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An improved dispersion law of thin metal film and application to the study of surface plasmon resonance phenomenon

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ABSTRACT

The dispersion law of metal film based on the Drude's free-electron theory is improved by employing the Fuchs–Sondheimer theory. In the new law, the thickness and surface roughness are taken into account to describe theoretically the dispersion of thin metal films. Then the improved model is used to analyze the surface plasmon resonance (SPR) of gold films. The results indicate that the thickness and surface roughness of gold film affect the resonance properties significantly. Specifically, when the film thickness is getting close to the electron mean-free-path (MFP), there is no obvious SPR phenomenon. In addition, the surface roughness of metal film can change the resonance angle of SPR. When the surface becomes smooth, resonance angle of SPR will decrease. It means that, for a certain metal the effects of film thickness and surface roughness should be considered in SPR technique.

1. Introduction

Surface plasmon resonance (SPR) phenomenon is a coupled electromagnetic field-charge density oscillation that occurs on the boundary of a dielectric and a metal whose real parts of dielectric constants have opposite signs [1,2]. For its high precision and sensitivity to the optical properties of dielectric above the metal films, SPR-based sensing devices have been successfully exploited in recent years [3,4]. Dispersion characteristic of thin metal (e.g., gold, silver, copper) film is a key factor affecting properties of SPR. So far, the dispersion of metal film is mainly based on the Drude's free-electron theory [5,6]. However, it was seen from the reported experimental results that the Drude model failed to describe the dispersion of nanoscale metal films. That is because Drude model is based on the assumption of 'semi-infinite' metal [7,8].

The well-known electrical conductivity theory of Fuchs–Sondheimer (FS) model describes the relationship between film conductivity and semi-infinite conductivity [9–11]. Though this model has its restriction for its simplifying assumption, the size-effects of film have been considered theoretically. So employing the FS theory, we propose an improved Drude–Lorentz model (called FS–DL model in this paper) for determining the dispersion law of thin metal films with the size-effects containing thickness and surface roughness taken into account. Furthermore, the FS–DL model was used to discuss the influences of thickness and surface roughness on SPR phenomenon in gold film.

2. Models of dispersion

2.1. The Drude and Drude–Lorentz models

The most commonly used model for metal permittivity is the classical Drude model for describing the electrical conduction properties of materials. In the Drude model [5], metals are characterized by free-electron gas that moves within the metal lattice. Under an applied electric field $\vec{E}=\vec{E}_0 e^{-i\omega t}$, the Drude model can be expressed as

$$m_e\frac{\partial^2\vec{r}}{\partial t^2}+m_e\Gamma_p\frac{\partial\vec{r}}{\partial t}=e\vec{E} \tag{1}$$

where e and m_e are the charge and the effective mass of the free electrons, and Γ_p is the damping rate (approximately 10^{14} rad/s). Solving (1) and using the Ohm's law $\vec{J}=\sigma\vec{E}$ yield the frequency-dependent electrical conductivity of metal

$$\sigma=\frac{Ne^2}{m_e(\Gamma_p-i\omega)} \tag{2}$$

here N is the number of free-charge in per unit volume. Meanwhile, the interaction of electromagnetic radiation with metal is

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described by the Maxwell equations. When complex permittivity was introduced, the relative dielectric permittivity of metal can be expressed as:

$$\varepsilon(\omega) = 1 + \frac{i\sigma}{\varepsilon_0 \omega} \tag{3}$$

Substituting (2) into (3), we get the metal dielectric permittivity of Drude model

$$\varepsilon(\omega) = 1 + \frac{iNe^2}{\varepsilon_0 \omega m(\Gamma_p - i\omega)} = 1 - \frac{{\omega_p}^2}{\omega^2 + i\Gamma_p \omega} \tag{4}$$

where $\omega_p = \sqrt{Ne^2/\varepsilon_0 m_e}$ is the plasma frequency of metal (approximately 10^{16} rad/s).

Although electron–electron interactions and frequency-dependent interband transitions are ignored in Drude model, it is still in good agreement with the experimentally measured permittivity data for the noble metals in the near infrared region [12,13]. In order to match experimental data more closely over a wide spectrum, the Drude model needs to be supplemented in the visible range by adding frequency-dependent contributions from interband transitions. So the Drude model was extended to the Drude–Lorentz model added with multiple additional Lorentzian terms [6]. And the expression of permittivity of metal can be extended to

$$\varepsilon(\omega) = 1 - \frac{{\omega_p}^2}{\omega^2 + i\Gamma_p \omega} - \sum \frac{f_n {\omega_n}^2}{(\omega^2 - {\omega_n}^2) + i\Gamma_n \omega} \tag{5}$$

where ω_n and Γ_n, stand for the oscillator strength and the spectral width of the Lorentz oscillators, respectively, and f_n can be interpreted as weighting factors.

Up to now, a wide variety of analytical model of dispersion of metal based on the Drude and Drude–Lorentz models have been developed [6–8]. Unfortunately, these models are not suitable for describing the dispersion properties of thin metal films because the size-effects such as thickness or surface roughness are not considered.

2.2. FS–DL model

We try to give an analytical description of dispersion law of thin metal film with size-effects taken into account. For this aim, the well-known FS theory is employed. The FS model has been used successfully by many authors to calculate the electrical resistivity and temperature coefficient of the resistivity of metal films. The reduced conductivity of films can be expressed in the following form [9–11]

$$\beta(k,p) = \frac{\sigma_f}{\sigma} = \begin{cases} 1 - \frac{3}{8k}(1-p) & (k \geq 1) \\ \frac{3}{4} k \frac{(1+p)}{(1-p)} \ln\left(\frac{1}{k}\right) & (k < 1) \end{cases} \tag{6}$$

where σ_f, σ are the conductivity of film and bulk, respectively, and $k = d/\lambda_0$ is the ratio of film thickness to the electron mean-free-path (MFP), p is a constant ranging from 0 to 1, to describe the degree of electrons scattering at the surface of the film with perfectly diffuse reflection and perfectly specular reflection. Namely, p is a specularity coefficient corresponding to the surface roughness of film [10].

For the case of metal films, the bulk conductivity σ in Eq. (3) can be replaced by the film conductivity σ_f. So the relative dielectric permittivity of film in the Drude model can be expressed as follows:

$$\varepsilon(\omega, k, p) = 1 - \frac{\beta(k,p){\omega_p}^2}{\omega^2 + i\Gamma_p \omega} \tag{7}$$

Considering the contribution from interband transitions, the improved Drude–Lorentz model (i.e. FS–DL model) of permittivity can be written as:

$$\varepsilon(\omega, k, p) = 1 - \frac{\beta(k,p){\omega_p}^2}{\omega^2 + i\Gamma_p \omega} - \sum \frac{f_n {\omega_n}^2}{(\omega^2 - {\omega_n}^2) + i\Gamma_n \omega} \tag{8}$$

Thus the size-effects dependent dispersion of metal film can be characterized.

2.3. Validation of the new approach

In order to evaluate the performance of our approach, we fit the permittivity of gold film published in Ref. [14] by FS–DL model. For comparison, the results obtained with Drude and Drude–Lorentz models were also given. Here we restrict the Drude–Lorentz and FS–DL models to two additional Lorentzian terms. To determine the best set of parameters, we employ the optimization scheme as performing the minimization of function

$$Z = \sum_{\omega_j} \left\{Re\left[\varepsilon_{\text{Model}}(\omega_j) - \varepsilon_{\text{Exp}}(\omega_j)\right]\right\}^2 + \left\{\text{Im}\left[\varepsilon_{\text{Model}}(\omega_j) - \varepsilon_{\text{Exp}}(\omega_j)\right]\right\}^2 \tag{9}$$

where ω_j are the discrete values of the frequency $\omega = 2\pi c/\lambda$ for which the permittivity is calculated. The results are presented in Table 1, and the real and imaginary parts of the permittivity calculated with Drude, Drude–Lorentz and FS–DL models are plotted in Fig. 1. Here, we take the thickness of gold films $d = 45$ nm as in Ref. [14].

From Fig. 1, one can see that the results of FS–DL model are almost coincide with that of Drude–Lorentz model, and agree well with the experimental values in the range of 400–800 nm. But

Table 1
Values of the parameters used for the optimization of dispersion models.

	Drude	Drude–Lorentz	FS–DL
λ_0 (nm)			30.8795
p			0.5707
ω_p (eV)	7.3325	8.7329	9.2593
Γ_p (eV)	0.1855	0.0445	0.0445
f_1		3.6126	3.6128
ω_1 (eV)		4.0095	4.0095
Γ_1 (eV)		0.4782	0.4783
f_2		1.4232	1.423
ω_2 (eV)		2.9227	2.9227
Γ_2 (eV)		0.8181	0.818
Res	480.71	1.9295	1.9295

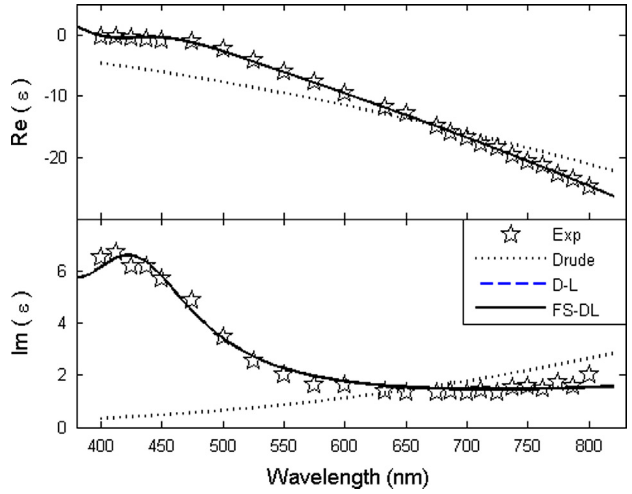

Fig. 1. Experimental data of real and imaginary parts of gold permittivity as published in Ref. [14], and results calculated with the Drude, Drude–Lorentz and FS–DL models.