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Optical Dispersion Models for Time-Domain Modeling of Metal-Dielectric Nanostructures

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We discuss second-order complex Padé approximants which give a systematic approach to time-domain modeling of dispersive dielectric functions. These approximants, which also reduce to the classical Drude, Lorentz, Sellmeier, critical points and other models upon appropriate truncation, are used to compare frequency domain (FD) versus time-domain (TD) simulations of local optical responses and the transmission-reflection spectra for a plasmonic nanostructure. A comparison is also made using auxiliary differential equations (ADE), and second order recursive convolution (RC) formulations embedded in finite-difference, finite-volume, and finite-element time-domain solvers.

Index Terms—Critical points, dispersive media, drude, FDTD methods, FETD, FVTD, Lorentz, Padé approximant, Sellmeier.

I. INTRODUCTION

In recent years, a great deal of work has been dedicated to accurate modeling and experimental studies of electromagnetic processes in optical nanoscaled metal-dielectric composites, or optical plasmonic metamaterials (OPM). In order to analytically or numerically study OPMs, including non-linear and multi-physical processes, an accurate analytical description of the dispersive behavior of both metals and dielectrics is required, particularly one which can be efficiently implemented in a numerical time domain solver.

We present a formalism based on a critical points model [1], which is essentially a [1/2]-order Padé approximant of dispersive electric susceptibility in the frequency domain. This formalization includes widely used dispersion models such as Drude, Lorentz, Sellmeier and the critical points model and allows for the unified formulas, which effectively implement the dispersion in time-domain solvers, with either an auxiliary differential equation (ADE) method or with recursive convolution (RC) methods. In particular, a new approach for the parameterization of RC methods is proposed, which allows for different 1st and 2nd order RC methods to be treated similarly; the approach also works for both-normal and over-damped Lorentz oscillators. We apply this technique to simulate gold nanostructures using a critical points model for gold [1], which costs less computationally due to its shorter local response and the lower number of dispersion terms than in a conventional Drude-Sellmeier model.

II. CONVENTIONAL DISPERSION MODELS

The FD dispersion of the isotropic electric susceptibility of noble metals, \( \chi(\omega) = \chi_0 + \chi_D(\omega) + \sum_{i=1}^{\text{Nmax}} \chi(i)_D(\omega) \), is described as a combination of the non-dispersive term \( \chi_0 \), the Drude (D) susceptibility, \( \chi_D(\omega) \), and a sum of additional terms, \( \chi(i)_D(\omega) \), with the number index denoting either the Lorentz (L), \( \chi_L(\omega) \), or the critical points (C) terms, \( \chi(C)_D(\omega) \) [1]. The dielectric functions of glasses and other transparent optical materials are often approximated using the Sellmeier (S) model, \( \chi(\omega) = \chi_0 + \sum_{i=1}^{\text{Nmax}} \chi(S)_i(\omega) \). The frequency dependencies are traditionally written as

\[
\chi_D(\omega) = -\frac{\omega_D^2}{(\omega^2 + i\omega \Gamma_D)}, \quad \chi_L(\omega) = \frac{\omega_L^2}{(\omega^2 + (\omega_L - i\omega \Gamma_L))}, \quad \chi(S)_i(\omega) = \frac{\omega_S^2}{(\omega_S^2 - \omega^2)} \quad \text{and} \quad \chi(C)_i(\omega) = \frac{2f_{C,i}\omega_C \cos \varphi_C + (i\omega - \Gamma_C) \sin \varphi_C}{\Gamma_C^2 + \omega_C^2 - \omega^2 - 2i\omega \Gamma_C}.
\]

where \( \omega_D, \omega_L, \omega_S, \omega_C, \Gamma_D, \Gamma_L, \Gamma_C, f_{C,i} \) and \( \varphi_C \) are constant parameters.

Here we introduce a convenient formalism for universal treatment of these models (including their ADE and RC numerical implementations) and their brief numerical analysis.

III. ADE-BASED AND RC-BASED UNIFICATION

Dispersion terms (1)–(4) can be represented as particular cases of a [1/2] Padé approximant, which can be expanded as a sum of two single poles (ignoring the case of multiple poles)

\[
\chi_i(\omega) = \frac{(a_{0,i} - \omega a_{1,i})/(b_{0,i} - \omega b_{1,i} - \omega^2)}{a_{2,i}/2 + e^{i\varphi_i}} / \left( d_{i} - (\omega + \epsilon i) \right)
\]

The first representation is useful for the description of the time domain isotropic local material response for each term \( P_i(\omega) = \varepsilon_0 \chi_i(\omega)E(\omega) \) with an ODE

\[
\ddot{P}_i + b_{1,i}\dot{P}_i + b_{0,i}P_i = \varepsilon_0 (a_{0,i}E + a_{1,i}\dot{E})
\]

which can be implemented with ADE methods in time-domain solvers. The second representation in (5) gives a simple formula for the time-domain susceptibility

\[
\chi(t) = a_0 e^{-\gamma t} \sin(\delta t - \varphi) U(t)
\]

where \( U(t) \) is the Heaviside step function. The polarization is then given as a convolution integral

\[ P_i(t) = \varepsilon_0 \int_0^t \chi(t)E(t - \tau)d\tau \]
The latter can be approximated using RC methods. Expression (7) brings more physical meaning to the optical responses of each model: \(\alpha_i\)—amplitude, \(\varphi_i\)—phase (which is zero if \(\delta_i \in i\mathbb{R}\) to guarantee \(\chi_i \in \mathbb{R}\), \(\gamma_i\)—damping, \(\delta_i\)—frequency if \(\delta_i \in \mathbb{R} \Leftrightarrow b_i^2 \leq 4\beta_0\delta_i\) and over-damping otherwise.

The formulas and coefficients for ODE and time-convolution forms (1)–(4) are shown in Tables I and II.

### IV. NUMERICAL IMPLEMENTATION

#### A. Auxiliary Differential Equation Method Parameters

The ADE method is applied to universal ODE (6) by approximating it with second order accuracy using finite differences. A bilinear scheme is used because of its known stability when coupled with Yee FDTD for Lorentz media [2]

\[
(F^{n+1} - 2F^n + F^{n-1})/\tau^2 + b_1(F^{n+1} - F^{n-1})/2\tau + b_0(F^{n+1} + F^{n-1})/4
= \alpha (E^{n+1} - E^{n-1})/2\tau + \alpha_0(E^{n+1} + E^{n-1})/4, \tag{9}
\]

#### B. Recursive Convolution Methods

RC rules are usually built for Debye (one real pole) and Lorentz media (two conjugate poles), for a specific RC method, for example, for the first order PCRC method introduced by Luebbers [3]. The Drude model is either regarded as a separate case or as a sum of a conductivity term and Debye relaxation. The generalization for a rational dielectric function including multiple poles, is discussed in [4] for PCRC and PLRC methods, however direct implementation of each exponential pole requires complex operations if conjugate poles occur. The usual approach for the Lorentz and critical points models using RC methods is to take the imaginary part of a complex exponent, i.e. \(\chi(t) = a_\text{C}e^{-\gamma t} \sin(\delta t - \varphi)U(t) = \text{Im}[a_\text{C}e^{-\gamma t + i\delta t - i\varphi}U(t)]. \) Yet in this case the generality of any [1/2] Pade approximant is lost, since this trick only works if \(\delta_1 \in \mathbb{R}\). Also the update of the complex recursive accumulator requires more flops than its conversion to real functions. The RC methods published for critical points model so far are of the first order [5], [6].

In this section we present formulas with real coefficients for RC implementations which are unified for any [1/2] Padé approximant (5) and for basic RC methods, i.e. for second order accurate methods such as TRC [7], PCRC2 [8], PLRC [9] and first order accurate methods PCRC [3] and RRC [10].

We start from simple algebraic lemmas, which give generalized recurrent formulas for RC methods:

**Lemma 1**: If an RC method for the exponential susceptibility \(\chi(t) = \alpha e^{\beta t}U(t)\) approximates the convolution integral with the sum \(F^n = \sum_{i=0}^{n-1} d_i^n\chi_i, d_i = \xi_0 E_i\), and all the approximation coefficients \(\chi_i\), except for the first two, can be found recursively, \(\chi_{i+1} = e^{\beta_t} \chi_i, i \geq 1\), then the polarization satisfies the recursive rule

\[
P^{n+1} = e^{\beta t} F^n + \chi_0 d^{n+1} + [\chi - e^{\beta t} \chi_0] d^n. \tag{10}
\]

**Lemma 2**: If an RC method satisfies the prerequisite of Lemma 1 then polarization for the 1/2 Padé approximant (5), where susceptibility is a composition of two exponential terms \(\chi(t) = \chi^+ + \chi^-\), \(\chi^\pm(t) = \alpha^\pm e^{\beta^\pm t}U(t)\), \(\alpha^\pm = -i\alpha e^{\beta_t/2}, \beta^\pm = -\gamma \mp i\delta\), can be evaluated recursively by the formula

\[
P^{n+1} = \beta_1 P^n + \beta_0 P^{n-1} + \alpha_2 d^n + \alpha_1 d^n + \alpha_0 d_0^{n+1} \tag{11}
\]

with \(\alpha_0 = [e^{\beta^+ t} \chi^+ - e^{\beta^- t} \chi^-] - \alpha_0 \beta_0, \alpha_1 = [\chi^+ - \chi^-] - \alpha_0 \beta_1, \alpha_2 = \chi^+ - \chi^0, \beta_1 = 2e^{\beta^+ t} \cos \delta t, \beta_0 = -e^{-\beta^+ t} \) being real coefficients.

The recursive rule (11) can be easily obtained for an ADE scheme after grouping terms, i.e. in (9), providing further generalized treatment for ADE and RC methods.

The assumption of Lemma 1 holds practically for all RC methods and is easy to apply after determining the first two approximation coefficients for the exponential case, summarized in Tables III, IV for a number of RC methods.

#### C. Dispersion Errors

One advantage of the generalized representation is the straightforward analysis of numerical aspects. For example, for bilinear ADE method the investigated stability condition coincides with the non-dispersive CFL stability condition if \(\sin \varphi \leq 0\) and \(\alpha_0 \beta_1 - \alpha_0 \beta_0 \geq 0\); in particular, the inequalities hold for any Drude, Sellmeier, Lorentz media, and the latter is in agreement with [2]. A study of stability for all Table I

---

**Table I**

<table>
<thead>
<tr>
<th>( a )</th>
<th>( b )</th>
<th>( b_0 )</th>
<th>( b_1 )</th>
<th>( b_2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \alpha_0 \cos \varphi - \gamma_0 \sin \varphi )</td>
<td>( -\alpha_0 \sin \varphi )</td>
<td>( \delta_0^2 + \gamma_0^2 )</td>
<td>( 2\gamma_0 )</td>
<td></td>
</tr>
<tr>
<td>D</td>
<td>( \alpha_0^2 )</td>
<td>0</td>
<td>0</td>
<td>( \Gamma_D )</td>
</tr>
<tr>
<td>L</td>
<td>( f_1 \alpha_0^2 )</td>
<td>0</td>
<td>( \alpha_0^2 )</td>
<td>( \Gamma_L )</td>
</tr>
<tr>
<td>S</td>
<td>( f_2 \alpha_0^2 )</td>
<td>( \alpha_0^2 )</td>
<td>0</td>
<td>( \Gamma_S )</td>
</tr>
<tr>
<td>C</td>
<td>( 2f_0 \alpha_0 (\cos \varphi - \gamma_0 \sin \varphi) )</td>
<td>( 2f_0 \alpha_0 (\cos \varphi - \gamma_0 \sin \varphi) )</td>
<td>( \Gamma_C^2 + \alpha_0^2 )</td>
<td>( 2f_0 \alpha_0 )</td>
</tr>
</tbody>
</table>

**Table II**

<table>
<thead>
<tr>
<th>( a )</th>
<th>( b )</th>
<th>( \gamma )</th>
<th>( \delta )</th>
<th>( \frac{\varphi}{\alpha} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \frac{1}{\sqrt{a^2 - b_0 b_1 + b_0^2 a_1}^{1/2}} )</td>
<td>( \frac{1}{b_1} )</td>
<td>( \sqrt{b_0 - \frac{b_0^2 - 4b_0^2 a_1}{b_1}} )</td>
<td>( \frac{\text{Arg}[a_0 - a_1]}{(\gamma + i\text{Re} \delta)} )</td>
<td></td>
</tr>
<tr>
<td>D</td>
<td>( -2i a_0^2/\Gamma_D )</td>
<td>( \frac{1}{\Gamma_D} )</td>
<td>( \frac{1}{2} \Gamma_D )</td>
<td>0</td>
</tr>
<tr>
<td>L</td>
<td>( f_1 \alpha_0^2/\delta )</td>
<td>( \frac{1}{\Gamma_L} )</td>
<td>( \sqrt{\alpha_0^2 - \frac{1}{4} \Gamma_L^2} )</td>
<td>0</td>
</tr>
<tr>
<td>S</td>
<td>( f_0 \alpha_0 )</td>
<td>0</td>
<td>( \alpha_0 )</td>
<td>0</td>
</tr>
<tr>
<td>C</td>
<td>( 2f_0 \alpha_0 )</td>
<td>( \Gamma_C )</td>
<td>( \alpha )</td>
<td>( \varphi )</td>
</tr>
</tbody>
</table>

**Table III**

<table>
<thead>
<tr>
<th>TRC</th>
<th>PCRC2</th>
<th>PLRC</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \chi_0 )</td>
<td>( \frac{1}{2} \alpha t )</td>
<td>( \alpha \beta^{-1} \left(e^{\beta t/2} - 1\right) )</td>
</tr>
<tr>
<td>( \chi_0 )</td>
<td>( \alpha \tau e^{\beta t} )</td>
<td>( \alpha \beta^{-1} \left(e^{\beta t/2} - 1\right) )</td>
</tr>
</tbody>
</table>

**Table IV**

<table>
<thead>
<tr>
<th>RRC</th>
<th>PCRC</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \lambda_0 )</td>
<td>( \alpha \tau )</td>
</tr>
</tbody>
</table>
RC methods may be carried out as in [11] for PLRC using coefficients in (11).

Calculation of dispersion error is usually complicated by numerous derivations for each model and each method, e.g. [8], [12], [13], and the resulting expressions are quite tedious. Here we end up with simple generalized expressions in terms of relative errors, truncated to a second order term.

The numerical susceptibility can be written by substituting the harmonic solutions $P_n = P_0 e^{-i \omega_n \tau}$, $E_n = E_0 e^{-i \omega_n \tau}$ into (11)

$$\chi_{\text{num}}(\omega) = \frac{P_0}{E_0} = \frac{\alpha_0 e^{-i \omega \tau} + \alpha_1 + \alpha_0 e^{i \omega \tau}}{e^{-i \omega \tau} - \beta_1 - \beta_0 e^{i \omega \tau}}.$$ (12)

By truncating it for different methods to a 2nd order term we obtain $(\Omega = 2 \tan(\omega \pi/2)/\pi$ and $\delta_{\text{num}} = (\chi_{\text{num}} - \chi)/\chi$)

$$\chi_{\text{num}}(\omega) = \frac{P_0}{E_0} = \frac{\alpha_0 e^{-i \omega \tau} + \alpha_1 + \alpha_0 e^{i \omega \tau}}{e^{-i \omega \tau} - \beta_1 - \beta_0 e^{i \omega \tau}}.$$ (12)

$$\delta_{\text{ADE}}(\omega) \approx \frac{\omega^2}{12} \left( 1 - \frac{2 \gamma}{\omega} \right) \left( 1 - \frac{2 \delta}{\omega} \right)$$

$$\delta_{\text{RC}}(\omega) \approx \frac{\omega^2}{12} \left( 1 - \frac{2 \gamma}{\omega} \right) \left( 1 - \frac{2 \delta}{\omega} \right)$$

$$\delta_{\text{PCRC}}(\omega) \approx \frac{\omega^2}{12} \left( 1 - \frac{2 \gamma}{\omega} \right) \left( 1 - \frac{2 \delta}{\omega} \right)$$

Besides just proving the theoretical second order of accuracy this brings a few interesting facts. First, the PLRC relative error of non-dispersive if the time step is adjusted for each frequency so that $\omega \tau = \text{const}$, this should be used for CW runs. Second, for any system having a zero instantaneous response $(\chi(0) = 0 \Leftrightarrow \sum a_i \sin(\omega_i \tau_i) = 0)$, in particular for any number of Drude, Lorentz, Sellmeier terms, the sine term in (14) vanishes in the overall numerical error and PCRC becomes twice more accurate than PLRC, in terms of susceptibility relative error.

D. Coupling With Maxwell’s Time-Domain Solvers

First we couple the local polarization recurrence (11), obtained by either some ADE approximation or by some RC method, with Yee’s FDTD scheme. The update equation for the electric field written in some fixed coordinate system is shown here

$$\tau \nabla \times \mathbf{H}^{n+\frac{1}{2}} = \varepsilon_0 c_0 (\mathbf{E}^{n+1} - \mathbf{E}^n) + \sum_i \left( \mathbf{P}^{n+1}_i - \mathbf{P}^n_i \right)$$

$$\mathbf{P}^{n+1}_i = \beta_i^2 \mathbf{P}^n_i + \beta_i \mathbf{P}^{n+1}_i + \varepsilon_0 \left( \alpha_2 \mathbf{E}^{n+1} + \alpha_1 \mathbf{E}^n + \alpha_0 \mathbf{E}^{n+1} \right).$$

The system can be reorganized for effective computation as

$$\mathbf{E}^{n+1} = \xi_1^{-1} \left[ \xi_0 \mathbf{E}^n + \tau \varepsilon_0 c_0 \nabla \times \mathbf{H}^{n+\frac{1}{2}} - \sum_i \mathbf{P}^{n+1}_i \right]$$

$$\mathbf{P}^{n+1}_i = \beta_i^2 \mathbf{P}^n_i + \beta_i \mathbf{P}^{n+1}_i + \varepsilon_0 \left( \alpha_2 \mathbf{E}^{n+1} + \alpha_1 \mathbf{E}^n + \alpha_0 \mathbf{E}^{n+1} \right) \mathbf{E}^n$$

$$i = I + i_k$$

where $\xi_i = \xi_0 + \alpha_2^2 \beta_i$, $\eta_i = \alpha_2 + \alpha_2 \beta_i$, $\xi_1 = \varepsilon_1 + \sum \alpha_i^2$, and $\xi_0 = \xi_1 - \sum \eta_i \eta_i$.

A brief comparison can be done to the conventional realization of RC methods for Lorentz media: even the simplest PCRC method [3] requires 10 flops to update the complex recursive accumulator $\Psi^{n+1}_i = C \Psi^n_i + \Delta \chi_0 \mathbf{E}^n$, while in (16) it is updated in 7 flops for all RC methods listed in Tables III, IV. The memory allocation is the same: 1 additional float store per pole.

Since the Drude model has $\beta_0^2 = 0$ we resolve the singularity 0/0 in Tables III, IV by expanding the numerator in the Taylor series.

E. Drude and Over-Damped Lorentz Models

Although the previously discussed approach works for Drude and over-damped Lorentz models directly, a more efficient implementation can be obtained if an additional expansion to two real exponential poles is performed using (5) and then each pole is treated separately. The zero pole of the Drude model is traditionally implemented directly to Yee’s scheme as a conductivity term with $\sigma = \varepsilon_0 \omega \mu_0$. Each of the non-zero real poles $\beta_i^0$ can be evaluated using again either ADE approach applied for the ODE of the first order or recursive sum (10); both end up with the recurrence $\mathbf{P}^{n+1}_i = \beta_i^0 \mathbf{P}^n_i + \alpha_2^0 \mathbf{E}^{n+1} + \alpha_1^0 \mathbf{E}^n$. Then the final set of equations for the conductive ($\sigma$) dispersive media that has $i_c$ conjugate poles and $i_r$ real poles can be written as

$$\mathbf{E}^{n+1} = \xi_1^{-1} \left[ \xi_0 \mathbf{E}^n + \tau \varepsilon_0 c_0 \nabla \times \mathbf{H}^{n+\frac{1}{2}} - \sum_i \mathbf{P}^{n+1}_i \right]$$

$$\mathbf{P}^{n+1}_i = \beta_i^0 \Psi^n_i + \Phi^0_i + \left( \eta^{0}_i + \left( \beta_i^0 - 1 \right) \eta^{1}_i \right) \mathbf{E}^n$$

$$\mathbf{P}^{n+1}_i = \beta_i^0 \Psi^n_i + \left( \eta^{1}_i - \left( \beta_i^0 - 1 \right) \eta^{1}_i \right) \mathbf{E}^n$$

$$i \in I_c$$

$$i \in I_c$$

$$i \in I_r$$

where $\xi_1 = \varepsilon_1 + (\sigma/\varepsilon_0) + \sum \alpha_i^2$, $\xi_0 = \xi_1 - \sigma/\varepsilon_0$, and sets of indices denoted as $I_c = 1 + i_c$, $I_r = r + i_r + 1$, $I = I_c \cup I_r$. Note that the formula for implementation of a real pole is obtained simply as a reduction of (16) for $\alpha_0^0 = \beta_0^0 = 0$.

V. NUMERICAL EXPERIMENTS

In our numerical experiments we use the proposed implementation of ADE and RC methods in FDTD and finite-volume time domain (FVTD) solvers. Finite-element (FETD) simulations were done using commercial FE software (COMSOL MULTIPHYSICS, RF module) for transient propagation of an in-plane wave. Local polarization was simultaneously solved using a separate coefficient form of the coupled PDE model (6) for each of the D, L or C terms, i.e. using the ADE approach.

2D time-domain simulations were performed for spectroscopic analysis of a periodic 2D nanostructured sample Fig. 1(a). The dispersive function of gold was approximated as a sum of a Drude term and four Lorentz oscillators (D4L model) or as a sum of a Drude term and two critical points terms (D2CP model). The detailed parameters of both models are given in Table V.

For both TE and TM polarizations, the sample is illuminated with a Gaussian pulse (761-nm wavelength carrier, 3.15 FWHM, 11 fs offset) with the computational domain being $2 \mu$m long. The field probes located at the beginning and end of the domain are post-processed with FFT to get the numerical reflectance and transmittance spectra. The latter is compared in Fig. 1(b), (c) to the spectra obtained from a semi-analytical tool based on spatial harmonic analysis [14]. To compare the numerical accuracy of all three methods we use a uniform mesh in the entire domain and the Courant number is set to 1/2.
For the TE case, FDTD and FVTD methods reach satisfactory accuracy for reflectance and transmittance by resolving the object width with 4 numerical cells (dx = 2.5 nm). In FETD the same level of accuracy is reached by taking higher order of elements in the ADE domain with twice larger mesh size, dx = 5 nm, see Fig. 2.

TM incidence is complicated by longer plasmonic echo and two components of the electrical field involved in the dispersion relation. The nanostructure sample has resonance at about 650 nm. The starting point of accuracy is an FDTD run with a 5-nm space step, which performs quite well. FVTD simulations converge much slower and reach the same level of accuracy only with a 1.25-nm mesh size. For the FETD solver it is again sufficient to increase the order of elements, while having the same coarse mesh with 5-nm step, see Fig. 2.

VI. CONCLUSION

A systematic approach to time-domain modeling of dispersive dielectric functions is proposed based on 2nd order Pade approximants, which includes Drude, Lorentz, Sellmeier and critical points models. The approach provides a generalized set of update equations for TD solvers with real coefficients depending on the method, e.g., ADE, PLRC, PCRC, PCRC2, TRC, thus unifying the computational costs of the methods. Different dispersive methods provide different dispersion errors, which are analyzed theoretically. The analysis shows that PCRC2 relative dispersion error is about twice less than that of PLRC method. The approach is applied to 2D simulations of a periodic metamaterial with FDTD, FVTD and FETD methods. Additionally, we see that due to its larger number of dispersion terms and smaller damping constants, the D4L model of gold requires about four times longer simulation time than the D2CP model.

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