# Towards more general constitutive relations for metamaterials: A checklist for consistent formulations

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When the period of unit cells constituting metamaterials is no longer much smaller than the wavelength but only smaller, local material laws fail to describe the propagation of light in such composite media when considered at the effective level. Instead, nonlocal material laws are required. They have to be derived by approximating a general response function of the electric field in the metamaterial at the effective level that is accurate but cannot be handled practically. But how to perform this approximation is not obvious at all. Indeed, many approximations can be perceived and one should be able to decide as quickly as possible which of these possible material laws are mathematically and physically meaningful at all. Here, at the example of a second order Padé approximation of the general response function of the electric field, we present a checklist of each possible constitutive relation that has to pass in order to be physically and mathematically liable. As will be shown, only two out of these nine Padé approximations pass the checklist. The work is meant to be a guideline applicable to decide which constitutive relation actually makes any sense at all. It is an essential ingredient for future research on composite media as any possible constitutive relation to be discussed should pass it.

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## I. INTRODUCTION

Electromagnetic metamaterials (MMs) are artificial structures made of subwavelength inclusions. These inclusions are called meta-atoms and they are mostly arranged in a periodic manner. The purpose of constructing MMs is to control the light propagation in a way inaccessible with natural materials. A referential purpose, but of course not the only one, would be to achieve a material where the real parts of both the permittivity and the permeability can be simultaneously negative at some frequencies [1]. Because of their properties, MMs enable many applications. For instance, super lenses [2,3], cloaking devices [4], medical devices [5], and many others (see, e.g., [6–10]) could be mentioned.

Due to the mesoscopic features of MMs, their homogenization was always a prime theoretical challenge. The homogenization consists of linking the mesoscopic structure of the actual MM to a macroscopic homogeneous material, i.e., replacing a MM by a hypothetical homogeneous material described by some effective material parameters [11]. Many homogenization techniques can be found in literature (see, e.g., [12–15]). However, local constitutive relations have been, mostly, considered in the homogenization and it was frequently assumed that the MM possesses a weak spatial dispersion (WSD). An effective description with at most bi-anisotropic constitutive relations would be the most advanced model to describe the MM at the level of such local constitutive relations [16]. Local constitutive relations are only justified as long as the period of the meta-atoms' arrangement is much smaller than the wavelength of light [17].

Unfortunately, most MMs do have a period only smaller than the wavelength, and period and operational wavelength are actually often in the same order of magnitude. This implies that WSD is insufficient to capture the properties of most actual MMs. A natural extension to capture the physics of MMs at the effective level, therefore, are nonlocal constitutive relations [18,19]. There, the induced response does not just depend on the electric and magnetic field at the same spatial locations but also on the fields at points further apart. Alternatively, the response depends, besides on the fields, also on the gradients of the field at the same location, a notion that is usually called strong spatial dispersion (SSD) [20,21]. This kind of spatial dispersion was introduced into solid state physics by Pekar [22], who expressed the relationship between a specific dipole moment of polarization of the crystal and the electric field intensity in the form of a differential equation. This consideration led to higher order Maxwell's equations. In the present research, we show that a similar assumption provides also higher order equations. Note that we take both terms, nonlocality and SSD, as synonymous here. Moreover, constitutive relations accommodating SSD or nonlocality, respectively, were already discussed by several authors, see, e.g., [23–28]. It was unambiguously shown that

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they frequently capture the properties of actual MMs much more accurately than ordinary local constitutive relations. But how to come to meaningful nonlocal constitutive relations for the effective description? It is clear that any description has to depart from a general nonlocal response function $\mathbf{R}(\omega, \mathbf{r} - \mathbf{r}')$ [also written in spatial Fourier space as $\widehat{\mathbf{R}}(\omega, \mathbf{k})$] that expresses the response at the effective level as a convolution. Here, the response function links the electric field to the electric displacement. It constitutes an exact description but the use of this nonlocal response function at the effective level, however, is cumbersome and not handy. It has to be approximated if it shall be of any practical use. But how to approximate it in a suitable manner?

The general nonlocal response function can be expanded, as one possible path towards meaningful nonlocal constitutive relations, by a Taylor polynomial [26,29]. Truncating the Taylor polynomial to the fourth order, forcing the coefficients of the Taylor polynomials to obey certain relations, and while omitting odd order terms that vanish for MMs with an inversion symmetry, as we do assume also here, the following constitutive relation can be derived:

$$\begin{aligned}\widehat{\mathbf{D}}(\omega, \mathbf{k}) = \varepsilon(\omega)\widehat{\mathbf{E}}(\omega, \mathbf{k}) - \mathbf{k} \times [\alpha(\omega)\mathbf{k} \times \widehat{\mathbf{E}}](\omega, \mathbf{k}) \\ + \mathbf{k} \times \mathbf{k} \times [\gamma(\omega)\mathbf{k} \times \mathbf{k}\times]\widehat{\mathbf{E}}(\omega, \mathbf{k}).\end{aligned} \tag{1}$$

In the real space, the corresponding expression to Eq. (1) is given by

$$\begin{aligned}\mathbf{D}(\omega, \mathbf{r}) = \varepsilon(\omega)\mathbf{E}(\omega, \mathbf{r}) + \nabla \times \alpha(\omega)\nabla \times \mathbf{E}(\omega, \mathbf{r}) \\ + \nabla \times \nabla \times [\gamma(\omega)\nabla \times \nabla\times]\mathbf{E}(\omega, \mathbf{r}).\end{aligned} \tag{2}$$

The material parameters $\varepsilon(\omega)$, $\alpha(\omega)$, and $\gamma(\omega)$ are anisotropic diagonal matrices, which implies that in general they do not commute with the curl operators. There is a physical meaning behind each of these terms. The first term corresponds to a local electric response, and indeed the permittivity $\varepsilon(\omega)$ appears here. It is a local response because the induced electric displacement field $\mathbf{D}(\omega, \mathbf{r})$ depends locally on the electric field $\mathbf{E}(\omega, \mathbf{r})$. The second term is associated with a weak spatial dispersion [30]. However, while it obviously depends on spatial derivatives of the electric field, a suitable gauge transformation to Maxwell's equations can be applied that forces the appearance of this second term as an artificial local magnetic response [31]. This implies that the magnetic field depends locally on the magnetic induction, where the response is mediated by the permeability that is explicitly expressed in terms of this parameter $\alpha(\omega)$. That finding is important as it shows that an artificial magnetism in MMs is a consequence of the weak spatial dispersion in the electric response. The third term in Eqs. (1) and (2) is special and indeed constitutes the suggested extension to capture effects due to strong spatial dispersion [26]. It is a consequence of the fourth order term in the Taylor polynomial. This term cannot be transformed to emerge in some local constitutive relation and is truly nonlocal. The choice of the specific functional dependency of that term can be motivated by the apparent similarity to the term that was used to capture the artificial magnetism, i.e., the material parameter is sandwiched between an equal number of curl operators.

To make practical use of such constitutive relation, interface conditions need to be derived that connect the fields inside the MM to the fields in a medium adjacent to an interface. Essential to nonlocal material laws is the appearance of multiple modes [32]. While in a local material and for a given polarization of the electromagnetic field, the dispersion relation says that for a given frequency and transverse wave vector component there is only a single forward and a single backward propagating mode, nonlocal constitutive relations lead to multiple solutions. Therefore, not just ordinary interface conditions are needed but some additional (see, e.g., [33,34]). Evidently, several methods for deriving these interface conditions exist. For instance, in the multipole expansion theory, they were obtained for a medium characterized by an electric quadrupolar and octupolar response (see, e.g., [35,36]). For our purposes, it is convenient to obtain these interface conditions using a weak formulation to Maxwell's equations (see, e.g., [37]).

The main physical motivation behind the use of the weak formulation is to consider it as a "physical model" to generate interface conditions. Moreover, our advanced constitutive relations are required to reduce to the usual interface condition in the absence of any nonlocality. This helps us to justify from a physical perspective the correctness of these interface conditions. Notice that this approach is not much different from the usual approach to derive interface conditions in electrodynamics. There, a "pillbox approach" is mostly suggested in which Maxwell's equations are solved in a spatial domain corresponding to a pillbox that is placed in its center directly at the interface. For local media we obtain from solving Maxwell's equations in such pillbox volume the continuity of tangential electric and magnetic field and normal component of electric displacement and magnetic induction as the interface condition. Now, with the nonlocal constitutive relations we do exactly the same. It is just that the resulting differential equation is too complex to find a closed form solution as it is possible for a local material. Instead, we consider its weak form. Passing to the weak formulation is frequently done and well known in mathematics and physics, and the interface or boundary conditions, which arise when partially integrating in order to switch between the weak and the strong formulation, are regarded as "natural" boundary conditions coming in addition to the boundary conditions contained in the weak formulation. Note also that finite-element methods are based on the weak formulation of the underlying differential equation.

However, the choice of the specific model for the constitutive relation based on a Taylor polynomial that led to Eq. (1) leaves the impression as being somewhat arbitrary. Many other approximations to the general nonlocal response function could have been considered and could have been applied to the homogenization [38]. A decision concerning a most appropriate model for the homogenization is always the question of how well the homogeneous model can capture the response from an actual MM. But of course, prior to any consideration it is of utmost importance to know which model for a constitutive relation is eligible at all. It is the purpose of this contribution to establish, at the example of a specific but systematic approach to generate nonlocal constitutive relations more complicated than those of the bi-anisotropic material, a checklist that can be used to validate whether a specific constitutive relation is admissible to homogenize a MM or not.

This suggested checklist has three entries. First, the dispersion relation of a nonlocal media, as mentioned, gives rise to multiple modes to be excited when an interface between an ordinary material and a nonlocal MM is illuminated with a plane wave. Therefore, we require us to derive from the combination of constitutive relations and possible weak formulations the same number of interface conditions as modes supported in the MM. Neither more (overdetermined) nor less (underdetermined) interface conditions are admissible. Second, we require that the nonlocal models resort to the local models in the limiting case of a vanishing nonlocality. Third, reciprocity relations, expressed here in terms of the Casimir-Onsager relations, need to be fulfilled by the constitutive relations. As we show, many of the possible models one could imagine actually do not cope with these requirements and, indeed, expression (1) is already an excellent choice to express the fourth order term in the nonlocality.

We demonstrate the checklist, exemplary, while considering a more general expansion of the non-local response kernel, namely a Padé approximation. The Padé approximation has the advantage of expanding the nominator and the denominator or a rational function independently. Moreover, we can admit models where the material parameters are not written specifically between symmetric order of curl operators, as written in Eq. (1). This offers a path to derive quite diverse nonlocal constitutive relations that may or may not capture the response from an actual MM quite well. However, before being considered for this task, these constitutive relations have to pass the suggested checklist.

This paper is organized as follows. In Sec. II we recall the Maxwell equations in the specific setting of interest. In Sec. III we talk about the concept of homogenization, by showing the difference between the local and nonlocal homogenization notions. In Sec. IV we present constitutive relations for what we call Padé MMs obtained from a Padé-type approximation of the nonlocal response function. The nine possible cases to perform the Padé-type approximation constitute the constitutive relations expressing the properties of the MMs at the effective level. In Sec. V we give the solutions to the dispersion relations for the nine cases, by treating separately both transverse electric (TE) and transverse magnetic (TM) polarizations. For the sake of readability, a detailed derivation of the dispersion relations is presented in an Appendix. In Sec. VI we set the function space we need in the present research, and we explain the essence of the weak formulation. Our main contribution is given in Sec. VII, in which we present the criteria we set for the checklist in order to analyze the validity of each case. These criteria are based on mathematical and physical first principles. This necessarily requires also a derivation of the interface conditions that is done in Sec. VII A as well. In Sec. VIII we discuss and summarize our findings. At the end of the paper we present in an Appendix the detailed coefficients given in the solutions to the dispersion relations.

With this work we unlock the opportunities to consider in the future other constitutive relations. Basically many formulations can be postulated but only those that pass the presented checklist should be considered.

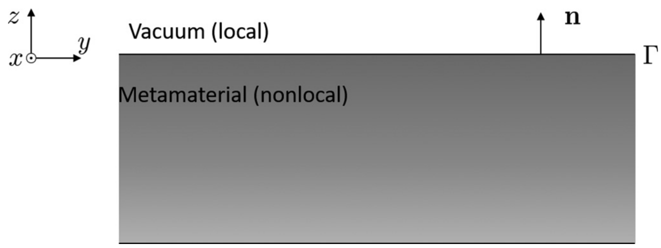


FIG. 1. Illustration of the domain in which the light propagates, the upper-half space is occupied by vacuum and the lower-half space is occupied by a homogenized MM. The surface separating the two half-spaces is denoted $\Gamma$. The normal $\mathbf{n}$ is outward directed from the homogenized MM.

## II. GENERAL SETTING

For the sake of simplicity, we assume that the propagation of light takes place in the entire space $\mathbb{R}^3$, where the upper-half space $\mathbb{R}^3_+$ is occupied by vacuum and the lower-half space $\mathbb{R}^3_-$ is occupied by a MM. The interface between them is denoted by $\Gamma$. The unit normal $\mathbf{n}$ is outwardly directed from the homogeneous MM (see Fig. 1).

We recall Maxwell's equations with no external charges and currents, which govern the propagation of light in a material

$$\nabla \times \tilde{\mathbf{E}} + \frac{1}{c}\frac{\partial \tilde{\mathbf{B}}}{\partial t} = 0, \quad \nabla \cdot \tilde{\mathbf{B}} = 0, \tag{3a}$$

$$\nabla \times \tilde{\mathbf{H}} - \frac{1}{c}\frac{\partial \tilde{\mathbf{D}}}{\partial t} = 0, \quad \nabla \cdot \tilde{\mathbf{D}} = 0. \tag{3b}$$

This system of equations includes four-vector fields: the electromagnetic field given by the pair $(\tilde{\mathbf{E}}, \tilde{\mathbf{H}})$ and the electromagnetic induction given by the pair $(\tilde{\mathbf{D}}, \tilde{\mathbf{B}})$. They depend on the position $\mathbf{r}$ and the time $t$ in $\mathbb{R}^3 \times \mathbb{R} \to \mathbb{C}^3$, but this dependency is omitted here for brevity. The speed of light is given by $c$ and the time frequency is given by $\omega > 0$. For a harmonic time dependency, Maxwell's system is written as

$$\nabla \times \mathbf{E} - ik_0\mathbf{B} = 0, \quad \nabla \cdot \mathbf{B} = 0, \tag{4a}$$

$$\nabla \times \mathbf{H} + ik_0\mathbf{D} = 0, \quad \nabla \cdot \mathbf{D} = 0. \tag{4b}$$

Here $k_0$ refers to the wave number of the external monochromatic light, it is given by the relation $k_0 = \frac{\omega}{c}$. The electromagnetic MMs we are considering are supposed to be, only for simplicity, centrosymmetric, such that no optical activity emerges in the principal axes. A typical example of such structure is the Fishnet MM (see, e.g., [39,40]). Providing the relation between the electromagnetic fields and the electromagnetic induction is required to solve the system of equation. These relations are the desired constitutive relation if considered at the effective level. For the mesoscopic, i.e., for the actual, MM, the usual local constitutive relations are considered and we assume that the MM itself is made from a nonmagnetic media.

## III. HOMOGENIZATION OF METAMATERIALS

The modeling of phenomena occurring in composite materials leads generally to partial differential equations whose coefficients are strongly oscillating. In our case, these coefficients refer to the spatially dependent permittivity that describes the properties of the actual MM in real space. These oscillations can generate problems in the analytical and numerical resolution of these equations. Moreover, we do not wish to solve Maxwell's equations exactly anytime a device made from MMs is considered; but we wish to consider MMs actually on an equal footing as ordinary materials. The theory of homogenization serves to overcome these difficulties by replacing problems with strongly oscillating coefficients by approximate problems whose coefficients are constant, and therefore much simpler to process numerically. For an introduction to this theory we refer the reader to see for instance [41–43]. In the case of MMs, the homogenization process has to be done in order to associate the MM to its effective properties. The principle consists of defining a hypothetical homogeneous material, which is characterized by effective properties. We require that in the homogeneous material: (a) the propagation of light has to be analogous as in the actual MM and (b) the reflection and transmission coefficients on an interface, separating a MM to an adjacent known material, have to be also analogous for both the homogeneous material and the actual MM. For the description of MMs at the effective level, either local or nonlocal constitutive relations can be used.

### A. Local homogenization of metamaterials

Most frequently, local constitutive relations are considered to homogenize MMs, where the electromagnetic field and the electromagnetic induction are linked to each other through local constitutive relations. This is a mere extension of how we treat natural materials in Maxwell's equations. The functional dependency of $\mathbf{D}$ and $\mathbf{H}$ is a linear combination of the macroscopic fields $\mathbf{E}$ and $\mathbf{B}$, that read

$$\mathbf{D}(\omega,\mathbf{r}) = \mathbf{E}(\omega,\mathbf{r}) + \mathbf{P}(\mathbf{E})(\omega,\mathbf{r}), \tag{5a}$$

$$\mathbf{H}(\omega,\mathbf{r}) = \mathbf{B}(\omega,\mathbf{r}) - \mathbf{M}(\mathbf{B})(\omega,\mathbf{r}), \tag{5b}$$

where $\mathbf{P}$ is the polarization and $\mathbf{M}$ is the magnetization. They shall explicitly depend only on $\mathbf{E}$ and $\mathbf{B}$ at the same spatial location and also not on derivatives. Please note, for the centrosymmetric materials as considered here, there is no electromagnetic cross coupling, i.e., $\mathbf{P}$ does not depend on $\mathbf{B}$ and $\mathbf{M}$ does not depend on $\mathbf{E}$. For intrinsically nonmagnetic materials, we have $\mathbf{M} \equiv 0$. To apply such constitutive relations, the size of the meta-atoms constituting the MM has to be much smaller than the wavelength, something that does not apply to most MMs frequently considered especially at optical frequencies. Most MMs do have a period only smaller than the wavelength and both of them are in the same order of magnitude. This implies that the WSD is insufficient to capture the properties of most actual MMs, and going beyond local homogenization with WSD is then needed for a proper homogenization.

A typical example of the local homogenization is the asymptotic homogenization (see, e.g., [14,15,44–48]). The concept considers a small parameter $\delta$ referring to the period of the meta-atoms' arrangement. Homogenizing the Maxwell system consists in studying the asymptotic behavior of its solution when $\delta$ tends to 0.

### B. Nonlocal homogenization of metamaterials

To further advance the effective description, nonlocal constitutive relations with SSD are required. We consider the general nonlocal material law of a homogeneous medium written in the real space in the following form:

$$\mathbf{D}(\omega,\mathbf{r}) = \int_{\mathbb{R}^3} \mathbf{R}(\omega,\mathbf{r}-\mathbf{r}')\mathbf{E}(\omega,\mathbf{r}')\,d\mathbf{r}', \tag{6}$$

where the kernel $\mathbf{R}(\omega,\mathbf{r}-\cdot)$ represents the response function of the electric Field $\mathbf{E}$. If the response tensor $\mathbf{R}(\omega,\mathbf{r}-\cdot)$ contains distributional terms, the nonlocal material law (6) can be written as a dual pairing, i.e., as a distributional action, as follows:

$$\mathbf{D}(\omega,\mathbf{r}) = \langle \mathbf{R}(\omega,\mathbf{r}-\cdot), \mathbf{E}(\omega,\cdot)\rangle. \tag{7}$$

In general, the formula of the response tensor is unknown, hence it is not clear how to evaluate the electromagnetic field on the interface separating two disparate media. Finally, while the expression is certainly correct, it is unhandy for any practical purpose. Therefore, this expression of the response kernel needs to be simplified in order to reach a constitutive relation of practical utility. How this can be done will be discussed in the next section.

## IV. PADÉ METAMATERIALS

### A. Constitutive relations

In the spatial frequency space the nonlocal material law (6) is written in the following form:

$$\widehat{\mathbf{D}}(\omega,\mathbf{k}) = \widehat{\mathbf{R}}(\omega,\mathbf{k})\widehat{\mathbf{E}}(\omega,\mathbf{k}). \tag{8}$$

In general, several methods exist to approximate an unknown function [like $\widehat{\mathbf{R}}(\omega,\mathbf{k})$ here], see for instance [49]. Notably, we can approximate it using polynomials. Taylor approximation is a widely used method for this purpose. However, this approach can be generalized using rational functions, i.e., the quotient of two polynomials. It is well known that for some functions an approximation with a rational function offers a higher accuracy than polynomial approximation (see [50]). This benefit clearly motivates the consideration of a Padé approximation in this paper.

Notice that the concept of an approximation with rational functions has been used as early as the beginning of the nineteenth century [51], even before Henri Padé published in 1892 an article concerning the approximate representation of a function by rational fractions [52]. After that publication, this kind of approximation of functions became well known under the name of Padé, and it was widely used for computing approximations for both physical and mathematical problems (see for instance [53]).

In [26] a specific approximation of the nonlocal response function based on a Taylor approximation up to the fourth order was considered. In the present research we consider a rational approximation of the response function, written as

$$\widehat{\mathbf{R}}(\omega, \mathbf{k}) = [I - \widehat{\mathbf{q}}(\omega, \mathbf{k})]^{-1}[\varepsilon(\omega)I + \widehat{\mathbf{p}}(\omega, \mathbf{k})], \tag{9}$$

where $I$ refers to the identity matrix, $\widehat{\mathbf{p}}(\omega, \cdot)$ and $\widehat{\mathbf{q}}(\omega, \cdot)$ are $\mathbb{C}^{3\times 3}$ matrices, polynomially depending on $\mathbf{k}$ and vanish at $\mathbf{k} = 0$. They are assumed to take the following expressions:

$$\widehat{\mathbf{p}}(\omega, \mathbf{k}) \in \{[-\alpha(\omega)\mathbf{k} \times \mathbf{k}\times],\ [-\mathbf{k} \times \alpha(\omega)\mathbf{k}\times],\ [-\mathbf{k} \times \mathbf{k} \times \alpha(\omega)]\}, \tag{10a}$$

$$\widehat{\mathbf{q}}(\omega, \mathbf{k}) \in \{[-\gamma(\omega)\mathbf{k} \times \mathbf{k}\times],\ [-\mathbf{k} \times \gamma(\omega)\mathbf{k}\times],\ [-\mathbf{k} \times \mathbf{k} \times \gamma(\omega)]\}, \tag{10b}$$

with $\alpha(\omega)$ and $\gamma(\omega)$ representing the material parameters corresponding to the polynomials $\widehat{\mathbf{p}}$ and $\widehat{\mathbf{q}}$, respectively. Together with the electric permittivity $\varepsilon(\omega)$, they are assumed to be $\mathbb{C}^{3\times 3}$ anisotropic diagonal matrices with smooth and bounded entries depending on the frequency $\omega$, defined by

$$\varepsilon = \begin{pmatrix} \varepsilon_{xx} & 0 & 0 \\ 0 & \varepsilon_{yy} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{pmatrix}, \quad \alpha = \begin{pmatrix} \alpha_{xx} & 0 & 0 \\ 0 & \alpha_{yy} & 0 \\ 0 & 0 & \alpha_{zz} \end{pmatrix},$$
$$\gamma = \begin{pmatrix} \gamma_{xx} & 0 & 0 \\ 0 & \gamma_{yy} & 0 \\ 0 & 0 & \gamma_{zz} \end{pmatrix}. \tag{11}$$

After backward Fourier transform, we see that the polynomial-matrices $\widehat{\mathbf{p}}$ and $\widehat{\mathbf{q}}$ amount to one of the following differential operators:

$$\mathbf{p}(\omega, i\nabla) \in \{[\alpha(\omega)\nabla \times \nabla\times], [\nabla \times \alpha(\omega)\nabla\times],\ [\nabla \times \nabla \times \alpha(\omega)]\}, \tag{12a}$$

$$\mathbf{q}(\omega, i\nabla) \in \{[\gamma(\omega)\nabla \times \nabla\times], [\nabla \times \gamma(\omega)\nabla\times],\ [\nabla \times \nabla \times \gamma(\omega)]\}. \tag{12b}$$

Thereafter, the nonlocal material law amounts to the following differential equation:

$$[I - \mathbf{q}(\omega, i\nabla)]\mathbf{D}(\omega, \mathbf{r}) = [\varepsilon(\omega)I + \mathbf{p}(\omega, i\nabla)]\mathbf{E}(\omega, \mathbf{r}). \tag{13}$$

According to the choices of $\mathbf{p}(\omega, i\nabla)$ and $\mathbf{q}(\omega, i\nabla)$, Eq. (13) leads to nine different formulations that we call in the following "cases."

Let us now justify the assumptions on the material parameter of being anisotropic diagonal matrices. The anisotropy of a given material is the property of being directionally dependent. More precisely, anisotropic materials admit different properties in different directions. They can be represented by nine element tensors, i.e., $\mathbb{C}^{3\times 3}$ matrices. However, for a suitable choice of coordinate system, we can only have nonzero diagonal elements in the material parameters tensors, whereas all the off-diagonal elements are zero. For this reason, instead of full anisotropic tensors, we consider only diagonal matrices that simplify computations. Furthermore, if the three values for the diagonal elements are equal, we say that the material is isotropic. Under an isotropy assumption, the material parameters are simply scalars. This fact implies that the three different formulas of the operator $\mathbf{p}(\omega, i\nabla)$ expressed in (12a) will be identical; the same will hold also for the operator $\mathbf{q}(\omega, i\nabla)$ expressed in (12b). Hence, we get only one wavelike equation instead of several formulations.

Notice that in this paper we adopted the naming *Padé metamaterials* because we used a rational fraction expansion for the nonlocal response function of the electric field inside MMs. This kind of expansions, i.e., of Padé type, were already considered in the literature, for example:

(i) When expressing the properties of metals within a hydrodynamic Drude model, e.g., applicable to study for plasmonic nanostructures (see for instance [54,55]), where the nonlocal response function is given by

$$\widehat{\mathbf{R}}(\omega, \mathbf{k})_{\text{Hydro}} = 1 - \frac{\omega_p^2}{\omega^2 + i\omega\gamma - \beta^2|\mathbf{k}|^2}, \tag{14}$$

for $\omega_p$ being the plasma frequency of the free electrons, $\gamma$ is the damping constant, and $\beta$ is a nonlocal term proportional to the Fermi velocity.

(ii) When expressing the properties of wire media [56,57], which constitutes an example of complex artificial electromagnetic materials, consisting of metallic nanowires embedded in a dielectric host medium. See for instance [58], where a nonlocal homogenization theory was applied to homogenize a wire medium and it was compared to a homogenization theory specifically derived for the wire medium in [59]. We can see clearly in [59] that the permittivity of a nanowire medium is of Padé type, having the following form:

$$\varepsilon_{\text{m}} = \varepsilon_{\text{bulk}} + \frac{i\omega_p^2\tau(R_b - R)}{\omega(\omega\tau + i)(\omega\tau R + iR_b)}. \tag{15}$$

The first term of the permittivity, denoted by $\varepsilon_{\text{bulk}}$, corresponds to the permittivity of bulk gold. The second term represents additional effects, with $\omega_p$ being the plasma frequency, $R_b$ and $R$ refer to the mean-free path and the effective mean-free path of the electrons respectively, $\tau$ refers to the relaxation time of the conducting electrons.

In the next subsection we express one wave equation that governs the propagation of light in a medium characterized by each of these constitutive relations.

### B. Wave equations

We recall Maxwell's equation for the electric field

$$\nabla \times \nabla \times \mathbf{E}(\omega, \mathbf{r}) = k_0^2\mathbf{D}(\omega, \mathbf{r}). \tag{16}$$

By substituting the general nonlocal material law (13) in the wave equation (16), we obtain

$$[I - \mathbf{q}(\omega, i\nabla)][\nabla \times \nabla \times \mathbf{E}(\omega, \mathbf{r})] = k_0^2[\varepsilon(\omega)I + \mathbf{p}(\omega, i\nabla)]\mathbf{E}(\omega, \mathbf{r}). \tag{17}$$

Hence, the general wavelike equation for homogenized MMs is given by

$$\nabla \times \nabla \times \mathbf{E}(\omega, \mathbf{r}) = k_0^2\varepsilon(\omega)\mathbf{E}(\omega, \mathbf{r}) + \big[k_0^2\mathbf{p}(\omega, i\nabla) + \mathbf{q}(\omega, i\nabla)(\nabla \times \nabla\times)\big]\mathbf{E}(\omega, \mathbf{r}). \tag{18}$$

Notice that from Eq. (13) it is clear that the relation between the fields $\mathbf{D}$ and $\mathbf{E}$ is given through a differential equation; and from Eq. (18) we can write a closed formula for the Padé

TABLE I. Wavelike equations modeling the propagation of light in media, where the upper-half space is vacuum and the lower-half space is a MM. These equations are obtained by means of a Padé-type approximation of the nonlocal response function to an incident electric field.

| | | |
|---|---|---|
| Case 1 : $\nabla\times\nabla\times\mathbf{E}=$ $k_0^2\tilde\varepsilon\mathbf{E}+k_0^2\nabla\times\tilde\alpha\nabla\times\mathbf{E}$ $+\nabla\times\nabla\times\tilde\gamma\nabla\times\nabla\times\mathbf{E}$ | Case 2 : $\nabla\times\nabla\times\mathbf{E}=$ $k_0^2\tilde\varepsilon\mathbf{E}+k_0^2\nabla\times\tilde\alpha\nabla\times\mathbf{E}$ $+\nabla\times\tilde\gamma\nabla\times\nabla\times\nabla\times\mathbf{E}$ | Case 3 : $\nabla\times\nabla\times\mathbf{E}=$ $k_0^2\tilde\varepsilon\mathbf{E}+k_0^2\nabla\times\tilde\alpha\nabla\times\mathbf{E}$ $+\tilde\gamma\nabla\times\nabla\times\nabla\times\nabla\times\mathbf{E}$ |
| Case 4 : $\nabla\times\nabla\times\mathbf{E}=$ $k_0^2\tilde\varepsilon\mathbf{E}+k_0^2\nabla\times\nabla\times\tilde\alpha\mathbf{E}$ $+\nabla\times\nabla\times\tilde\gamma\nabla\times\nabla\times\mathbf{E}$ | Case 5 : $\nabla\times\nabla\times\mathbf{E}=$ $k_0^2\tilde\varepsilon\mathbf{E}+k_0^2\nabla\times\nabla\times\tilde\alpha\mathbf{E}$ $+\nabla\times\tilde\gamma\nabla\times\nabla\times\nabla\times\mathbf{E}$ | Case 6 : $\nabla\times\nabla\times\mathbf{E}=$ $k_0^2\tilde\varepsilon\mathbf{E}+k_0^2\nabla\times\nabla\times\tilde\alpha\mathbf{E}$ $+\tilde\gamma\nabla\times\nabla\times\nabla\times\nabla\times\mathbf{E}$ |
| Case 7 : $\nabla\times\nabla\times\mathbf{E}=$ $k_0^2\tilde\varepsilon\mathbf{E}+k_0^2\tilde\alpha\nabla\times\nabla\times\mathbf{E}$ $+\nabla\times\nabla\times\tilde\gamma\nabla\times\nabla\times\mathbf{E}$ | Case 8 : $\nabla\times\nabla\times\mathbf{E}=$ $k_0^2\tilde\varepsilon\mathbf{E}+k_0^2\tilde\alpha\nabla\times\nabla\times\mathbf{E}$ $+\nabla\times\tilde\gamma\nabla\times\nabla\times\nabla\times\mathbf{E}$ | Case 9 : $\nabla\times\nabla\times\mathbf{E}=$ $k_0^2\tilde\varepsilon\mathbf{E}+k_0^2\tilde\alpha\nabla\times\nabla\times\mathbf{E}$ $+\tilde\gamma\nabla\times\nabla\times\nabla\times\nabla\times\mathbf{E}$ |

approximation to the dielectric function, given by

$$\begin{aligned}\mathbf{D}[\mathbf{E}](\omega,\mathbf{r}) &= \mathbf{R}(\omega,i\nabla)\mathbf{E}(\omega,\mathbf{r})\\ &\approx \big[\varepsilon(\omega)I+\mathbf{p}(\omega,i\nabla)\\ &\quad+\left(k_0^2\right)^{-1}\mathbf{q}(\omega,i\nabla)(\nabla\times\nabla\times)\big]\mathbf{E}(\omega,\mathbf{r}),\end{aligned}\tag{19}$$

here $I$ refers to the identity operator. By combining each possible expression for $\mathbf{p}(\omega,i\nabla)$ with each possible expression for $\mathbf{q}(\omega,i\nabla)$, given in (12a) and (12b), we get nine equations modeling the propagation of light in a homogeneous MM.

In a domain with upper-half space vacuum and lower-half space MM, the wavelike equations generated from the Padé-type response functions, named from case 1 to case 9, are given in Table I.

We draw the attention of the reader that the material parameters defined in the entire space and in the presence of an interface $\Gamma$ have no longer smooth entries due to their discontinuity on this surface; we marked them with "~" and they are given by

$$\tilde\varepsilon=\begin{cases}1, & z>0,\\ \varepsilon, & z<0,\end{cases}\qquad \tilde\alpha=\begin{cases}0, & z>0,\\ \alpha, & z<0,\end{cases}$$
$$\tilde\gamma=\begin{cases}0, & z>0,\\ \gamma, & z<0.\end{cases}\tag{20}$$

Under the anisotropy assumption, these coefficients are always $\mathbb{C}^{3\times3}$ diagonal matrices, such that the parameters $\varepsilon$, $\alpha$, and $\gamma$, given by (11), characterize a MM in the absence on any surface and they are written without "~".

We stress that the second order terms in all equations in Table I describe the local effect, i.e., the effect produced using local constitutive relations. These terms can be regrouped by means of the parameter $\tilde\mu=(1-k_0^2\tilde\alpha)^{-1}$. We notice that for the formulas corresponding to cases 1, 2, and 3 in Table I, the parameter $\tilde\mu$ represents the magnetic permeability. For the other cases, we do not have an explicit physical explanation. The nonlocal effect is given through the fourth order derivative terms, and it is represented by the parameter $\tilde\gamma$.

We emphasize that the present approach, i.e., Padé-type approximation of the response function, can be considered as a mathematical justification to the different locations of the material parameters expressed in Table I. In other words, this approach shows that these models are not randomly chosen. Notably, due to the anisotropy of the material parameters, they do not commute with the curl operators. Together with the fact of approximating the nonlocal response function $\mathbf{R}$ by means of fractional functions, it is clear that we can easily end up at a large number of formulations. We highlight that possibly not all of these formulations are useful. Therefore, a checklist needs to be established that allows us to conclude which formulations are correct and which are not. Certainly, Padé-type approximation is not the only way for generating different formulations, and any future suggestions for a more general constitutive relation should respect such sort of checklist to end up with meaningful and sound results.

We remark that the first wavelike equation in Table I, denoted as case 1, coincides implicitly with the wave equation obtained in [26,34,39] using a Taylor approximation of the response function truncated at the fourth order terms. Namely, for centrosymmetric MMs the constitutive relation in the real space is given by

$$\begin{aligned}D_i(\omega,\mathbf{r}) &= a_{ij}E_j(\omega,\mathbf{r})+c_{ijlm}\nabla_l\nabla_m E_j(\omega,\mathbf{r})\\ &\quad+e_{ijklmn}\nabla_k\nabla_l\nabla_m\nabla_n E_j(\omega,\mathbf{r}).\end{aligned}\tag{21}$$

The expansion coefficients are assumed to satisfy the following formulas:

$$a_{ij}\overset{!}{=}\varepsilon(\omega),\tag{22a}$$
$$c_{ijlm}\nabla_l\nabla_m E_j\overset{!}{=}\{\nabla\times[\alpha(\omega)\nabla\times\mathbf{E}]\}_i,\tag{22b}$$
$$e_{ijklmn}\nabla_k\nabla_l\nabla_m\nabla_n E_j\overset{!}{=}\{\nabla\times\nabla\times[\gamma(\omega)\nabla\times\nabla\times\mathbf{E}]\}_i.\tag{22c}$$

Then the obtained wavelike equation reads

$$\begin{aligned}\nabla\times\nabla\times\mathbf{E} &= k_0^2[\varepsilon(\omega)\mathbf{E}+\nabla\times\alpha(\omega)\nabla\times\mathbf{E}+\nabla\\ &\quad\times\nabla\times\gamma(\omega)\nabla\times\nabla\times\mathbf{E}].\end{aligned}\tag{23}$$

On the other hand, if we choose the operators $\mathbf{p}(\omega,i\nabla)$ and $\mathbf{q}(\omega,i\nabla)$ as follows:

$$\mathbf{p}(\omega,i\nabla)=\nabla\times\alpha(\omega)\nabla\times,\tag{24a}$$
$$\mathbf{q}(\omega,i\nabla)=\nabla\times\nabla\times\gamma(\omega),\tag{24b}$$

and we scale the parameter $\gamma(\omega)$, we get exactly the same equation as in case 1 of the Padé approximation. This equation was studied in detail in Refs. [26,34,39], and it provided good results (reflection and transmission coefficients) when compared to those obtained from a direct solution of Maxwell's equations for the actual MM (see, e.g., [60,61]). Our main concern in this paper is to study the other cases in Table I and to compare them to case 1.

Before presenting the established checklist, analyzing the wavelike equations obtained in the present paper, and solving the interface problems; we solve first the bulk problems. Namely, we compute and solve the dispersion relations which are required to define the propagating eigenmodes within a MM.

## V. DISPERSION RELATIONS

For a plane wave propagating within a medium, the dispersion relation has to be satisfied to guarantee that the plane wave indeed solves Maxwell's equations [62]. The dispersion relation expresses the functional dependency of the wave vector components of the wave and its frequency for a given medium, i.e., $k_z(k_x, k_y, k_0, \varepsilon, \alpha, \gamma)$. For a homogeneous material we can obtain the dispersion relation by plugging a plane wave ansatz in the wave equation. Hence, we get an algebraic system of equations instead of differential equations. For nontrivial solutions, the determinant of the corresponding matrix has to be zero. It is a polynomial with indeterminants the wave vector's components. The eigenvalues are the roots of the dispersion relation and the associated eigenmodes are the modes sustained inside the homogeneous material. In Fourier space, the general wavelike Eq. (18) is written as

$$\underbrace{\left\{k_0^2[\varepsilon(\omega)I + \widehat{\mathbf{p}}(\omega,\mathbf{k})] + [I - \widehat{\mathbf{q}}(\omega,\mathbf{k})](\mathbf{k}\times\mathbf{k}\times)\right\}}_{\mathcal{W}(\omega,\mathbf{k})}\widehat{\mathbf{E}}(\omega,\mathbf{k}) = 0, \tag{25}$$

for $\widehat{\mathbf{p}}(\omega,\mathbf{k})$ and $\widehat{\mathbf{q}}(\omega,\mathbf{k})$ having one of the forms expressed in (10a) and (10b). The vanishing of the determinant of the wave operator $\mathcal{W}(\omega,\mathbf{k})$ represents the dispersion relation. In vacuum, the dispersion relation is given by $k_z^2 = k_0^2 - (k_x^2 + k_y^2)$. It has two solutions $k_z = \pm\sqrt{k_0^2 - (k_x^2 + k_y^2)}$ which represent the forward and the backward modes, respectively. In a bulk MM described by the effective material parameters (11), we will see that the dispersion relations are polynomials of order four, which gives rise to multiple, exactly four, modes propagating per polarization. Precisely, in the presence of interfaces, for example a MM slab with finite thickness, there will be two forward modes excited at the first interface and two backward modes at the second interface, instead of only one at each interface in the absence of strong spatial dispersion.

Without loss of generality, we assume that the incident wave propagates in the $yz$ plane with the $z$ direction as a principal propagation direction. Hence we set $k_x = 0$ and the wave vector is written $\mathbf{k} = (0, k_y, k_z)^T$. Furthermore, along an interface $\Gamma$, the system is invariant under translation. Consequently, the wave vectors for all plane waves at the interface share the same $y$ component that we commonly denote by $k_y$. Furthermore, we decompose the incident wave into its transverse electric (TE) and transverse magnetic (TM) polarized modes. Due to the absence of optical activity in centrosymmetric MMs, polarization is preserved for the reflected and transmitted fields, such that:

(i) TE polarization:

$$\mathbf{E} = \mathbf{E}_0\exp(i\mathbf{k}\cdot\mathbf{r}),\ \text{with}\quad \mathbf{E}_0 = (E_x, 0, 0)^T\ \text{and}\quad \mathbf{k} = (0, k_y, k_z)^T.$$

(ii) TM polarization:

$$\mathbf{E} = \mathbf{E}_0\exp(i\mathbf{k}\cdot\mathbf{r}),\ \text{with}\quad \mathbf{E}_0 = (0, E_y, E_z)^T\ \text{and}\quad \mathbf{k} = (0, k_y, k_z)^T.$$

In what follows, we treat the TE and TM polarizations separately, i.e., we plug the corresponding polarized plane wave into the wavelike equation for each case in Table I and seek for the solutions $k_z(k_x, k_y, k_0, \varepsilon, \alpha, \gamma)$. In all nine cases the dispersion relations are polynomials of order 4 that depend on the polarization of the field. The corresponding solutions specify the forward and backward propagating eigenmodes inside such MMs. The solutions to the wave equation for the nine studied cases are summarized in Table II. In fact, only five different functional dependencies of $k_z(k_y)$ emerge in these $2\times 9$ resulting dispersion relations, leading to identical isofrequency contours. These five types are

*a. Dispersion of type A:* $k_z^2(k_y) = -\frac{1}{2}(q_0 + q_1)k_y^2 + p_0 \pm \sqrt{l_0 + \left(p_0 + \frac{q_0 - q_1}{2}k_y^2\right)^2}$,

*b. Dispersion of type B:* $k_z^2(k_y) = -k_y^2 + p_0 \pm \sqrt{l_0 + p_0^2 + 2(p_1 - p_0)k_y^2}$,

*c. Dispersion of type C:* $k_z^2(k_y) = -\frac{1}{2}(q_0 + q_1) + p_0 \pm \sqrt{l_0 + \left(p_0 - \frac{q_0 + q_1}{2}k_y^2\right)^2 + 2q_0p_1k_y^2 - q_0q_1k_y^4}$,

*d. Dispersion of type D:* $k_z^2(k_y) = -k_y^2 + p_0 \pm \sqrt{l_0 + p_0^2}$,

*e. Dispersion of type E:* $Q_0(k_y)k_z^2(k_y) = P_0(k_y) \pm \sqrt{[P_0(k_y^2)]^2 + P_1(k_y^2)}$.

Here the coefficients $p_0, p_1, q_0, q_1, l_0, P_0, P_1$, and $Q_0$ are products and ratios of the material parameters and depend on the polarization and on the case. For the sake of readability, we summarize the exact coefficients of the dispersion relations in the Appendix.

Concerning the first case, the dispersion relations (later the interface conditions as well) for both TE and TM polarization are identical to the wave equation considered by the Taylor approach that has been previously introduced in [26], with a normalization of the parameter expressing the nonlocal effects. The reader can return to [26,34,39] for detailed discussions. Here we repeat this dispersion relation for completeness. While eight out of nine Padé approximants yield a polynomial type of dispersion relations, the TM polarization of case 6 seems to be of special type, namely type E. It is the only case where the functional dependency of $k_z(k_y)$ is of Padé type as well, leading to peculiar isofrequency contours, inaccessible with the other eight cases. The above types of dispersion relations repeat also in a few occasions. The same type of dispersion relations will eventually reproduce the same bulk properties. In cases "4 and 7" and "5 and 8" the dispersion relations as well as their corresponding coefficients

TABLE II. Summary of the different types of dispersion relations for the wavelike equations defined in Table I, with respect to the transverse electric (TE) and the transverse magnetic (TM) polarized modes.

| $\boldsymbol{p}(\omega,\boldsymbol{k})$ \ $\boldsymbol{q}(\omega,\boldsymbol{k})$ | $\boldsymbol{k}\times\boldsymbol{k}\times\gamma$ | $\boldsymbol{k}\times\gamma\boldsymbol{k}\times$ | $\gamma\boldsymbol{k}\times\boldsymbol{k}\times$ |
|---|---|---|---|
| $\boldsymbol{k}\times\alpha\boldsymbol{k}\times$ | Case 1<br>TM: A TE: B | Case 2<br>TM: A TE: C | Case 3<br>TM: C TE: B |
| $\boldsymbol{k}\times\boldsymbol{k}\times\alpha$ | Case 4<br>TM: C TE: D | Case 5<br>TM: C TE: A | Case 6<br>TM: E TE: D |
| $\alpha\boldsymbol{k}\times\boldsymbol{k}\times$ | Case 7<br>TM: C TE: D | Case 8<br>TM: C TE: A | Case 9<br>TM: C TE: D |

are pairwise identical. However, this does not forcibly mean that the corresponding pairs of cases are equivalent, as their interface conditions differ. Consequently, light at the interface will couple differently, leading to different electromagnetic response, i.e., reflection and transmission coefficients. To reach to these interfaces, we need to say something more general on the function space setting and the possible weak formulations that we discuss in the next section.

## VI. FUNCTION SPACES SETTING AND WEAK DERIVATIVES

The wavelike equations expressed in Table I are strong formulations for Padé MMs. $\boldsymbol{C}^4(\mathbb{R}^3)$ solutions[1] do not exist. However, due to the discontinuity of $\tilde{\varepsilon}(\omega)$, $\tilde{\alpha}(\omega)$, and $\tilde{\gamma}(\omega)$ on the surface $\Gamma$ separating the upper-half space vacuum to the lower-half space MM (see Fig. 1), the derivatives in Table I are no longer considered as classical derivatives. Namely, these equations are understood in the generalized sense (see, e.g., [63]) and their solutions are generalized solutions, which are linear functionals acting continuously on smooth compactly supported functions. The space of generalized functions[2] is denoted $\mathcal{D}'(\mathbb{R}^3)$. This space represents the dual space of compactly supported smooth functions, i.e., $\boldsymbol{C}_0^\infty(\mathbb{R}^3)$.

Locally integrable functions on $\mathbb{R}^3$ [i.e., functions $f \in L^1_{\mathrm{loc}}(\mathbb{R}^3)$] provide generalized functions $F_f$, called regular generalized functions, acting on $\Psi \in C_0^\infty(\mathbb{R}^3)$ by

$$F_f[\Psi] := \int_{\mathbb{R}^3} f(\boldsymbol{r})\Psi(\boldsymbol{r})\, d\boldsymbol{r}.$$

In the same manner, we can define the associated generalized functions for a subset of $L^1_{\mathrm{loc}}(\mathbb{R}^3)$, which is the space of locally square integrable functions, i.e., $f \in L^2_{\mathrm{loc}}(\mathbb{R}^3)$. In this paper we deal with functions located in this space.

To solve the interface problems corresponding to equations in Table I, we need to write them in a weaker formulation [64]. The essence of this operation consists of formally multiplying the strong form[3] with a test function, i.e., compactly supported smooth functions, and integrating over the domain on which the equations are defined. In the generalized sense, the definition of generalized derivatives mimics partial integration. For example, for $\boldsymbol{E} \in \boldsymbol{L}^2_{\mathrm{loc}}(\mathbb{R}^3)$, $\nabla\times\boldsymbol{E} \in \mathcal{D}'(\mathbb{R}^3)$, and

$$(\nabla\times\boldsymbol{E})[\boldsymbol{\Phi}] := \int_{\mathbb{R}^3} \mathbf{E}\cdot(\nabla\times\boldsymbol{\Phi})\, d\mathbf{r}, \quad \forall\boldsymbol{\Phi} \in \mathbf{C}_0^\infty(\mathbb{R}^3).$$

Let us now consider a term appearing in cases 4, 5, and 6 in Table I, given by $\nabla\times\nabla\times\tilde{\alpha}\mathbf{E}$, and show how to define the weak derivation. For $\mathbf{F} := \tilde{\alpha}\mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3)$ and $\nabla\times\nabla\times\tilde{\alpha}\mathbf{E} \in \mathcal{D}'(\mathbb{R}^3)$, by definition we have

$$(\nabla\times\nabla\times\mathbf{F})[\boldsymbol{\Phi}] := \int_{\mathbb{R}^3} \mathbf{F}\cdot(\nabla\times\nabla\times\boldsymbol{\Phi})\, d\mathbf{r}, \quad \forall\boldsymbol{\Phi} \in \mathbf{C}_0^\infty(\mathbb{R}^3).$$

On the other hand, we have

$$\int_{\mathbb{R}^3} \mathbf{F}\cdot(\nabla\times\nabla\times\boldsymbol{\Phi})\, d\mathbf{r} = \int_{\mathbb{R}^3} \tilde{\alpha}\mathbf{E}\cdot(\nabla\times\nabla\times\boldsymbol{\Phi})\, d\mathbf{r} = \int_{\mathbb{R}^3_-} \alpha\mathbf{E}\cdot(\nabla\times\nabla\times\boldsymbol{\Phi})\, d\mathbf{r}.$$

Thus, the weak derivative of $\nabla\times\nabla\times\tilde{\alpha}\mathbf{E}$ reads

$$(\nabla\times\nabla\times\mathbf{F})[\boldsymbol{\Phi}] = \int_{\mathbb{R}^3_-} \alpha\mathbf{E}\cdot(\nabla\times\nabla\times\boldsymbol{\Phi})\, d\mathbf{r}, \quad \forall\boldsymbol{\Phi} \in \mathbf{C}_0^\infty(\mathbb{R}^3).$$

According to the same principle, we define the weak derivatives in all the wavelike equations in Table I (see Sec. VII).

Always dealing with the term $\nabla\times\nabla\times\tilde{\alpha}\mathbf{E}$, we stress that due to the discontinuity of $\tilde{\alpha}\mathbf{E}$ at the surface $\Gamma$, it is not correct to consider the following weak derivative:

$$(\nabla\times\nabla\times\mathbf{F})[\boldsymbol{\Phi}] = \int_{\mathbb{R}^3_-} (\nabla\times\alpha\mathbf{E})\cdot(\nabla\times\boldsymbol{\Phi})\, d\mathbf{r}, \quad \forall\boldsymbol{\Phi} \in \mathbf{C}_0^\infty(\mathbb{R}^3),$$

because it is not possible to define the corresponding regular generalized function to $\nabla\times\mathbf{F}$ since $\nabla\times\mathbf{F} = \nabla\times\tilde{\alpha}\mathbf{E} \notin \mathbf{L}^1_{\mathrm{loc}}(\mathbb{R}^3)$.

After explaining the principle of generalized derivatives and weak formulation, we solve the interface problems for the different cases presented in Table I by deriving the interface conditions. For the sake of shortness, we combine this task with the checklist for each model in Table I in the next section.

[1] $\boldsymbol{C}^4(\mathbb{R}^3)$: the space of vector functions, with components functions continuously differentiable up to order 4.

[2] In some references they are called distributions (see, e.g., [63]).

[3] The governing partial differential equation, in which the order of derivatives is not reduced as in the weak form.

## VII. SELECTION CRITERIA AND DISCUSSIONS

To decide which case can possibly describe the propagation of light in homogeneous MMs, we shall present mathematical and physical criteria that intervene basically in the checking process.

(1) At first we need to solve the interface problem for each case, which reveals the main principle one has to respect in such study. We highlight that the number of unknown field amplitudes at the interface $\Gamma$ is defined through the dispersion relations by counting the number of eigenmodes (cf. Sec. V). To define their amplitudes, the number of eigenmodes must coincide with the number of interface conditions that we can derive at the interface $\Gamma$.

(2) Second, as the nonlocal approach should always contain the local approach as a limit, we have to analyze the resulting reflection and transmission coefficients calculated from the Fresnel equations. To this end, we check the limit of these coefficients when $\gamma \to 0$. More precisely, they have to be the same as the reflection and transmission coefficients produced by the WSD approach. In case of mismatch, the case corresponding to the chosen weak formulation cannot be considered for further applications.

(3) The third criterion that must be fulfilled in the effective description of optical MMs is the conformity with the Casimir-Onsager reciprocity principle [65], which imposes some symmetry conditions for the nonlocal response function $\widehat{\mathbf{R}}(\omega, \mathbf{k})$.

In the following subsection we derive first of all the interface conditions for all nine cases and verify their conformity with our first criteria. Afterwards, we impose in the following subsections the other criteria and rule out an increasing number of cases to possibly express constitutive relations.

### A. Interface conditions analysis

The number of eigenmodes excited in a half-space or a slab MM is determined from the dispersion relations (cf. Sec. V and [33]). When spatial dispersion occurs, the classical interface conditions are not sufficient to compute the amplitudes of all eigenmodes. For every constitutive relation proposed in Table I, light-matter interaction is modeled differently. Therefore, one has to derive additional interface conditions for each case separately. In the literature they are not often calculated analytically, but rather introduced on phenomenological grounds (see, e.g., [19]). For that reason, solving the interface problems by exploiting the weak formulation of each wavelike equation in Table I represents one of our main contributions in this investigation.

Before starting, we recall Green's formula for the $\nabla\times$ derivative (see, e.g., [66]). For $\Omega$ a bounded domain in $\mathbb{R}^3$ with a Lipschitz boundary $\partial\Omega = \Gamma$, we define the space $\mathbf{H}(\mathbf{curl}, \Omega)$ as follows:

$$\mathbf{H}(\mathbf{curl}, \Omega) = \{\mathbf{u} \in \mathbf{L}^2(\Omega),\ \nabla \times \mathbf{u} \in \mathbf{L}^2(\Omega)\}.$$

For $\mathbf{u}$ and $\mathbf{v}$ two vector fields in the space $\mathbf{H}(\mathbf{curl}, \Omega)$, we have the following Green's formula:

$$\int_\Omega \mathbf{u} \cdot (\nabla \times \mathbf{v})\, d\mathbf{r} = \int_\Omega (\nabla \times \mathbf{u}) \cdot \mathbf{v}\, d\mathbf{r} + \int_\Gamma (\mathbf{u} \times \mathbf{n}) \cdot \mathbf{v}_t\, d\mathbf{s}, \tag{26}$$

where $\mathbf{v}_t$ represents the tangential component of the vector field $\mathbf{v}$ on the surface $\Gamma$ and $d\mathbf{s}$ represents the area of the surface element on $\Gamma$. The normal vector $\mathbf{n}$ is outwardly directed from $\Omega$.

For $\mathbf{E}$ a vector field defined in the upper-half space $\mathbb{R}^3_+$, we denote by $\mathbf{E}_+$ its trace on the surface $\Gamma$. Conversely, for $\mathbf{E}$ a vector field defined in the lower-half space $\mathbb{R}^3_-$, we denote by $\mathbf{E}_-$ its trace on the surface $\Gamma$.

The strategy of deriving the interface conditions consists in writing the corresponding weak formulations of the wavelike equations. A weak formulation requires formally writing the equations in the function space $\mathcal{D}'(\mathbb{R}^3)$ (cf. Sec. VI). Then, we can derive the additional interface conditions by re-establishing the strong formulations via partial integration.

We emphasize that because of reasons linked to the discontinuity of the material parameters on the surface $\Gamma$, it is not always possible to write symmetric weak formulations. Namely, the bilinear form associated with the respective weak formulation is not always symmetric, i.e., the order of derivatives applied on the vector field $\mathbf{E}$ is not always the same as the order of derivatives applied on the test function $\mathbf{\Phi}$. Actually, our plan is not necessarily shifting all the $\nabla\times$ derivatives located before the parameters $\alpha$ and $\gamma$ to the test function $\mathbf{\Phi}$.

Some cases will be already excluded due to violation of the first criterion. Regarding the other cases, they need to fulfill the second and the third criteria. As previously mentioned, case 1 coincides implicitly with the model given by the Taylor approach studied in [34], in this paper we will just recall its corresponding interface conditions for a self-contained paper.

#### 1. Interface conditions for case 1

We recall the wavelike equation corresponding to case 1:

$$\nabla \times \nabla \times \mathbf{E} = k_0^2\tilde{\varepsilon}\mathbf{E} + k_0^2\nabla \times \tilde{\alpha}\nabla \times \mathbf{E} + \nabla \times \nabla \times \tilde{\gamma}\nabla \times \nabla \times \mathbf{E}. \tag{27}$$

*a. Weak formulation.* Equation (27) is understood in the generalized sense. We suppose that $\mathbf{E}$ has the following regularities:

$$\mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3), \quad \nabla \times \mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3), \quad \text{and} \quad \nabla \times \nabla \times \mathbf{E} \in \mathbf{L}^2_{\mathrm{loc}}(\mathbb{R}^3_-), \tag{28}$$

which, in particular, implies that $\mathbf{E}$, $\nabla \times \mathbf{E}$, and $\nabla \times \nabla \times \mathbf{E}$ cannot contain delta distributions at the interface. For $\mathbf{\Phi} \in \mathbf{C}_0^\infty(\mathbb{R}^3)$, we define the weak formulation corresponding to the Eq. (27) as follows:

$$\int_{\mathbb{R}^3} (\nabla \times \mathbf{E}) \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r} = k_0^2 \int_{\mathbb{R}^3_+} \mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} \varepsilon\mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} (\alpha\nabla \times \mathbf{E}) \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r} + \int_{\mathbb{R}^3_-} (\gamma\nabla \times \nabla \times \mathbf{E}) \cdot (\nabla \times \nabla \times \mathbf{\Phi})\, d\mathbf{r}. \tag{29}$$

We say that $\mathbf{E}$ is a weak solution to the wavelike Eq. (27) if it has the regularities (28) and satisfies the weak formulation (29).

*b. Interface conditions.* (See [34])

$$(\mathbf{E}_+ - \mathbf{E}_-) \times \mathbf{n} = 0, \tag{30a}$$

$$(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + k_0^2(\alpha\nabla \times \mathbf{E}_-) \times \mathbf{n} + (\nabla \times \gamma\nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0, \tag{30b}$$

$$(\gamma\nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0. \tag{30c}$$

The third term in the second interface condition (30b) is purely coming from nonlocality. The interface condition (30c) represents the one what we call in this context "the additional interface condition." In contrast to Maxwell's equations with the classical constitutive relations, we only have two interface conditions given by

$$(\mathbf{E}_+ - \mathbf{E}_-) \times \mathbf{n} = 0, \tag{31a}$$

$$(\nabla \times \mathbf{E}_+ - \mu^{-1}\nabla \times \mathbf{E}_-) \times \mathbf{n} = 0. \tag{31b}$$

Here $\mu$ represents the magnetic permeability, that can be written as follows: $\mu = (1 - k_0^2\alpha)^{-1}$ (cf. Sec. IV B).

### *2. Analysis of case 2*

The wavelike equation corresponding to case 2 reads

$$\nabla \times \nabla \times \mathbf{E} = k_0^2\tilde{\varepsilon}\mathbf{E} + k_0^2\nabla \times \tilde{\alpha}\nabla \times \mathbf{E} + \nabla \times \tilde{\gamma}\nabla \times \nabla \times \nabla \times \mathbf{E}. \tag{32}$$

*a. Weak formulation.* In the generalized sense, for $\mathbf{E}$ verifying the following regularities:

$$\mathbf{E} \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3), \quad \nabla \times \mathbf{E} \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3), \quad \text{and} \quad \nabla \times \nabla \times \nabla \times \mathbf{E} \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3_-), \tag{33}$$

and for $\mathbf{\Phi} \in \mathbf{C}_0^\infty(\mathbb{R}^3)$, the weak formulation of the wavelike Eq. (32) reads

$$\begin{aligned}\int_{\mathbb{R}^3} (\nabla \times \mathbf{E}) \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r} = k_0^2 \int_{\mathbb{R}^3_+} \mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} \varepsilon\mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} (\alpha\nabla \times \mathbf{E}) \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r} \\ + \int_{\mathbb{R}^3_-} (\gamma\nabla \times \nabla \times \nabla \times \mathbf{E}) \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r}.\end{aligned} \tag{34}$$

We say that $\mathbf{E}$ is a weak solution to the wavelike Eq. (32) if it has the regularities (33) and satisfies the weak formulation (34).

*b. Interface conditions.* First interface condition: It represents a natural interface condition obtained from the fact that $\nabla \times \mathbf{E} \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3)$ and not from the weak formulations. This is why it represents also the first interface condition for all the other cases. The proof will be presented only in this paragraph, and for the other cases we cite it without showing the proof again.

For $\mathbf{E} \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3)$ and $\nabla \times \mathbf{E} \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3)$ we have

$$\forall \mathbf{\Phi} \in \mathbf{C}_0^\infty(\mathbb{R}^3) : \int_{\mathbb{R}^3} (\nabla \times \mathbf{E}) \cdot \mathbf{\Phi}\, d\mathbf{r} = \int_{\mathbb{R}^3} \mathbf{E} \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r}. \tag{35}$$

Integrating by parts on each half-space, we get

$$\begin{aligned}\int_{\mathbb{R}^3} (\nabla \times \mathbf{E}) \cdot \mathbf{\Phi}\, d\mathbf{r} &= \int_{\mathbb{R}^3_+} (\nabla \times \mathbf{E}) \cdot \mathbf{\Phi}\, d\mathbf{r} + \int_{\mathbb{R}^3_-} (\nabla \times \mathbf{E}) \cdot \mathbf{\Phi}\, d\mathbf{r} \\ &= \int_{\mathbb{R}^3_+} \mathbf{E} \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r} + \int_{\mathbb{R}^3_-} \mathbf{E} \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r} + \int_\Gamma (\mathbf{E}_+ \times \mathbf{n} - \mathbf{E}_- \times \mathbf{n}) \cdot \mathbf{\Phi}_t\, ds \\ &= \int_{\mathbb{R}^3} \mathbf{E} \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r} + \int_\Gamma (\mathbf{E}_+ \times \mathbf{n} - \mathbf{E}_- \times \mathbf{n}) \cdot \mathbf{\Phi}_t\, ds.\end{aligned} \tag{36}$$

Since $\mathbf{\Phi}$ is arbitrary, we get from Eqs. (35) and (36) the first interface condition, given by

$$(\mathbf{E}_+ - \mathbf{E}_-) \times \mathbf{n} = [\mathbf{E} \times \mathbf{n}] = 0, \tag{37}$$

where $[\cdot]$ represents the jump across the surface $\Gamma$.

Second interface condition: For $\mathbf{E}$ a weak solution to (32) with the regularities $\mathbf{E} \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3)$, $\nabla \times \nabla \times \mathbf{E} \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3)$, by taking first a test function $\mathbf{\Phi} \in \mathbf{C}_0^\infty(\mathbb{R}^3_+)$ and then in $\mathbf{C}_0^\infty(\mathbb{R}^3_-)$, we can show that

$$\nabla \times \nabla \times \mathbf{E} = k_0^2\mathbf{E}, \quad \text{for a.e. } x \in \mathbb{R}^3_+, \tag{38a}$$

$$\nabla \times \nabla \times \mathbf{E} = k_0^2\varepsilon\mathbf{E} + k_0^2\nabla \times \alpha\nabla \times \mathbf{E} + \nabla \times \gamma\nabla \times \nabla \times \nabla \times \mathbf{E}, \quad \text{for a.e. } x \in \mathbb{R}^3_-. \tag{38b}$$

For more details, we cite Ref. [34]. We integrate by parts Eq. (34) in a way that we reconstruct in the integrand Eqs. (38a) and (38b). Then we write volume integrals on the left-hand side and surface integrals on the right-hand side, as follows:

$$\int_{\mathbb{R}^3} (\nabla \times \nabla \times \mathbf{E}) \cdot \mathbf{\Phi}\, d\mathbf{r} - \int_{\mathbb{R}^3_+} k_0^2 \mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} - \int_{\mathbb{R}^3_-} \big( k_0^2 \varepsilon \mathbf{E} + k_0^2 \nabla \times \alpha \nabla \times \mathbf{E} + \nabla \times \gamma \nabla \times \nabla \times \nabla \times \mathbf{E} \big) \cdot \mathbf{\Phi}\, d\mathbf{r}$$
$$= \int_\Gamma \big[ (\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + k_0^2 (\alpha \nabla \times \mathbf{E}_-) \times \mathbf{n} + (\gamma \nabla \times \nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n} \big] \cdot \mathbf{\Phi}_t\, d\mathbf{s}. \tag{39}$$

Using Eqs. (38a) and (38b), the left-hand side in (39) vanishes. Since $\mathbf{\Phi}$ is arbitrary, we get the second interface condition

$$(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + k_0^2 (\alpha \nabla \times \mathbf{E}_-) \times \mathbf{n} + (\gamma \nabla \times \nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0. \tag{40}$$

We remark that the number of interface conditions for case 2 is less than the number of the reflected and transmitted fields. More precisely, from the dispersion relation we know that there are two transmitted fields at the interface $\Gamma$ into $\mathbb{R}^3_-$ and one reflected field into $\mathbb{R}^3_+$. Hence, there are three unknowns, whereas we got only two equations (interface conditions) to identify them. This system of equations is underdetermined and can, therefore, lead to ambiguous solutions. In addition, case 2 does not admit any other weak formulation that may give the necessary number of interface conditions. Thus, the corresponding case is not adequate for describing the propagation of light in Padé MMs. For the same reason, case 8 will be excluded from the investigation of Padé MMs.

### *3. Analysis of case 3*

The wavelike equation corresponding to case 3 is given by

$$\nabla \times \nabla \times \mathbf{E} = k_0^2 \tilde{\varepsilon} \mathbf{E} + k_0^2 \nabla \times \tilde{\alpha} \nabla \times \mathbf{E} + \tilde{\gamma} \nabla \times \nabla \times \nabla \times \nabla \times \mathbf{E}. \tag{41}$$

*a. Weak formulation.* In the generalized sense, for $\mathbf{E}$ verifying the following regularities:

$$\mathbf{E} \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3), \quad \nabla \times \mathbf{E} \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3), \quad \text{and} \quad \nabla \times \nabla \times \nabla \times \nabla \times \mathbf{E} \in \mathbf{L}^2_{\text{loc}}(\mathbb{R}^3_-), \tag{42}$$

and for $\mathbf{\Phi} \in \mathbf{C}_0^\infty(\mathbb{R}^3)$, we define the weak formulation corresponding to the wavelike Eq. (41) as follows:

$$\int_{\mathbb{R}^3} (\nabla \times \mathbf{E}) \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r} = k_0^2 \int_{\mathbb{R}^3_+} \mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} \varepsilon \mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} (\alpha \nabla \times \mathbf{E}) \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r}$$
$$+ \int_{\mathbb{R}^3_-} (\gamma \nabla \times \nabla \times \nabla \times \nabla \times \mathbf{E}) \cdot \mathbf{\Phi}\, d\mathbf{r}. \tag{43}$$

We say that $\mathbf{E}$ is a weak solution to the wavelike Eq. (41) if it has the regularities (42) and satisfies the weak formulation (43).

*b. Interface conditions.* We follow the same principle as in case 2. By means of the equations

$$\nabla \times \nabla \times \mathbf{E} = k_0^2 \mathbf{E}, \quad \text{for a.e. x} \in \mathbb{R}^3_+, \tag{44a}$$

$$\nabla \times \nabla \times \mathbf{E} = k_0^2 \varepsilon \mathbf{E} + k_0^2 \nabla \times \alpha \nabla \times \mathbf{E} + \gamma \nabla \times \nabla \times \nabla \times \nabla \times \mathbf{E}, \quad \text{for a.e. x} \in \mathbb{R}^3_-. \tag{44b}$$

We need to apply just one partial integration on the weak formulation (43) to recover Eqs. (44a) and (44b). Then we obtain

$$\int_{\mathbb{R}^3_+} \big( \underbrace{\nabla \times \nabla \times \mathbf{E} - k_0^2 \mathbf{E}}_{=0} \big) \cdot \mathbf{\Phi}\, d\mathbf{r} + \int_{\mathbb{R}^3_-} \big( \underbrace{\nabla \times \nabla \times \mathbf{E} - k_0^2 \varepsilon \mathbf{E} - k_0^2 \nabla \times \alpha \nabla \times \mathbf{E} - \gamma \nabla \times \nabla \times \nabla \times \nabla \times \mathbf{E}}_{=0} \big) \cdot \mathbf{\Phi}\, d\mathbf{r}$$
$$= \int_\Gamma \big[ (\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + k_0^2 (\alpha \nabla \times \mathbf{E}_-) \times \mathbf{n} \big] \cdot \mathbf{\Phi}_t\, d\mathbf{s} = 0. \tag{45}$$

For an arbitrary test function $\mathbf{\Phi}$, we get the second interface condition

$$(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + k_0^2 (\alpha \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0. \tag{46}$$

Because of the same reason as in case 2, which is a lack of additional interface conditions, case 3 will be excluded form the study. This inconsistency is due to the material parameters $\gamma$ that represents the nonlocal effect, which is located in the first position in the fourth order differential operator. Cases 6 and 9 share the same nature of the fourth order term. Hence, case 9 will be immediately excluded in turn. Whereas, because of the nature of the second order term in case 6, we have a chance to keep it initially in the investigation.

#### *4. Analysis of case 4*

We recall the wavelike equation corresponding to case 4:

$$\nabla \times \nabla \times \mathbf{E} = k_0^2 \tilde{\varepsilon} \mathbf{E} + k_0^2 \nabla \times \nabla \times \tilde{\alpha} \mathbf{E} + \nabla \times \nabla \times \tilde{\gamma} \nabla \times \nabla \times \mathbf{E}. \tag{47}$$

*a. Weak formulation.* Equation (47) is understood in the generalized sense. For $\mathbf{E}$ verifying regularities (28) and for $\mathbf{\Phi} \in \mathbf{C}_0^\infty(\mathbb{R}^3)$, we define the weak formulation corresponding to case 4 as follows:

$$\begin{aligned}\int_{\mathbb{R}^3} (\nabla \times \mathbf{E}) \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r} &= k_0^2 \int_{\mathbb{R}^3_+} \mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} \varepsilon \mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} (\alpha \mathbf{E}) \cdot (\nabla \times \nabla \times \mathbf{\Phi})\, d\mathbf{r} \\ &\quad + \int_{\mathbb{R}^3_-} (\gamma \nabla \times \nabla \times \mathbf{E}) \cdot (\nabla \times \nabla \times \mathbf{\Phi})\, d\mathbf{r}. \end{aligned} \tag{48}$$

We say that $\mathbf{E}$ is a weak solution to the wavelike Eq. (47) if it has the regularities (28) and satisfies the weak formulation (48).

*b. Interface conditions.* We follow the same principle as in cases 2 and 3. By means of the equations

$$\nabla \times \nabla \times \mathbf{E} = k_0^2 \mathbf{E}, \quad \text{for a.e. } \mathrm{x} \in \mathbb{R}^3_+, \tag{49a}$$

$$\nabla \times \nabla \times \mathbf{E} = k_0^2 \varepsilon \mathbf{E} + k_0^2 \nabla \times \nabla \times \alpha \mathbf{E} + \nabla \times \nabla \times \gamma \nabla \times \nabla \times \mathbf{E}, \quad \text{for a.e. } \mathrm{x} \in \mathbb{R}^3_-, \tag{49b}$$

after partial integration, the weak formulation (48) leads to the following formula:

$$\begin{aligned}&\int_{\mathbb{R}^3_+} \big(\underbrace{\nabla \times \nabla \times \mathbf{E} - k_0^2 \mathbf{E}}_{=0}\big) \cdot \mathbf{\Phi}\, d\mathbf{r} + \int_{\mathbb{R}^3_-} \big(\underbrace{\nabla \times \nabla \times \mathbf{E} - k_0^2 \varepsilon \mathbf{E} - k_0^2 \nabla \times \nabla \times \alpha \mathbf{E} - \nabla \times \nabla \times \gamma \nabla \times \nabla \times \mathbf{E}}_{=0}\big) \cdot \mathbf{\Phi}\, d\mathbf{r} \\ &= \int_\Gamma \big[(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + k_0^2 (\nabla \times \alpha \mathbf{E}_-) \times \mathbf{n} + (\nabla \times \gamma \nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n}\big] \cdot \mathbf{\Phi}_t\, ds \\ &\quad + \int_\Gamma \big[k_0^2 (\alpha \mathbf{E}_-) \times \mathbf{n} + (\gamma \nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n}\big] \cdot (\nabla \times \mathbf{\Phi})_t\, d\mathbf{s} = 0. \end{aligned} \tag{50}$$

The left-hand side is null because of Eqs. (49a) and (49b). To get out the second interface conditions, we choose the test function $\mathbf{\Phi}$ given by

$$\mathbf{\Phi}(\mathbf{r}) = [\Phi_1(x, y)\eta(z), \Phi_2(x, y)\eta(z), 0]^T, \tag{51}$$

where $\Phi_1, \Phi_2$ are arbitrary functions in $\mathbf{C}_0^\infty(\mathbb{R}^2)$ and $\eta(z) \in \mathbf{C}_0^\infty(\mathbb{R})$, satisfying $\eta|_{\{z<\mathrm{d}\}} = 1$. On the surface $\Gamma$ we have

$$\mathbf{\Phi}|_\Gamma = (\Phi_1, \Phi_2, 0)^T, \quad \nabla \times \mathbf{\Phi}|_\Gamma = (0, 0, \partial_x \Phi_2 - \partial_y \Phi_1)^T.$$

Then we obtain

$$(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + k_0^2 (\nabla \times \alpha \mathbf{E}_-) \times \mathbf{n} + (\nabla \times \gamma \nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0. \tag{52}$$

To extract the third interface conditions, we choose the test function $\mathbf{\Phi}$ this time in the form

$$\mathbf{\Phi}(\mathbf{r}) = [\Phi_2(x, y) z \eta(z), -\Phi_1(x, y) z \eta(z), 0]^T, \tag{53}$$

such that $\Phi_1, \Phi_2 \in \mathbf{C}_0^\infty(\mathbb{R}^2)$ and $\eta(z) \in \mathbf{C}_0^\infty(\mathbb{R})$, satisfying $\eta|_{\{z<\mathrm{d}\}} = 1$. On the surface $\Gamma$ we have

$$\mathbf{\Phi}|_\Gamma = (0, 0, 0)^T,$$
$$\nabla \times \mathbf{\Phi}|_\Gamma = (\Phi_1, \Phi_2, 0)^T.$$

Since $\Phi_1$ and $\Phi_2$ are arbitrary, we have

$$k_0^2 (\alpha \mathbf{E}_-) \times \mathbf{n} + (\gamma \nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0. \tag{54}$$

Now we set all the interface conditions for the case 4 as follows:

$$(\mathbf{E}_+ - \mathbf{E}_-) \times \mathbf{n} = 0, \tag{55a}$$

$$(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + k_0^2 (\nabla \times \alpha \mathbf{E}_-) \times \mathbf{n} + (\nabla \times \gamma \nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0, \tag{55b}$$

$$k_0^2 (\alpha \mathbf{E}_-) \times \mathbf{n} + (\gamma \nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0. \tag{55c}$$

The first criterion is satisfied by the case 4 since we have three interface conditions as required.

#### *5. Analysis of case 5*

The wave equation corresponding to case 5 reads

$$\nabla \times \nabla \times \mathbf{E} = k_0^2\tilde{\varepsilon}\mathbf{E} + k_0^2\nabla \times \nabla \times \tilde{\alpha}\mathbf{E} + \nabla \times \tilde{\gamma}\nabla \times \nabla \times \nabla \times \mathbf{E}. \tag{56}$$

*a. Weak formulation.* Equation (56) is understood in the generalized sense. For $\mathbf{E}$ verifying regularities (33) and for $\mathbf{\Phi} \in \mathbf{C}_0^\infty(\mathbb{R}^3)$, we define the weak formulation corresponding to case 5:

$$\int_{\mathbb{R}^3}(\nabla \times \mathbf{E}) \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r} = k_0^2 \int_{\mathbb{R}^3_+} \mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} \varepsilon\mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} (\alpha\mathbf{E}) \cdot (\nabla \times \nabla \times \mathbf{\Phi})\, d\mathbf{r}$$
$$+ \int_{\mathbb{R}^3_-} (\gamma\nabla \times \nabla \times \nabla \times \mathbf{E}) \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r}. \tag{57}$$

We say that $\mathbf{E}$ is a weak solution to the wavelike Eq. (56) if it has the regularities (33) and satisfies the weak formulation (57).

*b. Interface conditions.* We follow the same process as in case 4. By taking the choices (51) and (53) for the test function $\mathbf{\Phi}$, we get the following interface conditions:

$$(\mathbf{E}_+ - \mathbf{E}_-) \times \mathbf{n} = 0, \tag{58a}$$
$$(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + k_0^2(\nabla \times \alpha\mathbf{E}_-) \times \mathbf{n} + (\gamma\nabla \times \nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0, \tag{58b}$$
$$k_0^2(\alpha\mathbf{E}_-) \times \mathbf{n} = 0. \tag{58c}$$

We obtained three interface conditions as required, then the first criterion is satisfied by the case 5.

#### *6. Analysis of case 6*

We recall the wave equation corresponding to case 6:

$$\nabla \times \nabla \times \mathbf{E} = k_0^2\tilde{\varepsilon}\mathbf{E} + k_0^2\nabla \times \nabla \times \tilde{\alpha}\mathbf{E} + \tilde{\gamma}\nabla \times \nabla \times \nabla \times \nabla \times \mathbf{E}. \tag{59}$$

The nature of the second order term in this case gives the possibility to keep it initially in the study, even if the nonlocal material parameter $\tilde{\gamma}$ is located in the first position as in cases 3 and 9. In such equations, the effect of the nonlocal parameter does not appear explicitly in the interface conditions, but it is present on the level of the dispersion relation.

*a. Weak formulation.* In the generalized sense, for $\mathbf{E}$ verifying regularities (42) and for $\mathbf{\Phi} \in \mathbf{C}_0^\infty(\mathbb{R}^3)$, we define the weak formulation corresponding to case 6:

$$\int_{\mathbb{R}^3}(\nabla \times \mathbf{E}) \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r} = k_0^2 \int_{\mathbb{R}^3_+} \mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} \varepsilon\mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} (\alpha\mathbf{E}) \cdot (\nabla \times \nabla \times \mathbf{\Phi})\, d\mathbf{r}$$
$$+ \int_{\mathbb{R}^3_-} (\gamma\nabla \times \nabla \times \nabla \times \nabla \times \mathbf{E}) \cdot \mathbf{\Phi}\, d\mathbf{r}. \tag{60}$$

We say that $\mathbf{E}$ is a weak solution to the wavelike Eq. (59) if it has the regularities (42) and satisfies the weak formulation (60).

*b. Interface conditions.* Analogous to cases 2, 4, and 5, we derive the interface conditions corresponding to case 6, given by

$$(\mathbf{E}_+ - \mathbf{E}_-) \times \mathbf{n} = 0, \tag{61a}$$
$$(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + k_0^2(\nabla \times \alpha\mathbf{E}_-) \times \mathbf{n} = 0, \tag{61b}$$
$$k_0^2(\alpha\mathbf{E}_- \times \mathbf{n}) = 0. \tag{61c}$$

#### *7. Analysis of case 7*

We recall the wave equation corresponding to case 7:

$$\nabla \times \nabla \times \mathbf{E} = k_0^2\tilde{\varepsilon}\mathbf{E} + k_0^2\tilde{\alpha}\nabla \times \nabla \times \mathbf{E} + \nabla \times \nabla \times \tilde{\gamma}\nabla \times \nabla \times \mathbf{E}. \tag{62}$$

*a. Weak formulation.* Equation (62) is understood in the generalized sense. For $\mathbf{E}$ verifying regularities (28) and for $\mathbf{\Phi} \in \mathbf{C}_0^\infty(\mathbb{R}^3)$, we define the weak formulation corresponding to case 7:

$$\int_{\mathbb{R}^3}(\nabla \times \mathbf{E}) \cdot (\nabla \times \mathbf{\Phi})\, d\mathbf{r} = k_0^2 \int_{\mathbb{R}^3_+} \mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} \varepsilon\mathbf{E} \cdot \mathbf{\Phi}\, d\mathbf{r} + k_0^2 \int_{\mathbb{R}^3_-} (\alpha\nabla \times \nabla \times \mathbf{E}) \cdot \mathbf{\Phi}\, d\mathbf{r}$$
$$+ \int_{\mathbb{R}^3_-} (\gamma\nabla \times \nabla \times \mathbf{E}) \cdot (\nabla \times \nabla \times \mathbf{\Phi})\, d\mathbf{r}. \tag{63}$$

We say that $\mathbf{E}$ is a weak solution to the wavelike Eq. (62) if it satisfies the weak formulation (63) and has the following regularities (28).

*b. Interface conditions.* Analogous to cases 2, 4, 5, and 6, we derive the interface conditions corresponding to case 7, given by

$$(\mathbf{E}_+ - \mathbf{E}_-) \times \mathbf{n} = 0, \tag{64a}$$

$$(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + (\nabla \times \gamma \nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0, \tag{64b}$$

$$(\gamma \nabla \times \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0. \tag{64c}$$

The first criterion in the checklist consisted on deriving the right number of interface conditions, which are necessary for computing the amplitudes of all the propagating modes in MMs. The dispersion relation of each case reveals that we need exactly three interface conditions on each surface. Cases 1, 4, 5, 6, and 7 survived, because of fulfilling this criterion. The weak formulation of cases 2, 3, 8, and 9 are not adequate to give the additional interface condition. Hence, the systems of equations necessary for defining the amplitudes of all the propagating modes are underdetermined, which implies that these cases will be rejected. In the next subsection we study reflection and transmission coefficients for the remaining cases and continue to consider only those that agree in the limiting case of a vanishing nonlocality with expressions obtained from a WSD.

### B. Reflection and transmission coefficients analysis

We recall that the main motivation for proceeding with the nonlocal analysis is to overcome the limitations exhibited when using the local theory. This fact implies that the local approach must be contained in the nonlocal approach. Meaning, by taking the limit of the parameters representing the nonlocality to zero, we have to recover the local models. On the level of the mathematical models this can be clearly seen from equations in Table I. However, for more accurate analysis we need to check the limit of the corresponding interface conditions and also the produced reflected and transmitted modes. Already, by using the local approach, we know that we have only one reflected and one transmitted mode in each half-space. Besides, for the nonlocal models we have one reflected mode together with two transmitted modes. This fact implies that by taking the limit of the material parameter $\gamma$ to zero, one of that two transmitted modes must vanish. Actually, we can also observe that on the level of the interface conditions. In other words, one of the three interface conditions vanish when $\gamma$ tends to zero. For this reason, we present a quick check for the remaining cases 1, 4, 5, 6, and 7. Then, we need to recall the principle of Fresnel equations that serve to compute the amplitudes of the reflected and transmitted modes, and we give their formulas for the surviving cases.

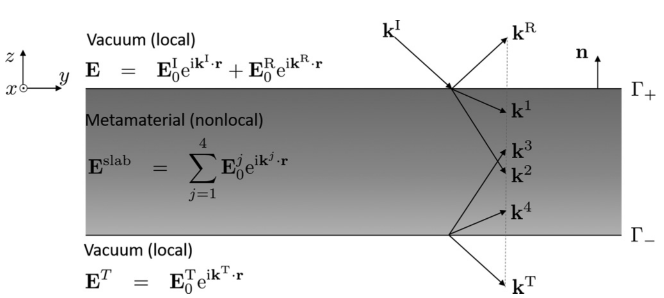


FIG. 2. Propagating modes inside a slab MM. We have two forward modes defined on the the surface $\Gamma^+$ and the other backward modes are defined on the surface $\Gamma^-$.

*a. Analysis of the remaining cases using the second criterion. a. Analysis of case 1.* We take the limit of $\gamma$ to zero in the interface conditions corresponding to case 1, we get

$$(\mathbf{E}_+ - \mathbf{E}_-) \times \mathbf{n} = 0,$$
$$(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + k_0^2(\alpha \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0.$$

We notice that using the notation $\mu = (1 - k_0^2\alpha)^{-1}$ (cf. Sec. IV B), the limit interface conditions take the same formulas as in the presence of WSD, as follows:

$$(\mathbf{E}_+ - \mathbf{E}_-) \times \mathbf{n} = 0,$$
$$(\nabla \times \mathbf{E}_+ - \mu^{-1} \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0.$$

Which implies that the second criterion is fulfilled by the first case.

*b. Analysis of case 4.* In the presence of WSD, the wavelike Eq. (47) reads

$$\nabla \times \nabla \times \mathbf{E} = k_0^2 \tilde{\varepsilon} \mathbf{E} + k_0^2 \nabla \times \nabla \times \tilde{\alpha} \mathbf{E}.$$

Its corresponding interface conditions are given by

$$(\mathbf{E}_+ - \mathbf{E}_-) \times \mathbf{n} = 0,$$
$$(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + k_0^2(\nabla \times \alpha \mathbf{E}_-) \times \mathbf{n} = 0.$$

On the other hand, the limiting interface conditions for case 4, when $\gamma$ tends to zero read

$$(\mathbf{E}_+ - \mathbf{E}_-) \times \mathbf{n} = 0,$$
$$(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} + k_0^2(\nabla \times \alpha \mathbf{E}_-) \times \mathbf{n} = 0,$$
$$k_0^2(\alpha \mathbf{E}_-) \times \mathbf{n} = 0.$$

We can clearly see that the presence of the material parameter $\alpha$ in the third interface condition prevents it to vanish. Meanwhile, the dispersion relation for $\gamma \to 0$ states that there is only one transmitted field in the half-space filled with the MM. In this case, the system of equations is overdetermined and

TABLE III. Substitution of the coefficients $b_j^{(\mathrm{TE})}$ and $c_j^{(\mathrm{TE})}$, for $1 \leqslant j \leqslant 4$, corresponding to TE polarization for cases 1 and 7; with $(\mathbf{k}^j)^2 = (k_y)^2 + (k_z^j)^2$.

| | Case 1 | Case 7 |
|---|---|---|
| $b_j^{(\mathrm{TE})}$ | $-\left[1 - k_0^2\alpha_y - \gamma_x(\mathbf{k}^j)^2\right]k_z^j$ | $-\left[1 - \gamma_x(\mathbf{k}^j)^2\right]k_z^j$ |
| $c_j^{(\mathrm{TE})}$ | $\gamma_x(\mathbf{k}^j)^2$ | $\gamma_x(\mathbf{k}^j)^2$ |

leads to unphysical results. From a physical point of view, the third interface condition states that the tangential component of $\mathbf{E}_-$ is zero at the interface. Linking this with the first (natural) interface condition, would suggest that the tangential component of $\mathbf{E}_+$, i.e., in vacuum is zero. Since the total field in the incidence half-space is $\mathbf{E}^I + \mathbf{E}^R$, the transversal components of the incident and reflected fields totally cancel, independent of the angle of incidence, the permittivity contrast between the two half-spaces and polarization. The result is even stranger when the MM is lossy, i.e., when $\operatorname{Im} \varepsilon > 0$, where some energy is absorbed in the MM half-space. However, here we obtain a total reflection phenomenon, without incorporating loss. This is clearly a contradiction to the classical reflection and transmission phenomena between two local materials.

This case represents a counterexample that shows that even if we have the exact number of interface conditions in the presence of SSD, that gives the exact number of reflected and transmitted modes in turn, it is not possible to reproduce the local forward and backward modes after taking the limit of the nonlocal parameter to zero. To conclude, case 4 does not fulfill the second criterion, and will, therefore, be excluded.

For the same reason of no conformity with the modes produced by the WSD when taking the limit of $\gamma$ to zero, cases 5 and 6 will be excluded from the investigation of Padé MMs.

*c. Analysis of case 7.* When we take the limit of $\gamma$ to zero, i.e., in the presence of WSD, the wavelike Eq. (62) reads

$$\nabla \times \nabla \times \mathbf{E} = k_0^2 \tilde{\varepsilon} \mathbf{E} + k_0^2 \tilde{\alpha} \nabla \times \nabla \times \mathbf{E}.$$

Its corresponding interface conditions are given by

$$(\mathbf{E}_+ - \mathbf{E}_-) \times \mathbf{n} = 0,$$
$$(\nabla \times \mathbf{E}_+ - \nabla \times \mathbf{E}_-) \times \mathbf{n} = 0.$$

They represent, exactly, the same limiting interface conditions for case 7 when $\gamma$ tends to zero. Which implies that the second criterion is fulfilled in this case.

*2. Fresnel coefficients for cases 1 and 7.* We denote the incident, reflected, and transmitted fields by the superscripts I, R, and T, respectively, and we refer to each one of them by $*$. Inside the slab MM, we have four propagating linearly independent eigenmodes. Two modes are forward and the other two ones are backward, we refer to each one by $\mathbf{E}^j$, for $j \in \{1, 2, 3, 4\}$. The total field in the incidence half-space is given by $\mathbf{E}^I + \mathbf{E}^R$. Inside the MM slab, the total field is the sum of the propagating eigenmodes. It is written $\mathbf{E}^{\text{slab}} = \sum_{j=1}^4 \mathbf{E}^j$. In the transmission plane, we have just one transmitted field $\mathbf{E}^T$ (see Fig. 2).

The reflection ($\rho$) and transmission ($\tau$) coefficients represent the ratios of the amplitudes of the reflected and transmitted waves to the incident wave. We compute the amplitudes of these fields by using Fresnel formulas. We start by plugging the plane wave ansatzes into the interface conditions defined on the surfaces $\Gamma_+$ and $\Gamma_-$ . For both polarizations, we have

(i) TE polarization:

$$\mathbf{E}^* = \mathbf{E}_0^* \exp(i\mathbf{k}^* \cdot \mathbf{r}), \text{ with } \mathbf{E}_0^* = (E_x^*, 0, 0)^T \text{ and } \mathbf{k}^* = (0, k_y, k_z^*)^T.$$
$$\mathbf{E}^j = \mathbf{E}_0^j \exp(i\mathbf{k}^* \cdot \mathbf{r}), \text{ with } \mathbf{E}_0^j = \left(E_x^j, 0, 0\right)^T \text{ and } \mathbf{k}^j = \left(0, k_y, k_z^j\right)^T.$$

(ii) TM polarization:

$$\mathbf{E}^* = \mathbf{E}_0^* \exp(i\mathbf{k}^* \cdot \mathbf{r}), \text{ with } \mathbf{E}_0^* = (0, E_y^*, E_z^*)^T \text{ and } \mathbf{k}^* = (0, k_y, k_z^*)^T.$$
$$\mathbf{E}^j = \mathbf{E}_0^j \exp(i\mathbf{k}^* \cdot \mathbf{r}), \text{ with } \mathbf{E}_0^j = \left(0, E_y^j, E_z^j\right)^T \text{ and } \mathbf{k}^j = \left(0, k_y, k_z^j\right)^T.$$

Using the divergence free equation for the electric displacement field $\mathbf{D}$, we get a relation between the $y$ and $z$ components of the electric field $\mathbf{E}$, that depends on the medium and is given by

$$E_y^* = -\frac{k_z^*}{k_y} E_z^*, \qquad \text{in } \Omega_- \cup \Omega_+, \tag{65a}$$

$$E_y^j = -\frac{\varepsilon_z k_z^*}{\varepsilon_y k_y} E_z^j, \qquad \text{in } \Omega_\delta. \tag{65b}$$

Then we get an algebraic system in the form

$$\mathcal{A} \ E \ = \mathcal{F}. \tag{66}$$

In total we have six unknowns for each polarization, such that $E = (E_{(P)}^R, E_{(P)}^1, E_{(P)}^2, E_{(P)}^3, E_{(P)}^4, E_{(P)}^T)^T$. The index $(P)$ refers to the $x$ component of the electric field for TE polarization and to the $z$ component of the electric field for TM polarization. The Fresnel matrix $\mathbb{A}$ is a $6 \times 6$ matrix, obtained from the fact that we have on each surface of the slab three interface conditions leading to three equations. The nonhomogeneity is described by the vector $\mathcal{F}$, which is relative to the incident plane wave.

In the sequel we give Fresnel matrices for TE and TM polarizations separately. Each matrix combines Fresnel coefficients for cases 1 and 7, followed with tables precising the explicit formulas of some coefficients. Fresnel formulas of case 1 are computed and written clearly in [39].

TABLE IV. Substitution of the coefficients $b_j^{(\mathrm{TM})}$ and $c_j^{(\mathrm{TM})}$, for $1 \leqslant j \leqslant 4$, corresponding to TM polarization for cases 1 and 7.

| | Case 1 | Case 7 |
|---|---|---|
| $b_j^{(\mathrm{TM})}$ | $-\big[1-\gamma_z(k_y)^2-\gamma_y\big(k_z^j\big)^2\big]\big[(k_y)^2+\frac{\varepsilon_z}{\varepsilon_y}\big(k_z^j\big)^2\big]$ | $-\big[1-\gamma_z(k_y)^2-\gamma_y\big(k_z^j\big)^2\big]\big[(k_y)^2+\frac{\varepsilon_z}{\varepsilon_y}\big(k_z^j\big)^2\big]$ |
| $c_j^{(\mathrm{TM})}$ | $\gamma_y\big[(k_y)^2+\frac{\varepsilon_z}{\varepsilon_y}\big(k_z^j\big)^2\big]k_z^j$ | $\gamma_y\big[(k_y)^2+\frac{\varepsilon_z}{\varepsilon_y}\big(k_z^j\big)^2\big]k_z^j$ |

*a. Fresnel matrix for TE polarization*

$$\mathcal{A}^{(\mathrm{TE})}=\begin{pmatrix} 1 & -1 & -1 & -1 & -1 & 0\\ k_z^R & b_1^{(\mathrm{TE})} & b_2^{(\mathrm{TE})} & b_3^{(\mathrm{TE})} & b_4^{(\mathrm{TE})} & 0\\ 0 & c_1^{(\mathrm{TE})} & c_2^{(\mathrm{TE})} & c_3^{(\mathrm{TE})} & c_4^{(\mathrm{TE})} & 0\\ 0 & -e^{ik_z^1d} & -e^{ik_z^2d} & -e^{ik_z^3d} & -e^{ik_z^4d} & 1\\ 0 & c_1^{(\mathrm{TE})}e^{ik_z^1d} & c_2^{(\mathrm{TE})}e^{ik_z^2d} & c_3^{(\mathrm{TE})}e^{ik_z^3d} & c_4^{(\mathrm{TE})}e^{ik_z^4d} & 0\\ 0 & b_1^{(\mathrm{TE})}e^{ik_z^1d} & b_2^{(\mathrm{TE})}e^{ik_z^2d} & b_3^{(\mathrm{TE})}e^{ik_z^3d} & b_4^{(\mathrm{TE})}e^{ik_z^4d} & k_z^T \end{pmatrix}, \tag{67}$$

and $\mathcal{F}^{(\mathrm{TE})}=(-1,-k_z^I,0,0,0,0)^T$. The coefficients $b_j^{(\mathrm{TE})}$ and $c_j^{(\mathrm{TE})}$ for both cases 1 and 7 and $1\leqslant j\leqslant 4$ are given in Table III.

*b. Fresnel matrix for TM polarization*

$$\mathcal{A}^{(\mathrm{TM})}=\begin{pmatrix} k_z^R & -\frac{\varepsilon_z}{\varepsilon_y}k_z^1 & -\frac{\varepsilon_z}{\varepsilon_y}k_z^2 & -\frac{\varepsilon_z}{\varepsilon_y}k_z^3 & -\frac{\varepsilon_z}{\varepsilon_y}k_z^4 & 0\\ (\mathbf{k}^R)^2 & b_1^{(\mathrm{TM})} & b_2^{(\mathrm{TM})} & b_3^{(\mathrm{TM})} & b_4^{(\mathrm{TM})} & 0\\ 0 & c_1^{(\mathrm{TM})} & c_2^{(\mathrm{TM})} & c_3^{(\mathrm{TM})} & c_4^{(\mathrm{TM})} & 0\\ 0 & -\frac{\varepsilon_z}{\varepsilon_y}k_z^1e^{ik_z^1d} & -\frac{\varepsilon_z}{\varepsilon_y}k_z^2e^{ik_z^2d} & -\frac{\varepsilon_z}{\varepsilon_y}k_z^3e^{ik_z^3d} & -\frac{\varepsilon_z}{\varepsilon_y}k_z^4e^{ik_z^4d} & 1\\ 0 & c_1^{(\mathrm{TM})}e^{ik_z^1d} & c_2^{(\mathrm{TM})}e^{ik_z^2d} & c_3^{(\mathrm{TM})}e^{ik_z^3d} & c_4^{(\mathrm{TM})}e^{ik_z^4d} & 0\\ 0 & b_1^{(\mathrm{TM})}e^{ik_z^1d} & b_2^{(\mathrm{TM})}e^{ik_z^2d} & b_3^{(\mathrm{TM})}e^{ik_z^3d} & b_4^{(\mathrm{TM})}e^{ik_z^4d} & (\mathbf{k}^T)^2 \end{pmatrix}, \tag{68}$$

and $\mathcal{F}^{(\mathrm{TM})}=[-1,-(\mathbf{k}^I)^2,0,0,0,0]^T$. The coefficients $b_j^{(\mathrm{TM})}$ and $c_j^{(\mathrm{TM})}$ for both cases 1 and 7 and $1\leqslant j\leqslant 4$ are given in Table IV. In both polarizations, the notation $(\mathbf{k}^*)^2$ means the sum $(k_y)^2+(k_z^*)^2$.

For both cases 1 and 7, taking the limit of the nonlocal parameter $\gamma$ towards zero, we get Fresnel matrices identical to those obtained by considering local constitutive relations. At the level of reflection and transmission coefficients, everything seems to be fine with cases 1 and 7. Then we have to move to the third and final criterion to end up the checking process.

To summarize this section, the second criterion consists on analyzing the reflection and transmission coefficients. As the models produced by the WSD are implicitly included in those obtained in the presence of SSD, then we checked whether it is really the case for the remaining proposed cases. Cases 1 and 7 survived, because we could reproduce the same equations as in the presence of WSD. Contrary to cases 4, 5, and 6, their weak formulations led to well posed systems; but the produced interface condition are not valid for computing the right reflection and transmission coefficients corresponding to the WSD.

### C. Casimir-Onsager reciprocity

Before setting the last criterion in the checklist, we draw the attention of the reader to the fact that in this study we assume that the unit cell of the MM is made of reciprocal constituents. Henceforth, the MMs by themselves are reciprocal as well. Now we check the requirement of time-reversal symmetry for the equations ingredients. In fact, it holds that the displacement field $\tilde{\mathbf{D}}(t,\mathbf{r})$ and the electric field $\tilde{\mathbf{E}}(t,\mathbf{r})$ are symmetric under time inversion, but antisymmetric under space inversion (parity); and the magnetic flux $\tilde{\mathbf{B}}(t,\mathbf{r})$ and the magnetic field $\tilde{\mathbf{H}}(t,\mathbf{r})$ are antisymmetric under time inversion, but symmetric under space inversion [67]. These symmetries of the fields have consequences on the symmetry of the nonlocal response function in Fourier space.

Due to this fact and to Eq. (8), we deduce that the response function $\mathbf{R}(\omega,\mathbf{r})$ must contain some symmetry as well. In the spatial frequency space it holds [68]

$$\widehat{R}_{ij}(\omega,\mathbf{k})=\widehat{R}_{ji}(\omega,\mathbf{k}), \tag{69}$$

which represents the Casimir-Onsager reciprocity principle for centrosymmetric structures. In the remaining case 7, we argue with Casimir-Onsager reciprocity that this case can only exist if $\alpha$ is a scalar, i.e., $\alpha_{ii}=\alpha_{jj}$, for $(i,j)\in\{x,y,z\}\times\{x,y,z\}$.

Let us first look at the term proportional to $\alpha$, i.e., $\gamma = 0$. The general expression of the nonlocal response function $\mathbf{R}$ is

$$D_i(\omega, \mathbf{r}) = R_{ij}(\omega, i\nabla)E_j(\omega, \mathbf{r}) = \varepsilon_{ij}E_j(\omega, \mathbf{r}) + c_{ijlm}\partial_l\partial_m E_j(\omega, \mathbf{r}). \tag{70}$$

According to symmetry condition (69), it must hold that

$$c_{ijlm} = c_{jilm}. \tag{71}$$

We further require that the electric field $\mathbf{E}$ is at least a $\mathcal{C}^2(\mathbb{R}^3_-)$ function. Consequently, according to the equality of mixed partials (Schwarz's theorem), the second-order derivatives can be interchanged which renders

$$c_{ijlm}\partial_l\partial_m E_j = c_{ijlm}\partial_m\partial_l E_j = c_{ijml}\partial_l\partial_m E_j.$$

In the first equality we put the fact that $\mathbf{E} \in \mathcal{C}^2(\mathbb{R}^3_-)$ and in the second equation we simply relabeled the indices. In fact, we have

$$c_{ijlm} = c_{jilm} = c_{ijml} = c_{jiml}. \tag{72}$$

These are the fundamental symmetry conditions for the fourth-rank tensor $c_{ijlm}$.

*1. Study of case 1 with $\tilde{\gamma} = 0$.* Let $\mathbf{D}(\omega, \mathbf{r}) = \tilde{\varepsilon}\mathbf{E}(\omega, \mathbf{r}) + \nabla \times \tilde{\alpha}\nabla \times \mathbf{E}(\omega, \mathbf{r})$. Inside the homogenized MM, this constitutive relation requires that

$$c_{ijlm}\partial_l\partial_m E_j \overset{!}{=} [\nabla \times (\alpha\nabla \times \mathbf{E})]_i. \tag{73}$$

We develop the right-hand side in (73), we get

$$\nabla \times (\alpha\nabla \times \mathbf{E}) = \begin{pmatrix} \alpha_{yy}\partial_x\partial_z E_z - \alpha_{yy}\partial_z\partial_z E_x + \alpha_{zz}\partial_x\partial_y E_y - \alpha_{zz}\partial_y\partial_y E_x \\ \alpha_{xx}\partial_y\partial_z E_z - \alpha_{xx}\partial_z\partial_z E_y + \alpha_{zz}\partial_x\partial_y E_x - \alpha_{zz}\partial_x\partial_x E_y \\ \alpha_{xx}\partial_y\partial_z E_x - \alpha_{xx}\partial_y\partial_y E_z + \alpha_{yy}\partial_x\partial_z E_x - \alpha_{yy}\partial_x\partial_x E_z \end{pmatrix}, \tag{74}$$

where it has been assumed that $\alpha$ is a diagonal matrix. Comparing the coefficients yields

$$\begin{pmatrix} \underline{c_{xzxz} = \alpha_{yy}}\,, & \underline{c_{xxzz} = -\alpha_{yy}}\,, & c_{xyxy} = \alpha_{zz}\,, & c_{xxyy} = -\alpha_{zz} \\ \underline{\underline{c_{yzyz} = \alpha_{xx}}}\,, & \underline{\underline{c_{yyzz} = -\alpha_{xx}}}\,, & c_{yxyx} = \alpha_{zz}\,, & c_{xxyy} = -\alpha_{zz} \\ \underline{\underline{c_{zyzy} = \alpha_{xx}}}\,, & \underline{\underline{c_{zzyy} = -\alpha_{xx}}}\,, & \underline{c_{zxzx} = \alpha_{yy}}\,, & \underline{c_{zzxx} = -\alpha_{yy}} \end{pmatrix}.$$

The coefficients in the same number of underlines are related to the same material parameter $\alpha_{ii}$. From the comparison, we note that the assumption in Eq. (73) is compatible with the fundamental symmetry constraints in Eq. (72). For example we systematically obtain equalities such as $c_{xyxy} = c_{yxyx} = \alpha_{zz}$ or $c_{yzyz} = c_{zyzy} = \alpha_{xx}$.

We also note that there are constraints that impose $c_{iijj} = c_{jjii}$ for all $(i, j) \in \{x, y, z\} \times \{x, y, z\}$ and $i \neq j$, which are not part of the fundamental symmetry constraints. For instance, we have $c_{xxzz} = c_{zzxx} = -\alpha_{yy}$. Furthermore, the fact that such terms differ by a minus sign as well, i.e., $c_{ijij} = -c_{iijj}$ for all $(i, j) \in \{x, y, z\} \times \{x, y, z\}$ and $i \neq j$ suggests that the assumption in Eq. (73) is of higher symmetry than simply spatial inversion symmetry. The first constraints, i.e., $c_{iijj} = c_{jjii}$ for all $(i, j) \in \{x, y, z\} \times \{x, y, z\}$ and $i \neq j$ renders the crystal of a tetragonal system, i.e., of fourfold symmetry. They define the symmetry classes $C_4$ and $D_{4h}$. We stress that the fishnet MM is of $D_{2h}$ symmetry (only), while we assume Eq. (73) to hold. We technically try to describe a system with lower symmetry ($D_{2h}$) with coefficients of higher symmetry ($D_{4h}$). We highlight that the second constraint $c_{ijij} = -c_{iijj}$ for all $(i, j) \in \{x, y, z\} \times \{x, y, z\}$ and $i \neq j$, is not linked to a symmetry.

One can prove that the symmetry condition is satisfied by the fourth order term, i.e., when $\tilde{\gamma} \neq 0$. We follow the same principles as in the second order term, i.e., symmetry condition (69) and the mixed partial derivatives, we can examine the assumption

$$e_{ijklmn}\nabla_k\nabla_l\nabla_m\nabla_n E_j \overset{!}{=} \{\nabla \times \nabla \times [\gamma(\omega)\nabla \times \nabla \times \mathbf{E}]\}_i. \tag{75}$$

Due to having long formulas, we do not present the computational details in this paper.

*2. Study of case 7 with $\tilde{\gamma} = 0$.* Let $\mathbf{D}(\omega, \mathbf{r}) = \tilde{\varepsilon}\mathbf{E}(\omega, \mathbf{r}) + \tilde{\alpha}\nabla \times \nabla \times \mathbf{E}(\omega, \mathbf{r})$.

Here $\tilde{\alpha}$ is positioned on the left. This supposition imposes that

$$c_{ijlm}\partial_l\partial_m E_j \overset{!}{=} (\alpha\nabla \times \nabla \times \mathbf{E})_i. \tag{76}$$

We develop the right-hand side in (76), we get

$$\alpha\nabla \times \nabla \times \mathbf{E} = \begin{pmatrix} \alpha_{xx}\partial_x\partial_y E_y - \alpha_{xx}\partial_y\partial_y E_x + \alpha_{xx}\partial_x\partial_z E_z - \alpha_{xx}\partial_z\partial_z E_x \\ \alpha_{yy}\partial_x\partial_y E_x - \alpha_{yy}\partial_x\partial_x E_y + \alpha_{yy}\partial_y\partial_z E_z - \alpha_{yy}\partial_z\partial_z E_y \\ \alpha_{zz}\partial_x\partial_z E_x - \alpha_{zz}\partial_x\partial_x E_z + \alpha_{zz}\partial_y\partial_z E_y - \alpha_{zz}\partial_y\partial_y E_z \end{pmatrix}. \tag{77}$$

Comparing the coefficients yields

$$\begin{pmatrix} \underline{\underline{c_{xyxy} = \alpha_{xx}}}\,, & \underline{\underline{c_{xxyy} = -\alpha_{xx}}}\,, & \underline{\underline{c_{xzxz} = \alpha_{xx}}}\,, & \underline{\underline{c_{xxzz} = -\alpha_{xx}}} \\ \underline{c_{yxyx} = \alpha_{yy}}\,, & \underline{c_{yyxx} = -\alpha_{yy}}\,, & \underline{c_{yzyz} = \alpha_{yy}}\,, & \underline{c_{yyzz} = -\alpha_{yy}} \\ c_{zxzx} = \alpha_{zz}\,, & c_{zzyy} = -\alpha_{zz}\,, & c_{zzyy} = \alpha_{zz}\,, & c_{zzyy} = -\alpha_{zz} \end{pmatrix}.$$

From this comparison, we read out that the assumption in Eq. (76) yields that $c_{xyxy} = \alpha_{xx}$, $c_{yxyx} = \alpha_{yy}$ and $c_{xzxz} = \alpha_{xx}$, $c_{zxzx} = \alpha_{zz}$ hold simultaneously. Following the fundamental symmetry constraints (72), it must hold that

$$\alpha_{xx} = \alpha_{yy} = \alpha_{zz}.$$

Consequently, the system has to be isotropic, otherwise physical symmetries are violated.

Please notice that cases 1 and 7 are not equivalent, even if $\alpha$ is a scalar. This important point comes down to the fact that these two cases are physically different since they lead to different boundary conditions [see (30b) and (64b)]; even if their corresponding constitutive relations are identical. For more details, we draw the attention of the reader to Ref. [21].

## VIII. CONCLUSION

In this contribution we investigated the propagation of light in mediums constituted of MMs, where the unit cells are centrosymmetric and we considered nonlocal constitutive relations. By means of a second-order Padé-type approximation of the response functions, we modeled the light-matter interaction with nine different constitutive relations that lead to different wave equations. The main reason for having this diversity is the nature of the material parameters, which are supposed to be anisotropic diagonal matrices. That means they do not commute with the differential operators, then they do not express necessarily the same light-matter interaction. Hence, a checklist process was of major necessity to decide which formulation is suitable for describing the considered optical phenomena. This analysis is mandatory prior to checking the validity of a model to adequately homogenize a MM. A valid model must simultaneously fulfill the following three criteria:

(1) The first criterion concerns the analysis of interface conditions posed on surfaces separating MMs from vacuum. They are obtained by means of the weak formulations corresponding to each case and they are necessary for computing the reflected and transmitted fields. These propagating modes represent the solutions to the dispersion relation relative to each case with respect to TE and TM polarizations. To compute the amplitudes of the reflected and transmitted fields, we derive the corresponding Fresnel formulas. For well defined systems, the number of propagating modes within MMs must coincide with the number of the derived interface conditions. Cases 1, 4, 5, 6, and 7 led to weak formulations that revealed the required number of interface conditions. For cases 2, 3, 8, and 9 it was not the case; the associated systems of equations obtained by using Fresnel formulas are underdetermined due to the lack of additional interface conditions. Thus, they are rejected from the investigation for not fulfilling the first criterion.

(2) The second criterion is called the reflection and transmission coefficients analysis. In principle, by taking the limit of the nonlocal material parameters to zero, we require us to reproduce the reflection and transmission coefficients produced by the WSD, as the constitutive relations reduce to those of the local models. More precisely, by following the local approach we get only two propagating fields at each interface. One of them is reflected and the other one is transmitted. In the nonlocal analysis approach, we have three fields propagating away from an interface, one reflected and two transmitted. By taking the limit of the nonlocal material parameter to zero, one of the two transmitted fields must vanish, which is not the case for cases 4, 5, and 6. Namely, for these three cases, if the second transmitted mode vanishes, it implies one of two facts: (a) either the material parameter related to the second order terms in the wavelike equations is zero, which does not even reflect models with WSD, (b) or the tangential components of the transmitted electric field are zero. The latter result would imply a total reflection that is independent from the angle of incidence and the permittivities of the two materials. Both significations are not true. This fact undeniably rises from the nonvanishing additional interface condition, which makes the system of equations being too restrictive and overdetermined. For cases 1 and 7 this criterion is fulfilled.

(3) The third and last criterion was based on the requirement of fulfilling the Casimir-Onsager reciprocity. It is a symmetry constraint, which is satisfied by the first case, whereas, for case 7, it makes sense only when the material parameter $\alpha$ is a scalar.

At the end of the checklist, the only considered cases are case 7 under the claim of being isotropic and case 1, in which the material parameters are sandwiched between an equal number of curl operators. It represents the model studied previously [26] by means of a Taylor approximation of the response function. It showed a good description of the propagation of light in MMs compared to the WSD. The use of Padé-type approximation for the response function opened the path to derive several representation of the light-matter interaction; it allowed us to set a solid background to study and check the validity of other models, not necessarily obtained by following the same approach. For further investigations, the Padé-type approximation confirms that going to higher order spacial dispersion is required. However, other constitutive relations could have been suggested as well but prior being considered in detail, they have to necessarily pass the checklist that we have put forward in this contribution. Therefore, our work is quite general and important in the ongoing endeavor to homogenize MMs with advanced and nonlocal constitutive relations.

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### APPENDIX: EXACT COEFFICIENTS OF THE DISPERSION RELATIONS

In this Appendix we shall summarize the resulting coefficients that appear in every case. While the physical discussion is made in Sec. V, here we merely write down the exact solutions. For each case, the solution and its corresponding coefficients are written first in TM and then in TE polarization.

#### 1. Case 1 [$\hat{\mathbf{p}}(\omega,\mathbf{k}) = -\mathbf{k}\times\boldsymbol{\alpha}\mathbf{k}\times$ and $\hat{\mathbf{q}}(\omega,\mathbf{k}) = -\mathbf{k}\times\mathbf{k}\times\boldsymbol{\gamma}$]

*a. TM polarization.*

$$k_z^2(k_y) = -\frac{1}{2}\left(q_0^{\mathrm{TM}} + q_1^{\mathrm{TM}}\right)k_y^2 + p_0^{\mathrm{TM}} \pm \sqrt{l_0^{\mathrm{TM}} + \left(p_0^{\mathrm{TM}} + \frac{q_0^{\mathrm{TM}} - q_1^{\mathrm{TM}}}{2}k_y^2\right)^2}, \tag{A1}$$

with

$$q_0^{\mathrm{TM}} = \frac{\epsilon_y}{\epsilon_z}, \quad q_1^{\mathrm{TM}} = \frac{\gamma_z}{\gamma_y}, \quad p_0^{\mathrm{TM}} = \frac{-1}{2\gamma_y\mu_x}, \quad l_0^{\mathrm{TM}} = \frac{k_0^2\epsilon_y}{\gamma_y}.$$

*b. TE polarization.*

$$k_z^2(k_y) = -k_y^2 + p_0^{\mathrm{TE}} \pm \sqrt{l_0^{\mathrm{TE}} + \left(p_0^{\mathrm{TE}}\right)^2 + 2\left(p_1^{\mathrm{TE}} - p_0^{\mathrm{TE}}\right)k_y^2}, \tag{A2}$$

with

$$p_0^{\mathrm{TE}} = \frac{-1}{2\gamma_x\mu_y}, \quad p_1^{\mathrm{TE}} = \frac{-1}{2\gamma_x\mu_z}, \quad l_0^{\mathrm{TE}} = \frac{k_0^2\epsilon_x}{\gamma_x}.$$

#### 2. Case 2 [$\hat{\mathbf{p}}(\omega,\mathbf{k}) = -\mathbf{k}\times\boldsymbol{\alpha}\mathbf{k}\times$ and $\hat{\mathbf{q}}(\omega,\mathbf{k}) = -\mathbf{k}\times\boldsymbol{\gamma}\mathbf{k}\times$]

*a. TM polarization.*

$$k_z^2(k_y) = -\frac{1}{2}\left(q_0^{\mathrm{TM}} + q_1^{\mathrm{TM}}\right) + p_0^{\mathrm{TM}} \pm \sqrt{l_0^{\mathrm{TM}} + \left(p_0^{\mathrm{TM}} + \frac{q_0^{\mathrm{TM}} - q_1^{\mathrm{TM}}}{2}k_y^2\right)^2}, \tag{A3}$$

with

$$q_0^{\mathrm{TM}} = \frac{\epsilon_y}{\epsilon_z}, \quad q_1^{\mathrm{TM}} = 1, \quad p_0^{\mathrm{TM}} = \frac{-1}{2\gamma_x\mu_x}, \quad l_0^{\mathrm{TM}} = \frac{k_0^2\epsilon_y}{\gamma_y}.$$

*b. TE polarization.*

$$k_z^2(k_y) = -\frac{1}{2}\left(q_0^{\mathrm{TE}} + q_1^{\mathrm{TE}}\right) + p_0^{\mathrm{TE}} \pm \sqrt{l_0^{\mathrm{TE}} + \left(p_0^{\mathrm{TE}} - \frac{q_0^{\mathrm{TE}} + q_1^{\mathrm{TE}}}{2}k_y^2\right)^2 + 2q_0^{\mathrm{TE}}p_1^{\mathrm{TE}}k_y^2 - q_0^{\mathrm{TE}}q_1^{\mathrm{TE}}k_y^4}, \tag{A4}$$

with

$$q_0^{\mathrm{TE}} = 1, \quad q_1^{\mathrm{TE}} = \frac{\gamma_z}{\gamma_y}, \quad p_0^{\mathrm{TE}} = \frac{-1}{2\gamma_y\mu_y}, \quad p_1^{\mathrm{TE}} = \frac{-1}{2\gamma_y\mu_z}, \quad l_0^{\mathrm{TE}} = \frac{k_0^2\epsilon_x}{\gamma_y}.$$

#### 3. Case 3 [$\hat{\mathbf{p}}(\omega,\mathbf{k}) = -\mathbf{k}\times\boldsymbol{\alpha}\mathbf{k}\times$ and $\hat{\mathbf{q}}(\omega,\mathbf{k}) = -\boldsymbol{\gamma}\mathbf{k}\times\mathbf{k}\times$]

*a. TM polarization.*

$$k_z^2(k_y) = -\frac{1}{2}\left(q_0^{\mathrm{TM}} + q_1^{\mathrm{TM}}\right) + p_0^{\mathrm{TM}} \pm \sqrt{l_0^{\mathrm{TM}} + \left(p_0^{\mathrm{TM}} - \frac{q_0^{\mathrm{TM}} + q_1^{\mathrm{TM}}}{2}k_y^2\right)^2 + 2q_0^{\mathrm{TM}}p_1^{\mathrm{TM}}k_y^2 - q_0^{\mathrm{TM}}q_1^{\mathrm{TM}}k_y^4}, \tag{A5}$$

with

$$q_0^{\mathrm{TM}} = \frac{\epsilon_y\gamma_z}{\epsilon_z\gamma_y}, \quad q_1^{\mathrm{TM}} = 1, \quad p_0^{\mathrm{TM}} = \frac{-1}{2\gamma_y\mu_x}, \quad p_1^{\mathrm{TM}} = \frac{-1}{2\gamma_z\mu_x}, \quad l_0^{\mathrm{TM}} = \frac{k_0^2\epsilon_y}{\gamma_y}.$$

*b. TE polarization.*

$$k_z^2(k_y) = -k_y^2 + p_0^{\mathrm{TE}} \pm \sqrt{l_0^{\mathrm{TE}} + \left(p_0^{\mathrm{TE}}\right)^2 + 2\left(p_1^{\mathrm{TE}} - p_0^{\mathrm{TE}}\right)k_y^2}, \tag{A6}$$

with

$$p_0^{\mathrm{TE}} = \frac{-1}{2\gamma_x\mu_y}, \quad p_1^{\mathrm{TE}} = \frac{-1}{2\gamma_x\mu_z}, \quad l_0^{\mathrm{TE}} = \frac{k_0^2\epsilon_x}{\gamma_x}.$$

### 4. Case 4 [$\hat{\mathbf{p}}(\omega, \mathbf{k}) = -\mathbf{k} \times \mathbf{k} \times \boldsymbol{\alpha}$ and $\hat{\mathbf{q}}(\omega, \mathbf{k}) = -\mathbf{k} \times \mathbf{k} \times \boldsymbol{\gamma}$]

*a. TM polarization.*

$$k_z^2(k_y) = -\frac{1}{2}\left(q_0^{\mathrm{TM}} + q_1^{\mathrm{TM}}\right) + p_0^{\mathrm{TM}} \pm \sqrt{l_0^{\mathrm{TM}} + \left(p_0^{\mathrm{TM}} - \frac{q_0^{\mathrm{TM}} + q_1^{\mathrm{TM}}}{2}k_y^2\right)^2 + 2q_0^{\mathrm{TM}}p_1^{\mathrm{TM}}k_y^2 - q_0^{\mathrm{TM}}q_1^{\mathrm{TM}}k_y^4}, \tag{A7}$$

with

$$q_0^{\mathrm{TM}} = \frac{\epsilon_y}{\epsilon_z}, \qquad q_1^{\mathrm{TM}} = \frac{\gamma_z}{\gamma_y}, \qquad p_0^{\mathrm{TM}} = \frac{-1}{2\gamma_y\mu_y}, \qquad p_1^{\mathrm{TM}} = \frac{-1}{2\gamma_y\mu_z}, \qquad l_0^{\mathrm{TM}} = \frac{k_0^2\epsilon_y}{\gamma_y}.$$

*b. TE polarization.*

$$k_z^2(k_y) = -k_y^2 + p_0^{\mathrm{TE}} \pm \sqrt{l_0^{\mathrm{TE}} + \left(p_0^{\mathrm{TE}}\right)^2}, \tag{A8}$$

with

$$p_0^{\mathrm{TM}} = \frac{-1}{2\gamma_x\mu_x}, \quad l_0^{\mathrm{TM}} = \frac{k_0^2\epsilon_x}{\gamma_x}.$$

### 5. Case 5 [$\hat{\mathbf{p}}(\omega, \mathbf{k}) = -\mathbf{k} \times \mathbf{k} \times \boldsymbol{\alpha}$ and $\hat{\mathbf{q}}(\omega, \mathbf{k}) = -\mathbf{k} \times \boldsymbol{\gamma}\mathbf{k}\times$]

*a. TM polarization.*

$$k_z^2(k_y) = -\frac{1}{2}\left(q_0^{\mathrm{TM}} + q_1^{\mathrm{TM}}\right) + p_0^{\mathrm{TM}} \pm \sqrt{l_0^{\mathrm{TM}} + \left(p_0^{\mathrm{TM}} - \frac{q_0^{\mathrm{TM}} + q_1^{\mathrm{TM}}}{2}k_y^2\right)^2 + 2q_0^{\mathrm{TM}}p_1^{\mathrm{TM}}k_y^2 - q_0^{\mathrm{TM}}q_1^{\mathrm{TM}}k_y^4}, \tag{A9}$$

with

$$q_0^{\mathrm{TM}} = \frac{\epsilon_y}{\epsilon_z}, \quad q_1^{\mathrm{TM}} = 1, \quad p_0^{\mathrm{TM}} = \frac{-1}{2\gamma_x\mu_y}, \quad p_1^{\mathrm{TM}} = \frac{-1}{2\gamma_x\mu_z}, \quad l_0^{\mathrm{TM}} = \frac{k_0^2\epsilon_y}{\gamma_x}.$$

*b. TE polarization.*

$$k_z^2(k_y) = -\frac{1}{2}\left(q_0^{\mathrm{TE}} + q_1^{\mathrm{TE}}\right)k_y^2 + p_0^{\mathrm{TE}} \pm \sqrt{l_0^{\mathrm{TE}} + \left(p_0^{\mathrm{TE}} + \frac{q_0^{\mathrm{TE}} - q_1^{\mathrm{TE}}}{2}k_y^2\right)^2}, \tag{A10}$$

with

$$q_0^{\mathrm{TE}} = 1, \quad q_1^{\mathrm{TE}} = \frac{\gamma_z}{\gamma_y}, \quad p_0^{\mathrm{TE}} = \frac{-1}{2\gamma_y\mu_x}, \quad l_0^{\mathrm{TE}} = \frac{k_0^2\epsilon_x}{\gamma_y}.$$

### 6. Case 6 [$\hat{\mathbf{p}}(\omega, \mathbf{k}) = -\mathbf{k} \times \mathbf{k} \times \boldsymbol{\alpha}$ and $\hat{\mathbf{q}}(\omega, \mathbf{k}) = -\boldsymbol{\gamma}\mathbf{k} \times \mathbf{k}\times$]

*a. TM polarization.*

$$Q(k_y)k_z^2(k_y) = P_0\left(k_y^2\right) \pm \sqrt{[P_0(k_y)]^2 + P_1\left(k_y^2\right)}, \tag{A11}$$

where

$$\begin{aligned} Q_0(k_y) &= 2\delta k_y - 2p_0^{\mathrm{TM}}, \\ P_0(k_y) &= -\delta k_y^4 + \left(p_0^{\mathrm{TM}} + p_1^{\mathrm{TM}}\right)k_y^2 + n_1, \\ P_1(k_y) &= 2\left(p_1^{\mathrm{TM}}k_y^4 + n_0k_y^2 + n_0n_1\right)\left(2\delta k_y^2 - 2p_0^{\mathrm{TM}}\right), \end{aligned}$$

with

$$\delta = (\gamma_y - \gamma_z)(\mu_y - \mu_z), \quad p_0^{\mathrm{TM}} = k_0^2\gamma_y\mu_y\epsilon_z\mu_z, \quad p_1^{\mathrm{TM}} = k_0^2\gamma_z\mu_z\epsilon_y\mu_y, \quad n_0 = k_0^2\epsilon_y\mu_y, \quad n_1 = k_0^2\epsilon_z\mu_z.$$

*b. TE polarization.*

$$k_z^2(k_y) = -k_y^2 + p_0^{\mathrm{TE}} \pm \sqrt{l_0^{\mathrm{TE}} + \left(p_0^{\mathrm{TE}}\right)^2}, \tag{A12}$$

with

$$p_0^{\mathrm{TE}} = \frac{-1}{2\gamma_x\mu_x}, \quad l_0^{\mathrm{TE}} = \frac{k_0^2\epsilon_x}{\gamma_x}.$$

### 7. Case 7 [$\hat{\mathbf{p}}(\omega, \mathbf{k}) = -\alpha\mathbf{k}\times\mathbf{k}\times$ and $\hat{\mathbf{q}}(\omega, \mathbf{k}) = -\mathbf{k}\times\mathbf{k}\times\gamma$]

The dispersion relations for both TE and TM polarizations for this case, are identical to case 4.

### 8. Case 8 [$\hat{\mathbf{p}}(\omega, \mathbf{k}) = -\alpha\mathbf{k}\times\mathbf{k}\times$ and $\hat{\mathbf{q}}(\omega, \mathbf{k}) = -\mathbf{k}\times\gamma\mathbf{k}\times$]

The dispersion relations for both TE and TM polarizations for this case, are identical to case 5.

### 9. Case 9 [$\hat{\mathbf{p}}(\omega, \mathbf{k}) = -\alpha\mathbf{k}\times\mathbf{k}\times$ and $\hat{\mathbf{q}}(\omega, \mathbf{k}) = -\gamma\mathbf{k}\times\mathbf{k}\times$]

*a. TM polarization.*

$$k_z^2(k_y) = -\frac{1}{2}\left(q_0^{\mathrm{TM}} + q_1^{\mathrm{TM}}\right) + p_0^{\mathrm{TM}} \pm \sqrt{l_0^{\mathrm{TM}} + \left(p_0^{\mathrm{TM}} - \frac{q_0^{\mathrm{TM}} + q_1^{\mathrm{TM}}}{2}k_y^2\right)^2 + 2q_0^{\mathrm{TM}}p_1^{\mathrm{TM}}k_y^2 - q_0^{\mathrm{TM}}q_1^{\mathrm{TM}}k_y^4}, \tag{A13}$$

with

$$q_0^{\mathrm{TM}} = \frac{\epsilon_y\gamma_z}{\epsilon_z\gamma_y}, \quad q_1^{\mathrm{TM}} = 1, \quad p_0^{\mathrm{TM}} = \frac{-1}{2\gamma_y\mu_y}, \quad p_1^{\mathrm{TM}} = \frac{-1}{2\gamma_z\mu_z}, \quad l_0^{\mathrm{TM}} = \frac{k_0^2\epsilon_x}{\gamma_x}.$$

*b. TE polarization.*

$$k_z^2(k_y) = -k_y^2 + p_0^{\mathrm{TE}} \pm \sqrt{l_0^{\mathrm{TE}} + \left(p_0^{\mathrm{TE}}\right)^2}, \tag{A14}$$

with

$$p_0^{\mathrm{TM}} = \frac{-1}{2\gamma_x\mu_x}, \quad l_0^{\mathrm{TM}} = \frac{k_0^2\epsilon_x}{\gamma_x}.$$

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