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Field Determination Near Plasmonic Structures by the Feynman-Kac Stochastic Representation

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Abstract—Feynman-Kac stochastic representation for elliptic equations is used to construct the solution of Helmholtz equation in domains containing plasmonic structures. The method involves initiation of Brownian motion at an interior point in the plasmonic structure and relating the field there to that on the structure boundary (equivalent surface) through the Feynman-Kac formula. The field exterior to the plasmonic structure can be related to the field on the equivalent surface through a variety of numerical methods depending on the complexity of the exterior domain. Finally, the field on the equivalent surface is determined by the imposition of boundary conditions. Comparison is shown here with the exact solution for the total field on the surface of a circular nano-cylinder and a nano-sphere driven by localized sources and where the permittivity is described by the Drude model.

Index Terms—Feynman-Kac representation, Plasmonic nano-materials, Elliptic equations, Stochastic technique, Drude dispersion model.

I. INTRODUCTION

In nano-plasmonic devices, electromagnetic waves interact with conduction electrons at metallic interfaces and lead to unique electrical properties that are attractive in a number of applications such as sub-wavelength imaging, solar-cells, environmental monitoring, national security, biosensing, photo thermal therapy, drug delivery, etc [1], [2], [3], [4]. Computational modeling of wave propagation inside a plasmonic structure could be very time consuming due to the necessity of employing adaptively refined meshes in and around the structures for treating the highly evanescent and singular fields of the plasmon or a polariton [5]. In this paper we demonstrate a technique that bypasses the construction of a mesh and directly generates the field at selected points in the structure without having to determine the fields at all other points. The technique is based on the Feynman-Kac stochastic representation for elliptic equations [6], [7] that becomes particularly attractive for plasmonic nanomaterials [8], wherein the real part of permittivity assumes negative values. The technique has ability to handle material inhomogeneities as well as non-linearities. Furthermore the technique is amenable to complete parallelization. The Feynman-Kac representation was previously considered by several authors for wave propagation problems. However, its utility was limited to sizes below the first resonance [9], [10], [11]. Even the eikonal/transport approach of [12] for treating the Helmholtz equation by random walks is limited to separable geometries in standard coordinate systems and

it requires analytical continuation of trajectories traversing a complex space. The approach presented here is free from such limitations and has the advantage of being directly amenable to integration with traditional numerical methods such as the integral equation methods or finite element methods for exterior regions in domains containing arbitrary plasmonic geometric shapes. In § II we give a brief introduction to the stochastic representation. In § III we outline procedures for coupling the interior and exterior regions via integral equation or series representation. Finally in § IV we demonstrate the stochastic technique by comparing the numerical solutions for 2D and 3D geometries.

II. STOCHASTIC REPRESENTATION

Consider the scalar elliptic boundary value problem (BVP)

$$\frac{1}{2}\nabla^2\psi + c\psi = 0, x \in D, \quad \psi(x_0) =: \psi_0(x_0), x_0 \in \partial D, \quad (1)$$

where x denotes the position vector¹ and D denotes the interior of the domain of interest having a boundary ∂D . The scalar quantity c is allowed to be a function of x to include inhomogeneous media and depends on the frequency of operation and the electrical parameters of the medium. In the case of lossy and/or plasmonic materials c is also complex-valued. Let $\tau_D^x = \inf(t > 0 : X_t^x \in \partial D)$, that is, τ_D^x is the *first exit ‘time’* (a random quantity that depends on the particular trajectory of the Brownian motion; units of [m²]) from the domain D of a 3-D Brownian motion X_t^x [13, p. 164] that starts at $x \in \mathcal{R}^3$ at time $t = 0$ as illustrated in Fig. 1. Define

Fig. 1. Exit time of Brownian motion from a closed domain.

the following additional quantities

$$c_t = \int_0^t c(X_s^x) ds, \quad w(x) = \mathbb{E}_x [\exp(c_{\tau_D^x})], \quad (2)$$

where c_t is a random variable that depends on the particular realization of Brownian motion X_t^x and $\mathbb{E}_x$ stands for the

¹For ease of notation, we do not explicitly use bold letters to denote vectors. However, it should become clear from the context whether a given quantity is a scalar or a vector.

expectation operator *given* x . For a homogeneous medium, the function $w(x)$ measures the mean growth of the exponential of the random variable $c_{\tau_D^x}$. We make the following assumptions: (i) D is a bounded connected open domain, (ii) ψ_0 and c are bounded and continuous; c is Hölder continuous [14, p. 152], and (iii) $|w(x)| < \infty$ for all $x \in D$. Then it can be shown by an application of Itô-Doebelin formula and the theory of *local martingales* that [6], [7]

$$\psi(x) = \mathbb{E}_x \left[\psi_0(X_{\tau_D^x}^x) \exp(c_{\tau_D^x}) \right]. \quad (3)$$

Formula (3) is known as the Feynman-Kac stochastic representation for density functions associated with diffusion type random processes. It states that the solution to BVP (1) at x is obtained by first initiating a 3-D Brownian motion, X_t^x , at x , determining the random exit time, τ_D^x , for that particular trajectory of Brownian motion, determining the value of the given data $\psi_0(x_0)$ at $x_0 = X_{\tau_D^x}^x$, and finally taking the expectation of $\psi_0(X_{\tau_D^x}^x) \exp(c_{\tau_D^x})$ over independent Brownian motions, all starting at x . For wave propagation problems in a dielectric (possibly lossy) medium, $c = k_0^2 \epsilon_s / 2$, where ϵ_s is the relative complex permittivity and k_0 is the wavenumber in free-space. It is easy to see that when ϵ_s is constant, the exit time τ_D^x is proportional to the *quadratic variation* of the Brownian path. In an ordinary material with the real part of permittivity satisfying $\Re(\epsilon_s) > 0$, the condition $|w(x)| < \infty$ is only met below the first resonance frequency as demonstrated in [11], [15]. However, in a plasmonic material, a surface plasmon is excited when $\Re(\epsilon_s) < 0$. Consequently $\Re(c_{\tau_D^x}) < 0$ and $|w(x)| < \infty$ is automatically met at all times. So for plasmonic materials the stochastic representation (3) remains valid for *any* domain size. It is important to realize that the space-time trajectory for Helmholtz equation here is governed by the Brownian motion in *real* space even for lossy media, a distinct advantage over the eikonal/transport equation procedure of [12], which requires trajectories in complex space.

In the next section we discuss how the stochastic representation can be used to determine the field in domains containing plasmonic structures by considering preliminary validation examples of a circular cylinder and a sphere both excited by localized sources. The essential idea is to couple the stochastic representation governing the interior fields with a numerical scheme (eg., integral equation, finite element, or analytical) describing the exterior fields and subsequently determining the surface fields by imposing boundary conditions.

III. WAVE PROPAGATION IN PLASMONIC STRUCTURES

For the sake of demonstration, the relative complex permittivity of the nanoparticle in the examples considered below is assumed to follow the Drude dispersion model [16]

$$\epsilon_s(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega^2 + \gamma^2} - j \frac{\gamma \omega_p^2}{\omega(\omega^2 + \gamma^2)} =: \epsilon_{rs} - j \epsilon_{is} \quad (4)$$

where ω_p is the *plasma frequency*, γ is the damping constant, ϵ_∞ is the relative permittivity at infinite frequency. Commonly used values for the parameters are [5] $\epsilon_\infty = 5$, $\omega_p = 1.4433 \times 10^{16}$ rad/s, and $\gamma = 10^{14}$ /s. For the above set of parameters, $\epsilon_{rs} < 0$ when the free-space wavelength λ_0 falls in the range $300 \text{ nm} < \lambda_0 < 600 \text{ nm}$.

A. 2D Example

The 2D example we consider is that of a circular nanocylinder of radius a with center at $(0, 0)$. The axis of the cylinder is along z . A magnetic line source with current density $\mathbf{M} = \hat{\mathbf{z}} V_0 \delta(x - 0) \delta(y - b)$, $b > a$ excites the cylinder. The analytical solution of this 2D, TE_z problem can be obtained via cylindrical harmonics [17], but is omitted here due to space limitations.

We consider a numerical solution by coupling the stochastic representation (3) for the interior tangential magnetic field, $\psi = H_z$, with an integral equation involving surface H_z and the tangential electric field $E_\tau = (j\eta_0/k_0) \partial H_z / \partial n$ and utilizing the exterior Green's function:

$$\frac{1}{2} H_z + \oint_{\partial D} \left[\frac{\partial H_z}{\partial n'} g_0 - H_z \frac{\partial g_0}{\partial n'} \right] ds' = H_z^i, \quad (5)$$

where $g_0(\boldsymbol{\rho}; \boldsymbol{\rho}') = 1/(4j) H_0^{(2)}(k_0 |\boldsymbol{\rho} - \boldsymbol{\rho}'|)$ is the 2D free-space Green's function, η_0 is the intrinsic impedance of free-space, n' is the outward normal on ∂D , and H_z^i is the incident magnetic field of the line source in free-space. Note that two coupled integral equations for the two unknowns H_z and E_τ are needed for a homogeneous cylinder in a purely integral equation technique. Also, volume integral equations or finite element methods will need to be considered for treating material inhomogeneities. We bypass the interior region integral equation(s) or finite methods here completely by using the fields determined from (3).

The unknowns H_z and E_τ on the surface are both expanded in terms of overlapping rooftop basis functions $b_n(s') = 1 - |s' - s_n|/\ell$, $s_n \leq s' \leq s_{n+1}$, where s' is the arc length parameter, $\ell = s_{n+1} - s_n$ is the semi-support of the basis functions, $n = 1, 2, \dots, N$. If $\hat{\mathbf{n}}$ denotes a unit inner normal to ∂D and if ψ^- denotes the field at the interior point $x = x_0 + \hat{\mathbf{n}}\Delta$, then (3) can be used to arrive at

$$\psi^- = \mathbf{A} \psi_0, \quad (6)$$

where $\mathbf{A}$ is an $N \times N$ matrix with entries

$$A_{mn} = \mathbb{E}_m [b_n(s_D) \exp(k_0^2 \epsilon_s \tau_D^m)], \quad (7)$$

and s_D is the arc length on the outer boundary ∂D at the exit time τ_D^m for a particle released at the interior point x_m . The interaction matrix $\mathbf{A}$ captures the non-local relationship between ψ^- and $\{\psi_0\}$ and depends only on the shape and electric parameters of the plasmonic particle and on the basis functions $b_n(s)$ used to represent the surface field. The surface field ψ_0 is finally determined by imposing continuity of tangential fields. Continuity of the tangential electric field across the boundary ∂D implies that $\partial H_z / \partial n(x_0^+) = (1/\epsilon_s) \partial H_z / \partial n(x_0^-)$. Using (6) for interior fields translates this to

$$\left\{ \frac{\partial H_z}{\partial n}(x_0^+) \right\} = \frac{1}{\epsilon_s \Delta} (\mathbf{I} - \mathbf{A}) \{H_z(x_0)\} \quad (8)$$

on employing a first order backward difference formula² for the normal derivative, where $\mathbf{I}$ is an identity matrix. When substituted into (5), one finally gets a matrix equation for the unknown H_z of the form

$$\left[\frac{1}{2} \mathbf{I} - \mathbf{C}' + \frac{1}{\epsilon_s \Delta} \mathbf{B} (\mathbf{I} - \mathbf{A}) \right] \{H_z\} = \{H_z^i\}, \quad (9)$$

²Other higher order approximations may be considered similarly by relating the field on multiple interior spheres to the surface field and employing higher order elements.

where

$$\begin{bmatrix} B_{mn} \\ C'_{mn} \end{bmatrix} = \int_{\partial D} b_n(s') \begin{bmatrix} g_0(\rho_m; s') \\ \frac{1}{k_0} \frac{\partial g_0}{\partial n'}(\rho_m; s') \end{bmatrix} k_0 ds'. \quad (10)$$

B. 3D Example

The 3D example we consider is that of a vertical electric dipole (VED) with current density $\mathbf{J} = I_0 \ell_0 \delta(x) \delta(y) \delta(z - b)$ placed over a plasmonic nanosphere of radius a . The center of the sphere is at the origin and $b > a$. The mode excited is TM_r and the fields H_ϕ and E_θ in a source-free region can be obtained in terms of the Debye potential ψ as

$$H_\phi = -\frac{1}{\mu_0} \frac{\partial \psi}{\partial \theta}, \quad E_\theta = -\frac{j}{\omega \epsilon_0 \mu_0 \epsilon_s r} \frac{\partial^2}{\partial r \partial \theta} (r\psi), \quad (11)$$

where ψ satisfies the homogeneous Helmholtz equation $\nabla^2 \psi + k_0^2 \epsilon_s \psi = 0$ inside the sphere and $\nabla^2 \psi + k_0^2 \psi = 0$ exterior to the sphere. The analytical solution for ψ can be expressed in terms of spherical harmonics [17] but is omitted here for space limitations.

For arbitrary geometries one may once again expand the field on the surface in terms of sub-domain basis functions as is done in subsection III-A. Here we will demonstrate the technique using entire domain basis functions involving Legendre polynomials. Accordingly, we expand the potential on the sphere $r = a$ and on an interior spherical contour $r < a$ as

$$\begin{bmatrix} \psi(a, \theta) \\ \psi^-(r, \theta) \end{bmatrix} = -\mu_0 H_0 \sum_{n=0}^{\infty} \begin{bmatrix} C_n \\ C_n^-(r) \end{bmatrix} P_n(\cos \theta) \quad (12)$$

with the unknown coefficients related to the surface fields as

$$\begin{bmatrix} C_n \\ C_n^-(r) \end{bmatrix} = \frac{2n+1}{2} \int_0^\pi \begin{bmatrix} \psi(a, \theta) \\ \psi^-(r, \theta) \end{bmatrix} P_n(\cos \theta) \sin \theta d\theta, \quad (13)$$

where $P_n(\cdot)$ is the Legendre polynomial of degree n and $H_0 = j k_0 I_0 \ell_0 / 4\pi b$. Equation (13) follows from (12) on using the orthogonality properties of Legendre polynomials on the surface of a sphere [17]. We also express the potential ψ^+ on an exterior spherical contour $a < r = \text{constant} < b$ as

$$\psi^+(r, \theta) = -\mu_0 H_0 \sum_{n=0}^{\infty} (2n+1) h_n^{(2)}(\zeta_b) P_n(\cos \theta) \left[j_n(\zeta^+) + a_n j_n(\zeta_a) \frac{h_n^{(2)}(\zeta^+)}{h_n^{(2)}(\zeta_a)} \right], \quad (14)$$

where $\zeta^+ = k_0 r$, $\zeta_a = k_0 a$, $\zeta_b = k_0 b$, $j_n(\cdot)$, $h_n^{(2)}(\cdot)$ are spherical Bessel and Hankel functions, respectively, of order n . In (14) we have expressed the total external field ψ^+ as the sum of a known incident field and a scattered field with unknown coefficients a_n . Continuity of H_ϕ at $r = a$ implies

$$C_n = (2n+1)(1+a_n) j_n(\zeta_a) h_n^{(2)}(\zeta_b), \quad n = 0, 1, \dots \quad (15)$$

Our goal here is to use (3) and the appropriate boundary conditions to determine the unknown coefficients C_n (and hence a_n). To this end, we originate a Brownian motion at $u = -\cos \theta$ on an interior contour $r = a - \Delta$, $\Delta > 0$, relate ψ^- to ψ via (3), note the exit time on $r = a$ at the point $\theta = \theta_e$ (corresponding $v = -\cos \theta_e$), use $c_{\tau_D}^x = k_0^2 \epsilon_s \tau_D^2(u, v; \omega) / 2$

in (13), and the orthogonality relations of $P_n(\cos \theta)$ on the unit sphere to finally arrive at

$$C_n^-(a - \Delta) =: C_n^- = \frac{(2n+1)}{2} \sum_{m=0}^{\infty} d_{nm} C_m, \quad (16)$$

$$d_{nm} = \int_{-1}^1 F_m(-u) P_n(-u) du, \quad (17)$$

$$F_m(-u) = \mathbb{E}_u \left[P_m(-v) e^{\frac{k_0^2 \epsilon_s \tau_D(u, v; \omega_k)}{2}} \right] \quad (18)$$

where the k th realization of the Brownian motion is indexed by ω_k . Truncating the upper limit of the summation to a reasonable number M , equation (16) may be cast in a matrix form as $\mathbf{C}^- = \mathbf{A} \mathbf{C}$. It is seen that the entries of the interaction matrix $\mathbf{A}$ (of size $(M+1) \times (M+1)$) as given in (17) and (18) can be computed entirely by the geometry of the plasmonic structure, its electrical parameters and the choice of expansion functions. Using a first-order backward difference formula for approximating $\partial/\partial r$, the electric field E_θ^- at the boundary ($r = a^-, \theta$) is equal to

$$E_\theta^- = \frac{j H_0}{\omega \epsilon_0 \epsilon_s a \Delta} \sum_{n=0}^{\infty} [(a + \Delta) C_n - a C_n^-] \frac{dP_n(\cos \theta)}{d\theta}. \quad (19)$$

From (11) and (14), the electric field E_θ^+ at $(r = a^+, \theta)$ can be written as

$$E_\theta^+ = \frac{j H_0}{\omega \epsilon_0 \epsilon_b a} \sum_{n=0}^{\infty} (2n+1) j_n(\zeta_a) h_n^{(2)}(\zeta_b) [R_{nj}(\zeta_a) + a_n R_{nh}(\zeta_a)] \frac{dP_n(\cos \theta)}{d\theta}, \quad (20)$$

with $R_{nj}(\zeta) = [\zeta j_n(\zeta)]' / j_n(\zeta)$ and $R_{nh}(\zeta) = [\zeta h_n^{(2)}(\zeta)]' / h_n^{(2)}(\zeta)$, a prime denoting derivative with respect to the argument of Bessel functions. Enforcing the continuity of E_θ at $r = a$, we obtain from (16), (19) and (20) that

$$D_n C_n - \frac{(2n+1)a}{2} \sum_{m=0}^{\infty} d_{nm} C_m = Q_n, \quad n = 0, 1, 2, \dots \quad (21)$$

where

$$D_n = (a + \Delta) - \Delta \epsilon_s R_{nh}(\zeta_a), \quad (22)$$

$$Q_n = \Delta \epsilon_s (2n+1) j_n(\zeta_a) h_n^{(2)}(\zeta_b) [R_{nj}(\zeta_a) - R_{nh}(\zeta_b)] \quad (23)$$

For a pre-determined M , the linear system (21) of size $(M+1) \times (M+1)$ can be solved for evaluating the coefficients C_n . Asymptotic analysis reveals that a reasonable upper limit is $M \approx \lceil 2|k_0 a \sqrt{\epsilon_s}| \rceil$, where $\lceil \cdot \rceil$ is the ceiling operator.

IV. NUMERICAL RESULTS AND CONCLUSIONS

Figures 2 and 3 show a comparison for the surface magnetic fields between the stochastic and the analytical solutions for $a = 400 \text{ nm}$ and $\lambda_0 = 450 \text{ nm}$. For the cylinder we choose $N = 180$, $b = a + \lambda_0$, whereas for the sphere we choose $M = 30$, $b = 1.1a$. The stochastic solution is generated with Monte Carlo averaging using N_r realizations. Brownian

motion is generated numerically using walk-on-cubes [18] with a lattice size $\Delta = a/100$. In d dimensions, time stepping for this random walk is set to $\Delta t = \Delta^2/d$ [18]. Note that the time step is proportional to the square of lattice size, a feature that is characteristic of random walks associated with diffusion processes [19]. In the results presented, symmetry of the geometries was not exploited in the generation of **A**. Excellent agreement with the analytical solution is seen for both the real and imaginary parts of the stochastic solution. Notice that $|H_z| \approx 2|H_z^i|$ in the lit region in Fig. 2, mimicking the behavior of a perfectly conducting (PEC) cylinder. However, unlike the PEC cylinder, the field is significant in the deep shadow region due to the presence of a surface plasmon [20]. Other quantities of interest such as the extinction cross section can be computed once the surface fields are known.

The procedure described here is applicable to arbitrary plasmonic geometries (including multiple particles) and material properties. If N basis functions are used to represent the surface field, a linear system of order $N \times N$ must be solved to determine it. All of the material inhomogeneities and non-linearities will be reflected in the quantity $c_{\tau_D}^x$ that appears in stochastic representation. Since the exit time plays a central role in the determination of field, significant insight can be gained from the availability of probability density functions of τ_D^x for several canonical geometries. We plan to present these in a future publication. Integration of the stochastic technique with other numerical methods such as frequency domain finite element method will greatly enhance the capabilities of the latter in the modeling of plasmonic devices and these will be pursued in the future. Other averaging techniques that can converge faster than Monte-Carlo approach will also be explored in the future.

Fig. 2. Tangential magnetic field on the surface of a circular cylinder excited by a line source at $b = a + \lambda_0$. Analytical solution H_z^a versus stochastic solution H_z^{st} . Arc length $s = 0$ in deep shadow at $x = 0, y = -a$.

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Fig. 3. Tangential magnetic field on the surface of a plasmonic nanosphere excited by a VED placed at $\theta = 0, b = 1.1a$ with $\mu_0 H_0 = -10^{-7}$. Analytical solution H_ϕ^a versus stochastic solution H_ϕ^{st} . Note that $H_\phi = 0$ at $\theta = 0, 180^\circ$.

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