}}{2} \left[\int_0^a y^{-1/2} \exp(-1/y) dy - \int_0^a y^{-1/2} dy \right]$$

$$= \frac{(\pi)^{1/2}}{2} (2(a)^{1/2} \exp(-1/a) - 4(\pi)^{1/2}$$

$$\times \{1 - \Phi[(2/a)^{1/2}]\} - 2(a)^{1/2}).$$

Hence

$$f(a) = \pi - 2\pi\{1 - \Phi[(2/a)^{1/2}]\} - (\pi a)^{1/2}[1 - \exp(-1/a)]. \tag{C1}$$

For all $x > 0$,

$$\left(\frac{1}{x} - \frac{1}{x^3} \right) \gamma(x) < 1 - \Phi(x) < \frac{1}{x} \gamma(x),$$

where $\gamma(x) = \exp(-x^2/2)/(2\pi)^{1/2}$. Hence, if x is large,

$$1 - \Phi(x) \approx \exp(-x^2/2)/[x(2\pi)^{1/2}]$$

is a good approximation. If $a > 0$ is small, $(2/a)^{1/2}$ is large, and we have

$$1 - \Phi[(2/a)^{1/2}] \approx (a/2)^{1/2} \exp(-1/a)/(2\pi)^{1/2}$$

$$= \frac{1}{2} (a/\pi)^{1/2} \exp(-1/a).$$

Hence

$$f(a) \approx \pi - (\pi a)^{1/2} \exp(-1/a) - (\pi a)^{1/2}$$

$$+ (\pi a)^{1/2} \exp(-1/a) = \pi - (\pi a)^{1/2}. \tag{C2}$$

Concerning $F_\Delta(\epsilon)$ and $F_\Delta(1 - \epsilon)$, we give the following estimates without proofs:

$$F_\Delta(1 - \epsilon) \approx 1,$$

$$F_\Delta(\epsilon) < \epsilon,$$

when $\epsilon > 0$ is small.

APPENDIX D

In this appendix we will deduce a general expression for the mean and variance for a general n-wave-mixing process.

Assume that we have an n-wave-mixing process using N different multimode laser sources and taking f_v photons from laser v; $v = 1, \ldots, N$. Let

$$F_k = \sum_{v=1}^k f_v, \qquad k \geqslant 1,$$

and define F_0 by $F_0 \equiv 0$. Furthermore, let

$$I_j = \sum_u \left\{ \prod_{k=1}^N \left[\prod_{x=2F_{k-1}+1}^{2F_k} a_k[u(x)] \right] \right.$$

$$\times \exp\left(i \sum_{y=F_{k-1}+1}^{F_k} \{\theta_k[u(2y-1)] - \theta_k[u(2y)]\} \right)$$

$$\left. \times H_j[u(1), u(2), \ldots, u(2F_N)] \right\}.$$

The outer summation is over the indices $u(1), u(2), \ldots, u(2F_N)$ and $u(v) \epsilon Z$, $v = 1, \ldots, 2F_N$.

As usual $[a_k{}^2(r)]_r$ is a sequence of independent identically distributed $\Gamma(l, m_k)$ variables and $[\theta_k(r)]_r$ is a sequence of independent identically distributed $R(0, 2\pi)$ variables, $k = 1, \ldots, N$. If $m \neq n$, $\theta_m(r)$ is independent of $\theta_n(s)$ and $a_m(r)$ is independent of $a_n(s)$ for all r and s, $a_m(r)$ and $\theta_n(s)$ are independent for all m, n, r, and s, and H_j is a (possibly complex) deterministic function.

Now we define the permutation operators $P_n(2m)$ and $\tilde{P}_n(2m)$. $P_n(2m)$ operates on a function $H(k_1, \ldots, k_{2N})$. More specifically, $P_n(2m)$ permutates the variables $k_{n+1}, \ldots, k_{n+2m}$ in the following way: k_{n+2} is replaced by any one of the variables $k_{n+2r-1}, r = 1, \ldots, m$. This can be done in m different ways. Next, k_{n+4} is replaced by any of the remaining $m - 1$ variables. We continue this procedure until k_{n+2m} is replaced by the only remaining variable:

Definition:

$$P_n(2m)[H(k_1, \ldots, k_n, k_{n+1}, \ldots, k_{n+2m}, k_{n+2m+1}, \ldots, k_{2N})]$$

$$= \left(\prod_{j=1}^m \sum_{\substack{i_j \in A_n(m) \\ i_1 \neq \ldots \neq i_j}} \right) H(k_1, \ldots, k_n, k_{n+1}, k_{i_1}, k_{n+3}, k_{i_2},$$

$$\ldots, k_{n+2m-1}, k_{i_m}, k_{n+2m+1}, \ldots, k_{2N}),$$

where $A_n(m) = \{n + 2r - 1; r = 1, \ldots, m\}$ and

$$\prod_{j=1}^m \sum_{\substack{i_j \in A_n(m) \\ i_1 \neq \ldots \neq i_j}} = \sum_{\substack{i_m \in A_n(m) \\ i_1 \neq \ldots \neq i_m}} \cdots \sum_{i_3 \in A_n(m)} \sum_{\substack{i_2 \in A_n(m) \\ i_1 \neq i_2 \neq i_3}} \sum_{\substack{i_1 \in A_n(m) \\ i_1 \neq i_2}}.$$

The operator $\tilde{P}_n(2m)$ operates on a product of two functions

$$H_1(s_1, \ldots, s_n, k_{n+1}, \ldots, k_{n+2m}, t_{n+2m+1}, \ldots, t_{2N})$$

$$\times H_2(u_1, \ldots, u_n, k_{n+1}, \ldots, k_{n+2m}, v_{n+2m+1}, \ldots, v_{2N}),$$

where the variables at places $n + 1, \ldots, n + 2m$ in H_1 equal the variables at the same places in H_2. The remaining variables s, t, u, and v are arbitrary. As with $P_n(2m)$, $\tilde{P}_n(2m)$ permutates only the variables $k_{n+1}, \ldots, k_{n+2m}$ and leaves the others unaffected. The permutation works as follows: in H_1, k_{n+2} is replaced by any one of the variables $k_{n+r}, r = 1, \ldots, 2m$, which can be done in $2m$ different ways. Next, still in H_1, k_{n+4} is replaced by any of the $2m - 1$ remaining variables. The procedure of replacing $k_{n+2r}, r = 1, \ldots, m$

(in H_1) by any one of the variables not yet used continues until k_{n+2m} have been assigned a value among the now $m + 1$ remaining variables. Now, we permute the k variables in H_2 in a similar way: k_{n+1} is replaced by any one of the m remaining variables, then k_{n+3} is replaced, and so on until k_{n+2m-1} has been assigned the last variable. Definition:

$$\tilde{P}_n(2m)[H_1(s_1, \ldots, s_n, k_{n+1}, \ldots, k_{n+2m}, t_{n+2m+1}, \ldots, t_{2N})$$

$$\times H_2(u_1, \ldots, u_n, k_{n+1}, \ldots, k_{n+2m}, v_{n+2m+1}, \ldots, v_{2N})]$$

$$= \left(\prod_{j=1}^{2m} \sum_{\substack{i_j \in \tilde{A}_n(m) \\ i_1 \neq \ldots \neq i_j}}\right)[H_1(s_1, \ldots, s_n, k_{n+1}, k_{i_1}, k_{n+3}, k_{i_2},$$

$$\ldots, k_{n+2m-1}, k_{i_m}, t_{n+2m+1}, t_{2N})$$

$$\times H_2(u_1, \ldots, u_n, k_{i_{m+1}}, k_{n+2}, k_{i_{m+2}}, k_{n+4},$$

$$\ldots, k_{i_{2m}}, k_{n+2m}, v_{n+2m+1}, \ldots, v_{2N})],$$

where $\tilde{A}_n(m) = \{n + r; r = 1, \ldots, 2m\}$. Using these two permutation operators, we make the following two statements:

$$E(I_1) = \left(\prod_{v=1}^{N} m_v^{f_v}\right) \sum \left\{\left[\prod_{n=1}^{N} P_{2F_{n-1}}(2f_n)\right]\right.$$

$$\left.\times H_1[u(1), \ldots, u(2N)]\right\}, \tag{D1}$$

$$E(I_1\bar{I}_2) = \left(\prod_{v=1}^{N} m_v^{2f_v}\right) \sum \left\{\left[\prod_{n=1}^{N} \tilde{P}_{2F_{n-1}}(2f_n)\right]\right.$$

$$\left.\times H_1[u(1), \ldots, u(2N)]H_2\overline{[u(1), \ldots, u(2N)]}\right\}, \tag{D2}$$

where the summation in the first case is over the indices $u(1)$, $u(3), \ldots, u(2N-1)$ and in the second case is over $u(1), u(2)$, $\ldots, u(2N)$. As usual, $u(x) \in Z, x = 1, \ldots, 2N$. We have not proved these statements rigorously, except in the special case where $f_n \leq 2$ and N is arbitrary. However, general considerations indicate that Eqs. (D1) and (D2) are correct for an arbitrary case as long as the stochastic variables belong to the stated distributions. Finally, we have

$$C(I_1, I_2) = E(I_1\bar{I}_2) - E(I_1)\overline{E(I_2)}$$

and in particular

$$V(I_1) = E(|I_1|^2) - |E(I_1)|^2.$$

We note that in a general case, where the mode amplitudes are not exponentially distributed, there will be correction terms to Eqs. (D1) and (D2). The summations in the correction terms will run over fewer indices than the main terms, as given in Eqs. (D1) and (D2), and thus will normally be smaller. Thus, as long as only the phases are uniformly distributed over the interval $(0, 2\pi)$, Eqs. (D1) and (D2) can be expected to be reasonable starting points for calculating the noise.

APPENDIX E

In this appendix we give approximate analytical expressions for the noise $\sigma(I)/\langle I \rangle$ in the different experimental approaches to broadband CARS calculated as outlined in the paper and Appendixes A–D. The present expressions are appropriate for the conditions normally encountered in CARS, e.g., such as those given in Table 3. The complete analytical expressions, which, e.g., are valid for much shorter laser excitation pulses, can be obtained from the authors.

Non-Raman Resonant Case

Conventional Coherent Anti-Stokes Raman Scattering
Multimode pump laser:

$$\frac{\sigma(I)}{\langle I \rangle} = \frac{1}{(2\pi)^{1/4}}\left[\frac{(4 + \sqrt{2})\Omega_a\Gamma_a}{\Gamma^2} + \frac{\Omega_b}{(W^2 + 2\Gamma_a^2)^{1/2}}\right.$$

$$\left.+ \frac{8\pi}{T(W^2 + \Gamma_a^2)^{1/2}} + \frac{2\pi}{WT}\right]^{1/2}. \tag{E1}$$

Single-mode pump laser:

$$\frac{\sigma(I)}{\langle I \rangle} = \frac{1}{(2\pi)^{1/4}}\left(\frac{\Omega_b}{W}\right)^{1/2}. \tag{E2}$$

Dual-Broadband Coherent Anti-Stokes Raman Scattering
Multimode pump laser:

$$\frac{\sigma(I)}{\langle I \rangle} = \frac{1}{(2\pi)^{1/4}}\left[\sum_{x=a,b,c} \frac{\Omega_x\Gamma_x}{\Gamma} \frac{1}{(\Gamma^2 - \Gamma_x^2)^{1/2}}\right.$$

$$\left.+ \frac{2\pi}{T}\sum_{x=a,b,c}\left(\frac{1}{W^2 + \Gamma_x^2} - \frac{1}{\Gamma^2}\right)^{1/2} + \frac{2\pi}{WT}\right]^{1/2}. \tag{E3}$$

Single-mode pump laser:

$$\frac{\sigma(I)}{\langle I \rangle} = \frac{1}{(2\pi)^{1/4}}\left[\sum_{x=a,b} \frac{\Omega_x\Gamma_x}{\Gamma} \frac{1}{(\Gamma^2 - \Gamma_x^2)^{1/2}} + \frac{2\pi}{WT}\right]^{1/2}. \tag{E4}$$

Rotational Dual-Broadband Coherent Anti-Stokes Raman Scattering
Multimode pump laser:

$$\frac{\sigma(I)}{\langle I \rangle} = \frac{1}{(2\pi)^{1/4}}\left[\frac{1}{\sqrt{2}}\frac{\Gamma}{\Gamma_a}\frac{\Omega_a}{(W^2 + \Gamma_b^2)^{1/2}} + \frac{\Omega_b\Gamma_b}{\Gamma^2} + \frac{2\pi}{WT}\right]^{1/2}. \tag{E5}$$

Single-mode pump laser [to first order in $(W/\Gamma)^{1/2}$]

$$\frac{\sigma(I)}{\langle I \rangle} = \frac{1}{(2\pi)^{1/4}}\left(\frac{\Omega_a}{W}\right)^{1/2}. \tag{E6}$$

Raman Resonant Coherent Anti-Stokes Raman Scattering

It is now assumed that there is a single Lorentzian Raman resonance centered under the instrument function and (with the exception of RDBC) that we have chosen a diode with which the non-Raman-resonant anti-Stokes signal would have its maximum. Furthermore, the noise is calculated under the conditions given at the end of Appendix B. In particular it should be noted that the bandwidth of the pump laser is assumed to be much larger than the Raman linewidth (Γ_r) in the multimode case. Only terms to first

order in $\Gamma_r/\Gamma_{\text{laser}}$ (~0.1) are included. Furthermore, only zeroth-order terms in $\Gamma_{\text{pump laser}}/\Gamma_{\text{dye laser}}$ (~0.01) are included. The symbol F_δ, which is introduced in the multimode expressions, is ~0.1 if normalization, as in the second half of Appendix B, is employed and is 1 otherwise.

Conventional Coherent Anti-Stokes Raman Scattering
Multimode pump laser:

$$\frac{\sigma(I)}{\langle I \rangle} = \left\{ \frac{1}{(2\pi)^{1/2}} \frac{\Omega_a}{\Gamma_a} \left\{ \left(1 + \frac{\Gamma_a^2}{W^2}\right)^{1/2} + \left(1 + \frac{\Gamma_a^2}{\Gamma_b^2}\right)^{1/2} \right. \right.$$
$$\times \left[1 + 2\left(1 + \frac{\Gamma_a^2}{2W^2 + \Gamma_a^2}\right)^{1/2} \right] \right\}$$
$$\times F_\delta + \frac{1}{(2\pi)^{1/2}} \frac{\Omega_b}{\Gamma_a} \left(1 + \frac{\Gamma_a^2}{\Gamma_b^2}\right)$$
$$+ (2\pi)^{1/2} \frac{1}{T\Gamma_a} \left[\left(1 + \frac{\Gamma_a^2}{W^2}\right)^{1/2} + 4\left(1 + \frac{\Gamma_a^2}{\Gamma_b^2}\right)^{1/2} \right.$$
$$\times \left. \left(1 + \frac{\Gamma_a^2}{2W^2 + \Gamma_a^2}\right)^{1/2} \right] + \frac{1}{T\Gamma_r} \right\}^{1/2}.$$

Single-mode pump laser:

$$\frac{\sigma(I)}{\langle I \rangle} = \frac{1}{(2\pi)^{1/2}} \left(\frac{\Omega_b}{\Gamma_r}\right)^{1/2}.$$

Dual-Broadband Coherent Anti-Stokes Raman Scattering
Multimode pump laser:

$$\frac{\sigma(I)}{\langle I \rangle} = \left[\frac{1}{(2\pi)^{1/2}} \frac{\Omega_c}{\Gamma_c} \left(1 + \frac{\Gamma_c^2}{W^2}\right)^{1/2} F_\delta \right.$$
$$+ \left. \frac{(2\pi)^{1/2}}{T\Gamma_c} \left(1 + \frac{\Gamma_c^2}{W^2}\right)^{1/2} + \frac{1}{T\Gamma_r} \right]^{1/2}.$$

Single-mode pump laser [to first order in $(\Omega_a/\Gamma_a)^{1/2}$]:

$$\frac{\sigma(I)}{\langle I \rangle} = \frac{1}{(T\Gamma_r)^{1/2}}.$$

Rotational Dual Broadband Coherent Anti-Stokes Raman Scattering
Multimode pump laser:

$$\frac{\sigma(I)}{\langle I \rangle} = \left[\frac{1}{(2\pi)^{1/2}} \frac{\Omega_c}{\Gamma_c} \left(1 + \frac{\Gamma_c^2}{W^2}\right) F_\delta \right.$$
$$+ \left. \frac{(2\pi)^{1/2}}{T\Gamma_c} \left(1 + \frac{\Gamma_c^2}{W^2}\right)^{1/2} + \frac{1}{2\pi} \frac{\Omega_a}{\Gamma_r} \right]^{1/2}.$$

Single-mode pump laser [to first order in $(\Gamma_r/\Gamma_a)^{1/2}$]:

$$\frac{\sigma(I)}{\langle I \rangle} = \frac{1}{(2\pi)^{1/2}} \left(\frac{\Omega_a}{\Gamma_r}\right)^{1/2}.$$

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Note added in proof: The calculations for CC and RDBC are done assuming that the two photons taken from the same laser come from the same beam or from two beams correlated in time. For uncorrelated beams the noise in Tables 4 and 5 will decrease by a factor of $\sqrt{2}$ for RDBC.[33,36]

REFERENCES

1. 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).
2. D. A. Greenhalgh and S. T. Whittley, "Mode noise in broadband CARS spectroscopy," Appl. Opt. **24**, 907 (1985).
3. R. J. Hall, "The statistical behaviour of nonresonant CARS intensities," Opt. Commun. **56**, 127 (1985).
4. 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).
5. 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).
6. R. J. Hall, "Theoretical analysis of non-thermal pump effects in broadband CARS spectroscopy," Opt. Quantum Electron. **18**, 319 (1986).
7. G. S. Agarwal and R. L. Farrow, "Theoretical modeling of two-color CARS spectra measured with a frequency-doubled, multimode pump laser," J. Opt. Soc. Am. B **3**, 1596 (1986).
8. R. J. Hall and D. A. Greenhalgh, "Noise properties of single-pulse CARS spectroscopy with multimode pump sources," J. Opt. Soc. Am. B **3**, 1637 (1986).
9. 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).
10. S. Kröll, M. Aldén, T. Berglind, and R. J. Hall, "Noise characteristics of single shot broadband Raman resonant CARS with single- and multimode lasers," Appl. Opt. **26**, 1068 (1987).
11. R. J. Hall and A. C. Eckbreth, "Coherent anti-Stokes Raman spectroscopy (CARS) application to combustion diagnostics," Laser Appl. **5**, 213 (1984).
12. M. Pealat, P. Bouchardy, M. Lefebvre, and J.-P. Taran, "Precision of multiplex CARS temperature measurements," Appl. Opt. **24**, 1012 (1985).
13. 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).
14. D. R. Snelling, T. Parameswaran, and G. J. Smallwood, "Noise characteristics of single-shot broadband CARS signals," Appl. Opt. **26**, 4298 (1987).
15. A. C. Eckbreth and T. J. Anderson, "Dual broadband CARS for simultaneous, multiple species measurements," Appl. Opt. **24**, 2731 (1985).
16. 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).
17. M. Aldén, P.-E. Bengtsson, and H. Edner, "Rotational CARS generation through a multiple four-color interaction," Appl. Opt. **25**, 4493 (1986).
18. W. B. Roh, P. W. Schreiber, and J. P. E. Taran, "Single-pulse coherent anti-Stokes Raman scattering," Appl. Phys. Lett. **29**, 174 (1976).
19. M. A. Yuratich, "Effects of laser linewidth on coherent anti-Stokes Raman spectroscopy," Mol. Phys. **38**, 625 (1979).
20. 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).
21. R. E. Teets, "Accurate convolutions of coherent anti-Stokes Raman spectra," Opt. Lett. **9**, 226 (1984).
22. D. A. Greenhalgh and R. J. Hall, "A closed form solution for the CARS intensity convolution," Opt. Commun. **57**, 125 (1986).
23. F. Y. Yueh and E. J. Beiting, "Analytical expressions for coherent anti-Stokes Raman spectral (CARS) profiles," Comp. Phys. Commun. **42**, 65 (1986).

24. M. Aldén, Department of Atomic Physics, Combustion Centre, Lund Institute of Technology, Box 118, S-221 Lund, Sweden (personal communication).
25. G. L. Eesley, *Coherent Raman Spectroscopy* (Pergamon, New York, 1981), Chap. 3B.
26. M. D. Levenson, *Introduction to Nonlinear Laser Spectroscopy* (Academic, New York, 1982).
27. See, e.g., in several of the contributions in the special issue on stimulated Raman and Brillouin scattering for laser beam control, J. Opt. Soc. Am. B 3(10) (1986), and references therein.
28. L. A. Westling and M. G. Raymer, "Intensity autocorrelation measurements and spontaneous FM phase locking in a multimode pulsed dye laser," J. Opt. Soc. Am. B **3,** 911 (1986).
29. D. S. King and R. R. Cavanagh, "Streak-camera analysis of XeCl- and N_2-pumped dye-laser outputs," Opt. Lett. **8,** 18 (1983).
30. R. E. Teets, "CARS signals: phase matching, transverse modes, and optical effects," Appl. Opt. **25,** 855 (1986).
31. J. Ducuing and N. Bloembergen, "Statistical fluctuations in nonlinear optical processes," Phys. Rev. **133,** A1493 (1964).
32. A. M. Mood, *Introduction to the Theory of Statistics* (McGraw-Hill, New York, 1974), Chap. 5.
33. M. Aldén, P. E. Bengtsson, H. Edner, S. Kröll, and D. Nilsson, "Rotational CARS: comparison of different techniques with emphasis on temperature accuracies," to be submitted to Appl. Opt.
34. L. A. Westling, M. G. Raymer, and J. J. Snyder, "Single-shot spectral measurements and mode correlations in a multimode pulsed dye laser," J. Opt. Soc. Am. B **1,** 150 (1984).
35. Z. W. Li, C. Radzewicz, and M. G. Raymer, "Temporal smoothing of multimode dye laser pulses," Opt. Lett. **12,** 416 (1987).
36. S. Kröll and D. Sandell, "A model for calculating the noise due to the stochastic nature of multimode laser radiation in nonlinear optical processes," in *Proceedings of the International Conference on Nonlinear Optics*, V. J. Corcoran, ed. (Society for Optics and Quantum Electronics, McLean, Va., to be published).