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Numerical simulation of light propagation in silver nanowire films using time-harmonic inverse iterative method

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The interaction between light and silver nanowires (Ag NWs) in a thin film is simulated by solving Maxwell's equations numerically. Time-harmonic inverse iterative method is implemented to overcome the problem of negative permittivity of silver, which makes the classical finite-difference time-domain iteration unstable. The method is validated by showing the correspondence between the plasmonic resonance of an Ag NW from a two dimensional simulation and the analytical solution. In agreement with previous experimental studies, the simulation results show that the transmissivity of the Ag NW films is higher than expected from the geometric aperture. The cause of this phenomenon is studied using TE/TM modes analysis for Ag NW films with different surface coverage of parallel-aligned Ag NWs. Furthermore, 3D simulation of Ag NW films with randomly arranged Ag NWs is performed by parallel computation on high performance computers. A binder layer is taken into account for a preliminary comparison between the simulation and experimental results. The agreements and disagreements between the simulated and measured spectra are discussed. © 2013 AIP Publishing LLC [<http://dx.doi.org/10.1063/1.4801919>]

I. INTRODUCTION

Nanostructured metal electrodes were introduced in recent years¹ as a good option for transparent electrodes in optoelectronic devices. They have the potential to replace indium tin oxide (ITO) in applications involving flexible substrate, such as organic solar cells (OPV) and OLEDs, by overcoming the drawbacks of ITO, including a brittle nature, difficulty in using on flexible substrates and a costly step of vacuum sputtering deposition.² Solution-processed silver nanowire (Ag NW)^{3–5} films are a low-cost realization of this kind of electrode. Ag NW films can be fabricated with a low processing temperature and their coating properties are highly reliable.⁶ The processing procedure ends with randomly distributed Ag NWs that form a mesh in the film. These Ag NW films are called percolation type electrodes, because their conductivity is gained from the interconnected Ag NW network and their transparency is mainly due to the space between nanowires.

Optical properties, specifically how light is transmitted, absorbed and reflected, are essential for Ag NW films when they are used as transparent electrodes. Experimental results^{1,6} have shown that the transmittance of such films is high in the visible spectrum, exceeding the geometric aperture. However, how the Ag NWs interact with light in Ag NW films is still an open question. Numerical simulation of this interaction is an efficient way to explore the optical behavior of such films and to find out what kind of role

plasmonic effects play in the process. Combined with the analysis of the electrical characteristics, it can also be beneficial to adjust the conductivity and transparency of the film.

Methods used to simulate light propagation in systems involving metallic nanostructures can be divided into two categories. The first category consists of methods which solve Maxwell's equations directly under certain discretization of the structure. Many methods in this category, such as the FDTD method incorporating a dispersive model,^{4,7} the discontinuous Galerkin time-domain method (DGTD),⁸ and the finite element method (FEM),^{1,9} have been implemented to simulate the optical properties of Ag NWs in 2D or simple 3D structures. However, no result regarding complex 3D structures, like a randomly distributed mesh of Ag NWs, has been presented so far. Simulation of Ag NW films with randomly distributed Ag NWs is a challenging problem because of its multi-scale nature. Here, "multi-scale" means that the size of the film can be in micrometer range while the Ag NWs inside are in nanometer range (~ 30 nm) in actual fabricated Ag NW films.⁶ Therefore, discretizing the film finely enough in the Ag NWs' scale will lead to a huge number of grid points. This necessitates the numerical solver to be less computationally intensive and parallelizable. The classical FDTD method¹¹ does not model dispersive materials (e.g., silver), and the standard time iteration scheme becomes unstable when the material's permittivity is negative. One common solution to this problem is to incorporate a dispersive model into the FDTD discretization.^{12–14} However, it introduces additional unknowns to the field update equations which consequently increase the demand for computational

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resources. On the other hand, fitting the experimental data of the material parameters to the dispersive models causes some loss of accuracy. We implemented a time-harmonic inverse iteration method (THIIM)¹⁰ to solve the time-harmonic Maxwell's equations. THIIM also solves the problem of classical FDTD method and has two advantages compared with FDTD incorporating a dispersive model. First, it performs a “back iteration” in metallic regions to deal with the negative permittivity of silver, which makes the method computationally as intensive as the classical FDTD method. Second, the experimental material parameters can be used directly in simulations. Furthermore, the algorithm can be easily implemented for parallel computation, which makes it more suitable for the kind of problem we deal with here.

The second category of methods dealing with light propagation through metallic nanostructures is characterized by a newly formulated scattering matrix method.¹⁵ The structure is divided into slices such that in each slice the material is homogenous in the direction of division. The electric and magnetic fields are expanded in each slice according to a selected basis. The scattering matrix depending on this expansion is then calculated iteratively slice by slice. This method is efficient for regular structures which can easily be divided into several slices. However, for complicated structures like Ag NW films, one may need to divide the Ag NW layer finely enough to get an accurate solution. Moreover, the randomly aligned Ag NWs can result in a complicated field distribution in the slices, which means a large number of basis functions are needed to resolve the field. This will lead to more computational cost, since at each step of iteration, the inverse of a matrix at the order of the number of the basis functions needs to be calculated.

This paper is structured as follows: In Sec. II, we present our simulation method briefly; in Sec. III, we will show and discuss the simulation results for the optical properties of Ag NW films, including results for 2D test models and more realistic 3D models; finally in Sec. IV, we will give a conclusion of this work and an outlook for future work.

II. MODEL AND APPROACH

The Ag NW films are modeled as a three-layer structure which is shown in Fig. 1. The Ag NWs are placed on a glass substrate and surrounded by air, so that the three layers are a glass layer, an Ag NW layer and a thin air layer. A plane-wave light source illuminates the film from the top with the wave vector perpendicular to its top surface. The time-harmonic Maxwell's equations are given as

$$\begin{cases} i\omega\hat{E} = \frac{1}{\epsilon}\nabla \times \hat{H} - \frac{\sigma}{\epsilon}\hat{E} \\ i\omega\hat{H} = -\frac{1}{\mu}\nabla \times \hat{E}, \end{cases} \quad (1)$$

where ω is the angular frequency of the plane wave, and $\hat{E}$ and $\hat{H}$ are the time-independent components of the electric and magnetic fields, respectively. They are related to the time-dependent electric and magnetic fields in the following way:

FIG. 1. A sketch showing the cross section of the computational domain in x - z plane. The dark regions in the middle layer denote the cross sections of Ag NWs. They are in different shapes, because the Ag NWs are aligned in different directions. Also, the Ag NWs can intersect with each other.

$$\vec{E} = \hat{E}e^{i\omega t}, \quad \vec{H} = \hat{H}e^{i\omega t}.$$

The boundary conditions are as follows: For top and bottom surfaces, we use absorbing boundary conditions with perfectly matched layers (PML),¹⁶ and for the sides, we apply periodic boundary conditions.

The approach we adopt to solve time-harmonic Maxwell's equations given in Eq. (1) is THIIM,^{10,17} which introduces a “back iteration” in regions with negative permittivity $\epsilon < 0$. The computational domain is discretized into Yee lattice.¹⁸ For grid points located in regions with positive permittivity $\epsilon > 0$, the method gives an iterative scheme which is derived from the classical FDTD discretization

$$\frac{e^{i\omega\tau}\hat{E}_h^{n+1} - \hat{E}_h^n}{\tau} = \frac{1}{\epsilon}\nabla_h \times \hat{H}_h^{n+1/2}e^{i\omega\tau/2} - \frac{\sigma}{\epsilon}e^{i\omega\tau}\hat{E}_h^{n+1} + S_E, \quad (2)$$

$$\frac{e^{i\omega\tau/2}\hat{H}_h^{n+1/2} - e^{-i\omega\tau/2}\hat{H}_h^{n-1/2}}{\tau} = -\frac{1}{\mu}\nabla_h \times \hat{E}_h^n + S_H, \quad (3)$$

where S_E and S_H are the source terms introduced by the incident plane wave. Because it solves the Maxwell's equations in the frequency domain instead of the time domain, it can be called a finite-difference frequency-domain (FDFD) discretization. For grid points in regions with negative permittivity $\epsilon < 0$, the iteration scheme for electric field (2) is modified to a “back iteration” scheme

$$\frac{e^{i\omega\tau}\hat{E}_h^n - \hat{E}_h^{n+1}}{\tau} = \frac{1}{\epsilon}\nabla_h \times \hat{H}_h^{n+1/2}e^{i\omega\tau/2} - \frac{\sigma}{\epsilon}\hat{E}_h^{n+1} + S_E, \quad (4)$$

to warrant the convergence of the iterative solver. A fixed point $(\hat{E}_h, \hat{H}_h)$ is then reached to approximate the solution of time-harmonic Maxwell's equations given in Eq. (1). For a detailed derivation and mathematical proof of the method, we refer the readers to Refs. 10 and 17.

The quantities of interest at the end of the simulations are the transmittance, absorptance, and reflectance of the Ag NW films. Assuming the incident wave has fixed wavelength λ and power $P_{inc}(\lambda)$, we calculate the time-averaged reflected and transmitted power of a film by integrating the electromagnetic flow over the top and bottom surface of the structure, which is given by

$$P_R(\lambda) = \frac{1}{2} \left| \int_{S_T} (\hat{E}(\lambda) \times \hat{H}(\lambda)) \cdot \vec{n} dS_T \right| \quad \text{and} \quad (5)$$

$$P_T(\lambda) = \frac{1}{2} \left| \int_{S_B} (\hat{E}(\lambda) \times \hat{H}(\lambda)) \cdot \vec{n} dS_B \right|,$$

where S_T and S_B are the top and bottom surfaces of the film, respectively, $\hat{E}(\lambda)$ and $\hat{H}(\lambda)$ are the electric and magnetic fields obtained by solving Maxwell's equation in the frequency domain, $\hat{E}(\lambda) \times \hat{H}(\lambda)$ denotes the Poynting vector which represents the directional energy flux density and $\vec{n}$ is the outward directed norm vector of the surfaces. Reflectance and transmittance are defined as $R(\lambda) = P_R(\lambda)/P_{inc}(\lambda)$ and $T(\lambda) = P_T(\lambda)/P_{inc}(\lambda)$, respectively. Subsequently, absorptance A can be calculated through the relation $A(\lambda) = 1 - R(\lambda) - T(\lambda)$. Since we are mainly interested in the application of Ag NW films in solar cells, the overall transmittance, reflectance and absorptance of an Ag NW film are studied under solar irradiance. They are defined as photon flux weighted values

$$\bar{R} = \int_{\lambda} R(\lambda) p(\lambda) d\lambda, \quad \bar{T} = \int_{\lambda} T(\lambda) p(\lambda) d\lambda \quad \text{and} \quad (6)$$

$$\bar{A} = \int_{\lambda} A(\lambda) p(\lambda) d\lambda,$$

where $p(\lambda)$ is the probability density function of the photons' distribution over the solar spectrum.

III. RESULTS AND DISCUSSION

We begin the simulation by a 2D model of Ag NW films with a single Ag NW or multiple Ag NWs aligned in parallel. The diameter of the Ag NWs is fixed as 30 nm. Our first simulation was a 100 nm wide film that contains a single Ag NW aligned parallel to the y-axis. If we assume infinite length and constant structure along the y-axis, the problem can be reduced to a 2D problem on the x-z plane. For an incident wave of TM polarization, the transmittance, reflectance, and absorptance spectrum of the film are shown in Fig. 2. The lowest value of transmittance or the highest value of absorptance is located at around 340 nm where plasmonic resonance occurs.^{5,12} This behavior is mainly governed by the localized surface plasmons, which are defined as non-propagating excitations of the conduction electrons of metallic nanostructures coupled to the electromagnetic field.¹⁹ The strength of this excitation can be characterized by a scalar α which is called polarizability. It relates the dipole moment of the excited resonance and the incident electric field by

$$\mathbf{p} = \pi a^2 \epsilon_0 \epsilon_m \alpha \mathbf{E}_0, \quad (7)$$

where $\mathbf{p}$ is the dipole moment, a is the diameter of the nanowire, ϵ_m is the permittivity of the dielectric around, and $\mathbf{E}_0$ is the incident electric field. Analytical expression for polarizability of an infinite-length single Ag NW placed in isotropic and non-absorbing medium can be derived by solving the Laplace equation for the potential as discussed in Ref. 19. It gives

FIG. 2. Simulation results under incident wave of TM polarization for a 100 nm wide film with a 30 nm-diameter Ag NW inside. The spectrum of transmittance (black circles), absorptance (red squares), and reflectance (blue diamonds). The inset plot shows the absolute value of polarizability calculated from the analytical expression for an infinite-length Ag NW of 30 nm diameter, when it is placed in free space and under incident wave of TM polarization.

$$\alpha = \frac{\epsilon_{Ag} - \epsilon_m}{\epsilon_{Ag} + \epsilon_m}, \quad (8)$$

where ϵ_{Ag} is the complex permittivity of silver, and ϵ_m takes the same value as in Eq. (7). For reference, the absolute value of polarizability as a function of wavelength is plotted as inset in Fig. 2. We notice that the plasmonic resonance is located at the same wavelength (340 nm) for both simulation and analytical results. This indicates that the simulation technique is suitable for modeling the plasmonic effects. For wavelengths larger than 400 nm, the absolute values of polarizability are rather small compared with its peak. This explains why absorptance is low in the higher wavelength range in Fig. 2(a). Plots of the amplitude of the electric field for different wavelengths are shown in Fig. 3. The patterns clearly show how the plasmonic resonance enhances the electric field around the Ag NW.

As mentioned in the literature,^{1,19} incident wave of TE mode does not excite plasmonic resonances. Therefore, it gives a totally different result for transmittance, absorptance, and reflectance. Comparison between the transmittance of the film with a single Ag NW under incident wave of TE and TM mode is shown in Fig. 4. Solar irradiance (dashed line in Fig. 4) is taken into account to calculate the weighted average of transmittance by Eq. (6). The weighted average

FIG. 3. The amplitude of the modeled optical electric field ($|E|$, unit: N/C) for three different wavelengths: 300 nm, 340 nm, and 700 nm.

FIG. 4. Transmittance comparison between TE (curve with blue squares) and TM (curve with red circles) modes for a single Ag NW model. AM1.5 solar irradiance is plotted as the dashed line for reference.

transmittance of the film under incident wave of TE mode is 70.8%, while the value under incident wave of TM mode is 94.6%, which gives average weighted transmittance of 82.7%. This value exceeds the film's geometric aperture of 70%. The additional 12.7% transmittance can be attributed to two aspects: one is the amount of light transmitted through the Ag NW because the Ag NW is very thin and silver is not totally opaque at the wavelength range we are considering, the other is high transmittance at large wavelengths caused by plasmonic effects. Furthermore, the first aspect can be estimated by simulation results with incident wave of TE mode, and the second can be estimated from TM mode results.

The second set of simulations we consider was a $1\ \mu\text{m}$ width film with the Ag NWs aligned equidistantly and in parallel along the y-direction. Three cases were studied with 10, 20, and 30 Ag NWs placed in the film. Fig. 5(a) shows the spectrum of the transmittance under circularly polarized incidence for the three cases. As the distance between the wires decreases, the dip of the transmittance spectrum is broader and red-shifted. This is caused by the coupling between plasmonic resonances of adjacent Ag NWs, which can be observed from the patterns of the resulting optical electric field presented in Fig. 5(b). Table I gives the comparison between the geometric aperture and the films' transmittance for the three cases. Results for different modes of polarization are given to reveal how much of the excess transmittance over geometric aperture is from the transmission through Ag NWs and how much is from the plasmonic effect. We can see that when there are 10 or 20 Ag NWs in the film, the plasmonic effect is the main reason for the high transmittance of the film. However, for 30 Ag NWs, the difference between transmittance under TE and TM modes decreases, which means that the plasmonic effect plays a less important role in improving the film's transmissivity as the density of Ag NWs increases.

Moreover, we deal with a more realistic model using a 3D structure, since solution-processed Ag NW films exhibit a randomly distributed Ag NW network inside. We take 500 Ag NWs, all with a length of $10\ \mu\text{m}$, and place them randomly in a $20\ \mu\text{m} \times 20\ \mu\text{m}$ film. Due to the computational

FIG. 5. (a) Transmittance spectra for $1\ \mu\text{m}$ wide Ag NW films with different number of parallel-aligned and equally spaced Ag NWs inside. This result is obtained under incident wave of circular polarization, and it can also be considered as an average of TE and TM mode results. (b) Coupling between plasmonic resonances of Ag NWs. Cases with 10, 20, and 30 wires are presented from top to bottom. Only half of the $1\ \mu\text{m}$ wide region is shown here. The left column is a sketch of the structure and the right column shows the patterns for the amplitude of electric field ($|E|$, unit: N/C) at incident wavelength of 340 nm. This result is obtained under incident wave of TM polarization.

limitation, simulation is restricted to a $1\ \mu\text{m} \times 1\ \mu\text{m}$ computational domain, using artificial periodic boundary condition for its extension. Fig. 6(a) shows the cross section of the structure in the x-y plane. The computation is carried out on LIMA cluster in Erlangen and SuperMUC cluster in Munich. The model discussed here involves the number of grid points at an order of 10^8 . It takes around 45 h running with 768 processors to finish one simulation over the spectrum with 80 sample wavelengths. Fig. 6(b) shows the spectrum of transmittance, absorbance and reflectance of this film. With 30.6% of the area occupied by silver, the film gives a transmittance of 80.14%. The value is 2.7% lower than what we get earlier from 10 parallel Ag NWs equidistantly placed in a $1\ \mu\text{m}$ wide film (30% occupied by silver and 82.7% of the incident light transmitted). The intersecting random Ag NWs

TABLE I. Film transmittance for $1\ \mu\text{m}$ wide Ag NW films with different number of parallel-aligned and equally spaced Ag NWs inside comparing to their geometric aperture. NUM = number of Ag NWs, GA = geometric aperture (%), and TT = transmittance (%).

NUM	GA	TT (TE)	TT (TM)	TT (circular)
10	70	70.8	94.6	82.7
20	40	41.6	87.8	64.7
30	10	25	36	30.5

FIG. 6. (a) Cross section in the x - y plane of the $1\ \mu\text{m} \times 1\ \mu\text{m}$ film with an Ag NW network inside. (b) Simulation results for the same structure: the spectrum of transmittance (black circles), absorptance (red squares), and reflectance (blue diamonds).

result in a broader dip of transmittance. However, the Ag NW film's overall transmissivity does not reduce a lot.

To compare simulation with measurement, we improve the model by taking a polymer binder into account. The reason is that the Ag NWs are not in direct contact with air in actual fabricated Ag NW films. Instead, they are cladded by a polymer layer so that the Ag NWs could be fixed on the substrate and also stabilized from environmental influence.⁶ We model the binder as a 6 nm thick ring around each Ag NW with a refractive index of 1.5. This layer has an effect on the absorption peak, which becomes broader and red-shifted comparing with the case without the polymer layer (see Fig. 7(a)).

The spectrum of the transmittance and absorptance of a sample Ag NW film with $\sim 30\%$ surface coverage of Ag NWs is measured. We carry out simulations with both 2D and 3D model to compare with the measured spectra. To begin with, we simulated a 2D model with parallel-aligned Ag NWs. The surface coverage of Ag NWs is adjusted to be 25%. The simulated spectra are compared with the experimental results in Fig. 7(b). We can observe that the simulated absorptance spectrum fits well to the experimental result. However, the simulated transmittance is lower than the measured transmittance for wavelengths above 450 nm.

FIG. 7. (a) Influence of the polymer binder. A film with parallel-aligned Ag NWs is simulated. The black circles show transmittance (solid line) and absorptance (dashed line) without the binder, while the red squares show the spectra for the same film when each Ag NW inside is cladded by a 6 nm thick ring of polymer; (b) Comparison between experiment and simulation. The solid lines represent the transmittance spectra, while the dashed lines represent the absorptance spectra. The black circles give the experiment results, and the red squares and blue diamonds show the simulation results for parallel-aligned Ag NWs and random Ag NWs, respectively. The layer thickness of the measured sample is about 80–90 nm with the possibility that 2 or 3 Ag NWs are on top of each other. The film for simulation has a thickness of 50 nm. It includes one layer of Ag NWs with 30 nm diameter and some space filled with air or cladding materials.

This deviation can be explained by the difference in the geometry of the Ag NWs. In the measured sample, the Ag NWs are randomly oriented, so that they polarize the incident light in a random way. This condition does not apply to the parallel aligned Ag NWs though. We can already observe the influence of polarization on the transmittance spectrum from Fig. 3 by looking into the TE/TM modes. This explanation is confirmed by a further 3D simulation of a $1\ \mu\text{m} \times 1\ \mu\text{m}$ film containing randomly distributed Ag NWs, which has a more similar transmittance spectrum to the real case. The film has 28.5% covered by Ag NWs. Its absorptance and transmittance spectrum are also shown in Fig. 7(b). Both spectra show a similar trend comparing with the experimental results. In Fig. 7(b), it shows that the simulated and experimental transmittance spectrum both possess a peak (absorptance dip) at around 320 nm and double dips

(absorptance peaks) at around 360 nm and 380 nm, respectively. Also, both the simulated and experimental results show that the transmittance decreases slowly from 500 nm on. The difference in values has three reasons. Firstly, it is difficult to obtain an accurate evaluation of the Ag NWs' surface coverage in actual fabricated films. Secondly, for reason of simplicity, the binder was modeled by a ring around the Ag NWs. But in reality, the polymer binder is distributed on the whole glass substrate with a varying thickness. And thirdly, the Ag NWs do not intersect each other in actual fabricated films. Instead, one Ag NW surmounts another when they meet. To achieve a more accurate comparison between experiments and simulation, we still need to make efforts in the following two directions. One is to retrieve useful information (e.g., surface coverage of Ag NWs) of actual fabricated Ag NW films though microscopy techniques, for example, transmission electron microscopy (TEM), and the other is to refine the model for a better approximation of the reality by improving the modeling of the binder as well as the Ag NWs' geometry.

IV. CONCLUSIONS

In this paper, we simulated optical properties of Ag NW films, by using an efficient technique, THIM. The algorithm can be implemented in parallel to meet the high computational demand of the problem. We obtained reasonable results for 2D test models, which confirm the reliability of the method. Also, we studied the reason for the films' high transmissivity by TE and TM mode analysis. Including parallel computation on HPC, the algorithm is well-suited for simulation of 3D large-scale structure with complex geometry of Ag NWs. Simulation results are in agreement with the experimental results from the following two points of view: (1) Both experimental and simulation results show that for Ag NW percolation type electrode layers the transmittance is much higher than expected from surface aperture; (2) After we consider a polymer binder into the model, the simulated and experimental spectra show a similar trend, as well as similar positions of dips and peaks. Our future work will include establishing a solid connection between real fabricated films and the simulation model, as well as continuing the study of the Ag NW films' functionality as transparent

electrodes by integrating their optical properties and electric characteristics.

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