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LETTER TO THE EDITOR

Relation of the refractive index to the dielectric constant containing Doppler-like spatial dispersion

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Abstract. It is suggested that in obtaining the complex refractive index from a transverse dielectric function $\tilde{\epsilon}_T(k, \omega)$, it is always better to treat spatial dispersion by expanding $\tilde{\epsilon}_T(k, \omega)$ about the free-space value of k, namely ω/c, rather than about $k = 0$, as in the customary procedure due to Ginzburg, since this mistreats Doppler-like spatial dispersion. The proposed modification recovers correct results already in zero-order and is: (i) computationally advantageous as it needs only first derivatives where second derivatives are needed in the usual method, (ii) equivalent to an infinite-order expansion about $k = 0$, (iii) it is intuitively the obvious procedure to follow if k is approximately identified as a photon momentum, and (iv) it avoids any formal difficulties arising from the high symmetry of the $k = 0$ neighbourhood.

In the elementary theory of refractive index and dielectric constant (e.g., see Friedel, chapter 1 of Abelès 1972), it is customary to take

$$n^2(\omega) = \tilde{\epsilon}_T(\omega) \tag{1}$$

where $\tilde{\epsilon}_T(\omega)$ is the transverse dielectric function while $n(\omega)$ is the complex refractive index. The real and imaginary parts of $n(\omega)$ are adequate to describe transmission, absorption and other aspects of the interaction of light with matter. If we introduce the time-dependent electric and displacement vectors $E(r, t)$ and $D(r, t)$ of macroscopic Maxwell theory we can introduce a more general dielectric tensor by the Fourier transformed relation

$$D_i(k, \omega) = \sum_j \tilde{\epsilon}_{ij}(k, \omega)E_j(k, \omega), \qquad i = 1, 2, 3 \tag{2}$$

where $\tilde{\epsilon}_{ij}$ contains a k, ω dependence with k and ω as free variables. The k is referred to as spatial dispersion (Pekar 1957). Let us consider the usual method of extracting a refractive index from this k, ω dependent dielectric function (see chapter 6 of Abelès 1972, Ginzburg 1958, Agranovich and Ginzburg 1966). Taking E, D to be normal homogeneous waves, it is easy to establish the wave equation

$$\text{curl curl } E + \frac{1}{c^2} \frac{\partial^2 D}{\partial t^2} = 0$$

and hence, for the homogeneous case,

$$\sum [\omega^2 \tilde{\epsilon}_{ij}(k, \omega) - c^2 k^2(\delta_{ij} - k_i k_j/k^2)]E_j(k, \omega) = 0. \tag{3}$$

The condition for the existence of a non-trivial solution of the above set of equations,

namely the vanishing of the determinant of (3), leads us to a dispersion relation connecting ω, k and $\tilde{\epsilon}_{ij}$. Taking the transverse part of $\tilde{\epsilon}_{ij}$ and considering only isotropic media this becomes (we suppress vector and tensor notation except when needed to avoid confusion),

$$\omega^2 \tilde{\epsilon}_T(k, \omega) - c^2 k^2 = 0. \tag{4}$$

If the k dependence in $\tilde{\epsilon}_T(k, \omega)$ is neglected, using (6) we can easily recover the simplest result (1). When spatial dispersion is important, i.e. when the k dependence is non-negligible, Ginzburg (1958) introduced the Taylor expansion about $k = 0$:

$$\tilde{\epsilon}_T(k, \omega) = \tilde{\epsilon}_T^0(\omega) + k^2(\partial^2 \tilde{\epsilon}_2/\partial k^2)_{k=0}. \tag{5}$$

The linear term vanishes for isotropic media and we shall restrict ourselves to second-order expansions. The refractive index is defined by

$$ck = \omega n(\omega) u \tag{6}$$

and connects the photon momentum in the direction of the unit vector u with ω and depends only on ω. Substituting for k from (6) into (5) we have, from (4), the Fresnel equation (Landau and Lifshitz 1960),

$$\tilde{\epsilon}_T^0(\omega) - (1 - \tilde{\epsilon}_T^{(2)}(\omega))n^2(\omega) = 0$$

where

$$\tilde{\epsilon}_T^{(2)}(\omega) = k_0^2 \left(\frac{\partial^2 \tilde{\epsilon}_T(k, \omega)}{\partial k^2}\right)_{k=0}, \qquad k_0 = \frac{\omega}{c} \tag{7}$$

for determining $n(\omega)$ from the dielectric constant. Ginzburg (1958), Agranovich and Ginzburg (1966) have discussed expansions of $\tilde{\epsilon}_T^{-1}$ as well as other expansions but *always at $k = 0$*. These authors themselves point out that there could be some formal difficulties in the passage to the limit $k = 0$ in $\tilde{\epsilon}_T(k, \omega)$ although this limit is physically well defined. $k = 0$ is indeed a very special limit as at this point the distinctions between longitudinal and transverse functions begin to disappear (Ambegoakar and Kohn 1960). In addition $k = 0$ is usually a point of high degeneracy with special consequences to optical selection rules. However all studies of optical spatial dispersion known to the author follow the Ginzburg prescription (e.g., Hopfield and Thomas 1963, for a recent review see Mills and Burstein 1974, § 6). Consider the free-space photon momentum k_0 which is usually quite small compared with the characteristic momenta prevailing in solids but not necessarily in fluids. The expansion of $\tilde{\epsilon}_T(k, \omega)$ about k_0 up to second order is

$$\tilde{\epsilon}_T(k, \omega) = \tilde{\epsilon}_T(k_0, \omega) + (k - k_0)\left(\frac{\partial \tilde{\epsilon}_T}{\partial k}\right)_{k=k_0} + \frac{1}{2}(k - k_0)^2 \left(\frac{\partial^2 \tilde{\epsilon}_T}{\partial k^2}\right)_{k=k_0}. \tag{8}$$

On substituting from (6) this becomes

$$\tilde{\epsilon}_T(k, \omega) = (\tilde{\epsilon} - \tilde{\epsilon}_1 + \tilde{\epsilon}_2) + (\tilde{\epsilon}_1 - 2\tilde{\epsilon}_2)n(\omega) + \tilde{\epsilon}_2 n^2(\omega) \tag{9}$$

where

$$\tilde{\epsilon} = \tilde{\epsilon}_T(k_0, \omega) \tag{10}$$

$$\tilde{\epsilon}_1 = k_0(\partial \tilde{\epsilon}_T/\partial k)_{k=k_0} \tag{11}$$

$$\tilde{\epsilon}_2 = \frac{1}{2}k_0^2(\partial^2 \tilde{\epsilon}_T/\partial k^2)_{k=k_0}. \tag{12}$$

Due to the isotropy of the medium (i.e., $\partial \tilde{\epsilon}_T / \partial k$ at $k = 0$ is zero) the first and second derivatives at k_0 are related and this gives, to the order of our expansions:

$$\tilde{\epsilon}_1 = 2\tilde{\epsilon}_2 \tag{13}$$

and hence (9) gives

$$(\tilde{\epsilon} - \tfrac{1}{2}\tilde{\epsilon}_1) - (1 - \tfrac{1}{2}\tilde{\epsilon})n^2(\omega) = 0 \tag{14}$$

as the Fresnel equation for determining the refractive index inclusive of spatial dispersion. Equation (14), unlike equation (7), requires the numerical computation of *only the first derivative* of the dielectric function rather than the second derivative required in the Ginzburg theory. Even in those exceptional cases where the second derivative is easily available, use of the first derivative at a finite value of k, namely k_0, is equivalent to an infinite-order summation of the expansion about $k = 0$, and also avoids any formal difficulties which arise owing to the high symmetry of the $k = 0$ neighbourhood.

The microscopic theory of the dielectric function $\tilde{\epsilon}(k, \omega)$ leads to the linear response result (Kubo 1957, Bonch-Bruevich and Tyablikov 1962, § 14):

$$\tilde{\epsilon}_{ij}(k, \omega) = \left(1 - \frac{\omega_0^2}{\omega^2}\right)\delta_{ij} - \frac{1}{\omega^2} \langle\langle J_i, J_j \rangle\rangle_{\omega + i\epsilon} \tag{15}$$

where (15) contains the space and time Fourier transform of the retarded current–current Green function denoted by $\langle\langle J(r, t), J(r', t') \rangle\rangle$ and ω_0 is the plasma frequency of the system. Only the paramagnetic part of the current enters into the Green function. If the isolated particles (or quasi-particles) in the system are in the centre of mass momentum states l and internal energy states μ or ν, the transverse part of (15) can be reduced to the form ($\hbar = e = 1$)

$$\tilde{\epsilon}_T(k, \omega) = 1 - \frac{\omega_0^2}{\omega^2} - \frac{\omega_0^2}{\omega^2}\frac{1}{2N} \sum_{l,\mu,\nu} \frac{(\epsilon_\mu(l) - \epsilon_\nu(l+k))f_{\mu\nu}(l, k)\langle n_{\mu,l} - n_{\nu,l+k} \rangle}{\omega + \epsilon_\mu(l) - \epsilon_\nu(l+k) - M_{\mu\nu}(l, k, \omega)} \tag{16}$$

where $\epsilon_\mu(l)$ is the energy of the particle in the μth internal state and centre of mass momentum l (see equation (17)). In (16) $M_{\mu\nu}(l, k, \omega)$ is a mass operator which shifts and broadens the energy levels of the individual particles when the inter-particle potential is included in the Hamiltonian. The level populations $\langle n_{\mu,l} \rangle$ and the generalized oscillator strengths (see equation (18)) appear in (16) and this is a perfectly general result for an N particle system applicable to a wide range of problems. In particular (16) corresponds to (1.18–1.25) of Adler (1962) in the case of band electrons if $M_{\mu\nu}$ is neglected and if μ, ν are interpreted as band indices. Equation (16) also corresponds to microscopic results for excitons, fluid or plasma dielectric functions (Dharma-wardana 1975) if the terms are suitably interpreted. Equation (16) provides a response function inclusive of particle dynamics and a correct treatment of momentum conservation in a simple rigorous manner. The extensive use of 'quasistatic' approximations in pressure and Stark broadening theories (see the review by Griem 1974, also Berman 1972, Berman and Lamb 1971 for attempts to overcome quasistatic approximations in the theories of lasers, Jones 1974 for attempts to construct a Doppler corrected polarizability) shows that the simplicity of the rigorous theory has not been generally appreciated. Note that *ad hoc* attempts to include particle dynamics, e.g. in the theory of dilute gases, usually corrects only the energy denominators although the

centre of mass momentum l and the scattered photon momentum k appear both in the numerator and the denominator of the last term in (16). We have,

$$\epsilon_\mu(l) - \epsilon_\nu(l+k) = \epsilon_\mu - \epsilon_\nu + \frac{l^2}{2m} - \frac{(l+k)^2}{2m} = \epsilon_{\mu\nu} - \frac{l \cdot k}{m} - \frac{k^2}{2m} \tag{17}$$

$$f_{\mu\nu}(l, k) = \frac{2m}{N}\left(\epsilon_{\mu\nu} - \frac{l \cdot k}{m} - \frac{k^2}{2m}\right)\langle\mu|R \cdot u_k|\nu\rangle \tag{18}$$

where R is a dipole operator and u_k a polarization unit vector for the mode k. Also, in general $M_{\mu\nu}(l, k, \omega)$ is of the form

$$M_{\mu\nu}(l, k, \omega) = a + bk + c'kk + \dots \tag{19}$$

but b may be zero for an expansion about $k = 0$ owing to symmetry features. However, from (17) we see that the Doppler term occurs even for 'isotropic' systems and this asymmetry would exist in the energy denominators and numerators of the terms in the mass operator, so that b could in fact be non-zero. Since spatial dispersion is very much a 'non-local' characteristic, the mass operator which brings in exchange and correlation corrections to the local field should always be considered in spatial dispersion problems and any asymmetries in $M_{\mu\nu}$ will have to be correctly incorporated. This is all the more important since spatial dispersion is usually a weak phenomenon. Given equations (17), (18) and (19) we see that the Ginzburg method would lead to the *wrong result* for systems with a Doppler-like dispersion term. Even if we separately expand the numerator and denominators (instead of expanding $\tilde{\epsilon}_T(k, \omega)$ itself) as in Hopfield and Thomas (1963), about $k = 0$, a convenient Fresnel equation is not obtained and linear derivatives (which contain the Doppler term) cannot in any case be ignored.

By contrast, the expansion of $\tilde{\epsilon}_T(k, \omega)$ about $k = k_0$ is easily ascertained not to suffer from any of these difficulties. The usual Doppler profile is recovered in zero-order itself, without even having to consider the first derivative $\tilde{\epsilon}_1$. What we suggest is that the simplest method of treating spatial dispersion in every case is to expand $\tilde{\epsilon}_T(k, \omega)$ about the free-space value when in most cases the zero-order term $\tilde{\epsilon}_T(k_0, \omega)$ would have picked up an infinite series of terms in the expansion about $k = 0$ and hence would have adequately accounted for spatial dispersion. Once a refractive index is calculated in terms of the bare-photon value k_0, this could be used to define a dressed-photon wavevector which can be used in $\tilde{\epsilon}_T(k, \omega)$ and the process could be iterated if desired. However, since spatial dispersion effects are quite small such iterations would hardly ever be necessary. Even in spatial dispersion effects arising from the presence of a surface (Hopfield and Thomas 1963, Bullough 1970, Agarwal *et al* 1971) it would seem that it is physically reasonable to expand around $k = k_0$ rather than $k = 0$ since the former corresponds at least to the photon momentum 'on the vacuum side' of the surface. In effect, although k, ω are free variables in equation (2) we use an intuitive picture where k and ω are tied to a photon—a point of view entirely appropriate for extracting a refractive index.

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