

AD-A062 250

CALIFORNIA UNIV IRVINE DEPT OF PHYSICS
A SURFACE PLASMON DISPERSION RELATION IN THE PRESENCE OF SPATIA--ETC(U)
SEP 78 A A MARADUDIN
TR-78-16
F/G 20/10
N00014-76-C-0121
NI

UNCLASSIFIED

[OF]

AD
A062250



END
DATE
FILMED

3 --79

DDC

REPORT DOCUMENTATION PAGE

LEVEL 13

INSTRUCTIONS
FOR COMPLETING FORM
REPORT NUMBER

1. REPORT NUMBER

2. GOVT ACCESSION NO.

4. TITLE (and Subtitle)

A Surface Plasmon Dispersion Relation
in the Presence of Spatial Dispersion

5. TYPE OF REPORT & PERIOD COVERED

FINAL

6. PERFORMING ORG. REPORT NUMBER

TR-78-16

8. CONTRACT OR GRANT NUMBER(s)

N00014-76-C-0121

7. AUTHOR(s)

A. A. Maradudin

9. PERFORMING ORGANIZATION NAME AND ADDRESS

University of California, Irvine
Irvine, CA 92717

10. PROGRAM ELEMENT, PROJECT, TASK
AREA & WORK UNIT NUMBERS

NR 392-001

11. CONTROLLING OFFICE NAME AND ADDRESS

Office of Naval Research, Physics Program
800 N. Quincy St.
Arlington, VA 22217

12. REPORT DATE

31 SEPT. 1978

13. NUMBER OF PAGES

25

14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)

15. SECURITY CLASS. (of this report)

UNCLASSIFIED

15a. DECLASSIFICATION/DOWNGRADING
SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Approved for public release; distribution unlimited

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

DDC

DEC 15 1978

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Surfaces, Dispersion relation, Surface Plasmons, Spatial dispersion

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

A dispersion relation for surface plasmons (retardation neglected) is obtained in the presence of spatial dispersion. The physical system considered is one of vacuum separated by a plane interface from a dielectric medium occupying the half-space $x_2 > 0$, and characterized by a nonlocal dielectric function $\epsilon(\mathbf{k}, \omega)$, assumed to be symmetric in x_2 and ω . Here $\mathbf{k}$ is a two-dimensional wave vector whose components are parallel to the interface, and ω is the frequency of the electromagnetic field in the medium. The dispersion relation has the form $1 + k_x \chi(\mathbf{k}, \omega | x_2=0, x_2=0) = 0$, and an explicit prescription for obtaining the function $\chi(\mathbf{k}, \omega | x_2, x_2')$ is presented. The use of the dispersion relation is illustrated by applying it to two examples: (1) a local dielectric constant; and (2) the nonlocal dielectric function used previously by Maradudin and Mills. In both cases previously obtained results are recovered.

K-Parallel

DD FORM 1 JAN 73 1473

EDITION OF 1 NOV 65 IS OBSOLETE

UNCLASSIFIED

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

AD A062250

DDC FILE COPY

"Reproduction in whole or in part is
permitted for any purpose of the
United States Government"

⑥ A SURFACE PLASMON DISPERSION RELATION
IN THE PRESENCE OF SPATIAL DISPERSION²

⑩ A. A. Maradudin

Department of Physics
University of California
Irvine, California 92717

⑫ 26 p.

⑪ 31 Sep 78

⑨ Final rept.

⑭ TR-78-16

DISTRIBUTION STATEMENT A
Approved for public release
Distribution Unlimited

DDC
RECEIVED
DEC 15 1978
B

* This research was supported in part by the Office of
Naval Research under Contract No. N00014-76-C-0121

Technical Report #78-16

⑮

403 659

set

ABSTRACT

A dispersion relation for surface plasmons (retardation neglected) is obtained in the presence of spatial dispersion. The physical system considered is one of vacuum separated by a plane interface from a dielectric medium occupying the half-space $x_3 > 0$, and characterized by a nonlocal dielectric function $\epsilon(\vec{k}_{\parallel}\omega|x_3x'_3)$, assumed to be symmetric in x_3 and x'_3 . Here $\vec{k}_{\parallel}$ is a two-dimensional wave vector whose components are parallel to the interface, and ω is the frequency of the electromagnetic field in the medium. The dispersion relation has the form $1 + k_{\parallel}\chi(\vec{k}_{\parallel}\omega|x_3=0, x'_3=0) = 0$, and an explicit prescription for obtaining the function $\chi(\vec{k}_{\parallel}\omega|x_3x'_3)$ is presented. The use of the dispersion relation is illustrated by applying it to two examples: (1) a local dielectric constant; and (2) the nonlocal dielectric function used previously by Maradudin and Mills. In both cases previously obtained results are recovered.

ACCESSION for		
NTIS	NTIS Section	☒
DDC	DDC Section	☐
UNANNOUNCED		☐
JUSTIFICATION		
BY		
DISTRIBUTION/AVAILABILITY CODES		
Dist.	AVAIL. and/or	SPECIAL
A		

I. Introduction

Several years ago Ritchie and Marusak⁽¹⁾ published a derivation of a dispersion relation for surface plasmons⁽²⁾ in the presence of spatial dispersion. Their result has the simple form

$$1 + \frac{k_{\parallel}}{\pi} \int_{-\infty}^{\infty} \frac{dk_3}{k^2 \epsilon(\vec{k}, \omega)} = 0 \quad . \quad (1.1)$$

Here $\epsilon(\vec{k}, \omega)$ is the frequency and wave vector dependent dielectric constant of the material out of which the semi-infinite medium is composed, along whose planar interface with vacuum the surface plasmon propagates. In Eq. (1.1) $k_{\parallel}$ is the magnitude of the projection of the three-dimensional wave vector $\vec{k}$ on the plane of the surface, and k_3 is the component of $\vec{k}$ normal to the surface.

A central assumption in the derivation of Eq. (1.1) is that the electrons in the semi-infinite dielectric medium are reflected specularly from the surface. It is this assumption which ultimately leads to the appearance of the bulk dielectric constant $\epsilon(\vec{k}, \omega)$ in Eq. (1.1).

However, in any real solid it is unlikely that all of the electrons are reflected specularly from the surface. It is more likely that some fraction are scattered specularly and the rest diffusely, or that more general boundary conditions obtain. In any case, it would seem to be desirable to have a dispersion relation for surface plasmons which does not depend, in its derivation, on some particular assumption about the nature of the interaction of an electron with the boundary. Moreover, it

would also seem to be desirable to have such a dispersion relation which is not based on the assumption that the dielectric properties of the medium supporting the surface plasmon are due to a specific collective excitation in the medium, viz. an electron plasma, so that it applies to systems in which these properties are associated with other electric dipole excitations such as IR active phonons and excitons, for example, or combinations of them.

It seemed to be worthwhile, therefore, to try to obtain a dispersion relation for surface plasmons in the presence of spatial dispersion of a form similar to that of Eq. (1.1), but without invoking the restrictive assumption of specular reflection of electrons at the boundary of the dielectric, in which the central role is played by the nonlocal dielectric constant of the medium, about which a minimum number of assumptions are made.

In this paper we present the derivation of such a dispersion relation. It is obtained in Section II, and its use illustrated by application to systems for which the dispersion relation is already known in Section III. A discussion of the results obtained is given in Section IV.

II. The Dispersion Relation

We assume a dielectric medium which occupies the semi-infinite region $x_3 > 0$. The region $x_3 < 0$ is occupied by vacuum. Because the system possesses infinitesimal translational invariance in directions parallel to the surface, the macroscopic electric field and the displacement in it can be expressed in the forms

$$\vec{E}(\vec{x}, t) = \vec{E}(\vec{k}_{\parallel} | x_3) e^{i\vec{k}_{\parallel} \cdot \vec{x}_{\parallel} - i\omega t} \quad (2.1a)$$

$$\vec{D}(\vec{x}, t) = \vec{D}(\vec{k}_{\parallel} | x_3) e^{i\vec{k}_{\parallel} \cdot \vec{x}_{\parallel} - i\omega t} \quad , \quad (2.1b)$$

where $\vec{k}_{\parallel} = \hat{x}_1 k_1 + \hat{x}_2 k_2$, $\vec{x}_{\parallel} = \hat{x}_1 x_1 + \hat{x}_2 x_2$, and $\hat{x}_1$ and $\hat{x}_2$ are two mutually perpendicular unit vectors in the plane of the dielectric-vacuum interface. Within the dielectric medium the relation between $\vec{D}(\vec{k}_{\parallel} | x_3)$ and $\vec{E}(\vec{k}_{\parallel} | x_3)$ is assumed to be

$$\vec{D}(\vec{k}_{\parallel} | x_3) = \int_0^{\infty} dx'_3 \epsilon(\vec{k}_{\parallel} | x_3 x'_3) \vec{E}(\vec{k}_{\parallel} | x'_3) \quad x_3 \geq 0 \quad , \quad (2.2)$$

where $\epsilon(\vec{k}_{\parallel} | x_3 x'_3)$ is assumed to be symmetric in the variables x_3 and x'_3 .

Thus, we assume the nonlocal dielectric tensor of the semi-infinite medium to be isotropic. This assumption is similar to and, because of the lower symmetry of the semi-infinite medium, may be as restrictive as the combined assumption by Ritchie and Marasak of an isotropic, nonlocal, bulk dielectric tensor and specular reflection of electrons from the boundary. The assumption of isotropy, however, can be removed from both the present theory and that of Ritchie and Marusak. The result in each case is a

78 10 23 143

more complicated dispersion relation. The assumption of specular reflection from the boundary, on the other hand, is essential in the Ritchie-Marusak approach, while it is not in the present treatment.

The Maxwell equation

$$\nabla \cdot \vec{D}(\vec{x}, t) = 0 \quad (2.3)$$

is now combined with Eq. (2.2) and with the equation of the electrostatic approximation

$$\vec{E}(\vec{x}, t) = - \nabla \varphi(\vec{x}, t) \quad , \quad (2.4)$$

where $\varphi(\vec{x}, t)$ is a scalar potential, to yield an integro-differential equation for $\varphi(\vec{k}_{\parallel\omega} | x_3)$, the Fourier coefficient of $\varphi(\vec{x}, t)$:

$$k_{\parallel}^2 \int_0^{\infty} dx'_3 \epsilon(\vec{k}_{\parallel\omega} | x_3 x'_3) \varphi(\vec{k}_{\parallel\omega} | x'_3) - \frac{d}{dx_3} \int_0^{\infty} dx'_3 \epsilon(\vec{k}_{\parallel\omega} | x_3 x'_3) \frac{d}{dx'_3} \varphi(\vec{k}_{\parallel\omega} | x'_3) = 0 \quad . \quad (2.5)$$

To solve Eq. (2.5) we begin by formally expanding both $\epsilon(\vec{k}_{\parallel\omega} | x_3 x'_3)$ and $(d/dx_3) \epsilon(\vec{k}_{\parallel\omega} | x_3 x'_3)$ in double series of functions complete and orthonormal in the semi-infinite interval $(0, \infty)$, and localized in the vicinity of the interface at $x_3 = 0$:

$$\epsilon(\vec{k}_{\parallel\omega} | x_3 x'_3) = \sum_{m,n=0}^{\infty} a_{mn}(\vec{k}_{\parallel\omega}) \varphi_m(x_3) \varphi_n(x'_3) \quad (2.6a)$$

$$\frac{d}{dx_3} \epsilon(\vec{k}_{\parallel\omega} | x_3 x'_3) = \sum_{m,n=0}^{\infty} b_{mn}(\vec{k}_{\parallel\omega}) \varphi_m(x_3) \varphi_n(x'_3) \quad (2.6b)$$

Although our final result will be independent of any particular choice for the $\{\varphi_m(x_3)\}$, a convenient choice, for definiteness, is the set of Laguerre functions defined by

$$\varphi_n(x_3) = \beta^{\frac{1}{2}} e^{-\frac{1}{2}\beta x_3} \frac{L_n(\beta x_3)}{n!}, \quad (2.7)$$

where $L_n(x_3)$ is the n^{th} Laguerre polynomial, and β is a real, positive parameter with the dimensions of an inverse length.

Because of our assumption that $\epsilon(\vec{k}_{\parallel\omega} | x_3 x'_3)$ is symmetric in x_3 and x'_3 , it is the case that the coefficients $\{a_{mn}(\vec{k}_{\parallel\omega})\}$ are symmetric in the subscripts m and n .

We next expand $\varphi(\vec{k}_{\parallel\omega} | x_3)$ and $(d/dx_3)\varphi(\vec{k}_{\parallel\omega} | x_3)$ in terms of the $\{\varphi_m(x_3)\}$:

$$\varphi(\vec{k}_{\parallel\omega} | x_3) = \sum_{n=0}^{\infty} A_n(\vec{k}_{\parallel\omega}) \varphi_n(x_3) \quad (2.8a)$$

$$\frac{d}{dx_3} \varphi(\vec{k}_{\parallel\omega} | x_3) = \sum_{n=0}^{\infty} B_n(\vec{k}_{\parallel\omega}) \varphi_n(x_3), \quad (2.8b)$$

so that

$$A_n(\vec{k}_{\parallel\omega}) = \int_0^{\infty} dx_3 \varphi_n(x_3) \varphi(\vec{k}_{\parallel\omega} | x_3) \quad (2.9a)$$

$$B_n(\vec{k}_{\parallel\omega}) = \int_0^{\infty} dx_3 \varphi_n(x_3) \frac{d}{dx_3} \varphi(\vec{k}_{\parallel\omega} | x_3) \quad (2.9b)$$

When we substitute Eqs. (2.6) and (2.8) into Eq. (2.5) and use the orthonormality of the $\{\varphi_n(x_3)\}$, we find the latter takes the form

$$k_{\parallel}^2 \sum_{n=0}^{\infty} a_{mn}(\vec{k}_{\parallel\omega}) A_n(\vec{k}_{\parallel\omega}) = \sum_{n=0}^{\infty} b_{mn}(\vec{k}_{\parallel\omega}) B_n(\vec{k}_{\parallel\omega}) \quad (2.10)$$

The coefficients $\{A_n(\vec{k}_{\parallel\omega})\}$ and $\{B_n(\vec{k}_{\parallel\omega})\}$ are not independent, however. If we integrate by parts the integral in Eq. (2.9b) we obtain the result that

$$B_n(\vec{k}_{\parallel\omega}) = -\varphi_n(0)\varphi(\vec{k}_{\parallel\omega} | 0+) - \int_0^{\infty} dx_3 \varphi(\vec{k}_{\parallel\omega} | x_3) \frac{d}{dx_3} \varphi_n(x_3) \quad (2.11)$$

where the notation $\varphi(\vec{k}_{\parallel\omega} | 0+)$ is intended to emphasize that it is the potential on the medium side of the interface that is being considered. If we substitute Eq. (2.8a) into the second term on the right hand side of Eq. (2.11), we obtain finally the relation

$$B_n(\vec{k}_{\parallel\omega}) = -\varphi_n(0)\varphi(\vec{k}_{\parallel\omega} | 0+) - \sum_{p=0}^{\infty} e_{np} A_p(\vec{k}_{\parallel\omega}) \quad (2.12)$$

where

$$e_{np} = \int_0^{\infty} dx_3 \left(\frac{d}{dx_3} \varphi_n(x_3) \right) \varphi_p(x_3) \quad (2.13a)$$

We note for future reference that e_{np} can be expressed equivalently as

$$e_{np} = \int_0^{\infty} dx_3 \int_0^{\infty} dx'_3 \varphi_p(x_3) \frac{d}{dx_3} \delta(x_3 - x'_3) \varphi_n(x'_3) = -\varphi_p(0) \varphi_n(0) - e_{pn} \quad (2.13b)$$

On combining Eqs. (2.10) and (2.12) we find that the coefficient $B_n(\vec{k}_{||\omega})$ is given by

$$B_n(\vec{k}_{||\omega}) = -k_{||}^2 \varphi(\vec{k}_{||\omega} | 0+) \sum_{p=0}^{\infty} \left[k_{||}^2 \tilde{a}^{-1} + \tilde{e} \tilde{a}^{-1} \tilde{b} \right]_{np}^{-1} \varphi_p(0) \quad (2.14)$$

where for convenience we have suppressed the argument $(\vec{k}_{||\omega})$ of the matrices $\tilde{a}(\vec{k}_{||\omega})$ and $\tilde{b}(\vec{k}_{||\omega})$ on the right hand side of this equation. It follows from this result and Eqs. (2.2), (2.4), (2.6a) and (2.9b) that

$$D_3(\vec{k}_{||\omega} | 0+) = k_{||}^2 \varphi(\vec{k}_{||\omega} | 0+) \sum_{m,n=0}^{\infty} \varphi_m(0) \left[k_{||}^2 \tilde{a}^{-1} + \tilde{e} \tilde{a}^{-1} \tilde{b} \right]_{mn}^{-1} \varphi_n(0) \quad (2.15)$$

We now turn to the vacuum region $x_3 < 0$. In this region the function $\varphi(\vec{k}_{||\omega} | x_3)$ is readily found to have the form

$$\varphi(\vec{k}_{||\omega} | x_3) = A e^{k_{||} x_3} \quad x_3 < 0 \quad (2.16)$$

where A is an arbitrary constant, while the displacement component $D_3(\vec{k}_{||\omega} | x_3)$ becomes

$$D_3(\vec{k}_{||\omega} | x_3) = -A k_{||} e^{k_{||} x_3} \quad x_3 < 0 \quad (2.17)$$

The boundary conditions in the problem are the continuity of $\varphi(\vec{x}, t)$ and $D_3(\vec{x}, t)$ across the surface $x_3 = 0$. The first of these yields the relation $\varphi(\vec{k}_{||}\omega|0+) = A$; the use of this result together with Eqs. (2.15) and (2.17) in the second boundary condition yields the dispersion relation for surface plasmons in the form

$$1 + k_{||} \sum_{m,n=0}^{\infty} \varphi_m(0) \chi_{mn}(\vec{k}_{||}\omega) \varphi_n(0) = 0, \quad (2.18a)$$

where

$$\chi_{mn}(\vec{k}_{||}\omega) = \left[k_{||}^2 \vec{a}^{-1} + \vec{e} \vec{a}^{-1} \vec{b} \vec{a}^{-1} \right]_{mn}^{-1}. \quad (2.18b)$$

It is useful to point out that $\chi_{mn}(\vec{k}_{||}\omega)$ is symmetric in m and n . For this purpose it suffices to show that the matrix $k_{||}^2 \vec{a}^{-1} + \vec{e} \vec{a}^{-1} \vec{b} \vec{a}^{-1}$ is a symmetric matrix. Inasmuch as the matrix $\vec{a}^{-1}$ is symmetric because the matrix $\vec{a}$ is, it is necessary to show only that the matrix $\vec{X} = \vec{e} \vec{a}^{-1} \vec{b} \vec{a}^{-1}$ is symmetric.

To show this we first relate the coefficients $\{b_{mn}(\vec{k}_{||}\omega)\}$ to the coefficients $\{a_{mn}(\vec{k}_{||}\omega)\}$. We see from Eq. (2.6b) that

$$b_{mn}(\vec{k}_{||}\omega) = \int_0^{\infty} dx_3 \int_0^{\infty} dx'_3 \varphi_m(x_3) \frac{d}{dx_3} \epsilon(\vec{k}_{||}\omega|x_3 x'_3) \varphi_n(x'_3). \quad (2.19)$$

An integration by parts yields

$$\begin{aligned} b_{mn}(\vec{k}_{||}\omega) = & - \int_0^{\infty} dx'_3 \varphi_m(0) \epsilon(\vec{k}_{||}\omega|0 x'_3) \varphi_n(x'_3) - \\ & - \int_0^{\infty} dx_3 \int_0^{\infty} dx'_3 \left(\frac{d}{dx_3} \varphi_m(x_3) \right) \epsilon(\vec{k}_{||}\omega|x_3 x'_3) \varphi_n(x'_3). \end{aligned} \quad (2.20)$$

We now substitute Eq. (2.6a) into the right hand side of this equation, and use Eq. (2.13a) to obtain

$$\begin{aligned}
b_{mn}(\vec{k}_{\parallel}\omega) &= - \sum_{r=0}^{\infty} \left\{ \varphi_m(0) \varphi_r(0) + e_{mr} \right\} a_{rn}(\vec{k}_{\parallel}\omega) \\
&= \sum_r e_{rm} a_{rn}(\vec{k}_{\parallel}\omega) .
\end{aligned} \tag{2.21}$$

The second equality in Eq. (2.21) follows from the use of Eq. (2.13b).

It follows from Eq. (2.21) that the matrix X can be written equivalently as

$$\vec{X} = \vec{e} \vec{a}^{-1} (\vec{e}^T \vec{a})^{-1} = \vec{e} \vec{a}^{-1} \vec{e}^T = \vec{X}^T, \tag{2.22}$$

where $\vec{M}^T$ denotes the transpose of the matrix $\vec{M}$. The matrix $\vec{X}(\vec{k}_{\parallel}\omega)$ is therefore symmetric, and can be written alternatively as

$$\chi_{mn}(\vec{k}_{\parallel}\omega) = \left[k_{\parallel}^2 \vec{a}^{-1} + \vec{e} \vec{a}^{-1} \vec{e}^T \right]_{mn}^{-1}. \tag{2.18c}$$

Equation (2.18) may have some interest of its own in connection with purely numerical studies of surface plasmons in spatially dispersive media. However, at this point we direct our efforts to re-expressing Eq. (2.18) in a form in which the $\{\varphi_m(x_3)\}$ no longer appear explicitly or implicitly.

We begin by introducing the function $\chi(\vec{k}_{\parallel}\omega | x_3 x'_3)$, which is defined by

$$\chi(\vec{k}_{\parallel}\omega | x_3 x'_3) = \sum_{m,n=0}^{\infty} \varphi_m(x_3) \chi_{mn}(\vec{k}_{\parallel}\omega) \varphi_n(x'_3) \quad x_3, x'_3 > 0. \tag{2.23}$$

In terms of this function the dispersion relation (2.18) becomes

$$1 + k_{\parallel} \chi(\vec{k}_{\parallel}\omega | \infty) = 0. \tag{2.24}$$

To obtain the function $\chi(\vec{k}_{\parallel}\omega|x_3x'_3)$ or more accurately, the equations determining it, we return to Eq. (2.18c), which we rewrite as

$$k_{\parallel}^2 \sum_p a_{mp}^{-1} \chi_{pn} + \sum_{prs} e_{mp} a_{pr}^{-1} e_{sr} \chi_{sn} = \delta_{mn}. \quad (2.25)$$

We now multiply both sides of this equation by $\varphi_m(x_3)\varphi_n(x'_3)$ and sum on m and n from zero to infinity. We recall the orthonormality and completeness conditions

$$\delta_{mn} = \int_0^{\infty} dx_3 \varphi_m(x_3) \varphi_n(x_3) \quad (2.26a)$$

$$\sum_{m=0}^{\infty} \varphi_m(x_3) \varphi_m(x'_3) = \delta(x_3 - x'_3). \quad (2.26b)$$

We next note that

$$\sum_{m,n=0}^{\infty} \varphi_m(x_3) a_{mn}^{-1}(\vec{k}_{\parallel}\omega) \varphi_n(x'_3) = \epsilon^{-1}(\vec{k}_{\parallel}\omega|x_3x'_3), \quad (2.27)$$

where $\epsilon^{-1}(\vec{k}_{\parallel}\omega|x_3x'_3)$ is the inverse of $\epsilon(\vec{k}_{\parallel}\omega|x_3x'_3)$ in the sense that

$$\begin{aligned} \int_0^{\infty} dx'_3 \epsilon^{-1}(\vec{k}_{\parallel}\omega|x_3x'_3) \epsilon(\vec{k}_{\parallel}\omega|x'_3x'_3) \\ = \int_0^{\infty} dx'_3 \epsilon(\vec{k}_{\parallel}\omega|x_3x'_3) \epsilon^{-1}(\vec{k}_{\parallel}\omega|x'_3x'_3) = \delta(x_3 - x'_3). \end{aligned} \quad (2.28)$$

We also use the relations

$$\sum_{m,n=0}^{\infty} \varphi_m(x_3) e_{mn} \varphi_n(x'_3) = - \left[\frac{d}{dx_3} \delta(x_3 - x'_3) + \delta(x_3) \delta(x'_3) \right] \quad (2.29a)$$

$$\sum_{m,n=0}^{\infty} \varphi_m(x_3) e_{nm} \varphi_n(x'_3) = \frac{d}{dx_3} \delta(x_3 - x'_3). \quad (2.29b)$$

The result of all of this is to transform Eq. (2.25) into

$$\begin{aligned}
 & k_{\parallel}^2 \int_0^{\infty} dx_3'' \epsilon^{-1}(\vec{k}_{\parallel\omega} | x_3 x_3'') \chi(\vec{k}_{\parallel\omega} | x_3'' x_3') - \\
 & - \frac{d}{dx_3} \int_0^{\infty} dx_3'' \epsilon^{-1}(\vec{k}_{\parallel\omega} | x_3 x_3'') \frac{d}{dx_3''} \chi(\vec{k}_{\parallel\omega} | x_3'' x_3') - \\
 & - \delta(x_3) \int_0^{\infty} dx_3'' \epsilon^{-1}(\vec{k}_{\parallel\omega} | 0 x_3'') \frac{d}{dx_3''} \chi(\vec{k}_{\parallel\omega} | x_3'' x_3') = \delta(x_3 - x_3'). \quad (2.30)
 \end{aligned}$$

Finally, we see that the function $\chi(\vec{k}_{\parallel\omega} | x_3 x_3')$ which satisfies the integro-differential equation

$$\begin{aligned}
 & k_{\parallel}^2 \int_0^{\infty} dx_3'' \epsilon^{-1}(\vec{k}_{\parallel\omega} | x_3 x_3'') \chi(\vec{k}_{\parallel\omega} | x_3'' x_3') - \\
 & - \frac{d}{dx_3} \int_0^{\infty} dx_3'' \epsilon^{-1}(\vec{k}_{\parallel\omega} | x_3 x_3'') \frac{d}{dx_3''} \chi(\vec{k}_{\parallel\omega} | x_3'' x_3') = \delta(x_3 - x_3'), \quad (2.31a)
 \end{aligned}$$

together with the boundary condition

$$\int_0^{\infty} dx_3'' \epsilon^{-1}(\vec{k}_{\parallel\omega} | 0 x_3'') \frac{d}{dx_3''} \chi(\vec{k}_{\parallel\omega} | x_3'' x_3') = 0, \quad (2.31b)$$

is a solution of Eq. (2.30). As we are seeking electric fields localized in the vicinity of the surface $x_3 = 0$, the boundary condition which must be imposed on $\chi(\vec{k}_{\parallel\omega} | x_3 x_3')$ at infinity is that

$$\lim_{x_3 \rightarrow \infty} \chi(\vec{k}_{\parallel\omega} | x_3 x_3') = 0. \quad (2.31c)$$

Equation (2.24), together with the prescription for obtaining $\chi(\vec{k}_{\parallel\omega} | x_3 x_3')$ given by Eqs. (2.31), is the central result of this section: the surface plasmon dispersion relation. In the next section we illustrate its use by applying it to two cases for which the surface plasmon dispersion relation is known.

III. Examples

In this section we solve Eqs. (2.31) for two choices of $\epsilon(\mathbf{k}_{\parallel}\omega|\mathbf{x}_3\mathbf{x}'_3)$, one local and the other nonlocal, for which the surface plasmon dispersion relation is already known, to illustrate the use of the dispersion relation, Eq. (2.24).

A. A Local Dielectric Tensor

We first consider the case in which $\epsilon(\mathbf{k}_{\parallel}\omega|\mathbf{x}_3\mathbf{x}'_3)$ is given by

$$\epsilon(\mathbf{k}_{\parallel}\omega|\mathbf{x}_3\mathbf{x}'_3) = \epsilon(\omega)\delta(\mathbf{x}_3 - \mathbf{x}'_3), \quad (3.1)$$

where $\epsilon(\omega)$ is independent of $\mathbf{k}_{\parallel}$ and of the coordinates $\mathbf{x}_3$ and $\mathbf{x}'_3$.

It follows that

$$\epsilon^{-1}(\mathbf{k}_{\parallel}\omega|\mathbf{x}_3\mathbf{x}'_3) = \frac{1}{\epsilon(\omega)} \delta(\mathbf{x}_3 - \mathbf{x}'_3). \quad (3.2)$$

The equation satisfied by $\chi(\mathbf{k}_{\parallel}\omega|\mathbf{x}_3\mathbf{x}'_3)$ in this case becomes

$$\frac{k_{\parallel}^2}{\epsilon(\omega)} \chi(\mathbf{k}_{\parallel}\omega|\mathbf{x}_3\mathbf{x}'_3) - \frac{1}{\epsilon(\omega)} \frac{d^2}{dx_3^2} \chi(\mathbf{k}_{\parallel}\omega|\mathbf{x}_3\mathbf{x}'_3) = \delta(\mathbf{x}_3 - \mathbf{x}'_3) \quad (3.3a)$$

together with the boundary condition

$$\frac{d}{dx_3} \chi(\mathbf{k}_{\parallel}\omega|\mathbf{x}_3\mathbf{x}'_3) \Big|_{x_3=0} = 0. \quad (3.3b)$$

The solution of Eq. (3.3a) which vanishes as $\mathbf{x}_3 \rightarrow \infty$ is

$$\chi(\mathbf{k}_{\parallel}\omega|\mathbf{x}_3\mathbf{x}'_3) = -\epsilon(\omega) \left\{ \frac{e^{-k_{\parallel}|\mathbf{x}_3 - \mathbf{x}'_3|}}{-2k_{\parallel}} + A e^{-k_{\parallel}\mathbf{x}_3} \right\}, \quad (3.4)$$

where the first term is the particular integral and the second is the complementary function. The coefficient A is determined

with the aid of Eq. (3.3b), with the result that

$$\chi(\mathbf{k}_{\parallel} \omega | x_3 x'_3) = \frac{\epsilon(\omega)}{2k_{\parallel}} \left\{ e^{-k_{\parallel} |x_3 - x'_3|} + e^{-k_{\parallel} (x_3 + x'_3)} \right\}. \quad (3.5)$$

When Eq. (3.5) is substituted into Eq. (2.24), we obtain as the surface plasmon dispersion relation

$$1 + \epsilon(\omega) = 0, \quad (3.6)$$

a well-known result for this case. ⁽³⁾

B. A Nonlocal Dielectric Constant

The second example we consider is based on the nonlocal dielectric function

$$\epsilon(\mathbf{k}_{\parallel} \omega | x_3 x'_3) = \epsilon_0 \delta(x_3 - x'_3) + i \frac{\omega_p^2}{2D\Gamma} e^{-\Gamma |x_3 - x'_3|} \quad x_3, x'_3 \geq 0, \quad (3.7)$$

which was used by Birman and Sein⁽⁴⁾, Maradudin and Mills⁽⁵⁾, and Agarwal, Pattanayak, and Wolf⁽⁶⁾, in studies of various optical optical properties of semi-infinite media in the presence of spatial dispersion. There are some unphysical features of the

model underlying this form for $\epsilon(\mathbf{k}_{\parallel} \omega | x_3 x'_3)$, as will be discussed in the following section. However, because results based on its use are available it serves as a useful example to illustrate Eq. (2.24), which is not restricted by these unphysical features. In Eq. (3.7) ϵ_0 is the optical frequency dielectric constant, which is assumed to be real and frequency independent; ω_p is a plasma frequency; D is a positive constant which defines the curvature of the exciton or transverse optical phonon dispersion curve at $\mathbf{k} = 0$; and Γ is given by

$$\Gamma = \left(\frac{\omega^2 - \omega_0^2}{D} - k_{\parallel}^2 + \frac{i\omega\gamma}{D} \right)^{\frac{1}{2}}, \quad (3.8)$$

so that $\text{Re } \Gamma > 0$, $\text{Im } \Gamma > 0$. The frequency ω_0 in Eq. (3.8) is that of the exciton or transverse optical phonon at $\vec{k} = 0$.

The equation for the inverse dielectric function $\epsilon^{-1}(\vec{k}_{\parallel}\omega|x_3x'_3)$, Eq. (2.28), in the present case takes the form

$$\begin{aligned} \epsilon^{-1}(\vec{k}_{\parallel}\omega|x_3x'_3) + \frac{i\omega_p^2}{2\epsilon_0 D \Gamma} \int_0^{\infty} dx'_3 e^{i\Gamma|x_3-x'_3|} \epsilon^{-1}(\vec{k}_{\parallel}\omega|x'_3x'_3) = \\ = \epsilon_0^{-1} \delta(x_3-x'_3). \end{aligned} \quad (3.9)$$

We apply the operator $(d^2/dx_3^2) + \Gamma^2$ to both sides of this equation to transform it into

$$\left(\frac{d^2}{dx_3^2} + \alpha^2 \right) \epsilon^{-1}(\vec{k}_{\parallel}\omega|x_3x'_3) = \frac{1}{\epsilon_0} \left(\frac{d^2}{dx_3^2} + \Gamma^2 \right) \delta(x_3-x'_3), \quad (3.10)$$

where

$$\alpha = \left(\frac{\omega^2 - \omega_L^2}{D} - k_{\parallel}^2 + \frac{i\omega\gamma}{D} \right)^{\frac{1}{2}} \quad (3.11a)$$

and

$$\omega_L^2 = \omega_0^2 + (\omega_p^2/\epsilon_0), \quad (3.11b)$$

so that $\text{Re } \alpha > 0$, $\text{Im } \alpha > 0$. We now use the fact that

$$\begin{aligned} \left(\frac{d^2}{dx_3^2} + \alpha^2 \right) \left\{ \delta(x_3 - x'_3) + \frac{\Gamma^2 - \alpha^2}{2i\alpha} e^{i\alpha|x_3-x'_3|} \right\} = \\ = \left(\frac{d^2}{dx_3^2} + \Gamma^2 \right) \delta(x_3 - x'_3) \end{aligned} \quad (3.12)$$

to write the solution of Eq. (3.10) in the form

$$\epsilon^{-1}(\vec{k}_{\parallel} \omega | x_3 x'_3) = \frac{1}{\epsilon_0} \delta(x_3 - x'_3) + \frac{\omega_p^2}{\epsilon_0 D} \frac{e^{i\alpha|x_3 - x'_3|}}{2i\alpha} + A e^{i\alpha x_3}. \quad (3.13)$$

The coefficient A is obtained by requiring that the expression (3.13) also satisfy the original integral equation (3.9). In this way we obtain finally the result that

$$\begin{aligned} \epsilon^{-1}(\vec{k}_{\parallel} \omega | x_3 x'_3) &= \frac{1}{\epsilon_0} \delta(x_3 - x'_3) + \\ &+ \frac{\omega_p^2}{\epsilon_0 D} \frac{1}{2i\alpha} \left[e^{i\alpha|x_3 - x'_3|} + \frac{\alpha - \Gamma}{\alpha + \Gamma} e^{i\alpha(x_3 + x'_3)} \right] \\ &= \frac{1}{\epsilon_0} \delta(x_3 - x'_3) + f(\vec{k}_{\parallel} \omega | x_3 x'_3). \end{aligned} \quad (3.14)$$

We note the result that

$$\left(\frac{d^2}{dx_3^2} + \alpha^2 \right) f(\vec{k}_{\parallel} \omega | x_3 x'_3) = \frac{\omega_p^2}{\epsilon_0 D} \delta(x_3 - x'_3). \quad (3.15)$$

The integral equation (2.31a) for the function $\chi(\vec{k}_{\parallel} \omega | x_3 x'_3)$ now takes the form

$$\begin{aligned} \frac{1}{\epsilon_0} \left(\frac{d^2}{dx_3^2} - k_{\parallel}^2 \right) \chi(\vec{k}_{\parallel} \omega | x_3 x'_3) &+ \frac{d'}{dx_3} \int_0^{\infty} dx_3'' f(\vec{k}_{\parallel} \omega | x_3 x_3'') \times \\ &\times \frac{d}{dx_3''} \chi(\vec{k}_{\parallel} \omega | x_3'' x'_3) - k_{\parallel}^2 \int_0^{\infty} dx_3'' f(\vec{k}_{\parallel} \omega | x_3 x_3'') \chi(\vec{k}_{\parallel} \omega | x_3'' x'_3) = \\ &= -\delta(x_3 - x'_3). \end{aligned} \quad (3.16)$$

We now apply the operator $(d^2/dx_3^2) + \alpha^2$ to both sides of this equation, and use Eq. (3.15), to obtain

$$\left(\frac{d^2}{dx_3^2} - k_{\parallel}^2\right)\left(\frac{d^2}{dx_3^2} - \beta^2\right)\chi(\vec{k}_{\parallel}\omega|x_3x'_3) = -\epsilon_0\left(\frac{d^2}{dx_3^2} + \alpha^2\right)\delta(x_3-x'_3), \quad (3.17)$$

where

$$\beta = \left(\frac{\omega_0^2 - \omega^2}{D} + k_{\parallel}^2 - \frac{i\omega\gamma}{D}\right)^{\frac{1}{2}} = -i\Gamma, \quad (3.18)$$

so that $\text{Re } \beta > 0$, $\text{Im } \beta < 0$. The solution of Eq. (3.17) which vanishes at infinity is given by

$$\begin{aligned} \chi(\vec{k}_{\parallel}\omega|x_3x'_3) = & -\frac{\epsilon_0}{2\beta - k_{\parallel}^2} \left[(\alpha^2 + k_{\parallel}^2) \frac{e^{-k_{\parallel}|x_3-x'_3|}}{2k_{\parallel}} - \right. \\ & \left. - (\alpha^2 + \beta^2) \frac{e^{-\beta|x_3-x'_3|}}{2\beta} \right] + C_1 e^{-k_{\parallel}x_3} + C_2 e^{-\beta x_3}. \end{aligned} \quad (3.19)$$

The two coefficients C_1 and C_2 in this solution are determined from the two equations obtained by requiring that the solution (3.19) satisfy the integro-differential equation (3.16) on the one hand, and from the boundary condition (2.31b) on the other. After some tedious analysis the following result is obtained:

$$\begin{aligned} \chi(\vec{k}_{\parallel}\omega|x_3x'_3) = & \frac{\epsilon(\omega)}{2k_{\parallel}} \left[e^{-k_{\parallel}|x_3-x'_3|} + \right. \\ & \left. + \frac{ik_{\parallel} + \alpha}{ik_{\parallel} - \alpha} \frac{ik_{\parallel} - \Gamma}{ik_{\parallel} + \Gamma} e^{-k_{\parallel}(x_3+x'_3)} \right] + \frac{(\epsilon(\omega) - \epsilon_0)}{2i\Gamma} e^{i\Gamma|x_3-x'_3|}, \end{aligned} \quad (3.20)$$

where

$$\epsilon(\omega) = \epsilon_0 + \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\omega\gamma} \quad (3.21)$$

is the bulk dielectric constant at $\vec{k} = 0$, and where Eq. (3.18) has been used.

Substitution of Eq. (3.20) into Eq. (2.24) yields as the dispersion relation for surface plasmons

$$1 + \frac{\epsilon(\omega)}{2} \left[1 + \frac{ik_{\parallel} + \alpha}{ik_{\parallel} - \alpha} \frac{ik_{\parallel} - \Gamma}{ik_{\parallel} + \Gamma} \right] + \frac{k_{\parallel}}{2i\Gamma} (\epsilon(\omega) - \epsilon_0) = 0. \quad (3.22)$$

This equation can be rearranged into the form

$$1 + \epsilon(\omega) = ik_{\parallel} \left[\epsilon(\omega) \frac{\alpha - \Gamma}{(k_{\parallel} + i\alpha)(k_{\parallel} - i\Gamma)} + \frac{\epsilon(\omega) - \epsilon_0}{2\Gamma} \right]. \quad (3.23)$$

Inasmuch as the right hand side of this equation is explicitly proportional to $k_{\parallel}$, in obtaining the frequency of the surface plasmon to lowest nonvanishing order in $k_{\parallel}$ it suffices to set $k_{\parallel} = 0$ in the expression in braces. We thus obtain the approximate dispersion relation

$$1 + \epsilon(\omega) \cong ik_{\parallel} \left[\epsilon(\omega) \left(\frac{1}{\Gamma} - \frac{1}{\alpha} \right) + \frac{\epsilon(\omega) - \epsilon_0}{2\Gamma} \right]_{k_{\parallel}=0}. \quad (3.24)$$

At this point we neglect the intrinsic damping in the dielectric medium and let γ tend to zero through positive values. In this limit we find that

$$\begin{aligned} \Gamma(k_{\parallel} = 0) &= iD^{-\frac{1}{2}}(\omega_0^2 - \omega^2)^{\frac{1}{2}} & \omega < \omega_0 \\ &= D^{-\frac{1}{2}}(\omega^2 - \omega_0^2) & \omega > \omega_0 \end{aligned} \quad (3.25)$$

$$\begin{aligned} \alpha(k_{\parallel} = 0) &= iD^{-\frac{1}{2}}(\omega_L^2 - \omega^2)^{\frac{1}{2}} & \omega < \omega_L \\ &= D^{-\frac{1}{2}}(\omega^2 - \omega_L^2)^{\frac{1}{2}} & \omega > \omega_L \end{aligned} \quad (3.26)$$

We next note that the equation $\epsilon(\omega) + 1 = 0$ is the surface plasmon dispersion relation in the absence of spatial dispersion. Consequently the frequency of the surface plasmon in the absence of spatial dispersion, ω_s , is the solution of the equation

$$\epsilon(\omega_s) + 1 = 0 \quad (3.27)$$

We thus write the solution of Eq. (3.24) in the form

$$\omega = \omega_s + \Delta\omega_s, \quad (3.28)$$

where $\Delta\omega_s$ is of first order in $k_{\parallel}$. Substituting Eq. (3.28) into Eq. (3.24) and linearizing the resulting equation in $\Delta\omega_s$ with the aid of Eq. (3.27) we obtain

$$\Delta\omega_s \cong \frac{ik_{\parallel}}{\epsilon'(\omega_s)} \left[\frac{1}{\alpha} - \frac{3+\epsilon_o}{2\Gamma} \right]_{\substack{k_{\parallel} \rightarrow 0 \\ \omega = \omega_s}} \quad (3.29)$$

If we denote the static ($\omega=0$) value of the bulk dielectric constant at $\vec{k} = 0$ by ϵ_s , we have the relation

$$\epsilon_s = \epsilon_o + (\omega_p^2/\omega_o^2) \quad (3.30)$$

With the substitution of Eq. (3.21) (with $\gamma=0$) into Eq. (3.27) we find that the frequency of the surface plasmon in the absence of spatial dispersion is given by

$$\omega_s^2 = \omega_o^2 + \frac{\omega_p^2}{1+\epsilon_o}, \quad (3.31a)$$

or equivalently

$$\omega_s = \left[\frac{1 + \epsilon_s}{1 + \epsilon_o} \right]^{\frac{1}{2}} \omega_o \quad (3.31b)$$

It is readily determined that

$$\omega_o < \omega_s < \omega_L \quad (3.32)$$

These inequalities, together with Eqs. (3.25) and (3.26), enable us to express $\Delta\omega_s$ given by Eq. (3.29) in the form

$$\begin{aligned} \Delta\omega_s = & \frac{D^{\frac{1}{2}} k_{\parallel}}{\epsilon'(\omega_s)} \left[\frac{1}{(\omega_L^2 - \omega_s^2)^{\frac{1}{2}}} - \frac{i(\epsilon_o + 3)}{2} \frac{1}{(\omega_s^2 - \omega_o^2)^{\frac{1}{2}}} \right] \\ & - \frac{D^{\frac{1}{2}} (1 + \epsilon_o)^{\frac{1}{2}} k_{\parallel}}{\omega_p \epsilon'(\omega_s)} \left[\epsilon_o^{\frac{1}{2}} - i \frac{\epsilon_o + 3}{2} \right], \end{aligned} \quad (3.33)$$

which agrees completely with the result for $\Delta\omega_s$ obtained by Maradudin and Mills⁽⁵⁾ on letting the speed of light tend to infinity in the surface polariton dispersion relation they obtained on the basis of the dielectric constant (3.7).

IV. Discussion

In this paper we have obtained an explicit dispersion relation for surface plasmons at the interface between vacuum and a semi-infinite dielectric medium occupying the half-space $x_3 > 0$ and characterized by a nonlocal dielectric constant $\epsilon(\vec{k}_{\parallel} \omega | x_3 x'_3)$. It should be remarked that the derivation of Eqs. (2.24) and (2.31) presented here is somewhat artificial. It should be possible to obtain these results more directly, but we have not attempted to do so. It also appears as if the use of Eqs. (2.24) and (2.31) to obtain the surface plasmon dispersion relation may involve lengthier calculations than are required in a more conventional approach based on a direct solution of the equations of electrostatics. Nevertheless, it still seems useful to have an explicit dispersion relation, Eq. (2.24), with a definite prescription for obtaining the function $\chi(\vec{k}_{\parallel} \omega | x_3 x'_3)$ entering it, Eq. (2.31). For example, in the case of dielectric constants of more complicated form than those considered in the preceding section Eq. (2.31) can serve as the basis for approximate determinations of $\chi(\vec{k}_{\parallel} \omega | x_3 x'_3)$, e.g. by variational methods.

The fact that the method developed here yields the same surface plasmon frequency, Eqs. (3.28), (3.31), and (3.33), as was obtained by a rather different method by Maradudin and Mills⁽⁵⁾ is of some independent interest for the following reason. The dielectric constant (3.7) is obtained by partially Fourier transforming the bulk dielectric constant

$$\epsilon(\vec{k}, \omega) = \epsilon_0 + \frac{\omega_p^2}{\omega_0^2 + Dk^2 - \omega^2 - i\omega\gamma} \quad (4.1)$$

according to

$$\epsilon(\vec{k}_{\parallel}\omega|x_3x'_3) = \int_{-\infty}^{\infty} \frac{dk_3}{2\pi} \epsilon(\vec{k},\omega) e^{ik_3(x_3 - x'_3)}, \quad (4.2)$$

and restricting x_3 and x'_3 to be positive. Recently Horing⁽⁷⁾ used $\epsilon(\vec{k},\omega)$ as given by Eq. (4.1) in the Ritchie-Marusak dispersion relation, Eq. (1.1), and obtained an expression for the frequency of a surface plasmon which differs from the Maradudin-Mills result, Eqs. (3.28), (3.31), and (3.33). This difference is not very surprising in fact. As we have noted in the Introduction, underlying the Ritchie-Marusak dispersion relation is the assumption that electrons are reflected specularly from the surface of the solid. The dielectric constant (3.7), on the other hand, represents the so-called dielectric approximation, in which the bulk dielectric constant is partially Fourier transformed according to Eq. (4.2), after which x_3 and x'_3 are restricted to be positive, so that the resulting $\epsilon(\vec{k}_{\parallel}\omega|x_3x'_3)$ is no longer a function of $x_3 - x'_3$, but depends on x_3 and x'_3 separately. It is known that the dielectric approximation does not conserve particle number⁽⁸⁾, and causes the surface to act as a source or sink of energy⁽⁹⁾, neither of which is the case when specular reflection is assumed. Thus a different physical situation is being considered in Horing's work from that assumed in the work of Maradudin and Mills. Consequently, it is not surprising that different surface plasmon frequencies are obtained in these two calculations. However, this example points up the desirability of having a dispersion relation for surface plasmons which is valid for more general physical situations than

is represented by the assumption of specular reflection at the surface. Equations (2.24) and (2.31) provide such a dispersion relation.

Acknowledgement

I am grateful to Professor N. J. M. Horing for informing me of his work and for a discussion of it. The present paper is a direct result of that work.

References

1. R. H. Ritchie and A. L. Marusak, Surface Science 4, 234 (1966).
2. In this paper we use the term "surface plasmon" to denote a surface localized electromagnetic wave in the limit that the effects of retardation are neglected.
3. See, for example, the discussion by D. L. Mills and E. Burstein, Repts. on Progress in Physics 37, 817 (1974).
4. J. L. Birman and J. J. Sein, Phys. Rev. B6, 2482 (1972).
5. A. A. Maradudin and D. L. Mills, Phys. Rev. B7, 2787 (1973).
6. G. S. Agarwal, D. N. Pattanayak, and E. Wolf, Phys. Rev. B10, 1447 (1974).
7. N. J. M. Horing, Proc. Seventh Int. Vacuum Conf. and the Third Int. Conf. on Solid Surfaces, edited by R. Dobrozemsky, F. Rüdenauer, F. P. Viehböck, and A. Breth (F. Berger & Söhne, Vienna, 1977), p. 419.
8. J. T. Foley and A. J. Devaney, Phys. Rev. B12, 3104 (1975).
9. M. F. Bishop and A. A. Maradudin, Phys. Rev. B14, 3384 (1976).