Power generation in coherent anti-Stokes Raman spectroscopy with focused laser beams

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(Received 1 June 1977)

Focusing effects for nonlinear power generation are explicitly discussed for coherent anti-Stokes Raman spectroscopy (CARS). It is shown that very tight focusing only increases CARS power generation by about a factor of 4, while an extra λ^{-2} dependence introduced by focusing implies CARS signals scale with wavelength in the same manner as do the signals in normal spontaneous Raman scattering. Axial power generation in a gas phase medium is illustrated, and a modified plane wave approximation function is developed to analyze this behavior for loose focusing cases.

INTRODUCTION

In recent years the technique of coherent Anti-Stokes Raman spectroscopy (CARS) has received considerable attention, as it affords an enormously increased signal-to-noise ratio and superb discrimination against background fluorescence, compared to the more conventional technique of spontaneous Raman scattering.[1–3,5] Expressions for the efficiency of generation of CARS emission by four wave mixing relative to stimulated Raman scattering has been theoretically developed on the assumption of collinear plane waves.[1–5] Estimates based on these expressions predict very high Raman efficiencies (as much as 1% peak power conversion).[1,2] More recent theoretical calculations, made on the assumption of focused collinear Gaussian waves, predict an enhancement in Raman efficiency should occur due to focusing effects. Bjorklund[6] and Regnier[7] in particular have performed theoretical calculations based on the method of Ward and New[8] which demonstrate several important features of focused Gaussian beams used in the generation of CARS emission. This paper recasts Bjorklund's general expression for various nonlinear power generation to a form which is used to provide further insight concerning focusing effects for CARS. Additionally, the development of a closed form modified plane wave expression, which to a good approximation qualitatively reproduces Bjorklund's results for loose focusing, is discussed. It is shown that this approximation is useful as a simple method for predicting the relative magnitude of the CARS power generated at any distance along the axis perpendicularly away from the input point of the pump beams to the focusing medium.

CARS POWER-GAUSSIAN FIELD TREATMENT

In Bjorklund's focusing paper[6] the fundamental electric fields are given by the theory of Boyd and Gordon[9] as

$$E_n(r) = E_{n0}\exp(ik_nz)\,(1+i\epsilon)^{-1}\exp[-k_n(x^2+y^2)/b(1+i\epsilon)] \ . \tag{1}$$

Above, b is the confocal parameter and ϵ is a normalized coordinate along the direction of propagation (the z axis arbitrarily), defined as

$$\epsilon = 2(z-f)/b \ ,$$

in which f is the position of the minimum beam waist along the axis.

It is shown that for the four wave mixing process $2\omega_1-\omega_2$ the resultant CARS field generated at ω_3 is given by

$$E_3(r) = i\frac{3N\chi}{2k_3}\pi k_0^2 bE_{10}^2E_{20}\exp(ik'z)\int_{-\xi}^{\epsilon} d\epsilon' \times \frac{\exp[-i(b/2)\,\Delta k\,(\epsilon'-\epsilon)]}{(1+i\epsilon')\,(k''-ik'\epsilon')H}\exp\left[\frac{-(x^2+y^2)}{bH}\right] , \tag{2}$$

where $k''=2k_1+k_2$, $k'=2k_1-k_2$, $N\chi$ is the bulk third order nonlinear susceptibility, $\Delta k=k_3-k'$, and H is given by

$$H=\frac{1+\epsilon'^2}{k''-ik'\epsilon'}-i\frac{\epsilon'-\epsilon}{k'} \ .$$

If one assumes the pump beams are essentially undepleted as a result of CARS power generation, the maximum field amplitudes of the pump beams E_{n0} can be shown to be[10] related to the pump power P_n through the expression

$$|E_{n0}|^2=\frac{16P_n}{cd_0^2} \ , \tag{3}$$

where c is the speed of light and d_0 is the minimum beam waist radius. All variables are expressed in cgs units. Thus, in an experiment where the total power P_n is held constant and the tightness of the focusing (the value of d_0) is varied E_{n0} does not remain constant, but rather varies as d_0^{-1}.

The total power generated at the anti-Stokes frequency is then given by an integral of the form

$$P_3\propto\int_0^\infty 2\pi R\,|E_3(r)|^2\,dR \ . \tag{4}$$

Equations (2) and (3) can then be used to write (4) as (phase matched case, $\Delta k=0$)

$$P_3=\left(\frac{3\pi^2\omega_3}{c^2n}\right)^2(N\chi)^2P_1{}^2P_2 \times\left\{\frac{32}{\pi^3}\left(\frac{b}{d_0^3}\right)^2\int_0^\infty 2\pi R\left|\int_{-\xi}^{\epsilon}\frac{\exp[-R^2/bH]d\epsilon'}{(1+i\epsilon')H(k''-ik'\epsilon')}\right|^2dR\right\} \tag{5}$$

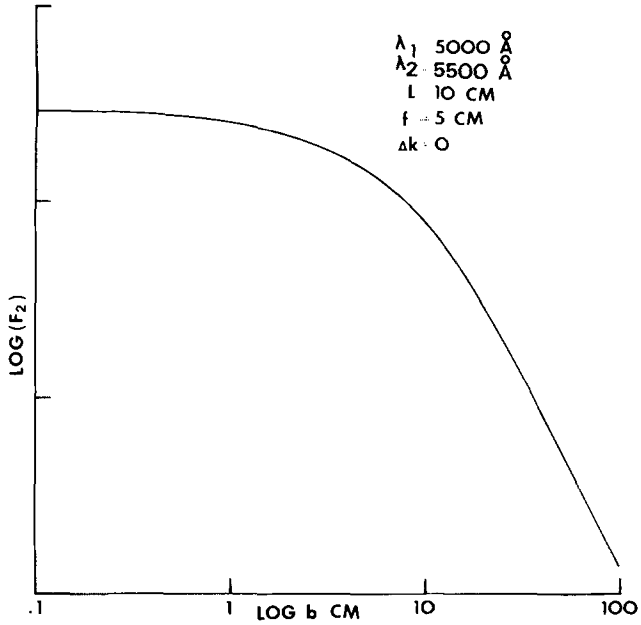

FIG. 1. The focusing enhancement. F_2 as described by Bjorklund[6] is plotted as a function of the confocal parameter b.

In the above expression for the CARS process P_1 and P_2 correspond to the laser and Stokes pump beam powers, respectively, and n is the refractive index. The expression for focused pump beams is cast in the above form for comparison to the expression derived for plane wave fields[1,2,5]

$$P_3 = \left(\frac{3\pi^2\omega_3}{c^2 n}\right)^3 (N\chi)^2 P_1{}^2 P_2 \frac{A_3}{A_1^2 A_2} z^2 \ . \tag{6}$$

Above, A_i is the cross-sectional area of each laser beam, assumed constant for the plane wave case. Examination of Eqs. (5) and (6) shows the focusing effects are determined by an expression of the form

$$F = \frac{32}{\pi^3}\left(\frac{b}{d_0^3}\right)^2 \int_0^\infty 2\pi R \left| \int_{-\xi}^{\epsilon} \frac{\exp(-R^2/bH)\,d\epsilon'}{(1+i\epsilon')H(k''-ik'\epsilon')} \right|^2 dR \ . \tag{7}$$

Using the relationship $d_0^2 = b\lambda/2\pi$ Eq. (7) can be written as

$$F = \frac{64}{\lambda^2}\left\{\frac{4}{\lambda b}\int_0^\infty 2\pi R \left| \int_{-\xi}^{\epsilon} \frac{\exp(-R^2/bH)\,d\epsilon'}{(1+i\xi')H(k''-ik'\epsilon')} \right|^2 dR\right\} , \tag{8}$$

where the expression above in brackets is just the function

$$F_2\left(b\Delta k, \frac{b}{L}, \frac{f}{L}, \frac{k''}{k'}\right)$$

as defined by Bjorklund. It is clear from Eq. (8) that a wavelength enhancement effect should be expected in CARS power generation with focused pump beams that is not anticipated by a plane wave analysis.

CALCULATIONS

The function F_2 has been evaluated as a function of the confocal parameter b over a range of focusing conditions, e.g., loose ($b/L \sim 1$) to very tight ($b/L \to 0$). The results are presented in Fig. 1 with the conclusion that the effect of tight focusing in CARS power generation with $\Delta k = 0$ is relatively minor, i.e., the generated power is increased by a factor of about 4 over loose focusing. However, this conclusion is somewhat misleading, as can be seen when one considers the confocal parameters associated with CARS experiments as they are commonly performed in the laboratory. For example, a commercially available YAG laser (e.g., Chromatix 1000 laser) has a confocal parameter of about 3.2×10^2 cm, which corresponds to a b/L value of 32 for a cell of 10 cm pathlength. The value of the function F_2 associated with this confocal parameter is about 1.34×10^{-3}. If the output beam from this laser is then focused with a 50 cm focal length lens, the resulting confocal parameter has a value of 31.25 cm, which gives a value of about 0.13 for the function F_2. The resultant focusing enhancement relative to the unfocused laser output is therefore on the order of about two orders of magnitude. If a 25 cm focal length lens is used, the enhancement relative to the unfocused beam is about a factor of 700, and with a 10 cm focal length lens the enhancement is more than three orders of magnitude. For shorter focal lengths than 10 cm the additional enhancement does not dramatically increase further, and thus for a typical CARS experiment a 10 cm focal length lens seems an ideal choice for optimizing the focusing enhancement.

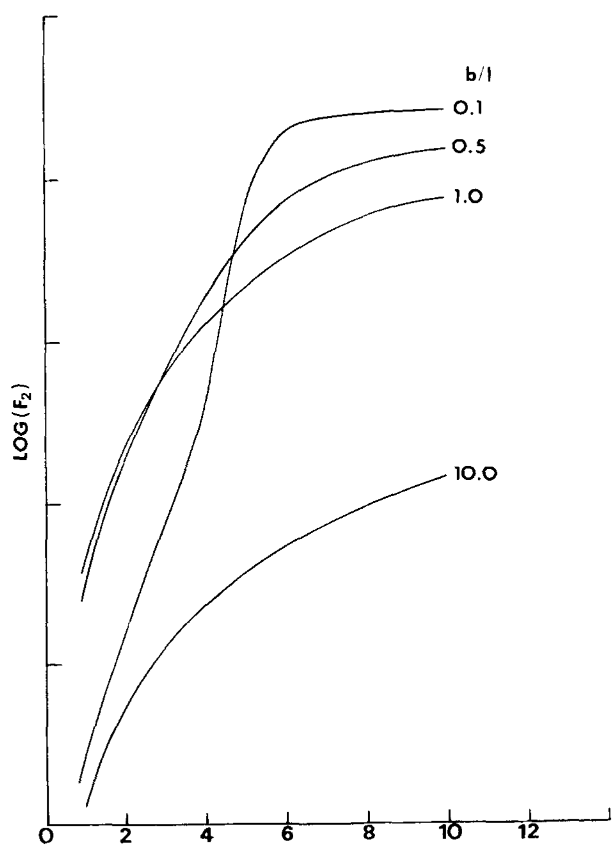

FIG. 2. Rigorous theory treatment. The $\log_{10}$ of the function F [Eq. (8)] is plotted for various values of b/L as a function of axial pathlength away from the input window. The focal point is at $X = 5$ cm.

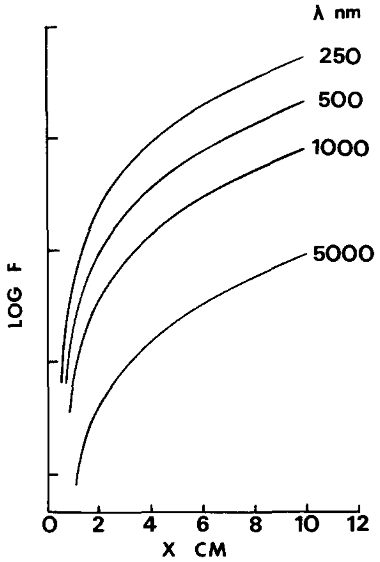

FIG. 3. Rigorous theory treatment. The $\log_{10}$ of the function F [Eq. (8)] is plotted for various values of λ for $b/L = 1.0$, as a function of axial pathlength away from the input window. The focal point is at $X = 5$ cm.

The development of the generated CARS power as a function of the distance from the input point of the pump beams to the medium is presented in Fig. 2 for various values of the ratio b/L. It is clear from Fig. 2 that power generation increases dramatically as one approaches the location of the minimum beam waist, consistent with the expectation that maximum power generation should occur about the region of highest field density. The wavelength enhancement effect is illustrated by evaluation of the function $F = F_2/\lambda^2$ for a fixed value of b/L as presented in Fig. 3. Here the conclusion is that a significant wavelength enhancement in CARS power generation is to be expected as the wavelength is changed from the infrared to the ultraviolet region of the electromagnetic spectrum.

CARS POWER-MODIFIED PLANE WAVE ANALYSIS

The plane wave differential gain expression[10] may be integrated to give (phase matched case)

$$E_3 = i\,\frac{3\pi\omega}{8cn}\,N\chi\int_0^z E_1^2(z)E_2(z)\,dz\,, \tag{9}$$

In the loose focusing limit $b/L \sim 1$ it is reasonable to assume Gaussian and plane wave field amplitudes are interchangeable. It should be stressed that this approximation must be carefully applied, as the following treatment neglects diffraction effects and hence is not applicable for tight focusing (i.e., $b/L \lesssim 1.0$). Then the field amplitudes may be written as

$$E_n(z) \sim \left(\frac{16\pi P_n}{c}\right)^{1/2}\left(\pi\frac{b}{k_n}\left\{1+\left[\frac{2(z-f)}{b}\right]^2\right\}\right)^{-1/2}. \tag{10}$$

On the assumption that (i) the laser and Stokes beams are axially concentric with identical beam waists, (ii) the pump beam power is essentially undepleted as a result of CARS power generation, and (iii) the focal point f is located at the center of a cell of length L, Eq. (9) may then be written as

$$E_3 \sim i\left(\frac{3\pi\omega_3 N\chi}{8cn}\right)\left(\frac{8\pi}{c}\right)^{3/2} P_1 P_2^{1/2}\int_0^z \frac{dz}{\pi\frac{b}{k}\left[1+\left(\frac{2(z-L/2)}{b}\right)^2\right]^{3/2}} \tag{11}$$

This integral may be evaluated in closed form to yield

$$E_3 \sim i\left(\frac{3\pi\omega_3 N\chi}{8cn}\right)\left(\frac{8\pi}{c}\right)^{3/2} P_1 P_2^{1/2}\left[\frac{b}{2}\left(\frac{k}{\pi b}\right)^{3/2}\right.$$

$$\left.\times\frac{z-L/2}{\sqrt{\left(\frac{b}{2}\right)^2+\left(z-\frac{L}{2}\right)^2}}+\frac{L/2}{\sqrt{\left(\frac{b}{2}\right)^2+\left(\frac{L}{2}\right)^2}}\right]. \tag{12}$$

If one uses the relations $I_3 = c|E_3|^2/8\pi$, $I_3 = P_3/A_3$, $A_i = (\pi b/k)\{1+[2(z-L/2)/b]^2$, and assumes A_3 is approximately linearly proportional to A_1 (an assumption which is verified by numerical analysis), the loose focusing expression for the CARS power may be written as

$$P_3 \propto C\left[\left(\frac{\omega}{\pi bc}\right)^2\left(\frac{b}{2}\right)^2+\left(z-\frac{L}{2}\right)^2\right]\left[\frac{(z-L/2)^2}{\sqrt{\left(\frac{b}{2}\right)^2+\left(z-\frac{L}{2}\right)^2}}+\frac{(L/2)^2}{\sqrt{\left(\frac{b}{2}\right)^2+\left(\frac{L}{2}\right)^2}}+\frac{2(z-L/2)(L/2)}{\sqrt{\left(\frac{b}{2}\right)^4+\left(\frac{b}{2}\right)^2\left[\left(z-\frac{L}{2}\right)^2+\left(\frac{L}{2}\right)^2+\left(z-\frac{L}{2}\right)^2\left(\frac{L}{2}\right)^2\right]}}\right] \tag{13}$$

where

$$C \equiv \left(\frac{3\pi^2\omega_3}{c^2 n}\right)^2 (N\chi)^2 P_1^2 P_2\,.$$

The function $f = P_3/C$ has been evaluated as a function of z for various cases of focusing as previously chosen by varying b/L. It is seen that the function f reproduces the qualitative behavior of the function F_2 very well for loose focusing ($b/L \gtrsim 1.0$). For tight focusing ($b/L < 1.0$) the method is not applicable, as can be seen by comparison of Figs. 2 and 4 and as expected, since as previously mentioned, diffraction effects are neglected in the approximation. In summary, the approximation should be useful for estimations of the axial power buildup to be expected with loosely focused beams. It also correctly predicts the additional λ^{-2} wavelength dependence in the CARS power generation expression. Note that this additional inverse squared wavelength dependence predicts the CARS efficiency using focused laser beams should vary with λ^{-4} just as in normal spontane-

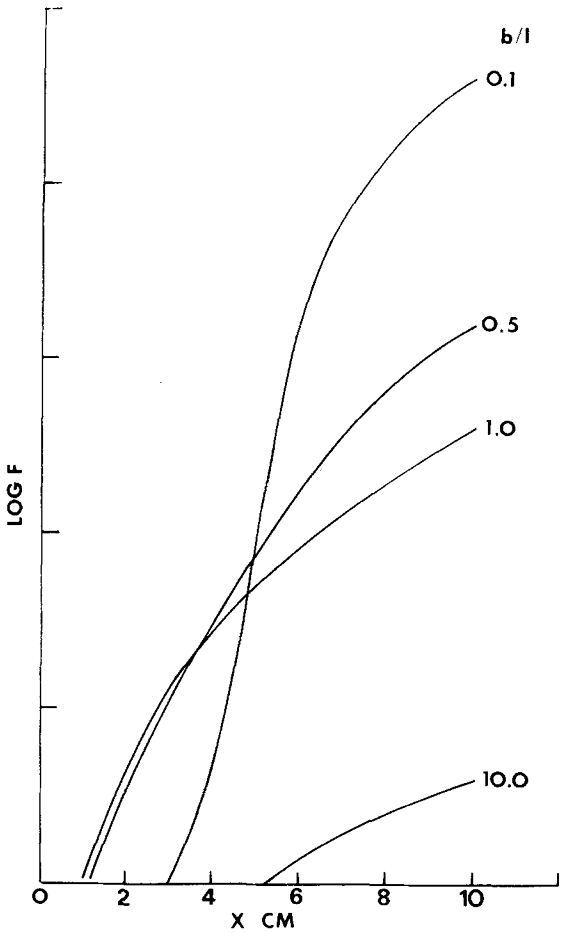

FIG. 4. Modified plane wave theory treatment. The function $f=P_3/C$ [see Eq. (13)] has been plotted for various values of b/L as a function of axial pathlength away from the input window. The focal point is at $X=5$ cm. Note the disagreement for $b/L=1.0$ compared to the rigorous theory treatment (Fig. 2) in which diffraction effects are included.

ous Raman scattering. Note that the modified plane wave treatment gives a more realistic fit to the case of loose focusing than does the plane wave treatment [Eq. (6)], which predicts a parabolic power increase along the axial cell length.

CONCLUSIONS

It has been shown (i) that for CARS power generation the effect of tightening the focus of the pump beams at fixed wavelength is minimal, (ii) there is an enhancement to be expected as one goes from long to shorter wavelengths, and (iii) a modified plane wave analysis is instructive for studying axial power generation, in the loose focus limit ($b/L>1.0$). It should be noted that the efficiency, whether computed assuming plane wave or Gaussian pump beams, is generally meaningful only for low conversion efficiencies, as the effects due to pump beam depletion have not been considered here. Additional loss terms due to thermal lensing, power saturation, beam quality, and other similarly important effects have not been included. They are beyond the scope or intent of this paper.

[1]R. F. Begley, A. B. Harvey, and R. L. Byer, Appl. Phys. Lett. 25, 387 (1974).
[2]R. F. Begley, A. B. Harvey, R. L. Byer, and B. S. Hudson, J. Chem. Phys. **61**, 2466 (1974).
[3]N. Bloembergen, *Non-Linear Optics* (Benjamin, New York, 1965).
[4]A. Yariv, *Quantum Electronics* (Wiley, New York, 1967).
[5]P. R. Regnier, and J-P. E. Taran, Appl. Phys. Lett. **23**, 240 (1973); P. R. Regnier and J-P. E. Taran, AIAA J. **12**, 826 (1974).
[6]G. Bjorklund, IEEE J. Quantum. Electron. **11**, 287 (1975).
[7]P. R. Regnier, Ph.D. thesis, Centre d'Orsay-University of Paris-Sud, 29 October, 1973, available as European Space Agency Technical Translation No. ESATT 200, November 1975.
[8]J. F. Ward and G. H. C. New, Phys. Rev. **185**, 57 (1969).
[9]G. D. Boyd and J. P. Gordon, Bell Syst. Tech. J. **40**, 489 (1961).
[10]R. W. DeWitt, A. B. Harvey, and W. N. Tolles, "Theoretical Development of Third Order Susceptibility as Related to CARS," NRL Memorandum Report No. 3260, Naval Research Laboratory, Washington, D. C. 20375 (1976).