Optical near-field scattering in a tip-sample system: A Green’s function approach

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Abstract. The optical near-field contribution to the total scattered intensity for a spherical metallic nanoparticle close to a dielectric planar interface is analytically calculated using a Green’s function formalism. This system is suited for probing a specimen of interest on a dielectric substrate with a taper optical metallic nanoantenna. Based on the effective polarizability model we find a Green’s function in the dipolar approximation, that allows one take into account the presence of the dielectric substrate. In particular, we provide evidence that the latter noticeably influences the near-field contribution and it should necessarily be included in the generalized formalism for calculating the Green’s functions for Raman scattering and fluorescence. A comparative analysis of the analytical model and finite-difference time domain based numerical simulation is performed.

1. Introduction
In classical optics an incident radiation polarizes an object under study and the induced polarization gives rise to emitted radiation through, for example, Rayleigh or Raman scattering. According to a near-field paradigm the interaction between optical radiation and matter is achieved by means of an intermediate object, often referred to as an optical antenna, tip or probe, and that is brought to the close proximity of the matter [1]. Light-matter interactions at the nanoscale have led to fast development of optical near-field microscopy methods, which allow one to probe a specimen with a sub-wavelength spatial resolution less than the Abbe diffraction limit and single molecule sensitivity due to the giant field enhancement [2,3]. Thus, optical antennas play a crucial role in near field optical spectroscopy and microscopy [4]. They represent nano-sized devices enabling to transfer freely propagating electromagnetic waves into localized modes and vice versa [5]. In general the near-field contributes to the total scattered field \( E_{sc} \), but it rapidly vanishes in the far-field zone. Within the frameworks of the Green's function formalism, a tip-sample system can be represented as follows:

\[
G = G_0 + G_{sc},
\]

where \( G_0 \) and \( G_{sc} \) are the Green's functions for freely propagating and scattered electromagnetic waves, respectively. The scattered field in the tip-sample system can be, in general, expanded to a series, namely:

\[
E_{sc} = E_s + E_i + E_{st} + E_{ts} + E_{stt} + \ldots
\]
where $E_s$ and $E_t$ are fields emitted or scattered from a sample and a tip, respectively; $E_{st}$ is the field scattered by the sample and then re-emitted by the tip; $E_{st}$ is the scattered field from the sample under illumination coming from the tip [1]; $E_{st}$ is a contribution to the scattered field as shown in figure 1(a). As a rule the sample represents an object under study on a substrate (glass, mica, etc.). Novotny’s group has analytically calculated a Green's function for tip-enhanced Raman scattering from a single walled carbon nanotube [6,7] (figure 1(b)). However, this approach does not take into account the Rayleigh scattering from the substrate. This is important for probing a specimen with a scattering-type antennas which allow one to localize and enhance incident optical radiation due to the lighting rod effect. Figure 1(c) demonstrates a general problem statement - a pointed metallic tip is brought to the dielectric substrate and is aside illuminated. Our aim is to determine a Green's function for near-field scattering with the tip-sample system and to compare it with the numerical simulation based on the finite-difference time domain (FDTD) method.

![Figure 1. Schematic of optical radiation scattering in the tip-sample system.](image)

2. Results and discussion

For finding the Green's function for Rayleigh scattering we replace the tapered nanoantenna (see figure 1) with a metallic nanosphere with a radius $\rho$ and a permittivity $\varepsilon_m$, that is placed at the distance $\Delta$ from a planar interface between two dielectric media with permittivities $\varepsilon_1$ and $\varepsilon_2$, respectively. This is schematically shown in figure 2.

![Figure 2. A schematic for optical near-field scattering.](image)
In order to calculate a field distribution near the nanoparticle we need to solve the inhomogeneous Helmholtz equation for an electric field which can be directly obtained from Maxwell’s equations [8]:

\[ \nabla \times \nabla \times E(r) - k_0^2 \varepsilon(r) E(r) = i \omega \mu_0 j_e(r) , \]

where \( k_0 \) is the wavevector for the incident radiation in vacuum, \( \omega \) is an angular frequency of the incident radiation and \( j_e(r) \) is an induced current density. The solution of Eq. (3) in the formalism of Green’s function reads as:

\[ E(r, \omega) = E_0(r, \omega) + i \omega \mu_0 \int \vec{G}(r, r_0, \omega) \vec{j}_e(r_0) dV_0 , \]

where \( \vec{G}(r, r_0; \omega) \) is a dyadic Green’s function. The current density of an electric dipole with moment \( p = \alpha E_0 \) located at \( r = r_0 \) is

\[ j_e(r) = -i \omega p \delta(r - r_0) . \]

The Green’s function in Eq. (4) includes, in general, these for freely propagating and scattered electromagnetic waves, that is, \( G_0 \) and \( G_{sc} \). In particular, the last term considers the scattering from the substrate. With the image method we readily derive

\[ \vec{G}(r, r_0, \omega) = \left( \mathbb{I} + \frac{1}{k^2} \nabla \nabla \right) \left( \frac{e^{-i k |r - r_0|}}{4 \pi |r - r_0|} - \frac{e^{-i k |r - r_0|}}{4 \pi |r - r_0|} \right) , \]

where \( r_0' \) is a vector position of an image dipole.

It is possible to incorporate the influence of the image field into an effective polarizability according to [9]

\[ p_{\text{eff}} = \alpha [E_0 + E_{\text{image}}] = \alpha_{\text{eff}} E_0 , \]

here \( E_{\text{image}} \) is a local field of the image dipole, which can be written as

\[ E_{\text{image}} = \frac{1}{4 \pi \varepsilon_0} \left( \frac{3(\Delta p)(r' - r_0)}{|r' - r_0|} - \frac{p_{\text{eff}}}{|r' - r_0|} \right) , \]

The polarizability of the small metallic sphere \( \alpha \) in Eq. (7) reads as [10]

\[ \alpha = 4 \pi \varepsilon_0 \rho \left( \begin{array}{ccc} \varepsilon_m - 1 & 0 & 0 \\ \varepsilon_m + 2 & 0 & 0 \\ 0 & \varepsilon_m - 1 & 0 \\ 0 & \varepsilon_m + 2 & f_e / 2 \end{array} \right) , \]

here \( f_e \) is a complex field enhancement factor depending on tip material and geometry, \( \varepsilon_m \) is a dielectric function of the sphere. Then effective polarizability reads as

\[ \alpha_{\text{eff}}^\parallel = \frac{\alpha^\parallel}{1 - \alpha^\parallel b \frac{1}{16 \pi \varepsilon_0 (\Delta + \rho)^3}} , \]

\[ \alpha_{\text{eff}}^\perp = \frac{\alpha^\perp}{1 - \alpha^\perp b \frac{1}{32 \pi \varepsilon_0 (\Delta + \rho)^3}} , \]
where \( b = \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + \varepsilon_1} \) is a quasi-static Fresnel-reflection coefficient. Changes in the local fields caused by the substrate are equivalent to the modification of tip polarizability. Thus, we can rewrite Eq. (4) as follows:

\[
E(\mathbf{r}, \omega) = E_0(\mathbf{r}, \omega) + \frac{\alpha_0^2}{\varepsilon_0 c^2} \mathcal{G}_0(\mathbf{r}, \mathbf{r}_s; \omega) \alpha_{\mathbf{r}_s} E_0(\mathbf{r}_s, \omega) \cdot \epsilon_{\mathbf{r}_s}, \omega \cdot \omega.
\]  

Eq. (12)

One of the most common experimental schemes in near-field imaging is the inverted optical configuration, in which the sample is illuminated with the highly focused laser beam with in-plane polarization. Since the antenna mainly interacts with longitudinal lobes we may drop X- and Y-components for the scattered field. In this case we substitute \( \rho = \Delta + \rho \), and the component of the dyadic Green’s function is written as:

\[
\mathcal{G}^{(0)}(\mathbf{r}) = \frac{e^{-ik(\Delta + \rho)}}{4\pi(\Delta + \rho)} \left( \delta_{\mathbf{x}} + il \frac{\delta_{\mathbf{z}}}{k(\Delta + \rho)} + 2 \frac{\delta_{\mathbf{z}}}{k^2(\Delta + \rho)^2} \right).
\]  

Eq. (13)

Since \( k(\Delta + \rho) \ll 1 \) we drop the first and the second terms in this relationship. By substituting Eq.(10) and Eq.(13) into Eq. (12) and neglecting \( E_0 \) compared to the enhanced field, we can readily derive

\[
E_\epsilon(\mathbf{r}, \omega) = \frac{f_s E_\epsilon(\mathbf{r}_s) e^{-i\rho^3}}{1 - f_s b \rho^3 (\Delta + \rho)}.
\]  

Eq. (14)

Figure 3 shows a comparative analysis of the field enhancement obtained with Eq.(14) and that calculated with FDTD method. For this purpose we used the following tip parameters: \( \rho = 20 \) nm, \( f_s = -2.9 + 11.8i \), \( \varepsilon_2 = 1.5 \) and \( \varepsilon_1 = 1 \). An excitation wavelength was \( \lambda = 632.8 \) nm. For numerical simulation the dielectric function for gold was taken from Ref. [11]. In a region where the approximation \( k(\Delta + \rho) \ll 1 \) is fulfilled analytical results (black curve) are in a good agreement with the numerical solution of Maxwell’s equations (blue dots).

Figure 3. Dependence of the field enhancement vs the tip-substrate distance calculated with Eq.(14) and the FDTD method. A red curve corresponds to the field enhancement when the substrate is absent.
6. Conclusion
In conclusion, we would like to highlight that the field enhancement in the tip-substrate system is caused by two processes: 1) interference between the external and the reflected optical fields; and 2) interaction between the dipole moment of the nanoantenna and the substrate. The first one depends on the material of the substrate only, whereas the second one depends on the parameters of both the tip and the substrate. As a result the field enhancement factor for the tip-substrate system noticeably differs from that for the case when the substrate is not taken into account. This is important for consideration of scattering-type problems in which plasmon-enhanced optical fields are negligible.

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References
[1] Novotny L 2011 Physics today 47-52
[2] Hayazawa N, Yano T and Kawata S 2012 Journal of Raman Spectroscopy 43 1177-1182
[3] Kharintsev S S, Noskov A I, Hoffman G 2007 Nanotechnology 18 315502
[4] Zhu X and Ohtsu M 2000 World scientific 205
[5] De Angelis F, Das G, Candeloro P et al. 2009 Nature Nanotechnology 5 67-72
[6] Cancado L G, Jorio A, Ismach A, Joselevich E, Hartschuh A and Novotny L 2009 Phys. Rev. Lett. 103 186101-4
[7] Maximiliano R V, Beams R, Novotny L, Jorio A and Cancado L G 2012 Phys. Rev. B 85 235334-2-8
[8] Novotny L and Hecht B 2006 Cambridge University Press 559
[9] Novotny L and Stranick S J 2006 Annu. Rev. Phys. Chem. 57 303-331
[10] Bouhelier A, Beversluis M, Hartschuh A and Novotny L 2003 Phys. Rev. Lett. 90 013903
[11] Johnson P B and Christy R W 1972 Phys. Rev. B 6 4370-4379