Non-Linear Modeling of Active or Passive Optical Lamellar Nanostructures

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ABSTRACT

A universal approach to modeling the refraction of a monochromatic plane wave incident on an optical metamaterial arranged of arbitrary (passive, active, or plasmonic) layers of linear or feebly non-linear elemental materials is examined. The approach is built on an alternative formulation of the method of single expression, which works well upon different combinations (non-linear lossy dielectric, graded-index Kerr medium, and gain medium with metal) upon either p-polarized or s-polarized light. We show that although the formulation for s-polarized light is straightforward, the p-polarization case is more complicated, since solving an implicit material equation is required. Numerical validation of the proposed method indicates good agreement with the results taken from other numerical solvers (based on finite element and mode matching methods).

Keywords: method of single expression, non-linear optics, optical metamaterials

1. INTRODUCTION

The latest progress in the numerical analysis [1], fabrication and optical characterization [2] of low-loss nanostructured optical lamellar media with an effective hyperbolic dispersion has been inspired by the intriguing properties of the perfect focusing device with compensated losses [3]. Most recently, other promising optical effects, made possible through the use of hyperbolic metamaterials (HMMs), has come into focus. It was reported that HMMs can have a unique broadband singularity in the photonic density of states (PDOS) [4]. For example, because spontaneous emission depends appreciably on the available photonic density of states, the emission was predicted to be enhanced close to the surface of HMM. Not only could such materials enhance the spontaneous emission (e.g., of a quantum dot), but could also cause the photons to be emitted in a given direction.

For these reasons, there is considerable interest in accurate, computationally inexpensive, and hence presumably one-dimensional tools for modeling the dynamics of plane waves in a most general HMM structure. The approach should include an adequate treatment of the layers with a weak non-linearity, gain, gradients of the refractive index, losses, and plasmonic effects. A method of single expression (MSE, [5, 6]) has a promise to satisfy all of the above requirements. Here, we are extending this practical concept to be capable of modeling any planar lamellar nano-structures with effective hyperbolic dispersion. The proposed alternative formulation of MSE could be used to design HMMs that are optimized to have specific absorption, reflection and transmission spectra and given non-linear responses to light waves. Subsequently, the development of a nanophotonic modeling tool shall be undertaken, with a goal of substituting or complementing our current software tools for linear cases [7, 8].

A lamellar structure has been used to validate the approach with numerical simulations of a multilayer stack of silver-dielectric layers with nanoscale thicknesses. Full-wave electromagnetic simulations have been completed using a commercial finite-element software and our SHA technique [7, 9], and the validation results are in good agreement with those obtained from the proposed method.

2. THE METHOD OF SINGLE EXPRESSION

2.1 Nomenclature

Throughout the paper we use the following free-space constants: wavelength \( \lambda [\text{m}] \), wavenumber \( k = 2\pi/\lambda [\text{rad/m}] \),

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the speed of light, \( c = 299792458 \text{ m/s} \), permeability \( \mu_0 = 4\pi \times 10^{-7} \text{ [H/m]} \), permittivity \( \varepsilon_0 = \varepsilon^2 / \mu_0 \text{ [F/m]} \), and impedance \( z_0 = \mu_0 c \Omega \). We shall also note that the normalized 2D space \((kx \rightarrow x, ky \rightarrow y)\) and the corresponding derivatives (denoted as superscript indices in parenthesis) are used in the paper. Figure 1 shows a general set-up of the problem for two distinct linear polarizations of light; s-polarized incidence is shown in panel (a), while the p-polarization case is shown in (b).

![Fig. 1. General set-up of the problem for two distinct linear polarizations of light; (a) s-polarized, and (b) p-polarized incidence.](image)

2.2 Alternative formulation for s-polarization

We start with the monochromatic scalar wave equation \( d^{(x,x)} + d^{(y,y)} + \varepsilon d = 0 \), which is written for s-polarized light (\( \hat{d} = \varepsilon_0 \hat{E} \)). Following the MSE we consider a test function written as,

\[
d = \exp[u(x) + \nu(x) y], \quad \{u, \nu \} \in \mathbb{C},
\]

which is a different form of the original MSE formulation [1].

Here, a possible non-linearity of permittivity is introduced through additional dependence on \( l = \text{Re} u \); hence, the complex permittivity, \( \varepsilon(l; x) = \varepsilon_i(x) + \chi(l; x) \), allows for lossy or gain elemental materials with sharp or continuous changes in linear permittivity \( \varepsilon_i \) and non-linear susceptibility \( \chi \).

We shall note that \( \nu = i \sqrt{\varepsilon} \sin \phi \) is defined by a position-dependent angle \( \phi(x) \) and a position-dependent permittivity \( \varepsilon(x) \), but in accord with Snell’s law, this part is invariant through the slab, i.e. \( \sqrt{\varepsilon} \sin \theta_i = \sqrt{\varepsilon} \sin \phi = \sqrt{\varepsilon} \sin \theta_i \), we finally arrive at \( k_i = \sqrt{\varepsilon} \sin \theta_i \):

\[
d = \exp[u(x) + t \nu(x) y],
\]

converting the scalar wave equation \( d^{(x,x)} + d^{(y,y)} + \varepsilon d = 0 \) into a classical Riccati equation

\[
u^{(x,x)} + \left(u^{(x)}\right)^2 + \varepsilon(l; x) - k_i^2 = 0.
\]

2.3 Alternative formulation for p-polarization

In contrast with s-polarized incidence, for the p-polarization, there is a complication due to an implicit material equation [10, 11]. First, using \( h = \exp[u(x) + tk_y y] \), we write

\[
\begin{align*}
[\nabla \cdot (\varepsilon^{-1} \nabla h)] + h &= 0, \\
\varepsilon^{(x)} &= \varepsilon_i^{(x)} + \left[\chi \left(\nabla |h| \varepsilon^2 \right)^2\right]^{(x)}
\end{align*}
\]

which is equivalent to
\[
\begin{align*}
\left\{ \begin{array}{l}
(u^{(s)}(z) + (u^{(i)}(z))^2 - (\ln\epsilon^{(s)}(z) u^{(s)}(z) + e - k^2) = 0 \\
(g^{(s)}) = |\nabla h^{(s)}(z)| = \left(\text{e}^{\Re u} \sqrt{|u^{(s)}(z)|^2 + k^2} \right)^{(s)} \\
(e^{(s)}) = e^{i(s)}(z) + \chi^{(s)}(z) g^{(s)}(z) + \chi^{(s)}/|e^{(s)}| 
\end{array} \right.
\end{align*}
\]

where the logarithmic derivative is \((\ln\epsilon)^{(s)}(z) = \epsilon^{(s)}/\epsilon\).

2.4 Cubic non-linearity for p-polarization

As an example, we may consider a cubic non-linearity \((g^2 = |\nabla h|^2 = \left|u^{(s)}(z) + k^2\right|^2 \text{e}^{2Re u})\)

\[
\epsilon = e^s + 3\chi(z^2) g^2 |\epsilon|^{-2}.
\]

Then, the above gives \(\left(\epsilon + \epsilon - e^s\right)^{(s)} = 3\chi(z^2) g^2 (g^2)^{(s)}\), or \(\alpha \ln\epsilon^{(s)} + \ln\epsilon^{(s)}\beta = \gamma\), where \(\alpha = e + \beta\), \(\beta = e - e^s\), and \(\gamma = e^s + 3\chi(z^2) g^2 (g^2)^{(s)}\), and we finally arrive at,

\[
\epsilon^{(s)}(z) = e^{(\gamma \alpha - \beta^2)}/\left|\alpha^2 - |\beta|\right|^2.
\]

Hence, the general scheme degenerates into the following sequence,

\[
\begin{align*}
\left\{ \begin{array}{l}
(u^{(s)}(z) + (u^{(i)}(z))^2 - (\ln\epsilon^{(s)}(z) u^{(s)}(z) + e - k^2) = 0 \\
(g^{(s)}) = \text{e}^{2Re u} \left|u^{(s)}(z) + \overline{u}^{(s)}(z) + 2\text{Re} u^{(s)}\right| |u^{(s)}(z)|^2 + k^2 \right) \\
\ln\epsilon^{(s)}(z) = (\gamma \alpha - \beta^2)/\left|\alpha^2 - |\beta|\right|^2 
\end{array} \right.
\end{align*}
\]

Only if the change of \(\epsilon\) is adiabatic, i.e. \((\ln\epsilon)^{(s)}(z) \to 0\), then (8) can be reduced to (3).

2.5 Initial conditions for linear polarized light

At the substrate side of the slab at \(y = 0\), we take either \(d = d_s = d_{\max} \exp(-ik_{\text{sub}} x_{\max})\) for s-polarization, or \(h = h_s = h_{\max} \exp(-ik_{\text{sub}} x_{\max})\) for p-polarization, and the general initial condition for \(u\) is \(u(x_{\max}) = -ik_{\text{sub}} x_{\max}\), where \(k_{\text{sub}} = \sqrt{e_{\text{sub}} - k^2}\).

At the illumination side, we may use the Sommerfeld radiation condition,

\[
\begin{align*}
d_i = \frac{1}{2} \left(d - m_{\text{sup}}^{-1} d^{(s)}(z) \right) \\
d_i = \frac{1}{2} \left(d + m_{\text{sup}}^{-1} d^{(s)}(z) \right),
\end{align*}
\]

thus splitting the incident and the reflected fields \((d_i, d_r)\), and obtaining their magnitudes. Finally, using (2) in (9) for \(y = 0\), we arrive at:

\[
\begin{align*}
d_i = \frac{1}{2} \left(1 - m_{\text{sup}}^{-1} u^{(s)}(z) \right) \exp u \\
d_r = \frac{1}{2} \left(1 + m_{\text{sup}}^{-1} u^{(s)}(z) \right) \exp u
\end{align*}
\]
3. VALIDATION TEST

The proposed method is implemented using the explicit Runge-Kutta scheme. A metal-dielectric multilayer structure arranged of alternating layers of silver and Vanadium Pentoxide ($V_2O_5$) as shown in Fig. 2 is designed for the validation test. The thickness of each silver layer is 20 nm. The thickness of the external $V_2O_5$ layers is 28 nm, while the thickness of other V2O5 layers is 64 nm; the entire structure is immersed in air. The plane wave of the incident light propagates normally to the interface from left to right. $V_2O_5$ exhibits strong cubic nonlinearity with the value of $\chi^{(3)}$ about $31 \times 10^{-8}$ e.s.u. and a linear refractive index of 2.37 at a wavelength of 532 nm [12]. The optical constants of silver are taken from [13].

![Fig. 2. Metal-dielectric multilayer structure arranged of alternating layers of silver and Vanadium Pentoxide ($V_2O_5$) for the validation test. The thickness of each silver layer is 20 nm. The thickness of the external $V_2O_5$ layers is 28 nm, while the thickness of other V2O5 layers is 64 nm. The entire structure is immersed in air.](image)

The linear case, where $V_2O_5$ layers were treated as dielectric layers without any nonlinearity, has been tested first. The resulting transmittance and reflectance spectra are shown in Fig. 3(a). Full-wave electromagnetic simulations have been also carried out using our SHA approach [9], and the validation results are in good agreement with those obtained from the proposed method. Simulations considering cubic nonlinearity have been carried out at a wavelength of 532 nm. Fig. 3(b) shows transmittance and reflectance vs. the E-field magnitude. The result obtained from a commercial finite element method (FEM) software package (COMSOL Multiphysics) is also included in Fig. 3(b) as a reference. Note that as shown in the figure the nonlinear solver in FEM fails to obtain a valid solution for the input electric field magnitude larger than $2.45 \times 10^7$ V/m. From the figure we can clearly see there is strong nonlinear effect due to the change of the strength of the input field. The results obtained from the proposed method and those obtained from FEM solver match quite well with each other.

![Fig. 3. Comparison of (a) transmittance and reflectance spectra, and (b) input field strength dependent transmittance and reflectance obtained by using the proposed method and other methods.](image)

Furthermore, the transmittance and reflectance spectra with different angle of incidence are calculated using the proposed method and SHA approach. The results which are shown in Fig. 4(a) indicate a very good agreement between the two methods. We also show the input electric field dependent spectra with different angle of incidence in Fig. 4(b).
Most interestingly, the structure we studied above shows a bistability behavior for sufficiently large input electric field magnitude. Fig. 5 shows this behavior and the arrows show the abrupt changing of the reflectance or transmittance when the magnitude of the input electric field increases or decreases. The phenomenon has been studied in nonlinear materials for a long time and similar behavior was discussed for multilayer structures [6, 14]. With our proposed method, we are able to capture this bistability behavior precisely.

Another example we want to show in this paper is an active metal-dielectric multilayer HMM which consists of 16 layers of alternating 6-nm-thick silver layers and 50-nm-thick active dielectric layers on a glass substrate. The size-dependent sliver loss has been taken into account by setting the loss factor to two [15]. For the active dielectric medium, we take the experimentally measured gain values of the organic dye (Rh800) mixed in epoxy [16]. The refractive index of epoxy is 1.65. The emission peak of Rh800 is about 720 nm. The gain coefficient of a material is proportional to the imaginary part of the refractive index, we therefore modeled the active dielectric with a complex dielectric function. The effective anisotropic permittivities are shown in Fig. 6. At a wavelength around 730 nm, we can clearly see that the multilayer structure shows a hyperbolic dispersion, namely the effective permittivities are having different signs in x and y directions. The “without gain” case is studied by simply taking pure epoxy as the material of the dielectric layers.
A general approach to modeling the propagation of a plane wave through an arbitrary lamellar nanostructure has been obtained using a modified method of single expression. The new formulation presents either the electric or magnetic scalar wave as $\exp[i(k_x x + k_y y)]$, thus transforming the wave equation into the general dispersion identities (Eqs. (3) and (4)) for s-polarized and p-polarized light respectively. For the trivial case of homogeneous media both equations reduce to the standard dispersion relation, $\omega^{(s)} = k_s = \sqrt{\varepsilon - k^2}$. We note that in contrast to the s-polarization case, which is quite straightforward, the p-polarization case is complicated with an implicit material equation, which can be converted into an explicit form within a selected numerical scheme.

By controlling the parameters of the numerical scheme, (e.g., by reducing the minimal step-size) the convergence of the simulation approach has been additionally analyzed. This study has also shown that it is possible to apply the MSE to very different combinations of the elemental materials in a given lamellar structure, thus allowing for an arbitrary
mixture of layers (metals, dielectrics with gain, dielectrics with non-linear optical properties of other origin) with gradient-index structures, and giving a tool for designing hyperbolic metamaterials for near field optical imaging and other applications.

5. ACKNOWLEDGEMENT

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REFERENCES