117 (1969); Ref. 11.
[28]R. H. Parmenter, Phys. Rev. 86, 552 (1952).
[29]J. C. Slater, *Handbuch der Physik*, edited by S. Flügge (Springer, Berlin, 1956), p. 27.
[30]J. M. Ziman, *Principles of the Theory of Solids* (Cambridge U. P., Cambridge, England, 1965), p. 84.

PHYSICAL REVIEW B VOLUME 6, NUMBER 6 15 SEPTEMBER 1972

Optics of Polaritons in Bounded Media

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(Received 13 July 1970; revised manuscript received 13 December 1971)

Using a simple polarization picture of a medium in which there is spatial dispersion and using the integral equation method of Ewald–Oseen, some rigorous results are obtained for the optics of a bounded spatially dispersive medium. In particular, an extinction theorem for polaritons, and rigorous additional boundary conditions are obtained. The normal-incidence propagation of transverse modes for an isotropic crystal bounded by a plane is analyzed. The rigorous boundary conditions obtained by this method, in general, do not agree with other popular choices, based upon the same constitutive relations.

I. INTRODUCTION

The purpose of this paper is to give a simple derivation of results needed for an analysis of wave propagation in a bounded medium in which there is a nonlocal constitutive equation. Physically the nonlocality can arise from an exciton absorption lying close to the frequency of the propagating wave. This produces a wave-vector-dependent dielectric function and leads to propagation of the mixed exciton-photon modes known as polaritons.[1–3]

When an incident plane wave impinges upon such a medium from vacuum, several polaritons can be excited in the medium. In a particular geometry these propagate parallel to the incident wave, but with different phase velocities. The total field consists of incident plus reflected fields in vacuum, plus the polariton fields in the medium. To determine the amplitudes of all fields, we need a complete set of boundary conditions. In addition the polariton dispersion relation for waves propagating in the medium is required. Finally, in order that the incident wave shall not propagate in the medium, we require an extinction condition.

The method we use to solve this problem is based on the Ewald-Oseen integral equation formulation of optics, plus a polarization picture of the spatially dispersive medium.[4] In this framework the results can be derived in a particularly transparent fashion, and a comparison with the treatments of the usual "local" optics can be made.

The extinction theorem, and the boundary conditions were first obtained by one of us[5] using a somewhat different method. That derivation along with detailed numerical analysis and comparison of calculated and experimental reflectivity will be published separately. The calculated reflectivity is in satisfactory agreement with experiment.

The results of our analysis have been used in recent theoretical work on Raman scattering[6] in the polariton picture. There, a quantum-mechanical treatment of polariton scattering inside the crystal, plus the polariton reflectivity at the crystal boundary was needed in order to compute cross sections for Raman scattering.

Besides being relatively simple and familiar, the method we use to analyze wave propagation is rigorous. It does not require, for example, that the surface boundary conditions be assumed, but yields them as a result of the theory. We return to this point later.

II. SPATIAL DISPERSION

Some field vectors needed in our work are $\vec{E}(\vec{r}, t)$ the *macroscopic* electric field and $\vec{P}(\vec{r}, t)$ the dielectric polarization at $(\vec{r}, t)$. The time and space Fourier transforms are defined as

$$\vec{P}(\vec{r}, \omega) = (1/(2\pi)^{1/2}) \int \vec{P}(\vec{r}, t)\, e^{-i\omega t}\, dt \tag{2.1}$$

and

$$\vec{P}(\vec{k}, \omega) = (1/(2\pi)^{3/2}) \int \vec{P}(\vec{r}, \omega)\, e^{-i\vec{k}\cdot\vec{r}}\, d\vec{r}\,. \tag{2.2}$$

The inverse of (2.2) is

$$\vec{P}(\vec{r}, \omega) = (1/(2\pi)^{3/2}) \int \vec{P}(\vec{k}, \omega) e^{i\vec{k}\cdot\vec{r}} d\vec{k} . \qquad (2.3)$$

Similar definitions apply to other field vectors.

The constitutive relation which characterizes a spatially dispersive medium is

$$\vec{P}(\vec{k}, \omega) = \chi(\vec{k}, \omega) \vec{E}(\vec{k}, \omega) , \qquad (2.4)$$

with $\chi(\vec{k}, \omega)$ the Fourier transform of the macroscopic susceptibility. In writing (2.4) we assume a nontrivial wave-vector dependence. Nonlocality is immediately apparent when (2.4) is Fourier transformed. We have

$$\vec{P}(\vec{r}, \omega) = (1/(2\pi)^{3/2}) \int \chi(\vec{r}' - \vec{r}, \omega) \vec{E}(\vec{r}', \omega) d\vec{r}' , \qquad (2.5)$$

with

$$\chi(\vec{r}, \omega) = (1/(2\pi)^{3/2}) \int \chi(\vec{k}, \omega) e^{i\vec{k}\cdot\vec{r}} d\vec{k} . \qquad (2.6)$$

Then (2.5) proves that the polarization at $\vec{r}$ in the medium is determined in a nonlocal fashion by the electric field.

To be specific and useful, we shall take a particular form for $\chi(\vec{k}, \omega)$ which is appropriate to the spatial-dispersion effects associated with incident frequency ω close to an exciton-absorption band.[7] Let

$$\chi(\vec{k}, \omega) = \frac{\epsilon_0 - 1}{4\pi} + \frac{F}{\omega_0^2 - \omega^2 - i\omega\Gamma + Bk^2} , \qquad (2.7)$$

where ϵ_0 is some effective background dielectric coefficient; Γ a real positive damping constant; ω_0 is the exciton resonance frequency; $B \equiv (\hbar\omega_0/m^*)$, where m^* is the exciton effective mass; and $F = \alpha_0 \omega_0^2$, where α_0 is an oscillator strength.

Carrying out the Fourier transformation [Eq. (2.6)], we find the kernel function $\chi(\vec{r}, \omega)$ needed in (2.5). The details are elementary and are given in Appendix A. Thus we have

$$\chi(\vec{r}, \omega) = \chi_0 \delta(\vec{r}) + \chi_1 G_+(\vec{r}, \omega) , \qquad (2.8)$$

where

$$\chi_0 \equiv [(\epsilon_0 - 1)/4\pi](2\pi)^{3/2}, \qquad (2.9)$$

$$(2\pi)^3 \delta(\vec{r}) = \int e^{i\vec{k}\cdot\vec{r}} d\vec{k} , \qquad (2.10)$$

$$\chi_1 \equiv \pi F/B(2\pi)^{1/2} , \qquad (2.11)$$

$$G_+(\vec{r}, \omega) = e^{ik_+ r}/r \equiv G_+(r) . \qquad (2.12)$$

For brevity we may suppress the ω in the argument as in (2.12). The quantity k_+ is a complex wave number defined as follows. Write the second term in (2.7) as

$$\frac{F}{\omega_0^2 - \omega^2 - i\omega\Gamma + Bk^2} \equiv \frac{F}{C(\omega) + Bk^2} . \qquad (2.13)$$

Then this term has simple poles at complex wave numbers given by

$$k_\pm = (|C(\omega)|/B)^{1/2} \exp\{i[\tfrac{1}{2}(\theta \pm \pi)]\} , \qquad (2.14)$$

where

$$C(\omega) \equiv \omega_0^2 - \omega^2 - i\omega\Gamma = |C| e^{i\theta} , \qquad (2.15)$$

$$|C| = [(\omega_0^2 - \omega^2) + \omega^2\Gamma^2]^{1/2} , \qquad (2.16)$$

$$\tan\theta = -\omega\Gamma/(\omega_0^2 - \omega^2) , \qquad (2.17)$$

with

$$-\pi \leqslant \theta \leqslant 0 . \qquad (2.18)$$

The range (2.18) is chosen so that θ will be a continuous function of ω as $0 \leqslant \omega < \infty$. We may write the real and imaginary parts of k_+ as

$$k_+ \equiv i\delta + \epsilon$$

and then

$$\delta = (|C|/B)^{1/2} \cos\tfrac{1}{2}\theta \geqslant 0 , \qquad (2.19)$$

$$\epsilon = -(|C|/B)^{1/2} \sin\tfrac{1}{2}\theta \geqslant 0 . \qquad (2.20)$$

The important inequalities on the right-hand side of (2.19) and (2.20) follow (2.18). It is also very useful to write the susceptibility [Eq. (2.7)] in the form

$$\chi(k, \omega) = \frac{1}{(2\pi)^{3/2}} \left(\chi_0 + \frac{4\pi\chi_1}{k^2 - k_+^2} \right) . \qquad (2.21)$$

From (2.8) it is clearly seen that the second term $\chi_1 G_+(r, \omega)$ is responsible for nonlocal behavior. Notice the form of $G_+(r)$ from (2.12). The range of nonlocality is δ^{-1}, with δ defined in (2.19). Clearly δ depends upon ω. For example, if $\theta = 0$ for all ω and $\Gamma = 0$, then

$$\delta^{-1} = (B/|\omega_0^2 - \omega^2|)^{1/2} ,$$

so that $\delta^{-1} \to \infty$ as $\omega \to \omega_0$. Thus at resonance, neglecting damping, the polarization at any point is determined by the field at all points in the medium; this is the maximum possible nonlocality. Still taking $\Gamma = 0$, when ω is very far from resonance so $|\omega_0^2 - \omega^2| \gg 0$, then $\delta^{-1} \to 0$ and the theory becomes local ($G_+ \to 0$). For $B = 0$, or infinite exciton mass, a local theory results for all frequencies. Nonlocality is thus seen to depend on the finite exciton mass and the frequency ω.

Although we only concern ourselves here with a susceptibility (2.7) corresponding to a single resonance, multiple resonances can be simply incorporated into the work. A sum of terms such as $\chi_1 G_+$ will then arise in the generalization of (2.8). Similarly, tensorial effects can be incorporated if needed.

The function $G_+(r, \omega)$ is the Green function of a Helmholtz equation[8]:

$$\nabla^2 G_+(\vec{r} - \vec{r}') + k_+^2 G_+(\vec{r} - \vec{r}') = -4\pi\delta(\vec{r} - \vec{r}') . \qquad (2.22)$$

In our case k_+ is given in (2.14) and

$$k_+^2 = -C(\omega)/B . \qquad (2.23)$$

In the sequel we always take the constitutive equation in the medium to be (2.4), with (2.7) defining the susceptibility: This will include the case of a bounded crystal. We assume also that for all cases the nonlocal relationship (2.8) applies in the medium, including points right up to the surface.

III. THE INTEGRAL EQUATION: POLARIZATION FRAMEWORK

The treatment of local optics by the integral equation method is discussed in many places.[4,9] We give a brief review here to establish notation and to bring out new points applicable to spatial dispersion.

The polarization $\vec{P}$ of our medium can be assumed to originate in some pseudo-oscillators which represent the polaritons. This polarization is assumed to be the source of the electric field. Then if $\vec{E}_L(\vec{r}, t)$ is the local field and $\vec{E}^{(i)}(\vec{r}, t)$ is the incident field [at $(\vec{r}, t)$], then

$$\vec{E}_L(\vec{r}, t) = \vec{E}^{(i)}(\vec{r}, t) + \int_{-\sigma(\vec{r})}^{\Sigma} dV' \, \vec{\nabla}\times\vec{\nabla}\times\left(\frac{\vec{P}(\vec{r}\,', t-R/c)}{R}\right) . \tag{3.1}$$

In (3.1) we omit a small sphere $\sigma(\vec{r})$ centered at the field point $\vec{r}$. The integral is taken over the interior of the crystal, bounded by the surface Σ, and omitting $\sigma(\vec{r})$. Also $R = |\vec{r} - \vec{r}\,'|$. Taking all fields time harmonic as $e^{i\omega t}$, Eq. (3.1) becomes

$$\vec{E}_L(\vec{r}, \omega) = \vec{E}^{(i)}(\vec{r}, \omega) + \int_\sigma^\Sigma dV' \, \vec{\nabla}\times\vec{\nabla}\times\vec{P}(\vec{r}\,', \omega)\, G_0(R) , \tag{3.2}$$

with

$$G_0(R) \equiv e^{ik_0 R}/R , \tag{3.3}$$

where

$$k_0 \equiv \omega/c . \tag{3.4}$$

Since $G_0(\vec{r} - \vec{r}\,')$ satisfies the wave equation

$$\nabla^2 G_0(\vec{r} - \vec{r}\,') + k_0^2 G_0(\vec{r} - \vec{r}\,') = -4\pi\delta(\vec{r} - \vec{r}\,') , \tag{3.5}$$

we may take the operator $\vec{\nabla}\times\vec{\nabla}\times$ ouside the integral to obtain

$$\vec{E}_L(\vec{r}, \omega) + \tfrac{8}{3}\pi \vec{P}(\vec{r}, \omega) = \vec{E}^{(i)}(\vec{r}, \omega) + \vec{\nabla}\times\vec{\nabla}\times \int_\sigma^\Sigma \vec{P}(\vec{r}\,', \omega)\, G_0(R)\, dV' . \tag{3.6}$$

To proceed we need to employ the constitutive relation (2.5). But this requires that we eliminate the local field $\vec{E}_L$ appearing in (3.6) in favor of the macroscopic field $\vec{E}$. We assume that the Lorentz–Lorenz expression applies

$$\vec{E}_L(\vec{r}, \omega) = \vec{E}(\vec{r}, \omega) + \tfrac{4}{3}\pi \vec{P}(\vec{r}, \omega) . \tag{3.7}$$

Substituting this in (3.6) gives

$$\vec{E}(\vec{r}, \omega) + 4\pi \vec{P}(\vec{r}, \omega) = \vec{E}^{(i)}(\vec{r}, \omega) + \vec{\nabla}\times\vec{\nabla}\times \int_{\sigma(\vec{r})}^\Sigma \vec{P}(\vec{r}\,', \omega)\, G_0(R)\, dV' , \tag{3.8}$$

which is the familiar equation. An *a posteriori* plausibility argument for the use of the Lorentz–Lorenz expression for the present situation of a polarization produced by exciton polaritons will be given below. But it would be of interest to study this question by microscopic theory.

Now we multiply both sides of (3.8) by $(2\pi)^{-3/2} \times\chi(\vec{r}\,'' - \vec{r})$ and integrate the variable $\vec{r}$ over the medium. When the expression (2.8) is substituted for the susceptibility, and (2.5) is employed to recognize the polarization, we obtain the basic integrodifferential equation

$$\begin{aligned}\vec{P}(\vec{r}\,'', \omega) &+ 4\pi \frac{\chi_0}{(2\pi)^{3/2}} \vec{P}(\vec{r}\,'', \omega) + 4\pi \frac{\chi_1}{(2\pi)^{3/2}} \int^\Sigma \vec{P}(\vec{r}, \omega)\, G_+(\vec{r}\,'' - \vec{r})\, d\vec{r} \\ &= \frac{\chi_0}{(2\pi)^{3/2}} \vec{E}^{(i)}(\vec{r}\,'', \omega) + \frac{\chi_1}{(2\pi)^{3/2}} \int^\Sigma G_+(\vec{r}\,'' - \vec{r})\, E^{(i)}(\vec{r}, \omega)\, d\vec{r} + \frac{\chi_0}{(2\pi)^{3/2}} \vec{\nabla}\times\vec{\nabla}\times \int_\sigma^\Sigma \vec{P}(\vec{r}\,', \omega)\, G_0(\vec{r}\,' - \vec{r}\,'')\, d\vec{r}\,' \\ &\quad + \frac{\chi_1}{(2\pi)^{3/2}} \int^\Sigma G_+(\vec{r}\,'' - \vec{r})\, \vec{\nabla}\times\vec{\nabla}\times' \int_\sigma^\Sigma \vec{P}(\vec{r}\,', \omega)\, G_0(\vec{r}\,' - \vec{r})\, d\vec{r}\, d\vec{r}\,' .\end{aligned} \tag{3.9}$$

Local optics[4,9] is recovered if χ_1 is set equal to zero.

IV. CONDITION FOR SOLUTION OF INTEGRAL EQUATION: TRANSVERSE WAVES

The integral equation (3.9) has both longitudinal and transverse solutions,[5] but in the interest of simplicity we shall treat only the case of transverse wave solutions here.

As an incident wave we take a wave $E^{(i)}$ satisfying the vacuum wave equation

$$\vec{\nabla}^2 \vec{E}^{(i)}(\vec{r}, \omega) + k_0^2 \vec{E}^{(i)}(\vec{r}, \omega) = 0 , \tag{4.1}$$

where k_0 is given in (3.4). In the medium we take the total polarization as the sum of two plane waves, each one satisfying a wave equation with undetermined wave vector. Thus we have

$$\vec{P}(\vec{r}, \omega) = \vec{P}_1(\vec{r}, \omega) + \vec{P}_2(\vec{r}, \omega) \tag{4.2}$$

and

$$\vec{\nabla}^2 \vec{P}_j + k_j^2 \vec{P}_j = 0\ , \quad j = 1, 2\ . \tag{4.3}$$

k_1 and k_2 will be determined by (3.9).

At this point the analysis, although straightforward, is somewhat untidy, and it is easy to lose the thread of the argument. Actually we follow in principle the kind of analysis used in local optics.[4] In effect, when (4.2) is substituted into (3.9), we find that (3.9) contains terms (fields) which propagate at four different velocities. That is, there are terms which satisfy the wave equation with four different wave vectors: k_1, k_2, k_0, and k_+. As these are independent, each term must vanish separately. The vanishing of these terms produces (i) the dispersion relation which determines k_1 and k_2, (ii) the extinction theorem which eliminates the incident wave in the medium, and (iii) the exact additional boundary conditions ("a.b.c.").

In order to obtain this result a certain number of manipulations of (3.9) are needed to transform the expressions from volume integrals to surface integrals, etc. These are not very different from the corresponding manipulations for local optics and are of limited physical interest. The details are given in Appendix B.

The result is that after (4.2) is substituted into (3.9), we obtain

$$\begin{aligned} 0 = -\sum_{j=1}^{2} \Bigg[\vec{P}_j \left(1 + 4\pi\chi(k_j, \omega) - 4\pi\chi(k_j, \omega) \frac{k_j^2}{k_j^2 - k_0^2} \right) \Bigg] \\ + \left(E^{(i)} + \sum_j \frac{A_0(\vec{P}_j)}{k_j^2 - k_0^2} \right) \chi(k_0, \omega) \\ + \Bigg[\chi_1 \left(\frac{2}{\pi} \right)^{1/2} \sum_j \frac{S_+(\vec{P}_j)}{k_j^2 - k_+^2} \left(\frac{k_j^2}{k_j^2 - k_0^2} - 1 \right) \\ + \frac{\chi_1}{(2\pi)^{3/2}(k_0^2 - k_+^2)} \left(S_+(\vec{E}^{(i)}) + \sum_j \frac{S_+(\vec{A}_0(\vec{P}_j))}{k_j^2 - k_0^2} \right) \Bigg]. \end{aligned} \tag{4.4}$$

All the objects in (4.4) have been previously defined with the exception of: the surface integral

$$\vec{S}_0(\vec{P}_j) = \int^{\Sigma} dS' \left(\vec{P}_j(\vec{r}', \omega) \frac{\partial G_0}{\partial n} (\vec{r}' - \vec{r}'') - G_0(\vec{r}' - \vec{r}'') \frac{\partial \vec{P}_j}{\partial n} (\vec{r}', \omega) \right), \tag{4.5}$$

the vector

$$\vec{A}_0(\vec{P}_j) = \vec{\nabla} \times \vec{\nabla} \times \vec{S}_0(\vec{P}_j) \tag{4.6}$$

and

$$\vec{S}_+(\vec{A}_0(\vec{P}_j)) = \int^{\Sigma} dS \left(\vec{A}_0(\vec{P}_j) \frac{\partial G_+}{\partial n} - G_+ \frac{\partial \vec{A}_0(\vec{P}_j)}{\partial n} \right), \tag{4.7}$$

and

$$\vec{S}_+(\vec{E}^{(i)}) = \int^{\Sigma} dS \left(\vec{E}^{(i)} \frac{\partial G_+}{\partial n} - G_+ \frac{\partial \vec{E}^{(i)}}{\partial n} \right). \tag{4.8}$$

Now we explore the consequences of the vanishing of (4.4). In (4.4) the object within the first set of square brackets satisfies the wave equation at k_j^2, within the second set of large parentheses at k_0^2, and within the last satisfies it at k_+^2.

V. DISPERSION EQUATION

The first set of square brackets in (4.4) contains terms at k_1^2 and k_2^2. Since, in general, $P_j \neq 0$, we have

$$1 + 4\pi\chi(k, \omega) - 4\pi\chi(k, \omega) k^2/(k^2 - k_0^2) = 0\ . \tag{5.1}$$

The solutions of this equation are the roots k_1 and k_2. The same equation evidently determines both, or in familiar form, the allowed wave vectors satisfy

$$(k_j/k_0)^2 = 1 + 4\pi\chi(k_j, \omega)\ . \tag{5.2}$$

Now if k_j solves (5.2), then for the electric field associated with the polarization $\vec{P}_j$, we have

$$\begin{aligned} \vec{E}(k_j, \omega) &= \chi^{-1}(k_j, \omega) \vec{P}(k_j, \omega) \\ &= \frac{4\pi k_0^2 \vec{P}_j(k_j, \omega)}{k_j^2 - k_0^2}\ . \end{aligned} \tag{5.3}$$

It is important to realize that each polarization $\vec{P}_j$ has its own associated electric field, and that to each k_j [solution of (5.2)] there is a susceptibility $\chi(k_j, \omega)$.

The dispersion (5.2) is a direct consequence of the use of the Lorentz–Lorenz local-field expression. The dispersion (5.2) is also the result, which one obtains directly from the well-known treatment using the Maxwell differential equations (see Appendix C). Thus the plausibility for using the Lorentz–Lorenz local-field correction is the agreement produced between the present integral equation result, and that of the usual treatment. To make the point clearer (if redundant), if one leaves the exact local-field correction to be determined[10] by requiring that the integral-equation method produces the same dispersion (5.2) as the usual differential-equation method, then one will obtain the Lorentz–Lorenz form. Although this is an *a posteriori* argument for the local-field correction used, a microscopic theory is of course to be preferred.

We have from (5.2) and (2.7) or (2.13) explicitly

$$\left(\frac{k}{k_0} \right)^2 = \epsilon_0 + \frac{4\pi F}{C(\omega) + Bk^2}\ . \tag{5.4}$$

The solutions of this equation are given in general as the roots k_1, k_2:

$$\left(\frac{k}{k_0} \right)^2 = \frac{1}{2} \left(\epsilon_0 - \frac{C(\omega)}{Bk_0^2} \right) \pm \frac{1}{2} \left[\left(\epsilon_0 + \frac{C(\omega)}{Bk_0^2} \right)^2 + \frac{16\pi F}{Bk_0^2} \right]^{1/2}. \tag{5.5}$$

Equation (5.5) determines the two phase velocities or complex refractive indices

$$n_1 = k_1/k_0 , \quad n_2 = k_2/k_0 \tag{5.6}$$

for the two propagating transverse waves (polaritons). In the region close to $\omega \approx \omega_0$, the solutions k_1 and k_2 of (5.5) differ appreciably from the solutions for the uncoupled exciton and photon fields. The form (5.5) has been used in various computations. For large ω, (5.5) can be approximately solved by expanding the radical, since in the second term

$$(16\pi F/Bk_0^2) \sim \omega^{-2} \to 0 . \tag{5.7}$$

Hence in this domain $\omega \gg \omega_0$, the roots are

$$n_1^2 \cong \epsilon_0 , \quad n_2^2 \sim -C(\omega)/Bk_0^2$$
or
$$n_1^2 \sim \epsilon_0 , \quad n_2 \sim k_+/k_0 . \tag{5.8}$$

In writing (5.8) recall the definition of k_+ as given in (2.14), or the equivalent statement [Eq. (2.23), where $k_+^2 = -C(\omega)/B$]. It is also useful at times to define a complex wave number by

$$\gamma \equiv k_+/k_0 , \tag{5.9}$$

so that for $\omega \gg \omega_0$, γ is a root of (5.5).

VI. EXTINCTION THEOREM FOR POLARITONS

The second set of large parentheses in (4.4) contains terms propagating at k_0. Setting it equal to zero

$$\vec{E}^{(i)} + \frac{\vec{A}_0(\vec{P}_1)}{k_1^2 - k_0^2} + \frac{\vec{A}_0(\vec{P}_2)}{k_2^2 - k_0^2} = 0 . \tag{6.1}$$

Thus, the incident wave is extinguished by an electric field composed of the sum of two fields. The sources of the latter are just the polariton polarizations. This is the generalization of the usual extinction theorem of local optics.[4]

VII. ADDITIONAL BOUNDARY CONDITIONS

The last set of square brackets in (4.4) contains terms propagating at k_+. This bracketed expression can be set equal to zero but may first be simplified using the extinction theorem (6.1). Then

$$S_+(\vec{E}^{(i)}) + \sum_j \frac{S_+(\vec{A}_0(\vec{P}_j))}{k_j^2 - k_0^2} = \int^{\Sigma} dS \left[\left(\vec{E}^{(i)} + \sum_j \frac{\vec{A}_0(\vec{P}_j)}{k_j^2 - k_0^2} \right) \times \frac{\partial G_+}{\partial n} - G_+ \frac{\partial}{\partial n} \left(\vec{E}^{(i)} + \sum_j \frac{\vec{A}_0(\vec{P}_j)}{k_j^2 - k_0^2} \right) \right] = 0 .$$

Hence the last term in (4.4) becomes

$$\sum_j \frac{S_+(\vec{P}_j)}{k_j^2 - k_+^2} \frac{k_0^2}{k_j^2 - k_0^2} = 0 . \tag{7.1}$$

This expression relates the boundary values of the polarizations and their normal derivatives on Σ in a very particular fashion. Only the particular "a. b. c." of (7.1) are the exact consequences of classical electrodynamics, in the integral equation framework, and the existence of transverse polaritons via the constitutive relations Eqs. (2.4) and (2.7). To appreciate the content of (7.1) as well as of the remainder of the analysis, we shall analyze a familiar physical situation in Sec. VIII.

VIII. NORMAL INCIDENCE ON A PLANE SURFACE

A case of considerable physical importance which has been studied in the literature experimentally and theoretically[7] is a plane wave normally incident from vacuum upon a semi-infinite crystal bounded by a plane surface Σ. In this case, we can produce two propagating parallel polaritons in the crystal, whose phase velocity dispersion is given by $n_1(\omega)$ and $n_2(\omega)$ of (5.5). The plane incident wave, the plane reflected wave, and the two propagating polariton waves in the crystal comprise the entire wave field to be determined. By contrast, in local optics, there is only one propagating polarization wave, and the incident and reflected wave.

Take the incident plane wave at normal incidence from the vacuum, traveling in direction $-z$. The surface Σ is the xy plane. The incident electric field is transverse and is

$$\vec{E}^{(i)}(\vec{r}, t) = \vec{E}^{(i)}(\vec{k}^{(i)}, \omega)\, e^{i(\vec{k}^{(i)} \cdot \vec{r} - \omega t)} , \tag{8.1}$$

$$\vec{k}^{(i)} \cdot \vec{E}^{(i)} = 0 . \tag{8.2}$$

For the polarization waves in the medium, take

$$\vec{P}_j(r, t) = \vec{P}_j(k_j, \omega)\, e^{i(\vec{k}_j \cdot \vec{r} - \omega t)} , \quad j = 1, 2 . \tag{8.3}$$

For (8.2), and (8.3),

$$|k^{(i)}| = k_0 = \omega/c , \quad |k_j| = n_j k_0 . \tag{8.4}$$

The surface integrals required in the work are $\vec{S}_0(\vec{P}_j)$ and $\vec{S}_+(\vec{P}_j)$ defined in (4.5) and (4.8). The first of these is evaluated in standard works[11] by the method of stationary phase (recall k_0 is real), and its value is

$$\vec{S}_0(\vec{P}_j) \cong -2\pi \vec{P}_j(k_j, \omega)(k_0 + k_j)(e^{ik_0 r}/k_0) . \tag{8.5}$$

The second integral may be evaluated by elementary methods using complex integration (details are given in Appendix D). The result is then given an asymptotic expansion to find

$$\vec{S}_+(\vec{P}_j) \cong -2\pi \vec{P}_j(k_j, \omega)(k_+ + k_j)(e^{ik_+ r}/k_+) , \tag{8.6}$$

which is the same form as (8.5), irrespective of k_+ being complex, while k_0 is real. Finally, we require the vector $\vec{A}_0(\vec{P}_j)$ defined in (4.6); but, since all waves are transverse,

$$\vec{A}_0(\vec{P}_j) = \vec{\nabla} \times \vec{\nabla} \times \vec{S}_0(P_j) = k_0^2 S_0(P_j) . \tag{8.7}$$

In (8.5) and (8.6) the field point is a point *within* the crystal, with coordinates $(0, 0, -r)$.

To find the reflected field, the various integrals need to be evaluated at a field point *outside* the crystal which as in the standard treatment[4] is equivalent to reversing signs of $k^{(i)}$, k_1, and k_2.

The results [Eqs. (8.5)–(8.7)] can now be substituted into (6.1) and (7.1). The extinction theorem [Eq. (6.1)] becomes

$$\vec{\mathrm{E}}^{(i)}(\vec{\mathrm{r}},\omega)=\frac{2\pi k_0^2}{k_1^2-k_0^2}\vec{\mathrm{P}}_1(k_1,\omega)(k_0+k_1)\frac{e^{ik_0r}}{k_0}+\frac{2\pi k_0^2}{k_2^2-k_0^2}\vec{\mathrm{P}}_2(k_2,\omega)(k_0+k_2)\frac{e^{ik_0r}}{k_0}\ . \tag{8.8}$$

But in our geometry (8.1) is

$$\vec{\mathrm{E}}^{(i)}(\vec{\mathrm{r}},\omega)=\vec{\mathrm{E}}^{(i)}(k_0,\omega)e^{ik_0r}\ . \tag{8.9}$$

Hence (8.8) simplifies to

$$\vec{\mathrm{E}}^{(i)}(k_0,\omega)=\tfrac{1}{2}(n_1+1)\vec{\mathrm{E}}_1(k_1,\omega)+\tfrac{1}{2}(n_2+1)\vec{\mathrm{E}}_2(k_2,\omega)\ . \tag{8.10}$$

We have also used (5.3) and the definition of refractive indices (5.6). Equation (8.10) is the extinction theorem written in terms of the electric fields.

Turning to (7.1) we have after substituting (8.5)–(8.7), using (5.3) and the definition (5.9) of a complex refractive index $\gamma\equiv k_+/k_0$,

$$\frac{\vec{\mathrm{E}}_1(k_1,\omega)}{n_1-\gamma}+\frac{\vec{\mathrm{E}}_2(k_2,\omega)}{n_2-\gamma}=0\ . \tag{8.11}$$

This is the additional boundary condition needed to determine all fields.

Finally, the reflected field is given as

$$\vec{\mathrm{E}}^{(R)}(k_0,\omega)=-\tfrac{1}{2}(n_1-1)\vec{\mathrm{E}}_1(k_1,\omega)-\tfrac{1}{2}(n_2-1)\vec{\mathrm{E}}_2(k_2,\omega)\ . \tag{8.12}$$

Clearly (8.10)–(8.12) suffice to determine all the needed fields in terms of the single incident field.[11a] Consequently, the reflectivity of a semi-infinite crystal can be computed as a function of frequency, once the parameters of the susceptibility are given. Also, the work can be generalized[5] to the lamella geometry[7] which has been used to discuss experiments on reflectivity in some II-VI compounds. These generalizations, detailed numerical calculations, and comparison with the experiment will be reported elsewhere.[5]

IX. DISCUSSION AND CONCLUSION

The theory of crystal optics including spatial dispersion, for example, polaritons has been intensively discussed in recent years.[12] A considerable literature exists on various aspects of wave propagation, reflection, and related matters, although serious theoretical questions remain even in matters of central importance. In particular, controversy exists concerning the proper additional boundary conditions to be used in order to completely specify all the fields.[13,14]

One simple type of boundary condition has been much used, apparently first due to Pekar.[1] This "a.b.c." prescribes that the total polarization due to the additional waves (polaritons) must vanish at the crystal surface Σ. In our notation this requires

$$\vec{\mathrm{P}}_1(x,y,0)+\vec{\mathrm{P}}_2(x,y,0)=0\ , \tag{9.1}$$

where $(x, y, 0)$ is the coordinate of any point on the surface Σ. In order to compare this boundary condition with the "*a.b.c.*" of our theory [Eq. (8.11)], we must rewrite the latter. Our boundary condition (8.11) becomes, after using (5.3),

$$\frac{\vec{\mathrm{P}}_1(k_1,\omega)}{(n_1-\gamma)(n_1^2-1)}+\frac{\vec{\mathrm{P}}_2(k_2,\omega)}{(n_2-\gamma)(n_2^2-1)}=0\ , \tag{9.2}$$

to be compared with (9.1) in Fourier transform

$$\vec{\mathrm{P}}_1(k_1,\omega)+\vec{\mathrm{P}}_2(k_2,\omega)=0\ . \tag{9.3}$$

Now (9.2) and (9.3) will be equal if

$$(n_1-\gamma)(n_1^2-1)=(n_2-\gamma)(n_2^2-1)\ . \tag{9.4}$$

In general, condition (9.4) is not satisfied. The refractive indices n_1 and n_2 are dispersive, and as (9.4) can be regarded as an implicit equation for the frequency, it does not appear possible to solve (9.4). However, a general statement can be made of a very approximate nature far from resonance, i.e., for $\omega\gg\omega_0$. Then taking the background dielectric constant $\epsilon_0\sim1$, one may have $n_1^2\sim1$, $n_2\sim\gamma$. In this limit, (9.2) approaches (9.3).

Since, as will be shown elsewhere,[5] the present "a.b.c." gives results in at least as good agreement with experiment (albeit using *different* material parameters such as Γ), as other boundary conditions, the appeal to experimental agreement does not appear decisive. In fact the values of material parameters (particularly the damping constant Γ) used with the present "a.b.c." appear in better agreement with independent measurements, than those values required for other theories.[5] However, still more measurements of reflectivity in different crystals and the material parameters of the crystals (such as: damping constant Γ, oscillator strength α_0, effective mass m^*, and frequency ω_0) are required.

The present analysis can be generalized to include multiple resonances, and tensorial (anisotropic) effects. The propagation of longitudinal waves can be studied also. But here we have restricted ourselves to the simplest cases which illustrate the new results.

We conclude by pointing out that the analysis presented here is rigorous. The only inputs needed are: (i) the integral equation formulation of optics based upon the polarization picture of the medium [Eq. (3.1)]; (ii) the Lorentz-Lorenz local-field expression (3.7), assumed to apply to the polarization and local field produced by polaritons; and (iii) the constitutive equations (2.4) and (2.7). Granting these, the rest of the analysis proceeds straightforwardly, and in particular without the need to assume, or guess additional boundary conditions.

Note added in manuscript. Subsequent to submission of this work for publication, there have been several related publications. A brief paper[15] gave the results for the additional boundary conditions which were obtained by use of a different method[15,16] for solving Eq. (3.1) of this paper. The results of Ref. 15 include longitudinal and transverse modes and are identical to that obtained by solution of Eq. (3.9) of this paper. Also Wolf and co-workers[17] gave results for the mode structure of the electromagnetic field in a spatially dispersive medium and showed, *inter alia*, that for the plane-parallel slab and the semi-infinite medium with plane boundary, the electric field can be expressed as a superposition of plane waves. This established the validity of the plane-wave *ansatz* made here, and also in Ref. 5. A treatment of the extinction theorem in local optics along the lines of Ref. 16 has now also appeared,[18] but free from several assumptions regarding continuity of the vector potentials made in Ref. 16. More recently the electrodynamics of bounded spatially dispersive media has been reexamined[19] using a different form for the susceptibility than used in the present paper and in Refs. 15 and 17; this changes the mode structure of the electromagnetic field and modifies the "a. b. c." This will be published separately[20] along with a proposed experiment which could decide amongst the possibilities.[21]

ACKNOWLEDGMENT

One of us (J. L. B.) wishes to thank Professor M. Hulin, Professor M. Balkanski, and Professor P. Nozières for their hospitality at the Université de Paris. He also wishes to warmly thank Professor E. Wolf for correspondence and continuing discussions. Finally, it is a pleasure to acknowledge the collaboration with Dr. R. Zeyher and Professor W. Brenig in the recent work.

APPENDIX A: FOURIER TRANSFORM OF $\chi(k,\omega)$

We write the macroscopic susceptibility as

$$\chi(k,\omega)=\frac{\chi_0}{(2\pi)^{3/2}}+\frac{F}{C(\omega)+Bk^2}\,, \tag{A1}$$

where χ_0 is defined in (2.9) and $C(\omega)$ in (2.15). The Fourier transform of $\chi(k,\omega)$ is defined via (2.3). Since a definition of the Dirac δ function can be taken as

$$(2\pi)^{-3}\int e^{i\vec{k}\cdot\vec{r}}\,d\vec{k}=\delta(\vec{r}) \tag{A2}$$

the integral of the first term is simply $\chi_0\delta(\vec{r})$. For the second term, we have the transform

$$\begin{aligned}\left(\frac{1}{2\pi}\right)^{3/2}\int e^{i\vec{k}\cdot\vec{r}}\left(\frac{F}{C+Bk^2}\right)d\vec{k}
&=\left(\frac{1}{2\pi}\right)^{3/2}(2\pi)\int_{-1}^{1}dx\int_0^\infty\frac{e^{ikrx}k^2\,dk\,F}{C+Bk^2}\\
&=\frac{F}{(2\pi)^{1/2}}\int_0^\infty\frac{(e^{ikr}-e^{-ikr})k^2\,dk}{ikr(C+Bk^2)}\\
&=\frac{F}{(2\pi)^{1/2}(ir)}\int_{-\infty}^{\infty}\frac{e^{ikr}k\,dk}{C+Bk^2}\,.\end{aligned}$$

Now we treat this integral as a complex integral, closing the contour by a semicircle at infinity. The denominator can be written as

$$C+Bk^2=B[k+i(C/B)^{1/2}]\,[k-i(C/B)^{1/2}]\,,$$

with simple poles at

$$k=\pm i(C/B)^{1/2}=k_\pm$$

as defined in (2.14). But as $r>0$, we close the contour by a semicircle in the upper-half plane. Hence the integral becomes

$$\frac{F}{(2\pi)^{1/2}(iBr)}\oint\frac{e^{ikr}k\,dk}{(k-k_+)(k-k_-)}=\frac{(2\pi i)F}{(2\pi)^{1/2}(iBr)}\,\frac{e^{ik_+r}k_+}{2k_+}=\chi_1G_+(r)\,. \tag{A3}$$

In obtaining (A3) we used the fact that k_+ has a positive imaginary part, and thus contributes to the residue in the upper-half plane, while k_- has a negative imaginary part; χ_1 is given in (2.11) and $G_+(r)$ in (2.12). One also easily verifies that with the normalization chosen, the Fourier inverse theorem (2.2) is also satisfied.

APPENDIX B: REDUCTION OF EQ. (3.9) TO EQ. (4.4) FOR *T* WAVES

To reduce (3.9) to (4.4) we require a number of intermediate results. The incident wave $\vec{E}^{(i)}$ is taken to satisfy (4.1). The trial polarization waves satisfy (4.2) and (4.3), and in addition in the transverse case,

$$\vec{\nabla}\times\vec{\nabla}\times\vec{P}_j=(\vec{\nabla})\vec{\nabla}\cdot\vec{P}_j-\nabla^2\vec{P}_j=-\nabla^2\vec{P}_j=k_j^2\vec{P}_j\,. \tag{B1}$$

Then using (4.3) and the equation satisfied by G_0 [namely, Eq. (3.5)], we have an integral

$$\int^{\Sigma}\vec{P}_j(\vec{r}\,',\omega)\,G_0(\vec{r}\,'-\vec{r}\,'')\,d\vec{r}\,'=\frac{4\pi\vec{P}_j(\vec{r}\,'',\omega)}{k_j^2-k_0^2}+\frac{\vec{S}_0(\vec{P}_j)}{k_j^2-k_0^2}\,, \tag{B2}$$

where $S_0(P_j)$ is defined in (4.5). We obtained (B2) by using Green's theorem.[4] Other integrals of this form which occur when (4.2) is substituted in (3.9) are

$$\int^{\Sigma}\vec{P}_j(\vec{r}\,',\omega)G_+(\vec{r}\,'-\vec{r}\,'')\,d\vec{r}\,'=\frac{4\pi\vec{P}_j(\vec{r}\,'',\omega)}{k_j^2-k_+^2}+\frac{\vec{S}_+(\vec{P}_j)}{k_j^2-k_+^2}\,, \tag{B3}$$

with $\vec{S}_+(\vec{P}_j)$ given in (4.5) by merely substituting $G_+(\vec{r}'-\vec{r}'')$ for $G_0(\vec{r}'-\vec{r}'')$; and finally,

$$\int^{\Sigma}\vec{E}^{(i)}(\vec{r}',\omega)G_+(\vec{r}'-\vec{r}'')\,d\vec{r}' = \frac{4\pi\vec{E}^{(i)}(\vec{r}'',\omega)}{k_0^2-k_+^2}+\frac{\vec{S}_+(\vec{E}^{(i)})}{k_0^2-k_+^2} , \quad \text{(B4)}$$

with $\vec{S}_+(\vec{E}^{(i)})$ defined in (4.8).

The surface integrals

$$\vec{S}_0(\vec{P}_j) , \quad \vec{S}_+(\vec{P}_j) , \quad \vec{S}_+(\vec{E}^{(i)})$$

each satisfy a wave equation, with the propagation vector determined by the Green's function. Thus we have

$$\nabla^2\vec{S}_0(\vec{P}_j)+k_0^2\vec{S}_0(\vec{P}_j)=0 , \quad \text{(B5)}$$

$$\nabla^2\vec{S}_+(\vec{P}_j)+k_+^2\vec{S}_+(\vec{P}_j)=0 , \quad \text{(B6)}$$

$$\nabla^2\vec{S}_+(\vec{E}^{(i)})+k_+^2\vec{S}_+(\vec{E}^{(i)})=0 . \quad \text{(B7)}$$

When (B2)–(B4) are substituted into (3.9), some additional manipulations are required. These are

$$\vec{\nabla}\times\vec{\nabla}\times\int^{\Sigma}\vec{P}_j(\vec{r}',\omega)\,G_0(\vec{r}'-\vec{r}'')\,d\vec{r}' = \frac{4\pi k_j^2\,\vec{P}_j(\vec{r}'',\omega)}{k_j^2-k_0^2}+\frac{\vec{\nabla}\times\vec{\nabla}\times\vec{S}_0(\vec{P}_j)}{k_j^2-k_0^2} . \quad \text{(B8)}$$

The last term in (B8) is the vector $\vec{A}_0(\vec{P}_j)$ defined in (4.6). Now when (B8) is put into (3.9) we find

$$\int^{\Sigma}G_+(\vec{r}''-\vec{r})\,\vec{\nabla}\times\vec{\nabla}\times\int_{\sigma}^{\Sigma}\vec{P}_j(\vec{r}',\omega)\,G_0(\vec{r}'-\vec{r})\,d\vec{r}\,d\vec{r}'$$

$$=\frac{4\pi k_j^2}{k_j^2-k_0^2}\,\frac{4\pi\vec{P}_j(\vec{r}'',\omega)}{k_j^2-k_+^2}+\frac{4\pi k_j^2}{k_j^2-k_0^2}\,\frac{\vec{S}_+(\vec{P}_j)}{k_j^2-k_+^2}+\frac{4\pi\vec{A}_0(\vec{P}_j)}{(k_j^2-k_0^2)\,(k_0^2-k_+^2)}+\frac{\vec{S}_+(\vec{A}_0(\vec{P}_j))}{(k_j^2-k_0^2)\,(k_0^2-k_+^2)} , \quad \text{(B9)}$$

with $\vec{S}_+(\vec{A}_0(\vec{P}_j))$ given in (4.7). The result (B9) is also obtained by using the Green theorem. Again it is easily verified that

$$\nabla^2\vec{S}_+(\vec{A}_0(\vec{P}_j))+k_+^2\vec{S}_+(\vec{A}_0(\vec{P}_j))=0 . \quad \text{(B10)}$$

Now all the intermediate results (B1)–(B10) can be substituted into (3.9), and the expression (2.21) can be used to identify $\chi(k,\omega)$. When all terms are gathered we obtain (4.4).

The grouping of terms in (4.4) according to the propagation vectors follows from the wave equations satisfied by $\vec{P}_j$, $\vec{A}_0(\vec{P}_j)$ and $\vec{S}_+(\vec{P}_j)$, $\vec{S}_+(\vec{E}^{(i)})$, $\vec{S}_+(\vec{A}_0(\vec{P}_j))$: following the intermediate results (B5)–(B7) and (B10).

APPENDIX C: WAVE PROPAGATION

For an infinite homogeneous insulator without currents, we have

$$\vec{\nabla}\times\vec{\nabla}\times\vec{E}(\vec{r},t)=-\frac{1}{c^2}\,\frac{\partial^2\vec{D}(\vec{r},t)}{\partial t^2} .$$

Taking fields as

$$\vec{E}(\vec{r},t)=\vec{E}(k,\omega)\,e^{i\vec{k}\cdot\vec{r}-i\omega t}$$

and using (2.4), we have

$$k^2\vec{E}(k,\omega)-\vec{k}(\vec{k}\cdot\vec{E}(k,\omega))=k_0^2[1+4\pi\chi(k,\omega)]\,\vec{E}(k,\omega) ,$$

with $(k_0)=\omega/c$. But for transverse waves, we have $\vec{k}\cdot\vec{E}=0$ and

$$(k/k_0)^2=1+4\pi\chi(k,\omega) , \quad \text{(C1)}$$

as in (5.2).

APPENDIX D: EVALUATION OF AN INTEGRAL $\vec{S}_+(\vec{P}_j)$

In the coordinate system given in Sec. VIII, the crystal surface is the xy plane Σ. The incident wave vector $\vec{k}^{(i)}$ has components $(0, 0, -k_0)$, and the wave vector of the propagating polarizations is $(0, 0, -k_j)$. A general vector in plane Σ is $\vec{r}'=(x', y', 0)$. For a field point inside the crystal, take $\vec{r}=(0, 0, -r)$. For $\vec{P}_j(r, t)$ take (8.3).

The integral $\vec{S}_+(\vec{P}_j)$ in this coordinate system is

$$\vec{S}_+(\vec{P}_j)=i\vec{P}_j(k,\omega)\int_{-\infty}^{+\infty}\int_{-\infty}^{\infty}dx'\,dy'\,\frac{e^{ik_+R}}{R}\times\left[k_+\left(1+\frac{i}{k_+R}\right)\left(\frac{r}{R}\right)+k_j\right] , \quad \text{(D1)}$$

with

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

Now as k_+ has a positive imaginary part, the integrand in (D1) converges as $|r|\to\infty$. We can evaluate it in terms of known functions by two elementary transformations. Let

$$x'=\rho\cos\theta , \quad y'=\rho\sin\theta ,$$

then

$$\xi\equiv(\rho^2+r^2)^{1/2}$$

and (D1) becomes

$$\vec{S}_+(\vec{P}_j)=2\pi i\vec{P}_j(\mathrm{k}_j,\omega)\int_r^{\infty}d\xi\,e^{ik_+\xi}\left(\frac{ir}{\xi^2}+\frac{k_+r}{\xi}+k_j\right) ,$$

or in terms of standard exponential integrals,[22] we have

$$\vec{S}_+(P_j)=2\pi i\vec{P}_j(k_j,\omega)[i\mathcal{E}_2(-ik_+r)+(k_+r)\,E_1(-ik_+r)-k_je^{ik_+r}/ik_+] ,$$

where various arguments are complex. Making an asymptotic approximation and only keeping the leading term, we have

$$\vec{S}_+(\vec{P}_j)=-2\pi\vec{P}_j(k_j,\omega)\,(k_++k_j)\left(\frac{e^{ik_+r}}{k_+}\right) . \quad \text{(D2)}$$

Actually (D2) is the same as one would obtain in the usual method,[11] but since k_+ is complex the present discussion seems more apt.

For $\vec{S}_0(\vec{P}_j)$ we ca.. replace k_+ in (D2) by k_0.

For points outside the crystal [field points $(0, 0, r)$] replace wave vectors by their negatives, as in the usual treatment.[4]

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[1]S. I. Pekar, Zh. Eksperim. i Teor. Fiz. 33, 1022 (1958) [Sov. Phys. JETP 6, 785 (1958)]; Fiz. Tverd. Tela 17, 1301 (1962) [Sov. Phys. Solid State 4, 953 (1962)].

[2]J. J. Hopfield, Phys. Rev. 112, 1555 (1958).

[3]V. M. Agranovich and V. L. Ginzburg, *Spatial Dispersion in Crystal Optics and The Theory of Excitons* (Wiley, New York, 1966). This gives a comprehensive review of the literature (see pp. 307–313) as well as the authors' point of view on many topics.

[4]See M. Born and E. Wolf, *Principles of Optics*, 3rd ed. (Pergamon, New York, 1965) for a discussion, review, and recent references, especially pp. 98–109.

[5]J. J. Sein, Ph.D. thesis (New York University, 1969) (unpublished).

[6]B. Bendow and J. L. Birman, Phys. Rev. B 1, 1678 (1970).

[7]J. J. Hopfield and D. G. Thomas, Phys. Rev. 132, 563 (1963).

[8]P. M. Morse and W. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill, New York, 1953), Chaps. 7 and 13.

[9]A. S. Pine [Phys. Rev. 139, A901 (1965)] discusses linear and nonlinear local optics by this method.

[10]For example, one might take $\vec{E}_L = \vec{E} + (4\pi/3)\gamma\vec{P}$ [as in R. Guertin and F. Stern, Phys. Rev. 134, A427 (1964)], with γ being a scalar $(0 \le \gamma \le 1)$, taking into account the extended charge distribution (eigenfunction) of the polariton. One then finds $\gamma = 1$ for consistency, of Appendix C and the dispersion equation (containing γ) which replaces Eq. (5.2).

[11]Reference 4, p. 753.

[11a]In (8.9)–(8.12) the $\vec{E}$ fields can be taken to have only one independent transverse component so these can be regarded as scalar equations in E_j.

[12]In addition to Refs. 1–3, see, for example, J. J. Hopfield, J. Phys. Soc. Japan Suppl. 21, 77 (1966).

[13]This question is discussed at great length in Ref. 3, Sec. 10, especially pp. 205–208.

[14]See G. D. Mahan and G. Obermaiar, Phys. Rev. 183, 834 (1969); and T. S. Kettrup and I. Balslev, Phys. Rev. B 3, 1457 (1971).

[15]J. J. Sein, Phys. Letters 32A, 141 (1970).

[16]J. J. Sein, Opt. Commun. 2, 170 (1970).

[17]G. S. Agarwal, D. N. Pattanayak, and E. Wolf, Phys. Rev. Letters 27, 1022 (1971); Opt. Commun. 4, 255 (1971); 4, 260 (1971); and private communications.

[18]J. de Goede and P. Mazur, Physica 58, 568 (1972).

[19]R. Zeyher and J. L. Birman, Bull. Am. Phys. Soc. 17, 369 (1972).

[20]R. Zeyher, J. L. Birman, and W. Brenig (unpublished).

[21]W. Brenig, R. Zeyher, and J. L. Birman (unpublished).

[22]W. Magnus, F. Oberhettinger, and R. P. Soni, *Formulas and Theorems for the Special Functions of Mathematical Physics*, 3rd ed. (Springer Verlag, New York, 1966), p. 342.