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Optical response of a nonlinear dielectric film

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We present a theoretical discussion of the optical reflectivity and transmissivity of a dielectric slab of arbitrary thickness, with index of refraction that contains a term proportional to the intensity of the optical wave in the slab. An exact solution is found for the problem, in terms of a single parameter whose value is determined through a straightforward numerical search procedure. We present numerical studies of the power dependence of the transmissivity, with emphasis on bistability, and the multivalued behavior of the transmissivity considered as a function of incident power.

I. INTRODUCTION

For many years, there has been very great interest in the theoretical and experimental study of the propagation of electromagnetic radiation in materials whose index of refraction varies with the amplitude or intensity of the radiation field. Of particular interest is the regime of strong nonlinearity, where a cavity filled with a nonlinear medium may exhibit bistability, or other dramatic manifestations of the nonlinear response characteristic.

The description of the interface between a linear medium and a nonlinear medium has also been the focus of a number of studies. In the recent literature, these have been descriptions of surface polaritons which propagate along the interface between a linear medium, and a nonlinear dielectric.[1] The properties of these waves may be modified substantially by the nonlinear response of the one partner to the interface; one may also "bind" waves to the interface by the nonlinearity itself, though it is not clear if the conditions required can be met by real materials.[2]

In the mathematical models of nonlinear surface polaritons that admit analytic solutions, the basic equation reduces to that which describes a one-dimensional field with ϕ^4 nonlinearity; simple well-known closed-form "kink" solutions exist, provided the field amplitude vanishes at infinity, a condition realized in the description of a surface polariton localized at the interface between a semi-infinite nonlinear medium, and a structure with linear response.

If we consider electromagnetic propagation in a nonlinear film of finite thickness, the condition just described cannot be invoked, and no closed-form analytic solutions of the relevant wave equation exist. This paper is devoted to the analysis of the simplest problem involving the nonlinear optical response of a nonlinear film: The calculation of the reflectivity or transmissivity of a thin film with index of refraction dependent on field intensity, with the incident beam normally incident on the sample. This problem is of interest because of its close relationship to the Fabry-Perot interferometer, which exhibits bistability when a nonlinear medium is incorporated into the cavity. We are unaware of any simple exact solution of the optical reflectivity of the nonlinear thin film, for the case where the film thickness is comparable to the optical wavelength. The slowly varying envelope approximation has been applied to the limit where the thickness of the nonlinear medium is large compared to the wavelength,[3] a limit appropriate to the Fabry-Perot device. Marburger and Felber[4] address the problem considered here, but simplified the analysis very considerably by imposing boundary conditions which suppose the nonlinear medium is bounded by perfect mirrors. Band has presented a brief discussion of an exact solution to the problem by a rather complicated method.[5] In the end our analysis can be implemented very simply, as the reader will appreciate.

We examine the case where the thin film is surrounded by vacuum (or linear dielectric media). The index of refraction of the film is supposed to vary with field strength, according to the law $n^2(I)=n^2(1+\lambda I)$, where $I=|E|^2$ is the field intensity, a behavior appropriate to a liquid, or solid material with an inversion center. In essence, we assume it is the dielectric constant which contains a term proportional to the intensity. The solution of the relevant wave equation can be expressed in terms of certain elliptic integrals,[6] which cannot be expressed in terms of elementary functions when, as is the case here, we have a finite rate of transport of energy normal to the film surface.

In general, as we shall see, the solution of the nonlinear wave equation may be written in terms of four parameters (the same number required in the general solution of the linear problem), which are to be deduced from the electromagnetic boundary conditions applied to each surface. Since some of these constants are imbedded within the elliptic integral in a nontrivial manner, the problem of solving for the reflectivity is formidable. We show here how three of the four parameters may be eliminated. The analysis then reduces to the search for a single real number consistent with the boundary conditions and, in fact, there are bounds on this number. Once the appropriate elliptic function is evaluated, a numerical solution of the problem is then readily achieved.

II. GENERAL DISCUSSION

A. Basic wave equation

Our interest is in the geometry illustrated in Fig. 1. A plane electromagnetic wave of frequency ω, with electric vector parallel to the x axis is incident on a film of thickness d. The wave vector in vacuum is $k_0=\omega/c$, and R is the amplitude of the reflected wave, measured with respect to E_0. Similarly T is the amplitude of the transmitted wave, expressed as a fraction of that of the incident wave. Throughout the discussion, we only need consider the one Cartesian component of the electric field parallel to the x axis. We ignore harmonic generation, since higher harmonics will have small amplitude in the absence of phase matching, and confine our attention to the propagation of the component of the wave frequency ω. In the film, the electric field obeys

$$\frac{d^2E}{dz^2}+\frac{\omega^2}{c^2}n^2(1+\lambda\,|E\,|^2)E=0\ , \tag{2.1}$$

where the nonlinear coefficient λ may be either positive or negative.

In this paper, both the linear index of refraction and λ are assumed real. Thus, we ignore absorption, an assumption appropriate to the thin films explored in our numerical calculations, in appropriate spectral regimes. Explicit inclusion of absorption is a nontrivial complication to the present analysis, and those of other authors.

We measure the field in the film in units of the incident field by writing

$$E=E_0\epsilon(z)e^{i\phi(z)}\ , \tag{2.2}$$

where both $\epsilon(z)$ and $\phi(z)$ are real. Let $k^2=\omega^2n^2/c^2$, $\tilde{\lambda}=\lambda\,|E_0\,|^2$, and separate Eq. (2.1) into real and imaginary parts, using Eq. (2.2). This gives

$$\frac{d^2\epsilon}{dz^2}-\epsilon\left[\frac{d\phi}{dz}\right]^2+k^2(1+\tilde{\lambda}\epsilon^2)\epsilon=0\ , \tag{2.3a}$$

and

$$2\frac{d\epsilon}{dz}\frac{d\phi}{dz}+\epsilon\frac{d^2\phi}{dz^2}=0\ . \tag{2.3b}$$

At this point, the incident field may be assumed to have unit amplitude always, and the intensity dependence of the reflectivity may be explored by calculating the variation of the amplitude R with $\tilde{\lambda}$, noting that $\tilde{\lambda}=\lambda\,|E_0\,|^2$.

Note that Eq. (2.3b) may be written

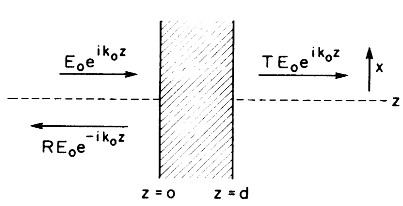

FIG. 1. Geometry considered in this paper. Plane wave of amplitude E_0 strikes a nonlinear film of thickness d, to be reflected and transmitted.

$$\frac{d}{dz}\left[\epsilon^2\frac{d\phi}{dz}\right]=0\ , \tag{2.4}$$

which means that

$$\frac{d\phi}{dz}=\frac{W}{\epsilon^2}\ , \tag{2.5}$$

where W is a constant.

If we were examining the case where the film is semi-infinite, to consider fields localized near the interface as in the theory of surface polaritons,[1] the fact that $\epsilon\to 0$ as $z\to\infty$ requires $W=0$. We have no reason to make this choice, and in fact we require $W\neq 0$. The time-averaged energy flow S normal to the film surface is, within the film,

$$S=\frac{c^2}{8\pi\omega}\mathrm{Re}\left[\frac{1}{i}E^*\frac{dE}{dz}\right]=\frac{c^2\,|E_0\,|^2}{8\pi\omega}\epsilon^2\frac{d\phi}{dz}$$
$$=\frac{c^2\,|E_0\,|^2}{8\pi\omega}W\ . \tag{2.6}$$

Conservation of energy then requires, with $k_0=\omega/c$,

$$W=k_0\,|\,T\,|^2\ , \tag{2.7}$$

where $|\,T\,|^2$ is the transmission coefficient of the film, as illustrated in Fig. 1. Quite clearly, we are required to choose $W\neq 0$.

We may use Eq. (2.5) to eliminate ϕ from Eq. (2.3a),

$$\frac{d^2\epsilon}{dz^2}-\frac{W^2}{\epsilon^3}+k^2(1+\tilde{\lambda}\epsilon^2)\epsilon=0\ . \tag{2.8}$$

This last equation may be integrated once to give

$$\left[\frac{d\epsilon}{dz}\right]^2+\frac{W^2}{\epsilon^2}+k^2\epsilon^2+\tfrac{1}{2}k^2\tilde{\lambda}\epsilon^4=A\ , \tag{2.9}$$

with A a constant of integration. If we let

$$I(z)=\epsilon^2(z)\ , \tag{2.10}$$

then integration of Eq. (2.9) yields

$$\int_{I(0)}^{I(z)}\frac{1}{(AI-k^2I^2-\frac{1}{2}k^2\tilde{\lambda}I^3-W^2)^{1/2}}dI=\pm 2(z-z_0)\ , \tag{2.11}$$

and we also have

$$\phi(z)=\phi(0)+W\int_0^z\frac{1}{I(z')}dz'\ . \tag{2.12}$$

These expressions provide us with an implicit expression for the most general solution for the electromagnetic field within the film. We shall see that the integral of Eq. (2.11) may be evaluated in terms of certain Jacobi elliptic functions.

The solution contains four parameters which must be deduced from the boundary conditions at the film surfaces. These are A and W, along with $\phi(0)$ and $I(0)$. In the linear theory, of course, four parameters exists also. One considers two propagating waves in the film, one from left to right and the second from right to left. Each is described by a complex amplitude.

B. Boundary conditions: General problem

We now turn to examination of the boundary conditions at the film surfaces. These are continuity of the electric field and its normal derivative at $z=0$ and $z=d$. At $z=0$, we have

$$1+R=\epsilon(0)e^{i\phi(0)}\,, \tag{2.13a}$$

$$1-R=\frac{1}{ik_0}\left[\frac{d\epsilon}{dz}\right]_{z=0}e^{i\phi(0)}+\frac{1}{k_0}e^{i\phi(0)}\epsilon(0)\left[\frac{d\phi}{dz}\right]_{z=0}, \tag{2.13b}$$

and at $z=d$

$$Te^{ik_0d}=\epsilon(d)e^{i\phi(d)}\,, \tag{2.14a}$$

$$Te^{ik_0d}=\frac{1}{ik_0}\left[\frac{d\epsilon}{dz}\right]_{z=d}e^{i\phi(d)}+\frac{1}{k_0}e^{i\phi(d)}\epsilon(d)\left[\frac{d\phi}{dz}\right]_{z=d}. \tag{2.14b}$$

Constraints on the four constants in the general solution follow from Eqs. (2.13) and (2.14). For example, upon adding Eq. (2.13a) to Eq. (2.13b), we find

$$2=\epsilon(0)e^{i\phi(0)}+\frac{1}{k_0}\left[\frac{d\epsilon}{dz}\right]_{z=0}e^{i\phi(0)}+\frac{i}{k_0}e^{i\phi(0)}\epsilon(0)\left[\frac{d\phi}{dz}\right]_{z=0}, \tag{2.15}$$

which when separated into real and imaginary parts gives

$$2k_0=k_0\epsilon(0)\cos\phi(0)+\left[\frac{d\epsilon}{dz}\right]_{z=0}\sin\phi(0)+\epsilon(0)\left[\frac{d\phi}{dz}\right]_{z=0}\cos\phi(0) \tag{2.16}$$

and

$$\left[\frac{d\epsilon}{dz}\right]_{z=0}\cos\phi(0)=k_0\epsilon(0)\sin\phi(0)+\epsilon(0)\left[\frac{d\phi}{dz}\right]_{z=0}\sin\phi(0)\,. \tag{2.17}$$

If Eq. (2.16) is multiplied by $\sin\phi(0)$, Eq. (2.17) by $\cos\phi(0)$ and subtracted from Eq. (2.16), we find

$$2k_0\sin\phi(0)=\left[\frac{d\epsilon}{dz}\right]_{z=0} \tag{2.18}$$

while multiplying Eq. (2.16) by $\cos\phi(0)$, Eq. (2.17) by $\sin\phi(0)$, then adding gives

$$2k_0\cos\phi(0)=k_0\epsilon(0)+\epsilon(0)\left[\frac{d\phi}{dz}\right]_{z=0}. \tag{2.19}$$

If Eqs. (2.18) and (2.19) are squared and added, we have

$$4k_0^2=\left[\left[\frac{d\epsilon}{dz}\right]_{z=0}\right]^2+\epsilon^2(0)\left[k_0+\left[\frac{d\phi}{dz}\right]_{z=0}\right]^2. \tag{2.20}$$

Similar procedures may be applied to Eqs. (2.14), to obtain relations between the various amplitudes and derivatives at $z=d$. We have, upon equating the right-hand side of Eq. (2.14a) to that of Eq. (2.14b),

$$ik_0\epsilon(d)=\left[\frac{d\epsilon}{dz}\right]_{z=d}+i\epsilon(d)\left[\frac{d\phi}{dz}\right]_{z=d}, \tag{2.21}$$

which upon separating real and imaginary parts requires simply

$$\left[\frac{d\epsilon}{dz}\right]_{z=d}=0 \tag{2.22a}$$

and

$$\frac{1}{k_0}\left[\frac{d\phi}{dz}\right]_{z=d}=1\,. \tag{2.22b}$$

We assume $\epsilon(d)\neq 0$.

The constraints just outlined will prove most useful in our numerical solution of the nonlinear problem. We shall see how this is done shortly. For instance, consider Eq. (2.18). If we know the three parameters A, W, and $I(0)=\epsilon^2(0)$, then from Eq. (2.9) we may evaluate $(d\epsilon/dz)_{z=0}$. Equation (2.18) then allows us to choose the fourth parameter $\phi(0)$.

C. Case $\tilde{\lambda}=0$

When the nonlinearity is ignored by setting $\tilde{\lambda}=0$, of course the reflectivity of the film is given by a well-known elementary solution. It will be informative to see how this result emerges from the analysis just presented, which has an unfamiliar appearance when applied to the linear problem.

With $\tilde{\lambda}=0$ and $I=\epsilon^2$, Eq. (2.9) becomes

$$\frac{1}{4}\left[\frac{dI}{dz}\right]^2+W^2+k^2I^2=AI\,. \tag{2.23}$$

The boundary condition at $z=d$, Eq. (2.14a) combined with Eq. (2.7) gives

$$I(d)=\frac{1}{k_0}W\,, \tag{2.24}$$

while Eq. (2.22a) requires

$$\left[\frac{dI}{dz}\right]_{z=d}=0\,. \tag{2.25}$$

Thus, when all quantities in Eq. (2.23) are evaluated at $z=d$, we are led to require

$$A=k_0W(1+n^2)\,. \tag{2.26}$$

We may integrate Eq. (2.23),

$$\int_{W/k_0}^{I(z)}\frac{k}{(AI-W^2-k^2I^2)^{1/2}}dI=\pm 2k(z-d)\,, \tag{2.27}$$

which gives

$$\sin^{-1}\left[\frac{2k^2I(z)-A}{(A^2-4k^2W^2)^{1/2}}\right]-\sin^{-1}\left[\frac{2k^2I(d)-A}{(A^2-4k^2W^2)^{1/2}}\right]=\pm 2k(z-d)\ . \tag{2.28}$$

The argument of the second term on the left-hand side of Eq. (2.28) may be reduced to unity by using the relations between the various quantities, so the solution reduces to

$$\sin^{-1}\left[\frac{2k^2I(z)-A}{(A^2-4k^2W^2)^{1/2}}\right]-\frac{\pi}{2}=\pm 2k(z-d) \tag{2.29}$$

or

$$\cos^{-1}\left[\frac{2k^2I(z)-A}{(A^2-4k^2W^2)^{1/2}}\right]=\pm 2k(z-d)\ . \tag{2.30}$$

The solution is independent of which sign is chosen on the right-hand side of Eq. (2.30). After some algebra, we find

$$I(z)=\frac{W}{k_0n^2}[1+(n^2-1)\cos^2k(z-d)]\ , \tag{2.31}$$

where W is still undetermined.

We shall determine W through use of Eq. (2.20). Before we do this, we simplify Eq. (2.20) through use of Eq. (2.5), Eq. (2.8) with $\tilde{\lambda}=0$, and Eq. (2.26). One finds the condition

$$W(n^2+3)-k_0(n^2-1)I(0)=4k_0 \tag{2.32}$$

from which, using Eq. (2.31), one finds W. Then the final expression for $I(z)$ becomes

$$I(z)=\frac{4[1+(n^2-1)\cos^2k(z-d)]}{n^4+2n^2+1-(n^2-1)^2\cos^2(kd)}\ . \tag{2.33}$$

One may verify that this result agrees with that derived by matching the appropriate plane waves to the reflected and transmitted wave at the boundary. The reflection amplitude R is

$$R=\frac{2i(n^2-1)\sin(kd)}{(n+1)^2\exp(-ikd)-(n-1)^2\exp(ikd)}\ . \tag{2.34}$$

We now turn to the nonlinear problem.

D. Nonlinear film

We now consider the case where $\tilde{\lambda}\neq 0$. We begin by writing Eq. (2.9) in terms of the intensity $I(z)=\epsilon^2(z)$. One has

$$\left[\frac{1}{2}\frac{dI}{dz}\right]^2+W^2+k^2I^2+\tfrac{1}{2}k^2\tilde{\lambda}I^3=AI\ . \tag{2.35}$$

Note that we still have, from Eqs. (2.14a) and (2.7),

$$I(d)=\frac{1}{k_0}W\ , \tag{2.36}$$

while the relationship between A and W now becomes, noting that we still have $(dI/dz)_d=0$ from Eq. (2.22a),

$$A=k_0(n^2+1)W+\tfrac{1}{2}n^2\tilde{\lambda}W^2\ . \tag{2.37}$$

For a given value of W, one thus can determine A from this relation.

To proceed, it will prove useful to separate the case where $\tilde{\lambda}>0$ from that for which $\tilde{\lambda}<0$.

1. Case $\tilde{\lambda}>0$

It will prove helpful in what follows to examine the roots of Eq. (2.35) at $z=d$, where dI/dz vanishes. These values of $I(d)$, which we will denote by $I_1(d)$, $I_2(d)$, and $I_3(d)$ satisfy

$$\tfrac{1}{2}k^2\tilde{\lambda}I^3(d)+k^2I^2(d)-AI(d)+W^2=0\ . \tag{2.38}$$

We know that W/k_0 is a root of this equation, so Eq. (2.38) is replaced by

$$\left[I(d)-\frac{1}{k_0}W\right][aI^2(d)+bI(d)+c]=0\ , \tag{2.39}$$

where

$$a=\tfrac{1}{2}k^2\tilde{\lambda}\ , \tag{2.40a}$$

$$b=k_0n^2(k_0+\tfrac{1}{2}\tilde{\lambda}W)\ , \tag{2.40b}$$

and

$$c=-k_0W\ . \tag{2.40c}$$

The three roots are then

$$I_1(d)=\frac{W}{k_0}\ , \tag{2.41a}$$

$$I_2(d)=\frac{1}{\tilde{\lambda}}\left\{\left[\left[1+\frac{\tilde{\lambda}W}{2k_0}\right]^2+\frac{2\tilde{\lambda}W}{n^2k_0}\right]^{1/2}-\left[1+\frac{\tilde{\lambda}W}{2k_0}\right]\right\} \tag{2.41b}$$

and

$$I_3(d)=-\frac{1}{\tilde{\lambda}}\left\{\left[\left[1+\frac{\tilde{\lambda}W}{2k_0}\right]^2+\frac{2\tilde{\lambda}W}{n^2k_0}\right]^{1/2}+\left[1+\frac{\tilde{\lambda}W}{2k_0}\right]\right\}. \tag{2.41c}$$

Notice that $I_1(d)>I_2(d)>I_3(d)$.

Now we may integrate Eq. (2.35), expressing the result in terms of the three roots just obtained. We integrate from $z=d$, into an arbitrary point within the film,

$$\int_{W/k_0}^{I(z)}\frac{dI}{\{[I_1(d)-I][I_2(d)-I][I_3(d)-I]\}^{1/2}}=\pm(2\tilde{\lambda})^{1/2}k(z-d)\ . \tag{2.42}$$

The integral on the left-hand side can be expressed in terms of the inverse of a Jacobi elliptic function.[6] We have

$$\frac{1}{[I_1(d)-I_3(d)]^{1/2}}\mathrm{cn}^{-1}\left[\frac{[I(z)-I_2(d)]^{1/2}}{[I_1(d)-I_2(d)]^{1/2}}\left|\frac{I_1(d)-I_2(d)}{I_1(d)-I_3(d)}\right.\right]=\pm\left[\frac{\tilde{\lambda}}{2}\right]^{1/2}k(z-d)\ . \tag{2.43}$$

Hence we have

$$I(z)=I_2(d)+[I_1(d)-I_2(d)]\mathrm{cn}^2\left[\frac{\tilde{\lambda}^{1/2}}{\sqrt{2}}[I_1(d)-I_3(d)]^{1/2}k(d-z)\left|\frac{I_1(d)-I_2(d)}{I_1(d)-I_3(d)}\right.\right]\ . \tag{2.44}$$

The result in Eq. (2.44) is an analogue of Eq. (2.31). The only unknown parameter in the expression is W, as in Eq. (2.31). Indeed, one may verify explicitly that as $\tilde{\lambda}\to 0$, Eq. (2.44) reduces to Eq. (2.31).

The one parameter W may be determined by requiring that Eq. (2.20), derived at the boundary $z=0$, be satisfied. With the various relations outlined above, this may be rearranged to read

$$\tfrac{1}{2}n^2\tilde{\lambda}I^2(0)+(n^2-1)I(0)+4-(n^2+3)\frac{W}{k_0}-\tfrac{1}{2}n^2\tilde{\lambda}\frac{W^2}{k_0^2}=0\ . \tag{2.45}$$

We may now proceed as follows. Given a guess for the one parameter W which enters Eq. (2.44), we may use the expression to calculate $I(0)$, then check to see if Eq. (2.45) is satisfied. Notice that W is positive definite, and since $|T|^2\le 1$ always, we must have $W\le k_0$. Hence W is bounded. Thus, once one has a program which calculates the Jacobi elliptic function which appears in Eq. (2.44), the search for values of W consistent with Eq. (2.45) is straightforward. In our numerical work, under the conditions outlined in Sec. III, we find regimes where several values of W emerge as the solution. This occurs in regimes where the film may switch between states of differing transmissivity. We shall elaborate on this in Sec. III.

Note that the Jacobi elliptic function $\mathrm{cn}(x\,|\,y)$ which appears in Eq. (2.44) is bounded between -1 and $+1$. This means that the intensity $I(z)$ at any point in the film is bounded above by $I_1(d)$, and below by $I_2(d)$.

2. Case $\tilde{\lambda}<0$

While the analysis for the case $\tilde{\lambda}<0$ proceeds along the lines just described, nonetheless in the end the functional dependence of the intensity I with position z in the film differs from that displayed in Eq. (2.44). We begin by noting that Eq. (2.35) is replaced by

$$\left[\frac{1}{2}\frac{dI}{dz}\right]^2+W^2+k^2I^2-\tfrac{1}{2}k^2|\tilde{\lambda}|I^3=AI\ , \tag{2.46}$$

and then the relation between A and W is

$$A=k_0(n^2+1)W-\tfrac{1}{2}n^2|\tilde{\lambda}|W^2\ . \tag{2.47}$$

At $z=d$, where the boundary condition $(dI/dz)_d=0$ applies, Eq. (2.46) becomes a cubic for the three possible values $I(d)$,

$$\tfrac{1}{2}k^2|\tilde{\lambda}|I^3(d)-k^2I^2(d)+AI(d)-W^2=0\ , \tag{2.48}$$

and we arrange the roots as follows:

$$I_1(d)=\frac{1}{|\tilde{\lambda}|}\left\{\left[1-\frac{|\tilde{\lambda}|W}{2k_0}\right]+\left[\left[1-\frac{|\tilde{\lambda}|W}{2k_0}\right]^2-\frac{2|\tilde{\lambda}|W}{n^2k_0}\right]^{1/2}\right\}, \tag{2.49a}$$

$$I_2(d)=\frac{W}{k_0}\ , \tag{2.49b}$$

and

$$I_3(d)=\frac{1}{|\tilde{\lambda}|}\left\{\left[1-\frac{|\tilde{\lambda}|W}{2k_0}\right]-\left[\left[1-\frac{|\tilde{\lambda}|W}{2k_0}\right]^2-\frac{2|\tilde{\lambda}|W}{n^2k_0}\right]^{1/2}\right\}. \tag{2.49c}$$

We have labeled the roots so that once again

$$I_1(d)>I_2(d)>I_3(d)\ ,$$

$$\text{for } n^2\ge\tfrac{1}{2}|\tilde{\lambda}|\frac{W}{k_0}+\left[2|\tilde{\lambda}|\frac{W}{k_0}\right]^{1/2},$$

a condition we shall assume holds.

Now Eq. (2.46) can be integrated,

$$\int_{W/k_0}^{I(z)}\frac{1}{\{[I-I_1(d)][I-I_2(d)][I-I_3(d)]\}^{1/2}}dI=\pm(2|\tilde{\lambda}|)^{1/2}k(z-d)\ . \tag{2.50}$$

The integral once more can be expressed in terms of Jacobi elliptic functions,

$$\frac{1}{[I_1(d)-I_3(d)]^{1/2}}\mathrm{cn}^{-1}\left[\left[\frac{[I_1(d)-I_2(d)][I(z)-I_3(d)]}{[I_2(d)-I_3(d)][I_1(d)-I_3(d)]}\right]^{1/2}\Bigg|\,\frac{[I_2(d)-I_3(d)]}{[I_1(d)-I_3(d)]}\right]=\pm\left[\frac{|\tilde{\lambda}|}{2}\right]^{1/2}k(d-z)\ . \tag{2.51}$$

This may be inverted to give

$$I(z)=\frac{N(z)}{D(z)}\ , \tag{2.52a}$$

where

$$N(z)=[I_1(d)-I_2(d)]I_3(d)+I_1(d)[I_2(d)-I_3(d)]\mathrm{cn}^2\left[\frac{|\tilde{\lambda}|^{1/2}}{\sqrt{2}}[I_1(d)-I_3(d)]^{1/2}k(d-z)\,\Bigg|\,\frac{[I_2(d)-I_3(d)]}{[I_1(d)-I_3(d)]}\right] \tag{2.52b}$$

and

$$D(z)=[I_1(d)-I_2(d)]+[I_2(d)-I_3(d)]\mathrm{cn}^2\left[\frac{|\tilde{\lambda}|^{1/2}}{\sqrt{2}}[I_1(d)-I_3(d)]^{1/2}k(d-z)\,\Bigg|\,\frac{[I_2(d)-I_3(d)]}{[I_1(d)-I_3(d)]}\right]\ . \tag{2.52c}$$

Notice that $I(z)$ satisfies the inequality $I_3(d)\leq I(z)\leq I_2(d)$. To complete the solution, and find the one parameter W which enters Eq. (2.52), we proceed precisely as in the case $\tilde{\lambda}>0$. We now turn our attention to numerical studies of the reflectivity, as a function of $\tilde{\lambda}$.

III. RESULTS AND DISCUSSION

We have carried out calculations of the transmissivity of films of various thicknesses, as a function of incident laser power, using the approach developed in Sec. II. It should be noted that the computer program required for this purpose is remarkably elementary, once a subroutine for calculating the Jacobi elliptic function is available.

The parameter $\tilde{\lambda}$ is proportional to the incident laser power and, once the fields are scaled as described in Sec. II, a plot of the transmissivity as a function of $\tilde{\lambda}$ is equivalent to a plot of this quantity as a function of laser power, as we have seen.

In the thin-film limit, where the film thickness d is small compared to the wavelength $\lambda_f=2\pi c/n\omega$ of (low-power) radiation in the film, we find no bistability, though the reflectivity and transmissivity indeed is power dependent. This conclusion contradicts that reached by Band,[5] whose calculations indicate the presence of bistability for very thin InSb films. Unfortunately, we have no idea of the source of the discrepancy, since the two calculations proceed along very different lines. For the case $d=0.1\lambda_f$, our results are displayed in Fig. 2. When $\tilde{\lambda}>0$, for the range of values of $\tilde{\lambda}$ explored, the transmissivity decreases monotonically with laser power, as displayed in Fig. 2(a). As the power increases, the average index of refraction increases, to produce greater impedance mismatch between the film and the surrounding vacuum, so the reflectivity increases, while $|T|^2$ falls. We find the opposite behavior in Fig. 2(b), where calculations for $\tilde{\lambda}<0$ are displayed.

When the film thickness increases, so that one is close to the point where an integral number of half wavelengths may fit into the film, we find bistability, or as we shall see a progressive increase in laser power allows an ever increasing number of possible states for the film to lock into. Figure 3(a) shows $|T|^2$ as a function of $\tilde{\lambda}$, for $d=0.4\lambda_f$ and several choices of the linear index of refraction n. We see a region of bistable behavior for $n>6$.

One may understand these results on physical grounds, as follows. First note that for all the curves, there is a power for which $|T|^2$ rises to exactly unity. In the graph, for all choices of the linear index of refraction n, it appears as if $|T|^2$ equals unity for the same laser power. If one recalls that $|T|^2=1$ for a half wave plate for which $d=\frac{1}{2}\lambda_f$, one may understand that increasing the laser power increases the average index of refraction in

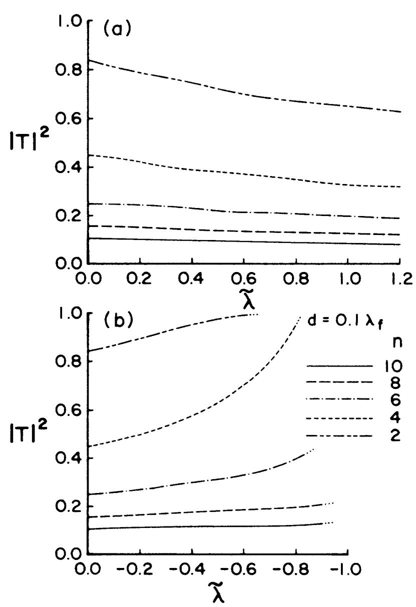

FIG. 2. For the cases (a) $\tilde{\lambda}>0$ and (b) $\tilde{\lambda}<0$, and various choices of linear index of refraction, we show the transmissivity of the film as a function of laser power, for $d=0.1\lambda_f$.

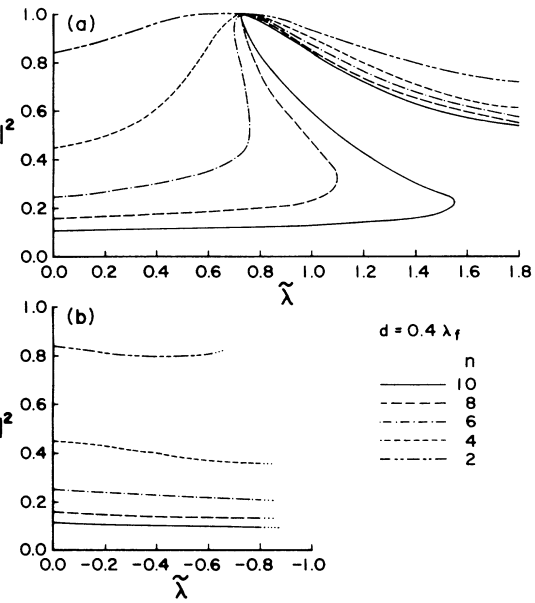

FIG. 3. For the cases (a) $\tilde{\lambda}>0$ and (b) $\tilde{\lambda}<0$, and various choices of linear index of refraction, we show the transmissivity of the film as a function of laser power, for $d=0.4\lambda_f$.

the medium, with the result that the wavelength is shortened. There is then a point where precisely one-half wavelength fits into the film for which $d=0.4\lambda_f$, and also $\tilde{\lambda}>0$.

We may estimate the value of $\tilde{\lambda}$ for which $|T|^2=1$ as follows. The average index of the medium is $\bar{n}=n(1+\langle\tilde{\lambda}\rangle)$, and $|T|^2=1$ when $\bar{\lambda}_f=\lambda_f n/\bar{n}=2d$. This gives the estimate $\langle\tilde{\lambda}\rangle\cong\frac{1}{4}$. From Eq. (2.33), when $kd=\pi$ and $n^2\gg 1$, the average intensity in the film is $\bar{I}\cong\frac{1}{2}I_0$, so $\langle\tilde{\lambda}\rangle\cong\frac{1}{2}\tilde{\lambda}$. This gives the estimate $\tilde{\lambda}\cong\frac{1}{2}$ independent of the linear index n for the point where $|T|^2=1$, a result approximately correct but wrong in detail because this simple argument overlooks the role of spatial variation in the intensity.

For powers equal to that where $|T|^2=1$, we have a solution for which exactly one-half wavelength fits into our film, as just described. For greater powers, the average index increases further, the wavelength in the medium shortens to move one off the half wave condition, and $|T|^2$ associated with this solution decreases as we increase the power further.

When the linear index n is small, a large fraction of the laser power is transmitted into the film, and at low power, $|T|^2$ is large. An increase in laser power shortens $\bar{\lambda}_f$, to the point where the condition $\bar{\lambda}_f=2d$ is realized, and $|T|^2$ is a single-valued function of $\tilde{\lambda}$, rising smoothly to unity then falling off.

We have seen that, on the basis of the crude argument presented above, the value of $\tilde{\lambda}$ for which $|T|^2=1$ is insensitive to the linear index n. This point is reinforced by the exact results of Fig. 3(a). Now if the linear index n is *large*, $|T|^2$ is small at low powers, and rather little of the incident radiation enters the film at low powers. Thus, at low powers, $\langle\tilde{\lambda}\rangle$ is very small, and an increase in power has rather little effect on the average index $\bar{n}$. Indeed, for $n=10$, we see $|T|^2$ varies little with $\tilde{\lambda}$ in Fig. 3(a), in the low-power regime.

We now understand why larger linear indices of refraction favor bistability. For n large, by the time $\tilde{\lambda}$ increases to the critical value required to make $|T|^2=1$, the low-power solution does not allow $\bar{n}$ to increase to the point where the condition $\bar{\lambda}_f=2d$ is realized. The film does not admit sufficient power for this to happen. But we also have seen that there is a second solution which just allows a half wavelength in the film; the second solution has high-field intensity within the film, and a large value of $|T|^2$ or small reflectivity as a consequence. For small n, the low-power solution evolves continuously and monotonically into that for which $|T|^2=1$, but this does not occur for large values of n, and we find bistability.

If $d=0.4\lambda_f$, and $\tilde{\lambda}<0$, the physical arguments just presented suggest no bistability should occur, since an increase in laser power increases rather than decreases the average wavelength. Our calculations show this is indeed the case, as we see from Fig. 3(b). Also, if we start with $d=0.6\lambda_f$, the argument suggests no bistability is present for $\tilde{\lambda}>0$, while for $\tilde{\lambda}<0$ we again should realize a laser power where precisely a half wavelength will fit into the film, and bistability will occur. In Figs. 4(a) and 4(b), for the two signs of $\tilde{\lambda}$, we show calculations of $|T|^2$, and the results are in accord with our expectations.

We see we are able to understand the origin of the bistability, on the basis of rather simple physical arguments. These arguments, admittedly very simple, do not allow for bistable behavior in the thin-film limit $d\ll\lambda_f$, as the exact calculations in Fig. 2 confirm. This raises further question in our mind on the correctness of the conclusions in Ref. 5.

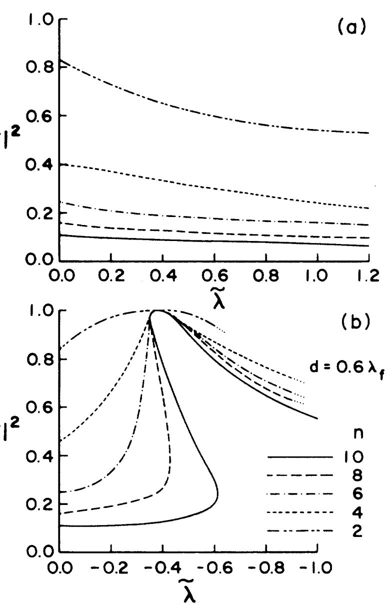

FIG. 4. For the cases (a) $\tilde{\lambda}>0$ and (b) $\tilde{\lambda}<0$, and various choices of the linear index of refraction, we show the transmissivity of the film as a function of laser power, for $d=0.6\lambda_f$.

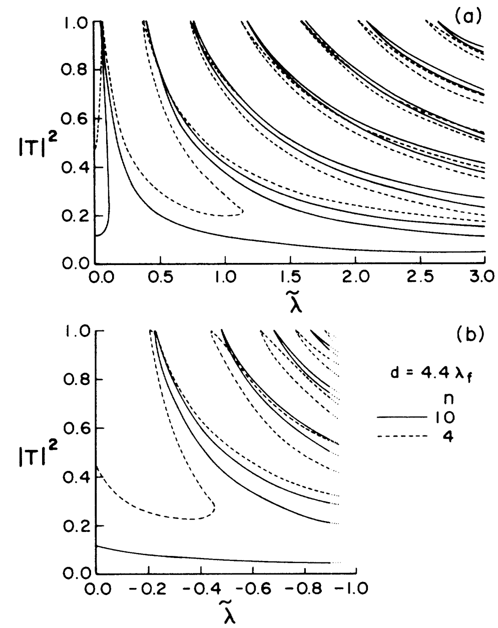

FIG. 5. For the cases (a) $\tilde{\lambda}>0$ and (b) $\tilde{\lambda}<0$, and for two values of the linear index of refraction and two choices of the linear index of refraction, we show the transmissivity of the film as a function of laser power, for $d=4.4\lambda_f$.

Very complex behavior is realized for thicker films, where many half wavelengths nearly fit. As the laser power increases, we find a progressively greater number of possible states, reminiscent of the cascading sequence of bifurcations encountered often in nonlinear problems. This is illustrated in Figs. 5(a) and 5(b), where for both signs of $\tilde{\lambda}$ and $d=4.4\lambda_f$, we plot $|T|^2$ as a function of $\tilde{\lambda}$. For $\tilde{\lambda}=3$, we find 13 possible states of the film. Results qualitatively similar to this may be found in Ref. 4. For $n=4$ and $\tilde{\lambda}=0.9$, where we have five possible states, we show in Fig. 6 the intensity distribution $I(z)$ within the film. On general grounds, from properties of the Jacobi elliptic functions, one sees that $I(z)$ should be periodic in z. For $\tilde{\lambda}=0.9$, we are in the regime of strong

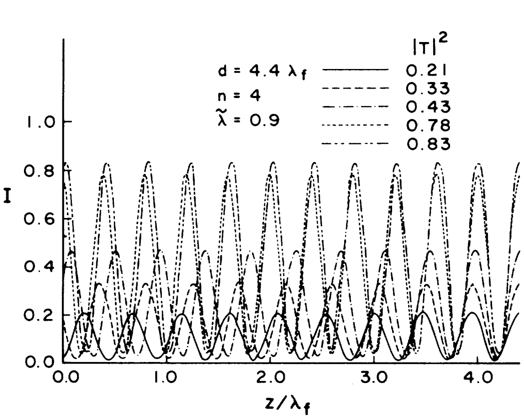

FIG. 6. For $n=4$, $\tilde{\lambda}=0.9$, and $d=4.4\lambda_f$, we show the spatial variation of the field within the film, for each of the five solutions compatible with the boundary conditions.

nonlinearity, but the spatial variation of $I(z)$ is remarkably sinusoidal.

The multivalued nature of $|T|^2$, considered as a function of $\tilde{\lambda}$, is intimately related to the geometrical resonances in the thin film, as our discussion of the cases $d=0.4\lambda_f$ and $d=0.6\lambda_f$ suggest. The case of a semi-infinite film may be solved analytically, as we show in the Appendix. For both signs of $\tilde{\lambda}$, $|T|^2$ is a single-valued function of $\tilde{\lambda}$, as one sees from this discussion. The thin film thus acts very much as a Fabry-Perot cavity.

It is then intriguing to inquire how the single-valued reflectivity appropriate to $d=\infty$ is recovered by beginning with finite d, then letting d be increased progressively. In fact, by this limiting process, in our model, there is no unique limit as $d\to\infty$ since, at any finite d we shall encounter the multivalued character of $|T|^2$.

In any physically realizable film, absorption is present, and when d is much greater than the absorption length, quite clearly the semi-infinite limit will be realized. It would be most intriguing to extend our study to include absorption, but this represents a nontrivial extension of the analysis. The discussion here is applicable only to films whose thickness d is small compared to the absorption length in the material.

ACKNOWLEDGMENT

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APPENDIX: REFLECTIVITY OF THE SEMI-INFINITE NONLINEAR DIELECTRIC

Here we have only the boundary conditions at $z=0$, which are stated in Eqs. (2.13) of the text. We may solve for the reflectivity R

$$R=\frac{ik_0\epsilon(0)-(d\epsilon/dz)_0-i\epsilon(0)(d\phi/dz)_0}{ik_0\epsilon(0)+(d\epsilon/dz)_0+i\epsilon(0)(d\phi/dz)_0} . \tag{A1}$$

In the semi-infinite medium, we have a solution where the amplitude of the field $\epsilon(z)$ is a constant ϵ. Thus, we have $(d\epsilon/dz)=0$ everywhere, and $d^2\epsilon/dz^2=0$ also. Then $d\phi/dz$ is a constant, with value given in Eq. (2.3a),

$$\left[\frac{d\phi}{dz}\right]_0^2=k^2(1+\tilde{\lambda}\epsilon^2) . \tag{A2}$$

The solution is then a pure sinusoid, with the wavelength controlled by the effective power-dependent dielectric constant $n^2(1+\tilde{\lambda}\epsilon^2)$; in the end we must obtain ϵ^2 in a self-consistent manner.

With the above remarks in mind, Eq. (A1) reduces to

$$R=\frac{k_0-(d\phi/dz)}{k_0+(d\phi/dz)} , \tag{A3}$$

or

$$R=\frac{1-n(1+\tilde{\lambda}\epsilon^2)^{1/2}}{1+n(1+\tilde{\lambda}\epsilon^2)^{1/2}} . \tag{A4}$$

We may determine ϵ^2 as follows. We know

$$\frac{d\phi}{dz}=\frac{W}{\epsilon^2}=k\epsilon^2(1+\tilde{\lambda}\epsilon^2)^{1/2} , \tag{A5}$$

and energy conservation also requires

$$W=k_0(1-|R|^2) . \tag{A6}$$

Upon combining Eqs. (A5) and (A6) one finds, assuming $1+\tilde{\lambda}\epsilon^2>0$, the condition

$$\epsilon^2=\frac{2}{[1+n(1+\tilde{\lambda}\epsilon^2)^{1/2}]^2} , \tag{A7}$$

an equation that has one unique solution for each choice of $\tilde{\lambda}$. Thus, for the semi-infinite medium, there is no bistability in our model, when $1+\tilde{\lambda}\epsilon^2>0$, and the dielectric constant is a positive number.

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[1]For a rather general discussion, which contains a number of special models explored in earlier papers, see K. M. Leung, Phys. Rev. B **32**, 5093 (1985). A review of the field has been presented by A. A. Maradudin, in *Proceedings of the Second International School on Condensed Matter Physics, Varna, Bulgaria* (World Scientific, New York, 1982).

[2]For example, one model has dielectric tensor with $\epsilon_{xx}=\epsilon_{xx}^{(0)}(1+\lambda|E_x|^2)$ and $\epsilon_{zz}=\epsilon_{zz}^{(0)}$ independent of field, with z normal to the interface and x the propagation direction of a p-polarized surface polariton. For $\lambda<0$, $\epsilon_{xx}^{(0)}$ and $\epsilon_{zz}^{(0)}$ positive, it is argued that the interface may bind a surface polariton when the dielectric constant of the adjacent material is also positive. One may show that for these waves to exist, there must be a spatial regime where $1+\lambda|E_x|^2<0$, so the nonlinearity must be sufficiently strong to actually change the sign of the dielectric tensor element ϵ_{xx}, in the nonlinear medium, while ϵ_{zz} retains the positive value $\epsilon_{zz}^{(0)}$. It will prove difficult to realize such a material in practice.

[3]Y. B. Band, in *Optical Bistability II,* edited by C. M. Bowden *et al.* (Plenum, New York, 1984).

[4]J. H. Marburger and F. S. Felber, Phys. Rev. A **17**, 335 (1978).

[5]Y. B. Band, J. Appl. Phys. **56**, 656 (1984).

[6]L. M. Milne-Thomson, *Jacobian Elliptic Functions Tables* (Dover, New York, 1950).