

Analysis of four wave mixing in slab waveguide under the undepleted pump approximation

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The four-wave mixing in a slab waveguide is investigated theoretically by using the singular perturbation technique with multiple space scales. In order to apply the perturbation technique, we first introduce the perturbation parameter concerned with the nonlinear coefficients and the multiple space scales in the propagation direction. Substituting the expansion of the field and the multiple space scales into the Maxwell's equation, we can get the governing equations to each order of the perturbation. From the solvability conditions to have nontrivial solutions for each perturbation solutions, it can be shown that the coupled-mode equations for four waves that are asymptotically correct in the limit of weak nonlinearity are obtained. The dependence of the coupling coefficient on the waveguide width and the field distribution of the phase conjugate wave are discussed numerically. We show that the coupling coefficients have the maximum for the optimum waveguide width. © 1997 American Institute of Physics. [S0021-8979(97)05506-0]

I. INTRODUCTION

Nonlinear guided optical phenomena have attracted the attention of many researchers who are working to establish all-optical signal processing. A nonlinear medium shows interesting phenomena such as self-focusing, generation of self-phase modulation, optical bistability, second harmonic generation, optical amplification, and so on. Recently, optical phase conjugation has been discussed theoretically and experimentally and these are useful for wave-front aberration correction, spatial information processing, and frequency filtering. Four-wave mixing in a waveguide has many advantages due to the waveguide effect.

Degenerate four-wave mixing has been studied in bulk media. Also, many applications have been predicted and demonstrated.¹ On the other hand, since the field is concentrated in the film or the core region in a waveguide structure, nonlinear guided wave phenomena have been attractive for many years.² Yariv *et al.*³ and Hellworth⁴ investigated phase conjugation by four wave mixing in a multimode nonlinear waveguide. After that, several papers reported studies of four wave mixing in waveguides and the experimental results for degenerate four wave mixing with guided waves were reviewed.⁵ Also, degenerate four wave mixing by long range surface plasmons at a metal boundary was analyzed and it was shown that large signal levels can be obtained.⁶ Karaguleff and Stegeman⁷ also studied theoretically degenerate four-wave mixing by guided waves and gave the numerical estimates for polydiacetylenes. Tomita *et al.*⁸ extended the method of Ref. 7 and obtained the phase conjugate gain. It was shown that the relative effective gain is large for a TE mode with a dielectric substrate but is small for a TM mode with metal substrates. Liu⁹ has pointed out that a large number of modes is needed for sufficient resolution of the phase

conjugation image signal and proposed an alternative way of using arrays of single-mode waveguides to avoid these constraints.

In this paper, four-wave mixing in a multimode waveguide that consists of a weakly Kerr-type nonlinear dielectric film bounded by semi-infinite linear dielectrics is investigated by a singular perturbation technique with multiple space scales.^{10,11} In order to apply the perturbation technique, we first introduce the perturbation parameter concerned with the nonlinear coefficients and the multiple space scales in the propagation directions. Substituting the expansion of the field and the multiple space scales into Maxwell's equations, we can get the governing equations to each order. The perturbation is carried up to the first order. From the solvability conditions for nontrivial solutions, the coupled-mode equations are derived. After applying the undepleted approximation, simpler coupled-mode equations are obtained. The coupling properties are evaluated numerically. When we consider four wave mixing in the waveguide structure, it is important that the phase conjugate wave is distinguished from the pump wave, because the direction of the phase conjugate wave and the pump wave are the same. In this paper, different modes and frequencies are adopted in order to distinguish the phase conjugate wave from the pump wave.

II. FORMULATION OF PROBLEM

Consider the symmetric slab waveguide with thickness $2d$ as shown in Fig. 1. The slab waveguide consists of a thin, optically nonlinear dielectric film bounded by semi-infinite linear dielectrics. The optically nonlinearity is of the Kerr type and self-focusing. Waves 1 and 2 identify to the pump waves and waves 3 and 4 are, respectively, the probe wave and phase conjugate wave. The geometry considered here corresponds to the collinear case. It is assumed that the four waves have the different frequencies $\omega_1 \sim \omega_4$ and different mode number, so it is possible to distinguish the conjugate

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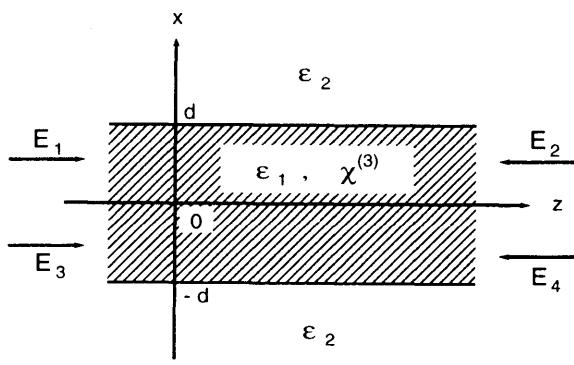


FIG. 1. Geometry of the problem.

wave from the other waves. All four waves are TE polarized. In this case, the wave equation is written as follows:

$$\nabla^2 E_y = \mu_0 \epsilon_0 \epsilon(x) \frac{\partial^2}{\partial t^2} E_y + \mu_0 \frac{\partial^2}{\partial t^2} P_{NL}, \quad (1)$$

where

$$\epsilon(x) = \begin{cases} \epsilon_2, & |x| > d \\ \epsilon_1, & |x| < d \end{cases} \quad (2)$$

and P_{NL} is the nonlinear polarization, which is expressed in the general case as follows:

$$P_{NL} = \epsilon_0 \sum_{l,m,n} D_3 \chi^{(3)}(\omega_l; \omega_l, \omega_m, \omega_n) E_l E_m E_n \times \exp \left[j \left(\sum \omega_l t - \sum \beta_l z \right) \right] \quad (3)$$

and $\chi^{(3)}$ is the nonlinear susceptibility and D_3 is the degeneracy factor.

In order to apply the perturbation technique^{10,11} we first introduce the perturbation parameter concerned with the nonlinear coefficients and the multiple space scales in the propagation direction $z_0 = z$, $z_1 = \delta z$. Then the electromagnetic field components are expanded by using the perturbation parameter as follows:

$$\Psi(x, z) = \sum_{m=0}^1 \delta^m \Psi^{(m)}(x, z_0, z_1, x_2), \quad (4)$$

where $\Psi(x, z)$ denotes the electromagnetic field components H_x, E_y, H_z . Substituting the expansion of the field (4) and the multiple space scales into Eq. (1) and equating the coefficients for each powers of the perturbation parameter to zero, we can get the following governing equations for each order of the perturbation:

$O(\delta^0)$:

$$\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial z_0^2} - \mu_0 \epsilon_0 \epsilon(x) \frac{\partial^2}{\partial t^2} \right) E_y^{(0)} = 0, \quad (5)$$

$O(\delta^1)$:

$$\begin{aligned} & \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial z_0^2} - \mu_0 \epsilon_0 \epsilon(x) \frac{\partial^2}{\partial t^2} \right) E_y^{(1)} \\ & = -2 \frac{\partial^2}{\partial z_0 \partial z_1} E_y^{(0)} + \mu_0 \frac{\partial^2}{\partial t^2} P_{NL}^{(0)}. \end{aligned} \quad (6)$$

The boundary conditions at the planes $x = \pm d$ require the continuity of the tangential electric and magnetic fields. In what follows, the perturbation solutions are investigated to each order.

III. PERTURBATION SOLUTIONS

A. $O(\delta^0)$

From Eq. (5), it is shown that the governing equation of the zero-order problem corresponds to the symmetric linear slab waveguide. In this paper, the following four guided waves are considered as the zero-order solutions:

$$E_y^{(0)} = \sum_{i=1}^4 A_i(z_1) \psi_i^{(0)}(x) \exp \{ j[\omega_i t + (-1)^i \beta_i z_0] \}, \quad (7)$$

where $A_1 \sim A_4$ are the complex amplitudes of $E_1 \sim E_4$ and $\psi_i^{(0)}(x)$ are the modal functions of the linear slab waveguide as follows:

$$\psi_i^{(0)}(x) = N_i \begin{cases} \frac{x}{|x|} \sin \alpha_i d e^{-\gamma_i(|x|-d)}, & |x| > d \\ \sin \alpha_i x, & |x| < d \end{cases} \quad i=1,2, \quad (8a)$$

$$\psi_i^{(0)}(x) = N_i \begin{cases} \cos \alpha_i d e^{-\gamma_i(|x|-d)}, & |x| > d \\ \cos \alpha_i x, & |x| < d \end{cases} \quad i=3,4, \quad (8b)$$

N_i is the normalization coefficient, which is defined in such way that $|A_i|^2$ equals the power carried by the guided wave, and α_i, γ_i satisfy the following dispersion equations:

$$\tan \alpha_i d = -\frac{\alpha_i}{\gamma_i} \quad i=1,2, \quad (9a)$$

$$\tan \alpha_i d = \frac{\gamma_i}{\alpha_i} \quad i=3,4. \quad (9b)$$

B. $O(\delta^1)$

Although waves other than the zero-order solutions Eq. (7) can be generated due to the nonlinear polarization Eq. (3), it is assumed that the order of the coupling to other waves is higher than the second order. This means that only the coupling among the four guided waves in Eq. (7) are considered.

Solving Eq. (6) by using Eq. (7), the first order solutions are given by

$$E_{y,i}^{(1)} = \phi_{p_i}^{(1)} + \phi_{g_i}^{(1)}, \quad (10)$$

where $\phi_{p_i}^{(1)}$ and $\phi_{g_i}^{(1)}$ are, respectively, the particular solution and the solution to the homogeneous equation as follows:

$$\phi_{pi}^{(1)} = \left(j\beta_i \frac{dA_i}{dz_1} U_i(x) + V_i(x, z_0, z_1) \right) \times \exp\{j[\omega_i t + (-1)^i \beta_i z_0]\} \quad i=1 \sim 4, \quad (11)$$

$$\phi_{gi}^{(1)} = \exp\{j[\omega_i t + (-1)^i \beta_i z_0]\} \times \begin{cases} C_i e^{-\gamma_i(|x|-d)}, & |x| > d \\ B_i \sin \alpha_i x, & |x| < d \end{cases} \quad i=1,2, \quad (12)$$

$$\phi_{gi}^{(1)} = \exp\{j[\omega_i t + (-1)^i \beta_i z_0]\} \times \begin{cases} E_i e^{-\gamma_i(|x|-d)}, & |x| > d \\ D_i \cos \alpha_i x, & |x| < d \end{cases} \quad i=3,4. \quad (13)$$

The expressions of the solutions $V_i(x, z_0, z_1)$ depend on the condition assumed. In this section, the degenerate case, which is one of the simplest cases, is shown although the general case can be obtained. In what follows, the solution with the frequencies ω_1 is shown. The other solutions with the frequencies ω_2 to ω_4 can be derived in the same way. From the phase matching condition, the nonlinear polarization $P_{NL}^{(0)}$ is given as follows:

$$P_{NL}^{(0)} = \epsilon_0 \chi^{(3)} \left(6A_3 A_4 A_2^* \psi_3 \psi_4 \psi_2 + 3 \sum_{i=1}^4 \sigma_{i1} |A_i|^2 A_i \psi_i^2 \psi_1 \right) \times \exp[j(\omega_1 t - \beta_1 z_0)], \quad (14)$$

where $\sigma_{11}=1$, $\sigma_{i1}=2(i \neq 1)$. By using Eqs. (14) and (6), the solution $U_1(x)$ and $V_1(x, z_0, z_1)$ are obtained by the usual mathematical methods as follows:

$$U_1 = \begin{cases} -N_1 \frac{x \sin \alpha_1 d}{\gamma_1} e^{-\gamma_1(|x|-d)}, & |x| > d \\ -N_1 \frac{x \cos \alpha_1 x}{\alpha_1}, & |x| < d \end{cases}, \quad (15)$$

$$V_1 = \begin{cases} 0, & |x| > d \\ -k^2 \chi^{(3)} \left\{ A_3 A_4 A_2^* N_3 N_4 N_2 F_1(x) + 3 \sum_{i=1}^4 G_{i1}(x) N_i^2 N_1 |A_i|^2 A_1 \right\}, & |x| < d \end{cases} \quad (16)$$

where

$$F_1(x) = 6[a_1 x \cos \alpha_1 x + a_2 \sin(\alpha_1 + 2\alpha_3)x + a_3 \times \sin(\alpha_1 - 2\alpha_3)x], \quad (17a)$$

$$a_1 = -\frac{1}{4\alpha_1}, \quad (17b)$$

$$a_2 = -\frac{1}{16\alpha_3(\alpha_1 + \alpha_3)}, \quad (17c)$$

$$a_3 = \frac{1}{16\alpha_3(\alpha_1 - \alpha_3)}, \quad (17d)$$

$$G_{11}(x) = \frac{\sin 3\alpha_1 x - 12\alpha_1 x \cos \alpha_1 x}{32\alpha_1^2}, \quad (17e)$$

$$G_{21}(x) = 2G_{11}(x), \quad (17f)$$

$$G_{31}(x) = \frac{1}{3}F_1(x), \quad (17g)$$

$$G_{41}(x) = G_{31}(x). \quad (17h)$$

The expressions for $U_i(x), V_i(x)$ ($i=2-4$) are listed in the Appendix.

Applying the boundary condition by using the zero-order dispersion equation (9), the set of the linear equations in the unknown coefficients E_1 and D_1 is obtained. The linear equations are singular, since the homogeneous parts satisfy the zero-order dispersion equation. Then, the solutions have finite amplitudes only when the following coupled-mode equations for A_1 are satisfied.

$$\begin{aligned} \frac{d}{dz_1} A_1 = & -j\mu_0 \epsilon_0 \omega_1^2 \chi^{(3)} \left(\frac{N_2 N_3 N_4}{N_1} A_3 A_4 A_2^* [\gamma_1 F_1(d) \right. \\ & \left. + F_1'(d)] + 3 \sum_{i=1}^4 (\gamma_i G_{i1}(d) + G_{i1}'(d)) N_i^2 |A_i|^2 A_1 \right) \\ & \times \left(\beta_1 \left(\frac{\sin \alpha_1 d}{\gamma_1} - \frac{\cos \alpha_1 d}{\alpha_1} \right) \cdot (1 + \gamma_1 d) \right)^{-1} \\ \equiv & -jf_1 A_3 A_4 A_2^* - j \sum_{i=1}^4 g_{i1} |A_i|^2 A_1 \end{aligned} \quad (18)$$

and the asterisk * indicates the complex conjugate and the prime denotes differentiation with respect to the argument.

By the same procedure, the coupled-mode equations for $A_2 \sim A_4$ can be obtained as follows:

$$\frac{d}{dz_1} A_2 = jf_2 A_3 A_4 A_1^* + j \sum_{i=1}^4 g_{i2} |A_i|^2 A_2, \quad (19)$$

$$\frac{d}{dz_1} A_3 = -jf_3 A_1 A_2 A_4^* - j \sum_{i=1}^4 g_{i3} |A_i|^2 A_3, \quad (20)$$

$$\frac{d}{dz_1} A_4 = jf_4 A_1 A_2 A_3^* + j \sum_{i=1}^4 g_{i4} |A_i|^2 A_4, \quad (21)$$

where the coefficients f_i, g_{i1} whose expressions are listed in the Appendix include the parameters of the nonlinearity, the waveguide width, and the frequency. The coupled-mode equations obtained at the above can be solved exactly. Karaguleff *et al.*⁷ used the simplest approximation to obtain the governing equation. In strictly speaking, their governing equations are not the coupled-mode equations, since the effect of the probe beam is not included. The expressions of Eqs. (18)–(21) seem to be almost the same as those for the slowly varying amplitude approximation. In this paper, the slowly varying amplitude approximation [$|d^2 A_i / dz^2| \ll |\beta_i (dA_i / dz)|$ ($i=1-4$)] is not applied. The amplitudes A_i are the function of the propagation direction z , and the deviation from the linear case. The behavior of the amplitudes is determined by the boundary condition. The results obtained here are the first order for the asymptotic expansion. If we carry up the higher order, we can obtain the coupled-mode equations precisely. In the linear directional coupler, the coupled-mode equations¹¹ obtained by the singular perturbation technique are in complete agreement with those¹² from the asymptotic expansion of the exact dispersion equa-

tion. So, the results in this paper are asymptotically correct in the limit of weak nonlinearity, since the wave equation and the boundary condition satisfy exactly.

In this paper, the undepleted approximation ($|A_1|^2, |A_2|^2 \gg |A_3|^2, |A_4|^2$) is applied in order to obtain the simple solution and make clear the physical phenomena. For the undepleted pump approximation, the coupled-mode equations lead to the following equations:

$$\frac{d}{dz_1} A_3^* = j f_3 A_1^* A_2^* A_4 + j (g_{13} |A_1|^2 + g_{23} |A_2|^2) A_3^*, \quad (22)$$

$$\frac{d}{dz_1} A_4 = j f_4 A_1 A_2 A_3^* + j (g_{14} |A_1|^2 + g_{24} |A_2|^2) A_4. \quad (23)$$

After a simplifying transformation of the parameter in Eqs. (23) and (22), we can get the following equations:

$$\frac{d}{dz_1} a_3^* = j \kappa a_4, \quad (24)$$

$$\frac{d}{dz_1} a_4 = j \kappa^* a_3^*, \quad (25)$$

where

$$A_3 = \sqrt{f_3} a_3(z_1) \exp[-j \{g_{13} |A_1|^2 + g_{23} |A_2|^2\} z_1], \quad (26)$$

$$A_4 = \sqrt{f_4} a_4(z_1) \exp[j \{g_{14} |A_1|^2 + g_{24} |A_2|^2\} z_1], \quad (27)$$

$$\kappa = \sqrt{f_3 f_4} A_1^* A_2^*. \quad (28)$$

When the boundary conditions $a_3(0)$ and $a_4(L)$ are given, the solutions to Eqs. (24) and (25) are obtained as follows:

$$a_3^* = d_1 e^{j|\kappa|z_1} + d_2 e^{-j|\kappa|z_1} \quad (29)$$

$$a_4 = \frac{|\kappa|}{\kappa} (d_1 e^{j|\kappa|z_1} - d_2 e^{-j|\kappa|z_1}), \quad (30)$$

where the coefficients d_1 and d_2 are

$$d_1 = \frac{1}{2 \cos|\kappa|L} \left(\frac{\kappa}{|\kappa|} a_4(L) + a_3(0) e^{-j|\kappa|L} \right) \quad (31a)$$

$$d_2 = \frac{-1}{2 \cos|\kappa|L} \left(\frac{\kappa}{|\kappa|} a_4(L) - a_3(0) e^{j|\kappa|L} \right) \quad (31b)$$

IV. NUMERICAL RESULTS

In this section, we present numerical results for the coupling coefficients and the field distributions. PTS is chosen as the nonlinear material because of its high nonlinear susceptibility. The nonlinear susceptibility $\chi^{(3)}$ of PTS is $1.8 \times 10^{-16} \text{ m}^2/\text{W}$, and $\sqrt{\epsilon_1} = 1.88$. The relative refractive index in the linear cladding is $\sqrt{\epsilon_2} = 1.55$, which corresponds to glass. The power of the pump waves assumed is $|A_1|^2 = |A_2|^2 = 10^8 \text{ W/m}$.

At first, the degenerate four wave mixing is considered. Figure 2 shows the coupling coefficients as a function of the waveguide width for various wavelengths. It is found that the coupling coefficients varies with the waveguide width and reaches a maximum at the suitable width. Comparing with plane wave coupling in an unbounded nonlinear medium, the coupling coefficient for the plane wave in the unbounded

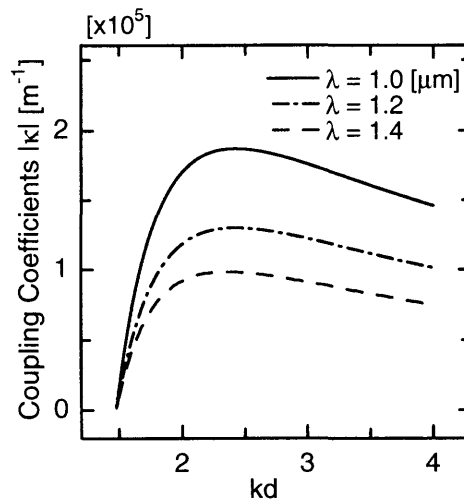


FIG. 2. Coupling coefficient for degenerate four wave mixing as functions of the normalized waveguide width.

medium is constant, whereas the maximum values of the coupling coefficients for the waveguide structure are about twice of that of the unbounded structure. This result shows that a waveguide structure is more suitable than an unbounded structure for a given input power, because the waveguide structure has the possibility to be smaller than the unbounded structure.

Figure 3 shows the coupling coefficients for the nearly degenerate four wave mixing as a function of the waveguide width for various changes in the normalized angular frequency. The actual expressions of the solutions for the nearly degenerate four wave mixing case can be obtained by the same procedure in the previous section. The wavelength λ is $1 \mu\text{m}$, and $\omega_1 = \omega_2 = \omega$, $\omega_3 = \omega - (c/d)\Omega$, $\omega_4 = \omega + (c/d)\Omega$ (c is the light velocity in free space). It is found that the maximum values of the coupling coefficients for nearly degenerate four wave mixing are smaller than those of the degenerate four wave mixing case, and the optimum width to give

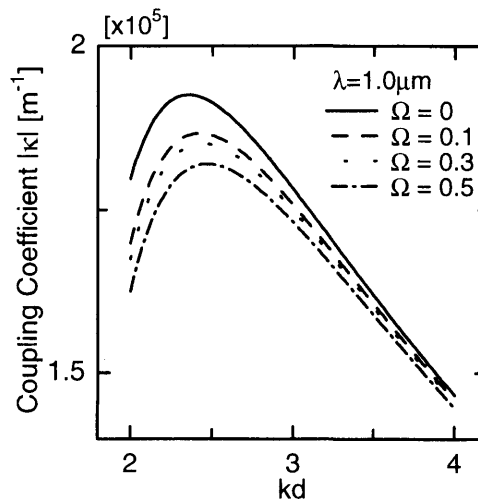


FIG. 3. Coupling coefficient for nearly degenerate four wave mixing as functions of the normalized waveguide width.

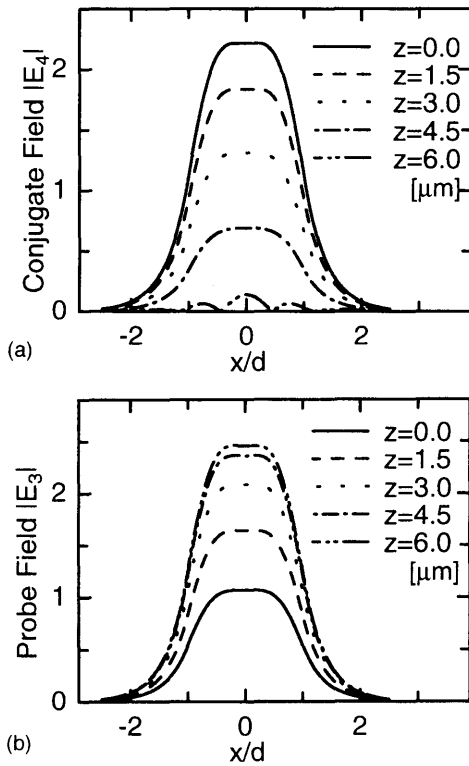


FIG. 4. The field distribution at the arbitrary z plane: (a) the conjugate wave, (b) the probe wave.

the maximum coupling coefficient deviates from that of the degenerate case ($\Omega=0$) as the difference in frequency Ω increases.

Figures 4(a) and 4(b) show the field distributions for the phase conjugate wave and probe wave at an arbitrary plane a distance z into the medium, respectively. The sample length L is $6 \mu\text{m}$, the wavelength λ is $1 \mu\text{m}$, and $d=0.385 \mu\text{m}$. $A_4(L)=0$ is adopted as the initial condition. From these figures, it is found that the phase conjugate field is increasing and the probe wave is decreasing as the waves propagate, respectively. The power for the phase conjugate wave is obtained from the pump wave. It is found from these figures that the efficiency, which is defined by the power ratio of the phase conjugate wave to the probe wave at $z=0$, is about 220% in this case. This efficiency seems to be large for such a thin sample. As we can see from Eq. (28), the efficiency is a function of the pump power. If the pump power is small, the efficiency also becomes small. In this paper, the large pump power is adopted in order to obtain the large nonlinear effect. In the Fig. 4(a), the conjugate field at $z=L$ is not zero. This field equals the first order solution E_y^I of the conjugate wave. The initial condition $A_4(L)=0$ leads to that the zero-order solution with a zero conjugate wave. It is difficult to obtain the initial condition that the total field of the conjugate wave is zero.

V. CONCLUSIONS

Four-wave mixing in a multimode slab waveguide for the undepleted pump approximation has been investigated by using the singular perturbation technique.

From the numerical results, it is found that the coupling coefficients have the maximum (optimum) for the suitable waveguide width. For the nearly degenerate four wave mixing, it is shown that the maximum values of the coupling coefficients become smaller and the optimum width required for the maximum coupling coefficient deviates from that of the degenerate case as the difference in frequencies increases. The results in this paper are asymptotically correct in the limit of weak nonlinearity, since the wave equation and the boundary condition satisfy exactly. The analytical steps of the technique presented in this paper is straightforward and the coupled-mode equations are obtained systematically.

The exact solutions to Eqs. (18)–(21) will be presented elsewhere.

ACKNOWLEDGMENT

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APPENDIX

In this section, the coefficients in the text are listed.

$$U_2 = -U_1, \quad (\text{A1})$$

$$V_2 = \begin{cases} 0, & |x| > d \\ -k^2 \chi^{(3)} \left\{ A_3 A_4 A_1^* N_3 N_4 N_1 F_2(x) \right. \\ \left. + 3 \sum_{i=1}^4 G_{i2}(x) N_i^2 N_2 |A_i|^2 A_2 \right\}, & |x| < d \end{cases}, \quad (\text{A2})$$

where

$$F_2(x) = F_1(x), \quad (\text{A3a})$$

$$G_{12}(x) = 2G_{11}(x), \quad (\text{A3b})$$

$$G_{22}(x) = G_{11}(x), \quad (\text{A3c})$$

$$G_{i2}(x) = G_{i1}(x) \quad i=3,4, \quad (\text{A3d})$$

$i=3$

$$U_3 = \begin{cases} -N_3 \frac{|x| \cos \alpha_3 d}{\gamma_3} e^{-\gamma_3(|x|-d)}, & |x| > d \\ N_3 \frac{x \sin \alpha_3 x}{\alpha_3}, & |x| < d \end{cases}, \quad (\text{A4})$$

$$V_3 = \begin{cases} 0, & |x| > d \\ -k^2 \chi^{(3)} \left\{ A_1 A_2 A_4^* N_1 N_2 N_4 F_3(x) \right. \\ \left. + 3 \sum_{i=1}^4 G_{i3}(x) N_i^2 N_3 |A_i|^2 A_3 \right\}, & |x| < d \end{cases}, \quad (\text{A5})$$

where

$$F_3(x) = 6 \{ c_1 x \sin \alpha_3 x + c_2 \cos(2\alpha_1 + \alpha_3)x + c_3 \cos(2\alpha_1 - \alpha_3)x \}, \quad (\text{A6a})$$

$$c_1 = \frac{1}{4\alpha_3}, \quad (\text{A6b})$$

$$c_2 = \frac{1}{16\alpha_1(\alpha_1 + \alpha_3)}, \quad (\text{A6c})$$

$$c_3 = \frac{1}{16\alpha_1(\alpha_1 - \alpha_3)}, \quad (\text{A6d})$$

$$G_{13}(x) = \frac{1}{3}F_3(x), \quad (\text{A6e})$$

$$G_{23}(x) = G_{13}(x), \quad (\text{A6f})$$

$$G_{33}(x) = \frac{12\alpha_3x \sin \alpha_3x - \cos 3\alpha_3x}{32\alpha_3^2}, \quad (\text{A6g})$$

$$G_{43}(x) = 2G_{33}(x), \quad (\text{A6h})$$

$i=4$

$$U_4 = -U_3 \quad (\text{A7})$$

$$V_4 = \begin{cases} 0, & |x| > d \\ -k^2\chi^{(3)} \left\{ A_1A_2A_3^*N_1N_2N_3F_4(x) \right. \\ \left. + 3 \sum_{i=1}^4 G_{i4}(x)N_i^2N_4|A_i|^2A_4 \right\}, & |x| < d \end{cases}, \quad (\text{A8})$$

where

$$F_4(x) = F_3(x), \quad (\text{A9a})$$

$$G_{i4}(x) = G_{i3}(x) \quad i=1,2, \quad (\text{A9b})$$

$$G_{34}(x) = 2G_{33}(x), \quad (\text{A9c})$$

$$G_{44}(x) = G_{33}(x). \quad (\text{A9d})$$

The coefficients f_i and g_{ij} are given as follows:

$$f_2 = f_1, \quad (\text{A10})$$

$$f_3 = \mu_0\epsilon_0\omega_3^2\chi^{(3)} \left(\frac{N_1N_2N_4}{N_3} [\gamma_3F_3(d) + F_3'(d)] \right) \cdot \left[\beta_3(\gamma_3d+1) \left(\frac{\cos \alpha_3d}{\gamma_3} + \frac{\sin \alpha_3d}{\alpha_3} \right) \right]^{-1}, \quad (\text{A11})$$

$$f_4 = f_3, \quad (\text{A12})$$

$$g_{i2} = g_{i1}, \quad (\text{A13})$$

$$g_{i3} = 3k^2\chi^{(3)}N_i^2 \{ \gamma_3G_{i3} + G_{i3}' \} \cdot \left[\beta_3(\gamma_3d+1) \left(\frac{\cos \alpha_3d}{\gamma_3} + \frac{\sin \alpha_3d}{\alpha_3} \right) \right]^{-1}, \quad (\text{A14})$$

$$g_{i4} = g_{i3}. \quad (\text{A15})$$

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