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A liquid crystal variable retarder-based reflectance difference spectrometer for fast, high precision spectroscopic measurements

Chunguang Hu^{*}, Pengfei Xie, Shuchun Huo, Yanning Li^{*}, Xiaotang Hu

State Key Laboratory of Precision Measuring Technology and Instruments, Tianjin University, Tianjin, 300072, China

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ABSTRACT

We present a liquid crystal variable retarder (LCVR)-based design for reflectance difference (RD) spectrometry, which offers one high quality RD spectrum in the visible range within several seconds. The measurement principle and the instrument development of this design are provided. As the LCVR is a key component, investigations are focused on its wavelength-dependent optical characteristics, the systematic errors induced by its imperfections, and the temperature effect on the measurement results. With careful calibration and data correction, the qualities of corrected RD spectra, defined as standard deviations of the RD signals as a function of wavelength and time, are better than 3×10^{-4} and 1.3×10^{-3} , respectively.

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1. Introduction

Reflectance difference spectroscopy (RDS), also called reflectance anisotropy spectroscopy, has high surface sensitivity and is widely used in the fields of semiconductor, thin film growth, biological detection, and so on [1]. A common instrument of the RD spectrometry was developed in the 1980s [2]. It is based on a photoelastic modulator (PEM) with a working frequency of several tens of kHz. Thus, the measurement time at one wavelength goes down to millisecond scale. This type of instrument can also acquire RD spectra in a broad wavelength range by using a wavelength-scanning monochromator, but it is time consuming [3]. In order to realize fast spectroscopic measurements, a rotating-compensator (RC)-based RD spectrometer was developed, which offers an RD spectrum from 1.5 to 4.5 eV within 10 s [4]. However, the optical imperfections of a super-achromatic compensator and the mechanical rotation-induced measurement errors complicate the procedures for signal calculation and system calibration and correction. Precise synchronization between the detector and the rotation action of the compensator is needed by using a sophisticated control mechanism.

A liquid crystal variable retarder (LCVR) allows a controllable phase retardation [5], which depends on the applied electric field, but is compact in size with respect to the PEM. The LCVR has been an alternative to the polarization modulation in various designs of the ellipsometry/polarimetry [6–8]. Many of those designs are of single-wavelength measurements as the retardation value of the LCVR is a nonlinear function of the applied voltage. Here, we propose an LCVR-based spectroscopic RD

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spectrometer with the following reasons: First, there is no optical element mechanically rotated during measurement when the LCVR is applied as phase retardation modulator. Errors related to the mechanical rotation are avoided. At the same time, the structure of the instrument becomes simple thanks to the compact size of the LCVR. Second, the retardation of the LCVR can either be modulated at a low frequency or be fixed. This permits the LCVR to work with a linear array detector for fast spectroscopic measurements. Third, the control protocol is easy because only two steps are needed sequentially and repeatedly: set the LCVR to a desired retardation value and read the intensity spectrum from the detector.

In this paper, the mathematical modeling as well as the data collection and reduction are presented in Section 2. A detailed instrumentation is given in Section 3. The electro-optical characteristics of the LCVR as a function of wavelength and applied voltage are measured in Section 4. At the end, the defects of the LCVR-induced systematic errors, the temperature influence on the measurement precision, and the measurement repeatability are explored in Section 5.

2. Measurement principle

An LCVR-based RD spectrometer (LC-RDS) discussed below follows a configuration of polarizer-LCVR sample analyzer. Similar to the PEMbased design, one LCVR device is used as phase retardation modulator. It is noted that this design cannot measure all polarization properties of the sample, e.g., depolarization. Using Muller matrix approach, the intensity imposing on one pixel of the array detector, i.e., one wavelength channel, can be expressed as Eq. (1) [3]. Here, the azimuthal orientations of the polarizer, the LCVR, the sample, and the analyzer are set at 0, 45°, 0, and 45°, respectively, and it is supposed that the sample has no depolarization effect.

^{*} Corresponding authors. Tel.: +86 22 5801 8712; fax: +86 22 2740 6941. *E-mail addresses*: cghu@tju.edu.cn (C. Hu), drynli@tju.edu.cn (Y. Li).

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$$I = I_0 (1 + N\cos\delta + S\sin\delta), \tag{1}$$

where $N = -\cos 2\psi$, $S = \sin 2\psi \sin \Delta$, and ψ and Δ are ellipsometric parameters of the sample. I_0 is the base intensity and should be constant during measurement. δ is the retardation of the LCVR. Eq. (1) is the same as the one achieved from the one-PEM-based RD spectrometer [3]. However, a nonlinear relationship between the retardation and the applied voltage exists in the LCVR device. A sinusoidal modulation of the applied voltage does not lead to a sinusoidal change of the retardation. The way for data reduction in the PEM-based RD spectrometer is not fit for the LVCR-based design. As an alternative solution, scanning the retardation step by step is carried out in our design.

In principle, only three intensity signals of *I* at different retardation values of the LCVR are needed to solve Eq. (1). In order to optimize the measurement quality, the least square method is applied. The best approximations of *N* and *S* are deduced using Eq. (2) according to a set of *I* acquired at *n* different values of δ .

$$\begin{pmatrix} I_1\\I_2\\\vdots\\I_n \end{pmatrix} = \begin{pmatrix} 1 & \cos\delta_1 & \sin\delta_1\\1 & \cos\delta_2 & \sin\delta_2\\\vdots & \vdots & \vdots\\1 & \cos\delta_n & \sin\delta_n \end{pmatrix} \begin{pmatrix} I_0\\I_0N\\I_0S \end{pmatrix}, \quad (n>>3).$$
(2)

One gets the RD signal of $\Delta r/r$ as below [4]

$$\frac{\Delta r}{r} = 2 \frac{N + iS}{1 + \sqrt{1 - N^2 - S^2}}.$$
(3)

Here, Δr is the difference in reflectance of normal incidence plane-polarized light between two orthogonal directions in the surface plane. *r* is the mean reflectance. Both *N* and *S* are normally much less than 1, and the real and imaginary parts of the RD signal approximately equal to *N* and *S*, respectively. Since the intensity spectrum is measured in parallel using a linear array detector, the RD spectra are gained by a parallel calculation for all wavelengths.

3. Instrumentation

The schematic diagram of LC-RDS is shown in Fig. 1. A white light source (Ocean optics, DH-2000-BAL) emits light, which passes through a linear polarizer (Edmund optics, Glan-Thompson type) and an LCVR (Thorlabs, LCC1113-A) and then reaches to the sample. The reflected



Fig. 1. (Color online) Scheme of the liquid crystal variable retarder-based reflectance difference spectrometer. See Ref. [5] for details of the LCVR.

beam goes through an analyzer, which is the second linear polarizer and is fed into a spectrometer (Ocean optics, QE65Pro) via a multimode fiber. Intensity spectrum is recorded by a detector array in the spectrometer. Different to the ellipsometer, the angle between the incident beam and the reflected beam is less than 5° for RD spectrometer. It is a basic requirement to obtain the in-plane optical anisotropy of the sample.

The control procedure for one measurement is performed as follows: 1) change the retardation of the LCVR, start an integration of the detector afterwards, and read out the intensity spectrum when the integration is completed. 2) Repeat the first step at *n* different retardations of the LCVR. 3) According to the collected intensity spectra and the corresponding retardations of the LCVR, the RD spectra are deduced using Eqs. (2) and (3).

4. Calibration

The retardation δ of the LCVR in Eq. (2) has to be known before data reduction. Since the commercial LCVR used in our setup is only calibrated at one wavelength by the manufacturer, it is necessary to determine the retardation value of the LCVR as a function of applied voltage and wavelength [9,10]. However, it is reported that the fast axis orientation of the LCVR might be also dependent on the voltage [11]. We follow references [6] and [12] and do a calibration with the steps below:

- 1) The fast axis of the polarizer is placed to be orthogonal to the one of the analyzer, in which case there is no light passing through the two-element structure.
- 2) The LCVR is placed between the polarizer and the analyzer. By rotating the LCVR, its azimuthal axis is determined when no light or a minimum intensity is measured from the three-element constructed setup. The mathematical expression is

$$I = I_0 (1 - \cos\delta) \sin^2(2L) \tag{4}$$

where *L* is the fast axis orientation of the LCVR with respect to the active axis of the polarizer. Note that this step does not work well when the retardation is close to a value of 2π or an integral multiple of 2π .

3) The analyzer is fixed at a place where its fast axis is parallel to the polarizer. The retardation of the LCVR is determined by fitting the intensities as a function of the rotation angle of the LCVR according to Eq. (5).

$$I = I_0 \Big[2 - (1 - \cos\delta) \sin^2(2L) \Big] \tag{5}$$



Fig. 2. (Color online) Orientation of the fast axis of the LCVR vs. wavelength and voltage. The value of the fast axis is the indication number of the rotation stage in unit of degree.

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