Electromagnetic-field imbalance in surface plasmon polariton and its role in slow propagation and field-matter interaction

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We present the results of a theoretic study of the electromagnetic field imbalance in surface plasmon polaritons (SPPs), which reveal that the magnetic field components induced by the electric fields normal and parallel to a metal surface cancel each other in SPPs, resulting in an imbalance. A group velocity analysis shows that this imbalance contributes significantly to the slow propagation of SPPs. We also analyze the enhanced spontaneous emission and nonlinearity in plasmonic cavities, and the results indicate that the electromagnetic field imbalance must be considered to correctly estimate the interaction strength. © 2019 Optical Society of America

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1. INTRODUCTION

The electric field of surface plasmon polaritons (SPPs) exponentially decays with distance from a metal surface because they induce charge oscillation, which is known as a charge density wave (CDW), and is thus confined to a tiny modal cross-section beyond the diffraction limit [1,2]. SPPs also exhibit slow propagation and a high density of states because of the strong dispersion of metal. Consequently, the SPP-emitter interaction is enhanced in a manner similar to that of a light emitter in a dielectric structure [3,4]. To date, the properties of SPPs have been investigated in various plasmonic structures, such as metal plates, periodic grooves (propagating SPP), particles (localized SPP) [5], and graphene-included structures [6]; further, enhanced spontaneous emission (SE) [7–16], Raman scattering [17,18], stimulated emission [19], single-photon emission [20,21], and Rabi splitting in the strong-coupling regime [22] have been studied with the SPPs.

Recently, the imbalance between electric and magnetic fields of SPPs has attracted attention. Herein, we refer to this situation as EM imbalance. In our previous study on enhanced SE, the Purcell factor showing the enhancement fraction was expressed via the EM imbalance factor \( \Theta \equiv \text{magnetic energy/electric energy} \), which approaches zero for SPPs on a metal plate [16]. Similar expressions were reported for different normalization conditions of the electric field [23], for localized SPPs [24], and for a dissipating rate [25]. Moreover, Johnson et al. investigated the EM imbalance in free plasma, which explained why it is caused by an increased kinetic energy of oscillating charges, resulting in the slow propagation of the electromagnetic field [26]. A recent study on the EM imbalance in SPPs and surface phonon polaritons [27] showed that the magnetic energy can be quite small in a small metal sphere, which indicates that the degree of the imbalance depends on the size of a plasmonic cavity, which confined the SPP or surface phonon polariton.

In this paper, we extend these studies to investigate the mechanism of extreme EM imbalance (\( \Theta \approx 0 \)) and of slow propagation of light over a metal plate. Moreover, we again discuss the Purcell factor as well as the enhanced nonlinearity to show that they are underestimated unless the EM imbalance is taken into account. In Section 2, to clarify the mechanism, we show analytically and numerically the electromagnetic energy of SPPs and the kinetic energy of the CDW on a metal plate and in a metal sphere. In Section 3, we discuss the slow propagation and analyze the group velocity by using Johnson’s method. In Section 4, we analyze quantum mechanically the Purcell factor and compare the results with experimental results. Finally, we discuss the enhanced nonlinearity in a coupled plasmonic cavity [28], for which the EM imbalance exerts a much greater influence.

2. MECHANISM OF ELECTROMAGNETIC IMBALANCE

A. Surface Plasmon Polariton on a Metal Plate

Figure 1(a) shows a metal plate model in which SPPs with wave vector \( k \) travel along the metal– dielectric interface. The permittivity of the metal is approximated as \( \epsilon_m = \epsilon_\infty - \epsilon_\infty \omega_p^2 / \omega^2 \). The dielectric side has refractive index \( n \), and its absorption is ignored. The electromagnetic fields \( \mathbf{E}^+ \) and \( \mathbf{B}^+ \) of SPPs vary exponentially in the \( z \) direction.
normal to the interface, as shown in Fig. 1(b). This is characterized by the imaginary propagation constant \( ik \); \( k \) is negative on the metal side (\( z < 0 \)) and positive on the dielectric side (\( z > 0 \)) [1,2]. The CDW is characterized by the current density \( \mathbf{J}^+ = i \varepsilon_\infty \varepsilon_0 (\omega_c^2 / \omega) \mathbf{E}^+ \) and charge density \( \rho^+ = -\int \mathbf{v} \cdot i \varepsilon_\infty \varepsilon_0 (\alpha_p^2 / \omega) \mathbf{E}^+ \, dr \) on the metal surface, where \( \varepsilon_0 \) is the permittivity in vacuum. We now present an example numerical calculation of the CDW, assuming \( n_{\text{Au}} = 0.215 + i3.22 \) at a wavelength \( \lambda = 612 \, \text{nm} \) on the metal (Au) side and \( n = 1.5 \) on the dielectric side [29]. In the Drude model, \( n_{\text{Au}} \) is characterized by the plasma frequency \( \omega_p \) or the corresponding wavelength \( \lambda_p = 2\pi c / \omega_p = 181 \, \text{nm} \), where \( c \) is the speed of light in vacuum and \( \varepsilon_\infty = 1 \). Figure 1(c) shows the calculated distributions of Re(\( \mathbf{J}^+ \)) and Re(\( \rho^+ \)) of the CDW excited near the surface, where the SPPs propagate in the \( \lambda \) direction. Figure 1(d) illustrates the relation among the magnetic field \( B_y \), \( \equiv |\mathbf{B}^+| \), the rotating electric field \( E_{\text{Fara}} \) induced through Faraday’s law, the net parallel electric field \( E_x \equiv |\mathbf{E}^+ \cdot \mathbf{k} / |k| \), and the normal electric field \( E_z \equiv |\mathbf{E}^+ \times \mathbf{k} / |k| \). The electric-field component \( E_y \) is canceled by the superposition of \( E_{\text{Fara}} \) in a uniform medium, whereas it has nonzero values in SPPs and forms the CDW because of the rapid exponential decay in the \( z \) direction.

The electric-field amplitudes \( E_x \) and \( E_z \) are obtained analytically by solving Maxwell’s equations for \( \mathbf{E}^+ \), which are expressed as

\[
(E_x, E_z) \propto \left( \frac{\kappa}{\varepsilon_m}, \frac{1}{|\varepsilon_m|} \right), \quad z < 0, \tag{1a}
\]

\[
(E_x, E_z) \propto \left( \frac{\kappa}{n^2}, \frac{1}{n} \right), \quad z > 0, \tag{1b}
\]

where \( \kappa = -[k^2 - \varepsilon_m (\omega / c)^2]^{1/2} \) at \( z < 0 \), \( \kappa = [k^2 - n^2 (\omega / c)^2]^{1/2} \) at \( z > 0 \), and \( \kappa = [k^2 - \varepsilon_m (\omega / c)^2]^{1/2} / \varepsilon_m = 0 \) [2]. Maxwell’s equation \( \mathbf{B}^+ = \nabla \times \mathbf{E}^+ \) also relates \( B_y \) to \( E_x \) and \( E_z \) as follows:

\[
B_y = \frac{1}{\omega} |k E_z| - |k E_x|, \tag{1c}
\]

Equation (1c) shows that the magnetic field components induced by \( E_x \) and \( E_z \) partly cancel each other and \( B_y \) decrease.

Figure 2(a) shows the distributions of electric, magnetic, and kinetic energy densities in the \( z \) direction:

\[
w_\text{E} = \frac{\varepsilon_\infty \varepsilon_0 |E_x|^2}{2} \quad (z < 0), \quad \frac{n^2 \varepsilon_0 |E_z|^2}{2} \quad (z > 0), \tag{2a}
\]

\[
w_\text{B} = \frac{|B_y|^2}{2 \mu_0}, \tag{2b}
\]

\[
w_\text{kin} = \frac{|J_x|^2}{2 \varepsilon_\infty \varepsilon_0 \omega_p^2}, \tag{2c}
\]

and \( |E_x / E_z| \) on the metal surface at different wavelengths \( \lambda_x (\equiv 2 / k) \) and frequencies \( \omega \) normalized by the resonance frequency \( \omega_x \equiv \omega_p / (\varepsilon_\infty + n^2)^{1/2} \) [1,2]. The energies \( W_\text{E}, W_\text{B}, W_\text{kin} \) are also shown in Fig. 2(b), which are given by the volume integrals of the energy densities. We also define the sum of the electric field and electric charge energies as \( W_\text{EE} \equiv W_\text{E} + W_\text{kin} \) and the total energy \( W \equiv W_\text{E} + W_\text{B} + W_\text{kin} \) in this case,

\[
\Theta \equiv W_\text{B} / W_\text{EE}. \tag{3}
\]

By using the relation \( \varepsilon_m = \varepsilon_\infty - \varepsilon_\infty \alpha_p^2 / \omega^2 \), \( W_\text{EE} \) may be written in the well-known form \( \frac{\partial}{\partial \omega} \int \bar{E} |\mathbf{E}^+|^2 \, dr \), where \( \bar{E} = \partial (\varepsilon_m / \omega) / \partial \omega \) in the metal and \( \bar{E} = n^2 \) in the dielectric.

**Fig. 1.** (a) Uniform metal–dielectric interface in (\( x, y \)) plane. Regions \( z < 0 \) and \( z > 0 \) are filled with metal (Au) and dielectric (\( n = 1.5 \)), respectively. The vector \( \mathbf{x} \) in the (\( x, y \)) plane represents the projection of the position vector \( \mathbf{r} \). (b) Field distribution of SPPs in the z direction. (c) Current density (arrows) and charge density (colors) of CDW near Au surface, when SPPs propagate in the \( x \) direction. (d) Schematic illustration showing how CDW is excited. \( E_{\text{Fara}}, E_x, \) and \( E_z \) are the rotating electric field induced by \( B_y \) due to Faraday’s law, the net parallel electric field, and the normal electric field, respectively.
not change; consequently, Θ and $W_{VE}/W$ approach zero and $W_{VE}$ because of the magnetic field cancelation.

### B. Surface Plasmon Polariton Around a Metal Sphere

Previous work investigated the EM imbalance in a metal sphere [27]. We check the correspondence between SPPs on a metal plate and SPPs on a metal sphere, focusing on the field patterns and Θ for high-order SPP modes on the metal sphere. Figure 3(a) shows the EM fields of the fundamental ($l = 1$) and higher-order ($l = 2, 3$) modes excited around a 20 nm diameter Au sphere [32] (see Appendix A for analysis method). Standing waves of the modes have 2f nodes and antinodes. Figure 3(b) gives the wavelength of the standing wave, $\lambda_s$, node volumes $V_{mode}$ [14,15], and Θ. As $l$ increases, $\lambda_s$ decreases ($\omega \rightarrow \omega_z$), and then Θ decreases in the same manner as the relation $\lambda_s \propto \Theta$ on a metal plate.

### 3. PROPAGATION OF SURFACE PLASMON POLARITON

#### A. Surface Plasmon Polariton on a Metal Plate

According to a study by Johnson [26], the optical group velocity $v_{gr,pl}$ is interpreted as the modified phase velocity $v_{ph,pl}$ in a uniform free plasma (the superscript pl denotes a free plasma), which is multiplied by the ratio of the propagating energy density $w_{pl}^{E}$ and $w_{kin}$ to the total energy density $W_{E}^{\text{pl}} + W_{B}^{\text{pl}} + W_{kin}^{\text{pl}}$ giving

$$v_{ph,pl} = \frac{w_{E}^{\text{pl}} + w_{B}^{\text{pl}} + W_{kin}^{\text{pl}}}{w_{pl}^{E}},$$

where $w_{E}^{\text{pl}} = w_{E}^{\text{pl}} + w_{B}^{\text{pl}}$ must hold. In Eq. (5), the propagating energy density is less than the total energy density by a factor of $2w_{kin}$ because the electrons do not propagate by themselves, but their oscillation does. In addition, only the part of $w_{pl}$ that is equal to $w_{B}^{\text{pl}}$ can propagate. By applying this relation to SPPs on a metal plate, we obtain

$$v_{ph} = \frac{W - 2W_{kin}^{\text{pl}}}{W} = \frac{2\Theta}{1 + \Theta} v_{ph}^{\text{pl}}.$$

To verify Eq. (6), we calculate $2\Theta/(1 + \Theta)$ and $v_{ph}/v_{ph}^{\text{pl}}$ independently by using the model of Fig. 1(a). Here, the former was obtained from the energies calculated in the same manner as used in Section 2A, and the latter was evaluated from the ($\omega, k$) dispersion relation as $(\partial \omega / \partial k)/(\omega / k)$. We confirm the relation $v_{ph}/v_{ph}^{\text{pl}} = 2\Theta/(1 + \Theta)$, as shown in Fig. 4. This means that the EM imbalance on a metal plate is essentially the same as that for a free plasma. Equation (6) indicates that an extremely low group velocity of the SPP occurs when Θ takes quite small values at frequencies close to $\omega_z$.

### B. Surface Plasmon Polariton in a Thin Metal Film

We now consider a thin metal film and its antisymmetric SPP mode, as shown in Fig. 5(a), which undergoes slow propagation...
over a wider range of \( k \) than does an SPP on a metal plate [1,16]. To examine the EM imbalance, we simply calculate \( \Theta \) and the energy flux in the \( x \) direction: \( s_d = (E^+ \times B^+) \cdot k/k \) in the dielectric and an analogous expression for \( s_m \) in the metal (Au). Figure 5(d) shows the sum of the energy flux normalized by the energy density, \( S_d = \int \int s_d \mathrm{d}x \mathrm{d}z \) and \( S_m = \int \int s_m \mathrm{d}x \mathrm{d}z \), where \( w = w_0 + w_1 + w_{\text{kin}} \) and the group velocity \( v^g \) (= \( S_d + S_m \)), for the first antisymmetric SPP mode [1,2]; its fields, dispersion curve, and \( \Theta \) are shown in Figs. 5(b) and 5(c). \( S_d \) and \( S_m \) have opposite signs because of the backward scattering of light by the conduction electrons in the metal. At \( k < 0.06 \text{ nm}^{-1} \) in the plot of Fig. 5(d), \( S_d \) decreases and \( S_m \) increases as \( k \) increases. They eventually cancel out at \( k = 0.06 \text{ nm}^{-1} \), where \( v^g \) goes to zero.

At \( k > 0.06 \text{ nm}^{-1} \), both approach zero, which is attributed to the weakened magnetic field. In the thin metal layer, the cancelation of forward and backward energy fluxes and the decreases in the fluxes themselves result in slow light propagation over a wide range of \( k \).

In an asymmetric structure \((n_1 > n_2)\), because the propagation constant \( k_x \) in the \( z \) direction has a real part, the SPP transports its energy toward the dielectric with the larger index \( n_1 \), being coupled into the far field (e.g., in the case of the Kretschmann–Raether [K-R] configuration). According to [33], the group velocity \( \partial \omega_x / \partial k \) in the \( z \) direction takes a quite small value at a frequency close to \( \omega_s \), as \( \partial \omega_x / \partial k \) does in Figs. 5(c) and 5(d). It indicates that the reduction of the magnetic field affects the energy transport in the \( z \) direction and the far field coupling in the K-R configuration.

4. FIELD-MATTER INTERACTION

A. Electric Field Enhancement by EM Imbalance

To determine how \( \Theta \) affects the field-matter interaction, we first examine quantized electric field \( \hat{E} \) with \( \Theta < 1 \), and the expectation value of the intensity operator \( \hat{E} \hat{E}^\dagger \) with the Fock state \( |N \rangle \). The Hamiltonian \( H \) for a plasmonic cavity mode is generally expressed with the vector potential \( \mathbf{A} (r, t) \) as

\[
H = \frac{1}{2} \int \int \left[ \frac{\partial \mathbf{E}(r, t)}{\partial t} \times \mathbf{B}(r, t) - \mathbf{A}(r, t) \right] \mathrm{d}V,
\]

where \( \mathbf{B} = -\mathbf{\nabla} \times \mathbf{E} \). By introducing the normalized electric field \( \mathbf{f}^+(r) \), satisfying \( \int \int \left[ \frac{\partial \mathbf{E}(r, t)}{\partial t} \right] \mathrm{d}V \mathbf{f}^+(r) \mathrm{d}r = 1 \) and \( \mathbf{E}^+(r, t) = \mathbf{D}^+(r) e^{i \omega t} \), where \( D \) is the amplitude coefficient, \( \mathbf{A} \) is expressed as

\[
\mathbf{A} (r, t) = \frac{1}{D} \mathbf{f}^+(r) e^{i \omega t} + \text{c.c.},
\]

where c.c. is the complex conjugate of the first term. By using the mode volume \( V_{\text{mode}} \equiv \int \int \left[ \frac{\partial \mathbf{E}(r, t)}{\partial t} \mathbf{E}^+(r, t) \right] \mathrm{d}V / \max \left[ \mathbf{E}(r, t) \mathbf{E}^+(r, t) \right] \), we obtain

Next, 2\( W_{\text{EE}} \) = \( \int \int \left[ \frac{\partial \mathbf{E}(r, t)}{\partial t} \mathbf{E}^+(r, t) \right] \mathrm{d}V \mathbf{f}^+(r) \mathrm{d}r = |D|^2 \), and \( 2\mathbf{W}_{\text{BB}} = \int \int \left[ (1/\mu_0) \mathbf{E} \times \mathbf{B} \right] \mathrm{d}V \mathbf{f}^+(r) \mathrm{d}r \). By introducing the parameter \( \Theta \equiv \mathbf{W}_{\text{BB}} / \mathbf{W}_{\text{EE}} \), we obtain \( 1/(1 + \Theta) = \mathbf{W}_{\text{EE}} / (\mathbf{W}_{\text{EE}} + \mathbf{W}_{\text{BB}}) \) and \( \Theta / (1 + \Theta) = \mathbf{W}_{\text{BB}} / (\mathbf{W}_{\text{EE}} + \mathbf{W}_{\text{BB}}) \). For quantization, \( D e^{-i \omega t} \) and \( D e^{i \omega t} \) in Eq. (8) are replaced with the variables \( p \) and \( q \), which are defined as

\[
p(t) = \sqrt{1 + \Theta D} \cos(\omega t),
\]

\[
q(t) = \sqrt{1 + \Theta D} \sin(\omega t) / \omega.
\]

From Eqs. (7)–(10), we find that \( H \) satisfies the following Hamilton equations: \( \dot{p} = (p^2 + \omega^2 q^2) / 2 \), \( \dot{q} = p \), and \( \dot{\mathbf{H}} / \partial q = \omega^2 q = -\mathbf{p} \). Upon applying canonical field quantization, the classical variables \( p \) and \( q \) are identified with the quantum operators \( \hat{Q} \) and \( \hat{P} \), and introducing the annihilation operator \( \hat{a} \) defined as \( \hat{a} \mathbf{E}^+ \equiv (\omega \hat{Q} + i \hat{P}) / \sqrt{2\hbar} \) and the creation operator \( \hat{a} \mathbf{E} \equiv (\omega \hat{Q} - i \hat{P}) / \sqrt{2\hbar} \), we obtain the following quantized fields [34]:

\[
\hat{E}_\mathbf{A} = \frac{\omega \hat{Q} + i \hat{P}}{\sqrt{2\hbar}}, \quad \hat{E}_\mathbf{B} = \frac{\omega \hat{Q} - i \hat{P}}{\sqrt{2\hbar}}.
\]
where $H.c.$ is the Hermitian conjugate of the first term.

Using Eq. (11a), $\langle N | \hat{E}^2 | N \rangle$ is expressed as

$$\langle N | \hat{E}^2 | N \rangle = \frac{2}{1 + \Theta} \left( \frac{\hbar}{\omega} \right)^2 \epsilon_0 \bar{W} \left( \frac{N + 1}{2} \right) \left( 1 - \frac{\delta \rho}{\delta \mu} \right) \left( | \hat{T}^+ |^2 \right)$$

in which $2/(1 + \Theta) = (2 \bar{W}_{EE}/W)$ shows the relative enhancement of the electric energy.

### B. Spontaneous Emission Enhancement by SPP

To analyze the enhanced SE, we assume a two-level system, and the interaction between the emitter and SPP electric field is expressed as $V = -\mu \cdot \hat{E}$, where $\mu$ is the electric dipole near the metal surface. The emission rate into a single SPP mode at $\omega$ is $\Gamma_{SPP} = \frac{2}{\pi \kappa} \left( \frac{\omega_0}{\hbar} \right) \mu \cdot \mathbf{f}^+(r) S(\Omega, Q)$, where $\Omega$ and $r$ are the frequency and position of the dipole, respectively, and $S(\Omega, Q)$ is the Lorentzian spectrum with the $Q$ factor. The Purcell factor $F$ is $F = \frac{\Gamma_{SPP}}{\Gamma_{SE}}$, where $\Gamma_{SE}$ is the intrinsic SE rate ($= n \mu^2 Q^2/3 \pi \epsilon_0 c^3$) in a bulk host material. When $\mu \cdot \mathbf{f}^+(r) = \mu \times \mathbf{f}^+(r)$ and $\omega = \Omega$, $F = \frac{2}{\pi \kappa} \left( \frac{\omega_0}{\hbar} \right) \left( \lambda n^3 Q/V_{mode} \right)$. For a dipole coupled to an SPP near the metal surface, $2/(1 + \Theta) \approx 2$, and $F$ becomes twice as large as the conventional value given by $F = (3/4 \pi^2) \left( \lambda n^3 Q/V_{mode} \right)$.

Figure 6 shows the measured lifetime of a fluorescent material (Eu compound) lying in front of an Au plate and the theoretical lifetime ($\Gamma^{-1} = \gamma^{-1}[(\Gamma_{SPP} + F_{non-SPP})/2]^{-1}$), where $\gamma$ and $\eta$ are the intrinsic decay rate of the fluorescent material without the metal plate and the quantum yield ($= \eta/\gamma$, respectively; $\Gamma_{non-SPP}$ is the emission rate into SPP modes and averaged over the randomly oriented emitters; $\Gamma_{SPP}$ is the same but for non-SPP modes).

![Fig. 6](image)

**Fig. 6.** Comparison between theoretically estimated lifetime with $2/(1 + \Theta) = 1.60$ (solid line) and measured lifetime (circles) of optically excited Eu compound in Au dielectric (fatty acid) layers, which were taken from [8]. The dashed line gives the theoretical values with $2/(1 + \Theta) = 1$. The error bar for the measured lifetime is $\pm 4\%$ [8,10].

The error bar for the theoretical lifetime (solid line) shows inaccuracies of $\gamma^{-1}$ and $\eta$; $\gamma^{-1} = 536 \pm 8 \mu s$ and $\eta = 0.688 \pm 0.090$. The dashed line has the same fitting errors as shown by the error bars in the solid line.

In this theoretical estimation, the SPP decay due to the absorption in Au was considered. The details of calculation and parameters for the theoretical estimate are summarized in Appendix B. In Fig. 6, the lifetime estimated without EM imbalance; namely, $2/(1 + \Theta) = 1$ (dashed line), slightly deviates from the measured lifetime (circles) with a 30% difference at most, whereas our estimate with the imbalance (solid line) fits much better.

### C. Nonlinearity Enhancement by Surface Plasmon Polaron

This section shows that the effects of the EM imbalance increase in nonlinear multiphoton processes. We assume coupled plasmonic cavities and analyze the double resonance with nonlinear coupling coefficients $\beta_1$ and $\beta_2$ by using the coupled-mode theory [28]. For simplicity, we consider a nonlinearity only in a dielectric around the cavities. We first evaluate the frequency change $\delta \omega$ induced by a small change in $\epsilon$ of the dielectric. In a plasmonic cavity, the electromagnetic field ($\mathbf{E}^+, \mathbf{B}^+$) and electric polarization $\mathbf{P}^+$ satisfy the following equation [37]:

$$\omega^2 \iint \mathbf{d}r \left( | \mathbf{B}^+ \cdot \epsilon_0 |^2 + (\frac{\delta \rho}{\delta \mu}) \mathbf{P}^+ \right) + \left( \frac{\delta \rho}{\delta \mu} \right) \left( \mathbf{P}^+ \cdot \mathbf{B}^+ \right)$$

where $\epsilon = \epsilon_{\infty} = 1$ and $\delta(\rho) = 1$ in a metal, $\epsilon = n^2$ and $\delta(\rho) = 0$ in a dielectric, and the superscript “−” is the complex conjugate of the same with “+”. Consider a small change $\epsilon \rightarrow \epsilon + \delta \epsilon$ and $\omega \rightarrow \omega + \delta \omega$; in a first-order approximation [38], we obtain

$$\delta \omega \omega = -\int_{\text{in dielectric}} \delta \epsilon \epsilon_0 | \mathbf{E}^+ |^2 \mathbf{d}r \left( \frac{1}{\mu_0} | \mathbf{B}^+ \cdot \epsilon_0 |^2 + \left( \frac{\delta \rho}{\delta \mu} \right) \mathbf{P}^+ |^2 \right)$$

By using $\Theta$, which is

$$\Theta = \frac{\int | \mathbf{B}^+ |^2 \mathbf{d}r}{\left( \mathbf{E} \cdot \epsilon_0 | \mathbf{E}^+ |^2 + \left( \frac{\delta \rho}{\delta \mu} \right) \mathbf{P}^+ |^2 \right) \mathbf{d}r}$$

and $\epsilon_0 | \mathbf{E}^+ |^2 + \left( \frac{\delta \rho}{\delta \mu} \right) \mathbf{P}^+ |^2 \mathbf{d}r = \epsilon_0 | \mathbf{E}^+ |^2 + \left( \frac{\delta \rho}{\delta \mu} \right) \mathbf{P}^+ |^2 \mathbf{d}r = \mathbf{E} | \mathbf{E}^+ |^2 | \mathbf{E}^+ |^2 \mathbf{d}r$, we obtain

$$\frac{\delta \omega \omega}{\omega} = \frac{2}{1 + \Theta} \int_{\text{in dielectric}} \delta \epsilon \epsilon_0 | \mathbf{E}^+ |^2 \mathbf{d}r$$

where the right-hand side is multiplied by a factor $2/(1 + \Theta)$ in comparison with the conventional formula [38].

In accordance with [28] (also see Appendix C), by introducing the optical susceptibility $\chi_{ijk}$ in the dielectric and relating $\delta \mathbf{E}^+$ to the electric polarization induced by $\chi_{ijk} \beta_1$ and $\beta_2$ are expressed as
where $\mathbf{u}_l^+ = (u^+_{l,1}, u^+_{l,2}, u^+_{l,3})$ is the normalized electric field of mode $l$ ($l = 1, 2$) and is defined as $\mathbf{u}_l^+ = \mathbf{E}_l^+ / \sqrt{|\varepsilon_0\varepsilon|\mathbf{E}_l^+|d\mathbf{r}|}$. Note that $\mathbf{u}_l^+$ is the complex conjugate of $\mathbf{u}_l^-$. $\Theta_l$ is that for $\mathbf{u}_l^+$. As $\Theta_l$ approaches unity, the term $\sqrt{2/(1 + \Theta_l)}$ also approaches unity, and Eqs. (17a) and (17b) give the known formulas without the EM imbalance [28].

Figure 7(a) shows a model of two metal spheres with diameter $2R$ and the center distance $a$ set at $0.10 \times 2\pi c/\omega_{p,1}$ and $0.12 \times 2\pi c/\omega_{p,1}$, and different plasma frequencies $\omega_{p,1}$ and $\omega_{p,2} = 2\omega_{p,1}$ [39]. Figure 7(b) shows the results of Eqs. (17a) and (17b) calculated for SPPs excited around this model. The values estimated by using the formulas without EM imbalance [28] are shown in parentheses. The values with EM imbalance are almost six times greater than those without EM imbalance.

5. CONCLUSION

This study reveals that the magnetic field components in SPPs induced by electric fields normal and parallel to a metal surface partly cancel each other, resulting in EM imbalance and slow propagation of the SPPs. This knowledge is valuable for understanding light propagation in plasmonic waveguides. In the theoretical analysis of the Purcell factor near a plasmonic structure and of the enhanced nonlinearity in coupled plasmonic cavities, we show that the EM imbalance must be considered to avoid underestimating the interaction. Although we studied only two cases of field-matter interactions, the EM imbalance should have several times greater effects in any multiphoton interaction.

We would finally suggest three studies of interest, relevant to the EM imbalance. The first one is that the electric field enhancement and slow propagation can increase the sensitivity in sensing; dielectric and metal-included multilayers have been compared for sensing applications [40]. The enhanced nonlinearities can increase the sensitivity in SPP-enhanced Raman scattering and other nonlinear sensing, although they have not yet been discussed. The second one is regarding assembled SPP nanocavities. The energy exchange between neighboring cavities may be decreased by the reduced magnetic field. Because that energy exchange deviates their resonance frequencies [41], the modification of the EM imbalance can be a scheme to design assembled cavities. The third one is the further investigation of SPP’s radiative decay rate. SPP modes in a metal sphere have small radiative decay rates [32]. Only low energy dipole or quadrupole modes contribute to the far-field radiation in the SPP excited around a metal nanoplat [42]. We expect the small energy flux induced by the reduced magnetic field to be related to such small radiative decay rates.

APPENDIX A: SPP AROUND A SMALL METAL SPHERE

Electromagnetic fields around a metal sphere have been theoretically analyzed in the spherical coordinates $(r, \theta, \phi)$ [43]. For SPPs, connecting the electromagnetic fields inside and outside a metal sphere with radius $R$, and, with the boundary conditions of $B_{1,\phi}^+$ and $E_{1,\phi}^+$, we obtain

$$B_{1,\phi}^+ = (1 + 2)^{1/2} (k R) j_1(k_m R) \frac{d}{d\theta} P_1(\cos \theta),$$

$$E_{1,\phi}^+ = (1 + 2)^{1/2} (k R) j_1(k_m R) \frac{d}{d\theta} P_1(\cos \theta),$$

inside the metal ($r < R$), and

$$B_{1,\phi}^- = (1 + 2)^{1/2} (k R) j_1(k_m R) \frac{d}{d\theta} P_1(\cos \theta),$$

$$E_{1,\phi}^- = (1 + 2)^{1/2} (k R) j_1(k_m R) \frac{d}{d\theta} P_1(\cos \theta),$$

outside of the metal ($r > R$), where $k^2 = n^2 \mu_0 \omega^2$ and $k_m^2 = \varepsilon_m \mu_0 \omega^2$. The functions $\beta^{(1)}(x), j_1(x)$, and $P_1(x)$ are the first spherical Hankel function, first spherical Bessel function, and Legendre polynomial, respectively. The frequency of mode $l$ is obtained by solving the boundary condition of $\text{Re}(E_{1,\phi}^+)$ at $r = R$ neglecting the small discontinuity of $\text{Im}(E_{1,\phi}^-)$. $\text{Im}(E_{1,\phi}^-)/\text{Re}(E_{1,\phi}^+)$ is less than 0.003 for an Au sphere with $R < 10$ nm.

APPENDIX B: FLUORESCENCE LIFETIME OF MATERIAL LYING IN FRONT OF A METAL PLATE

Considering the relaxation of an excited fluorescent material through emission into SPP and non-SPP modes, and, through nonradiative processes, the lifetime $\Gamma^{-1}$ of the emitter is

$$\Gamma^{-1} = \gamma^{-1} \times [(\bar{F}_{\text{SPP}} + \bar{F}_{\text{non-SPP}}) \eta + (1 - \eta)]^{-1}.$$
an accuracy of a few tenths of a percent [9]. The authors estimated this numerical factor was obtained by solving the complex dispersion equation [16]. In the calculation of $F_{p,∥}$, $F_{∥}$, and $F_{p,∥}$, we assumed an Au/dielectric interface, and the emission into dielectric modes above the light line ($\omega > c k / n$) was considered. The parameter values for estimating $\Gamma^{-1}$ in Fig. 6 were taken from [10,29]: $n_{\text{Au}} = 0.215 + i 3.22$ for the Eu$^{3+}$ emission wavelength of $\lambda = 612$ nm (equivalently, $\lambda_d = 181$ nm, $c/\gamma_{\text{damp}} = 1$, and $c/\gamma_{\text{damp}} = 797$ nm, where $\gamma_{\text{damp}}$ is the damping rate of light), and $n = 1.5$ for the dielectric material. For the fluorescent material, $\gamma^{-1} = 536 \pm 8$ ps and $\eta = 0.688 \pm 0.090$, which were estimated by fitting the measured lifetimes at $d > 200$ nm in Fig. 6, where emission into non-SPP modes is dominant without influence of the factor $2/(1 + \Theta)$. The value of $2/(1 + \Theta)$ is theoretically estimated by the formula presented in Section 2; $2/(1 + \Theta) = 1.60$

The lifetime measurement of Eu$^{3+}$ compound on a fatty acid/mirror structure was conducted by Drexhage [8,9] and by Kuhn et al. [7], for several types of mirrors, including Au, Ag, and Cu plates, and a fatty acid/air interface; the measured lifetimes in Fig. 6 were taken from Drexhage’s work [8]. In their studies, the fatty acid layers were deposited by using the Langmuir–Blodgett technique, their thickness achieved within an accuracy of a few tenths of a percent [9]. The authors estimated that the inaccuracy of the measured lifetimes was within 3%–4% [10]. Details of the measurement and fluorescence decay curve can be found in [44]. The measured lifetimes were theoretically analyzed in the classical electrodynamics theory by Chance et al. [10] and Barnes [11]. The theoretical lifetime, which was numerically estimated from a decay rate of the classical electric dipole moment, induced by the electromagnetic field excited at the dipole position, almost agreed with the measured one, with parameters $\gamma^{-1}$ and $\eta$, and random dipolar orientation. It should be noted that their theoretical lifetime includes the emission enhancement effect due to EM imbalance, although it cannot be separated from other enhancement effects by their analysis method [16]. In this study, we used the quantum analysis with the modal expansion, in which the emission enhancement due to EM imbalance is explicitly expressed by the factor $2/(1 + \Theta)$.

### APPENDIX C: DEVIATIONS OF $\beta_1$ AND $\beta_2$ FOR COUPLED MODES

Relating $\delta|E|^2 = (E^* \cdot \delta P^* + \delta E' \cdot \delta P) / 2$ in Eq. (16) to the nonlinear polarization $\delta P_p = \sum_{i+j+k} \epsilon_{0 \epsilon_{ij}} E_i^* E_j E_k^*$, where $E_i^*$ is the electric field of mode $i$, and leaving only the terms satisfying $\alpha_2 = 2 \alpha_1$, we obtain

$$\delta \alpha_2 = \frac{2}{1 + \Theta} \frac{\int_{\text{in,dielectric}} \sum_{i+j+k} \epsilon_{0 \epsilon_{ij}} E_i^* E_j E_k^* |E_i^2| \, dr + C}{\alpha_1^2}$$

(C1)

Equation (17b) is derived by comparing Eq. (C1) to the corresponding formula for dielectric cavity modes in [28], which we multiplied by $\sqrt{2/(1 + \Theta)} (I = 1, 2)$, so that $|\alpha_j|^2$ represents the electromagnetic energy of mode $j$. Note that $[I (1 + \Theta)]^2 / 2 \int_{\text{in,dielectric}} \sum_{i+j+k} \epsilon_{0 \epsilon_{ij}} E_i^* E_j E_k^* |E_i|^2 \, dr$ is the electromagnetic energy.

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