A Dynamic Approach to the Lumped Impedance Representation of A Nanoparticle

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Abstract: Recent research on plasmonic nanoparticles have emphasized their use in the circuits and device-based applications by exploiting control of the plasmon resonance. The resonance behavior of the nanoparticle can be explained by its lumped-impedance representation at optical frequencies, which is derived using voltage-current (VI) laws. The VI model uses quasi-static approximation, which is inherently restricted, and can only work well for small radii (R < 10 nm) nanoparticles. In this paper, an extended approach is proposed by solving for the full dipole equation, including the radiation damping for the dominant mode of the oscillating nanoparticle. The proposed approach employs taking all time-dependent fields of a dipole and introduced the concept of radiation impedance, which can store and absorb radiated power. Closed form expressions for the quantities, such as internal and external impedance's are derived using voltages and currents, giving good understanding of the behavior of a metallic nanoparticle. A comparison with the Mie solution, demonstrates that our proposed impedance model extends the range of the impedance model to the larger radii nanoparticles.

Index Terms: Plasmonics, nanoparticles, plasmon resonance, dipole, nanophotonics, impedance and lumped circuits.

1. Introduction

There has been a growing interest in the study of electromagnetic waves interaction with metallic nanostructures at optical frequency [1], [2], [3], [4], [5]. In these nanostructures, nanoparticles are the most used structures. They form the basic building block, responsible for the potential applications in sensor, biomedical, photonics and electronics [6], [7], [8], [9]. The analytical solution to the nanoparticle optical scattering, absorption and extinction power is derived from the infinite series expansion, consisting of the vector spherical harmonics. The solution is well known, however it lacks the much needed physical insight of the problem. In order to develop the better understanding of the qualitative behavior, several studies are carried out to approximate the behavior of the nanoparticle [10], [11]. The first such general approximation is Wiscombe’s criterion, where the solution is truncated for a suitable finite number of terms (n_{max}) [12]. Several methods, since then have been proposed for the criteria of choosing n_{max}, however any approximate solution based on the truncated series is still large enough to lay sound conceptual foundation and good understanding of the localized surface plasmon resonance [13].

A nanoparticle, which is rotationally symmetric nanostructure, present a linear dynamic geometric problem. Its analytical solution also known as Mie solution is often used as a benchmark to calibrate various numerical field solvers. The energy flow during the resonance, computed using 3D Mie solution for a 10 nm nanoparticle is plotted in Fig 1a. The resonance phenomena in metallic nanoparticle reminds us of the valuable property of the electric circuit, where lumped
Fig. 1: (a) A vector field plot of the Poynting vector in $xy$ plane for a nanoparticle of radius $10\text{nm}$, showing strong dipole response. The Dynamic VI model retains the quasi-static term (near-field term $1/r$) of Quasi-static model, and also includes the far-field ($1/r^2$) and retardation-field ($1/r^3$) terms. (b) Resonance wavelength is plotted against the changes in radius. The Quasi-static VI model [17] is compared with Mie solution [25]. The infinite series of Mie solution is truncated and number of terms preserved are determined by Wiscombes criterion for a maximum value of $n_{\text{max}} = kR + 4(kR)^{1/3} + 2$ ranging between 4 and 9, where $k = \omega/\sqrt{\varepsilon_{\text{m}}}$ [12].
approach based on full time-dependent model of the fundamental mode. These refinement in the
theory leads to major changes in the VI model to the impedance representation and extend the
validity of the approach to include larger radii nanoparticles.

The rest of the paper is organized into four sections. In Section II, the theoretical preliminaries
of the Quasi-static VI model are discussed. The proposed derivation of the extended impedance
model (Dynamic VI model) of a nanoparticle is discussed in Section III. Simulation results are
presented in Section IV by comparing proposed Dynamic VI model with the Quasi-static VI model,
full-wave Finite-Difference Time-Domain (FDTD) simulation and the Mie solution. Finally, the paper
is concluded in Section V.

2. Quasi-static Approach to Lumped Circuit Representation of a
Nanoparticle

The analytical field solution also known as Mie solution uses plain wave and describe the scat-
tering, absorption and extinction cross-sections of a sphere in terms of infinite series expansion.
The expression for scattering cross-section highlights the complexity of the field approach, and
can be written as

\[ Q_{sca} = \frac{2\pi}{k_0^2} \sum_{n=1}^{\infty} (2n + 1)(|a_n|^2 + |b_n|^2), \]

where the coefficients \( a_n \) and \( b_n \), can be written as

\[ a_n = \frac{j_n(k_1 R)[k_2 R j_n(k_2 R)]' - j_n(k_2 R)[k_1 R j_n(k_1 R)]'}{j_n(k_1 R)[k_2 R h_n^{(2)}(k_2 R)]' - h_n^{(2)}(k_2 R)[k_1 R j_n(k_1 R)]'}, \]

\[ b_n = \frac{j_n(k_2 R)[k_1 R j_n(k_1 R)]' - c_1 j_n(k_1 R)[k_2 R j_n(k_2 R)]'}{h_n^{(2)}(k_2 R)[k_1 R j_n(k_1 R)]' - c_1 j_n(k_1 R)[k_2 R h_n^{(2)}(k_2 R)]'}. \]

Where \( j_n \) is the spherical bessel function of first kind and the prime at square bracket is differ-
entiation with respect to argument. Note that (1) is a multi-variable infinite series expansion of
\( Q_{sca} \), where the coefficients \( a_n \) and \( b_n \) are the function of frequency, radius of the nanoparticle
and the dielectric function dependent wave vector of the nanoparticle \( (k_1) \) and the surrounding
medium \( (k_2) \) [25]. In general, the circuit theory is often used as analogous to the field theory
for circuit analysis at low frequency [14], [20]. The underlying assumption is a special case
of field theory, where circuit operating at low frequencies are much smaller compared to the
wavelength and hence the physical size of the component becomes insignificant [21]. This lumped
circuit approximation at low frequency is know to be universal, however emulating similar practice
at higher frequency becomes very complex. The circuit analysis becomes significantly more
complicated for metal at optical frequency, where they also exhibit negative permittivity.

An early approach to model the nanostructures as nanocircuit elements i.e. nanocapacitors,
nanoinductors and nanoresistors is based on Quasi-static VI model [17]. It is of some interest to
note that the VI model adopt the same classical method used in circuit theory for the analysis of
voltages and currents at low frequencies. However, when carefully applied to a mathematically
well conditioned problem, it can be extended to much higher frequencies. In solving classic
problem of scattering from a metallic nanoparticle, Quasi-static VI model uses lumped circuit
approximation, i.e. \( R << \lambda_0 \) in describing the fields inside and outside the sphere. Considering
an electromagnetic wave \( (E_o) \) incident on a sphere with size much smaller than the wavelength,
the wave will excite fields inside \( (E_{in}) \) and outside \( (E_{ext} = E_o + E_{dip}) \) the sphere, where \( E_{dip} \) is
the dipole field created by the initial excitation. The dipole field valid in \( r > R \) will create the potential
inside and outside the nanoparticle and under quasi-static approximation is given as

\[ E_{dip} = \frac{3r \cdot \hat{p} \cdot \hat{r} - \hat{p}}{4\pi \varepsilon_0 r^3}, \]
where \( \mathbf{p} \) is the polarizability of the nanoparticle and \( \mathbf{r} \) is the position vector. Note that (4) represent the dipole field under quasi-static approximation, where only the near field effects are taken into consideration. The incident electromagnetic field creates the residual field inside the sphere and is responsible for generating the displacement current circulating in the sphere. In a similar manner, the displacement current of the fringe is derived by integrating the dipole field given by (4). For the voltages, the residual field inside the sphere is responsible for creating the potential difference between the upper and lower hemispherical surfaces, thus giving the average potential (i.e. \(<V_{\text{sph}}>\) and \(<V_{\text{fringe}}>\)) available for computing effective impedance’s.

The equivalent impedance’s can be expressed as the ratio of the voltages and currents as follows

\[
<Z_{\text{sph}}> = \frac{<V_{\text{sph}}>}{<I_{\text{sph}}>} = \frac{1}{-i\omega\epsilon\pi R} , \quad <Z_{\text{fringe}}> = \frac{<V_{\text{fringe}}>}{<I_{\text{fringe}}>} = \frac{1}{i\omega 2\pi R} .
\]

The compact form of \(<Z_{\text{sph}}>\) and \(<Z_{\text{fringe}}>\) clearly highlights the importance of the closed form solution offered by the Quasi-static VI model. The developed model modifies the complex dynamical behavior into much simpler impedance-based system, where linear system theory can be used for analysis. However, the accuracy of the derivation is far from ideal. Fig 1b plots resonance wavelength against the changes in the radius of a gold nanoparticle. It shows that for the larger radii, Quasi-static VI approach does not accurately predict the resonant wavelength of the nanoparticle. In the next section, an extended approach by including the effects of the full dipole equation is proposed, thereby extending the range of VI approach to the larger radii nanoparticles.

3. Proposed Dynamic VI Model (Includes Near-Field, Far-Field and the Retardation Effects)

Let us start with the derivation of the accurate expression for the impedance representation of a nanoparticle and compare the results with the Quasi-static VI model, FDTD simulation and the Mie solution. It is evident from the initial assumption of the Quasi-static VI model that any increase in the radius of the sphere will adversely effect the accuracy of the results. Without loss of generality, let us consider a uniform plane wave incident on a nanoparticle with radius \( R \) and complex permittivity \( \epsilon(\omega) \). The initial excitation will generate the electric field \( \mathbf{E}_{\text{int}} \) inside the sphere, which is parallel to the incident E-field [22]. The internal field \( \mathbf{E}_{\text{in}} \) can be expressed as follows

\[
E_{\text{int}} = \frac{3\epsilon_o \mathbf{E}_o}{(\epsilon + 2\epsilon_o)} .
\]

Similarly, the full time-dependent field of an oscillating dipole can be written as [22]

\[
E_{\text{dip}} = \frac{1}{4\pi \epsilon_o} \left[ (1 - ikr) \frac{3\mathbf{r} \cdot \mathbf{p} - \mathbf{p}}{r^3} + k^2 \frac{\mathbf{p} - \mathbf{r} \cdot \mathbf{p}}{r} \right] .
\]

The polarizability \( \mathbf{p} \) of the dipole field responsible for the dynamic response of the external field is given as \( \mathbf{p} = 4\pi \epsilon_o R^2 \mathbf{E}_o [(\epsilon - \epsilon_o)/(\epsilon + 2\epsilon_o)] \), where \( R \) is the radius of the nanoparticle. Note that (7) represent the fully retarded solution of the dipole field. It retains the quasi-static term (near-field \( 1/r^3 \) term), and also includes the \( 1/r \) (far-field) and \( 1/r^2 \) (retardation-field) terms. In order to derive the expression for the voltage, let us consider the two hemispherical surfaces i.e. the upper and lower hemispherical surfaces of the nanoparticle. The potential difference between the two hemispherical surfaces can be expressed as

\[
<V_{\text{sph}}> = <V_{\text{fringe}}> = R \frac{(\epsilon - \epsilon_o)}{(2\epsilon + \epsilon_o)} \mathbf{E}_o .
\]

The voltages defined above for both inside and outside the nanoparticle are same. Likewise, the incident electric field, which excites the surface charges and become source of the impressed
displacement current \( (I_{imp}) \), the displacement current of sphere \( (I_{sph}) \), and the fringe displacement current \( (I_{fringe}) \). The integration of the respective field over the upper hemisphere, leads to the following expression of the currents

\[
< I_{imp} > = -i\omega (\epsilon - \epsilon_o) 2\pi R^2 E_o , \quad < I_{sph} > = -i\omega \frac{(\epsilon - \epsilon_o)}{(2\epsilon + \epsilon_o)} 2\pi R^2 E_o , \quad (9)
\]

\[
< I_{fringe} > = i\omega \pi R^5 \frac{(\epsilon - \epsilon_o)}{2(\epsilon + \epsilon_o)} \left( 1 - i\frac{kR}{R} \right)^2 R^3 + k^2 R^3 E_o . \quad (10)
\]

Note that the above three equations obeys Kirchoff's current law i.e. \( < I_{imp} > = < I_{sph} > + < I_{fringe} > \). This form of voltage and current representation is similar to the circuit analysis, where they can form simple set of equations, and can be used to derive values of the equivalent circuit elements. It is important to note that in the proposed model, the displacement currents are derived using the full dipole equations. These accurate derivations of the currents will provide better understanding of the resonance phenomena and will include the much needed accuracy to the VI approach, thereby giving an improved modeling tool for synthesizing of the next generation nanophotonic circuits.

The above derivation now allow us to define voltages and current along with the associated impedance's. Therefore, the internal \( < Z_{sph} > \) and external impedance \( < Z_{fringe} > \) of an oscillating metallic nanoparticle can be written as

\[
< Z_{sph} > = \frac{< V_{sph} >}{< I_{sph} >} = \frac{1}{-i\omega \pi R} , \quad < Z_{fringe} > = \frac{< V_{fringe} >}{< I_{fringe} >} = \frac{1}{i\omega \pi \epsilon_o R^4 \left( 1 - i\frac{kR}{R} \right)^2 R^3 + k^2 R^3} . \quad (11)
\]

The equivalent nanocircuit elements of the spherical and fringe branch are lumped elements i.e. nanoinductor, nanocapacitor and nanoresistor and are given as following.

\[
L_{sph} = \left( -\omega^2 \pi R \Re \{\epsilon\} \right)^{-1} , \quad G_{sph} = \pi \omega R \Im \{\epsilon\} , \quad (12)
\]

\[
C_{fringe} = C_{fringe1} + C_{fringe2} = 2\pi \epsilon_o R + \pi \epsilon_o k^2 R^3 , \quad G_{fringe} = 2\pi \omega \epsilon_o k R^2 . \quad (13)
\]

Where \( L_{sph} \) and \( G_{sph} \) are the self-inductance and conductance, responsible for the internal impedance of the nanoparticle. The \( C_{fringe} \) and \( G_{fringe} \) are the capacitance and conductance representing the external impedance of the nanoparticle. Note that the fringe capacitance has two parts, one is quasi-static \( (C_{fringe1}) \) and the other is dynamic \( (C_{fringe2}) \). Fig. 2 shows the proposed...
dynamic nanocircuit model illustrating the addition to the Quasi-static VI model to provide much needed accuracy to the impedance representation.

It is important to note that the derivation of the two new components \( C_{\text{fringe}} \) and \( G_{\text{fringe}} \) highlights the importance of the proposed Dynamic VI model. The new lumped component \( C_{\text{fringe}} \) is proportional to \( R^3 \) and accounts for the energy stored in the far-field. Note that the it is this far-field term, which will survive the longest and is mainly responsible for the radiated power. In addition, \( G_{\text{fringe}} \) represents the frequency-dependent leakage current through the dielectric. It reduces as we go away from the nanoparticle and actually accounts for the retardation effect and is contributed by the \( 1/r^2 \) term. The concept of external impedance is analogous to the radiated impedance, which is mainly responsible for radiated power loss and storage outside the nanoparticle. Note that the addition of these components brings them close to the exact solution (Mie solution) and give the much needed accuracy to the VI approach to represent nanostructures as circuit elements.

3.1. Convergence to Quasi-static VI model (under quasi-static conditions)

In this section, we will show that under the quasi-static conditions our proposed Dynamic VI model converges to the Quasi-static VI model [17]. It is noted here that the internal impedance of the two models are same as we can see from (5) and (11). In order to prove the convergence of the external impedance, we will apply quasi-static approximation to our proposed Dynamic VI model i.e. \( (R << \lambda_0) \). Let us first simplify (11) as follows

\[
Z_{\text{fringe}}|_{\text{Dynamic VI}} = \frac{1}{i\omega\pi\epsilon_o R^4 \left[ (1 - ikR) \frac{2}{R^3} + \frac{k^2}{R^2} \right]} = \frac{1}{i\omega\pi\epsilon_o \left( 2R - \frac{i2R^2\pi}{\lambda} + \frac{R^3\pi}{\lambda^2} \right)} .
\]  

(14)

The quasi-static approximation i.e. \( R << \lambda_0 \) reduces (14) to

\[
Z_{\text{fringe}}|_{\text{Dynamic VI}} \simeq \frac{1}{i\omega 2\pi \epsilon_o R} \quad \text{and} \quad Z_{\text{sph}}|_{\text{Dynamic VI}} = \frac{1}{-i\omega\epsilon_o R} .
\]  

(15)

Comparing (15) with (5), it can be easily verified that under the restriction on the size \( (R << \lambda_0 \) and \( R << \lambda_0/\sqrt{\text{Re}(\epsilon)/\epsilon_o} \), impedances of our proposed Dynamic VI model converges to the Quasi-static VI model.

3.2. Necessary Conditions for Resonance \((\text{Re}\{\epsilon\} = -2\epsilon_o)\) using Circuit Theory

In this section, the well-know resonance condition is derived using our proposed Dynamic VI model. In circuit theory, the necessary condition for resonance is when the two reactances in the resonance circuit are equal in magnitude. In a parallel resonance circuit involving the inductance and capacitance, the necessary resonance condition (at \( \omega = \omega_0 \)) can be expressed as

\[
X_L = X_C , \quad \text{which in our case is given by} \quad \omega_o L_{\text{sph}} = \frac{1}{\omega_o C_{\text{fringe}}} .
\]  

(16)

Let us use values of \( L_{\text{sph}} \) (12) and \( C_{\text{fringe}} \) (13) in (16) and the relationship can be written as

\[
\frac{1}{\omega_o \pi R \text{Re}\{\epsilon\}} = \frac{1}{\omega_o (2\pi\epsilon_o R + \pi\epsilon_o k^2 R^3)} = \frac{1}{\omega_o 2\pi\epsilon_o R \left( 1 + \frac{2\pi^2 R^2}{\lambda^2} \right)} .
\]  

(17)

Under quasi-static condition \( (R << \lambda) \), (17) reduces to the necessary resonance condition as

\[
\frac{1}{\omega_o \pi R \text{Re}\{\epsilon\}} \simeq \frac{1}{\omega_o 2\pi\epsilon_o R} \quad \Rightarrow \text{Re}\{\epsilon\} = -2\epsilon_o .
\]  

(18)
3.3. Scattering Cross-section using Circuit Theory

The scattering cross-section is defined as the ratio between total energy scattered per second (also termed as scattered power $P_{sca}$) and the power density of the incident wave ($P_{in}$) and is given as

$$Q_{sca} = \frac{P_{sca}}{P_{in}}, \text{ where } P_{in} = \frac{R_{imp}^2 \eta_o}{\text{Area of sphere}}, \text{ and } P_{sca} = I_{imp}^2 \frac{Y_{fringe}}{Y_{fringe} + Y_{sph}} |Z_{fringe}|^2, \quad (19)$$

where $Y_{fringe} = 1/Z_{fringe}$ and $Y_{sph} = 1/Z_{sph}$ are defined by (11) and $\eta_o$ is the intrinsic impedance of the medium. Note that (19) will be used in next section to simulate Dynamic VI model and compare it with Quasi-static VI model, FDTD simulation and Mie solution.

4. Simulation Results

The accuracy of the proposed Dynamic VI model is evaluated by comparing it with the Mie solution, which exactly solves the problem of electromagnetic field scattering, absorption and extinction from a nanoparticle. We start our simulation by employing a plain wave incident on a sphere with the wavelengths varying from 450 nm to 1000 nm. It is important to note that in this range of the frequency, metals behave as plasma and possess complex permittivity with negative real part and a significant imaginary part [23]. Typical simulation uses Drude or Drude-Lorentz, which are closest to the experimentally measured data of Johnson and Christy [23]. In our simulation, we used a modified Drude-Lorentz model [24], which reproduces the Johnson and Christy data with analytical model and defines the relative permittivity as

$$\epsilon(\lambda) = \epsilon_\infty - \frac{1}{\lambda_p^2} + \sum_{i=1,2} A_i \left\{ \frac{e^{i\phi_i}}{1/\lambda_i - 1/\lambda - i/\gamma_i} + \frac{e^{-i\phi_i}}{1/\lambda_i + 1/\lambda + i/\gamma_i} \right\}. \quad (20)$$

Where $\epsilon_\infty$ is the high-frequency limit dielectric constant, $\lambda_p = 2\pi c/\lambda_p$ is the plasma wavelength, $\gamma_p = 2\pi c/\Gamma_p$ is the damping expressed as wavelength, $\lambda_i = 2\pi c/\omega_i$ is the interband transition wavelength, $\phi_i$ is the phase and $A_i = C_i/\omega_i$ is the dimensionless critical point amplitudes [24]. Note that the first two expression are the standard Drude model contributions with others accounting for the interband transitions.

In order to evaluate our model, we next compared accuracy of our Dynamic VI model with the Mie solution. Firstly, we consider the radius variations from 5 to 100 nm. Fig. 3a shows the change in the resonance wavelength against the peak resonance wavelength for different radii of the nanoparticle. It illustrates that our proposed Dynamic VI model closely matches with the Mie solution for radius $R < 50nm$. We here observe that the solution in predicting the resonance frequency given by our dynamic model departs from the Mie solution for radius $R > 50nm$, which is quite expected. The reason for this break-away point comes from the fact that Mie solution accounts for all modes. However, the proposed approach employs taking all time-dependent fields for the dipole mode only, which in the Mie solution corresponds to the value of $n = 1$ (dipole mode). Therefore, the higher order modes in Mie solution becomes significant for nanoparticles with $R > 50$ nm, thereby contributing to the red-shift in Mie solution results. Please note that in our simulation, Mie solution is truncated for a maximum value of $n$ based on Wiscombe’s criteria (in our simulation maximum value of $n$ ranges from 4 to 9) [12]. As shown in Fig. 3a, the increase in the accuracy compared to the quasi-static model is accounted by the additional new terms i.e. $C_{fringe,2}$ and $G_{fringe}$ derived and included in our proposed Dynamic VI model. Fig. 3a also highlight the importance of taking the extended approach by using the full dipole equations for our Dynamic VI model derivation. Note that the inclusion of the far-field and the retardation-field to the quasi-static term encompasses the full field of a point dipole approximation and sufficiently covers the fundamental mode of the oscillating metallic nanoparticle.

Next, the simulations were carried out using FDTD, Mie solution and the proposed Dynamic VI model for the scattering cross-section area of a nanoparticle, which is an important property...
while calculating the response of the system. For the simulations, we used (19) for the proposed Dynamic VI model and four different nanoparticle sizes varying from 5 nm to 75 nm are selected. Without loss of generality, we normalized all the results of scattering cross-section to their maximum value, which automatically scales it to a maximum value of unity as shown in Fig. 4. Note that the proposed Dynamic VI model response closely matches well with the exact Mie solution with an almost overlapping resonance peak till $R = 50$ nm. We also observe that the model predicts the Quality factor of the Dipole mode well (as Dynamic VI model employs taking all time-dependent fields of the dipole mode). However, for the larger radii nanoparticles ($R > 50$ nm), as the higher order modes start to influence the response, a red-shift is observed in Mie solution simulation for nanoparticle with $R = 75$ nm radius, compared to Dynamic VI model. The reason for red shift comes from the fact that Mie solution accounts for all modes. However, the proposed approach is for dipole mode and employs taking all time-dependent fields for the dipole mode only, which in the Mie solution corresponds to the value of $n = 1$.

The simulation results clearly show that a refinement in the theory clearly leads to important changes in the VI impedance model and extend the validity of the approach. Fig. 3b plots the capacitive and inductive reactances of proposed Dynamic VI model using (5) at resonance against the changes in the radius of nanoparticle. Note that at resonance the two are exactly equal in magnitude and opposite to each other. In fact, the capacitance and inductance signifies the amount of energy stored in each cycle of the resonance of a nanoparticle.

Fig. 3: (a) Resonance wavelength plotted against changes in the radius. The proposed Dynamic VI model shows good approximation of the analytical solution. (b) Capacitive and inductive reactances of the proposed Dynamic VI model plotted against the changes in the radius.

Fig. 4: Comparison of scattering cross-section areas calculated using Mie solution [25], Full wave FDTD and Dynamic VI model for 5, 25, 50 and 75 nm radius nanoparticles with all the results normalized to maximum value of respective cross-sectional areas. The data points are FDTD, while solid lines are Mie solution results, with Dynamic VI model plotted using dashed lines.
5. Conclusion
In this paper, we propose a Dynamic VI impedance model of nanoparticle, by including the effects of full dipole equations. The proposed approach gives fully retarded solution and includes the quantities such as internal and external (radiation) impedance derived using voltages and currents for the fundamental (dipole) mode. A comparison with the Mie solution (analytic solution) and FDTD, demonstrates that our proposed impedance model extend the range of the impedance model to the larger radii (5 nm < R < 50 nm) nanoparticles.

References