Amplifiers involving two-photon energy-extraction schemes*

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Amplifiers based on two-photon decay channels of inverted metastable species are examined from a rate-equation and practical-feasibility viewpoint. Approximate analytical solutions, within the rate-equation approximation, predict that resonant parametric generation coupled with anti-Stokes–stimulated Raman scattering (ASRS) is a strong competitor to straight two-photon emission. Expressions for initial growth rates indicate that large linear chromatic dispersion, proper linear absorptions, and resonant enhancements can initially favor two-photon emission, but that ASRS will ultimately dominate in most practical situations if inversion depletion does not occur early. For an amplifier based on degenerate two-photon processes, this situation has led to the proposal of running the system as an odd-harmonic generator, first extracting energy via two-photon emission followed by greater-than-100% conversion to the third, fifth, seventh, etc., harmonics. This type of amplifier response has important potential applications for laser-induced thermonuclear fusion as well as for the production of coherent vacuum-uv soft-x-ray systems. An examination of practical constraints provides further analytical relationships between various physical properties of prospective metastable species. Combining these results, several two-photon schemes are pointed out. Based on current technology, atomic iodine, which is inverted to the $^2P_{1/2}$ state, appears to be the best medium for experiments, but other materials, such as atomic oxygen, show greater promise if absolute population inversions of high-density material are created at high efficiency.

I. INTRODUCTION

Quantum amplifiers based on two-photon processes were discussed very early in the development of nonlinear optics,[1] and the first discussions of multiple-quantum transitions predate the laser by about thirty years.[2]

Despite this early history, the development of practical amplifiers utilizing multiple-quantum transitions has been slow. Only the possibility of laser-induced thermonuclear fusion, and its requirement for very large lasers, has recently increased the interest in amplifiers based on multiple-quantum transitions. This interest stems from several aspects of two-photon systems. First, one can naively compare two-photon emission to single-photon emission by considering the former as an emission process with an instantaneous intensity-dependent cross section, as opposed to the more usual intensity-independent cross section of the latter process. Thus, for single-photon emission, the saturation energy, the maximum stored energy, and the stimulated gain coefficient of the amplifier are related through the cross section, while for two-photon emission the corresponding quantities are decoupled. Furthermore, because of the intrinsic nonlinearity of the amplification process, the output pulse can be tailored in both time and space. Because amplification usually takes place at a frequency different from that of the input driving laser, many options for target isolation are now available that did not exist before.[3] As an additional advantage, several different methods can now be imagined for producing frequency-modulated pulses (pulses whose center frequency changes simultaneously with its time-evolution; frequency tailoring), where the frequency change of interest is toward the blue for increasing time, occurs either continuously or stepwise, or possibly assumes an even more complicated form. Such a pulse, if it could sweep from the infrared to the ultraviolet, should have a very important impact on the successful demonstration of laser-induced thermonuclear fusion, because many implosion symmetry problems can be alleviated[4] by initiating and preliminarily heating the plasma with infrared light. However, a progressive increase in laser frequency during hydrodynamic shock and adiabatic compression of the target-implosion cycle implies that laser energy should be deposited closer and closer to the supersolidly dense central core, minimizing the difficulties of energy transport and of decoupling the corona from the center of the pellet,[5,6] as well as minimizing energy losses due to superthermal electrons and ions.[6]

We will consider two somewhat different but related two-quantum decay channels: (a) two-photon emission (TPE), and (b) anti-Stokes–stimulated Raman scattering (ASRS). The two processes are

schematically shown in Fig. 1. Both are of the same order of nonlinearity, involving a polarization that is dependent on the cube of the optical electric fields, within the perturbation-expansion limit for nonlinear polarization.[7] To date, both spontaneous[8] and enhanced[9] two-photon emission have been predicted and observed, whereas stimulated two-photon emission has not been demonstrated to our knowledge. Likewise, both spontaneous[10] and stimulated[11] anti-Stokes Raman scattering have been reported.

We shall take two different approaches toward examining two-photon amplifiers. First, we will be dealing with the propagation and pulse evolution in such an amplifier during both the initial growth phase and through the unsaturated-gain, pulse-distortion phase of amplification. Of particular concern will be the competition between TPE and ASRS and the effect of intermediate states on this competition. This concern exists because Manley-Rowe energy-balance considerations dictate that an amplifier operating purely in the ASRS mode has an energy gain limited to ω_{AS}/ω_L, whereas a similar amplifier based purely on TPE is limited in gain only by the amount of energy stored. Furthermore, once ASRS dominates, the input laser field decays and thereby further diminishes the probability for TPE. This decline in probability is to be contrasted with TPE that amplifies both the input laser field and the complementary field, leading to the possibility of later domination of ASRS.

The second approach consists of formulating a series of constraint equations in terms of phenomenological parameters and then combining these constraints to arrive at bounds on the experimentally adjustable parameters, written in terms of purely material parameters. We thus find some useful scaling conditions for evaluating prospective amplification schemes and materials that could be used as the active media. Several promising materials will be compared.

Note that systems which display large cross sections for two-quantum decay generally involve metastable states and thus do not decay readily via the single-photon radiative channel. Therefore, there is less tendency for these systems to amplify light of low intensity at any frequency, and more tendency for resistance to superfluorescent depumping. Intrinsically, we should thus be able to store more energy per unit volume and to use the input-driving laser as a trigger for the release of this energy. These systems are frequently limited by nonradiative decay channels such as collisional deactivation or energy-exchange processes, and these channel rates should thus be minimized. Furthermore, because light in the visible is desired, we are, of necessity, dealing with electronic or vibrational-electronic transitions. As a final comment, because the light intensities within the amplifying media must be high, other nonlinear modes of response, e.g., of vibrational, rotational, or Stark-effect origin, must be minimized.

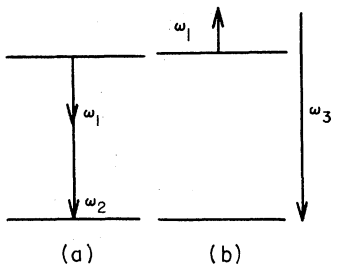

FIG. 1. Two two-quantum decay channels considered in this paper: (a) two-photon emission, and (b) anti-Stokes-stimulated Raman scattering. Here ω_1 refers to the input laser, ω_2 refers to the complementary two-photon emission, and ω_3 is the anti-Stokes–Raman frequency.

Finally, we will propose a method of operating a two-quantum amplifier in which the pulse from a driving laser is first amplified at its original frequency and then continues to be amplified while shifting the pulse-center frequency sequentially through several odd harmonics. Such an amplification system can not only affect the laser-induced fusion problem, but, if optimized, may well represent a means of efficiently converting available lasers into high-power coherent vacuum ultraviolet and soft x-ray sources.

II. PULSE-EVOLUTION RATE EQUATIONS

We begin by writing down the material and field equations for the simplest case, namely, a two-level system that is relatively far removed from all intermediate states. Further, we assume that single-photon radiative decay rates can be neglected. Therefore, the second-order Bloch equations describe, within perturbation theory,[12] the material system in which the general third-order susceptibility α is used. Maxwell's equations describe the evolution of the fields:

$$\frac{\partial \Delta}{\partial t} + \Gamma'(1+\Delta) + \gamma_{\text{pump}}(t) = \frac{1}{\hbar\omega_0}\,\alpha \mathcal{E}^2 \frac{\partial Q}{\partial t}\ , \tag{1a}$$

$$\frac{\partial^2 Q}{\partial t^2} + \Gamma \frac{\partial Q}{\partial t} + \omega_0^2 Q = -\tfrac{1}{2}\alpha \mathcal{E}^2 \Delta, \tag{1b}$$

$$\nabla^2 \mathcal{E} - \frac{\epsilon}{c^2}\frac{\partial^2 \mathcal{E}}{\partial t^2} + 2\beta \frac{\partial \mathcal{E}}{\partial z} = \frac{4\pi}{c^2}\frac{\partial^2 P^{\text{NL}}}{\partial t^2}\ , \tag{1c}$$

where P^{NL} is the nonlinear polarizability and is related to α by the usual relationship $P^{NL} = N\alpha\mathcal{E}Q$. The term Q is the amplitude of the driven excitation, which for the case of vibrational Raman scattering corresponds to the amplitude of molecular vibration. Also, $\Delta \equiv \Psi_1\Psi_1^* - \Psi_0\Psi_0^*$, where Ψ_0 and Ψ_1 are the probability amplitudes describing system occupations in the ground state and in the electronically or vibration-electronically excited state, respectively. If N is the density of atoms in the amplifying medium, then $N\Delta$ represents the difference in the population densities. Finally, Γ and Γ' are assumed to be the reciprocals of T_2 and T_1 (normal Bloch notation), respectively, whereas β and γ are the optical linear-absorption coefficient and the time-dependent external pumping function, respectively.

In the usual manner,[13] the fields are expressed in terms of slowly varying complex amplitudes by

$$\mathcal{E}_L = E_1(z,t)e^{ik_1 z - i\omega_1 t}\ , \tag{2a}$$

$$\mathcal{E}_{AS1} = E_3(z,t)e^{ik_3 z - i\omega_3 t}\ , \tag{2b}$$

$$\mathcal{E}_{TPE} = E_2(z,t)e^{ik_2 z - i\omega_2 t}\ , \tag{2c}$$

$$\mathcal{E}_{AS2} = E_4(z,t)e^{ik_4 z - i\omega_4 t}\ , \tag{2d}$$

$$Q = q(z,t)e^{ik_0 z - i\omega_0 t}\ , \tag{2e}$$

where the wave vectors and frequencies are chosen to satisfy conditions corresponding to the conservation of energy and momentum, respectively,

$$\omega_1 + \omega_2 = \omega_0,\quad k_1 + k_2 = k_0\ , \tag{3a}$$

$$\omega_1 + \omega_0 = \omega_3,\quad k_1 + k_0 = k_3\ , \tag{3b}$$

$$\omega_2 + \omega_0 = \omega_4,\quad k_2 + k_0 = k_4\ , \tag{3c}$$

where $\hbar\omega_0$ is the energy gap between the relevant material states. The usual dispersion relations of the electromagnetic waves for a linear medium further relate k_i and ω_i, namely, $k_i = n_i\omega_i/c$. We are assuming that a strong field, called the input-pump laser, at frequency ω_1 stimulates TPE at ω_1 and ω_2 as well as ASRS at ω_3. The buildup of ω_2 then further leads to ASRS, being stimulated at ω_4. We implicitly ignore any higher-order ASRS where $\omega = \omega_{1,2} + n\omega_0$, and n is a positive integer greater than one.

To take into account the possible enhancement of either TPE or ASRS at one of the frequencies due to a partially resonant intermediate state, we will generalize Eq. (1) slightly by letting α_{ij} represent the magnitude of α corresponding to each pair of photons involved in the specific coupling terms. Combining these features with the above, we arrive at the following set of coupled first-order equations for the complex amplitudes:

$$\frac{1}{2ik_1}\nabla_T^2 E_1 + \frac{\partial E_1}{\partial z} + \frac{n_1}{c}\frac{\partial E_1}{\partial t} + \beta_1 E_1 = \frac{2\pi i N\omega_1^2}{c^2 k_1}(\alpha_{21} q E_2^* + \alpha_{31} E_3 q^*)\ , \tag{4a}$$

$$\frac{1}{2ik_2}\nabla_T^2 E_2 + \frac{\partial E_2}{\partial z} + \frac{n_2}{c}\frac{\partial E_2}{\partial t} + \beta_2 E_2 = \frac{2\pi i N\omega_2^2}{c^2 k_2}(\alpha_{21} q E_1^* + \alpha_{24} q^* E_4)\ , \tag{4b}$$

$$\frac{1}{2ik_3}\nabla_T^2 E_3 + \frac{\partial E_3}{\partial z} + \frac{n_3}{c}\frac{\partial E_3}{\partial t} + \beta_3 E_3 = \frac{2\pi i N}{c^2 k_3}\alpha_{31}\omega_3^2 q E_1\ , \tag{4c}$$

$$\frac{1}{2ik_4}\nabla_T^2 E_4 + \frac{\partial E_4}{\partial z} + \frac{n_4}{c}\frac{\partial E_4}{\partial t} + \beta_4 E_4 = \frac{2\pi i N}{c^2 k_4}\alpha_{24}\omega_4^2 q E_2\ , \tag{4d}$$

$$\frac{\partial q}{\partial t} + \frac{\Gamma}{2}q = \frac{i\Delta}{4\omega_0}(\alpha_{21}E_1E_2 + \alpha_{24}E_4E_2^* + \alpha_{31}E_3E_1^*)\ , \tag{4e}$$

$$\frac{\partial \Delta}{\partial t} + (\Gamma' - \gamma) = -\Gamma'\Delta - \frac{1}{\hbar\Gamma'}\mathrm{Im}(\alpha_{21}E_1E_2q^* + \alpha_{24}E_4E_2^*q^* + \alpha_{31}E_3E_1^*q^*)\ . \tag{4f}$$

To simplify Eqs. (4), a change of variables is made which introduces more physically significant parameters in terms of stimulated emission, while simultaneously converting the variables to dimensionless units:

$$\begin{aligned} &g_R \equiv 8\pi^2 N\alpha_{31}^2\omega_1^2/c^2\omega_0 k_1\Gamma,\\ &\mathcal{E}_i \equiv (c/8\pi)^{1/2}(2k_1a^2g_R)^{1/2}E_i\ ,\\ &z^* \equiv z/2k_1a^2,\quad t^* \equiv ct/2k_1a^2n_1\ ,\\ &\nabla_T'^2 \equiv a^2\nabla_T^2,\quad \bar{\beta}_i \equiv 2k_1a^2\beta_i\ ,\\ &S = 4n_1/\hbar\omega_1(4k_1^2a^4)g_R N\Gamma'\ . \end{aligned} \tag{5}$$

The significance of these parameters is easily recognized. The term g_R is the steady-state stimulated ASRS gain coefficient in cm/W, scaled to an anti-Stokes frequency and to the k vector of the input-pump laser; $\mathcal{E}_1$ is then the square root of the laser intensity, normalized by a diffraction length and the gain coefficient to be dimensionless. Therefore, the field $\mathcal{E}_1$ becomes the square root of the log of the steady-state ASRS gain, scaled to ω_1, k_1, which occurs over one Rayleigh range. Because in ASRS the diffraction of the pump light to twice its initial diameter reduces the log of the gain by 4, we observe essentially no gain beyond

the distance $2k_1a^2$, where a is the beam radius of the input pump laser. Hence, $\mathcal{E}_1$ is further the square root of the maximum achievable unsaturated exponential gain coefficient for ASRS. The term z^* denotes the distance propagated down the amplifying medium normalized to the pump diffraction length, whereas t^* is similarly made dimensionless. Further, the linear absorption coefficient β is normalized to a pump diffraction length, and transverse derivatives ∇_T^2 are converted to be dimensionless by multiplying this term with the square of the pump-beam radius. Finally, we introduce S as a dimensionless saturation parameter whose significance will become clear shortly.

We are introducing the steady-state ASRS parameters because we will restrict ourselves to steady-state conditions. Steady state can be expressed in two ways: In Eq. (4e), steady-state implies that $\partial q/\partial t \ll \frac{1}{2}\Gamma q$, where q is a time-independent complex amplitude. Correspondingly, the duration of the laser-pump pulse τ_p is sufficiently long for q to become time independent. Mathematically,[14] this can be stated as

$$\tau_p > |\mathcal{E}_1|^2 z^* T_2 = (\ln G_{AS})/\Gamma . \tag{6}$$

Under these conditions, the magnitude of q is

$$q = \frac{i\Delta}{2\Gamma\omega_0}\left(\alpha_{21}E_1E_2 + \alpha_{24}E_4E_2^* + \alpha_{31}E_3E_1^*\right) . \tag{7}$$

Substituting Eqs. (5) and (7) into Eqs. (4), we arrive at the rate equations for the normalized electromagnetic fields and at the normalized population difference equation below:

$$\frac{\partial \mathcal{E}_1}{\partial z^*} + \frac{\partial \mathcal{E}_1}{\partial t^*} + \bar{\beta}_1 \mathcal{E}_1 - i\nabla_T'^2 \mathcal{E}_1 = \Delta\left\{\left[\left(\frac{\alpha_{21}}{\alpha_{31}}\right)^2 |\mathcal{E}_2|^2 - |\mathcal{E}_3|^2\right]\mathcal{E}_1 + \frac{\alpha_{21}\alpha_{24}}{\alpha_{31}^2}\mathcal{E}_2^{*2}\mathcal{E}_4 e^{i\varphi_{4221}z^*} - \frac{\alpha_{24}}{\alpha_{31}}\mathcal{E}_4^*\mathcal{E}_3\mathcal{E}_2 e^{-i\varphi_{4123}z^*}\right\}, \tag{8a}$$

$$\left(\frac{\partial \mathcal{E}_2}{\partial z^*} + \frac{n_2}{n_1}\frac{\partial \mathcal{E}_2}{\partial t^*} + \bar{\beta}_2\mathcal{E}_2\right)\frac{k_2}{k_1} - i\nabla_T'^2\mathcal{E}_2 = \frac{\Delta\omega_2^2}{\omega_1^2}\left\{\left[\left(\frac{\alpha_{21}}{\alpha_{31}}\right)^2 |\mathcal{E}_1|^2 - \left(\frac{\alpha_{24}}{\alpha_{31}}\right)^2 |\mathcal{E}_4|^2\right]\mathcal{E}_2 + \left(\frac{\alpha_{21}}{\alpha_{31}}\right)\mathcal{E}_3\mathcal{E}_1^{*2}e^{i\varphi_{3211}z^*} - \left(\frac{\alpha_{24}}{\alpha_{31}}\right)\mathcal{E}_3^*\mathcal{E}_1\mathcal{E}_4 e^{i\varphi_{4123}z^*}\right\}, \tag{8b}$$

$$\left(\frac{\partial \mathcal{E}_3}{\partial z^*} + \frac{n_3}{n_1}\frac{\partial \mathcal{E}_3}{\partial t^*} + \bar{\beta}_3\mathcal{E}_3\right)\frac{k_3}{k_1} - i\nabla_T'^2\mathcal{E}_3 = \frac{\Delta\omega_3^2}{\omega_1^2}\left[|\mathcal{E}_1|^2\mathcal{E}_3 + \left(\frac{\alpha_{21}}{\alpha_{31}}\right)\mathcal{E}_1^2\mathcal{E}_2 e^{-i\varphi_{3211}z^*} + \left(\frac{\alpha_{24}}{\alpha_{31}}\right)\mathcal{E}_4\mathcal{E}_1\mathcal{E}_2^* e^{i\varphi_{4123}z^*}\right], \tag{8c}$$

$$\left[\frac{\partial \mathcal{E}_4}{\partial z^*} + \left(\frac{n_4}{n_1}\right)\frac{\partial \mathcal{E}_4}{\partial t^*} + \bar{\beta}_4\mathcal{E}_4\right]\frac{k_4}{k_1} - i\nabla_T'^2\mathcal{E}_4 = \frac{\Delta\omega_4^2}{\omega_1^2}\left[\left(\frac{\alpha_{24}}{\alpha_{31}}\right)^2 |\mathcal{E}_2|^2\mathcal{E}_4 + \left(\frac{\alpha_{24}}{\alpha_{31}}\right)\mathcal{E}_1^*\mathcal{E}_2\mathcal{E}_3 e^{-i\varphi_{4123}z^*} + \frac{\alpha_{21}\alpha_{24}}{\alpha_{31}^2}\mathcal{E}_2^2\mathcal{E}_1 e^{i\varphi_{4221}z^*}\right], \tag{8d}$$

$$\frac{c}{2k_1a^2\Gamma' n_1}\frac{\partial \Delta}{\partial t^*} = \frac{\gamma}{\Gamma'} - 1 - \Delta\left(1 + S\left|\mathcal{E}_3\mathcal{E}_1^* + \frac{\alpha_{24}}{\alpha_{31}}\mathcal{E}_4\mathcal{E}_2^* + \frac{\alpha_{21}}{\alpha_{31}}\mathcal{E}_1\mathcal{E}_2\right|^2\right). \tag{8e}$$

The term n_i in Eq. (8) represents the refractive index at frequency ω_1, and the φ's take into account the phase mismatching in the various parametric processes due to the linear chromatic dispersion of the nonlinear medium, and are defined as follows:

$$\varphi_{3211} \equiv \frac{(n_3\omega_3 - n_2\omega_2 - 2n_1\omega_1)}{c}(2k_1a^2), \tag{9a}$$

$$\varphi_{4221} \equiv \frac{(n_4\omega_4 - 2n_2\omega_2 - n_1\omega_1)}{c}(2k_1a^2), \tag{9b}$$

$$\varphi_{4123} \equiv \frac{(n_4\omega_4 + n_1\omega_1 - n_2\omega_2 - n_3\omega_3)}{c}(2k_1a^2) . \tag{9c}$$

Equations (8) will be sufficiently general to encompass almost all cases of interest for the rest of this paper.

III. INITIAL GROWTH OF OPTICAL FIELDS

By using the rate equations derived in the previous section, we can establish which waves will grow first. This is significant because, for ASRS, the energy gain, $G_E = \omega_{AS}/\omega_L = \omega_3/\omega_1$, whereas G_E for TPE is limited only by energy storage, as mentioned in the Introduction. Also, because the waves at ω_1 decrease as ω_3 grows, ASRS eliminates any possibility of subsequent TPE, while the reverse is not true. Hence, for a high-gain system, we shall desire that TPE be optimized initially. This will lead us to several kinds of boundary conditions for the fields and for the linear chromatic dispersion of the medium, as well as for the effect of resonant enhancements.

Starting from Eq. (8), we first assume that we are not changing the inversion significantly and

that the system is initially totally inverted, or $\Delta \equiv 1$ and $\partial\Delta/\partial t^* = 0$. Next, we assume $|\mathcal{E}_1| > |\mathcal{E}_2| \gg |\mathcal{E}_3|, |\mathcal{E}_4|$, and that $\mathcal{E}_1 = \mathcal{E}_i' e^{i\varphi_i}$. We are then led to a set of approximate equations which describe the initial growth, similar to those for normal Raman scattering.[15] For $\psi = (\varphi_2 + 2\varphi_1 - \varphi_3)$ and $\zeta = (2\varphi_2 + \varphi_1 - \varphi_4)$, the phase equations can be approximately solved immediately, namely, φ_1 = constant and φ_2 = constant, which implies

$$\psi = 2\tan^{-1}(e^{-\delta z^*}), \quad \text{for } \varphi_{3211} z^* \ll \psi,$$

or

$$\psi = -(\delta/\varphi_{3211})\cos(\varphi_{3211} z^*), \quad \text{for } \varphi_{3211} z^* \gg \psi,$$

while

$$\zeta = 2\tan^{-1}(e^{-\xi z^*}), \quad \text{for } \varphi_{4221} z^* \ll \zeta,$$

or

$$\zeta = -(\xi/\varphi_{4221})\cos(\varphi_{4221} z^*), \quad \text{for } \varphi_{4221} z^* \gg \zeta,$$

where

$$\delta = \frac{k_1\omega_3^2}{k_3\omega_1^2}\left(\frac{\alpha_{21}}{\alpha_{31}}\right)\frac{\mathcal{E}_{20}'\mathcal{E}_{10}'^2}{\mathcal{E}_{30}'}$$

and

$$\xi = \frac{k_1\omega_4^2}{k_4\omega_1^2}\left(\frac{\alpha_{21}\alpha_{24}}{\alpha_{31}^2}\right)\frac{\mathcal{E}_{20}'^2\mathcal{E}_{10}'}{\mathcal{E}_{40}'}.$$

For purposes of describing the initial growth of the waves, we will approximate ψ and ζ as

$$\psi' \equiv \psi - \varphi_{3211} z^* = Az^* + B \tag{10a}$$

and

$$\zeta' \equiv \zeta - \varphi_{4221} z^* = Cz^* + D. \tag{10b}$$

We can now solve the amplitude equations leading to the following initial growth solutions:

$$\frac{\mathcal{E}_1'}{\mathcal{E}_{10}'} \sim \exp\left\{\left[\left(\frac{\alpha_{21}}{\alpha_{31}}\right)^2 \mathcal{E}_{20}'^2 - \bar{\beta}_1\right] z^*\right\}, \tag{11a}$$

$$\frac{\mathcal{E}_2'}{\mathcal{E}_{20}'} \sim \exp\left\{\left[\frac{k_1\omega_2^2}{k_2\omega_1^2}\left(\frac{\alpha_{21}}{\alpha_{31}}\right)^2 \mathcal{E}_{10}'^2 - \bar{\beta}_2\right] z^*\right\}, \tag{11b}$$

$$\frac{\mathcal{E}_3'}{\mathcal{E}_{30}'} = \frac{k_1\omega_3^2}{k_3\omega_1^2 A}\left(\frac{\alpha_{21}}{\alpha_{31}}\right)\frac{\mathcal{E}_{20}'\mathcal{E}_{10}'^2}{\mathcal{E}_{30}'}[\sin(Az^* + B) - \sin B] + 1 \quad \text{for } \psi' \approx 0$$

$$= \exp\left[\left(\frac{k_1\omega_3^2}{k_3\omega_1^2}\mathcal{E}_{10}'^2 - \bar{\beta}_3\right) z^*\right] \quad \text{for } \psi' \gg 0, \tag{11c}$$

and similarily for $\mathcal{E}_4'/\mathcal{E}_{40}'$. Two solutions exist for $\mathcal{E}_3'/\mathcal{E}_{30}'$ and $\mathcal{E}_4'/\mathcal{E}_{40}'$ because there are two processes that can contribute initially: (a) straight ASRS and (b) the parametric mixing of waves 1 and 2 to produce either wave 3 or 4. This parametric mixing is enhanced by the real two-photon absorption state, and can compete initially with the exponential gain from optical noise as long as the coherence lengths are sufficient. Note that resonance enhancement of the mixing is the same whether the total population is in the upper or lower state. Finally, note that the parametric process as defined removes no energy from the medium. Its major contribution is to increase the input to the exponential-gain ASRS process, allowing ASRS to compete even more favorably with TPE.

Having obtained the initial growth solutions of Eq. (11), we can now define a set of distances z_i, which will represent the normalized distance required for the ith wave to grow by a gain G. Hence, these propagation lengths are a measure of the growth rate of each wave. The magnitudes of these distances are obtained from the following:

$$z_1^{cr} = \frac{\ln G}{(\alpha_{21}/\alpha_{31})^2 \mathcal{E}_{20}'^2 - \bar{\beta}_1}, \tag{12a}$$

$$z_2^{cr} = \frac{\ln G}{(\alpha_{21}/\alpha_{31})^2 \mathcal{E}_{10}'^2 (k_1\omega_2^2/k_2\omega_1^2) - \bar{\beta}_2}, \tag{12b}$$

$$z_3^{cr} = \frac{1}{A}\arcsin\left[\left(\frac{\alpha_{31}}{\alpha_{21}}\right)(G-1)A\,\frac{\mathcal{E}_{30}'}{\mathcal{E}_{20}'\mathcal{E}_{10}'^2}\frac{k_3\omega_1^2}{k_1\omega_3^2} + \sin B\right] - \frac{B}{A} \quad \text{for } \psi' \approx 0$$

$$= \frac{\ln G}{(k_1\omega_3^2/k_3\omega_1^2)\,\mathcal{E}_{10}'^2 - \bar{\beta}_3} \quad \text{for } \psi' \gg 0. \tag{12c}$$

Now, the important question to answer, for ω_1 being the input pump laser, is whether ω_2 or ω_3 grows first. This can be assessed by taking the ratio of z_3/z_2. If this ratio is greater than 1, TPE initially dominates, whereas if the ratio is less than 1, ASRS always dominates. For negligible $\bar{\beta}_i$,

$$\frac{z_3^{cr}}{z_2^{cr}} \sim \left(\frac{\alpha_{21}}{\alpha_{31}}\right)^2 \frac{\mathcal{E}_{20}'^2}{A\ln G}\left\{\arcsin\left[\left(\frac{\alpha_{31}}{\alpha_{21}}\right)(G-1)A\,\frac{\mathcal{E}_{30}'}{\mathcal{E}_{20}'\mathcal{E}_{10}'^2}\frac{k_3\omega_1^2}{k_1\omega_3^2}\sin B\right] - B\right\} \quad \text{for } \psi' \approx 0$$

$$\sim \frac{n_3(\omega_2/\omega_1)}{n_2(2+\omega_2/\omega_1)}\left(\frac{\alpha_{21}}{\alpha_{31}}\right)^2 \quad \text{for } \psi' \gg 0. \tag{13}$$

We therefore infer that optimum initial dominance of TPE implies $A \gg 1$;

$$\frac{\mathcal{G}'_{20}}{\mathcal{G}'_{30}} \gg 1, \quad \frac{\alpha_{21}}{\alpha_{31}} \gg 1, \quad \frac{\omega_2}{\omega_1} \gg 1.$$

It thus emerges that it is important to have large phase mismatches for the parametric mixing processes, some input light above usual noise at the TPE conjugate frequency; a large discrepancy in the conjugate frequencies where the pump is the lower of the two; and, finally, some resonant enhancement of TPE over ASRS. This kind of resonant enhancement is possible only if the intermediate states lie between the initial and final states, which consequently implies that the matrix element between the intermediate and final states should be much larger than between the initial and intermediate states. This ordering is caused by a desire to avoid premature radiative deactivation of the initial (inverted) state while making the product of the two matrix elements as large as possible. Linear absorption at ω_3 ($\bar{\beta}_3 \neq 0$) will also help until absorption saturation occurs.

IV. SOLUTION OF NONDEGENERATE TWO-PHOTON AMPLIFICATION WITHOUT ASRS OR INVERSION DEPLETION

Under conditions where ASRS can be neglected, we can begin with Eqs. (8a) and (8b), letting $\mathcal{G}_3 \equiv \mathcal{G}_4 \equiv 0$. This problem is analytically solvable if inversion depletion is negligible, or if $\partial\Delta/\partial t = 0$ and $\Delta \equiv 1$. Equations (8) then reduce to

$$\frac{\partial \mathcal{G}_1}{\partial z^*} = \left(\frac{\alpha_{21}}{\alpha_{31}}\right)^2 |\mathcal{G}_2|^2 \mathcal{G}_1 \tag{14a}$$

and

$$\frac{\partial \mathcal{G}_2}{\partial z^*} = \left(\frac{\alpha_{21}}{\alpha_{31}}\right)^2 \frac{k_1\omega_2^2}{k_2\omega_1^2} |\mathcal{G}_1|^2 \mathcal{G}_2, \tag{14b}$$

where we assumed $n_1 = n_2$. The term z^* refers to normalized distance in the coordinate system moving with the pulse, and diffraction and linear absorption are negligible. Defining $I'_{i,2} = |\mathcal{G}_{1,2}|^2$, and integrating directly, we arrive at the solution

$$\frac{I'_2}{I'_{20}} = \frac{e^{I'_{10}z^*}}{1 - R(e^{I'_{10}z^*} - 1)}, \tag{15}$$

where

$$R = \frac{2I'_{10}}{I'_{20}}\left(\frac{\alpha_{21}}{\alpha_{31}}\right)^2.$$

We then find the solution for I'_1, namely,

$$\frac{I'_1}{I'_{10}} = \frac{1}{2}\left(\frac{\alpha_{31}}{\alpha_{21}}\right)^2 \frac{1+R}{1 - R(e^{I'_{10}z^*} - 1)} \frac{k_2\omega_1^2}{k_1\omega_2^2}. \tag{16}$$

Equation (16) does not show the behavior directly, which was found for the initial growth of I'_1 in Eq. (11a). However, by the symmetry of the equations, we can find a solution for I'_1/I'_{10} of the same form as Eq. (15).

Even for ASRS gains of e^{36}, we reduce the pulse width only by a factor of 6, in contrast with a case where, for example, $I'_{10} \gg I'_{20}$ and/or $(\alpha_{21}/\alpha_{31})^2 \gg 1$, and the denominator of Eq. (15) approaches zero for $e^{I'_{10}z^*} \sim 1$, implying very extreme pulse sharpening. Of course, we cannot neglect population-inversion depletion, under these circumstances [Eq. (8e)], and such saturation will tend to flatten the top of the pulse. Thus, by utilizing nonlinear pulse sharpening and saturation flat-topping, it should be possible to control the pulse shape within limits set primarily by the reproducibility of experimental conditions, while pulse shortening by factors of 20–100 should be feasible, as long as the pulse duration does not become comparable to T_2.

V. SPECIAL CASE–DEGENERATE TWO-PHOTON EMISSION

Because of its special significance, as well as its qualitative and quantitative differences, the degenerate system will be treated separately from the more general case discussed above. In Eqs. (2)–(4) we must equate waves 1 and 2 as well as waves 3 and 4 for the degenerate case. Hence,

$$2\omega_1 = \omega_0, \quad 2k_1 = k_0, \tag{17a}$$

$$\omega_1 + \omega_0 = 3\omega_1 = \omega_3, \quad k_1 + k_0 = k_3, \tag{17b}$$

$$\omega_0 + \omega_3 \equiv \omega_5 = 5\omega_1, \quad k_0 + k_3 = k_5. \tag{17c}$$

In addition, for reasons that will become obvious shortly, we will introduce the second anti-Stokes mode, which in this case corresponds to the fifth harmonic of the input laser. Proceeding as previously from Eqs. (8), we then can find the rate equations in the limit where a steady state in time is established for q, namely

$$q = (i\Delta/2\Gamma\omega_0)(\alpha_{11}E_1^2 + \alpha_{31}E_3E_1^* + \alpha_{53}E_5E_3^*).$$

However, we note in passing that the procedure used in Ref. 14 no longer applies for establishing the laser-pulse duration [analogously to Eq. (6)] for which this steady-state amplitude is reached. The procedure is inadequate because growth or depletion of the laser input can no longer be neglected. While previously the anti-Stokes or complementary two-photon mode was regarded as small to establish Eq. (6), we must here treat the complementary two-photon mode (which is the same as the input laser wave) as a strong wave. This problem has been addressed elsewhere for the case of Raman scattering.[16]

Utilizing the rate equations [Eqs. (8)] applicable for the degeneration Sec. III, we first find the initial growth response for the degenerate case. For $|\mathcal{E}_1| \gg |\mathcal{E}_3| \gg |\mathcal{E}_5|$, we find

$$\frac{\partial \mathcal{E}_1}{\partial z^*} \sim \left[\left(\frac{\alpha_{11}}{\alpha_{31}}\right)^2 |\mathcal{E}_1|^2 - \bar{\beta}_1\right] \mathcal{E}_1, \tag{18a}$$

$$\frac{\partial \mathcal{E}_3}{\partial z^*} \sim \left(\frac{k_1\omega_3^2\alpha_{11}}{k_3\omega_1^2\alpha_{31}} |\mathcal{E}_1|^2 - \bar{\beta}_3\right)\mathcal{E}_3 + \frac{k_1\omega_3^2\alpha_{11}^2}{k_3\omega_1^2\alpha_{31}^2}\mathcal{E}_1^3 e^{i\theta_{3111}z^*}, \tag{18b}$$

$$\frac{\partial \mathcal{E}_5}{\partial z^*} \sim \frac{\alpha_{53}\alpha_{11}}{\alpha_{31}^2}\left(\frac{k_1\omega_5^2}{k_5\omega_1^2}\right)\mathcal{E}_3\mathcal{E}_1^2 e^{-i\theta_{5311}z^*} - \bar{\beta}_5'\mathcal{E}_5, \tag{18c}$$

where again we have neglected diffraction and assumed $\Delta \equiv 1$. These equations can be solved analogously to those of Sec. III by assuming $\mathcal{E}_n = \mathcal{E}_n' e^{i\theta_n}$. Then

$$|\mathcal{E}_1'|^2 \sim \frac{|\mathcal{E}_{10}'|^2 \bar{\beta}_1}{(\alpha_{11}/\alpha_{31})^2 |\mathcal{E}_{10}'|^2 - e^{2\bar{\beta}_1 z^*}[(\alpha_{11}/\alpha_{31})^2 |\mathcal{E}_{10}'|^2 - \bar{\beta}_1]} \tag{19}$$

and

$$\frac{\partial \theta_1}{\partial z^*} = 0,$$

which for $\bar{\beta}_1 \to 0$ implies

$$|\mathcal{E}_1'|^2 \sim \frac{|\mathcal{E}_{10}'|^2}{1 - 2(\alpha_{11}/\alpha_{31})^2 |\mathcal{E}_{10}'|^2 z^*}. \tag{20}$$

Letting $\Psi \equiv 3\theta_1 - \theta_3$, we see

$$\frac{\partial \Psi}{\partial z^*} = -\frac{k_1\omega_3^2}{k_3\omega_1^2}\frac{\alpha_{11}^2}{\alpha_{31}^2}\frac{\mathcal{E}_1'^3 \sin(\Psi - \theta_{3111}z^*)}{\mathcal{E}_3'}, \tag{21}$$

similarily leading to $\Psi' \equiv \Psi - \theta_{3111}z^* = (Az^* + B)$, whereas

$$\frac{\partial \mathcal{E}_3'}{\partial z^*} \approx \left(\frac{k_1\omega_3^2\alpha_{11}}{k_3\omega_1^2\alpha_{31}} |\mathcal{E}_1'|^2 - \bar{\beta}_3\right)\mathcal{E}_3' + \frac{k_1\omega_3^2\alpha_{11}^2}{k_3\omega_1^2\alpha_{31}^2}\mathcal{E}_1'^3 \cos(Az^* + B). \tag{22}$$

Therefore, if $\mathcal{E}_1'$ is approximately constant, we obtain in analogy with the analysis performed for the nondegenerate case,

$$\frac{\mathcal{E}_3'}{\mathcal{E}_{30}'} = \frac{3n_1}{n_3}\left(\frac{\alpha_{11}}{\alpha_{31}}\right)^2 \frac{\mathcal{E}_{10}'^3}{A\mathcal{E}_{30}'}[\sin(Az^* + B) - \sin B] + 1 \quad \text{for } \psi' \approx 0$$

$$= \exp\left[\left(\frac{3n_1\alpha_{11}}{n_3\alpha_{31}} |\mathcal{E}_{10}'|^2 - \bar{\beta}_3\right) z^*\right] \quad \text{for } \psi' \gg 0. \tag{23}$$

Continuing the analogy, the approximate equations for the $\mathcal{E}_5'$ wave are

$$\frac{\partial \mathcal{E}_5'}{\partial z^*} = \frac{\alpha_{53}\alpha_{11}}{\alpha_{31}^2}\frac{5n_1}{n_5}\mathcal{E}_3'\mathcal{E}_{10}'^2 \cos(Cz^* + D), \tag{24}$$

where $Cz^* + D = \eta' = \eta - \theta_{5311}z^*$. We now see two approximate solutions, depending on the third-harmonic generation mechanism, namely,

$$\frac{\mathcal{E}_5'}{\mathcal{E}_{50}'} = 1 + \frac{5n_1}{n_5}\frac{\alpha_{53}\alpha_{11}}{\alpha_{31}^2 C}\frac{\mathcal{E}_{10}'^2\mathcal{E}_{30}'}{\mathcal{E}_{50}'}[\sin(Cz^* + D) - \sin D] - \frac{15n_1^2}{n_3n_5}\frac{\alpha_{53}\alpha_{11}^3}{\alpha_{31}^4}\frac{\mathcal{E}_{10}'^5}{\mathcal{E}_{50}'}\frac{\cos[(A+C)z^* + (B+D)] - \cos(B+D)}{A+C}$$

$$+ \frac{\cos[(A-C)z^* + (B-D)] - \cos(B-D)}{A-C} \quad \text{for } \psi' \approx 0.$$

$$= \frac{\alpha_{53}\alpha_{11}}{2\alpha_{31}^2}\frac{5n_1}{n_5}\frac{\mathcal{E}_{30}'\mathcal{E}_{10}'^2}{\mathcal{E}_{50}'}\exp\left[\left(\frac{3n_1}{n_3}|\mathcal{E}_{10}'|^2 - \bar{\beta}_3\right)z^*\right]\left[\left(\frac{3n_1}{n_3}|\mathcal{E}_{10}'^2| - \bar{\beta}_3\right)^2 - C^2\right]^{-1}$$

$$\times\left[\left(\frac{3n_1}{n_3}|\mathcal{E}_{10}'|^2 - \bar{\beta}_3\right)[\cos(Cz^* + D) - \cos D] + C[\sin(Cz^* + D) - \sin D]\right] + 1 \quad \text{for } \psi' \gg 0. \tag{25}$$

Now, from Eqs. (20) and (23), we can obtain the amplifier length z_n^{cr} required for the third-harmonic wave to have experienced a gain G, where we will normalize to z_1^{cr}:

$$z_1^{cr} = \frac{\alpha_{31}}{\alpha_{11}}\frac{1}{2|\mathcal{E}_{10}'|^2}\left(1 - \frac{1}{G^2}\right) \tag{26a}$$

and

$$\frac{z_3^{cr}}{z_1^{cr}} \approx \frac{2}{3}\frac{\mathcal{E}_{30}'}{\mathcal{E}_{10}'}\frac{n_3}{n_1}G^2, \quad \Psi' \approx 0$$

$$\approx \frac{G^2 \ln G}{G^2 - 1}\left(\frac{\alpha_{11}}{\alpha_{31}}\right)^2 \frac{2|\mathcal{E}_{10}'|}{(3n_1/n_3)(\alpha_{11}/\alpha_{31})|\mathcal{E}_{10}'|^2 - \bar{\beta}_3}, \quad \psi' \gg 0. \tag{26b}$$

Note that $z_3^{cr}/z_1^{cr} \gg 1$ implies either $\mathcal{E}_{30}'/\mathcal{E}_{10}' \gg 1$ and $\alpha_{11}/\alpha_{31} \gg 1$ or the presence of linear absorption

at $3\omega_1$, where

$$\bar{\beta}_3 \sim \frac{3n_1}{n_3}\frac{\alpha_{11}}{\alpha_{31}}|\mathcal{E}'_{10}|^2,$$

and the presence of large linear chromatic dispersion, causing short coherence lengths for third-harmonic generation.[17] Of course, this latter situation is subject to saturation of the linear absorption.

Assuming that conditions for pure degenerate two-photon amplification can be found, we next must consider the pulse shortening that occurs in such an amplification process[18] as well as the effects of saturation due to depletion of the population inversion. From Eqs. (8a) and (8d), we can obtain approximate solutions for the case where

$$\frac{\gamma}{\Gamma'} - 1 - \Delta \gg \frac{\partial \Delta}{\partial t^*}\frac{c}{2k_1 a^2 \Gamma'}.$$

The solution for Δ is then

$$\Delta = \frac{\gamma/\Gamma' - 1}{1 + S(\alpha_{11}/\alpha_{31})^2|\mathcal{E}'_1|^4}. \tag{27}$$

An equation for $|\mathcal{E}'_1|$ which crudely includes saturation is then

$$\frac{\partial|\mathcal{E}'_1|^2}{\partial z^*} = \frac{2(\alpha_{11}/\alpha_{31})^2(\gamma/\Gamma' - 1)|\mathcal{E}'_1|^4}{1 + S(\alpha_{11}/\alpha_{31})^2|\mathcal{E}'_1|^4}. \tag{28}$$

The solution of Eq. (33) can be expressed analytically, namely,

$$\left|\frac{\mathcal{E}'_1}{\mathcal{E}'_{10}}\right|^2 = \frac{(\gamma/\Gamma' - 1)z^*}{S|\mathcal{E}'_{10}|^2} + \frac{1}{2} - \frac{1}{2S(\alpha_{11}/\alpha_{31})^2|\mathcal{E}'_{10}|^4} + \left[\left(\frac{(\gamma/\Gamma' - 1)z^*}{S|\mathcal{E}'_{10}|^2} + \frac{1}{2} - \frac{1}{2S(\alpha_{11}/\alpha_{31})^2|\mathcal{E}'_{10}|^4}\right)^2 + \frac{1}{S(\alpha_{11}/\alpha_{31})^2|\mathcal{E}'_{10}|^4}\right]^{1/2}. \tag{29}$$

Equation (29) reduces to the more familiar solution for the case of no saturation given by Eq. (20). We note that under highly saturated conditions within the steady-state response, the growth rate changes from a highly singular type to approximately linear growth in z^*. Thus, while an apparent large potential for pulse shortening in time does exist for a gain of the form given by Eq. (20), a saturated gain like that of Eq. (29) will not lead to this condition, and radical pulse narrowing in time will no longer occur. For straight exponential-gain processes, the pulse duration of the SRS scattered light t_s will shorten in accordance with

$$t_s(z) \sim \frac{t_s(0)}{[g_{ss} I_L^{\max}(0) z]^{1/2}} \equiv \frac{t_s(0)}{\gamma^{1/2}}.$$

For the degenerated TPA, the similar expression is

$$t_L(z) \sim t_L(0)\left(\frac{1 - 2\gamma}{1.582 - 2\gamma}\right)^{1/2},$$

where $0 < \gamma \leq \frac{1}{2}$, which quantifies the greater pulse-shortening capability of this very nonlinear gain mechanism.

We note in passing that for a transient response, the pulse narrows to a width comparable to $1/\Gamma$; the pulse narrowing will then slow down compared with that implied above. In the transient regime, the gain experienced by the pulse will be less the shorter the pulse, leading to a restoring force that will tend to stabilize the pulse width.

Another case to be treated is that in which both $\mathcal{E}_1$ and $\mathcal{E}_3$ waves are growing, which modifies the $\mathcal{E}_3$ growth rate. Returning to Eq. (22), but substituting Eq. (20) for $\mathcal{E}_1$, we then arrive at a new approximate growth equation for $\mathcal{E}'_3$ namely,

$$\frac{\mathcal{E}'_3}{\mathcal{E}'_{30}} = 1 + \frac{3n_1}{n_3}\frac{|\mathcal{E}'_{10}|}{|\mathcal{E}'_{30}|}\frac{1}{[1 - 2(\alpha_{11}/\alpha_{31})^2|\mathcal{E}'_{10}|^2 z^*]^{1/2}} - 1 \quad \text{for } \psi' = 0$$

$$= \frac{1}{1 - 2(\alpha_{11}/\alpha_{31})^2|\mathcal{E}'_{10}|^2 z^*}\left(1 + \frac{3n_1\alpha_{31}}{2n_3\alpha_{11}}\right) \quad \text{for } \psi' \gg 0. \tag{30}$$

Note that for phase matching where $\Psi' = 0$, and for the parametric production process, the third-harmonic wave grows more slowly with z^* than the input laser field $\mathcal{E}'_1$, but once the exponential gain associated with anti-Stokes Raman scattering at $3\omega_1$ becomes important, the third harmonic grows faster in z^* than $\mathcal{E}'_1$, implying that for a long enough amplifier, the third harmonic will always overtake the first harmonic. From Eqs. (18c) and (25), we also see that the production of the fifth harmonic will become significant if the third-harmonic waves becomes sufficiently large. In analogy with Eq. (30), it can be shown that the fifth-harmonic production rate in the exponential-gain limit is still faster in z^* than that of the third harmonic, implying that the fifth-harmonic intensity will eventually overtake that of both the first and third har-

monic. We have, of course, neglected numerous times the feedback energy to prior odd harmonics in Eq. (8), which occurs when $|\mathcal{E}_1| \sim |\mathcal{E}_3| \sim |\mathcal{E}_5|$, but we still expect this type of behavior.

These statements apply even if we assume that the third-order susceptibility is the same for all photon-production processes. However, as the photon energy increases, we approach the uv linear-absorption bands (intermediate states) of the atomic system, leading in general to an enhanced cross section for the production of the higher harmonics.

Two different methods emerge from this discussion for the operation of a degenerate two-photon amplifier. The first involves phase mismatching during third-harmonic generation via the parametric process, combined with linear absorption at the third harmonic and possibly resonant enhancement of $\mathcal{E}_1'$ amplification by $\alpha_{11}/\alpha_{31} \gg 1$. This only leads to a degenerate two-photon amplifier and pulse-duration shortener. The second method consists of allowing the odd-harmonic generation processes to dominate, leading to greater-than-100% conversion of the previous odd harmonic to the next. If, for example, all odd harmonics are not absorbed by the atomic system while significant cancellations in the sum over intermediate states can be avoided, this kind of amplifier may perhaps be useful for efficient generation of coherent x rays. In laser fusion, one can also conceive of including some linear absorption in one of the higher odd harmonics, thus terminating the process at the preceding odd harmonic. Further, by combining these two proposed methods, we might be able to simultaneously amplify while pulse-shortening an input (driver) laser beam in one amplifier, followed by sequential odd-harmonic generation in a second amplifier, again with both energy and intensity gain, as diagramed in Fig. 2. The resultant discretely frequency-chirped pulse might have most of its energy at $13\omega_1 = \omega_{13}$, while the initial rise of the pulse would be at frequency ω_1, as schematically indicated in Fig. 2.

VI. GENERAL CONSIDERATIONS

Starting from rather simple concepts, we will attempt to find some bounds on experimentally adjustable parameters as well as some quantitative relationships that will allow us to evaluate amplifying media and relevant schemes. As a first condition, we recognize that the single-photon radiative loss rate must be sufficiently small so that the required energy can be stored before superfluorescent depumping occurs. It is also desirable to avoid complicated isolation schemes. Because a gain of $\sim e^7$ is usually assumed as marginally stable in the absence of reflecting surfaces, we conclude that

$$N l \sigma_I / \Gamma < g_s \approx 7 , \tag{31}$$

where σ_I / Γ is the total single-photon cross section, Γ is the corresponding linewidth, and g_s is the steady-state ASRS gain coefficient.

Note that the radiative-depumping rate is scaling as the excited-state density times the length of the medium, which is identical to the scaling of the total energy storage per unit beam area. Because large total energy storage per unit area implies low total-loss rates, it is equally important to examine nonradiative losses, which usually scale with some power of the excited-state density N. However, because no simple expression can be written for general applicability, we will just have to remember that an upper bound exists for the excited-state density N.

In a system where the second-order Bloch equations apply, a steady-state response of the system implies that the minimum laser-pulse duration is bounded[14] and dependent on the magnitude of the nonlinear gain G_{NL} (representing TPE or ASRS); for example,

$$t_L > (\ln G_{\mathrm{NL}})/\Gamma , \tag{32}$$

as indicated by Eq. (6). This condition arises from the desire to avoid higher-than-minimum intensity levels as well as to minimize the length of the nonlinear medium.

Sufficient energy must be stored in the system per unit beam area to avoid saturation of the amplifier, at least until near the end of the last e-folding length, or

$$\text{system energy gain} \equiv G_E < 1 + \frac{\text{usable stored energy}}{\text{laser energy input}} ,$$

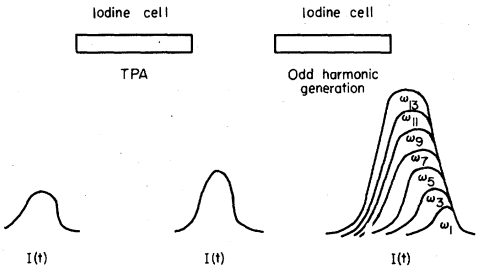

FIG. 2. Schematic diagram of the possible sequential operation of two different degenerate-two-photon amplifiers: the first leading to amplification at the input (fundamental) frequency and the second leading to a frequency sweep in time from the fundamental through the thirteenth harmonic.

leading to the condition

$$G_E < 1 + \frac{\hbar\omega_0 Nl}{I_L t_L (\ln G_{\rm NL})} . \tag{33}$$

The final empirical relationship relates, e.g., the onset of laser-induced damage to components, the breakdown in various gaseous media, or large F-center production rates to the laser energy density; namely,

$$I_L t_L G_E < \mathcal{K}_{\rm BKD}(\lambda) , \tag{34}$$

where $\mathcal{K}_{\rm BKD}$ is a constant depending only on the relevant wavelength. This condition is to be applied only over the pulse-duration range 50 psec $< t_L <$ 3 nsec. We are implicitly neglecting multiphoton-induced processes, such as self-focusing, which clearly dominate for very short pulse durations and may be important for the shorter wavelengths, where two- or three-photon absorption leads either to resonances with states near the ionization level or to the ionization continuum.

By simply manipulating the above four conditions, we can immediately derive two conditions that must be satisfied:

$$\frac{(G_E - 1) I_L t_L (\ln G_{\rm NL})}{\hbar\omega_0} < Nl < g_s \Gamma/\sigma_I , \tag{35a}$$

$$(\ln G_{\rm NL})/\Gamma < t_L < \mathcal{K}_{\rm BKD}/I_L G_E . \tag{35b}$$

We can further eliminate the center parameter in both of the above relationships, to derive two upper bounds on laser intensity, namely,

$$I_L < \begin{cases} \dfrac{\hbar\omega_0 g_s \Gamma^2}{\sigma_I (G_E - 1)(\ln G_{\rm NL})^2} \\ \dfrac{\mathcal{K}_{\rm BKD}\Gamma}{G_E (\ln G_{\rm NL})} . \end{cases} \tag{36}$$

Whether total energy storage or breakdown dictates the limitation depends on whether the ratio of two parameters, which we shall define as $\mathcal{K}_{\rm sat}/\mathcal{K}_{\rm BKD}$, is either less than or greater than unity, respectively, where $\mathcal{K}_{\rm sat} \equiv \hbar\omega_0 g_s \Gamma/\sigma_I (\ln G_{\rm NL})$.

While we succeeded in obtaining bounds [Eqs. (35) and (36)], those bounds are not given in terms of material parameters only. By substituting the pertinent form of $G_{\rm NL}$, similar limits can be derived in which material parameters appear exclusively. We will demonstrate this possibility for the case of unsaturated steady-state anti-Stokes–stimulated Raman scattering, where $G_{\rm NL}$ is given by[14]

$$G_{\rm NL} \equiv G_{\rm AS} = \exp\left(\frac{16\pi^2 (\partial\sigma_R/\partial\Omega) I_L N l}{\hbar\omega_1 \Gamma k_3^2}\right) , \tag{37}$$

and $\partial\sigma_R/\partial\Omega$ is the Raman cross section per unit solid angle. Solving Eq. (37) for Nl and substituting, we can find immediately the following two relationships:

$$I_L > \frac{k_3^2 \hbar\omega_1 \sigma_1 (\ln G_{\rm AS})}{16\pi^2 g_s (\partial\sigma_R/\partial\Omega)} , \tag{38a}$$

and

$$t_L < \hbar\omega_0 \Gamma k_3^2 \hbar\omega_1 / 16\pi^2 (\partial\sigma_R/\partial\Omega) I_L^2 (G_E - 1) . \tag{38b}$$

Finally, Eqs. (38) can then be used to eliminate I_L from all other relationships, leading to the desired set of bounds on experimental parameters that are expressed solely in terms of material parameters. These are summarized in Table I.

We can even go one step further by eliminating the center parameter in any of the three relationships of Table I to find an upper bound on the ratio of the single- and two-photon cross sections, namely,

$$\frac{\sigma_I/\Gamma}{\partial\sigma_R/\partial\Omega} < \begin{cases} \dfrac{16\pi^2 g_s^2 \Gamma}{k_3^2 \sigma_I (G_E - 1)(\ln G_{\rm AS})^3} \left(\dfrac{\omega_0}{\omega_1}\right) \\ \dfrac{16\pi^2 g_s \mathcal{K}_{\rm BKD}}{k_3^2 \hbar\omega_1 G_E (\ln G_{\rm AS})^2} . \end{cases} \tag{39}$$

If breakdown is negligible, this implies

$$\begin{aligned} \frac{(\sigma_I/\Gamma)^2}{\partial\sigma_R/\partial\Omega} &< \frac{4 g_s^2 \lambda_3^2}{(G_E - 1)(\ln G_{\rm AS})^3} \left(\frac{\omega_0}{\omega_1}\right) \\ &= 2\times 10^{-11} \left(\frac{\omega_0}{\omega_1}\right) \lambda_3^2(\mu) \\ &= 2\times 10^{-3} \frac{\bar{k}_0}{\bar{k}_1 (\bar{k}_0 + \bar{k}_1)^2} . \end{aligned} \tag{40}$$

Thus, large ω_0 and large ω_1 must be associated

TABLE I. Constraints for anti-Stokes–stimulated Raman scattering (unsaturated and steady-state).

$$\frac{(G_E - 1) k_3^2 \sigma_I (\ln G_{\rm AS})^3}{16\pi^2 g_s \Gamma (\partial\sigma_R/\partial\Omega)} \left(\frac{\omega_1}{\omega_0}\right) < Nl < \frac{g_s \Gamma}{\sigma_I}$$

$$\frac{k_3^2 \hbar\omega_1 \sigma_I (\ln G_{\rm AS})}{16\pi^2 g_s (\partial\sigma_R/\partial\Omega)} < I_L < \begin{cases} \dfrac{\hbar\omega_0 g_s \Gamma^2}{\sigma_I (G_E - 1)(\ln G_{\rm AS})^2} \\ \dfrac{\mathcal{K}_{\rm BKD}\Gamma}{G_E (\ln G_{\rm AS})} \end{cases}$$

$$\frac{(\ln G_{\rm AS})}{\Gamma} < t_L < \begin{cases} \dfrac{16\pi^2 g_s^2 \Gamma (\partial\sigma_R/\partial\Omega)}{(G_E - 1) k_3^2 \sigma_I^2 (\ln G_{\rm AS})^2} \left(\dfrac{\omega_0}{\omega_1}\right) \\ \dfrac{16\pi^2 g_s \mathcal{K}_{\rm BKD} (\partial\sigma_R/\partial\Omega)}{k_3^2 G_E \hbar\omega_1 \sigma_I (\ln G_{\rm AS})} \end{cases}$$

with small σ_I and large $\partial\sigma_R/\partial\Omega$, and vice versa, leading to the conclusion that infrared lasers and modest electronic-transition energies will more readily satisfy the constraints. This finding is in agreement with the conclusions of the previous section.

To treat the nondegenerate two-photon emission problem, we have to use the more complicated form of the gain (Eq. 15)

$$G_{\mathrm{NL}} \equiv G_{\mathrm{NDTP}} = e^{B}\left[1 + \frac{2I_{10}}{I_L}\left(\frac{\alpha_{21}}{\alpha_{31}}\right)^2 \frac{k_1\omega_2^2}{k_2\omega_1^2}(1 - e^{B})\right]^{-1}, \tag{41}$$

where B is given by

$$B = 16\pi^2 \frac{\partial\sigma_R}{\partial\Omega}\frac{I_L N l}{\hbar\omega_1\Gamma}\left(\frac{\alpha_{21}}{\alpha_{31}}\right)^2 \frac{\omega_2^2 k_2}{k_1 k_3^2 \omega_1^2},$$

and we are no longer using dimensionless units. This form of gain leads to very similar conditions on I_L, t_L, and Nl.

Because the degenerate two-photon amplifier does not involve exponential gain but rather a more singular gain, we will examine this latter case more carefully. The gain expression is given by

$$G_{\mathrm{NL}} \equiv G_{\mathrm{DTP}} = \left(1 - \frac{32\pi^2(\alpha_{11}/\alpha_{31})^2(\partial\sigma_R/\partial\Omega)I_L N l}{\hbar\omega_0\Gamma k_1^2}\right)^{-1}. \tag{42}$$

By following the same procedure given earlier for ASRS and tabulating the resulting bounds, we arrive at the results presented in Table II. Note that both the minimum intensity and the minimum Nl product have been reduced by a factor of 2 $(\ln G_{\mathrm{AS}}) \times (1 - 1/G_{\mathrm{DTP}})$, which for $G_{\mathrm{DTP}} \sim G_{\mathrm{AS}} \sim e^{10}$ amounts to a factor of ~20. Furthermore, the maximum usable pulse duration has been increased by the same factor. However, this result is consistent with the results of the previous section, because the anti-Stokes competition is so effective for the degenerate TPA, primarily due to the efficient parametric generation of third-harmonic waves which act as an input to the exponential gain. Thus, we see that energy extraction for the degenerate TPE case involves much more desirable experimental parameters than the nondegenerate TPE case.

The maximum achievable energy gain G_E vs wavelength is plotted in Fig. 3 for both TPE and ASRS for the $^2P_{1/2} \rightarrow {}^2P_{3/2}$ transition of atomic iodine. As indicated previously, best performance is achieved for either the degenerate TPE process or for long-wavelength inputs. The nonresonant character of this system enables us to plot such a simple set of curves. For other systems, the intermediate states will usually complicate the picture.

TABLE II. Constraints for degenerate two-photon amplifier.

$$\left(1 - \frac{1}{G_{\mathrm{DTP}}}\right)\frac{(G_E - 1)k_1^2\sigma_I(\ln G_{\mathrm{DTP}})^2}{32\pi^2 g_s\Gamma(\partial\sigma_R/\partial\Omega)(\alpha_{11}/\alpha_{31})^2}\left(\frac{\omega_1}{\omega_0}\right) < Nl < \frac{g_s\Gamma}{\sigma_I}$$

$$\frac{\hbar\omega_1\sigma_I k_1^2(1 - 1/G_{\mathrm{DTP}})}{32\pi^2 g_s\Gamma(\partial\sigma_R/\partial\Omega)(\alpha_{11}/\alpha_{31})^2} < I_L < \begin{cases} \dfrac{\hbar\omega_0 g_s\Gamma^2}{\sigma_I(G_E - 1)(\ln G_{\mathrm{DTP}})^2} \\ \dfrac{\mathcal{K}_{\mathrm{BKD}}\Gamma}{G_E(\ln G_{\mathrm{DTP}})} \end{cases}$$

$$\frac{\ln G_{\mathrm{DTP}}}{\Gamma} < t_L < \begin{cases} \dfrac{32\pi^2 g_s^2\Gamma(\partial\sigma_R/\partial\Omega)(\alpha_{11}/\alpha_{31})^2}{k_1^2\sigma_I^2(G_e - 1)(\ln G_{\mathrm{DTP}})}\left(\dfrac{\omega_0}{\omega_1}\right)\left(1 - \dfrac{1}{G_{\mathrm{DTP}}}\right)^{-1} \\ \dfrac{32\pi^2 g_s\mathcal{K}_{\mathrm{BKD}}(\partial\sigma_R/\partial\Omega)(\alpha_{11}/\alpha_{31})^2}{\hbar\omega_1 k_1^2\sigma_I G_E}\left(1 - \dfrac{1}{G_{\mathrm{DTP}}}\right)^{-1} \end{cases}$$

In Tables III and IV, we have estimated the relevant material parameters to predict the bounds on experimental parameters. Note that a CO_2 frequency upconverter using excited atomic iodine ($^2P_{1/2}$) appears feasible, but that an HF upconverter with I* does not appear interesting. Also, while a degenerate two-photon amplifier using

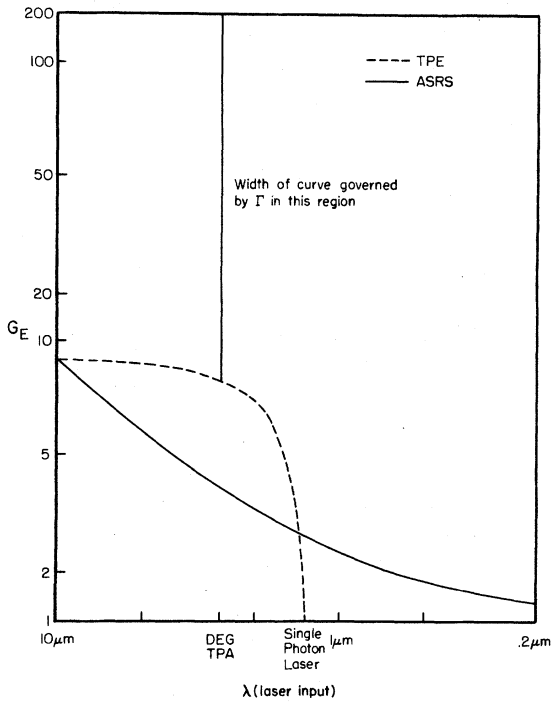

FIG. 3. Plot of the maximum energy gain realizable vs the input wavelength for two-photon emission and first-order anti-Stokes–stimulated Raman scattering calculated from inverted atomic iodine to the $^2P_{1/2}$ state.

TABLE III. Estimated material parameters for various plausible systems.

Parameter	I* ($^3P_{1/2}$) and CO_2	I*($^3P_{1/2}$) and HF	Degen TPA I*($^3P_{1/2}$)	Degen TPA O*(^1S)	Degen TPA S*(^1S)	$N_2^*(A)$ and Nd glass
λ_{pump} (mμm)	10.6	2.75	2.63	1.12	1.485	1.06
λ_{TPE} (mμm)	1.5	2.52	...	...	...	~0.33
λ_{AS} (mμm)	1.17	0.89	0.877	0.374	0.495	~0.2
λ_{2AS} (mμm)	1.054	0.67	0.526	0.224	0.297	~0.16
α_{11}/α_{31}	~1	~1	~1	~1	~1	$\sim 10^{-4}$
$\partial\sigma_R/\partial\Omega$	10^{-28}	10^{-28}	10^{-28}	$< 3\times 10^{-29}$	$\sim 10^{-29}$	$\sim 10^{-30}$
Γ (s^{-1}) (pressure broadened with inert gas)	1.9×10^{11}	1.9×10^{11}	1.9×10^{11}	1.9×10^{11}	1.9×10^{11}	1.9×10^{11}
σ_I/Γ (cm^2)	2×10^{-19}	2×10^{-19}	2×10^{-19}	2×10^{-20}	$\sim 10^{-19}$	$\sim 3\times 10^{-22}$
$\hbar\omega_{pump}$ (J)	1.9×10^{-20}	7.3×10^{-20}	7.5×10^{-20}	1.77×10^{-19}	1.33×10^{-19}	1.9×10^{-19}
$\hbar\omega_0$ (J)	1.5×10^{-19}	1.5×10^{-19}	1.5×10^{-19}	3.54×10^{-19}	2.66×10^{-19}	$\sim 10^{-18}$
G_E	~10	~7	100	100	10	5
G_{NL}	e^{10}	e^{10}	e^{10}	e^{10}	e^{10}	e^{10}
g_s	7	7	7	7	7	7
$\mathcal{H}_{BKD}$ (J/cm^2)	~1	~5	~5	~10	~10	~5

either I* or excited atomic oxygen ($^1S_0$-$^1D_2$) appears appealing, the corresponding amplifier using excited atomic sulfur ($^1S_0$-$^1D_2$) is not. Of greater interest is the possibility of using the $^1D_2$ state in the atomic-oxygen, -sulfur, or -selenium systems as an intermediate state for two-photon emission between $^1S_0$ and the ^3P manifold.[19] However, absolute inversion is required in these cases.

If atomic iodine is produced by photolysis involving CF_3I as the production mechanism of I*, one could go one step further. This possibility exists because the optical absorption of flashlamp light at ~2650 Å is nonsaturable, leading to a relationship between the diameter of the system and the density of initial gas; namely $d = 2\eta/N\sigma_{2650}$, where η is the fraction of available iodine inverted to I* (1–10% typically) and $\sigma_{2650} = 5.4\times 10^{-19}$ cm^2.

Furthermore, a relationship exists which guarantees that diffraction will not cause the beam of input diameter $d \equiv 2a$ to increase beyond the diameter of the tube after traveling through the inverted medium of length l, namely, $d^2 > 10\lambda l$. The two above condition on d plus the conditions on the product Nl can be used to establish what minimum d and l and maximum N are possible, namely,

$$d > 5\lambda_1 (Nl)_{min}\sigma_{2650}/\eta, \tag{43a}$$

$$l > 2.5(Nl)^2_{min}(\sigma_{2650})^2\lambda_1/\eta^2, \tag{43b}$$

$$N < 2\eta^2/5(\sigma_{2650})^2\lambda_1(Nl)_{min}. \tag{43c}$$

Predictions for the CO_2 frequency upconverter and for the degenerate two-photon amplifier are summarized in Table V for the situation considered in Tables III and IV where $\eta = 0.01$.

TABLE IV. Bounds on experimental parameters dictated by Table III.

Parameter	I*($^3P_{1/2}$) and CO_2	I*($^3P_{1/2}$) and HF	Degen TPA I*($^3P_{1/2}$)	Degen TPA O*(^1S)	Degen TPA S*(^1S)	$N_2^*(A)$ and Nd Glass
I_L (W/cm^2) <	1.5×10^9	1.5×10^9	10^8	2×10^9	4×10^9	2×10^{10}
I_L (W/cm^2) >	2×10^8	1.2×10^9	7×10^7	3×10^8	2×10^9	10^{10}
t_L (psec) <	300	100	72	400	95	100
t_L (psec) >	50	50	50	50	50	50
Nl (cm^{-2}) <	5×10^{19}	3.5×10^{19}	3.5×10^{19}	3.5×10^{20}	7×10^{19}	2×10^{19}
Nl (cm^{-2}) >	4×10^{18}	2×10^{20}	2.5×10^{19}	4×10^{19}	3.5×10^{19}	2×10^{19}
l (cm) <	$95d^2$ (cm)	$365d^2$	$380d^2$	$892d^2$	$673d^2$	$943d^2$

TABLE V. Predicted minimum medium diameter, length of amplifier, and maximum inversion density for I* production by photolysis of CF_3I, when $\eta = 1\%$.

	CO_2	Degen TPA
$d \geq$	1.2 cm	1.8 cm
$l \geq$	124 cm	1200 cm
$N \leq$	$3.5\times10^{16}/cm^3$	$2.1\times10^{16}/cm^3$

VII. CONCLUSIONS

By liberally utilizing approximations, we have been able to examine analytically many features of an amplifier based on a two-quantum transition. While anti-Stokes Raman scattering appears the most likely process in most cases, conditions seem to exist that might allow two-photon amplification to dominate. Furthermore, while ideal amplifying media and driver lasers have not been identified, a few prospective systems have been investigated mathematically which appear to be capable of generating both high-energy and high-intensity pulses for laser fusion application.

In Tables III and V, we see that, theoretically, atomic iodine appears to be capable of acting in an odd-harmonic generator mode at an energy gain of ~100 starting with an input 2.63-μm laser intensity of ~10^8 W/cm^2 if inversion densities of 2×10^{16} are created in approximately a 2-cm diameter by 2-m-long container. Also, atomic iodine can be employed as an energy gain 10 "afterburner" on a CO_2 laser system if an inversion density of 3×10^{16} is established in approximately a 1.2-cm diameter by 1.2-m-long container, if the input CO_2 intensity is ~10^9 W/cm^2. Both of these proposals appear quite reasonable. Experiments to verify that this performance can be obtained are currently in preparation.

Some difficulties which are anticipated in realizing practical systems are related to maintaining good enough spatial coherence. Some numerical calculations have been performed indicating that pulse narrowing in space is to be expected, but it can be controlled. However, since two-photon emission and self-focusing are of the same order of nonlinearity, only careful experiments under conditions where pulse amplification takes place will be convincing. Recently, extensive work has been carried out experimentally on normal-vibrational-stimulated Raman scattering in gases under geometry and laser-pump conditions which are close to those required to realize the amplifiers discussed in this paper.[20] Both spatial and temporal breakup of the input-laser pulse have been observed. Most of the observations are in good agreement with theory, and there are reasons for optimism that conditions exist where such problems can be avoided.

Another set of uncertainties which should be pointed out in connection to this work deals with the effects of T_2 dephasing of the inverted atomic system in times comparable to the duration of the amplified input-laser pulse, as well as the effects of inversion depletion through the amplification process. Furthermore, the details of how the many odd-harmonic waves interact with each other at high signal levels and the details of how different degrees of phase mismatching effect the high signal conditions, must be explored. Numerical solutions to these problems are currently being found and will be reported elsewhere.

While much emphasis has been placed so far on atomic iodine, there are certainly other media with potentially better properties. We have mentioned just a few above. Extensive discussions of these other candidates are not really merited at this time because even the methods of inverting any of these media to sufficiently high inversion densities have not been reported. Because of the amount of work currently in progress, it is likely that this situation will change in the not-too-distant future.

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