RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 31, NUMBER 1 JANUARY 1985

Minimum power requirements for efficient four-wave mixing and self-focusing of electromagnetic beams in glasses and fluids

R. W. Hellwarth

Departments of Electrical Engineering and Physics, University of Southern California, Los Angeles, California 90089-0484

(Received 12 September 1984)

We derive general expressions for the efficiency of cw phase conjugation by degenerate four-wave mixing and for the threshold power P_{cr} for cw self-trapping of coherent electromagnetic beams in fluids and glasses in which these effects arise predominantly from driven nuclear motions (molecular vibrations, molecular reorientation, elastic deformations, etc.). Apart from their dependence on beam frequency ω, medium temperature T, and refractive index n, these expressions are functions only of the integrated (polarized and depolarized) light scattering strengths versus scattering angle in the medium and the attenuation coefficient α. There is no other dependence on the scattering mechanism. When attenuation is entirely due to scattering, the expressions simplify and suggest that unprecedented low powers (in the microwatt regime for microwaves) can produce self-focusing and strong phase conjugation, as well as other beam mixing effects, when beam geometries are optimized in the scattering medium. In this case we find, for example, $P_{cr} \sim nk_BT\omega^2/c\alpha$ (k_B is Boltzmann's constant and c is the velocity of light) provided that there is significant beam diffraction in length α^{-1}. Our results also apply to some other media such as electron plasma.

I. INTRODUCTION

The connection between spontaneous inelastic scattering of electromagnetic waves and coherent nonlinear propagation effects has long been exploited in nonlinear optics. Recently, Smith, Ashkin, and Tomlinson[1] demonstrated that a liquid suspension of 234-nm latex spheres in water, designed for high scattering ($\alpha \sim 15$ cm^{-1}), had a remarkably high nonlinear index ($\sim 10^5$ times that of CS_2) at the 515-nm argon laser wavelength. They observed cw phase-conjugation efficiency of order 0.5% using degenerate four-wave mixing with two 50-mW pump beams in the "artificial Kerr medium."[1] Ashkin, Dziedzic, and Smith[2] demonstrated cw self-focusing of 515-nm beams in less than the attenuation length α^{-1}(~ 2 mm) in similar suspensions, observing a threshold power of order 1 W, independent of beam diameter as is expected if the refractive index change equals a nonlinear index n_2 times the optical intensity I. They noted importantly that n_2 was proportional to the attenuation α cm^{-1} due to scattering.[2] Smith, Maloney, and Ashkin[3] estimated the constant of proportionality in terms of the difference between the refractive indices of a suspended particle and the liquid, finding satisfactory agreement with experiment. We generalize their expression in the course of this paper, in which we study the question: what limits, if any, are placed on the phase-conjugation efficiency and self-focusing thresholds of a fluid or glass that scatters light with arbitrary angular asymmetry and depolarization function, by virtue of the competition between the nonlinear coupling (proportional to the scattering strength) and the negative effects of the beam attenuation from the same scattering. We obtain a general expression for all components of the third-order nonlinear optical susceptibility tensor in terms of integrated (over frequency) scattering cross sections alone, independent of the scattering mechanism. Using this expression in standard treatments, we conclude that the same cw nonlinear effects will be found in any fluid or glass, having the same depolarized and polarized scattering coefficients $\sigma_\perp(\theta)$ and $\sigma_\parallel(\theta)$ cm^{-1}, regardless of the scattering mechanism, be it orientational or vibrational fluctuations, opalescence, concentration fluctuations in mixtures, aerosols,[4] etc. For example, efficient phase conjugation of 1-cm radiation is predicted with pump beam intensities less than a picowatt per square centimeter.

Here, we show that any transparent medium in thermal equilibrium at temperature T which attenuates an electromagnetic beam more by scattering than by absorption will phase conjugate a beam by degenerate four-wave mixing,[5] achieving power reflectivity R of order unity with relatively small pump powers. Specifically, we will show that, in such scattering with the usual beam configuration of Fig. 1, and with pump intensities I_1 and I_2 low enough so that R does not exceed ~ 0.5,[6]

$$R = \xi_1 \eta^2 I_1 I_2 / I_0^2 \ , \tag{1}$$

where ξ_1 is a dimensionless function of the degree of depolarization and anisotropy of scattering only, which we will show is of order unity when light scattering is isotropic (but possibly orders-of-magnitude larger for anisotropic scatter-

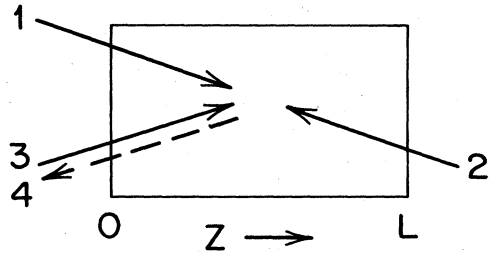

FIG. 1. Schematic of beam geometry for phase conjugation by degenerate four-wave mixing.

ing);

$$I_0 \equiv n^2 k_B T \omega^3 / c^2 \ , \tag{2}$$

where n is the refractive index, k_B Boltzmann's constant, ω the electromagnetic beam (angular) frequency, c the velocity of light in vacuum, and

$$\eta \equiv 3^{3/2} e^{-\alpha L/2}(1-e^{-\alpha L})/2 \tag{3}$$

achieves its maximum value of unity when the interaction length L in the scattering medium is equal to α^{-1} (i.e., one attenuation length), where α is the intensity attenuation coefficient for beams of frequency ω. This may be expressed as the sum of the parts α_d arising from absorption and σ arising from scattering:

$$\alpha = \alpha_d + \sigma \ . \tag{4}$$

Note that σ, sometimes called the total scattering cross section per unit volume, equals the fraction of photons scattered per unit length to photons which are generally of slightly different frequency. When the scattered photons have frequencies differing from ω by shifts of order $\pm\Delta$, the response time of the phase-conjugating mirror, or of the self-focusing, will be of order Δ^{-1}.

From (1) we expect to achieve efficient phase conjugation in scattering media with lower intensities at longer wavelengths. For example, (1) predicts pump intensities of order 10^{-4} W/cm^2 for strong conjugation of 10-μm radiation, and predicts intensities of order 10^{-13} W/cm^2 for conjugating 1-cm microwave radiation. Realization of media with convenient scattering lengths σ^{-1}, and having $\alpha_d < \sigma$, should be possible. For example, liquid chloroform has an absorption length ~ 6 mm at the carbon dioxide laser wavelength of 10.6 μm, and, being a polar liquid, should be capable of suspending enough micron-size rods (e.g., gold) to attenuate such a beam in less than 6 mm by scattering from orientational fluctuations. A convenient interaction length L of several millimeters or less in such a suspension should achieve phase conjugation with efficiencies approaching those in (1), at heating rates $2\alpha_d I_1$ much less than are met in dye lasers. The response time of such a medium will be the orientational relaxation time of the scattering rods, which should be, for example, of the order-of-magnitude of milliseconds for micron-size rods.[7]

We also show that, assuming the critical power P_{cr} for self-focusing of monochromatic linearly polarized radiation is given by $5.14\, c^2/(\omega^2 n n_2)$,[8] then

$$P_{cr} = \frac{1.24 n k_B T \omega^2 \xi_2}{c\sigma} \ , \tag{5}$$

where ξ_2 is a dimensionless function of scattering anisotropy and depolarization only, which equals unity for isotropic polarized scattering (the case studied in Refs. 1–3). From (5) we would expect for example, to produce self-focusing of X band (3 cm) microwaves from a 2-m-diam antenna, in such a medium having $\sigma = 5$ km^{-1}, with 14 μW of power.

II. THEORY

We will consider only fluids and glasses. These have the simplest, though nontrivial symmetry. To establish (1) for such media we first write the usual formula for the complex phase-conjugate-amplitude reflectivity r, which is proportional to $\eta\chi^{(3)}/\alpha$.[5] We then relate the appropriate third-order nonlinear susceptibility $\chi^{(3)}$ to the real scalar response functions $a(t, \vec{x})$ and $b(t, \vec{x})$ which describe nuclear motion in the Born-Oppenheimer regime in which $\chi^{(3)}$ is negligibly affected by electronic excitations. (Purely electronic nonlinearities contribute negligibly to $\chi^{(3)}$ in the regime of interest here.) It is well known that the a and b functions can be determined from the exponential "Raman" gain at frequency ω that is stimulated by a "pump" beam at frequency ν, if specified at all ω and for all relative beam directions and polarizations.[9] It is also well known that this Raman gain is linearly related to ordinary Raman scattering cross sections.[10] We will show that the nonlinear $\chi^{(3)}$ that governs cw phase conjugation is proportional to a weighted average $\bar{\sigma}$, depending on beam polarization, of ordinary inelastic scattering cross sections at angles θ and $\pi-\theta$. The "isotropic scattering" limit, where these cross sections depend only on beam polarization, and not otherwise on the scattering wave vector $\vec{k}$, has been treated previously.[9] Here, we will sometimes be interested in media caused to be highly scattering by particles having size of the order of a wavelength, for which we must extend the theory to anisotropic scattering. Since α is also proportional to σ in the media of interest here, the maximum magnitude of the reflectivity $r \propto \chi^{(3)}/\alpha$ does not depend on the scattering mechanism, and we find the simple limit of (1).

We consider the plane-wave description of phase conjugation by degenerate four-wave mixing depicted in Fig. 1 and assume the angle between beams 1 and 3 to be much less than one radian. The optical field $\mathscr{E}_i$ $(i = x,y,z)$ in the medium can be represented by

$$\mathscr{E}_i(\vec{x}t) = \mathrm{Re}\{e_{1i}E_1 \exp(i\vec{k}_1\cdot\vec{x} - \alpha z/2) + e_{2i}E_2 \exp[i\vec{k}_2\cdot\vec{x} - \alpha(L-z)/2] + e_{3i}E_3 \exp(i\vec{k}_3\cdot\vec{x} - \alpha z/2) + e_{4i}E_4 \exp(i\vec{k}_4\cdot\vec{x} + \alpha z/2)\} \ . \tag{6}$$

The complex polarization vectors (which must be proper vectors of the linear dielectric tensor) are normalized so that $e_{1i}^* e_{1i} = 1$, etc. (Repeated space indices are assumed to be summed throughout.) Using the usual convention, the component of the nonlinear polarization density $\mathscr{P}_4^{NL}(\vec{x},t)$ along vector e_{4i}, that generates the phase conjugate wave E_4 has complex amplitude

$$P_4 = 6\chi^{(3)} E_1 E_2 E_3^* \exp[i(\vec{k}_1 + \vec{k}_2 - \vec{k}_3)\cdot\vec{x} - \alpha(L+z)/2] \ , \tag{7}$$

where, in terms of the usual nonlinear susceptibility tensor c_{ijkl} (Ref. 11)

$$\chi^{(3)} = c_{ijkl}(-\omega, \omega, \omega, -\omega) e_{4i}^* e_{1j} e_{2k} e_{3l}^* \ . \tag{8}$$

In the absence of $\chi^{(3)}$, E_1, E_2, E_3, and E_4 are constants. With $\chi^{(3)}$, transient and steady-state (cw) solutions for these amplitudes have been found in a variety of approximations, with and without nonlinear components other than (7) which may be pertinent.[5] For our purposes it will suffice to

assume the conjugate wave E_4 disturbs the others negligibly, nor do they disturb each other. This approximation has been found to be accurate for power reflectivities $R=|r|^2=|E_4/E_3|^2$ up to ~ 0.5.[6] We also consider only the steady-state solution quantitatively, for which Maxwell's equations give directly[5]

$$r=-8\pi i\omega\chi^{(3)}E_1E_2\eta/(3^{1/2}nc\alpha) \ . \tag{9}$$

A useful expression of the part of the third-order polarization density $\mathscr{P}_i^{\rm NL}$ arising from nuclear motions in fluids and glasses is obtained by noting that it is the quantum average $\langle\chi_{ij}(\vec{x},t)\,\mathscr{E}_j(\vec{x},t)\rangle$ calculated to third order in $\mathscr{E}_i$ using the time-dependent interaction potential

$$-\tfrac{1}{2}\int d^3x'\,\mathscr{E}_i(\vec{x}',t)\chi_{ij}(\vec{x}',t)\,\mathscr{E}_j(\vec{x}',t)$$

(in esu). Clearly $\mathscr{P}_i^{\rm NL}(\vec{x},t)$ will have only terms parallel to $\mathscr{E}_i(\vec{x},t)$ and $\mathscr{E}(\vec{x}',t')(t'<t)$, the only vectors available. This average results in a $\mathscr{P}_i^{\rm NL}$ of the form

$$\mathscr{P}_i^{\rm NL}(\vec{x},t)=\int_{-\infty}^{\infty}dt'\int d^3x'[\,\mathscr{E}_i(\vec{x},t)\,\mathscr{E}_j^2(\vec{x}',t')a(t-t',\vec{x}-\vec{x}')+\mathscr{E}_i(\vec{x}',t')\,\mathscr{E}_j(\vec{x},t)\,\mathscr{E}_j(\vec{x}',t')b(t-t',\vec{x}-\vec{x}')] \ . \tag{10}$$

The functions a and b may be calculated directly from the quantum average, or its hydrodynamic or classical limits, in the tradition of Brillouin, Born, and Plazcek. For our purposes it is more useful to use (10) to obtain both an expression for $\chi^{(3)}$ and for the stimulated gain function, thereby linking $\chi^{(3)}$ to the spontaneous scattering functions. Inserting (6) in (10) and identifying P_4, one has, by comparing with (7), that

$$\begin{aligned}24\chi^{(3)}=&\ (42,13)[2A_0(13)+B_0(23)]\\&+(41,23)[2A_0(23)+B_0(13)]\\&+(43,12)[B_0(13)+B_0(23)] \ ,\end{aligned} \tag{11}$$

where $(42,13)\equiv e_{4i}^*e_{2i}e_{1j}e_{3j}^*$, etc., and $A_0(13)$ represents the Fourier transform

$$A_\Delta(\vec{k})\equiv\int_{-\infty}^{\infty}dt\int d^3x\,\exp(i\Delta t-i\vec{k}\cdot\vec{x})a(t,\vec{x}) \tag{12}$$

evaluated at $\Delta=0$ and $\vec{k}=\vec{k}_1-\vec{k}_3$. Similarly $B_0(23)\equiv B_0(\vec{k}_2-\vec{k}_3)$, etc.

In the regime of interest here, where the light scattering is predominantly to frequency shifts much smaller than both $k_BT/\hbar$ and the laser frequency ω itself, the coefficients in (11) are related to the total polarized and depolarized differential light scattering cross sections per unit volume $\sigma_\parallel(\theta)$ and $\sigma_\perp(\theta)$ at scattering angle θ (and its complement $\bar{\theta}\equiv\pi-\theta$) by the relation (A8) derived in the Appendix. Substituting (A8) in (11) gives

$$24\chi^{(3)}=c^4\bar{\sigma}/(\omega^4k_BT) \ , \tag{13}$$

where

$$\begin{aligned}\bar{\sigma}\equiv&\ (42,13)[\sigma_\parallel(\theta)-2\sigma_\perp(\theta)+\sigma_\perp(\bar{\theta})]\\&+(41,23)[\sigma_\parallel(\bar{\theta})-2\sigma_\perp(\bar{\theta})+\sigma_\perp(\theta)]\\&+(43,21)[\sigma_\perp(\theta)+\sigma_\perp(\bar{\theta})] \ .\end{aligned} \tag{14}$$

Using this result in (9) for the amplitude reflection r, and taking the absolute square to obtain the phase-conjugate power reflectivity R, gives the result (1) with

$$\xi_1=(8\pi^2\bar{\sigma}/\alpha)^2/27 \ . \tag{15}$$

Note, from the angle dependence (A3) of the scattering cross section, we have that the total scattering cross section per unit volume σ (the scattering attenuation coefficient) is related to the differential cross sections by the integral over solid angle Ω:

$$\sigma=\int d\Omega\,[\sigma_\perp(\theta)(1+\cos^2\phi)+\sigma_\parallel(\theta)\sin^2\phi] \ , \tag{16}$$

where ϕ is the angle between one possible incident (linear) polarization vector and the direction of scattering.

When the scattering particles are much smaller than a wavelength, the polarized and depolarized scattering cross sections (per unit volume per unit solid angle) $\sigma_\parallel$ and $\sigma_\perp$ are constants independent of scattering angle θ, and (16) gives the familiar expression $8\pi(\sigma_\parallel+2\sigma_\perp)/3$. When all beam polarizations are linear and parallel in this "isotropic" scattering limit, (15) gives ($\rho\equiv\sigma_\perp/\sigma_\parallel$)

$$\xi_1\rightarrow 3[6\pi/(9+18\rho)]^2 \ , \tag{17}$$

nearly two for the interesting case of scattering from orientational fluctuations in fluids, for which $\rho=\frac{3}{4}$.

The change in refractive index for a linearly polarized plane wave is easily seen from (10) to be n_2I where

$$n_2=8\pi^2[A_0(0)+B_0(0)]/n^2c \ . \tag{18}$$

With the relations (A8) this may be expressed as

$$n_2=\frac{3\sigma c^4}{8\xi_2nk_BT\omega^4} \ , \tag{19}$$

where the parameter

$$\xi_2=3\sigma/[8\pi\sigma_\parallel(0)] \ , \tag{20}$$

which is a function of scattering depolarization and anisotropy only, becomes unity for isotropic polarized scattering, the case studied in Refs. 1–3. The exact expressions (19) and (20) reduce to the approximate expression (4) of Ref. 3 for a suspension of small spherical particles, in the limit where ξ_2 approaches unity and the refractive index of the particles is close to that of the suspending liquid. It is (19) and (20) that give directly our result (5) for the threshold power for self-focusing or self-trapping of a Gaussian beam when the attenuation length α^{-1} is much larger than the distance in which normal diffraction becomes significant.

We note that any nonlinear effect in transparent fluids or glasses that arises mainly from nuclear motions and can be described with the nonlinear susceptibility tensor $c_{ijkl}(-\omega,\omega,\omega,-\omega)$, can be predicted from light scattering data by using (A8) to evaluate the nonlinear polarization density (10). To obtain the time dependence of such effects, the details of the frequency dependence of the light scattering must also be known. Saturation effects (i.e., $\chi^{(n)}$ for $n>3$) cannot be obtained from light scattering and must be considered separately for each material.

There are other classes of scattering media, such as an electron plasma, or a multicomponent plasma of charged

particles, for which the nonlinear polarization density is approximately of the form (10). Since the relation (A1) does not depend on the type of excitations responsible for the scattering, the results we have derived here apply to these other media as well.

In conclusion, we have derived expressions (1) for the efficiency of cw phase conjugation by degenerate four-wave mixing, and (5) for the threshold of cw self-trapping of beams in a fluid or glass, in which electronic transitions and nonlinearities have no effect on light propagation (and also in certain plasma), and by which light is scattered with arbitrary function of scattering angle and state of scattered polarization. The results suggest that these and other nonlinear effects can be produced with unprecedented low powers with proper beam geometry in any medium in which beam attenuation arises more from scattering than from absorption.

ACKNOWLEDGMENTS

We would like to acknowledge the support of the Air Force Office of Scientific Research through Contract No. F49620-83-C-0045 and of the National Science Foundation under Grant No. ECS-8114754.

APPENDIX

When a material in thermal equilibrium is transparent enough so that there is a well-defined differential scattering cross section $d^2\sigma_{\alpha\beta}/d\Omega\, d\omega$ [per unit volume per unit solid angle per (angular) frequency interval] to scatter a photon from proper polarization state $e_{\alpha i}$ and wave vector $\vec{k}_\alpha$ to proper state $e_{\beta i}$ and wave vector $\vec{k}_\beta$, then[10]

$$\frac{d^2\sigma_{\alpha\beta}}{d\Omega\, d\omega_\beta}=\frac{g_{\alpha\beta}\hbar\omega_\alpha\omega_\beta^2 n_\alpha n_\beta}{I_\alpha 8\pi^3 c^2\{1-\exp[-\hbar(\omega_\alpha-\omega_\beta)/k_BT]\}} \quad , \tag{A1}$$

where $g_{\alpha\beta}$ is the stimulated exponential gain per unit length experienced by the β beam due to the presence of intensity I_α in the α beam. $|\vec{k}_\alpha|=n_\alpha\omega/c$, etc., $\hbar=h/2\pi$.

If one uses Eq. (10) to calculate the stimulated gain $g_{\alpha\beta}$ in a fluid or glass, one finds[12] ($I_\alpha=n_\alpha c|E_\alpha|^2/8\pi$)

$$g_{\alpha\beta}=\mathrm{Im}\pi\omega_\beta D_\Delta(\alpha\beta)|E_\alpha|^2 n_\beta c \quad , \tag{A2}$$

where

$$D_\Delta(\alpha\beta)\equiv 2A_\Delta(\alpha\beta)|e_{i\beta}^* e_{i\alpha}|^2+B_\Delta(\alpha\beta)(1+|e_{i\beta}e_{i\alpha}|^2) \quad . \tag{A3}$$

Here, $\Delta\equiv\omega_\alpha-\omega_\beta$ is the Stokes shift and the argument $(\alpha\beta)$ signifies $\vec{k}=\vec{k}_\alpha-\vec{k}_\beta$. Substituting (A2) in (A1) gives for the Born-Oppenheimer regime in fluids and glasses

$$\frac{d^2\sigma_{\alpha\beta}}{d\Omega\, d\omega_\beta}=\frac{\hbar\omega_\alpha\omega_\beta^3\,\mathrm{Im}D_\Delta(\alpha\beta)}{\pi c^4[1-\exp(-\hbar\Delta/k_BT)]} \quad . \tag{A4}$$

The usual integrated differential polarized scattering cross section is defined for scattering angle θ in optically inactive media by

$$\sigma_{\parallel}(\theta)=\int_0^\infty d\omega_\beta(d^2\sigma_{\alpha\alpha}/d\Omega\, d\omega_\beta) \quad , \tag{A5}$$

in which both incident and scattered polarizations are "π," i.e., perpendicular to the scattering plane. The depolarized cross section $\sigma_\perp(\theta)$ is defined similarly, with one polarization perpendicular to, and the other lying in, the scattering plane. Note that, for optically active media, one may need to employ the cross sections for right-to-right and right-to-left circularly polarized beams; this was one motivation for writing (A1) to (A4) for arbitrary proper polarizations.

From (A3) we see that, for optically inactive fluids and glasses,

$$\sigma_\perp(\theta)=\int_{-\infty}^{\omega_\alpha}\frac{d\Delta\hbar\omega_\alpha\omega_\beta^3\,\mathrm{Im}B_\Delta(\vec{k}_\Delta)}{\pi c^4[1-\exp(-\hbar\Delta/k_BT)]} \quad , \tag{A6}$$

where we have recognized that $\vec{k}$ is, in general, a function of the scattered frequency ω at fixed scattering angle θ. We can neglect this dependence because all shifts Δ of interest here are less than $\theta\omega$. Since $B_\Delta(\vec{k})$ is a function only of the magnitude of $\vec{k}$ we can therefore use $B_\Delta(k_0)$ in (A6), where $k_0\equiv|\vec{k}_\alpha-\vec{k}_\beta|_{\Delta=0}$.

Strongly scattering media of interest here also scatter mainly to shifts Δ much less than both $k_BT/\hbar$ and ω. Since the Kramers-Kronig relation between the real and imaginary parts of A_Δ (and B_Δ) gives ($\mathrm{Re}A_0=A_0$)

$$A_0=\int_{-\infty}^{\infty}\mathrm{Im}A_\Delta d\Delta/\pi\Delta \quad , \tag{A7}$$

and similarly for B_0, we have from (A6) that ($\omega\equiv\omega_\alpha$)

$$\sigma_\perp(\theta)\cong\omega^4 k_BTB_0(k_0)/c^4 \quad . \tag{A8}$$

The relation (A8) also holds for $\sigma_\parallel(\theta)$ but with $B_0(k_0)$ replaced by $2[A_0(k_0)+B_0(k_0)]$. These are the desired relations between the differential scattering cross sections and the nonlinear coefficients in (11) which we need to write the important relations (13) and (19).

[1]P. W. Smith, A. Ashkin, and W. J. Tomlinson, Opt. Lett. **6**, 284 (1981).

[2]A. Ashkin, J. M. Dziedzic, and P. W. Smith, Opt. Lett. **7**, 276 (1982).

[3]P. W. Smith, P. J. Maloney, and A. Ashkin, Opt. Lett. **7**, 347 (1982).

[4]A. J. Palmer, Opt. Lett. **5**, 54 (1980).

[5]*Optical Beam Phase Conjugation,* edited by R. A. Fisher (Academic, New York, 1983).

[6]Generalizations of Eq. (1) to include pump beam depletion exist. These predict, for example, gain ($R>1$) as has been observed in a variety of systems (see Ref. 5). However, no general treatment exists that takes into account self-focusing, optical Kerr effect and the other nonlinear effects expected to become strong when $R>1$.

[7]F. Perrin, J. Phys. Radium **7**, 1 (1956).

[8]R. Y. Chiao, E. Garmire, and C. H. Townes, Phys. Rev. Lett. **2**, 138 (1965); *ibid.* **14**, 1056(E) (1965).

[9]R. W. Hellwarth, J. Cherlow, and T. T. Yang, Phys. Rev. B **11**, 964 (1975).

[10]R. W. Hellwarth, Phys. Rev. **130**, 1850 (1963).

[11]P. D. Maker and R. W. Terhune, Phys. Rev. **137**, A801 (1965).

[12]R. W. Hellwarth, *Progress in Quantum Electrons* (Pergamon, New York, 1977), Vol. 5, Pt. I.