Theory of Degenerate Four-Wave Mixing in Saturable Absorbing Media with the Inclusion of Pump Propagation Effects

ALEXANDER L. GAETA, MARK T. GRUNEISEN, AND ROBERT W. BOYD

Abstract—**The phase-conjugate reflectivity attainable by degenerate four-wave mixing in an idealized saturable absorbing medium is calculated for arbitrary values of the laser intensity, laser frequency, and absorption path-length of the nonlinear medium. The influence of pump-wave absorption, including saturation and standing-wave effects, is included in the theory. The treatment is based on an analytic expression for the intensity distribution of the counterpropagating waves. The theoretical predictions differ substantially from those based on theories that ignore pump absorption effects. The results of the present calculation are used to predict the minimum laser intensity, absorption path-length, and detuning required to obtain unit reflectivity and self-oscillation.**

I. Introduction

DEGENERATE four-wave mixing (DFWM) has proven to be a versatile technique for the generation of the phase conjugate of an optical wavefront [1]. Since the initial theoretical treatments of DFWM in Kerr media [2] and saturable absorbers [3], there has been considerable interest in extending these theories to include effects such as the polarization properties of the interaction [4]–[6], depletion of the pump waves due to nonlinear coupling [7], detuning of the frequency of the signal wave from that of the pumps [8], [9], nonlinear refractive index changes [10], and the use of pump waves of unequal input intensities [11], [12]. The highest phase conjugate reflectivities have been achieved through DFWM utilizing the nonlinear response of saturable absorbing media [13], [14]. Since absorption effects are an inherent aspect of this type of nonlinearity, attenuation of the interacting waves plays a key role in determining the phase conjugate reflectivity. However, such effects are very difficult to treat theoretically because the standing-wave nature of the optical field within the medium causes the saturated absorption coefficient to vary spatially in distances of the order of an optical wavelength. For this reason, most previous theories have either ignored the effects of pump wave absorption [3] or have treated it in an approximate manner [11], [15].

Recently, we presented a theory of DFWM in saturable absorbing media in which the effects of pump wave propagation were included in an exact manner, including both the effects of saturated absorption and of the standing wave nature of the two counterpropagating pump waves [16]. We assumed that the probe and conjugate waves were weak relative to the pump waves because only in this limit is the output wave guaranteed to be the phase conjugate of the input probe wave. This theory was applied to the case in which the optical fields were tuned to exact resonance with the optical transition, and it was found that the theoretical predictions for the phase-conjugate reflectivity were profoundly modified by the inclusion of pump absorption effects. It was also found that for line center operation the phase conjugate reflectivity could not exceed unity and in fact rarely exceeded 10 percent. In the present paper, we apply this method to the case in which the optical waves are detuned from the optical resonance, thus allowing an increased value of the phase conjugate reflectivity. For this case, the calculation is computationally more difficult, but again an analytic solution for the intensity of the two pump waves at every point within the nonlinear medium exists, making it possible to derive simple coupled-amplitude equations for the probe and conjugate waves in terms of nonlinear absorption and coupling coefficients whose spatial dependence is known. These coupled amplitude equations can then be solved numerically in a simple, noniterative manner to determine the predicted phase conjugate reflectivity.

II. Theory

The geometry that we consider in this paper is shown in Fig. 1. Forward and backward going pump waves of amplitude A_f and A_b interact in a nonlinear medium with a weak probe wave of amplitude A_p to form a phase conjugate wave of amplitude A_c. The total electric field within the nonlinear medium can hence be represented as

$$\mathcal{E}(\vec{r}, t) = E(\vec{r}) \exp(-i\omega t) + \text{c.c.} \tag{1}$$

where the field amplitude $E(\vec{r})$ can be expressed as

$$E(\vec{r}) = A_f(\vec{r})\, e^{i\vec{k}\cdot\vec{r}} + A_b(\vec{r})\, e^{-i\vec{k}\cdot\vec{r}} + A_p(\vec{r})\, e^{i\vec{k}'\cdot\vec{r}} + A_c(\vec{r})\, e^{-i\vec{k}'\cdot\vec{r}}. \tag{2}$$

The nonlinear response of the material system is next modeled by solving the Bloch equations in steady state

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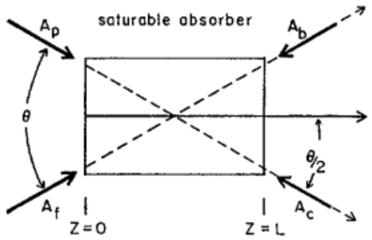

Fig. 1. Geometry of the degenerate four-wave mixing process.

for a two-level atomic system subjected to the field given by (2). This nonlinear response acts as a source term in the wave equation governing the spatial evolution of field (2). We showed in our previous paper [16] that in the limit where the signal and conjugate fields are much weaker than the two pump fields, the slowly varying field amplitudes obey the equations

$$\frac{dA_f}{dz} = -\alpha_f A_f \tag{3a}$$

$$\frac{dA_b}{dz} = \alpha_b A_b \tag{3b}$$

$$\frac{dA_p^*}{dz} = -\alpha^* A_p^* - \kappa^* A_c \tag{3c}$$

$$\frac{dA_c}{dz} = \alpha A_c + \kappa A_p^* \tag{3d}$$

where

$$\alpha_{f,b} = \frac{1}{2}\alpha_0(1 + i\delta\omega T_2)\left[\frac{1}{C}\left(1 + \frac{C - B}{2I_{f,b}}\right)\right] \tag{4a}$$

$$\alpha = \frac{1}{2}\alpha_0(1 + i\delta\omega T_2)\left[\frac{B}{C^3}\right] \tag{4b}$$

$$\kappa = -\alpha_0 \frac{(1 + i\delta\omega T_2)A_f A_b}{C^3 E_s^2} \tag{4c}$$

$$B = 1 + I_f + I_b \tag{4d}$$

$$C = \{[1 + I_f + I_b]^2 - 4I_f I_b\}^{1/2} \tag{4e}$$

$$I_{f,b} = \frac{|A_{f,b}|^2}{E_s^2} \tag{4f}$$

$$\alpha_0 = \frac{4\pi N\omega\mu^2 T_2}{\hbar c[1 + (\delta\omega T_2)^2]} \tag{4g}$$

$$E_s^2 = \frac{[1 + (\delta\omega T_2)^2]\hbar^2}{4\mu^2 T_1 T_2}. \tag{4h}$$

Here, N is the atomic number density, μ is the dipole transition moment, T_1 is the population relaxation time, T_2 is the dipole dephasing time, $\delta\omega = \omega - \omega_0$ is the detuning of the laser from the atomic resonance frequency ω_0, and $k = \omega/c$. Note that α_0 is the unsaturated, intensity absorption coefficient for a wave detuned by an amount $\delta\omega$ from line center and that $I_{f,b}$ gives the intensity of the forward or backward pump wave normalized by the saturation intensity for waves detuned by $\delta\omega$.

We first obtain a solution for the intensity distribution of the pump waves within the saturable absorber [17]. An analytic solution of (3a) and (3b) can be written in terms of the difference function

$$D(z) = I_f(z) - I_b(z).$$

The solution is

$$|D(z)| - |D(0)| + \log\left[\frac{F(z)}{F(0)}\right] = \mp\alpha_0 z$$

$$\text{for } D(z)D(0) > 0 \tag{5a}$$

and

$$D(z) - D(0) \mp \log\left[\frac{F(z)F(0)}{2(1 + K)}\right] = -\alpha_0 z$$

$$\text{for } D(z)D(0) < 0 \tag{5b}$$

where in either case

$$F(z) = |D(z)| + [D^2(z) + 2(1 + K)]^{1/2} \tag{5c}$$

and K is a constant of integration independent of z and given by

$$K = I_f(z) + I_b(z) - [D^2(z) + 2\{I_f(z) + I_b(z)\} + 1]^{1/2}. \tag{6}$$

In these equations, we take the minus sign for $D(0) > 0$ and the plus sign otherwise.

The coupling constant κ that appears in (3c) and (3d) is dependent on the complex amplitudes of the pump waves and, hence, is dependent on their phases. For $\delta\omega \neq 0$ these phases vary with z due to the intensity-dependent refractive index experienced by the pump waves [18]. We express the complex field amplitudes of the pump waves by

$$A_{f,b}(z) = |A_{f,b}(z)| \exp[i\phi_{f,b}(z)] \tag{7}$$

and derive from the Bloch equations the differential equation satisfied by $\phi_{f,b}(z)$. The derivation is analogous to that presented in [16], and yields

$$\frac{\partial\phi_{f,b}}{\partial z} = \mp\frac{\alpha_0\delta\omega T_2}{4I_{f,b}}\left[1 - \frac{1 \mp (I_f - I_b)}{[(1 + I_f + I_b)^2 - 4I_f I_b]^{1/2}}\right] \tag{8}$$

where plus and minus refer to backward and forward, respectively.

We now make the variable transformations

$$A_p'^* = A_p^* e^{i\phi(z)/2} \quad \text{and} \quad A_c' = A_c e^{i\phi(z)/2} \tag{9}$$

where

$$\phi(z) = \phi_f(z) - \phi_b(z) \tag{10}$$

and use (3c) and (3d) to arrive at a new set of coupled-amplitude equations,

$$\frac{\partial A_p'^*}{\partial z} = -\alpha''^* A_p'^* - \kappa'^* A_c' \tag{11a}$$

$$\frac{\partial A_c'}{\partial z} = \alpha' A_c' + \kappa' A_p'^* \tag{11b}$$

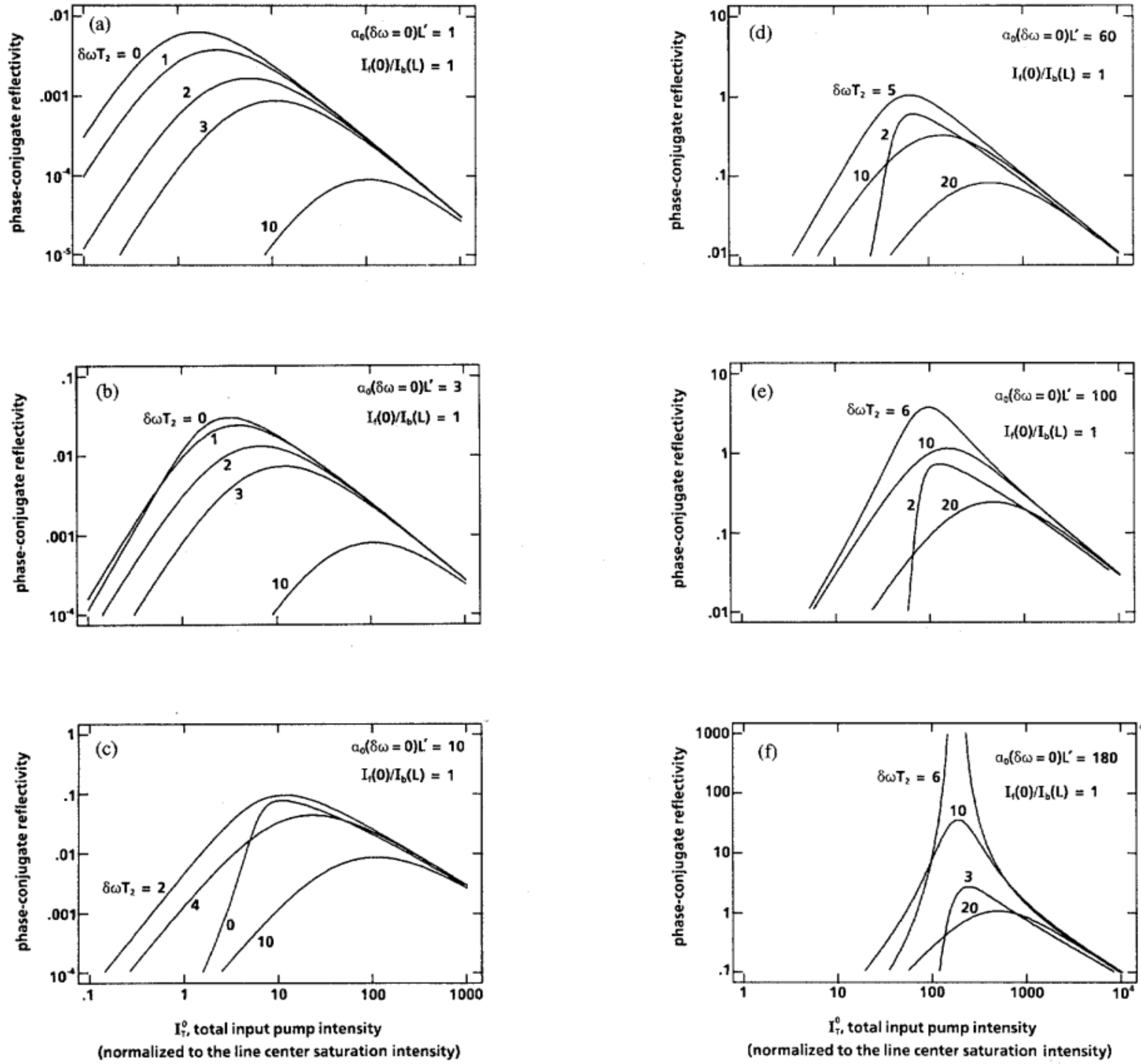

Fig. 2. Phase-conjugate reflectivity plotted as a function of the total input pump intensity normalized by the line-center saturation intensity. Equal input pump intensities are assumed. Each plot pertains to a different value of the unsaturated, line-center pump attenuation $\alpha_0(\delta\omega = 0)L'$, in the range 1.0 to 180, and in each plot several different values of the normalized laser detuning from line center, $\delta\omega T_2$, are shown.

where

$$\alpha' = \alpha - \frac{i}{2}\frac{\partial \phi}{\partial z} \tag{11c}$$

$$\alpha'' = \alpha + \frac{i}{2}\frac{\partial \phi}{\partial z} \tag{11d}$$

$$\kappa' = \kappa \frac{|A_f A_b|}{A_f A_b}. \tag{11e}$$

Our new absorption constants, α' and α''^*, and coupling constant, κ', are known functions of $I_f(z)$ and $I_b(z)$. Thus, we are easily able to integrate these new coupled-amplitude equations numerically and thereby calculate the phase-conjugate reflectivity as

$$R = \frac{|A_c(0)|^2}{|A_p(0)|^2}. \tag{12}$$

III. Results

Figs. 2–4 show the predicted values of the phase-conjugate reflectivity over a broad range of values of laser detuning, absorption path-length, and input pump intensity. In all cases the two input pump intensities are assumed to be equal. The laser detuning from line-center is normalized by the homogeneous linewidth and is thus given by the value of $\delta\omega T_2$. The absorption path-length is given in terms of the unsaturated, line-center attenuation, $\alpha_0(\delta\omega = 0)L'$, where $L' = L/\cos(\theta/2)$. The pump intensity is given in terms of the total input intensity normalized by the line-center saturation intensity, that is, by $I_T^0 = 4\mu^2[|A_f|^2 + |A_b|^2]T_1T_2/\hbar^2$.

Fig. 2 shows the predicted phase-conjugate reflectivity plotted as a function of the total normalized input intensity of the two pump fields. Each plot comprising Fig. 2

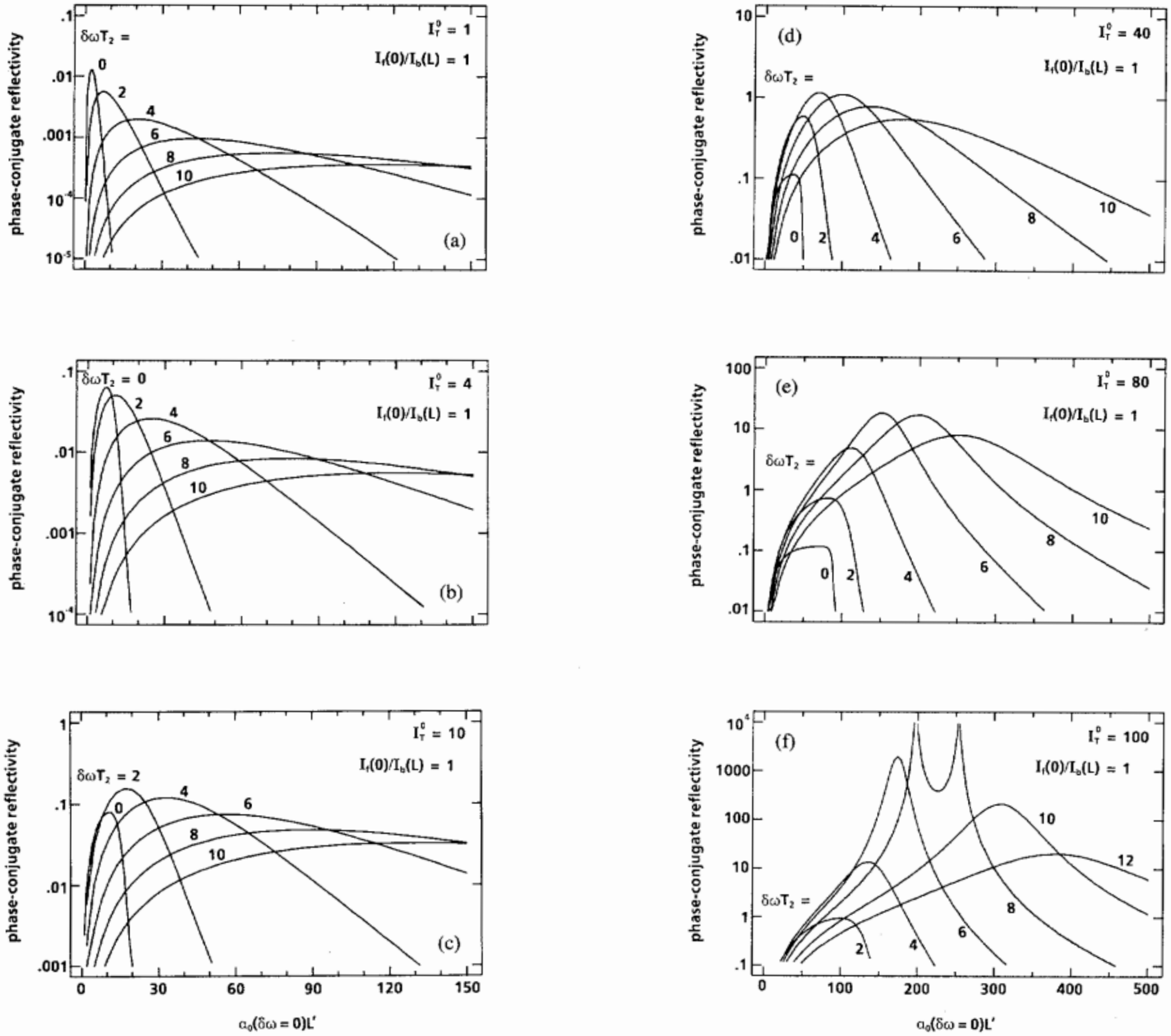

Fig. 3. Phase-conjugate reflectivity plotted as a function of the unsaturated, line-center, pump attenuation, $\alpha_0(\delta\omega = 0)L'$. Equal pump intensities are assumed. Each plot pertains to a different value of the total input pump intensity normalized by the line-center saturation intensity, I_T^0, in the range 1.0 to 100, and in each graph several different values of the normalized laser detuning from line-center, $\delta\omega T_2$, are shown.

shows the reflectivity for several different values of the normalized laser detuning $\delta\omega T_2$ for a fixed value of the line-center attenuation of the medium given by $\alpha_0(\delta\omega = 0)L'$. Fig. 2(a) shows that for $\alpha_0(\delta\omega = 0)L' = 1$, the maximum reflectivity is achieved by tuning the laser to line center. For $\alpha_0(\delta\omega = 0)L' = 3$ [Fig. 2(b)] the maximum reflectivity still occurs for line-center operation, but the maximum reflectivity is four times larger. For $\alpha_0(\delta\omega = 0)L' = 10$, the maximum reflectivity occurs for a detuning of $\delta\omega T_2 = 2$. However, to achieve this larger maximum reflectivity the laser intensity must be increased from that appropriate for $\alpha_0(\delta\omega = 0)L' = 1$ or 3. Fig. 2(d) shows that for $\alpha_0(\delta\omega = 0)L' = 60$, unit reflectivity can be obtained for $\delta\omega T_2 = 5$ and $I_T^0 = 60$. We have determined that $\alpha_0(\delta\omega = 0)L' = 60$ is the smallest absorption path length for which unit reflectivity can be achieved. For still larger values of $\alpha_0(\delta\omega = 0)L'$ [Fig. 2(e)], the maximum reflectivity exceeds unity, but larger values of $\delta\omega T_2$ and of I_T^0 are required to achieve these reflectivities. Fig. 2(f) shows that for $\alpha_0(\delta\omega = 0)L' = 180$, infinite reflectivity, implying the possibility of self oscillation, can occur for $\delta\omega T_2 = 6$ and $I_T^0 = 180$. We have compared our results for the phase-conjugate reflectivity with those obtained by Brown [19] using a purely numerical method and have found complete agreement.

Fig. 3 shows the predicted reflectivity plotted as a function of the line-center attenuation, $\alpha_0(\delta\omega = 0)L'$. In each plot the total laser intensity normalized by the line-center saturation intensity, I_T^0, is held fixed, and curves are plotted for several different values of the detuning $\delta\omega T_2$. These results are useful in determining the values of $\alpha_0(\delta\omega = 0)L'$ and $\delta\omega T_2$ that give maximum reflectivity for a fixed value of the pump intensity. In general, higher reflectivities can be achieved through the use of higher pump intensities, although larger values of the absorption path-length and of the detuning are then required. For the case

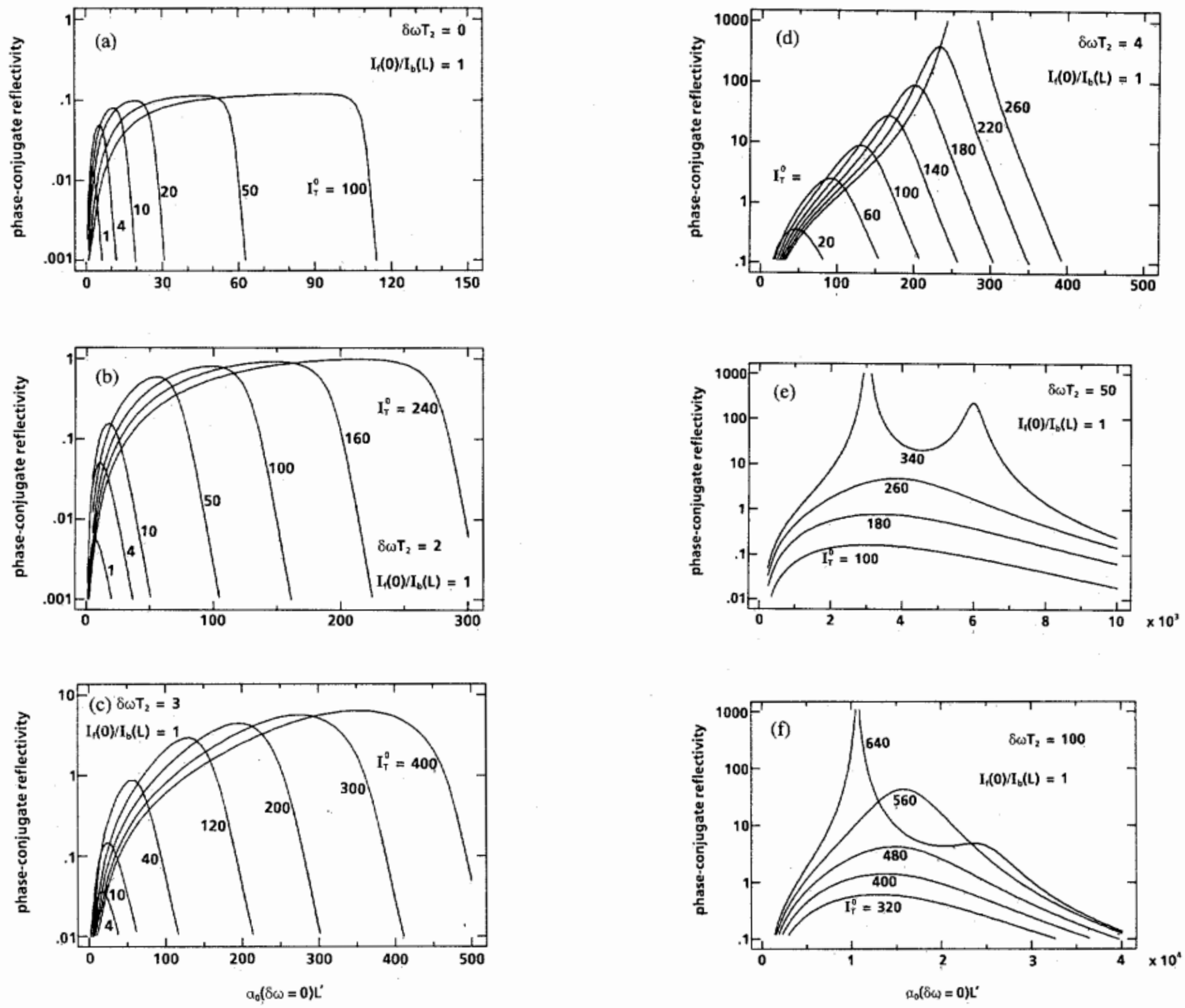

Fig. 4. Similar to Fig. 3, except that each plot pertains to a different value of the laser detuning from line-center, $\delta\omega T_2$, and in each plot several different values of the total input pump intensity normalized to the line-center saturation intensity are shown.

$I_T^0 = 1$ [Fig. 3(a)], the maximum reflectivity is ~ 0.01, and occurs for line-center operation and $\alpha_0(\delta\omega = 0)L' = 2$. For $I_T^0 = 4$, the optimum detuning is again zero, but the maximum reflectivity of ~ 0.05 occurs for $\alpha_0(\delta\omega = 0)L' = 8$. For $I_T^0 = 10$ [Fig. 3(c)] the optimum detuning is given by $\delta\omega T_2 = 2$, and the reflectivity exceeds 10 percent for $\alpha_0(\delta\omega = 0)L' = 20$. $I_T^0 = 40$ is the minimum total intensity required to achieve a reflectivity of unity [Fig. 3(d)]. A detuning of $\delta\omega T_2 = 4$ and an absorption path length of $\alpha_0(\delta\omega = 0)L' = 80$ are required to achieve unit reflectivity for this pump intensity. Still higher reflectivities can be obtained for $I_T^0 = 80$ [Fig. 3(e)]. For $I_T^0 = 100$ [Fig. 3(f)], the solutions predict infinite reflectivity for $\delta\omega T_2 = 8$ and $\alpha_0(\delta\omega = 0)L' = 250$.

Each graph in Fig. 4 shows the reflectivity plotted as a function of $\alpha_0(\delta\omega = 0)L'$ for a fixed value of the laser detuning, and each curve corresponds to a given value of the normalized laser intensity. These results are relevant to a phase conjugate mirror pumped at a fixed frequency. For line-center operation, Fig. 4(a) shows that the reflectivity does not appreciably exceed 10 percent for reasonable operating conditions. Fig. 4(b) shows that a reflectivity of unity can be obtained for a laser tuned two linewidths from line center. The total pump intensity must exceed $I_T^0 = 100$ to achieve unity reflectivity, and for such intensities, the reflectivity is relatively insensitive to the value of $\alpha_0(\delta\omega = 0)L'$. Fig. 4(c) shows that reflectivities approaching 10 are possible for a detuning of three linewidths, although larger values of I_T^0 and $\alpha_0(\delta\omega = 0)L'$ are required to achieve these larger reflectivities. Self-oscillation is possible when the detuning exceeds four linewidths [Fig. 4(d)–(f)].

IV. Conclusions

The qualitative behavior of the solutions displayed in the last section can be understood in terms of simple physical arguments. In order to obtain a large phase conjugate reflectivity, it is necessary that a large number of atoms participate in the nonlinear optical process and, therefore, $\alpha_0(\delta\omega = 0)L'$ should be chosen to be as large as possible.

TABLE I
MINIMUM VALUES OF ABSORPTION PATH-LENGTH, LASER INTENSITY, AND LASER DETUNING REQUIRED TO PRODUCE A PHASE-CONJUGATE REFLECTIVITY OF UNITY AND OF INFINITY. THE CONDITIONS UNDER WHICH THESE MINIMUM VALUES APPLY FOR EACH CASE ARE SHOWN IN BRACKETS.

	$R = 1$	$R \to \infty$
$(\alpha_0(\delta\omega = 0)L)_{min}$	60 $[\delta\omega T_2 = 5, I_T^0 = 60]$	180 $[\delta\omega T_2 = 6, I_T^0 = 180]$
$(I_T^0)_{min}$	40 $[\delta\omega T_2 = 4, \alpha_0(\delta\omega = 0)L = 80]$	100 $[\delta\omega T_2 = 8, \alpha_0(\delta\omega = 0)L = 200]$
$(\delta\omega T_2)_{min}$	2 $[I_T^0 = 200, \alpha_0(\delta\omega = 0)L = 200]$	4 $[I_T^0 = 260, \alpha_0(\delta\omega = 0)L = 260]$

On the other hand, it is undesirable that absorption be allowed to deplete any of the waves and, hence, the saturated absorption coefficient $\alpha = \alpha_0[1 + I_T^0 + (\delta\omega T_2)^2]^{-1}$ should be chosen to be as small as possible. The best compromise between these two conflicting requirements is obtained by detuning the interacting several-linewidth waves from line center so that dispersive and not absorptive effects dominate, and by adjusting the number density and detuning such that

$$\alpha L' = \frac{\alpha_0(\delta\omega = 0)L'}{1 + I_T^0 + (\delta\omega T_2)^2} \approx 1. \qquad (13)$$

In addition, it is desirable that each atom be driven into its nonlinear regime, which requires that the two contributions I_T^0 and $1 + (\delta\omega T_2)^2$ in the denominator of (13) be comparable or that

$$I_T^0 \approx 1 + (\delta\omega T_2)^2. \qquad (14)$$

The results shown in Figs. 2–4 are consistent with these simple predictions. In Fig. 2, each curve peaks at a value of I_T^0 given approximately by $I_T^0 = 2.0\,(\delta\omega T_2)^2$. The value of $\delta\omega T_2$ which gives the maximum reflectivity is given approximately by $\delta\omega T_2 = 0.7\,[\alpha_0(\delta\omega = 0)L']^{1/2}$. In Fig. 3, each curve peaks at a value of $\alpha_0(\delta\omega = 0)L'$ given approximately by $\alpha_0(\delta\omega = 0)L' = 1 + I_T^0 + (\delta\omega T_2)^2$. The value of the detuning that maximizes the reflectivity is given approximately by $\delta\omega T_2 = 0.7\,[I_T^0]^{1/2}$. In Fig. 4, each curve is again seen to peak for values of $\alpha_0(\delta\omega = 0)L'$ given approximately by $\alpha_0(\delta\omega = 0)L' = 1 + I_T^0 + (\delta\omega T_2)^2$. The fact that the maximum reflectivity is obtained when condition (13) is satisfied illustrates that the pump absorption effects cannot be ignored in an optimized phase conjugate mirror.

The theoretical results presented here can be summarized in terms of the minimum values of $\alpha_0(\delta\omega = 0)L'$, I_T^0 and $\delta\omega T_2$ required to produce a given value of the phase conjugate reflectivity. Such results are given in Table I for the minimum values required to produce reflectivities of unity and of infinity. For each case, the values of the other input parameters required to produce the desired output are given in brackets.

In conclusion, we have presented a theory of degenerate four-wave mixing in saturable absorbing media that includes the influence of pump absorption including saturation and standing-wave effects. Our predictions differ substantially from those based on theories that ignore the effects of pump absorption. Our results are consistent with a simple physical interpretation and can be used to determine the minimum absorption path length $\alpha_0(\delta\omega = 0)L'$ and pump intensity I_T^0 required to produce a given phase conjugate reflectivity.

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