Henneberger Replies: Addressing optical properties of a bounded medium, a way to circumvent, at all, the explicit consideration of surface effects is to match bulk solutions of Maxwell's equation (ME) valid for medium and vacuum separately via Maxwell's boundary conditions (MBC). This is justified as long as the optical wavelength λ is larger than the range a of surface effects. This "MBC approach" applies without conceptual problems if spatial dispersion is neglected. In the commented paper (CP) [1] I presented a method to generalize this approach to the inclusion of spatial dispersion still without additional boundary conditions (ABC) besides MBC.

In two preceding Comments by Nelson and Chen (CNC) [2] and by Zeyher (ZC) [3] the authors (A) allege errors in CP and (B) insist on the general validity of Pekar's ABC in the vicinity of the excitonic resonance. Below I will show that (A) the alleged errors result merely from incorrect references to CP and misinterpretation of both its ideas and results, and (B) the reasoning to obtain Pekar's ABC is neither general nor convincing.

(A) Let me first repeat the basic ideas of CP following the notation there: Because of the nonlocality brought about by spatial dispersion, global solutions of ME (6) of CP existing in the infinite bulk medium are used. Looking for solutions valid in the infinite bulk medium may appear rather drastic; however, only this makes the procedure independent of any claim on the behavior of the boundary (e.g., step functions or better). Since $\mathrm{Im}\{q_\alpha(\omega)\} \neq 0$ in an absorbing medium, global solutions $e^{\pm i q_\alpha x}$ describing quasi freely propagating waves as solutions of the homogeneous ME (6) do not exist on account of their asymptotic behavior. Therefore one is forced to consider sources $s(x,\omega)$ in a region $|x| < a$ in order to generate propagating waves for $|x| > a$ having the correct asymptotic behavior. These sources, though well defined, are reference sources to be put in a medium filling the full space (instead of the half space). On the other hand, since $s(x,\omega)$ is arbitrary in the beginning Eqs. (6)–(10) of CP, the method is completely general insofar as any linear combination of solutions of the homogeneous equation Eq. (6) can be obtained for $|x| > a$ choosing s_α in Eq. (10).

This MBC approach is to be contrasted with all those approaches which consider explicitly the surface providing, e.g., an expression for the susceptibility $\chi(x,x')$ of the spatially inhomogeneous system comprising the bounded medium and vacuum. Supposing $\chi(x,x')$ given, ME can be solved without stressing any ABC's. Both CNC and ZC use as a starting point the so-called dielectric approximation (DA) $\chi_0(x,x') = \Theta(x)\Theta(x')\chi_b(x-x')$ (compare Ref. [5] of ZC), where χ_b is the bulk susceptibility and the surface is considered via step functions. To χ_0 corrections $\Delta\chi(x,x')$ are added accounting surface effects more realistically. The surface response induced by $\Delta\chi$, $\Delta s(x) = q_0^2 \int dx' \, \Delta\chi(x,x')E(x')$ can be used to rewrite the homogeneous ME ($\Box_0 = \frac{\partial^2}{\partial x^2} + q_0^2$)

$$\Box_0 E(x) + q_0^2 \theta(x) \int_0^\infty dx' \, \chi_b(x-x')E(x') = -\Delta s(x) . \tag{1}$$

This equation must not be confused with ME (6) of CP being a reference ME for a medium filling the full space: (i) On its left-hand side (lhs) the step function $\Theta(x)$ is missing and the lower limit of integration is $-\infty$ instead of 0. (ii) The source term $s(x)$ on its right-hand side (rhs) appears independent of and different from the surface response $\Delta s(x)$ as an additional inhomogeneity being localized within a layer $|x| < a$.

Both Comments identify erroneously Eq. (1) of this Reply and Eq. (6) of CP: Nelson and Chen present directly their result for Δs [rhs of Eq. (2) of CNC] and Zeyher reconstructs Δs from his assumption for $\Delta\chi$ [expression (3) of ZC]. Both CNC and ZC obtain up to a prefactor $\Delta s(x) \sim e^{i q_{\mathrm{ex}} x}$ and conclude from that a contradiction to the assumption $s(x) = s_0\delta(x)$ made in CP. However, rewriting Eq. (6) of CP in such a way that its lhs corresponds to that of Eq. (1) also yields $\Delta s(x) \sim e^{i q_{\mathrm{ex}} x}$ for the Hopfield model (HM) dielectric function Eq. (13) of CP. Thus, there is no contradiction between the spatially extended surface response on the one hand and the assumption of localized sources made in CP on the other hand, and the controversy concerns merely the prefactor in the surface response describing the reflection of field fluctuations at the boundary.

A way to address this problem following Ref. [2] of ZC in a slightly generalized form is to regard the equation for the HM excitonic susceptibility $\Box_{\mathrm{ex}}\chi(x,x') = -\tilde{\Delta}\delta(x-x')$ as a propagation equation for excitons, where $\Box_{\mathrm{ex}} = \partial^2/\partial x^2 + q_{\mathrm{ex}}^2$ and $q_{\mathrm{ex}}(\omega)$ considers electron-phonon self-energy by a finite damping γ. Its solution for $x, x' > 0$, taking reflection at $x = 0$ into account, is $2q_{\mathrm{ex}}\chi(x,x') = i\tilde{\Delta}[e^{i q_{\mathrm{ex}}|x-x'|} + re^{i q_{\mathrm{ex}}(x+x')}]$. It yields for any given $r(\omega)$ the correct polariton dispersion of the bulk and, depending on the choice of $r(\omega)$, an ABC $(1+r)P'(0) + iq_{\mathrm{ex}}(1-r)P(0) = 0$, sometimes called generalized Pekar's ABC. Hence, the Pekar ABC is equivalent to putting $r(\omega) = -1$.

Nelson and Chen claim to having derived the Pekar ABC and that this is explicitly stated in their Ref. [2]. However, neither is this statement proved in their Comment nor made in their Ref. [2]. There (p. 15 387) they write "we conclude that the Pekar boundary condition cannot apply to a Frenkel exciton ... the same conclusion applies approximately to Wannier excitons" and summarize "we believe, it is incorrect to use Pekar's boundary condition macroscopically." This, so far, would not contradict but rather confirm the result of CP.

ZC states that the approach of CP is incorrect with respect to (a) a proper treatment of $\chi(x,x')$ and (b) solving ME using the obtained χ. The fact is, however, that (a) CP never addressed a proper treatment of $\chi(x,x')$ but just, on the contrary, figured out a way to avoid its consideration at all, providing a solution referring solely to χ_b, and (b) Eq. (6) of CP differs from Eq. (1) of this

Reply in its mathematical structure (lhs) as well as in the meaning of the sources s and the surface response Δs (rhs). Particularly $\Delta s = 0$ reduces Eq. (1) to the DA, whereas $s(x) = 0$ in Eq. (6) of CP leads inevitably to $E(x) \equiv 0$, because a global solution of the homogeneous Eq. (6) of CP does not exist in contrast to the reasoning in point (b) of ZC.

(B) Zeyher in his Ref. [2] in a sense "derives" the Pekar ABC, $r(\omega) = -1$, assuming ideal reflection of an exciton at the boundary. However, this assumption is neither generally valid nor convincing from a theoretical point of view. For example, in a paper by Ting, Frankel, and Birman (TFB) (compare Ref. [5] of CP) $r(\omega) = +1$ is derived assuming ideal reflection for the electron and the hole separately. This demonstrates clearly that consideration of exciton reflection by introducing $r(\omega)$ in χ merely displaces the ABC problem solving ME for the field, to the $r(\omega)$ problem solving the equation of motion for the susceptibility χ.

It is to be stated, however, that χ is not appropriate to consider the intuitive idea going back to Pekar on exciton reflection at the boundary. Quite general and regardless of the simplifying treatment of electron-hole pairs as $1s$ excitons, both exciton-photon and exciton-phonon interaction are crucial insofar as they describe just the effects under consideration, namely, formation of polaritons and absorption, respectively. In a description appropriate for this many-body system (compare Ref. [10] of CP) the response to the effective field, $\chi \sim \delta j/\delta A_{\text{eff}}$, has not an autonomous physical meaning concerning propagation of quasiparticles but enters the Dyson equation for the photon Green's function (GF) as self-energy. The photon GF, however, coincides up to prefactors with the exciton GF, both having their poles at the polariton dispersion and describing the same physics, namely, the propagation of field fluctuations as (damped) polaritons. Clearly speaking, what propagates through the crystal towards the boundary are damped polaritons being either reflected, or transmitted as vacuum photons. This problem should be addressed by the Dyson equation for the photon GF reflecting most clearly the underlying physics of field fluctuations.

It can be proved easily that adding $\Delta\chi \sim re^{iq_{\text{ex}}(x+x')}$ to χ_0 is neither necessary nor consistent to describe exciton reflection at the boundary: Assuming $r = 0$, i.e., $\Delta\chi = 0$ and, hence, $\chi = \chi_0$, the photon GF can be exactly calculated within the DA. Since it yields reflection coefficients $r_\alpha(\omega, x') \neq 0$ for each polariton branch α and any polariton wave carries excitonic contributions too, this result contradicts the assumption. Also the generalized MBC approach of CP considers exciton reflection at the boundary. For that reason the photon GF has been explicitly given in the last part of CP.

In summary, the MBC approach presented in CP does not exhibit the bad mistakes alleged in both Comments, which result merely from confusing Eq. (1) of this Reply with Eq. (6) of CP and comparing quantities Δs and s that must not be compared directly. In spite of that, it is true that the idealization of the MBC approach of CP has a restricted range of validity, $\lambda \gg a \to 0$, and may fail in specific situations, where it cannot replace a more serious consideration of surface effects. This is explicitly stated already in CP. The reasoning to obtain the Pekar ABC does obviously not fill this gap but, for the very restricted case of the HM and half-space geometry, it yields an alternative idealization $r(\omega) = -1$ instead. Surprisingly, in this specific case the idealization proposed in CP corresponds choosing $r(\omega) = +1$. Thus, the reasoning of TFB to obtain their ABC, although being completely independent of and different from that of CP, confirms and supports this proposal. Moreover, as I have been informed only recently, Davidov and Eremko [4] already in 1973 used the method proposed in CP within the HM. The MBC approach of CP generalizes these results and presents the underlying principle: It describes the wave penetrating the medium from the surface by Eq. (12) of CP, and, thus, provides a solution of the ABC problem for arbitrary layer geometries in terms of the exact bulk susceptibility. For half-space geometry its result fulfills the ABC of TFB which, however, is not sufficient to fix the problem in any realistic case, i.e., where more than one resonance is considered in χ_b. Quite generally, it does not provide a further ABC in that sense, that it requires the behavior of the polarization at the boundary *a priori*, but, as it should be, fixes it *a posteriori* depending on both the medium properties and the layer geometry. This point should have been addressed by the title of CP, which apparently has led to that confusion, that it claimed to having solved the surface problem completely. For this purpose one would need a Hamiltonian of the system comprising both medium and vacuum and addressing the surface consistently on the same many-body footing on which the properties of all the quasiparticles (electron-hole pairs, phonons, photons) and their interactions have been obtained for the bulk. As long as a treatment of this problem seems out of reach, a pragmatic way would be to consider in CP at least the finite range of the source region, using in Eq. (10) $s_\alpha(\omega, a)$ for $a \neq 0$ instead of $s_0(\omega)$. Fitting then a from experimental data may prove whether the idealization of the MBC approach applies.

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