Noise characteristics of single-shot broadband CARS signals

Snelling, D. R.; Parameswaran, T.; Smallwood, G. J.

Publisher's version / Version de l'éditeur:

Applied Optics, 26, 19, pp. 4298-4302, 1987

Noise characteristics of single-shot broadband CARS signals

D. R. Snelling, T. Parameswaran, and G. J. Smallwood

Spectrally resolved measurements of noise in resonant nitrogen coherent anti-Stokes Raman spectroscopy (CARS) are presented for three pump laser bandwidths. The experimental noise curves are compared with those calculated from a simple model involving statistically independent Stokes modes. The sources of noise in both resonant and nonresonant CARS spectra are discussed.

I. Introduction

Single-pulse broadband coherent anti-Stokes Raman spectroscopy (CARS) has become an important diagnostic technique,[1–7] particularly for combustion measurements of temperature and species concentration. The single-pulse (10-ns) capability is crucial for the study of turbulent combustion environments such as gas turbine combustors, internal combustion engines, and turbulent flames.

Noise in the CARS signal limits the accuracy of these single-pulse measurements; and pulse-to-pulse variation in the output of the broadband dye laser is the predominant source of this noise. A picture of CARS noise has emerged[8,9] whereby the greater the number of dye laser modes contributing to the excitation of the CARS signal the lower the resultant CARS noise becomes. In particular, a model of dye laser noise based on statistically independent modes with random phases[8] has recently been adduced[10–12] to explain the observed dependence of resonant and nonresonant CARS noise[6,10] on the pump laser bandwidth. A spectral dependence of the amplitude fluctuations observed in the temporal profile of the broadband dye laser has been proposed[3,9,10] as an additional source of noise. CARS pulses shorter than the laser intramode beat periods may also lead to incomplete temporal averaging.[12] The benefit of lengthening the CARS pulse has also been discussed.[9]

Calculations of average (spectral) noise levels in resonant (Raman resonant N_2 spectra) and nonresonant CARS spectra[8,10,11] are in semiquantitative agreement with experimental results.[6,8,10] In particular it has been shown[10,11] that the decrease in N_2 resonant CARS spectral noise with increasing pump laser bandwidth (see Table I which is reproduced from Ref. 10) is attributable to the greater number of dye laser modes contributing as the pump laser bandwidth increases. Kroll *et al.*[11] have developed a simple explicit semiquantitative model for estimating the average relative noise of various broadband CARS techniques. We have very briefly reported[10] numeric calculations where the CARS noise was obtained by computing the quantity:

$$\frac{\sigma}{\langle I\rangle} = \left(\sum_k \langle I_k\rangle^2\right)^{1/2} \sum_k \langle I_k\rangle, \qquad (1)$$

which represents[8] the noise attributable to K statistically independent modes of intensity $\langle I_k\rangle$. In Eq. (1), σ is the standard deviation, the $\langle\,\rangle$ brackets indicate a time average, and $\langle I\rangle = \Sigma\langle I_k\rangle$. In this analysis the following assumptions are made.[8,12] The Stokes and pump lasers are statistically independent. For pump lasers with a finite bandwidth, the medium samples a large number of fluctuations in the intensities in a single-pulse CARS experiment. Therefore, the pump laser may be crudely treated as a coherent source with a constant intensity equal to the time average.

With the above assumptions the effect of the pump laser can be included in the calculation of $\langle I_k\rangle$ by convolving the monochromatic CARS intensity with the appropriate pump laser bandwidth, and the noise in the CARS spectrum can then be attributed to the modes in the dye laser [Eq. (1)].

Spectrally averaged (15-cm^{-1} bandwidth) values of the fractional noise $\sigma/\langle I\rangle$, calculated in this way, showed the same trends as the experimental data in Table I; however, the calculated values were somewhat lower.[10] The dye mode spacing was taken to be 0.01 cm^{-1} (approximately the longitudinal mode spacing in our dye laser), and the $\langle I_k\rangle$ was calculated as the pump convolved CARS intensities at the 0.01-cm^{-1} interval as modulated by the slit function.[10]

The authors are with National Research Council of Canada, Division of Mechanical Engineering, Ottawa, Ontario K1A 0R6, Canada.

Received 28 February 1987.
0003-6935/87/194298-05$02.00/0.

Table I. Noise Statistics: Standard Deviation σ Expressed as Percent Calculated for 15-cm^{-1} Bandwidth

Spectrum type	Pump laser configuration: Single mode	Multimode (0.10 cm^{-1})	Multimode (0.69 cm^{-1})
CO_2 nonresonant spectra	6.6	8.4	9.1
N_2 flame spectra $T = 1580$ K	22.4(22.0)	16.1(15.6)	11.1(10.8)
The above values are reproduced from Ref. 10			
Calculated for 70-cm^{-1} Bandwidth			
CO_2 nonresonant spectra	7.7	10.3	10.5

In this paper we present for the first time spectrally resolved measurements of CARS noise for pump laser bandwidths of 0.1 and 0.69 cm^{-1} as well as single-mode pump laser operation. The observed noise, with the component attributable to detector shot noise subtracted, is compared with theoretical calculations based on Eq. (1).

II. Results and Discussion

The theoretical noise was calculated on the basis of Eq. (1) as outlined in the previous section. The dye laser mode spacing was assumed to be constant, independent of wavelength. An asymmetric Voigt instrument function[10] was used to calculate the I_k values from the pump-convolved CARS intensities generated at the assumed dye laser mode spacing. The theoretical CARS computer program used included the effects of cross-coherence and collisional narrowing.[10] The summation in Eq. (1) was performed over all the I_k values falling under the envelope of the Voigt instrument function, and $\sigma/\langle I\rangle$ was calculated as a function of frequency (cm^{-1}). $\sigma/\langle I\rangle$ calculated in this way represents the expected fractional noise (single standard deviation) associated with a given detector element (pixel) of the diode array tuned to that frequency. The mode spacing (0.02 cm^{-1}) was chosen to give the best fit to the experimental single-mode data.

This is consistent with the method of calculating noise from experimental spectra[6,10] where we first divide the individual spectra by an averaged spectrum (to remove the dependence on diode sensitivity) and then divide by the integrated intensity from all detector elements (pixels) in the analysis bandwidth. This ensures that fluctuations in the spectrally integrated CARS signal do not contribute to the measured noise of individual pixels.[6,10] It should be noted, however, that the experimental noise defined in this way is a weak function of the analysis bandwidth; for example, a decrease in the analysis bandwidth from 70 to 30 cm^{-1} produced a 13% reduction in the measured nonresonant CARS noise.[6]

The experimental CARS spectra were recorded in a hydrogen/air fueled flat-flame burner for pump laser bandwidths of 0.1 and 0.69 cm^{-1} and for single-mode pump laser operation.[10] The method of calculating the spectrally averaged noise has been discussed previously[6,10] where the quantity Y_{ij}, which represents the normalized CARS signal, is defined. i and j represent the diode index and spectrum index, respectively. The standard deviation of the spectra Y_{ij} was calculated for each diode to yield a percentage noise N_i, which represents the shot-to-shot variability of the CARS noise. This noise can be corrected for detector shot noise by assuming that the detector shot noise component σ_D and the corrected experimental noise σ_C are uncorrelated and give a net noise σ_N given by

$$\sigma_N^2 = \sigma_D^2 + \sigma_C^2. \tag{2}$$

We have previously measured the detector shot noise component, which is given by[10]

$$\sigma_D^2 = \sigma_0^2 + kC, \tag{3}$$

where σ_0^2 is the dark noise and kC is the shot noise component for a diode signal of C counts. Measured values of σ_0 and k[10] were used to obtain the corrected experimental noise σ_C, which is plotted in the accompanying figures. At very low CARS intensities the noise is dominated by detector shot noise, and the calculation of σ_C becomes uncertain as can be seen in Figs. 1 and 2.

The single-mode pump laser noise curves are shown in Fig. 1 with the experimental (400-pulse average) CARS spectrum. A mode spacing of 0.02 cm^{-1} was chosen to give the best fit to the single-mode experimental data, and the theoretical noise was calculated for a temperature of 1550 K and 1-atm pressure, the conditions appropriate to the experimental spectra. The spectral shapes of the theoretical and experimental noise curves are quite similar. In particular, the observed spectral coincidence of noise peaks with peaks in the CARS spectrum is predicted by theory as is the falloff in noise in the lower intensity regions of the CARS spectrum beyond the $V = 1$–0 bandhead and in the vicinity of the 3–2 band. It is interesting to note that both the theoretical and experimental noise spectra exhibit considerably more modulation than do the corresponding CARS spectra. Heuristically, the noise peaks can be thought of as resulting from fewer dye laser modes contributing to the summation in Eq. (1) due to the discontinuous narrow (typically 0.03–0.05 cm^{-1}) Raman resonances in the pump laser convolved CARS spectrum. In the lower intensity regions of the CARS spectrum, where the nonresonant contribution is more important, the pump laser convolved CARS spectrum is relatively more continuous leading to the participation of many more dye laser modes in Eq. (1). In the limit of a continuous featureless nonresonant spectrum the theoretical noise becomes 7.9% independent of frequency and pump laser bandwidth. This is in good agreement with the experimental value of 7.7% obtained from nonresonant CARS spectra using a single-mode pump laser and an analysis bandwidth of 70 cm^{-1} (Table I). (As noted above and observed previously[6] the experimental noise is a weak function of the analysis bandwidth.) Thus the experimental noise observed in both the resonant N_2 flame spectra and the

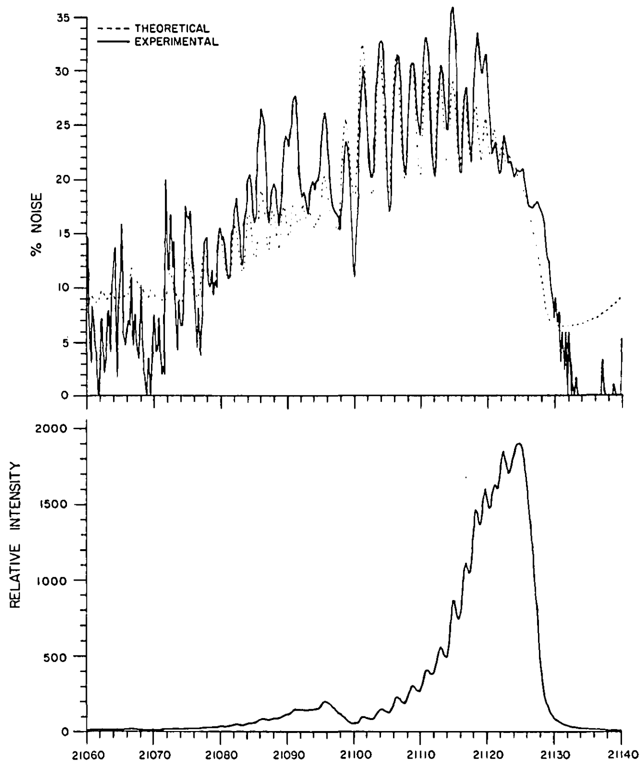

Fig. 1. Top: CARS noise (single standard deviation) for single-mode pump laser: dotted curve, theoretical; solid curve, observed. Bottom: 400 pulse average single-mode CARS spectrum. The theoretical calculations were based on an assumed dye laser mode spacing of 0.02 cm^{-1}. The equivalent width of the asymmetric Voigt instrument function used in the calculations was 1.89 cm^{-1}.

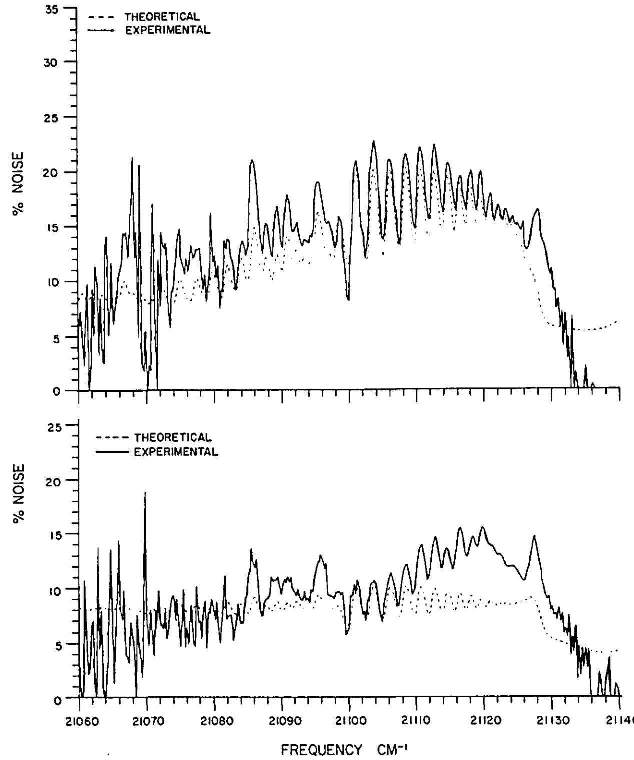

Fig. 2. Theoretical (dotted curve) and observed (solid curve) CARS noise (single standard deviation). Top: 0.10-cm^{-1} multimode pump laser. Bottom: 0.69-cm^{-1} multimode pump laser. The theoretical calculations were based on an assumed dye laser mode spacing of 0.02 cm^{-1}. The equivalent width of the asymmetric Voigt instrument function used in the calculations was 2.04 cm^{-1}.

nonresonant CO_2 spectra are in satisfactory agreement with Eq. (1) if a dye laser mode spacing of 0.02 cm^{-1} is assumed.

The limited number of previous comparisons of calculated and observed noise[8,10,11] have used spectrally averaged values. The structure in the spectral noise curves (Figs. 1 and 2) indicates the importance of specifying the exact bandwidth in such comparisons.

The experimental and theoretical noise curves (using the same 0.02-cm^{-1} dye laser mode spacing assumed for the single-mode pump laser comparison) for pump laser bandwidths of 0.1 and 0.69 cm^{-1} are shown in Fig. 2. The 0.1-cm^{-1} results are in satisfactory agreement, but, for the larger pump laser bandwidth, the calculated curve underestimates the observed noise. That there is an additional source of noise in the CARS spectra to that represented by Eq. (1) is not surprising, since this equation predicts the noise in nonresonant CARS spectra to be independent of laser bandwidth, which is in disagreement with the experimental data reproduced in Table I. From a more detailed analytic analysis, Hall and Greenhalgh[12] also conclude that the noise in nonresonant CARS spectra should be largely independent of pump laser bandwidth. However, the assumption of uncorrelated pump and Stokes laser beams inherent in this analysis and in the derivation of Eq. (1) may be invalid for the multimode pump laser results in Fig. 2. In fact, intensity correlations have been observed between the Stokes and pump lasers in that the temporal profile of the Stokes laser attempts to follow that of the multimode pump laser.[6] These correlations between the temporal profiles of the pump and Stokes lasers, which vary over the Stokes spectral profile, have been adduced[3,6,9,10] to explain the observed increase in the nonresonant CARS noise with increasing pump laser bandwidths.

The dye laser used in our experiments is an oscillator/amplifier configuration with an amplifier gain of ~4. Both the oscillator and amplifier are pumped slightly off-axis. The high-gain near-saturated amplifier, which dominates the output of our dye laser, has a temporal profile that follows that of the pump laser.[6] We have observed[6] a spectral dependence to the Stokes laser temporal amplitude profiles which can account for one source of noise. The temporal behavior of the Stokes laser may be a function of the pumping geometry,[12] which can vary from one CARS system to another.

We can estimate this temporal contribution to the observed nonresonant noise by assuming that the single-mode noise is attributable to mode noise only [Eq. (1)] and subtracting this component from the multimode noise. Assuming that the temporal σ_T and mode noise σ_M contributions are uncorrelated the net noise σ_N is given by

$$\sigma_N^2 = \sigma_M^2 + \sigma_T^2. \quad (4)$$

For the 70-cm^{-1} bandwidth σ_T is 6.7% (0.1-cm^{-1} multimode pump laser) and 7.1% (0.69-cm^{-1} multimode pump laser).

This temporal noise component can also qualitatively account for the differences observed between the calculated and theoretical spectra in Fig. 2. While a detailed comparison is not warranted, Eq. (4) can be used to obtain the net calculated noise with the values of σ_T given above. It can be readily shown that this would increase the calculated noise by 1–2% (0.1-cm^{-1} multimode pump) and 2–3% (0.69-cm^{-1} multimode pump) in the central higher-intensity region of the CARS resonant spectrum. This would improve the agreement between calculated and observed noise, particularly for the 0.69-cm^{-1} data.

Returning to the question of mode noise as represented by Eq. (1) we can see that, although the spectral forms of the theoretical noise curves are in good agreement with the single-mode experiment, the absolute fit was obtained by selecting a Stokes (dye) laser mode spacing of 0.02 cm^{-1}. It has been suggested that CARS generation is dominated by the longitudinal TEM_{00} modes.[8,13] The longitudinal mode spacing in our dye laser[6,10] is ~0.009 cm^{-1} or about one-half of the mode spacing assumed to fit the data in Fig. 1. By deliberately inducing some spherical aberration in the dye laser we observed a drop in noise that was attributed to the greater participation of higher-order transverse modes in the CARS generation with the aberrated beam.[10] The contribution of these higher-order modes in the absence of deliberately induced aberration is unclear. However, their participation in the CARS generation would further increase the discrepancy noted above. The larger Stokes laser mode spacing required to fit the data in Fig. 1 may result from the systematic absence of modes in the dye laser output. A more likely explanation is that mode correlations, such as those observed in a multimode pulsed dye laser,[14] may also contribute to the reduction in the effective number of independent modes contributing to Eq. (1). These mode correlations have been interpreted in terms of a simple model that calculates the effect of gain competition on the modes.[14]

While the above results show that at atmospheric pressure the single-mode resonant spectra are considerably noisier than the 0.69-cm^{-1} multimode spectra, this difference is expected to decrease with increasing pressure. The broadening of the Raman resonances leads to the contribution of a greater number of Stokes laser modes, and, in the limit of completely overlapping lines, the number of modes contributing is independent of pump laser bandwidth. The effect of pressure on the calculated single-mode noise is demonstrated in Fig. 3 where theoretical noise curves are shown for pressures of 1 and 5 atm. It can be seen that, as expected, there is a dramatic decrease in noise with increasing pressure down to a level approaching

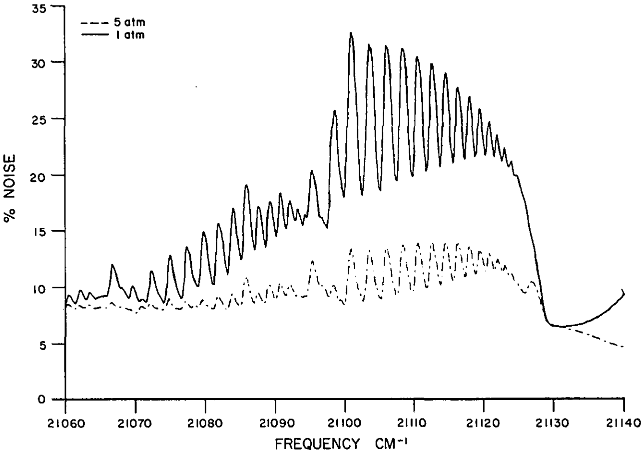

Fig. 3. Theoretical CARS noise for single-mode pump laser: solid curve, 1 atm; dot–dash curve, 5 atm. The theoretical calculations were based on an assumed dye laser mode spacing of 0.02 cm^{-1}.

the 1-atm 0.69-cm^{-1} pump laser bandwidth results shown in Fig. 2. The calculations show that there is little difference between the single-mode and 0.69-cm^{-1} multimode noise at 5-atm pressure.

III. Conclusions

We have shown that the spectral form of the CARS noise with varying pump laser bandwidth can be interpreted in terms of Stokes laser mode noise [Eq. (1)] and an additional term, which we have attributed to temporal variations in the multimode laser pulses. The Stokes laser mode spacing, which is required to fit calculated and experimental noise curves, is about one-half of the expected longitudinal mode spacing in our Stokes laser and may be indicative of missing modes or, more likely, of mode correlations, which reduce the number of independent modes. While the results indicate that at atmospheric pressure a multimode pump laser will lead to substantially lower noise in resonant nitrogen CARS spectra, the advantage over a single-mode pump source is greatly reduced at higher pressure (5 atm). At higher pressures, as the Raman spectrum becomes less structured, the resonant nitrogen CARS spectral noise is expected to approach the nonresonant limit where a single-mode pump source now leads to lower CARS noise.

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," in *Laser Applications, Vol. 5*, J. F. Ready and R. K. Erif, Eds. (Academic, New York, 1984), pp. 213–309.
3. 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).
4. R. L. Farrow, R. P. Lucht, G. L. Clark, and R. E. Palmer, "Species Concentration Measurement using CARS with Nonresonant Susceptibility Normalization," Appl. Opt. **24**, 2241 (1985).

5. 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).
6. 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).
7. M. Pealat, P. Bourchardy, M. Lefebvre, and J. P. Taran, "Precision of Multiplex CARS Temperature Measurements," Appl. Opt. **24**, 1012 (1985).
8. D. A. Greenhalgh and S. T. Whittley, "Mode Noise in Broadband CARS Spectroscopy," Appl. Opt. **24**, 907 (1985).
9. A. C. Eckbreth and J. H. Stufflebeam, "Considerations for the Application of CARS to Turbulent Reacting Flows," Exp. Fluids **3**, 301 (1985).
10. D. R. Snelling, G. J. Smallwood, R. A. Sawchuk, and T. Parameswarm, "Precision of Multiplex CARS Temperatures using both Single-Mode and Multimode Pump Lasers," Appl. Opt. **26**, 99 (1987).
11. S. Kroll, M. Alden, 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).
12. 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).
13. R. E. Teets, presented at *First International Laser Science Conferences* (Dallas, 1985).
14. 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).

Meetings Calendar *continued from page 4292*

1988

March

13–15 2nd Int. Conf. on Artificial Intelligence Systems, Chicago *Int. Academy of Cytology, 5841 Maryland Ave., House Mail Box #449, Chicago, IL 60637*

13–18 Advances in Semiconductors & Semiconductor Structures Mtg., Orlando *SPIE, P.O. Box 10, Bellingham, WA 98227*

13–18 Optics & Optoelectronics Symp. Southeast, Orlando *SPIE, P.O. Box 10, Bellingham, WA 98227*

16–18 7th Ann. IEEE Phoenix Conf. on Computers & Communications, Scottsdale *IEEE, E. 42nd St., New York, NY 10017*

20–25 4th Int. Congr. on Advances in Non-Impact Printing Technologies, New Orleans *SPSE, 7003 Kilworth Lane, Springfield, VA 22151*

23–25 **Optical Bistability Top. Mtg.,** Aussois, France *OSA Mtgs. Dept., 1816 Jefferson Pl., N.W., Wash., DC 20036*

28–30 **LEOS/OSA Top. Mtg. on Integrated & Guided-Wave Optics,** Santa Fe *OSA Mtgs. Dept., 1816 Jefferson Pl., N.W., Wash., DC 20036*

29–31 Optical Storage of Documents & Images Mtg., Wash., DC *TOC, P.O. Box 14817, San Francisco, CA 94114*

April

4–8 Tech. Symp. Southeast on Optics, Electro-Optics, & Sensors, Orlando *SPIE, P.O. Box 10, Bellingham, WA 98227*

5–8 Process Diagnostics Mtg., Reno *A. Hays, Div. 1831, Sandia Natl. Labs., Albuquerque, NM 87185*

5–8 Materials Research Soc. Mtg., Reno *MRS, 9800 McKnight Rd., Ste. 327, Pittsburgh, PA 15237*

12–14 **Optical Interference Coatings Top. Mtg., Tucson** *OSA Mtgs. Dept., 1816 Jefferson Pl., N.W., Wash., DC 20036*

13–15 4th Int. Conf. on Metrology & Properties of Eng. Surfaces, Gaithersburg *T. Vorburger, A117 Metrology Bldg., NBS, Gaithersburg, MD 20899*

14 Spectroradiometric Measurement Mtg., Teddington *G. Freeman, Natl. Physical Lab., Teddington, Middlesex TW11 OLW, England*

19–21 Int. Aerospace & Ground Conf. on Lightning & Static Electricity, Oklahoma City *D. MacGorman, Natl. Severe Storm Lab., 1313 Halley Circle, Norman, OK 73069*

19–22 Analytica 88, Munich *G. Kallman, Kallman Assocs., 5 Maple Court, Ridgewood, NJ 07450*

20–27 Optec '87, Hannover *Hannover Fairs USA Inc., 103 Carnegie Ctr., P.O. Box 7066, Princeton, NJ 08540*

25–29 **Conf. Lasers & Electro-Optics, Anaheim** *OSA Mtgs. Dept., 1816 Jefferson Pl., N.W., Wash., DC 20036*

26–28 Fibre Optics '88, Olympia *Sira Ltd. Conf. Off., South Hill, Chislehurst, Kent BR7 5EH, England*

May

2–6 Int. Conf. on Nonlinear Optical Phenomena, Ashford Castle, Ireland *NLO '88, P.O. Box 245, McLean, VA 22101*

9–13 New Mexico State U. Short Courses in Applied Optics, Las Cruces *L. Radziemski, Physics Dept., NMSU, Las Cruces, NM 88003*

continued on page 4312