Influence of polarization direction, incidence angle, and geometry on near-field enhancement in two-layered gold nanowires*

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The influences of polarization direction, incidence angle, and geometry on near-field enhancements in two-layered gold nanowires (TGNWs) have been investigated by using the vector wave function method. When the polarization direction is perpendicular to the incidence plane, the local field factor (LFF) in TGNW decreases first and then increases with the increase in the incidence angle. The minimum LFF is observed at an incidence angle of 41°. It is found that the increase in the dielectric constant of the inner core leads to a decrease in the LFF. With the increase in the inner core radius, the LFF in TGNW increases first and then decreases, and the maximum LFF is observed at an inner core radius of 27 nm. On the other hand, when the polarization direction is parallel to the incidence plane, the collective motions of the induced electrons are enhanced gradually with the decrease in the incidence angle, and hence the near-field enhancement is increased.

Keywords: gold nanowire, localized surface plasmon resonance, near-field enhancement

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1. Introduction

Noble metal nanowires have attracted extensive interest due to their potential applications in nano-electronics, nano-optical devices, and biomedicine.[1−6] The Au nanowire has been used as a good plasmon biosensor.[3] Polarizing glass containing Ag nanowires can provide a significantly enhanced third-order nonlinear susceptibility.[4] Neubrech *et al.*[5] observed significant antenna-like plasmon resonances in Au and Cu nanowires, which have been applied as metallic nano waveguides.[6] Underlying these applications is localized surface plasmon resonance (LSPR) in the metal nanoparticles, the collective oscillations of conduction electrons induced by the interacting electromagnetic field, which can also induce a large electric field near the surface of the nanoparticle.[7]

Recently, many reports have shown that the optical properties of metal nanowire arrays are highly sensitive to the incidence angle and the polarization direction.[8−10] For example, the Cu nanowire arrays illuminated by a polarized light can present distinct anisotropic optical properties.[8] Podolskiy[9] reported that both the structure and the incidence angle have great effects on the propagating surface plasmon polariton modes in metal nanowires. Hendren *et al.*[10] also found that the optical properties of gold nanotube arrays are sensitive to the polarization state and the incidence angle. It follows that the plasmon resonances of a single metal nanowire can be controlled by the polarization direction and the incidence angle.[11] However, the influences of incidence angle and polarization direction on the near-field enhancements of the metal nanowires were seldom reported,[12] especially on the core–shell structure metal nanowires. Giannini *et al.*[12] studied the light scattering from metal wires with arbitrary cross sections by using the Green's theorem surface integral equations, and found that the near fields of the metal nanowires depend on the cross section, the incidence angle, and the polarization direction.

In this paper, we investigate the near-field properties of two-layered gold nanowires (TGNWs). The

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electric field enhancements in TGNWs have been calculated by using the vector wave function method. We focus on the influences of the incidence angle and the polarization direction on the near fields of TGNWs. In addition, the effect of the geometry on the near-field enhancement in TGNWs will be discussed.

2. Electromagnetic scattering of the model

The geometry of the TGNW is depicted in Fig. 1. The TGNW model consists of a particle with a dielectric core of radius r_1 and an outer Au shell of radius r_2. For a long nanowire, the values of r_1 and r_2 are much smaller compared with the length. Therefore, the gold nanowire can be considered as an infinitely long cylinder.[13] The permittivities of the core, the outer shell, and the embedding medium are ε_1, ε_2, and ε_3, respectively.

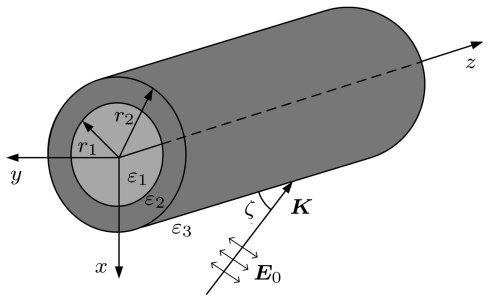


Fig. 1. The geometry of a core–shell structured gold nanowire illuminated by an electromagnetic plane wave at the incidence angle ζ. The z axis coincides with the axis of the nanowire, and the x–z plane is the incidence plane.

For metal nanostructures, ε_2 should be modified by replacing the ideal Drude part in the dielectric function with a size-dependent one because of the scattering of the conduction electrons in the particle surfaces, and is expressed as[14,15]

$$\varepsilon_2 = 1 - \frac{\omega_{\rm p}^2}{\omega^2 + {\rm i}\omega\gamma} + \chi_\infty, \tag{1}$$

where the background susceptibility χ_∞ arises from the electron polarizability and the interband transition, and $\omega_{\rm p}$ is the bulk plasma frequency. The modified collision frequency γ can be expressed as

$$\gamma = \gamma_{\rm f} + \frac{V_{\rm f}}{a}, \tag{2}$$

where $\gamma_{\rm f}$ is the bulk collision frequency, $V_{\rm f}$ is the Fermi velocity, and the reduced electron mean free path a equals the shell thickness $r_2 - r_1$. The parameters can be obtained by fitting the dielectric function to a particular frequency range of bulk dielectric data for Au.[16]

The infinitely long TGNW is illuminated by a homogeneous plane wave $\boldsymbol{E}_{\rm i}$ propagating in direction $\boldsymbol{K}$. There are two possible polarization states of the incident light, i.e., the electric field perpendicular to the incidence plane (x–z plane) and the one parallel to the incidence plane. When the incident light polarization is perpendicular to the incidence plane, the incident electric field $\boldsymbol{E}_{\rm i}$ can be expanded as[11,17]

$$\boldsymbol{E}_{\rm i} = -{\rm i}\sum_{n=-\infty}^{\infty} E_n \boldsymbol{M}_n^{(1)}, \tag{3}$$

where $E_n = E_0\,(-{\rm i})^n/(k\sin\zeta)$, and the incidence angle ζ is the angle between the incident light and the axis of the TGNW. The electric fields in core $\boldsymbol{E}_1$, shell $\boldsymbol{E}_2$, and embedding medium $\boldsymbol{E}_3$ can be given as[11,18]

$$\boldsymbol{E}_1 = \sum_{n=-\infty}^{\infty} E_n \left[t_n^{(1)}\boldsymbol{N}_{n,1}^{(1)} + q_n^{(1)}\boldsymbol{M}_{n,1}^{(1)}\right], \tag{4}$$

$$\boldsymbol{E}_2 = \sum_{n=-\infty}^{\infty} E_n [t_n^{(2)}\boldsymbol{N}_{n,2}^{(1)} + q_n^{(2)}\boldsymbol{M}_{n,2}^{(1)} - p_n^{(2)}\boldsymbol{N}_{n,2}^{(2)} - w_n^{(2)}\boldsymbol{M}_{n,2}^{(2)}], \tag{5}$$

$$\boldsymbol{E}_3 = \sum_{n=-\infty}^{\infty} E_n \left[b_{n{\rm I}}\boldsymbol{N}_n^{(3)} + {\rm i}a_{n{\rm I}}\boldsymbol{M}_n^{(3)}\right], \tag{6}$$

where $a_{n{\rm I}}$, $b_{n{\rm I}}$, $t_n^{(1)}$, $q_n^{(1)}$, $t_n^{(2)}$, $q_n^{(2)}$, $p_n^{(2)}$, and $w_n^{(2)}$ are the scattering coefficients, and $\boldsymbol{M}_{n,1}^{(1)}$, $\boldsymbol{M}_{n,2}^{(2)}$, $\boldsymbol{M}_n^{(3)}$, $\boldsymbol{N}_{n,1}^{(1)}$, $\boldsymbol{N}_{n,2}^{(2)}$, and $\boldsymbol{N}_n^{(3)}$ are the vector cylindrical harmonics functions.[17,18] Maxwell's boundary conditions are applied to resolve the unknown expansion coefficients of the scattered and the interior waves. The obtained extinction efficiency $Q_{\rm ext,I}$, scattering efficiency $Q_{\rm sca,I}$, and absorption efficiency $Q_{\rm abs,I}$ can be expressed as[17]

$$Q_{\rm ext,I} = \frac{2}{kr_2}{\rm Re}\left(a_{0{\rm I}} + 2\sum_{n=1}^{\infty} a_{n{\rm I}}\right), \tag{7}$$

$$Q_{\rm sca,I} = \frac{2}{kr_2}\left[|a_{0{\rm I}}|^2 + 2\sum_{n=1}^{\infty}\left(|b_{n{\rm I}}|^2 + |a_{n{\rm I}}|^2\right)\right], \tag{8}$$

$$Q_{\rm abs,I} = Q_{\rm ext,I} - Q_{\rm sca,I}. \tag{9}$$

When the incident light polarization is parallel to the incidence plane, the incident electric field $\boldsymbol{E}_{\rm i}$ can be

expanded as[11,17]

$$\boldsymbol{E}_{\rm i} = \sum_{n=-\infty}^{\infty} E_n N_n^{(1)}. \tag{10}$$

Then the electric fields in core $\boldsymbol{E}_1$, shell $\boldsymbol{E}_2$, and embedding medium $\boldsymbol{E}_3$ can be expressed as[11,18]

$$\boldsymbol{E}_1 = \sum_{n=-\infty}^{\infty} E_n \left[t_n^{(1)} \boldsymbol{N}_{n,1}^{(1)} + q_n^{(1)} \boldsymbol{M}_{n,1}^{(1)} \right], \tag{11}$$

$$\boldsymbol{E}_2 = \sum_{n=-\infty}^{\infty} E_n [t_n^{(2)} \boldsymbol{N}_{n,2}^{(1)} + q_n^{(2)} \boldsymbol{M}_{n,2}^{(1)} - p_n^{(2)} \boldsymbol{N}_{n,2}^{(2)} - w_n^{(2)} \boldsymbol{M}_{n,2}^{(2)}], \tag{12}$$

$$\boldsymbol{E}_3 = -\sum_{n=-\infty}^{\infty} E_n \left[b_{n\rm II} \boldsymbol{N}_n^{(3)} + {\rm i} a_{n\rm II} \boldsymbol{M}_n^{(3)} \right], \tag{13}$$

where $a_{n\rm II}$ and $b_{n\rm II}$ are the scattering coefficients. The obtained extinction efficiency $Q_{\rm ext,II}$, scattering efficiency $Q_{\rm sca,II}$, and absorption efficiency $Q_{\rm abs,II}$ can be expressed as[11,17]

$$Q_{\rm ext,II} = \frac{2}{kr_2} {\rm Re} \left(b_{0\rm II} + 2 \sum_{n=1}^{\infty} b_{n\rm II} \right), \tag{14}$$

$$Q_{\rm sca,II} = \frac{2}{kr_2} \left[|b_{0\rm II}|^2 + 2 \sum_{n=1}^{\infty} \left(|b_{n\rm II}|^2 + |a_{n\rm II}|^2 \right) \right], \tag{15}$$

$$Q_{\rm abs,II} = Q_{\rm ext,II} - Q_{\rm sca,II}. \tag{16}$$

3. Results and discussion

First, we investigate the state when the polarization direction is perpendicular to the incidence plane. Figure 2 shows a contour plot of the absorption spectra of TGNWs in water ($\varepsilon_3 = 1.7689$) as a function of incidence angle ζ. Here, r_1 and r_2 are fixed at 30 nm and 35 nm, respectively. The dielectric constant of the SiO_2 core is fixed at 2.04.

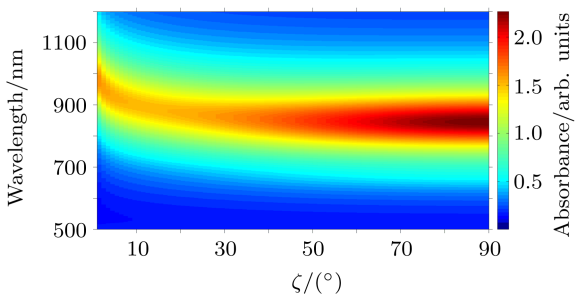


Fig. 2. (colour online) Contour plot of the absorption spectra of TGNWs in water as a function of incidence angle ζ. The polarization direction is perpendicular to the incidence plane.

It is found that the LSPR of TGNW shows a red shift with the decrease in the incidence angle. When the incidence angle $\zeta \neq \pi/2$, the scattering cross section should be an ellipse. The decrease in the incidence angle will increase the transmission distance of the incident light in TGNW, and hence increases the major axis of the cross-section ellipse. In this case, the phase retardation should be increased. The increase in the phase retardation decreases the number of electrons that collectively oscillate at the dipole resonance wavelength, and hence decreases the red shift of the LSPR.[19]

Figure 3 shows contour plots of the near field enhancement distributions in TGNWs. The incidence angles are (a) 90°, (b) 30°, (c) 20°, and (d) 10°. The calculation wavelengths correspond to the dipole plasmon resonances of TGNWs. For the different incidence angles, the large electrical field outside the particle is along the incident polarization direction and is only located within a few nm of the shell surface, which can be interpreted by the dipole plasmon resonance.[20] Large electric fields in the Au shell can be found at the two poles perpendicular to the polarization direction. According to the plasmon hybridization theory,[21–23] the LSPR of TGNW can be understood based on the interaction between the plasmons of a capillary with radius r_1 and a gold nanowire with radius r_2. The interaction results in a splitting of the plasmon resonances into a low energy mode (symmetric) and a high energy mode (antisymmetric). For the low energy mode, the same kind of charge on the inner and outer surfaces of the Au layer leads to the field lines inside the Au shell repelling each other in the poles along the incident polarization direction and bunching at the poles perpendicular to the polarization direction. In addition, it is observed that the repellence between the field lines inside the Au shell is reduced and the E-field in the inner core is decreased with the decrease in the incidence angle.

Figure 4(a) shows the variations in the local field factors (LFFs) of TGNWs with the incidence angle ζ. The LFF is defined to be the maximum of the electric field enhancement $\left|\boldsymbol{E}/E_{\rm i}\right|_{\rm max}$ near the surface of the particle, which is calculated at the dipole plasmon wavelength. It is found that with the decrease in the incidence angle, the LFF of TGNW decreases first from ~ 3.3018 at $\zeta = 90°$ to ~ 3.1425 at $\zeta = 41°$ and then increases to ~ 3.6416 at $\zeta = 10°$.

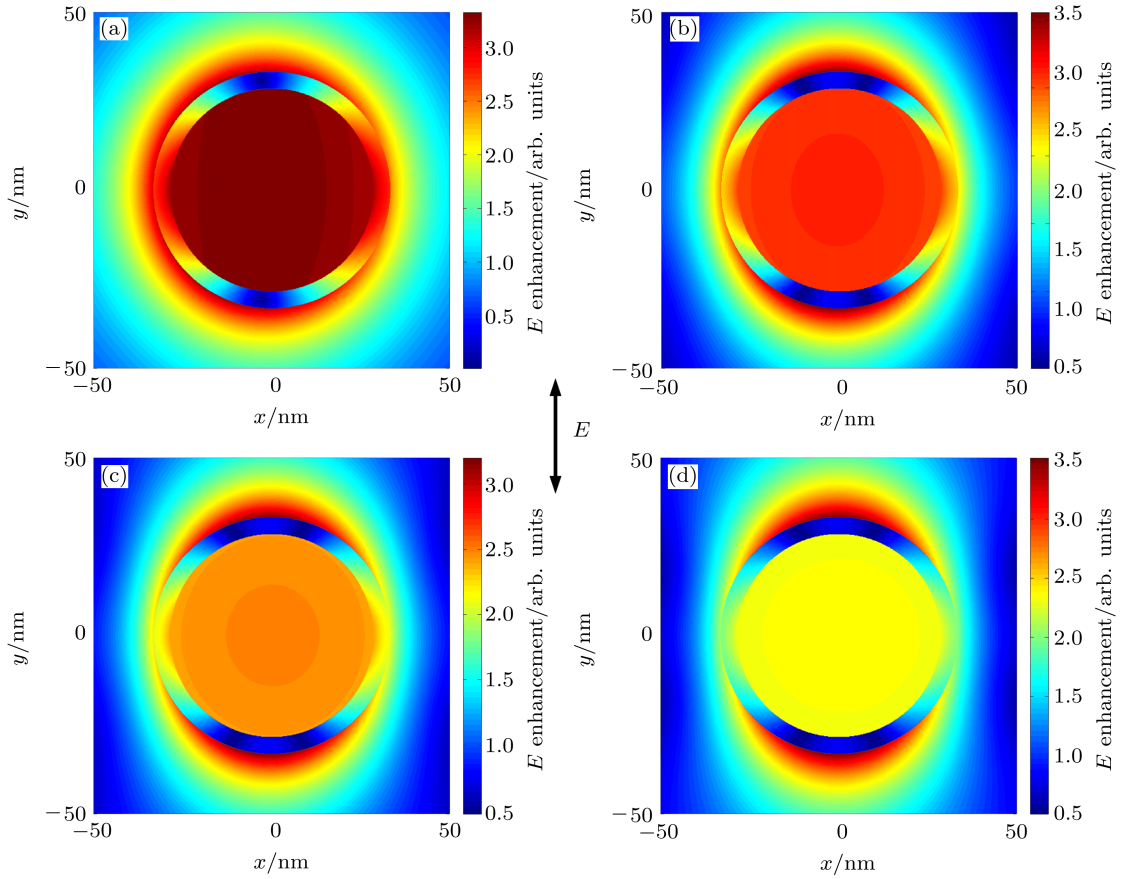


Fig. 3. (colour online) Contour plots of the E-field enhancement distributions of TGNWs. The incidence angles are (a) 90° (at ∼845 nm), (b) 30° (at ∼860 nm), (c) 20° (at ∼874 nm), and (d) 10° (at ∼898 nm). The polarization direction is perpendicular to the incidence plane.

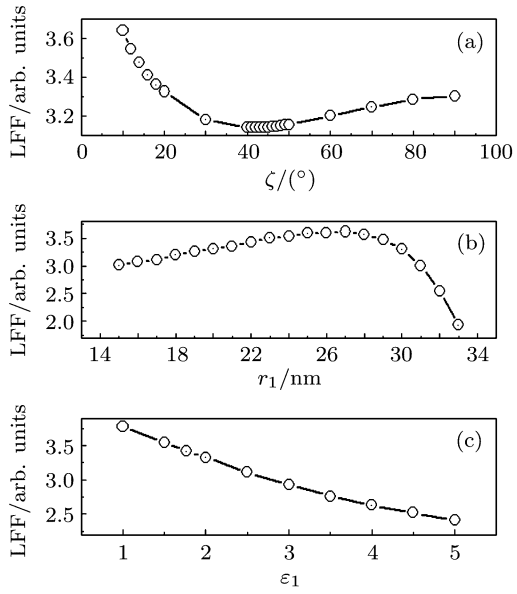


Fig. 4. (colour online) The dependences of the LFF of TGNW on (a) the incidence angle ζ, (b) the inner core radius r_1, and (c) the dielectric constant of the inner core ε_1. The polarization direction is perpendicular to the incidence plane.

With the decrease in the incidence angle, the increased major axis of the cross-section ellipse will cause two counteracting effects on the near-field enhancement: an increase in the conduction electrons available for plasmon resonance and a decrease in the electrons collectively oscillating at the dipole resonance wavelength due to phase retardation.[19] The increased conduction electrons increase the LFF, while the phase retardation reduces the LFF. For $\zeta < 41°$, the effect of the increased conduction electrons on the near field outweighs that of the phase retardation, which causes the appearance of the minimal LFF. Figure 4(b) shows the dependence of the LFF of TGNW on r_1. Here, r_2 and ζ are fixed at 35 nm and $\pi/2$, respectively. It is found that with the increase in the core radius, the LFF increases first from ∼ 3.0148 at $r_1 = 15$ nm to ∼ 3.6177 at $r_1 = 27$ nm and then decreases to ∼1.9344 at $r_1 = 33$ nm. The increased r_1 suppresses the separation between the inner and the outer surfaces of the Au shell. Then, the influence of the induced charge in the inner surface on the near field is enhanced, and hence the near field is increased. On the other hand, the increased core radius decreases

the shell thickness. When the Au shell is thin, the number of conduction electrons that are available for plasmon resonance will decrease with the increase in the core radius, and hence the near field is reduced.[19] When $r_1 > 27$nm, the effect of the decrease in the induced charge is stronger than that of the decreased separation, and the LFF is decreased. Figure 4(c) shows the variation in the LFF of TGNW as a function of ε_1. Here, r_1, r_2, and ζ are fixed at 30 nm, 35 nm, and $\pi/2$, respectively. With the increasing ε_1, the LFF decreases from ~3.7777 at $\varepsilon_1 = 1$ to ~ 2.4082 at $\varepsilon_1 = 5$. The increased ε_1 should reduce the induced charge in the inner surface of the Au shell, which leads to the decreased near field.[24]

Finally, we discuss the state when the polarization direction is parallel to the incidence plane. When $\zeta = \pi/2$, the light with its polarization direction along the axis of TGNW cannot induce the collective motions of conduction electrons. When $\zeta \neq \pi/2$, the angle between the polarization direction and the axis of TGNW is equal to $(\pi/2 - \zeta)$. In this case, plasmon resonances can be induced in TGNWs. Figure 5 shows the contour plot of the absorption spectra of TGNWs in water as a function of incidence angle ζ. With the increase in incidence angle ζ, the angle between the polarization direction and the axis of TGNW decreases, and then the plasmon resonances in TGNWs are gradually reduced. When ζ is above 40°, the dipole resonance peak almost disappears due to the weak strength. Figure 6 shows the contour plots of the E-field enhancements for TGNWs. The incidence angles are (a) 30°, (b) 20°, and (c) 10°. In Fig. 6(a), the collective motions of the induced electrons in the Au shell are very weak, and the scattering properties dominate the E-field distribution. The induced charge in the inner surface of the Au shell is very small, and hence the E-field in the core is very weak. With the decrease in the incidence angle, the plasmon resonance in TGNW and the collective motion of the induced electrons is enhanced gradually. Then, the E-field in the inner core increases, as shown in Figs. 6(b) and 6(c). The decreased incidence angle should increase the phase retardation, which causes the near field to be reduced. Therefore, the near-field enhancement in TGNW is small even for the small incidence angle.

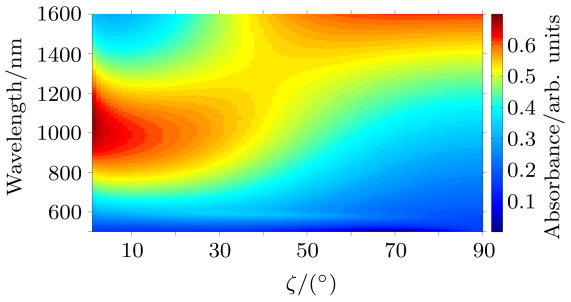


Fig. 5. (colour online) Contour plot of the absorption spectra for TGNWs as a function of incidence angle ζ. The polarization direction is parallel to the incidence plane.

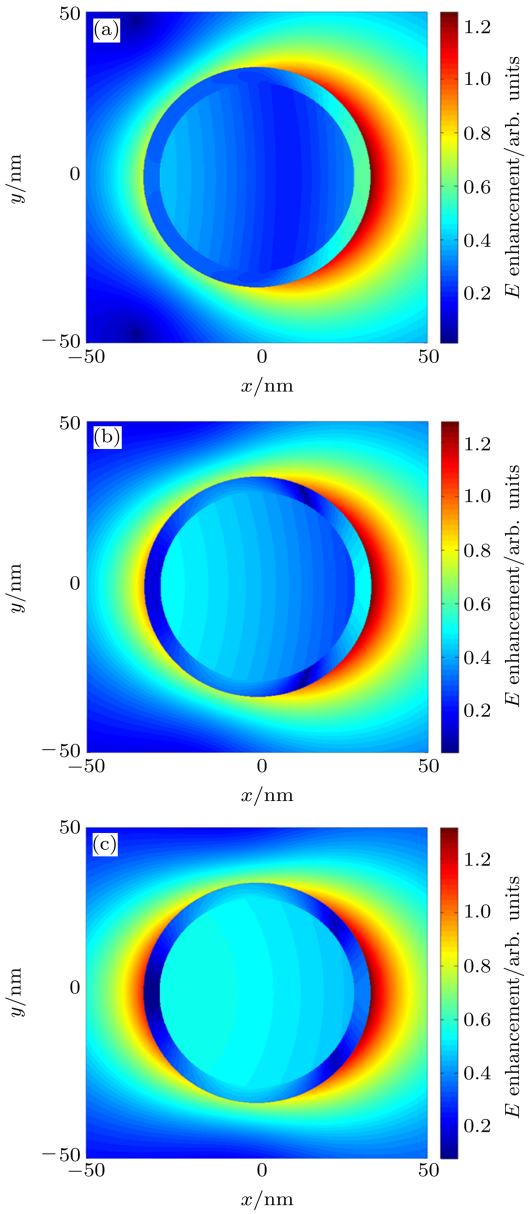


Fig. 6. (colour online) Contour plots of the E-field enhancement distributions of TGNWs. The incidence angles are (a) 30° (at ~1039 nm), (b) 20° (at ~994 nm), and (c) 10° (at ~986 nm). The polarization direction is parallel to the incidence plane.

We should denote that if the length of the nanowire is comparable to the radius of the nanowire, the above results will be different. For a finite length cylinder, when the incident polarization is not perpendicular to the axis of the gold nanowire, the collective motions of the conduction electrons in the two cross sections will occur and have a large effect on the plasmon resonances in the gold nanowire. When the incident polarization is perpendicular to the axis of the gold nanowire, the localizations from the two cross sections will also affect the plasmon resonance in the gold nanowire.

4. Conclusion

The near-field enhancement properties of TGNWs have been investigated by using the vector wave function method. When the polarization direction is perpendicular to the incidence plane, it is found that with the increase in the incidence angle, the LFF of TGNW decreases first and then increases. The minimum LFF is observed at an incidence angle of 41°. With the increase in the inner core radius, the LFF of TGNW increases first and then decreases, and the maximum can be observed in TGNW with $r_1 = 27$ nm and $r_2 = 35$ nm. In addition, the LFF in TGNW is also found to be sensitive to the dielectric constant of the inner core, i.e., it decreases with increasing ε_1. When the polarization direction is parallel to the incidence plane, the collective motion of the induced electrons is enhanced gradually with the decrease in the incidence angle. The near-field enhancements in TGNWs may be useful for further applications in nano-optical devices and biomedicine.

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