Exact results on optical bistability with surface plasmons in layered media

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Optical bistability with surface plasmons in layered structures on a nonlinear substrate is studied using Leung's exact results for the nonlinear p-polarized surface waves. Explicit results for the bistable behavior of the reflected field are given for two different geometries involving excitation of short-range and long-range surface plasmons. The excitation of long-range plasmons results in a much lower bistability threshold.

I. INTRODUCTION

Excitation of surface plasmons at metal surfaces is known to be important in a large class of problems[1,2] involving linear and nonlinear interactions between electromagnetic fields and the atoms or molecules adsorbed on the metal surface. Similarly nonlinear processes in various materials can be enhanced by using suitable geometries involving metal films and nonlinear materials. The efficiency or cross section for a nonlinear process is, in general, enhanced if surface plasmons are excited. The enhancement is due to large local fields[3] present near the metal surface if surface plasmons are excited. In a very interesting paper Wysin *et al.*[4] suggested that this local field enhancement can be quite useful in the study of the bistable behavior of the light reflected from a nonlinear interface. They presented a detailed analysis of the fields produced in the Kretschmann geometry. Martinot *et al.*[5] suggested different geometrical arrangements and have now observed[6] optical bistability with surface plasmons. The nonlinearity in the experiment of Martinot *et al.* is of thermal origin. Various approximate[4,5,7,8] schemes for calculating the reflected fields in the geometry involving a metal film on a nonlinear substrate exist. The surface waves in the nonlinear medium make the nonlinear medium inhomogeneous since the effective dielectric function of the nonlinear medium depends on the intensity of the field in the medium. The intensity of the surface wave decays as one moves away from the interface. This electric-field-induced inhomogeneity in the medium is the major source of difficulty in as far as an exact calculation of the fields is concerned. In the case of weak nonlinearity such that one can neglect the transverse spatial derivatives of the field-dependent dielectric function, one can use an approximate nonlinear wave equation for the magnetic field.[9] In a recent paper Leung[10] showed how the exact calculation of the p-polarized fields in the nonlinear medium can be reduced to quadratures and he presented exact results for the nonlinear surface polaritons. The purpose of this paper is twofold—(a) to use the exact analysis of Leung in the context of optical bistability with surface plasmons and to check the validity of the earlier theoretical calculations and (b) to present results for optical bistability with long-range surface plasmons.[11] The long-range surface plasmons (LRSP's) are specially attractive since the local field enhancement is much higher[12] when LRSP's are excited. Note that this extra enhancement has been extensively used[13] in the study of the nonlinear optical processes such as harmonic generation.

In this paper we present the exact results for bistability with surface plasmons in layered structures. Thus in Sec. II we recall some of the results of Leung which are used subsequently in our discussion of optical bistability with surface plasmons. In Sec. III we obtain the exact expression for the reflection coefficient from a multilayered structure on a nonlinear Kerr substrate. In Sec. IV we consider the Kretschmann geometry and compare the exact results thus obtained with the previous approximate[4,7] ones. We show that the results of Ref. 4 underestimate, whereas our previous calculations overestimate, the bistability threshold. Moreover, we present the exact numerical results for bistability with LRSP's and compare thresholds for bistability in the two cases when short- and long-range surface plasmons are excited.

II. EXACT SOLUTIONS FOR P-POLARIZED SURFACE WAVES IN NONLINEAR MEDIUM

Consider a semi-infinite isotropic nonlinear Kerr medium with dielectric function $\epsilon_f=\epsilon_{f0}+\alpha\,|E\,|^2$ having the plane interface along $z=0$. Assuming the x dependence of the fields to be $\sim e^{ik_x x}$ and with no variation along y, we can write Maxwell's equation for p-polarized field components in the form

$$B_y'(\xi)=i\epsilon_f E_x(\xi)\ , \tag{2.1a}$$

$$\eta B_y(\xi)=-\epsilon_f E_z(\xi)\ , \tag{2.1b}$$

$$E_x'(\xi)-i\eta E_z(\xi)=iB_y(\xi)\ , \tag{2.1c}$$

where

$$\xi=\frac{\omega z}{c},\quad \eta=\frac{k_x c}{\omega}\ .$$

The system of equations (2.1) can be reduced to a second-order nonlinear differential equation of the form[10]

$$\left[\frac{B_y'}{\epsilon_f}\right]'=\left[\frac{\eta^2}{\epsilon_f}-1\right]B_y\ , \tag{2.2}$$

which with additional relations

$$\epsilon_f = \epsilon_{f0} + \alpha |E|^2 , \tag{2.3a}$$

$$|E|^2 = |E_x|^2 + |E_z|^2 = + \left|\frac{B_y'}{\epsilon_f}\right|^2 + \eta^2 \left|\frac{B_y}{\epsilon_f}\right|^2 , \tag{2.3b}$$

can be solved for B_y and its derivative B_y'. For B_y and B_y' we get the following expressions:

$$B_y^2 = \frac{\epsilon_f}{2\eta^2 - \epsilon_f} (\epsilon_f I - J) , \tag{2.4a}$$

$$(B_y')^2 = \epsilon_f^2 I - \eta^2 B_y^2 , \tag{2.4b}$$

where

$$I = \frac{\epsilon_f - \epsilon_{f0}}{\alpha}, \quad J = \frac{(\epsilon_f - \epsilon_{f0})^2}{2\alpha} . \tag{2.4c}$$

In the next section we look at the boundary-value problem where by satisfying the relevant boundary conditions, we obtain the expression for the reflection coefficient from a multilayered medium on a nonlinear substrate.

III. REFLECTION FROM A MULTILAYERED STRUCTURE ON A NONLINEAR SUBSTRATE

Consider a stratified medium consisting of N layers occupying a region $-h < z < 0$ on a nonlinear Kerr substrate spanning $z > 0$ (see Fig. 1). Let all the layers occupying $z < 0$ be linear. Let a p-polarized plane monochromatic wave be incident at an angle θ on the interface $z = -h$ from the left-hand side. The magnetic and electric field amplitudes at the interface $z = 0$ on the linear-medium side can be calculated from a knowledge of the characteristic matrix[14] of the stratified medium:

$$\begin{bmatrix} B_y \\ E_x \end{bmatrix}_{z=-h^+} = M \begin{bmatrix} B_y \\ E_x \end{bmatrix}_{z=0^-} . \tag{3.1}$$

Here M is the characteristic matrix of the stratified medium occupying $-h < z < 0$ and is given by

$$M = M_1(d_1) M_2(d_2) \cdots M_N(d_N) , \tag{3.2}$$

where M_j is the characteristic matrix of the jth layer with width d_j $(j = 1, N)$:

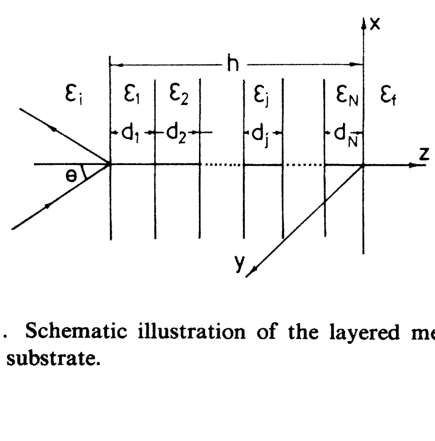

FIG. 1. Schematic illustration of the layered medium on a nonlinear substrate.

$$M_j(d_j) = \begin{bmatrix} \cos(k_{jz} d_j) & -\frac{i}{p_j}\sin(k_{jz} d_j) \\ -ip_j \sin(k_{jz} d_j) & \cos(k_{jz} d_j) \end{bmatrix} . \tag{3.3}$$

Here

$$p_j = \frac{k_{jz}}{k_0 \epsilon_j} , \quad k_0 = \frac{\omega}{c}$$

is the z component of the wave vector and ϵ_j is the dielectric k_{jz} constant of the jth medium. k_{jz} is related to the angle of incidence θ by the following expression:

$$k_{jz} = k_0 (\epsilon_j - \epsilon_i \sin^2\theta)^{1/2} ,$$

where ϵ_i is the refractive index of the medium from which the wave is incident.

The field components B_y and E_x at $z = -h$ are related to the amplitudes of the incident and reflected magnetic fields B_+ and B_-, respectively, as follows:

$$B_y = B_+ + B_- , \quad E_x = p_i (B_+ - B_-) , \tag{3.4}$$

where subscript i refers to the medium from which the field is incident.

Making use of the results of the preceding section, i.e., Eq. (2.4), we can write

$$\begin{bmatrix} B_y \\ E_x \end{bmatrix}_{z=0^+} = \begin{bmatrix} B_{y,\mathrm{NL}}(0) \\ E_{x,\mathrm{NL}}(0) \end{bmatrix} , \tag{3.5}$$

where

$$B_{y,\mathrm{NL}}(0) = \left[\frac{\epsilon_f(0)}{2\eta^2 - \epsilon_f(0)} [\epsilon_f(0) I(0) - J(0)] \right]^{1/2} , \tag{3.5a}$$

$$E_{x,\mathrm{NL}}(0) = i \left[I(0) - \eta^2 \left[\frac{B_{y,\mathrm{NL}}(0)}{\epsilon_f(0)} \right]^2 \right]^{1/2} , \tag{3.5b}$$

$$I(0) = E^2(0), \quad J(0) = \frac{\alpha [E(0)]^4}{2} . \tag{3.5c}$$

We now require continuity of the tangential field components B_y and E_x at $z = 0$:

$$\begin{bmatrix} B_y \\ E_x \end{bmatrix}_{z=0^-} = \begin{bmatrix} B_y \\ E_x \end{bmatrix}_{z=0^+} . \tag{3.6}$$

Equations (3.1)–(3.6) yield $B_\pm$ as parametrically given functions of $I(0)$:

$$B_\pm = \tfrac{1}{2} \{ [m_{11} B_{y,\mathrm{NL}}(0) + m_{12} E_{x,\mathrm{NL}}(0)] \pm \frac{1}{p_i} [m_{21} B_{y,\mathrm{NL}}(0) + m_{22} E_{x,\mathrm{NL}}(0)] \} , \tag{3.7}$$

Here $m_{k,l}$ $(k,l = 1,2)$ are the elements of M. Introducing $\widetilde{B}_{\mathrm{NL}}$ and $\widetilde{E}_{\mathrm{NL}}$ in place of $B_{y,\mathrm{NL}}(0)$ and $E_{x,\mathrm{NL}}(0)$ by

$$\widetilde{B}_{\mathrm{NL}} = \frac{B_{y,\mathrm{NL}}(0)}{[I(0)]^{1/2}}, \quad \widetilde{E}_{\mathrm{NL}} = \frac{E_{x,\mathrm{NL}}(0)}{[I(0)]^{1/2}} , \tag{3.8}$$

one can rewrite (3.7) as

$$\alpha|B_\pm|^2=(\tfrac{1}{4})\alpha I\,|(m_{11}\tilde{B}_{\rm NL}+m_{12}\tilde{E}_{\rm NL})\pm\frac{i}{p_i}(m_{21}\tilde{B}_{\rm NL}+m_{22}\tilde{E}_{\rm NL})|^2\,, \tag{3.9}$$

$$\tilde{B}_{\rm NL}=\left[\frac{(\epsilon_{f0}+\alpha I)\left[\epsilon_{f0}+\frac{\alpha I}{2}\right]}{2\eta^2-(\epsilon_{f0}+\alpha I)}\right]^{1/2}, \tag{3.9a}$$

$$\tilde{E}_{\rm NL}=i\left[1-\frac{\eta^2\left[\epsilon_{f0}+\frac{\alpha I}{2}\right]}{(\epsilon_{f0}+\alpha I)[2\eta^2-(\epsilon_{f0}+\alpha I)]}\right]^{1/2}. \tag{3.9b}$$

In (3.9) we have written I instead of $I(0)$. Henceforth αI is treated as a parameter and $\alpha|B_+|^2$, $\alpha|B_-|^2$, and corresponding $R=|B_-/B_+|^2$ are calculated for given values of αI. It will be shown in the next section that R versus $\alpha|B_+|^2$ curves show hysteresis when the system has a positive (negative) angle detuning from the surface plasmon resonance and the nonlinearity constant α is positive (negative).

In a previous work[8] we had used the approximate nonlinear wave equation for the magnetic field

$$\frac{d^2B_y}{dz^2}=k_0^2(g^2-\alpha'|B_y|^2)B_y\,,\qquad g^2=\epsilon_i\sin^2\theta-\epsilon_{f0}\,, \tag{3.10}$$

with the solution

$$B_y=\left[\frac{2}{\alpha'}\right]^{1/2}\frac{g\exp[ic+izk_0(\epsilon_{f0})^{1/2}\sin\theta]}{\cosh k_0 g(z_0-z)}\,, \tag{3.11}$$

where z_0 and c are constants. Using Eq. (3.10) with solutions (3.11) we found out that the reflected and incident intensities can be expressed as

$$U_{r,i}=\frac{U_t}{4}\left|\frac{1}{1+U_t}\left[m_{11}\pm m_{21}\frac{k_0\epsilon_i}{k_{iz}}\right]-\frac{ig}{\epsilon_{f0}}\frac{1-U_t}{1+U_t}\left[m_{12}\pm m_{22}\frac{k_0\epsilon_i}{k_{iz}}\right]\right|^2, \tag{3.12}$$

where

$$U_i=\frac{\alpha'}{8g^2}|B_+|^2,\quad U_r=\frac{\alpha'}{8g^2}|B_-|^2\,,\qquad U_t=\frac{\alpha'}{8g^2}|B_t|^2\,. \tag{3.13}$$

Here $B_\pm$ are the incident and reflected fields as before and B_t is the transmitted field.

IV. EXACT NUMERICAL RESULTS ON OPTICAL BISTABILITY WITH SHORT- AND LONG-RANGE SURFACE PLASMONS

The general idea underlying optical bistability in resonant systems is that the system under consideration should possess a sharp resonance, the location of which depends on field-dependent parameters. In a slightly detuned system a rise in the intensities may affect corresponding changes in these parameters thereby causing the system to "sweep over" the resonance. For appropriate detuning and parameter values this may result in a functional relation between the reflected (or transmitted) field and the incident field which is multivalued. This in general leads to hysteresis. This is true for various systems such as nonlinear vibrators[15] and nonlinear Fabry Perot resonators.[16]

Keeping the aforementioned in mind we study the linear characteristics namely the R versus θ dependence where θ is the angle of incidence. Detuning the system slightly we examine the behavior for increasing intensities. We present results for two important geometries so that different types of surface plasmons can be excited.

A. Optical bistability with surface plasmons in Kretschmann geometry

We first consider the excitation of surface plasmons in the Kretschmann geometry and the role of surface plasmons in the bistable behavior of the reflected field. The nonlinear medium is taken to be CS_2. This is the system previously studied by Wysin *et al.*[4] and by us using various approximation schemes.[8] We use the same set of parameters as those used by Wysin *et al.*, i.e., we use $\epsilon_i=3.6$, $\epsilon_1=-57.8+i0.6$ at $\lambda=1.06$ μm; $\epsilon_{f0}=2.25$.

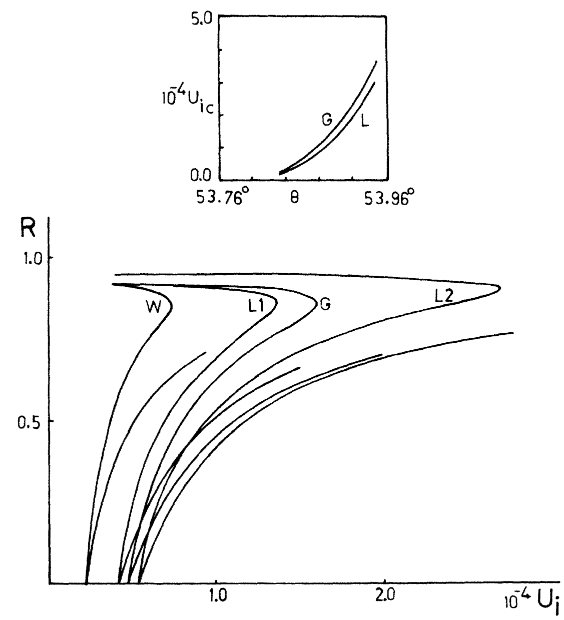

FIG. 2. Reflection coefficient as a function of the incident field intensity in Kretschmann geometry. Curves marked W and G are from Ref. 4 and Ref. 7, respectively. Curves marked $L1$ and $L2$ are the exact results for different angles of incidence, namely, $\theta=53.90°$ ($L1$) and $\theta=53.94°$ ($L2$). The inset gives the switching intensity as a function of the angle of incidence. G—from Ref. 7 and L—exact result.

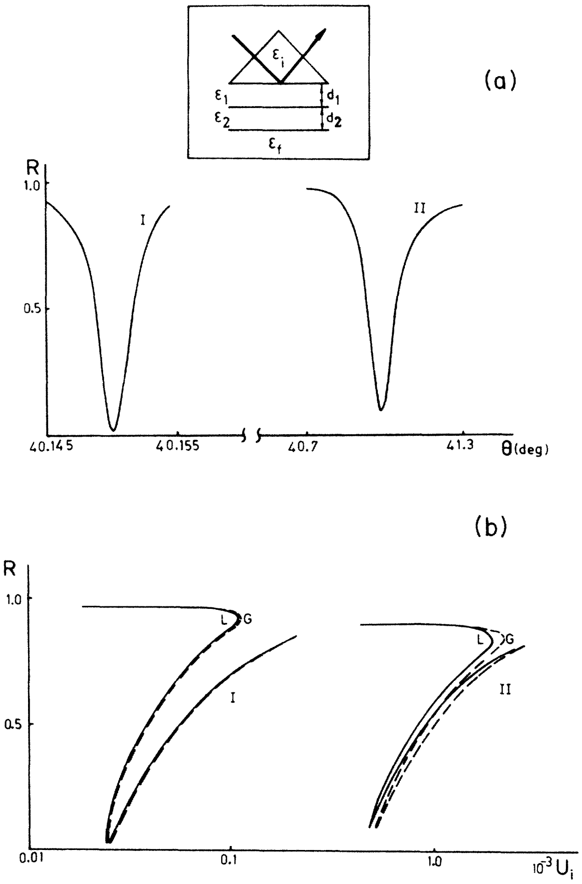

FIG. 3. (a) Reflection coefficient as a function of θ for low intensities in Sarid configuration (prism-index matching liquid—Ag-CS_2). The inset shows the geometry with medium parameters $\epsilon_i=6.145$; $\epsilon_1=\epsilon_{f0}=2.54$, $\epsilon_2=-67.03+2.44i$, and $\lambda=10\,600$ Å. CS_2 parameters are taken from the work of Hellwarth (Ref. 19). Curve marked I corresponds to LRSP with $d_1=35\,000$ Å and $d_2=160$ Å, whereas II to SP with $d_1=100$ Å and $d_2=500$ Å. (b) Reflection coefficient as a function of the normalized intensity U_i in Sarid configuration. Parameters used are the same as in (a). Curve marked I correspond to LRSP and II to SP: ——, exact results; — — —, approximate results using (3.12).

The reflectivity as a function of the incident field intensity U_i, defined by[17]

$$U_i=\frac{\alpha\,|B_+|^2}{8\epsilon_{f0}(\epsilon_i\sin^2\theta-\epsilon_{f0})}\ , \tag{4.1}$$

is shown in Fig. 2; we also show in this figure the results obtained in previous works. The inset in the figure gives the switching intensities as a function of the angle of incidence. The results for the switching intensities are also compared with those obtained previously using the solution (3.12). It is clear from this figure that the approximate solution of Ref. 4 underestimates and the solution of Ref. 7 somewhat overestimates the threshold intensities.

B. Optical bistability with long-range surface plasmons

We next consider what happens if the metal film is very thin. We consider the geometry used by Sarid and coworkers.[11,13] It is known that in this geometry it is possible to excite both long- and short-range surface plasmons. The long-range surface plasmons (LRSP's) are known to produce much higher enhancement of the local fields,[12] i.e., fields near the metal surface. Therefore the bistability threshold is expected to be much lower due to higher effective fields in the Kerr medium. The results of our numerical calculations for this geometry are presented in Fig. 3. We choose two sets of parameter values to make the system (a) pseudosymmetric so that LRSP's can be excited; (b) highly symmetric where only surface plasmons (SP's) (short range) are excited. Before studying the bistability in such cases one needs to have the detailed structure of the linear reflectivity in these two cases so that the positions and widths of the resonances are known. Figure 3(a) exhibits these resonances in the linear case. One finds the resonance at $\theta_{\mathrm{LRSP}}=40.1501°$ with a half-width at half maximum (which is inverted) $\Delta=0.0035°$ for (a) and $\theta_{\mathrm{SP}}=40.985°$ with half-width at half maximum (which is inverted) $\Delta=0.15°$ for (b). For computing the bistable behavior, we set the initial incident angle at detuning equal to 2Δ. Figure 3(b) shows the bistable character of the reflected field in the two cases. A comparison of the threshold intensities for the two cases clearly shows that one can lower the threshold by at least one order of magnitude if one is using geometries so that long-range surface plasmons are excited. We also show in Fig. 3(b) the results obtained using the approximate hyperbolic secant solution [Eq. (3.12)]. The approximate solutions are strikingly close to the exact solutions for the reflected field for the case (a).

V. CONCLUSIONS

In conclusion we have presented the exact results for optical bistability with surface plasmons in layered media. Explict numerical results have been given for the Kretschmann and Sarid geometries. Field-induced inhomogeneities are fully taken into account. Our analysis reveals that LRSP bistability occurs at a much lower intensity threshold than short-range surface plasmon bistability. We have analyzed the case when the angle of incidence is fixed and the intensity is varied. It has been pointed out that a wave-guide system on a nonlinear substrate can show angular as well as frequency bistability.[18] We hope to present an analysis of this system, in terms of the exact solutions of Leung, elsewhere.

ACKNOWLEDGMENT

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[1]See for example, articles by P. F. Liao, T. F. Heing, H. W. K. Tom, Y. R. Shen, A. Wokaun, H. P. Lutz, and A. P. King, in *Surface Studies with Lasers,* edited by F. R. Aussenegg, A. Leitner, and M. E. Lippitsch (Springer, Berlin, 1983).

[2]See for example, articles by A. A. Maradudin and E. Burstein, in *Surface Polaritons,* edited by V. M. Agranovich and D. Mills (North-Holland, Amsterdam, 1982).

[3]H. Raether, in *Surface Polaritons,* Ref. 2, p. 331.

[4]G. M. Wysin, H. J. Simon, and R. T. Deck, Opt. Lett. **6**, 30 (1981).

[5]P. Martinot, S. Laval, and A. Koster, J. Phys. **45**, 597 (1984); P. Martinot, A. Koster, S. Laval, and W. Carvalho, Appl. Phys. **B 29**, 172 (1982).

[6]P. Martinot, A. Koster, and S. Laval, IEEE J. Quantum Electron **QE-21**, 1140 (1985).

[7]S. Dùtta Gupta and G. S. Agarwal, J. Opt. Soc. Am. **B3**, 236 (1986).

[8]G. S. Agarwal and S. Dutta Gupta, in *Optical Bistability 3,* edited by H. M. Gibbs, P. Mandel, N. Peyghambarian, and S. D. Smith (Springer, Berlin 1986), p. 117; some very preliminary results on bistability with long-range surface plasmons were presented here. Figure 3(b) of the present paper gives better results.

[9]G. I. Stegeman and C. T. Seaton, Opt. Lett. **9**, 235 (1984).

[10]K. M. Leung, Phys. Rev. A **32**, 5093 (1985).

[11]D. Sarid, Phys. Rev. Lett. **47**, 1927 (1981).

[12]S. Sarid, R. T. Deck, A. E. Craig, R. K. Hickernell, R. S. Jameson, and J. J. Fasano, Appl. Opt. **21**, 3993 (1982); G. S. Agarwal, Phys. Rev. B **31**, 3534 (1985).

[13]J. C. Quail, J. G. Rako, H. J. Simon, and R. J. Deck, Phys. Rev. Lett. **50**, 1987 (1983); D. Sarid, R. T. Deck, and J. J. Fasano, J. Opt. Soc. Am. **72**, 1345 (1982); R. T. Deck and D. Sarid, *ibid.* **72**, 1613 (1982).

[14]M. Born and E. Wolf, *Principles of Optics* (Macmillan, New York, 1965), p. 62.

[15]V. V. Migulin, V. E. Medvegev, E. P. Mustel, and V. N. Parigin, *Osnovi Teorii Kolebanii* (Nauka, Moscow, 1978), p. 112; J. J. Stoker, *Nonlinear Vibrations in Mechanical and Electrical Systems* (Interscience, New York, 1950), p. 83.

[16]F. S. Felber and J. H. Marburger, Appl. Phys. Lett. **28**, 731 (1976); J. H. Marburger and F. S. Felber, Phys. Rev. A **17**, 335 (1978).

[17]We have used somewhat odd normalization for incident intensity. This is for comparison with the results of Ref. 7.

[18]P. Arlot, E. Pic, R. Reinisch, and G. Vitrant, in *Optical Bistability 3* (Ref. 8).

[19]R. W. Hellwarth, in *Progress in Quantum Electronics* (Pergamon, London, 1977), Vol. 5, pp. 1—68.