Theoretical Study of Plasmon Excitation of a Drude Metal Nanowire Coupled with Optically Dynamic Shell

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***Abstract*—In this paper, fundamental theoretical understanding of the details of the transient dynamics of the plasmon excitation in structure, consisting of metal nanowire coupled with the optically dynamic medium is presented. The excited plasmon dynamics in the metal nanowire by means of a rigorous mathematical approach that based on the analytical solution in the Laplace transform domain and its accurate inversion into time domain by virtue of residues evaluation at singular points that correspond to the excited plasmons of the structure are described. This problem is a 2D model of nanolaser and has applications in the most growing areas of nanophotonics and nanotechnologies which demand devices that can generate coherent plasmonic fields in subwavelength scale.**

Keywords— surface plasmons; time domain analysis; transient dynamics; nanoshell

I. Introduction

Nanostructures are the subject of strong interest due to the potential of a powerful light localization beyond the diffraction limit via the excitation of surface delocalized and localized plasmons [1-3]. They can be considered as a plasmon resonance biosensor based on multiple-beam interference [4] or as a novel modulator to control the intensity of the transmitted surface plasmon polaritons through a nanowire array [5] or as device designs that allow for the rapid sensing and detection of a wide range of important chemical and biological objects [6]. Possible future nanophotonic technologies demand devices that can generate stimulated emission through the excitation of the surface plasmons (spaser-based nanolaser) [7-9]. However it is challenging problem due to the extremely strong absorption losses in metal at optical frequencies. The suggestion to compensate the losses by an optical gain using dye molecules in presence of metal nanoparticles [10] or using nanoparticles with gold core and dye-doped silica shell [11] has been tested in experiments recently. Other configurations include metal nanostrips in a gain-material shell [12] and combinations of metal wires and quantum wires [13, 14]. For these applications an accurate frequency and time domain modeling that provides a valuable insight into fundamental processes is of great importance.

The main goal of this paper is to apply very efficient and accurate time-domain method for investigation of the transient processes in a metal nanowire coupled with a dynamic medium. This can reveal peculiarities of non-stationary processes and give possibility to formulate recommendations for applications in new ultrafast nanophotonics technologies for generation, control and switching of localized surface plasmons.

The problem is formulated in a differential-equation form with corresponding initial and boundary conditions. Abrupt time change of refractive index allows deriving a semi-analytical solution. Applying the Laplace transform directly to the wave equation, one can derive an analytical solution of the problem in the frequency domain. Then one can recover the time-domain electromagnetic field using the computation of the inverse Laplace transform via the residue evaluation at singular points associated with eigenvalues of the structure. This approach guarantees accurate inverse transformation with controllable accuracy and allows us to extract and interpret physical phenomena easily. This method has been already successfully applied to a variety of time-domain problems with various nanophotonic configurations [15-17].

II. Problem Formulation and Methodology

We investigate a circular plasma infinitely-long cylinder of radius a_1 with optically dynamic shell of the radius a_2 (see Fig. 1). This structure embedded in a dielectric environment with index of permittivity n_3.

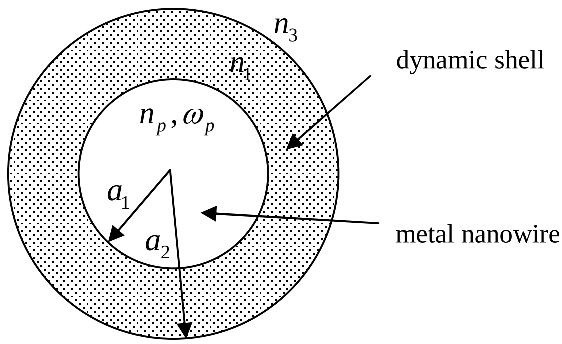

Fig. 1. Metal nanowire with optically dynamic shell.

Initial value of the permittivity of dynamic medium is n_1. Frequency dependent plasma permittivity ε_p is described by the Drude model

$$\varepsilon_p = 1 - \omega_p^2 \cdot (\omega(\omega - i\gamma))^{-1}, \qquad (1)$$

where ω_p corresponds to the frequency of plasma and γ represents the material absorption. The polar system of coordinates (ρ, φ) is introduced co-axially with the cylinder.

Such kind of nanowires can support plasmons only in the H-polarization case and the z-component of their magnetic field can be presented in the following form [18]:

$$h_0 = \begin{cases} A_k J_k(n_p k_0 \rho)\cos(k\varphi)e^{-i\omega_0 t^*}, \ \rho < a_1, \\ B_k J_k(n_1 k_0 \rho)\cos(k\varphi)e^{-i\omega_0 t^*} + \\ \qquad + C_k H_k^{(2)}(n_1 k_0 \rho)\cos(k\varphi)e^{-i\omega_0 t^*}, \ a_1 < \rho < a_2, \\ D_k H_k^{(2)}(k_0 \rho)\cos(k\varphi)e^{-i\omega_0 t^*}, \ \rho > a_2. \end{cases} \qquad (2)$$

here $k_0 = \omega_0 / c$ is the wavenumber, c is the velocity of light in vacuum, $n_p = \sqrt{\varepsilon_p}$, ε_p is defined in (1), the time dependence is $e^{i\omega_0 t}$, and ω_0 is the corresponding eigenfrequency of the plasmon. The associated time dependence for this initial field can be expressed as $e^{i\omega_0 t^*}$, where t^* is the moment of instantaneous excitation of the localized surface plasmon, and in our consideration $t^* < 0$.

The unknown coefficients A_k, B_k, C_k, D_k are determined from the boundary conditions requiring the continuity of the tangential components of the total electric and magnetic fields at each boundary of the our structure. To find all coefficients, we use the following set of equations:

$$\begin{aligned} &A_k J_k(n_p k_0 a_1) - B_k J_k(n_1 k_0 a_1) - C_k H_k^{(2)}(n_1 k_0 a_1) = 0, \\ &n_1 A_k J_k'(n_p k_0 a_1) - n_p B_k J_k'(n_1 k_0 a_1) - n_p C_k H_k'^{(2)}(n_1 k_0 a_1) = 0, \\ &B_k J_k(n_1 k_0 a_2) + C_k H_k^{(2)}(n_1 k_0 a_2) - D_k H_k^{(2)}(k_0 a_2) = 0, \\ &B_k J_k'(n_1 k_0 a_2) + C_k H_k'^{(2)}(n_1 k_0 a_2) - n_1 D_k H_k'^{(2)}(k_0 a_2) = 0. \end{aligned} \qquad (3)$$

Solution of (3) corresponds to the eigenfrequency value of the structure (ω_0).

At the initial (zero) moment of time, the refractive index (square root of dielectric permittivity) of the dynamic shell of the metal nanowire changes in value from n_1 to n_2 in response to some external mechanism (not included in our model) as illustrated schematically in Fig. 2. Now we consider the effects that couple the initial plasmon of the structure with new refractive index with particular emphasis on the transient processes occurring in such a single dynamic nanoshell.

In general, there are two classes of approaches for solving such transient electromagnetic problems. One method dwells on solving Maxwell's equations directly in time domain by using a time-step scheme and is typified by the finite difference time domain method. Such approaches are computationally intensive and do not allow to see the key physical phenomena. In contrast, the investigation to be pursued here is to solve Maxwell's equations analytically in the frequency domain and

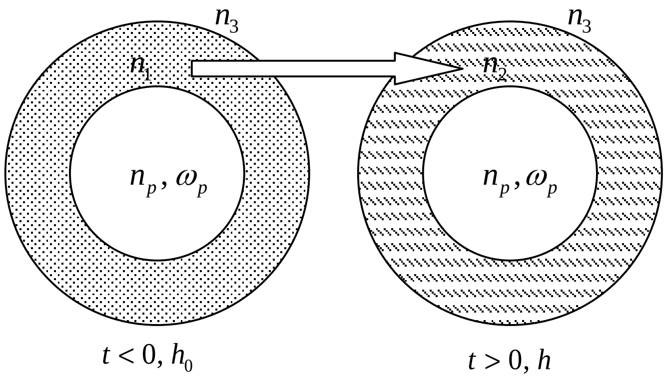

Fig. 2. Schematic illustration of the studied problem.

then to recover the time-domain electromagnetic field via the inverse frequency transform.

In this paper we have to get a fundamental understanding and to describe theoretically details of the transient dynamics of the plasmon excitation in structure, consisting of metal nanowire coupled with an optically active medium. For this we investigate transient response of the localized surface plasmon to a change of the refractive index of the dynamic shell. This formulation of the problem assumes deriving the analytical solution that can show physical phenomenon in detail.

Non-stationary expression for the field in a plasma medium with dielectric permittivity described by the Drude model (1) has the following form:

$$\begin{aligned} &rot rot \vec{h} + \frac{1}{c^2}\varepsilon_\infty \frac{\partial^2}{\partial t^2}\vec{h} + \frac{1}{c^2}\frac{\omega_p^2}{\gamma}\frac{\partial}{\partial t}\vec{h} \\ &\quad - \frac{1}{c^2}\frac{\omega_p^2}{\gamma}\frac{\partial^2}{\partial t^2}\int_0^t e^{-\gamma(t-t')}\vec{h}(t')dt' = 0. \end{aligned} \qquad (4)$$

Using the fact that the magnetic field of the localized plasmon has only one nonzero element ($z-$ component), we can obtain expression for the transformed field as

$$\begin{aligned} &\Delta h + \frac{1}{c^2}\varepsilon_\infty \frac{\partial^2}{\partial t^2}h + \frac{1}{c^2}\frac{\omega_p^2}{\gamma}\frac{\partial}{\partial t}h \\ &\quad - \frac{1}{c^2}\frac{\omega_p^2}{\gamma}\frac{\partial^2}{\partial t^2}\int_0^t e^{-\gamma(t-t')}h(t')dt' = 0, \ \rho < a_1, \end{aligned} \qquad (5)$$

$$\Delta h - \frac{n_2^2}{c^2}\frac{\partial^2 h}{\partial t} = 0, \ a_1 < \rho < a_2, \qquad (6)$$

$$\Delta h - \frac{1}{c^2}\frac{\partial^2 h}{\partial t} = 0, \ \rho > a_2, \qquad (7)$$

where h corresponds the z-component of the magnetic field,

$$\Delta = \frac{\partial^2}{\partial \rho^2} + \frac{1}{\rho}\frac{\partial}{\partial \rho} + \frac{1}{\rho^2}\frac{\partial^2}{\partial \varphi^2}. \qquad (8)$$

Applying the Laplace Transform, $H(p) = \int_0^\infty h(t)e^{-pt}dt$ to equations (5)-(7), we can write the image functions as

$$\Delta H+(\varepsilon_\infty+\frac{\omega_p^2}{p(p+\gamma)})H-(\frac{1}{c^2}(\varepsilon_\infty(p+i\omega_0)+\frac{\omega_p^2}{\gamma}))A_kJ_k(n_pk_0\rho)\cos k\varphi,\quad \rho<a_1, \qquad (9)$$

$$\Delta H+\frac{n_2^2}{c^2}p^2H+\frac{n_2^2}{c^2}(p+i\omega_0\frac{n_1^2}{n_2^2})(B_kJ_k(n_1k_0\rho)+C_kH_k^{(2)}(n_1k_0\rho))\cos k\varphi,\quad a_1<\rho<a_2, \qquad (10)$$

$$\Delta H+\frac{p_2^2}{c^2}H+\frac{1}{c^2}(p+i\omega_0)D_kH_k^{(2)}(k_0\rho)\cos k\varphi,\ \rho>a_2. \qquad (11)$$

The solution of (9)-(11) is a superposition of the initial value problem and contributions of the boundary terms [15, 16]

$$H=\frac{1}{p-i\omega_0}A_kJ_k(n_pk_0\rho)\cos k\varphi e^{-i\omega t^*}+A_k^1I_k(\tilde{n}_pq\rho)\cos k\varphi e^{-i\omega t^*},\quad \rho<a_1, \qquad (12)$$

$$H=\frac{n_2^2p+in_1^2\omega_0}{n_2^2p^2+n_1^2\omega_0^2}(B_kJ_k(n_1k_0\rho)+C_kH_k^{(2)}(n_1k_0\rho))\cos k\varphi e^{-i\omega t^*}+$$
$$+(B_k^1I_k(n_2q\rho)+C_k^1K_k(n_2q\rho))\cos k\varphi e^{-i\omega t^*},\quad a_1<\rho<a_2, \qquad (13)$$

$$H=\frac{1}{p-i\omega_0}D_kH_k^{(2)}(k_0\rho)\cos k\varphi e^{-i\omega t^*}+D_k^1K_k(q\rho)\cos k\varphi e^{-i\omega t^*},\quad \rho>a_2, \qquad (14)$$

Here $q=p/c$, $I_k(..),K_k(..)$ are the modified Bessel functions, and

$$\tilde{n}_p=\sqrt{\varepsilon_\infty+\frac{1}{c^2}\frac{\omega_p^2}{q(q+\gamma/c)}}\,. \qquad (15)$$

New coefficients A_k^1,B_k^1,C_k^1,D_k^1 are found from the boundary conditions. The set of equations for determining the unknown coefficients has the following form:

$$A_k^1I_k(\tilde{n}_pq\rho)-B_k^1I_k(n_2q\rho)-C_k^1K_k(n_2q\rho)=$$
$$=-\frac{ip\omega_0(n_1^2-n_2^2)}{(p^2n_2^2+\omega_0^2n_1^2)(p-i\omega_0)}A_kJ_k(n_pk_0\rho), \qquad (16)$$

$$B_k^1I_k(n_2q\rho)+C_k^1K_k(n_2q\rho)-D_k^1K_k(q\rho)=$$
$$=-\frac{ip\omega_0(n_1^2-n_2^2)}{(p^2n_2^2+\omega_0^2n_1^2)(p-i\omega_0)}D_kH_k^{(2)}(k_0\rho), \qquad (17)$$

$$\frac{1}{\tilde{n}_p}A_k^1I_k'(\tilde{n}_pq\rho)-\frac{1}{n_2}(B_k^1I_k'(n_2q\rho)+C_k^1K_k'(n_2q\rho))=$$
$$=\frac{ip^2(n_1^2-n_2^2)}{n_p(p^2n_2^2+\omega_0^2n_1^2)(p-i\omega_0)}A_kJ_k'(n_pk_0\rho), \qquad (18)$$

$$\frac{1}{n_2}(B_k^1I_k'(n_2q\rho)+C_k^1K_k'(n_2q\rho))-D_k^1K_k'(q\rho)=$$
$$=\frac{ip^2(n_1^2-n_2^2)}{n_p(p^2n_2^2+\omega_0^2n_1^2)(p-i\omega_0)}D_kH_k'^{(2)}(k_0\rho). \qquad (19)$$

Further we perform the inverse transform to the time domain via the Mellin formula, $h(t)=1/(2\pi i)\int_{-i\infty}^{+i\infty}H(p)e^{pt}dp$. Expressions (16)–(19) have singular points at $p=i\omega_0$, $p=\pm i\omega_0\,n_1/n_2$, and also singular points corresponding to the localized surface plasmons given by the zeros of determinants of (16)–(19) (for $k=0,1,2,...$),

$$\Delta=\begin{vmatrix} I_k(\tilde{n}_pq\rho) & -I_k(n_2q\rho) & -K_k(n_2q\rho) & 0\\ 0 & I_k(n_2q\rho) & K_k(n_2q\rho) & K_k(q\rho)\\ 1/\tilde{n}_p\cdot I_k'(\tilde{n}_pq\rho) & -1/n_2\cdot I_k'(n_2q\rho) & -1/n_2\cdot K_k'(n_2q\rho) & 0\\ 0 & 1/n_2\cdot I_k'(n_2q\rho) & 1/n_2\cdot K_k'(n_2q\rho) & K_k'(q\rho)\end{vmatrix} \qquad (20)$$

After the time change of the refractive index of dynamic shell, the field inside the wire is represented by only the first term of (12) and outside the nanoshell by the first term of (14) in similar manner (for details see [17]). Inside the dynamic shell the field is represented by the expression in the first bracket of (13).

III. Numerical results

We consider a silver nanowire with radius $a_1=25$ nm and dynamic shell with radius $a_2=35$ nm. In this investigation we use the following parameters of the Drude model: $\omega_p=1.4525\cdot10^{16}$ Hz, $\gamma=7.0656\cdot10^{13}$ Hz, $\varepsilon_\infty=5.2573$ [19]. We also consider the normalized time, $T=tca^{-1}$. The normalized frequency of the initial surface plasmon mode is $w_0=\omega_0ac^{-1}=0.4464+i*0.0046$ ($n_1=1.45$, $k=1$) that is found from the set of equations (3).

The time domain field is recovered using the Cauchy residue theorem. This algorithm has been already successfully used for different of time domain problems [15-17].

Nonzero contribution comes only from the solutions of the determinants (20), which correspond to the eigenfrequencies of the nanowire. All these frequencies are complex, their imaginary parts determine the velocities of decay of the oscillations. For each azimuthal index $k=const$, expression (20) has one solution, which is associates with the localized surface plasmon, and infinite number of solutions corresponding to the bulk plasmons. Using these analytical solutions, it is possible to evaluate the contribution of each excited resonant frequency to the total field.

To this end, we write the values of residues separately in a few singular points assuming that $n_2=1.452$. Singular points are the eigenfrequencies of the corresponding surface plasmon $\tilde{\omega}_0ac^{-1}=\pm0.4462+i*0.0045$, and the residues at each of these frequencies are equal $-0.994+i*0.2861$ for (+) and $0.0025+i*0.0032$ for $(-)$. The values correspond to the normalized amplitude of the initial field. The frequencies $\omega_1ac^{-1}=\pm2.3308+i*0.5643$ corresponds to the bulk plasmons with field variation along the radius, and residues are $-0.0036-i*0.0034$ for (+) and $-0.0026+i*0.002$ for $(-)$. Calculating the residues at each bulk plasmon frequency,

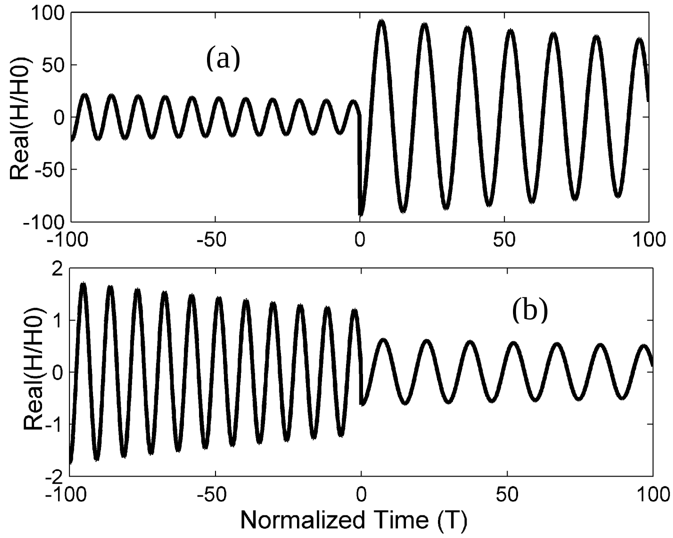

Fig. 3. The transient response inside the silver nanowire with dynamic medium: (a) $n_1 = 2.8$, $n_2 = 1.5$; (b) $n_1 = 1.5$, $n_2 = 2.5$.

we see that their contribution to the total field is negligibly small. Only the surface plasmon has the amplitude comparable to that of the initial plasmon. Regarding to this, the overall effect of the abrupt change in refractive index transforms the initial plasmon to a transformed one with similar field pattern and a slight shift of the eigenfrequency. The poles contribute to the steady state solution while the branch point fields makes single contribution to the transient solution.

Figure 3 illustrates the transient response of the magnetic field inside the silver nanowire with the dynamic shell in the steady-state regime. Before the zero moment of time the initial field is observable. Change in the refractive index of the shell at the zero moment of time disturbs the field. To illuminate clearer the phenomena associated with the change in frequency and amplitudes we consider relatively large changes in refractive index. Although they are impractical they do reveal the key phenomena very clearly. Changing of the amplitude follows from initial conditions of displacement continuity at zero moment of time. Therefore it is concluded that the time variation of the refractive index of the dynamic shell of nanowire does not change the near field pattern.

IV. CONCLUSIONS

Transformation of the surface plasmon field of a silver nanowire with an dynamic shell due to time variations of shell's dielectric permittivity has been studied by means of an analytical formulation that clearly reveals the key mechanisms of the problem. Accurate time-domain inversion has made it possible to analyze transient and steady state regimes. The maximum amplitude of the new structure is observed for the plasmon with the same number of angular field variations as the initial plasmon. Therefore, it is concluded that changes of the index of permittivity of the dynamic shell lead to a transient regime in which a change of frequency is observed but the field pattern is preserved.

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