Influence of surface roughness on surface plasmon resonance phenomenon of gold film

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We experimentally investigate the effects of the surface roughness of gold thin films on the properties of surface plasmon resonance. By annealing at different temperatures, film samples with different surface morphologies are obtained. Specifically, due to the diffusion of the gold atoms towards the films' surface, the surface root-mean-square roughness decreases with the increasing annealing temperature. Then, we measure the surface plasmon resonance of the samples. The results show that the resonance angle of the surface plasmon resonance is sensitive to the root-mean-square roughness, and it gradually decreases by reducing the surface root-mean-square roughness.

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The surface plasmon resonance (SPR) phenomenon is a coupled electromagnetic field-charge density oscillation that occurs on the boundary between a dielectric and a metal[1,2]. For its high precision and sensitivity to the optical properties of dielectrics above metal films (e.g., gold, silver), SPR-based sensing devices have been successfully exploited in recent years[3,4]. The properties of the SPR in theory mainly depend on the characteristics of the thin film. At present, the dependence of the SPR on film thickness has been investigated widely.

However, the effects of the surface roughness of gold thin films on the properties of SPR have barely been studied[5]. In fact, there are discrepancies between most reported experimental measurements and the theoretical calculations of SPR even for films with a specific thickness. This inconsistency may result from researchers' ignorance of the influence of surface roughness on theoretical SPR, as discussed in Ref. [6], in which a new dispersion law of metal thin films was exploited. Therefore, it is necessary to experimentally study the effects of surface roughness on the SPR properties of metal thin films.

In this work, we prepared gold films with a specific thickness under the same deposition conditions. By annealing at different temperatures, we obtained film samples with different surface morphologies. Then, the effects of the surface roughness of samples on the properties of SPR curves were examined.

Instead of depositing them on prisms, the gold films were made on K9 cover glass substrates using sputtering deposition, which makes it is more convenient to anneal the gold films at different temperatures. The cover glasses, which had a diameter of 22 mm, were cleaned in order to deposit the target. The deposit of the gold films was carried out using a commercial sputtering system with a DC sputtering power of 23 W at a rate of 2 nm/min for 22 min. Then, the thickness of these films was examined by x ray diffraction (PANalytical–Empyrean).

To obtain samples with different surface roughness, these gold films were annealed in a muffle furnace at four different temperatures (100°C, 200°C, 300°C, and 400°C) for 15 min. The surface roughness of the samples was measured using an atomic force microscope (AFM, Bruker-V Multimode) in contact mode, and characterized with root-mean-square roughness (RMS-roughness). We found that the surface roughness of the sample without annealing was almost the same as that annealed at 100°C. So in this work, we selected four samples annealed at 100°C, 200°C, 300°C, and 400°C (corresponding to samples 1, 2, 3, and 4, respectively) for later measurements.

In order to examine the influence of the surface roughness on the SPR of gold films, we use the angle modulation in the SPR measurement depicted in Fig. 1. The incident angle is regulated by rotating the right-angle prism. When the coupling condition of the incident light has been satisfied, there will be a resonant angle of the measured reflected light intensity versus the incident angle. The angle-modulation method is commonly and widely used in SPR techniques; the details can be viewed in Refs. [1,2]. Here, we employ a TM-polarized laser with a wavelength of 632.8 nm as the incident light beam. The cover glass with the gold film is attached at the inclined plane of the right-angle prism (K9 glass) by an index-matching fluid.

As shown in Fig. 2, the reflection intensity versus the angle position of the film is scanned for angle region

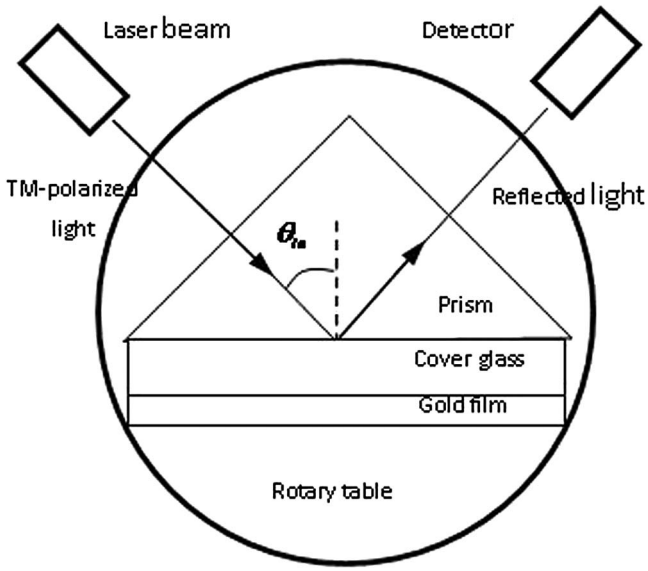

Fig. 1. Configuration of the angle-modulation SPR measurement.

0.2°–2°. An x ray scan on a gold film in the small-angle region shows the oscillation associated with the thickness of the film. The thickness of the film can be calculated according to the following formula[7,8]:

$$d = \frac{\lambda}{2(\sin\theta_{n+1} - \sin\theta_n)} \approx \frac{\lambda}{2(\theta_{n+1} - \theta_n)}, \qquad (1)$$

where d, λ, and θ_n are the thickness of film, the wavelength of the x ray, and the angle position of the n-class interference peak, respectively. The thicknesses of the gold film samples annealed at the temperatures of 100°C, 200°C, 300°C, and 400°C are measured. We find that the thickness of samples is almost constant in the error range, which is probably related to the shot annealing duration (for 15 min). Here, we measure the thickness of the gold films to be $d = 57.50$ nm.

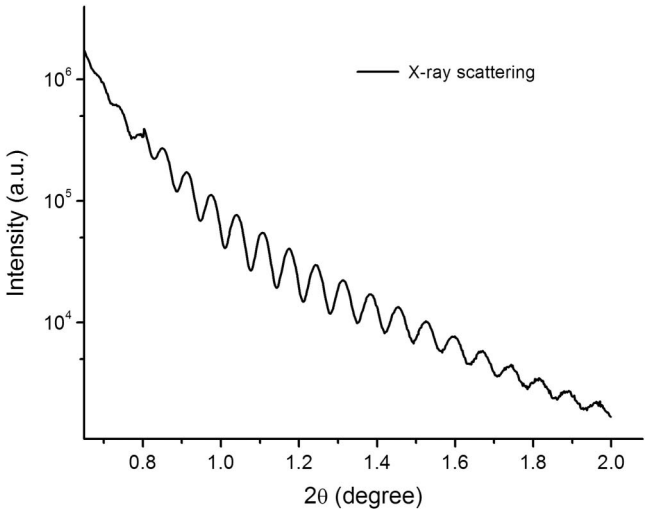

Fig. 2. Reflection Intensity of small-angle x ray scattering from the gold film.

The surface morphologies of the gold films scanned over 250 nm × 250 nm by the AFM in contact mode are shown in Fig. 3. These images correspond to the samples annealed at the different temperatures: 100°C, 200°C, 300°C, and 400°C (denoted as samples 1, 2, 3, and 4). From these images, the differences in the morphologies are clearly observed, and the surface RMS-roughness of each sample is 1.260, 0.906, 0.700, and 0.415 nm, corresponding to samples 1, 2, 3, and 4, respectively. We can see that with the increasing annealing temperature, the surface RMS-roughness decreases, i.e., the film surface becomes smooth. This phenomenon results from the diffusion of the gold atoms toward the surface when annealed[9,10]. Due to the slow diffusion rate and less mobility at a low temperature, the surface morphologies of the sample annealed at 100°C and the one that was not annealed are the same.

By the angle-modulating method, we examine the influence of the surface roughness of the gold films on the SPR phenomenon. The SPR curves of gold film samples with different surface roughness are shown in Fig. 4, and a quantitative analysis of the dependence of the SPR phenomenon on the surface roughness of the samples is summarized in Table 1.

From the results in Table 1, we can see that the dependence of the resonance depth on the RMS-roughness of the surface is not evident. The resonance angle versus the RMS-roughness of theses samples is plotted in Fig. 5. We found that the resonance angle increases linearly with the RMS-roughness. So we fitted these data with a linear formula:

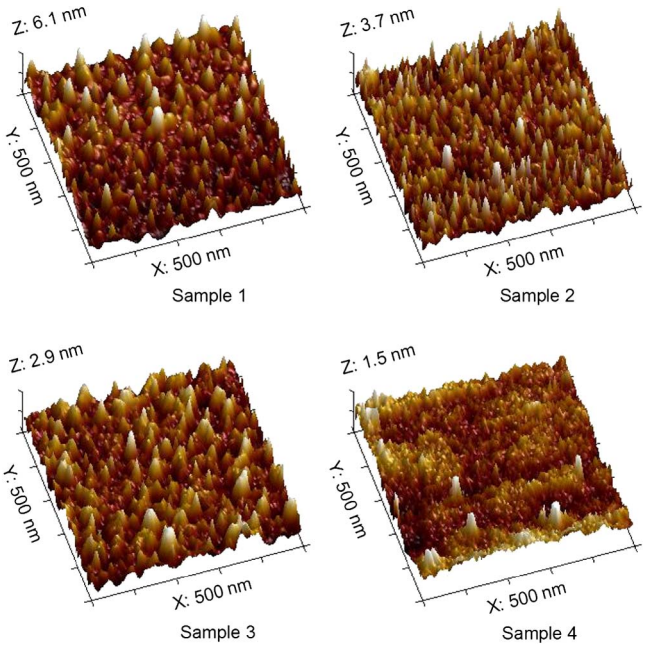

Fig. 3. Surface morphology images of samples detected by AFM.

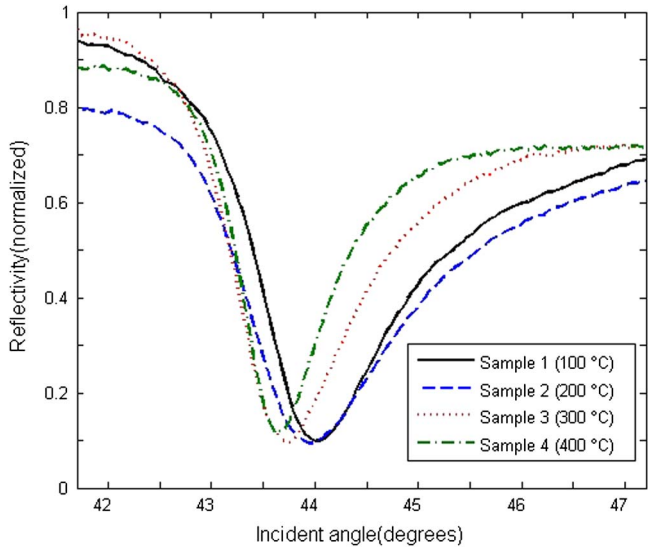

Fig. 4. SPR measurement of samples with different surface roughness.

Table 1. Results of Surface RMS-Roughness and SPR of Samples

Sample	Annealing temperature (°C)	RMS-roughness (nm)	Resonance angle (°)	Resonance depth
1	100	1.260	44.04	0.0976
2	200	0.906	43.94	0.0935
3	300	0.700	43.75	0.0947
4	400	0.415	43.65	0.1160

$$y = A + Bx. \tag{2}$$

Here, x and y are the RMS-roughness and resonance angle, respectively. There are two undetermined constants A and B, and the fitting results are listed in the inset table of Fig. 5. The value of the slope $B = \frac{dy}{dx} = 0.48475°/\text{nm}$ indicates that the resonance angle is very sensitive to the surface RMS-roughness. These results can be explained according to the calculations reported in Ref. [6].

In the Ref. [6], an improved dispersion law of metal thin films (FS-DL model) was presented, and the permittivities of gold film with different surface roughness characterized by specularity were calculated. By employing the FS-DL model, the dependence of the specularity on the SPR was discussed. The results show that the resonance angle decreases by increasing the specularity from 0 to 1, which corresponds to the surface's change from rough to smooth. In the present work, our experimental measurements prove the calculations in Ref. [6], i.e., the resonance angle will decrease when the surface of the gold film becomes smooth.

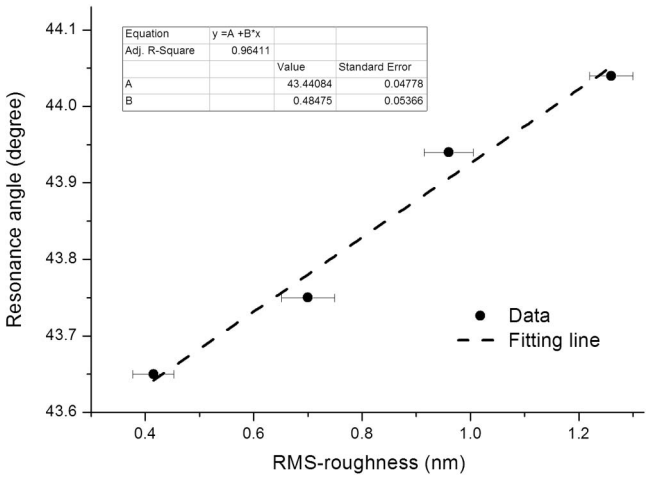

Fig. 5. Data of resonance angle versus RMS-roughness and the results of the fitting.

We investigate the effects of surface roughness on the SPR of gold films. The results show that the resonance angle of the SPR is sensitive to the surface roughness of the gold film. Specifically, the resonance angle gradually decreases with the reducing RMS-roughness, and the change rate of the resonance angle with RMS-roughness is about 0.485°/nm. These results are accord with the reported theoretical calculations. Therefore, the influence of the surface roughness of films on the SPR should be taken into account in the SPR technique.

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