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Nonlocal effect in surface plasmon polariton of ultrathin metal films

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ABSTRACT

Using the nonlocal conductivity based on quantum response theory, we study the optical properties of p-polarized wave in quartz–metal–film–air structures, especially the influence of nonlocal effect on the surface plasmon polaritons (SPPs) resonance. In absorption spectrum, the resonant peak of SPP is found, and the dependence of the resonant peak on film thickness shows that nonlocal effect in the SPP resonance is enhanced significantly with the decrease of film-thickness, especially in the less than 20 nm metal film. We calculate the surface charge density as a function of frequency, and find that the frequencies at the charge and absorption peaks are the same. This clearly confirms that the absorption peak stems from SPP resonance excitation, and SPPs absorb the energy of the electromagnetic wave via charge oscillations. In the case of SPP resonance, the charge and electric field on the down-surface of thin film are always greater than that on the up-surface; however, the situation is just opposite in the case of no SPP resonance. This implies that the SPP resonance occurs near the down-surface of the film. Moreover, due to the nonlocal response of electric current to the electric field, the energy flow and electric current show anomalous oscillations, and with the increase of film thickness the anomalous oscillations exhibit obvious attenuation.

1. Introduction

Recently, with the rapid progress in nanofabrication and measurement techniques, optical properties in metallic nanostructures have been extensively investigated. One of the main driving forces of this is their unique optical properties such as local field enhancement, negative refractive index, and surface-enhanced Raman spectroscopy. These unique properties mainly stem from the resonance excitations of surface plasmon polariton (SPP) and play very important role in the promising applications of nanosensing [1–3], bio-imaging [4–7], and optical cloaking [8,9].

In nanostructures, surface effect leads that in metals the response of electric current $\mathbf{J}$ to an electric field $\mathbf{E}$ should be a nonlocal form as

$$\mathbf{J}(\mathbf{r}\omega) = \int \sigma(\mathbf{r}, \mathbf{r}', \omega)\mathbf{E}(\mathbf{r}', \omega)\, \mathbf{dr}' \tag{1}$$

in place of the usual local form $\mathbf{J}(\mathbf{r}, \omega) = \sigma(\omega)\mathbf{E}(\mathbf{r}, \omega)$. In fact, the nonlocal response Eq. (1) is a more generalized form and valid for all cases. It is the nonlocal response to induce the SPP excitation. This is because via the nonlocal response a transverse external field may induce in surface regions of metal current fields $\mathbf{J}(\mathbf{r}, \omega)$, which have nonzero divergence, and further induce charge oscillation. In earlier studies [10–12] of SPPs, the local response or dielectric constant is used, and Maxwell's equations are solved only in the 'bulk' regions in which there are no induced charges, while the surface regions where the induced charges exist are excluded. However, due to strong surface effect, in some nanostructures such 'bulk' regions nearly do not exist. Various nonlocal-response theories and methods have been applied to investigate properties of nanostructures [13–28]. In the studies of field distribution, scattering cross section, and Hamaker coefficients, the obvious differences of the results based on local and nonlocal response theories have been reported [29–38].

The purpose of this paper is to study the optical properties of p-polarized wave in quartz–film–air structure, especially in the case of the resonant excitations of SPP. Combining absorption spectrum with the charge and electric field distributions, we systematically investigate SPP excitations, and demonstrate that the absorption peak originates from the SPP resonant excitations near the down-surface of the film. The absorption coefficient and surface charge are calculated as functions of frequency, both the absorption and charge peaks are found, and the two peaks correspond to the same frequency. This clearly confirms that SPP just is aided by charge oscillation to absorb the energy of

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electromagnetic wave. At the frequency corresponding to the peak, the charge and electric field on down-surface are greater than the ones on up-surface. These imply that the resonance excitations occur near the down-surface of the film. To study the dependence of optical properties on the film-thickness, we calculate the absorption spectrum and electric current as functions of the film-thickness, and find that the difference between the results calculated by nonlocal and local theories decreases with increasing film-thickness. This indicates that the nonlocal effect on the optical properties is obvious in the ultrathin film systems, but with the increase of the film-thickness the nonlocal effect would be weakened. In addition, due to the nonlocal response of electric current to the electric field, the electric current and energy flow show anomalous oscillations in the direction perpendicular to the film.

2. Theoretical model and the calculation method

2.1. Nonlocal conductivity in real space

In order to calculate nonlocal response of ultrathin metal film, we need to find a nonlocal conductivity of real space in the direction perpendicular to the metal film surface. Using a method similar to the one offered by Agarwal et al. [39], we can get the nonlocal dynamic conductivity by the inverse Fourier transform of the conductivity in wave vector space as follows:

$$\sigma(\omega, q_x, z, z') = \int \sigma(\omega, q_x, q_z)\exp(iq_z(z-z'))\frac{dq_z}{2\pi} \tag{2}$$

where ω is the angular frequency, q_x and q_z respectively are the wave vector components parallel and perpendicular to metal film surface (see Fig. 1). Since we are interested in p-polarized wave in this work, we need transverse σ_T and longitudinal σ_L conductivities. The σ_T and σ_L can be written as [40]

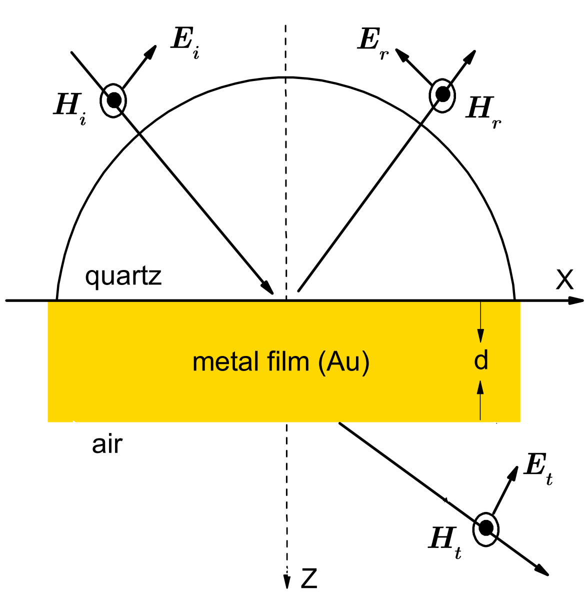

Fig. 1. Schematic view of the quartz–film–air structure for p-polarized incident wave.

$$\sigma_T(\mathbf{q},\omega) = \frac{iNe^2}{\omega m}\left(\frac{3}{8}\left[\left(\frac{q}{2k_F}\right)^2 + 3\left(\frac{\omega+i/\tau}{qv_F}\right)^2 + 1\right] - \frac{3k_F}{16q}\left[1-\left(\frac{q}{2k_F}-\frac{\omega+i/\tau}{qv_F}\right)^2\right]^2 \ln\left\{\frac{\frac{q}{2k_F}-\frac{\omega+i/\tau}{qv_F}+1}{\frac{q}{2k_F}-\frac{\omega+i/\tau}{qv_F}-1}\right\} - \frac{3k_F}{16q}\left[1-\left(\frac{q}{2k_F}+\frac{\omega+i/\tau}{qv_F}\right)^2\right]^2 \ln\left\{\frac{\frac{q}{2k_F}+\frac{\omega+i/\tau}{qv_F}+1}{\frac{q}{2k_F}+\frac{\omega+i/\tau}{qv_F}-1}\right\}\right) \tag{3}$$

and

$$\sigma_L(\mathbf{q},\omega) = \frac{3\omega}{4\pi i}\frac{\omega_p^2}{q^2v_F^2}\left(\frac{1}{2}+\frac{k_F}{4q}\left[1-\left(\frac{q}{2k_F}-\frac{\omega+i/\tau}{qv_F}\right)^2\right] \ln\left\{\frac{\frac{q}{2k_F}-\frac{\omega+i/\tau}{qv_F}+1}{\frac{q}{2k_F}-\frac{\omega+i/\tau}{qv_F}-1}\right\} + \frac{k_F}{4q}\left[1-\left(\frac{q}{2k_F}+\frac{\omega+i/\tau}{qv_F}\right)^2\right] \ln\left\{\frac{\frac{q}{2k_F}+\frac{\omega+i/\tau}{qv_F}+1}{\frac{q}{2k_F}+\frac{\omega+i/\tau}{qv_F}-1}\right\}\right) \tag{4}$$

Here, ω is the angular frequency, v_F is the velocity of fermi, k_F is the wave vector of fermi, N is the density of electron, e is the electronic charge magnitude, τ is the electron relaxation time, m is the electron mass, $q=\sqrt{q_x^2+q_z^2}$ is the wave vector, $\omega_p=\sqrt{Ne^2/m\varepsilon_0}$ is the plasma frequency, and ε_0 is the vacuum dielectric constant. The corresponding parameter values for gold are $v_F=1.4\times10^8\ \mathrm{cm\ s^{-1}}$, $k_F=1.21\times10^8\ \mathrm{cm^{-1}}$, $N=5.98\times10^{22}\ \mathrm{cm^{-3}}$, $e=1.6\times10^{-19}\ \mathrm{C}$, $\tau=5.5449\times10^{-14}\ \mathrm{s}$, and $m=9.1\times10^{-31}\ \mathrm{kg}$. Substituting Eqs. (3) and (4) into Eq. (2), respectively, we can obtain the nonlocal transverse $\sigma_T(\omega, q_x, z, z')$ and longitudinal $\sigma_L(\omega, q_x, z, z')$ conductivities of real space, which are abbreviated as $\sigma_T(z, z')$ and $\sigma_L(z, z')$ in the following parts.

2.2. The fields in metal film

For the p-polarized incident wave case, there are transverse and longitudinal electric fields in metal film. Because of the different responses of electric current to transverse and longitudinal electric fields, we need to distinguish them. In this case, Maxwell equations can be written as

$$\mathbf{E} = \mathbf{E}_L + \mathbf{E}_T \tag{5}$$

$$\nabla\cdot\mathbf{E}_T = 0 \tag{6}$$

$$\nabla\times\mathbf{E}_T = i\omega\mu_0\mathbf{H} \tag{7}$$

$$\nabla\cdot\mathbf{E}_L = \frac{\rho}{\varepsilon_0} \tag{8}$$

$$\nabla\times\mathbf{E}_L = 0 \tag{9}$$