Hermitian description of localized plasmons in dispersive dissipative subwavelength spherical nanostructures

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The canonical quantization procedure in dispersive and lossy media assumes the eigenstates of the system to be collective excitations of the electromagnetic field and reservoir degrees of freedom. In this paper, we show that in low loss limit, the collective plasmonic modes in a quasistatic approximation separate from the reservoir. As an example, we consider the localized surface plasmon on a spherical metallic nanoparticle in a vacuum and find the macroscopic longitudinal electric near field per plasmon in the cases of gold and silver. Using our canonical approach, we calculate the correction to the electric near field per plasmon obtained from phenomenological quantization. The canonical conjugated variable to the electric near field is determined.

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I. INTRODUCTION

The latest achievements in nanotechnology make it possible to manufacture subwavelength metallic structures, which can be used for plasmon excitation [1,2]. The amplification of plasmons on such subwavelength structures allows the creation of nanolasers [3,4], spasers [5,6], and subwavelength optical transition lines [7–9]. Therefore, the problem of interaction of the plasmons with molecules [10–12], semiconductor medium [13,14], quantum dots [13], and quantum wells [14] is important. In many cases, only a few plasmon quanta are excited [15–20] on the structure, which is why the quantum properties of plasmons play a considerable role [21–24]. One of the main quantum characteristics of the systems is the electric field per plasmon. This quantity appears to be valuable for such key characteristics of plasmonic structures as the interaction constant between the field and the matter [25] and threshold pump level [6].

The quantum description of an electromagnetic field was developed in the 20th century [26,27]; nowadays it is the main study approach for the description of numerous physical phenomena. The most convenient way to quantize the electromagnetic field in a vacuum is to use the Coulomb gauge to eliminate the longitudinal component of the electric field [28]. This is possible due to Maxwell's first equation in vacuum $\mathrm{div}\mathbf{E} = -\Delta U = 0$, and the absence of the canonical conjugate variable to scalar potential U. However, in the medium, the longitudinal electromagnetic field is derived by the equation $\varepsilon_0 \Delta U = \mathrm{div}\mathbf{P}$, where $\mathbf{P}$ is the total medium polarization, so it cannot be eliminated in the same way as in a vacuum. In macroscopic electromagnetic field quantization, the problem has not been investigated appropriately [29–31]. The most consistent approach is to quantize the medium polarization and the field simultaneously [32–34]. This approach demands the choice of a model of the medium. One of the simplest and most usable models for this purpose is the Lorentz model [35]. It assumes that the medium consists of dumped harmonic oscillators. Their relaxation is provided by including additional degrees of freedom (reservoir) [36]. The Hamiltonian of the system "field + dipole oscillators + reservoirs" is Hermitian, and its quantization can be performed in the standard way by the introduction of creation and annihilation operators. The eigenmodes of the system are collective oscillations of field and medium and can be determined by the Fano diagonalization method [36,37]. First, the described procedure was used for the bulk medium [32–34], then it was generalized to nonuniform media [37]. To obtain the exact solution in the nonuniform case, it is necessary to use the Green functions formalism and the noise current approach [38–40] which was justified in [37] from the first principles. However, it is very difficult to give the physical interpretation of each mode of the system, and to separate the field modes from the system. As a result, this approach cannot be applied to find the number of excited plasmons or the electric near field per plasmon of plasmonic nanoparticle.

The widely used phenomenological approach [41–44] to plasmon quantization is much simpler but not canonical. It treats the localized plasmons as the harmonic oscillators whose eigenfrequencies are the frequencies of the plasmon resonances and determines the electric field per plasmon through the specific normalized condition for each plasmon mode. Namely, the total electric field energy per plasmon is supposed to be equal to the quantum of the oscillator's energy. This condition seems to be reasonable, although it has not been derived from the canonical procedure. Therefore, the phenomenological quantization has some drawbacks. In particular, there is no way to describe the Joule losses consistently. For example, the near electric field per plasmon obtained from this method does not depend on the imaginary part of the permittivity. Therefore, the canonical verification of the phenomenological plasmon quantization method is a current and important problem.

The paper is organized in the following way. In Sec. II we describe the model. In Sec. III we canonically quantize the plasmons, describing the permittivity through the Lorentz model using the approach of [32]. The difficulty here is the system under consideration is inhomogeneous. Therefore we cannot treat the longitudinal and the transverse electromagnetic fields independently as was performed in Ref. [32]. We show that the electric field per plasmon is fully described by the permittivity defined from the solution for bulk medium. In Sec. IV, we show that in the low loss limit, the quantum of plasmon electric field has the same value as the quantum obtained from phenomenological theory [41–44]. Also, we find the correction to the quantity obtained from phenomenological theory. We find in the Coulomb gauge the oscillations of medium subsystem response for quantization of scalar potential, separately we quantize the vector potential corresponding to photon subsystem. These quantum subsystems interact due to excitation of plasmonic resonances in the nanosphere. The excitation of one of the subsystems causes the excitation of the other. This fact, as we show, relates to the retardation character of the electromagnetic field outside the nanosphere. We derive a canonically conjugated variable to electric field. Finally, we replace the model permittivity by the real values for gold and silver to obtain the electric near field per plasmon for real materials.

II. DESCRIPTION OF THE MODEL

As mentioned earlier, to quantize the electromagnetic field in a medium, it is necessary to consider the medium as a part of the system "field + medium oscillators + reservoir." We consider a subwavelength sphere with radius R consisting of Lorentz oscillators, placed in a vacuum. The Lagrangian of the system is

$$\begin{aligned} L = &\int d^3\mathbf{r}\left\{\frac{\varepsilon_0[\dot{\mathbf{A}}(\mathbf{r},t)+\mathrm{grad}U(\mathbf{r},t)]^2}{2}-\frac{[\mathrm{rot}\mathbf{A}(\mathbf{r},t)]^2}{2\mu_0}\right\}+\int_{r<R} d^3\mathbf{r}\left\{\kappa\frac{\dot{\mathbf{P}}(\mathbf{r},t)^2}{2}-\kappa\omega_{\mathbf{P}0}^2\frac{\mathbf{P}(\mathbf{r},t)^2}{2}\right\}\\ &+\int_{r<R} d^3\mathbf{r}\left\{\int_0^\infty d\Omega\left[\frac{\dot{\mathbf{Y}}_{\mathbf{P}}(\mathbf{r},\Omega,t)^2}{2}-\Omega^2\frac{\mathbf{Y}_{\mathbf{P}}(\mathbf{r},\Omega,t)^2}{2}\right]\right\}\\ &+\int_{r<R} d^3\mathbf{r}\{U(\mathbf{r},t)\mathrm{div}[\mathbf{P}(\mathbf{r},t)]+\dot{\mathbf{P}}(\mathbf{r},t)\mathbf{A}(\mathbf{r},t)\}-\int_{r<R} d^3\mathbf{r}\left\{\int_0^\infty d\Omega[V_{\mathbf{P}}(\Omega)\mathbf{P}(\mathbf{r},t)\dot{\mathbf{Y}}_{\mathbf{P}}(\mathbf{r},\Omega,t)]\right\}, \end{aligned} \tag{1}$$

where $\mathbf{A}(\mathbf{r},t)$ is vector potential, $U(\mathbf{r},t)$ is scalar potential, $\mathbf{P}(\mathbf{r},t)$ is induced medium polarization density, $\omega_{\mathbf{P}0}$ is the frequency of the dipole harmonic oscillators, κ is the ratio of the mass and charge density of the dipole harmonic oscillators, $\mathbf{Y}_{\mathbf{P}}(\mathbf{r},\Omega,t)$ are reservoir variables (e.g., phonons), and $V_{\mathbf{P}}(\Omega)$ is a coupling constant between dipole harmonic oscillators and reservoir. The physical meaning of each part of the Lagrangian (1) is as follows:

$$\int d^3\mathbf{r}\left\{\varepsilon_0\frac{[\dot{\mathbf{A}}(\mathbf{r},t)+\mathrm{grad}U(\mathbf{r},t)]^2}{2}-\frac{1}{\mu_0}\frac{[\mathrm{rot}\mathbf{A}(\mathbf{r},t)]^2}{2}\right\}$$

is the Lagrangian of electromagnetic field;

$$\int_{r<R} d^3\mathbf{r}\left\{\kappa\frac{\dot{\mathbf{P}}(\mathbf{r},t)^2}{2}-\kappa\omega_{\mathbf{P}0}^2\frac{\mathbf{P}(\mathbf{r},t)^2}{2}\right\}$$

is the dipole harmonic oscillators part, which models the polarization;

$$\int_{r<R} d^3\mathbf{r}\left\{\int_0^\infty d\Omega\left[\frac{\dot{\mathbf{Y}}_{\mathbf{P}}(\mathbf{r},\Omega,t)^2}{2}-\Omega^2\frac{\mathbf{Y}_{\mathbf{P}}(\mathbf{r},\Omega,t)^2}{2}\right]\right\}$$

is the reservoir part, comprising a continuum of harmonic oscillators, which are used to model losses;

$$\int_{r<R} d^3\mathbf{r}\{U(\mathbf{r},t)\mathrm{div}[\mathbf{P}(\mathbf{r},t)]+\dot{\mathbf{P}}(\mathbf{r},t)\mathbf{A}(\mathbf{r},t)\}$$

is the interaction part, which includes interaction between the electromagnetic field and induced polarization. $-\mathrm{div}\mathbf{P}(\mathbf{r},t)$ and $\dot{\mathbf{P}}(\mathbf{r},t)$ are the polarization charge density and polarization current density, respectively. The final part

$$-\int_{r<R} d^3\mathbf{r}\left\{\int_0^\infty d\Omega[V_{\mathbf{P}}(\Omega)\mathbf{P}(\mathbf{r},t)\dot{\mathbf{Y}}_{\mathbf{P}}(\mathbf{r},\Omega,t)]\right\}$$

is the interaction part, which includes the interaction between induced dipole moments and the reservoir. The reservoir is modeled by a continuum of harmonic oscillators, which provide the dissipation of the energy of the dipole harmonic oscillators [36]. Indeed, when an exited harmonic oscillator is coupled to the continuum harmonic oscillators, which are not exited in the initial moment of time, the oscillators energy is fully transferred into the continuum harmonic oscillators [45–48]. The exited harmonic oscillator loses the energy, but the total energy of the system is conserved.

III. LOCALIZED PLASMONS QUANTIZATION

Following the standard approach to QED for nonrelativistic phenomena, we choose the Coulomb gauge $\mathrm{div}\mathbf{A}(\mathbf{r},t)=0$, so that the vector potential is purely a transverse field. Now we enter into momentum space and write it according to the Fourier transformation as

$$\mathbf{A}(\mathbf{r},t)=\sum_{\lambda=1,2}\int d^3\mathbf{k}e^{i\mathbf{k}\cdot\mathbf{r}}\mathbf{e}(\lambda,\mathbf{k})A(\lambda,\mathbf{k},t), \tag{2}$$

where the vectors $\mathbf{e}(\lambda,\mathbf{k})$ are polarization vectors that satisfy the following relations

$$\mathbf{k}\cdot\mathbf{e}(\lambda,\mathbf{k})=0,$$
$$\mathbf{e}(\lambda,\mathbf{k})\cdot\mathbf{e}^*(\lambda',\mathbf{k})=\delta_{\lambda\lambda'}.$$

We expand the variables $\mathbf{P}$, $\mathbf{Y}_{\mathbf{P}}$, scalar potential U, and electric near field $\mathbf{E}_{\mathrm{near}}=-\mathrm{grad}U$ in Laplace spherical harmonics.

$$\mathbf{P}(\mathbf{r},t)=\sum_{l,m}P_{lm}(t)\begin{cases}R\mathrm{grad}[(r/R)^l\Psi_{lm}(\theta,\varphi)], & r<R\\ 0, & r>R\end{cases}, \tag{3}$$

$$\mathbf{Y}_{\mathbf{P}}(\mathbf{r},\Omega,t)=\sum_{l,m}Y_{lm}(\Omega,t)\begin{cases}R\mathrm{grad}[(r/R)^l\Psi_{lm}(\theta,\varphi)], & r<R\\ 0, & r>R\end{cases}, \tag{4}$$

$$U(\mathbf{r},t)=\sum_{l,m}U_{lm}(t)\begin{cases}(r/R)^l\Psi_{lm}(\theta,\varphi), & r<R\\ (R/r)^{l+1}\Psi_{lm}(\theta,\varphi), & r>R\end{cases}, \tag{5}$$

$$\mathbf{E}_{\text{near}}(\mathbf{r},t) = -\sum_{l,m} E_{lm}(t)\begin{cases} R\,\text{grad}[(r/R)^l \Psi_{lm}(\theta,\varphi)], & r < R \\ R\,\text{grad}[(R/r)^{l+1}\Psi_{lm}(\theta,\varphi)], & r > R \end{cases}, \tag{6}$$

where $\Psi_{lm}(\theta,\varphi)$ are the spherical functions. Note that the electric near field $\mathbf{E}_{\text{near}} = -\text{grad}U$ is not the total electric field $\mathbf{E} = -\text{grad}U - \dot{\mathbf{A}}$. The set of the Laplace spherical harmonics is not the basis for the polarization $\mathbf{P}$ and the reservoir $\mathbf{Y_P}$. For an instant this decomposition does not include the whispering gallery modes. However, it describes all the modes in a quasistatic approximation [7,41,49]. Therefore the expansions (3–6) are not exact, but they have the small parameter R/λ [50,51], where λ is the characteristic wavelength in a vacuum. From the physical point of view, the expansion assumes that we can omit all retardation effects on the sphere scale R.

We insert expansions (2–6) into the Lagrangian (1) and obtain

$$\begin{aligned} L = {} & \frac{1}{2}\sum_{l,m}\left\{\kappa R^3 l \dot{P}_{lm}^2(t) - \kappa\omega_{\mathbf{P}0}^2 R^3 l P_{lm}^2(t)\right\} + \frac{1}{2}\sum_{l,m}\left\{\varepsilon_0 R(2l+1)U_{lm}^2(t) - 2R^2 l U_{lm}(t)P_{lm}(t)\right\} \\ & + \frac{1}{2}\sum_{l,m}\left\{\int_0^{+\infty} d\Omega\big[R^3 l \dot{Y}_{lm}^2(\Omega,t) - \Omega^2 R^3 l Y_{lm}^2(\Omega,t)\big]\right\} - \frac{1}{2}\sum_{l,m}\left\{\int_0^{+\infty} d\Omega[2R^3 l V_{\mathbf{P}}(\Omega)P_{lm}(t)Y_{lm}(\Omega,t)]\right\} \\ & + \sum_{\lambda=1,2}\int_{\text{half}} d^3\mathbf{k}\left\{\varepsilon_0|\dot{A}(\lambda,\mathbf{k},t)|^2 - \frac{1}{\mu_0}\mathbf{k}^2|A(\lambda,\mathbf{k},t)|^2\right\} + \sum_{\lambda=1,2}\sum_{l,m}\int d^3\mathbf{k}\{\dot{P}_{lm}R^3\Lambda_{lm}(\lambda,\mathbf{k})A(\lambda,\mathbf{k},t)\}, \end{aligned} \tag{7}$$

where $\Lambda_{lm}(\lambda,\mathbf{k}) = \oint_{r=R} d\mathbf{S}\{e^{i\mathbf{k}\cdot\mathbf{r}}\mathbf{e}(\lambda,\mathbf{k})\Psi_{lm}(\theta,\varphi)\}/R^2$ is a new interaction constant, which includes the interaction between the transverse vector potential and polarization current. The interaction is nonlocal in space because of the chosen Coulomb gauge [52]. We discuss this coupling term in the Appendix B. This interaction results in impossibility of independent consideration of longitudinal and transverse components of electromagnetic field as it was performed in [32]. Therefore more accurate consideration of transverse and longitudinal modes is required.

To obtain (7), we use the Gauss's flux theorem and the fact that function $U(\mathbf{r},t)$ is continuous and piecewise continuously differentiable. The "half" in the integration over $\mathbf{k}$ in (7) denotes that the integration is restricted to half the space (for example, $k_z > 0$). The integration over half space is sufficient because of $A^*(\lambda,\mathbf{k},t) = A(\lambda,-\mathbf{k},t)$, which results from the representation of the real function. Here and after we will associate the vector potential with the electromagnetic field in a vacuum, because in empty space only this term remains in the Lagrangian. We will associate the scalar potential with the near field, because it obeys the Poisson equation.

We join the medium oscillations and the electric near field into collective oscillations to obtain the plasmon. We use the Euler-Lagrange equation [53] for $U_{lm}(t)$ to eliminate it from the Lagrangian (7)

$$\frac{\delta L}{\delta U_{lm}(t)} = 0 \Rightarrow U_{lm}(t) = \frac{1}{\varepsilon_0}\frac{l}{2l+1}RP_{lm}(t) \tag{8}$$

and finally we obtain

$$\begin{aligned} L = {} & \sum_{l,m}\frac{\kappa R^3 l}{2}\left\{\dot{P}_{lm}^2(t) - \left[\omega_{\mathbf{P}0}^2 + \frac{1}{\varepsilon_0\kappa}\frac{l}{2l+1}\right]P_{lm}^2(t)\right\} + \sum_{l,m}\frac{R^3 l}{2}\left\{\int_0^{+\infty} d\Omega\big[Y_{lm}^2(\Omega,t) - \Omega^2 Y_{lm}^2(\Omega,t)\big]\right\} \\ & - \sum_{l,m} R^3 l\left\{\int_0^{+\infty} d\Omega[V_{\mathbf{P}}(\Omega)P_{lm}(t)\dot{Y}_{lm}(\Omega,t)]\right\} + \sum_{\lambda=1,2}\int_{\text{half}} d^3\mathbf{k}\left\{\varepsilon_0|\dot{A}(\lambda,\mathbf{k},t)|^2 - \frac{1}{\mu_0}\mathbf{k}^2|A(\lambda,\mathbf{k},t)|^2\right\} \\ & + \sum_{\lambda=1,2}\sum_{l,m}\int d^3\mathbf{k}\{R^3\Lambda_{lm}(\lambda,\mathbf{k})\dot{P}_{lm}(t)A(\lambda,\mathbf{k},t)\}. \end{aligned} \tag{9}$$

The elimination of the scalar potential leads to the change of the dipole harmonic oscillators frequencies. The change relates to the additional electrostatic forces. The particular value of the frequency change is defined by the shape of the body. Therefore, the surface plasmonic resonance frequencies differ from the frequency of the bulk plasmonic resonance [54–56].

The Lagrangian (9) can be used to obtain the canonical conjugate variables

$$\begin{aligned} Q_{\mathbf{P}lm}(t) &= \kappa R^3 l \dot{P}_{lm}(t) + \sum_{\lambda=1,2}\int d^3\mathbf{k}R^3\Lambda_{lm}(\lambda,\mathbf{k})A(\lambda,\mathbf{k},t), \\ Q_{\mathbf{Y}lm}(\Omega,t) &= R^3 l \dot{Y}_{lm}(\Omega,t) - R^3 l V_{\mathbf{P}}(\Omega)P_{lm}(t) \\ Q_{\mathbf{A}}(\lambda,\mathbf{k},t) &= \varepsilon_0\dot{A}(\lambda,\mathbf{k},t) \end{aligned} \tag{10}$$

and the Hamiltonian

$$H = \frac{1}{2}\sum_{l,m}\left\{\frac{Q^2_{\mathbf{P}lm}(t)}{\kappa R^3 l} + \kappa R^3 l\left[\omega^2_{\mathbf{P}0} + \frac{1}{\varepsilon_0\kappa}\frac{l}{2l+1} + \int_0^{+\infty} d\Omega V^2_{\mathbf{P}}(\Omega)\right]P^2_{lm}(t)\right\}$$
$$+\frac{1}{2}\sum_{l,m}\left\{\int_0^{+\infty} d\Omega\left[\frac{Q^2_{\mathbf{Y}lm}(\Omega,t)}{R^3 l} + \Omega^2 R^3 l Y^2_{lm}(\Omega,t)\right]\right\} + \frac{1}{2}\sum_{l,m}\left\{\int_0^{+\infty} d\Omega[2V_{\mathbf{P}}(\Omega)P_{lm}(t)Q_{\mathbf{Y}lm}(\Omega,t)]\right\}$$
$$+\sum_{\lambda=1,2}\int_{\text{half}} d^3\mathbf{k}\left\{\frac{|Q(\lambda,\mathbf{k},t)|^2}{\varepsilon_0} + \frac{1}{\mu_0}\mathbf{k}^2|A(\lambda,\mathbf{k},t)|^2\right\} - \sum_{\lambda=1,2}\sum_{l,m}\int d^3\mathbf{k}\left\{\frac{\Lambda_{lm}(\lambda,\mathbf{k})}{\kappa l}Q_{\mathbf{P}lm}(t)A(\lambda,\mathbf{k},t)\right\}$$
$$+\frac{1}{2}\sum_{l,m}\frac{R^3}{\kappa l}\left[\sum_{\lambda=1,2}\int d^3\mathbf{k}\Lambda_{lm}(\lambda,\mathbf{k})A(\lambda,\mathbf{k},t)\right]^2. \tag{11}$$

The first term of the Hamiltonian (11) is the energy of the collective medium oscillations, including the energy of scalar potential. This part of the Hamiltonian is described by two sets of variables $\{P_{lm}(t), Q_{\mathbf{P}lm}(t)\}$ and $\{Y_{lm}(\Omega,t), Q_{\mathbf{Y}lm}(\Omega,t)\}$.

It is not obvious which part corresponds to the plasmonic modes. The solution of the problem is to find the canonical transformation, which makes the medium part of the Hamiltonian diagonal. Such a transformation can be obtained by using the method of Fano diagonalization [32,36,37,57]:

$$P_{lm}(t) = \int_0^{\infty} d\Omega\left[\frac{2l+1}{l}\sqrt{\frac{2\varepsilon_0}{\pi R^3 l}}\frac{\sqrt{\mathrm{Im}\varepsilon(\Omega)}}{\left|\varepsilon(\Omega)+\frac{l+1}{l}\right|}\sqrt{\Omega}Z_{lm}(\Omega,t)\right]$$
$$Q_{\mathbf{P}lm}(t) = \kappa\int_0^{\infty} d\Omega\left[\frac{2l+1}{l}\sqrt{\frac{2R^3 l\varepsilon_0}{\pi}},\frac{\sqrt{\mathrm{Im}\varepsilon(\Omega)}}{\left|\varepsilon(\Omega)+\frac{l+1}{l}\right|}\sqrt{\Omega}Q_{\mathbf{Z}lm}(\Omega,t)\right], \tag{12}$$

where $Z_{lm}(\Omega,t)$ and $Q_{\mathbf{Z}lm}(\Omega,t)$ are new canonical conjugated variables, and $\varepsilon(\Omega)$ is the permittivity of the bulk medium, consisting of the same harmonic oscillators. The permittivity is derived in Appendix A. If at some frequencies the permittivity is negative, we may observe the localized surface plasmon. The canonical transformation (12) changes the Hamiltonian (11) to

$$H = \sum_{l,m}\int_0^{+\infty} d\Omega\left\{\frac{Q^2_{\mathbf{Z}lm}(\Omega,t)}{2} + \frac{\Omega^2 Z^2_{lm}(\Omega,t)}{2}\right\} + \sum_{\lambda=1,2}\int_{\text{half}} d^3\mathbf{k}\left\{\frac{|Q(\lambda,\mathbf{k},t)|^2}{\varepsilon_0} + \frac{1}{\mu_0}\mathbf{k}^2|A(\lambda,\mathbf{k},t)|^2\right\}$$
$$-\sum_{\lambda=1,2}\sum_{l,m}\int d^3\mathbf{k}\int_0^{\infty} d\Omega\left\{\frac{2l+1}{l}\sqrt{\frac{2R^3}{\pi l}}\frac{\sqrt{\Omega\varepsilon_0\mathrm{Im}\varepsilon(\Omega)}}{\left|\varepsilon(\Omega)+\frac{l+1}{l}\right|}\Lambda_{lm}(\lambda,\mathbf{k})Q_{\mathbf{Z}lm}(\Omega,t)A(\lambda,\mathbf{k},t)\right\}$$
$$+\sum_{l,m}\int_0^{+\infty} d\Omega\left\{\frac{(2l+1)^2}{\pi l^3}R^3\frac{\Omega\varepsilon_0\mathrm{Im}\varepsilon(\Omega)}{\left|\varepsilon(\Omega)+\frac{l+1}{l}\right|^2}\right\}\left[\sum_{\lambda=1,2}\int d^3\mathbf{k}\Lambda_{lm}(\lambda,\mathbf{k})A(\lambda,\mathbf{k},t)\right]^2. \tag{13}$$

As mentioned, the physical meaning of the first term is the energy of the collective medium oscillations, so we call it the localized plasmon energy. The second term can be called the energy of the electromagnetic field in a vacuum, because it is the only part of the Hamiltonian that remains in empty space. The third part of the Hamiltonian is the interaction energy between the electromagnetic field in a vacuum and plasmons. The final part of (13) which contains the $\mathbf{A}^2$ term is the minimal light-matter coupling Hamiltonian [58]. Note that this term is not present in Lagrangian (9) and appears after transition to Hamiltonian by using appropriated generalized momentum conjugated by generalized coordinate, see Ref. [53]. This term is substantially connected with interaction of electromagnetic field with medium despite that the "plasmon" operator is absent. The multiplier proportional to $\mathrm{Im}\varepsilon$ and Λ_{lm} indicates this fact. This term is absent when medium is absent.

In Fig. 1 we show the dependence of the interaction constant between the electromagnetic field in a vacuum and plasmons on the frequency. The form of the permittivity

$$\varepsilon(\omega) = 1 + \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\gamma\omega}, \tag{14}$$

which is used in Fig. 1 can be obtained from the Eq. (A7) if the function $V_{\mathbf{P}}(\Omega)$ is set to be the constant $\sqrt{2\gamma\kappa}$. One can see the resonance behavior of the interaction constant when $\mathrm{Re}(\varepsilon(\omega)) + (l+1)/l = 0$.

The canonical quantization of the photons and plasmons can be performed in a standard way by replacing Poisson brackets by commutators [28]

$$[\hat{P}_{lm}(t), \hat{Q}_{\mathbf{P}l'm'}(t)] = i\hbar\delta_{ll'}\delta_{mm'}$$
$$[\hat{Y}_{lm}(\Omega,t), \hat{Q}_{\mathbf{Y}l'm'}(\Omega',t)] = i\hbar\delta(\Omega-\Omega')\delta_{ll'}\delta_{mm'}$$
$$[\hat{A}(\lambda,\mathbf{k},t), \hat{Q}(\lambda',\mathbf{k}',t)] = i\hbar\delta(\mathbf{k}-\mathbf{k}')\delta_{\lambda\lambda'}$$
$$[\hat{Z}_{lm}(\Omega,t), \hat{Q}_{\mathbf{Z}l'm'}(\Omega',t)] = i\hbar\delta(\Omega-\Omega')\delta_{ll'}\delta_{mm'} \tag{15}$$

When the procedure is finished, the variables can be written in terms of the creation and annihilation operators:

$$\hat{A}(\lambda,\mathbf{k},t) = \sqrt{\frac{\hbar}{2\varepsilon_0 ck}}[\hat{a}^+(\lambda,-\mathbf{k},t) + \hat{a}(\lambda,\mathbf{k},t)],$$

$$\hat{Q}(\lambda,\mathbf{k},t) = i\sqrt{\frac{\hbar ck}{2\varepsilon_0}}[\hat{a}^+(\lambda,-\mathbf{k},t) - \hat{a}(\lambda,\mathbf{k},t)], \tag{16}$$

$$\hat{Z}_{lm}(\Omega,t) = \sqrt{\frac{\hbar}{2\Omega}}[\hat{d}^+_{lm}(\Omega,t) + \hat{d}_{lm}(\Omega,t)],$$

$$\hat{Q}_{\mathbf{Z}lm}(\Omega,t) = i\sqrt{\frac{\hbar\Omega}{2}}[\hat{d}^+_{lm}(\Omega,t) - \hat{d}_{lm}(\Omega,t)], \tag{17}$$

where $\hat{a}^+(\lambda,\mathbf{k},t)$ and $\hat{a}(\lambda,\mathbf{k},t)$ are photon creation and annihilation operation of wave vector $\mathbf{k}$ and polarization λ, and $\hat{d}^+_{lm}(\Omega,t)$ and $\hat{d}_{lm}(\Omega,t)$ are plasmon creation and annihilation operators of mode lm and frequency Ω. The Hamiltonian can be written in terms of introduced operators as

$$\begin{aligned}\hat{H} =& \sum_{l,m}\int_0^{+\infty} d\Omega\hbar\Omega\hat{d}^+_{lm}(\Omega,t)\hat{d}_{lm}(\Omega,t) + \sum_{\lambda=1,2}\int d^3\mathbf{k}\hbar ck\hat{a}^+(\lambda,\mathbf{k},t)\hat{a}(\lambda,\mathbf{k},t) - i\sum_{\lambda=1,2}\sum_{l,m}\int d^3\mathbf{k}\int_0^{\infty} d\Omega\hbar\Omega \\ &\times \left\{\frac{2l+1}{l\sqrt{2\pi l}}R^{3/2}\frac{\sqrt{\mathrm{Im}\varepsilon(\Omega)}}{\left|\varepsilon(\Omega)+\frac{(l+1)}{l}\right|}[\hat{d}^+_{lm}(\Omega,t) - \hat{d}_{lm}(\Omega,t)]\left[\frac{\Lambda^*_{lm}(\lambda,-\mathbf{k})}{\sqrt{ck}}\hat{a}^+(\lambda,-\mathbf{k},t) + \frac{\Lambda_{lm}(\lambda,\mathbf{k})}{\sqrt{ck}}\hat{a}(\lambda,\mathbf{k},t)\right]\right\} \\ &+ \sum_{l,m}\int_0^{+\infty} d\Omega\hbar\Omega R^3\frac{(2l+1)^2}{2\pi l^3}\frac{\mathrm{Im}\varepsilon(\Omega)}{\left|\varepsilon(\Omega)+\frac{l+1}{l}\right|^2}\left\{\sum_{\lambda=1,2}\int d^3\mathbf{k}\left[\frac{\Lambda^*_{lm}(\lambda,-\mathbf{k})}{\sqrt{ck}}\hat{a}^+(\lambda,-\mathbf{k},t) + \mathrm{H.c.}\right]\right\}^2.\end{aligned} \tag{18}$$

This Hamiltonian describes the processes of emitting, scattering and absorbing photons by plasmons at the quantum level. It can be seen from the Hamiltonian (18) that the power of absorbed electromagnetic energy is proportional to the volume of the plasmon particle.

The polarization modes $\hat{P}_{lm}(t)$ can be obtained from (12) and (17)

$$\hat{P}_{lm}(t) = \int_0^{\infty} d\Omega\frac{2l+1}{l}\sqrt{\frac{\hbar\varepsilon_0}{\pi R^3 l}}\frac{\sqrt{\mathrm{Im}\varepsilon(\Omega)}}{\left|\varepsilon(\Omega)+\frac{l+1}{l}\right|} \times(\hat{d}^+_{lm}(\Omega,t) + \hat{d}_{lm}(\Omega,t)). \tag{19}$$

Substitution of (19) into (8) leads us to the quantum longitudinal electric near field of the plasmon:

$$\hat{E}_{lm}(t) = \int_0^{\infty} d\Omega\mathcal{E}_l(\Omega)(\hat{d}^+_{lm}(\Omega,t) + \hat{d}_{lm}(\Omega,t)), \tag{20}$$

where we define electric near field per plasmon on frequency Ω

$$\mathcal{E}_l(\Omega) = \sqrt{\frac{\hbar}{\pi R^3 l\varepsilon_0}}\frac{\sqrt{\mathrm{Im}\varepsilon(\Omega)}}{\left|\varepsilon(\Omega)+\frac{l+1}{l}\right|}. \tag{21}$$

Note that the idea of the calculation of the electric field per plasmon is similar to one used in the phenomenological quantization approach. Namely, using the commutation relations (15) we diagonalize the plasmon part of the full Hamiltonian (11). Then we set the quantum of the energy of the plasmon to be $\hbar\omega$ and obtain the dimensional prefactors. It is important that the expression (21) is valid for arbitrary losses.

The developed formalism allows us to calculate the electric near field per plasmon $\mathcal{E}_l(\Omega)$ [see (21)] for a sphere made from real gold and silver (Fig. 2). In other words the permittivity of gold and silver taken from Ref. [59] were inserted in Eq. (21). Then the quantity $\mathcal{E}_l(\Omega)$ was plotted as a function of frequency. One can see that these curves are Lorentz-like in the frequency region before the interband transitions.

IV. LOW LOSS LIMIT: PHENOMENOLOGICAL ELECTRIC NEAR FIELD PER PLASMON

In Sec. II we obtained the Hamiltonian (18) of the surface plasmon are in the limit $R/\lambda \ll 1$. In this section, we will show that the quantization procedure described above in the low loss limit, $\mathrm{Im}\varepsilon(\Omega) \ll \Omega(\partial\mathrm{Re}\varepsilon(\Omega)/\partial\Omega)$, leads to the same result for the electric near field per plasmon, as predicted by phenomenological theory.

When the imaginary part of permittivity tends to zero, the interaction between dipole harmonic oscillators and the reservoir tends to zero too. In this case, there is only one pair of annihilation and creation operators for each plasmon mode. As a result, the Hamiltonian of plasmon takes the following form:

$$\hat{H}_{\text{plasmons}} = \sum_{l,m}\hbar\omega_l\hat{d}^+_{0lm}(t)\hat{d}_{0lm}(t), \tag{22}$$

where ω_l is defined by the pole of (21), namely $\mathrm{Re}\varepsilon(\omega_l) = -(l+1)/l$. Here $\hat{d}_{0lm}(t)$ and $\hat{d}^+_{0lm}(t)$ are plasmon creation and annihilation operators, respectively, subscript 0 points to zero loss limit. The pole of (19) in the limit is close to the real axes, so that

$$\hat{P}_{0lm}(t) = P_{0lm}(\hat{d}^+_{0lm}(t) + \hat{d}_{0lm}(t)), \tag{23}$$

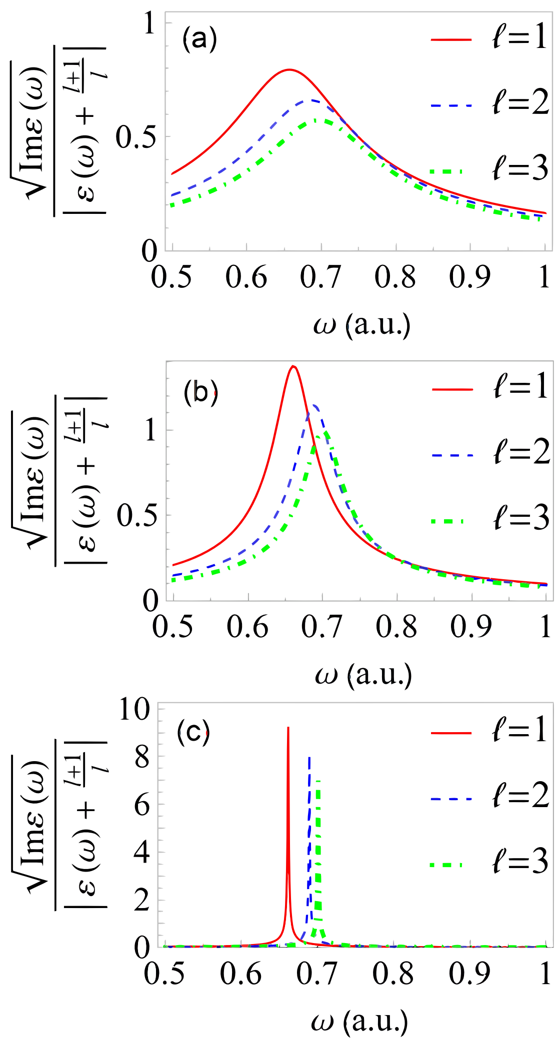

FIG. 1. Dependence of interaction constant on the frequency for different plasmon modes. The permittivity of the sphere has Lorentzian form $\varepsilon(\omega) = 1 + \omega_p^2/(\omega_0^2 - \omega^2 - i\gamma\omega)$ with $\omega_p/\omega_0 = 0.75$ and (a) $\gamma/\omega_0 = 0.3$, (b) $\gamma/\omega_0 = 0.1$, and (c) $\gamma/\omega_0 = 0.002$. Decrease of γ leads to decrease of the loss.

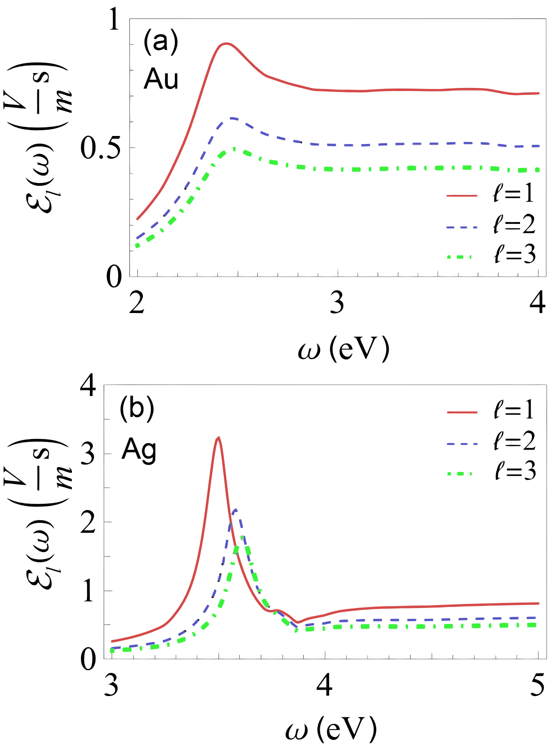

FIG. 2. Dependence of electric near field per plasmon Eq. (21) on frequency for different plasmon modes for (a) gold and (b) silver spheres. The permittivity data is taken from Ref. [59].

where P_{0lm} is a polarization per plasmon. Transferring (19) to (23) is nontrivial because integrating over frequencies in both expressions includes operators. To perform this transfer, we assume the commutation relation conservation condition

$$\left[\hat{P}_{lm}^{(-)}(t), \hat{P}_{lm}^{(+)}(t)\right]\Big|_{\frac{\mathrm{Im}\varepsilon(\omega)}{\omega\partial\mathrm{Re}\varepsilon(\omega)/\partial\omega}\ll 1} = \left[\hat{P}_{0lm}^{(-)}(t), \hat{P}_{0lm}^{(+)}(t)\right], \tag{24}$$

where $^{(+)}$ and $^{(-)}$ denote the annihilation and creation part of expressions (19) and (23). We substitute (23) and (19) into (24) and obtain

$$\begin{aligned} P_{0lm}^2 &= \int_0^\infty d\Omega \left[\left(\frac{2l+1}{l}\right)^2 \frac{\hbar\varepsilon_0}{\pi R^3 l} \frac{\mathrm{Im}\varepsilon(\Omega)}{\left(\mathrm{Re}\varepsilon(\Omega) + \frac{l+1}{l}\right)^2 + \mathrm{Im}\varepsilon(\Omega)^2}\right] \\ &\simeq \int_0^\infty d\Omega \left[\left(\frac{2l+1}{l}\right)^2 \frac{\hbar\varepsilon_0}{\pi R^3 l} \frac{\mathrm{Im}\varepsilon(\omega_l)}{(\partial\mathrm{Re}\varepsilon(\Omega)/\partial\Omega|_{\Omega=\omega_l})^2(\Omega-\omega_l)^2 + \mathrm{Im}\varepsilon(\omega_l)^2}\right] \\ &\simeq \left(\frac{2l+1}{l}\right)^2 \frac{\hbar\varepsilon_0}{R^3 l|\partial\mathrm{Re}\varepsilon(\Omega)/\partial\Omega|_{\Omega=\omega_l}} \left(1 - \frac{\mathrm{Im}\varepsilon(\omega_l)}{2\pi\omega_l|\partial\mathrm{Re}\varepsilon(\Omega)/\partial\Omega|_{\Omega=\omega_l}}\right). \end{aligned} \tag{25}$$

In the second equality we used the condition $\mathrm{Im}\varepsilon(\omega)/[\omega\partial\mathrm{Re}\varepsilon(\omega)/\partial\omega] \ll 1$. In the same way we can obtain

$$\hat{E}_{lm}(t) = \sqrt{\frac{\hbar}{|\partial\mathrm{Re}\varepsilon(\Omega)/\partial\Omega|_{\Omega=\omega_l} R^3 l\varepsilon_0}} \left(1 - \frac{\mathrm{Im}\varepsilon(\omega_l)}{2\pi\omega_l|\partial\mathrm{Re}\varepsilon(\Omega)/\partial\Omega|_{\Omega=\omega_l}}\right)(\hat{d}_{0lm}^+(t) + \hat{d}_{0lm}(t)). \tag{26}$$

Expression (26) in the absence of losses is equal to that obtained in the phenomenological approach [41–43]. The electric near field per plasmon is

$$E_0 = \sqrt{\frac{\hbar}{|\partial\mathrm{Re}\varepsilon(\Omega)/\partial\Omega|_{\Omega=\omega_l} R^3 l\varepsilon_0}}. \tag{27}$$

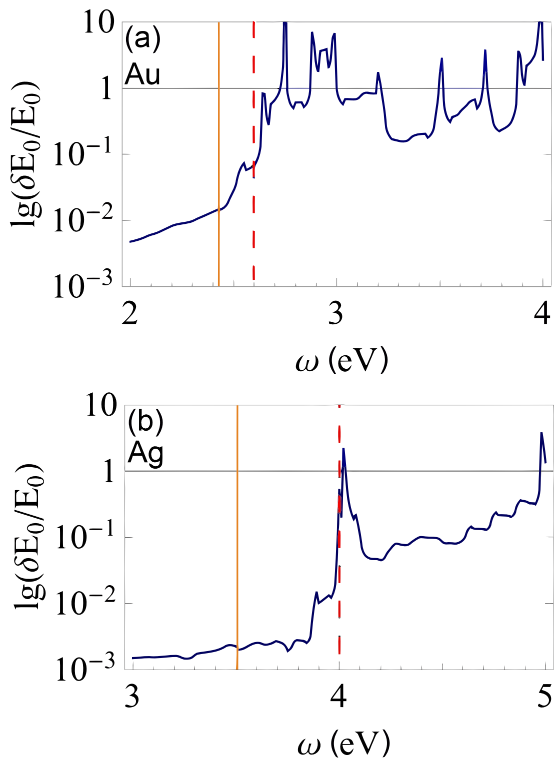

FIG. 3. Correction to the electric near field per plasmon due to the imaginary part of permittivity depending on frequency for (a) gold and (b) silver. The vertical dashed lines mark the interband transition frequency. The vertical solid lines mark the frequency of dipole plasmonic resonance. The permittivity data is taken from Ref. [59].

We recall that, in the phenomenological consideration, the near electric field per plasmon introduced by the equation

$$\varepsilon_0 \int d^3\mathbf{r} \frac{\partial(\omega \mathrm{Re}\varepsilon(\mathbf{r},\omega))}{\partial \omega}\bigg|_{\omega=\omega_l} |E(\mathbf{r})|^2 = \hbar\omega_l \tag{28}$$

leads to the same electric near field per plasmon.

We obtain the correction to the expression (27) due to the imaginary part of the permittivity by noting that from (26) we have

$$\frac{\delta E_0(\omega)}{E_0(\omega)} = \frac{\mathrm{Im}\varepsilon(\omega)}{2\pi\omega|\partial \mathrm{Re}\varepsilon(\omega)/\partial\omega|}. \tag{29}$$

The Equation (27) is applicable when $\delta E_0/E_0 \ll 1$. In consideration, which leads us to the (29) we used the Lorentz model. Now we use the data taken from Ref. [59] to verify the validity of the condition $\delta E_0/E_0 \ll 1$ for real gold and silver spheres. It is possible to replace Lorentz model Eq. (A7) by the real dielectric functions. The reason is as follows. First, if in (A7) we set the function to be constant then we obtain the Drude-Lorentz model with one resonance Eq. (14). Second, as is mentioned in Appendix A, if we consider the number of oscillators instead of one we obtain dielectric permittivity in the form of sum Eq. (14). Finally, in the visible range domain it is possible to fit actual permittivity of gold and silver with the sum of Drude-Lorentz oscillators [60,61]. Thus, the replacement of the model permittivity with the real permittivity of gold and silver is possible.

The correction (29) is shown in Fig. 3. It can be neglected in the range $\omega < 2.6$ eV and $\omega < 4$ eV for gold and silver, respectively. One can see from Fig. 3 that the correction to the electric near field per plasmon becomes sufficient when the interband transitions start to play an important role.

One can see from (10), (19), (20), and commutators (15) that the amplitudes of the polarization current $\hat{j}_{lm}(t) = \dot{\hat{P}}_{lm}(t)$ and amplitudes of the electric near field $\hat{E}_{lm}(t)$ obey the commutation relation

$$[\hat{E}_{lm}(t), \hat{j}_{l'm'}(t)] = \frac{i\hbar\delta_{ll'}\delta_{mm'}}{\varepsilon_0\kappa(2l+1)R^3}, \tag{30}$$

in other words the electric near field $\hat{E}_{lm}(t)$ and the polarization current $\hat{j}_{lm}(t)$ are canonical conjugate variables. Note that this commutation relation has been previously found in Ref. [44] without rigorous derivation.

As mentioned the pure plasmon part of the Hamiltonian (18) in the absence of the loss is the set of the harmonic oscillators (22). The operators in the interacting term [the third term in Eq. (18)], [see Eqs. (12) and (17)]

$$\hat{Q}_{\mathbf{P}lm}(t) = i\kappa \int_0^{+\infty} d\Omega \frac{2l+1}{l}\sqrt{\frac{\hbar R^3 l\varepsilon_0}{\pi}} \frac{\Omega\sqrt{\mathrm{Im}\varepsilon(\Omega)}}{\left|\varepsilon(\Omega)+\frac{l+1}{l}\right|} \times (\hat{d}^+_{lm}(\Omega,t) - \hat{d}_{lm}(\Omega,t)) \tag{31}$$

in low loss limit takes the form

$$\hat{Q}_{\mathbf{P}0lm}(t) = Q_{\mathbf{P}0lm}[\hat{d}^+_{0lm}(t) - \hat{d}_{0lm}(t)] \tag{32}$$

This transition can be done similarly to the Eqs. (23) and (24). Namely,

$$\left[\hat{Q}^{(-)}_{\mathbf{P}lm}(t), \hat{Q}^{(+)}_{\mathbf{P}lm}(t)\right]_{\frac{\mathrm{Im}\varepsilon(\omega)}{\omega\partial\mathrm{Re}\varepsilon(\omega)/\partial\omega}\ll 1} = \left[\hat{Q}^{(-)}_{\mathbf{P}0lm}(t), \hat{Q}^{(+)}_{\mathbf{P}0lm}(t)\right] \tag{33}$$

One can obtain from this

$$\hat{Q}_{\mathbf{P}0lm}(t) = i\kappa\frac{2l+1}{l}\omega_l\sqrt{\frac{\hbar R^3 l\varepsilon_0}{|\partial\mathrm{Re}\varepsilon(\Omega)/\partial\Omega|_{\Omega=\omega_l}}} \times (\hat{d}^+_{0lm}(t) - \hat{d}_{0lm}(t)) \tag{34}$$

The integral over Ω in the last term of Eq. (18) in the low loss limit can be transformed according to Eq. (25)

$$\frac{1}{\pi}\int_0^{+\infty} d\Omega \frac{\Omega\mathrm{Im}\varepsilon(\Omega)}{\left|\varepsilon(\Omega)+\frac{l+1}{l}\right|^2} \simeq \frac{\omega_l}{|\partial\mathrm{Re}\varepsilon(\Omega)/\partial\Omega|_{\Omega=\omega_l}}. \tag{35}$$

Substituting all these expressions into the Hamiltonian (18) we can obtain

$$\hat{H} = \sum_{l,m}\hbar\omega_l\hat{d}^+_{0lm}(t)\hat{d}_{0lm}(t) + \sum_{\lambda=1,2}\int d^3\mathbf{k}\hbar ck\hat{a}^+(\lambda,\mathbf{k},t)\hat{a}(\lambda,\mathbf{k},t) - i\sum_{\lambda=1,2}\sum_{l,m}\int d^3\mathbf{k}\left\{\frac{2l+1}{l\sqrt{l}}\frac{\hbar\omega_l R^{3/2}}{\sqrt{\left|\frac{\partial\mathrm{Re}\varepsilon(\omega_l)}{\partial\omega}\right|}}[\hat{d}^+_{0lm}(t) - \hat{d}_{0lm}(t)]\right.$$
$$\left.\times\left[\frac{\Lambda^*_{lm}(\lambda,-\mathbf{k})}{\sqrt{2ck}}\hat{a}^+(\lambda,-\mathbf{k},t) + \mathrm{H.c.}\right]\right\} + \sum_{l,m}\frac{(2l+1)^2}{l^3}\frac{\hbar\omega_l R^3}{\left|\frac{\partial\mathrm{Re}\varepsilon(\omega_l)}{\partial\omega}\right|}\left\{\sum_{\lambda=1,2}\int d^3\mathbf{k}\left[\frac{\Lambda^*_{lm}(\lambda,-\mathbf{k})}{\sqrt{2ck}}\hat{a}^+(\lambda,-\mathbf{k},t) + \mathrm{H.c.}\right]\right\}^2. \tag{36}$$

Thus, in the low loss limit, it is possible to introduce a discrete plasmon spectrum. One can see applying the Fermi golden rule that the Hamiltonian (36) provides the same plasmon radiative decay rate as calculated in Ref. [44]. The last term of the Hamiltonian (36) is rigorously derived first. The presence of the term in the Hamiltonian may lead to the decoupling effect, which has been recently studied in Refs. [62,63].

V. CONCLUSIONS

The problem of canonical quantization of plasmons on the subwavelength spherical nanoparticle has been investigated. By using the Lorentz microscopic model of medium and Fano diagonalization method, we transformed the plasmonic part of the Hamiltonian into a diagonal form, and quantized it in the standard way. The permittivity appears to be the only parameter that defines all characteristics of the system. The polarization and scalar potential are fully defined by the plasmonic structure. The quantization of plasmonic structure polarization causes the quantization of the scalar potential in the Coulomb gauge. The obtained Hamiltonian describes the plasmon oscillations and the plasmon and photon interaction. This describes the transition of the quanta of plasmons into those of the photons, and vice versa. It is shown that polarization current is a canonically conjugated variable to the electric near field.

It was shown that in the low loss limit, $\mathrm{Im}\varepsilon(\Omega) \ll \Omega(\partial \mathrm{Re}\varepsilon(\Omega)/\partial\Omega)$, the reservoir may be eliminated and Hamiltonian of the plasmons becomes equal to a set of independent harmonic oscillators. The obtained electric near field per plasmon in zero order in the imaginary part of permittivity coincides with the field obtained from phenomenological theory. The first order gives the correction, which cannot be obtained using phenomenological theory. The described method can be extended to subwavelength plasmonic nanoparticles of arbitrary shape.

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APPENDIX A: INTRODUCTION OF DIELECTRIC PERMITTIVITY

In this appendix, we consider the bulk medium, consisting of the Lorentz oscillators, as a classical system, and introduce the permittivity. The Lagrangian of the system is

$$\begin{aligned} L = &\int d^3\mathbf{r}\left\{\frac{\varepsilon_0[\dot{\mathbf{A}}(\mathbf{r},t)+\mathrm{grad}U(\mathbf{r},t)]^2}{2} - \frac{[\mathrm{rot}\mathbf{A}(\mathbf{r},t)]^2}{2\mu_0}\right\} + \int d^3\mathbf{r}\left\{\kappa\frac{\dot{\mathbf{P}}(\mathbf{r},t)^2}{2} - \kappa\omega_{\mathbf{P}0}^2\frac{\mathbf{P}(\mathbf{r},t)^2}{2}\right\} \\ &+ \int d^3\mathbf{r}\left\{\int_0^\infty d\Omega\left[\frac{\dot{\mathbf{Y}}_{\mathbf{P}}(\mathbf{r},\Omega,t)^2}{2} - \Omega^2\frac{\mathbf{Y}_{\mathbf{P}}(\mathbf{r},\Omega,t)^2}{2}\right]\right\} + \int d^3\mathbf{r}\{U(\mathbf{r},t)\mathrm{div}[\mathbf{P}(\mathbf{r},t)] + \dot{\mathbf{P}}(\mathbf{r},t)\mathbf{A}(\mathbf{r},t)\} \\ &- \int d^3\mathbf{r}\left\{\int_0^\infty d\Omega[V_{\mathbf{P}}(\Omega)\mathbf{P}(\mathbf{r},t)\dot{\mathbf{Y}}_{\mathbf{P}}(\mathbf{r},\Omega,t)]\right\}. \end{aligned} \tag{A1}$$

The physical meaning of each part of (A1) is given after expression (1). The equations of motion can be evaluated from (A1) by using the Euler-Lagrange equations [53]

$$\begin{aligned} \ddot{\mathbf{A}}(\mathbf{r},t) + c^2\mathrm{rotrot}\mathbf{A}(\mathbf{r},t) &= [\dot{\mathbf{P}}(\mathbf{r},t) - \varepsilon_0\mathrm{grad}\dot{U}(\mathbf{r},t)]/\varepsilon_0, \\ \Delta U(\mathbf{r},t) &= \mathrm{div}\mathbf{P}(\mathbf{r},t)/\varepsilon_0, \\ \kappa\ddot{\mathbf{P}}(\mathbf{r},t) + \kappa\omega_{\mathbf{P}0}^2\mathbf{P}(\mathbf{r},t) &= -\dot{\mathbf{A}}(\mathbf{r},t) - \mathrm{grad}U(\mathbf{r},t) - \int_0^{+\infty} d\omega[V_{\mathbf{P}}(\Omega)\dot{\mathbf{Y}}_{\mathbf{P}}(\mathbf{r},\Omega,t)], \\ \ddot{\mathbf{Y}}_{\mathbf{P}}(\mathbf{r},\Omega,t) + \Omega^2\mathbf{Y}_{\mathbf{P}}(\mathbf{r},\Omega,t) &= V_{\mathbf{P}}(\Omega)\dot{\mathbf{P}}(\mathbf{r},t), \end{aligned} \tag{A2}$$

where c is the speed of light in vacuum. In the frequency domain, Equations (A2) take the form

$$\begin{aligned} \omega^2\mathbf{A}(\mathbf{r},\omega) - c^2\mathrm{rotrot}\mathbf{A}(\mathbf{r},\omega) &= [i\omega\mathbf{P}(\mathbf{r},\omega) - i\omega\varepsilon_0\mathrm{grad}U(\mathbf{r},\omega)]/\varepsilon_0, \\ \Delta U(\mathbf{r},\omega) &= \mathrm{div}\mathbf{P}(\mathbf{r},\omega)/\varepsilon_0, \\ \kappa\left(\omega_{\mathbf{P}0}^2 - \omega^2\right)\mathbf{P}(\mathbf{r},\omega) &= i\omega\mathbf{A}(\mathbf{r},\omega) - \mathrm{grad}U(\mathbf{r},\omega) + \int_0^{+\infty} d\Omega[i\omega V_{\mathbf{P}}(\Omega)\mathbf{Y}_{\mathbf{P}}(\mathbf{r},\Omega,\omega)], \\ (\Omega^2-\omega^2)\mathbf{Y}_{\mathbf{P}}(\mathbf{r},\Omega,\omega) &= -i\omega V_{\mathbf{P}}(\Omega)\mathbf{P}(\mathbf{r},\omega). \end{aligned} \tag{A3}$$

The final equation leads to

$$\mathbf{Y}_{\mathbf{P}}(\mathbf{r},\Omega,\omega) = -\frac{i\omega}{\Omega^2-(\omega+i0\Omega)^2}V_{\mathbf{P}}(\Omega)\mathbf{P}(\mathbf{r},\omega). \tag{A4}$$

Substitution of (A4) into the third equation of (A3) leads to

$$\kappa\left(\omega_{\mathbf{P}0}^2 - \omega^2 - \omega^2\frac{1}{\kappa}\int_0^{+\infty} d\Omega\left[\frac{V_{\mathbf{P}}^2(\Omega)}{\Omega^2-(\omega+i0\Omega)^2}\right]\right)\mathbf{P}(\mathbf{r},\omega) = \mathbf{E}(\mathbf{r},\omega), \tag{A5}$$

where we define $\mathbf{E}(\mathbf{r},\omega) = i\omega\mathbf{A}(\mathbf{r},\omega) - \text{grad}U(\mathbf{r},\omega)$. Equation (A5) and the first equation of (A3) allow us to obtain to the Helmholtz equation for the electric field

$$\text{rotrot}\mathbf{E}(\mathbf{r},\omega) - \left(\frac{\omega}{c}\right)^2\left[1 + \frac{1}{\varepsilon_0\kappa}\frac{1}{\omega_{\mathbf{P}0}^2 - \omega^2 - \omega^2\frac{1}{\kappa}\int_0^{+\infty} d\Omega \frac{V_{\mathbf{P}}^2(\Omega)}{\Omega^2-(\omega+i0\Omega)^2}}\right]\mathbf{E}(\mathbf{r},\omega) = 0. \tag{A6}$$

The Helmholtz equation (A6) allows us to introduce the permittivity of the bulk medium

$$\varepsilon(\omega) = 1 + \frac{1}{\varepsilon_0\kappa}\frac{1}{\omega_{\mathbf{P}0}^2 - \omega^2 - \omega^2\frac{1}{\kappa}\int_0^{+\infty} d\Omega \frac{V_{\mathbf{P}}^2(\Omega)}{\Omega^2-(\omega+i0\Omega)^2}}. \tag{A7}$$

One can see from (A2) and (A3) and as mentioned in Ref. [32], if one considers the number of dipole harmonic oscillators interacting with reservoirs, then the dielectric permittivity is the sum of Eq. (A7). This fact plays an important role if one tries to fit the actual permittivity with the model one.

APPENDIX B: TRANSVERSE POLARIZATION CURRENT IN THE QUASISTATIC APPROXIMATION

In this appendix, we discuss the interaction between the transverse electromagnetic field and polarization current. The interaction between medium and transverse field is introduced through the term proportional $\mathbf{A}\dot{\mathbf{P}}$ in Lagrangian (1). Note that the medium polarization is a discontinued function due to finite volume of the metallic sphere. As a result, the transverse part of the current, $\mathbf{j}_t = \dot{\mathbf{P}} - \varepsilon_0\text{grad}\dot{U}$, is not zero because the terms $\dot{\mathbf{P}}$ and $\varepsilon_0\text{grad}\dot{U}$ do not cancel each other. Indeed, if we substitute Eqs. (3), (5), and (8) into $\mathbf{j}_t = \dot{\mathbf{P}} - \varepsilon_0\text{grad}\dot{U}$ we obtain

$$\mathbf{j}_t(\mathbf{r},t) = \sum_{l,m}\dot{P}_{lm}(t) \times \begin{cases} \frac{l+1}{2l+1}R\,\text{grad}\left[\left(\frac{r}{R}\right)^l\Psi_{lm}(\theta,\varphi)\right], & r < R \\ -\frac{l}{2l+1}R\,\text{grad}\left[\left(\frac{R}{r}\right)^{l+1}\Psi_{lm}(\theta,\varphi)\right], & r > R \end{cases}. \tag{B1}$$

One can see from the expression that this is indeed a transverse current $\text{div}\mathbf{j}_t(\mathbf{r},t) = 0$ and this current is not vanishing. No one summand is vanishing. That is the current, which induces the vector potential. Indeed, from Lagrangian (1) and the Euler-Lagrange equation we obtain in Coulomb gauge wave equation for the vector potential in the form (see also [52])

$$\Delta\mathbf{A}(\mathbf{r},t) - \ddot{\mathbf{A}}(\mathbf{r},t)/c^2 = -\mu_0\mathbf{j}_t(\mathbf{r},t), \tag{B2}$$

which demonstrates the coupling between transverse current and vector potential.

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