Optical Near-Field Study of Ag Nanowires by the Differential Method

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ABSTRACT

The optical response of subwavelength silver nanowires arranged periodically on silica has been analyzed numerically by the differential method improved by the S matrix algorithm. Our results improve the capacity of this rigorous method to give a description of various phenomena occurring in near and far-field around the periodic grating. This renders possible to determine the positions of plasmon’s resonance according to the choice of materials used and the geometrical properties. We study the behavior of the diffracted light by the nano-structure in both single nanowire case and grating nanowires case. The influence of the exact grating period and the induced modification of the spacer nanowire dependence are then discussed. Moreover, we present mappings of the electromagnetic field located at 50 nm above the nanowires.

Keywords: Near-Field; Differential Method; Nanowires; Plasmons; Silver

1. Introduction

In recent years, surface plasmons (SPs) have gained attention because of their potential applications in nanophotonic and plasmonic devices [1-7]. Surface plasmons are electron charge density waves confined to and traveling along the surface between a metal and a dielectric. These electronic excitations are a consequence of the collective oscillation of the conduction band electrons in the metal [8]. They manifest themselves in strong field enhancement near the metal surface. In the same way, various photonic crystal structures with nanostructured metals have been realized so far. In addition to periodically modulated metal surfaces, including surface corrugation [9,10], regular arrangements of individual metal nanoparticles on dielectric substrates [11] are prominent examples of such polaritonic crystal structures. The particular optical properties of metallic nanostructures can be attributed to the excitation surface plasmons [12].

The properties of the local enhanced field for a single isolated particle depend on the particle parameters, the properties of the incident irradiation and the surrounding medium [13-15].

As for nanoparticles array they can be additionally modified by the interparticles coupling effect [16-18].

Surface enhanced effects, like surface enhanced Raman scattering or surface enhanced fluorescence [13,19-21] take advantage of both, the near-field and the far-field of resonantly excited metal nanoparticles: on one hand, the enhanced near-fields lead to a higher excitation efficiency and on the other hand, the nanoparticles act as a transmitting antenna and enhance the coupling of atomic or molecular resonances to the optical far-field. For the first one, the induced near electromagnetic field has properties of an evanescent wave, its amplitude decreases rapidly with the distance from the metal surface. Thus, the size of the field enhanced area is governed only by the size of the optically illuminated metal structure and the diffraction limit can be overcome.

The waveguiding of surface plasmon polaritons (SPPs) along one-dimensional structures has been attracting intensive attention. A number of ideas, such as planar metal waveguides [22], metallic nanohole grating [23], dielectric-loaded SPP waveguides [24] and metal nanowires [25] have been suggested for this aim. For all these structures, silver nanowires that can be routinely fabricated with smooth
surfaces and uniform diameters are one of the most considered systems [25,26].

We have compared experimental results obtained by Schider et al. [27], with our simulations performed by the Differential Method [28-34] improved by S-algorithm [35,36], Li remarks [37] and the Redheffers star product [38] when the number of matrix multiplications can be further reduced remarkably. The versatility of the differential method is demonstrated by considering complex configurations. In this respect, this method can be an easy-to-implement alternative to more conventional method such as FDTD or finite elements. The nanowire gratings strongly depend on the polarization direction of the incident light. The excitation of dipolar plasmon mode is predominant when the polarization direction is perpendicular to the wire axis.

The paper is organized as follows. In Section 2, the numerical method we use to model the optical and plasmonic properties of the gratings is introduced. The results of our calculations of the extinction properties of the nanostructures are presented and discussed in Section 3 distinguishing the noninteracting particles case from the grating configuration both in far and near-field. Finally, the conclusion is drawn in Section 4.

2. Numerical Method

To our knowledge, scalar electromagnetic theory, which has been widely used to compute the characteristics of diffractive gratings, is valid only for structures that are large compared to the incident wavelength, and the diffracted field can not be computed near the surface vicinity. For near-field, on sub-wavelength features, it is necessary to solve the full vector form of Maxwell’s equations under suitable boundary conditions [39,40]. There are some reported numerical calculation methods based on rigorous electromagnetic theory, such as time-domain finite difference method [41-44], boundary element, finite element method, Fourier modal method, etc.

Among these methods, the differential method [28-31] is one of the macroscopic methods allowing the calculation of the intensity of light diffracted by an object. It is also based on Maxwell equations and the different boundary conditions at interfaces. The principle of this technique consists of establishing the propagation equations of the electromagnetic field in different regions of space and calculating its components amplitudes.

This method has been performed with an S-algorithm; taking into account Li remarks [35-37] which is aimed for achieving unconditional numerical stability with S-algorithms is to avoid the exponentially growing functions in every step of the matrix recursion, and assuming a periodic arrangement of metal wires. This algorithm is used to reduce the numerical instabilities linked with the existence of increasing exponential functions during the numerical integration process, that is exponential functions linked with evanescent orders, the electric and magnetic fields above the surface are presented as an expansion over Rayleigh waves. To find their complex amplitudes, the propagation equations are to be solved with boundary conditions at the two surfaces of the metal film. When the amplitude are calculated, the transmitted intensity as well as the electric and magnetic field distributions can be obtained by summation over all the modes. A finite number of modes in the calculations was chosen according to the stability of the numerical procedure. The numerical result was checked by calculation of the energy conservation and the convergence of the extinction while increasing the number of modes.

We have studied a one dimensional structure (Figure 1) consisting of silver film of thickness $h$ with a periodical set of rectangular wires ($D$ is a grating period, $w$ is the wire width and $h$ is the wire height). The structure is deposited on a substrate of glass ($n = 1.458$).

In all calculations below, the structure is illuminated in normal incidence with $P$-polarized light. In order to obtain good results, we injected into the simulation program a great number of modes (i.e. numbers of vectors) to correct the numerical errors.

The input parameters of the method are the geometrical sizes of the structure and the dielectric susceptibilities of the constituent materials. The latter are treated as spatially local and frequency dependent, which is especially important in the case of metals. The dielectric function of silver was taken from Ref. [45].

3. Results and Discussion

3.1. Noninteracting Nanowires

3.1.1. Far-Field Case

Silver is a paradigmatic case for studying surface plasmons and there are a large amount of literatures on surface polariton plasmons (SPPs) by gratings [12,46-49] in the visible region. The reason for that is because at these wavelengths the imaginary part of the permittivity ($\varepsilon_r$) is small, then the SPPs is well defined [12,50].

Figure 1. Schematic of a nanostructured film with the notations used for the numerical work.
We measure the optical extinction which is defined as $\text{Ext.} = \log(T_0/T)$, where $T_0$ and $T$ denote the optical transmission through the bare substrate and the nanowire sample deposited on the substrate, respectively.

For instance, Schider et al. found, by fabricating one-dimensional silver nanowires ($D = 2250$ nm) with a cross section of 83 nm in width ($w$) and 25 nm in height ($h$) on a quartz substrate, that the optical response of regularly patterned nanowires strongly depends on the polarization of the incident light [27]. The extinction spectra produced by differential method (Figure 2(b)), are in excellent agreement with experimentally obtained results [27] shown in (Figure 2(a)); for our simulated results, the extinction maximum is located near 490 nm and the full width at half maximum (FWHM) is approximately 180 nm. In the same case, experimental results show that the SPs resonance wavelength is near 495 nm and the FWHM is near 140 nm.

The extinction slightly grows with increased wavelengths due to the absorption of silver. Note that the position of an extinction peak indicates the resonance wavelength while the spectral width exhibits the damping of plasmon resonance induced by nanowires.

Starting from these two figures (Figures 2(a)-(b)), we notice a light shift between the experimental and numerical wavelength of resonance, this can be explained by the conditions of manufacture of the samples (defects, asperities) and experimental measurement.

However, to make sure that we always work within the framework of the individual nanowires, we calculated the spectrum of optical extinction for different periods $D = 2250$ nm, $3375$ nm and $9000$ nm respectively; where the corresponding curves are represented in Figure 3. We notice that the plasmon resonance peak remains almost unchanged; this confirms the absence of mutual effects between the nanowires which validates the judicious choice of the period used in our calculations as confirmed by Schider [27].

### 3.1.2. Near-Field Case

To understand the main features in the optical near-field, we present the distributions of the electromagnetic field located at 50 nm above the individual nanowire. $D = 2250$ nm, $w = 83$ nm and $n = 25$ nm, at the resonance plasmons wavelength $\lambda = 490$ nm.

In our calculations, we must specify that the various components of the electromagnetic field do not converge at the same speed. In $P$ polarization for example, the component of the magnetic field $H_z$ is obtained most quickly. Indeed this component is continuous everywhere since it
is necessarily parallel to each interface. As for the components of the electric field $E_x$ and $E_y$, they have strong amplitudes at the metal interfaces. This leads to instabilities that are more significant, and in order to obtain the electromagnetic field map, we need to use a significant number of modes.

The field map presented in the Figure 4 shows the intensity of the electric field $|E|^2$. The real part of the complex electric field and the values obtained are standardized compared to the incidentals values. In Figure 4, we represent only the value of the intensity amplified 5 times to clearly highlight the amplification above the nanowires.

The distribution of the electromagnetic intensity at the wavelength 490 nm reveals an extremum of the electric field above the nanowires. This strong localized field enhancement leads us to say that the SPs are excited on the level of the metal/air interface, this intensity is significant and develops in a considerable way as it can reach ~30 times the intensity injected.

One notices also the presence of an amplification on both sides of both edges. This clearly shows the damping according to axis $x$ on the quartz substrate.

To explain this phenomenon, we compared our simulated results based on the differential method below with the analytical SPs coupling theory. For the latter, we used the following equations for an air-metal system deposited on a quartz substrate exposed to an incident $P$-polarized light. The dispersion relation of the excited SPs under resonance conditions is given by [12]:

$$
|E|^2 = k_0 \frac{E_x E_w}{\sqrt{E_x^2 + E_y^2}}
$$

where $k_{sp}$ and $k_0$ denote the wave vectors of the SPs and the incident light respectively, and $E_w$ and $E_x$ are the dielectric constants of metal and adjacent medium on the top of wire. Propagating SPs are transformed into a radiative mode when the following condition is satisfied [12]:

$$
\alpha_n = k_0 \sin \theta \pm nK
$$

where $k^{(n)}_s$ is the wave vector of the n-th diffracted light, $K = \frac{2\pi}{D}$ is the grating vector and $n$ is an integer. We have found that the first diffracted waves are plane waves: $k^{(n)}_s < k_0$ in our case ($n = 0, 1, 2, 3, 4$), the following ones are evanescent waves: $k^{(n)}_s > k_0$ (case $n > 4$). For the diffracted order $n = 5$, i.e. diffracted evanescent waves and $k^{(n)}_s - k_{sp}$, the incident wave is resonantly coupled the SPs (the electromagnetic field is considerably amplified).

With our method we know that the electric and magnetic fields above the grating are expanded as a sum over Rayleigh waves [28]. In particular, for $P$ polarization the magnetic field above the grating can be written as follows [51-53]:

$$
H_z(x,y) = \sum_{n=0}^{\infty} A_n \exp[j(\alpha_n x - x_n y)]
$$

$$
\alpha_n = k_0 n_1 \sin \theta + \frac{2\pi}{D} \quad (4)
$$

$$
x_n = \sqrt{\left(k_0^2 n_2^2 - \alpha_n^2\right)} \quad \text{if} \quad k_0 n_2 \geq \alpha_n \quad (5)
$$

$$
x_n = \sqrt{\left(\alpha_n^2 - k_0^2 n_2^2\right)} \quad \text{if} \quad k_0 n_2 \geq \alpha_n \quad (6)
$$

Using Equation (5), we evaluated the order which contributes to coupling the diffracted light with the SPs, and we found this for $n > 4$. This result confirms that our calculations with differential method are in good agreement compared to the analytic theory.

### 3.2. Grating Nanowires

#### 3.2.1. Far-Field Case

The second part of this work is devoted to the study of closely positioned nanowires. Figure 5(a) and Figure 5(b) show the extinction spectra as a function of widths of the silver nanowires from $w = 76$ nm to $w = 150$ nm with fixed grating period $D = 350$ nm. The thickness of the silver nanowires is chosen to be $h = 25$ nm from experimental and simulated results respectively.

We clearly see:

1) The increase in the extinction value when the distance between nanowires decreases.
Figure 5. Extinction spectra of silver nanowire gratings from (a) the experimental curves from reference [27] and (b) from our simulated curves. The nanowire width is varied from \( w = 76 \text{ nm} \) to \( 150 \text{ nm} \), at fixed \( h = 25 \text{ nm} \) and \( D = 350 \text{ nm} \).

This strong effect of coupling demonstrates that the collective interaction of the nanowires with the light field leads to interesting diffraction phenomena which strongly modify the optical properties of the nanowires with respect to the individual nanowire case.

2) When the period is fixed and the width of the nanowires is increased, we noticed that there is a red-shift of the resonance peaks due to the dipolar plasmons effect.

We can also see on Figure 5(b) a minimum peak on \( \lambda = 510 \text{ nm} \) which corresponds to Wood anomalies defined by the following relation \( \lambda = n_{\text{substrate}} \times D \).

Furthermore, we have presented in the Figure 6, a comparison between the experimental and simulated curves of plasmons resonances peaks for different widths. We observe a good agreement between the two results. This confirms the capacity of the presented method to really represent what physically occurs with a very high degree of accuracy.

3.2.2. Near-Field Case

In this last section, the intensities have been normalized with the incoming plane wave intensity. We plotted the electric field \( |E_z|^2 \) intensity according to \( x \) axis shown in Figure 7 (for \( D = 350 \text{ nm} \), \( w = 83 \text{ nm} \) and \( h = 25 \text{ nm} \)) at a resonance plasmons wavelength of \( \lambda = 514 \text{ nm} \).

In addition, in Figure 7, we represented only the intensity value amplified 5 times for clear representation of the amplification above the nanowires.

Figure 6. Resonance peak positions as a function of the nanowires width.

Figure 7. The electric field \( E_z \) distribution in the near-field above the metallic nanowires grating at the wavelength \( \lambda = 514 \text{ nm} \).
The calculated results showed that for SPs excitations, the enhancement in the near-field distribution was concentrated around the metal/dielectric interface which can reach until ~198 times the injected intensity. Interference of the SPs field can be visible between the adjacent nanowires.

We thus proceed as for the case of individual nanowires near-field (Section 3.1.2.). We have determined the order responsible of the generation of the plasmons on the nanowires grating using the analytical theory and the differential method. We found in both cases the order \( n = 1 \). This is valid when \( D < \lambda \) because the scattering is mostly prohibited and only possible in the direction of the zeroth grating order.

As expected, the electric near-field intensity maps around the grating nanowires show different field distributions compared to the case of an individual nanowires, especially on the right and the left edges of nanowires. The widths values at the amplifications for each edge are estimated at ~30 nm and ~18 nm for grating and individual nanowires respectively. The first value is more important than the other because of the mutual effect due to the close nanowires.

4. Conclusions

In conclusion, we have presented a numerical study of the performance of periodic metallic nanowires for sensing purposes. Our results show that their extinction spectra are highly sensitive to variations of the grating period.

As already mentioned, the optical response of a periodic arrangement of metallic nanowires can differ substantially from that of noninteracting individual metallic particles. It turns out that the period of the grating structure is the crucial parameter for the modification of the isolated nanowire plasmon response. This can be interpreted as a manifestation of the nanowire-nanowire interaction in both far-field and near-field regimes.

We have also presented the enhancement of electric field above the nanowires which is more important in the grating nanowires configuration (~198 times) than the individual one (~30 times).

We demonstrated analytically and numerically that the surface plasmons in both individual and grating nanowires cases are generating by the 5-th and the first diffractive order respectively.

Finally, this study shows the potential of using transmitted SPs waves in a variety of optical applications, such as optical imaging system, optical biosensor, polarizer, filters, and modulators and other light sources.

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