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Nonlinear Carrier Responses in Gold Thin Films Induced by Intense Terahertz Waves

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Abstract: Terahertz transmittances of the gold thin-films with thicknesses ranging from 1 to 12 nm were investigated. As terahertz field becomes intense, the transmittance of the terahertz field decreases, suggesting the increase of the carrier density.

OCIS codes: (300.6495) Spectroscopy, terahertz; (310.6188) Thin films, Spectral properties; (320.7120) Ultrafast phenomena

1. Introduction

Gold (Au) has been extensively studied in the field of microelectronics and bio-sensing, since many attractive electromagnetic properties of Au nanostructures were recently reported; the effective sheet conductivity of Au thin film dramatically decreases with the thickness less than 2 nm [1]. This suggests that the nano-scale size and morphology play key roles on the electronic properties. In the present study, we investigate the carrier dynamics in Au thin film with different thicknesses using intense terahertz electric field.

2. Experiments

The Au thin film was evaporated on the high resistivity Si substrate with Si (111) — 7 x 7 surface in the chamber with the ultrahigh vacuum of ~ 10^{-7} Pa at room temperature. The crystallization process was confirmed using reflection high-energy electron diffraction, and the polycrystal and flat Au films were successfully prepared. The thicknesses of the obtained Au films were 1.3, 1.9, 5.8, and 11.5 nm. The details of the experimental setup are reported elsewhere [2,3]. Figure 1(a) shows the SEM image of the specimen with 1.9 nm thickness, which indicates the flatness of the specimen. The flatness enabled us to investigate the terahertz responses of Au thin films precisely. The experimental configuration of specimen from the side view is shown in Fig. 1(b).

A Ti:Sapphire amplifier system (repetition rate: 1 kHz, pulse duration: 130 fs, center wavelength: 800 nm, pulse energy: ~ 1.6 mJ/pulse) is employed to generate intense terahertz waves and to detect these terahertz waves. In generating terahertz fields, tilted laser pulses are irradiated onto a $LiNbO_3$ via Cherencov-type phase matching process [4]. The generated terahertz waves are incident on the specimen. Then, the transmitted waves are forwarded to a 0.4 mm-thick GaP crystal using off-axis parabolic mirrors and finally observed by the electro-optic (EO) sampling. The maximum-terahertz field was ~ 340 kV/cm. The intensity is tuned by the wire grid polarizers set in front of the specimen. The transmittance of the specimen is obtained by normalizing the transmitted spectrum of the specimen (Au on Si) by that of the reference (Si) (see Fig. 1 (b)).

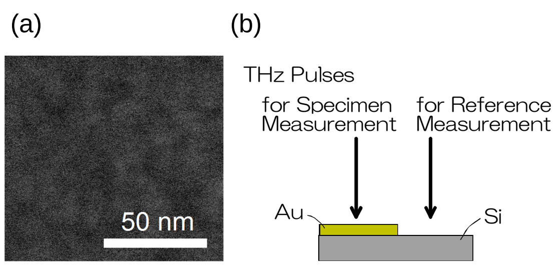

Fig. 1 (a) SEM image of Au thin film surface with 1.9 nm thickness. (b) Side view of the specimen for measuring terahertz transmittance of the reference and signal.

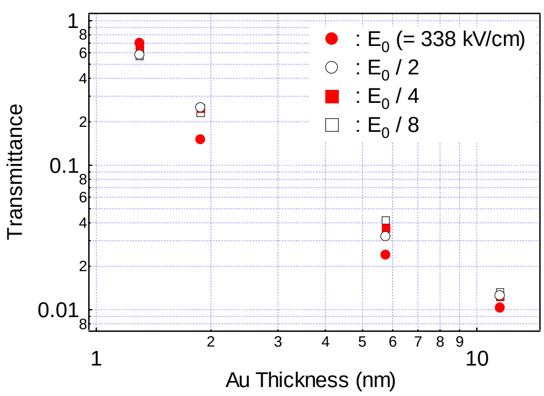

Fig. 2 Transmittance of terahertz waves as a function of the Au film thickness with several terahertz intensities.

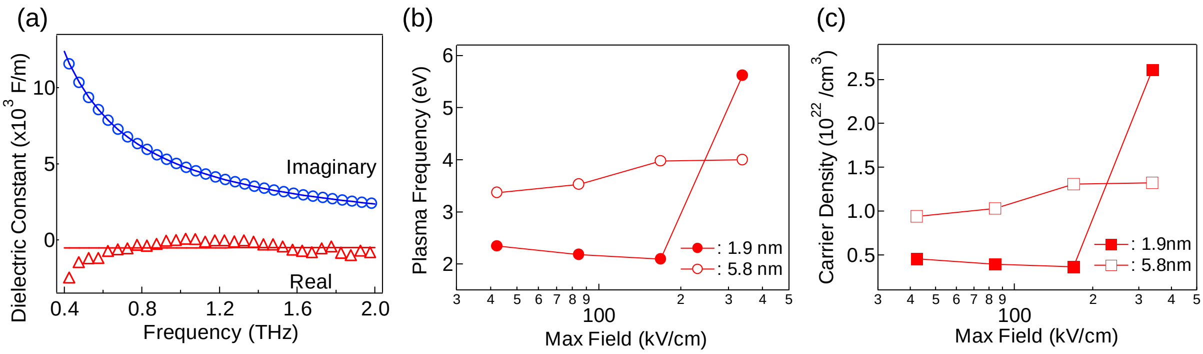

Fig. 3 (a) Complex dielectric constant of gold film with the thickness of 1.9 nm. Solid curves indicate the best-fit to the experimental data obtained by Drude analysis. (b) The estimated plasma frequencies and (c) the carrier densities of the Au films with the thicknesses of 1.9 and 5.8 nm as a function of maximum terahertz electric field.

3. Results and Discussion

Figure 2 shows the Au film thickness dependence of the transmittance with different terahertz fields. Except for the case of 1.3 nm-thick film, the transmittance becomes lower with the intense terahertz field illumination. To clarify the reason, Drude analysis was carried out for the complex dielectric constant obtained from the terahertz transmittance. For example, the complex dielectric constant obtained for the Au film with 1.9 nm thickness is shown in Fig. 3(a). In the Drude model, the plasma frequency of the free electron gas is expressed as

$$\omega_{\mathrm{p}} = \sqrt{\frac{ne^2}{\varepsilon_0 m^*}}, \tag{1}$$

where n is the carrier density, m^* is the effective mass of the carrier, e is the elementary charge, and ε_0 dielectric constant in vacuum. The plasma frequency of the Au film vs. the maximum electric field is shown in Fig. 3(b). The solid and open circles show the plasma frequencies of the 1.9 and 5.8 nm-thick Au films, respectively. In the weak terahertz field region, the plasma frequencies of 1.9 nm- and 5.8-nm thick films are similar to those obtained in Ref. [5]. In contrast to the weak terahertz field region, the plasma frequency is strongly enhanced by the intense terahertz field especially in the case of 1.9 nm-thick Au film. Assuming that the effective mass is constant in Eq. (1), the enhancement of the plasma frequency depends on the increase of the carrier density. The carrier density of the Au film vs. the maximum electric field is shown in Fig. 3(c). The solid and open squares show the carrier densities of the 1.9 and 5.8 nm-thick Au films, respectively. The carrier density of the 1.9 nm-thick Au film at 338 kV/cm terahertz field is larger than that of 5.8 nm-thick Au film. This enhancement can be explained by the penetration depth of terahertz electric field. The penetration depth of terahertz wave into the specimen was estimated to be 2 nm from Fig. 2. Therefore, the carriers in the 1.9 nm-thick film is more susceptible to the influence of terahertz electric field than that in the 5.8 nm-thick film.

In case of the 1.3 nm thick Au film, the clear enhancement was not observed, probably because of some drastic change in electronic structures taking place between 1 and 2 nm such as insulator-to-metal transition [1].

4. Conclusions

We investigated the transmittance of the Au thin films with thickness ranging from 1 to 12 nm by intense terahertz electric fields. In the case of the thickness with 1.9 – 11.8 nm, the transmittance became lower by the irradiation of intense terahertz fields. The Drude analysis clearly shows that the carrier density is enhanced by the intense terahertz field.

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