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Comparison of gold and silver dispersion laws suitable for FDTD simulations

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Abstract We show that it is possible to increase the accuracy of gold and silver permittivity description by using the Drude–critical points model rather than the widely used Drude–Lorentz model. We also show the effect of this improvement on the extinction efficiency and near-field intensity precision.

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1 Introduction

Following the ever increasing use of the Finite-Difference Time-Domain (FDTD) method for electromagnetic simulations [1], there has been a growing interest in the description of metal permittivities by means of analytical models. Indeed, the accuracy of spectroscopic studies performed using the FDTD method strongly relies on the accuracy of the permittivity description by means of causal analytical laws over the range of wavelengths of interest. For this reason, metals like gold or silver are generally described using the Drude model with the addition of one or more Lorentzian terms [2–7].

In a recent paper [8], Hao and Nordlander also proposed an improved analytical model (called L4) for the dielectric response of Au and Ag. This model consists of four Lorentzian terms (which may reduce to Drude terms) and a constant conductivity term, and aims at fitting Au and Ag dielectric data from in the 180–2000 nm range. In particular, it was shown that its accuracy was better in the 200–1000 nm range than that of the Drude model with up to five Lorentzian terms, as previously published in [9].

Nevertheless, it is known that increasing the number of terms used to describe the permittivity of dispersive media also increases the memory requirements of the FDTD code, whatever the chosen implementation [10]. For this reason, the critical points (CP) model recently introduced for the description of the permittivity of gold in the 200–1000 nm range [11, 12] is an interesting alternative to the Lorentz model, as only two additional terms to the Drude model are required for obtaining good accuracy.

In this paper, we will compare the efficiency of both L4 and Drude–2CP models for the description of gold and silver dielectric functions. In Sect. 2, we will briefly recall the two models and the consequences in terms of memory requirements. In Sect. 3, we will compare the results for both models in terms of dielectric function description, extinction efficiency and near-field intensity. Finally, we will conclude in Sect. 4.

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2 Dielectric functions models

2.1 L4 model

A model including 4 Lorentzian-pole pairs (L4) was recently published [8] for the description of Au and Ag dielectric

functions for wavelengths between 180 and 2000 nm. This model can be expressed as

$$\epsilon(\omega) = \epsilon_\infty + \frac{\sigma/\epsilon_0}{\imath\omega} + \sum_{p=1}^{4} \frac{C_p}{\omega^2 + A_p \imath\omega + B_p}, \tag{1}$$

and its numerical implementation using the auxiliary differential equation (ADE) method or the recursive convolution (RC) method can be easily derived using previous works [1, 2, 5, 10, 13, 14].

While this model is certainly of interest for the study of highly tunable structures [15, 16], the reduced wavelength range 200–1000 nm is generally sufficient for studying plasmon resonances of typical Ag and Au nanoparticles, as emphasized by examples given by Hao and Nordlander themselves [8]. In order to reduce the computational effort (time, of course, but mainly memory in the case of the FDTD) it is advantageous to use as few Lorentzian terms as possible, even at the cost of a smaller spectral width. This is the reason why many authors used only one or two Lorentzian terms in addition to the Drude one [2, 3]. Moreover, depending on the way dispersion is taken into account in the FDTD code, parameters given in Table 1 of [8] may rapidly increase the memory requirements. Both ADE and RC methods require the introduction of additional fields (**P** or $\boldsymbol{\psi}$) for each additional term in the permittivity description. For the particular case of the recursive convolution method with Lorentzian terms, a key requirement is (using notation of [8]) $|A_p/2|^2 < |B_p|$. If this is not verified (as it can be seen on several lines of Table 1 of [8] for Au and Ag), we then face the over-damped case [17], which can be treated at the cost of the introduction of further additional vector fields, which does not minimize the overall computational effort. As a side note concerning values given in Table 1 of [8], it would be interesting to know if $\epsilon_\infty = 1$ was set by the authors, or if it was the result of the optimization process. Not constraining ϵ_∞ should facilitate the overall optimization process.

2.2 Drude–critical points model

Recently, a critical points (CP) model has been used together with the Drude model for the description of the permittivity of gold in the 200–1000 nm range [11, 12]. The D+2CP model can be expressed as:

$$\begin{aligned}\epsilon_{\mathrm{DCP}}(\omega) &= \epsilon_\infty - \frac{\omega_D^2}{\omega^2 + i\gamma\omega} \\ &\quad + \sum_{p=1}^{2} A_p \Omega_p \left(\frac{e^{i\phi_p}}{\Omega_p - \omega - i\Gamma_p} + \frac{e^{-i\phi_p}}{\Omega_p + \omega + i\Gamma_p} \right). \end{aligned} \tag{2}$$

It can also easily be applied to the case of Ag, as it will be shown in Sect. 3.

Its numerical implementation using the RC method has been published previously [18, 19], and it was found that in this case there is no concern with the relative values of the parameters. On the other hand, one needs to check that $|\epsilon_\infty/(\epsilon_\infty + \chi_0)| < 1$ (see [18, 19] for the meaning of χ_0). It was shown that each CP term requires the same amount of memory as a Lorentz term.

If one wishes to use the ADE method rather than the RC method, then the temporal relationship between the polarization vector **P** and the electric field **E** has to be made explicit as

$$\begin{aligned}&\left(\frac{\partial^2}{\partial t^2} + 2\Gamma_p \frac{\partial}{\partial t} + \Omega_p^2 \right) \mathbf{P_p}(t) \\ &\quad = 2A_p \Omega_p \left(\sqrt{\Omega_p^2 + \Gamma_p^2} \sin(\theta_p - \phi_p) - \sin\phi_p \frac{\partial}{\partial t} \right) \mathbf{E}, \end{aligned} \tag{3}$$

with $\tan(\theta_p) = \frac{\Omega_p}{\Gamma_p}$, and one of the many possible discretization schemes has to be applied [5, 10, 13]. Equation (3) is very similar to the equation of a damped, driven harmonic oscillator, but with the addition of a term proportional to the derivative of the electric field. A simple physical interpretation for this term has not yet been found.

3 Comparison of the models

In this section we will compare the quality of the fits achieved using the L4 and the D+2CP models, respectively. L4 parameters were taken from [8], and the D+2CP parameters were obtained using a simulated annealing procedure. Results for Au and Ag are displayed in Table 1. We decided to use our own values for gold, rather than using values previously published [12] where ϕ_1 and ϕ_2 were set to $-\pi/4$.

Table 1 Parameters for the Drude+2CP fit of the dielectric functions of Au and Ag for $200 < \lambda < 1000$ nm (experimental data from [20])

	ϵ_∞	ω_D (rad · s^{-1})	γ (rad · s^{-1})	A_1	ϕ_1	Ω_1 (rad · s^{-1})	Γ_1 (rad · s^{-1})	A_2	ϕ_2	Ω_2 (rad · s^{-1})	Γ_2 (rad · s^{-1})
Au	1.1431	1.3202E16	1.0805E14	0.26698	−1.2371	3.8711E15	4.4642E14	3.0834	−1.0968	4.1684E15	2.3555E15
Ag	15.833	1.3861E16	4.5841E13	1.0171	−0.93935	6.6327E15	1.6666E15	15.797	1.8087	9.2726E17	2.3716E17

3.1 Gold

In Fig. 1 we have plotted the real and imaginary parts of the dielectric function of gold as tabulated in [20], as well as the description achieved using the D+2CP and the 4L models, respectively. We can see that both models lead to a satisfying agreement with experimental data.

We have then used the three permittivity descriptions for the computation of the extinction efficiency and the near-field intensity 5 nm above a gold sphere of radius 80 nm using Mie theory. Results are presented in Fig. 2, and both analytical models give results in good agreement with those obtained using tabulated data.

3.2 Silver

The dielectric function of silver is studied in Fig. 3. It can be seen that both the D+2CP and the L4 models are close to experimental data, except for $\lambda < 300$ nm, where the D+2CP model performs better. Indeed, the L4 model predicts positive values for the real part of the permittivity for $\lambda < 300$ nm.

These differences of behavior are emphasized when studying the extinction efficiency of a silver sphere of radius 10 nm and the near-filed intensity 5 nm above such a sphere, as shown in Fig. 4. The 4L model predicts an extinction efficiency of 60, and a near-field intensity of 592 (a.u.) at $\lambda = 354$ nm, whereas the D+2CP model gives, respectively, 3 and 3.6, and values obtained using tabulated data are 7 and 9. So, in this case, both analytical models fail to give an accurate value of the extinction efficiency and the near-filed intensity, but the D+2CP provides at least order of magnitude agreement. This discrepancy is actually related to the fact that for $\lambda = 354$ nm, the 4L model predicts $\epsilon_{\mathrm{Ag}} = -2.0764 + 0.0250i$, which is very close to the well-know resonance condition $\epsilon = -2$ for a small sphere [21], whereas the experimental value is $\epsilon_{\mathrm{Ag}} = -2.0036 + 0.2838i$. The almost vanishing imaginary part of the 4L model explains the difference.

It should be also noted that both models lead to a negative imaginary part of the dielectric function in some parts of the spectrum, which may lead to artificially increasing fields. Nevertheless, this would be a problem for the FDTD method only for plane-wave studies, in which case adapted Drude parameters should actually be used rather than models specifically developed for spectroscopic studies.

3.3 Other metals: aluminum and chromium

In order to further emphasize the general applicability of the D+2CP model, we have calculated parameters for two other

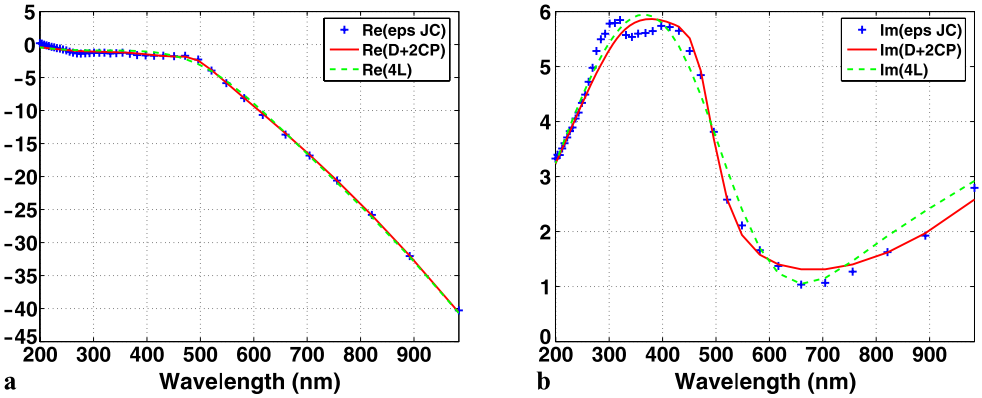

Fig. 1 Tabulated gold dielectric function [20] and comparison with the D+2CP and L4 models. (**a**) Real part. (**b**) Imaginary part

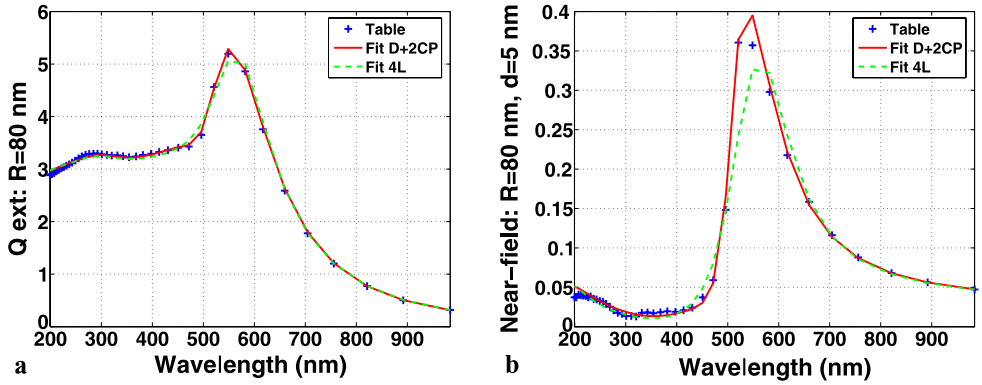

Fig. 2 Study of a gold sphere of radius 80 nm using tabulated data and the D2CP and L4 models. (**a**) Extinction efficiency. (**b**) Near-field intensity 5 nm above the sphere

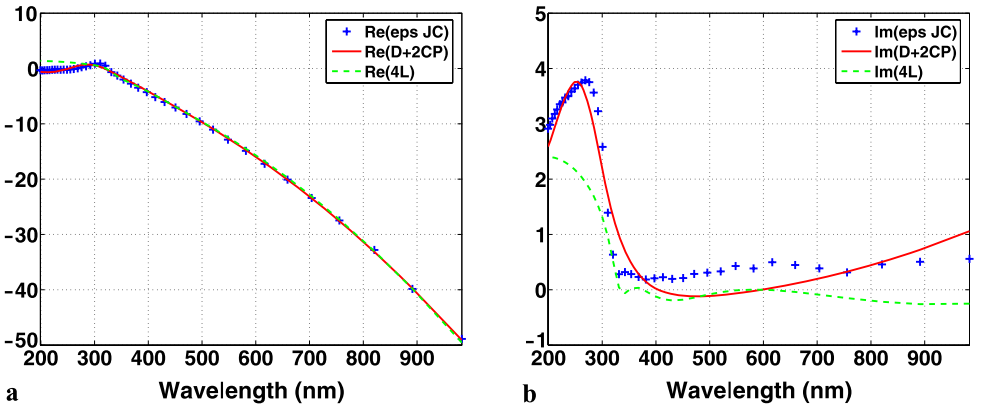

Fig. 3 Tabulated silver dielectric function [20] and comparison with the D+2CP and L4 models. (**a**) Real part. (**b**) Imaginary part

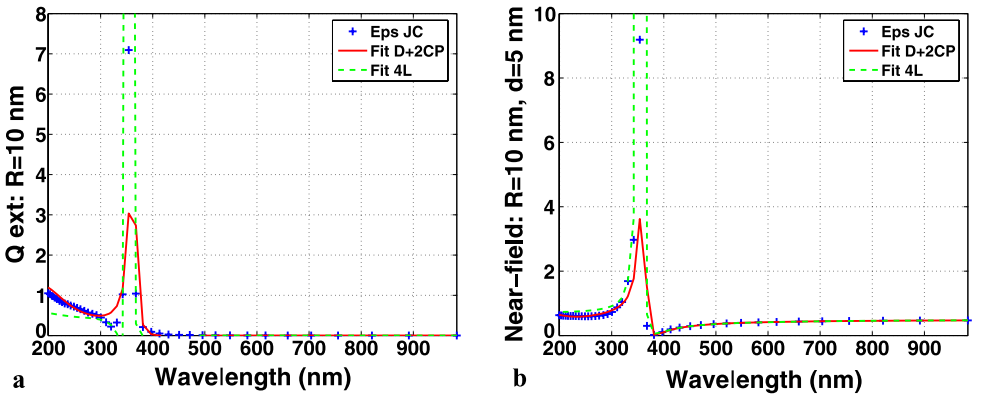

Fig. 4 Study of a silver sphere of radius 80 nm using tabulated data and the D2CP and L4 models. (**a**) Extinction efficiency. (**b**) Near-field intensity 5 nm above the sphere

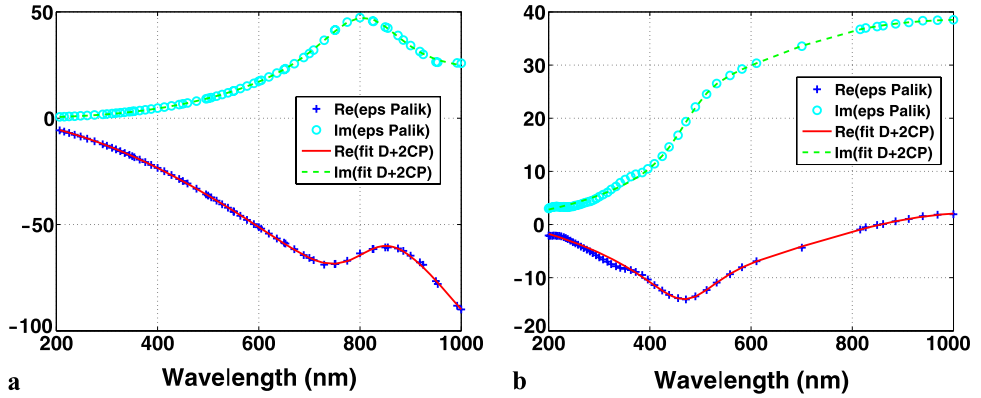

Fig. 5 Tabulated values [22] and fit using the D+2CP model of Al and Cr. (**a**) Aluminum dielectric function. (**b**) Chromium dielectric function

Table 2 Parameters for the Drude+2CP fit of the dielectric functions of Al and Cr for $200 < \lambda < 1000$ nm (experimental data from [22])

	ϵ_∞	ω_D (rad · s^{-1})	γ (rad · s^{-1})	A_1	ϕ_1	Ω_1 (rad · s^{-1})	Γ_1 (rad · s^{-1})	A_2	ϕ_2	Ω_2 (rad · s^{-1})	Γ_2 (rad · s^{-1})
Al	1.0000	2.0598E16	2.2876E14	5.2306	−0.51202	2.2694E15	3.2867E14	5.2704	0.42503	2.4668E15	1.7731E15
Cr	1.1297	8.8128E15	3.8828E14	33.086	−0.25722	1.7398E15	1.6329E15	1.6592	0.83533	3.7925E15	7.3567E14

metals: aluminum and chromium. Both of them were previously studied in a reduced range of wavelengths [18, 19], but in Table 2 we give parameters in order to fit tabulated values available in [22] for $200 < \lambda < 1000$ nm.

It can be verified in Fig. 5 that the agreement with tabulated data is good.

4 Conclusions

We have shown that the Drude–critical points model was able to accurately describe the dielectric functions of gold and silver, with fewer parameters to determine than the L4 model, and also a lower memory footprint when used in the

FDTD method framework. In the particular case of silver, the accuracy of D+2CP is clearly better than that of L4, and this may be of great importance if one wishes to study plasmon propagation length [23], for example. On the other hand, as it was already previously emphasized [11], parameters given here are mainly valid for bulk structures, and an accurate modelling of plasmonic devices should take into account the dielectric functions modifications induced by deposition processes or nanostructuration [24, 25].

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