**Differential Equations for Localized Plasmons in the Random Phase Approximation**

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Differential equations are derived for localized bulk and surface plasmons in metal nanostructures in the random phase approximation (RPA) at the high frequency condition. A differential equation for the scalar potential in the RPA gives a position-dependent Drude-like dielectric function. Using the dielectric function, a differential equation for the vector potential is derived in the Lorentz gauge. A corrected RPA differential equation for the scalar potential is then derived by using the differential equation for the vector potential and the Lorentz condition. The vector potential contribution to the electric field in the Lorentz gauge is found negligible compared with the scalar potential one in metal nanostructures. This indicates that the scalar potential plays important roles in analyzing the localized plasmons in metal nanostructures as reported previously. The corrected RPA differential equations are found equivalent to the Maxwell equations using the position-dependent Drude-like dielectric function.

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I. INTRODUCTION

Surface plasmons can couple with photons to produce collective excitations called localized surface plasmons (LSPs) [1–4]. LSPs can concentrate optical waves into regions that are much smaller than their free space radiation wavelengths. They can also much enhance their local electric fields near metal nanostructures at the surface plasmon excitation energies. These effects can use LSPs to fabricate nanoscale photonic devices and to perform optical measurements with extremely high detection sensitivity.

It has recently been reported that 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 optical effects and the quantum tunneling effects between the metal nanostructures [5–12].

A new field called graphene plasmonics has also been 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, 14]. Topological plasmonics in graphene have been very recently proposed, which show a possibility to develop topologically robust chiral plasmonic devices [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]. Thus the plasmonics area is still interesting and developing.

Theoretical studies of the interaction between the external electric field or the incident electrons and the surface plasmon have been reported [20–24]. However, in these studies, the coupling phenomena between bulk and surface plasmons, which play important roles in the plasmon excitation process, have not been sufficiently studied. The coupling phenomena are caused by the many-body interaction between the electrons near the surfaces.

The author has developed a theory of localized plasmons using the random phase approximation (RPA) at high frequency conditions, where the coupling phenomena are properly considered in metal nanostructures [25]. In this theory, the local electron density in the metal nanostructures plays an essential role in the plasmon excitation. A theory has also been developed to calculate the light emission intensity from a single metal nanostructure by considering the retardation of the scalar potentials for localized plasmons [26]. Then, the theory has been developed to investigate localized plasmon excitations for multiple metal nanostructures [27]. These theories, the integral equation formulas are mostly used, which are not very convenient for numerical calculations. Differential equation formulas are more useful to investigate localized plasmons in complex metal nanostructures.

In this study, differential equations for localized plasmons are derived in the RPA at the high frequency condition by developing our previous theories in the Lorentz gauge. The validity of the RPA differential equations is then investigated for metal nanostructures. The corrected RPA differential equations are finally compared with the Maxwell equations.

II. DIFFERENTIAL EQUATIONS FOR LOCALIZED PLASMONS

A. Scalar potential and dielectric function in the RPA

Considering an effective scalar potential $\varphi_{\text{eff}}(\mathbf{r}, \omega)$ and an external scalar potential $\varphi_{\text{ext}}(\mathbf{r}, \omega)$ within the electron gas in metal nanostructures at frequency $\omega$, previous studies [25, 26] have shown that they satisfy the following integral equation in the RPA under the high frequency condition, i.e., the plasmon excitation condition:

$$
\varphi_{\text{eff}}(\mathbf{r}, \omega) = \varphi_{\text{ext}}(\mathbf{r}, \omega) + \frac{e^2}{m_e \omega^2} \int d\mathbf{r}_1 n(\mathbf{r}_1) \nabla_1 G_0(\mathbf{r} - \mathbf{r}_1, \omega) \times \nabla_1 \varphi_{\text{eff}}(\mathbf{r}_1, \omega)
$$

(1)

$$
n(\mathbf{r}_1) = 2 \sum_n |\psi_n(\mathbf{r}_1)|^2 \delta(E_F - E_n),
$$

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where \( \psi_n(\mathbf{r}_1) \) is the normalized single-electron wave function in the metal nanostructure, \( \theta \) is the step function, and \( E_F \) and \( E_n \) are the Fermi energy and the single electron energy, respectively. \( n(\mathbf{r}_1) \) is the local electron density in the metal nanostructure, \( m_e \), \( -e \) are the electron mass and charge, and \( \nabla_1 \) represents the gradient with respect to \( \mathbf{r}_1 \), \( G_0(\mathbf{r} - \mathbf{r}_1, \omega) \) is the retarded Green’s function of the Helmholtz equation in Gaussian units. The derivation of Eq. (1) is briefly described in Appendix.

An integration by parts of Eq. (1) with respect to \( \mathbf{r}_1 \) gives

\[
\varphi_{\text{eff}}(\mathbf{r}, \omega) = \varphi(\mathbf{r}, \omega) + \int d\mathbf{r}_1 G_0(\mathbf{r} - \mathbf{r}_1, \omega) \nabla_1 \cdot [n(\mathbf{r}_1)\nabla \varphi_{\text{eff}}(\mathbf{r}_1, \omega)],
\]

where \( \rho(\mathbf{r}_1, \omega) \) is defined as the induced electric charge density, and is given by

\[
\rho(\mathbf{r}_1, \omega) = -\frac{e^2}{m_e \omega^2} \nabla_1 \cdot [n(\mathbf{r}_1)\nabla \varphi_{\text{eff}}(\mathbf{r}_1, \omega)].
\]

The operation of \( (\nabla^2 + \omega^2/c^2) \) on both sides of Eq. (2) and the use of Eq. (A4) give the following differential equation for the effective scalar potential:

\[
(\nabla^2 + \omega^2/c^2) \varphi_{\text{eff}}(\mathbf{r}, \omega) = -4\pi \{ \rho(\mathbf{r}, \omega) + \rho_{\text{ext}}(\mathbf{r}, \omega) \},
\]

where \( \rho_{\text{ext}}(\mathbf{r}, \omega) \) is the external charge density.

Considering the quasi-static approximation; \( \omega/c \to 0 \), the following equation is derived from Eq. (4):

\[
\nabla \cdot \left\{ 1 - \frac{4\pi e^2}{m_e \omega^2} n(\mathbf{r}) \right\} \mathbf{E}(\mathbf{r}, \omega) = D(\mathbf{r}, \omega) = 0,
\]

\[
\nabla \cdot \mathbf{D}(\mathbf{r}, \omega) = 4\pi \rho_{\text{ext}}(\mathbf{r}, \omega),
\]

where \( \mathbf{E}(\mathbf{r}, \omega) \) and \( \mathbf{D}(\mathbf{r}, \omega) \) are the electric field and the electric displacement, respectively. Then the following position-dependent Drude-like dielectric function is derived from Eq. (5) as reported previously [25]:

\[
\varepsilon(\mathbf{r}, \omega) = \left[ 1 - \frac{\omega_p^2(\mathbf{r})}{\omega^2} \right],
\]

where \( \omega_p(\mathbf{r}) \) is considered to be the position-dependent bulk plasmon frequency.

In the quasi-static approximation; \( \omega/c \approx k \to 0 \), the wave number \( k \) of the incident electric field is much smaller than the Fermi wave number \( k_F \) of the electrons. In this case, the bulk plasmon excitation energy is much larger than the single electron excitation energy at the wave number \( k \) and the exchange-correlation interactions between the electrons are not significant at \( k \ll k_F \) since the size of the exchange-correlation holes is comparable to the inverse of \( k_F \). The above analysis indicates that the position dependent dielectric function \( \varepsilon(\mathbf{r}, \omega) \) using the position-dependent bulk plasmon frequency \( \omega_p(\mathbf{r}) \) is justified in the quasi-static approximation.

## B. Differential equations for scalar and vector potentials in the Lorentz gauge

The Maxwell equations for the electromagnetic fields \( \mathbf{E}(\mathbf{r}, \omega) \) and \( \mathbf{B}(\mathbf{r}, \omega) \) can be represented by the effective scalar potential \( \varphi_{\text{eff}}(\mathbf{r}, \omega) \) and the vector potential \( \mathbf{A}(\mathbf{r}, \omega) \). When the Lorentz gauge and Gaussian units are used, the electromagnetic fields after the Fourier transformation for \( \omega \) are given by

\[
\mathbf{E}(\mathbf{r}, \omega) = -\nabla \varphi_{\text{eff}}(\mathbf{r}, \omega) + \frac{i\omega}{c} \mathbf{A}(\mathbf{r}, \omega),
\]

\[
\mathbf{B}(\mathbf{r}, \omega) = \mathbf{rot} \mathbf{A}(\mathbf{r}, \omega),
\]

with the Lorentz condition:

\[
\nabla \cdot \mathbf{A}(\mathbf{r}, \omega) = i\frac{\omega}{c} \varphi_{\text{eff}}(\mathbf{r}, \omega).
\]

Substituting Eqs. (6), (7) and (8) into the following Maxwell equations,

\[
\mathbf{rot} \mathbf{H}(\mathbf{r}, \omega) = \frac{4\pi}{c} \mathbf{J}_{\text{ext}}(\mathbf{r}, \omega) - \frac{i\omega}{c} \mathbf{D}(\mathbf{r}, \omega),
\]

\[
\mathbf{B}(\mathbf{r}, \omega) = \mu \mathbf{H}(\mathbf{r}, \omega),
\]

the following differential equation is derived for the vector potential:

\[
(\nabla^2 + \omega^2/c^2) \mathbf{A}(\mathbf{r}, \omega) = -\frac{4\pi}{c} \left[ \mathbf{J}_L(\mathbf{r}, \omega) + \mathbf{J}_{\text{ext}}(\mathbf{r}, \omega) \right],
\]

\[
\mathbf{J}_L(\mathbf{r}, \omega) = i\frac{e^2}{\omega m_e} \left[ -\nabla \varphi_{\text{eff}}(\mathbf{r}, \omega) + \frac{i\omega}{c} \mathbf{A}(\mathbf{r}, \omega) \right] = \sigma(\mathbf{r}, \omega) \mathbf{E}(\mathbf{r}, \omega),
\]

\[
\sigma(\mathbf{r}, \omega) = i\frac{e^2}{\omega m_e},
\]

where \( \mathbf{J}_L(\mathbf{r}, \omega) \) is the current density induced by the plasmon excitations, \( \mathbf{J}_{\text{ext}}(\mathbf{r}, \omega) \) is the external current density and \( \sigma(\mathbf{r}, \omega) \) is the position-dependent electric conductivity. Non-magnetic metals (\( \mu = 1 \)) are considered in this study. Equation (10) becomes the same as Eq. (17) given in our previous article [26], when the term related to \( \omega_p^2(\mathbf{r})/\omega^2 \) in \( \varepsilon(\mathbf{r}, \omega) \) is moved to the right side of the equation.

The vector potential \( \mathbf{A}(\mathbf{r}, \omega) \) in Eq. (10) and the effective scalar potential \( \varphi_{\text{eff}}(\mathbf{r}, \omega) \) in Eq. (4), however, do not satisfy the Lorentz condition of Eq. (8) by the vector potential term \( i\omega \mathbf{A}(\mathbf{r}, \omega)/c \) in \( \mathbf{J}_L(\mathbf{r}, \omega) \) of Eq. (10). In order to get the scalar potential that satisfies the Lorentz condition, taking the divergence of both sides of Eq. (10) and using the Lorentz condition of Eq. (8), the following differential equation is derived for the corrected scalar potential \( \varphi_{\text{eff}}(\mathbf{r}, \omega) \):
\[ \left( \nabla^2 + \frac{\omega^2}{c^2} \right) \phi_{\text{eff}}^L(r, \omega) = -4\pi \left[ \rho_L(r, \omega) + \rho_{\text{ext}}(r, \omega) \right], \]
\[
\rho_L(r, \omega) = \frac{e^2}{m_\omega} \omega \rho_{\text{ext}}(r, \omega) \times \nabla \cdot \left\{ n(r) \left[ -\nabla \phi_{\text{eff}}^L(r, \omega) + \frac{i\omega}{c} A(r, \omega) \right] \right\} \]
\[
= \frac{e^2}{m_\omega} \omega \nabla \cdot \left[ n(r) E(r, \omega) \right],
\] 
where \( \rho_L(r, \omega) \) is the corrected induced charge density and the external charge conservation law; \( \nabla \cdot \mathbf{j}_{\text{ext}}(r, \omega) - i\omega \rho_{\text{ext}}(r, \omega) = 0 \) was used. The corrected induced charge density \( \rho_L(r, \omega) \) and the induced current density \( \mathbf{j}_L(r, \omega) \) satisfy also the charge conservation law; \( \nabla \cdot \mathbf{j}_L(r, \omega) - i\omega \rho_L(r, \omega) = 0 \).

Equations (10) and (11) are the differential equations for localized plasmons in the corrected RPA. These are useful to perform numerical calculations for complex metal structures, which are not limited to metal nanostructures, by using the finite element method or the finite difference method because the electric field has the scalar and vector potential terms in the Lorentz gauge. The corrected scalar potential \( \phi_{\text{eff}}^L(r, \omega) \) and the vector potential \( \mathbf{A}(r, \omega) \) should have the following asymptotic values at \( |r| \to \infty \):

\[
\phi_{\text{eff}}^L(r, \omega) = \phi_{\text{ext}}(r, \omega), \quad \mathbf{A}(r, \omega) = \mathbf{A}_{\text{ext}}(r, \omega), \quad \nabla \cdot \mathbf{A}_{\text{ext}}(r, \omega) = \frac{i\omega}{c} \phi_{\text{ext}}(r, \omega).
\]

\[ (12) \]

C. Validity of the RPA differential equations

The corrected induced charge density in Eq. (11) indicates that the vector potential term \( i\omega \mathbf{A}(r, \omega) / c \) is added to the charge density in Eq. (3). The vector potential term is accompanied with the local electron density \( n(r) \), indicating that the contribution of this term can be analyzed for the metal nanostructures. When a plane wave-like electric field \( \mathbf{E}_z \) is incident on a nanostructure with the size \( a \), the following scalar and vector potentials are approximately given in the dipole approximation by

\[
\phi_{\text{eff}}^L(r, \omega) \approx -E_z z, \quad \mathbf{A}(r, \omega) \approx \frac{i\omega}{2c} E_z z \mathbf{z},
\]

where the Lorentz condition of Eq. (8) was used to get the vector potential. Then, the electric field in Eq. (11) is given by

\[ \mathbf{E}(r, \omega) = E_z \left( 1 + \frac{\omega^2 z^2}{2c^2} \right) \approx E_z \left[ 1 + \frac{(ka)^2}{2} \right], \]

where \( \omega \approx ck \) and \( k \) is the wave number of the incident electric field.

When the condition \( ka \ll 1 \) is satisfied, indicating that the wavelength of the electric field is much larger than the size of the nanostructures, the vector potential contribution to the electric field, i.e. the second term in Eq. (14) can be ignored in metal nanostructures. The same conclusion was derived for the electric fields produced by the electric dipole moments reported previously [26, 27] (not shown). Then, using Eqs. (10) and (11), the differential equations are approximately given for metal nanostructures by

\[
\begin{align*}
\left[ \nabla^2 + \frac{\omega^2}{c^2} \right] \phi_{\text{eff}}(r, \omega) & = -4\pi \left[ \rho(r, \omega) + \rho_{\text{ext}}(r, \omega) \right], \\
\left[ \nabla^2 + \frac{\omega^2}{c^2} \right] \mathbf{A}(r, \omega) & = -4\pi \frac{e}{c} \left[ \mathbf{j}(r, \omega) + \mathbf{j}_{\text{ext}}(r, \omega) \right],
\end{align*}
\]

where \( \rho(r, \omega) \) and \( \mathbf{j}(r, \omega) \) are given by

\[
\begin{align*}
\rho(r, \omega) & = -\frac{e^2}{m_\omega} \nabla \cdot \left[ n(r) \nabla \phi_{\text{eff}}(r, \omega) \right], \\
\mathbf{j}(r, \omega) & = -\frac{ie^2 n(r)}{\omega m_e} \nabla \phi_{\text{eff}}(r, \omega).
\end{align*}
\]

The above differential equations are useful for numerical calculations because the scalar potential can be independently calculated without considering the vector potential term as reported previously [26, 27]. The scalar and vector potentials in Eqs. (15) and (16) satisfy the Lorentz condition. The induced charge and current densities in Eqs. (17) and (18) satisfy also the charge conservation law; \( \nabla \cdot \mathbf{j}(r, \omega) - i\omega \rho(r, \omega) = 0 \).

D. Relationship between the corrected RPA differential and Maxwell equations

Here the corrected RPA differential equations that consider the vector potential term in the electric field are compared with the Maxwell equations. The equation (11) can be transformed into the following equation:

\[ \nabla \cdot \left[ \varepsilon(r, \omega) \mathbf{E}(r, \omega) \right] = \nabla \cdot \mathbf{D}(r, \omega) = 4\pi \rho_{\text{ext}}(r, \omega), \]

where \( \varepsilon(r, \omega) \) is the position-dependent Drude-like dielectric function in Eq. (6) but \( \mathbf{E}(r, \omega) \) is the electric field with the vector potential term in the Lorentz gauge, not that in Eq. (5). Then, Eq. (19) gives the following relation:

\[ \mathbf{D}(r, \omega) = \varepsilon(r, \omega) \mathbf{E}(r, \omega). \]

The above equation reveals that the corrected RPA differential equations in Eqs. (10) and (11) are equivalent to the Maxwell equations using the position-dependent Drude-like dielectric function \( \varepsilon(r, \omega) \).

Then, using the dielectric function in Eq. (20) and the Maxwell equations without external charges, i.e. \( \nabla \cdot \mathbf{D}(r, \omega) = 0 \), the following differential equations are derived:

\[
\begin{align*}
\nabla^2 & \mathbf{E}(r, \omega) + \frac{\omega^2}{c^2} \varepsilon(r, \omega) \mathbf{E}(r, \omega) \\
& = -\frac{4\pi e^2}{m_\omega c^2} \nabla \left\{ \mathbf{E}(r, \omega) \cdot \nabla n(r) \right\},
\end{align*}
\]

\[
\begin{align*}
\nabla^2 & \mathbf{H}(r, \omega) + \frac{\omega^2}{c^2} \varepsilon(r, \omega) \mathbf{H}(r, \omega) \\
& = -\frac{4\pi e^2}{cm_\omega c^2} \nabla n(r) \times \mathbf{E}(r, \omega).
\end{align*}
\]
The causality of the one-particle Green’s function for a single electron. $\psi_n(\mathbf{r})$ is the normalized single-electron wave function in the metal nanostructure, which satisfies the following Schrödinger equation:

$$
\left[ -\frac{\hbar^2}{2m_e} \nabla^2 + V(\mathbf{r}) \right] \psi_n(\mathbf{r}) = E_n \psi_n(\mathbf{r}).
$$

In Eqs. (A2) and (A3), $\theta(E_F - E_n)$ is the step function, $E_F$ is the Fermi energy, $m_e$ is the electron mass and $E_n$ is the single-electron energy in the nanostructure with the local potential energy $V(\mathbf{r})$. $G_0(\mathbf{r}_3 - \mathbf{r}_1, \omega)$ is the retarded Green’s function, which satisfies the following Helmholtz equation in Gaussian units:

$$
\left( \nabla^2 + \frac{\omega^2}{c^2} \right) G_0(\mathbf{r} - \mathbf{r}_1, \omega) = -4\pi \delta(\mathbf{r} - \mathbf{r}_1),
$$

where $c$ is the light velocity and $\delta(\mathbf{r} - \mathbf{r}_1) \equiv \delta(x - x_1)\delta(y - y_1)\delta(z - z_1)$. The vector potential contribution to the electron interaction can be neglected because the electron interactions via the scalar and the vector potentials are approximately given by $e^2G_0(\mathbf{r}_3 - \mathbf{r}_1, \omega)$ and $e^2G_0(\mathbf{r}_3 - \mathbf{r}_1, \omega)(v_p/c)^2$, respectively, where $v_p/c \approx 5 \times 10^{15}$, $c \approx 3.0 \times 10^8$ ms$^{-1}$ and $v_p$ is the Fermi velocity of the electrons in metal nanostructures, for example, $v_p \approx 1.4 \times 10^6$ ms$^{-1}$ for Ag metal.

An induced scalar potential $\varphi_{\text{ind}}(\mathbf{r}, \omega)$ in the electron gas, which is induced by an external scalar potential $\varphi_{\text{ext}}(\mathbf{r}, \omega)$, is given by

$$
\varphi_{\text{ind}}(\mathbf{r}, \omega) = e^2 \int \int d\mathbf{r}_1 d\mathbf{r}_2 G_0(\mathbf{r} - \mathbf{r}_1, \omega) \times P(\mathbf{r}_1, \mathbf{r}_2, \omega) \varphi_{\text{ext}}(\mathbf{r}_2, \omega).
$$

Then, an effective scalar potential $\varphi_{\text{eff}}(\mathbf{r}, \omega)$ that is induced by the external scalar potential satisfies

$$
\varphi_{\text{eff}}(\mathbf{r}, \omega) = \varphi_{\text{ext}}(\mathbf{r}, \omega) + \varphi_{\text{ind}}(\mathbf{r}, \omega)
$$

$$
= \varphi_{\text{ext}}(\mathbf{r}, \omega) + e^2 \int \int d\mathbf{r}_1 d\mathbf{r}_2 \times G_0(\mathbf{r} - \mathbf{r}_1, \omega) P(\mathbf{r}_1, \mathbf{r}_2, \omega) \varphi_{\text{eff}}(\mathbf{r}_2, \omega),
$$

where Eq. (A1) is used to derive the above self-consistent integral equation for the effective scalar potential $\varphi_{\text{eff}}(\mathbf{r}, \omega)$. A high-frequency condition of $|E_n - E_{n'}|/h \omega \ll 1$ is considered valid for collective excitations such as plasmons, which have excitation energies that are much larger than single-electron excitation energies. The right side denominator of Eq. (A2) can then be approximated as:

$$
\frac{1}{\omega + (E_n - E_{n'})/\hbar} \approx \frac{1}{\omega} \left[ 1 - \frac{E_n - E_{n'}}{\hbar \omega} \right],
$$

where $\omega$ is assumed to include the infinitesimal imaginary term $i\eta$.

Equation (1) was then derived by using integration by parts, Eq. (A3) to calculate the second term of Eq. (A7) and the completeness of the electron wave functions; $\sum_n \psi_n^*(\mathbf{r}_1) \psi_n(\mathbf{r}_2) = \delta(\mathbf{r}_1 - \mathbf{r}_2)$ [25, 26].
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