Scattering solution to the problem of additional boundary conditions

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Maxwell's boundary conditions (MBCs) were long known to be insufficient to determine the optical responses of spatially dispersive medium. Supplementing MBCs with additional boundary conditions (ABCs) has become a normal yet controversial practice. Here, the problem of ABCs is solved by analyzing some subtle aspects of a physical surface. A generic theory is presented for handling the interaction of light with the surfaces of an arbitrary medium and applied to study the traditional problem of exciton polaritons. We show that ABCs can always be adjusted to fit the theory in the examples studied here but they can by no means be construed as intrinsic surface characteristics, which are instead captured by a *surface scattering amplitude*. Methods for experimentally extracting the spatial profile of this quantity are proposed.

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Introduction. A light wave incident upon a dielectric gets partly reflected and partly transmitted. The textbook approach [1,2] to determining the reflection and transmission amplitudes, E_r and E_t, respectively, proceeds by writing down the waves in the vacuum and those in the dielectric and joining them with Maxwell's boundary conditions (MBCs) at the separation. For instance, for a normally incident beam [Fig. 1(a)] of frequency ω and wave number $k_0 = \omega/c$, where c is the speed of light in vacuum, one may write, omitting the time dependence $e^{-i\omega t}$, for the electric field $E(z<0) = e^{ik_0z} + E_r e^{-ik_0z}$ and $E(z>0) = E_t e^{ikz}$. Here, k satisfies $\epsilon = (k/k_0)^2$, with ϵ being the dielectric constant. MBCs dictate the continuity of $E(z)$ and its derivative $E'(z)$, which determines E_r and E_t.

In 1957, Pekar claimed that MBCs were insufficient to determine the optical responses of a system of excitons [3], for which ϵ is not a constant but a function of k. In the Lorentz oscillator model (LOM) [4,5], for example, one takes

$$\epsilon(k) \approx \epsilon_b + Q^2/(k^2 - q^2). \tag{1}$$

Here, ϵ_b denotes the background permittivity, $Q = \sqrt{2M\Delta/\hbar^2}$ and $q = \sqrt{2M(\omega + i\gamma - \omega_{\rm ex})/\hbar}$, with Δ, $\omega_{\rm ex}$, M, and γ being the exciton longitudinal-transverse splitting, transition energy, effective mass, and damping rate, respectively, and $\hbar$ is the reduced Planck constant. Waves propagating through such a medium fulfill a dispersion relation given by

$$\epsilon(k) = (k/k_0)^2, \tag{2}$$

which admits two solutions, k_1 and k_2, representing waves propagating to the right and two other solutions, $k_3 = -k_1$ and $k_4 = -k_2$, for waves propagating to the left. Pekar hence wrote $E(z>0) = \sum_{j=1,2} E_j e^{ik_jz}$, where E_j is the amplitude for the jth transmitted wave, ending up with three unknowns, E_r, E_1, and E_2, but only two MBCs. He imposed an additional boundary condition (ABC), that the exciton polarization vanishes at the surface, to determine all the amplitudes.

The practice of supplementing MBCs with ABCs has since been widely—and sometimes unwittingly—adopted for dealing with spatially dispersive media [6–8]. Though popular, ABCs are unjustified and disputed [9–14]. Indeed, a whole zoo of ABCs, besides Pekar's one, have been proposed [15], while no *a priori* criteria exist regarding which ABC should be selected for a given physical system. The parameters accompanying the ABCs are irrelevant to the physical parameters of a system and hence meaningless. They are more of an experimental fitting machinery than a theoretical device. Efforts to remove ABCs without extra assumptions or approximations have failed so far [16–23].

We contend that ABCs arise from an incomplete view on physical boundaries [24,25]. Here, we rectify this view and resolve the problem. We derive an ABC-free macroscopic theory for the optical responses of bounded media and exemplify it with the long-standing problem of exciton polaritons within the LOM—extensions [10,26] are discussed in the Supplemental Material [27]—for three setups, S_1, S_2, and S_3 (see Fig. 1). In both S_1 and S_2 a semi-infinite medium (SIM) is considered, but in S_1 the light source is placed outside the medium whereas in S_2 it is inside. In S_3 a slab of medium is considered with light incident from outside. The physical effects of a surface are shown totally contained in a *surface scattering amplitude* $\mathcal{R}(z)$, which gauges the response of the surface to incoming polarization waves generated at a distance z from it [28]. We show that for each setup an ABC can be contrived to fit the theory, but no single one applies to all setups even with the same material. S_1 is shown to be insensitive to the full profile of $\mathcal{R}(z)$ but only probes some average, whereas S_2 and S_3 detect $\mathcal{R}(z)$ in full and can be used for experimentally extracting it.

Macroscopic limit of a physical surface. For clarity, let us consider a SIM with a surface lying in the x-y plane. Microscopically, the system (i.e., the medium plus the vacuum) divides into three regions: the vacuum $z<0$, the surface region $0 \leqslant z < d_s$, and the bulk region $z > d_s$, where d_s de-

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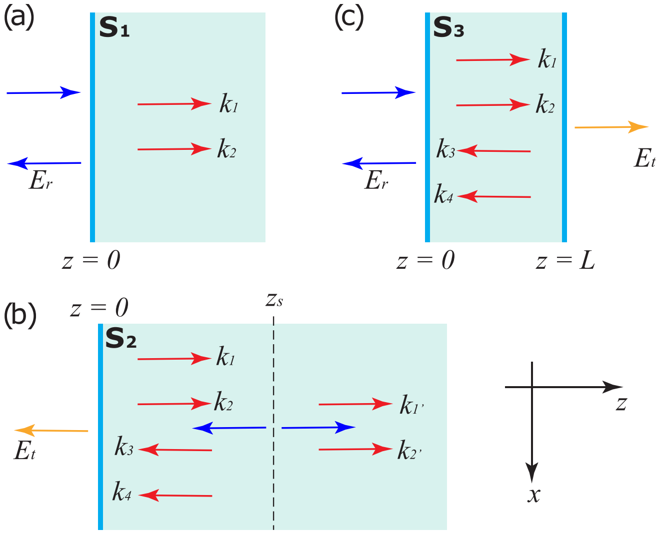

FIG. 1. Schematic of setups: (a) Setup S_1, where light normally incidents from a vacuum upon a semi-infinite medium (SIM). (b) Setup S_2, where light is radiated from a source located at $z = z_s > 0$ inside the SIM. (c) Setup S_3, where light incidents from a vacuum on a slab. The wave numbers k_α solve Eq. (2).

notes the thickness of the surface region. Macroscopically, on a scale $\Lambda \gg d_s$ where MBCs make sense [1], the surface region appears extremely thin and is traditionally treated as a geometrical separation between the vacuum and the bulk region. Physically, this region, however thin, is where physical quantities undergo rapid yet regular variations [2].

The dielectric responses may be analyzed by looking at the electric polarization $e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}\mathbf{p}(z)$ induced by an electric field $e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}\mathbf{E}(z)$ present in the system, where translational symmetry along the surface is assumed, $\mathbf{r} = (x, y)$, and $\mathbf{k}$ is a planar wave vector. $\mathbf{p}$ vanishes in the vacuum. In the bulk region it can be related to $\mathbf{E}$ by a susceptibility function $S(z, z')$, i.e., $p_\mu(z > d_s) = P_\mu(z) = \sum_\nu \int_0^\infty dz' S_{\mu\nu}(z, z')E_\nu(z')$, where $\mu, \nu = x, y, z$ label the components of the fields and we have suppressed the possible dependence of $S_{\mu\nu}$ on $(\omega, \mathbf{k})$ to simplify the notation. Since the atomistic environment in the bulk region is indistinguishable from that in an infinite medium, $S(z, z')$ must satisfy the same dynamical equations as the susceptibility function for the infinite medium without any boundaries. $S(z, z')$ inevitably contains some parameters, which appear in the general solution to the equations (see below). These parameters characterize surface scattering effects and are not determined by the equations.

To determine the polarization in the surface region, the obvious but often impractical option is to solve the full dynamical equations in this region, which requires microscopic details of a surface that is unknown in reality. Fortunately, the macroscopic limit can be determined without such details. To show this, we can introduce some phenomenological functions $w_\mu(z)$ such that $p_\mu(z) = w_\mu(z)P_\mu(z)$. By definition, $w_\mu(z)$ smoothly evolves from zero to unity as z travels across the surface region from the vacuum into the bulk region. The exact form of w_μ depends on the microscopic details of a surface. Nevertheless, on a macroscopic scale where the surface region appears infinitely thin (i.e., $d_s/\Lambda \to 0_+$), the microscopic variations become irrelevant after standard coarse graining [1,2] and $w_\mu(z)$ degenerates into the Heaviside step function $\theta(z)$. In this way, $\mathbf{p}(z)$ is fixed also in the surface region and hence determined throughout the system. Generalization of the reasoning to other geometries such as a slab is straightforward.

Interaction with light. The polarization charge density is obtained as $\rho(z) = -\nabla\cdot\mathbf{p}(z)$, where $\nabla = (i\mathbf{k}, \partial_z)$, and the current density as $\mathbf{j}(z) = -i\omega\mathbf{p}(z)$. For isotropic materials, for which $S_{\mu\nu}(z, z') = \delta_{\mu\nu}S(z, z')$ with $\delta_{\mu\nu}$ being the Kronecker symbol, $\mathbf{P}(z)$ aligns with $\mathbf{E}(z)$. Under normal incidence ($\mathbf{k} = \mathbf{0}$), we may take $\mathbf{E}(z) = [E(z), 0, 0]$ and $\mathbf{p}(z > 0) = [P(z), 0, 0]$. Then the difference between $\mathbf{p}$ and $\mathbf{P}$ is immaterial since $P_z = 0$. Substituting ρ and $\mathbf{j}$ in Maxwell's equations gives [27]

$$E(z) = E_{\rm in}(z) - 4\pi k_0^2 \int dz'\, G(z - z')P(z'), \tag{3}$$

$$P(z) = \int dz'\, S(z, z')E(z'), \tag{4}$$

where the integrals are carried out over the medium only, the Green's function $G(z) = \frac{1}{2ik_0}e^{ik_0|z-z'|}$ generates "outgoing" waves, and $E_{\rm in}(z) = e^{ik_0 z}$ for a radiation source located outside the medium (e.g., in S_1 and S_3) but differs otherwise (e.g., in S_2) (see below). In Eq. (4), z is confined to the medium. For nondispersive SIM, for which $S(z, z') = S\delta(z - z')$ with $\delta(z)$ being the Dirac function, Eq. (3) reproduces the textbook result $E_r = (\sqrt{\epsilon} - 1)/(\sqrt{\epsilon} + 1)$ with $\epsilon = 1 + 4\pi S$ being the dielectric constant [27].

Excitons by the Lorentz oscillator model. For LOM we write $S(z, z') = S_b\delta(z - z') + \tilde{S}(z, z')$, where $S_b = (\epsilon_b - 1)/4\pi$ represents the background response and $\tilde{S}$ accounts for the excitonic response. As aforementioned, the dynamical equation governing $\tilde{S}(z, z')$ is the same as that for an infinite medium, which can be established from the second term of Eq. (1) as

$$\left(\partial_z^2 + q^2\right)\tilde{S}(z, z') = -\frac{Q^2}{4\pi}\delta(z - z'). \tag{5}$$

Its solution for SIM, vanishing at infinity, has the form

$$\tilde{S}(z, z') = S_\infty\left(e^{iq|z-z'|} + \mathcal{R}(z')e^{iq(z+z')}\right), \tag{6}$$

where $S_\infty = \frac{iQ^2}{8\pi q}$ and $\mathcal{R}(z)$ is the surface characteristic quantity—the *surface scattering amplitude* (SSA). The first term in Eq. (6) describes outgoing waves generated by an electric field localized at z' and is the inverse Fourier transform of the second term in Eq. (1). In the widely used dielectric approximation [29,30], only this term is included. The second term in Eq. (6) describes polarization waves reflected from the surface. $\mathcal{R}(z)$ encodes surface scattering effects and serves as a fingerprint for distinguishing one surface from another. It cannot be determined in a macroscopic theory, but can be extracted from a microscopic surface model [27] or, as shown below, from a measured optical response.

Connection with ABCs in SIM. The excitonic polarization reads $\tilde{P}(z) = \int_0^\infty dz'\tilde{S}(z, z')E(z')$. Using Eq. (6), one gets

$$\begin{pmatrix} iq\tilde{P}(0) \\ \tilde{P}'(0) \end{pmatrix} = iqS_\infty \begin{pmatrix} R + 1 \\ R - 1 \end{pmatrix} \int_0^\infty dz e^{iqz}E(z), \tag{7}$$

where R is an average of $\mathcal{R}(z)$, defined by

$$\int_0^\infty dz e^{iqz} E(z)\mathcal{R}(z) = R \int_0^\infty dz e^{iqz} E(z). \tag{8}$$

R depends on both $\mathcal{R}(z)$ and $E(z)$, the latter being specific to the way the system is excited. Equation (7) implies that

$$\tilde{P}'(0) = \kappa \tilde{P}(0), \quad \kappa = iq\frac{R-1}{R+1}, \tag{9}$$

which has the same form as a general ABC [10,25,31]. A big caveat here is that R (and κ) cannot be interpreted as a surface characteristic (material parameter). Indeed, R does not just vary from one surface to another but, depending on the details of the way the system is excited, could take on different values even for the same surface. In what follows, we show this for both the SIM and slab geometry. It shall be seen that Eq. (9) (and $\tilde{S}$) needs to be modified for a slab merely due to the existence of two surfaces allowing waves to travel back and forth. This again underlines that R is not a surface property.

Results for S_1 *and* S_2. We begin with S_1, where light incidents from outside [Fig. 1(a)]. Equations (3) and (4) are solved by the ansatz that in the medium $P(z) = \sum_j P_j e^{ik_j z}$ and $E(z) = \sum_j E_j e^{ik_j z}$, where $\mathrm{Im}(k_j) \geqslant 0$. One obtains

$$E_j = \frac{4\pi k_0^2}{k_j^2 - k_0^2} P_j, \quad P_j = S_j E_j, \quad S_j = \frac{\epsilon(k_j) - 1}{4\pi}, \tag{10}$$

where $\epsilon(k)$ is given by Eq. (1), and

$$\sum_{j=1,2} E_j \left(\frac{1}{k_j - q} + \frac{\mathcal{R}_j(q)}{k_j + q} \right) = 0, \tag{11}$$

$$\sum_{j=1,2} E_j (k_j + k_0) = 2k_0, \tag{12}$$

with $\mathcal{R}_j(q) = -i(q+k_j)\int_0^\infty dz' \, \mathcal{R}(z') e^{i(q+k_j)z'}$. Equation (10) does not explicitly involve $\mathcal{R}_j$ and has the same form as for an infinite medium, in consistency with the Ewald-Oseen extinction theorem [23,32]. Hence, k_j are the roots of Eq. (2) as proposed by Pekar. Equations (11) and (12) uniquely fix the amplitudes E_j. The former alone takes care of boundary effects while the latter can be shown equivalent to the MBCs.

With the above ansatz Eq. (8) gives $\sum_j \frac{E_j \mathcal{R}_j(q)}{k_j + q} = R \sum_j \frac{E_j}{k_j + q}$, by which Eq. (11) can be rewritten as

$$\sum_{j=1,2} E_j \left(\frac{1}{k_j - q} + \frac{R}{k_j + q} \right) = 0. \tag{13}$$

This equation is equivalent to Eq. (9). The equivalence between Eqs. (11) and (13) fixes R, yielding

$$R = \frac{(k_2+q)(k_1-q)\mathcal{R}_1 - (k_2-q)(k_1+q)\mathcal{R}_2}{(\mathcal{R}_2 - \mathcal{R}_1)(k_2-q)(k_1-q) + 2q(k_1-k_2)}. \tag{14}$$

The optical responses are obtained by solving Eqs. (10)–(12). An example is shown in the inset of Fig. 2, where the reflection $|E_r|^2$ is plotted for $\mathcal{R}(z) = -e^{-sz}$ with $\mathrm{Re}(s) > 0$ mimicking real materials. Clearly, Eqs. (11) and (13) yield identical results as long as R is calculated by Eq. (14).

We see that S_1 is not sensitive to the whole profile of $\mathcal{R}(z)$ but only probes the average R. To experimentally extract

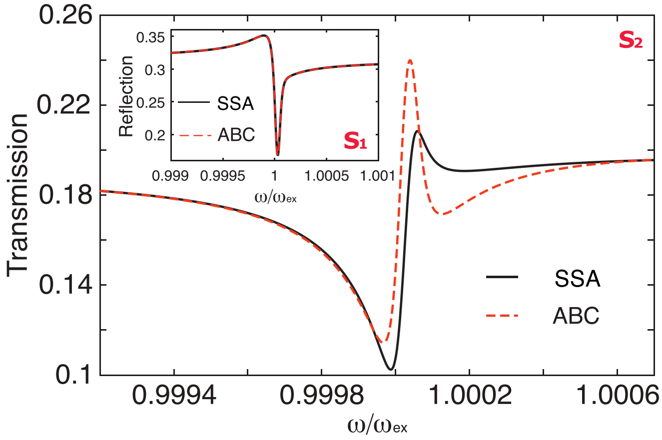

FIG. 2. Transmission $|E_t|^2 = |E(0)|^2$ of light into vacuum for S_2 with $\mathcal{R}(z) = -e^{-sz}$, where $sQ^{-1} = 0.05 + 0.32i$ and $z_s Q = 4.75$ are chosen merely for the sake of illustration, by SSA theory [i.e., Eqs. (15)–(18)] and ABC [i.e., Eqs. (15)–(17), (9), and (14)]. Inset: Reflection $|E_r|^2$ for S_1, with $E_r = E(0) - 1$, by SSA theory [Eqs. (11) and (12)] and ABC [Eqs. (9) and (14)]. The same $\mathcal{R}(z)$ is used. Other parameters are the same as in Ref. [10] and listed in the Supplemental Material [27].

$\mathcal{R}(z)$, we must analyze setups which detect the full $\mathcal{R}(z)$. S_2 and S_3 suffice for this purpose. In S_2 light is incident from a source located at $z = z_s > 0$ inside the medium [Fig. 1(b)], for which we have [27] $E_{\mathrm{in}} = e^{ik_0|z-z_s|}$. Now Eqs. (3) and (4) are solved by the ansatz that $P(0 < z < z_s) = \sum_\alpha P_\alpha e^{ik_\alpha z}$, $P(z > z_s) = \sum_j P_j e^{ik_j z}$, and $E(0 < z < z_s) = \sum_\alpha E_\alpha e^{ik_\alpha z}$ as well as $E(z > z_s) = \sum_j E_j e^{ik_j z}$. There are no restrictions on k_α but $\mathrm{Im}(k_j) \geqslant 0$. One finds that $E_{\alpha/j} = \frac{4\pi k_0^2}{k_{\alpha/j}^2 - k_0^2} P_{\alpha/j}$, $P_{\alpha/j} = S_{\alpha/j} E_{\alpha/j}$ with $S_{\alpha/j} = \frac{\epsilon(k_{\alpha/j}) - 1}{4\pi}$, which has the same form as Eq. (10), consistent with the extinction theorem. Both k_α and k_j obey Eq. (2), yielding four α modes with amplitudes E_α and two j modes with amplitudes E_j, which are determined by

$$\sum_\alpha E_\alpha (k_\alpha + k_0) = 0, \tag{15}$$

$$\sum_j e^{ik_j z_s} E_j (k_j \pm k_0) - \sum_\alpha e^{ik_\alpha z_s} E_\alpha (k_\alpha \pm k_0) = 2k_0, \tag{16}$$

$$\sum_j e^{ik_j z_s} \frac{E_j}{k_j \pm q} - \sum_\alpha e^{ik_\alpha z_s} \frac{E_\alpha}{k_\alpha \pm q} = 0, \tag{17}$$

$$\sum_\alpha E_\alpha \left(\frac{1}{k_\alpha - q} + \frac{\mathcal{R}_\alpha(z_s, q)}{k_\alpha + q} \right) + \sum_j E_j \frac{\mathcal{R}_j(z_s, q)}{k_j + q} = 0, \tag{18}$$

where $\mathcal{R}_\alpha(z_s, q) = -i(k_\alpha + q)\int_0^{z_s} dz' \, \mathcal{R}(z') e^{i(k_\alpha + q)z'}$ and $\mathcal{R}_j(z_s, q) = -i(k_j + q)\int_{z_s}^\infty dz' \, \mathcal{R}(z') e^{i(k_j+q)z'}$. These equations can be rewritten as $M(z_s)E = \psi$, where $M(z_s)$ is a square matrix with elements provided in the Supplemental Material [27], E is a column vector with elements E_α and E_j, while ψ is a column vector with all elements vanishing except two, both of which are $2k_0$.

Inserting the ansatz for S_2 into Eq. (8) and using Eq. (17) reveal that $\sum_\alpha E_\alpha \frac{\mathcal{R}_\alpha(z_s,q)}{k_\alpha+q} + \sum_j E_j \frac{\mathcal{R}_j(z_s,q)}{k_j+q} = \sum_\alpha E_\alpha \frac{R}{k_\alpha+q}$, by which Eq. (18) can be transformed into Eq. (13) but with j replaced by α. Thus, the ABC Eq. (9) remains valid. Nevertheless, there is a crucial difference resting with the fact that here R generally is not equal to the average given in Eq. (14). Instead, it takes on a completely different value [27] and varies with z_s. Exemplifying this discrepancy, we have calculated the transmission (of light into vacuum) as $|E_t|^2 = |E(0)|^2$ for $\mathcal{R}(z) = -e^{-sz}$ first by the proper theory [i.e., Eqs. (15)–(18)] and then by the ABC [Eqs. (15)–(17) and (13) with $j \to \alpha$] with R given by Eq. (14). The results are displayed in Fig. 2 and they are clearly different. This shows that experimentalists cannot use the value of R (equivalent to κ in the ABC), which they have painstakingly measured with S_1, to make predictions regarding the outcome for S_2 even though the same system is experimented with. The situation is made worse by the dependence of R on z_s, which nullifies their effort to predict what would happen if the radiation source is displaced. Only for $\mathcal{R}(z)$ being a constant is the value of R the same for both S_1 and S_2 (and equal to this constant).

Slab systems: S_3. The slab has two surfaces located at $z = 0$ and $z = L > 0$, respectively [Fig. 1(c)]. The function $\tilde{S}(z, z')$ now contains an extra contribution due to polarization waves reflected from the surface at $z = L$,

$$\tilde{S}(z, z') = S_\infty\big(e^{iq|z-z'|} + \underline{\mathcal{R}}(z')e^{iq(z+z')} + \underline{\mathcal{R}}(\bar{z}')e^{iq(\bar{z}+\bar{z}')}\big), \quad (19)$$

where $\bar{z} = L - z$ and $\bar{z}' = L - z'$, and the slab is assumed symmetric under reflections about its midplane $z = L/2$. Accounting for multiple reflections of polarization waves by the two surfaces, one may show that [27]

$$\underline{\mathcal{R}}(z) = \frac{\mathcal{R}(z) + e^{2iq(L-z)}\mathcal{R}(L)\mathcal{R}(L-z)}{1 - \mathcal{R}^2(L)e^{2iqL}}, \quad (20)$$

which can never be a constant unless $\mathcal{R}(z) \equiv 0$. $\underline{\mathcal{R}}(z)$ tends to $\mathcal{R}(z)$ for $L \to \infty$. While $\mathcal{R}(z)$ characterizes a single surface, $\underline{\mathcal{R}}(z)$ represents a cumulative effect of both surfaces.

Equations (3) and (4) are solved by the same ansatz as for S_1 but with no restrictions on wave numbers k_α here. In agreement with the extinction theorem [23,32], Eq. (10) remains valid (with $j \to \alpha$) so k_α are all four roots of Eq. (2). Equations (11) and (12) are now augmented as follows,

$$\sum_\alpha E_\alpha(k_\alpha + k_0) = 2k_0, \quad \sum_\alpha e^{ik_\alpha L}E_\alpha(k_\alpha - k_0) = 0, \quad (21)$$

$$\sum_\alpha E_\alpha\left(\frac{1}{k_\alpha - q} + \frac{\mathcal{R}^+_\alpha(q, L)}{k_\alpha + q}\right) = 0, \quad (22)$$

$$\sum_\alpha e^{ik_\alpha L}E_\alpha\left(\frac{\mathcal{R}^-_\alpha(q, L)}{k_\alpha - q} + \frac{1}{k_\alpha + q}\right) = 0. \quad (23)$$

Here, $\mathcal{R}^\pm_\alpha(q, L) = -i(q \pm k_\alpha)\int_0^L dz'\,\underline{\mathcal{R}}(z')e^{i(q\pm k_\alpha)z'}$ depends on both q and L. It can be shown [27] that, in the semi-infinite limit $L \to \infty$, E_α vanishes for modes with $\mathrm{Im}(k_\alpha) < 0$ and the results for S_1 are restored.

Mistaking that ABCs represent surface characteristics, one might apply Eq. (9) to the slab by imposing that [10]

$$\tilde{P}'(0) = \kappa\tilde{P}(0), \quad \tilde{P}'(L) = -\kappa\tilde{P}(L). \quad (24)$$

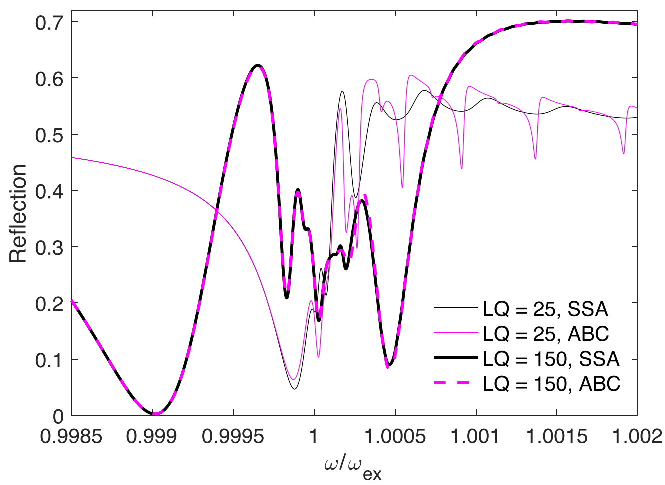

FIG. 3. Slab reflection $|E_r|^2$ for the same $\mathcal{R}(z)$ as in Fig. 2. SSA: Eqs. (21)–(23). ABC: Eqs. (21) and (24), κ by Eq. (9), and R by Eq. (14). Oscillations are due to multiple reflections [cf. Eq. (20)].

which, however, contradict Eqs. (22) and (23). As an illustration, in Fig. 3 we display the reflection $|E_r|^2$ calculated for the same $\mathcal{R}(z)$ as in Fig. 2. The SSA theory [i.e., Eqs. (21)–(23)] produces obviously different results from the ABC [i.e., Eqs. (21) and (24)] for not so thick slabs. The ABC produces unphysical results for certain thicknesses [27]. For thick slabs the SSA and the ABC produce close results owing to the effective decoupling of the surfaces. The oscillations seen in Fig. 3 stem from the multiple-reflection effect described in Eq. (20). They can also be understood as due to the quantization of the center-of-mass motions of excitons [9,10,31].

Notwithstanding, a generalization of Eq. (24) can be shown to be compatible with the SSA theory. To see this, we use Eq. (19) to obtain the exciton polarization and find

$$\begin{pmatrix} iq\tilde{P}(0) \\ iq\tilde{P}(L) \\ \tilde{P}'(0) \\ \tilde{P}'(L) \end{pmatrix} = iqS_\infty \begin{pmatrix} 1 + R_0, & e^{iqL}R_L \\ -R_0e^{iqL}, & (1 + R_L) \\ R_0 - 1, & -R_Le^{iqL} \\ R_0e^{iqL}, & (1 - R_L) \end{pmatrix} \times \begin{pmatrix} \int_0^L dze^{iqz}E(z) \\ \int_0^L dze^{iq(L-z)}E(z) \end{pmatrix}, \quad (25)$$

which is the analog of Eq. (7). Here, $R_{0/L}$ are counterparts of R, defined by

$$\int_0^L dze^{iqz}\underline{\mathcal{R}}(z)E(z) = R_0\int_0^L dze^{iqz}E(z), \quad (26a)$$

$$\int_0^L dze^{iq(L-z)}\underline{\mathcal{R}}(L-z)E(z) = R_L\int_0^L dze^{iq(L-z)}E(z). \quad (26b)$$

Eliminating the integrals from Eq. (26) yields

$$\tilde{P}'(0) = \kappa_0\tilde{P}(0) + \gamma\tilde{P}(L), \quad \tilde{P}'(L) = -\kappa_L\tilde{P}(L) - \gamma\tilde{P}(0), \quad (27)$$

which, equivalent to Eqs. (22) and (23), replaces (24). Here,

$$\begin{pmatrix} \kappa_0 \\ \kappa_L \\ \gamma \end{pmatrix} = \frac{iq}{D}\begin{pmatrix} (1 - R_0)(1 + R_L) - R_0R_Le^{2iqL} \\ (1 - R_L)(1 + R_0) - R_0R_Le^{2iqL} \\ 2R_0R_Le^{iqL} \end{pmatrix}, \quad (28)$$

with $D = R_0 R_L e^{2iqL} - (1 + R_0)(1 + R_L)$. In general, $\kappa_0 \neq \kappa_L$ except for $L \to \infty$. This asymmetry lies in the setup, not the slab, reminding that κ_0 and κ_L are not surface characteristics. As $R(z_s)$ for S_2 varies with z_s, R_0 and R_L vary with L.

Experimental relevance of $\mathcal{R}(z)$. Either S_2 or S_3 can be used to experimentally extract $\mathcal{R}(z)$. Let us take S_2 for instance. We expand $\mathcal{R}(z)$ against a set of N basis functions. The problem boils down to determining the expansion coefficients. To this end, we experimentally measure the transmission amplitude of light into the vacuum [see Fig. 1(b)] for each of N values of z_s and theoretically express the amplitudes in terms of the coefficients, which produce N equations that can be solved to obtain the coefficients. This scheme requires precise implantation and control of the interior radiation source. Using S_3 avoids this but a multitude of samples need to be fabricated [27]. More practical methods using oblique incidence, ellipsometry, and interference may be developed.

Conclusions. A macroscopic theory is presented for the optical responses of a bounded dispersive medium without invoking ABCs. ABCs are shown bearing no direct relation to the physically meaningful parameters of the system but can be deduced, if needed, from the SSA $\mathcal{R}(z)$. The SSA is an intrinsic property of a surface, which, unlike any ABCs, reflects the generally long-range contribution of the surface to the optical response. Our results call for a reappraisal of innumerable experiments that have been fitted with ABCs (see, for instance, Refs. [5–8]). An interesting direction may be to apply the present theory to ordinary and topological metamaterials, which have recently attracted much attention due to their strongly dispersive and anisotropic electrodynamic responses [33–37].

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[1] L. D. Landau and M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon Press, Oxford, UK, 1984).
[2] R. P. Feynman, R. B. Leighton, and M. Sands, *The Feynman Lectures on Physics*, Vol. II (California Institute of Technology, Pasadena, 2013), https://www.feynmanlectures.caltech.edu/II_toc.html.
[3] S. I. Pekar, Zh. Eksp. Teor. Fiz. **33**, 1022 (1957).
[4] J. J. Hopfield, Phys. Rev. **112**, 1555 (1958).
[5] A. Stahl and I. Balslev, *Electrodynamics of the Semiconductor Band Edge* (Springer, Berlin, 1987).
[6] F. Garcia-Moliner and F. Flores, *Introduction to the Theory of Solid Surfaces* (Cambridge University Press, New York, 1979).
[7] F. Forstmann and R. R. Gerhardts, *Metal Optics Near the Plasma Frequency* (Springer, Berlin, 1986).
[8] N. A. Mortensen, Nanophotonics **10**, 2563 (2021).
[9] J. Tignon, T. Hasche, D. S. Chemla, H. C. Schneider, F. Jahnke, and S. W. Koch, Phys. Rev. Lett. **84**, 3382 (2000).
[10] E. A. Muljarov and R. Zimmermann, Phys. Rev. B **66**, 235319 (2002).
[11] H.-Y. Deng, New J. Phys. **21**, 043055 (2019).
[12] H.-Y. Deng, Ann. Phys. **418**, 168204 (2020); D. Brown and H.-Y. Deng, J. Chem. Phys. **155**, 114109 (2021).
[13] H.-Y. Deng, K. Wakabayashi, and C.-H. Lam, Phys. Rev. B **95**, 045428 (2017).
[14] H.-Y. Deng, Phys. Rev. B **95**, 125442 (2017); J. Phys.: Condens. Matter **29**, 455002 (2017); Philos. Mag. **101**, 729 (2020); Eur. J. Phys. **41**, 035203 (2020).
[15] See, for example, R. J. Churchill and T. G. Philbin, Phys. Rev. B **95**, 205406 (2017), and references therein.
[16] R. Zeyher, J. L. Birman, and W. Brenig, Phys. Rev. B **6**, 4613 (1972); W. Brenig, R. Zeyher, and J. L. Birman, *ibid.* **6**, 4617 (1972).
[17] V. M. Agranovich and V. I. Yudson, Opt. Commun. **7**, 121 (1973).
[18] R. Ruppin and R. Englman, Phys. Rev. Lett. **53**, 1688 (1984); R. Ruppin, J. Phys. France **47**, 259 (1986).
[19] B. Chen and D. F. Nelson, Phys. Rev. B **48**, 15365 (1993).
[20] K. Henneberger, Phys. Rev. Lett. **80**, 2889 (1998).
[21] D. F. Nelson and B. Chen, Phys. Rev. Lett. **83**, 1263 (1999).
[22] R. Zeyher, Phys. Rev. Lett. **83**, 1264 (1999).
[23] R. Schmidt and S. Scheel, Phys. Rev. A **93**, 033804 (2016).
[24] V. M. Agranovich and V. L. Ginzburg, *Spatial Dispersion in Crystal Optics and the Theory of Excitons* (Interscience, London, 1966).
[25] D. L. Johnson and P. R. Rimbey, Phys. Rev. B **14**, 2398 (1976).
[26] K. Victor, V. M. Axt, and A. Stahl, Z. Phys. B **92**, 35 (1993).
[27] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.106.195301 for detailed derivations and a microscopic theory for the calculation of the SSA for excitons.
[28] The readers should not confuse our SSA with the so-called *d* parameters introduced by P. J. Feibelman, which are sometimes referred to as surface response functions. They have a totally different physical meaning and are irrelevant in the macroscopic limit; see, for example, P. J. Feibelman, Prog. Surf. Sci. **12**, 287 (1982); J. M. Pitarke, V. M. Silkin, E. V. Chulkov, and P. M. Echenique, Rep. Prog. Phys. **70**, 1 (2007); Y. Yang *et al.*, Nature (London) **576**, 248 (2019). A brief discussion is given in the Supplemental Material [27].
[29] G. S. Agarwal, D. N. Pattanayak, and E. Wolf, Phys. Rev. Lett. **27**, 1022 (1971).
[30] J. L. Birman and J. J. Sein, Phys. Rev. B **6**, 2482 (1972).
[31] V. A. Kiselev, B. S. Razbirin, and I. N. Uraltsev, Phys. Status Solidi B **72**, 161 (1975).
[32] D. N. Pattanayak and E. Wolf, Opt. Commun. **6**, 217 (1972); R. Schmidt and S. Scheel, J. Phys. B: At., Mol. Opt. Phys. **51**, 044003 (2018).

[33] J. B. Pendry, A. J. Holden, W. J. Steward, and I. Youngs, Phys. Rev. Lett. **76**, 4773 (1996).
[34] P. A. Belov, R. Marques, S. I. Maslovski, I. S. Nefedov, M. Silveirinha, C. R. Simovski, and S. A. Tretyakov, Phys. Rev. B **67**, 113103 (2003).
[35] M. G. Silveirinha, C. A. Fernandes, and J. R. Costa, New J. Phys. **10**, 053011 (2008); M. G. Silveirinha, *ibid.* **11**, 113016 (2009).
[36] B. Yang, Q. Guo, B. Tremain, R. Jun, L. E. Barr, Q. Yan, W. Gao, H. Liu, Y. Xinag, J. Chen, C. Fang, A. Hibbins, L. Lu, and S. Zhang, Science **359**, 1013 (2018).
[37] H. Cheng, W. Gao, Y. Bi, W. Liu, Z. Li, Q. Guo, Y. Yang, O. You, J. Feng, H. Sun, J. Tian, S. Chen, and S. Zhang, Phys. Rev. Lett. **125**, 093904 (2020).
[38] P. F. Bedaque and U. van Kolck, Annu. Rev. Nucl. Part. Sci. **52**, 339 (2002).
[39] A. Bansal, S. Binder, A. Ekström, G. Hagen, G. R. Jansen, and T. Papenbrock, Phys. Rev. C **98**, 054301 (2018).