Comment on "Additional Boundary Conditions: An Historical Mistake"

In a Letter [1] Henneberger proposes a method of finding the transmission and reflection of a light wave from a crystal near an exciton resonance (the additional boundary condition or ABC problem). His method is macroscopic, formulated in real space, assumes no ABC, but does assume a surface source in the electric field wave equation. Though we agree with his physical reasoning, our derivation [2] shows that Henneberger's starting point is in error and so his results are also.

The error is seen by comparing Henneberger's assumed starting equation with a comparable one obtained from our derivation. Equation (6) from his work [1] can be written for $x \geq 0$ as

$$\frac{\partial^2 E(x,\omega)}{\partial x^2} + q_0^2 \epsilon_b E(x,\omega) + \epsilon_0 q_0^2 \int_0^\infty \chi^{\mathrm{ex}}(x - x', \omega) E(x', \omega)\, dx' = s_0(\omega)\delta(x), \tag{1}$$

where $q_0 \equiv \omega/c$, ϵ_b is the background dielectric permittivity, $\chi^{\mathrm{ex}}(x,\omega)$ is the excitonic susceptibility, Henneberger's approximation $s(x,\omega) \approx s_0(\omega)\delta(x)$ for the surface source is inserted, and ϵ_0 is the permittivity of free space (SI units).

Our derivation begins at a more fundamental level than an assumed susceptibility and operates entirely in wave-vector space [2]. However, a real-space transformation for the excitonic polarization, Eqs. (88a,b) of Ref. [2], can be reexpressed and put in the wave equation for $x > 0$ with the result

$$\frac{\partial^2 E(x,\omega)}{\partial x^2} + q_0^2 \epsilon_b E(x,\omega) + \epsilon_0 q_0^2 \int_0^\infty \chi^{\mathrm{ex}}(x - x', \omega) E(x', \omega)\, dx' = -\frac{1}{4}\left[p^{\mathrm{ex}(0)} + \frac{1}{iq_{\mathrm{ex}}}\left(\frac{\partial P^{\mathrm{ex}}}{\partial x}\right)^{(0)}\right] \times \exp(iq_{\mathrm{ex}}x), \tag{2}$$

where P^{ex} is the excitonic polarization, (0) refers to a surface layer value of the quantity, and q_{ex} is the same physical quantity as used by Henneberger (but with its frequency dependence corrected). Note that the surface layer terms depend nontrivially upon the space coordinate x and bring in *two* surface distributions to characterize the surface layer. Two arise from the Fourier transformation when the wave-vector dispersion is second order. The wave-vector-space method evaluates the surface distributions so they do not appear explicitly in our final solution though they do affect it.

Henneberger's reference to our paper suggests that our work is specialized to a particular dielectric function model. Quite to the contrary we presented a first-principles method applicable to any electromagnetic wave problem involving a bounded medium, particularly a nonlocal medium. Our method's unique characteristic is that it uses *no* boundary conditions, neither Maxwell boundary conditions nor an ABC. Our work [2] solves the macroscopic ABC problem completely. Here macroscopic means that the surface layer is very thin compared to a wavelength of the light with the result that surface layer effects can be represented as ideally thin surface distributions. Our derivation found that the fully macroscopic solution is equivalent to the use of the Pekar ABC $P^{\mathrm{ex}}(x = 0) = 0$. This is stated in our paper. We then carried the study a step further by deriving a microscopic surface layer from quantum mechanics for the case of a Frenkel exciton (small, tightly bound). The more interesting problem of a Wannier exciton (large, loosely bound) is much more difficult and was not treated. We chose to present the fully macroscopic result as merely a special case of the result applying to the Frenkel exciton. This choice has perhaps made our paper less readable and led to the misimpression referred to. It is worth noting for contrast to our result that Henneberger's result is equivalent to the use of the Ting-Frankel-Birman ABC [3] $(\partial P^{\mathrm{ex}}/\partial x) = 0$ at $x = 0$, a fact not mentioned in Ref. [1].

We emphasize that the form of the surface layer terms in Eq. (2) is quite involved and cannot be anticipated at the beginning of a real-space approach by any reasoning that we are aware of. Thus our wave-vector-space solution has not led us to finding an equivalent real-space approach. Henneberger's assumption that a simple phenomenological source $s(x,\omega) \approx s_0\delta(x)$ is adequate is a further demonstration of this difficulty.

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