I. INTRODUCTION

Surface plasmons can couple with photons to produce collective excitations called surface plasmon-polaritons (SPPs) and localized surface plasmons (LSPs) [1-5]. SPPs and LSPs can concentrate optical waves into regions that are much smaller than their wavelengths and they can also greatly enhance their local electric fields at surface plasmon excitation energies. These effects make it possible to use SPPs and LSPs to fabricate nanoscale photonic devices and to dramatically enhance detection sensitivities for optical measurements. A new field called graphene plasmonics has recently developed, where the electron density in graphene can be varied over a wide range by gating it to tune the surface plasmon resonance frequency [6-8].

Many theoretical studies of SPPs and LSPs, including those based on the hydrodynamic method [9-12], the semi-classical method [13-15], random-phase approximation (RPA) [16-18] and the time-dependent local density approximation [19-22], have been reported. Theoretical studies of the interaction between the external electric field and the incident electrons and the surface plasma have also been reported [3, 12, 23-28]. These studies, however, have mainly focused on surface plasmon excitation, and the coupling phenomena between bulk and surface plasmons, which play important roles in the plasmon excitation process, have not been widely studied.

The author previously studied these coupling phenomena when calculating the inelastic scattering cross-sections of incident electrons based on bulk and surface plasmon excitations in metal thin films [18], and has recently developed a theory of localized plasmons under high frequency conditions using RPA, where the coupling between the bulk and surface plasmons is properly considered in metal nanostructures [29]. In this theory, the local electron density in the metal nanostructures plays essential roles in the plasmon excitation. This treatment has been recently applied to a study of surface plasmon excitation in Na nanoclusters [30] and developed further to study nonlinear optical responses in metal nanoparticles [31] by other groups.

In the author’s previous study [29], the quasi-static scalar potential was used for the metal nanostructures, where the light velocity is assumed to have an infinite value. However, retardation of the scalar potential caused by a finite light velocity must be considered in the study of light emission from metal nanostructures. In this study, a theory of light emission by localized plasmon excitations is derived by generalization of the previous theory of localized plasmons for metal nanostructures to consider the retardation of the scalar potentials. The emitted light intensities are analytically calculated for metal nanoparticles with step function-like electron densities at their surfaces when electric fields and electrons are incident on those surfaces.

II. LIGHT EMISSION BY LOCALIZED PLASMON EXCITATION

A. Retarded scalar potential

Considering an effective scalar potential \( \varphi_{\text{eff}}(r_2, \omega) \) within the electron gas in metal nanostructures, an electron charge density \( \rho_{\text{ind}}(r_1, \omega) \) induced by the effective scalar potential is given in the frequency \( \omega \) expression by [9, 29]

\[
\rho_{\text{ind}}(r_1, \omega) = \int d^2 r e^2 P_0(r_1, r_2, \omega) \varphi_{\text{eff}}(r_2, \omega),
\]

where the electron charge is \(-e\) and the polarization of the free electron gas \( P_0(r_1, r_2, \omega) \) is given by

\[
P_0(r_1, r_2, \omega) = \frac{2}{\hbar} \sum_{n,n'} \psi_n^*(r_1) \cdot \psi_n(r_2) \cdot \psi_{n'}(r_1) \cdot \psi_{n'}^*(r_2) \times \frac{\theta(E_F - E_n) - \theta(E_F - E_{n'})}{\omega + \omega_F - E_{n'} - E_n} / \hbar + i \eta,
\]

where the infinitesimal imaginary term \( i \eta \) is required by the causality of the one-particle Green’s function for a single electron [32]. \( \psi_n(r) \) is the single-electron wave func-
tion in the metal nanostructure, which satisfies the following Schrödinger equation:

$$\left[ -\frac{\hbar^2}{2m_e} \nabla^2 + V(r) \right] \psi_n(r) = E_n \psi_n(r). \quad (3)$$

In Eqs. (2) and (3), $\theta(E_F - E_n)$ is the step function, $E_F$ is the Fermi energy, $m_e$ is the electron mass and $E_n$ is the electron energy in the nanostructure with the local potential energy $V(r)$. Then a retarded induced scalar potential $\varphi_{\text{ind}}(r, \omega)$ is given by

$$\varphi_{\text{ind}}(r, \omega) = \int \text{d}r_1 G_0(r - r_1, \omega) \rho_{\text{ind}}(r_1),$$

$$= \int \text{d}r_1 \text{d}r_2 G_0(r - r_1, \omega) \times e^2 \rho_0(r_1, r_2, \omega) \varphi_{\text{eff}}(r_2, \omega). \quad (4)$$

$G_0(r - r_1, \omega)$ is the retarded Green’s function, which satisfies the following Helmholtz equation in Gaussian units:

$$\left( \nabla^2 + \frac{\omega^2}{c^2} \right) G_0(r - r_1, \omega) = -4\pi \delta(r - r_1),$$

$$G_0(r - r_1, \omega) = \frac{\exp(i\frac{\omega}{c} \cdot |r - r_1|)}{|r - r_1|} \quad (5)$$

where $\delta(r - r_1) \equiv \delta(x - x_1)\delta(y - y_1)\delta(z - z_1)$ and $c$ is the light velocity. When an external scalar potential $\varphi_{\text{ext}}(r, \omega)$ is considered, the retarded effective scalar potential satisfies

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

$$= \varphi_{\text{ext}}(r, \omega) + \int \text{d}r_1 \text{d}r_2 e^2 G_0(r - r_1, \omega) P_0(r_1, r_2, \omega) \varphi_{\text{eff}}(r_2, \omega). \quad (6)$$

The above self-consistent integral equation is equivalent to that calculated in the RPA when $c \to \infty$, which gives the well-known dielectric function for the homogeneous electron gas with translational invariance [32]. Equation (6) agrees with that given in the previous article when neglecting the retardation effect, namely, $c \to \infty$ [29].

A high-frequency condition of $|E_n - E_{n'}|/\hbar \omega \ll 1$ is considered to be valid for collective excitations such as plasmons, which have excitation energies that are much larger than single-electron excitation energies [29]. The right side denominator of Eq. (2) can then be approximated as:

$$\frac{1}{\omega} + (E_n - E_{n'})/\hbar \approx \frac{1}{\omega} \left[ 1 - \frac{(E_n - E_{n'})}{\hbar \omega} \right], \quad (7)$$

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

Using a similar method to that given previously [29], calculation of Eq. (6) using Eq. (7) leads to

$$\varphi_{\text{eff}}(r, \omega) = \varphi_{\text{ext}}(r, \omega) + \frac{\epsilon^2}{m_e \omega^2} \int \text{d}r_1 n_0(r_1) \times \nabla_1 G_0(r - r_1, \omega) \cdot \nabla_1 \varphi_{\text{eff}}(r_1), \quad \text{or}$$

$$n_0(r_1) = 2 \sum_n \psi_n^*(r_1) \cdot \psi_n(r_1) \cdot \theta(E_F - E_n),$$

where $n_0(r_1)$ is the local electron density in the metal nanostructure. $\nabla_1$ represents the gradient with respect to $r_1$. Equation (8) was derived using integration by parts, and Eq. (3) was used to calculate the second term of Eq. (7) and the completeness of the electron wave functions:

$$\sum_n \psi_n^*(r_1) \cdot \psi_n(r_2) = \delta(r_1 - r_2).$$

An integration by parts of Eq. (8) with respect to $r_1$ and the use of Eq. (5) lead to

$$\left[ 1 - \frac{4\pi \epsilon^2}{m_e \omega^2} n_0(r) \right] \varphi_{\text{eff}}(r, \omega)$$

$$+ \frac{\epsilon^2}{m_e \omega^2} \int \text{d}r_1 \varphi_{\text{eff}}(r_1, \omega) \nabla_1 G_0(r - r_1, \omega) \cdot \nabla_1 n_0(r_1)$$

$$= \varphi_{\text{ext}}(r, \omega). \quad (9)$$

When the light velocity is assumed to be infinite, i.e. $c \to \infty$, which indicates neglect of the retardation effect or the quasi-static approximation, Eq. (9) agrees with the equation that was given previously [29]:

$$\left[ 1 - \frac{4\pi \epsilon^2}{m_e \omega^2} n_0(r) \right] \varphi_{\text{eff}}(r, \omega)$$

$$+ \frac{\epsilon^2}{m_e \omega^2} \int \text{d}r_1 \varphi_{\text{eff}}(r_1, \omega) \nabla_1 \frac{\epsilon^2}{|r - r_1|} \cdot \nabla_1 n_0(r_1)$$

$$= \varphi_{\text{ext}}(r, \omega). \quad (10)$$

Another integration by parts of Eq. (8) leads to

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

$$- \frac{\epsilon^2}{m_e \omega^2} \int \text{d}r_1 G_0(r - r_1, \omega) \nabla_1 \cdot [n_0(r_1) \nabla_1 \varphi_{\text{eff}}(r_1, \omega)],$$

$$= \varphi_{\text{ext}}(r, \omega) + \int \text{d}r_1 G_0(r - r_1, \omega) \rho(r_1, \omega), \quad (11)$$

where $\rho(r_1, \omega)$ is the induced electric charge density, and is given by

$$\rho(r_1, \omega) \equiv -\frac{\epsilon^2}{m_e \omega^2} \nabla_1 \cdot [n_0(r_1) \nabla_1 \varphi_{\text{eff}}(r_1, \omega)]. \quad (12)$$

### B. Retarded vector potential

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

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

$$B(r, \omega) = \text{rot} A(r, \omega), \quad (13)$$

with the Lorentz condition:

$$\nabla \cdot A(r, \omega) - \frac{i \omega}{c} \varphi_{\text{eff}}(r, \omega) = 0. \quad (14)$$
The previous study [29] showed that the electric displacement \( D(\mathbf{r}, \omega) \) can be expressed using the local electron density-dependent dielectric function \( \varepsilon(\mathbf{r}, \omega) \):

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

\[
\varepsilon(\mathbf{r}, \omega) = \left[ 1 - \frac{\omega_p^2(\mathbf{r})}{\omega^2} \right], \quad \omega_p^2(\mathbf{r}) = \frac{4\pi\varepsilon_0^2}{m_e^*}n(\mathbf{r}),
\]

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

By substituting Eqs. (13), (14) and (15) into the following Maxwell equation,

\[
\mathbf{E}(\mathbf{r}, \omega) = \frac{4\pi}{c} j_{\text{ext}}(\mathbf{r}, \omega) - \frac{i\omega}{c} D(\mathbf{r}, \omega),
\]

the following equation is derived:

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

the following equation is derived:

\[
\nabla^2 \mathbf{A}(\mathbf{r}, \omega) + \frac{\omega_p^2}{c^2} \varepsilon(\mathbf{r}, \omega) \mathbf{A}(\mathbf{r}, \omega)
\]

\[
= \frac{i\omega}{c} \mathbf{j}_{\text{eff}}(\mathbf{r}, \omega) - \frac{4\pi}{c} j_{\text{ext}}(\mathbf{r}, \omega),
\]

\[
= -4\pi \left[ \mathbf{j}(\mathbf{r}, \omega) + j_{\text{ext}}(\mathbf{r}, \omega) \right],
\]

\[
\therefore \mathbf{J}(\mathbf{r}, \omega) = -\frac{i\omega}{c} \mathbf{J}_{\text{eff}}(\mathbf{r}, \omega),
\]

where \( j_{\text{ext}}(\mathbf{r}, \omega) \) is the external current density and \( \mathbf{j}(\mathbf{r}, \omega) \) is the current density induced by the plasmon excitations. Non-magnetic metals (\( \mu = 1 \)) are considered in this study. The retarded vector potential is then given by

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

\[
= \frac{1}{c} \int_{\mathbf{r}_1} \mathbf{G}_m(\mathbf{r} - \mathbf{r}_1, \omega) \left[ \mathbf{j}(\mathbf{r}_1, \omega) + j_{\text{ext}}(\mathbf{r}_1, \omega) \right],
\]

where \( \mathbf{G}_m(\mathbf{r} - \mathbf{r}_1, \omega) \) is the retarded Green’s function that satisfies

\[
\left[ \nabla^2 + \frac{\omega_p^2}{c^2} \varepsilon(\mathbf{r}, \omega) \right] \mathbf{G}_m(\mathbf{r} - \mathbf{r}_1, \omega) = -4\pi \delta(\mathbf{r} - \mathbf{r}_1).
\]

\( G_m(\mathbf{r} - \mathbf{r}_1, \omega) \) agrees with \( G_0(\mathbf{r} - \mathbf{r}_1, \omega) \) of Eq. (5) outside the metal nanostructures because \( \varepsilon(\mathbf{r}, \omega) = 1 \). The induced current density of Eq. (17) and the induced electric charge density of Eq. (12) satisfy the continuity equation for the electric charge:

\[
\nabla \cdot \mathbf{J}(\mathbf{r}, \omega) - i\omega \rho(\mathbf{r}, \omega) = 0.
\]

C. Electromagnetic fields from electric dipole moments

Only the induced electric charge and the induced current densities are considered in this section because only these densities contribute to light emission by the localized plasmon excitations. Using the induced electric charge density from Eq. (12), the electric dipole moment \( \mathbf{p}(\omega) \), its first order derivative \( \mathbf{p}(\omega) \) and its second order derivative \( \mathbf{p}(\omega) \) with respect to time are given by

\[
\mathbf{p}(\omega) = \int \mathbf{r}_1 \rho(\mathbf{r}_1, \omega) d\mathbf{r}_1,
\]

\[
= -\frac{e^2}{m_e^* \omega} \int d\mathbf{r}_1 \mathbf{r}_1 \nabla_1 \cdot [\mathbf{n}(\mathbf{r}_1) \mathbf{V}_{\text{eff}}(\mathbf{r}_1, \omega)],
\]

\[
= -\frac{1}{\omega^2 c} \int d\mathbf{r}_1 \mathbf{n}(\mathbf{r}_1) \mathbf{V}_{\text{eff}}(\mathbf{r}_1, \omega),
\]

\[
\mathbf{p}(\omega) = -i\omega \mathbf{p}(\omega) = \int d\mathbf{r}_1 \mathbf{r}_1(\mathbf{r}, \omega),
\]

\[
= -\frac{e^2}{m_e^* \omega} \int d\mathbf{r}_1 \mathbf{n}(\mathbf{r}_1) \mathbf{V}_{\text{eff}}(\mathbf{r}_1, \omega),
\]

\[
\mathbf{p}(\omega) = -\omega^2 \mathbf{p}(\omega) = \frac{e^2}{m_e^* \omega} \int d\mathbf{r}_1 \mathbf{n}(\mathbf{r}_1) \mathbf{V}_{\text{eff}}(\mathbf{r}_1, \omega).
\]

Following the method from the literature [33], the electromagnetic fields are calculated in the electric dipole approximation. The retarded Green’s function \( G_0(\mathbf{r} - \mathbf{r}_1, \omega) \) is then expanded as

\[
G_0(\mathbf{r} - \mathbf{r}_1, \omega) = \sum_{l=0}^{\infty} (2l + 1) h_l^{(1)} \left( \frac{\omega}{c} \right) j_l \left( \frac{\omega}{c} \right) P_l(\cos \theta),
\]

where \( h_l^{(1)} \left( \frac{\omega}{c} \right) \) is the spherical Hankel function of the first kind, \( j_l \left( \frac{\omega}{c} \right) \) is the spherical Bessel function, \( P_l(\cos \theta) \) is the Legendre polynomial, and \( \theta \) is the angle between \( \mathbf{r} \) and \( \mathbf{r}_1 \). The spherical Hankel function of the first kind is expressed by

\[
h_l^{(1)} \left( \frac{\omega}{c} \right) = \left( \frac{\omega}{c} \right) j_l \left( \frac{\omega}{c} \right) \left[ \frac{1}{d \mathbf{r}} \right] \frac{\exp(i\omega r/c)}{i\omega r/c},
\]

and the spherical Bessel function can be approximated by

\[
\sim \left( \frac{\omega}{c} \right) \frac{1}{2\mathbf{r}} \left( \frac{2\mathbf{r}}{c} \right) P_l(\cos \theta),
\]

when \( \omega r/c \approx k r_1 \ll 1 \) (where \( k \) is the wave number of the light) is assumed for metal nanostructures, which are much smaller in size than the wavelength of the emitted light. Then, the induced electric potential \( \varphi_{\text{ind}}(\mathbf{r}, \omega) \) is given by

\[
\varphi_{\text{ind}}(\mathbf{r}, \omega) = \int d\mathbf{r}_1 G_0(\mathbf{r} - \mathbf{r}_1, \omega) \rho(\mathbf{r}_1, \omega),
\]

\[
= \sum_{l=0}^{\infty} \left( \frac{2l + 1}{(2l + 1)!} \right) \left( \frac{\mathbf{r}}{r} \right)^l \left[ \frac{\exp(i\omega r/c)}{r} \right]
\]

\[
\times \left\{ \left( \frac{1}{c} \cdot \mathbf{J}_{\text{eff}}(\mathbf{r}_1) \mathbf{V}_{\text{eff}}(\mathbf{r}_1, \omega) \right) \right\},
\]

In the above equation, the term \( l = 0 \) vanishes when we use \( P_0(\cos \theta) = 1 \) and the Gauss theorem for the \( \mathbf{r}_1 \) integration. For the term \( l = 1 \) (an electric dipole moment), the induced scalar potential \( \varphi_{\text{ind}}^{(1)}(\mathbf{r}, \omega) \) is given by

\[
\varphi_{\text{ind}}^{(1)}(\mathbf{r}, \omega) = \exp \left( \frac{1}{i\omega c} \frac{1}{r^2} \right) \left[ \frac{e}{r} \cdot \mathbf{p}(\omega) + \frac{e}{c r} \cdot \mathbf{p}(\omega) \right].
\]
where the dipole moments in Eq. (21) and \( P_l(\cos \theta') = \cos \theta' \) were used; \( \mathbf{e}_r = r/r \) is the unit vector in the \( r \) direction. Because the ratio of the higher terms of \( l \geq 2 \) (i.e. multipole moments) to that of Eq. (24) is an order of \((kr_1)^{l-1} \ll 1\), only the induced scalar potentials of Eq. (24) can be considered in this study, indicating the electric dipole approximation.

Similar calculations for the induced vector potential \( \mathbf{A}_{\text{ind}}(\mathbf{r}, \omega) \) outside the metal nanostructures give

\[
\mathbf{A}_{\text{ind}}(\mathbf{r}, \omega) = \frac{1}{c} \int \mathrm{d} \mathbf{r}_1 \mathcal{G}_0(\mathbf{r} - \mathbf{r}_1, \omega) \mathbf{j}(\mathbf{r}_1, \omega),
\]

\[
= \frac{1}{c} \sum_{l=0}^{\infty} \frac{(2l+1)!}{(2l+1)^l} \left( -\frac{r}{r} \right)^l \left\{ \frac{1}{r} \frac{\mathrm{d}}{\mathrm{d}r} \right\}^l \frac{\exp(i\omega r/c)}{r} \\
\times \int \mathrm{d}r_1 r_1^l P_l(\cos \theta) \left\{ -\frac{\mathbf{e}_r \times \mathbf{p}(\omega)}{\omega m_e} \nabla \varphi_{\text{eff}}(\mathbf{r}_1, \omega) \right\}. 
\]

(25)

For the term \( l = 0 \), the induced vector potential is given by

\[
\mathbf{A}_{\text{ind}}^{(0)}(\mathbf{r}, \omega) = \frac{\exp(i\omega r/c)}{cr} \mathbf{p}(\omega). 
\]

(26)

Because the ratio of the higher terms of \( l \geq 1 \) (i.e. the magnetic dipole and the multipole moments) to that of Eq. (26) is an order of \((kr_1)^{l-1} \ll 1\), only the induced vector potentials of Eq. (26) can be considered in this study.

Using Eqs. (13), (24) and (26), the induced electric fields are given by

\[
\mathbf{E}_{\text{ind}}(\mathbf{r}, \omega) = -\nabla \varphi_{\text{ind}}^{(1)}(\mathbf{r}, \omega) + i\frac{\omega}{c} \mathbf{A}_{\text{ind}}^{(0)}(\mathbf{r}, \omega),
\]

\[
= \exp\left( i\frac{\omega}{c} r \right) \left\{ -\frac{\mathbf{p}(\omega)}{r^3} + \frac{3\mathbf{e}_r \times \mathbf{p}(\omega)}{r^3} \right\} + \frac{\mathbf{e}_r \times \mathbf{p}(\omega)}{c^2 r^2} + \frac{3\mathbf{e}_r \times \mathbf{p}(\omega)}{c^2 r^2},
\]

and the induced magnetic fields are given by

\[
\mathbf{H}_{\text{ind}}(\mathbf{r}, \omega) = \mathrm{rot} \mathbf{A}_{\text{ind}}^{(0)}(\mathbf{r}, \omega),
\]

\[
= -\exp\left( i\frac{\omega}{c} r \right) \left\{ \frac{\mathbf{e}_r \times \mathbf{p}(\omega)}{c^2 r} + \frac{\mathbf{e}_r \times \mathbf{p}(\omega)}{c^2 r} \right\}.
\]

(27)

(28)

The electromagnetic far fields \((\propto 1/r)\) and near fields \((\propto 1/r^2, 1/r^3)\) of Eqs. (27) and (28) in the \( \omega \) expression reproduce the well-known ones in the dipole approximation [33].

D. Intensity of light emission

Using the electromagnetic fields given in Eqs. (27), and (28) in the time \((t)\) expression, the Poynting vector is given by

\[
\mathbf{S}(\mathbf{r}, t) = \frac{c}{4\pi} \mathbf{E}_{\text{ind}}(\mathbf{r}, t) \times \mathbf{H}_{\text{ind}}(\mathbf{r}, t).
\]

(29)

Then, the energy of the electromagnetic waves emitted from the metal nanostructures per unit solid angle \( \Omega \) over the time \((t)\) interval from \(-\infty \) to \( \infty \) at \( r \to \infty \) is given by

\[
\frac{dI_T}{d\Omega} = \lim_{r \to \infty} \int_{-\infty}^{\infty} \frac{c^2}{4\pi^2} \mathbf{E}_{\text{ind}}(\mathbf{r}, t) \times \mathbf{H}_{\text{ind}}(\mathbf{r}, t),
\]

\[
= \lim_{r \to \infty} \int_{-\infty}^{\infty} d\omega \left\{ \mathbf{e}_r \times \Re\{\mathbf{E}_{\text{ind}}(\mathbf{r}, \omega) \times \mathbf{H}_{\text{ind}}(\mathbf{r}, \omega)\} \right\},
\]

(30)

and per unit frequency is given by

\[
\frac{d^2I_T}{d\Omega d\omega} = \lim_{r \to \infty} \int_{-\infty}^{\infty} \frac{c^2}{4\pi^2} \mathbf{E}_{\text{ind}}(\mathbf{r}, \omega) \times \mathbf{H}_{\text{ind}}(\mathbf{r}, \omega).
\]

(31)

where \( \Re \) represents the real part of the equation.

Dividing Eq. (31) by \( h\omega \), the photon number with energy \( h\omega \) per unit solid angle \( \Omega \) is given by

\[
I_{\text{ph}}(\Omega, \omega) = \frac{\mathbf{e}_r \times \mathbf{p}(\omega)}{4\pi^2 c^3 h\omega} |\mathbf{E}_{\text{ind}}(\mathbf{r}, \omega) \times \mathbf{H}_{\text{ind}}(\mathbf{r}, \omega)|^2,
\]

(32)

Substitution of Eqs. (27) and (28) into Eq. (32) and calculations performed using the formulas for the vectors give

\[
I_{\text{ph}}(\Omega, \omega) = \frac{\mathbf{e}_r \times \mathbf{p}(\omega)}{4\pi^2 c^3 h\omega} |\mathbf{E}_{\text{ind}}(\mathbf{r}, \omega) \times \mathbf{H}_{\text{ind}}(\mathbf{r}, \omega)|^2.
\]

(33)

and then,

\[
I_{\text{ph}}(\Omega, \omega) = \frac{1}{4\pi^2 c^3 h\omega} |\mathbf{E}_{\text{ind}}(\mathbf{r}, \omega) \times \mathbf{H}_{\text{ind}}(\mathbf{r}, \omega)|^2.
\]

(34)

Only the electromagnetic far-fields \((\propto 1/r)\) contributed to the light emission. This reproduces the well-known formula for the light intensity emitted from the electric dipole moments \( \mathbf{p}(\omega) \) [33].

Equation (34) is the general formula used to calculate the intensity of the light emission by localized plasmon excitation in the electric dipole approximation. This indicates also that the light emission intensity can be calculated if the electric field \( \mathbf{E}(\mathbf{r}_1, \omega) \approx -\nabla \varphi_{\text{eff}}(\mathbf{r}_1, \omega) \) derived by solving the Maxwell equation with the use of the dielectric function of Eq. (15) and the local electron density \( n_0(\mathbf{r}_1) \) are known inside the metal nanostructure in the case of the quasi-static approximation. The approximation is considered to be valid inside the nanostructures whose sizes are much smaller than the wavelength of light.

E. Light emissions from metal nanospheres

1. Dipole moments in metal nanospheres

In this section, \( \mathbf{p}(\omega) \) in Eq. (34) is calculated for the metal nanospheres to investigate the validity of the
present formulas. Using the polar coordinates \((r, \theta, \phi)\), the gradient of \(\varphi_{\text{eff}}(r, \omega)\) is given by

\[
\nabla \varphi_{\text{eff}}(r, \omega) = e_z \left( \sin \theta \cos \phi \frac{\partial}{\partial r} + \frac{\cos \theta \cos \phi}{r} \frac{\partial}{\partial \theta} - \frac{\sin \phi}{r \sin \theta} \frac{\partial}{\partial \phi} \right) \varphi_{\text{eff}}(r, \omega)
\]

\[+ e_y \left( \sin \theta \sin \phi \frac{\partial}{\partial r} + \frac{\cos \theta \sin \phi}{r} \frac{\partial}{\partial \theta} + \frac{\cos \phi}{r \sin \theta} \frac{\partial}{\partial \phi} \right) \varphi_{\text{eff}}(r, \omega)
\]

\[+ e_z \left( \cos \theta \frac{\partial}{\partial r} - \frac{\sin \theta}{r} \frac{\partial}{\partial \phi} \right) \varphi_{\text{eff}}(r, \omega),
\]

where \(e_x, e_y, e_z\) are the unit vectors for the \(x, y, z\) axes, respectively. When the system does not depend on the azimuth angle \(\phi\), which indicates rotational symmetry around the \(z\) axis, \(\vec{p}(\omega)\) is given by

\[
\vec{p}(\omega) = -e_z \frac{2\pi e^2}{m_e} \int_0^\infty r^2 dr \int_0^\pi \sin \theta d\theta \times n_0(r) \left[ \cos \theta \frac{\partial \varphi_{\text{eff}}(r, \omega)}{\partial r} - \frac{\sin \theta}{r} \frac{\partial \varphi_{\text{eff}}(r, \omega)}{\partial \phi} \right],
\]

(35)

because the \(x\) and \(y\) components vanish when the integration for \(\phi\) is performed. When \(n_0(r)\) has spherical symmetry, which depends only on \(r\), integration by parts for \(\theta\) in Eq. (35) gives

\[
\vec{p}(\omega) = -e_z \frac{2\pi e^2}{m_e} \int_0^\infty r^2 dr \int_0^\pi \sin \theta d\theta \times n_0(r) \left\{ \cos \theta \left[ \frac{\partial \varphi_{\text{eff}}(r, \omega)}{\partial r} + \frac{2}{r} \varphi_{\text{eff}}(r, \omega) \right] \right\}.
\]

(36)

The effective and external scalar potentials can be expanded using the normalized spherical harmonics \(Y_{l,m}(\theta, \phi)\) with the complete orthonormal property as

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

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

(37)

where

\[
Y_{l,m}(\theta, \phi) = \varepsilon \left[ \frac{2l+1}{4\pi} \frac{(l-|m|)!}{(l+|m|)!} \right]^{1/2} P^m_l(\cos \theta) e^{im\phi},
\]

where \(\varepsilon = (-1)^m\) for \(m > 0\), \(\varepsilon = 1\) for \(m \leq 0\), and \(P^m_l(\cos \theta)\) is the associated Legendre function, where \(l\) and \(m\) are integers. Because the effective scalar potential does not depend on \(\phi\), \(\varphi_{\text{eff}}(r, \omega)\) from Eq. (37) can be expressed as

\[
\varphi_{\text{eff}}(r, \omega) = \sum_{l=0}^\infty \left[ \frac{2l+1}{4\pi} \right]^{1/2} \varphi_{l,0}^{l,m}(r, \omega) P^0_l(\cos \theta),
\]

(38)

where only the term \(m = 0\) contributes to the effective scalar potential. By substituting Eq. (38) into Eq. (36), the following result is obtained:

\[
\vec{p}(\omega) = -e_z \frac{2\pi e^2}{m_e} \left\{ \frac{4\pi}{3} \int_0^\infty r^2 dr \times n_0(r) \left[ \frac{\partial \varphi_{l,0}^{l,m}(r, \omega)}{\partial r} + \frac{2}{r} \varphi_{l,0}^{l,m}(r, \omega) \right] \right\},
\]

(39)

where the term \(l = 1\) (the electric dipole term) only contributes to the calculation because \(P^1_l(\cos \theta) = \cos \theta\) and because of the orthogonal property of \(P^l_0(\cos \theta)\),

\[
\int_{-1}^1 P^0_l(\cos \theta) P^0_m(\cos \theta) d(\cos \theta) = \frac{2}{2l+1} \delta_{n,1},
\]

where \(\delta_{n,1}\) is the Kronecker’s delta function. The terms of \(l \neq 1\), however, contribute to the dipole moments when \(n_0(r)\) has the \(\theta\) dependence such as in a spheroidal nanostucture, which is known from Eq. (35). Equation (39) is the general formula used to calculate the electric dipole moments in metal nanospheres.

2. Model calculations

Consider a model of a metal nanosphere with spherical symmetry and radius \(a\), where the electron density is assumed to have a step function shape, \(n_0(r) = n_0 (a - r)\) to know the analytical results. Equation (10) can be used to calculate the effective scalar potential because the areas of \(r\) and \(r_1\) are confined to the metal nanospheres by the local electron density term \(n_0(r)\) and the condition of \(\omega/c \cdot |r - r_1| \approx k |r - r_1| \ll 1\) is satisfied, indicating that \(G_0(r - r_1, \omega) \approx 1/|r - r_1|\). The validity of the approximation has been confirmed by the actual calculation using Eq. (9) (not shown in this paper). By expanding \(\varphi_{\text{eff}}(r, \omega)\) and \(\varphi_{\text{ext}}(r, \omega)\) based on the normalized spherical harmonics and using the relationship of

\[
\frac{dn_0 (a - r)}{dr} = -n_0 \delta(r - a),
\]

Eq. (10) can be analytically solved. After calculations similar to those of the previous study [29], the following results are obtained:

(1) Outside the nanosphere: \(\lambda \gg r > a\) (\(\lambda\) is the wavelength of the light)

\[
\varphi_{\text{eff}}^{l,m}(r, \omega) - \varphi_{\text{ext}}^{l,m}(r, \omega) = \varphi_{\text{ind}}^{l,m}(r, \omega)
\]

\[
= \frac{\omega_p^2 \omega^2 \left[ \frac{l}{2l+1} \right] \left[ \frac{l}{2l+1} \right]^{1/2} P^0_l(\cos \theta) e^{im\phi}}{\omega^2 - \omega_p^2 \left[ \frac{l}{2l+1} \right]^{1/2} P^0_l(\cos \theta) e^{im\phi}}.
\]

The pole of the above equation gives the localized surface plasmon frequency:

\[
\omega = \omega_p \sqrt{\frac{l}{2l+1}}, \quad \omega_p = \sqrt{\frac{4\pi e^2 n_0}{m_e}}.
\]

(2) Inside the nanosphere: \(0 \leq r \leq a\)

\[
\varphi_{\text{eff}}^{l,m}(r, \omega) = \frac{\omega^2}{\omega^2 - \omega_p^2 \left[ \frac{l}{2l+1} \right]} \left[ \frac{l}{a} \right] \varphi_{\text{ext}}^{l,m}(r, \omega)
\]

\[+ \frac{\omega^2}{\omega^2 - \omega_p^2} \left[ \frac{l}{a} \right] \varphi_{\text{ext}}^{l,m}(a, \omega).
\]

(40)
The poles of the above equation not only give the localized surface plasmon frequency, but also give the bulk plasmon frequency: \( \omega = \omega_p \). It is noted that the second term of the bulk plasmon excitation vanishes at the nanosphere surface, where \( r = a \), based on the coupling term of \(- (r/a)^4 \varphi_{\text{ext}}^1(a, \omega) \). The disappearance of the bulk plasmon term is also derived at the surfaces of metal thin films (not shown in this work) [18]. This means that the scalar potential caused by bulk plasmon excitation is shielded by the induced surface charges caused by surface plasmon excitation outside the metal nanostructures.

Substitution of Eq. (41) for \( l = 1, m = 0 \) into Eq. (39) and integration by parts for \( r \) lead to

\[
\hat{p}(\omega) = -e_z \frac{e^2 r_0}{m_e} \left( \frac{4\pi}{3} \right)^{1/2} \frac{\omega^2 \omega^2}{\omega^2 - \omega_p^2/3} \varphi_{\text{ext}}^1(a, \omega).
\]  

(42)

It is noted that the bulk plasmon excitation term vanishes because of the coupling term and thus only the surface plasmon excitation for the dipole (\( l = 1 \)), \( \omega = \omega_p/\sqrt{3} \), contributes to the electric dipole moments. The coupling phenomenon has not been discussed so far for the electric dipole moment. The dipole moment is determined by the external scalar potential at the metal nanosphere surface \( r = a \).

Next, the following two cases with rotational symmetry around the \( z \)-axis are considered using this model.

(a) Consider a case where an external electric field \( \mathbf{E}(r, \omega) = e_z E_0 \) is incident on the nanosphere, indicating white light irradiation in the dipole approximation. The external scalar potential is given by

\[
\varphi_{\text{ext}}(r, \omega) = -E_0 z = -E_0 r \cos \theta,
\]

and then

\[
\varphi_{\text{ext}}^1(r, \omega) = - \left( \frac{4\pi}{3} \right)^{1/2} \frac{\omega^2 \omega^2}{\omega^2 - \omega_p^2/3} E_0 r.
\]  

(43)

Substitution of Eq. (43) at \( r = a \) into Eq. (42) then leads to

\[
\hat{p}(\omega) = e_z \frac{a^2 E_0}{3} \frac{\omega^2 \omega^2}{\omega^2 - \omega_p^2/3}.
\]  

(44)

The above electric dipole moment agrees with that derived by direct solution of the Maxwell equation in the quasi-static approximation [1]. The same dipole moment was also obtained when the electric field \( \mathbf{E}(r, \omega) = e_z E_0 \omega^2 / (\omega^2 - \omega_p^2/3) \) derived by solving the Maxwell equation with the Drude dielectric function of \( 1 - \omega_p^2/\omega^2 \) in the quasi-static approximation was used to calculate the dipole moment as described in the last paragraph of section II.D.

To consider the damping of the plasmon, \((\omega + i\eta)^2\) (where \( \eta \) is the infinitesimal imaginary term) cannot be merely replaced by \((\omega + i\Gamma)^2\) due to the violation of the detailed balance condition of the electrons in the metal nanosphere [34], where \( \Gamma \) is the finite damping frequency caused by the finite lifetime of the single electron in the metal nanosphere [2]. To consider the damping correctly, \((\omega + i\eta)^2\) should be replaced by \((\omega + i\Gamma)^2\) [35, 36]. However, the plasmon damping caused by intra- and inter-band excitations of individual electrons induced by the plasmon oscillation, which is called Landau damping [32], should be studied further by calculation of the imaginary terms in Eq. (2) [37].

By substituting Eq. (44) into Eq. (34) and considering the plasmon damping \( \Gamma \), the photon number with energy \( h\omega \) per unit solid angle of \( \Omega \) is given by

\[
I_{\text{ph}}(\Omega, \omega) = \frac{\omega^3 E_0^2 a^0 \sin^2 \theta}{36\pi^2 c^3 \hbar} \frac{\omega^2}{(\omega^2 + \omega_p^2/3) + \omega^2 \Gamma^2},
\]  

(45)

where \( \theta \) is the angle between \( e \) and \( e_z \), as shown in Fig. 1. The light intensity of Eq. (45) has a maximum value near the surface plasmon excitation frequency, \( \omega = \omega_p/\sqrt{3} \), and the typical angular dependence of \( \sin^2 \theta \) for light emission from an electric dipole moment.

(b) Consider a case where an electron with velocity \( v \) is incident on the nanosphere along the \( z \)-axis direction, \( e_z \), at the position \( X_0 \) on the \( x-y \) plane, as shown in Fig. 1. The external scalar potential \( \varphi_{\text{ext}}^1(a, \omega) X_0 \) in Eq. (42) is given by

\[
\varphi_{\text{ext}}^1(a, \omega) X_0 = \frac{-ie \sqrt{12\pi} \int d\mathbf{k}_\perp}{\pi v} \sum_{l,m} j_1 \left[ a \sqrt{k_\perp^2 + (v/c)^2} \right] \frac{1}{k_\perp^2 + [1 - (v/c)^2](v/c)^2} \exp(-i\mathbf{k}_\perp \cdot \mathbf{X}_0),
\]  

(46)

where \( \mathbf{k}_\perp \) is the wave number vector perpendicular to the \( z \)-axis direction of the electron. The term \( [1 - (v/c)^2] \) indicates the relativistic effect caused by the retardation of the scalar potential. Detailed calculations of Eq. (46) are shown in Appendix. Using Eqs. (34) and (42), the following result is then obtained:

FIG. 1. Geometrical arrangement between the metal nanosphere and an incident electron.
\[
I_{ph}(\Omega, \omega)X_0 = \frac{e^2 \omega_p^4 a^2 \sin^2 \theta}{4 \pi^3 \epsilon_0^3 v^2} \int dk_\perp \frac{j_1}{k_\perp^2} \frac{[a \sqrt{k_\perp^2 + (\omega/v)^2}]^2}{[k_\perp^2 + 1 - (v/c)^2]} \exp(-i k_\perp \cdot X_0) \frac{(\omega^2 - \omega_p^2/3)^2}{(\omega^2 - \omega_p^2)^2},
\]

\[
e^2 \omega_p^4 a^2 \sin^2 \theta \int_0^\infty k_\perp dk_\perp \frac{j_1}{k_\perp^2} \frac{[a \sqrt{k_\perp^2 + (\omega/v)^2}]^2}{[k_\perp^2 + 1 - (v/c)^2]} \exp(-i k_\perp \cdot X_0) \frac{(\omega^2 - \omega_p^2/3)^2}{(\omega^2 - \omega_p^2)^2},
\]

where \( J_0(k_\perp X_0) \) is the Bessel function of the first kind given by

\[
J_0(k_\perp X_0) = \frac{1}{2\pi} \int_0^{2\pi} d\phi' \exp(-i k_\perp X_0 \cos \phi').
\]

Equation (47) shows the photon number when the electron passes at position \( X_0 \). When the incident electrons with the two-dimensional density \( N_e \) on the \( x-y \) plane are incident on the nanosphere, the photon number with the energy \( h\omega \) per unit solid angle of \( \Omega \) is given by

\[
I_{ph}(\Omega, \omega) = N_e \int dX_0 I_{ph}(\Omega, \omega)X_0 = \frac{2N_e e^2 \omega_p^4 a^2 \sin^2 \theta}{\pi^3 \epsilon_0^3 v^2} \int_0^\infty k_\perp dk_\perp \frac{j_1}{k_\perp^2} \frac{[a \sqrt{k_\perp^2 + (\omega/v)^2}]^2}{[k_\perp^2 + 1 - (v/c)^2]} \exp(-i k_\perp \cdot X_0) \frac{(\omega^2 + \omega \Gamma^2)}{(\omega^2 - \omega_p^2/3)^2 + \omega^2 \Gamma^2},
\]

where the plasmon damping was taken into consideration. The result of Eq. (48) has also a maximum value near the surface plasmon excitation frequency, \( \omega = \omega_p/\sqrt{3} \), and the typical angular dependence of \( \sin^2 \theta \). The frequency dependence, however, is modified by the frequency dependence of the external scalar potential of the incident electron. For the non-relativistic condition, where \( v/c \ll 1 \), the \( k = \sqrt{k_\perp^2 + (\omega/v)^2} \) dependence of Eq. (48) coincides with that of the inelastic scattering cross section of the plane wave incident electron by the surface plasmon excitation for \( l = 1 \), which was previously derived by the quantum mechanical method [29]. This indicates that the light emission is accompanied with the inelastic scattering of the incident electron by the surface plasmon excitation.

### III. CONCLUSIONS

The theory of light emission caused by localized plasmon excitation was derived by developing our existing theory for localized plasmons for metal nanostructures to consider the retardation of the scalar potentials. Using the retarded scalar and vector potentials, the electromagnetic fields produced by the electric dipole moments were calculated for the metal nanostructures. The electromagnetic far-fields gave the frequency and angular dependences of the emitted light intensity from the metal nanostructures.

The light emission intensities were analytically calculated by application of the theory to the cases where electric fields and electrons were incident on metal nanospheres with step function-like electron densities at their surfaces. It was shown that only the surface plasmon excitation for \( l = 1 \) contributed to the light emission because of the coupling term between the surface and bulk plasmon excitations. The results showed the validity of the present theory. Moreover, the result of Eq. (34) showed that the light emission intensity can be calculated for the metal nanostructures if the electric fields and the local electron densities are known inside the metal nanostructures in the quasi-static approximation.

The results presented here were limited to the electric dipole approximation. The theory, however, can be developed to study light emissions in the multipole approximation, when the higher order terms are taken into account to calculate the retarded Green’s function of Eq. (22). The present formula can be also developed to the light emission from periodic metal nanostructures using the structural Green’s function method [38-40], which is applied to Eq. (11) using the periodic electric charge density and the Bloch’s theorem for the scalar potentials. These are problems that should be solved in future work. The theory presented here is considered useful for the study of light emission by localized plasmon excitation in metal nanostructures.

### Appendix A: External scalar potential caused by an incident electron

Consider the case where an incident electron with velocity \( v \) passes through position \( X_0 \) on the \( x-y \) plane, as shown in Fig. 1. The external scalar potential from the incident electron is given by [33]

\[
\varphi_{ext}(r, t)X_0 = \frac{-e}{\sqrt{(z - vt)^2 + [1 - (v/c)^2]}(X - X_0)^2},
\]

where the external charge density of \( \rho_{ext}(r, \omega) = -e\delta(r - r'(t)) \); here, \( r'(t) \) is the trajectory of the incident electron, and the retarded Green’s function of Eq. (5) in the time \( t \) expression was used to derive the above result. By expanding the external scalar potential using normalized spherical harmonics, \( \varphi_{ext}(r, \omega)X_0 \) is given by
The Fourier transformation of Eq. (A.2) for $X_0$ leads to

$$\int d{X_0} \varphi_m^{l,m}_{\text{ext}}(r, \omega) X_0 \exp(i k \cdot X_0) = -\frac{4 \pi e}{v} \int_0^\pi \sin \theta d\theta \int_0^{2\pi} d\phi \frac{Y_m^*(\theta, \phi) \exp(i \omega t)}{\sqrt{(z - v t)^2 + [1 - (v/c)^2]} (X - X_0)^2}, \quad (A.2)$$

where the orthogonal property of the associated Legendre function $P_m^l(\cos \theta)$ and the following relation were used:

$$\exp(i k \cdot r) = \sum_{l=0}^\infty (2l+1) (i)^l j_l(kr) P_l^0(\cos \theta), \quad (A.4)$$

where $k = (k_\perp, \frac{\omega}{v})$, and $\theta$ is the angle between $k$ and $r$. The inverse Fourier transform of Eq. (A.3) for $k_\perp$ leads to

$$\varphi_m^{l,0}_{\text{ext}}(r, \omega) X_0 = -\frac{e}{\pi v} \int d{k_\perp} \frac{[4 \pi (2l+1)^{1/2} (i)^l j_l(k r^2 + (\omega/v)^2)]}{k_\perp^2 + [1 - (v/c)^2] (\omega/v)^2} \exp(-i k_\perp \cdot X_0). \quad (A.5)$$

Equation (46) is then derived for $l = 1$ and $r = a$. 

[27] F. J. García de Abajo, Rev. Mod. Phys. 82, 209 (2010).