Spatial dispersion and the optical properties of a vacuum-dielectric interface

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Electromagnetic matching at a nonspecular dielectric surface can be described phenomenologically in terms of a specularity parameter U. This paper comments on the solution recently given by Johnson and Rimbey. It is pointed out that this solution is only partly correct. A preliminary discussion of the general form of the complete solution is briefly presented.

A rather general study of this problem has been recently reported in this journal.[1] The main point is that previous discussions based on additional boundary conditions tend to give a partial view of the problem and to obscure the fact that no specific form of additional boundary conditions can claim more generality than any other. Each correspond to a particular model of surface scattering of the quasiparticles, e.g., excitons. A simple phenomenological model describes this in terms of a specularity parameter, called U in Ref. 1, which can take values between -1 and $+1$.

While we entirely agree with the general discussion given in Ref. 1, we point out that the results presented therein as the general solution for arbitrary U are only partly correct. To be specific, assume that the dielectric is bounded by the plane $z=0$ and contained in $z>0$. One can then construct a fictitious electromagnetic field in a hypothetical medium filling up the entire space, and such that in $z>0$ it is equal to the real field. These extended fields *are not* the real ones for $z<0$. Rather, they have definite relationships to the field in $z>0$. These relationships are written in terms of U in (2.11) and (2.12) of Ref. 1, and therefore state that the extended electric field has "U symmetry," i.e., for P-mode geometry,

$$E_x(-z)=UE_x(+z), \quad E_z(-z)=-UE_x(+z), \quad z>0, \tag{1}$$

while the magnetic field has "U antisymmetry," i.e.,

$$B_y(-z)=-UB_y(+z). \tag{2}$$

Then $U=\pm1$ corresponds to specular or antispecular surface scattering, and the corresponding E field is symmetric or antisymmetric about $z=0$. It would seem natural to expect that in the general case the extended field will be a combination of symmetric and antisymmetric terms involving U. The extended electromagnetic field is given in this form in Eqs. (3.10) and (3.12) of Ref. 1. However, it is easy to see that this gives the correct solution *only* for the two extreme cases, $U=\pm1$. Consider, for example, the x and z components of (3.10), i.e.,

$$\begin{aligned} E_x(\vec{\mathrm{k}}) &= (1+U)\left(\frac{\kappa^2}{\omega\epsilon_l}+\frac{\omega\lambda^2}{\omega^2\epsilon_t-c^2k^2}\right)\frac{ic}{k^2}B_y^s(\vec{\kappa}) \\ &\quad +(1-U)\frac{ic^2}{\omega^2\epsilon_t-c^2k^2}\lambda E_x^s(\vec{\kappa}), \\ E_z(\vec{\mathrm{k}}) &= (1+U)\left(\frac{1}{\omega\epsilon_l}-\frac{\omega}{\omega^2\epsilon_t-c^2k^2}\right)\frac{ic}{k^2}B_y^s(\vec{\kappa}) \\ &\quad -(1-U)\frac{ic^2}{\omega^2\epsilon_t-c^2k^2}\kappa E_x^s(\vec{\kappa}). \end{aligned} \tag{3}$$

Here $\vec{\mathrm{k}}=(\vec{\kappa},\lambda)$ is the three-dimensional wave vector, λ is k_z, and ω dependence is understood, e.g., $\epsilon_l(\vec{\mathrm{k}},\omega)$, etc. Now, in order to obtain the z dependence of E one must evaluate the integrals

$$E_i(\vec{\kappa},z)=\frac{1}{2\pi}\int e^{i\lambda z}E_i(\vec{\kappa},\lambda)\,d\lambda. \tag{4}$$

The exponential factor guarantees convergence on closing through the upper or lower half-circle at infinity—in the λ plane—for $z \gtrless 0$. The point to notice is the occurrence of even or odd powers of λ in (3). Remember $k^2=\kappa^2+\lambda^2$ and $\epsilon_{l,t}$ are functions of k^2. Look, e.g., at E_x in (3). The first term is symmetric; it vanishes for $U=-1$. The second one is antisymmetric and vanishes for $U=+1$. Take $U=+1$; then E_x depends only on even powers of λ. The integral (4) yields the same for $z>0$ or $z<0$. On the other hand, the surviving terms in E_z are proportional to λ and the integrals for $z>0$ and $z<0$ have opposite sign. Thus condition (1) is obeyed for $U=+1$. An identical argument shows that it is also obeyed for $U=-1$, the second term in (3) being the only surviving one in this case. Of course condition (2) is just a consequence of (1) and need not be discussed. However, this argument shows explicitly that the sum of terms in (3) *does not* obey (1) for *any* other value of U. For example, when $U=0$ the field (3) *does not* vanish for $z<0$. The point is that, against all intuitive expectation, a combination of symmetric and antisymmetric terms cannot satisfy (1). It is impossible to diagonalize the inversion group except in its own natural basis of the two functions corresponding to $U = \pm 1$. The general solution for arbitrary U must contain extra terms not included in (3). The full derivation of these extra terms is rather involved. The paper discussed here[1] came to our attention as we were completing the study of this problem. We shall only indicate here the nature of the general result we have obtained for arbitrary U. A full discussion will soon appear elsewhere.

The complete extended electric field is of the form

$$\begin{aligned} E_x(\vec{k}) &= i\frac{4\pi}{c}\left[\left(-\frac{c}{\omega}\frac{\kappa(1+f_1)}{\epsilon_l}+\frac{\omega}{c}\frac{\lambda(f_2+\lambda/\kappa)}{k^2-\epsilon_t\omega^2/c^2}\right)\frac{j(\kappa)}{k^2}-\frac{\lambda}{k^2-\epsilon_t\omega^2/c^2}g(\kappa)\right], \\ E_z(\vec{k}) &= -i\frac{4\pi}{c}\left[\left(\frac{c}{\omega}\frac{\lambda(1+f_1)}{\epsilon_l}+\frac{\omega}{c}\frac{\kappa(f_2+\lambda/\kappa)}{k^2-\epsilon_t\omega^2/c^2}\right)\frac{j(\kappa)}{k^2}-\frac{\kappa}{k^2-\epsilon_t\omega^2/c^2}g(\kappa)\right]. \end{aligned} \tag{5}$$

Here $j(\kappa)$ or $g(\kappa)$ have the nature of symmetric or antisymmetric stimuli creating the extended field. The typical denominators of the electromagnetic Green's function are quite apparent in (5), as in (3). The important difference comes from the functions f_1 and f_2, which are functions of (κ, λ). Now, j and g, being constant with respect to λ, are Fourier transforms of $\delta(z)$ and represent *surface* stimuli, while f_1 and f_2, being functions of λ, are Fourier transforms of functions of z which represent *volume* stimuli. One finds[2] that f_1 and f_2 vanish identically for $U = \pm 1$, while j vanishes for $U = -1$ and g vanishes for $U = +1$. The crucial point is that unless $U = \pm 1$, *no extended field can be constructed to satisfy* (1) and (2) *without introducing volume terms*. These are subject to certain conditions. For example, they must be zero inside the real material. This determines the possible functional form of f_1 and f_2. The parameters entering these functions, as well as j and g —all of them functions of (κ, ω) — must satisfy certain subsidiary equations so that (1) and (2) are obeyed. The explicit form of these equations —which serve to eliminate the said parameters from the final physical results— depends on the model used for the dielectric functions ϵ_l and ϵ_t.[2]

Our result (5) follows from a more compact equation which, for a closer parallel with the results of Ref. 1, can be written in the following form:

$$\begin{aligned} -\vec{k}\times[\vec{k}\times\vec{E}(\vec{k})] &= \frac{\omega^2}{c^2}\vec{D}(\vec{k}) + i\frac{\omega}{c}(1+U)\hat{n}\times\vec{B}^s(\kappa) \\ &\quad + i(1-U)\vec{k}\times[\hat{n}\times\vec{E}^s(\kappa)] \\ &\quad + i\frac{4\pi\omega}{c^2}\vec{J}_b(\vec{\kappa},\lambda). \end{aligned} \tag{6}$$

This amounts to relating our j and g to E^s and B^s of Ref. 1.

The first three terms on the right-hand side are identical to those in (3.5) of Ref. 1, where, however, the fourth term of (6) is missing. Here $\vec{J}_b(\vec{\kappa}, \lambda)$ has the nature of a volume current distribution, similar to $f_1(\kappa, \lambda)$ in (5). It can be shown, although the argument is not trivial,[2] that it vanishes for $U = \pm 1$. In other words, consider Eq. (3.3) of Ref. 1. Its Fourier transform in real space, i.e.,

$$(\omega^2/c^2)\vec{D}(\vec{r}) = \vec{\nabla}\Lambda[\vec{\nabla}\Lambda\vec{E}_{\text{eff}}(\vec{r})], \tag{7}$$

is valid for all z if and only if $U = \pm 1$. Otherwise (7) holds only for $z > 0$. It was assumed in Ref. 1 that (7) holds for all z and U. The consequence of this is carried on to Eqs. (3.10) and (3.12) of Ref. 1 which have just been discussed here.

In conclusion, the general philosophy of Ref. 1 is, we believe, quite correct. The results therein contained are also correct for the extreme cases $U = \pm 1$, but it would be erroneous to use all of them uncritically for any other values of U. To be specific, Secs. III A and III B of Ref. 1 should be preceded by the caveat that U is restricted to the values $U = \pm 1$ only; the remainder of the article is correct as it stands and needs no such restriction except where it is already specified. It might also be in order to stress, since this point is not explicitly discussed by the authors, that an analysis of the type described in Ref. 1, where it is implicitly assumed that there are no free charge carriers, is *not, in general, applicable to conductors*. However, it happens to be valid also for conductors if and only if $U = +1$. We have also studied this problem and this will also be reported in a forthcoming publication.

[1] D. L. Johnson and P. R. Rimbey, Phys. Rev. B 14, 2398 (1976).

[2] F. García-Moliner and F. Flores, J. Phys. (Paris) (to be published).