Spectroscopic imaging ellipsometry for thin film detection on uniaxial crystal

Lichao Guan[1], Jiexiong Ding[1], Li Du[1], Achyut Adhikari[2], Anand Krishna Asundi[2]

1 School of Mechatronics Engineering, University of Electronic Science and Technology of China, Chengdu, China, 611731

2 Centre for Optical and Laser Engineering, School of Mechanical and Aerospace Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore, 639798

Corresponding Author / E-mail:lichaoguan@126.com, TEL: +86-028-61831052, FAX: +86-028-61831052

ABSTRACT:

Spectroscopic imaging ellipsometry (SIE) is a powerful technique devoted to the study of optical properties and thickness of thin films by measuring the change in polarization state of light reflected from the surface. SIE measures two quantities (ψ and Δ), which represent the amplitude ratio and phase angle of deflection of the p-polarized light and s-polarized light reflected from the sample surface. The SIE measurement of thin film is difficult when the substrate is uniaxial anisotropic crystal for two reasons - firstly the p-polarized and s-polarized components of reflective light are coupled which makes the data processing more complex and secondly an optical model is needed for SIE data processing with the substrate optical constants as known parameters in the model, but the substrate optical constants are not unique when the plane of the optical axis changes. Hence in the measurement of thin film on an anisotropic material using generalized ellipsometry, significant errors due to the complex calculation arise. The best approach is to measure the uniaxial substrate as an isotropic material by adjusting the optical axis in the incident plane. In this paper, the crystal optical axis is determined by rotating the sample using the SIE setup and the incident light is adjusted in the optical axis plane to eliminate the effects of uniaxial substrate. A uniaxial KDP (Potassium Dihydrogen Phosphate) crystal with thin oil film and a bare KDP substrate are prepared. A scheme to determine KDP crystal optical axis is proposed. Finally, the optical constants of the KDP substrate are determined, and the oil film thickness on KDP crystal is measured when the incident light is in crystal optical axis plane.

Key words: Spectroscopic imaging ellipsometry, uniaxial crystal, optical axis, thin film, KDP crystal

1. INTRODUCTION

Thin film detection is wildly used in mechanical manufacturing, integrated optics, solar cells and semiconductor industry, and there are liquid film, solid film and so on[1,2]. The thin film is covered on the materials surface for various purposes, such as protecting substrate from damaging, optical modulation and electrical modulation, there also has thin film need to clean up like the residual oil film on optical components after polishing[3]. So the measurement of thin film thickness is needed to control thin film volume or evaluate the surface quantity of optical components after polishing. There are various methods to measure the thickness or distribution of thin film, such as eddy current method, optical interferential method[4], X-ray method[5], AFM technology[6] and so on. However, there are many limits for these methods, such as the electroconductibility, reflection coefficient, and resolution. The spectroscopic ellipsometry (SE) technology[7] is a powerful tool to measure the optical constants and thickness of thin film, SE has the advantages of non-destructive, nano-layer resolution and less limitation of materials, it is wildly applied in the detection of graphene,

International Conference on Optical and Photonics Engineering (icOPEN 2017), edited by Anand Krishna Asundi,
Proc. of SPIE Vol. 10449, 1044921 · © 2017 SPIE · CCC code: 0277-786X/17/$18 doi: 10.1117/12.2270796

solar cells, protein interaction and various thin films. Spectroscopic imaging ellipsomtry(SIE)[8-9] is developed based on the conventional SE to improve the lateral resolution, which has a CCD camera after the analyzer. SIE technology combines the advantages of SE and the high lateral resolution of CCD camera which make its lateral resolution be less than $1\mu m$ and the vertical resolution less than $0.1nm$. The measurement results of SIE are fitted from the measured data, but the data processing is more complicated when the thin film is on uniaxial crystal due to the birefringence phenomenon.

In this paper, the optical axis plane of uniaxial crystal is noticed, the SIE data processing of uniaxial is the same as isotropic materials when the incident plane is parallel to the optical axis plane. The KDP crystal after magnetorheological (MRF) polishing is prepared as ambient-anisotropic thin film-uniaxial substrate sample as there is residual oil on its surface after polishing and surface cleaning, and SIE technology is applied to measure the thickness distribution of residual oil film. The optical axis plane of KDP crystal is determined by rotating the sample and the probe light of SIE is adjusted to be in optical axis plane, then the optical constants of KDP substrate and the thickness distribution of residual oil film on KDP crystal are measured.

2. PRINCIPLES AND ANALYSIS METHODS

2.1 Principles and analysis of uniaxial crystal measurement by SIE

SIE measurement is based on the ellipsometric changes when the incident elliptical polarized light is reflected on sample surface. The SIE measurement system has six parts as light source, polarizer, compensator, sample, analyzer and CCD camera, the incident light changes to elliptically light after polarizer and compensator, and the corresponding polarization states of reflected light from sample surface are detected by analyzer. The ratio ρ of the reflection coefficients is used to describe the polarization states change of reflected light, which can be described as $\rho = r_p / r_s = \tan(\psi)e^{i\Delta}$, where r_p and r_s refer to the reflection coefficients of the p and s-polarized light, psi (ψ) is amplitude ratio and delta (Δ) is the phase difference between two polarized light waves[10].

The Jones matrix of the corresponds to the light reflection on sample surface can be described as:

$$S = \begin{bmatrix} r_{pp} & r_{ps} \\ r_{sp} & r_{ss} \end{bmatrix} = r_{ss} \begin{bmatrix} r_{pp}/r_{ss} & r_{ps}/r_{ss} \\ r_{sp}/r_{ss} & 1 \end{bmatrix} = r_{ss} \begin{bmatrix} \rho_{pp} & \rho_{ps} \\ \rho_{sp} & 1 \end{bmatrix} \quad (1)$$

where $\rho_{pp} = r_{pp}/r_{ss} = \tan(\psi_{pp})e^{i\Delta_{pp}}$, $\rho_{ps} = r_{ps}/r_{ss} = \tan(\psi_{ps})e^{i\Delta_{ps}}$ and $\rho_{sp} = r_{sp}/r_{ss} = \tan(\psi_{sp})e^{i\Delta_{sp}}$.

When the sample is isotropic, the off-diagonal elements of the Jones matrix are zero, and the Jones matrix is defined by two parameters (ψ_{pp}, Δ_{pp}). However, the Jones matrix of anisotropic sample has six parameters $(\psi_{pp}, \psi_{ps}, \psi_{sp}, \Delta_{pp}, \Delta_{ps}, \Delta_{sp})$, which illustrates the p-polarized and s-polarized components of reflective light are coupled. Thus, the data processing for anisotropic sample is more complex than isotropic sample. For the uniaxial crystal, the light reflection on negative crystal surface is shown in Fig1, p is the interface between air and crystal, p1 is the optical axis plane and p2 is the incident plane. The off-diagonal elements of Jones matrix can become zero ($\psi_{ps} = \psi_{sp} = \Delta_{ps} = \Delta_{sp} = 0$) in some special orientations when the optical axis lies parallel to the incident plane ($\theta_4 = 0°$), which has been confirmed in reference [10].

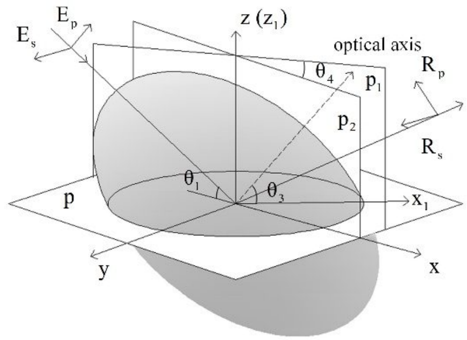


Fig1 Light reflection on a negative uniaxial crystal, θ_1 is the incident angle, θ_3 is the angle between optical axis and interface plane, θ_4 is the angle between optical axis plane and incident plane, E_p . E_s R_p R_s are p and s part of the incident light and reflected light.

For the isotropic thin film detection on uniaxial crystal, the optical parameters of substrate are needed in data processing, because the ψ and Δ are related to the optical properties of thin film and substrate, which can be described as $\tan(\psi)e^{i\Delta} = \rho(n_0, n_1, n_2, d_1, \phi_0, \lambda)$, n_0 , n_1 , n_2 , d_1 , ϕ_0 and λ are the refractive indexes of air, thin film and substrate, thin film thickness, incident angle and incident light wavelength. Here, if the incident plane is adjusted parallel to the optical axis during the measurement, the Jones matrix of uniaxial crystal is diagonal and the optical parameters of substrate would be unique when the incident angle is also fixed, then the detection of isotropic thin film on uniaxial crystal can be seen as on isotropic substrate.

2.2 Optical axis detection of uniaxial crystal by SIE

As shown in Fig2(a), the light travels on the interface of two isotropic material, and the refractive indexes are n_1 and n_2 . The incident light can be divided into s-polarized light and p-polarized light, where the electric components of incident light, reflective light and transmitted light can be described as E_p, E_s, R_p, R_s, D_p and D_s. The Fresnel formula of the reflective light are described by eq(2) and eq(3), where the θ_1 and θ_2 are the incident angle and refracted angle[11].

$$\frac{R_s}{E_s} = \frac{n_1 \cos\theta_1 - n_2 \cos\theta_2}{n_1 \cos\theta_1 + n_2 \cos\theta_2} = -\frac{\sin(\theta_1 - \theta_2)}{\sin(\theta_1 + \theta_2)} \tag{2}$$

$$\frac{R_p}{E_p} = \frac{n_2 \cos\theta_1 - n_1 \cos\theta_2}{n_2 \cos\theta_1 + n_1 \cos\theta_2} = -\frac{\tan(\theta_1 - \theta_2)}{\tan(\theta_1 + \theta_2)} \tag{3}$$

When the incident light is linearly polarized, the relationship between vibration azimuth angle(α_1) and electric components(E_p and E_s) of linearly polarized light is shown in Fig2 (b), where $E_s = E\sin\alpha_1$ and $E_p = E\cos\alpha_1$. The reflective light will also be linearly polarized which can be gain from eq(2) and (3), thus the vibration azimuth angle(α_2) can be written as eq(4).

$$\tan\alpha_2 = \frac{R_s}{R_p} = \frac{\cos(\theta_1 - \theta_2)}{\cos(\theta_1 + \theta_2)}\tan(-\alpha_1) \tag{4}$$

The θ_1 and θ_2 are different, so the vibration azimuth angle of reflective light is different from the incident light, but when the incident light is only s-polarized light ($\alpha_1 = 0°$) or p-polarized light ($\alpha_1 = 90°$), $\alpha_2 = 0°$ or $90°$, which illustrates that the reflective light is s-polarized light or p-polarized light.

When the s-polarized light is reflected on the uniaxial crystal, the reflected light would be elliptic polarized light instead of the s-polarized, because the crystal generates p- polarized component due to the birefringence phenomenon and the vibration azimuth angle of reflected light would change which is also confirmed in section 3.2 and 3.3. The situation is the same for p-polarized light. However, when the incident light orientation is along with the optical axis of uniaxial crystal, the reflection light would be the same with isotropic material. As shown in Fig1, if the incident angle (θ_1) is adjusted as θ_3 ($\theta_1 = \theta_3$), the s-polarized light and p-polarized light are reflected from crystal surface respectively, θ_4 can be changed by rotating the crystal using sample stage of SIE, then the vibration azimuth angle of reflected light can be detected by analyzer. The optical axis is in the incident plane only when the vibration azimuth angle of reflected s and p-polarized light are $0°$ and $90°$ at the same time, which can be used as the method to detect optical axis plane.

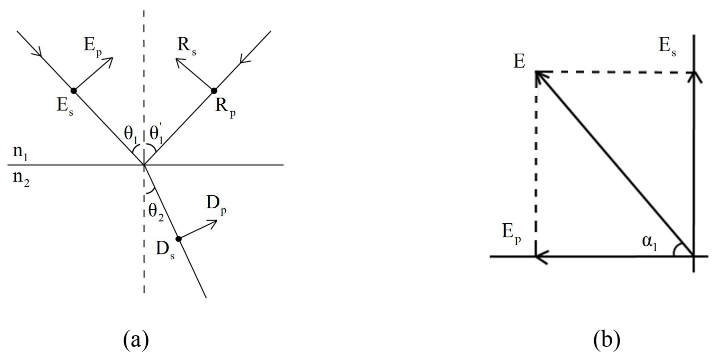


Fig2 (a)Light propagation in an isotropic material. (b)Vibration azimuth angle of linearly polarized light.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1 Introduction of KDP sample and SIE setup

Uniaxial KDP crystal is prepared for the thin film detection. KDP crystal is one of the important optical element in inertial confinement fusion(ICF) program[12] which is irreplaceable, and it is wildly used as Q-switches and light frequency converters due to its high laser damage threshold and high nonlinear efficiency. The common machine process of KDP crystal has three steps as single point diamond turning(SPDT)[13], magnetorheological finishing(MRF) and Mega sonic cleaning, the MRF polishing is used to finish the micro-scale ripples left after SPDT process and improve the crystal surface quality and the Mega sonic cleaning is applied to clean the polishing liquid adsorbed on the surface after polishing. However, the adsorbed polishing liquid is hardly to clean up and there will be nano-scale residual oil film on KDP surface which can be measured by SIE technology, and the quantitative characterization of residual oil film is urgent for evaluate the KDP surface quality and guide the cleaning process. One bare KDP substrate and one KDP sample with residual oil are prepared for SIE measurement, the bare substrate is only processed by SPDT and the sample is machined by three steps as SPDT, MRF and Mega sonic cleaning.

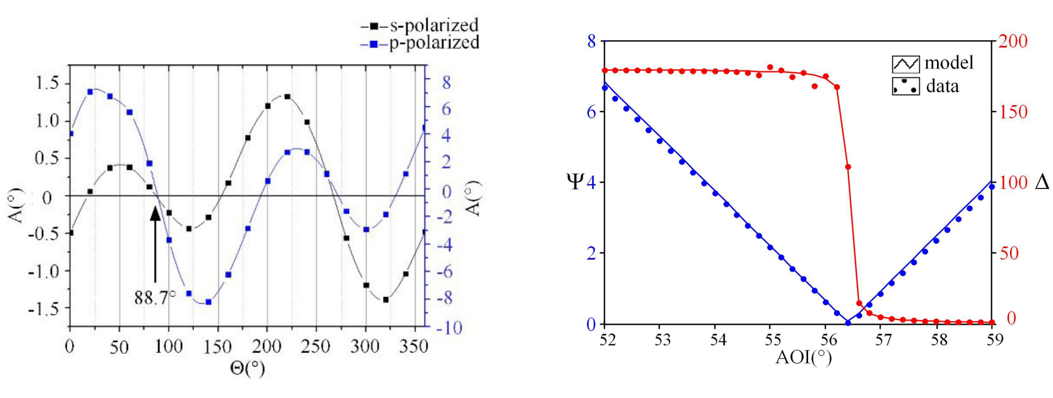
For the SIE detection of residual oil film on KDP crystal, the setup nanofilm_ep4 is applied with the incident light wavelength is set as 658nm. The traditional AOI (angle of incidence) mode and mapping mode are both used to determine the Ψ and Δ data by null ellipsometry. In the AOI mode, the ellipsometric parameters are recorded as the angle of incidence light changed, then the optical parameters can be fitted out with the best-fit optical model. The data are measured as the incident light and incident angle are fixed in mapping mode, then the map of results can be gained. In the given SIE setup, the reflected light from the sample must be linearly polarized, only when the incident light is in certain elliptical polarizations can meet the requirement. The certain polarizations are found by rotating the polarizer when the angle of compensator is kept $45°$ which is a commonly used angle. There are four different polarizer angles can satisfy the null condition, so there are four solutions. The four-zone mode is applied in this measurement that the data is recorded by averaging these four solutions, this method can get better solutions while it will take more time than one-zone mode.

3.2 Optical parameters measurement of KDP substrate

The s-polarized light and p-polarized light are used to detect the direction optical axis plane of KDP crystal by rotating the sample around the Z-axis where the sample surface is in x-y plane. There is $58.5°$ angle between the x-y plane and the optical axis which is measured by the KDP crystal machining company, and the incident light angle of setup is set as $58.5°$, then the reflected light vibration azimuth angle of s and p-polarized incident light from KDP surface are recorded by analyzer when the light intensity after analyzer is minimum. As shown in Fig3(a), the sample is rotated around Z axis from 0 to $360°$. The x-axis ($\theta[°]$)is the angels of sample rotation and y-axis ($A[°]$)is the analyzer angles where the rotating step is $20°$, the zero line of s-polarized light is set as $0°$ while p-polarized light is set as $90°$. The spots and lines are sampling data and fitting curves respectively. The fitting lines of s and p-polarized light cross the zero line at $88.7°$ simultaneously, which means the incident light is in the optical axis plane when the sample is rotated at this angle. Fig3 (a) also shows that there is a deflection of vibration azimuth angle when the optical axis is not in the incident plane. Then the optical parameters of KDP substrate is measured when the sample stage is rotated to $88.7°$ as the crystal is kept stable to the sample stage during all the measurement.

The fitting results of KDP substrate by AIO mode is shown in Fig3(b), the incident angle is changed from $52°$ to $59°$ as the step is $0.2°$, and the diameter of light spot is 1mm. As KDP crystal is weakly absorbing and transparent media, the classical Cauchy model[14] is applied to describe the optical dispersion when the light go through sample surface. The solid lines are the simulation results according to the Cauchy model while the spots are the measured data, the root mean square error(RSME) between the measured data and simulate line is 2.3 which means the optical model is suitable for the measurement (the fitting result is accuracy when the RMSE is less than 5). The refractive index (n_{KDP}) and extinction coefficient (k_{KDP}) are 1.5062 and 0.0028 as calculated respectively.

In mapping mode, the size of detection area is $440 \times 570 \mu m^2$ while the magnification factor is 10. The maps of measured data Δ and Ψ are shown in Fig4 (a) and (b), then the n_{KDP} map and k_{KDP} map are calculated by using the Cauchy model. The dark spots are the pollutions and residual crystal particles which are involved in SPDT process. The mean values of n_{KDP} and k_{KDP} are 1.506 and 0.003 which agree well with the results in AIO mode, and the results of mapping mode are used in the calculation of residual oil film.



(a) (b)

Fig3 Measurement result of uniaxial KDP crystal. (a) Detection result of optical axis of KDP crystal. (b) Fitting curves of ellipsometric parameters in AOI spectra mode, the spots are the measured data and the solid lines are the best-fit theoretical simulation.

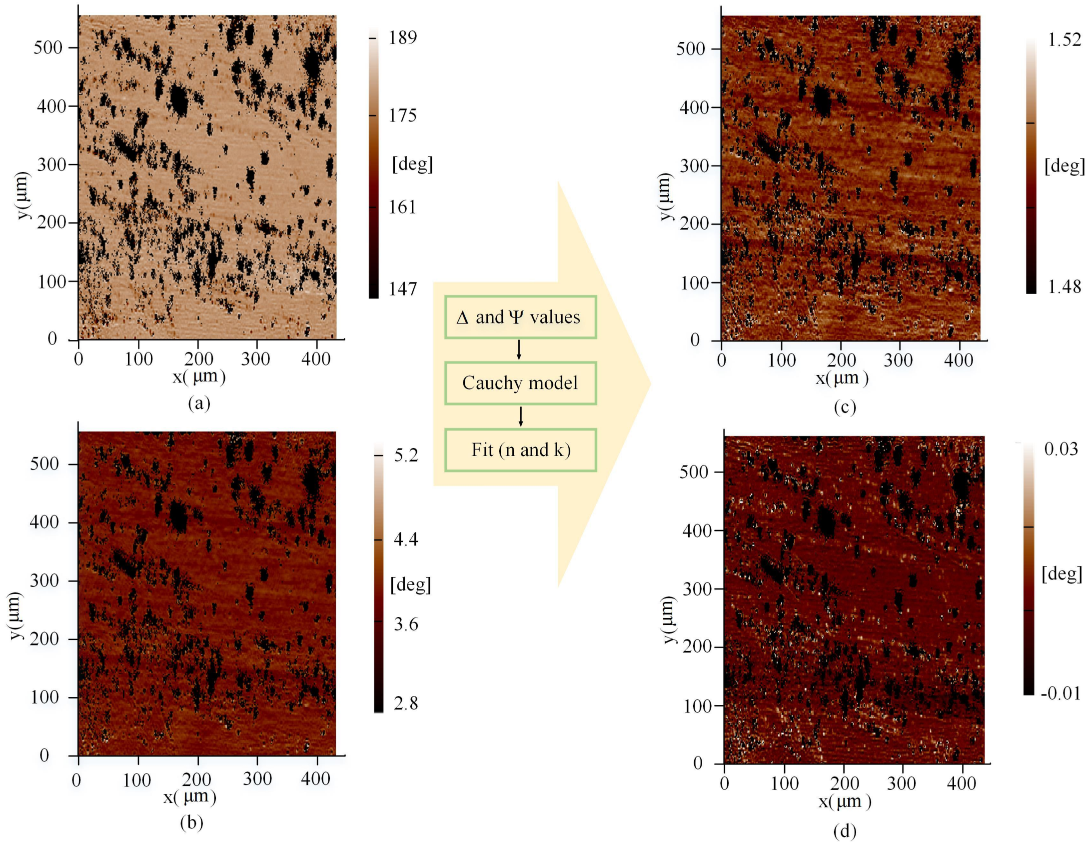


Fig4 Measurement result of optical parameters of KDP crystal by mapping mode. (a) Δ map. (b) Ψ map. (c) Refractive index (n_{KDP}) map of KDP substrate. (d) Extinction coefficient (k_{KDP}) map of KDP substrate.

3.3 Residual oil film measurement on KDP crystal

For the residual oil film measurement on KDP crystal, the optical axis of KDP is also detected in order to adjust the incident light in the optical axis plane where the optical parameters of KDP are the same as measured in section 3.2. The detection result of optical axis plane is shown in Fig5, the incident light is in optical axis plane when the sample rotation angle is 92.1°, so the measurement is done while the sample is rotated to 92.1°.

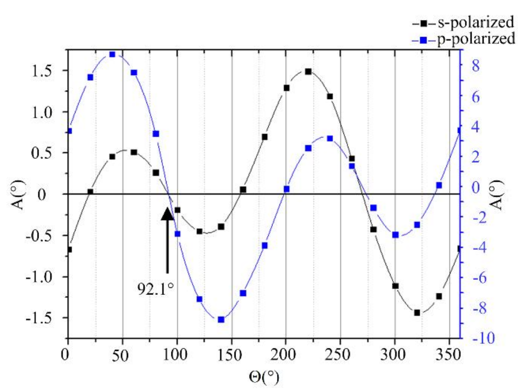


Fig5 Detection result of optical axis of KDP crystal with residual oil.

The mapping mode of SIE is applied for the residual oil detection. An air-oil film-KDP substrate optical model is built to calculate the thickness of oil film, the Cauchy model is used due to the transparent and weakly absorbing properties of KDP substrate and oil film, and Bruggeman EMA model is applied because the oil film surface is rough after mega sonic cleaning. The method for data processing is also shown in Fig 6, where the optical parameters of KDP substrate are substituted into the optical model system. The thickness map of residual oil in Fig 6(c) shows that the oil film thickness ranges from 0 to 15.6nm, and the mean value of oil thickness map is 7.77nm and distribution of residual oil is similar with Gaussian distribution which are shown in the statistic curve of oil film thickness.

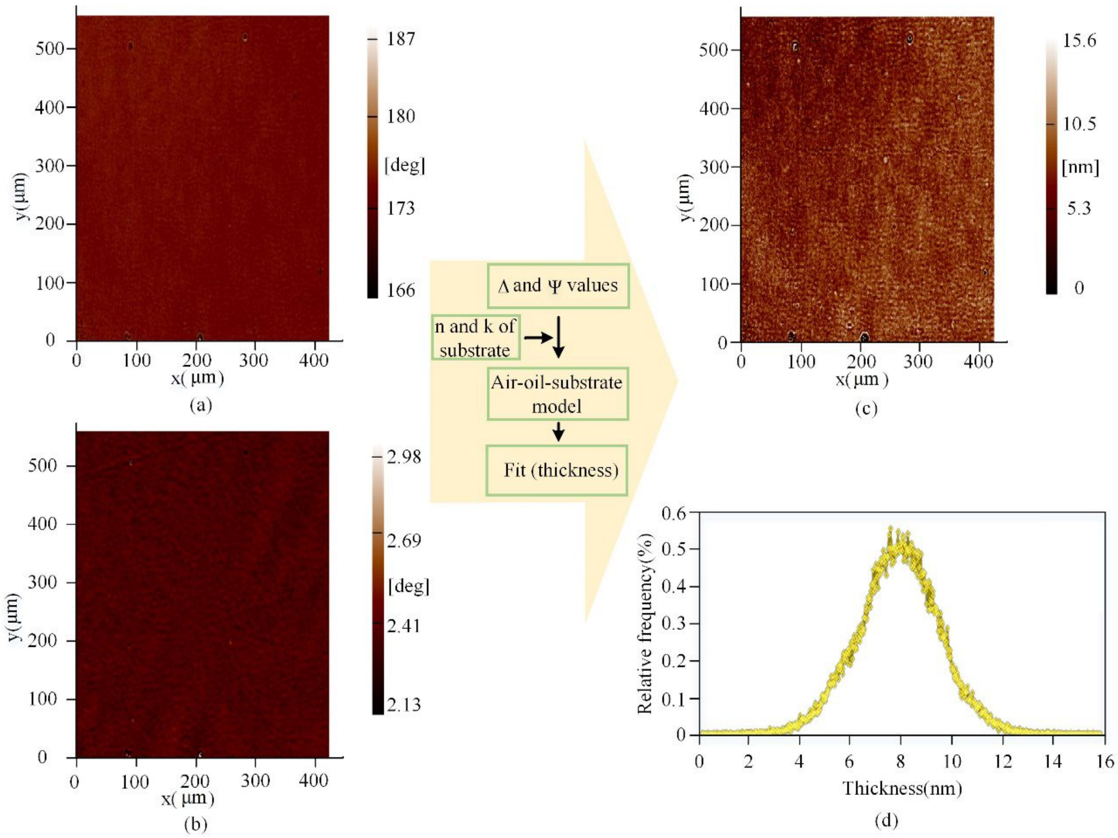


Fig6 The measurement result of residual oil film on KDP crystal by mapping mode. (a) Δ map. (b) Ψ map. (c) Thickness map of residual oil film. (d) Statistic curve of residual oil film thickness, the x-axis is the thickness and y-axis is the proportion of the pixels at each thickness in (c).

4. CONCLUSION

In this paper, the thickness map of isotropic thin film on uniaxial crystal are detected by SIE technology. The principles of SIE for uniaxial crystal and isotropic sample are analyzed and compared, and a special orientation is defined as the optical axis lies parallel to incident plane where the uniaxial crystal can be seen as isotropic material in SIE data processing. Then the light reflection on isotropic sample and uniaxial crystal are compared, the vibration azimuth angle of reflection of s and p-polarized light are unchanged at the same time only when the incident light is along to optical axis, which is the basis of the detection scheme for optical axis detection by rotating sample around Z-axis. Finally, the optical axis of uniaxial KDP crystal is decided by rotating sample, and the optical parameters of KDP substrate and thickness map of oil film on KDP crystal are measured by SIE.

The SIE technology is a powerful tool for thin film measurement. The measurement scheme proposed in this paper makes thin film measurement on uniaxial crystal the same as on isotropic sample, which predigests the data processing of SIE. The scheme can be wildly applied when the uniaxial crystal is used as substrate in SIE measurement.

Acknowledgments: Funding Support by Laboratory of Precision Manufacturing Technology, CAEP (Grant No. KF13002).

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