Rotating analyzer–fixed analyzer ellipsometer based on null type ellipsometer

Stoyan C. Russev
Department of Solid State Physics, Faculty of Physics, Sofia University, 1164 Sofia, Bulgaria

Tzanimir Vl. Arguirov
Laboratory of Thermodynamics and Physicochemical Hydrodynamics, Faculty of Chemistry, Sofia University, 1126 Sofia, Bulgaria

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The theory and design of an inexpensive rotating analyzer unit is presented, which allows a conventional null type ellipsometer to work as rotating analyzer–fixed analyzer automatic ellipsometer, without sacrificing the possibility to work in null mode. The mode switching is performed simply by adding or removing the rotating analyzer from its holder. It is shown that the rotating analyzer phase shift in rotating analyzer–fixed analyzer mode can be run-time determined from the measured Fourier coefficients. This avoids any need of recalibration procedure after mode switching and makes unnecessary plane of incidence synchronization, which further simplifies the needed hardware and reduces the errors connected with the phase shift instability of the output signal. The run-time phase shift calibration procedure and subsequent ellipsometric angles determination do not involve normalization of the output signal Fourier coefficients to the zeroth harmonic, eliminating in this way the influence of the dc component time drift. © 1999 American Institute of Physics. [S0034-6748(99)01807-9]

I. INTRODUCTION

Ellipsometry is well recognized as a powerful technique for nondestructive material characterization. It is based on the measurement of the light polarization state change, caused by the interaction with the sample.1 There is a large variety of ellipsometer arrangements which differ in their design, operation, and measurement capabilities.2 Among them the usual and oldest null type1 and rotating element types3–5 are probably the most often used and are commercially available. Both null and rotating analyzer ellipsometer types have their advantages and disadvantages in comparison to each other. The null type, because of the nature of nulling operation, does not involve errors connected with nonlinearity and drift of the electronic part1,6,7 and the optical element imperfections can be more easily accounted for by using two or four zone measurements.1 Rotating element type is faster; thus it is more suitable for kinetic measurements, and can operate without a wavelength dependent compensator, which is important for spectroscopic applications.3,8–10

The conventional null type ellipsometer is still widely used in many laboratories. In this article the conversion of the null polarizer-compensator-sample-analyzer (PCSA) type ellipsometer into the rotating analyzer-fixed analyzer (PCSrotA-A) type, without sacrificing the null mode operation, is considered. The two modes can be easily switched by simply removing the rotating analyzer from, or putting back to, its holder.

The rotating analyzer type polarization state detector (PSD) can be either, the rotating analyzer (rotA) or rotating analyzer–fixed analyzer (rotA-A) type.2 Both PSDs are equivalent in measurement capability, they are partial polarization state detectors, giving only two normalized components among the four components of the incident light Stokes vector.2 Nevertheless, rotA-A PSD in comparison to rotA PSD offers several advantages in comparison to rotA PSD:

1) rotA-A PSD produces a constant polarization state at the detector input, thus avoiding errors connected with the detector polarization dependent sensitivity2 and the need for their correction.2,7,10

2) The construction of the usual null type ellipsometer is very suitable for using both as null PCSA type ellipsometer and as PCSrotAA type by introducing a rotating analyzer unit (RAU) between the sample S and the analyzer A (i.e., in front of the analyzing arm of the null type ellipsometer). In the next section the performance of such an ellipsometric setup is considered.

3) The possibility of extracting more than two parameters from the output signal analyses for rotAA PSD arises from the abundance of the information contained in it, in comparison to the rotA PSD output signal. The latter contains only zeroth and second harmonics (three Fourier components and two after normalization), while the former output signal contains zeroth, second and fourth harmonics,2 thus giving four normalized Fourier components. As the analyzed polarization state is characterized by two parameters, the four Fourier coefficients offer the possibility of extracting two additional unknown parameters. One of these can be used to overcome the need of normalization of the Fourier coefficients. It means, that the measured polarization state parameters can be expressed by the combination of ratios of the second and fourth harmonic Fourier coefficients; thus it
is not necessary to normalize them to the zeroth harmonic. Consequently, only the ac component of the output signal can be treated; dc signal may be blocked at the very beginning. ac amplification and signal handling is always an easier task than dc treatment, as the temperature and time drift in the electronic part is mainly connected with dc drift. Due to $1/f$ noise, usually cutting the dc component increases the signal to noise ratio. Thus it offers potentially better long time stability and increases the precision of the ellipsometric measurement.

In Sec. III the theory of output signal data treatment for extracting the ellipsometric angles using only ac components of the output signal is considered.

II. EXPERIMENTAL SETUP

The principle of operation of the null type and rotating analyzer type ellipsometers are well known and only brief information for the setup is given in this section. The PCSrotAA output signal data treatment is considered in more detail in the following sections.

The output signal of the rotating analyzer type ellipsometer has cyclically varying intensity with a limited number of Fourier components, which carry the information for the light polarization state. Analog to digital (A/D) conversion of the output signal and subsequent numerical Fourier analysis is usually preferable to an all analog output signal treatment. Two series of synchronization pulses should be supplied from the rotating analyzer unit assembly; one pulse per revolution for the synchronization of the initial data point and circle dividing pulses for synchronization of A/D conversion with current rotating analyzer azimuth. The former is necessary for connecting the rotating analyzer azimuth to the plane of incidence and as mentioned above, can be avoided if the self calibration procedure further described is used.

Figure 1 shows schematically the combined null type–rotating analyzer type ellipsometer. The setup is based on the usual PCSA null type ellipsometer (LEF 3M, Novosibirsk, Russia). The RAU is introduced in front of the analyzing null ellipsometer arm. The RAU serves to rotate with constant speed (11 Hz) the rotating analyzer rotA and to produce the circle dividing pulses (1024 pulses/revolution in this case), which are fed to the electronic part for a synchronization of the A/D conversions. The mode switching is performed by

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FIG. 1. Null PCSA type ellipsometer with attached rotating analyzer unit (RAU). Mode switching between null PCSA type and PCSrotAA is performed by inserting to or removing from its holder the analyzer $A_r$. 

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introducing or removing the rotA from its holder. Another alternative would be to remove and put back the whole RA unit. In null mode the ellipsometer operates as usual, using its own null detection system. In rotA/A mode the output signal is fed to the preamplifier, S/H and A/D converter (12 bits), which samples the signal synchronously with the circle dividing pulses. The digitalized data are simultaneously transferred to the PC for real time fast Fourier analyses. The so-determined second and fourth harmonics Fourier coefficients (zeroth harmonic is not needed) are then used in a self calibration procedure to find the rotating analyzer phase shift and finally the ellipsometric angles Ψ and Δ. The computer controlled D/A converter (12 bits) is used to control the HV photomultiplier supply in order to fit the amplitude of the output signal to the A/D converter input range.

A/D conversions are transferred to the computer memory through direct memory access (DMA), while the main procedure performs the computation, ensuring ready results at the end of the current rotating analyzer cycle. The software provides computation of the ellipsometric angles for any settings of the ellipsometer optical elements azimuths (polarizer, compensator, and analyzer) with arbitrary predefined compensator parameters. Data averaging can be performed for any number of the rotating analyzer revolutions. The real time computed Fourier coefficients are averaged, but not the A/D conversions, which prevents the errors connected with source intensity and/or amplification drift.

The setup can also be used in a mode, described in Ref. 14 with no change in hardware and software. The difference is that PSD is rotAA type instead of a rotA type in Ref. 14 and that digital Fourier analyses instead of a lock-in based output signal treatment is applied.

### III. OUTPUT SIGNAL DATA TREATMENT IN PCSrotAA MODE

It is convenient to split the assembly in Fig. 1 into three parts: polarization state generator (PSG), sample S, and polarization state detector (PSD). The sample S is characterized by

\[ \rho = \tan(\Psi) \exp(i \Delta) = \chi_{\text{PSG}} / \chi_{\text{PSD}}, \]

where Ψ and Δ are ellipsometric angles to be determined, \( \chi_{\text{PSG}} \) and \( \chi_{\text{PSD}} \) are the polarization ratios at the PSG output and PSD input, respectively.\(^1\)

Polarization ratio \( \chi_{\text{PSG}} \) at the PSG output can be regarded further and as known, it can be either computed\(^1\) or measured in straightforward arm configuration with the PSD as explained below. It is interesting to note, that in the latter case no assumption for the ideality of the PSG optical elements has to be made and even their azimuthal calibration is not needed. It means that PCSrotA-A ellipsometer can work with only fixed analyzer azimuth calibration, if the rotA phase shift self calibration procedure is used and \( \chi_{\text{PSG}} \) is measured preliminarily in straightforward configuration. Therefore we concentrate only on the PSD (RAU plus the fixed analyzer of the null ellipsometer) and how the polarization ratio \( \chi_{\text{PSD}} \) is measured.

The output signal intensity \( J_{\text{out}} \) after rotating analyzer–fixed analyzer can be easily computed, using Jones formalism:\(^1\)

\[ J_{\text{out}} = J_0 [1 + c_2 \cos(2A_\rho) + s_2 \sin(2A_\rho) + c_4 \cos(4A_\rho) + s_4 \sin(4A_\rho)], \]

where \( A_\rho \) is the rotating analyzer azimuth with zero phase shift according to the fixed analyzer azimuth \( A \) (i.e., \( A_\rho = 0 \) in the \( A=0 \) plane). Let us denote with \( \chi^A_\text{PSD} \) the polarization ratio \( \chi_{\text{PSD}} \) in the coordinate system of the fixed analyzer. It is straightforward to obtain

\[ \chi^A_\text{PSD} = \frac{\sin(A) + \chi^A_\text{PSG} \cos(A)}{\cos(A) - \chi^A_\text{PSG} \sin(A)}, \]

The sign of the imaginary part is undefined from Eq. (3).

\[ \chi^A_\text{PSD} = \pm \sqrt{\chi^A_\text{PSG}^2 - s_2^2 / c_2^2}, \]

which reflects in ambiguity of the sign in the imaginary part of Eq. (4).

Presently, knowing the polarization states before and after the sample, the ellipsometric angles Ψ and Δ can be found using Eqs. (1), (3), and (4):

\[ \Psi = \arctan \left( \frac{\chi_{\text{PSG}}}{\chi_{\text{PSD}}} \right), \]

\[ \Delta = \arg(\chi_{\text{PSG}}) \mp \arg(\chi_{\text{PSD}}). \]

Equation (6b) expresses in general form the well known ambiguity in Δ determination. It is often stated that rotating element ellipsometers can measure only the absolute value of Δ and the sign is ambiguous. It is true in the most often used configurations when the PSG consists of only a polarizer (usually at \( P = \pm \pi / 4 \)) or generally when PSG produces linearly polarized light. In this case \( \arg(\chi_{\text{PSG}}) = 0 \) (or \( \pi \)) and Eq. (6b) gives \( \mp \Delta \) (0 ≤ Δ ≤ π), but in a null ellipsometer with a RA unit one can choose an arbitrary PSG configuration and then Eq. (6b) should be used. The free choice of \( \chi_{\text{PSG}} \) is another advantage of the present setup, because the reflected polarization can always be adjusted to the circular polarization state where the PSD has maximal sensitivity.\(^3\) This leads to the condition \( \chi_{\text{PSD}} = \pm j \), which in combination with Eq. (6a) gives two relations

\[ |\chi_{\text{PSG}}| = \tan(\Psi), \]

\[ \arg(\chi_{\text{PSG}}) = \Delta \mp \pi / 2, \]
for finding the polarizer \( P \) and the compensator \( C \) optimal settings. Usually there is no need to use exactly the optimal \( P \), \( C \) azimuths and consequently to solve Eq. (7). The control software of our setup constantly draws the polarization ellipsis, corresponding to \( \chi_{\text{PSG}} \) and \( \chi_{\text{PSD}} \). This allows easy adjustment of the optimal \( \chi_{\text{PSD}} \) for any investigated surface by visual observation of the polarization and changing the polarizer and/or compensator azimuths in order to achieve near to circular \( \chi_{\text{PSD}} \) polarization.

It should be noted that the presence of the compensator in the setup is a disadvantage when the instrument is used for spectroscopic measurements. PSRotAA is a more convenient configuration in this case. The need to remove the compensator from the optical scheme can be avoided by choosing the proper compensator azimuth; if the fast or slow compensator axes coincides with the polarizer azimuth, the PSG polarization is not influenced by the compensator parameters. Another alternative will be the preliminary determination of the compensator parameters for each used wavelength or measuring \( \chi_{\text{PSG}} \) in straightforward configurations for different wavelengths, as discussed above.

**IV. DETERMINATION OF THE ROTATING ANALYZER PHASE SHIFT**

As was already mentioned, the rotating analyzer azimuth \( A_r \) in Eq. (2) is measured according to the fixed analyzer \( A = 0 \) plane (i.e., \( A_r = 0 \) in this plane). This special case is not always present, because the switching between null and RA mode usually results in different rotating analyzer phase shift, \( A_{r0} \neq 0 \); see that the Fourier coefficients in Eq. (2) are:

\[
\begin{align*}
    c_2 &= c'_2 \cos(2A_{r0}) - s'_2 \sin(2A_{r0}), \\
    s_2 &= c'_2 \sin(2A_{r0}) + s'_2 \cos(2A_{r0}), \\
    c_4 &= c'_4 \cos(4A_{r0}) - s'_4 \sin(4A_{r0}), \\
    s_4 &= c'_4 \sin(4A_{r0}) + s'_4 \cos(4A_{r0}),
\end{align*}
\]

where primes refer to the actually measured Fourier coefficients when \( A_{r0} \neq 0 \).

Fourier coefficients in the coordinate system, connected with the fixed analyzer, are not independent and satisfy the following relations:

\[
\begin{align*}
    s_2 &= 2s_4, \\
    c_2 &= 1 + c_4.
\end{align*}
\]

Relation (9a) does not involve normalization of the Fourier coefficients, while relation (9b) holds true only for normalized Fourier coefficients \( c_2 \) and \( c_4 \). In order to keep the advantage of not using the dc component, only the former must be used in the derivation of the rotating analyzer phase shift.

Substitution of Eqs. (8b) and (8c) into Eq. (9a) leads to the fourth degree polynomial for \( x = \sin(2A_{r0}) \):

\[
P_4(x) = a_4x^4 + a_3x^3 + a_2x^2 + a_1x + a_0 = 0,
\]

with coefficients

\[
\begin{align*}
    a_4 &= -16(c'_4^2 + s'_4^2), \\
    a_3 &= 8(s'_2c'_4 - c'_2s'_4), \\
    a_2 &= 16(c'_4^2 + s'_4^2) - (c'_2^2 + s'_2^2), \\
    a_1 &= 4(s'_2c'_2 - 2c'_4s'_2), \\
    a_0 &= s'_2^2 - 4s'_4^2.
\end{align*}
\]

Although polynomial Eq. (10) can be solved analytically to directly find its four roots, some physical considerations help to find numerically the only proper root, that corresponds to the rotating analyzer phase shift.

The output signal from the RAU has an absolute minimum (zero intensity for an ideal RAU) when the rotating analyzer is crossed with the fixed analyzer. These points occur twice for every full rotation at rotating analyzer azimuth given by

\[
A_{r \min} = A - A_{r0} \pm \pi/2.
\]

In the coordinate system of the fixed analyzer \( A = 0 \) and consequently, the proper root of the polynomial Eq. (10) is

\[
x = \sin 2A_{r0} = \sin 2(-A_{r \min} \pm \pi/2) = \sin 2A_{r \min}.
\]

Rotating analyzer azimuth \( A_{r0} \) can also be found from the condition

\[
dJ_{\text{out}}(A_r)/dA_r = 0,
\]

which can be shown to be equivalent to Eq. (10).

Equation (14) will give four solutions in general in the interval \( 0 < A_r < \pi \). Two of them correspond to maximums of the output signal and can be easily discarded. The remaining two solutions correspond to minima—one of them corresponds to \( A_{r0} \) and is a global minimum independently of the polarization state \( \chi_{\text{PSD}} \). The other is generally a local one and depends on \( \chi_{\text{PSG}} \), for example, it is missing for the circle polarization at the PSD input. Another extreme case is linear polarization, in this case, the second minimum is also global (\( J = 0 \) for ideal PSD–A, is normal to the linear polarization plane). Consequently, the global minimum of the output signal determines the rotating analyzer phase shift unambiguously only if the input polarization state is not linear. As was discussed above, this situation can be avoided by the proper choice of \( \chi_{\text{PSG}} \).

As a first approximation \( A_{r \min} \) can be used:

\[
A_{r \min}^0 = 2\pi k_{\min}/N,
\]

where \( N \) is the number of sampled points for one revolution and \( k_{\min} \) is the point index \( (k = 0 - N - 1) \) at which the sampled signal has minimum value. This index can be easily determined at run time during sampling the output signal, by determination of the index of the minimum A/D value for \( N \) consequently sampled points. The starting point can be arbitrary; no initial synchronization is needed to start the measurements. The error in determination of \( A_{r \min} \) from Eq. (15) will be of the order of \( \pm 2\pi/N \) and is unacceptable. Moreover, Eq. (15) is based on a single point only; it is better to use all \( N \) points. Therefore, Eq. (15) is only the first approxi-
mation and the polynomial Eq. (10) has to be solved to more precisely find \( A_{r, \text{min}} \). The Newton–Raphson method\(^\text{15}\) is very convenient for iterative solution of Eq. (10):

\[
x_{n+1} = x_n - \frac{P_4(x_n) / dx}{P_\delta(x) / dx}|_{x=x_n},
\]

(16)

with initial approximate value, found from Eqs. (13) and (15)

\[
x_0 = \sin(2A_{r, \text{min}}) = \sin(4\pi k_{\text{max}}/N).
\]

(17)

As \( x_0 \) is very close to the polynomial root, usually 2–3 iterations of Eq. (16) are enough to achieve relative error less than 10\(^{-6}\). After the determination of the root \( x \) Eqs. (8) are used to find corrected Fourier coefficients using

\[
\sin(2A_{r, 0}) = x,
\]

\[
\cos(2A_{r, 0}) = \pm \sqrt{1 - x^2},
\]

\[
\sin(4A_{r, 0}) = \pm 2x \sqrt{1 - x^2},
\]

\[
\cos(4A_{r, 0}) = 1 - 2x^2.
\]

(18)

The signs of \( \cos(2A_{r, 0}) \) and \( \sin(4A_{r, 0}) \) are determined from the conditions [Eqs. (8) and (9)] or simply by using the initial approximation of the rotating analyzer phase shift \( A_{r, 0} \).

The primed Fourier coefficients in Eq. (8) are determined by

\[
c'_2 = \sum_{k=0}^{N-1} I_k \cos(4\pi k/N), \quad s'_2 = \sum_{k=0}^{N-1} I_k \sin(4\pi k/N),
\]

\[
c'_4 = \sum_{k=0}^{N-1} I_k \cos(8\pi k/N), \quad s'_4 = \sum_{k=0}^{N-1} I_k \sin(8\pi k/N),
\]

(19)

where \( I_k \) is the \( k \)th A/D converted value of the output signal, without any normalization to the dc component and the number of points \( N \). As only these nonnormalized Fourier coefficients are used, any dc offset of the signal amplification or A/D converter is not a source of errors.

The above proposed procedure for the determination of the rotating analyzer phase shift is fast enough to ensure run time operation. This allows ellipsometric angles to be determined for each RA period without any dead intervals of untreated output signal.

**V. DISCUSSION**

The modification described in the ellipsometric setup was intended to perform measurements on liquid surfaces. The random scattering in the ellipsometric angles, caused by the surface waves, was successfully overcome by continuously measuring and averaging in a suitable time interval. The improved long-time stability of the automated rotAA setting allows one to monitor kinetic processes with characteristic times of several hours, and relatively small change in the ellipsometric angles. An example is given in Fig. 2, where protein adsorption from the water solution at the air–water interface was measured ellipsometrically in rotAA mode (incidence angle 50°, wavelength 632.8 nm, polarizer azimuth \( P = 5^\circ \), compensator azimuth \( C = 0^\circ \), fixed analyzer azimuth \( A = 0^\circ \)). PSG configuration (\( P \) and \( C \) azimuths) ensures close to the optimal circular polarization state at the PSD input. This makes possible measurements of the ellipsometric angle \( \Delta \) even when it is near to 0° or 180° (as it is in our case). The average interval of 5 min was chosen according the rate of the protein adsorption. The photometric curves are given together with the long-time stability curves obtained on an unchangeable surface. We investigated the long-time stability of the setup in rotAA mode by measuring (incidence angle 70°, wavelength 632.8 nm) a glass surface for 9 h. The results are shown in Fig. 2. The maximum drift in measured ellipsometric angles is 0.013° for \( \Psi \) and 0.2° for \( \Delta \). It should be noted that these values characterize the resolution, but not the absolute precision of the measured ellipsometric angles.

The main advantage of the algorithm presented for the output signal data treatment in the rotAA mode is the run time determination of the rotating analyzer phase shift. This ensures mode switching with no need of calibration procedure and reduces the phase shift connected errors. The procedure for the evaluation of the ellipsometric angles and the above procedure do not involve the dc component of the output signal, thus reducing the errors, connected with dc drift.

Our experience in the usage of the above described ellipsometric setup shows that fast switching between two modes without moving the sample is an important advantage in kinetic measurements. Four zone measurements in null mode give better absolute precision, provided the intensity reflected from the sample is sufficient and no or very slow kinetic process is under consideration. Photometric rotAA mode has potentially better resolution\(^\text{16}\) and is faster in comparison to null mode, but the absolute precision is poorer. The combined use of the two modes give the advantage of exploiting both the precision of the null measurement and the resolution and speed of the photometric one. An example can
be found in Ref. 17, where the formation of Ag₂S films on silver surface in the presence of diluted H₂S gas is investigated. Null mode measurement is used for the precise determination of the ellipsometric angles of the structure (base point) before exposure to gas. Then the photometric rotAA mode is used to monitor their change during layer formation.

The advantage of the photometric mode is better manifested for low light intensities. We have used this mode for investigating the protein adsorption processes at the water/air and the water/oil interfaces. In these cases the reflected intensity is very low, especially for the water/oil/air system, where one has to measure the spot reflected from the internal interface and to separate it from the primarily reflected beam. Another problem is the inevitable liquid surface vibrations leading to additional difficulties in null mode measurements. The speed of a rotAA measurement gives another advantage; in a time needed for a single null measurement, several thousands of points can be measured and averaged in a rotAA mode. Thus the signal to noise ratio is greatly improved. The protein adsorption processes last several hours and consequently the long-time stability of the rotAA detection system is very important and is improved by the dc signal component exclusion in data treatment and run time phase shift determination. This is essential especially for low intensities, where the dc component is significantly influenced by external factors like temperature, diffused light, etc.

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