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A RETARDATION MODULATION ELLIPSOMETER FOR STUDYING FAST SURFACE TRANSIENTS

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Abstract - A high-speed ellipsometer has been constructed which can operate in microsecond orders. An example of the application to rapid growth of anodic films on GaAs is given.

1. Introduction

The ellipsometer with special advantages for high speed will be useful for studying rapid film growth and fast surface dynamics. We have tried to construct a high-speed ellipsometer having microsecond resolution. The use of an ADF four-crystal electrooptic modulator (EOM) for retardation modulation and a high-speed digitizer for data acquisition has made the high-speed operation possible/1,2/. This paper describes the ellipsometer system, the measurement method and the application to anodic film growth on GaAs.

2. Ellipsometer system

Two optical arrangements can be set up; $P$ (polarizer)$-Q$(EOM)$-S$(sample)$-A$(analyzer) and PQSQA. Here, $P$, $Q$, and $A$ also represent the azimuth angles of the optical components. The optical system is illustrated in Fig.1 for in situ measurements of anodic film growth. Since the PQSQA has been reported elsewhere /2,3/ and the PQSA has the advantage that the light beam propagates in the same optical path in the EOM independently of reflection from the sample surface, the PQSQA is described in this paper.

The optical components used in Fig.1 are as follows: a light source (L) of 1 mW He-Ne laser, pinholes (Ir), a quarter-wave plate (Qu) getting a circularly polarized light, a polarizer ($P$), the EOM having a bandwidth from dc to 25 MHz and the half-wavelength voltage of 115 V at 6328 $\AA$, an analyzer ($A$) and a photomultiplier(PM) with high-speed response and wide range linearity.

The electronic system is about the same as previously reported /2/. It consists of a function generator to generate modulation voltages of triangular waveform and timing triggers, a power amplifier to drive the EOM, the two channel digitizer with high-speed A-D converters, a sampling controller to control the timing of data acquisition and a minicomputer. The sampling controller determines the data-acquisition time $T$ in which $N$ data points of both the transmitted light intensity $I$ and the modulation voltage $V$ are acquired with the digitizer to obtain a data point $(\Psi, \Delta)$ of the sample. Next series of data acquisition of $N$ data points is repeated after intervals $T_i$. The memory capacity of the digitizer is 2 kwords/channel so that 2000/$N$ data points $(\Psi, \Delta)$ can be obtained in an experimental run. The intervals $T_i$ is programmable such that one value of $T_i$ is switched to the other in an experimental run according to the reaction speed of surface phenomena.
Fig.1 - The optical system and the electrolytic cell used in the in situ ellipsometric measurements of anodic film growth. L, He-Ne laser; Ir, pinhole; Qu, quarter-wave plate; P, polarizer; Q, EOM; S, sample; Pt, platinum cathode; A, analyzer; PM, photomultiplier; CCS, constant current source; SW, Hg-relay. Q replaces Q' when the PQSA arrangement is set up.

It has been shown that the highest speed of \( T = 4 \) \( \mu s \) is achieved with precision of \( 0.05^\circ \) in \( \Psi \) and \( 0.15^\circ \) in \( \Delta \) (root-mean square values) when a BaKl optical glass is used as a sample, and \( T_1 \) can be selected from \( 4 \) \( \mu s \) to 160 ms /2/.

3. Measurement method

The light intensity \( I \) at the photomultiplier in the PQSA arrangement with \( P = 0^\circ \) and \( Q = 45^\circ \) is expressed by

\[
I = I_0(1 + a_1 \cos \delta + a_2 \sin \delta + a_3 \cos^2 \delta + a_4 \sin^2 \delta)
\]

in which

\[
\delta = EV + C
\]

\[
a_1 = -(\cos 2\Psi - \cos 2\Delta - 2x_1\sin 2\Psi\cos \Delta \sin 2\Delta + x_4\sin 2\Psi\sin \Delta \sin 2\Delta)/a_0,
\]

\[
a_2 = (\sin 2\Psi \sin \Delta \sin 2\Delta + 2x_1\sin 2\Psi \cos \Delta \sin 2\Delta - x_4(\cos 2\Psi - \cos 2\Delta))/a_0,
\]

\[
a_3 = 2x_2\sin 2\Psi \cos \Delta \sin 2\Psi \sin \Delta \sin 2\Delta/a_0,
\]

\[
a_4 = x_4 \sin 2\Psi \cos \Delta \sin 2\Psi \cos \Delta \sin 2\Delta/a_0,
\]

\[
a_5 = 1 - \cos 2\Psi \cos 2\Delta - 2x_2\sin 2\Psi \cos \Delta \sin 2\Delta,
\]

to first order in \( x \). \( I_0 \) is the average light intensity. \( E \) is the proportionality constant in the electrooptic modulation and is determined by a best-fitting procedure to an \( I-V \) curve measured with \( A = 0^\circ \). \( C \) is a phase factor which varies slowly with temperature. The imperfection parameters \( x \)'s associated with the EOM arise from the window birefringence and axial misalignment of the ADP crystals in the EOM /3/.

\( \delta_0 \) is a phase factor resulting from the crystal birefringence and is sensitive to change in ambient temperature.

Consequently, the ellipsometric parameters \( \Psi \) and \( \Delta \) of the sample are derived from the coefficients \( a_1 \) and \( a_2 \) and \( C \) measured in calibration with \( A = 0^\circ \) if \( x \)'s are predetermined. In other words, two experimental runs are necessary whenever we want to get the ellipsometric parameters free from temperature fluctuation; one is the calibration with \( A = 0^\circ \) and the other is high-speed measurement with appropriate azimuth angle \( A \) immediately after or before the calibration.

It is understood from the similar calculation to that in the case of rotating-analyzer ellipsometers /4/ that the highest precision is achieved at \( A = \Psi \). We should be careful in setting \( A \) since \( \Psi \) of the sample changes with lapse of reaction time and deviates from the set azimuth angle in the transient measurements.
The coefficients $x$'s can be determined in the straight-through optical arrangement in which the following equations are valid from Eq.(1).

\[
\begin{align*}
    a_1 &= \cos 2A + 2x_1 \sin 2A + x_3 \sin 4A, \\
    a_2 &= x_4 \cos 2A + 2x_5 \sin 2A
\end{align*}
\]

(2)

are valid from Eq.(1). Figure 2 illustrates analyzer azimuth dependence of the coefficients $a_1$ and $a_2$ measured in this arrangement and the best-fitting curves calculated with the regression technique using Eq.(2). The best-fitting procedure yielded $x_{1F} = -0.009$, $x_{1P} = 0.005$, $x_{2P} = 0.005$ and $x_4 = -0.003$. These values are found small as compared with those in the previous PSQA arrangement/3/. $x_{1P}$ and $x_{1F}$ have been included to calculate $(\Psi, \Delta)$ in the present case.

![Fig.2 - Analyzer azimuth dependence of the coefficients $a_1$ and $a_2$ measured in the straight-through optical arrangement and the best-fitting curves for calibration of the imperfection parameters $x$'s.](image)

4. Application

As an example, the high-speed ellipsometry was applied to anodic film growth on GaAs. In most cases, anodization of GaAs has been performed with constant current densities below 1 mA/cm$^2$ to grow homogeneous films. With much higher current densities, the ellipsometers hitherto in use would not be able to follow the fast surface reactions.

P type GaAs with a carrier concentration of $3 \times 10^{18}$/cm$^3$ was used as a sample. The anodization was carried out under constant current condition in mixed solution of propylene glycol and aqueous solution of tartaric acid at pH 3/5/. The in situ ellipsometric measurement was made with use of the optical system as illustrated in Fig.1. The cell windows made of an annealed fused quartz have been found to have negligible birefringence. Figure 3 shows a typical response of $(\Psi, \Delta)$ with lapse of anodization time where the GaAs (100) surface was anodized with 40.8 mA/cm$^2$ for 3.92 s and the ellipsometric measurement was made under the condition $T = 40$ s, $T_i = 80$ s, $N = 40$, $\phi$ (incident angle) = 60° and $A = 26°$. The curves(A), (B), (C) and (D) indicate the film growth up to 880 Å calculated with the film refractive indices $n_f = 1.76, 1.78, 1.80$ and 1.82, respectively. Here, the substrate optical constant and the refractive index of the electrolyte are assumed to be 3.98 - 0.29i and 1.46, respectively. The experimental data suggest that $n_f$ tends to increase to the value of 1.8 found in the literature/6/ according as the film grows.
Fig. 3 - Response of (\(\psi\), \(\Delta\)) when the GaAs surface was anodized with the current density of 40.8 mA/cm\(^2\) for 3.92 s. The ellipsometric measurement was made with \(T = 40\ \mu\text{s}, N = 40, T_1 = 80\ \text{ms}\), \(\phi = 60^\circ\) and \(\epsilon = 26^\circ\). The solid curves (A), (B), (C) and (D) show the computed ellipsometry curves for grown films up to 880 Å thick with the refractive indices of 1.76, 1.78, 1.80 and 1.82, respectively.

5. Summary

A high-speed retardation modulation ellipsometer for studying rapid film growth and surface dynamics has been described; the optical and electronic system, the measurement method and the calibration of the imperfect parameters of the EOM has been shown in the PQSA optical arrangement with \(P = 0^\circ\) and \(Q = 45^\circ\). An example was given of the application to anodic film growth on GaAs anodized with high current density of 40.8 mA/cm\(^2\) for 3.92 s in which increasing tendency of the film refractive index was observed.

References