Quantum electrodynamics description of localized surface plasmons at a metal nanosphere

Kuniyuki Miwa and George C. Schatz

Department of Chemistry, Northwestern University, Evanston, Illinois 60208-3113
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A canonical quantization scheme for localized surface plasmons (LSPs) in a metal nanosphere is presented based on a microscopic model composed of electromagnetic fields, oscillators that describe plasmons, and a reservoir that describes excitations other than plasmons. The eigenmodes of this fully quantum electrodynamics theory show a spectrum that includes radiative depolarization and broadening, including redshifting from the quasi-static LSP modes, with increasing particle size. These spectral profiles correctly match those obtained with exact classical electrodynamics (Mie theory). The present scheme provides the electric fields per plasmon in both near- and far-field regions whereby its utility in the fields of quantum plasmonics and nano-optics is demonstrated.

Metal nanoparticles (MNPs) have been of great interest in nanotechnology owing to their unique properties originating from localized surface plasmon (LSP) resonances, the collective oscillations of conduction electrons in MNPs [1–4]. These resonances exhibit a tremendous potential for manipulating electromagnetic fields beyond the diffraction limit [5–8] and provide unique control of light [9, 10], energy [11–13], charge [14, 15], and heat [16, 17] at the nanoscale. A wide range of applications of nanoplasmonics has been reported including nanolasers [18–22], optical metamaterials [23], optical nonlinearities [24, 25], photoluminescence [26], photocatalysis [27], surface-tip-enhanced Raman spectroscopy [28–33], biosensing [34, 35], and photothermal therapy [36]. Parallel to this prominent progress, the quest for the quantum nature of plasmons and their interaction with matter has triggered a new branch of research named quantum plasmonics [37–41]. There have been widespread studies of quantum plasmonics [42–44] covering such quantum properties as strong coupling [45–48], entanglement [49, 50], squeezing [51], and Bose-Einstein condensation [52]. Quantum plasmonics drives progress in the field of integrated quantum photonics and nanoptics, providing a platform for many technological applications and devices operated at the quantum level, including single-photon sources [53], SPASER [54–56], transistors [57], ultra-compact circuits [58, 59], quantum information [60], and quantum computing devices [61].

The recent upsurge of interest in quantum plasmonics requires a quantum description of both electromagnetic fields and plasmons, which should be described in the natural context of quantum electrodynamics (QED) [62–66]. Quantization of electromagnetic fields has been developed since Dirac [62], however including plasmons with radiative damping and dissipation is a challenge. A canonical quantization procedure for electromagnetic fields in dispersive and dissipative homogeneous media was proposed by Huttner and Barnett [67, 68], which is based on prior work by Fano and Hopfield [69, 70]. This ‘microscopic’ approach has been extended to several inhomogeneous media subsequently [71, 72]. A different 'macroscopic’ approach was developed using the Green function formalism and the noise current method [73–76]. The quantum description of plasmons has been developed for bulk materials [77], metal surfaces [78], and MNPs [79, 80]. However, both the macroscopic Green function and microscopic Huttner-Barnett approaches have several drawbacks in the quantization process for LSPs in MNPs. Since the former offers a complicated procedure to calculate the electromagnetic fields, that are obtained indirectly from a phenomenologically introduced noise current operator, it is difficult to physically interpret each mode of the system [73–76]. Although the latter is the most prominent approach, the scheme becomes cumbersome to apply to an inhomogeneous medium. Indeed, the eigenmodes of the system have never been obtained even for a simple metal nanosphere. Quantized LSP modes were recently presented for a sphere by Shishkov et al. [81], but only within the quasi-static approximation. This approximation is valid only for a small particle (< 20 nm) as retardation effects become quite prominent otherwise [4, 82, 83].

A phenomenological approach to quantization of LSPs is widely used and much simpler [73, 83, 84, 85]. However, in this approach, no canonical formulation is obtained in dispersive and dissipative media [73]. Moreover, the effects of Joule losses cannot be described in a consistent way [81]. For example, eigenfrequencies of LSP resonances are calculated neglecting loss in the quantization procedure [37]. Also, the imaginary part of the permittivity does not affect the electric field generated by LSPs obtained by this approach [81]. In order to overcome these limitations, it is prerequisite to construct a rigorous approach to the quantization of LSPs which offers a canonical formulation in dispersive and dissipative inhomogeneous media.

In this Letter, we present a fully canonical quantization scheme for LSPs in a dispersive and dissipative metal nanosphere placed in vacuum. To quantize the electromagnetic fields and plasmons simultaneously, we utilize the Huttner-Barnett model and explore the eigenmodes of the system. Here, the plasmonic optical re-
Response of the metal is modeled with a set of harmonic oscillators that describe linear collective excitations of the electrons. In addition, we account for continuum reservoir degrees of freedom (electron-hole pair excitations and phonons) that are coupled to the plasmonic oscillator fields leading to damping. The reservoir is also responsible for the light absorption, such that diagonalization of the matter part of the Hamiltonian results in a set of dressed continuum fields that describe LSP modes in the quasi-static approximation. As a second step, the effects of radiation and retardation are investigated by exploring the eigenmodes of the total system composed of the vacuum electromagnetic field and the dressed oscillator field of the matter. Electric fields per plasmon are also calculated and correctly demonstrate both near- and far-field behavior.

Quantum theory can reproduce the exact classical theory from the classical electrodynamics, we find the developed theory provides a second quantized Hamiltonian for polarization density with mode \( l,m \) and frequency \( \omega \), \( \hat{b}_{l,m\omega}^\dagger \) is reservoir field annihilation (creation) operator for mode \( l,m \) and frequency \( \Omega \), and \( \hat{a}_{l,m\omega} \) is the annihilation (creation) operator of a transverse photon with wavenumber \( k_\parallel \) and polarization \( \lambda \). The vector potential is expanded onto a discrete basis of vector spherical harmonics and \( \nu = 1, \ldots, N \). \( \hat{P}_{lm} \) (\( \hat{\Pi}_{lm} \)) and \( \hat{Y}_{lm} \) (\( \hat{\Pi}_{lm} \)) are displacement (conjugate momentum) operators for the harmonic oscillator and reservoir fields, respectively. \( \hat{A}_{lm\lambda} \) is a vector potential operator for mode \( l,m \), wavenumber \( k_\parallel \), and polarization \( \lambda \). \( \Lambda_{\lambda}^{(\nu)} \) denotes the light-matter coupling strength. Detailed explanation is presented in Section III of SM.

The oscillator and reservoir parts of the Hamiltonian \( \hat{h}_{\text{mat}}^{(lm)} \) can be diagonalized by the Fano-type technique. Since a full frequency spectrum is investigated, we do not employ the rotating-wave approximation (RWA) to keep both co- and counter-rotating terms. The diagonalized expression is

\[
\begin{align*}
\hat{h}_{\text{mat}}^{(lm)} &= \int_0^\infty d\Omega \omega \hat{b}_{l,m\omega}^\dagger \hat{b}_{l,m\omega} + \int_0^{\infty} d\Omega \Omega \hat{b}_{l,m\omega}^\dagger \hat{b}_{l,m\omega} \\
&+ \int_0^{\infty} d\Omega \omega \hat{b}_{l,m\omega}^\dagger \hat{b}_{l,m\omega} \hat{b}_{l,m\omega} \hat{b}_{l,m\omega}^\dagger,
\end{align*}
\]

where \( \hat{b}_{l,m\omega} \) is the annihilation (creation) operator for polarization density with mode \( l,m \) and frequency \( \omega \), \( \hat{b}_{l,m\omega} \) is reservoir field annihilation (creation) operator for mode \( l,m \) and frequency \( \Omega \), and \( \hat{a}_{l,m\omega} \) is the annihilation (creation) operator of a transverse photon with wavenumber \( k_\parallel \) and polarization \( \lambda \). The vector potential is expanded onto a discrete basis of vector spherical harmonics and \( \nu = 1, \ldots, N \). \( \hat{P}_{lm} \) (\( \hat{\Pi}_{lm} \)) and \( \hat{Y}_{lm} \) (\( \hat{\Pi}_{lm} \)) are displacement (conjugate momentum) operators for the harmonic oscillator and reservoir fields, respectively. \( \hat{A}_{lm\lambda} \) is a vector potential operator for mode \( l,m \), wavenumber \( k_\parallel \), and polarization \( \lambda \). \( \Lambda_{\lambda}^{(\nu)} \) denotes the light-matter coupling strength. Detailed explanation is presented in Section III of SM.

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be independent of particle radius $R$ as expected for the quasistatic limit. The peak appears near 8.9 eV for Al, 3.5 eV for Ag, and 2.4 eV for Au, respectively. The energetic position of the peak in $\rho_{\text{gs}}(\omega)$ corresponds to the energy satisfying $\Re(\omega) = -2$, where $\epsilon(\omega)$ is the metal permittivity (Fig. S.1 in SM [88]). This relation denotes the so-called Fröhlich condition [1], which represents the condition for a resonance excitation of the dipolar LSPs for a small metal nanosphere and is valid within the quasi-static approximation. It is therefore concluded that the eigenmodes of the matter part of the system provide the LSP modes in the quasi-static approximation. Moreover, higher-order multipolar modes ($l > 1$) reproduce the corresponding LSP modes in the quasi-static approximation (Fig. S.2 in SM [88]).

We now consider the energy region where the imaginary part $\Im(\omega)$ of the permittivity is much smaller than $\omega|\partial \Re(\omega)/\partial \omega|$. In this region, the LSP modes can be separated from the reservoir and the eigenmodes of $\hat{h}_{\text{mat}}^{(lm)}$ offer a single discretized mode for each value of $l$ and $m$ (Section V in SM [88]). As a result, $\hat{h}_{\text{mat}}^{(lm)} \approx \hbar \omega_0 \hat{d}_{lm}^{(t)} \hat{d}_{lm}^{(r)}$, where the angular frequency $\omega_0$ of the eigenmode is given by $\Re(\omega_0) = -(l+1)/l$. It is shown that such an approximation, which we call the low-loss approximation, is valid in the range 3.0 eV $< \hbar \omega < 15.0$ eV for Al, $\hbar \omega < 4.0$ eV for Ag, and $\hbar \omega < 2.6$ eV for Au. It is noteworthy that the peak of the LSP modes appears in the energy range where the low-loss approximation is valid. Therefore, we employ this approximation hereafter.

To investigate the retardation and radiation effects, the total Hamiltonian $\hat{H} = \sum_{j,m} \hat{h}_{jm}$ with $\hat{h}_{jm} = \hat{h}_{\text{mat}}^{(lm)} + \hat{h}_{\text{em}}^{(lm)} + \hat{h}_{\text{int}}^{(lm)}$ is diagonalized by the Hopfield-Bogoliubov transformation [70]. By solving the problem numerically, a new set of eigenmodes is obtained, where the Hamiltonian $\hat{h}_{jm}$ is expressed in terms of the eigenoperator $\hat{X}_{\text{jm}}$ by $\hat{h}_{jm} = \sum_{j=1}^{N} \hbar \Omega_{jm} \hat{X}_{jm} \hat{X}_{jm}^\dagger$. The spectral function $\rho_{\text{full}}(\omega)$ of the LSP modes for the total system is obtained by $\rho_{\text{full}}(\omega) = -\Im G_{\text{full}}(\omega)/\pi$, where $G_{\text{full}}(\omega)$ is Fourier transform of the Green function $G_{\text{full}}(t,t') = (1/\hbar) \theta(t-t') \langle [\hat{d}_{lm}(t),\hat{d}_{lm}^\dagger(t')] \rangle$ and $\langle \ldots \rangle$ indicates the statistical average in the representation defined by the system evolution for the total Hamiltonian $\hat{H}$. The electric field $\mathbf{E}_{\text{lmj}}(r)$ per plasmon is calculated using $\mathbf{E}_{\text{lmj}}(r) = [\hat{X}_{\text{lmj}}(t), \mathbf{E}(r,t)]$ where the electric field operator $\mathbf{E}(r,t)$ is given by $\mathbf{E}(r,t) = -\nabla \phi(r,t) - \partial \mathbf{A}(r,t)/\partial t$ with $\phi(r,t)$ and $\mathbf{A}(r,t)$ the scalar and vector potential operators, respectively. Details of the calculation are provided in Section VI of SM [88].

FIG. 1. Spectral function $\rho_{\text{gs}}(\omega)$ for (a) Al, (b) Ag, and (c) Au nanosphere. Only the dipolar mode ($l = 1$) is considered. The permittivity $\epsilon(\omega)$ of Al is taken from the experimental data reported in Ref. 91 and Ag and Au in Ref. 92.

FIG. 2. Comparison of the spectral function $\rho_{\text{full}}(\omega)$ of the total system, the spectral function $\rho_{\text{gs}}(\omega)$ in the quasi-static approximation, and the extinction spectra $\sigma_{\text{ext}}$ obtained with Mie theory for aluminum nanoparticles with radius $R$ of (a)(d) 30 nm, (b)(e) 60 nm, and (c)(f) 90 nm, respectively. Red solid and green dotted lines indicate $\rho_{\text{gs}}(\omega)$ and $\rho_{\text{full}}(\omega)$. The blue dashed line represents $\sigma_{\text{ext}}$ calculated using Mie theory [89]. The dipolar (a-c) and quadrupolar modes (d-f) are displayed. The permittivity $\epsilon(\omega)$ of Al is taken from the experimental data [91].

Figure 2(a-c) shows the calculated results for $\rho_{\text{full}}(\omega)$ and $\rho_{\text{gs}}(\omega)$ for Al nanospheres with different radii $R$ in the case of a dipolar mode ($l = 1$). To confirm the validity of the results, we compare with the extinction spectra $\sigma_{\text{ext}}(\omega)$ calculated using Mie theory [89]. The permittivity $\epsilon(\omega)$ of Al is taken from experimental data [91]. The results $\rho_{\text{gs}}(\omega)$ in the quasi-static approximation exhibit a peak near 8.9 eV, where the peak position is independent of $R$. In $\rho_{\text{full}}(\omega)$, owing to the light-matter coupling, the peak reflects radiative depolarization and broadening, leading to a peak that is red shifted. As the radius $R$ increases, the linewidth and the amount of the red shift both increase. The results are in good agreement with the Mie theory result for $\sigma_{\text{ext}}(\omega)$. It is noteworthy that not only the dipolar ($l = 1$) mode but also the
The resonance energy extracted from the peak position in the Mie expression for $\sigma_{\text{ext}}(\omega)$ matches well with the energetic position of the peak in $\rho_{\text{full}}(\omega)$. In SM 88, the calculated results for Ag and Au nanospheres are obtained using the permittivity $\varepsilon(\omega)$ reported in Ref. 92. Radiative broadening and peak red shift due to the light-matter coupling can be observed. Owing to the large amount of the peak shift in $\rho_{\text{full}}(\omega)$, the linewidth and the amount of the peak shift in $\rho_{\text{full}}(\omega)$ are narrower and smaller than those in the Mie solution for $\sigma_{\text{ext}}(\omega)$. These problems can be solved by introducing the permittivity modeled with the well-known Drude model 93.

**FIG. 3.** Comparison of the spectral function $\rho_{\text{full}}(\omega)$ of the total system, the spectral function $\rho_{qs}(\omega)$ in the quasi-static approximation, and the extinction spectra $\sigma_{\text{ext}}$ obtained with Mie theory for Ag nanoparticles with radius $R$ of (a)(d) 30 nm, (b)(e) 60 nm, and (c)(f) 90 nm, respectively. Red solid and green dotted lines indicate $\rho_{\text{full}}(\omega)$ and $\rho_{qs}(\omega)$. The blue dashed line represents $\sigma_{\text{ext}}$ calculated using Mie theory 88. The dipolar (a-c) and quadrupolar modes (d-f) are displayed. The permittivity $\varepsilon(\omega)$ is parametrized utilizing the Drude model with the plasmon frequency $\omega_p$ and the damping term $\gamma$. The parameters ($\hbar\omega_p$, $\hbar\gamma$) are determined as (9.04 eV, 21.25 meV) that are used to model the permittivity of silver 93. The energetic position of the peak in $\rho_{\text{full}}(\Omega)$ is in good agreement with the resonance energy extracted from the peak position in the Mie solution for $\sigma_{\text{ext}}(\omega)$.

Figure 4 presents the calculated results for the electric field $E_{lmj}(r)$ per plasmon for Al nanospheres with various values of the radius $R$. $E_{lmj}(r)$ shows both near- and far-field behavior that is familiar for dipolar plasmon excitation. In the smaller nanosphere, the resonance energy appears at the higher energy and the spatial distribution of $|E_{lmj}(r)|$ oscillates with a high frequency in the far-field region. It is noteworthy that $E_{lmj}(r)$ is crucial for evaluating the interaction between the field and quantum emitters, playing an important role in determining Purcell effects associated with the emitter, governing the threshold pumping rate for the lasing oscillations, and many other properties 18, 22, 54, 56. This indicates that our theory provides a useful quantum electrodynamic platform for studying quantum plasmonics and nano-optics.

In conclusion, based on a microscopic model for the medium, we developed a fully canonical quantization scheme for the localized surface plasmons (LSPs) associated with a dispersive and absorptive metal nanosphere interacting with the vacuum electromagnetic field. The matter part of the Hamiltonian is first diagonalized with the Fano technique to determine eigenmodes representing the LSP modes in the quasi-static approximation. In the energy region where the imaginary part $\Im(\omega)$ of the permittivity is much smaller than $\omega|\partial\Re(\omega)/\partial\omega|$ of the quasi-static LSP modes are shown to be isolated from the reservoir. Moreover, using the Hopfield-Bogoliubov transformation, eigenmodes of the total system are obtained, wherein retardation and radiation effects are incorporated into the LSP modes. The obtained eigenmodes exhibit spectra in which radiative broadening and depolarization lead to significant redshifting relative to that of LSPs in the quasi-static approximation. The energetic position of the peak in the calculated spectra coincides with that obtained from the extinction spectra using Mie theory, which means that the theory matches Maxwell’s equations, including all multipoles, for a nanosphere. The calculated electric fields per plasmon demonstrate realistic behavior in both near- and far-fields, whereby the utility of the developed theory in quantum plasmonics and nano-optics is demonstrated.

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FIG. 4. Spatial distribution of electric field $E_{lmj}(r)$ per plasmon for Al nanosphere with the radius $R$ for (a,d) 30 nm, (b,e) 60 nm, and (c,f) 90 nm. Absolute values $|E_{lmj}(r)|$ of the electric fields are plotted. The angular frequency $\omega$ is set at the resonance excitation energy of (a,d) 6.06 eV, (b,e) 3.60 eV, and (c,f) 2.53 eV. The permittivity of Al is taken from Ref. 91.