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Electromagnetic fields in spatially dispersive media*

G. S. Agarwal
Tata Institute of Fundamental Research, Bombay 5, India

D. N. Pattanayak and E. Wolf
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627
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The structure of the electromagnetic field in a spatially dispersive model medium occupying a plane parallel slab is obtained, free of several customary *ad hoc* assumptions made in other theories. The model medium is characterized by a dielectric response function appropriate to the neighborhood of an isolated-exciton transition frequency. The exact mode expansion for the electromagnetic field in the slab is derived and it is found that, unlike in the case of an unbounded medium, a single plane wave cannot be generated in the slab. An elementary solution (a single mode) is found to consist, in general, of six plane waves (four transverse and two longitudinal ones), coupled by two linear relations. These relations are shown to be equivalent to two nonlocal boundary conditions (of the form encountered in connection with the Ewald-Oseen extinction theorem in molecular optics), which the nonlocal contribution to the induced polarization must satisfy on the faces of the slab. This result resolves a long-standing controversy about the nature of the so-called additional boundary conditions that are generally believed to be required for solving problems of interaction of an electromagnetic field with a spatially dispersive medium. The results are applied to the problem of refraction and reflection on a spatially dispersive model medium occupying a half-space and a generalization of the classic formulas of Fresnel are obtained. The behavior of the reflected and transmitted waves as functions of the angle of incidence and of the frequency are illustrated by several figures. Our results are shown to differ from those obtained by Pekar in a well-known paper. The difference is traced to the nature of the additional boundary conditions postulated by Pekar; they are found to be inconsistent with the additional boundary conditions that we derive as an exact consequence of Maxwell's theory. Comparisons with several other theories, especially with those of Sein and Birman and of Maradudin and Mills are also made.

I. INTRODUCTION

In this paper a new theory will be developed, relating to the nature of electromagnetic fields in spatially dispersive media. Even though spatial dispersion was discovered as early as 1957 by Pekar[1] (in the context of the theory of excitons, where it appears rather naturally), the subject is even now a rather controversial one. Many experiments on spatial dispersion effects have been carried out,[2] but the interpretation of the resulting data has been hampered by the lack of a satisfactory theory.

It may be useful to begin by a brief review of the concepts relating to electromagnetic fields both in ordinary and in spatially dispersive media. Let us consider first an electromagnetic field in an ordinary homogeneous isotropic nonmagnetic dielectric which, to begin with, will be assumed to fill the whole space. Let $\vec{E}(\vec{r},t)$ and $\vec{D}(\vec{r},t)$ represent the electric field vector and the electric displacement vector, respectively, at a point $\vec{r}$ in the medium, at time t. Let $\hat{\vec{E}}(\vec{r},\omega)$ and $\hat{\vec{D}}(\vec{r},\omega)$ denote the Fourier transform of these vectors, defined as

$$\hat{\vec{E}}(\vec{r},\omega)=\int_{-\infty}^{+\infty}\vec{E}(\vec{r},t)e^{+i\omega t}dt, \qquad (1.1)$$

with a similar expression for $\hat{\vec{D}}(\vec{r},\omega)$. The assumption of linearity, homogeneity, and isotropy implies that the (electric) constitutive relation in the frequency domain is of the form

$$\hat{\vec{D}}(\vec{r},\omega)=\hat{\epsilon}(\omega)\hat{\vec{E}}(\vec{r},\omega). \qquad (1.2)$$

If we take the Fourier frequency transform of (1.2) and use the convolution theorem, we obtain the following constitutive relation in the time domain:

$$\vec{D}(\vec{r},t)=\int_{-\infty}^{+\infty}\epsilon(t-t')\vec{E}(\vec{r},t')dt', \qquad (1.3)$$

where, of course, $\epsilon(t)$ is the Fourier transform of $\hat{\epsilon}(\omega)$. The principle of causality demands that $\epsilon(t)=0$ for $t<0$.

According to Eq. (1.3), the relation between $\vec{D}$ and $\vec{E}$ is *local* in the *spatial argument* $\vec{r}$, but *nonlocal* in the *time argument* t; i.e., the electric displacement vector $\vec{D}(\vec{r},t)$ at a point $\vec{r}$ in the medium depends on the value of the electric field at that point only, but its value at time t depends on the values of the electric field not only at that particular instant of time, but also on other (earlier) times $t'<t$. This nonlocality with regard to time is responsible for the phenomenon of ordinary dispersion: In the course of time a wave packet in the medium will be dispersed.

As is well known, a frequency dependence of the dielectric constant $\hat{\epsilon}(\omega)$ that describes well the response of most linear homogeneous isotropic media is of the form[3]

$$\hat{\epsilon}(\omega) = 1 + \sum_j \frac{4\pi \alpha_j \omega_j^2}{\omega_j^2 - \omega^2 - i\omega\Gamma_j} . \qquad (1.4)$$

It may be derived from classical considerations based on the Lorentz oscillator model, in which case the ω_j's are the natural frequencies of the oscillators, the α_j's are proportional to the product of the atomic polarizability and the number of oscillators per unit volume, and the Γ_j's are phenomenological damping constants. A formula essentially of the form (1.4) (the Kramer-Heisenberg formula) may also be derived quantum mechanically, in which case the ω_j's are the transition frequencies, the α_j's are essentially the oscillator strengths, and the Γ_j's are related to the lifetimes of the excited states of the atoms. In the vicinity of an isolated resonance $\omega = \omega_0$ (1.4) may usually be approximated by the formula

$$\hat{\epsilon}(\omega) = \epsilon_0 + \frac{4\pi\alpha\omega_0^2}{\omega_0^2 - \omega^2 - i\omega\Gamma} , \qquad (1.5)$$

where ϵ_0, which is assumed to be independent of ω, is the "background dielectric constant" associated with all transitions other than the transition under consideration ($\omega = \omega_0$).

Under certain circumstances[4–6] the constitutive relation connecting $\vec{D}(\vec{r}, t)$ and $\vec{E}(\vec{r}, t)$ will be nonlocal not only in the time argument, but also in the spatial argument ($\vec{r}$), even when the medium is macroscopically homogeneous.[7] In that case the medium is said to be *spatially dispersive* and the constitutive relation (1.2) is replaced by[8]

$$\hat{\vec{D}}(\vec{r}, \omega) = \int \hat{\epsilon}(\vec{r} - \vec{r}\,', \omega) \hat{\vec{E}}(\vec{r}\,', \omega) d^3\vec{r}\,' . \qquad (1.6)$$

Alternatively, if we introduce the spatial Fourier transform of the three functions appearing in (1.6), i.e.,

$$\tilde{\vec{D}}(\vec{k}, \omega) = \int \hat{\vec{D}}(\vec{r}, \omega) e^{-i\vec{k}\cdot\vec{r}} d^3 r , \qquad (1.7)$$

etc., and take the Fourier transform of (1.6) we obtain the constitutive relation in the form

$$\tilde{\vec{D}}(\vec{k}, \omega) = \tilde{\epsilon}(\vec{k}, \omega) \tilde{\vec{E}}(\vec{k}, \omega), \qquad (1.8)$$

where $\vec{k}$ and ω must be regarded as independent variables.

It is known that a medium exhibits such a spatially dispersive response when the frequency ω of the electromagnetic field is close to the exciton transition frequency of the medium or when the wavelength of the field is of the order of magnitude of the lattice constant of the medium. Although much of the current research on spatial dispersion was motivated by Pekar's work, some phenomena associated with spatial dispersion were known previously,[9] e.g., optical activity[10]; it may be understood by assuming that the dielectric constant is a linear function of $\vec{k}$, viz.,

$$\tilde{\epsilon}(\vec{k}, \omega) = \epsilon_1(\omega) + \text{terms linear in } \vec{k} . \qquad (1.9)$$

Some of the most important differences between the nature of the electromagnetic field in an ordinary medium and in a spatially dispersive medium may be appreciated by comparing monochromatic-plane-wave solutions of Maxwell equations in the two media. We have, from the first two Maxwell equations, viz.,

$$\nabla \times \vec{E} = -\frac{1}{c}\frac{\partial \vec{H}}{\partial t}, \quad \nabla \times \vec{H} = \frac{1}{c}\frac{\partial \vec{D}}{\partial t} , \qquad (1.10)$$

on eliminating the magnetic field $\vec{H}$, that

$$\nabla \times (\nabla \times \vec{E}) + \frac{1}{c^2}\frac{\partial^2 \vec{D}}{\partial t^2} = 0 . \qquad (1.11)$$

We also have the Maxwell equation

$$\nabla \cdot \vec{D} = 0 . \qquad (1.12)$$

Let us now consider a monochromatic-plane-wave solution of Eqs. (1.11) and (1.12), i.e., a solution of the form

$$\vec{E}(\vec{r}, t) = \tilde{\vec{E}}(\vec{k}, \omega) e^{i(\vec{k}\cdot\vec{r} - \omega t)} , \quad \vec{D}(\vec{r}, t) = \tilde{\vec{D}}(\vec{k}, \omega) e^{i(\vec{k}\cdot\vec{r} - \omega t)} . \qquad (1.13)$$

On substituting from (1.13) into (1.11) and (1.12) and on using the constitutive relation (1.8) we obtain the equations

$$\vec{k} \times [\vec{k} \times \tilde{\vec{E}}(\vec{k}, \omega)] + \frac{\omega^2}{c^2} \tilde{\epsilon}(\vec{k}, \omega) \tilde{\vec{E}}(\vec{k}, \omega) = 0 \qquad (1.14)$$

and

$$[\vec{k} \cdot \tilde{\vec{E}}(\vec{k}, \omega)] \tilde{\epsilon}(\vec{k}, \omega) = 0 . \qquad (1.15)$$

There are two possible cases to be distinguished, depending on whether $\tilde{\epsilon}(\vec{k}, \omega) \neq 0$ or $\tilde{\epsilon}(\vec{k}, \omega) = 0$.

(i) If

$$\tilde{\epsilon}(\vec{k}, \omega) \neq 0 \qquad (1.16)$$

we must have, according to (1.15),

$$\vec{k} \cdot \tilde{\vec{E}}(\vec{k}, \omega) = 0. \qquad (1.17)$$

Using this condition in (1.14) we obtain the relation

$$\vec{k}^2 = \left(\frac{\omega}{c}\right)^2 \tilde{\epsilon}(\vec{k}, \omega). \qquad (1.18)$$

(ii) If

$$\tilde{\epsilon}(\vec{k}, \omega) = 0 \qquad (1.19)$$

Eq. (1.15) is satisfied and from (1.14) we see that in this case

$$\vec{k} \times \tilde{\vec{E}}(\vec{k}, \omega) = 0 . \qquad (1.20)$$

In the first case ($\tilde{\epsilon} \neq 0$) Eq. (1.17) shows that the waves are necessarily *transverse* and (1.18) is the *dispersion equation* for such waves. For an ordinary, (i.e., spatially nondispersive), medium, the dielectric constant is independent of $\vec{k}$ and hence, for a given frequency ω, (1.18) becomes a quadratic equation in $\vec{k}$. For a spatially dispersive medium, $\tilde{\epsilon}$ depends on $\vec{k}$ and Eq. (1.18) will, in general, be of order higher than second in the components of the propagation vector $\vec{k}$. Hence more monochromatic plane waves of a given frequency can be generated in a spatially dispersive medium than in an ordinary one. Moreover, since the phase velocity may be defined as $v_p = c/\mathrm{Re}\,[\tilde{\epsilon}(\vec{k},\omega)]^{1/2}$, monochromatic plane waves of the same frequency ω propagate with the same velocity in an ordinary medium. But in a spatially dispersive medium, they are seen to propagate with different velocities, in general, even if they propagate in the same direction.

In the second case ($\tilde{\epsilon}=0$), we see from Eq. (1.20) that the waves are *longitudinal* and Eq. (1.19) is the *dispersion equation* for such waves.

We will now briefly discuss the functional form of the dielectric constant $\tilde{\epsilon}(\vec{k},\omega)$, that may be obtained from the simplest microscopic model that takes account of spatial dispersion. The model is a natural generalization of the classical Lorentz oscillator model. Many workers[9,11] have found from quantum-mechanical calculations that in many cases of interest, $\tilde{\epsilon}(\vec{k},\omega)$ is well approximated in the vicinity of a resonance by an expression of the form

$$\tilde{\epsilon}(\vec{k},\omega)=\epsilon_0+\frac{4\pi\alpha(\vec{k})\omega_0^2(\vec{k})}{\omega_0^2(\vec{k})-\omega^2-i\omega\Gamma(\vec{k})} . \qquad (1.21)$$

For the case of a dielectric resonance due to an exciton band at frequency $\omega=\omega_e$ and wave number $k\simeq 0$ one has, in the effective-mass approximation,

$$\hbar\omega_0(\vec{k})\simeq\hbar\omega_e+\frac{\hbar^2}{2m_e^*}\vec{k}^2 ,$$

or (1.22)

$$\omega_0^2(\vec{k})\simeq\omega_e^2+\frac{\hbar\omega_e}{m_e^*}\vec{k}^2 ,$$

where m_e^* is the effective mass of the exciton. [Actually certain care must be exercised in employing approximations such as are implicit in (1.22), since $\omega_0(\vec{k})$ is not continuous at $\vec{k}=0$.[12]] If the $\vec{k}$ dependence of $\alpha(\vec{k})$ and $\Gamma(\vec{k})$ may be ignored, we obtain for $\tilde{\epsilon}(\vec{k},\omega)$ the expression

$$\tilde{\epsilon}(\vec{k},\omega)=\epsilon_0+\frac{\chi}{\vec{k}^2-\mu^2(\omega)} , \qquad (1.23)$$

where

$$\chi=\frac{4\pi\alpha m_e^*\omega_e}{\hbar} , \quad \mu^2(\omega)=\frac{m_e^*}{\hbar\omega_e}(\omega^2-\omega_e^2+i\omega\Gamma). \qquad (1.24)$$

We will assume throughout this paper that $m_e^*>0$. The case when $m_e^*<0$ may be treated in a similar way. The parameters α, Γ, and ϵ_0 are, of course, also taken to be positive. In the formal limit $m_e^*\to\infty$, the expression (1.23) becomes $\vec{k}$ independent and, as expected, has the form (1.5).

For a medium with a dielectric constant $\tilde{\epsilon}(\vec{k},\omega)$ given by (1.23), the dispersion equations (1.18) and (1.19) imply that for a plane wave of any particular frequency ω, in the vicinity of the resonance frequency ω_e, the permissible values of $\vec{k}^2$ are

$$\vec{k}_t^2=\frac{1}{2}\left(\epsilon_0\frac{\omega^2}{c^2}+\mu^2(\omega)\right)\pm\left[\chi\frac{\omega^2}{c^2}+\frac{1}{4}\left(\epsilon_0\frac{\omega^2}{c^2}-\mu^2(\omega)\right)^2\right]^{1/2} , \qquad (1.25)$$

$$\vec{k}_l^2=\mu^2(\omega)-\frac{\chi}{\epsilon_0} . \qquad (1.26)$$

Here, and in our subsequent discussion, suffixes t and l label quantities associated with transverse and with longitudinal waves, respectively.

Strictly speaking, the preceding discussion applies unambiguously only to a field in an infinite unbounded medium. However, in all realistic problems the medium is bounded. For example, in order to check various theories relating to spatial dispersion and excitons one often analyzes measurements on the field that is reflected from a spatially dispersive medium which forms a plane parallel slab (possibly of great enough thickness to consider it as effectively occupying a half-space). It is clear that from such measurements one may infer information about the various parameters entering the expression for the dielectric constant of the medium, such as the effective mass of the excitons or the natural frequencies of the transitions. In past treatments of such problems a difficulty was encountered, which we will illustrate by the problem of reflection from a spatially dispersive half-space. Suppose that a plane monochromatic wave, with electric field

$$\hat{\vec{E}}_0(\vec{r},\omega)=\vec{e}_0e^{i\vec{k}_0\cdot\vec{r}} , \qquad (1.27)$$

is incident on this half-space from vacuum and let

$$\hat{\vec{E}}_r(\vec{r},\omega)=\vec{e}_re^{i\vec{k}\cdot\vec{r}} , \qquad (1.28)$$

$$\hat{\vec{E}}_t(\vec{r},\omega)=\sum_j\vec{e}_t^{(j)}e^{i\vec{k}_t^{(j)}\cdot\vec{r}} \qquad (1.29)$$

represent the reflected and the transmitted fields, respectively. (There are, of course, similar expressions for the magnetic fields.) Now, if the squares of the propagation vectors $\vec{k}_t^{(j)}$ that appear in Eq. (1.29) are assumed to satisfy the dispersion equations (1.18) and (1.19), one finds that the usual electromagnetic boundary conditions at a surface of discontinuity do not determine uniquely the reflected and the transmitted fields from the knowledge of the incident field. For this reason many previous workers[13] found it necessary to introduce "additional boundary conditions" (to be abbreviated as abc's), suggested by various physical models for the behavior of excitons in the medium; the question of the correct form of the abc's has been a rather controversial subject. It is clear that the different abc's lead to different solutions for the reflected and the transmitted fields.

We consider it highly unsatisfactory that, with the form of the constitutive relation being given, it should be apparently necessary to go beyond Maxwell's theory to solve the problem of refraction and reflection by a mixture of quasiclassical and quantum-mechanical considerations. In this paper we present a theory of electromagnetic fields in spatially dispersive media based entirely on Maxwell's electrodynamics.[14] In Sec. II we introduce a spatially dispersive dielectric model medium and set up the appropriate integro-differential equation for the electric field in the medium. In Sec. III we derive the general solution of this equation, in the form of a mode expansion, for the case when our medium forms a plane parallel slab. We find that the electromagnetic field in the slab may always be represented as a superposition of plane waves (both transverse and longitudinal), whose wave vectors obey dispersion relations that are identical with those appropriate to plane waves in the medium, of the same kind, occupying the whole infinite (unbounded) space. However, we also find that, unlike in the case of the unbounded medium, a *single* plane wave cannot be generated inside the slab. An elementary solution (mode) consists in general of six plane waves (four transverse and two longitudinal), coupled by two linear relations which we call *mode-coupling conditions*. In Sec. IV we apply these results to the problem of refraction and reflection of a monochromatic plane wave incident from vacuum onto a half-space occupied by the spatially dispersive medium and find that our mode-coupling conditions ensure a unique solution to the problem; i.e., no abc's are needed to solve this problem. Our solution is shown to differ from Pekar's. The reason for this difference is traced to the fact that the abc's assumed by Pekar are inconsistent with the mode-coupling conditions. We also present curves that illustrate the behavior of the transmitted and reflected waves. In Sec. V we discuss the relations between mode-coupling conditions, additional boundary conditions, and extinction theorems for spatially dispersive media of a more general class, with regard to both constitutive relations and geometry. In Sec. VI we compare our theory with other theories relating to the nature of the electromagnetic field in spatially dispersive media. In this connection we wish to point out at the outset that our theory is similar, both in spirit and in the main conclusions, to that developed recently by Maradudin and Mills.[15] Our main conclusions are also in agreement with results obtained by Sein[16] and Birman and Sein[17] from a self-consistent analysis. However, unlike all theories proposed previously, our analysis is free of any *ad hoc* assumptions, being a rigorous consequence of Maxwell's electromagnetic theory applied to our model medium.

II. MODEL MEDIUM AND BASIC EQUATIONS

In this section we will derive a basic integro-differential equation which must be satisfied by the electric field in a homogeneous isotropic nonmagnetic spatially dispersive dielectric medium whose dielectric constant in the $\vec{k}$, ω domain is of the form given by Eq. (1.23), viz.,

$$\tilde{\epsilon}(\vec{k},\omega)=\epsilon_0+\frac{\chi}{\vec{k}^2-\mu^2(\omega)}\ , \tag{2.1}$$

where

$$\chi=\frac{4\pi\alpha m_e^*\omega_e}{\hbar}\quad,\quad \mu^2(\omega)=\frac{m_e^*}{\hbar\omega_e}(\omega^2-\omega_e^2+i\omega\Gamma)\ . \tag{2.2}$$

On taking the Fourier $\vec{k}$ transform of (2.1), we find that

$$\hat{\epsilon}(\vec{r},\omega)=\epsilon_0\,\delta(\vec{r})+\frac{\chi}{4\pi}\,G_\mu(|\vec{r}|)\ , \tag{2.3}$$

where

$$G_\mu(|\vec{r}|)=\frac{e^{i\mu|\vec{r}|}}{|\vec{r}|}\ . \tag{2.4a}$$

In (2.4a), μ is that root of the second expression in (2.2) for which

$$\mathrm{Re}\,\mu>0,\quad \mathrm{Im}\,\mu>0, \tag{2.4b}$$

consistent with our choice of the Fourier kernel $e^{+i\omega t}$ in (1.1) and with the obvious physical requirement that the waves in the medium should decay and not grow in amplitude as the wave propagates. From (1.6) and (2.3) we then obtain the following constitutive relation:

$$\vec{D}(\vec{r},\omega)=\epsilon_0\vec{E}(\vec{r},\omega)+\frac{\chi}{4\pi}\int G_\mu(|\vec{r}-\vec{r}\,'|)\vec{E}(\vec{r}\,',\omega)\,d^3r'. \tag{2.5}$$

Since from now on we will confine our attention to Fourier frequency components only, we have omitted in Eq. (2.5), and in subsequent equations, the caret symbol; i.e., we write $\vec{D}(\vec{r},\omega)$ in place of $\hat{\vec{D}}(\vec{r},\omega)$, etc.

From (2.5) we see that the polarization $\vec{P}(\vec{r},\omega) = (1/4\pi)[\vec{D}(\vec{r},\omega)-\vec{E}(\vec{r},\omega)]$ may be expressed in the form

$$\vec{P}(\vec{r},\omega)=\vec{P}_L(\vec{r},\omega)+\vec{P}_{NL}(\vec{r},\omega), \tag{2.6}$$

where $\vec{P}_L$ and $\vec{P}_{NL}$ are contributions that are local and nonlocal, respectively, in the electric field, and are given by

$$\vec{P}_L(\vec{r},\omega)=\frac{\epsilon_0-1}{4\pi}\,\vec{E}(\vec{r},\omega)\,, \tag{2.7a}$$

$$\vec{P}_{NL}(\vec{r},\omega)=\frac{\chi}{(4\pi)^2}\int G_\mu(|\vec{r}-\vec{r}'|)\,\vec{E}(\vec{r}',\omega)\,d^3r'\,. \tag{2.7b}$$

$\vec{P}_{NL}$ represents what is customarily referred to as exciton polarization. Now, strictly speaking, the form (2.1) of the dielectric constant $\tilde{\epsilon}(\vec{k},\omega)$ applies only to the idealized case when the dielectric occupies the whole infinite (unbounded) space. The preceding relations apply, therefore, rigorously to this case only and the volume integrals in Eqs. (2.5) and (2.7b) extend then over the whole space. When dealing with the more realistic case of a dielectric occupying only a portion V of the whole space one has to resort to approximations, as the exact form of the dielectric constant for such cases is not known and is clearly exceedingly hard to determine, because of complicated effects arising from the presence of the boundary of the medium. It seems reasonable, however, to assume that as long as one is interested only in bulk effects, such as reflection and refraction, and not with the detailed nature of the field close to the boundary, one may, to a good approximation, retain the constitutive relation (2.5), with the integration extending over the volume V occupied by the dielectric.[18] One may expect that this approximation will be especially appropriate for media whose linear dimensions are large compared to the effective range of the Green's functions G_μ [of the order of $(\mathrm{Im}\mu)^{-1}$] that characterizes the size of the effective region of nonlocal response. We will therefore take as our model medium a spatially dispersive medium which is characterized by the constitutive relation

$$\vec{D}(\vec{r},\omega)=\epsilon_0\vec{E}(\vec{r},\omega)+\frac{\chi}{4\pi}\int_V G_\mu(|\vec{r}-\vec{r}'|)\vec{E}(\vec{r}',\omega)\,d^3r'. \tag{2.8}$$

We now eliminate the electric displacement vector $\vec{D}(\vec{r},\omega)$ between the Fourier time transform of Eq. (1.11) and the constitutive relation (2.8) and obtain the following integro-differential equation that the electric field in the spatially dispersive medium has to satisfy:

$$\nabla\times[\nabla\times\vec{E}(\vec{r},\omega)]-k_0^2\epsilon_0\vec{E}(\vec{r},\omega)=\frac{\chi k_0^2}{4\pi}\int_V G_\mu(|\vec{r}-\vec{r}'|)\times\vec{E}(\vec{r}',\omega)\,d^3r', \tag{2,9}$$

where

$$k_0=\omega/c\ . \tag{2.10}$$

Equation (2.9) is the basic equation from which our considerations start.

III. GENERAL SOLUTION OF EQ. (2.9) AND THE NATURE OF THE ELECTROMAGNETIC FIELD IN A SPATAILLY DISPERSIVE PLANE-PARALLEL SLAB

We will now obtain the general solution of the integro-differential equation (2.9) for the electric field, when the domain V occupied by the spatially dispersive medium is a plane parallel slab $0\le z\le d$. Equation (2.9) then takes the form

$$\nabla\times[\nabla\times\vec{E}(\vec{r},\omega)]-k_0^2\epsilon_0\vec{E}(\vec{r},\omega)=\frac{\chi k_0^2}{4\pi}\int\int_{-\infty}^{+\infty}dx'dy'\times\int_0^d dz'\,G_\mu(|\vec{r}-\vec{r}'|)\vec{E}(\vec{r}',\omega). \tag{3.1}$$

As we will see shortly, the form of this equation allows us to express the general solution as an *angular spectrum* of plane waves.[19(a)] For this purpose we first express the electric field as a two-dimensional Fourier integral with respect to the x and y variables:

$$\vec{E}(\vec{r},\omega)=\int\int_{-\infty}^{+\infty}\hat{\vec{E}}(u,v;z;\omega)\,e^{i(ux+vy)}\,du\,dv. \tag{3.2}$$

Next we express the kernel G_μ, defined by (2.4), of the integral transform in (3.1) in this form also. The explicit expression, which is a generalization of a well-known formula due to Weyl,[19(b)] is

$$G_\mu(|\vec{r}-\vec{r}'|)=\frac{i}{2\pi}\int\int_{-\infty}^{+\infty}\frac{1}{w_\mu}\times\exp[i(u(x-x')+v(y-y')+w_\mu|z-z'|)]\,du\,dv, \tag{3.3}$$

where

$$w_\mu=(\mu^2-u^2-v^2)^{1/2}\,, \tag{3.4a}$$

and the square root in (3.4a) is defined so that

$$\mathrm{Re}w_\mu>0,\quad \mathrm{Im}w_\mu>0\ . \tag{3.4b}$$

That the choice of the real and imaginary parts of w_μ, indicated by Eq. (3.4b), is possible, follows readily from (3.4a) and the definition of $\mu^2(\omega)$ given in (1.24), if we recall that $\Gamma>0$.

On substituting from (3.2) and (3.3) into (3.1), we find after a straightforward calculation that $\hat{\vec{E}}(u,v;z;\omega)$ satisfies the equation

$$(u^2+v^2-k_0^2\epsilon_0)\hat{\vec{E}}-\frac{\partial^2}{\partial z^2}\hat{\vec{E}}+\vec{L}=\frac{i\chi k_0^2}{2}\int_0^d\frac{e^{iw_\mu|z-z'|}}{w_\mu}\times\hat{\vec{E}}(u,v;z';\omega)\,dz', \tag{3.5}$$

where $\vec{L}$ is a vector with components

$$L_x=iu\xi,\quad L_y=iv\xi,\quad L_z=\frac{\partial\xi}{\partial z}\,;\qquad \xi=iu\hat{E}_x+iv\hat{E}_y+\frac{\partial\hat{E}_z}{\partial z}\ . \tag{3.6}$$

We note that if the $\vec{E}$ field were transverse ($\nabla\cdot\vec{E}=0$) then ξ and hence the vector $\vec{L}$ would vanish identically. However, as we shall see shortly, Eq. (3.1) admits transverse as well as longitudinal solutions.

In order to solve the integro-differential equation (3.5) we convert it first into a differential equation. This can easily be done if we note that the kernel of the integral transform in (3.5) satisfies the equation

$$\left(\frac{\partial^2}{\partial z^2}+w_\mu^2\right)\left(\frac{e^{iw_\mu|z-z'|}}{w_\mu}\right)=2\pi i\,\delta(z-z')\ , \qquad (3.7)$$

where δ is the Dirac δ function.

If we apply the operator $[(\partial^2/\partial z^2)+w_\mu^2]$ to both sides of (3.5) and use (3.7) we readily find that $\hat{\vec{E}}$ obeys the equation

$$\frac{\partial^4\hat{\vec{E}}}{\partial z^4}+(k_0^2\epsilon_0-u^2-v^2+w_\mu^2)\frac{\partial^2\hat{\vec{E}}}{\partial z^2}+[w_\mu^2(k_0^2\epsilon_0-u^2-v^2)-\chi k_0^2]\hat{\vec{E}}=\left(\frac{\partial^2}{\partial z^2}+w_\mu^2\right)\vec{L}. \qquad (3.8)$$

Equation (3.8), when separated into Cartesian components, gives a set of three coupled fourth-order linear differential equations for the components $\hat{E}_x$, $\hat{E}_y$, and $\hat{E}_z$ of $\hat{\vec{E}}$, the coupling arising from the presence of the vector $\vec{L}$ on the right-hand side of (3.8). These equations may readily be uncoupled by the use of a procedure well known in the theory of differential equations.[20] For this purpose we rewrite (3.8) in the form

$$\mathcal{L}\left(u,v;\ \frac{\partial}{\partial z}\ ;\ \omega\right)\hat{\vec{E}}(u,v;z;\omega)=0, \qquad (3.9)$$

where $\mathcal{L}$ is a 3×3 matrix operator, with the components

$$\mathcal{L}_{mn}=\left(\frac{\partial^4}{\partial z^4}+(k_0^2\epsilon_0-u^2-v^2+w_\mu^2)\frac{\partial^2}{\partial z^2}+[w_\mu^2(k_0^2\epsilon_0-u^2-v^2)-\chi k_0^2]\right)\delta_{mn}-\left(\frac{\partial^2}{\partial z^2}+w_\mu^2\right)l_m l_n, \qquad (3.10)$$

δ_{mn} being the Kronecker symbol and l_m, etc., are components of the vector

$$\vec{l}\equiv iu,\ iv,\ \frac{\partial}{\partial z}\ . \qquad (3.11)$$

Then the set of the uncoupled equation is

$$\mathfrak{M}\left(u,v;\ \frac{\partial}{\partial z}\ ;\omega\right)\hat{E}_m(u,v;z;\omega)=0\quad(m=x,y,z), \qquad (3.12)$$

where

$$\mathfrak{M}\left(u,v;\ \frac{\partial}{\partial z}\ ;\ \omega\right)=\begin{vmatrix}\mathcal{L}_{xx}, & \mathcal{L}_{xy}, & \mathcal{L}_{xz}\\ \mathcal{L}_{yx}, & \mathcal{L}_{yy}, & \mathcal{L}_{yz}\\ \mathcal{L}_{zx}, & \mathcal{L}_{zy}, & \mathcal{L}_{zz}\end{vmatrix} \qquad (3.13)$$

is the determinant of $\mathcal{L}$. The fact that the elements of $\mathcal{L}_{mn}$ of $\mathcal{L}$ are operators creates no problem, since any two elements of $\mathcal{L}$ commute with each other.

The general solution of (3.12) is

$$\hat{\vec{E}}(u,v;z;\omega)=\sum_j \vec{C}_j(u,v;z;\omega)\,e^{iw_jz}\ , \qquad (3.14)$$

where w_j are the w roots of the equation

$$\mathfrak{M}(u,v,w;\omega)=0 \qquad (3.15)$$

and $\vec{C}_j(u,v;z;\omega)$ is a polynomial in z whose degree depends on the multiplicity (if any) of the root $w=w_j$. If w_j has a multiplicity N, then $\vec{C}_j$ is a polynomial of degree $N-1$. From (3.10) and (3.11) one finds after a straightforward calculation that the equation (3.15) for w has the following more explicit form:

$$\mathfrak{M}_t^2(u,v,w;\omega)\,\mathfrak{M}_l(u,v,w;\omega)=0\ , \qquad (3.16)$$

where

$$\mathfrak{M}_t(u,v,w;\omega)=w^4-w^2(k_0^2\epsilon_0-u^2-v^2+w_\mu^2)+w_\mu^2(k_0^2\epsilon_0-u^2-v^2)-\chi k_0^2, \qquad (3.17)$$

$$\mathfrak{M}_l(u,v,w;\omega)=w^2-w_\mu^2+\frac{\chi}{\epsilon_0}\ . \qquad (3.18)$$

If we assume that the w roots of the equation $\mathfrak{M}_t=0$ and also of the equation $\mathfrak{M}_l=0$ are all distinct and note that each w root of the equation $\mathfrak{M}_t=0$ is a double root of the equation (3.16), it follows that the general solution (3.14) of (3.12) may be expressed in the form

$$\hat{\vec{E}}(u,v;z;\omega)=\sum_{j=1}^{4}\ [\vec{A}_j(u,v;\omega)+z\vec{B}_j(u,v;\omega)]\,e^{iw_jz}+\sum_{j=1}^{2}\vec{A}_j'(u,v;\omega)\,e^{iw_j'z}. \qquad (3.19)$$

Here the w_j and w_j' are the roots of the equations

$$\mathfrak{M}_t(u,v,w_j;\omega)=0, \qquad (3.20a)$$

$$\mathfrak{M}_l(u,v,w_j';\omega)=0 \qquad (3.20b)$$

and $\vec{A}_j$, $\vec{B}_j$, and $\vec{A}_j'$ are arbitrary vector functions of u, v, and ω.

It is clear from our derivation that any solution of the integro-differential equation (3.5) satisfies both the coupled set of differential equations (3.8) and the uncoupled set (3.12), and hence is necessarily expressible in the form (3.19). The converse, however, is not true, since in proceeding from (3.5) to (3.8) and (3.12) we differentiated several times. In order that (3.19) satisfy (3.5), several constraints must be imposed on the vector amplitudes $\vec{A}_j$, $\vec{B}_j$, and $\vec{A}_j'$. We determine these constraints in two stages. First we substitute from (3.19) into (3.8) and find after a long but straightforward calculation outlined in Appendix A that in order that each term on the right-hand side of (3.19) be a solution of (3.8) one must have

$$\vec{k}_j \cdot \vec{A}_j = 0, \tag{3.21}$$

$$\vec{k}_j' \times \vec{A}_j' = 0, \tag{3.22}$$

and that, in general,[21]

$$\vec{B}_j = 0\ , \tag{3.23}$$

where the (generally complex) vectors $\vec{k}_j$ and $\vec{k}_j'$ are defined as

$$\vec{k}_j \equiv u, v, w_j\ , \quad \vec{k}_j' \equiv u, v, w_j'\ . \tag{3.24}$$

Next we substitute from (3.19) into the integro-differential equation (3.5) and also use (3.21)–(3.23). We then obtain, after some calculations carried out in full in Appendix B, the following two additional constraints:

$$\sum_{j=1}^{4} \frac{\vec{A}_j}{w_j - w_\mu} + \sum_{j=1}^{2} \frac{\vec{A}_j'}{w_j' - w_\mu} = 0\ , \tag{3.25}$$

$$\sum_{j=1}^{4} \frac{\vec{A}_j}{w_j + w_\mu}\, e^{i w_j d} + \sum_{j=1}^{2} \frac{\vec{A}_j'}{w_j' + w_\mu}\, e^{i w_j' d} = 0. \tag{3.26}$$

From (3.2), (3.19), and (3.23) it follows that the *general solution* of our integro-differential equation (3.1) may be expressed in the form

$$\vec{E}(\vec{r}, \omega) = \iint_{-\infty}^{+\infty} \vec{\mathcal{E}}(\vec{r}, \omega; u, v)\, du\, dv, \tag{3.27}$$

where

$$\vec{\mathcal{E}}(\vec{r}, \omega; u, v) = \sum_{j=1}^{4} \vec{A}_j(u, v; \omega)\, e^{i\vec{k}_j \cdot \vec{r}} + \sum_{j=1}^{2} \vec{A}_j'(u, v; \omega)\, e^{i\vec{k}_j' \cdot \vec{r}}\ . \tag{3.28}$$

For each pair of the parameters (u, v) $(-\infty < u < \infty, -\infty < v < \infty)$ with ω fixed, the propagation vectors $\vec{k}_j$ and $\vec{k}_j'$ are given by (3.24), with w_j, w_j' being solutions of Eq. (3.20) and with the vector amplitudes $\vec{A}_j$, $\vec{A}_j'$ obeying the constraints (3.21), (3.22), (3.25), and (3.26). The constraint (3.21) implies that each of the four plane waves under the first summation sign in (3.28) is *transverse*. The constraint (3.22) implies that each of the two plane waves under the second summation sign in (3.28) is *longitudinal*. It may be verified that the values $\vec{k}_j^2$ and $\vec{k}_j'^2$ defined by (3.24) and (3.20) are identical with the values $\vec{k}_t^2$ and $\vec{k}_l^2$ given by Eqs. (1.25) and (1.26), respectively, so that each $\vec{k}_j^2$ satisfies the transverse dispersion relation (1.18) and each $\vec{k}_j'^2$ satisfies the longitudinal dispersion relation (1.19) for a plane wave in our spatially dispersive model medium, i.e.,

$$\tilde{\epsilon}(\vec{k}_j\, ; \omega) = \left(\frac{\vec{k}_j}{k_0}\right)^2, \quad \tilde{\epsilon}(\vec{k}_j', \omega) = 0 \quad (k_0 = \omega/c)\ . \tag{3.29}$$

It may be verified that for each pair of values of (u, v) and with ω fixed, $\vec{\mathcal{E}}(\vec{r}, \omega; u, v)$ given by (3.28) satisfies the original integro-differential equation (3.1) so that (3.27) is a true *mode representation* of the electric field in our spatially dispersive slab. It is important to appreciate that *each mode* [each $\vec{\mathcal{E}}(\vec{r}, \omega; u, v)$] *consists of a number of plane waves* (six in general, four of which are transverse and two longitudinal), *coupled by the two relations* (3.25) *and* (3.26). A single transverse or longitudinal plane wave cannot satisfy the integro-differential equation (3.1), since it is impossible to satisfy the constraints (3.25) and (3.26) with only one nonvanishing vector amplitude. We will refer to the relations (3.25) and (3.26) as *mode-coupling conditions* and will discuss them in detail in Sec. V. Here we only mention that these conditions play a crucial role in the understanding of the controversial subject of additional boundary conditions, referred to in the Introduction.

To complete the description of the electromagnetic field in the spatially dispersive slab we must also derive expressions for the electric displacement vector $\vec{D}$, for the magnetic field $\vec{H}$, and for the magnetic induction field $\vec{B}$. These may be obtained readily from Maxwell equations and from our mode expansion (3.27). The Fourier time transforms of the first Maxwell equation in (1.10) and of Eq. (1.11) give, if we also recall that we assumed the medium to be nonmagnetic,

$$\vec{B}(\vec{r}, \omega) = \vec{H}(\vec{r}, \omega) = \frac{1}{ik_0}\ \nabla \times \vec{E}(\vec{r}, \omega), \tag{3.30}$$

$$\vec{D}(\vec{r}, \omega) = \frac{1}{k_0^2}\ \nabla \times [\nabla \times \vec{E}(\vec{r}, \omega)]\ . \tag{3.31}$$

On substituting from (3.27) into (3.30) and (3.31), we obtain the following expressions for the electromagnetic field vectors:

$$\vec{B}(\vec{r}, \omega) = \vec{H}(\vec{r}, \omega) = \iint_{-\infty}^{+\infty} \vec{\mathcal{H}}(\vec{r}, \omega; u, v)\, du\, dv, \tag{3.32}$$

$$\vec{D}(\vec{r}, \omega) = \iint_{-\infty}^{+\infty} \vec{\mathfrak{D}}(\vec{r}, \omega; u, v)\, du\, dv, \tag{3.33}$$

where

$$\vec{\mathcal{H}}(\vec{r}, \omega; u, v) = \sum_{j=1}^{4} \left(\frac{\vec{k}_j}{k_0}\right) \times \vec{A}_j\, e^{i\vec{k}_j \cdot \vec{r}}\ , \tag{3.34}$$

$$\vec{\mathfrak{D}}(\vec{r}, \omega; u, v) = \sum_{j=1}^{4} \left(\frac{\vec{k}_j}{k_0}\right)^2 A_j e^{i\vec{k}_j \cdot \vec{r}}\ . \tag{3.35}$$

We note that the longitudinal plane waves do not contribute to $\vec{B}$, $\vec{H}$, or $\vec{D}$. This fact is consistent with Eqs. (3.30) and (3.31); these equations express the three fields in terms of $\nabla \times \vec{E}$ and for a longitudinal $\vec{E}$ wave $\nabla \times \vec{E} = 0$.

It is straightforward to verify that for each pair of parameters (u, v) the four vector fields $\vec{\mathcal{E}}$, $\vec{\mathfrak{D}}$, $\vec{\mathcal{H}}$, and $\vec{\mathfrak{B}}$ $(= \vec{\mathcal{H}})$ satisfy the four homogeneous Maxwell equations. Thus our solution, expressed by Eqs. (3.27), (3.32), and (3.33) as a superposition of these four vector fields for all possible values of u and v ($-\infty<u<\infty$, $-\infty<v<\infty$), is a true *mode representation of the complete electromagnetic field of frequency* ω, *in our spatially dispersive slab*.

We conclude this section with a few remarks about the nature of the six plane waves that consitute a typical u, v, ω mode. If we set

$$w_j=\alpha_j+i\beta_j, \tag{3.36a}$$

$$w_j'=\alpha_j'+i\beta_j', \tag{3.36b}$$

where α_j, β_j, α_j', β_j' are all real, then each of the four transverse plane waves belonging to the mode has the space dependence

$$e^{i(ux+vy+\alpha_j z)}e^{-\beta_j z} \quad (j=1,2,3,4), \tag{3.37}$$

and each of the two longitudinal modes belonging to the mode has the space dependence

$$e^{i(ux+vy+\alpha_j' z)}e^{-\beta_j' z} \quad (j=1,2). \tag{3.38}$$

Let us now examine more closely the parameters α_j, β_j, α_j', β_j', which determine the nature of these waves. It will be convenient to examine the longitudinal waves first.

We have, according to the "longitudinal" dispersion relation [Eqs. (3.20b) and (3.18)], if we substitute for w_μ^2 from (3.4a) and use the expression given in (1.24) for $\mu^2(\omega)$,

$$w_j'^2=a'+ib' \quad (a',\ b' \text{ real}), \tag{3.39}$$

where

$$a'=\frac{m_e^*}{\hbar\omega_e}(\omega^2-\omega_e^2)-u^2-v^2+\frac{\chi}{\epsilon_0}, \tag{3.40a}$$

$$b'=\frac{m_e^*\omega}{\hbar\omega_e}\Gamma. \tag{3.40b}$$

From (3.39) we see that the two roots w_1' and w_2' are connected by the relation $w_2'=-w_1'$, which, according to Eq. (3.36b), implies that

$$\alpha_2'=-\alpha_1', \quad \beta_2'=-\beta_1'. \tag{3.41}$$

Next, on squaring (3.36b) and comparing it with (3.39), we obtain the relations

$$\alpha_j'^2-\beta_j'^2=a', \quad 2\alpha_j'\beta_j'=b'. \tag{3.42}$$

Now in view of the assumptions that we made about the constants appearing in our expression for the nonlocal dielectric constant, it follows at once from (3.40b) that $b'>0$. The second relation (3.42) then implies that for each of the two values of j, α_j' and β_j' have the same sign; i.e., they are either both positive or both negative. And it is clear from (3.41) that both of these two eventualities occur: i.e., for one of the longitudinal waves $\alpha_j'>0$, $\beta_j'>0$; for the other $\alpha_j'<0$, $\beta_j'<0$. Recalling (3.38) it is clear that the first is a wave that propagates from the face $z=0$ of the slab towards the face $z=d$ and the other is a wave that propagates from the face $z=d$ towards the face $z=0$, and each of the two waves decays exponentially in amplitude as the wave progresses.

Let us now consider the four transverse waves. We have from the "transverse" dispersion relation [Eqs. (3.20a) and (3.17)], if we again substitute from (3.4a) and 1.24),

$$w_j^4-2Pw_j^2+Q=0 \quad (j=1,2,3,4), \tag{3.43}$$

where

$$2P=k_0^2\epsilon_0-2u^2-2v^2+\frac{m_e^*}{\hbar\omega_e}(\omega^2-\omega_e^2+i\omega\Gamma), \tag{3.44a}$$

$$Q=\left(\frac{m_e^*}{\hbar\omega_e}(\omega^2-\omega_e^2+i\omega\Gamma)-u^2-v^2\right)\times(k_0^2\epsilon_0-u^2-v^2)-\chi k_0^2. \tag{3.44b}$$

Since the quartic equation (3.43) involves w_j in second powers only, we may solve it at once for w_j^2 and obtain

$$w_j^2=P\pm(P^2-Q)^{1/2}. \tag{3.45}$$

Here and below we define for the sake of precision the positive square root of a complex number z as that particular square root whose imaginary part is non-negative. Let us label the two roots w_j for which the upper sign is taken in (3.45) by suffixes 1 and 2, and the other two roots, for which the lower sign is taken, by the suffixes 3 and 4. Thus we have

$$w_1^2=w_2^2=P+(P^2-Q)^{1/2}, \tag{3.46a}$$

$$w_3^2=w_4^2=P-(P^2-Q)^{1/2}. \tag{3.46b}$$

It is clear then that the four roots are

$$\begin{aligned} w_1&=+[P+(P^2-Q)^{1/2}]^{1/2},\\ w_2&=-[P+(P^2-Q)^{1/2}]^{1/2},\\ w_3&=+[P-(P^2-Q)^{1/2}]^{1/2},\\ w_4&=-[P-(P^2-Q)^{1/2}]^{1/2}. \end{aligned} \tag{3.47}$$

Since $w_2=-w_1$, $w_4=-w_3$, we have, if we recall Eq. (3.36a), that

$$\begin{aligned} \alpha_2&=-\alpha_1, \quad \beta_2=-\beta_1,\\ \alpha_4&=-\alpha_3, \quad \beta_4=-\beta_3. \end{aligned} \tag{3.48}$$

If now, by analogy with (3.39), we write

$$w_j^2=a_j+ib_j \quad (a_j,\ b_j \text{ real}), \tag{3.49}$$

we clearly have, on squaring (3.36a) and comparing it with (3.49),

$$\alpha_j^2-\beta_j^2=a_j, \quad 2\alpha_j\beta_j=b_j. \tag{3.50}$$

Since b_j is the imaginary part of w_j^2, we have from (3.46) and the second relation (3.50)

$$2\alpha_j\beta_j = \mathrm{Im}[P + (P^2 - Q)^{1/2}] \quad \text{if } j = 1, 2 , \qquad (3.51a)$$
$$= \mathrm{Im}[P - (P^2 - Q)^{1/2}] \quad \text{if } j = 3, 4 . \qquad (3.51b)$$

Now, we see at once from (3.44a) and our earlier assumptions about the constants appearing in that formula that $\mathrm{Im}P > 0$. Moreover, we have defined the positive square root of a complex number as that square root which has non-negative imaginary part. Hence it follows from (3.51a) that

$$\alpha_j\beta_j > 0 \quad (j = 1, 2) . \qquad (3.52a)$$

Further, from Eqs. (3.44) one may deduce by a straightforward but long calculation[22] that one also has $\mathrm{Im}[P - (P^2 - Q)^{1/2}] > 0$ (recalling our earlier assumption that $m_e^* > 0$) and (3.51b) then implies that

$$\alpha_j\beta_j > 0 \quad (j = 3, 4). \qquad (3.52b)$$

The conclusions one can draw from these relations are analogous to those that we reached in connection with the longitudinal waves. Equations (3.52) imply that for each of the four transverse waves α_j and β_j have the same signs; i.e., they are either both positive or both negative. And it is clear from Eqs. (3.48) that for two of the waves (α_j, β_j) are both positive, for the other two waves they are both negative. Hence two of the transverse waves are propagated from the face $z = 0$ of the slab towards the face $z = d$, the other two waves are propagated from the face $z = d$ towards the face $z = 0$, and each of the four transverse waves decays exponentially in amplitude as the wave progresses.

Also, since $w_2 = -w_1$, $w_4 = -w_3$, $w_2' = -w_1'$ there are also obvious geometric relations between the complex wave vectors $\vec{k}_2$ and $\vec{k}_1$, $\vec{k}_4$ and $\vec{k}_3$, and $\vec{k}_2'$ and $\vec{k}_1'$, which one could, of course, expect from symmetry considerations.

The general representation that we obtained in this section for the electromagnetic field in a spatially dispersive model medium forming a plane parallel slab may be used in analyzing a variety of problems of current research interest. In Sec. IV we will employ it to investigate refraction and reflection on a spatially dispersive half-space. The representation has also been used to study surface polaritons in a spatially dispersive medium occupying a half-space.[15,23]

IV. REFLECTION AND REFRACTION ON SPATIALLY DISPERSIVE HALF-SPACE

We will now apply the general theory developed in the preceding sections to the problem of refraction and reflection on a spatially dispersive medium whose dielectric constant in the $(\vec{k}, \omega)$ domain is given by Eq. (2.1). The medium will be assumed to occupy the half-space $z > 0$. As explained in the discussion following Eq. (2.7b), the constitutive relation connecting the electric displacement vector $\vec{D}$ and the electric field vector $\vec{E}$ in the $(\vec{r}, \omega)$ domain will be taken to be given by Eq. (2.8), where the volume V of integration is now the half-space $z > 0$ occupied by the dielectric. We will find that, contrary to statements frequently found in the literature, the problem can be solved entirely within the framework of Maxwell's theory; i.e., it is not necessary to introduce any boundary conditions beyond those implicit in Maxwell's equations.

Suppose that the sources of the incident electromagnetic field (assumed to be monochromatic, of frequency ω) are situated in the domain $z < z_0$, where $z_0 < 0$. Then at each point in the domain $z > z_0$, the electric vector of the incident field (i.e., of the field that would be generated by the sources in the absence of the spatially dispersive medium) may be represented in the form of an angular spectrum of plane waves[19(a)]:

$$\vec{E}_0(\vec{r}, \omega) = \iint_{-\infty}^{+\infty} \vec{A}_0(u, v; \omega)\, e^{i(ux+vy+w_0z)}\, du\, dv, \qquad (4.1)$$

where

$$w_0 = +(k_0^2 - u^2 - v^2)^{1/2} \quad \text{when } u^2 + v^2 \le k_0^2$$
$$= +i(u^2 + v^2 - k_0^2)^{1/2} \quad \text{when } u^2 + v^2 > k_0^2 , \qquad (4.2)$$

where

$$k_0 = \omega/c . \qquad (4.3)$$

We may expand the electric field of the reflected wave, in the domain $z_0 < z < 0$, in a similar way,

$$\vec{E}_r(\vec{r}, \omega) = \iint_{-\infty}^{+\infty} \vec{A}_r(u, v; \omega)\, e^{i(ux+vy-w_0z)}\, du\, dv . \qquad (4.4)$$

If the medium were occupying a slab $0 \le z \le d$, the electric field generated inside the slab would have the mode expansion given by Eqs. (3.27) and (3.28). In the present case, the medium occupies the half-space $0 \le z < \infty$ and it is clear that the appropriate mode expansion in this half-space is still given by Eq. (3.27), provided that in Eq. (3.28) we suppress the three plane waves (two transverse, one longitudinal) with $\vec{k}$ vectors whose z components have negative real part. For these three waves are evidently the waves that are reflected from the rear surface of the slab and must clearly be absent in the limiting case as $d \to \infty$, when the slab degenerates into the half-space. Thus we have the following mode expansion for the electric field inside the half-space:

$$\vec{E}(\vec{r}, \omega) = \iint_{-\infty}^{+\infty} \vec{\mathcal{E}}(\vec{r}, \omega; u, v)\, du\, dv , \qquad (4.5)$$

where

$$\vec{\mathcal{E}}(\vec{r}, \omega; u, v) = \sum_{j=1}^{2} \vec{A}_t^{(j)}(u, v; \omega)\, e^{i\vec{k}_t^{(j)}\cdot\vec{r}} + \vec{A}_l(u, v; \omega)\, e^{i\vec{k}_l\cdot\vec{r}} . \qquad (4.6)$$

In (4.6), the subscript t, l label quantities associated with transverse and longitudinal waves, respectively, so that

$$\vec{k}_t^{(j)} \cdot \vec{A}_t^{(j)} = 0 \quad (j = 1, 2) , \tag{4.7}$$

$$\vec{k}_l \times \vec{A}_l = 0 . \tag{4.8}$$

Here[24]

$$\vec{k}_t^{(j)} \equiv u, v, w_j , \quad \mathrm{Re} w_j > 0 \quad (j = 1, 2) , \tag{4.9}$$

$$\vec{k}_l \equiv u, v, w_l , \quad \mathrm{Re} w_l > 0 , \tag{4.10}$$

and the w_j's and w_l's are the roots of the dispersion relations (3.20) (with $w_j' \to w_l$), i.e., of the equations

$$\mathfrak{M}_t(u, v, w_j ; \omega) = 0, \quad \mathfrak{M}_l(u, v, w_l ; \omega) = 0, \tag{4.11}$$

where $\mathfrak{M}_t$ and $\mathfrak{M}_l$ are given by Eqs. (3.17) and (3.18), respectively.

The vector amplitudes $\vec{A}_t^{(j)}$ and $\vec{A}_l$ in Eq. (4.6) are not independent but are related by the mode-coupling condition (3.25), which now reduces to

$$\sum_{j=1}^{2} \frac{\vec{A}_t^{(j)}}{w_j - w_\mu} + \frac{\vec{A}_l}{w_l - w_\mu} = 0 . \tag{4.12}$$

The angular spectrum representation for the magnetic field in the half-space $z > 0$ is obtained at once from Eqs. (3.32) and (3.34) as

$$\vec{H}(\vec{r}, \omega) = \iint_{-\infty}^{+\infty} \vec{\mathcal{H}}(\vec{r}, \omega; u, v) \, du \, dv, \tag{4.13}$$

where

$$\vec{\mathcal{H}}(\vec{r}, \omega; u, v) = \sum_{j=1}^{2} \left(\frac{\vec{k}_t^{(j)}}{k_0} \right) \times \vec{A}_t^{(j)}(u, v; \omega) e^{i \vec{k}_t^{(j)} \cdot \vec{r}} . \tag{4.14}$$

Our main aim is to determine the amplitude vectors $\vec{A}_t^{(j)}$ $(j = 1, 2)$, $\vec{A}_l$, and $\vec{A}_r$ in terms of $\vec{A}_0$. For this purpose we make use of the usual boundary conditions of Maxwell's electromagnetic theory, according to which the tangential components (i.e., the components parallel to the boundary surface) of the electric and the magnetic fields are continuous across the surface bounding the medium. In the present case these conditions must be satisfied at every point (x, y) of the plane $z = 0$, i.e.,

$$\iint_{-\infty}^{+\infty} du \, dv \, e^{i(ux+vy)} \, \hat{z} \times \left(\vec{A}_0 + \vec{A}_r - \sum_{j=1}^{2} \vec{A}_t^{(j)} - \vec{A}_l \right) = 0 \tag{4.15}$$

and

$$\iint_{-\infty}^{+\infty} du \, dv \, e^{i(ux+vy)} \, \hat{z} \times \left(\vec{k}_0 \times \vec{A}_0 + \vec{k}_r \times \vec{A}_r - \sum_{j=1}^{2} \vec{k}_t^{(j)} \times \vec{A}_t^{(j)} \right) = 0 . \tag{4.16}$$

In Eqs. (4.15) and (4.16) $\hat{z}$ is the unit vector in the $+z$ direction and $\vec{k}_0$ and $\vec{k}_r$ are the wave vectors of the plane waves in the angular spectrum representations (4.1) and (4.4) of the incident and the reflected fields, respectively, i.e., the vectors

$$\vec{k}_0 \equiv u, v, w_0 , \quad \vec{k}_r \equiv u, v, -w_0 . \tag{4.17}$$

By taking the Fourier inverse of Eqs. (4.15) and (4.16) we see that *for each* $(u, v; \omega)$ *mode* the following relations must hold:

$$\hat{z} \times \left(\vec{A}_0 + \vec{A}_r - \sum_{j=1}^{2} \vec{A}_t^{(j)} - \vec{A}_l \right) = 0 , \tag{4.18}$$

$$\hat{z} \times \left(\vec{k}_0 \times \vec{A}_0 + \vec{k}_r \times \vec{A}_r - \sum_{j=1}^{2} \vec{k}_t^{(j)} \times \vec{A}_t^{(j)} \right) = 0. \tag{4.19}$$

Equations (4.18) and (4.19) imply that *each plane wave mode of the incident field couples to only one plane wave mode of the reflected field and to one mode of the transmitted field*, the latter consisting of two transverse and one longitudinal plane waves. All these modes (i.e. the incident, the reflected, and the transmitted mode) are labeled by the same mode parameters, namely, u, v, ω. From the linearity of the angular spectral representations (4.1), (4.4), and (4.5) of the three fields, it is clear that we may from now on restrict our analysis to a single mode; i.e., we take the incident field to consist of a single plane wave, with wave vector $\vec{k}_0$ (see Fig. 1).

We see from Eqs. (4.17), (4.9), and (4.10) that all the five wave vectors have the same x components, namely, u, and the same y components, namely, v. Hence it follows that

$$\vec{k}_l \times \hat{z} = \vec{k}_t^{(j)} \times \hat{z} = \vec{k}_0 \times \hat{z} \quad (j = 1, 2), \tag{4.20}$$

$$\vec{k}_r \times \hat{z} = \vec{k}_0 \times \hat{z} . \tag{4.21}$$

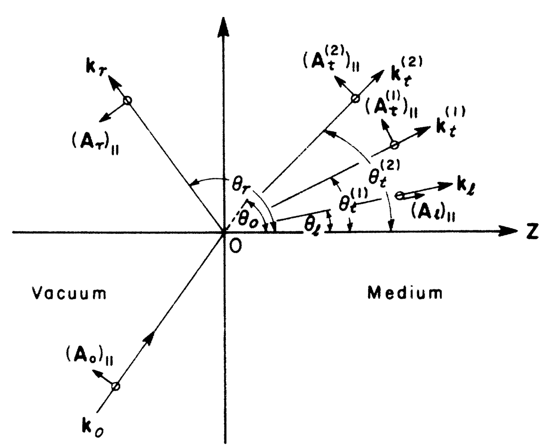

FIG. 1. Illustrating notation relating to refraction and reflection of a plane wave incident from vacuum on a spatially dispersive dielectric medium occupying the half-space $z > 0$. The wave vectors $\vec{k}_t^{(1)}$, $\vec{k}_t^{(2)}$, and $\vec{k}_l$ and also the angles $\theta_t^{(1)}$, $\theta_t^{(2)}$, and θ_l are, in general, complex, but for the purpose of illustration all these quantities are indicated as real. The figure represents the plane of incidence.

The consequences of these relations can be expressed in the form that is a natural generalization of the results well known in connection with spatially nondispersive media, by introducing complex angles of refraction. Let $\vec{k} = u, v, w$ be any one of the five wave vectors $\vec{k}_0$, $\vec{k}_r$, $\vec{k}_t^{(1)}$, $\vec{k}_t^{(2)}$, or $\vec{k}_l$ and let us introduce spherical polar coordinates k, θ, φ, with the polar axis in the direction of the z axis, via the relations

$$u = k \sin\theta \cos\varphi, \tag{4.22a}$$

$$v = k \sin\theta \sin\varphi, \tag{4.22b}$$

$$w = k \cos\theta\ . \tag{4.22c}$$

For all the five wave vectors the components u and v are real. For the three wave vectors $\vec{k}_t^{(1)}$, $\vec{k}_t^{(2)}$, and $\vec{k}_l$ the third component, w, is in general complex. From Eqs. (4.22) we clearly have

$$k = (u^2 + v^2 + w^2)^{1/2}\ . \tag{4.23a}$$

We choose that branch of the square root on the right-hand side of Eq. (4.23a) for which

$$\text{Im}k \geq 0\ . \tag{4.23b}$$

From Eqs. (4.22a) and (4.22b) it follows that $u^2 + v^2 = k^2 \sin^2\theta$ and, since u and v are real, so is $k^2 \sin^2\theta$. Of the two roots of this equation we choose the positive square root as defining $\sin\theta$, i.e.,

$$\sin\theta = \frac{+(u^2+v^2)^{1/2}}{k}\ . \tag{4.24}$$

In view of (4.23b), $\text{Im}\sin\theta \leq 0$. The cosine of the (generally complex) angle θ is given by Eq. (4.22c). Finally, from Eqs. (4.22a) and (4.22b) it follows, if Eq. (4.24) is also used, that the azimuthal angle φ is given by

$$\cos\varphi = \frac{u}{+(u^2+v^2)^{1/2}}\ , \quad \sin\varphi = \frac{v}{+(u^2+v^2)^{1/2}}\ , \tag{4.25}$$

and these two relations give a unique real azimuthal angle φ, defined in the range $0 \leq \varphi < 2\pi$.

If $\hat{x}$ and $\hat{y}$ denote unit vectors in the positive x and y directions, we have $|\vec{k}\times\hat{z}| = |v\hat{x} - u\hat{y}| = +(u^2+v^2)^{1/2}$ or, using (4.24),

$$|\vec{k}\times\hat{z}| = k \sin\theta\ . \tag{4.26}$$

Hence it follows from (4.20) that

$$k_l \sin\theta_l = k_t^{(j)} \sin\theta_j = k_0 \sin\theta_0, \tag{4.27}$$

where, or course, the subscripts and superscripts on the various symbols again label quantities associated with the appropriate waves. Further, we also have from (4.21), (4.17), and (4.22c) that $\cos\theta_r = -\cos\theta_i$, so that

$$\theta_r = \pi - \theta_0\ . \tag{4.28}$$

Let us now define complex refractive indices n_l and $n_t^{(j)}$ ($j = 1, 2$) for the longitudinal and transverse plane waves forming the $(u, v; \omega)$ mode in the spatially dispersive medium, by the formulas

$$n_l = \frac{k_l}{k_0}\ , \quad n_t^{(j)} = \frac{k_t^{(j)}}{k_0} \quad (j = 1, 2)\ . \tag{4.29}$$

In view of the inequality (4.23b) we have

$$\text{Im}n_l \geq 0, \quad \text{Im}n_t^{(j)} \geq 0\ . \tag{4.29a}$$

If we use Eqs. (1.25) and (1.26) we may express the three refractive indices in terms of the constants characterizing the macroscopic properties of the spatially dispersive medium. We then find that

$$n_t^{(j)} = \left\{\frac{1}{2}\left(\epsilon_0 + \frac{\mu^2}{k_0^2}\right) \pm \left[\frac{\chi}{k_0^2} + \frac{1}{4}\left(\epsilon_0 - \frac{\mu^2}{k_0^2}\right)^2\right]^{1/2}\right\}^{1/2} \quad (j = 1, 2), \tag{4.30}$$

$$n_l = \left(\frac{\mu^2}{k_0^2} - \frac{\chi}{\epsilon_0 k_0^2}\right)^{1/2}\ . \tag{4.31}$$

The frequency dependence of the three refractive indices near the exciton transition frequency ω_e is illustrated for a typical case in Fig. 2.

Using Eqs. (4.29), the relation (4.27) may be expressed in the form

$$n_l \sin\theta_l = n_t^{(j)} \sin\theta_t^{(j)} = \sin\theta_0 \quad (j = 1, 2)\ . \tag{4.32}$$

Moreover, on taking the scalar product of each term in Eqs. (4.20) and (4.21) with $\vec{k}_0$ and on using the well-known invariance property of a scalar triple product under cyclic permutation of the three vectors, it follows that

$$(\vec{k}_0 \times \hat{z}) \cdot \vec{k}_l = (\vec{k}_0 \times \hat{z}) \cdot \vec{k}_t^{(j)} = 0 \quad (j = 1, 2) \tag{4.33}$$

and

$$(\vec{k}_0 \times \hat{z}) \cdot \vec{k}_r = 0. \tag{4.34}$$

Now $\vec{k}_0 \times \hat{z}$ is a vector perpendicular to the plane containing the wave vector $\vec{k}_0$ of the incident wave and the normal to the boundary $z = 0$ of the medium; i.e., it is a vector perpendicular to the plane of incidence. Hence Eqs. (4.33) and (4.34) imply that *each of the four wave vectors* $\vec{k}_l$, $\vec{k}_t^{(1)}$, $\vec{k}_t^{(2)}$, and $\vec{k}_r$ *lies in the plane of incidence*. The relations (4.32), together with the fact that the propagation vectors $\vec{k}_l$, $\vec{k}_t^{(1)}$, and $\vec{k}_t^{(2)}$ lie in the plane of incidence, show that each of the three refracted plane waves generated inside the medium obeys the usual *Snell's law of refraction*. The relation (4.28), together with the fact that the wave vector of the reflected wave lies in the plane of incidence, shows that the reflected wave obeys the usual *law of reflection*.

Next we will obtain expressions for the vector amplitudes $\vec{A}_l$, $\vec{A}_t^{(1)}$, and $\vec{A}_t^{(2)}$ of the transmitted (refracted) plane waves and for the vector amplitude $\vec{A}_r$ of the reflected wave. For this purpose we rewrite the mode-coupling condition (4.12) in the form[25]

$$\sum_{j=1}^{2} \alpha_t^{(j)} \vec{A}_t^{(j)} + \alpha_l \vec{A}_l = 0, \tag{4.35}$$

where

$$\alpha_t^{(j)} = \frac{n_t^{(j)} \cos\theta_t^{(j)} + [(\mu^2/k_0^2) - \sin^2\theta_0]^{1/2}}{(n_t^{(j)})^2 - \mu^2/k_0^2}, \tag{4.36a}$$

$$\alpha_l = \frac{n_l \cos\theta_l + [(\mu^2/k_0^2) - \sin^2\theta_0]^{1/2}}{n_l^2 - \mu^2/k_0^2}. \tag{4.36b}$$

In writing down the expressions (4.36) we made use of Eqs. (4.22c), (4.24), and (4.29). As in usual treatments of refraction and reflection on a spatially nondispersive medium, it is convenient to consider separately the case when the electric field of the incident wave is linearly polarized either in the plane of incidence or at right angles to it. It will be seen that in the former case the electric field of the reflected and the transmitted waves will also be linearly polarized in the plane of incidence; in the latter case it will be linearly polarized at right angles to this plane. This result implies that the case when the incident wave has any other state of polarization may be treated by decomposing its electric field into these two states of polarization and applying our results separately to each of the two "partial" waves. From now on the components of the amplitude vectors that are parallel to the plane of incidence, will be denoted by suffix ∥ and those perpendicular to it by suffix ⊥. The choice of the positive directions for the parallel components is indicated in Fig. 1. The perpendicular components are at right angles to the plane of the figure.

A. Incident wave linearly polarized at right angle to plane of incidence $[(A_0)_\parallel = 0]$

In this case we have $(A_0)_\parallel = 0$ and we find from the relations (4.18) and (4.19) that

$$(\vec{A}_0)_\perp + (\vec{A}_r)_\perp = (\vec{A}_t^{(1)})_\perp + (\vec{A}_t^{(2)})_\perp, \tag{4.37}$$

and

$$[(\vec{A}_0)_\perp - (\vec{A}_r)_\perp] \cos\theta_0 = (\vec{A}_t^{(1)})_\perp n_t^{(1)} \cos\theta_t^{(1)} + (\vec{A}_t^{(2)})_\perp n_t^{(2)} \cos\theta_t^{(2)}. \tag{4.38}$$

Further, according to Eq. (4.8), $\vec{A}_l$ is parallel to $\vec{k}_l$. Now, we have already seen that $\vec{k}_l$ lies in the plane of incidence, and hence $\vec{A}_l$ also lies in the plane of incidence, so that

$$(\vec{A}_l)_\perp = 0. \tag{4.39}$$

Next, we take the perpendicular component of the mode-coupling condition (4.35) and use (4.39). This gives

$$\alpha_t^{(1)} (\vec{A}_t^{(1)})_\perp + \alpha_t^{(2)} (\vec{A}_t^{(2)})_\perp = 0. \tag{4.40}$$

On solving Eqs. (4.37), (4.38), and (4.40) for the perpendicular components of the three unknown vector amplitudes,[26] we obtain the results

$$\frac{(\vec{A}_t^{(j)})_\perp}{(\vec{A}_0)_\perp} = 2\cos\theta_0 \left((n_t^{(j)} \cos\theta_t^{(j)} + \cos\theta_0) - \frac{\alpha_t^{(j)}}{\alpha_t^{(3-j)}} (n_t^{(3-j)} \cos\theta_t^{(3-j)} + \cos\theta_0) \right)^{-1}, \tag{4.41}$$

$$\frac{(\vec{A}_r)_\perp}{(\vec{A}_0)_\perp} = \frac{\alpha_t^{(1)}(\cos\theta_0 - n_t^{(2)}\cos\theta_t^{(2)}) - \alpha_t^{(2)}(\cos\theta_0 - n_t^{(1)}\cos\theta_t^{(1)})}{\alpha_t^{(1)}(\cos\theta_0 + n_t^{(2)}\cos\theta_t^{(2)}) - \alpha_t^{(2)}(\cos\theta_0 + n_t^{(1)}\cos\theta_t^{(1)})}. \tag{4.42}$$

In addition, we also obtain for this case $[(\vec{A}_0)_\parallel = 0]$, from relations (4.18), (4.19), and (4.35) the following expressions for the parallel components:

$$(\vec{A}_t^{(1)})_\parallel = (\vec{A}_t^{(2)})_\parallel = (\vec{A}_l)_\parallel = (\vec{A}_r)_\parallel = 0, \tag{4.43}$$

as expected.

We note that according to (4.39) and one of the relations in (4.43) the vector amplitude of the longitudinal wave $\vec{A}_l = 0$, implying that *when the incident wave is polarized at right angles to the plane of incidence, no longitudinal wave is excited in the spatially dispersive medium.*

B. Incident wave linearly polarized in the plane of incidence $[(\vec{A}_0)_\perp = 0]$

We now have $(\vec{A}_0)_\perp = 0$ and from the relations (4.18) and (4.19) we find that

$$[(\vec{A}_0)_\parallel - (\vec{A}_r)_\parallel] \cos\theta_0 = (\vec{A}_t^{(1)})_\parallel \cos\theta_t^{(1)} + (\vec{A}_t^{(2)})_\parallel \cos\theta_t^{(2)} - (\vec{A}_l)_\parallel \sin\theta_l, \tag{4.44}$$

$$(\vec{A}_0)_\parallel + (\vec{A}_r)_\parallel = n_t^{(1)} (\vec{A}_t^{(1)})_\parallel + n_t^{(2)} (\vec{A}_t^{(2)})_\parallel. \tag{4.45}$$

The parallel component of the mode-coupling condition (4.35) gives

$$\alpha_t^{(1)} (\vec{A}_t^{(1)})_\parallel \cos\theta_t^{(1)} + \alpha_t^{(2)} (\vec{A}_t^{(2)})_\parallel \cos\theta_t^{(2)} - \alpha_l (\vec{A}_l)_\parallel \sin\theta_l = 0, \tag{4.46a}$$

$$\alpha_t^{(1)} (\vec{A}_t^{(1)})_\parallel \sin\theta_t^{(1)} + \alpha_t^{(2)} (\vec{A}_t^{(2)})_\parallel \sin\theta_t^{(2)} + \alpha_l (\vec{A}_l)_\parallel \cos\theta_l = 0. \tag{4.46b}$$

On solving the preceding four equations for the parallel components of the four unknown vector amplitudes we find that[26]

$$\frac{(\vec{A}_t^{(j)})_\parallel}{(\vec{A}_0)_\parallel} = 2\cos\theta_0\left(n_t^{(j)}\cos\theta_0+\cos\theta_t^{(j)} - \frac{\alpha_t^{(j)}}{\alpha_t^{(3-j)}}\,\frac{\cos(\theta_l-\theta_t^{(3-j)})}{\cos(\theta_l-\theta_t^{(3-j)})}\,(n_t^{(3-j)}\cos\theta_0+\cos\theta_t^{(3-j)}) + \frac{\alpha_t^{(j)}}{\alpha_l}\,\frac{\sin\theta_l}{\cos(\theta_l-\theta_t^{(3-j)})}\sin(\theta_t^{(j)}-\theta_t^{(3-j)})\right)^{-1}, \quad (4.47)$$

$$\frac{(\vec{A}_l)_\parallel}{(\vec{A}_0)_\parallel} = -2\cos\theta_0\left(\sin\theta_l + \frac{\alpha_l}{\alpha_t^{(2)}}\,\frac{\cos(\theta_l-\theta_t^{(1)})}{\sin(\theta_t^{(2)}-\theta_t^{(1)})}\,(n_t^{(2)}\cos\theta_0+\cos\theta_t^{(2)}) + \frac{\alpha_l}{\alpha_t^{(1)}}\,\frac{\cos(\theta_l-\theta_t^{(2)})}{\sin(\theta_t^{(1)}-\theta_t^{(2)})}\,(n_t^{(1)}\cos\theta_0+\cos\theta_t^{(1)})\right)^{-1}, \quad (4.48)$$

$$\frac{(\vec{A}_r)_\parallel}{(\vec{A}_0)_\parallel} = \frac{\alpha_t^{(1)}(n_t^{(2)}\cos\theta_0-\cos\theta_t^{(1)})\cos(\theta_l-\theta_t^{(1)}) - \alpha_t^{(2)}(n_t^{(1)}\cos\theta_0-\cos\theta_t^{(1)})\cos(\theta_l-\theta_t^{(2)}) + L}{\alpha_t^{(1)}(n_t^{(2)}\cos\theta_0+\cos\theta_t^{(1)})\cos(\theta_l-\theta_t^{(1)}) - \alpha_t^{(2)}(n_t^{(1)}\cos\theta_0+\cos\theta_t^{(1)})\cos(\theta_l-\theta_t^{(2)}) + L}, \quad (4.49)$$

where

$$L = \left(\frac{\alpha_t^{(1)}\alpha_t^{(2)}}{\alpha_l}\right)\frac{\sin(\theta_t^{(1)}-\theta_t^{(2)})\sin\theta_l}{\cos\theta_0} \quad (4.50)$$

In addition, we also obtain for this case $[(\vec{A}_0)_\perp = 0]$, from the relations (4.18), (4.19), and (4.35), the results

$$(\vec{A}_t^{(1)})_\perp = (\vec{A}_t^{(2)})_\perp = (\vec{A}_l)_\perp = (\vec{A}_r)_\perp = 0 . \quad (4.51)$$

The formulas (4.41), (4.42), and (4.39) and the formulas (4.47)–(4.49) are[27] *generalizations to a spatially dispersive medium* [characterized by a dielectric response function of the form (2.1)] of the well-known *Fresnel formulas for refraction and reflection.*[28] We will now examine some of the consequences of these formulas.

C. Normal incidence ($\theta_0 = 0$)

Let us consider the special case when a linearly polarized wave is incident *normally* on the half-

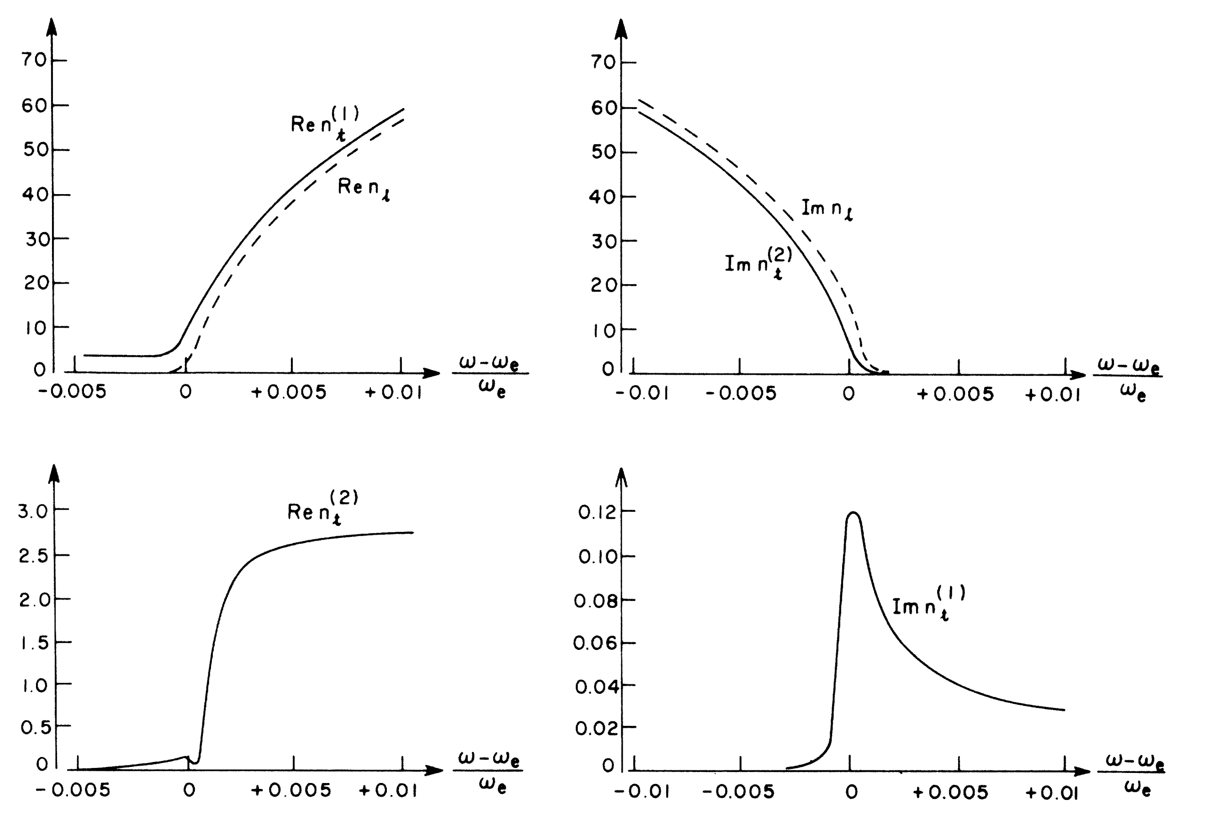

FIG. 2. Real and imaginary parts of the refractive indices $n_t^{(1)}$, $n_t^{(2)}$, and n_l as functions of the ratio $(\omega-\omega_e)/\omega_e$ near the exciton transition frequency $\omega=\omega_e$ for a spatially dispersive model medium of the kind considered throughout the main part of this paper. $\hbar\omega_e = 2.553$ eV ($\omega_e = 3.875\times 10^{15}$ sec^{-1}); $\hbar\Gamma = 5\times 10^{-5}$ eV, $4\pi\alpha = 0.0125$, $m_e^* = 0.9 m_e$ ($m_e = 9.109\times 10^{-31}$ kg is the electron mass) $\epsilon_0 = 8$.

space. Without loss of generality we may choose the x direction to coincide with the direction of polarization of the incident wave. Now, in the degenerate case of normal incidence the concept of the plane of incidence loses its meaning. However, we may readily derive the appropriate formulas by taking the limit $\theta_0 \to 0$ in the expressions for the general case. It will be convenient to proceed to the limit of the formulas for the case when the incident wave is polarized at right angles to the plane of incidence.

In the limit as $\theta_0 \to 0$, we have, from Eqs. (4.32) and (4.28), assuming that $n_t^{(j)}$ and n_l are both different from zero,

$$\theta_t^{(j)} = 0, \quad \theta_l = 0, \quad \theta_r = \pi, \tag{4.52}$$

and Eqs. (4.36a) and (4.36b) reduce to

$$\alpha_t^{(j)} = \frac{1}{n_t^{(j)} - \mu/k_0}, \tag{4.53a}$$

$$\alpha_l = \frac{1}{n_l - \mu/k_0}. \tag{4.53b}$$

The formulas (4.39) and (4.41)–(4.43) for the amplitude components may then readily be shown to be expressible in the form (if we recall that we have chosen the direction of polarization of the incident wave as the x direction)

$$\frac{(\vec{A}_t^{(j)})_x}{(\vec{A}_0)_x} = 2\left(n_t^{(j)} - \frac{\mu}{k_0}\right)(n_t^{(j)} - n_t^{(3-j)})(n^* + 1)^{-1}, \tag{4.54}$$

$$(\vec{A}_l)_x = 0, \tag{4.55}$$

$$\frac{(\vec{A}_r)_x}{(\vec{A}_0)_x} = (1 - n^*)(1 + n^*)^{-1}, \tag{4.56}$$

and

$$(\vec{A}_t^{(j)})_y = (\vec{A}_l)_y = (\vec{A}_r)_y = 0, \tag{4.57}$$

where

$$n^* = n_t^{(1)} + n_t^{(2)} - \frac{\mu}{k_0}. \tag{4.58}$$

Equations (4.55) and (4.57) show that, with normal incidence, no longitudinal wave is excited in the medium. Equations (4.56) and (4.57) imply that the reflection coefficient R for normal incidence is

$$R \equiv \left|\frac{(\vec{A}_r)_x}{(\vec{A}_0)_x}\right|^2 = \left|\frac{1 - n^*}{1 + n^*}\right|^2. \tag{4.59}$$

Equation (4.59) is formally identical with the formula for the reflection coefficient at normal incidence from an ordinary (i.e., spatially nondispersive) medium of refractive index n^*. Thus in the present problem we may regard n^*, defined by Eq. (4.58) together with Eqs. (4.30), as playing the role of an "effective" refractive index.

D. Numerical results

We will now present a number of curves, computed from the above formulas, which show the behavior of the reflected and refracted waves. All the curves refer to a model medium whose parameters have the values given in the caption[29] to Fig. 2.

Figure 3 shows the behavior of the amplitude ratios[30] $|(\vec{A}_r)_\parallel/(\vec{A}_0)_\parallel|$ and $|(\vec{A}_r)_\perp/(\vec{A}_0)_\perp|$ as functions of the angle of incidence θ_0, for the special case when the frequency of the incident wave is equal to the exciton transition frequency ω_e. Figure 4 shows the behavior of the corresponding ratios for the transmitted waves.

Figure 5 shows the behavior of the reflection coefficient $R_\perp = |(\vec{A}_r)_\perp/(\vec{A}_0)_\perp|^2$ as a function of the angle of incidence θ_0 at different frequencies ω, in the neighborhood of the exciton transition frequency ω_e, and Fig. 6 shows the behavior of $R_\perp$ as a function of the frequency ω for several values of the angle of incidence θ_0. In Figs. 7 and 8 the corresponding curves for the reflection coefficient $R_\parallel = |(\vec{A}_r)_\parallel/(\vec{A}_0)_\parallel|^2$ are given. The curves labeled by $\Delta\omega = 3.13 \times 10^{12}\ \text{sec}^{-1}$ in Figs. 5(b) and 7(b), which exhibit somewhat anomalous behavior, correspond to the longitudinal frequency[31] $\omega = \omega_l \simeq 3.87902 \times 10^{15}\ \text{sec}^{-1}$. The pronounced minima in Figs. 6 and 8 are seen to appear very close to this frequency also. The sets of curves representing $R_\perp$ and $R_\parallel$ are seen to differ considerably from each other. The difference is, of course, to some extent connected with the fact, established earlier in this section, that when the incident field is linearly polarized parallel to the plane of incidence, a longitudinal wave is generated in the medium (except at normal incidence), whereas no such wave is generated when it is linearly polarized at right angles to this plane. In particular, it should be noted that most of the

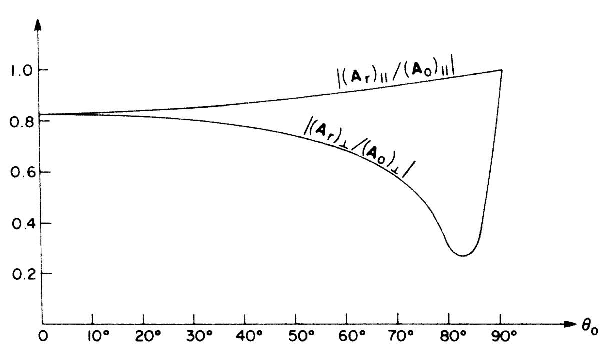

FIG. 3. Normalized amplitude of the reflected wave as a function of the angle of incidence θ_0, when the frequency ω of the incident wave coincides with the exciton transition frequency ω_e. (Calculated with the same values of the parameters as given in the caption to Fig. 2.)

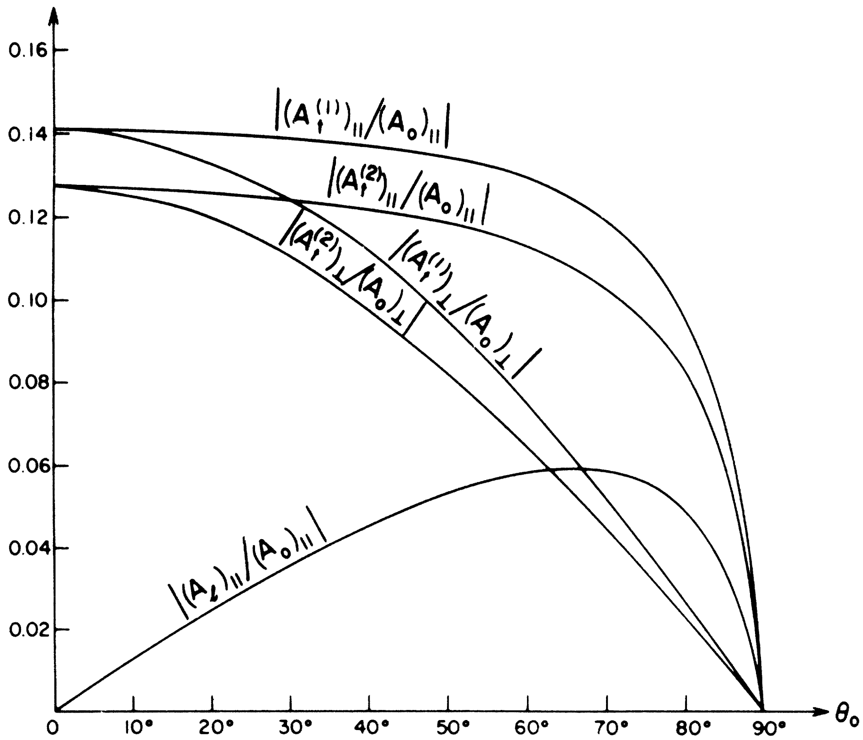

FIG. 4. Normalized amplitude of the transmitted waves as a function of the angle of incidence θ_0, when the frequency ω of the incident wave coincides with the exciton transition frequency ω_e. (Calculated with the same values of the parameters as given in the caption to Fig. 2.)

curves in Fig. 8, for $R_\parallel$ as functions of frequency, exhibit subsidary maxima and minima near the longitudinal frequency ω_l.

In Fig. 9 the behavior of the reflection coefficients $R_\parallel$ and $R_\perp$, as functions of the angle of incidence θ_0, is shown for an electromagnetic wave at the longitudinal frequency ω_l.

We conclude this section with a brief comparison of some of our results with those of Pekar.[1(b)] Although Pekar also derived the laws of refraction [Eqs. (4.27)] his expressions for the amplitudes of the transmitted and reflected waves differ from ours. In Pekar's theory the nonlocal polarization is assumed to obey the boundary condition

$$\vec{P}_{NL}(\vec{r}, \omega)|_{z=0} = 0, \tag{4.60}$$

which, in our terminology, implies the "mode-coupling condition"[32]

$$\sum_{j=1}^{2} \beta_t^{(j)} \vec{A}_t^{(j)} + \beta_l \vec{A}_l = 0, \tag{4.61}$$

with

$$\beta_t^{(j)} = [(n_t^{(j)})^2 - \mu^2/k_0^2]^{-1}, \tag{4.62a}$$

$$\beta_l = [n_l^2 - \mu^2/k_0^2]^{-1}. \tag{4.62b}$$

The condition (4.61) replaces, in the theory of Pekar, our mode-coupling condition (4.35) and his formulas for the amplitudes of the reflected and the transmitted waves follow from our Eqs. (4.39), (4.41), and (4.42) and from Eqs. (4.47)–(4.50) simply by letting

$$\alpha_t^{(j)} \to \beta_t^{(j)}, \quad \alpha_l \to \beta_l. \tag{4.63}$$

In the special case of normal incidence, the reflectivity R_p, calculated from Pekar's theory, is given by the formula

$$R_p = \left| \frac{1 - n_p^*}{1 + n_p^*} \right|^2, \tag{4.64}$$

where

$$n_p^* = (n_t^{(1)} n_t^{(2)} + \epsilon_0)(n_t^{(1)} + n_t^{(2)})^{-1}. \tag{4.65}$$

We see that the expression (4.64) for reflectivity at normal incidence, calculated from Pekar's theory,[33] is of the same form as that calculated from our theory [Eq. (4.59)], but the "effective refractive indices" n^* are different in the two cases, being given by Eq. (4.65) in Pekar's theory and by Eq. (4.58) in ours.[34]

In Fig. 10 we present reflectivity curves at resonance, (i.e., for $\omega = \omega_e$), calculated for a typical case from the two theories. It is seen that although the two theories predict the same general behavior of the curves, there are appreciable differences, of the order of 25%, in the predicted reflectivity values at small angles of incidence.

V. MODE-COUPLING CONDITIONS, ADDITIONAL BOUNDARY CONDITIONS, EXTINCTION THEOREM, AND GENERALIZATIONS

In Sec. III we obtained certain mode-coupling conditions [Eqs. (3.25) and (3.26)], which relate

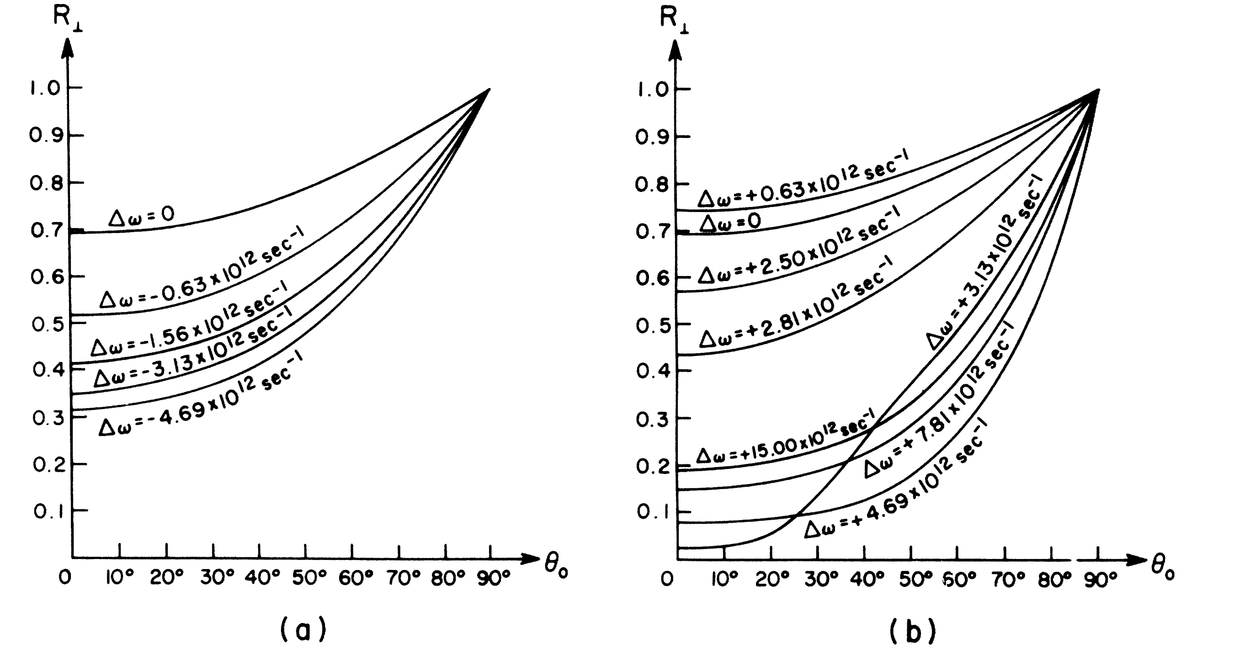

FIG. 5. Reflection coefficient $R_\perp$ as a function of the angle of incidence θ_0 at selected frequencies below [(a)] and above [(b)] the exciton transition frequency ω_e. The curves are labeled by the frequency difference $\Delta\omega = \omega - \omega_e$. (Calculated with the same values of the parameters as given in the caption to Fig. 2.)

all the plane waves that form a particular mode of the electromagnetic field inside the spatially dispersive slab. In Sec. IV we used these conditions to derive the solution of the problem of refraction and reflection on a spatially dispersive half-space. As mentioned in the Introduction this problem has in the past been generally treated with the help of so-called "additional boundary conditions" (abc's), suggested by various physical models relating to the behavior of excitons in the medium. The abc's are usually expressed in terms of the exciton part of the polarization which, in our terminology, is the nonlocal polarization [cf. Eqs. (2.7b) and (2.5)]:

$$\vec{P}_{NL}(\vec{r}, \omega) = (1/4\pi)[\vec{D}(\vec{r}, \omega) - \epsilon_0 \vec{E}(\vec{r}, \omega)]. \qquad (5.1)$$

We will now show that our mode-coupling conditions imply a precise form of the boundary conditions on the nonlocal polarization.

We express the $\vec{E}$ and $\vec{D}$ vectors in (5.1) in terms of their respective mode expansions (3.27), (3.28), and (3.33), (3.35) and obtain the following mode expansion for $\vec{P}_{NL}$ valid at each point $\vec{r}$ inside the slab:

$$\vec{P}_{NL}(\vec{r}, \omega) = \int\int_{-\infty}^{\infty} \vec{\mathcal{P}}_{NL}(\vec{r}, \omega; u, v)\, du\, dv, \qquad (5.2)$$

where

$$\vec{\mathcal{P}}_{NL}(\vec{r}, \omega; u, v) = \frac{1}{4\pi}\sum_{j=1}^{4}\left[\left(\frac{\vec{k}_j}{k_0}\right)^2 - \epsilon_0\right]\vec{A}_j e^{i\vec{k}_j\cdot\vec{r}} - \frac{\epsilon_0}{4\pi}\sum_{j=1}^{2}\vec{A}_j' e^{i\vec{k}_j'\cdot\vec{r}}. \qquad (5.3)$$

Now, we readily find from the dispersion relations (1.26) and (1.25), if we recall that $\vec{k}_j$ and $\vec{k}_j'$ correspond to $\vec{k}_t$ and $\vec{k}_l$, respectively,

$$-\epsilon_0 = \frac{\chi}{\vec{k}_j'^2 - \mu^2}, \qquad (5.4)$$

$$\left(\frac{\vec{k}_j}{k_0}\right)^2 - \epsilon_0 = \frac{\chi}{\vec{k}_j^2 - \mu^2}. \qquad (5.5)$$

Now, according to (3.24) and (3.4)

$$\vec{k}_j^2 = w_j^2 - w_\mu^2 + \mu^2, \quad \vec{k}_j'^2 = w_j'^2 - w_\mu^2 + \mu^2, \qquad (5.6)$$

so that Eqs. (5.4) and (5.5) may be written as

$$-\epsilon_0 = \frac{\chi}{w_j'^2 - w_\mu^2}, \qquad (5.7)$$

$$\left(\frac{\vec{k}_j}{k_0}\right)^2 - \epsilon_0 = \frac{\chi}{w_j^2 - w_\mu^2}. \qquad (5.8)$$

Using (5.7) and (5.8), the (u, v) mode (5.3) of the nonlocal polarization at frequency ω may be represented in the form

$$\vec{\mathcal{P}}_{NL}(\vec{r}, \omega; u, v) = \frac{\chi}{4\pi}\sum_{j=1}^{4}\frac{1}{w_j^2 - w_\mu^2}\vec{A}_j e^{i\vec{k}_j\cdot\vec{r}} + \frac{\chi}{4\pi}\sum_{j=1}^{2}\frac{1}{w_j'^2 - w_\mu^2}\vec{A}_j' e^{i\vec{k}_j'\cdot\vec{r}}. \qquad (5.9)$$

Now the vector amplitudes $\vec{A}_j$ and $\vec{A}_j'$ entering in (5.9) are not independent, but are related by the two mode-coupling conditions (3.25) and (3.26). On using these conditions one readily finds that on

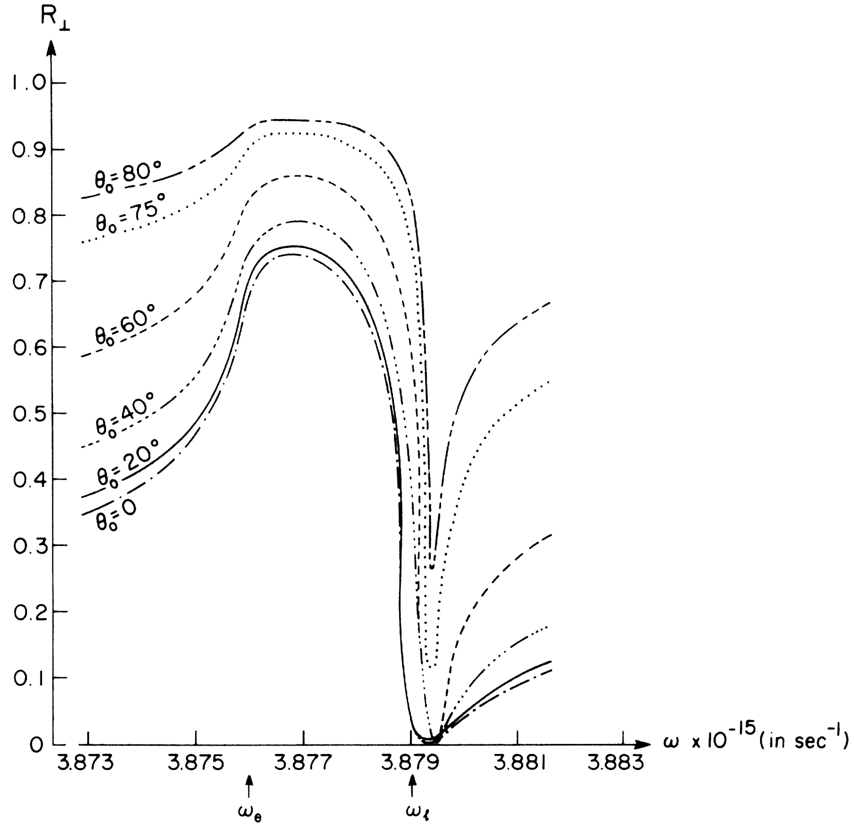

FIG. 6. Reflection coefficient $R_\perp$ as a function of the frequency ω for selected values of the angle of incidence θ_0. (Calculated with the same values of the parameters as given in the caption to Fig. 2.)

the boundaries of the slab $\vec{\mathcal{P}}_{\rm NL}$ and its normal derivatives must necessarily be related by the following two constraints:

$$\left(\vec{\mathcal{P}}_{\rm NL}(\vec{r},\ \omega;\ u,\ v)+\frac{1}{iw_\mu}\frac{\partial\vec{\mathcal{P}}_{\rm NL}(\vec{r},\ \omega;\ u,\ v)}{\partial z}\right)\bigg|_{z=0}=0\ , \tag{5.10a}$$

$$\left(\vec{\mathcal{P}}_{\rm NL}(\vec{r},\ \omega;\ u,\ v)-\frac{1}{iw_\mu}\frac{\partial\vec{\mathcal{P}}_{\rm NL}(\vec{r},\ \omega;\ u,\ v)}{\partial z}\right)\bigg|_{z=d}=0\ . \tag{5.10b}$$

The relations (5.10) are homogeneous *boundary conditions that each* (u, v, ω) *mode of the nonlocal polarization must satisfy on the two faces of the slab.* We stress that these boundary conditions were obtained as a direct consequence of our analysis regarding the nature of the electromagnetic field in a spatially dispersive medium and are therefore not additional postulates.

It should be noted that since, according to Eqs. (3.4) and (1.24),

$$w_\mu^2=(\mu^2-u^2-v^2)=\frac{m_e^*}{\hbar\omega_e}(\omega^2-\omega_e^2+i\omega\Gamma_e)-u^2-v^2\ , \tag{5.11}$$

the proportionality factor in (5.10) relating $\vec{\mathcal{P}}_{\rm NL}$ to its normal derivative on each of the faces of the slab is *frequency dependent.*

The boundary conditions (5.10) apply individually to each (u, v, ω) mode of the nonlocal polarization. From them one may readily derive boundary conditions on the total nonlocal polarization $\vec{P}_{\rm NL}(\vec{r}, \omega)$. This calculation is carried out in Appendix C and leads to the following result.

At each point $\vec{r}$ within the slab the nonlocal polarization $\vec{P}_{\rm NL}(\vec{r}, \omega)$ must satisfy the relations

$$\int\int_{-\infty}^{+\infty}\left(\vec{P}_{\rm NL}(\vec{r}\,',\ \omega)\frac{\partial G_\mu(|\vec{r}-\vec{r}\,'|)}{\partial z'}-G_\mu(|\vec{r}-\vec{r}\,'|)\frac{\partial\vec{P}_{\rm NL}(\vec{r}\,',\ \omega)}{\partial z'}\right)\bigg|_{z'=0}dx'\,dy'=0\ , \tag{5.12a}$$

and

$$\int\int_{-\infty}^{+\infty}\left(\vec{P}_{\rm NL}(\vec{r}\,',\ \omega)\frac{\partial G_\mu(|\vec{r}-\vec{r}\,'|)}{\partial z'}-G_\mu(|\vec{r}-\vec{r}\,'|)\frac{\partial\vec{P}_{\rm NL}(\vec{r}\,',\ \omega)}{\partial z'}\right)\bigg|_{z'=d}dx'\,dy'=0\ . \tag{5.12b}$$

The conditions (5.12a) and (5.12b) each have the form of a nonlocal boundary condition, which to some extent resembles the Ewald-Oseen extinction theorem of molecular optics (Ref. 28, pp. 100–104). For this reason, following Sein[16(a),(b)] we will refer to Eqs. (5.12) as *extinction theorems for nonlocal polarization.*

We will now establish several generalizations of these extinction theorems. First, we will derive an extinction theorem appropriate to a spatially dispersive medium, whose constitutive relation is again given by (2.8), but which now occupies a domain V, bounded by any closed surface Σ. For this purpose we note that the nonlocal polarization which, according to (5.1) and (2.8), is now given by

$$\vec{P}_{NL}(\vec{r}, \omega) = \frac{\chi}{(4\pi)^2}\int_V G_\mu(|\vec{r}-\vec{r}'|)\vec{E}(\vec{r}', \omega)\, d^3r' \tag{5.13}$$

satisfies the differential equation

$$(\nabla^2+\mu^2)\vec{P}_{NL}(\vec{r}, \omega) = -\left(\frac{\chi}{4\pi}\right)\vec{E}(\vec{r}, \omega). \tag{5.14}$$

Now, the Green's function $G_\mu(|\vec{r}-\vec{r}'|)$, defined by (2.4), is the outgoing solution of the equation

$$(\nabla^2+\mu^2)G_\mu(|\vec{r}-\vec{r}'|) = -4\pi\delta(\vec{r}-\vec{r}'). \tag{5.15}$$

where δ is the Dirac δ function. From (5.14) and (5.15) it follows in customary manner, with the help of Green's theorem, that for any point $\vec{r}$ inside the medium

$$\vec{P}_{NL}(\vec{r}, \omega) = \frac{\chi}{(4\pi)^2}\int_V G_\mu(|\vec{r}-\vec{r}'|)\vec{E}(\vec{r}', \omega)\, d^3r' - \frac{1}{4\pi}\int_\Sigma\left(\vec{P}_{NL}(\vec{r}', \omega)\frac{\partial G_\mu(|\vec{r}-\vec{r}'|)}{\partial n} - G_\mu(|\vec{r}-\vec{r}'|)\frac{\partial \vec{P}_{NL}(\vec{r}', \omega)}{\partial n}\right)dS, \tag{5.16}$$

where $\partial/\partial n$ denotes differentiation along the outward normal to Σ. It is clear that (5.16) is consistent with the expression (5.13) (that follows from our assumed constitutive relation) only if at each point $\vec{r}$ inside V

$$\int_\Sigma\left(\vec{P}_{NL}(\vec{r}', \omega)\frac{\partial G_\mu(|\vec{r}-\vec{r}'|)}{\partial n} - G_\mu(|\vec{r}-\vec{r}'|)\frac{\partial \vec{P}_{NL}(\vec{r}', \omega)}{\partial n}\right)dS = 0. \tag{5.17}$$

This formula, which is clearly a generalization of Eqs. (5.12), represents the *extinction theorem for nonlocal polarization* in a spatially dispersive medium whose constitutive relation is given by (2.8). We have now derived it solely from requirements of consistency.

The argument that led to (5.17) may be readily applied to obtain an analogous extinction theorem for any medium for which the constitutive relation is of the general form

$$\vec{D}(\vec{r}, \omega) = \epsilon_0\vec{E}(\vec{r}, \omega) + \int_V \epsilon_1(\vec{r}-\vec{r}'; \omega)\vec{E}(\vec{r}', \omega)\, d^3r', \tag{5.18}$$

where the "nonlocal part" ϵ_1 of the dielectric constant is the outgoing Green's function of some lin-

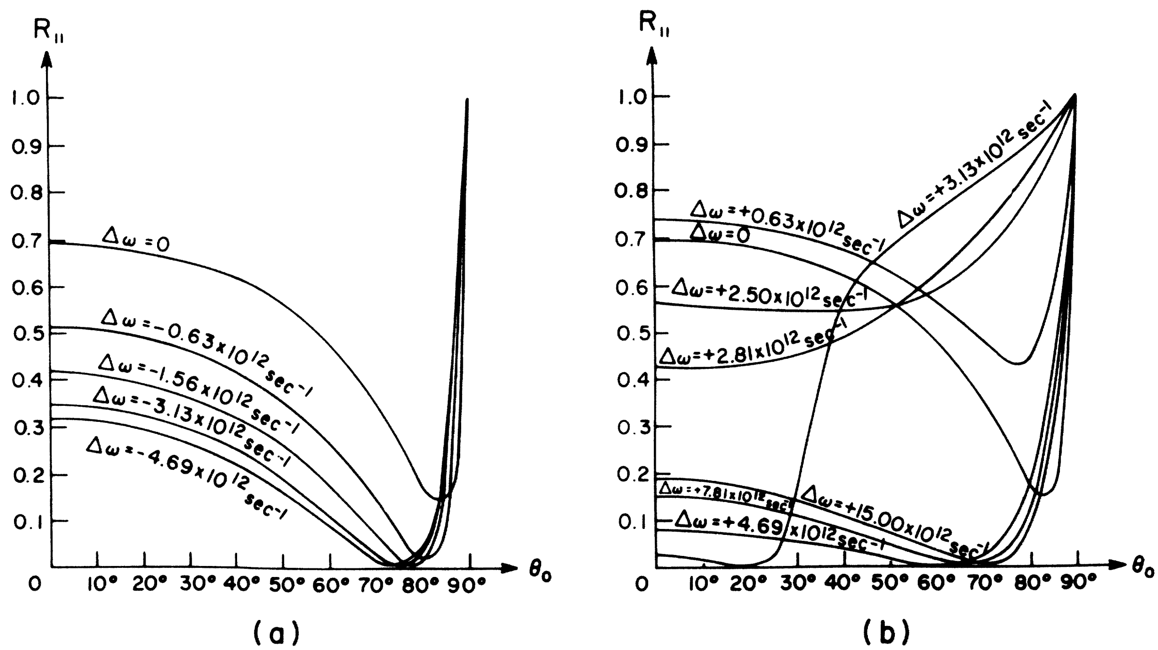

FIG. 7. Reflection coefficient $R_\parallel$ as a function of the angle of incidence θ_0 at selected frequencies below [(a)] and above [(b)] the exciton transition frequency ω_e. The curves are labeled by the frequency difference $\Delta\omega = \omega - \omega_e$. (Calculated with the same values of the parameters as given in the caption to Fig. 2.)

ear differential operator $\mathcal{L}$, i.e., the outgoing solution of the differential equation

$$\mathcal{L}\epsilon_1(\vec{r}-\vec{r}\,';\ \omega) = -4\pi\delta(\vec{r}-\vec{r}\,'), \tag{5.19}$$

$\mathcal{L}$ being assumed to operate on the coordinates of $\vec{r}$. In this case the nonlocal part

$$\vec{P}_{NL}(\vec{r},\ \omega) = \int_V \epsilon_1(\vec{r}-\vec{r}\,';\ \omega)\vec{E}(\vec{r}\,',\ \omega)\, d^3r' \tag{5.20}$$

of the polarization evidently satisfies the differential equation

$$\mathcal{L}\vec{P}_{NL}(\vec{r},\ \omega) = -\vec{E}(\vec{r},\ \omega). \tag{5.21}$$

From (5.19) and (5.21) one readily finds that

$$\vec{P}_{NL}(\vec{r},\ \omega) = \frac{1}{4\pi}\int_V \epsilon_1(\vec{r}-\vec{r}\,';\ \omega)\vec{E}(\vec{r}\,',\ \omega)\, d^3r' + \frac{1}{4\pi}\int_V [\epsilon_1(\vec{r}-\vec{r}\,';\ \omega)\mathcal{L}'\vec{P}_{NL}(\vec{r}\,',\ \omega) - \vec{P}_{NL}(\vec{r}\,',\omega)\,\mathcal{L}'\epsilon_1(\vec{r}-\vec{r}\,',\ \omega)]\, d^3r', \tag{5.22}$$

where the prime on $\mathcal{L}'$ indicates that the operator now acts on the coordinates of $\vec{r}\,'$ and the integration extends over the volume V occupied by the spatially dispersive medium. Now the second volume integral appearing on the right-hand side of (5.22) may be converted into a surface integral with the help of Green's theorem, generalized to the case of an arbitrary linear differential operator,[35]

$$\int_V [\Phi(\vec{r}\,')\mathcal{L}'\Psi(\vec{r}\,') - \Psi(\vec{r}\,')\mathcal{L}'\Phi(\vec{r}\,')]\, d^3r' = \int_\Sigma \vec{n}\cdot\vec{C}\{\Phi(\vec{r}\,');\ \Psi(\vec{r}\,')\} dS. \tag{5.23}$$

Here, $\vec{n}$ is the unit outward normal to the surface Σ bounding the volume V and $\vec{C}$ is the bilinear concomitant associated with the operator $\mathcal{L}$ and is defined by the relation

$$\Phi\mathcal{L}\Psi - \Psi\mathcal{L}^\dagger\Phi = \nabla\cdot\vec{C}\{\Phi,\Psi\}, \tag{5.24}$$

$\mathcal{L}^\dagger$ is the operator adjoint to $\mathcal{L}$ and $\Phi(\vec{r})$ and $\Psi(\vec{r})$ are arbitrary functions. On making use of (5.23) in (5.22), we see that each Cartesian component of $\vec{P}_{NL}(\vec{r},\ \omega)$ may be expressed in the form

$$[\vec{P}_{NL}(\vec{r},\ \omega)]_j = \frac{1}{4\pi}\int_V \epsilon_1(\vec{r}-\vec{r}\,';\ \omega)\vec{E}_j(\vec{r}\,',\ \omega)\, d^3r' + \frac{1}{4\pi}\int_\Sigma \vec{n}\cdot\vec{C}\{\epsilon_1(\vec{r}-\vec{r}\,',\ \omega); [\vec{P}_{NL}(\vec{r}\,',\ \omega)]_j\} dS, \tag{5.25}$$

where the subscript j labels Cartesian components. Evidently (5.25) is consistent with (5.20) only if at each point inside V

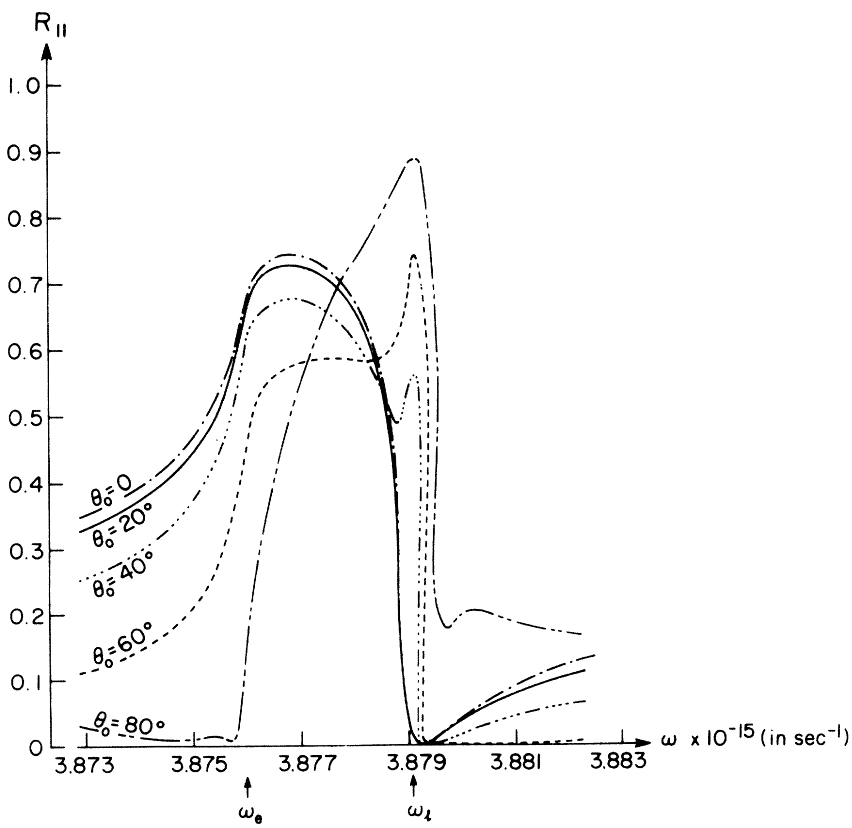

FIG. 8. Reflection coefficient $R_\parallel$ as a function of the frequency ω for selected values of the angle of incidence θ_0. (Calculated with the same values of the parameters as given in the caption to Fig. 2.)

$$\int_\Sigma \vec{n}\cdot\vec{C}\{\epsilon_1(\vec{r}-\vec{r}',\omega);\ [\vec{P}_{NL}(\vec{r}',\omega)]_j\}dS=0\,. \tag{5.26}$$

The formula (5.26) may be considered to be the *extinction theorem*[36] *for nonlocal polarization in a spatially dispersive medium whose constitutive relation is of the form* (5.18) (*with* ϵ_1 *being the outgoing solution of* (5.19).

It is clear from our analysis that electrodynamics in a spatially dispersive medium of the general class that we just considered is, for each frequency ω, completely specified by

1. Electrodynamic equations

$$\nabla\times(\nabla\times\vec{E})-k_0^2\vec{D}=0\,,$$

$$\vec{H}=\vec{B}=\frac{1}{ik_0}\nabla\times\vec{E}\,.$$

2. Constitutive relation

$$\vec{D}=\epsilon_0\vec{E}+4\pi\vec{P}_{NL}\,,$$

where $\vec{P}_{NL}$ is the solution of the equation

$$\mathcal{L}\vec{P}_{NL}=-\vec{E}\,.$$

subject to the extinction theorem (5.26).

3. Boundary conditions

Usual "continuity" conditions on appropriate components of the electromagnetic field vectors at a surface of discontinuity.

It may be worth noting that the extinction theorem (5.26) involves the $\mathcal{L}$ operator, which itself depends on the response properties of the medium.

Throughout the main part of this paper we have been concerned with a medium for which [as the comparison of (5.21) with (5.14) shows] the $\mathcal{L}$ operator is of the form

$$\mathcal{L}=\left(\frac{4\pi}{\chi}\right)(\nabla^2+\mu^2) \tag{5.27}$$

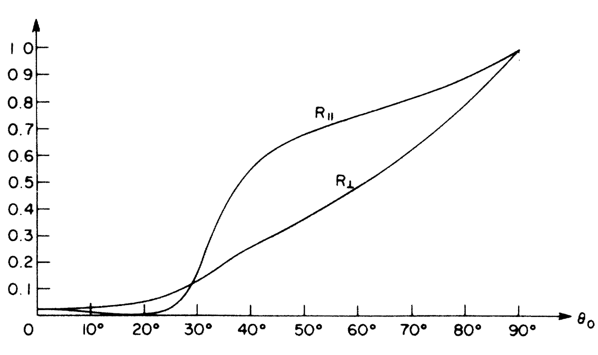

FIG. 9. Reflection coefficients $R_\parallel$ and $R_\perp$ as functions of the angle of incidence θ_0 for an electromagnetic wave at the longitudinal frequency $\omega=\omega_l\sim 3.879\times 10^{15}\ \text{sec}^{-1}$. (Calculated with the same values of the parameters as given in the caption to Fig. 2.)

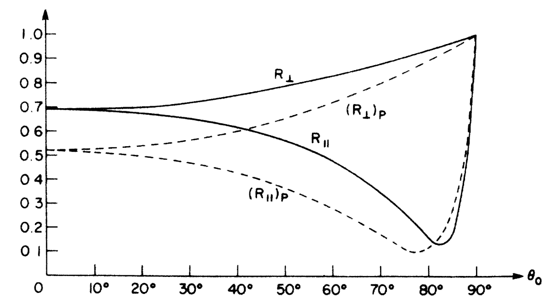

FIG. 10. Comparison of the reflection coefficients at the exciton transition frequency $\omega=\omega_e$ as functions of the angle of incidence θ_0, calculated from the present theory (solid lines) and from Pekar's theory (suffix P, dashed lines). (Calculated with the same values of the parameters as given in the caption to Fig. 2.)

and the nonlocal part of the dielectric constant is

$$\epsilon_1(\vec{r}-\vec{r}',\omega)=\left(\frac{\chi}{4\pi}\right)G_\mu(|\vec{r}-\vec{r}'|)\,, \tag{5.28}$$

as is seen by comparing (5.18) with (2.5). Now, the operator (5.27) is self-adjoint ($\mathcal{L}^\dagger=\mathcal{L}$) and the bilinear concomitant $\vec{C}$ defined by (5.24) is in this case readily seen to be

$$\vec{C}\{\Phi,\Psi\}=\left(\frac{4\pi}{\chi}\right)(\Phi\nabla\Psi-\Psi\nabla\Phi)\,. \tag{5.29}$$

If we substitute from (5.29) and (5.28) into the extinction theorem (5.26), it indeed reduces to (5.17).

Another case of considerable interest, which may be treated by our method, is that characterized by an $\mathcal{L}$ operator of the form

$$\mathcal{L}=4\pi\prod_{n=1}^{N}\frac{1}{\chi_n}(\nabla^2+\mu_n^2)\,. \tag{5.30}$$

This situation arises when several exciton bands are present or when in (1.21), instead of ignoring the $\vec{k}$ dependence of the damping function $\Gamma(\vec{k})$, one approximates it by the same kind of resonance form as the dielectric constant itself. We will not consider this case here.[37]

Experiments involving spatial dispersion are frequently carried out at normal incidence. If the scalar dielectric constant has the form (1.23), assumed throughout the main part of this paper, and if the incidence is normal, then according to Sec. IV only the mode $u=v=0$ is generated inside the slab and, moreover, its plane-wave constituents are all transverse ($\vec{A}_1'=\vec{A}_2'=0$). Equation (3.5) then reduces to (if we omit the caret on $\hat{\vec{E}}$)

$$\left(\frac{\partial^2}{\partial z^2}+k_0^2\epsilon_0\right)\vec{E}(z,\omega)=-4\pi\vec{P}_{NL}(z,\omega)\,, \tag{5.31}$$

where

$$\vec{P}_{NL}(z,\omega)=\frac{i\chi k_0^2}{8\pi}\int_0^d \frac{e^{iw_\mu|z-z'|}}{w_\mu}\vec{E}(z',\omega)\,dz'. \tag{5.32}$$

In this case the mode-coupling conditions (3.25) and (3.26) become [since according to (3.4) we now have $w_\mu=\mu$]

$$\sum_{j=1}^{4}\frac{\vec{A}_j}{w_j-\mu}=0,\quad \sum_{j=1}^{4}\frac{\vec{A}_j e^{iw_j d}}{w_j+\mu}=0 \tag{5.33}$$

and imply the boundary conditions

$$\left(\vec{P}_{NL}(z,\omega)+\frac{1}{i\mu}\frac{\partial\vec{P}_{NL}(z,\omega)}{\partial z}\right)\bigg|_{z=0}=0, \tag{5.34a}$$

$$\left(\vec{P}_{NL}(z,\omega)-\frac{1}{i\mu}\frac{\partial\vec{P}_{NL}(z,\omega)}{\partial z}\right)\bigg|_{z=d}=0, \tag{5.34b}$$

as is evident from Eqs. (5.10)

Finally, we briefly consider another case, which has been studied in detail by Mahan and Hopfield.[38] They considered the effects due to contributions to the dielectric constant that are linear in the wave vector, as is the case for crystals which do not possess inversion symmetry. In this case there is a splitting of the exciton band and the total nonlocal polarization is given by

$$\vec{P}_{NL}(z,\omega)=\vec{P}^{(+)}_{NL}(z,\omega)+\vec{P}^{(-)}_{NL}(z,\omega), \tag{5.35}$$

where $\vec{P}^{(+)}_{NL}(z,\omega)$ and $\vec{P}^{(-)}_{NL}(z,\omega)$ satisfy differential equations of the form[39]

$$\frac{\hbar^2}{m\epsilon(0)\beta^{(\pm)}}\left(\frac{\partial^2}{\partial z^2}\pm\frac{2mi\varphi}{\hbar^2}\frac{\partial}{\partial z}+\frac{m[(\hbar\omega)^2-\epsilon^2(0)+i\omega\Gamma_\pm]}{\epsilon(0)\hbar^2}\right)\times\vec{P}^{(\pm)}_{NL}(z,\omega)=-\vec{E}(z,\omega). \tag{5.36}$$

We may readily apply our method to determine the appropriate boundary conditions for the nonlocal polarization. For this purpose we first rewrite (5.36) in the form

$$\mathcal{L}^\pm\vec{P}^{(\pm)}_{NL}(z,\omega)=-\vec{E}(z,\omega), \tag{5.37}$$

where

$$\mathcal{L}^\pm=\frac{4\pi}{\chi^{(\pm)}}\left(\frac{\partial^2}{\partial z^2}\pm i\frac{2m\varphi}{\hbar^2}\frac{\partial}{\partial z}+\mu_\pm^2\right), \tag{5.38}$$

with

$$\chi^{(\pm)}=\frac{4\pi}{\hbar^2}m\epsilon(0)\beta^{(\pm)},\quad \mu_\pm^2=\frac{m}{\epsilon(0)\hbar^2}[(\hbar\omega)^2-\epsilon^2(0)+i\omega\Gamma_\pm]. \tag{5.39}$$

It is to be understood that each of the equations stands for two equations—one with the upper signs, the other with the lower signs.

According to Eqs. (5.35), (5.37), and (5.19), the "nonlocal" part of the dielectric constant $\hat{\epsilon}_1$ for this medium is given by

$$\hat{\epsilon}_1(z,\omega)=\hat{\epsilon}_1^{(+)}(z,\omega)+\hat{\epsilon}_1^{(-)}(z,\omega), \tag{5.40}$$

where $\hat{\epsilon}_1^{(+)}$ and $\hat{\epsilon}_1^{(-)}$ are the outgoing solutions of the differential equations

$$\mathcal{L}^\pm\hat{\epsilon}_1^{(\pm)}(z-z',\omega)=-4\pi\delta(z-z'). \tag{5.41}$$

The required solutions of (5.41) are readily found to be

$$\hat{\epsilon}_1^{(\pm)}(z-z',\omega)=\frac{4\pi i}{\nu_2^{(\pm)}-\nu_1^{(\pm)}}\exp(i\nu_2^{(\pm)}|z-z'|)\quad \text{when } z>z'$$
$$=\frac{4\pi i}{\nu_2^{(\pm)}-\nu_1^{(\pm)}}\exp(-i\nu_1^{(\pm)}|z-z'|)\quad \text{when } z<z', \tag{5.42}$$

where $\nu_1^{(\pm)}$ and $\nu_2^{(\pm)}$ are the roots of the equation

$$(\nu^{(\pm)})^2\pm\frac{2m\varphi}{\hbar^2}\nu^{(\pm)}-\mu_\pm^2=0, \tag{5.43}$$

with

$$\mathrm{Im}\,\nu_1^{(\pm)}<0,\quad \mathrm{Im}\,\nu_2^{(\pm)}>0. \tag{5.44}$$

The bilinear concomitant associated with the operators $\mathcal{L}^\pm$ may be shown to be

$$C^{(\pm)}\{\Phi,\Psi\}=\frac{4\pi}{\chi^{(\pm)}}\left[\left(\Phi\frac{\partial\Psi}{\partial z}-\Psi\frac{\partial\Phi}{\partial z}\right)\pm i\frac{2m\varphi}{\hbar^2}\Phi\Psi\right]. \tag{5.45}$$

With this form of the concomitant, the extinction theorem (5.26) for the half-space ($z>0$) occupied by the spatially dispersive medium reduces to

$$\left[\left(\hat{\epsilon}_1^{(\pm)}(z-z',\omega)\frac{\partial\vec{P}^{(\pm)}_{NL}(z',\omega)}{\partial z'}-\vec{P}^{(\pm)}_{NL}(z',\omega)\frac{\partial\hat{\epsilon}_1^{(\pm)}(z-z',\omega)}{\partial z'}\right)\pm i\frac{2m\varphi}{\hbar^2}\vec{P}^{(\pm)}_{NL}(z',\omega)\hat{\epsilon}_1^{(\pm)}(z-z',\omega)\right]\bigg|_{z'=0}=0 \tag{5.46}$$

and must be valid for all values of $z\geq 0$. On substituting for $\hat{\epsilon}_1^{(+)}$ and $\hat{\epsilon}_1^{(-)}$ the explicit expressions (5.42) and letting $z\to 0$ we find that

$$\left(\vec{P}^{(\pm)}_{NL}(z,\omega)+\frac{1}{iw^{(\pm)}}\frac{\partial\vec{P}^{(\pm)}_{NL}(z,\omega)}{\partial z}\right)\bigg|_{z=0}=0, \tag{5.47}$$

where

$$w^{(\pm)}=\mu^{(\pm)}\pm\frac{2m\varphi}{\hbar^2}. \tag{5.48}$$

Thus, the consistency of electrodynamics in a half-space occupied by the spatially dispersive medium under consideration demands that the two parts $\vec{P}^{(+)}_{NL}$ and $\vec{P}^{(-)}_{NL}$ of the nonlocal polarization obey the boundary conditions (5.47). These conditions differ from the abc's $\vec{P}^{(\pm)}_{NL}(0,\omega)=0$ of Mahan and Hopfield, but whether or not this difference is of practical significance in particular cases cannot be decided without numerical analysis.

VI. COMPARISON WITH OTHER THEORIES

Many other theories have been proposed relating to the nature of electromagnetic fields in spatially dispersive media. In this concluding section we will briefly compare some of the main features and results of our theory with those proposed by other authors.

Roughly speaking, the various theories may be divided into two groups.

A. Microscopic theories

These theories go beyond Maxwell's electromagnetic theory in that they involve microscopic models for the response of the medium to electromagnetic and mechanical excitations. To this group belong the pioneering investigations of Pekar[40] and those of Ginzburg and Agranovich,[4,5,13a] Hopfield and Thomas,[11d] Sugakov,[41] and Brenig, Zeyher, and Birman.[42]

B. Macroscopic theories

To this group belong theories in which the collective response of the medium is described phenomenologically by a response function that is a natural generalization of that for an ordinary (i.e., spatially nondispersive) medium. This group includes the theory presented in this paper and those of Ginzburg and Agranovich,[9] Sein,[16] Birman and Sein,[17] and Maradudin and Mills.[15]

Basically the different theories differ from each other in the form of the "additional boundary conditions" that they prescribe. However, even theories which prescribe the same abc's differ from each other in some aspects, especially in the nature of various assumptions which they contain. We will discuss some of these differences shortly. Most of the theories, whether microscopic or macroscopic, lead to abc's for the nonlocal polarization $\vec{P}_{NL}$ of the form

$$\vec{P}_{NL}(\vec{r},\omega)+\Lambda\frac{\partial\vec{P}_{NL}(\vec{r},\omega)}{\partial n}=0\ , \tag{6.1}$$

to be satisfied at each point $\vec{r}$ on the surface Σ (usually assumed to be a plane) bounding the spatially dispersive medium. $\partial/\partial n$ denotes differentiation along the outward normal to Σ and Λ is a parameter which, in general, depends on the particular theory. We have already pointed out at the end of Sec. IV in connection with the calculation of the reflectivity that the abc of Pekar's theory is $\vec{P}_{NL}(r,\omega)|_{z=0}=0$ [Eq. (4.60)] when $\vec{r}$ is on the boundary surface Σ, so that for Pekar's theory $\Lambda\equiv 0$. Under certain circumstances this special choice of Λ is also supported by considerations of Hopfield and Thomas and others, based on nonrigorous microscopic arguments as to the expected behavior of the exciton wave function at the boundary Σ. However, it seems to us doubtful—if, in fact it is at all possible, even in principle—that one may draw any conclusions about boundary conditions for the electromagnetic field from the behavior of the exciton wave function on the boundary. If $\Lambda\neq 0$ the knowledge of the dependence of Λ on the frequency is of importance, since this information is required for determining optical constants of the spatially dispersive medium from reflectivity measurements. Agranovich and Ginzburg considered this question to some extent. Their results imply that Λ is effectively independent of the frequency (cf. Sec. 10.4 of Ref. 5). (See, however, a recent paper by Agranovich and Yudson.[43]) This question was also considered by Sugakov,[41b] but it is difficult to interpret Sugakov's results (based on a microscopic model) in terms of the parameters of the macroscopic theory.

According to our theory, the boundary condition on the nonlocal polarization is not of the simple "local" form (6.1), but is a nonlocal condition, expressed by the extinction theorem (5.17), viz.,

$$\int_\Sigma\left(\vec{P}_{NL}(\vec{r}\,',\omega)\frac{\partial G_\mu(|\vec{r}-\vec{r}\,'|)}{\partial n}-G_\mu(|\vec{r}-\vec{r}\,'|)\frac{\partial\vec{P}_{NL}(\vec{r}\,',\omega)}{\partial n}\right)dS=0\,, \tag{6.2}$$

which must be satisfied at every point $\vec{r}$ inside the spatially dispersive medium. For the special case when the medium occupies the slab $0\leq z\leq d$, we have seen that (6.2) is a consequence of the fact that *each* $(u, v; \omega)$ *mode* in the slab obeys (local) boundary conditions of the form [Eq. (5.10)]

$$\vec{\mathcal{P}}_{NL}(\vec{r},\omega;u,v)-\frac{1}{iw_\mu}\frac{\partial\vec{\mathcal{P}}_{NL}(\vec{r},\omega;u,v)}{\partial n}=0\,, \tag{6.3}$$

for all points $\vec{r}$ situated on the faces of the slab, with w_μ being given by Eq. (5.11), viz.,

$$w_\mu=[\mu^2(\omega)-u^2-v^2]^{1/2} \tag{6.4a}$$

$$=\left(\frac{m_e^*}{\hbar\omega_e}(\omega^2-\omega_e^2+i\omega\Gamma)-u^2-v^2\right)^{1/2}, \tag{6.4b}$$

with $\mathrm{Re}\,w_\mu>0$, $\mathrm{Im}\,w_\mu>0$ [cf. (3.4b)]. We see then that in the special case when the spatially dispersive medium occupies a slab (or in the limit as $d\to\infty$ a half-space) the nonlocal polarization *of each* (u, v, ω) mode (not the nonlocal polarization itself) satisfies a boundary condition of the form (6.1), with

$$\Lambda=-\frac{1}{iw_\mu}\ , \tag{6.5}$$

w_μ being given by (6.4b). We stress that we have established this result as a rigorous consequence of Maxwell's electromagnetic theory alone, for a spatially dispersive medium whose constitutive relation has the form (2.8).

The present theory has much in common with the theories of Sein,[16] Birman and Sein,[17] and Maradudin and Mills.[15] In these investigations the same form of the constitutive relation is assumed as in our analysis. We will now briefly compare and contrast these theories with ours.

Sein[16] and Birman and Sein[17] take as their starting point a well-known integro-differential relation of molecular optics that couples the electric field with the induced polarization in an arbitrary medium. When the electric field is eliminated from this equation by the use of the dielectric constitutive relation, one obtains an integro-differential equation for the polarization field. To solve this equation Sein and Birman assumed that, irrespective of the exact shape of the spatially dispersive medium and irrespective of the exact nature of the incident monochromatic field, the polarization field may be expressed as the sum of plane waves, each propagating with a different velocity. The consistency of the solution of the integro-differential equation in this assumed form then leads to a set of relations involving the vector amplitudes and the wave vectors of the different plane waves. In this way Sein and Birman were led to a unique solution for the field inside the spatially dispersive medium, without introducing any additional boundary conditions. This approach clearly provides some insight into the structure of the field in the spatially dispersive medium, in the special circumstance when the plane-wave assumption is justified. That this is so in the special case when the medium occupies a plane parallel slab or a half-space has been demonstrated in the present paper (Sec. III). However, the plane-wave assumption appears not to be justified in general.

The other recently formulated macroscopic theory, that of Maradudin and Mills,[15] is intimately related to that presented in this paper. Maradudin and Mills base their analysis on the differential form of Maxwell equations, just as we do, and consider the interaction of a linearly polarized plane wave, with a spatially dispersive medium, that occupies the half-space $z > 0$. They take the electric field generated inside the medium to be of the form [their equation (3a)]

$$\vec{E}(\vec{r}, t) = \vec{E}(z) e^{ik_{\parallel}x} e^{-i\omega t} \tag{6.6}$$

(with similar expressions for the electric displacement vector and the magnetic field), the plane of incidence being taken as the xz plane. By substituting these expressions into Maxwell equations they are led to an integro-differential equation for $\vec{E}(z)$. Maradudin and Mills found that the solution of the equation is of the form

$$\vec{E}(z) = \vec{E}_1 e^{iq_1 z} + \vec{E}_2 e^{iq_2 z}, \tag{6.7a}$$

when the incident electric field is linearly polarized at right angles to the plane of incidence and is of the form

$$\vec{E}(z) = \vec{E}'_1 e^{iq'_1 z} + \vec{E}'_2 e^{iq'_2 z} + \vec{E}'_3 e^{iq'_3 z}, \tag{6.7b}$$

when the incident electric field is linearly polarized parallel to the plane of incidence. The quantities q_1, q'_1, etc., in Eqs. (6.7) were found to be given by the usual dispersion relations. Moreover, Maradudin and Mills showed that in Eq. (6.7a) the two vector amplitudes $\vec{E}_1$ and $\vec{E}_2$ are associated with transverse waves, that in Eq. (6.7b) two of the vector amplitudes are associated with transverse waves and one with a longitudinal wave, and that in each of the two cases the vector amplitudes are connected by a certain linear relation.

These results are in agreement with the solution that we presented in Sec. IV for refraction and reflection. (A discrepancy, probably due to an error in algebra, is noted in footnote 27.) The difference between the two treatments is that, while Maradudin and Mills assumed that the fields in the medium may be represented in the form (6.6), we showed as a consequence of the general mode representation obtained in Sec. III of the present paper that the fields in the medium must necessarily be of this form. For, as we demonstrated in Sec. IV when a monochromatic plane wave is incident on the spatially dispersive medium occupying a half-space, a single (u, v, ω) mode is excited in the medium. According to Eq. (4.6) the electric field of such a mode is of the form (suppressing the periodic term $e^{-i\omega t}$)

$$\vec{\mathcal{E}}(\vec{r}, \omega; u, v) = \vec{A}_t^{(1)}(u, v; \omega) e^{i\vec{k}_t^{(1)}\cdot\vec{r}} + \vec{A}_t^{(2)}(u, v; \omega) e^{i\vec{k}_t^{(2)}\cdot\vec{r}} + \vec{A}_l(u, v; \omega) e^{i\vec{k}_l\cdot\vec{r}}. \tag{6.8}$$

On substituting into Eq. (6.8) the expressions (4.9) and (4.10) for the wave vectors, (6.8) may be rewritten in the form

$$\vec{\mathcal{E}}(\vec{r}, \omega; u, v) = \vec{\mathcal{E}}(z) e^{i(ux+vy)}, \tag{6.9}$$

with

$$\vec{\mathcal{E}}(z) = \vec{A}_t^{(1)}(u, v; \omega) e^{iw_t^{(1)}z} + \vec{A}_t^{(2)}(u, v; \omega) e^{iw_t^{(2)}z} + \vec{A}_l(u, v; \omega) e^{iw_l z}. \tag{6.10}$$

The quantities $w_t^{(1)}$, $w_t^{(2)}$, and w_l are given by the dispersion relations and the vector amplitudes $\vec{A}_t^{(1)}$, $\vec{A}_t^{(2)}$, and $\vec{A}_l$ are related by the mode-coupling condition. Moreover, as we showed in Sec. IV, $\vec{A}_l = 0$ when the incident electric field is linearly polarized at right angles to the plane of incidence. If the plane of incidence is taken to be the xz plane as Maradudin and Mills have done, the second parameter which labels the mode, i.e., the parameter v, has then the value zero and the electric field mode (6.9) then reduces to the form (6.6) assumed by Maradudin and Mills. Detailed comparison shows that the electric field mode (6.9) is identical with the solution of Maradudin and Mills.

As regards the electromagnetic field within the spatially dispersive medium, our analysis goes well beyond that of Maradudin and Mills in that we determined, without any assumptions whatsoever, the general mode structure of the electromagnetic field in the spatially dispersive model medium occupying a plane parallel slab. On the other hand, Maradudin and Mills include in their paper a very thorough and elegant application of their analysis to the theory of surface waves, a subject not considered in the present paper.

We conclude by stressing that the macroscopic theories of Birman and Sein and Maradudin and Mills and our own lead—with different degrees of rigor—to the same basic conclusion: Namely, that within the framework of a macroscopic treatment the problem of refraction and reflection on a spatially dispersive medium occupying a half-space (or a plane parallel slab) can be solved completely within the framework of classical electromagnetic theory, without requiring the introduction of any additional boundary conditions.

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APPENDIX A: DERIVATION OF EQUATIONS (3.21)–(3.23)

It is clear from Eq. (3.19) that in order to determine the constraints on the amplitude vectors $\vec{A}_j$, $\vec{B}_j$, and $\vec{A}'_j$, we may consider separately solutions of the form

$$(\vec{A}_j + z\vec{B}_j)e^{iw_jz} \text{ and } \vec{A}'_j e^{iw'_jz} ,$$

since each of these two solutions is associated with a different propagation vector, namely (u, v, w_j) and (u, v, w'_j), respectively. We consider first the wave characterized by the second wave vector.

A. Longitudinality condition (3.22)

We substitute

$$\hat{\vec{E}} = \vec{A}'_j(u, v, \omega)e^{iw'_jz} \tag{A1}$$

in Eq. (3.8) and find that the left-hand side becomes

$$\mathfrak{M}_t(u, v, w'_j; \omega)\vec{A}'_j(u, v; \omega)e^{iw'_jz}, \tag{A2}$$

where $\mathfrak{M}_t$ is defined by Eq. (3.17). To determine the right-hand side of Eq. (3.8) we first evaluate the quantities $\hat{\vec{\xi}}$ and $\hat{\vec{L}}$, defined by Eq. (3.6). We find that when $\hat{\vec{E}}$ is given by (A1)

$$\vec{\xi} = i(\vec{k}'_j \cdot \vec{A}'_j)e^{iw'_jz}, \tag{A3}$$

$$\vec{L} = -(\vec{k}'_j \cdot \vec{A}_j)\vec{k}'_j e^{iw'_jz}, \tag{A4}$$

where

$$\vec{k}'_j \equiv u, v, w'_j . \tag{A5}$$

On substituting from (A2) and (A4) in (3.8) we find that

$$\mathfrak{M}_t(u, v, w'_j; \omega)\vec{A}'_j = (w_j^2 - w_\mu^2)(\vec{k}_j \cdot \vec{A}'_j)\vec{k}'_j . \tag{A6}$$

If we substitute for $\mathfrak{M}_t(u, v, w'_j; \omega)$ from (3.17) and make use of the dispersion relation (3.20b), (A6) is readily seen to imply that

$$\frac{\chi}{\epsilon_0}\left(\frac{\chi}{\epsilon_0} - \vec{k}'^2_j\right)\vec{A}'_j = -\frac{\chi}{\epsilon_0}(\vec{k}'_j \cdot \vec{A}'_j)\vec{k}'_j . \tag{A7}$$

If we multiply (A7) vectorially by $\vec{k}'_j$ and use the fact that $\chi \neq 0$ and $\chi/\epsilon_0 \neq \vec{k}'_j$, as is clear from the dispersion relation (3.20b), we deduce that

$$\vec{k}'_j \times \vec{A}'_j = 0 , \tag{A8}$$

which is the longitudinality condition (3.22).

B. Transversality condition (3.21) and vanishing of the $\vec{B}_j$ coefficients

Next, we consider solutions of the form

$$\hat{\vec{E}} = [\vec{A}_j(u, v; \omega) + z\vec{B}_j(u, v; \omega)]e^{iw_jz} . \tag{A9}$$

On substituting from (A9) in Eq. (3.8) and making use of the dispersion relation (3.20a) we find that the left-hand side of (3.8) becomes

$$2iw_j[(k_0^2\epsilon_0 - u^2 - v^2 + w_\mu^2) - 2w_j^2]\vec{B}_j e^{iw_jz} . \tag{A10}$$

The quantities $\vec{\xi}$ and $\vec{L}$, defined by Eq. (3.6), are now readily found to be given by

$$\vec{\xi} = [i(\vec{k}_j \cdot \vec{A}_j) + iz(\vec{k}_j \cdot \vec{B}_j) + B_{jz}]e^{iw_jz}, \tag{A11}$$

$$\vec{L} = \{ -(\vec{k}_j \cdot \vec{A}_j)\vec{k}_j + [-(\vec{k}_j \cdot \vec{B}_j)z + iB_{jz}]\vec{k}_j + i(\vec{k}_j \cdot \vec{B}_j)\hat{\vec{z}}\} e^{iw_jz}, \tag{A12}$$

where B_{jz} denotes the z component of the vector $\vec{B}_j$ and $\hat{\vec{z}}$ denotes the unit vector in the positive z direction. On substituting from (A10) and (A12) into Eq. (3.8) we obtain the relation

$$\begin{aligned} 2iw_j[(k_0^2\epsilon_0 - u^2 - v^2 + w_\mu^2) - 2w_j^2]\vec{B}_j &= (w_\mu^2 - w_j^2)\{[iB_{jz} - (\vec{k}_j \cdot \vec{B}_j)z - (\vec{k}_j \cdot \vec{A}_j)]\vec{k}_j \\ &\quad + i(\vec{k}_j \cdot \vec{B}_j)\hat{\vec{z}}\} - 2iw_j(\vec{k}_j \cdot \vec{B}_j)\vec{k}_j . \end{aligned} \tag{A13}$$

Equation (A13) must hold for all values of $z \geq 0$. This can only be so if the terms dependent on z and the terms independent of z vanish separately. The vanishing of the term depending on z gives at once

$$(\vec{k}_j \cdot \vec{B}_j)\vec{k}_j = 0 . \tag{A14}$$

Now, one may readily deduce from the dispersion relation (3.20a) that $\vec{k}_j \neq 0$. Hence, (A14) implies that

$$\vec{k}_j \cdot \vec{B}_j = 0 . \tag{A15}$$

On making use of (A15), (A13) simplifies to

$$2iw_j[(k_0^2\epsilon_0 - u^2 - v^2 + w_\mu^2) - 2w_j^2]\vec{B}_j = (w_\mu^2 - w_j^2) \times [iB_{jz} - (\vec{k}_j \cdot \vec{A}_j)]\vec{k}_j . \tag{A16}$$

On taking the scalar product of (A16) with $\vec{k}_j$ and making use of (A15), we find that

$$(w_\mu^2 - w_j^2)[iB_{jz} - (\vec{k}_j \cdot \vec{A}_j)]\vec{k}_j^2 = 0 . \tag{A17}$$

Now, one may readily deduce from the dispersion relation (3.20a) that w_j^2 cannot equal w_μ^2. Moreover, it is not difficult to see that $\vec{k}_j^2$ cannot vanish. For the vanishing of $\vec{k}_j^2$ would imply, according to (3.24), that $w_j^2 = -u^2 - v^2$. On substituting this expression for w_j^2 into the transverse dispersion relation (3.20a) and using Eq. (3.4a), we find that this would require that $\mu^2 = \chi/\epsilon_0$. However, μ^2 is complex, whereas χ/ϵ_0 was assumed to be real. Hence, Eq. (A17) implies that

$$\vec{k}_j \cdot \vec{A}_j = iB_{jz}. \tag{A18}$$

On using this result in (A16) it follows that

$$w_j[(k_0^2\epsilon_0 - u^2 - v^2 + w_\mu^2) - 2w_j^2]\vec{B}_j = 0. \tag{A19}$$

It can be shown from the dispersion relation (3.20a) that w_j cannot vanish for any real frequency ω. The vanishing of the square bracket implies that

$$w_j^2 = \tfrac{1}{2}(k_0^2\epsilon_0 - u^2 - v^2 + w_\mu^2) \quad (j = 1, 2, 3, 4). \tag{A20}$$

Hence, in this case the dispersion equation (3.20a) has two double roots and not four distinct roots. However, we have excluded such a degenerate case from our analysis [cf. remarks after Eq. (3.18)] and so we may conclude that, within the domain of validity of our theory, Eq. (A19) can only be satisfied if

$$\vec{B}_j = 0. \tag{A21}$$

Equation (A18) now implies that

$$\vec{k}_j \cdot \vec{A}_j = 0. \tag{A22}$$

Equations (A21) and (A22) are Eqs. (3.21) and (3.23) of the text.

APPENDIX B: DERIVATION OF MODE-COUPLING CONDITIONS (3.25) AND (3.26)

According to Eqs. (3.19) and (3.23), the general solution of Eq. (3.8) is

$$\hat{\vec{E}}(u, v; z; \omega) = \sum_{j=1}^{4} \vec{A}_j(u, v; \omega)e^{iw_jz} + \sum_{j=1}^{2} \vec{A}_j'(u, v; \omega)e^{iw_j'z}, \tag{B1}$$

where the $\vec{A}_j$ are any transverse vector fields [i.e., fields that obey the condition (3.21)] and $\vec{A}_j'$ are any longitudinal fields [i.e., fields that obey the condition (3.22)]. In order that (B1) also be a solution of the integro-differential equation (3.5), the $\vec{A}_j$'s and $\vec{A}_j'$'s must satisfy additional constraints that may readily be determined by substituting from (B1) into Eq. (3.5). The calculation may be shortened by recalling that for each term of the form (A1) the vector $\vec{L}$ is given by (A4). We then find that the left-hand side of (3.5) becomes

$$\sum_{j=1}^{4}(\vec{k}_j^2 - k_0^2\epsilon_0)\vec{A}_je^{iw_jz} + \sum_{j=1}^{2}(\vec{k}_j'^2 - k_0^2\epsilon_0)\vec{A}_j'e^{iw_j'z} - \sum_{j=1}^{2}(\vec{k}_j'\cdot\vec{A}_j')\vec{k}_j'e^{iw_j'z}, \tag{B2}$$

where $\vec{k}_j$ and $\vec{k}_j'$ are, of course, vectors with components u, v, w_j and u, v, w_j', respectively, and we have made use of the transversality condition (3.21). Now we have the vector identity $(\vec{k}_j'^2)\vec{A}_j' - \vec{k}_j'(\vec{k}_j'\cdot\vec{A}_j') = -\vec{k}_j'\times(\vec{k}_j'\times\vec{A}_j')$ and since, according to Eq. (3.22), $\vec{k}_j'\times\vec{A}_j' = 0$, two of the terms in (B2) cancel and (B2) simplifies to

$$\sum_{j=1}^{4}(\vec{k}_j^2 - k_0^2\epsilon_0)\vec{A}_je^{iw_jz} - \sum_{j=1}^{2}k_0^2\epsilon_0\vec{A}_j'e^{iw_j'z}. \tag{B3}$$

On substituting from (B1) into the right-hand side of Eq. (3.5) we see that the right-hand side of Eq. (3.5) will contain a sum of integrals of the form

$$I = \int_0^d \frac{e^{iw_\mu|z-z'|}}{w_\mu} e^{iw_jz'}dz'.$$

This integral may readily be evaluated. We have

$$I = \int_0^z \frac{e^{iw_\mu(z-z')}}{w_\mu}e^{iw_jz'}dz' + \int_z^d \frac{e^{iw_\mu(z'-z)}}{w_\mu}e^{iw_jz'}dz' = \frac{2e^{iw_jz}}{i(w_j^2 - w_\mu^2)} - \frac{e^{iw_\mu z}}{iw_\mu(w_j - w_\mu)} + \frac{e^{-iw_\mu(z-d)}e^{iw_jd}}{iw_\mu(w_j + w_\mu)}. \tag{B4}$$

Using this result, the right-hand side of (3.5) is found to be

$$\frac{i\chi k_0^2}{2}\left[\frac{2}{i}\sum_{j=1}^{4}\frac{\vec{A}_j}{(w_j^2 - w_\mu^2)}e^{iw_jz} + \frac{2}{i}\sum_{j=1}^{2}\frac{\vec{A}_j'}{w_j'^2 - w_\mu^2}e^{iw_j'z} - \frac{e^{iw_\mu z}}{iw_\mu}\left(\sum_{j=1}^{4}\frac{\vec{A}_j}{w_j - w_\mu} + \sum_{j=1}^{2}\frac{\vec{A}_j'}{w_j' - w_\mu}\right) + \frac{e^{-iw_\mu(z-d)}}{iw_\mu}\left(\sum_{j=1}^{4}\frac{\vec{A}_j}{w_j + w_\mu}e^{iw_jd} + \sum_{j=1}^{2}\frac{\vec{A}_j'}{w_j' + w_\mu}e^{iw_j'd}\right)\right]. \tag{B5}$$

On equating the left- and right-hand sides, given by Eqs. (B3) and (B5), respectively, and on rearranging terms we obtain the relation

$$\sum_{j=1}^{4}\left[\vec{k}_j^2 - k_0^2\left(\epsilon_0 + \frac{\chi}{w_j^2 - w_\mu^2}\right)\right]\vec{A}_je^{iw_jz} - \sum_{j=1}^{2}k_0^2\left(\epsilon_0 + \frac{\chi}{w_j'^2 - w_\mu^2}\right)\vec{A}_j'e^{iw_j'z} = \frac{e^{iw_\mu z}}{iw_\mu}\left(\sum_{j=1}^{4}\frac{\vec{A}_j}{w_j - w_\mu} + \sum_{j=1}^{2}\frac{\vec{A}_j'}{w_j' - w_\mu}\right) + \frac{e^{-iw_\mu(z-d)}}{iw_\mu}\left(\sum_{j=1}^{4}\frac{\vec{A}_j}{w_j + w_\mu}e^{iw_jd} + \sum_{j=1}^{2}\frac{\vec{A}_j'}{w_j' + w_\mu}e^{iw_j'd}\right). \tag{B6}$$

The left-hand side of (B6) may be readily shown to have zero value. To see this we note, first of all, that according to Eqs. (3.4) and (3.24) $w_j^2 - w_\mu^2 = \vec{k}_j^2 - \mu^2$, $w_j'^2 - w_\mu^2 = \vec{k}_j'^2 - \mu^2$, so that the left-hand side of (B6) may be rewritten in the form

$$\sum_{j=1}^{4}\left[\vec{k}_j^2 - k_0^2\left(\epsilon_0 + \frac{\chi}{\vec{k}_j^2 - \mu^2}\right)\right]\vec{A}_j e^{iw_j z} - \sum_{j=1}^{2} k_0^2\left(\epsilon_0 + \frac{\chi}{\vec{k}'^2 - \mu^2}\right)\vec{A}_j' e^{iw_j' z}. \quad \text{(B7)}$$

If we recall the expression (1.23) for $\tilde{\epsilon}(\vec{k}, \omega)$, we see that (B7) may be expressed as

$$\sum_{j=1}^{4}[\vec{k}_j^2 - k_0^2\tilde{\epsilon}(\vec{k}_j; \omega)]\vec{A}_j e^{iw_j z} - \sum_{j=1}^{2} k_0^2\tilde{\epsilon}(\vec{k}_j'; \omega)e^{iw_j' z}.$$

Now, the terms under each of the summation signs will vanish because of the dispersion relations (3.29). Hence, the left-hand side and consequently also the right-hand side of Eq. (B6) vanish and we obtain the relation

$$e^{iw_\mu z}\left(\sum_{j=1}^{4}\frac{\vec{A}_j}{w_j - w_\mu} + \sum_{j=1}^{2}\frac{\vec{A}_j'}{w_j' - w_\mu}\right) + e^{-iw_\mu z}\left(\sum_{j=1}^{4}\frac{\vec{A}_j}{w_j + w_\mu}e^{i(w_j + w_\mu)d} + \sum_{j=1}^{2}\frac{\vec{A}_j'}{w_j' + w_\mu}e^{i(w_j' + w_\mu)d}\right) = 0. \quad \text{(B8)}$$

Since (B8) holds for all values of z in the range $0 < z < d$ each of the two expressions in the large parentheses must separately vanish. We thus obtain the relations

$$\sum_{j=1}^{4}\frac{\vec{A}_j}{w_j - w_\mu} + \sum_{j=1}^{2}\frac{\vec{A}_j'}{w_j' - w_\mu} = 0, \quad \text{(B9)}$$

$$\left(\sum_{j=1}^{4}\frac{\vec{A}_j}{w_j + w_\mu}e^{iw_j d} + \sum_{j=1}^{2}\frac{\vec{A}_j'}{w_j' + w_\mu}e^{iw_j' d}\right)e^{iw_\mu d} = 0. \quad \text{(B10)}$$

Equation (B9) is the mode-coupling condition (3.25) and Eq. (B10), after dividing both sides by the factor $e^{iw_\mu d}$, becomes the mode-coupling condition (3.26).

APPENDIX C: DERIVATION OF EXTINCTION THEOREMS (5.12) FOR NONLOCAL POLARIZATION

We begin with Eq. (5.10a), which we now write in the form

$$\left(\vec{\mathcal{P}}_{NL}(\vec{r}', \omega; u, v) + \frac{1}{iw_\mu}\frac{\partial\vec{\mathcal{P}}_{NL}(\vec{r}', \omega; u, v)}{\partial z'}\right)\bigg|_{z'=0} = 0, \quad \text{(C1)}$$

it being understood that $z' = 0$ represents the limiting values as $z' \to 0$ from inside the plane parallel slab, i.e., z' approaches zero through positive values. Recalling that according to Eq. (3.24) $\vec{k}_j \equiv u, v, w_j$, $\vec{k}_j' \equiv u, v, w_j'$, it follows from Eq. (5.3) that the two terms on the left-hand side of (C1) may be expressed in the form

$$\vec{\mathcal{P}}_{NL}(\vec{r}', \omega; u, v)\big|_{z'=0} = \vec{U}(u, v; \omega)e^{i(ux' + vy')} \quad \text{(C2)}$$

and

$$\frac{\partial\vec{\mathcal{P}}_{NL}(\vec{r}', \omega; u, v)}{\partial z'}\bigg|_{z'=0} = \vec{V}(u, v; \omega)e^{i(ux' + vy')}, \quad \text{(C3)}$$

where $\vec{r}' = x', y', z'$ and

$$\vec{U}(u, v; \omega) = \frac{1}{4\pi}\sum_{j=1}^{4}\left(\frac{\vec{k}_j^2}{k_0^2} - \epsilon_0\right)\vec{A}_j(u, v; \omega) - \frac{\epsilon_0}{4\pi}\sum_{j=1}^{2}\vec{A}_j'(u, v; \omega), \quad \text{(C4)}$$

$$\vec{V}(u, v; \omega) = \frac{1}{4\pi}\sum_{j=1}^{4}(iw_j)\left(\frac{\vec{k}_j^2}{k_0^2} - \epsilon_0\right)\vec{A}_j(u, v; \omega) - \frac{\epsilon_0}{4\pi}\sum_{j=1}^{2}(iw_j')\vec{A}_j'(u, v; \omega). \quad \text{(C5)}$$

Now, we have, from Eq. (5.2), upon formally interchanging the order of integration with respect to u, v and the limiting procedure $z' \to 0^+$, and then substituting from (C2) and (C3),

$$\vec{P}_{NL}(\vec{r}', \omega)\big|_{z'=0} = \int\int_{-\infty}^{+\infty}\vec{U}(u, v; \omega)e^{i(ux' + vy')}\,du\,dv, \quad \text{(C6)}$$

$$\frac{\partial\vec{P}_{NL}(\vec{r}', \omega)}{\partial z'}\bigg|_{z'=0} = \int\int_{-\infty}^{+\infty}\vec{V}(u, v; \omega) \times e^{i(ux' + vy')}\,du\,dv. \quad \text{(C7)}$$

We note that (C6) implies that the boundary value $\vec{P}_{NL}(\vec{r}', \omega)|_{z'=0}$ and $\vec{U}(u, v, \omega)$ form a two-dimensional Fourier transform pair, and (C7) implies that the boundary value $(\partial\vec{P}_{NL}(\vec{r}', \omega)/\partial z')|_{z'=0}$ and $\vec{V}(u, v; \omega)$ also form such a pair.

Next we make use of the representation (3.3) of the Green's function $G_\mu(x - x', y - y', z - z') \equiv G_\mu(|\vec{r} - \vec{r}'|)$ with the special choice $\vec{r} = x, y, z$, $\vec{r}' = 0, 0, z'$, with $0 < z < d$, $0 < z' < d$, d being, as before, the thickness of the slab:

$$G_\mu(x, y, z - z') = \frac{i}{2\pi}\int\int_{-\infty}^{+\infty}\frac{1}{w_\mu} \times e^{i(ux + vy + w_\mu|z - z'|)}\,du\,dv, \quad \text{(C8)}$$

with

$$w_\mu = (\mu^2 - u^2 - v^2)^{1/2}, \quad \text{(C9)}$$

$\text{Re}\,w_\mu > 0$, $\text{Im}\,w_\mu > 0$. If we differentiate (C8) with respect to z' and interchange the orders of differentiation and integration we obtain

$$\frac{\partial G_\mu(x, y, z - z')}{\partial z'} = \mp\frac{1}{2\pi}\int\int_{-\infty}^{+\infty} \times e^{i(ux + vy + w_\mu|z - z'|)}\,du\,dv, \quad \text{(C10)}$$

where the upper or lower sign is taken on the right-hand side according as $z > z'$ or $z < z'$. Next, we proceed in (C8) and (C10) to the limit $z' \to +0$. If we again interchange the orders of the two limiting processes ($z' \to 0^+$ and integration) we obtain the formulas

$$G_\mu(x, y, z-z')|_{z'=0} = \iint_{-\infty}^{+\infty} g_\mu(u, v; z) \times e^{i(ux+vy)}\,du\,dv, \tag{C11}$$

$$\left.\frac{\partial G_\mu(x, y, z-z')}{\partial z'}\right|_{z'=0} = \int\int_{-\infty}^{+\infty} h_\mu(u, v, z) \times e^{i(ux+vy)}\,du\,dv, \tag{C12}$$

where

$$g_\mu(u, v; z) = \frac{i}{2\pi}\frac{1}{w_\mu} e^{iw_\mu|z|}, \tag{C13}$$

$$h_\mu(u, v; z) = \frac{1}{2\pi} e^{iw_\mu|z|}. \tag{C14}$$

Equation (C11) shows that $G_\mu(x, y, z-z')|_{z'=0}$ and $g_\mu(u, v, z)$ form a two-dimensional Fourier transform pair and (C12) implies that $\partial G_\mu(x, y, z-z')/\partial z')|_{z'=0}$ and $h_\mu(u, v, z)$ form also such a pair.

Next, let us multiply (C1) by $e^{-i(ux+vy)}$ and use Eqs. (C2) and (C3). We then obtain the relation

$$\vec{U}(u, v; \omega) + \frac{1}{iw_\mu}\vec{V}(u, v; \omega) = 0. \tag{C15}$$

If we multiply (C15) by $(1/2\pi)e^{iw_\mu z}$ and use (C13) and (C14) we find that

$$\vec{U}(u, v; \omega)h_\mu(u, v; z) - \vec{V}(u, v; \omega)g_\mu(u, v; z) = 0. \tag{C16}$$

Let us now take the two-dimensional Fourier transform of (C16), with respect to the variables u and v. If we recall the Fourier transform relations (C6), (C7) and (C11), (C12) and use the convolution theorem on Fourier transforms we obtain the identity

$$\int\int_{-\infty}^{+\infty}\left(\vec{P}_{NL}(\vec{r}', \omega)\frac{\partial G_\mu(|\vec{r}-\vec{r}'|)}{\partial z'} - G_\mu(|\vec{r}-\vec{r}'|)\frac{\partial \vec{P}_{NL}(\vec{r}', \omega)}{\partial z'}\right)\Bigg|_{z'=0} dx'\,dy' = 0, \tag{C17}$$

where we have again used the shortened notation $G_\mu(|\vec{r}-\vec{r}'|)$ for $G_\mu(x-x', y-y', z-z')$. The identity (C17), which is, of course, valid for every point $\vec{r}$ within the slab, is the required formula (5.12a) of the text.

If instead of starting from Eq. (5.10a) we start from Eq. (5.10b), we would obtain, in a strictly similar manner, the formula (5.12b).

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[1](a) S. I. Pekar, Zh. Eksp. Teor. Fiz. 33, 1022 (1957) [Sov. Phys.–JETP 6, 785 (1958)]. (b) S. I. Pekar, Zh. Eksp. Teor. Fiz. 34, 1176 (1958) [Sov. Phys.–JETP 7, 813 (1958)].

[2]See, for example, J. J. Hopfield and D. G. Thomas, Phys. Rev. 132, 563 (1963), and the papers quoted therein. See also D. D. Sell, R. Dingle, S. E. Strokowski, and J. V. DiLorenzo, Phys. Rev. Lett. 27, 1644 (1971).

[3]See, for example, E. Burstein, in *Dynamical Processes in Solid State Optics*, edited by R. Kubo and H. Kanimura (Benjamin, New York, 1967), p. 1, or F. Wooten, *Optical Properties of Solids* (Academic, New York, 1972), Chap. III.

[4]See, for example, V. M. Agranovich and V. L. Ginzburg, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1971), Vol. 9, p. 252.

[5]V. M. Agranovich and V. L. Ginzburg, *Spatial Dispersion in Crystal Optics and The Theory of Excitons* (Interscience, New York, 1966).

[6]A. A. Rukhadze and V. P. Silin, Usp. Fiz. Nauk 74, 223 (1961) [Sov. Phys. Uspekhi 4, 459 (1961)].

[7]If the medium is spatially dispersive but macroscopically inhomogeneous, the constitutive relation (1.6) must be generalized to read

$$\hat{\vec{D}}(\vec{r}, \omega) = \int \hat{\epsilon}(\vec{r}, \vec{r}'; \omega)\hat{\vec{E}}(\vec{r}', \omega)\,d^3\vec{r}'.$$

[8]It is known [cf. Ref. 6, p. 463, Eq. (2.11)] that, as a consequence of spatial dispersion, a medium that would normally be considered to be isotropic must, in general, be characterized by a tensorial rather than a scalar dielectric response function. However, as we are interested in the present paper only in phenomena which depend in an essential way on the nonlocal nature of the response function, we will ignore its tensorial character.

[9]See, for example, the review article by V. M. Agranovich and V. L. Ginzburg, Usp. Fiz. Nauk 77, 663 (1962) [Sov. Phys. Uspekhi, 5, 675 (1963)].

[10]See L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, London, 1960), p. 339, Eq. (83.5).

[11]For example: (a) S. I. Pekar, Zh. Eksp. Teor. Fiz. 36, 451 (1959) [Sov. Phys.–JETP 9, 314 (1959)]; Zh. Eksp. Teor. Fiz. 38, 1786 (1960) [Sov. Phys.–JETP 11, 1286 (1960)]; see, in particular, Eq. (7), p. 1288. (b) J. J. Hopfield, Phys. Rev. 112, 1555 (1958). (c) Ref. 9, p. 698, Sec. 4. (d) J. J. Hopfield and D. G. Thomas, Phys. Rev. 132, 563 (1963). (e) J. J. Hopfield and D. G. Thomas, Phys. Rev. 122, 35 (1961). (f) A. S. Davydov and V. A. Onishchuk, Dokl. Akad. Nauk SSR 178, 1285 (1968) [Sov. Phys. Doklady 13, 139 (1968)].

[12]See Ref. 5, Chap. 4.

[13]See, for example, Refs. 5 and 11d and (a) V. L. Ginzburg, Zh. Eksp. Teor. Fiz. 34, 1593 (1958) [Sov. Phys.–JETP 7, 1096 (1958)]. (b) S. I. Pekar, Fiz. Tverd. Tela 4, 1301 (1962) [Sov. Phys.–Solid State 4, 953 (1962)]. (c) T. Skettrup and I. Balslev, Phys. Rev. B 3, 1457 (1971). (d) V. S. Mashkevich, Zh. Eksp. Teor. Fiz. 40, 1803 (1961) [Sov. Phys. JETP 13, 1267 (1961)].

[14]A preliminary account of this investigation was given by G. S. Agarwal, D. N. Pattanayak, and E. Wolf [(a) Phys. Rev. Lett. 27, 1022 (1971); (b) Optics Commun. 4, 255 (1971); (c) Optics Commun. 4, 260 (1971); (d) Phys. Lett. 40A, 279 (1972)].

[15]A. A. Maradudin and D. L. Mills, Phys. Rev. B 7, 2787 (1973). We are grateful to Dr. D. L. Mills for

sending us a preprint of this article before publication.

[16]J. J. Sein, (a) Ph.D. dissertation (New York University, 1969) (unpublished); (b) Phys. Lett. 32A, 141 (1970); (c) J. Opt. Soc. Am. 62, 1037 (1972).

[17]J. L. Birman and J. J. Sein, Phys. Rev. B 6, 2482 (1972).

[18]The neglect of surface corrections to the form of the constitutive relations underlies practically all macroscopic theories of the electromagnetic field in material media, whether or not the media are spatially dispersive. For spatially dispersive media such neglect is implicit—for example, in the theories of Sein (Refs. 16b and 16c) and Birman and Sein (Ref. 17), Maradudin and Mills (Ref. 15), and Agranovich and Ginzburg (Ref. 5). A brief justification for this procedure is given on p. 2792 of Ref. 15. The same kind of approximation has also been made in recent work on surface plasmons in metals [e.g., G. A. Baraff, Phys. Rev. B 7, 580 (1973); J. Heinrichs, Phys. Rev. B 7, 3487 (1973)]. It seems very likely that even if a more accurate form of the constitutive relations that includes surface correction effects should become available, the solution of the appropriate electrodynamic equations would have to be based on some perturbation techniques, to which our solution would provide a natural first approximation. An analogy with the role of the Born approximation in scattering theory comes naturally to mind here.

[19](a) For a brief discussion of the angular spectrum representation of wave fields, see, for example, C. J. Bouwkamp, Rept. Progr. Phys. 17, 41 (1954); J. R. Shewell and E. Wolf, J. Opt. Soc. Am. 58, 1596 (1968); J. W. Goodman, *Introduction to Fourier Optics* (McGraw-Hill, New York, 1968), sec. 3.7. (b) H. Weyl, Ann. Phys. 60, 481 (1919); see also A. Banos, *Dipole Radiation in the Presence of a Conducting Half-space* (Pergamon, Oxford, 1966), Eq. (2.19).

[20]R. Courant and D. Hilbert, *Methods of Mathematical Physics* (Interscience, New York, 1962), Vol. II, Sec. 2, p. 14.

[21]The very special circumstances under which Eq. (3.23) may not be satisfied are noted in Sec. (B) of Appendix A. (In this connection see also Ref. 5, Sec. 2.3, and some of the references quoted therein.)

[22]In carrying out this calculation it is useful to employ the following identity, valid for any complex number $z = x + iy$ (x, y real):

$$\mathrm{Im}(z^{1/2}) = \pm (1/\sqrt{2})(r - x)^{1/2},$$

where $r = (x^2 + y^2)^{1/2}$.

[23]G. S. Agarwal, Optics Commun. 6, 221 (1972); Phys. Rev. B 8, 4768 (1973).

[24]The two w roots of the transverse dispersion relations, denoted here as w_1 and w_2, are not necessarily the roots labeled by subscripts 1 and 2 in Eqs. (3.47). w_1 and w_2 denote now those two roots of the set (3.47), which have positive real parts.

[25]The parameters $\alpha_t^{(j)}$ and α_l should not be confused with the quantity denoted by α (related to the polarizability) which appears in the expressions for the dielectric constant in Sec. I.

[26]We assume here, of course, that the determinant associated with this set of linear algebraic equations does not vanish.

[27]After a straightforward algebraic manipulation, the formulas (4.42) and (4.49) for the amplitude of the reflected waves may be expressed in the alternative forms

$$(\vec{A}_r)_\perp/(\vec{A}_0)_\perp = (q_\perp \cos\theta_0 + 1)/(q_\perp \cos\theta_0 - 1), \quad (4.42a)$$

$$(\vec{A}_r)_\parallel/(\vec{A}_0)_\parallel = (\cos\theta_0 - q_\parallel)/(\cos\theta_0 + q_\parallel), \quad (4.49a)$$

where

$$q_\perp = k_0/(w_\mu - w_1 - w_2),$$

$$q_\parallel = k_0\{-\omega_\mu + [k_\parallel^4 + k_\parallel^2(w_1^2 + w_2^2 + w_1 w_2) + w_1 w_2 w_l (w_1 + w_2)] \cdot [k_\parallel^2 (w_1 + w_2 - w_l)' + w_1 w_2 w_l]\}^{-1},$$

and

$$k_\parallel^2 = u^2 + v^2.$$

The quantities $q_\perp$ and $q_\parallel$ are the so-called surface impedances [cf. K. L. Kliewer and R. Fuchs, Phys. Rev. 172, 607 (1968)] and are given in terms of the field amplitudes by

$$q_\perp = E_y(0^-)/H_x(0^-) = E_y(0^+)/H_x(0^+),$$

$$q_\parallel = E_x(0^-)/H_y(0^-) = E_x(0^+)/H_y(0^+),$$

where the superscripts + and − refer to the limits taken from the two sides of the boundary ($z \to \pm 0$). Equation (4.42a) is identical (except for notation) with the corresponding formula [Eq. (3.24)] of Ref. 15. However, Eq. (4.49a) differs from the corresponding formula (3.36a) of that reference, although the two formulas are of the same general form. The discrepancy is probably due to an algebraic error made in the derivation of Eq. (3.36a) of Ref. 15, since that formula is dimensionally inconsistent.

[28]See, for example, M. Born and E. Wolf, *Principles of Optics* (Pergamon, Oxford, 1970), 4th ed., Sec. 1.5,

[29]These are actually the values of one of the components of the dielectric tensor of cadmium sulphide (CdS), but our results do not apply to CdS, except at normal incidence. Our assumption of a scalar dielectric constant excludes (for arbitrary angle of incidence) anisotropic media such as CdS.

[30]There is an error in the labeling of the curves in the corresponding figure in Ref. 14(b). The curves in Fig. 2 of that reference represent the reflectivities $|(\vec{e}_R)_\perp/(\vec{e}_0)_\perp|^2$ and $|(\vec{e}_R)_\parallel/(\vec{e}_0)_\parallel|^2$ and not the amplitude ratios $|(\vec{e}_R)_\perp/(\vec{e}_0)_\perp|$ and $|(\vec{e}_R)_\parallel/(\vec{e}_0)_\parallel|$. We are indebted to Dr. W. H. Carter for having drawn our attention to this error.

[31]The longitudinal frequency ω_l is defined by the usual expression

$$\omega_l = \omega_e[1 + (4\pi\alpha/\epsilon_0)]^{1/2}.$$

It is the root of the equation $\tilde{\epsilon}(0, \omega) = 0$, when damping is neglected. In view of the relation $\vec{k}_l^2 = (\mu^2/\epsilon_0)\tilde{\epsilon}(0, \omega)$ one has (if one again ignores damping) $\vec{k}_l^2 = 0$ when $\omega = \omega_l$.

[32]This is Eq. (22) of Ref. 1(b), written in our notation. The relation between "additional boundary conditions" and mode-coupling conditions is discussed in Secs. V and VI below.

[33]Reflection at normal incidence with Pekar's boundary conditions was studied in great detail both experimentally and theoretically by Hopfield and Thomas in Ref. 11d. Recently H. Grossman, J. Bielmann, and S. Nikitine [in Proceedings of the Taormina Conference on Polari-

tons, edited by E. Burstein (Pergamon, Oxford, to be published)] also described results of experiments at normal incidence and analyzed them on the basis of both Pekar's theory and ours.

[34]Even though $n^* \neq n_p^*$, in general, both these quantities have the same asymptotic limit (appropriate to a spatially nondispersive medium) as the effective exciton mass $m_e^* \to \infty$. To see this we note that the parameters χ and $\mu(\omega)$, defined by Eq. (1.24), are each proportional to m_e^*. We set

$$\chi = \chi_0 m_e^*, \quad \mu^2(\omega) = \mu_0^2(\omega)\, m_e^*,$$

where

$$\chi_0 = 4\pi\alpha\omega_e/\hbar, \quad \mu_0^2(\omega) = (1/\hbar\omega_e)(\omega^2 - \omega_e^2 + i\omega\Gamma).$$

From Eq. (4.30) we readily deduce that for large enough m_e^*

$$n_t^{(j)2} = \tfrac{1}{2}[\epsilon_0 + (\mu_0^2 m_e^*/k_0^2)] \pm \tfrac{1}{2}[\epsilon_0 - (\mu_0^2 m_e^*/k_0^2)]$$
$$\pm [(\chi_0 m_e^*/k_0^2)/(\epsilon_0 - \mu_0^2 m_e^*/k_0^2)] \pm O(1/m_e^*).$$

Using this formula in Eqs. (4.58) and (4.65) we find that as $m_e^* \to \infty$

$$n^* = n + O(1/\sqrt{m_e^*}), \quad n_p^* = n + O(1/\sqrt{m_e^*}),$$

where

$$n^2 = \epsilon_0 - (\chi_0/\mu_0^2).$$

On substituting for χ_0 and μ_0^2 the expression for n^2 becomes

$$n^2 = \epsilon_0 + 4\pi\alpha\omega_e^2/(\omega_e^2 - \omega^2 - i\omega\Gamma).$$

Comparison of this formula with Eq. (1.5) shows that this limiting value is precisely the value of the square of the refractive index ($n^2 = \epsilon$) for frequencies in the vicinity of an isolated resonance in a spatially nondispersive medium.

[35]See, for example, P. M. Morse and H. Feshbach, *Methods of Theoretical Physics* (New York, McGraw-Hill, 1953), Sec. 7.5.

[36]This generalized extinction theorem was first reported in Ref. 14(d). The corresponding equation in that reference [Eq. (9)] contains a misprint: The equal sign and the zero on the right-hand side were omitted. Also the subscript e on ω_e in the factor $(\hbar\omega_e/m_e^*)k^2$ in the denominator of Eq. (1) of that reference was inadvertently lost. Our generalized extinction theorem does not cover the special case when $\mathcal{L}$ is a linear differential operator which contains only first-order derivatives (as arises in the theory of optical activity). A different procedure has to be employed in this case.

[37]It is briefly discussed by D. N. Pattanayak, in *Proceedings of the Third Rochester Conference on Coherence and Quantum Optics*, edited by L. Mandel and E. Wolf (Plenum, New York, 1973), p. 359.

[38]G. D. Mahan and J. J. Hopfield, Phys. Rev. A 135, 428 (1964).

[39]Equation (5.36) corresponds to Eq. (3.27) of Ref. 38 when the Fourier transform of Eq. (3.4) of that reference is also used.

[40]S. I. Pekar: These are the papers referred to in Refs. 1, 11, and 13. See also G. Mahan and G. Obermair, Phys. Rev. 183, 834 (1969).

[41]V. I. Sugakov, (a) Fiz. Tverd. Tela 5, 2207 (1963) [Sov. Phys.–Solid State 5, 1607 (1964)]; (b) Fiz. Tverd. Tela 5, 2682 (1963) [Sov. Phys.–Solid State 5, 1959 (1964)].

[42]W. Brenig, R. Zeyher, and J. L. Birman, Phys. Rev. B 6, 4613 (1972).

[43]V. M. Agranovich and V. I. Yudson, Optics Commun. 7, 121 (1973).