Exact theory of pump-wave propagation and its effect on degenerate four-wave mixing in saturable-absorbing media

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An analytic solution for the intensity distribution of two counterpropagating pump waves within a saturable absorber is derived. From this distribution, the spatial variation of the nonlinear absorption and coupling constants that appear in the coupled-amplitude equations for the probe and the signal (i.e., conjugate) waves are determined. These coupled-amplitude equations are solved numerically in a noniterative manner, leading to a prediction for the phase-conjugate reflectivity. The results of the exact theory are compared with those of previously published theories. It is found that at large values of the input-pump intensities, the predicted phase-conjugate reflectivity is larger when pump-absorption effects are included in the theory.

INTRODUCTION

Degenerate four-wave mixing (DFWM) is a process that can generate the phase conjugate of an aberrated optical wave front. The highest reported phase-conjugate reflectivities have been achieved by means of DFWM using the nonlinear response of a two-level atom.[1] The first theoretical account of DFWM in such a system was that of Abrams and Lind,[2] in which the simplifying assumption was made that pump-beam attenuation that was due to absorption could be neglected. Their theory predicts that for line-center operation the reflectivity can be maximized by using a medium with a large absorption-path length and a large value of the laser intensity. However, for large absorption-path lengths, pump absorption cannot necessarily be neglected. More recently, Caro and Gower[3] have presented a treatment that includes in an approximate sense the effects of pump absorption. However, their treatment neglects the fact that the relative intensities of the two pump waves vary with position within the nonlinear medium. It is known from the work of Dunning and Steel[4] that the phase-conjugate reflectivity depends sensitively on the relative intensities of the two pump waves. While these theories have shed considerable light on the nature of DFWM in saturable absorbers, they are all somewhat inadequate in treating the case in which pump-wave absorption cannot be neglected.

In this paper we present a theory of DFWM in absorbing media that treats exactly the effects of pump-wave absorption. We derive an analytic expression for the intensity distribution of the two counterpropagating pump waves. The spatial dependence of the absorption and coupling constants that appear in the coupled-amplitude equations of the probe and signal waves is thereby determined. The coupled-amplitude equations for the probe and signal waves are then integrated numerically in a straightforward manner, and the phase-conjugate reflectivity is determined. Our results are in good agreement with those obtained by Brown[5] by a purely numerical method.

THEORY

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

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

where the field amplitude $E(\mathbf{r})$ is assumed to be composed of a strong- (pump) field contribution $E_0(\mathbf{r})$ and a weak- (probe and signal) field contribution $\Delta E(\mathbf{r})$ as

$$E(\mathbf{r}) = E_0(\mathbf{r}) + \Delta E(\mathbf{r}). \qquad (2)$$

This field must satisfy Maxwell's wave equation

$$\nabla^2\mathcal{E} - \frac{1}{c^2}\frac{\partial^2\mathcal{E}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2\mathcal{P}}{\partial t^2}, \qquad (3)$$

where $\mathcal{P}$ is the polarization, which can be represented as

$$\mathcal{P}(\mathbf{r}, t) = P(\mathbf{r})\exp(-i\omega t) + \text{c.c.} \qquad (4)$$

We assume that the nonlinear medium can be modeled as a collection of homogeneously broadened two-level atoms. By solving the Bloch equations in steady state, we obtain the following expression for the amplitude of the polarization:

$$P(\mathbf{r}) = \chi(E)E, \qquad (5)$$

where $\chi(E)$ is the intensity-dependent susceptibility given by[2]

$$\chi(E) = \frac{(-\alpha_0/k)(i - \delta\omega T_2)}{1 + (|E_0|^2/E_s{}^2)}, \qquad (6)$$

where $\alpha_0 = 4\pi N\omega\mu^2T_2\{\hbar c[1 + (\delta\omega T_2)^2]\}^{-1}$ is the weak-field frequency-dependent intensity-absorption coefficient, $E_s{}^2 = [1 + (\delta\omega T_2)^2]\hbar^2/(4\mu^2T_1T_2)$, N is the atomic density, μ is the dipole-matrix element, T_1 is the population-relaxation time,

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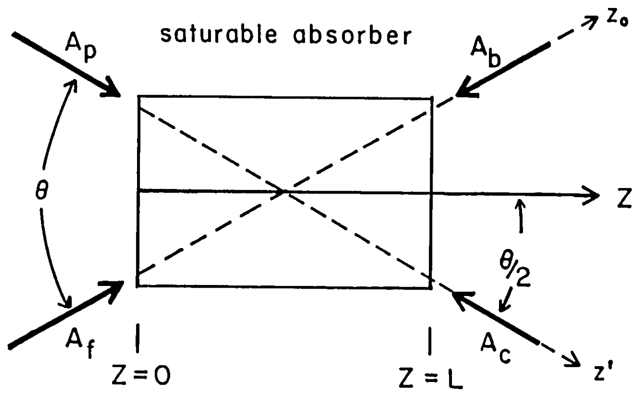

Fig. 1. Geometry of the degenerate four-wave mixing process.

T_2 is the dipole-dephasing time, $k = \omega/c$, and $\delta\omega = \omega - \omega_0$ is the detuning of the laser from the atomic-resonance frequency ω_0. In the weak-probe limit, we can expand $\chi(E)$ in a Taylor series about E_0, and express the polarization amplitude as

$$P(\mathbf{r}) = \chi_0 E_0 + \chi_0 \Delta E - \frac{\chi_0}{E_s^2} \frac{(E_0\Delta E^* + E_0^*\Delta E)(E_0 + \Delta E)}{1 + (|E_0|^2/E_s^2)}, \tag{7}$$

where $\chi_0 = \chi(E_0)$. We now assume that the strong component E_0 is composed of forward- and backward-going pump waves, counterpropagating along the z_0 axis, and that the weak component ΔE is composed of probe and conjugate components counterpropagating along the z'-axis:

$$E_0(z_0) = A_f(z_0)\exp(ikz_0) + A_b(z_0)\exp(-ikz_0) \tag{8a}$$

and

$$\Delta E(z') = A_p(z')\exp(ikz') + A_c(z')\exp(-ikz'). \tag{8b}$$

We now express χ_0 and $\chi_0[1 + (|E_0|^2/E_s^2)]^{-1}$ as complex Fourier series in z_0, and we retain only those terms in $P(\mathbf{r})$ that are phase matched to terms on the left-hand side of the wave equation (3):

$$P(\mathbf{r}) = \frac{-2i}{k}[\alpha_f A_f \exp(ikz_0) + \alpha_b A_b \exp(-ikz_0) + (\alpha A_p + \kappa A_c^*)\exp(ikz') + (\alpha A_c + \kappa A_p^*)\exp(-ikz')], \tag{9}$$

where we have introduced the following definitions:

$$\alpha_{f,b} = \tfrac{1}{2}\alpha_0(1 + i\delta\omega T_2)\left[\frac{1}{C}\left(1 + \frac{C - B}{2I_{f,b}}\right)\right], \tag{10a}$$

$$\alpha = \tfrac{1}{2}\alpha_0(1 + i\delta\omega T_2)[B/C^3], \tag{10b}$$

$$\kappa = -\alpha_0 \frac{(1 + i\delta\omega T_2)}{C^3} \frac{A_f A_b}{E_s^2}, \tag{10c}$$

$$B = 1 + I_f + I_b, \tag{10d}$$

$$C = \{[1 + I_f + I_b]^2 - 4I_f I_b\}^{1/2}, \tag{10e}$$

and

$$I_{f,b} = |A_{f,b}|^2/E_s^2. \tag{10f}$$

We now substitute our expression for $\mathcal{E}$ and $\mathcal{P}$ into the wave equation (3) and make the slowly varying envelope approximation, which leads to the following equations for the field amplitudes:

$$\frac{\partial A_f}{\partial z} = -\alpha_f A_f, \tag{11a}$$

$$\frac{\partial A_b}{\partial z} = -\alpha_b A_b, \tag{11b}$$

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

and

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

where $z = Z/\cos(\theta/2)$.

It can be seen from Eqs. (10) that, under the assumption of weak probe and conjugate fields, the nonlinear-absorption coefficients α, α_f, and α_b and coupling coefficient κ depend only on the amplitudes of the pump waves. We thus proceed to solve Eqs. (11a) and (11b) for the spatial evolution of the intensities of the pump waves within the nonlinear medium. These solutions are then used to determine the spatial dependence of the parameters α and κ that appear in the coupled-amplitude Eqs. (11c) and (11d) for the probe and signal waves. Explicitly, we find that the forward and backward intensities obey the equations

$$\frac{dI_f}{dz} = \frac{-\alpha_0}{2}\left[1 - \frac{1 - (I_f - I_b)}{\{[1 + I_f + I_b]^2 - 4I_f I_b\}^{1/2}}\right] \tag{12a}$$

and

$$\frac{dI_b}{dz} = \frac{\alpha_0}{2}\left[1 - \frac{1 + (I_f - I_b)}{\{[1 + I_f + I_b]^2 - 4I_f I_b\}^{1/2}}\right]. \tag{12b}$$

In order to solve these equations, we divide Eq. (12a) by Eq. (12b) to obtain an equation for I_f in terms of I_b. This equation is an exact differential equation, which, when solved, yields the relation

$$K = S(z) - [D^2(z) + 2S(z) + 1]^{1/2}, \tag{13}$$

where the constant of integration K is independent of z and where

$$S(z) \equiv I_f(z) + I_b(z) \tag{14a}$$

and

$$D(z) \equiv I_f(z) - I_b(z). \tag{14b}$$

We next form the sum and difference of Eqs. (12a) and (12b) to obtain differential equations for $S(z)$ and $D(z)$. Equation (13) is then used to decouple these equations, which we then solve. In particular, when $D(z)$ and $D(0)$ have the same sign, $D(z)$ obeys the equation

$$|D(z)| - |D(0)| + \log\left[\frac{F(z)}{F(0)}\right] = \mp\alpha z, \tag{15a}$$

and when $D(z)$ and $D(0)$ have opposite signs, $D(z)$ obeys the equation

$$D(z) - D(0) \mp \log\left[\frac{F(z)F(0)}{2(1 + K)}\right] = -\alpha z, \tag{15b}$$

where in either case

$$F(z) = |D(z)| + [D(z)^2 + 2(1 + K)]^{1/2}. \qquad (16)$$

In these equations, we take the minus when $D(0) > 0$ and the positive sign when $D(0) < 0$.

Using Eqs. (13) and (14), and evaluating Eqs. (15) at $z = L$, we obtain a transcendental equation for $I_b(0)$. We then employ a simple root-finding algorithm to solve for $I_b(0)$ and thus for $D(0)$, $S(0)$, and K. This information allows us to solve Eqs. (15) for $D(z)$ for arbitrary values of z. Once $D(z)$ is known, Eqs. (13) and (14) are used to calculate $I_f(z)$ and $I_b(z)$. Equations (12a) and (12b) have also been solved by Agrawal and Lax[6] and by Hermann[7] for the case of a Fabry–Perot resonator with plane-parallel mirrors. Our solution differs slightly from theirs in that we treat the case in which the two counterpropagating waves have arbitrary input intensities.

RESULTS

In this section, we graphically display the results of the analytic theory for several experimentally interesting cases. For simplicity, we assume the case of line-center operation ($\delta\omega = 0$) in all our examples. For this case, there is no variation in the phase of κ that is due to the refractive index experienced by the pump waves, and hence the correction for pump-wave dispersion that appears in the constant-pump theory of Grynberg *et al.*[8] vanishes. Our theory can be expanded to include pump-wave dispersion for the case of off-line-center operation.

The intensities of the forward- and backward-traveling pump waves are plotted as a function of position within the saturable absorber for a representative case in Fig. 2. In addition, the product of the two intensities is plotted. If standing-wave effects are ignored, this product can be shown to be constant.[4] Figure 2 shows that neither the pump-wave intensities nor their product can be assumed constant for cases of interest.

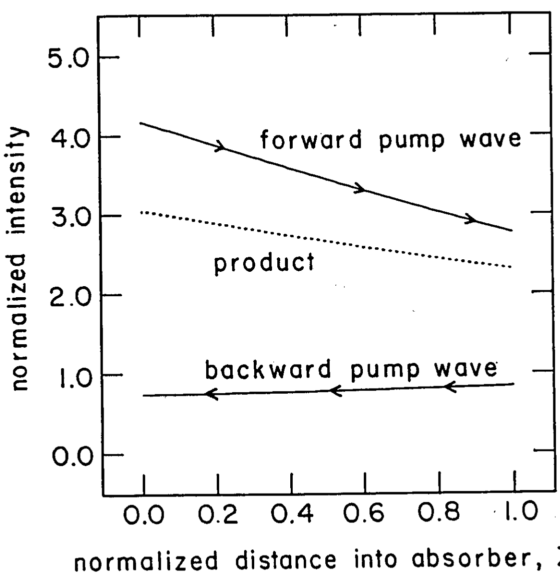

Fig. 2. Spatial distribution of the forward and backward pump waves and of their product for the case of unequal input-pump-beam intensities, with $I_f(0)/I_b(L) = 5.0$, $I_f(0) + I_b(L) = 5.0$, $\alpha_0 L' = \alpha_0 L/\cos(\theta/2) = 2.0$, and $\delta\omega = 0$.

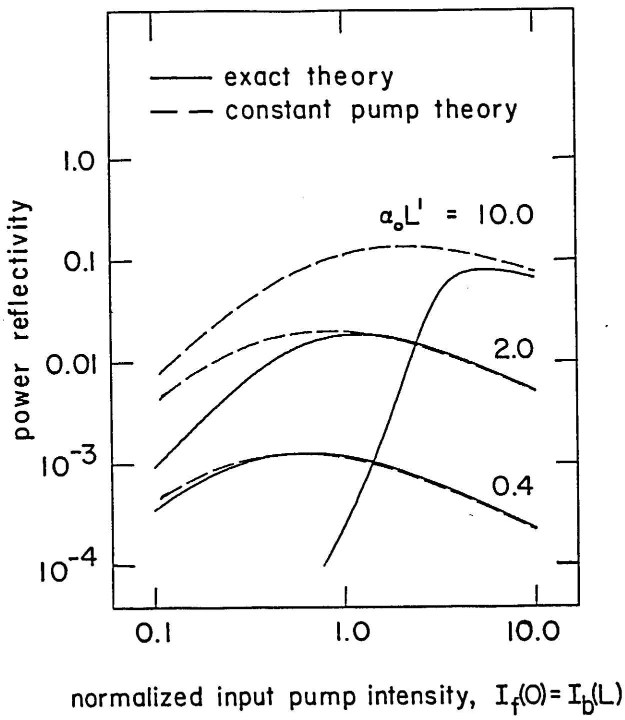

Fig. 3. Phase-conjugate reflectivity versus forward input-pump intensity for the case of equal-input-pump intensities for $\delta\omega = 0$. The results of the present theory are compared with those of the constant-pump-intensity theory of Abrams and Lind.[2]

Once the distribution of pump intensities is known, it is possible to calculate the absorption and coupling constants along the path of the probe and signal waves. By taking the conjugate-field amplitude at $z = L$ to be $A_c(L) = 0$ and choosing an arbitrary value for the amplitude $A_p(L)$ of the probe field at $z = L$, it is possible to integrate numerically the coupled-amplitude equations (11c) and (11d) from $z = L$ to $z = 0$. We then calculate the phase-conjugate power reflectivity as

$$R = \frac{|A_c(0)|^2}{|A_p(0)|^2}. \qquad (17)$$

Reflectivity results are shown for several pumping geometries in Figs. 3–6. The solid lines are plotted according to the theory presented in this paper. The broken lines refer to previous theories that ignore pump-wave absorption. The individual cases are labeled according to the length, $L' = L/\cos(\theta/2)$, of the medium measured along either of the propagation directions. The case of balanced pumping, i.e., equal-input pump-wave amplitudes $[A_f(0) = A_b(L)]$, is treated in Fig. 3. The phase-conjugate reflectivity is plotted as a function of pumping intensity normalized to the saturation intensity of the medium. The dashed lines represent predictions from the theory of Abrams and Lind,[2] in which the pump waves are assumed to have constant intensity. The peaked curves show the saturation nature of the process. At low intensities, our theory predicts a lower reflectivity than the constant-pump-wave theory as a result of our inclusion of absorption losses. At higher intensities, the medium saturates, becoming transparent to the pumping waves, and the two theories nearly agree. Close inspection shows that at these intensities ($I/I_s > 1$) our theory predicts slightly higher reflectivities than those predicted in the absence of pump-wave absorption. The effect of absorption is to attenuate the pump waves so that the pump intensities within the medium are closer to the saturation intensity, and under these conditions mixing occurs more efficiently. This effect is more pronounced for the case of unequal pumping shown in Fig. 4.

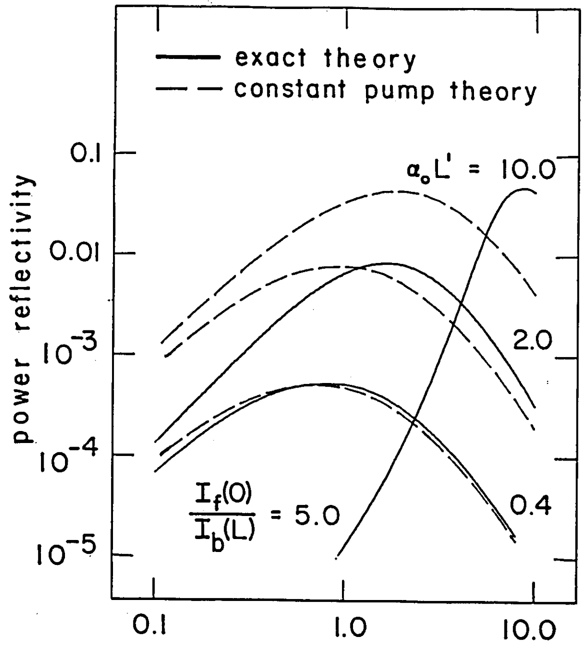

Fig. 4. Phase-conjugate reflectivity versus total pump intensity for the case of unequal pump intensities and $\delta\omega = 0$. The results of the present theory are compared with those of the constant-but-unequal pump-intensity theory of Dunning and Steel.[4]

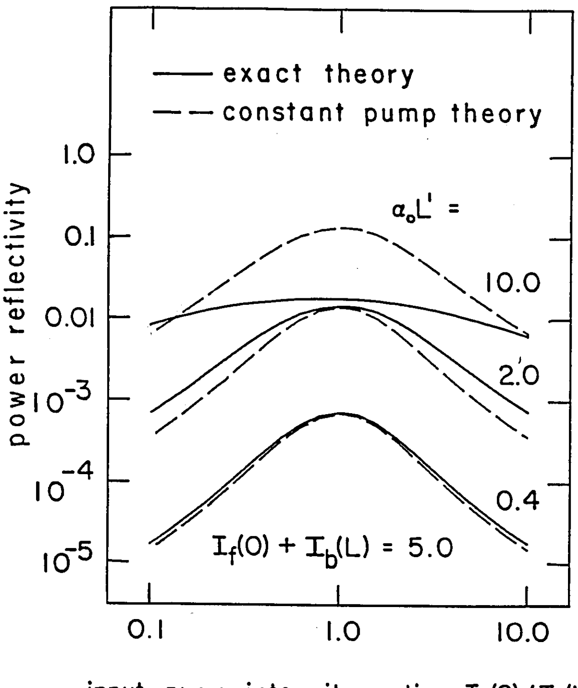

Fig. 5. Phase-conjugate reflectivity versus the input-pump-intensity imbalance, $I_f(0)/I_b(L)$, for the case $I_f(0) + I_b(L) = 5.0$ and $\delta\omega = 0$. The results of the present theory are compared with those of the constant-but-unequal pump-intensity theory of Dunning and Steel.[4]

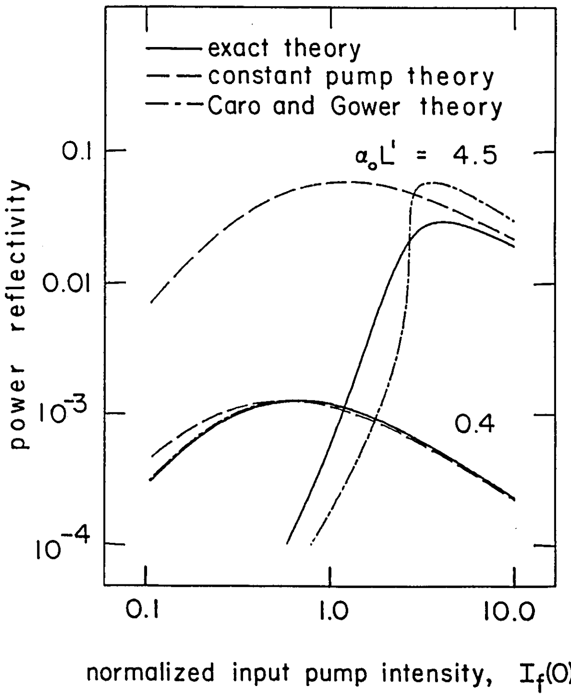

Fig. 6. Phase-conjugate reflectivity versus input-pump intensity for the case in which the backward pump wave is obtained by the retroreflection of the transmitted forward-going pump wave ($\delta\omega = 0$). The results of the present theory are compared with the constant-pump-intensity theory of Abrams and Lind and with the theory of Caro and Gower, which treats pump absorption in an approximate manner.

In this figure, our results are compared with those of Dunning and Steel,[4] who treated pump imbalance in the absence of pump-wave absorption.

Figure 5 shows the effect of varying the pumping ratio while keeping the total pump intensity constant. The optimum value of the ratio $I_f(0)/I_b(L)$ is seen to be approximately equal to one for all cases. Close examination of these curves reveals that the optimum ratio shifts slightly to lower values as $\alpha_0 L'$ is increased. A special case of the unequal-pumping geometry occurs when the second pump wave, I_b, is introduced by retroreflecting the forward wave after it passes through the absorber. Caro and Gower[3] introduced pump-wave absorption approximately to treat this geometry. In Fig. 6 we compare their theoretical results with those of Abrams and Lind[2] and with our own.

Brown[5] has also treated the effects of pump-wave absorption on the DFWM process in a system of two-level saturable absorbers for several cases, including the weak-probe limit treated here. An important difference between his and our treatments is the manner in which the two interfering pump waves are treated. Brown derived coupled-amplitude equations, identical to our Eqs. (11), and solved them numerically using an iterative procedure. For the cases that he treats, his results are in agreement with those of the theory presented here.

CONCLUSIONS

We have treated theoretically the case of two counterpropagating pump waves in a saturable absorber. Our analytic solutions for the forward and backward pump intensities show that, in general, the assumption that the product $I_f I_b$ is constant is not valid. Furthermore, we have used these results to extend the theory of DFWM to include the effects of saturable absorption on the pump waves. The theory is valid for arbitrary pump intensities in the weak-probe limit.

Theoretical values for the phase-conjugate reflectivity have been presented and are compared with the results of previous theories for several pumping geometries. We find that at large values of the input-pump intensities, the phase-conjugate reflectivity can be larger than that predicted by theories that ignore pump absorption. We also find that the optimum ratio of the forward-to-backward input-pump-wave intensities decreases from unity as the length of the absorbing medium is increased.

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