# Mixed polarization in determining the film thickness of a silicon sphere by spectroscopic ellipsometry*

Zhang Ji-Tao(张继涛)†, Wu Xue-Jian(吴学健), and Li Yan(李 岩)

*State Key Laboratory of Precision Measurement Technology & Instruments, Department of Precision Instruments and Mechanology, Tsinghua University, Beijing* 100084, *China*

(Received 15 June 2011; revised manuscript received 7 July 2011)

The effect of a spherical shape on the measurement result of spectroscopic ellipsometry (SE) is analyzed, and a method to eliminate this effect is proposed. Based on the simulation result of the SE measurement on a silicon sphere by ray tracking, we find that the sphere makes the parallel incident beam of the SE be divergent after reflection, and the measurement error of the SE caused by this phenomenon is explained by the mixed polarization theory. By settling an aperture in front of the detector of the SE, we can almost eliminate the error. For the silicon sphere with a diameter of 94 mm used in the Avogadro project, the thickness error of the oxide layer caused by the spherical shape can be reduced from 0.73 nm to 0.04 nm by using the proposed method. The principle of the method and the results of the experimental verification are presented.

**Keywords:** spectroscopic ellipsometry, silicon sphere, Avogadro constant, metrology

**PACS:** 07.60.Fs, 06.20.Jr, 06.20.–f **DOI:** 10.1088/1674-1056/21/1/010701

## 1. Introduction

The kilogram is the only base unit still defined by an artificial material, which is unstable and can be damaged or destroyed. Scientists all over the world have been devoted to the redefinition of the kilogram for nearly thirty years. One route to redefine the kilogram is by linking the unit of mass to a fundamental physical constant—the Avogadro constant ($N_{\rm A}$).[1−3] For this purpose, $N_{\rm A}$ should be determined with a relative uncertainty better than $2\times10^{-8}$. The X-ray crystal density (XRCD) method is considered to be one of the most accurate approaches to measure $N_{\rm A}$.[4,5] According to the XRCD method, the diameter of a ∼94 mm single crystal silicon sphere should be measured with an uncertainty better than 0.3 nm by using interferometers. However, when the single crystal silicon is exposed in air, an oxide layer grows on the surface because of natural oxidization. This layer causes an additional phase shift in the optical interferometer, so the measured diameter of the silicon sphere will contain an additional systematic error. For instance, an oxide layer of 1 nm thickness will result in an error of nearly 1.9 nm in the diameter,[6] which is too large to be accepted by the Avogadro project. To determine $N_{\rm A}$ with a relative uncertainty of $2\times10^{-8}$, the measurement uncertainty of the thickness of the oxide layer should be no more than 0.1 nm.

The layer has been measured by using many methods,[4,7−9] among which spectroscopic ellipsometry (SE) is frequently used. SE is indeed a powerful technique to determine the thickness of film.[10−12] However, except for some specific ellipsometries,[13] the basic principle of SE is based on the assumption that the shape of the film is a plane, which is in conflict with the fact that the silicon sphere has a spherical shape. To our knowledge, the effect of the shape of the silicon sphere on the measurement of SE has not yet been studied carefully so far.

In this paper, we evaluate the effect of the spherical shape on the SE result by using the ellipsometric theory for the first time. It is shown that the spherical shape will cause an effect of mixed polarization on the result of the SE, and the corresponding error of the measurement may be as large as 1 nm. A method named aperture-filtering method is proposed to eliminate this effect, and its validity has been confirmed by experiment.

*Project supported by the National Key Technology Research and Development Program of the Ministry of Science and Technology of China (Grant No. 2006BAF06B06) and the Tsinghua University Initiative Scientific Research Program, China (Grant No. 2009THZ06057).

†Corresponding author. E-mail: zjt@mail.tsinghua.edu.cn

© 2012 Chinese Physical Society and IOP Publishing Ltd

# 2. Effect of spherical shape on the SE

## 2.1. Divergent beam after reflection

Figure 1 shows the different behaviours of reflection on a plane sample (e.g., a silicon wafer) and a spherical sample (e.g., a silicon sphere) for the same incident parallel beam.

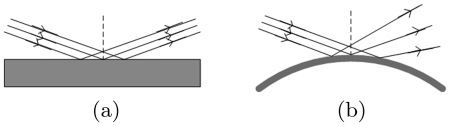



**Fig. 1.** Reflection of parallel rays on (a) a plane and (b) spherical samples.

It is clearly indicated in Fig. 1(b) that the outgoing rays are divergent after being reflected by a spherical sample. For a silicon sphere used in the determination of $N_{\rm A}$, we simulate the behaviour of the SE beam on the surface of the sphere by using the ray tracking method. Setting the diameter of the parallel incident beam of SE to 1 mm, the incident angle to 70° (this angle makes the ellipsometer have the highest sensitivity for silicon samples), and the diameter of the silicon sphere to 93.6 mm. Let the central ray of the incident beam hit the top point of the silicon sphere, and the receiver is set along the optical axis of the reflected rays. The simulation result is shown in Fig. 2. It shows that the shape of the SE incident beam is diverged to an ellipse after being reflected by the silicon sphere, and the size of the ellipse is enlarged as the receiver is moved far away from the top point of the sphere.

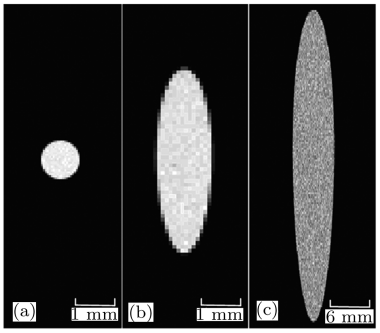



**Fig. 2.** Reflection of the incident parallel beam on the silicon sphere: (a) incident beam with a diameter of 1 mm; (b) reflected beam received at a distance of 30 mm from the top point of the sphere with a long axis of 5 mm and a short axis of 1.8 mm; (c) reflected beam received at a distance of 376.1 mm from the top point of the sphere with a long axis of 48.2 mm and a short axis of 6.7 mm.

For the common SE technique, it is assumed that the shape of the sample is a plane and homogeneous, which ensures that every ray of the SE beam has the same incident angle. However, things are different for the silicon sphere. In other words, only the central ray has a given incident angle, others will have offsets. Since the incident angle is an input parameter of the SE, the changes of the polarization state caused by the same sample are varied with the incident angles. Therefore, if the reflected beam from the silicon sphere is totally received by the detector of the SE, the mixture of the polarization state may cause additional systematic error in the SE result.

## 2.2. Mixed polarization

The basic principle of SE can be described by[10]

$$\rho = R_{\rm p}/R_{\rm s} = \tan\psi \exp({\rm i}\Delta), \tag{1}$$

where $\rho$ is the ratio of the Fresnel reflection coefficients $R_{\rm p}$ and $R_{\rm s}$. Since $\rho$ usually has a complex value, parameters $\psi$ and $\Delta$ vary for representing the magnitude and the phase shift, respectively, which are two parameters measured by the SE. The methods for determining $(\psi, \Delta)$ vary with the types of the SE. We here use a type of rotating-analyser ellipsometry (RAE),[14] whose setup is shown in Fig. 3.

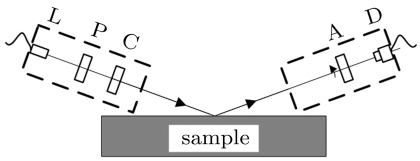



**Fig. 3.** Principle of the rotating-analyser type spectroscopic ellipsometer, where L denotes light source, P denotes polarizer, C denotes compensator, A denotes rotating analyser, and D denotes detector.

The principle of RAE can be described by the Jones matrix as

$$\begin{bmatrix} E_{\rm A} \\ 0 \end{bmatrix} = \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} \begin{bmatrix} \cos A & \sin A \\ -\sin A & \cos A \end{bmatrix} \begin{bmatrix} R_{\rm p} & 0 \\ 0 & R_{\rm s} \end{bmatrix} \times \begin{bmatrix} 1 & 0 \\ 0 & \exp(-{\rm i}\delta) \end{bmatrix} \begin{bmatrix} \cos P \\ \sin P \end{bmatrix}, \tag{2}$$

where $E_{\rm A}$ is the amplitude of the electric field after analyser A, $A$ and $P$ are the azimuth angles of the analyser and the polarizer, respectively. Usually, the azimuth angle of the polarizer is set to $P = 45°$, and the phase delay of C is set to $\delta = \pi/2$. According to

Eqs. (1) and (2), the intensity received by the detector can be deduced as

$$I = |E_{\rm A}|^2 = I_0 \left(1 - S_1 \cos 2A + S_2 \sin 2A\right), \quad (3)$$

where $S_1 = \cos 2\psi$, $S_2 = \sin 2\psi \cos \Delta$, and $I_0$ is a constant coefficient. In RAE, by rotating analyser A to different azimuth angles, the values of $S_1$ and $S_2$, represented by $(S_{1\rm m}, S_{2\rm m})$, can be obtained, so as the values of $(\psi, \Delta)$.

According to the simulation result in Subsection 2.1, the incident angle is varied with the position of the region illuminated by the SE beam on the silicon sphere. Therefore, the measured values of $S_1$ and $S_2$ should be described as functions of the spherical coordinates, $S'_{1\rm m}(\theta, \phi)$ and $S'_{2\rm m}(\theta, \phi)$, where $\theta$ and $\phi$ are the inclination and the azimuth in the spherical coordinate system, respectively. The measurement result of the SE is a combination of $S'_{1\rm m}(\theta, \phi)$ and $S'_{2\rm m}(\theta, \phi)$ from the whole illuminated region. To avoid confusion with the measurement of the plane sample, the measurement value on the silicon sphere is written as $(\bar{S}_{1\rm m}, \bar{S}_{2\rm m})$.

Divide the illuminated region of the silicon sphere into $N$ sub-regions equally, and assume that the rays in each sub-region have the same incident angle. Accordingly, the intensity received by the SE from each sub-region can be described as

$$\begin{aligned} I(\phi_i, \theta_i) = {} & \frac{I_0}{N}(1 - S'_{1\rm m}(\phi_i, \theta_i) \cos 2A \\ & + S'_{2m}(\phi_i, \theta_i) \sin 2A), \end{aligned} \quad (4)$$

where $i$ $(= 1, 2, \ldots, N)$ is the order of the sub-region. The total intensity received by the SE is

$$\begin{aligned} I &= \sum_{i=1}^{N} I(\phi_i, \theta_i) \\ &= I_0 \left(1 - \bar{S}_{1\rm m} \cos 2A + \bar{S}_{2\rm m} \sin 2A\right), \quad (5) \\ \bar{S}_{1\rm m} &= \frac{1}{N} \sum_{i=1}^{N} S'_{1\rm m}(\phi_i, \theta_i), \bar{S}_{2\rm m} \\ &= \frac{1}{N} \sum_{i=1}^{N} S'_{2\rm m}(\phi_i, \theta_i). \quad (6) \end{aligned}$$

It is shown in Eqs. (5) and (6) that the signal received by SE is a mixture of different polarization states. The phenomenon above is named as the mixed polarization in the SE measurement in this paper.

To evaluate the effect of the mixed polarization on the SE measurement, we first simulate the changes of the measured $(\psi, \Delta)$ along with that of the incident angle of SE, and then obtain the relationship between $\Delta$ and the thickness of the oxide layer. Assume that the sample is a 5 nm thickness silicon dioxide ($SiO_2$) film on a substrate of single crystal silicon (c-Si). The refractive indexes of the film and the substrate are supposed to be $n_{\rm SiO_2} = 1.462$ and $n_{\rm S_i} = 3.88 - 0.019{\rm i}$ at 633 nm, respectively, and the initial incident angle of SE is set to 70°. Let the incident angle be varied within the range of ±2° around 70°, we then calculate the corresponding changes of the elliptical angles $(\psi, \Delta)$ by Eq. (1), and the result is shown in Fig. 4. Furthermore, we calculate the relationship between offsets $(\delta_\psi, \delta_\Delta)$ of the initial elliptical angles $(\psi_0, \Delta_0)$ and divergent incident angle $\delta_{\rm i}$ of the SE incident beam at 70° by Eqs. (5) and (6), and the results are shown in Fig. 5. Finally, the relationship between the film thickness and elliptical angle $\Delta$ is presented in Fig. 6.

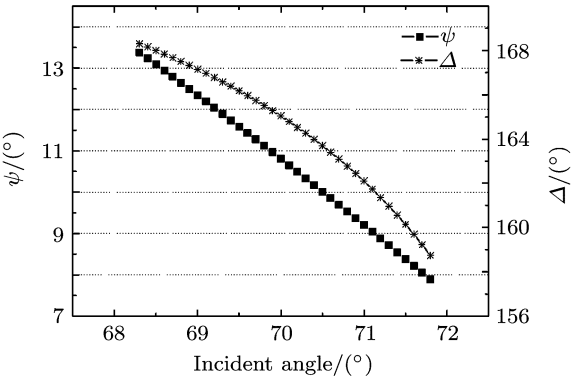



**Fig. 4.** Relationship between incident angle and elliptical angles $(\psi, \Delta)$ of the SE.

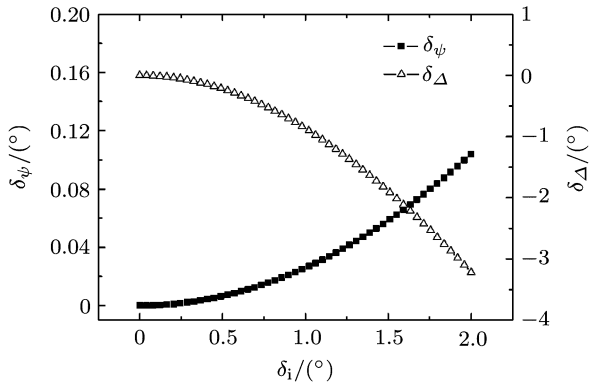



**Fig. 5.** Simulation results of the mixed polarization on the basis of the physical principle presented in Eqs. (5) and (6).

From Figs. 4 and 5 we note that the measured thickness of the ultra-thin film (e.g., less than 10 nm)

is sensitive to the change of $\Delta$ but not $\psi$. Consequently, we can usually determine the film thickness based on the measured value of $\Delta$. In addition, figure 6 shows that $\Delta$ is almost varying linearly with the film thickness. Therefore, we use a straight line to fit the discrete data shown in Fig. 6 by using the least square method. The fitted result is

$$\Delta = -2.74t + 178.95, \tag{7}$$

where $\Delta = \Delta_0 + \delta_\Delta$, the coefficient of correlation equals 0.99979, and the standard deviations of the fitted slope and the fitted intercept is 0.08 and 0.01, respectively.

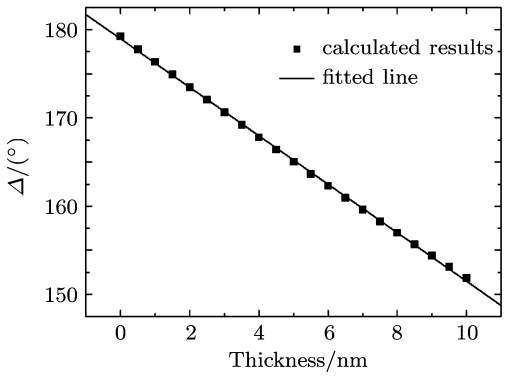



**Fig. 6.** Relationship between film thickness and elliptical angle $\Delta$.

Suppose the diameter of the incident beam (with the nominal incident angle of 70°) on the silicon sphere is 1 mm, it can be deduced from Subsection 2.1 that the incident angle is within the range of $[70° - 1.71°, 70° + 1.87°]$. According to Eqs. (5)–(7) and Figs. 4–6, the systematic error of the film thickness introduced by the mixed polarization is about 1 nm, which is too large to be accepted by the Avogadro project. We then develop an aperture-filtering method to reduce the effect of the mixed polarization.

### 2.3. Aperture-filtering method for eliminating the mixed polarization

The principle of the method for eliminating the effect of the mixed polarization is shown in Fig. 7. An aperture S with adjustable size is settled in the optical path of the reflected light. It acts like a filter: only the reflected rays around the top point of the sphere are allowed to pass and be detected, and the others are blacked. According to Subsections 2.1 and 2.2, the effect of the mixed polarization can be almost eliminated by means of choosing an appropriate position and size for the aperture. For instance, we set the distance between the aperture and the top point of the silicon sphere to 376.1 mm (just in front of the SE detector) and set the diameter of S to 4 mm. In this case, it can be calculated that the variation of the incident angles of the rays received by the detector is within the range of ±0.15°, and the corresponding value of $\delta_\Delta$ is −0.024°. According to Eq. (7), the systematic error of the film thickness caused by the mixed polarization is decreased to 0.01 nm, which is so small that it can be ignored.

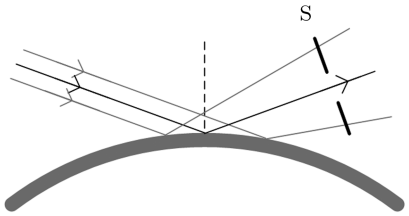



**Fig. 7.** Aperture-filtering method for eliminating the mixed polarization caused by the spherical shape of the silicon sphere.

The proposed method is inspired by the common apertures attached in most commercial SE. It should be noted that, however, according to the theoretical analysis and the simulation in Subsection 2.2, the position and the size of the aperture should be determined specifically. We verify this method by experiments in Section 3.

## 3. Experiments and results

The experimental setup is shown in Fig. 8. SE with a spectral range of 190–400 nm and an incident angle of 70° was used for the measurement, and the light source was a deuterium lamp. The incident arm (1) and receiving arm (2) were controlled by a precise goniometer, which had an angular accuracy better than 0.05°. An automatic collimating telescope (ACT) was applied to monitor the position of the sample. The process to adjust the position of the silicon sphere was as follows. First, set the incident and the receiving arms to a given angle (70° in this paper) by adjusting the goniometer. Second, by using a standard sample (e.g., a silicon wafer), we adjust the focus point of the ACT to the intersection of the optical axis between the incident and the receiving arms by monitoring detectors $D_1$ and $D_2$. At last, by monitoring the intensity of $D_1$ and the image of cross plate C in $D_2$, we adjust the top point of the silicon sphere

to the appropriate position. The intensity of $D_1$ is largest and the image of cross plate C is clearest at this position. With the above adjustments, we can make sure that the central light of the incident arm illuminates the top point of the sphere at the given incident angle accurately. Moreover, an aperture $S_3$ with adjustable diameter was settled in front of detector D, whose distance from the top point of the sphere was about 376 mm. The silicon sphere for the experiment was made of single crystal silicon with a purity better than 99.9999%, and its diameter was nearly 93.6 mm with a sphericity better than 150 nm.

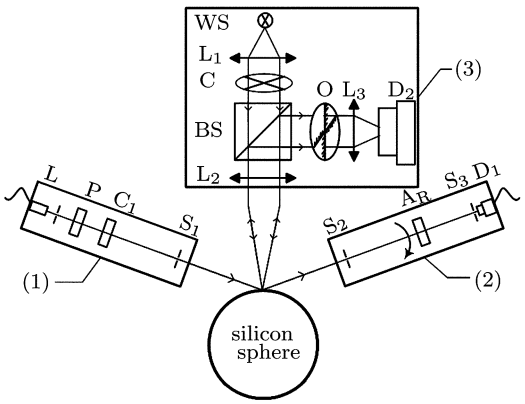



**Fig. 8.** Experimental set up for measuring oxide layer of silicon sphere by spectroscopic ellipsometer. (1) Incident arm includes light source (L), polarizer (P), compensator ($C_1$), and aperture ($S_1$). (2) Receiving arm includes apertures ($S_2$) and ($S_3$), rotating analyzer ($A_R$), and detector ($D_1$). (3) Automatic collimating system includes: white light source (WS), convergent lens ($L_1$), ($L_2$) and ($L_3$), cross plate (C), beam splitter (BS), scale plate (O), and CCD camera ($D_2$).

Two groups (A and B) of experiments were carried out on the surface of the silicon sphere by using SE with different sizes of aperture $S_3$. For group A, the diameter of $S_3$ was set to 10 mm (usually the effective apertures of the optical elements in the SE were larger than 10 mm), and for group B, the diameter of $S_3$ was set to 4 mm. The measurement result of each group was averaged over ten-time repetitions, which made sure that the random error of the SE was eliminated. The experimental standard deviations of the elliptical angles in the two groups were both less than 0.1°, as shown in Fig. 9.

Figure 10 shows the difference of the measured elliptical angles of groups A and B at the same point on the silicon sphere. It indicates that the mixed polarization caused by the spherical shape will cause an error as large as 2° into the result of the elliptical angle $\Delta$ within a broad spectral range. After converting to the film thickness by using Eq. (7), the corresponding systematic error of the thickness is nearly 0.73 nm. This is consistent with the simulation result in Subsection 2.2. The reason for the discrepancy between the experiment and the simulation results is that the effective apertures of the optical elements in the SE are usually less than the size of the ellipse shown in Fig. 2(c), which means that the real level of the mixed polarization will be slightly less than that in the simulation.

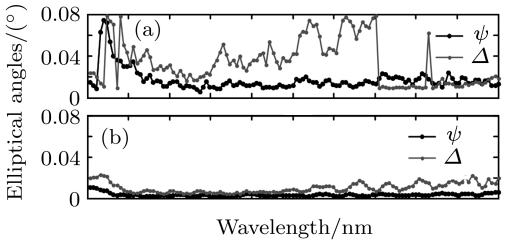



**Fig. 9.** Standard deviation of the elliptical angles of experiments (a) A and (b) B.

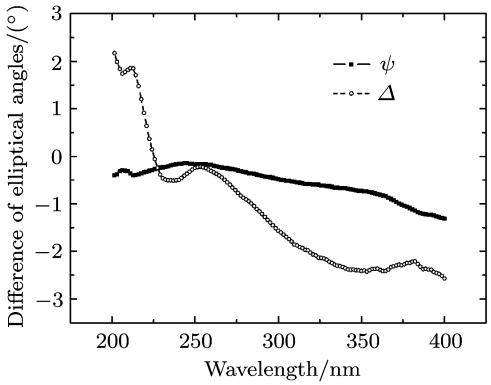



**Fig. 10.** Comparison results of the two groups of experiments, the curves show the difference of the elliptical angles ($\psi, \Delta$) between the two groups of experiments.

A potential shortcoming of this method is that the signal-noise ratio may be reduced, since fewer rays are detected. However, it has been shown in Fig. 8 that the standard deviation of $\Delta$ is less than 0.1°, which is approximately equivalent to 0.04 nm in the film thickness. This error is acceptable when considering the uncertainty target of 0.1 nm in the thickness of the oxide layer.

# 4. Conclusion

SE is a powerful tool for the determination of the film thickness. However, for some specific applications, measurement accuracy should be evaluated carefully. We have discussed the influence of the

spherical shape on the measurement of SE on the basis of determining the thickness of the oxide layer on a silicon sphere, and proposed a simple method to eliminate the effect of the mixed polarization caused by the spherical shape.

The error of a measurement usually consists of two parts, systematic error and random error. The effect of the spherical shape causes a systematic error in SE. Without considering it, the result of SE may be still precise but not accurate. The work presented in this paper has found that this effect cannot be ignored when considering a target uncertainty of 0.1 nm in measuring the thickness of the oxide layer in the Avogadro project.

The advantage of the proposed method is that it only selects the central part of the reflected rays for transmission, so that the influence of the polarization state of the rays is reduced. The method also has its limitation. Its working principle requires that the SE light source has enough intensity and the detector has a high sensitivity. An extension of the present study to improve the sensitivity of the detector is under way.

# References


[1] Davis R 2003 *Metrologia* **40** 299

[2] Mills I M, Mohr P J and Quinn T J 2005 *Metrologia* **42** 71

[3] Becker P 2003 *Metrologia* **40** 366

[4] Becker P 2001 *Rep. Prog. Phys.* **64** 1945

[5] Andreas B, Azuma Y, Bartl G, Becker P, Bettin H, Borys M, Busch I, Gray M, Fuchs P, Fujii K, Fujimoto H, Kessler E, Krumrey M, Kuetgens U, Kuramoto N, Mana G, Manson P, Massa E, Mizushima S, Nicolaus A, Picard A, Pramann A, Rienitz O, Schiel D, Valkiers S and Waseda A 2011 *Phys. Rev. Lett.* **106** 030801

[6] Fujii K, Tanaka M, Nezu Y, Nakayama K, Fujimoto H, de Bièvre P and Valkiers S 1999 *Metrologia* **36** 455

[7] Zhang J, Li Y, Wu X, Luo Z and Wei H 2010 *Opt. Express* **18** 7331

[8] Busch I, Danzebrink H U, Krumrey M, Borys M and Bettin H 2009 *IEEE Trans. Instrum. Meas.* **58** 891

[9] Kuramoto N, Fujii K, Azuma Y, Mizushima S and Toyoshima Y 2007 *IEEE Trans. Instrum. Meas.* **56** 476

[10] Azzam R M A and Bashara N M 1987 *Ellipsometry and Polarized Light* (Amsterdam: North-Holland)

[11] Fujiwara H 2007 *Spectroscopic Ellipsometry: Principles and Applications* (England: Wiley)

[12] Zhang J, Li Y and Luo Z 2010 *Acta Phys. Sin.* **59** 186 (in Chinese)

[13] Han C Y, Lee Z Y and Chao Y F 2009 *Appl. Opt.* **48** 3139

[14] Aspnes D E and Studna A A 1975 *Appl. Opt.* **14** 220