Theory of Localized Plasmons for Metal Nanostructures in Dielectrics

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A theory of localized bulk and surface plasmons for metal nanostructures in dielectrics is derived in the random phase approximation at the high frequency condition. The induced charge density in dielectrics is added to the induced charge density by the plasmons appeared in the previously reported integral equation for the scalar potential. The integral equation is composed of the local electron density in metal nanostructures, the local electric susceptibility of dielectrics and the retarded Green’s function. In the quasi-static approximation, the integral equation is transformed into the one that is composed of the local dielectric functions for metals and dielectrics and the Coulomb potential. Using a model where the local dielectric functions have step function shapes at the interfaces between the metals and dielectrics, it can be analytically solved in the quasi-static approximation. The localized surface plasmon frequencies and the light emission intensities are then derived for metal nanostructures such as ultrathin layer, nanowire, and nanosphere. [DOI: 10.1380/ejssnt.2018.329]

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I. INTRODUCTION

The localized surface plasmons (LSPs) can couple with photons to much enhance their local electric fields near metal nanostructures at the surface plasmon excitation energies [1–4]. These effects can use LSPs to fabricate nanoscale photonic devices and to perform very high-sensitive optical measurements.

The localized plasmons in multiple metal nanostructures reveal interesting phenomena, such as a red-shift of the surface plasmon frequency, large enhancement of the local electric fields, the nonlocal quantum effects and the quantum tunneling effects between the metal nanostructures [5–12].

The graphene plasmonics has been recently developed, where the two-dimensional (2D) electron density in graphene can be varied over a wide range by gating it to tune the plasmon resonance frequency [13–15]. Planar and edge plasmons in these 2D materials such as transition metal dichalcogenides [16–18] are also interesting because the plasmon resonance frequencies can be varied over a wide range by gating them to change the 2D electron density and changing the nanostructure sizes [19].

Many theoretical studies of the interaction between the external electric field or the incident electrons and the surface plasmon have been reported [20–24] up to now. The author has also developed theories of localized plasmons using the random phase approximation (RPA) at high frequency conditions [25–28]. In these theories, the local electron density in the metal nanostructures plays an essential role in the plasmon excitation, and the coupling between the localized bulk and surface plasmons is properly considered. These theories have been applied to calculate the light emission intensity from a single metal nanostructure by considering the retardation of the scalar potentials for localized plasmons [26] and to investigate localized plasmon excitations for multiple metal nanostructures [27]. Differential equation formula for the localized plasmons have been also derived, where the validity of the RPA formula has been also investigated [28].

These theories, however, are limited to metal nanostructures in vacuum or air where the dielectric polarizations at the metal surfaces can be ignored. They cannot be applied to metal nanostructures in dielectrics where the dielectric polarization cannot be ignored at the interfaces between metals and dielectrics.

In this study, a theory of localized plasmons for metal nanostructures that exist in dielectrics is derived in the RPA at the high frequency condition by considering the dielectric polarization in dielectrics. This is then applied to calculate localized surface plasmon frequencies for metal nanostructures in dielectrics and light emission intensities from the nanostructures using a model where the dielectric functions of metal nanostructures and dielectrics have step function shapes at their interfaces.

II. LOCALIZED PLASMONS FOR METAL NANOSTRUCTURES IN DIELECTRICS

Our previous studies [26–28] have shown that the following integral equation for the effective scalar potential \( \varphi_{\text{eff}}(r, \omega) \) and the external scalar potential \( \varphi_{\text{ext}}(r, \omega) \) for the metal nanostructures in vacuum or air is derived in the random phase approximation (RPA) at the high frequency condition:

\[
\varphi_{\text{eff}}(r, \omega) = \varphi_{\text{ext}}(r, \omega) + \int dr_1 G_0(r - r_1, \omega) \rho_m(r_1, \omega),
\]

where \( G_0(r - r_1, \omega) \) is the retarded Green’s function of the Helmholtz equation in frequency \( \omega \) and \( \rho_m(r_1, \omega) \) is the induced charge density in metal nanostructures, which are given by

\[
G_0(r - r_1, \omega) = \frac{\exp(i\omega|r - r_1|)}{|r - r_1|},
\]

\[
\rho_m(r_1, \omega) = \frac{e^2}{mc^2} \nabla_1 \cdot [n(r_1)E(r_1, \omega)],
\]

\[
E(r_1, \omega) = -\nabla_1 \varphi_{\text{eff}}(r_1, \omega) + \frac{i \omega}{c} A_{\text{eff}}(r_1, \omega).
\]

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\( \mathbf{E}(\mathbf{r}_1, \omega) \) is the electric field in the Lorentz gauge where \( A_{\text{eff}}(\mathbf{r}_1, \omega) \) is the effective vector potential, \( c \) is the velocity of light, \( m_e \) and \( -e \) are the electron mass and charge, respectively, and \( \nabla_1 \) represents the gradient with respect to \( \mathbf{r}_1 \). The Gaussian units are used in this article. \( n(\mathbf{r}_1) \) is the local electron density in metal nanostructures, which is given by

\[
n(\mathbf{r}_1) = 2 \sum_n |\Psi_n(\mathbf{r}_1)|^2 \theta(E_F - E_n),
\]

where \( \Psi_n(\mathbf{r}_1) \) is the normalized single-electron wave function with energy \( E_n \) in metal nanostructures, \( E_F \) is the Fermi energy and \( \theta(E_F - E_n) \) is the step function.

Equation (1) is valid for metal nanostructures in vacuum or air where the dielectric polarization can be ignored at the surfaces. To consider the case where metal nanostructures exist in dielectrics, the following induced charge density \( \rho_d(\mathbf{r}_1, \omega) \) should be added to \( \rho_m(\mathbf{r}_1, \omega) \) in Eq. (3):

\[
\rho_d(\mathbf{r}_1, \omega) = -\nabla_1 \cdot \mathbf{P}_d(\mathbf{r}_1, \omega) = -\nabla_1 \cdot [\chi_d(\mathbf{r}_1, \omega) \mathbf{E}(\mathbf{r}_1, \omega)],
\]

where \( \mathbf{P}_d(\mathbf{r}_1, \omega) \) is the dielectric polarization and the electric susceptibility \( \chi_d(\mathbf{r}_1, \omega) \) of the dielectrics is assumed to have the local position dependence and cubic symmetry, which can be expressed by the scalar. The electric susceptibility should be expressed by the tensor when the cubic symmetry is broken. Then the following integral equation is derived:

\[
\phi_{\text{eff}}(\mathbf{r}, \omega) = \phi_{\text{ext}}(\mathbf{r}, \omega) + \int d\mathbf{r}_1 G_0(\mathbf{r} - \mathbf{r}_1, \omega) \rho(\mathbf{r}_1, \omega),
\]

\[
\rho(\mathbf{r}_1, \omega) = \rho_m(\mathbf{r}_1, \omega) + \rho_d(\mathbf{r}_1, \omega) = \nabla_1 \cdot \left\{ \left[ \frac{\varepsilon^2 n(\mathbf{r}_1) + 1}{m_e \omega^2} - \chi_d(\mathbf{r}_1, \omega) \right] \mathbf{E}(\mathbf{r}_1, \omega) \right\}.
\]

By the charge conservation law \( i \omega \rho(\mathbf{r}_1, \omega) = \nabla_1 \cdot \mathbf{j}(\mathbf{r}_1, \omega) \), the induced current density is given by

\[
\mathbf{j}(\mathbf{r}_1, \omega) = i \omega \left[ \frac{\varepsilon^2 n(\mathbf{r}_1) + 1}{m_e \omega^2} - \chi_d(\mathbf{r}_1, \omega) \right] \mathbf{E}(\mathbf{r}_1, \omega).
\]

Then the following integral equation for the effective vector potential is derived:

\[
A_{\text{eff}}(\mathbf{r}, \omega) = A_{\text{ext}}(\mathbf{r}, \omega) + \frac{1}{c} \int d\mathbf{r}_1 G_0(\mathbf{r} - \mathbf{r}_1, \omega) \mathbf{j}(\mathbf{r}_1, \omega).
\]

The effective scalar potential in Eq. (6) and the effective vector potential in Eq. (8) satisfy the Lorentz condition \( \nabla \cdot A_{\text{eff}}(\mathbf{r}, \omega) = \frac{i}{\epsilon} \phi_{\text{eff}}(\mathbf{r}, \omega) \) when the external scalar potential \( \phi_{\text{ext}}(\mathbf{r}, \omega) \) and the external vector potential \( A_{\text{ext}}(\mathbf{r}, \omega) \) satisfy the Lorentz condition. Equations (6) and (8) can be used to study the localized plasmons for metal nanostructures in dielectrics, where the retardation effect for the localized plasmons are properly considered. The differential equation formulas of Eqs. (6) and (8) are also given in Appendix A.

Using the quasi-static approximation \( \omega/c \rightarrow 0 \) indicating that the retardation effect is ignored, i.e., in the case \( ka \ll 1 \) where \( k \) is the wave number of light and \( a \) is the typical size of metal nanostructures, which is considered valid for metal nanostructures [28]. Eq. (6) is transformed using \( \mathbf{E}(\mathbf{r}_1, \omega) \approx -\nabla_1 \phi_{\text{eff}}(\mathbf{r}_1, \omega) \), integration by parts and the relation \( \nabla_1^2 \frac{1}{r-\mathbf{r}_1} = -4 \pi \delta(\mathbf{r} - \mathbf{r}_1) \) into

\[
[\varepsilon_m(\mathbf{r}, \omega) + \varepsilon_d(\mathbf{r}, \omega) - 1] \phi_{\text{eff}}(\mathbf{r}, \omega) - \frac{1}{4 \pi} \int d\mathbf{r}_1 \mathbf{E}_{\text{eff}}(\mathbf{r}_1, \omega) \nabla_1 \left[ \frac{1}{|\mathbf{r} - \mathbf{r}_1|} \cdot \nabla_1 \right] [\varepsilon_m(\mathbf{r}, \omega) + \varepsilon_d(\mathbf{r}, \omega) - 2] = \phi_{\text{ext}}(\mathbf{r}, \omega),
\]

where \( \varepsilon_m(\mathbf{r}, \omega) \) and \( \varepsilon_d(\mathbf{r}, \omega) \) are local dielectric functions of metals and dielectrics respectively, which are given by

\[
\varepsilon_m(\mathbf{r}, \omega) = 1 - \frac{\omega_p^2(\mathbf{r})}{\omega^2}, \quad \varepsilon_p(r) = \sqrt{\frac{4 \pi e^2 n(r)}{m_e}},
\]

\[
\varepsilon_d(\mathbf{r}, \omega) = 1 + 4 \pi \chi_d(\mathbf{r}, \omega),
\]

where \( \omega_p(\mathbf{r}) \) is the local bulk plasmon frequency. The dielectric functions of dielectrics \( \varepsilon_d(\mathbf{r}, \omega) \) have generally complex values with finite imaginary parts near the optical absorption frequencies in the dielectrics.

The first and second terms in the left side of Eq. (9) are related to the localized bulk and surface plasmons, respectively. Equations (9) and (10) are used to study the localized plasmons for metal nanostructures in dielectrics such as metal ultrathin layer, nanowire, and nanosphere in dielectrics, using a model where the local dielectric functions have step function shapes at the interfaces between the metals and dielectrics.
III. APPLICATIONS TO METAL NANOSTRUCTURES

A. Surface plasmon frequencies for metal nanostructures

1. Metal ultrathin layer

In the quasi-static approximation, the localized plasmons are studied for a metal ultrathin layer in a dielectric, which has a thickness \( l \) in the \( z \) direction and spreads in the \( \rho = (x, y) \) plane as shown in Fig. 1. The local electron density in the metal layer and the local electric susceptibility of the dielectric are assumed to have the following step function shapes:

\[
\varepsilon_m(r_1, \omega) = 1 - \frac{4\pi e^2 n_0 [\theta(z_1) - \theta(z_1 - l)]}{m_e \omega^2} = 1 - \frac{\omega_e^2}{\omega^2} [\theta(z_1) - \theta(z_1 - l)],
\]

\[
\varepsilon_d(r_1, \omega) = 1 + 4\pi \chi_d^0(\omega) [\theta(-z_1) + \theta(z_1 - l)],
\]

\[
\omega_p = \sqrt{\frac{4\pi e^2 n_0}{m_e}},
\]

where \( n_0, \chi_d^0(\omega), \) and \( \theta(z_1) \) are the constant electron density, the homogeneous electric susceptibility, and the step function, respectively. The scalar potential and the Coulomb potential in Eq. (9) are expanded using the Fourier expansion with respect to \( \rho = (x, y) \) as

\[
\varphi_{\text{eff}}(\rho, z, \omega) = \frac{1}{(2\pi)^2} \int \frac{dk}{|r - r_1|} \varphi_{\text{ext}}^\rho(k, z, \omega) e^{ik \rho},
\]

\[
\frac{1}{(2\pi)^2} \int \frac{dk}{k} e^{-k|z - z_1|} e^{ik(\rho - \rho_1)}. \tag{12}
\]

After some calculations described in Appendix B, the effective scalar potential at \( z = 0 \) is given by

\[
\varphi_{\text{eff}}(k, 0, \omega) = \left[ 1 - \frac{\omega_e^2}{\omega^2} + 2\pi \chi_d^0(\omega) \right] \varphi_{\text{ext}}(k, 0, \omega) - \left[ \frac{\omega_e^2}{\omega^2} + 2\pi \chi_d^0(\omega) \right] e^{-kl} \varphi_{\text{ext}}(k, l, \omega)
\]

\[
\left[ 1 - \frac{\omega_e^2}{\omega^2} + 2\pi \chi_d^0(\omega) \right] - \left[ \frac{\omega_e^2}{\omega^2} + 2\pi \chi_d^0(\omega) \right] e^{-2kl}.
\tag{13}
\]

The effective scalar potential at \( z = l \) is given by

\[
\varphi_{\text{eff}}(k, l, \omega) = \left[ 1 - \frac{\omega_e^2}{\omega^2} + 2\pi \chi_d^0(\omega) \right] \varphi_{\text{ext}}(k, l, \omega) - \left[ \frac{\omega_e^2}{\omega^2} + 2\pi \chi_d^0(\omega) \right] e^{-kl} \varphi_{\text{ext}}(k, 0, \omega)
\]

\[
\left[ 1 - \frac{\omega_e^2}{\omega^2} + 2\pi \chi_d^0(\omega) \right] - \left[ \frac{\omega_e^2}{\omega^2} + 2\pi \chi_d^0(\omega) \right] e^{-2kl}.
\tag{14}
\]

The localized surface plasmons frequencies are given by the poles of Eq. (13) or Eq. (14):

\[
\omega_k^\pm = \omega_p \sqrt{\frac{1 + e^{-kl}}{2 + 4\pi \text{Re}\left[ \chi_d^0(\omega_k^\pm) \right] \left( 1 - e^{-kl} \right)}}.
\tag{15}
\]

where \( \text{Re} \) means the real part of the electric susceptibility.

The frequencies coincide with those reported by Ritchie et al. \cite{29} when \( \chi_d^0(\omega) = 0 \), i.e., in vacuum or air. The frequencies \( \omega_k^\pm \) correspond to the antisymmetric mode (+) with antisymmetric induced charge distribution in the \( z \)

\[
\omega_k^\pm = \omega_p \sqrt{\frac{1 + e^{-kl}}{2 + 4\pi \text{Re}\left[ \chi_d^0(\omega_k^\pm) \right] \left( 1 - e^{-kl} \right)}}.
\]

\[
\text{FIG. 1. Geometrical arrangement of a metal ultrathin layer in the dielectric.}
\]
direction and the symmetric mode (−) with the symmetric induced charge density, respectively. When a dielectric with semi-infinite thickness \( l \to \infty \) is considered in Eq. (15), the surface plasmon frequency becomes the well-known \( \omega_p / \sqrt{1 + \text{Re}[\varepsilon_d(\omega)]} \), where \( \varepsilon_d(\omega) = 1 + 4\pi\chi_d^0(\omega) \); the homogeneous dielectric function of the dielectric [1].

2. Metal nanowire

The localized plasmons are studied for a metal nanowire with the radius \( a \) in the \( \rho = (x, y) \) plane in a dielectric, whose center axis is parallel to the \( z \) direction as shown in Fig. 2. The local dielectric function of the nanowire and that of the dielectric are assumed to have the following step function shapes:

\[
\varepsilon_m(r_1, \omega) = 1 - \frac{\omega_p^2}{\omega^2} \theta(a_1 - r_1), \\
\varepsilon_d(r_1, \omega) = 1 + 4\pi\chi_d^0(\omega) \theta(r_1 - a).
\]

The scalar potential and the Coulomb potential in Eq. (9) are expanded using cylindrical coordinates as

\[
\varphi_{\text{eff}}(r, \omega) = \frac{1}{2\pi} \sum_{m=-\infty}^{\infty} e^{im\phi} \int_{-\infty}^{\infty} dq \varphi_{\text{ext}}^m(\rho, q, \omega) e^{iqz}, \\
\frac{1}{|r - r_1|} = \frac{1}{\pi} \sum_{m=-\infty}^{\infty} e^{im(\phi - \phi_1)} \int_{-\infty}^{\infty} dk I_m(k\rho_<) K_m(k\rho_> \rho_1) e^{ik(z - z_1)},
\]

where \( r = (\rho, z) \), \( m \) is integer, \( \phi \) is the azimuth angle in the \( \rho = (x, y) \) plane, \( \rho_< = \min(\rho, \rho_1) \), \( \rho_> = \max(\rho, \rho_1) \), and \( I_m(s) \) and \( K_m(s) \) are the modified Bessel functions of the first and second kinds, respectively [30, 31].

After some calculations described in Appendix B, the effective scalar potential at \( \rho = a \) is given by

\[
\varphi_{\text{eff}}^m(a, q, \omega) = \frac{\varphi_{\text{ext}}^m(a, q, \omega)}{1 - \omega_p^2 qaI_m(qa)K_m(qa)/\omega^2 + 4\pi\chi_d^0(\omega) [1 - qaI_m(qa)K_m(qa)]}.
\]

The localized surface plasmons frequencies are given by the poles of Eq. (18):

\[
\omega_q^m = \omega_p \sqrt{\frac{qaI_m(qa)K_m(qa)}{1 + 4\pi\text{Re}[\chi_d^0(\omega)] [1 - qaI_m(qa)K_m(qa)]}}.
\]

3. Metal nanosphere

The localized plasmons are studied for a metal nanosphere with the radius \( a \) in a dielectric as shown Fig. 3. The local dielectric function of the nanosphere and that of the dielectric are assumed to have the following step function shapes:

\[
\varepsilon_m(r_1, \omega) = 1 - \frac{\omega_p^2}{\omega^2} \theta(a - r_1), \\
\varepsilon_d(r_1, \omega) = 1 + 4\pi\chi_d^0(\omega) \theta(r_1 - a).
\]
The scalar potential and the Coulomb potential in Eq. (9) are expanded using polar coordinates as

\[ \varphi_{\text{ext}}(r, \omega) = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \varphi_{\text{ext}}^{lm}(r, \omega) Y_{lm}(\theta, \phi), \]

\[ \frac{1}{|r - r'|} = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \frac{4\pi}{2l+1} Y_{lm}(\theta, \phi) Y_{lm}^* (\theta_1, \phi_1) \frac{r_1^l r_>^l}{r_<^l}, \]

where \( Y_{lm}(\theta, \phi) \) is the normalized spherical harmonics, \( r_< = \min(r, r_1) \), and \( r_> = \max(r, r_1) \).

After some calculations described in Appendix B, the effective scalar potential at \( r = a \) is given by

\[ \varphi_{\text{eff}}^{lm}(a, \omega) = \frac{\varphi_{\text{ext}}^{lm}(a, \omega)}{(1 - \omega_0^2/\omega^2)^l/(2l+1) + (l+1)\varepsilon_0^a(\omega)/(2l+1)}, \]

\[ \varepsilon_0^a(\omega) = 1 + 4\pi \chi_0^a(\omega). \]

The localized surface plasmon frequencies are derived by the poles of Eq. (22):

\[ \omega_l = \omega_p \sqrt{l/(l + (l+1)\text{Re}[\varepsilon_0^a(\omega)])}. \]

B. Light emissions from metal nanostructures in dielectrics

Using the induced charge density in Eq. (6), the electric dipole moment is given using integration by parts by [26]

\[ p(\omega) = \int r \rho(r, \omega) dr = - \int dr \left[ \frac{\varepsilon_0^a(r)}{m_0\omega^2} - \chi^a(r, \omega) \right] E(r, \omega). \]

As the vector potential term \( \omega A_{\text{eff}}(r, \omega) \) in the electric field \( E(r, \omega) \) in Eq. (3) can be ignored for metal nanostructures as reported previously [28], the dipole moment is approximately given by

\[ p(\omega) \approx \int dr \left[ \frac{\omega_p^2(r)}{4\pi \omega^2} - \chi^a(r, \omega) \right] \nabla \varphi_{\text{eff}}(r, \omega) = - \int dr \varphi_{\text{eff}}(r, \omega) \nabla \left[ \frac{\omega_p^2(r)}{4\pi \omega^2} - \chi^a(r, \omega) \right], \]

\[ \tilde{p}(\omega) = -\omega^2 p(\omega) = \int dr \varphi_{\text{eff}}(r, \omega) \nabla \left[ \frac{\omega_p^2(r)}{4\pi} - \omega^2 \chi^a(r, \omega) \right], \]

where \( \tilde{p}(\omega) \) is the second order derivative with respect to time of the dipole moment.

Considering the case where metal nanostructures exist in dielectrics without optical absorption, which means that the light emission frequency is far from the optical absorption one in the dielectrics and the electric susceptibility has only real part, the emitted photon number or light emission intensity with energy \( h\omega \) per unit solid angle \( \Omega \) in the \( r \) direction from nanostructures is given in the electric dipole approximation by [26]

\[ I_{\text{ph}}(\Omega, \omega) = \frac{1}{4\pi^2 c^3 h\omega} |e_r \times \tilde{p}(\omega)|^2 = \frac{1}{4\pi^2 c^3 h\omega} \left| e_r \times \int dr \varphi_{\text{eff}}(r, \omega) \nabla \left[ \frac{\omega_p^2(r)}{4\pi} - \omega^2 \chi^a(r, \omega) \right] \right|^2, \]

where \( e_r = r/r \) is the unit vector in the \( r \) direction. When the optical absorption cannot be ignored in the dielectrics, the exponential decrease of the light emission intensity should be considered, which depends on the thickness of the dielectrics, the light emission direction and the frequency of the emitted light.

Using Eq. (26), the emitted photon numbers from the three nanostructures are calculated using the model where the local electron density \( n(r) \) and the electric susceptibility \( \chi^a(r, \omega) \) have step function shapes at the metal and dielectric interfaces as described in Sec. III A.
1. Metal ultrathin layer

Substituting Eqs. (13) and (14) into Eq. (25), \( \mathbf{p}(\omega) \) is given by

\[
\mathbf{p}(\omega) = e_z \lim_{k \to 0} \left\{ \frac{\varphi_{\text{ext}}(k, 0, \omega) - \varphi_{\text{ext}}(k, l, \omega)}{1 + 2\pi \chi_{\|}^2(0)(1 - e^{-kL})} - \omega_p^2/(2\omega^2) \right\} \left( \frac{\omega_p^2 \chi_{\|}^2(\omega)}{4\pi} + \omega^2 \chi_d^0(\omega) \right),
\]

where \( e_z \) is the unit vector in the \( z \) direction. The integration over \( \rho = (x, y) \) plane in Eq. (25) leads to \( k \to 0 \), indicating that only the localized surface plasmon contributes but the propagating surface plasmon with finite \( k \) does not contribute. The antisymmetric mode only contributes to the dipole moment because the symmetric mode does not generate the dipole charge in the \( z \) direction. When the external electric field \( \mathbf{E}_z \) has only \( z \)-component \( E_z \) as shown in Fig. 1, the external scalar potential \( \varphi_{\text{ext}}(0, z, \omega) \) is approximately given by \( \varphi_{\text{ext}}(0, z, \omega) \approx -E_z z \) in the quasi-static approximation, where \( A \) is the area of the layer.

Then the emitted photon number is given using Eq. (26) by

\[
I_{ph}(\Omega, \omega) = \frac{E_z^2 A L^2 \sin^2 \theta}{4\pi^2 c \hbar} \left| \frac{\omega_p^2 \chi_{\|}^2(\omega)}{4\pi} + \omega^2 \chi_d^0(\omega) \right|^2 \left( \frac{\omega^3 + \Gamma^2}{(\omega^2 - \omega_p^2)^2 + \omega^2 \Gamma^2} \right),
\]

where \( \theta \) is the angle between \( e_x \) and \( e_z \), \( 1 - \omega_p^2/\omega^2 \) in Eq. (27) is replaced by \( 1 - \omega_p^2/[\omega(\omega + i\Gamma)] \) to consider the damping of the localized plasmons with a damping frequency \( \Gamma \). The light emission originates from the surface plasmon and the dielectric polarization at the metal ultrathin layer and dielectric interfaces.

The resonant frequency of the emitted light becomes \( \omega_p \), which can be also derived by solving one-dimensional (1D) Laplace equation using the boundary conditions for the electric field and the electric displacement at the ultrathin layer and dielectric interfaces in the quasi-static approximation, where the Drude dielectric function \( 1 - \omega_p^2/\omega^2 \) for the ultrathin layer and the homogenous dielectric function \( \varepsilon_d^0(\omega) \) for the dielectric are used (not shown).

2. Metal nanowire

When the external electric field \( \mathbf{E}_x \) has only \( x \)-component \( E_x \) as shown in Fig. 2, substitution of Eq. (18) into Eq. (25) gives

\[
\mathbf{p}(\omega) = -e_x a \left( \frac{e^2 n_0}{m_e} + \omega^2 \chi_d^0(\omega) \right) \lim_{q \to 0} \sum_{m=-\infty}^{\infty} \varphi_{\text{eff}}^q(a, q, \omega) \int d\phi e^{i m \phi} \cos \phi \]

\[
\mathbf{p}(\omega) = -e_x 2\pi a \left( \frac{e^2 n_0}{m_e} + \omega^2 \chi_d^0(\omega) \right) \text{Re} \left[ \varphi_{\text{eff}}^1(a, 0, \omega) \right],
\]

where \( e_x \) is the unit vector in the \( x \) direction. The term of \( m = 1 \) (electric dipole) only contributes to the dipole moment, which is derived by using the translational invariance in the \( z \)-direction and the mirror symmetry in the \( y \)-direction of the system, giving \( \nabla n(r) = -e_x n_0 \delta(\rho - a) \cos \phi \) and \( \nabla \chi_d(r, \omega) = e_x \chi_d^0(\omega) \delta(\rho - a) \cos \phi \). When the symmetry is broken, the terms having other \( m \) also contribute to the dipole moment.

The integration over \( z \) direction in Eq. (25) leads to \( q \to 0 \), indicating that only the localized surface plasmon contributes but the propagating surface plasmon with finite \( q \) does not contribute. \( \varphi_{\text{eff}}^1(a, 0, \omega) \) is given by

\[
\varphi_{\text{eff}}^1(a, 0, \omega) = \frac{2 \varphi_{\text{ex}}^1(a, 0, \omega)}{[1 + e_d^0(\omega)]} - \omega_p^2/\omega^2,
\]

where the relation \( \lim_{q \to 0} [a\phi(qa)K_1(qa)] = 1/2 \) is used in Eq. (18). When the external scalar potential \( \varphi_{\text{ex}}^1(\rho, 0, \omega) \) is approximately given by \( \varphi_{\text{ex}}^1(\rho, 0, \omega) \approx -E_x z L \) that is derived by \( \varphi_{\text{ex}}^1(\rho, z, \omega) = -E_x z = -E_x \rho \cos \phi \) in the quasi-static approximation and \( L \) is the length of the nanowire, the emitted photon number is given using Eq. (26) by

\[
I_{ph}(\Omega, \omega) = \frac{4 E_x^2 a L^2 \sin^2 \theta}{c^3 \hbar} \left| \frac{\omega_p^2/(4\pi) + \omega^2 \chi_d^0(\omega)}{1 + e_d^0(\omega)} \right|^2 \left( \frac{\omega^3 + \Gamma^2}{(\omega^2 - \omega_p^2)^2 + \omega^2 \Gamma^2} \right),
\]

where \( \theta \) is the angle between \( e_x \) and \( e_z \), and the damping of the localized plasmons is also considered.

The resonant frequency of the emitted light is derived by \( \omega = \omega_p/\sqrt{1 + e_d^0(\omega)} \), which can be also derived by solving two-dimensional (2D) Laplace equation using the boundary conditions for the electric field and the electric.
displacement at the nanowire and dielectric interfaces in the quasi-static approximation, where the Drude dielectric function $1 - \omega_p^2/\omega^2$ for the ultrathin layer and the homogenous dielectric function $\varepsilon_d^0(\omega)$ for the dielectric are used [33].

3. Metal nanosphere

When the external electric field $E_z$ has only z-component $E_z$ as shown in Fig. 3, substitution of Eq. (22) into Eq. (25) gives

$$\mathbf{p}(\omega) = -e_z 2\pi a^2 \left[ \frac{\omega_p^2}{4\pi} + \omega^2 \chi_d^0(\omega) \right] \int_0^\infty \frac{\sin^2 \psi}{\psi} \varphi_{\text{eff}}^1(\omega, \psi) \frac{1}{\psi} d\psi$$

$$= -e_z \left( \frac{4\pi}{3} \right)^{1/2} a^2 \left[ \frac{\omega_p^2}{4\pi} + \omega^2 \chi_d^0(\omega) \right] \varphi_{\text{eff}}^1(\omega, \psi),$$

where $e_z$ is the unit vector in the z direction, $P_l^0(\cos \theta)$ is the associated Legendre function and the orthogonal property of $P_l^0(\cos \theta)$ is used because of $P_l^0(\cos \theta) = \cos \theta$, indicating that only the term of $l = 1$ (electric dipole) contributes the dipole moment. The rotational symmetry around the z-axis is also used, giving $m = 0$, $\nabla \psi = -e_z n_0 \delta(r - a) \cos \theta$, and $\nabla \chi_d(r, \omega) = e_z \chi_d^0(\omega) \delta(r - a) \cos \theta$. When the symmetry is broken, the terms having other $m$ and $l$ also contribute to the dipole moment.

When the external scalar potential is given by $\varphi_{\text{ext}}^1(a, \omega) = -(4\pi/3)^{1/2} E_0 a$ that is derived by $\varphi_{\text{ext}}^1(r, \omega) = -E_z r \cos \theta$ in the quasi-static approximation, the emitted photon number is given using Eq. (22) at $m = 0$, $l = 1$ and Eq. (26) by

$$I_{ph}(\Omega, \omega) = \frac{4d^6 E_z^2 \sin^2 \theta}{c^3 h} \left[ \frac{\omega_p^2/(4\pi) + \omega^2 \chi_d^0(\omega)}{1 + 2\varepsilon_d^0(\omega)} \right]^2 \frac{\omega^3 + \omega^4}{\{\omega^2 - \omega_p^2/[1 + 2\varepsilon_d^0(\omega)]\}^2 + \omega^2 \Gamma^2},$$

where $\theta$ is the angle between $e_r$ and $e_z$, the damping of the localized plasmons is also considered.

The resonant frequency of the emitted light is derived by $\omega = \omega_p/\sqrt{1 + 2\varepsilon_d^0(\omega)}$, which can be also derived by solving three-dimensional (3D) Laplace equation using the boundary conditions for the electric field and the electric displacement at the nanosphere and dielectric interfaces in the quasi-static approximation, where the Drude dielectric function $1 - \omega_p^2/\omega^2$ for the ultrathin layer and the homogenous dielectric function $\varepsilon_d^0(\omega)$ for the dielectric are used [1].

The resonant frequencies of the light emissions from the localized surface plasmons for the ultrathin layers, nanowires and nanospheres in vacuum [$\varepsilon_0^0(\omega) = 1$] are found to be expressed by $\omega_p/\sqrt{D}$, where $D$’s are the electron confinement dimensions for the ultrathin layer (one dimension: $D = 1$), nanowire (two dimension: $D = 2$) and nanosphere (three dimension: $D = 3$). This indicates that the larger induced charges produce the larger electric fields in the metal nanostructures with the smaller confinement dimensions, which produce the larger surface plasmon frequencies.

The results in this section were derived using the first order approximation; the quasi-static one that is valid when the nanostructure dimension is much smaller than the wavelength of light and the phase change of local electric field can be ignored, of those derived by the classical electrodynamic approach such as Mie theory [1, 34, 35] where the phase change of local electric field caused by the retardation effect is properly considered in a single metal structure. In order to consider the retardation effect on the localized plasmons in our approach, Eqs. (6) and (8) or Eq. (A1) should be solved instead of Eq. (9), which is a future subject to be studied.

IV. CONCLUSIONS

A theory of localized bulk and surface plasmons for metal nanostructures in dielectrics was derived in the random phase approximation at the high frequency condition. The induced charge density in dielectrics was added to the induced charge density by the plasmons, which appeared in the previously reported integral equation for the scalar potential [26, 27]. In the quasi-static approximation, the integral equation could be analytically solved using a model where the local dielectric functions had step function shapes at the interface between the metal nanostructures and dielectrics. The localized surface plasmon frequencies and the light emission intensities were then derived for the metal ultrathin layer, nanowire, and nanosphere.

The results reported here are limited to the single metal nanostructures. The present formula can be developed to study the localized plasmons for multiple metal nanostructures in dielectrics using the structural Green’s function method [36], which has been already applied to study the localized plasmons for multiple metal nanostructures in vacuum [27]. This will be reported in future study.

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Appendix A

The operation of \((\nabla^2 + \omega^2/c^2)\) on both sides of Eqs. (6) and (8) give the following partial differential equations for the effective scalar and vector potentials using the relation \((\nabla^2 + \omega^2/c^2)G_0(r - r_1, \omega) = -4\pi\delta(r - r_1)\):

\[
\begin{align*}
\left(\nabla^2 + \frac{\omega^2}{c^2}\right)\varphi_{\text{eff}}(r, \omega) &= -4\pi[p(r, \omega) + \rho_{\text{ext}}(r, \omega)], \\
\left(\nabla^2 + \frac{\omega^2}{c^2}\right)A_{\text{eff}}(r, \omega) &= \frac{4\pi}{c}[j(r, \omega) + j_{\text{ext}}(r, \omega)],
\end{align*}
\]

where \(\rho_{\text{ext}}(r, \omega)\) and \(j_{\text{ext}}(r, \omega)\) are the external charge and current densities, respectively. These partial differential equations are useful to perform numerical calculations for complex metal structures in dielectrics (not limited to metal nanostructures), where the retardation effect cannot be ignored.

Equation (A1) for the effective scalar potential can be transformed into the following equation using the Lorentz condition \(\nabla \cdot A_{\text{eff}}(r, \omega) = i\frac{\omega}{c}\varphi_{\text{eff}}(r, \omega)\):

\[
\nabla \cdot \{[\varepsilon_m(r, \omega) + \varepsilon_d(r, \omega) - 1]E(r, \omega)\} = \nabla \cdot D(r, \omega) = 4\pi\rho_{\text{ext}}(r, \omega).
\]

Then, Eq. (A2) gives the following relation:

\[
D(r, \omega) = [\varepsilon_m(r, \omega) + \varepsilon_d(r, \omega) - 1]E(r, \omega).
\]

The above equation reveals that the equations of Eq. (A1) are equivalent to the Maxwell equations using the local dielectric function \(\varepsilon_m(r, \omega) + \varepsilon_d(r, \omega) - 1\).

Then, using this dielectric function and the Maxwell equations without external charges, i.e., \(\nabla \cdot D(r, \omega) = 0\), the following partial differential equations are derived for the electric and magnetic fields:

\[
\begin{align*}
\nabla^2 E(r, \omega) + \frac{\omega^2}{c^2}[\varepsilon_m(r, \omega) + \varepsilon_d(r, \omega) - 1]E(r, \omega) &= \nabla \left\{\frac{E(r, \omega) \cdot \nabla [\varepsilon_m(r, \omega) + \varepsilon_d(r, \omega)]}{\varepsilon_m(r, \omega) + \varepsilon_d(r, \omega) - 1}\right\}, \\
\nabla^2 H(r, \omega) + \frac{\omega^2}{c^2}[\varepsilon_m(r, \omega) + \varepsilon_d(r, \omega) - 1]H(r, \omega) &= i\frac{\omega}{c}\nabla [\varepsilon_m(r, \omega) + \varepsilon_d(r, \omega)] \times E(r, \omega),
\end{align*}
\]

where the relation \(B(r, \omega) = H(r, \omega) = \text{rot}A_{\text{eff}}(r, \omega)\) was used for non-magnetic metals and dielectrics \((\mu = 1)\). The above equations can be used to any metal structures (not limited to metal nanostructures), which coincide with the previously reported ones when \(\varepsilon_d(r, \omega) = 1\) [28].

When the local dielectric functions are nearly constant, the right side terms of Eqs. (A4) and (A5) become almost zeros. Then the equations become the conventional Maxwell equations using the classical Drude dielectric function \(1 - \omega_p^2/\omega^2\) and the homogeneous dielectric function \(\varepsilon_0^0(\omega)\), which can be numerically calculated by using, for example, the finite-difference time-domain (FDTD) method [37, 38].

Appendix B

1. Metal ultrathin layer

Substituting Eqs. (11) and (12) into Eq. (9), the following equations are derived using the relation \(d\theta(z_1)/dz_1 = \delta(z_1)\), etc.:

(1) Outside the layer: \(z < 0\),

\[
[1 + 4\pi\chi_0^0(\omega)]\varphi_{\text{eff}}(k, z, \omega) - \left[\frac{\omega_p^2}{2\omega^2} + 2\pi\chi_0^0(\omega)\right] e^{kz} \varphi_{\text{eff}}(k, 0, \omega) + \left[\frac{\omega_p^2}{2\omega^2} + 2\pi\chi_0^0(\omega)\right] e^{k(z-1)} \varphi_{\text{ext}}(k, l, \omega) = \varphi_{\text{ext}}(k, z, \omega).
\]

(B1)

(2) Inside the layer: \(0 \leq z \leq l\),
\[
\left(1 - \frac{\omega_p^2}{\omega^2}\right) \phi_{\text{eff}}(k, z, \omega) + \left[\frac{\omega_p^2}{2\omega^2} + 2\pi \chi_d^0(\omega)\right] e^{-kz} \phi_{\text{eff}}(k, 0, \omega) + \left[\frac{\omega_p^2}{2\omega^2} + 2\pi \chi_d^0(\omega)\right] e^{k(z-l)} \phi_{\text{eff}}(k, l, \omega) = \phi_{\text{ext}}(k, z, \omega).
\]

(B2)

(3) Outside the layer: \( z > l \),

\[
[1 + 4\pi \chi_d^0(\omega)] \phi_{\text{eff}}(k, z, \omega) + \left[\frac{\omega_p^2}{2\omega^2} + 2\pi \chi_d^0(\omega)\right] e^{-kz} \phi_{\text{eff}}(k, 0, \omega) - \left[\frac{\omega_p^2}{2\omega^2} + 2\pi \chi_d^0(\omega)\right] e^{-k(z-l)} \phi_{\text{eff}}(k, l, \omega) = \phi_{\text{ext}}(k, z, \omega).
\]

(B3)

Equation (B2) coincides with Eq. (B1) at \( z = 0 \) and Eq. (B3) at \( z = l \), indicating that the bulk plasmon contribution of the first term in the left side of Eq. (B2) disappears at the interfaces between the metal and dielectric by the surface plasmon shielding effect [25–27]. Putting \( z = 0 \) and \( z = l \) in Eq. (B2), the scalar potentials in Eqs. (13) and (14) are derived.

2. Metal nanowire

Substituting Eqs. (16) and (17) into Eq. (9), the following equations are derived using the relation \( d\theta(a - r_1)/dr_1 = -\delta(r_1 - a) \), etc.:

(1) Outside the nanowire: \( \rho > a \),

\[
[1 + 4\pi \chi_d^0(\omega)] \phi_{\text{eff}}(\rho, q, \omega) - qa I'_m(qa) K_m(qa) \left[\frac{\omega_p^2}{\omega^2} + 4\pi \chi_d^0(\omega)\right] \phi_{\text{eff}}(a, q, \omega) = \phi_{\text{ext}}(\rho, q, \omega).
\]

(B4)

(2) Inside the nanowire: \( \rho \leq a \),

\[
\left[1 - \frac{\omega_p^2}{\omega^2}\right] \phi_{\text{eff}}(\rho, q, \omega) - qa I_m(qa) K'_m(qa) \left[\frac{\omega_p^2}{\omega^2} + 4\pi \chi_d^0(\omega)\right] \phi_{\text{eff}}(a, q, \omega) = \phi_{\text{ext}}(\rho, q, \omega).
\]

(B5)

where \( I'_m(s) \) and \( K'_m(s) \) are the first-order derivatives of \( I_m(s) \) and \( K_m(s) \).

Using the relation \( I_m(s)K'_m(s) - I'_m(s)K_m(s) = -1/s \), Eq. (B5) coincides with Eq. (B4) at \( \rho = a \), indicating that the bulk plasmon contribution disappears at the interfaces between the metal and dielectric by the surface plasmon shielding effect. Putting \( \rho = a \) in Eq. (B4), the scalar potential in Eq. (18) is derived.

3. Metal nanosphere

Substituting Eqs. (20) and (21) into Eq. (9), the following equations are derived using the relation \( d\theta(a - r_1)/dr_1 = -\delta(r_1 - a) \), etc.:

(1) Outside the nanosphere: \( r > a \),

\[
[1 + 4\pi \chi_d^0(\omega)] \phi_{\text{eff}}(r, \omega) - \frac{l}{2l + 1} \left(\frac{a}{r}\right)^{l+1} \left[\frac{\omega_p^2}{\omega^2} + 4\pi \chi_d^0(\omega)\right] \phi_{\text{ext}}(a, \omega) = \phi_{\text{ext}}(r, \omega).
\]

(B6)

(2) Inside the nanosphere: \( r \leq a \),

\[
\left[1 - \frac{\omega_p^2}{\omega^2}\right] \phi_{\text{eff}}(r, \omega) + \frac{l + 1}{2l + 1} \left(\frac{r}{a}\right)^l \left[\frac{\omega_p^2}{\omega^2} + 4\pi \chi_d^0(\omega)\right] \phi_{\text{ext}}(a, \omega) = \phi_{\text{ext}}(r, \omega).
\]

(B7)

Equation (B7) coincides with Eq. (B6) at \( r = a \), indicating that the bulk plasmon contribution disappears at the interfaces between the metal and dielectric by the surface plasmon shielding effect. Putting \( r = a \) in Eq. (B6), the scalar potential in Eq. (22) is derived.
