p-polarized nonlinear surface polaritons in materials with intensity-dependent dielectric functions

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We investigate the properties of p-polarized nonlinear surface polaritons (NLSP) propagating along the interfaces of optically nonlinear materials. We show that Maxwell's equations for the NLSP can be solved exactly in quadratures for optically isotropic media with dielectric functions which can be an arbitrary function of the field intensity. The required boundary conditions can be imposed readily, and a form of the dispersion relation for the NLSP is obtained without the need to solve for the field profile first. The general results are then applied to a specific model in which the material has a nonlinear dielectric function proportional to the electric field intensity. Both the self-focusing and self-defocusing cases are studied, as well as different values of the linear dielectric functions inside and outside the material. The physically allowed regions in parameter space and the nonlinear surface-plasmon resonance conditions are examined. The field profile in each region is also investigated.

I. INTRODUCTION

The primary concern in the area of nonlinear optics has been on nonlinear processes involving the generations and interactions of waves of different frequencies. Examples are harmonic generation, modulation and demodulation of light, mixing of light waves in parametric up and down conversions, and stimulated Brillouin and Raman scattering.[1] The theoretical treatment for these phenomena often rests on the assumption that the nonlinearities involved are sufficiently weak; the calculations are therefore almost exclusively perturbative in nature.

Recently there are some growing interests in the study of intrinsically nonlinear effects on the propagation of electromagnetic waves, all at a single frequency, along the interfaces of optically nonlinear media.[2] The conventional quasilinear approximation cannot be used. The solutions to Maxwell's equations must be calculated in a fully nonperturbative fashion and the necessary electromagnetic boundary conditions must be matched across the interfaces. A number of exact model calculations have revealed many remarkable results of these nonlinear electromagnetic waves which have *no counterpart* whatsover in the linear theory. With the electromagnetic energy flux as an additional physical parameter which actively changes the effective value of the dielectric functions of the nonlinear media, a host of *entirely new phenomena* are potentially possible.

Among these unexpected results is the existence of p-polarized (TM) nonlinear surface polaritons (NLSP) at the interface of two media whose dielectric functions at the polariton frequency are of the same sign.[3] The case studied by Agranovich *et al.*[3] consists of a linear medium and a nonlinear medium in contact at a single ($z=0$) plane. However, in order to obtain analytical solutions to the problem the component of the dielectric function for the nonlinear medium perpendicular to $\hat{\mathbf{z}}$, ϵ_{zz}, was taken to be field independent. Moreover, the other two field-dependent components of the dielectric functions were assumed not to be dependent on the z component of the electric field. With this simplified form of the nonlinear dielectric functions, p-polarized NLSP have also been studied in a variety of different configurations. The case in which both media are nonlinear was studied by Lomtev.[4] Fedyanin and Mihalache[5] investigated the surface localized modes in three-layer dielectric structures, and the waveguide modes in such structures were reported by Lederer, Langbein, and Ponath,[6] and by Stegeman and Seaton.[7] However, for the case in which the nonlinear dielectric is sandwiched between two linear-dielectrics, only the "zero-energy solution" was investigated.[6] The use of a somewhat oversimplified form for the nonlinear dielectric function in all these studies has recently been improved to include cases where ϵ_{zz} can also be nonlinear.[8,9] Moreover, the nonlinear dielectric functions can be arbitrary functions of the electric field intensity. However, like previous works, only the dependence on the electric field components parallel to the interface was considered. This somewhat artificial restriction will be eliminated in the present work.

The first experimental results that bear on the existence of p-polarized NLSP were reported by Chen and Carter.[10,11] They investigated the propagation of p-polarized NLSP at the interface of Ag and semiconducting nonlinear media. Both GaAs and Si have been studied. By measuring the intensity of the reflected laser beam as a function of the incident angle, the dispersion relation of the NLSP is deduced.[10,11] From the dependence of this relation on the intensity of the incident beam, the authors were able to obtain both the signs and the magnitudes of the degenerate third-order susceptibility for Si and GaAs. One can also see from their results that the effects of the nonlinearities that are possible in these two materials are rather small. In fact, their analyses of the data were based on the use of a nonlinear dispersion relation which was derived assuming weak nonlinearities.[10,11] A slightly more refined perturbative calculation which yields exactly the same expression was later carried out by Agranovich and Chernyak.[12]

However, a number of materials whose nonlinear coefficients are a few order of magnitudes larger than that of Si are currently available. They include 4-methoxybenzulidene-4′-n-butylaniline (MBBA) liquid crystal,[13] GaAs/$Ga_{1-x}Al_xAs$ multiple-quantum-well structure,[14] semiconductors such as InSb (Refs. 14 and 15) and HgCdTe (Ref. 16), and artificial Kerr media.[17,18] Quite larger relative changes in refractive indices can be induced in these materials with modest laser powers. The waves propagating in such media are then not just small extensions of the linear ones, but must be regarded as separate independent entities, with their own unique properties which cannot be treated using the conventional quasilinear approach.

In this work we investigate the properties of p-polarized NLSP propagating along the surface of materials whose dielectric functions depend on the electric field intensity. In Sec. II we show that Maxwell's equations for the NLSP can be solved exactly in quadratures for optically isotropic media with dielectric functions which can depend on the field intensity in quite an arbitrary way. The required boundary conditions can be imposed readily, and a form of the dispersion relation for the NLSP is obtained without having to solve for the field profile first. Section III is devoted to an application of the general result developed here to specific model in which the material has a nonlinear dielectric function proportional to the electric field intensity. Both the self-focusing and self-defocusing cases are studied, as well as different values of the linear dielectric functions inside and outside the material. The physically allowed regions in parameter space and the nonlinear surface-plasmon resonance conditions are examined. The field profile in each region is also investigated. In Sec. IV we conclude by pointing out the limitations of our present model and rooms for further studies. Results for the corresponding linear case are contained in the Appendix to facilitate making detailed comparisons with those of the NLSP. The corresponding investigations for the s-polarized NLSP are equally as interesting,[2,19,9] but will not be discussed here at all because they lie outside the scope of the present paper.

II. GENERAL RESULTS

Consider a nonlinear material with a plane surface perpendicular to one of its optical axes. Let this be the $z=0$ plane. We consider here a p-polarized electromagnetic wave propagating in the $\hat{\mathbf{x}}$ direction along the surface. The wave is assumed to have an x dependence of the form of a plane wave, $\sim e^{ik_x x}$, and no y dependence. The field components E_y, B_x, and B_z are all zero, and the remaining components obey the set of equations:

$$-\partial_z B_y(z,t)=\partial_t[\epsilon_{xx}E_x(z,t)] , \tag{2.1a}$$

$$ik_x B_y(z,t)=\partial_t[\epsilon_{zz}E_z(z,t)] , \tag{2.1b}$$

$$\partial_z E_x(z,t)-ik_x E_z(z,t)=-\partial_t B_y(z,t) , \tag{2.1c}$$

where the magnetic permeability μ has been taken to be unity. If we further assume the wave be monochromatic, $\sim e^{-i\omega t}$, and that ϵ's depend on the intensity $|\mathbf{E}|^2$ only, then the ϵ's do not depend on t. We can rewrite the above Maxwell's equations in the form

$$B_y'(\xi)=i\epsilon_{xx}E_x(\xi) , \tag{2.2a}$$

$$\eta B_y(\xi)=\epsilon_{zz}E_z(\xi) , \tag{2.2b}$$

$$E_x'(\xi)-i\eta E_z(\xi)=iB_y(\xi) , \tag{2.2c}$$

where

$$\xi\equiv\omega z/c , \tag{2.3}$$

$$\eta\equiv k_x c/\omega , \tag{2.4}$$

and the prime denotes differentiation with respect to the dimensionless variable ξ. We find it convenient here to express E_x and E_z in terms of B_y so that we obtain from Eqs. (2.2a)–(2.2c) the second-order nonlinear differential equation

$$\left[\frac{B_y'}{\epsilon_{xx}}\right]'=\left[\frac{\eta^2}{\epsilon_{zz}}-1\right]B_y . \tag{2.5}$$

Note that the dielectric functions depend on ξ only through their dependence on the electric field intensity

$$|\mathbf{E}|^2=|E_x|^2+|E_z|^2=\left|\frac{\eta B_y}{\epsilon_{zz}}\right|^2+\left|\frac{B_y'}{\epsilon_{xx}}\right|^2 .$$

Now assuming a lossless medium we see that B_y can only be determined up to a constant phase factor. Without loss of generality we can take B_y to be real and non-negative. Then B_y is always in phase with E_z but is always 90° out of phase with E_x.

For simplicity, and in order that our results will apply directly to the case studied experimentally,[11,12] we consider here only optically isotropic systems and put $\epsilon=\epsilon_{xx}=\epsilon_{zz}$. Our problem now is to solve for B from the following set of equations:

$$\left[\frac{B'}{\epsilon}\right]'=\left[\frac{\eta^2}{\epsilon}-1\right]B , \tag{2.6a}$$

$$\epsilon=\epsilon_0+\epsilon_2(E^2) , \tag{2.6b}$$

$$E^2=\eta^2\left[\frac{B}{\epsilon}\right]^2+\left[\frac{B'}{\epsilon}\right]^2 , \tag{2.6c}$$

where $\epsilon_2(E^2)$ is an arbitrary function of E^2 and vanishes when $E=0$, and we put $B\equiv B_y$ to simplify our notations. It turns out that our problem can be solved exactly in quadratures for arbitrary form of $\epsilon_2(E^2)$. To do so, let $I(\epsilon-\epsilon_0)$ be the inverse function of $\epsilon_2(E^2)$ so that in the linear limit, ϵ approaches ϵ_0, and $I(0)=0$. To simplify our analysis, we further assume that in the range of intensities of interest $\epsilon_2(I)$ is a monotonic function of I, and so $I(\epsilon-\epsilon_0)$ is uniquely defined. From Eqs. (2.6b) and (2.6c) we have

$$\left[\frac{B'}{\epsilon}\right]^2=I(\epsilon-\epsilon_0)-\eta^2\left[\frac{B}{\epsilon}\right]^2 . \tag{2.7}$$

Differentiating this equation once and making use of (2.6a), we obtain the result

$$\frac{2BB'}{\epsilon}\left[\frac{\eta^2}{\epsilon}-1\right]=I'-\eta^2\frac{B}{\epsilon^3}(\epsilon B'-\epsilon' B) . \tag{2.8}$$

Multiplying (2.8) by ϵ, we can put it into the form

$$\left[\left[\frac{2\eta^2-\epsilon}{\epsilon}\right]B^2\right]'=\epsilon I' , \tag{2.9}$$

which can immediately be integrated once to give

$$B^2=\frac{\epsilon}{2\eta^2-\epsilon}[\epsilon I(\epsilon-\epsilon_0)-J(\epsilon-\epsilon_0)] , \tag{2.10}$$

where

$$J(\epsilon-\epsilon_0)\equiv\int_0^{\epsilon-\epsilon_0}dx\, I(x) , \tag{2.11}$$

and the integration constant has been set equal to zero. This choice is appropriate when the material extends to infinity where $B=B'=0$, and therefore $I=0$ and $\epsilon=\epsilon_0$. Since ϵ is a function of B and B', (2.10) is a first integral of our problem.

In order to calculate B as a function of ξ it is difficult to work directly with (2.10), instead we use (2.6b) and (2.6c) to write

$$(B')^2=\epsilon^2 I(\epsilon-\epsilon_0)-\eta^2B^2 , \tag{2.12}$$

which then leads to

$$\xi=\mathrm{sgn}(B')\int_{B(0)}^{B}dB[\epsilon^2I(\epsilon-\epsilon_0)-\eta^2B^2]^{1/2} \tag{2.13a}$$

$$=\mathrm{sgn}(\epsilon')\int_{\epsilon(0)}^{\epsilon}d\epsilon[-2V(\epsilon)]^{-1/2} , \tag{2.13b}$$

where

$$V(\epsilon)\equiv-\frac{1}{2}\left[\frac{dB}{d\epsilon}\right]^{-2}[\epsilon^2I(\epsilon-\epsilon_0)-\eta^2B^2] , \tag{2.13c}$$

and $B(0)$ and $\epsilon(0)$ are, respectively, the magnetic field and the dielectric function at the surface, $\xi=0$. In (2.13c) we must use (2.10) to express B and $dB/d\epsilon$ in terms of ϵ. Note that with ϵ interpreted as the coordinate and ξ as the time, Eq. (2.13b) describes the classical motion of a unit mass moving in a one-dimensional potential V with zero total energy.[20] Performing the integral in (2.13b) and then inverting the result gives us $\epsilon=\epsilon(\xi)$, and finally putting it in (2.10) yields the magnetic field profile $B=B(\xi)$. Thus our problem has been reduced to quadratures. The values of $B(0)$ and $\epsilon(0)$ must be determined by boundary conditions which we will discuss next.

The boundary conditions are obtained from the requirement that the tangential field components be continuous across $z=0$, i.e.,[21]

$$[E_x]=[B_y]=0 , \tag{2.14}$$

or in terms of B_y:

$$[B]=0,\quad \left[\frac{B'}{\epsilon}\right]=0 . \tag{2.15}$$

To be more specific, let us consider the case where region I with $z<0$ is occupied by a linear isotropic medium with $\epsilon=\epsilon_1=$const, and region II with $z>0$ is occupied by the nonlinear medium of interest. Thus in region I (2.6) becomes

$$B''=(\eta^2-\epsilon_1)B \quad (z<0) . \tag{2.16}$$

In order for a localized mode to exist at the interface, it is clear that we must have $\eta^2-\epsilon_1>0$. The solution to (2.16) that approaches zero as $z\to-\infty$ is given by

$$B(\xi)=B(0)e^{\kappa_1\xi} \tag{2.17}$$

with

$$\kappa_1\equiv+(\eta^2-\epsilon_1)^{1/2} , \tag{2.18}$$

The boundary condition $[B]=0$ is satisfied automatically. The remaining boundary condition can be written with the help of (2.17) as

$$\frac{B'(0)}{\epsilon(0)}=\frac{B(0)\kappa_1}{\epsilon_1} . \tag{2.19}$$

Use of (2.19) in (2.7) gives

$$\left[\frac{B'(0)}{\epsilon(0)}\right]^2=\frac{B^2(0)\kappa_1^2}{\epsilon_1^2}=I(\epsilon(0)-\epsilon_0)-\eta^2\left[\frac{B(0)}{\epsilon(0)}\right]^2 . \tag{2.20}$$

We find it convenient to eliminate all explicit dependences on B by using (2.10). The result is some kind of dispersion relation connecting $\eta\equiv k_xc/\omega$ with the value of the dielectric function at the surface of the nonlinear medium, $\epsilon(0)$:

$$\frac{\epsilon(0)}{2\eta^2-\epsilon(0)}[\epsilon(0)I(\epsilon(0)-\epsilon_0)-J(\epsilon(0)-\epsilon_0)]=\left[\frac{\kappa_1^2}{\epsilon_1^2}+\frac{\eta^2}{\epsilon^2(0)}\right]I(\epsilon(0)-\epsilon_0) . \tag{2.21}$$

Solving for η gives the result

$$\eta^2=\frac{\epsilon_1\epsilon^2(0)\{[\epsilon(0)-\epsilon_1]I(\epsilon(0)-\epsilon_0)-J(\epsilon(0)-\epsilon_0)\}}{\epsilon(0)[\epsilon^2(0)-\epsilon_1^2]I(\epsilon(0)-\epsilon_0)-[\epsilon^2(0)+\epsilon_1^2]J(\epsilon(0)-\epsilon_0)} . \tag{2.22}$$

Note that if medium II is also linear than $J\equiv0$, and $\epsilon(0)=\epsilon$, and (2.22) reduces to the well-known result for the dispersion relation for p-polarized waves[22]

$$\eta^2\equiv\left[\frac{k_xc}{\omega}\right]^2=\frac{\epsilon\epsilon_1}{\epsilon+\epsilon_1} . \tag{2.23}$$

Equation (2.22) expresses the intensity-dependent dispersion relation for the p-polarized NLSP. However, instead of $\epsilon(0)$ or $B(0)$, it is the energy flux that is often more directly controllable experimentally. We now express the Poynting vector in terms of B using (2.2a) and (2.2b) as

$$\mathbf{S}(z)=\frac{c}{8\pi}\mathrm{Re}(\mathbf{E}\times\mathbf{H}^*)=\frac{c}{8\pi}\mathrm{Re}\left[\eta\frac{|B|^2}{\epsilon},0,-i\frac{B'B^*}{\epsilon}\right]$$
$$=\frac{c}{8\pi}\eta\frac{B^2}{\epsilon}\hat{\mathbf{x}}\ . \quad (2.24)$$

Note that S is entirely directed along $\hat{\mathbf{x}}$, as it should be. We are interested in the total energy flux per unit length in the y direction, S_x, which is defined by the equation

$$\int_{-\infty}^{+\infty} dz\,\mathbf{S}(z)=S_x\hat{\mathbf{x}}\ ,$$
$$=(S_x^{\mathrm{I}}+S_x^{\mathrm{II}})\hat{\mathbf{x}}\ , \quad (2.25)$$

where S_x^{I} and S_x^{II} are the contributions from regions I and II, respectively. Using (2.17), (2.10), and (2.24), we find

$$S_x^{\mathrm{I}}=\frac{c\eta}{8\pi}\int_{-\infty}^{0}dz\frac{B^2}{\epsilon}=\frac{c^2}{8\pi\omega}\eta\frac{B^2(0)}{2\epsilon_1\kappa_1}$$
$$=\frac{c\eta}{16\pi\epsilon_1\kappa_1}\frac{c}{\omega}\frac{\epsilon(0)}{2\eta^2-\epsilon(0)}$$
$$\times[\epsilon(0)I(\epsilon(0)-\epsilon_0)-J(\epsilon(0)-\epsilon_0)]\ . \quad (2.26)$$

To find S_x^{II}, which is defined as

$$S_x^{\mathrm{II}}=\frac{c}{8\pi}\eta\int_0^{+\infty}dz\frac{B^2}{\epsilon}=\frac{c}{8\pi}\eta\frac{c}{\omega}\int_0^{\infty}d\xi\frac{B^2}{\epsilon}\ , \quad (2.27)$$

there is no need to first calculate $B(\xi)$ and $\epsilon(\xi)$ and then perform the necessary integral over z. A more direct way is to express all relevant quantities in terms of ϵ. Therefore in (2.27) we make use of (2.13c) to write

$$d\xi=d\epsilon/\epsilon'=\pm[-2V(\epsilon)]^{-1/2}d\epsilon\ ,$$

and express B in terms of ϵ using (2.10). The resulting integral is given by

$$S_x^{\mathrm{II}}=\pm\frac{c}{8\pi}\eta\frac{c}{\omega}\int_{\epsilon(0)}^{\epsilon_0}d\epsilon\frac{\epsilon I(\epsilon-\epsilon_0)-J(\epsilon-\epsilon_0)}{(2\eta^2-\epsilon)[-2V(\epsilon)]^{1/2}}\ , \quad (2.28)$$

where the $+$ $(-)$ sign is to be used for the portion of the integral where ϵ' is positive (negative).

Since $\epsilon(0)$ can be written in terms of η by inverting (2.22), Eq. (2.28) expresses a relationship among S_x, k_x, and ω. For a given functional form of $\epsilon_2(x)$, the procedure to compute this relationship is as follows. For a fixed value of ω, we assume some value for $\epsilon(0)$.[23] This immediately specifies the value for η by (2.22). With this $\epsilon(0)$ we compute the integral in (2.29) to get S_x. We repeat this for other values of $\epsilon(0)$. In other words, we consider S_x and η as parametrically dependent on $\epsilon(0)$. In this way we can plot S_x as a function of η. In general, different curves will be generated depending on the value of ω.

However, note that if one works in a frequency region where the frequency dependences in ϵ_1, ϵ_0, and the form of $\epsilon_2(x)$ can be neglected, then the combination ωS_x depends on the wave vector k_x and frequency ω only through the dimensionless combination $(k_x c/\omega)^2\equiv\eta^2$. Thus all the curves collapse into a single one when ωS_x is plotted versus η. A specific example will be given in the following section.

III. SPECIFIC EXAMPLES

We now apply the results developed in Sec. II to a specific case of interest. We consider a material whose dielectric function has a nonlinear part proportional to the intensity, i.e.,

$$\epsilon_2(E^2)=\alpha E^2\ . \quad (3.1)$$

Both the self-focusing $(\alpha>0)$ and self-defocusing $(\alpha<0)$ cases will be considered, as well as different signs in ϵ_1 and ϵ_0.

The function $I(\epsilon-\epsilon_0)$ is then given by

$$I(\epsilon-\epsilon_0)=\frac{\epsilon-\epsilon_0}{\alpha}\ , \quad (3.2)$$

from which $J(\epsilon-\epsilon_0)$ defined in (2.11) can be obtained,

$$J(\epsilon-\epsilon_0)=\frac{(\epsilon-\epsilon_0)^2}{2\alpha}\ . \quad (3.3)$$

Equation (2.10) then gives B as a function ϵ in the form

$$B^2=\frac{\epsilon(\epsilon^2-\epsilon_0^2)}{2\alpha(2\eta^2-\epsilon)}\ . \quad (3.4)$$

The form of the "potential" $V(\epsilon)$ in (2.13c) can be obtained, using (2.13c), (3.2), and (3.4), with the result

$$V(\epsilon)=-\frac{1}{2}\frac{(\epsilon+\epsilon_0)\epsilon^2(2\eta^2-\epsilon)^2(\epsilon-\epsilon_0)^2(3\eta^2\epsilon-2\epsilon^2-\eta^2\epsilon_0)}{[\epsilon^2(2\eta^2-\epsilon)+\eta^2(\epsilon^2-\epsilon_0^2)]^2}\ . \quad (3.5)$$

Note that asymptotically away from the interface $(|z|\rightarrow+\infty)$ B and B' must go to 0 and so ϵ must go to ϵ_0. In that limit we find

$$V(\epsilon)\rightarrow-2(\eta^2-\epsilon_0)(\epsilon-\epsilon_0)^2,\quad \epsilon\rightarrow\epsilon_0\ . \quad (3.6)$$

It is clear that in order for ϵ to have a finite deviation from ϵ_0 for finite ξ we must have $\eta^2>\epsilon_0$. Combining this condition with a similar condition obtained previously for region I, i.e., $\eta^2>\epsilon_1$, we see that there is a long-wavelength cutoff for the existence of the present p-polarized NLSP, i.e., $\eta^2>\max(0,\epsilon_0,\epsilon_1)$. Below this cutoff the mode is no longer localized near the interface, but becomes radiative. From the Appendix we can see that this is true for the linear case as well.

The asymptotic form of $V(\epsilon)$ also determines the width of the NLSP in the direction perpendicular to the interface.[20] From (3.6) this width is given by $\frac{1}{2}(k_x-\omega\epsilon_0/c)^{-1/2}$ in region II. The corresponding width in region I is given by $(k_x-\omega\epsilon_1/c)^{-1/2}$ as can be seen from (2.17) and (2.18).

Putting (3.2) and (3.3) in (2.22) gives the result

$$\eta^2=\frac{\epsilon_1\epsilon^2(0)[\epsilon(0)-2\epsilon_1+\epsilon_0]}{\epsilon^3(0)+\epsilon^2(0)\epsilon_0-3\epsilon(0)\epsilon_1^2+\epsilon_1^2\epsilon_0}\ . \quad (3.7)$$

To analyze the behavior in the weak field limit we put

$$\epsilon(0)=\epsilon_0+\alpha\,|\,\mathbf{E}(0)\,|^2 \tag{3.8}$$

in (3.7) and expand the result to first order in $\alpha\,|\,E(0)\,|^2/\epsilon_0$ to obtain

$$\eta^2\cong\frac{\epsilon_1\epsilon_0}{\epsilon_1+\epsilon_0}\left[1+\frac{\epsilon_1\alpha\,|\,\mathbf{E}(0)\,|^2}{2\epsilon_0(\epsilon_1+\epsilon_0)}\right]. \tag{3.9}$$

The dispersion relation when expressed in terms of the degenerate nonlinear susceptibility $\chi^{(3)}$ is then given by

$$k_x\cong\frac{\omega}{c}\left[\frac{\epsilon_1\epsilon_0}{\epsilon_1+\epsilon_0}\right]^{1/2}\left[1+\frac{\pi\epsilon_1\chi^{(3)}\,|\,\mathbf{E}(0)\,|^2}{\epsilon_0(\epsilon_1+\epsilon_0)}\right]. \tag{3.10}$$

If one starts from (A3) for the p-polarized LSP, sets ϵ_{II} equal to the right-hand side of (3.8), and expands the result to first order in $\alpha\,|\,E(0)\,|^2/\epsilon_0$, Eq. (3.10) can also be obtained provided one uses an effective intensity which is half as large as the actual value.[11,12] This equation was used successfully by Chen and Carter to extract the magnitudes as well as the signs of $\chi^{(3)}$ for GaAs and Si from the experimentally determined intensity-dependent dispersion relations. However, we must point out that (3.10) was derived in the quasilinear limit with the expansion parameter $\chi^{(3)}\,|\,E(0)\,|^2/(\epsilon_1+\epsilon_0)$ which may not be small near very strong resonances where $\epsilon_1+\epsilon_0\approx 0$, even though $|\,\chi^{(3)}\,|\,|\,E(0)\,|^2$ may be small compared to unity. Thus (3.10) cannot be used in that case. The experiment of Chen and Carter[11,12] was conducted not very close to the plasmon resonance and the nonlinearities involved were sufficiently weak that nonlinear shifts of the plasmon resonance frequency described by (3.7) need not be considered.

A. Wave profiles

We see in Secs. II and III that in order for the existence of p-polarized NLSP we must have $\eta^2\geq\epsilon_1$, $\eta^2\geq\epsilon_0$. Thus we must work within the parameter regions given by the condition

$$\eta^2\geq\max(0,\epsilon_1,\epsilon_0) \tag{3.11}$$

To find the wave profiles we need to know more about the form of the "potential" $V(\epsilon)$ in (3.5). It is clear that $V(\epsilon)$ has simple roots at $-\epsilon_0$ and at

$$\epsilon_\pm\equiv[3\eta^2\pm\eta(9\eta^2-8\epsilon_0)^{1/2}]/4\,, \tag{3.12}$$

and has double roots at 0, ϵ_0, and $2\eta^2$. The cases $\epsilon_0>0$ and $\epsilon_0<0$ lead to different results and will be discussed separately.

Case (I): $\epsilon_0>0$

For $\epsilon_0>0$ the real roots of $V(\epsilon)$ are distributed as follows: $-\epsilon_0<0<\epsilon_-<\epsilon_0<\epsilon_+<2\eta^2$ regardless of the value of $\epsilon(0)$, as long as (3.11) is satisfied. However, because asymptotically B and B' must vanish and ϵ must approach ϵ_0 (or in other words, the "particle" must return to ϵ_0 eventually as "time"$\rightarrow+\infty$), the physically accessible region is given by $\epsilon_-\leq\epsilon\leq\epsilon_0$ for $\alpha<0$ and by $\epsilon_0\leq\epsilon\leq\epsilon_+$ for $\alpha>0$. Note that $\epsilon_\pm$, η, and thus the form of V all depend on $\epsilon(0)$, and therefore can all be modified by varying the field intensity. We also see that for $\epsilon_0>0$, $\epsilon(\xi)$ and therefore $\epsilon(0)$ must be positive. Since we have taken $B(0)$ to be non-negative one can see from (2.19) that

$$\text{sgn}[B'(0)]=\text{sgn}[\epsilon_1\epsilon(0)] \tag{3.13}$$

$$=\text{sgn}(\epsilon_1)\,. \tag{3.14}$$

(A) First we consider $\alpha>0$. In this case one can show from Eq. (3.14) that $B'(\xi)$ and $\epsilon'(\xi)$ always have the same sign.

(i) If ϵ_1 is positive then from (3.14) we have $B'(0)$ and therefore $\epsilon'(0)$ positive. The only possible classical motion with this initial condition in the nonlinear region is to have ϵ start from $\epsilon(0)$ at time $\xi=0$, increase to ϵ_+, turn around and eventually arrive at ϵ_0 as $\xi\rightarrow+\infty$ [Fig. 1(a)]. Thus in region I B increases from zero at $\xi=-\infty$ to $B(0)$ at $\xi=0$, and continues into region II with a change of slope, but still keeps on increasing until it reaches its maximum value

$$B_m=\frac{\epsilon_+}{2\alpha}\left[\frac{\epsilon_+^2-\epsilon_0^2}{2\eta^2-\epsilon_+}\right],$$

and then it decreases to zero exponentially as $\xi\rightarrow+\infty$ [Fig. 1(a)]. The presence of a peak within region II is clearly a manifestation of the intrinsic nonlinearities of the system, and the excitation of this wave will therefore require a minimum threshold power.

(ii) On the other hand, if ϵ_1 is negative then so is $\epsilon'(0)$. Thus after rising from zero at $\xi=-\infty$ to $B(0)$ at $\xi=0$, B decreases as soon as it enters region II and continues to decrease to zero as $\xi\rightarrow+\infty$. Therefore there is a cusp in B at the interface and no other peaks [Fig. 1(b)]. From the Appendix we see that this wave is qualitatively the

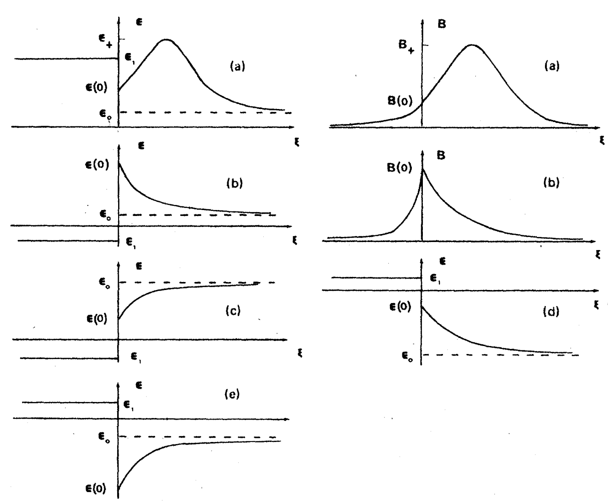

FIG. 1. Profiles of the dielectric function and the y component of the magnetic field along $\hat{z}$. The physical parameters are (a) $\epsilon_0>0$, $\alpha>0$, $\epsilon_1>0$; (b) $\epsilon_0>0$, $\alpha>0$, $\epsilon_1<0$; (c) $\epsilon_0>0$, $\alpha<0$, $\epsilon_1<0$, $\epsilon_s<\epsilon(0)<\epsilon_0$; (d) $\epsilon_0<0$, $\alpha>0$, $\epsilon_1>0$, $\epsilon_0<\epsilon(0)<\epsilon_s$; (e) $\epsilon_0<0$, $\alpha<0$, $\epsilon_1>0$. The magnetic field profiles for cases (c), (d), and (e) are qualitatively similar to that for case (b).

same as for the linear case. This is understandable since now ϵ_1 and ϵ_0 have opposite signs and thus p-polarized LSP are allowed.

(B) Next we consider $\alpha<0$. In this case one can show that $dB/d\epsilon$ vanishes at a single point ϵ_s which is given by $\eta^2[1+2\cos(\theta+4\pi/3)]$ with

$$\theta=\tfrac{1}{3}\cos^{-1}[1-(\epsilon_0/\eta^2)^2/2]\ .$$

Moreover ϵ_s lies between ϵ_- and ϵ_0, and $dB/d\epsilon$ is positive for $\epsilon_-<\epsilon<\epsilon_s$ and negative for $\epsilon_s<\epsilon<\epsilon_0$. At ϵ_s we have

$$\epsilon_s^3-3\eta^2\epsilon_s^2+\eta^2\epsilon_0^2=0\ , \tag{3.15}$$

and therefore V goes to $-\infty$. For $\epsilon_s<\epsilon\le\epsilon_0$ one can find a suitable solution for B from (3.4). However, that solution disappears as soon as ϵ is larger than ϵ_s, or equivalently that solution is not defined (or leads to complex values for ϵ) for $B>B_s$, where

$$B_s=\{\epsilon_s(\epsilon_0^2-\epsilon_s^2)/[2\alpha(2\eta^2-\epsilon_s)]\}^{1/2}\ .$$

(i) For $\epsilon_1>0$, we find from (3.14) that $B'(0)>0$. (a) If $\epsilon_s<\epsilon(0)<\epsilon_0$, then $\epsilon'(0)$ is negative. The particle will tend to move past ϵ_s and therefore we do not have any suitable solution. (b) The case where $\epsilon_-<\epsilon(0)<\epsilon_s$ can also be ignored.

(ii) For $\epsilon_1<0$, we have $B'(0)<0$. (a) If $\epsilon_s<\epsilon(0)<\epsilon_0$, then $dB/d\epsilon<0$, and so $\epsilon'(0)>0$. The profiles for ϵ and B are shown in Fig. 1(c). This mode is qualitatively similar to that found in the linear case, and thus we do not expect a minimum threshold power for it excitation. Note that ϵ_0 and ϵ_1 have opposite signs here so that p-polarized LSP can in fact exist in the zero intensity limit. (b) However, the case where $\epsilon_-<\epsilon(0)<\epsilon_s$ is clearly not allowed.

Case (II): $\epsilon_0<0$

For $\epsilon_0<0$, the distribution of the zeros of $V(\epsilon)$ is given by $\epsilon_0<\epsilon_-<0<-\epsilon_0<\epsilon_+<2\eta^2$ if $2\eta^2>-\epsilon_0$, and by $\epsilon_0<\epsilon_-<0<2\eta^2<\epsilon_+<-\epsilon_0$ if $2\eta^2<-\epsilon_0$. In either case, we only need to consider values of ϵ below ϵ_-. Thus ϵ, and therefore $\epsilon(0)$, are always negative. From (3.13) we find that

$$\mathrm{sgn}[B'(0)]=-\mathrm{sgn}(\epsilon_1)\ . \tag{3.16}$$

ϵ_s, which is determined by (3.15), exists and lies between ϵ_0 and ϵ_-. One can also show that $dB/d\epsilon$ is >0 for $\epsilon_0<\epsilon<\epsilon_s$, and is <0 for $\epsilon_s<\epsilon<\epsilon_-$. When $B<B_s$ one can find a real solution to ϵ which lies between ϵ_0 and ϵ_s. This is not so, however, for $B>B_s$.

(A) Consider first the case $\alpha>0$.

(i) In addition, if $\epsilon_1>0$, then from (3.15) we have $B'(0)<0$. (a) The situation where $\epsilon_s<\epsilon(0)<\epsilon_-$ is clearly not allowed. (b) On the other hand, if $\epsilon_0<\epsilon(0)<\epsilon_s$, then $\epsilon'(0)<0$. The results for ϵ and B as shown in Fig. 1(d) clearly may have a linear analog.

(ii) For $\epsilon_1<0$, we have $B'(0)>0$. (a) Again the case $\epsilon_s<\epsilon(0)<\epsilon_-$ needs not be considered. (b) But if $\epsilon_0<\epsilon(0)<\epsilon_s$, then $\epsilon'(0)$ is positive and thus ϵ can become larger than ϵ_s. This case must also be ignored.

(B) Next consider $\alpha<0$. ϵ is then never larger than ϵ_0, and therefore $dB/d\epsilon$ is always negative.

(i) For $\epsilon_1>0$, $B'(0)$ is negative and therefore $\epsilon'(0)$ is positive. The result for ϵ and B as shown in Fig. 1(e) clearly may have a linear analog.

(ii) For $\epsilon_1<0$, we have $B'(0)>0$ and so $\epsilon'(0)<0$. The classical motion is unbounded as the particle can move off to $-\infty$. This case does not correspond to any physically realizable situation and must be ignored.

B. Allowed physical regions and nonlinear surface plasmon

Next, we want to show that the modes discussed above do in fact exist, and we map out the regions in parameter space where they are allowed. To compute the allowed regions we need to simultaneously impose various conditions, and to check for internal consistencies. First we have the condition specified in (3.11) for the existence of surface modes. We find it more convenient to carry out our analysis in terms of the variables.

$$x\equiv\epsilon(0)/\epsilon_1 \ \text{ and } p\equiv\epsilon_0/\epsilon_1\ .$$

This condition can be broken down to three subcases. For $\max(0,\epsilon_1,\epsilon_0)=0$, i.e., ϵ_1 and ϵ_0 are both negative, we want only the regions where

$$(x-2+p)/(x^3+px^2-3x+p)\equiv r<0 \ \text{ and } p>0\ .$$

On the other hand, if $\max(0,\epsilon_1,\epsilon_0)=\epsilon_1$, i.e., ϵ_1 is positive and larger than ϵ_0, we must require $x^2r>1$ and $p<0$. And for $\max(0,\epsilon_1,\epsilon_0)=0$, i.e., ϵ_0 is positive and larger than ϵ_1, we want $x^2r>p>1$ if $\epsilon_1>0$, and $x^2r<p<0$ if $\epsilon_1<0$. The "resonance curve" is given by x^3+px^2-3x+p, or equivalently by $p=x(3-x^2)/(1+x^2)$, and is plotted as a solid curve in Fig. 2.

Next, we recall that to determine the possible wave profiles for each case, the form of $V(\epsilon)$ as well as the boun-

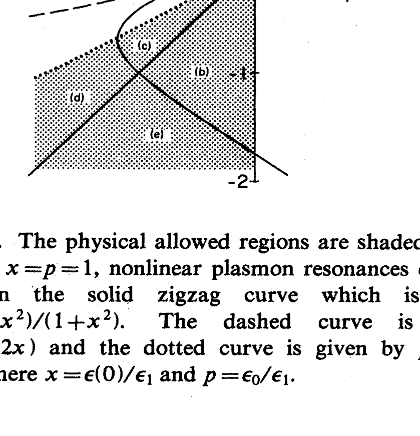

FIG. 2. The physical allowed regions are shaded. Except at the point $x=p=1$, nonlinear plasmon resonances occur everywhere on the solid zigzag curve which is given by $p=x(3-x^2)/(1+x^2)$. The dashed curve is given by $p=x(3-2x)$ and the dotted curve is given by $p=p_-$ [Eq. (3.19)], where $x=\epsilon(0)/\epsilon_1$ and $p=\epsilon_0/\epsilon_1$.

dary conditions must be known. However, all these quantities depend on $\epsilon(0)$ either explicitly or implicitly through their dependences on η. The wave profile deduced above must be consistent with the value of $\epsilon(0)$ assumed, i.e., the classical motions in potential V must start out at $\epsilon(0)$ in the nonlinear medium.

To be more specific, let us consider, for example, the case (I A i) in which ϵ is restricted to lie between ϵ_0 and ϵ_+. Thus we must require that $\epsilon_0 \le \epsilon(0) \le \epsilon_+$. To see if this condition is met, let us consider the situation where $\epsilon(0)=\epsilon_+$. Using Eqs. (3.12) and (3.7), we obtain a relationship between x and p which can be put into the form

$$(1+x^2)p^3+x(2x^3-x^2+2x-5)p^2 + x^2(4x^3-5x^2-4x+3)p + x^3(2x^3-3x^2-6x+9)=0 . \quad (3.17)$$

This equation turns out to be completely reducible yielding the result

$$(1+x^2)(p+x)\left[p-\frac{x(3-x^2)}{1+x^2}\right][p-x(3-2x)]=0 \quad (3.18)$$

The dashed curve in Fig. 2 is given by $p=x(3-2x)$.

There is one more problem left for cases (I B ii a) and (II A i b), where $\epsilon(0)$ must be larger than ϵ_s for the former, but less than ϵ_s for the latter. For given values of x and p, we must find the transition lines in parameter space which are determined by the condition $\epsilon(0)=\epsilon_s$. To do so, we substitute η from (3.7) into (3.15), and replace $\epsilon(0)$ by ϵ_s everywhere. Dividing the resulting equation by ϵ_1^3, we obtain a polynomial involving the parameters $x_s \equiv \epsilon_s/\epsilon_1$ and p. Again it is more convenient to consider p as the variable with coefficients which are functions of x_s. The resulting cubic equation is again completely reducible with the result

$$p^3+(x_s-2)p^2+x_s(x_s^2-3x_s+1)p+x_s^2(x_s^2-3x_s-3) = (p+x_s)(p-p_+)(p-p_-)=0 , \quad (3.19)$$

where

$$p_\pm=[1\pm 1-x_s(x_s^2-3x_s+3)]^{1/2} .$$

The solutions given by $p=-x_s$ and $p=p_+$ do not lie in the physically allowed region and can be ignored. The solution $p=p_-$ is plotted as a dotted curve in Figs. 2 and 3.

Combining all the above results, we finally obtain the complete map of physically allowed regions in parameter space for the various modes. Only the labeled regions shown in Fig. 2 are allowed.

For the linear case, surface-plasmon resonance happens only at the point $x=p=-1$, but the situation in the nonlinear case is rather rich. We find that, with the exception of the point $x=p=1$, plasmon resonance happens everywhere on the solid curve which is given by $p=x(3-x^2)/(1+x^2)$. As in the linear case, as the resonance is approached, $\eta \to +\infty$, and for a given value of $B(0)$, the electric fields at the interface become very large, and at the same time the wave forms along $\hat{z}$ become very narrow, and eventually collapse to the point $z=0$ in such a way that the energy flux remains finite.

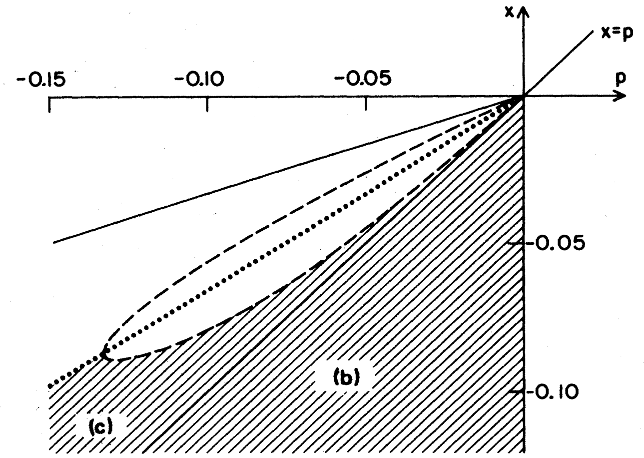

FIG. 3. Finer structures in the region where x and p are both small and negative.

For the case (I A i), Eq. (2.13b) with $V(\epsilon)$ given by (3.5) can in fact be integrated to obtain $\epsilon(\xi)$, which then gives $B(\xi)$ from (3.4). However, the resulting expression is too lengthy and will not be written out here. We believe that the integrals for the other cases can also be integrated, although we have not attempted to do so.

IV. DISCUSSION

There are a large number of problems left for future studies, some of them have already been pointed out by Maradudin.[2] Among these are those that can be readily treated by using the results developed here, or by straightforward extensions thereof. For example, we can easily study the effects due to saturations of the dielectric constants. It is important to consider these effects since the dielectric constant of a real material cannot increase (or decrease) indefinitely with increasing intensity, but must level off to some value depending on the particular material one is considering. By simply altering the appropriate boundary conditions we can also study the propagating of NLSP in layered dielectrics and waveguides.

Some problems, however, will require major modifications of our results. For example, instead of considering only the intensity dependence of the nonlinear dielectric function, one should consider all forms of electric field dependences that are allowed by crystal symmetries. Even if one confines to terms in the nonlinear polarization of order less than or equal to three, this problem has not been attempted. The effects due to dielectric losses have only been discussed within perturbation theory.[2] The effects of harmonic distortions, i.e., the presence of higher harmonic terms, have also not been treated. Most importantly, the question of stability of these nonlinear modes has not yet been addressed at all. A linear stability analysis similar to that applied to soliton studies should be extremely valuable.[24] As by-products of such an analysis the behavior of these waves in the presence of small perturbations such as surface irregularities, surface roughnesses, dielectric inhomogeneities, and additional external modulating fields can also be studied. It is possible that the nonlinear surface-guided wave discussed here may then lose some of its energy by emitting radiation away from the interface. Radiative solutions to the nonlinear Maxwell equations have in fact been discussed by Kaplan[25,26] in connection with the bistable reflection and refraction of light from nonlinear interfaces. It is clear that NLSP should play a major role in such phenomenon since experimentally the sum of reflected and transmitted energies at a nonlinear interface was observed to be less than the incident energy.[27]

The tremendous potential devices based on nonlinear interfaces have for fast, compact and relatively low-energy optical switching, and signal processing application has already been emphasized by Smith and Tomlinson.[27] Various waveguide configurations of nonlinear interface devices exhibiting optical triode and optical delimiter characteristics have been proposed and demonstrated experimentally using moderate-power cw lasers.[27,19] However, many important questions remain to be investigated. Much additional theoretical as well as experimental work will be needed to fully answer these questions.

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APPENDIX: *p*-polarized LSP

This appendix establishes some of the well-known results for the *p*-polarized LSP for comparisons with those of the corresponding NLSP. Such comparisons will help to bring out the unique features possessed by the nonlinear modes. Moreover, these result will be derived using our present approach, and in doing so, will illustrate in a simple way its usefulness.

Consider two linear media, one occupying region I, and the other region II. Their dielectric functions $\epsilon_{\rm I}$ and $\epsilon_{\rm II}$ are therefore constants, and without loss of generality we can take $\epsilon_{\rm I} > \epsilon_{\rm II}$. The first integral can be found from (2.16), with the result

$$\frac{(B')^2}{2} + V(B) = 0 , \tag{A1a}$$

where

$$V_{\rm I,II}(B) \equiv -\frac{(\eta^2 - \epsilon_{\rm I,II})}{2} B^2 . \tag{A1b}$$

The integration constant which plays the role of the total energy must be zero since asymptotically B and B' must vanish. It is clear that in order for B to assume nonzero values at finite ξ, we must require that $\eta^2 > \epsilon_{\rm I}$.

Next we must consider the boundary conditions in (2.15). The first condition implies that the "particle" jumps from $V_{\rm I}$ to $V_{\rm II}$ at time $\xi = 0$, where the particle is at the location $B_{\rm I} = B(0) = B_{\rm II}$. The second condition gives the relationship of the particle's velocity before and jump:

$$\frac{B'_{\rm I}}{\epsilon_{\rm I}} = \frac{B'_{\rm II}}{\epsilon_{\rm II}} . \tag{A2}$$

Note that both potentials are unbound for $|B| \to \infty$, since $B'_{\rm I}$ must be positive, or else the particle starting from $\xi = -\infty$ can never roll away from the origin down $V_{\rm I}$, and $B'_{\rm II}$ must be negative, so that the particle will roll up $V_{\rm II}$ to reach the origin eventually at $\xi = +\infty$. Thus we obtain the well-known fact that a necessary condition for the existence of *p*-polarized LSP is that $\epsilon_{\rm I}$ and $\epsilon_{\rm II}$ must have opposite signs.

In addition, if we square both sides of (A2) and then make use of (A1) we obtain in a simple way the well-known dispersion relation for *p*-polarized LSP:

$$\eta^2 = \frac{\epsilon_{\rm I}\epsilon_{\rm II}}{\epsilon_{\rm I} + \epsilon_{\rm II}} . \tag{A3}$$

The integrated Poynting vector along $\hat{\mathbf{x}}$ can be readily calculated to give

$$S_x = \frac{c^2 B^2(0)}{16\pi\omega} \left[\frac{\epsilon_{\rm I}^4 + \epsilon_{\rm II}^4}{-\epsilon_{\rm I}^3 \epsilon_{\rm II}^3} \right]^{1/2} . \tag{A4}$$

The electric field intensity is given by

$$E^2(\xi) = \frac{2\eta^2 - \epsilon_{\rm I,II}}{\epsilon_{\rm I,II}^2} B^2(0) e^{\pm 2\kappa_{\rm I,II}\xi} \tag{A5}$$

with

$$\kappa_{\rm I,II} \equiv (\eta^2 - \epsilon_{\rm I,II})^{1/2} . \tag{A6}$$

Since we have taken $\epsilon_{\rm I}$ to be larger than $\epsilon_{\rm II}$, in order to support a *p*-polarized wave $\epsilon_{\rm II}$ must be negative. The surface-plasmon resonance condition is then given by

$$\frac{1}{\epsilon_{\rm I}} \gtrsim -\frac{1}{\epsilon_{\rm II}} . \tag{A7}$$

From (A5), (A6), and (A3) we see that as we approach the resonance condition the field intensity grows very rapidly, at the same time the intensity becomes more and more localized at the interface, i.e., $\kappa_{\rm I,II}^{-1} \to 0$. As a result S_x remains finite there.

[1]For a review on the subject see, for example, Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).

[2]For a review on nonlinear surface polaritons, see A. A. Maradudin, *Proceedings of the 2nd International School on Condensed Matter Physics, Varna, Bulgaria* (Scientific, New York, 1982), and articles cited therein.

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[22]For recent discussions on linear surface polaritons see, for example, *Surface Polaritons,* edited by V. M. Agranovich and D. L. Mills (North-Holland, Amsterdam, 1982).
[23]Note that in general not all values for $E(0)$ are allowed. The physically allowed parameter regions will be computed for a specific model later in Sec. III B.
[24]See, for example, the review by A. R. Bishop, J. A. Krumhansl, and S. E. Trullinger, Physica **1D**, 1 (1980).
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