Journal of Physics: Conference Series **714** (2016) 012011 doi:10.1088/1742-6596/714/1/012011

Longitudinal polarizability and enhancement factor of a tapered optical gold nanoantenna

A R Gazizov[1,2], S S Kharintsev[1] and M Kh Salakhov[1,2]

[1] Kazan Federal University, 18 Kremlevskaya str., 420008 Kazan, Russian Federation
[2] Academy of Science of the Republic Tatarstan, 20 Bauman str., 420111 Kazan, Russian Federation

E-mail: Almaz.Gazizov@kpfu.ru

Abstract. This work focuses on the mechanism of electric field enhancement near a tapered optical antenna and the calculation of a complex field enhancement factor as a function of tip material, its curvature radius and cone angle. In this paper, an analytical model of longitudinal polarizability, taking into account retardation and dynamic polarization effects, is developed for evaluating the field enhancement factor.

1. Introduction

Optical antennas mediate energy between propagating electromagnetic waves and non-propagating near-fields [1]. The antennas are designed for effective concentration of optical field energy below the Abbe's diffraction limit and increasing the efficiency of tip-sample interactions [2]. There are the optical antennas that have found wide application in various microscopic techniques such as tip-enhanced Raman scattering (TERS) [3], antenna-assisted photoelectric cells [4], cancer treatment by the plasmonic probes [5], etc.

Physical properties of a taper optical antenna are characterised by three parameters: radius of curvature ρ, cone angle α and dielectric function $\varepsilon(\omega)$ [6]. Of the great importance is the field enhancement factor that depends on spatial distribution of induced currents. Despite the fact, that a lot of research groups worldwide have routinely used experimental tools of near-field microscopy and spectroscopy [7], some theoretical aspects in physics of the field enhancement still remain unsolved. In particular, it concerns a study of the field enhancement factor depending on antenna's parameters [8].

In Ref [9] authors have used a simple numerical model in which the tip apex is substituted for an anisotropic sphere. In this paper, we propose an analytical model of the tip apex, which suits to be used in the infrared region. We calculate the longitudinal polarizability of the nanoantenna in the approximation of the tip apex to be a prolate spheroid. The dynamic polarization and radiative damping of electric oscillations are taken into account by integration of retarded fields of elementary dipoles over the spheroid volume. Analytical results are compared to FDTD-based numerical simulation.

2. Results and discussion

Traditionally, the electric field around the tip of the nanoantenna is described with Green's function formalism [10-13]. By replacing the nanoantenna with a metallic nanosphere with a curvature radius

Journal of Physics: Conference Series **714** (2016) 012011 doi:10.1088/1742-6596/714/1/012011

of the tip apex ρ and the cone angle α the electric field in a tip-sample system can be written in the following form

$$\mathbf{E}(\mathbf{r},\omega) = \mathbf{E}_0(\mathbf{r},\omega) + \frac{\omega^2}{\varepsilon_0 c^2}\overset{\leftrightarrow}{\mathbf{G}}_0(\mathbf{r},\mathbf{r_0};\omega)\alpha_{\mathrm{eff}}\mathbf{E_0}(\mathbf{r_0},\omega), \tag{1}$$

where $\mathbf{E}_0$ is an incident laser field, ω is an angular frequency of the incident radiation, $\overset{\leftrightarrow}{\mathbf{G}}_0(\mathbf{r},\mathbf{r_0};\omega)$ is a Green's function in free space and α_{eff} is an effective polarizability, that contains information on the parameters of the optical antenna.

The effective polarizability of the small metallic sphere α_{eff} in equation (1) reads as [14]

$$\alpha_{\mathrm{eff}} = 4\pi\varepsilon_0\rho^3\begin{pmatrix} \frac{\varepsilon_{\mathrm{m}}-1}{\varepsilon_{\mathrm{m}}+2} & 0 & 0 \\ 0 & \frac{\varepsilon_{\mathrm{m}}-1}{\varepsilon_{\mathrm{m}}+2} & 0 \\ 0 & 0 & f_{\mathrm{e}}/2 \end{pmatrix} \tag{2}$$

$$\alpha_{\mathrm{eff},\perp} = 4\pi\varepsilon_0\rho^3\frac{\varepsilon_{\mathrm{m}}-1}{\varepsilon_{\mathrm{m}}+2} \tag{2a}$$

$$\alpha_{\mathrm{eff},\|} = 2\pi\varepsilon_0\rho^3\boldsymbol{f_e} \tag{2b}$$

where f_{e} is a complex field enhancement factor, ε_{m} is a dielectric function of the sphere. The expression for $\alpha_{\mathrm{eff},\|}$ originates from the requirement that the magnitude of the field at the surface of the tip is equal to the computationally calculated field set equal to $f_e E_0$ [15].

Modeling and simulating the complex field enhancement factor at various wavelengths are shown in figure 1. The parameters of the tapered tip are $\rho = 10$ nm, $\alpha = 30°$. For the numerical simulation the dielectric function of gold is approximated from Ref. [16] with a modified Drude model with a permittivity at infinite frequency of $\varepsilon_\infty = 7.5$, plasma frequency of $\omega_p = 1.38\cdot10^{16}$ rad/s and relaxation time of $\tau = 30$ fs.

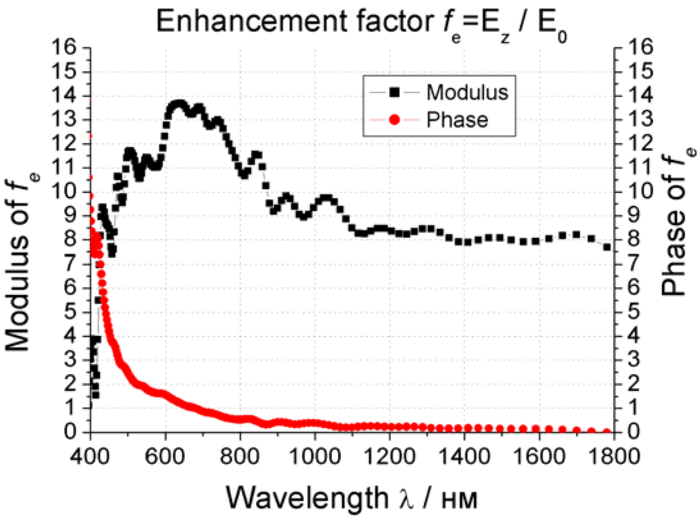

Figure 1. The computationally simulated enhancement factor modulus and phase for the gold tapered nanoantenna.

Since the simple sphere model does not provide anisotropy of the polarizability of the tapered optical antenna, we consider prolate spheroid as the model of the tip. Longitudinal polarizability of the metallic spheroidal particle in quasi-static approximation can be written as [17]:

$$\alpha_{\parallel} = \varepsilon_0 V \frac{\varepsilon_m - 1}{1 + n_{\parallel}(\varepsilon_m - 1)}, \tag{3}$$

where $n_{\parallel}$ is a geometric depolarization factor of ellipsoid. The depolarization factor is determined by the following expression for the prolate spheroid [17]:

$$n_{\parallel} = \frac{1-e^2}{e^2}\left(\frac{1}{2e}\ln\frac{1+e}{1-e} - 1\right), \tag{4}$$

where e is the eccentricity of the ellipse.

The quasi-static polarizability does not assume retardation processes such as dynamic polarization and self-radiation reaction in the taper nanoantenna, so the polarizability in equation (3) is not correct enough. To take these effects into account we use an idea that the electric field $\mathbf{E}$ inside the gold tip is a combination of the incident field $\mathbf{E}_0$ and depolarization field $\mathbf{E}_{dep}$. The polarization vector $\mathbf{P}$ can be expressed in these terms as [18]:

$$\mathbf{P} = \varepsilon_0(\varepsilon - 1)(\mathbf{E}_0 + \mathbf{E}_{dep}). \tag{5}$$

The depolarization field is a sum of all the retarded dipolar fields $\mathrm{d}\mathbf{E}_{dep}$ generated by elementary dipoles $\mathrm{d}\mathbf{p}(\mathbf{r}) = \mathbf{P}\mathrm{d}V(\mathbf{r})$ of each volume element $\mathrm{d}V$ inside the volume of the ellipsoid. Let the origin of spherical coordinate system be located at one of the foci of the ellipsoid and Z-axis be aligned with its major axis. To find the depolarization field we should integrate the Z-component of the field $\mathrm{d}\mathbf{E}_{dep,\parallel}$ generated at the origin by elementary dipole $\mathrm{d}p_{\parallel}(r)$ [18]:

$$\mathrm{d}E_{dep,\parallel} = \frac{1}{4\pi\varepsilon_0}\left[\frac{1}{r^3}(3\cos^2\theta - 1) + \frac{k^2}{2r}(\cos^2\theta + 1) + \mathrm{i}\frac{2}{3}k^3\right]\mathrm{d}p_{\parallel}(r), \tag{6}$$

where the retarded dipolar field generated by $\mathrm{d}p_{\|}(r)$ has been expanded into a power-series of (kr) up to the power k^3 [19]. Integration over the volume of prolate spheroid yields:

$$E_{\mathrm{dep}} = \frac{1}{4\pi\varepsilon_0}\left(-4\pi n_{\|} + 2\pi(1-n_{\|})k^2b^2 + \mathrm{i}\frac{8\pi}{9}k^3ab^2\right)P\,. \tag{7}$$

Here, a and b is a major and a minor semi-axis of prolate spheroid respectively. Inserting this result into equation (5) and solving it for P, one obtains

$$P_{\|} = \frac{\varepsilon - 1}{1 + (\varepsilon-1)n_{\|} + (\varepsilon-1)\dfrac{n_{\|}-1}{2}k^2b^2 - (\varepsilon-1)\mathrm{i}\dfrac{2}{9}k^3ab^2}\varepsilon_0 E_0\,, \tag{8}$$

$$\alpha_{\|} = \frac{\varepsilon - 1}{1 + (\varepsilon-1)n_{\|} + (\varepsilon-1)\dfrac{n_{\|}-1}{2}k^2b^2 - (\varepsilon-1)\mathrm{i}\dfrac{2}{9}k^3ab^2}\varepsilon_0 V\,. \tag{9}$$

We have obtained this result by using of an approximation of homogeneous field and polarization. As we see this polarizability in equation (9) differs from the polarizability of the quasi-static case in equation (3) by the denominator. The term proportional to k^2 is referred to the dynamic polarization, because this term vanishes in the case of $k = 0$ (static field). It also contributes only to the real part of the polarizability that corresponds to a change of effective depolarization factor of the spheroidal particle. This effect occurs due to the non-locality of the scattering process in time. The incident electromagnetic wave reaches different parts of the scatterer at different moments, so induced elementary dipoles are oscillating with different phases. Another reason for this term in the denominator is the excitation of electron plasma waves, which bound the field at the surface of the particle.

The term proportional to k^3 in the denominator is the radiation-damping correction of the quasi-static polarizability. It occurs due to the accelerated motion of charges inside the volume of the nanoantenna. It accounts for damping of the dipole by radiative losses and results in broadening and the reduction in intensity of the resonance band for the large particle. Since the antenna is a mediator between propagating electromagnetic waves and inductive localized field, one cannot consider it as a small volume particle and the analysis must contain the contribution of these two effects.

If we correct the result with the account of the retardation effect of the incident field analogous to the case of polarization of the sphere according to a power-series expansion of the Mie coefficients ${}^{\mathrm{e}}B_{\mathrm{l}}$ [18], for the complex enhancement factor of the field, we obtain:

$$f_{e,\|} = \frac{2}{3}\frac{1}{1-e^2}\frac{(\varepsilon_{\mathrm{m}}-1)(1-k^2p^2/10)}{1 + (\varepsilon_{\mathrm{m}}-1)n_{\|} + (0.7\varepsilon_{\mathrm{m}}-1)\dfrac{n_{\|}-1}{2}k^2b^2 - (\varepsilon_{\mathrm{m}}-1)\mathrm{i}\dfrac{2}{9}k^3ab^2} \tag{10}$$

here p is the ellipse parameter equal to the curvature radius of the tip apex $p = a(1-e^2)$. The eccentricity of the ellipse expressed in terms of the cone angle reads as $e = \sqrt{1-\tan^2\alpha}$.

Figure 2 shows a good agreement with the numerical simulation of the enhancement factor of the tapered antenna in the near-infrared region. Differences between both factors occur in the resonance band of the spheroidal particle. The tapered nanoantenna is a semi-infinite object and only its tip influences the polarizability. This means that surface plasmons excited at the tip apex are not strongly localized plasmons. This leads to a diffuse resonant band. In contrast, the spheroidal particle has a well-localized plasmon modes and a distinct resonance band. Hence, one cannot use this model to describe the resonance effects. The neglected processes become appreciable at shorter wavelengths and the small particle approximation of the tip becomes incorrect. Nevertheless, resonance effects vanish outside the resonance band at longer wavelengths and the small particle approximation remains valid.

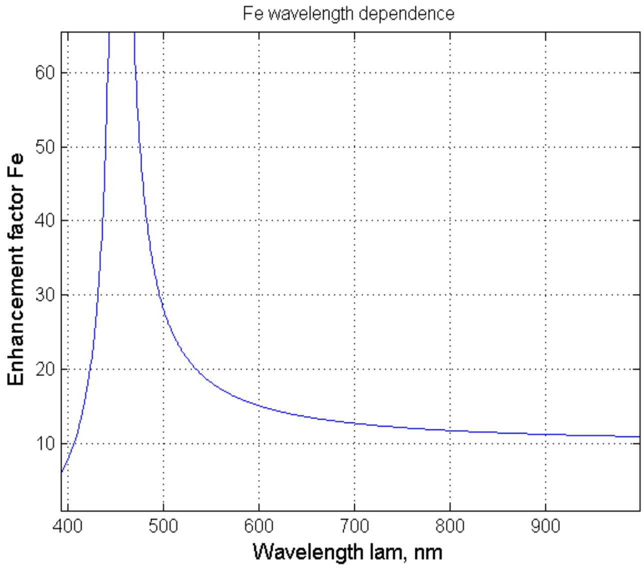

Figure 2. The absolute value of the enhancement factor of a gold spheroidal particle. The dielectric function of gold was taken from the modified Drude model with same parameters.

3. Conclusion

We have simulated the complex enhancement factor of the field for the gold tapered optical antenna at various wavelengths. We have proposed a prolate spheroid model to describe electric properties of the tip of the gold nanoantenna in the near infrared. This model accounts retardation of electric field by considering the dynamic polarization effect and radiative damping of electric oscillations in the framework of homogeneous polarization approximation. The model of polarizability remains valid at longer wavelengths.

Acknowledgments

Authors are grateful to Prof. A.I. Fishman for helpful discussions on the subject of near-field physics. This work was funded by the subsidy allocated to Kazan federal University for the state assignment in the sphere of scientific activities. This work was financially supported by the Russian Foundation for Basic Research (No. 15-42-02339). This work was done using equipment of Federal Center of Shared Equipment of Kazan Federal University.

References

[1] Novotny L and van Hulst N 2011 *Nature Photonics* **5** 83-90
[2] Maier S A and Atwater H A 2005 *J. Appl. Phys.* **98** 011101
[3] Georgi C and Hartschuh A 2010 *Appl. Phys. Lett.* **97** 143117
[4] Atwater H A and Polman A 2010 *Nature Mater.* **9** 205-213
[5] Bardhan R *et al* 2011 *Acc. Chem. Res.* **44** 936-946
[6] De Angelis F *et al* 2009 *Nature Nanotechnology* **5** 67-72
[7] Mino T, Saito Y and Verma P 2014 *ACS Nano* **8** 10187-10195

[8] Novotny L 2011 *Physics today* **64** 47-52
[9] Bouhelier A, Beversluis M R and Novotny L 2003 *Appl. Phys. Lett.* **82** 4596-4598
[10] Cançado L G, Jorio A, Ismach A, Joselevich E, Hartschuh A and Novotny L 2009 *Phys. Rev. Lett.* **103** 186101
[11] Maximiano R V, Beams R, Novotny L, Jorio A and Cançado L G 2012 *Phys. Rev.* B **85** 235434
[12] Novotny L and Hecht B 2006 *Principles of Nano-optics* (Cambridge: Cambridge University Press)
[13] Rogov A M, Gazizov A R, Kharintsev S S and Salakhov M Kh 2014 *J. Phys.: Conf. Ser.* **560** 012008
[14] Bouhelier A, Beversluis M, Hartschuh A and Novotny L 2003 *Phys. Rev. Lett.* **90** 013903
[15] Novotny L and Stranick S J 2006 *Annu. Rev. Phys. Chem.* **57** 303-331
[16] Johnson P B and Christy RW 1972 *Phys. Rev.* B **6** 4370-4379
[17] Landau L D and Lifshitz E M 1984 *Electrodynamics of continuous media* (Oxford: Butterworth-Heinemann)
[18] Meier M and Wokaun A 1983 *Optics Letters* **8** 581-583
[19] Born M and Wolf E 1980 *Principles of Optics*, 6th ed. (Oxford: Pergamon)