

# Optical performance of ultra-thin silver films under the attenuated total reflection mode

Ming ZHOU (✉)<sup>1</sup>, Sheng ZHOU<sup>1</sup>, Gang CHEN<sup>1</sup>, Yaopeng LI<sup>1</sup>, Dingquan LIU<sup>1,2</sup>

<sup>1</sup> Shanghai Institute of Technical Physics, Chinese Academy of Sciences, Shanghai 200083, China

<sup>2</sup> School of Physical Science and Technology, ShanghaiTech University, Shanghai 200031, China

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**Abstract** Ultra-thin silver films were deposited by thermal evaporation, and the dielectric functions of samples were simulated using Drude-Lorentz oscillators. When *s*-polarized incident light from the BK7 glass into thin silver film at 45° angle using attenuated total reflection (ATR) mode, we experimental observed that the reflection reach a minimum of 1.87% at 520 nm for thickness of *d*~6.3 nm silver film, and it reach a minimum of 10.1% at 500 nm for thickness of *d*~4.1 nm. Moreover, we simulated the absorption changes with incident angles at 520 nm for both *p*-polarized (TM wave) and *s*-polarized (TE wave) light using transfer matrix theory, and calculated the electric field distributions. The absorption as a function of incident angles of TM wave and TE wave showed different characteristics under ATR mode, TE wave reached the maximum absorption around the critical angle  $\theta_c \sim 41.1^\circ$ , while TM wave reached the minimum absorption.

**Keywords** silver films, transfer matrix theory, absorption, ellipsometer

## 1 Introduction

Thin silver films have been widely used in photoelectric devices, such as beam splitters [1], solar cells [2], sensors and detectors devices [3–5]. Ultra-thin silver film shows high transmittance at visible wavelengths and high reflection at infrared in D/M/D multilayer [6], and so it can obtain visible light from broadband spectrum. The ultra-thin layers provide an enhanced absorption effect using various materials [7,8]. Tischler et al. [7] used a 5 nm thick organic material and a planar mirror, obtained the absorbing larger than 97% for 591 nm wavelengths.

Driessen and de Dood [8] measured the absorption in 4.5 nm thick NbN film as a function of the angle of incidence, and absorption goes to a maximum value of 94% for *s*-polarized light at wavelength of 775 nm. As a kind of high permittivity materials, silver films also have high absorption, such as antireflection multilayer design [9]. If use attenuated total reflection (ATR) configuration consist of a prism to increases the momentum of the incoming photon [10], the light absorption should be strengthened, and this process would improve the application of thin silver films in sensors or detectors devices.

In this paper, we used thermal evaporation to fabricate ultra-thin silver films, and analyzed the surface morphologies using scanning electron microscopy (SEM) and atomic force microscopy (AFM). We reported that it is possible to obtain highly absorption of ultra-thin silver films (*d*~6.3 nm), when it was illuminated by TE wave under ATR mode beyond the critical angle. In this case, we calculated light reflectance, transmittance and absorption of ultra-thin silver under ATR mode using transfer matrix equation, and investigated the absorption various as a function of incident angles. Moreover, we discussed the physical phenomenon from the electric field distribution from thin film design software.

## 2 Experimental and theoretical method

2.1 Preparation process and optical constants characterize for ultra-thin silver films

Two different thickness silver films were coated on BK7 glasses, substrates were cleaned using alcohol mixed with ether. The deposition process was performed at room temperature, and the film thickness growth rate is ~1.2 nm/s. The chamber reaches high vacuum ( $\sim 1.5 \times 10^{-3}$  Pa) after 2 hours pumping. Tungsten was used as the evaporation source, and film thicknesses were controlled

by deposited time. Variable angle spectroscopic ellipsometer (VASE) was used to obtain parameters  $\Delta$  and  $\Psi$ , while transmittance ( $T$ ) was measured by spectrophotometer (Lambda 900) from 400 to 1000 nm. The ellipsometry measurement angles are  $55^\circ$  and  $65^\circ$ , transmittance measurement angle is  $0$ . The dielectric functions of ultra-thin silver films (Ag-I, Ag-II) were derived from combination of ellipsometry ( $\Delta$ ,  $\Psi$ ) and transmittance ( $T$ ). The physical model is linear combination of Drude oscillator and single Lorentz oscillator.

## 2.2 Reflectance measurement and theoretical analysis of ultra-thin silver films under ATR mode

BK7 substrate is attached under the bottom of BK7 triangular prism using index-matching fluid ( $n \sim 1.516$ ), and the simple configuration diagram is shown in Fig. 1. Ellipsometry is used to characterize the reflectance of broadband spectrum, and the actual reflectance is calculated through eliminating the residual reflected light at air/BK7 interfaces.

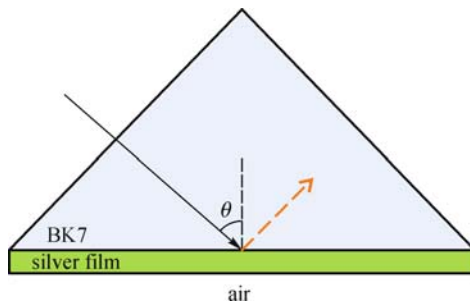


Fig. 1 ATR configuration (triangular prism) of reflection measurement

Suppose plane wave incident at angle of  $\theta$  using the configuration shown in Fig. 1, the transfer matrix equation at the BK7/silver film interface can be written as [11]

$$\begin{bmatrix} B \\ C \end{bmatrix} = \begin{bmatrix} \cos\delta & (i\sin\delta)/\eta \\ i\eta\sin\delta & \cos\delta \end{bmatrix} \begin{bmatrix} 1 \\ \eta_{\text{sub}} \end{bmatrix}, \quad (1)$$

where  $B$  and  $C$  respectively represent normalized tangential electric and magnetic fields at BK7/silver film interface,  $\delta = \frac{2\pi d}{\lambda} N \cos\theta$  is the phase factor of wave.  $\eta$  is admittance of silver film,  $\lambda$  is incident light wavelength, and  $N$  is the complex refractive index of thin film. Then the admittance of TM and TE wave can be written as [12]

$$\eta_p = N/\cos\theta, \quad (2)$$

$$\eta_s = N\cos\theta. \quad (3)$$

The reflectance and transmittance at oblique angle can be calculated as [12]

$$R = \left| \frac{\eta_0 B - C}{\eta_0 B + C} \right|^2, \quad (4)$$

$$T = \frac{4\eta_0 \text{Re}(\eta_{\text{sub}})}{|\eta_0 B + C|^2}, \quad (5)$$

where  $\eta_{\text{sub}}$ ,  $\eta_0$  indicate the admittance of substrate and air separately. The critical angle  $\theta_c \sim \arcsin(\eta_{\text{sub}}^{-1})$  can be calculated as  $41.1^\circ$  for BK7 substrate, when incident angle  $\theta$  is larger than critical angle  $\theta_c$ , transmittance becomes zero because  $\eta_{\text{sub}}$  is purely imaginary number. In that case, light absorption can be simplified written as [13]

$$A = 1 - R, \quad (6)$$

where  $R$  is the actual reflectance of the system, and light absorption could be calculated through the reflection measurement.

## 3 Results and discussion

Figure 2(a) shows the dielectric functions, and the simulated thicknesses are  $d_1 \sim 4.1$  nm (Ag-I) and  $d_2 \sim 6.3$  nm (Ag-II). Ultra-thin silver films show epsilon near zero (ENZ,  $\text{Re}(\epsilon) \sim 0$ ) wavelength [14] in visible spectrum, and the wavelengths are 470 nm (Ag-I), 480 nm (Ag-II). The transmittance curves of normal incidence are shown in Fig. 2(b), and the red dash lines give the simulated curves with optical constant from Fig. 2(a). Ag-I shows the lowest transmittance of 57.5% at 480 nm, and Ag-II shows the lowest transmittance of 49.7% at 495 nm. The wavelengths of the lowest transmittance are slightly longer than their ENZ wavelengths.

Figure 3 represents the SEM morphologies (silica wafers) and AFM morphologies of Ag-II. Ag-II shows the island uniform distributed surface, columnar structure and small sizes of grains can be found from the AFM pictures. The SEM surface of the ultra-thin silver film is similar with silver nanoparticles [15,16]. We chose part of region from SEM picture to analysis the size, the diameters of islands vary from 3.7 to 19.1 nm, and average diameter is 9.3 nm, which is much smaller than silver nanoparticles produced by spin-coated [2] or high temperature annealing method.

Figure 4 gives the reflectance of samples using ATR configuration. The black line gives reflectance of Ag-I, blue line show the reflectance of Ag-II. The incident angle is  $45^\circ$ , which is larger than the critical angle. The dash line gives the actual data after eliminating the residual reflection of air/BK7 interfaces. Light reflectance reach a minimum of 1.87% at 520 nm for silver film of  $d \sim 6.3$  nm, and the minimum reflectance is 10.1% for silver film of  $d \sim 4.1$  nm at 500 nm. The lowest reflectance indicates that there is a high absorption around their ENZ wavelengths, and the residual reflectance which caused by the prism

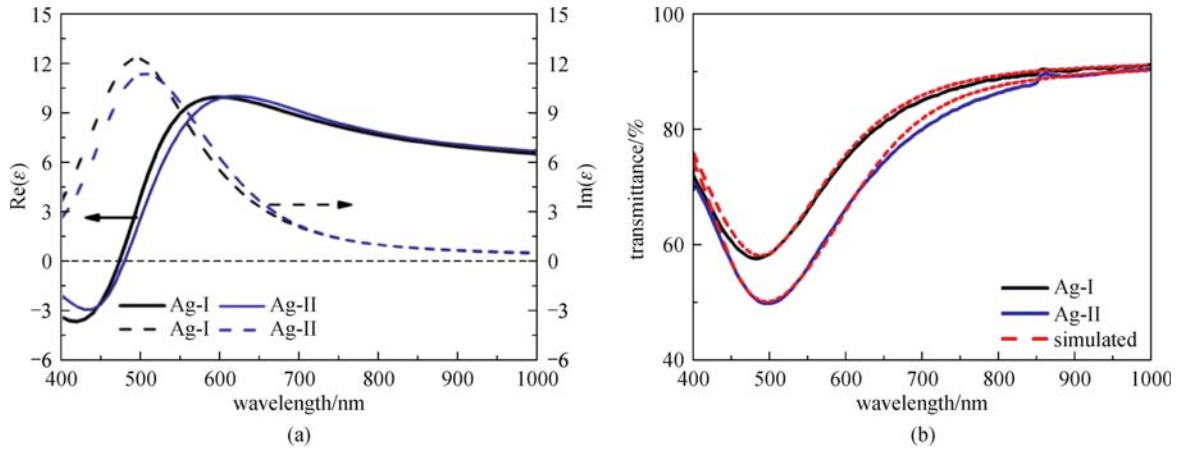


Fig. 2 Dielectric functions (a) and transmittances (b) of ultra-thin silver films (normal incidence)

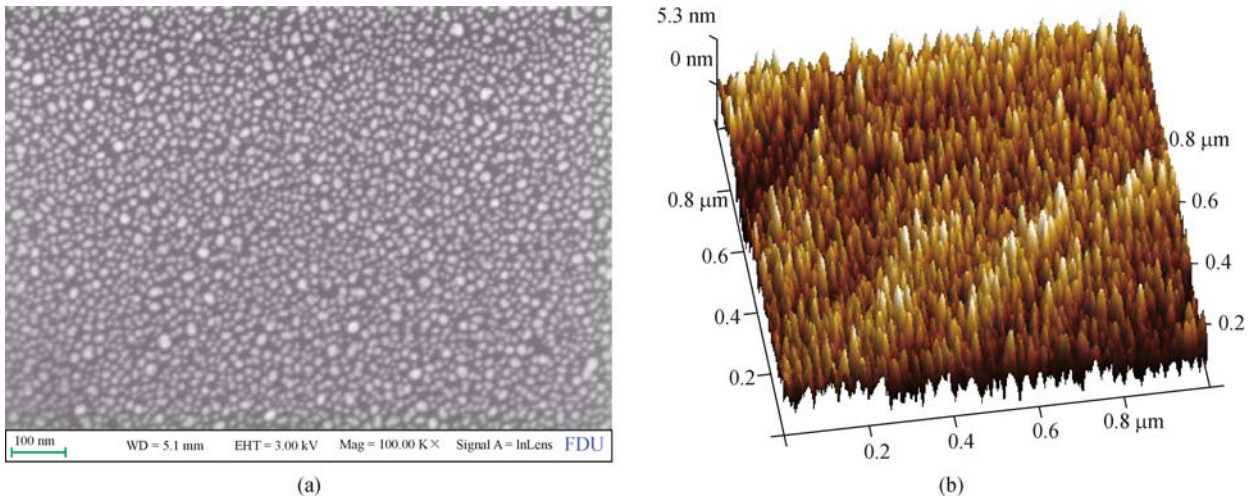


Fig. 3 SEM (a) and AFM (b) surface morphologies of Ag-II

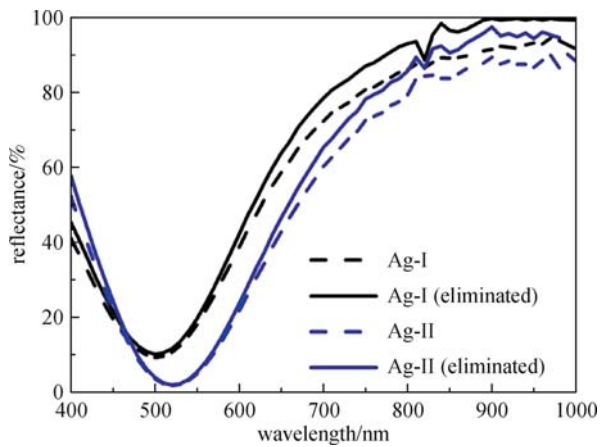


Fig. 4 Actual and measured reflectance using the ATR configuration of *s*-polarized

Using Eq. (6), we can calculate the absorption from the reflection measurement, and the absorptions as a function of wavelength for TE wave are shown in Fig. 5. For

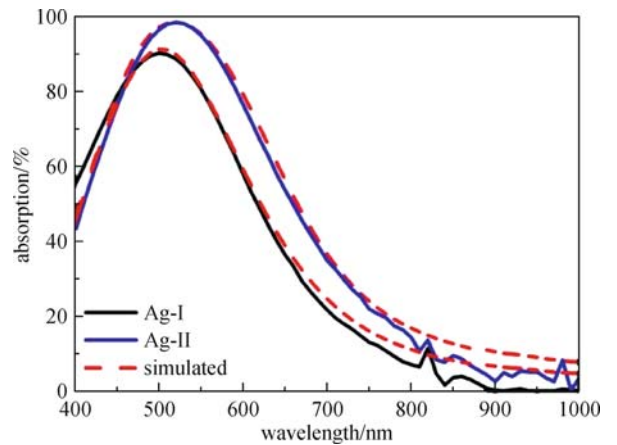
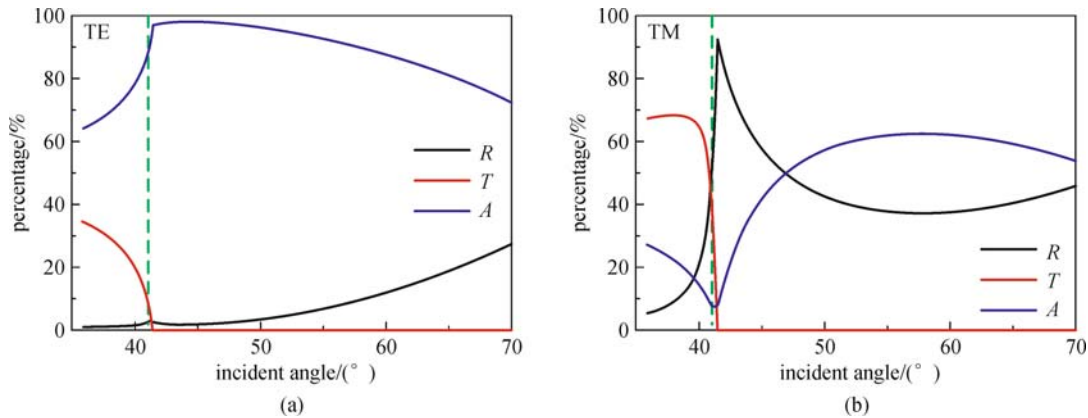


Fig. 5 Measured and calculated absorption of samples for *s*-polarized light

makes the actual reflectance to decrease a certain proportion.



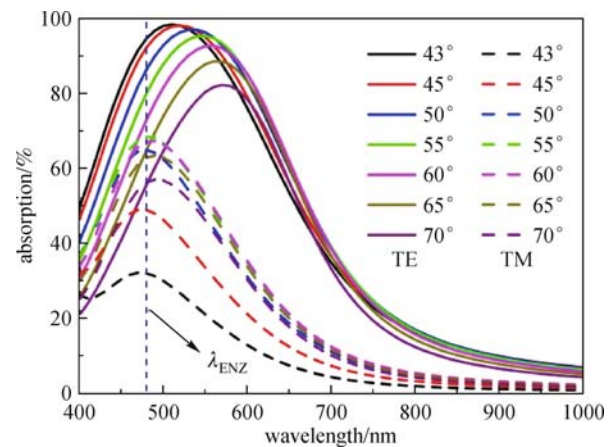
**Fig. 6** Absorption as a function of incident angles of Ag-II simulated by the transfer matrix method. (a) TE; (b) TM

incident angle of  $45^\circ$ , Ag-II shows the maximum absorption of 98.1% at 520 nm. Ag-I shows the maximum of 89.9% at 500 nm. The red dash lines represent the calculated results using Mathcad software. The red curves show a good agreement with measurement data, since there are a small amount of light losses scatted by small size of island distributed surface, the transfer matrix theory still can explain the optical performance of ultra-thin silvers films under ATR mode.

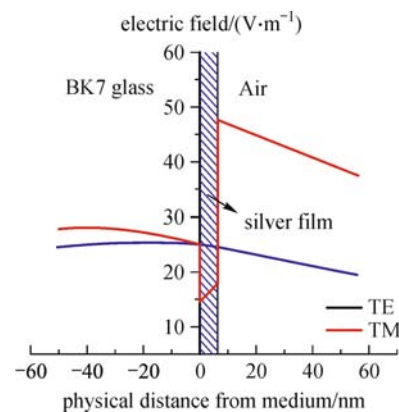
Since the experimental results can be explained by transfer matrix theory, we further theoretical analyzed the absorptions change with incident angles. Absorption of TE and TM waves are calculated at single wavelength 520 nm, as shown in Figs. 6(a) and 6(b). The symbol  $R$ ,  $T$  and  $A$  represent the reflectance, transmittance and absorption, respectively. As angle increases from  $36^\circ$  to  $70^\circ$  with interval  $0.5^\circ$ , transmittance ( $T$ ) becomes zero as incident angle  $\theta > \theta_c$ , and absorption of TE wave reduces after it reaches the maximum value around the critical angle  $\theta_c \sim 41.1^\circ$  (dash line). While absorption of TM wave increases after it reaches the minimum value  $\sim 7.5\%$ , and then it goes to a flat region  $\sim 62.0\%$ .

Figure 7 shows the simulated curves of absorption for Ag-II under TE (solid lines) and TM (dash lines) waves using different incident angles, and the angles are  $43^\circ$ ,  $45^\circ$ ,  $50^\circ$ ,  $55^\circ$ ,  $60^\circ$ ,  $65^\circ$  and  $70^\circ$ , respectively. The absorption peaks of TM wave are around its ENZ wavelength 480 nm, which similar with the “ENZ mode” described in previous works [17]. However, the absorption peak of TE wave shifts to longer wavelength gradually, and all the maximum absorption are larger than ENZ wavelength.

Figure 8 shows the total electric field of Ag-II which are calculated by Essential Macleod software. Wavelength 520 nm is chosen as the calculated parameter, for it represents the wavelength of absorption peak for  $45^\circ$  angle of incidence. TE wave excites high electric field of 25.0 V/m in the film. While the electric field excited by TM wave drops to 17.6 V/m inside the film, and then it increases to 47.6 V/m at silver/air interfaces. Since the



**Fig. 7** Absorption as a function of wavelength of Ag-II simulated by transfer matrix theory



**Fig. 8** Electric field distribution of TE and TM wave for Ag-II ( $\theta = 45^\circ$ , 520 nm)

surface plasmon resonance at BK7/air interface is excited only for the polarization direction parallel to the plane of incidence wave (TM wave), while we notice there is also a high electric field inside the film excited by TE wave.

## 4 Conclusions

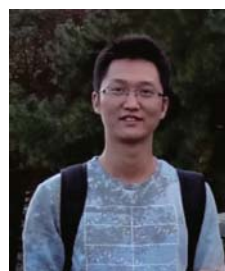
In this study, we investigate the optical performance of ultra-thin silver films under ATR mode, especially for light absorption, ultra-thin silver films show strong absorption under certain wavelength. Although ultra-thin silver films show islands distributed surface, the transfer matrix theory can explain the experimental results using optical constant which simulated by Drude-Lorentz oscillator. The two different thickness films show similar optical properties, thicker film ( $d \sim 6.3$  nm) obtains higher absorption. We suggest that the reason is the incident light excites high electric fields under ATR mode, which leads to the strong absorption. For TM wave, the high absorption need larger angle ( $\sim 60^\circ$ ), and the absorption curves of different incident angles appear similar with ENZ mode, and the absorption peaks are all around its ENZ wavelength. However, for TE wave, the highest absorption is around the critical angle ( $\sim 41.1^\circ$ ), and absorption peak decreases along with the increasing of incident angle.

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**Ming Zhou** obtained the Ph.D. degree from Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, and then works in Shanghai Institute of Technical Physics, Chinese Academy of Sciences. Dr. Zhou specializes field including design and preparation of optical thin film, metal thin films and infrared optical materials.



**Sheng Zhou** obtained the Master degree from University of Shanghai for Science and Technology, and then works in Shanghai Institute of Technical Physics, Chinese Academy of Sciences. His research interests including preparation of multi-channel narrowband filters and D/M/D multilayers.



**Gang Chen** works in Shanghai Institute of Technical Physics, Chinese Academy of Sciences. Now he is studying for Doctor's degree in Shanghai Institute of Technical Physics, Chinese Academy of Sciences. His research fields mainly engaged in preparation of narrowband filters, infrared filters, and beam splitters.



**Yaopeng Li** works in Shanghai Institute of Technical Physics, Chinese Academy of Sciences. Now he is studying for Master's degree in School of Information Science and Technology, Fudan University. He mainly engaged in preparation of all kinds of large size and high precision metal films.



**Prof. Dingquan Liu** is the director of the Department of Optical Coatings and Materials, Shanghai Institute of Technical Physics, Chinese Academy of Sciences. His mainly research field is thin film optics and technology, especially in infrared optical thin film and their applications. Now his research interests focus on: 1) nano bandwidth optical filters; 2) micro thin film optics; 3) polarization modulation based on thin films.