

APL–Supplementary Material

near-field multiple scattering effects of plasmonic nanospheres embedded into thin-film organic solar cells

Wei E.I. Sha, Wallace C.H. Choy, Yang G. Liu, and Weng Cho Chew

Department of Electrical and Electronic Engineering
the University of Hong Kong, Pokfulam Road, Hong Kong
Email: wsha@eee.hku.hk (W.E.I. Sha)

1 Volume Integral Equation Method

As a rigorous solution to Maxwell’s equations, a volume integral equation (VIE) method is developed to characterize the optical absorption of organic solar cells. The coupling between multiple nanospheres (NSPs), as well as the interaction between NSPs and multilayer device structure, is fully taken into account in our model.

Considering non-magnetic optical materials with an arbitrary inhomogeneity profile, the VIE can be written as

$$\mathbf{E}^i(\mathbf{r}) = \frac{\mathbf{J}(\mathbf{r})}{-i_0\omega(\epsilon(\mathbf{r}) - \epsilon_0)} - i_0\omega\mu_0 \int_v \overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}') \cdot \mathbf{J}(\mathbf{r}') d\mathbf{r}' \quad (1)$$

where i_0 is the imaginary unit, $\mathbf{E}^i(\mathbf{r})$ is the incident electric field of the light, $\epsilon(\mathbf{r})$ is the position-dependent permittivity of the inhomogeneous materials, \mathbf{J} is the volumetric polarization current to be solved, and $\overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}')$ is the dyadic Green’s tensor in free space. The widely adopted approach for solving the VIE is the discrete dipole approximation (DDA) method [1]. Due to the hypersingularity of the Green’s tensor and spurious discontinuity of tangential E -field induced by the scalar (piecewise constant) basis functions, the DDA method cannot accurately characterize the subwavelength plasmonic physics [2] and breaks down in the multilayered device structure with high-contrast metallic materials. Here, we develop an alternate algorithm to bypass the difficulties. In our model, the polarization currents are expanded using the roof-top vector basis functions [3] and thus the continuity of normal current is naturally satisfied at the material interfaces. Furthermore, the hypersingular Green’s tensor is smoothed by using the finite-difference approximation.

From the VIE (1), the scattered electric field generated by the volumetric polarization current \mathbf{J} can be written as

$$\mathbf{E}^s(\mathbf{r}) = i_0\omega\mu_0 \int_v \overline{\mathbf{G}}(\mathbf{r}, \mathbf{r}') \cdot \mathbf{J}(\mathbf{r}') d\mathbf{r}' \quad (2)$$

Considering the Cartesian coordinate system, we use the short notation (u_1, u_2, u_3) substituting for (x, y, z) , then we have

$$\begin{bmatrix} E_1^s \\ E_2^s \\ E_3^s \end{bmatrix} = \begin{bmatrix} L_{11} & L_{12} & L_{13} \\ L_{21} & L_{22} & L_{23} \\ L_{31} & L_{32} & L_{33} \end{bmatrix} \begin{bmatrix} J_1 \\ J_2 \\ J_3 \end{bmatrix} \quad (3)$$

where

$$L_{ij} = \begin{cases} L_{ii}^c + L_{ii}^q, & i = j \\ L_{ij}^q, & i \neq j \end{cases} \quad (4)$$

$$L_{ii}^c J_i = i_0 \omega \mu_0 \int_v g(\mathbf{r}, \mathbf{r}') J_i(\mathbf{r}') d\mathbf{r}' \quad (5)$$

$$L_{ij}^q J_j = \frac{i_0}{\omega \epsilon_0} \frac{\partial}{\partial u_i} \int_v g(\mathbf{r}, \mathbf{r}') \frac{\partial J_j(\mathbf{r}')}{\partial u_j'} d\mathbf{r}' \quad (6)$$

$g(\mathbf{r}, \mathbf{r}') = \frac{\exp(i_0 k_0 |\mathbf{r} - \mathbf{r}'|)}{4\pi |\mathbf{r} - \mathbf{r}'|}$ is the scalar green's function, $\bar{\mathbf{G}}(\mathbf{r}, \mathbf{r}') = \left[\bar{\mathbf{I}} + \frac{\nabla \nabla}{k_0^2} \right] g(\mathbf{r}, \mathbf{r}')$, and k_0 is the wave number of free space.

Using the rooftop basis functions to expand the unknown currents, we have

$$\mathbf{J}(\mathbf{r}) = \sum_{i=1}^3 \mathbf{u}_i \sum_{k,m,n} J_i^D(k, m, n) T_{k,m,n}^i \quad (7)$$

where $T_{k,m,n}^1$, $T_{k,m,n}^2$, and $T_{k,m,n}^3$ are the volumetric rooftop functions given by

$$\begin{aligned} T_{k,m,n}^1 &= \Lambda_k(u_1) \Pi_m(u_2) \Pi_n(u_3) \\ T_{k,m,n}^2 &= \Pi_k(u_1) \Lambda_m(u_2) \Pi_n(u_3) \\ T_{k,m,n}^3 &= \Pi_k(u_1) \Pi_m(u_2) \Lambda_n(u_3) \end{aligned} \quad (8)$$

The functions $\Lambda_k(u_1)$ and $\Pi_m(u_2)$ are defined by

$$\begin{aligned} \Lambda_k(u_1) &= \begin{cases} 1 - \frac{|u_1 - k\Delta u_1|}{\Delta u_1}, & |u_1 - k\Delta u_1| \leq \Delta u_1 \\ 0, & \text{else} \end{cases} \\ \Pi_m(u_2) &= \begin{cases} 1, & \left| u_2 - \left(m - \frac{1}{2} \right) \Delta u_2 \right| < \frac{\Delta u_2}{2} \\ 0, & \text{else} \end{cases} \end{aligned} \quad (9)$$

The cuboid cells are employed to discretize the structure to be modeled. Here, Δu_1 and Δu_2 are the grid sizes of each small cuboid along x and y directions, respectively. Other functions in (8) can be defined in the same way.

As a result, the discretized form for the operator L_{ii}^c in (5) can be written as

$$L_{ii}^{D,c} J_i^D = i_0 \omega \mu_0 g^D \otimes J_i^D \quad (10)$$

where \otimes denotes the discrete convolution

$$g^D \otimes J_i^D = \sum_{k,m,n} g^D(k - k', m - m', n - n') J_i^D(k', m', n') \quad (11)$$

and

$$g^D(k, m, n) = \int_0^{\Delta u_1} \int_0^{\Delta u_2} \int_0^{\Delta u_3} g(u_{1,k} - u'_1, u_{2,m} - u'_2, u_{3,n} - u'_3) du'_1 du'_2 du'_3 \quad (12)$$

Likewise, the operator $L_{12}^{D,q}$ in (6) can be discretized as

$$\begin{aligned} L_{12}^{D,q} J_2^D &= \frac{-j_0}{\omega \epsilon_0 \Delta u_1 \Delta u_2} [g^D(k+1, m, n) - g^D(k, m, n)] \\ &\otimes [J_2^D(k, m, n) - J_2^D(k, m-1, n)] \\ &= \frac{-j_0}{\omega \epsilon_0 \Delta u_1 \Delta u_2} \{ [g^D(k+1, m, n) - g^D(k, m, n)] \\ &\quad - [g^D(k+1, m-1, n) - g^D(k, m-1, n)] \} \otimes J_2^D(k, m, n) \end{aligned} \quad (13)$$

where the finite-difference method is used for the smooth approximation of the dyadic Green's function.

The computations of the discrete convolutions can be performed efficiently by means of cyclic convolutions and fast Fourier transform (FFT) [4], which is similar to the DDA method. As a traditional iterative solver of the resulting VIE matrix equation, the conjugate-gradient method converges very slowly and will produce the non-physical random errors in the calculation of optical absorption. To tackle the problem, we employ the fast and smoothly converging biconjugate gradient stabilized (BI-CGSTAB) method [5]. The FFT is adopted to accelerate the matrix-vector multiplications encountered in the BI-CGSTAB solver with computational complexity of $O(N \log N)$ and memory of $O(N)$.

2 The Biconjugate Gradient Stabilized Algorithm

The resulting VIE matrix equation can be expressed as

$$Ax = b$$

The procedure of the biconjugate gradient stabilized (BI-CGSTAB) algorithm is given as follows:

Give an initial guess x_0 , we have

$$\begin{aligned} r_0 &= b - Ax_0, \hat{r}_0 = r_0 \\ \rho_0 &= \alpha = \omega_0 = 1 \\ v_0 &= p_0 = 0 \end{aligned}$$

Iterate for $i = 1, 2, \dots, n$

$$\begin{aligned}\rho_i &= \langle \hat{r}_0, r_{i-1} \rangle \\ \beta &= (\rho_i / \rho_{i-1}) (\alpha / \omega_{i-1}) \\ p_i &= r_{i-1} + \beta(p_{i-1} - \omega_{i-1}v_{i-1}) \\ v_i &= Ap_i \\ \alpha &= \rho_i / \langle \hat{r}_0, v_i \rangle \\ s &= r_{i-1} - \alpha v_i \\ t &= As \\ \omega_i &= \langle t, s \rangle / \langle t, t \rangle \\ x_i &= x_{i-1} + \alpha p_i + \omega_i s \\ r_i &= s - \omega_i t\end{aligned}$$

Terminate when

$$\frac{\|r_i\|_2}{\|b\|_2} < \eta$$

where η is the tolerance that specifies the desired accuracy of solution.

References

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