THEORY OF NONLINEAR GUIDED AND SURFACE PLASMON–POLARITONS IN DIELECTRIC FILMS

S. BAHER* and M. G. COTTAM†

Department of Physics and Astronomy, University of Western Ontario, London, Ontario N6A 3K7, Canada
cottam@uwo.ca

Received 5 April 2002

Calculations are presented for the dispersion relations of nonlinear plasmon–polariton modes in a layered dielectric structure consisting of a film bounded on each side by another medium. At least one of the media is a metal or semiconductor (such as InSb) with a real, isotropic, frequency-dependent dielectric function, characteristic of an electron plasma. In addition, both media may have a Kerr-type nonlinearity in their dielectric functions. Hence the resulting plasmon–polaritons, which we study in s polarization, are nonlinear and are found to occur in multiple branches. The theory is illustrated by numerical solutions of the dispersion relations for several geometries and physical parameters, and a comparison is made with previous plasmon–polariton results for linear dielectric media as a special case.

Keywords: Nonlinear waves; surfaces; films; plasmon–polaritons.

1. Introduction

In recent years there have been extensive experimental and theoretical investigations of the various collective excitations (such as phonons, plasmons, magnons, etc.) that can propagate in multilayer structures composed of layers of different materials. In particular, the surfaces and interfaces in these structures give rise to localized and guided excitations (or waves) and to quantization effects for the bulk modes (see, for example, Ref. 1). Hence the properties are quite different from those in bulk (i.e. effectively infinite) media.

It is well known that polarization fields associated with the collective excitations modify the electromagnetic waves traveling through a dielectric medium. This leads to a dependence of the dielectric function $\varepsilon(\omega)$ on the frequency $\omega$ of the wave and hence to a coupling between an electromagnetic wave and the collective excitation. Mixed modes of this type are known generally as polaritons, and in multilayered materials they have properties that are distinct from those in bulk samples. For example, the polaritons within any material layer produce electric and magnetic fields that extend to influence the excitations in other layers. In recent years a strong interest has been developed in surface polaritons. These are associated with some form of guiding surfaces and are characterized by having their amplitudes localized in the vicinity of the terminating interface(s), and their associated fields decay (usually exponentially) in the normal directions. These modes can exist in the frequency ranges that are above, below and between bulk polariton bands.1–3

In the case of a degenerate electron gas, such as occurs in a semiconductor or metal, the collective excitations are called plasmons and the coupled electromagnetic modes are called plasmon–polaritons. These coupled modes have both photon and plasmon content, depending on the wave vector and the multilayer geometry.4

*Permanent address: Physics Department, University of Lorestan, Khoramabad, Iran.
†Corresponding author. Fax: (519) 661-2033.
PACS numbers: 42.65.Sf, 71.36.+c, 71.45.Gm, 73.43.Lp
Many papers have appeared which discuss the propagation of guided and/or surface electromagnetic (EM) waves in layered optical structures. Most of these papers are devoted to the cases where it is assumed that the dielectric response functions of all the constituent materials may be expressed in terms of a linear relation between the induced polarization and the macroscopic electric field (see, for example, Refs. 4 and 5). However, EM waves in layered dielectric media that exhibit nonlinear effects have been the subject of increased theoretical and experimental interests.\textsuperscript{5–17} For example, there may be new surface modes which have no counterpart in the case of interfaces between linear media. Also, the inclusion of the nonlinearity effects is of interest for various device applications (for example in all-optical switching devices).\textsuperscript{18,19} Investigations of nonlinear EM waves have been reported in the literature for a single-interface geometry (between two semi-infinite media, one linear and one nonlinear) and a two-interface geometry consisting of a dielectric thin film embedded between two semi-infinite media. In the latter case it was assumed that either the film or the bounding material was linear. A theory for EM waves in a general multilayer structure where all media may be nonlinear has only recently been developed.\textsuperscript{17}

When any layer is optically nonlinear, its dielectric constant changes from its linear (field-independent) value \( \varepsilon_{0j} \) to a nonlinear form that depends on the electric field vector \( \mathbf{E} \). Usually this dependence is characterized as \( \varepsilon_{0j} + \alpha_j |\mathbf{E}|^2 \), which is known as a Kerr-type nonlinearity, and \( \alpha_j \) is a nonlinearity coefficient. In previous theoretical papers devoted to the propagation of nonlinear EM waves in multilayer dielectric structures with planar interfaces, the nonlinear dielectric constants were generally considered to be frequency-independent. However, if there are the collective excitations (such as plasmons or optical phonons) in any optically active layer of the system, then the dielectric response of that layer generally has a characteristic frequency dependence. This will lead to a dispersion relation for the polaritons in terms of frequency versus wave vector. This is well known in the linear regime, as mentioned previously, but studies of the polaritons arising due to the frequency dependence of nonlinear dielectric functions are still required. This provides the motivation for our present work.

In this paper we present a theoretical study of s-polarized, or transverse-electric (TE), surface plasmon–polariton modes supported by a three-layer planar structure consisting of a dielectric film of one material bounded symmetrically on each side by another dielectric material. The dielectric functions of the media, which are taken to be real and homogeneous, may in general be both frequency-dependent and nonlinear. This generalizes recent work by Vassiliev and Cottam\textsuperscript{17} in which any frequency dependence of the dielectric functions was neglected. The dielectric functions that enter into the present problem are assumed to have the following form with a Kerr-type nonlinearity:

\[
\varepsilon_j(\omega) = \varepsilon_{0j}(\omega) + \alpha_j E^2. \tag{1}
\]

The linear term \( \varepsilon_{0j}(\omega) \) for layer \( j \) depends on the frequency \( \omega \) in a way that corresponds to an electron plasma in a metal or a doped semiconductor. \( E \) is the magnitude of the electric field, and the nonlinearity coefficient \( \alpha_j \) (which can be either positive or negative) is assumed here to be independent of \( \omega \), for simplicity. We investigate the propagation of plasmon–polariton modes in this symmetric layered system, considering the situation where either one or both of the media are nonlinear. Likewise, at least one of the media is assumed to have a frequency-dependent dielectric function.

This paper is structured as follows. In Sec. 2 our theoretical model is introduced by generalizing Ref. 17 to include the frequency dependence of the dielectric functions. This is of particular importance when \( \omega \) is comparable to the plasma frequency in any layer. We first present in Sec. 3 the special case of a nonlinear film bounded by a linear frequency-dependent dielectric material (an electron plasma). Then another special case, namely the linear frequency-dependent dielectric film bounded by nonlinear semi-infinite media, is discussed briefly in Sec. 4. The general case, in which both of the media are nonlinear and have frequency-dependent dielectric functions, is presented in Sec. 5. Our conclusions, including some discussion regarding possible extensions of the work, are given in Sec. 6.

2. Model and Theoretical Method

The propagation of light in a multilayer system is governed by Maxwell’s equations within each
medium, together with the application of standard boundary conditions at the interfaces (see, for example, Refs. 1–3). The bulk and surface EM waves in our system are assumed to be s polarized (or TE). The corresponding electric and magnetic fields, for propagation in a direction parallel to the interfaces (chosen, without loss in generality, as the x direction) with in-plane wave number k and frequency ω, can be expressed as

\[
E = \left[0, E_y(k, \omega, z), 0\right] \exp[i(kx-\omega t)], \quad (2a)
\]

\[
H = \left[H_x(k, \omega, z), 0, H_z(k, \omega, z)\right] \exp[i(kx-\omega t)]. \quad (2b)
\]

The assumed layer geometry and choice of coordinate axes for the symmetric two-interface system are shown in Fig. 1. Corresponding to an electron plasma (ignoring damping), we take the frequency dependence of the linear part of the dielectric functions as

\[
\varepsilon_{0j}(\omega) = 1 - \frac{\omega_p^2}{\omega^2}, \quad (3)
\]

where \(\omega_p\) is the plasma frequency for layer \(j\) (where \(j = 1, 2, 3\) and 1 being the same). Also, we henceforth assume that \(\alpha_j \geq 0\) for the nonlinearity coefficient in each layer, which corresponds to the so-called self-focusing case.

Substituting the above form of \(E\) and \(H\) into Maxwell equations, and eliminating the magnetic field terms, yields

\[
\varepsilon(\omega) \frac{\partial^2 E_j}{\partial t^2} - c^2 \nabla^2 E_j = 0 \quad (4)
\]

for a nonmagnetic material, where \(E_j\) is short for \(E_y(k, \omega, z)\) in layer \(j\). We seek solutions of Eq. (4) for the spatial dependence of the polariton amplitude in the z direction (perpendicular to the layers).

Substituting Eq. (2a) into Eq. (5) gives

\[
\left[\frac{\partial^2}{\partial z^2} + \frac{\omega^2}{c^2} \varepsilon_j(\omega) - k^2\right] E_j = 0. \quad (5)
\]

This has a first integral of the form

\[
\left(\frac{\partial E_j}{\partial z}\right)^2 + \frac{\omega^2}{c^2} \left(\varepsilon_{0j}(\omega) - \frac{c^2 k^2}{\omega^2}\right) E_j^2 + \frac{\alpha_j E_j^4}{2}\right) = C_j(\omega), \quad (6)
\]

where \(C_j\) is the constant of integration, and we have substituted for the nonlinear dielectric function.

There are two physically different situations, depending on whether the coefficient of \(E_j^2\) in Eq. (6) is positive or negative in layer 2. The condition that \(c^2 k^2/\omega^2 < \varepsilon_{02}(\omega)\) corresponds to guided waves and \(c^2 k^2/\omega^2 > \varepsilon_{02}(\omega)\) to surface waves in the film (layer 2). We shall furthermore assume that \(c^2 k^2/\omega^2 > \varepsilon_{01}(\omega)\), which is a necessary condition for decaying waves in the bounding layers. Hence we may apply the boundary conditions that \(E\) and \(\partial E/\partial z\) both tend to zero as \(|z| \to \infty\) and are continuous for all finite values of \(z\). It will also be convenient to take \(\varepsilon_{02}(\omega) > \varepsilon_{01}(\omega)\) so that we may cover both the cases of guided waves and surface waves in the film, depending on the value of \(\omega\).

Finally, to obtain the dispersion relation of the nonlinear modes, Eq. (6) must be integrated one more time with respect to \(z\). From the continuity of the field amplitude \(E(z)\) and its spatial derivative at the boundaries between layers, one can easily find the integration constants \(C_1(= C_3)\) and \(C_2\). In doing this integration we follow the general approach developed in Ref. 17, since it has the advantage over earlier methods of providing a systematic classification of the modes and allows the treatment of interfaces where both materials are nonlinear. From symmetry considerations it follows that the electric field strengths \(E_u \equiv E(d/2)\) and \(E_l \equiv E(-d/2)\) at the upper and lower boundaries of the film, respectively, are related by

\[
E_u = \pm E_l. \quad (7)
\]

While this does not necessarily imply that the overall fields \(E(z)\) are symmetric or antisymmetric functions (see the discussion in Refs. 15 and 17) of \(z\) with respect to the mid-plane of the film \((z = 0)\), we shall
focus on solutions of this type. Rearranging Eq. (6) gives

\[
\frac{u(E)dE}{\sqrt{C_2(\omega) - (\omega^2/\varepsilon^2)[(\varepsilon_02(\omega) - (\varepsilon^2k^2/\omega^2)]E^2 + \frac{1}{2}\alpha_2E^4}} = dz,
\]

(8)

where \(u(E)\) is either 1 or \(-1\) according to whether \(\partial E/\partial z\) is positive or negative, respectively. It is important when integrating Eq. (8) across the film thickness to take account of changes in \(u(E)\), as emphasized in Ref. 17. It was shown there how this can be done conveniently by introducing phase trajectory diagrams (i.e. plots of \(\partial E/\partial z\) versus \(E\)) to characterize and enumerate the nodes of \(E\). Depending on the value of \((and, in particular, on its sign), different cases may arise for the integration, leading to various Jacobian elliptic integrals.\(^{20}\) For the geometry under consideration it may readily be shown that

\[
\varepsilon_j(\omega) = \begin{cases} 
\varepsilon_02(0) + \alpha_2E^2 \equiv 1 + \alpha_2E^2, & j = 2 \text{ or } |z| \leq \frac{1}{2}d, \\
\varepsilon_01(\omega) \equiv 1 - (\omega_p^2/\omega^2), & j = 1 \text{ or } |z| \geq \frac{1}{2}d.
\end{cases}
\]

(10)

From Eq. (9) and the previous assumption that \(\alpha_2 > 0\), it follows that \(C_2(\omega) > 0\) applies for all \(\omega\). Using the method described in Ref. 17, the result of integrating Eq. (8) across the thickness of the nonlinear film [and then using the condition in Eq. (7) for symmetric and antisymmetric modes] gives the implicit dispersion relation for the plasmon–polaritons as

\[
b\ \text{cn}\left(\frac{1}{2}qd - JK\right)\bigg|m\bigg| = 1,
\]

(11)

where \(J\) is an integer. This involves a Jacobian elliptic function\(^{20}\) of the type \(\text{cn}(x|m)\), which is a periodic function of the argument \(x\) and has a shape and period controlled by the auxiliary parameter \(m\). The value of \(K\) is a quarter of the period of the \(\text{cn}\) function, and so a complete description is obtained if integer \(J\) takes the four values 0, 1, 2 and 3. The other quantities in Eq. (11) are defined by

\[
b(\omega) = \sqrt{(n^2 - \varepsilon_02(\omega) + Q(\omega))/2p_2},
\]

(12)

\[
m(\omega) = (n^2 - \varepsilon_02(\omega) + Q(\omega))/2Q(\omega),
\]

(13)

\[
q(\omega) = (\omega/c)\sqrt{Q(\omega)},
\]

(14)

where we introduce an effective refractive index \(n = ck/\omega\) and the dimensionless variable \(P_j = \alpha_jE_i^2/2\) (scaling with respect to the boundary field \(E_i\) at \(z = -d/2\)). Also, we define

\[
Q(\omega) = \sqrt{(n^2 - \varepsilon_02(0))^2 + 4p_2(\varepsilon_02(0) - \varepsilon_01(\omega) + p_2)}.
\]

(15)

It is easily verified that \(Q(\omega)\) is a real function of frequency, and thus \(b\), \(m\) and \(q\) are all real in this case. The quantity \(q\) plays the role of an effective \(z\) component of the wave number for the polaritons in the film.

The electric field amplitude in the nonlinear layer (the thin film) can also be obtained following Ref. 17 as

\[
E(z) = E_i b\ \text{cn}\left([qz + JK]|m\right), \quad |z| \leq d/2.
\]

(16)

The continuity of \(\partial E/\partial z\) at the \(z = -d/2\) boundary (where \(E = E_i\)) implies that physical solutions correspond to \(\partial E/\partial z\) being positive at this position,
and thus from Eq. (11) that
\[
\text{sn}\left(\frac{1}{2}q d - J K \left\lfloor m \right\rfloor \right) < 0. \tag{17}
\]

The implicit dispersion relation for the plasmon–polaritons, as given by Eqs. (11) and (17), must be solved numerically. It is convenient to re-express the result in dimensionless form using a reduced frequency \( \Omega = \omega / \omega_{p1} \) and a reduced wave vector \( \kappa = c k / \omega_{p1} \). The necessary condition \( c^2 k^2 / \omega^2 > \varepsilon_{01}(\omega) \) (see Sec. 2) implies that the physical solutions correspond to \( \Omega < \Omega_c \), where
\[
\Omega_c = \sqrt{\kappa^2 + 1}. \tag{18}
\]

Equation (18) is just the dimensionless form of the dispersion relation of bulk plasmon–polariton modes for a linear material.\(^1\,^3\)

The factor \( q d \) in the nonlinear plasmon–polariton dispersion relation, Eq. (11), can be re-expressed as \( q d = \Omega R \sqrt{Q} \). Here \( R \) denotes a dimensionless quantity related to the film thickness \( d \) and the plasma wavelength \( \lambda_{p1} \) (defined\(^2\) as \( \lambda_{p1} = 2 \pi c / \omega_{p1} \)) in the bounding medium as
\[
R = 2 \pi (d / \lambda_{p1}). \tag{19}
\]

Our numerical calculations show that the regime of the most physical interest occurs when \( d \) and \( \lambda_{p1} \) are comparable in magnitude. In the following examples we take parameters typical of a doped semiconductor such as InSb (for which \( \hbar \omega_{p1} \approx 96 \text{ meV} \) or \( \omega_{p1} / 2 \pi \approx 2.3 \times 10^{13} \text{ Hz} \), which is in the infrared\(^1\)) for the bounding medium. This corresponds to a plasma wavelength \( \lambda_{p1} \approx 13 \mu \text{m} \). We have made calculations for the cases \( d = \lambda_{p1} \) (or \( R \approx 6.28 \)) and \( d = 2 \lambda_{p1} \) (or \( R \approx 12.56 \)), taking \( \varepsilon_{02} = 1 \) for the film. For a metal \( \hbar \omega_{p1} \) is much larger, typically several eV, and so \( \lambda_{p1} \) is then of order 0.1 \( \mu \text{m} \).

Some results for the nonlinear plasmon–polaritons are shown in Figs. 2–5, where the spectra consist of multiple branches. First, in Fig. 2 the reduced frequency \( \Omega \) is plotted versus the reduced wave vector \( \kappa \), taking \( R \approx 6.28 \) and a fixed value of \( p_2 = 0.05 \) for the scaled nonlinearity coefficient. Due to the periodicity property of the Jacobian elliptic function there is a sequence of different curves representing the solutions associated with the plasmon–polariton modes. Here (and subsequently) the branches to the spectrum are labeled as \( A_n, B_n, C_n \) or \( D_n \), corresponding to \( J = 0, 1, 2 \) or 3, respectively. The integer subscript \( n(=1, 2, \ldots) \) labels the \( n \)th branch in each case. The curve for \( \Omega_c \), which represents the upper boundary of the physical region, is also shown. Second, in Fig. 3 the curves of frequency \( \Omega \) versus the nonlinearity (expressed in terms of \( \sqrt{p_2} \)) is plotted for the same value of \( R \), where a fixed value \( \kappa = 0.25 \) is chosen for the in-plane wave number.

![Fig. 2. Dispersion relation for plasmon polaritons, showing reduced frequency \( \Omega \) versus reduced wave vector \( \kappa \) for the case of a nonlinear film and linear bounding medium. \( R = 6.28 \) and \( p_2 = 0.05 \). The line labeled \( \Omega_c \) marks the upper bound of the plasmon–polariton region and the branches of the spectrum are labeled as explained in the text.](image1)

![Fig. 3. As for Fig. 2 but showing \( \Omega \) versus the nonlinearity (in terms of \( \sqrt{p_2} \)) for a fixed value of \( \kappa = 0.25 \), taking \( R = 6.28 \).](image2)
If the value of $R$ is increased, it is found that the branches in the spectrum generally become closer together. This is illustrated in Fig. 4, which is similar to Fig. 2 but for the case of $R \approx 12.56$. Next we illustrate in Fig. 5 the effect on the dispersion relation curves ($\Omega$ versus $\kappa$) of reducing the nonlinearity coefficient $p_2$. Compared with Fig. 2 for $p_2 = 0.05$, the cases of $p_2 = 0.005$ and $p_2 = 0$ are shown in Figs. 5(a) and 5(b), respectively, where the curves shift to the right (increasing $\kappa$) as $p_2$ is reduced.

The above results can be generalized rather straightforwardly to cases where the quantity $\varepsilon_{02}$ for the linear part of the dielectric function in the film has also a frequency dependence. In fact, we find that the results are qualitatively similar to those described above provided that the inequality $\omega_{p1} > \omega_{p2}$ applies. Note that our previous results simply represent the special case of $\omega_{p2} = 0$. Generally, it is found that the frequency dependence of the dielectric functions produces additional modes, as compared with the frequency-independent case.\textsuperscript{17} The resulting dispersion curves show features that are distinctive of the plasma frequencies $\omega_{p1}$ and $\omega_{p2}$.

4. Linear Film Bounded by Nonlinear Layer

This special case is, in effect, the reverse situation to the one just considered, i.e. the dielectric function of the film is now assumed to be linear (and may also be frequency-dependent) while the two bounding media have a nonlinear frequency-dependent dielectric function of the Kerr type. The special case of nonlinear EM waves in the absence of any frequency dependence of the dielectric functions (as occurs when $\omega_{p1} = \omega_{p2} = 0$) has been discussed in earlier work.\textsuperscript{7,10,17}

As in Sec. 3, the plasmon–polariton dispersion relations are obtained by integrating Eq. (8) across the film thickness (except that we now have $p_2 = 0$). The solutions for the electric field amplitude are then matched to those in the semi-infinite nonlinear bounding media for $|z| > d/2$, which are expressible in terms of hyperbolic functions.\textsuperscript{1} The calculations are again restricted to the case of self-focusing media (so $p_1 > 0$).

Suppose we now assume that $C_2(\omega) > 0$, which from Eq. (9) will be the case when $\varepsilon_{02}(\omega) > \varepsilon_{01}(\omega)$, or equivalently $\omega_{p1} > \omega_{p2}$, and $p_1$ is not too large. Provided also that $\varepsilon_{02}(\omega) < c^2 k^2/\omega^2$, the integration across the film thickness eventually leads to guided waves with sinusoidally varying amplitude with
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Fig. 6. Dispersion relation ($\Omega$ versus $\kappa$) of plasmon-polaritons for the case of a linear film and nonlinear bounding medium with $p_1 = 0.05$ (such that $C_2 > 0$): (a) $R = 6.28$, (b) $R = 12.56$. The physical solutions lie below the curves for $\Omega_{c1}$ and $\Omega_{c2}$.

respect to the $z$ coordinate. The other case, where $\varepsilon_{02}(\omega) > c^2 k^2 / \omega^2 > \varepsilon_{01}(\omega)$, corresponds to surface plasmon-polaritons in the film.

In practice, it is more convenient to adopt a general procedure (by analogy with that described in detail in Ref. 17) to include a nonlinear part for the dielectric function in both materials. Integration is carried out across the film as in Sec. 3, and it is found for the case where $C_2(\omega) > 0$ that the plasmon-polariton dispersion relation can be expressed in the implicit form

$$ b \cosh \left( \frac{1}{2} q d - JK \right) = \eta, \quad (20) $$

which is a generalization of Eq. (11). The $\eta$ is a parameter provides a classification of the nonlinear modes$^{17}$ and can take the values 0 (for which $J = 0$ or 1) and 1 (for which $J = 0, 1, 2$ and 3). The definitions of the frequency-dependent quantities $b, m$ and $q$ are formally the same as in Eqs. (12)–(14) provided that the definition of $Q(\omega)$ is generalized to

$$ Q(\omega) = \sqrt{n^2 - \varepsilon_{02}(\omega)^2} + 4p_2 [\varepsilon_{02}(\omega) - \varepsilon_{01}(\omega)] + p_2 - p_1. \quad (21) $$

Afterwards we may take the limiting case of $p_2 \to 0$ for the film in Eq. (20) to recover the dispersion relation for the special case treated in this section.

Restrictions on the reduced frequency arise as necessary conditions in the same manner as in Sec. 3, where we found a condition $\Omega < \Omega_c$, with $\Omega_c$ given by Eq. (18). More generally, we find that there are two characteristic frequencies of interest, $\Omega_{c1}$ and $\Omega_{c2}$, given by

$$ \Omega_{c1} = \sqrt{\kappa^2 + \left( \frac{\omega_{p2}}{\omega_{p1}} \right)^2}, \quad \Omega_{c2} = \sqrt{\kappa^2 + 1 \frac{1}{p_1 + 1}}. \quad (22) $$

The former frequency just defines the “light line”$^3$ when $\omega_{p2} = 0$ and the latter frequency reduces to $\Omega_c$ when $p_2 = 0$. In the present case a condition for nonlinear guided plasmon-polaritons in the film becomes $\Omega < \min(\Omega_{c1}, \Omega_{c2})$.

Numerical examples of the plasmon-polariton dispersion relations, taking $p_2 \approx 0$ in the film and $p_1 = 0.05$ in the bounding material, are shown in Figs. 6(a) and 6(b) for $R = 6.28$ and 12.56, respectively. Both media are plasmas with the ratio $\omega_{p2}/\omega_{p1} = 0.5$, and the curves for $\Omega_{c1}$ and $\Omega_{c2}$ are included. It is again found that the branches to the spectrum become closer together as $R$ is increased.

5. General Case of Film and Bounding Layers All Nonlinear

In this general case the dispersion relations can be calculated for self-focusing nonlinear media (i.e. $p_1 > 0$ and $p_2 > 0$) with frequency-dependent dielectric functions (i.e. both plasma frequencies $\omega_{p1}$ and $\omega_{p2}$ may be nonzero). The procedure is a direct extension of that in the previous two sections, and follows the
approach in Ref. 17. Basically we need to integrate 
Eq. (8) across the film thickness, keeping track of any 
changes in sign associated with the $u(E)$ term. This 
requires an analysis of the behavior of the integration 
constant $C_2(\omega)$, which incorporates properties of the 
bounding medium as well as the film, and is defined 
in Eq. (21). We shall describe only the final results 
along with numerical examples.

First, if the frequency ranges and parameters are 
such that $C_2(\omega) > 0$, we find results for the sym-
metric and antisymmetric modes similar to those 
in Sec. 4. In particular, the formal expression in 
Eq. (20) for the dispersion relation in terms of cn 
functions still holds. However, two different cases 
may now arise. In case I, which is for frequen-
cies such that $c^2k^2/\omega^2 < \varepsilon_{02}(\omega)$, we conclude 
that the physical region of nonlinear plasmon–polaritons 
corresponds to reduced frequency $\Omega$ being less than 
$\min(\Omega_{c1}, \Omega_{c2})$. The other situation (case II) occurs 
when $c^2k^2/\omega^2 > \varepsilon_{02}(\omega)$, and we conclude that $\Omega$
must then satisfy the inequalities $\Omega_{c1} < \Omega < \Omega_{c2}$. 
Case I is analogous to the situation considered in 
Sec. 4 (see also Fig. 6), while case II only gives rise 
to physical modes for certain restricted ranges of the 
material parameters.

In Figs. 7(a) and 7(b) we show some examples of 
dispersion relation curves resulting from numerical 
calculations for case I ($C_2 > 0$) as described above. 
Here we plot $\Omega$ versus $\kappa$ for $R = 12.56$ in the former 
figure and $\Omega$ versus $\sqrt{p_2}$ for $R = 6.28$ in the latter, 
taking the ratios $\omega_{p2}/\omega_{p1} = 0.8$ and $p_1/p_2 = 0.5$ in both cases.

When $C_2(\omega) < 0$ a similar procedure is used for 
calculations of the dispersion relation. The formal 
results now involve elliptic functions$^{20}$ of the type 
dn$(x|m)$, as discussed in Ref. 17 for special cases. 
Specifically, in the present applications to plasmon–
polaritons a sufficient condition for $C_2 < 0$ is that 
$\omega_{p2}/\omega_{p1} > 1$ and $p_1/p_2 > 1$, as can be seen from 
Eqs. (3) and (9). In this case it is again found 
that the nonlinear plasmon–polaritons correspond to 
$\Omega$ being less than $\min(\Omega_{c1}, \Omega_{c2})$. By analogy with 
Eq. (20) the implicit dispersion relation is

$$b \ dn \left( \frac{1}{2} a d - J K \right) m = \eta',$$

where now $J = 0$ or 1, while $\eta'$ can take the values 
a or 1 where

$$a(\omega) = \sqrt{[n^2 - \varepsilon_{02}(\omega) - Q(\omega)]/[2p_2]}.$$  

Here $b(\omega)$ and $Q(\omega)$ are defined formally as in 
Eqs. (12) and (21), while the definitions of $m(\omega)$ and 
$q(\omega)$ are modified to

$$m(\omega) = 2Q(\omega)/[n^2 - \varepsilon_{02}(\omega) + Q(\omega)],$$

$$q(\omega) = (\omega/c)\sqrt{[n^2 - \varepsilon_{02}(\omega) + Q(\omega)]/2}.$$  

As an example, in Fig. 8 we plot $\Omega$ versus $\kappa$ for 
$R = 6.28$ and $p_2 = 0.05$, taking the ratios $\omega_{p2}/\omega_{p1} = 1.25$ and $p_1/p_2 = 1.05$. 

Fig. 7. Frequencies of plasmon–polaritons for the general case of a nonlinear film and nonlinear bounding medium, both being electron plasmas for case I ($C_2 > 0$). The plots show (a) $\Omega$ versus $\kappa$ for $R = 12.56$ and (b) $\Omega$ versus $\sqrt{p_2}$ for $R = 6.28$, taking the ratios $\omega_{p2}/\omega_{p1} = 0.8$ and $p_1/p_2 = 0.5$ in both cases.
Fig. 8. Dispersion relation (Ω versus κ) of plasmon–polaritons for the general case of a nonlinear film and nonlinear bounding medium, both being electron plasmas, for $C_2 < 0$. The parameters are $R = 6.28$ and $p_2 = 0.05$, taking the ratios $\omega_{p2}/\omega_{p1} = 1.25$ and $p_1/p_2 = 1.05$.

If only one of the ratios $\omega_{p2}/\omega_{p1}$ and $p_1/p_2$ is greater than 1, while the other lies between 0 and 1, it is still possible to have $C_2(\omega) < 0$ at some values of the frequency. The limitations on Ω are more complicated, and we do not discuss this behavior here.

6. Conclusions

In this paper the spectra of nonlinear $s$-polarized surface plasmon–polaritons have been investigated for different cases of a three-layered (two-interface) structure. It was assumed that at least one layer can be a doped semiconductor or a metal, thus having a dielectric function with a frequency-dependent part (involving the plasma frequency), and at least one other layer (the same or a different one) has a Kerr-type nonlinearity in the dielectric function. These calculations represent new theoretical predictions for the nonlinear plasmon–polaritons in this type of geometry, generalizing earlier works where the frequency dependence of the dielectric functions was ignored.

We started with the special cases of an L/NL/L structure and an NL/L/NL structure (denoting L = linear medium and NL = nonlinear medium), before generalizing to the case where all media are nonlinear (and frequency-dependent). In the case of self-focusing nonlinear media, i.e. $\alpha > 0$ for all layers, we obtained the implicit dispersion relations for the surface plasmon–polariton modes and solved them numerically. Our results were shown to lead to multiple branches for the plasmon–polariton modes, as seen from plots of the plasmon–polariton frequencies versus nonlinearity coefficients. As a check on the validity of this approach, we verified that we recover the previous calculations for nonlinear (frequency-independent) dielectrics and also for the linear (but frequency-dependent) dielectrics as special cases.

The results of this investigation are capable of being tested experimentally, for example by Raman scattering of light (in semiconductors) or by optical techniques such as the attenuated total reflection (ATR) method. Both experimental methods have already been applied to study linear polaritons in layered structures. On the theoretical side, it would be of interest and practical application to extend the calculations to cases where the plasmons are damped and/or where there is optical absorption. In both cases the procedures would need to be generalized to solve for complex frequency-dependent dielectric functions and complex electric field vectors for the $s$-polarized surface plasmon–polaritons. Other types of nonlinear polaritons, such as phonon–polaritons, can also be studied.

Acknowledgments

S. B. is grateful to the Ministry of Science and Technology of Iran and to the University of Lorestan for a scholarship. Partial support (to M. G. C.) for this project from the NSERC of Canada is acknowledged.

References