# Surface plasmon–polaritons on ultrathin metal films*

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We discuss the surface plasmon–polaritons used for ultrathin metal films with the aid of linear response theory and make comparisons with the known result given by Economou E N. In this paper we consider transverse electromagnetic fields and assume that the electromagnetic field in the linear response formula is the induced field due to the current of the electrons. It satisfies the Maxwell equation and thus we replace the current (charge) term in the Maxwell equation with the linear response expectation value. Finally, taking the external field to be zero, we obtain the dispersion relation of the surface plasmons from the eigenvalue equation. In addition, the charge-density and current-density in the $z$ direction on the surface of ultrathin metal films are also calculated. The results may be helpful to the fundamental understanding of the complex phenomenon of surface plasmon-polaritons.

**Keywords:** surface plasmon-polariton, linear response theory, dispersion relation, ultrathin metal film

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## 1. Introduction

There has been a great deal of recent interest in surface plasmons both theoretically[1−3] and experimentally.[4−6] Experimental studies of surface plasmons have focused on their excitation energies and dispersions, which are accessible by a number of spectroscopic techniques such as electron energy loss spectroscopy, optical absorption and transmission, and surface enhanced Raman spectroscopy. Theoretical understanding of surface plasmons has been based on either classical models[1,7−10] or quantum-mechanical calculations.[11−14] Linear response theory combined with time-dependent local density approximation[15,16] provides one of the most accurate approaches to surface plasmons. It has been successfully applied to describe the plasmon energies and dispersions of metal surfaces[12,13] and thin films.[17,18] For bulk metals and their surfaces, this approach has been very successful in the description of collective electronic excitations, which usually dominate the dynamic response of a many-electron system.

In the present work we study the linear response of ultrathin metal films to the applied field of a plane electromagnetic wave. We consider thin metal films that represent so-called two-dimensional electron systems. In such systems the motion of electrons is quantized in one direction and free in two others. Thus, we can theoretically obtain the dispersion relation of surface plasmons for an ultrathin film metal by calculating the charge–, current–current response function within the linear response theory. In doing this, we must solve the Maxwell equation in the surface. In most of the literature,[19−21] the theories for surface plasmon of metals are based on the Maxwell equations for the field with time dependence $\mathrm{e}^{-\mathrm{i}\omega t}$ in the metal, that is to say $\boldsymbol{\nabla}\times\boldsymbol{E}=\mathrm{i}\omega\mu_0\boldsymbol{H}, \boldsymbol{\nabla}\times\boldsymbol{H}=-\mathrm{i}\omega\mu_0\boldsymbol{E}+\boldsymbol{J}, \boldsymbol{\nabla}\cdot\boldsymbol{E}=\rho/\varepsilon_0$ and $\boldsymbol{\nabla}\cdot\boldsymbol{H}=0$. Substituting the charge conservation and the property of conductors (Ohm law), that is, $\partial\rho/\partial t+\boldsymbol{\nabla}\cdot\boldsymbol{J}=0$ and $\boldsymbol{J}=\sigma\boldsymbol{E}$, into the Maxwell equations, one can obtain the field equation in the metal: $\boldsymbol{\nabla}^2\boldsymbol{E}+(\varepsilon_{\mathrm{M}}\omega^2/c^2)\boldsymbol{E}=0$ with $\varepsilon_{\mathrm{M}}=1+\mathrm{i}\sigma/\omega\varepsilon_0$, which was used widely in the literature. However, it is unreasonable that this field equation is applied in the ultrathin metal near the surface. In this case the Ohm law in such a form $\boldsymbol{J}=\sigma\boldsymbol{E}$ is not valid; it is only applicable to bulk and long-wave case. Since there is no symmetry of translation, the Ohm law should be written in the following form $\boldsymbol{J}(\boldsymbol{r},\omega)=\int\overleftrightarrow{\sigma}(\boldsymbol{r},\boldsymbol{r}',\omega)\boldsymbol{E}(r',\omega)\mathrm{d}\boldsymbol{r}'$, where $\overleftrightarrow{\sigma}(\boldsymbol{r},\boldsymbol{r}',\omega)$

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is the non-local conductivity tensor. So, our idea is to use the linear response theory to obtain the non-local conductivity tensor and construct the field equations in the metals. Then we solve the equation accompanying the boundary condition between the metal and dielectrics. In addition, we assume that the electromagnetic field $\boldsymbol{A}$ in the linear response formula is the induced field due to the current of the electrons and satisfies the Maxwell equation. Theoretically this idea can be realized by replacing the current term in the Maxwell equation by the linear response expectation value. Finally, taking the external field to be zero, we will obtain the dispersion relation from the eigenvalue equation.

## 2. Formula and derivation

As shown in Fig. 1, we consider an ultrathin metal film with thickness $L$ extending in the $x$–$y$ plane and assume $\boldsymbol{r}$ denotes two-dimensional vector parallel to $x$–$y$ plane. For this system in the presence of external fields, the Hamiltonian of the system is in general given by

$$H = H_0 + H'(t). \tag{1}$$

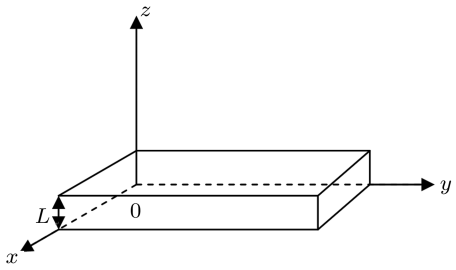



**Fig. 1.** Schematic view of an ultrathin metal film with a thickness $L$ used in the calculation.

The second term $H'(t)$ in Eq. (1) represents the interaction with the external fields and accounts for the electron–electron interaction. The first term describes the unperturbed Hamiltonian in the absence of external fields and refers to the kinetic energy of electrons with momentum $\boldsymbol{p}$ at the location $(\boldsymbol{r}, z)$

$$H_0 = \frac{\boldsymbol{p}^2}{2m} + U(\boldsymbol{r}, z). \tag{2}$$

In the following, we assume that $H_0$ has a complete and orthogonal set of eigenstates, $\{\psi^0_{\boldsymbol{k}_t,n}(\boldsymbol{r}, z)\}$, because the motion of an electron along the surface is completely free, we have

$$\Psi^0_{n,\boldsymbol{k}_t}(\boldsymbol{r}, z) = \frac{1}{\sqrt{S}} \mathrm{e}^{\mathrm{i}\boldsymbol{k}_t\cdot\boldsymbol{r}} \Phi_n(z),$$

$$E_{n,\boldsymbol{k}_t} = \varepsilon_n + \frac{\hbar^2 k_t^2}{2m^*}, \quad n = 1, 2, \ldots, \tag{3}$$

where $m^*$ is the effect mass of electron, $n = 1, 2, \ldots$ denotes the discrete quantum number describing the different quantum channels (or transverse modes) at fixed energy; $\boldsymbol{k}_t$ is the wave vector parallel to the surface, and $k_t = |\boldsymbol{k}_t|$. $S$ is the surface area of the system. The wave functions $\Phi_n(z)$ and energy eigenvalues $\varepsilon_n$ are the solutions of one-dimensional Kohn–Sham equation

$$-\frac{\mathrm{d}^2}{\mathrm{d}z^2}\Phi_n(z) + V_{\mathrm{eff}}(z)\Phi_n(z) = \varepsilon_n \Phi_n(z). \tag{4}$$

Here, we note that $\{\Phi_n(z)\}$ is real. In Eq. (4), $V_{\mathrm{eff}}(z)$ is the effective one-electron potential and consists of the usual electrostatic potential plus an exchange–correlation potential. In the existent documents, the electrostatic potential can be obtained by solving the Poisson equation and the exchange–correlation potential is described by the local Slater approximation and local Wigner interpolation.[22] In the present work we adopted the infinite square well model and assume that the quantum well has been grown along the $z$ direction and is translational invariant in the $x$–$y$ plane, namely,

$$V_{\mathrm{eff}}(z) = \begin{cases} 0, & 0 < z < L, \\ \infty, & \text{otherwise.} \end{cases} \tag{5}$$

According to this hypothesis, $\Phi_n(z)$ and $\varepsilon_n$ from Eq. (4) then yield

$$\Phi_n(z) = \sqrt{\frac{2}{L}} \sin\left(\frac{n\pi z}{L}\right), \quad \varepsilon_n = \frac{n^2\pi^2\hbar^2}{2m^* L^2}. \tag{6}$$

In terms of the set of eigenstates (see Eq. (3)), we write field operator $\Psi(\boldsymbol{r}, z)$ and its conjugation in the so-called second quantized form as follows:

$$\hat{\Psi}(\boldsymbol{r}, z) = \sum_{\boldsymbol{k}_t, n} \hat{a}_{\boldsymbol{k}_t,n} \Psi^0_{n,\boldsymbol{k}_t}(\boldsymbol{r}, z),$$

$$\hat{\Psi}^\dagger(\boldsymbol{r}, z) = \sum_{\boldsymbol{k}_t, n} \hat{a}^\dagger_{\boldsymbol{k}_t,n} \Psi^{*0}_{n,\boldsymbol{k}_t}(\boldsymbol{r}, z). \tag{7}$$

Here, $\hat{a}^\dagger_{\boldsymbol{k}_t,n}$ and $\hat{a}_{\boldsymbol{k}_t,n}$ are creation and annihilation operators, respectively.

Next, we derive an expression for the induced current in terms of the current–current correlation function. The operator of the electrical current density is defined as

$$\hat{\boldsymbol{J}}(\boldsymbol{r}, z) = \hat{\boldsymbol{J}}_{\mathrm{d}}(\boldsymbol{r}, z) + \hat{\boldsymbol{J}}_{\mathrm{p}}(\boldsymbol{r}, z), \tag{8}$$

$$\hat{\boldsymbol{J}}_{\mathrm{p}}(\boldsymbol{r}, z) = \frac{e\hbar}{2\mathrm{i}m^*}[\hat{\Psi}^\dagger(\boldsymbol{r}, z)\nabla\hat{\Psi}(\boldsymbol{r}, z)$$

$$- \left(\nabla\hat{\Psi}^{\dagger}(\boldsymbol{r},z)\right)\hat{\Psi}(\boldsymbol{r},z)], \tag{9}$$

$$\hat{\boldsymbol{J}}_{\rm d}(\boldsymbol{r},z) = -\frac{e^2}{m^*c}\hat{\Psi}^{\dagger}(\boldsymbol{r},z)\boldsymbol{A}\hat{\Psi}(\boldsymbol{r},z), \tag{10}$$

where $\hat{\boldsymbol{J}}_{\rm d}(\boldsymbol{r},z)$ denotes the diamagnetic current, and $\hat{\boldsymbol{J}}_{\rm p}(\boldsymbol{r},z)$ is called the paramagnetic current and given by

$$\hat{\boldsymbol{J}}_{\rm p}(\boldsymbol{r},z) = \frac{1}{S}\sum_{\boldsymbol{q_t}}(\boldsymbol{\rho}^{J}_{\boldsymbol{q}_t} + \boldsymbol{\rho}^{J_z}_{\boldsymbol{q}_t}){\rm e}^{{\rm i}\boldsymbol{q}_t\cdot\boldsymbol{r}}, \tag{11}$$

$$\boldsymbol{\rho}^{J}_{\boldsymbol{q}_t} = \frac{e\hbar}{2m^*}\sum_{\boldsymbol{k}_t,n,m}(2\boldsymbol{k}_t+\boldsymbol{q}_t)\Phi_{1nm}(z) \times \hat{a}^{\dagger}_{\boldsymbol{k}_t,n}\hat{a}_{\boldsymbol{k}'_{\rm t}+\boldsymbol{q}_{\rm t},m}, \tag{12}$$

$$\boldsymbol{\rho}^{J_z}_{\boldsymbol{q}_t} = \frac{e\hbar}{2{\rm i}m^*S}\sum_{\boldsymbol{k}_t,n,m}\Phi_{2nm}(z) \times \hat{a}^{\dagger}_{\boldsymbol{k}_t,n}\hat{a}_{\boldsymbol{k}'_{\rm t}+\boldsymbol{q}_{\rm t},m}\boldsymbol{e}_z, \tag{13}$$

where

$$\Phi_{1nm}(z) = \Phi^*_n(z)\Phi_m(z), \tag{14}$$

$$\Phi_{2nm}(z) = \Phi^*_n(z)\frac{{\rm d}}{{\rm d}z}\Phi_m(z) - \Phi_m(z)\frac{{\rm d}}{{\rm d}z}\Phi^*_n(z),$$
$$\boldsymbol{q}_t = \boldsymbol{k}_t - \boldsymbol{k}'_t, \quad q_t = |\boldsymbol{q}_t|. \tag{15}$$

The first sum in Eqs. (12) and (13) runs over the occupied energy levels only, and the second sum runs over both occupied and unoccupied levels.

Using the definition in Eq. (8) of the current density, the interaction term $H'(t)$ for transverse fields (note that $\varphi = 0$) can be expressed as

$$H'(t) = -\int \hat{\boldsymbol{J}}(\boldsymbol{r},z)\cdot\boldsymbol{A}(\boldsymbol{r},z,t)\,{\rm d}\boldsymbol{r}\,{\rm d}z. \tag{16}$$

The diamagnetic current term $\hat{\boldsymbol{J}}_{\rm d}(\boldsymbol{r},z)$ leads to a term in the interaction Hamiltonian which is of the second order in $\boldsymbol{A}(\boldsymbol{r},z,t)$; thus it is not included if we restrict ourselves to the interactions proportional to $\boldsymbol{A}(\boldsymbol{r},z,t)$. In a weak magnetic field, we can neglect the effect of the diamagnetic current term. In addition, in the frequency domain we have $\boldsymbol{A}(\boldsymbol{r},z,t) = \boldsymbol{E}(\boldsymbol{r},z,t)/{\rm i}\omega$ and therefore the external perturbation in Eq. (16) becomes

$$H'(t) = -\frac{1}{{\rm i}\omega}\int \hat{\boldsymbol{J}}(\boldsymbol{r},z)\cdot\boldsymbol{E}(\boldsymbol{r},z,t)\,{\rm d}\boldsymbol{r}\,{\rm d}z. \tag{17}$$

For the response of the total current density to the electromagnetic fields, it reads

$$\begin{aligned}&\delta\boldsymbol{J}(\boldsymbol{r},z,t)\\ &= -\frac{e^2\langle n_0(\boldsymbol{r},z)\rangle}{{\rm i}\omega m^*}\boldsymbol{E}(\boldsymbol{r},z,t)\\ &\quad + \frac{1}{\hbar\omega}\int_{-\infty}^{t}{\rm d}t'\left\langle\left[\hat{\boldsymbol{J}}^{\rm I}(\boldsymbol{r},z,t), \int {\rm d}\boldsymbol{r}'\,{\rm d}z'\hat{\boldsymbol{J}}^{\rm I}_{\rm p}(\boldsymbol{r}',z',t')\right.\right.\\ &\quad \left.\left.\cdot\,\boldsymbol{E}(\boldsymbol{r}',z',t')\right]\right\rangle_0,\end{aligned} \tag{18}$$

where $\boldsymbol{J}^{\rm I}_{\rm p}$ is of the form in interaction representation. $\langle[\ ]\rangle$ denotes the thermodynamic average for the systems described by $H_0$. The term $n_0(\boldsymbol{r},z)$ is the particle density in the ultrathin metal films, which vanishes at the boundaries: $z = 0, L$, and is given by

$$\begin{aligned}n_0(\boldsymbol{r},z) &= 2\sum_{E_{n,\boldsymbol{k}_t}<E_f}|\Psi_{n,\boldsymbol{k}_t}(\boldsymbol{r},z)|^2\\ &= \frac{1}{\pi}\sum_n|\Phi_n(z)|^2(E_{f_n}-\varepsilon_n).\end{aligned} \tag{19}$$

Factor 2 in Eq. (19) accounts for spin degeneracy. In general, the Fermi energy of ultrathin metal films differs from that of the semi-infinite surface, especially when the thickness $L$ is small. In our calculation, $E_{f_n}$is determined by the charge neutrality condition

$$\int_0^L n_0(\boldsymbol{r},z)\,{\rm d}\boldsymbol{r}\,{\rm d}z = \bar{n}_0 L, \tag{20}$$

where $\bar{n}_0$ denotes the average electron density. Collecting things now we have

$$\delta J^z(q_t,\omega,z) = -\frac{e^2\bar{n}_0}{{\rm i}\omega m^*}E_z(q_t,\omega,z) + \int {\rm d}z'\,[\Pi_{jy}(q_t,\omega,z,z')E_y(z') + \Pi_{jz}(q_t,\omega,z,z')E_z(z')] \tag{21}$$

with

$$\Pi_{jy}(q_t,\omega,z,z') = \frac{e^2\hbar^2}{4\omega S m*^2}\sum_{\boldsymbol{k}_t,n,m}\frac{f(E_{\boldsymbol{k}_t,n}) - f(E_{\boldsymbol{k}_t+\boldsymbol{q}_t,m})}{\hbar\omega - \hbar\omega_{m,n,\boldsymbol{k}_t,\boldsymbol{q}_t} + {\rm i}\eta}(q_t+2k_t)\Phi_{1nm}(z)\Phi_{1nm}(z'), \tag{22}$$

$$\Pi_{jz}(q_t,\omega,z,z') = \frac{{\rm i}e^2\hbar^2}{4\omega S m*^2}\sum_{\boldsymbol{k}_t,n,m}\frac{f(E_{\boldsymbol{k}_t,n}) - f(E_{\boldsymbol{k}_t+\boldsymbol{q}_t,m})}{\hbar\omega - \hbar\omega_{m,n,\boldsymbol{k}_t,\boldsymbol{q}_t} + {\rm i}\eta}\Phi_{2nm}(z)\Phi_{2nm}(z'), \tag{23}$$

where $\hbar\omega_{m,n,\boldsymbol{k}_t,\boldsymbol{q}_t} = E_{\boldsymbol{k}_t+\boldsymbol{q}_t,m} - E_{\boldsymbol{k}_t,n}$.

The operator of the electrical charge density $\hat{\rho}(\boldsymbol{r}, z)$ is defined as

$$\hat{\rho}(\boldsymbol{r}, z) = -e\Psi^{\dagger}(\boldsymbol{r}, z)\hat{\Psi}(\boldsymbol{r}, z). \tag{24}$$

In terms of $\hat{a}^{\dagger}_{\boldsymbol{k}_t,n}$ and $\hat{a}_{\boldsymbol{k}_t,n}$, $\hat{\rho}(\boldsymbol{r}, z)$ becomes

$$\hat{\rho}(\boldsymbol{r}, z) = -\frac{e}{S}\sum_{\boldsymbol{k}_t,\boldsymbol{k}'_t,n,m} \Phi^{*}_{n}(z)\Phi_{m}(z)\,\mathrm{e}^{\,\mathrm{i}\boldsymbol{q}_t\cdot\boldsymbol{r}}\hat{a}^{\dagger}_{\boldsymbol{k}_t,n}\hat{a}_{\boldsymbol{k}'_t+\boldsymbol{q}_t,m}. \tag{25}$$

Similarly, we have

$$\delta\rho(\boldsymbol{r}, z, t) = -\frac{1}{\hbar\omega}\int_{-\infty}^{t}\mathrm{d}t'\left\langle\left[\hat{\rho}^{\mathrm{I}}(\boldsymbol{r}, z, t), \int \mathrm{d}\boldsymbol{r}'\,\mathrm{d}z'\hat{\boldsymbol{J}}^{\mathrm{I}}_{\mathrm{p}}(\boldsymbol{r}', z', t')\cdot\boldsymbol{E}(\boldsymbol{r}', z', t')\right]\right\rangle_0. \tag{26}$$

Hence, the induced charge density becomes, in the Fourier domain,

$$\delta\rho(q_t, \omega, z) = \int \mathrm{d}z'\left[\Pi_{\rho y}(q_t, \omega, z, z')E_y(z') + \Pi_{\rho z}(q_t, \omega, z, z')E_z(z')\right] \tag{27}$$

with

$$\Pi_{\rho y}(q_t, \omega, z, z') = \frac{\mathrm{i}e^2\hbar}{2\omega S m^*}\sum_{\boldsymbol{k}_t,n,m}\frac{f(E_{\boldsymbol{k}_t,n}) - f(E_{\boldsymbol{k}_t+\boldsymbol{q}_t,m})}{\hbar\omega - \hbar\omega_{m,n,\boldsymbol{k}_t,\boldsymbol{q}_t} + \mathrm{i}\eta}(q_t + 2k_t)\Phi_{1nm}(z)\Phi_{1nm}(z'), \tag{28}$$

$$\Pi_{\rho z}(q_t, \omega, z, z') = -\frac{e^2\hbar}{2\omega S m^*}\sum_{\boldsymbol{k}_t,n,m}\frac{f(E_{\boldsymbol{k}_t,n}) - f(E_{\boldsymbol{k}_t+\boldsymbol{q}_t,m})}{\hbar\omega - \hbar\omega_{m,n,\boldsymbol{k}_t,\boldsymbol{q}_t} + \mathrm{i}\eta}\Phi_{1nm}(z)\Phi_{2nm}(z'). \tag{29}$$

We have completed the derivation of the equations for the charge- and current-responses. Below we discuss the transverse plasmon mode dispersion. To determine the dispersion relation, we must specify the boundary condition on the field vectors at $z = 0$ and $L$. The boundary conditions imply that the tangent components of the electric fields ($E_y$) and magnetic ($H_x$) fields should be continuous, yielding

$$E_y(z)|_{z=0^+,L^-} = E_y(z)|_{z=0^-,L^+}, \tag{30}$$

$$H_x(z)|_{z=0^+,L^-} = H_x(z)|_{z=0^-,L^+}. \tag{31}$$

Using the identity $\boldsymbol{\nabla}\cdot\boldsymbol{E} = \delta\rho/\varepsilon_0$, we obtain

$$\mathrm{i}q_tE_y + \frac{\mathrm{d}E_z}{\mathrm{d}z} = \begin{cases}\dfrac{\delta\rho}{\varepsilon_0}, & \text{in ultrathin metal film,}\\ 0, & \text{otherwise.}\end{cases} \tag{32}$$

We consider the Maxwell wave equation and replace the current density term and charge density term by $\delta\boldsymbol{J}$ and $\delta\rho$, thus we obtain the Maxwell wave equation in the following form

$$\frac{\mathrm{d}^2}{\mathrm{d}z^2}E_z(q_t, \omega, z) - \left(q_t^2 - \frac{\omega^2}{c^2}\varepsilon(z)\right)E_z(q_t, \omega, z) = \int \mathrm{d}z'\left[\Pi_{zy}(q_t, \omega, z, z')E_y(z') + \Pi_{zz}(q_t, \omega, z, z')E_z(z')\right] \tag{33}$$

with

$$\Pi_{zy}(q_t, \omega, z, z') = \frac{1}{\varepsilon_0}\frac{\mathrm{d}\Pi_{\rho y}(q_t, \omega, z, z')}{\mathrm{d}z} - \mathrm{i}\mu_0\omega\Pi_{jy}(q_t, \omega, z, z'), \tag{34}$$

$$\Pi_{zz}(q_t, \omega, z, z') = \frac{1}{\varepsilon_0}\frac{\mathrm{d}\Pi_{\rho z}(q_t, \omega, z, z')}{\mathrm{d}z} - \mathrm{i}\mu_0\omega\Pi_{jz}(q_t, \omega, z, z'), \tag{35}$$

where a simple dielectric function of the form $\varepsilon(z) = 1 - \omega_{\mathrm{p}}^2/\omega^2$ is used, and $\omega_{\mathrm{p}} = e^2n_0(z)/m^*\varepsilon_0$ is the eigenfrequency of plasmon in bulk material.

Thus equation (32) can be casted into

$$\frac{\mathrm{d}E_z}{\mathrm{d}z} + \mathrm{i}q_tE_y = \frac{1}{\varepsilon_0}\int \mathrm{d}z'\left[\Pi_{\rho y}(q_t, \omega, z')E_y(z') + \Pi_{\rho z}(q_t, \omega, z, z')E_z(z')\right]. \tag{36}$$

Outside the ultrathin metal film, field amplitudes decay exponentially away from the metal interface with field maxima occurring at the surface, so the electric fields components are characterized by the following equation

$$E_z = \begin{cases} E_{z1}\,\mathrm{e}^{-k_{z1}z+\mathrm{i}q_t y}, & z \geq L, \\ E_{z2}\,\mathrm{e}^{-k_{z2}z+\mathrm{i}q_t y}, & z \leq 0, \end{cases} \quad k_{zi} = \left(q_t^2 - \frac{\omega^2}{c^2}\varepsilon_i\right)^{1/2}, \quad i = 1, 2, \tag{37}$$

where $\varepsilon_i$ denotes dielectric constant of different dielectrics. Hence the field amplitude ($E_y$) outside the ultrathin metal films can be obtained via Eq. (32)

$$E_y = \begin{cases} \dfrac{k_{z1}}{\mathrm{i}q_t}E_{z1}\,\mathrm{e}^{-k_{z1}z+\mathrm{i}q_t y}, & z \geq L; \\ -\dfrac{k_{z2}}{\mathrm{i}q_t}E_{z2}\,\mathrm{e}^{-k_{z1}z+\mathrm{i}q_t y}, & z \leq 0. \end{cases} \tag{38}$$

Thus, $H_x$ can be calculated via $\boldsymbol{\nabla} \times \boldsymbol{E} = -\partial \boldsymbol{B}/\partial t$

$$H_x = \begin{cases} \dfrac{1}{q_t}\varepsilon_0\omega\left(1 - \frac{e^2\rho_0}{m^*\varepsilon_0\omega^2}\right)E_z - \dfrac{1}{q_t}J_z, & \text{in ultrathin metal film}, \\ \dfrac{\varepsilon\omega}{q_t c}E_z, & \text{else}. \end{cases} \tag{39}$$

Discretizing Eqs. (33) and (36) with an equal interval of $z$ and $z'$, hence, we have

$$\sum_{k'}^{N} M_{kk'}^{1z}(\omega, q_t, z, z')E_z(q_t, z_{k'}) + \sum_{k'}^{N} M_{kk'}^{1y}(\omega, q_t, z, z')E_y(q_t, z_{k'}) = \delta_{1k}E_{z2} + \delta_{Nk}E_{z1}\,\mathrm{e}^{-k_{z1}L}, \tag{40}$$

$$\sum_{k'}^{N} M_{kk'}^{2z}(\omega, q_t, z_{k'})E_z(q_t, z_{k'}) + \sum_{k'}^{N} M_{kk'}^{2y}(\omega, q_t, z_{k'})E_y(q_t, z_{k'}) = \delta_{Nk}E_{z2}, \tag{41}$$

where $k, k' = 1, 2, \ldots, N$, and the values of $N$ in this model are determined by the particular situations. $M_{kk'}^{1z}$, $M_{kk'}^{2z}$, $M_{kk'}^{1y}$ and $M_{kk'}^{2y}$ have the following forms

$$M_{kk'}^{1y} = \Pi_{zy}(\omega, q_t, z_k, z'_{k'})(\Delta z)^3, \tag{42}$$

$$M_{kk'}^{2y} = \begin{cases} \Pi_{\rho y}(\omega, q_t, z_k, z'_{k'})(\Delta z)^3/\varepsilon_0 - \mathrm{i}q_t, & k = k', \\ \Pi_{\rho y}(\omega, q_t, z_k, z'_{k'})(\Delta z)^3/\varepsilon_0, & \text{else}, \end{cases} \tag{43}$$

$$M_{kk'}^{1z} = \begin{cases} \Pi_{zy}(\omega, q_t, z_k, z'_{k'})(\Delta z)^3 + 2 - [\omega^2\varepsilon(z)/c^2 - \mathrm{q_t^2}](\Delta z)^2, & k = k', \\ \Pi_{zy}(\omega, q_t, z_k, z'_{k'})(\Delta z)^3 - 1, & k = k' \pm 1, \\ \Pi_{zy}(\omega, q_t, z_k, z'_{k'})(\Delta z)^3, & \text{else}, \end{cases} \tag{44}$$

$$M_{kk'}^{2z} = \begin{cases} \Pi_{\rho y}(\omega, q_t, z_k, z'_{k'})(\Delta z)^3 + 1, & k = k', \\ \Pi_{\rho y}(\omega, q_t, z_k, z'_{k'})(\Delta z)^3 - 1, & k = k' \pm 1, \\ \Pi_{\rho y}(\omega, q_t, z_k, z'_{k'})(\Delta z)^3, & \text{else}. \end{cases} \tag{45}$$

Let $A = [M_{mm'}^{1y}]_{N\times N}, B = [M_{mm'}^{1z}]_{N\times N}, C = [M_{mm'}^{2y}]_{N\times N}$, and $D = [M_{mm'}^{2z}]_{N\times N}$, we obtain

$$\bar{M}\begin{pmatrix} E_{z1} \\ E_{z2} \end{pmatrix} = 0, \tag{46}$$

with

$$\bar{D} = [\bar{D}_{kk'}]_{2N\times 2N} = \begin{pmatrix} A & B \\ C & D \end{pmatrix}^{-1}, \tag{47}$$

$$\bar{M} = \begin{pmatrix} (\bar{D}_{1,N+1} + \bar{D}_{1,2N})\,\mathrm{e}^{-k_{z1}L} & \bar{D}_{1,1} - \dfrac{\mathrm{i}k_{z2}}{q_t} \\ \left(\bar{D}_{N,N+1} + \bar{D}_{N,2N} + \dfrac{\mathrm{i}k_{z1}}{q_t}\right)\mathrm{e}^{-k_{z1}L} & \bar{D}_{N,1} + \dfrac{\mathrm{i}k_{z2}}{q_t}\,\mathrm{e}^{-k_{z1}L} \end{pmatrix}. \tag{48}$$

The frequency of plasmon excitation can be determined by $|\bar{M}| = 0$. Since $\mathrm{Im}[|\bar{M}|] \neq 0$ for all real $\omega$, the excitation is of damping. In principle we can use $|\bar{M}\omega_{\mathrm{p}} - \mathrm{i}\gamma_{\mathrm{p}}| = 0$ to obtain the plasmon frequency $\omega_{\mathrm{p}}$ and damping rate $\gamma_{\mathrm{p}}$. However, it is difficult to solve this equation, here; we do not attempt to obtain the exact solution for $\omega_{\mathrm{p}}$. Our interest is to find an approximate estimation of $\omega_{\mathrm{p}}$, and give a qualitative discussion about the plasmon excitation. Therefore we use

$$\mathrm{Re}\left|\bar{M}\right| = 0. \tag{49}$$

With the help of Eq. (46), the field amplitudes ratio is therefore

$$\frac{E_{z1}}{E_{z2}} = \frac{q_t D_{11} - \mathrm{i}k_{z2}}{q_t(D_{1,N+1} + D_{1,2N})}\,\mathrm{e}^{k_{z1}L}. \tag{50}$$

So, for given $E_{z1}$ and $E_{z2}$, the other electric field amplitude can be determined by Eq. (38). The electric field solution is obtained by solving Eq. (46). Substituting these results into Eqs. (21) and (27), the $z$-dependence of the induced charge-, current-density (averaged over the $x$–$y$ plane) can be qualitatively analysed.

## 3. Results and discussion

So far, we have finished the derivation of the equations for dynamic charge- and potential-responses. Below we present some discussion our findings and then give a simple example for the application of our theory. As an example, we consider some ultrathin metal films with different thicknesses in the vacuum. Here, we take Bohr radius $a_0 = 4\pi\varepsilon_0\hbar^2/m^*e^2$, $\Delta = \pi^2\hbar^2/2m^*a_0^2$ and $1/a_0$ as the units of length, energy and wave number, respectively.

In Fig. 2, we show the calculated dispersion relation of the surface plasmon within the linear response theory (solid line) compared with the classic case given by Economou E N (dash line). From these profiles, we find that the plasmon dispersion calculated with different methods shows the local classical behaviour at long wavelengths, but with increasing of the wave number, there are some differences between them.

In Figs. 3(a) and 3(b), we present plots of the distributions of the induced charge density and current density for various thicknesses of the ultrathin metal film. Here, we only consider the physical quantities averaged over the $x$–$y$ plane, namely, $\rho(z)$ and $J_z(z)$ given by Eqs. (21) and (27). Figure 3(a) corresponds to the case of $L = 30$. In this case, the charge distribution is considerable only around the surface of metal and otherwise almost zero. Moreover, it is symmetric with respect to the centre of the thin metal film. In addition to two peaks of charge distribution on both sides of the metal film, the charge density shows Friedel oscillations near the surface. But the current density is antisymmetric and is equal in magnitude but opposite in sign. From the curves of them, we see that the induced charge density and current density vanish at the boundaries $z = 0$ and $L$. The results are due to the unperturbed electron wave functions in Eq. (6) vanishing at the boundary.

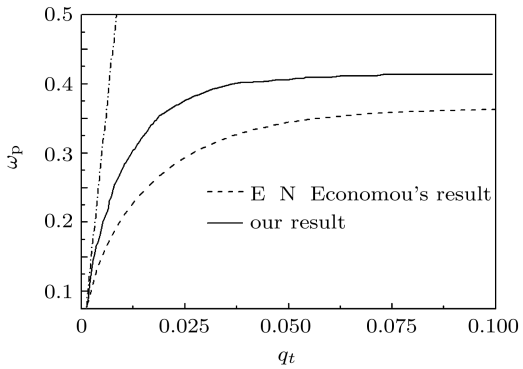



**Fig. 2.** Dispersion relation of surface plasmon in ultrathin metal film calculated in terms of linear response theory for $L = 70$. Dashed line: E N Economou's result. Solid line: our result.

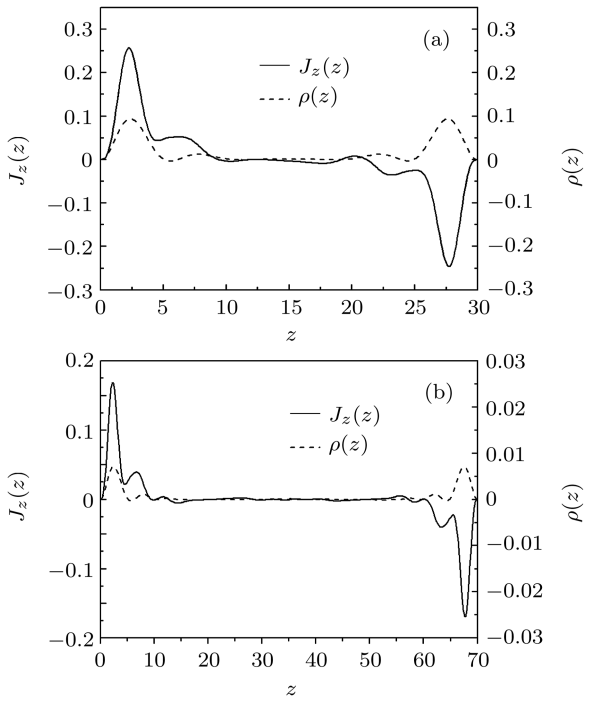



**Fig. 3.** The plots of the induced charge density and current density (averaged over the $x$–$y$ plane) for $L = 30$ (a) and $L = 70$ (b). Solid line: current density; dashed line: charge density.

In Fig. 3(b), we present the charge density and current density as functions of $z$ for $L = 70$. From the profiles of them, one can find that they also show the same behaviours as the results given in Fig. 3(a). Comparing the peak strength for different thicknesses, we find that the peak strength decreases with the increase of the thickness of the ultrathin metal film. Furthermore, the peak in the charge density is right corresponding to the peak (or valley) of the current density.

## 4. Conclusion

In conclusion, we have theoretically obtained the dispersion relation of surface plasmon in ultrathin metal films and the induced charge density and current density within the general linear response theory and found intriguing qualitative differences with the corresponding classical case. We conclude from the above results that there is a considerable amount of charge distribution on both sides of the surfaces of the ultrathin metal film and that there are many fluctuations near the surface region and two significant peaks in the curve. We emphasize the fact that the charge distribution is symmetric with respect to the centre of the system. The current density displays similar behaviour to that of charge density, but it is antisymmetric and the positive (negative) current peak corresponds to the largest charge peak.

## References


[1] Economou E N 1969 *Phys. Rev.* **182** 2

[2] Ngai K L, Economou E N and Cohen M H 1970 *Phys. Rev. Lett.* **24** 61

[3] Yang P F, Gu Y and Gong Q H 2008 *Chin. Phys.* B **17** 3880

[4] Chiang T C 2000 *Surf. Sci. Rep.* **39** 181

[5] Moskovits M 1985 *Rev. Mod. Phys.* **57** 783

[6] Echenique P M, Berndt R, Chulkov E V, Fauster Th, Goldmann A and Höfer U 2004 *Surf. Sci. Rep.* **52** 219

[7] Ritchie R H 1957 *Phys. Rev.* **106** 874

[8] Bennett A J 1970 *Phys. Rev.* B **1** 203

[9] Schwartz C and Schaich W L 1982 *Phys. Rev.* B **26** 7008

[10] Dharamvir K, Singla B, Pathak K N and Paranjape V V 1993 *Phys. Rev.* B **48** 12 330

[11] Tsuei K D, Plummer E W and Feibelman P J 1989 *Phys. Rev. Lett.* **63** 2256

[12] Tsuei K D, Plummer E W, Liebsch A, Kempa K and Bakshi P 1990 *Phys. Rev. Lett.* **64** 44

[13] Eguiluz A G 1983 *Phys. Rev. Lett.* **51** 1907

[14] Eguiluz A G 1985 *Phys. Rev.* B **31** 3303

[15] Zangwill A and Soven P 1980 *Phys. Rev.* A **21** 1561

[16] Stott M J and Zaremba E 1980 *Phys. Rev.* A **21** 12

[17] Dobson J F 1992 *Phys. Rev.* B **46** 10163

[18] Schaich W L and Dobson J F 1994 *Phys. Rev.* B **49** 14700

[19] Liu B C, Yu L, Lu Z X and Zhang K 2010 *Chin. Phys.* B **19** 097303

[20] Zhang H F, Cao D, Tao F, Yang X H, Wang Y, Yan X N and Bai L H 2010 *Chin. Phys.* B **19** 027301

[21] Lee H J and Cho S H 1997 *J. Plasma Phys.* **58** 409

[22] Rocca M 1995 *Surf. Sci. Rep.* **22** 1