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Review: An Optical Surface Probe by Reflectance Anisotropy Spectroscopy

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ABSTRACT

Reflectance anisotropy spectroscopy (RAS) is an optical technique to produce surface and interface information. RAS measures the difference in reflectance of light linearly polarised along two orthogonal axes in the surface at near normal incidence as a function of photon energy. The quantity obtained by RAS is the so-called reflectance anisotropy. Since only the surface is anisotropic, the measured reflectance anisotropy is connected only with the atomic composition of the surface, not of the bulk. This review presents theoretical as well as experimental procedure to explain RAS technique.

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INTRODUCTION

Optical techniques have played an important role in solid state physics for a considerable time. Ellipsometry has a history of over 100 years [1-3]. Ellipsometry has a surprisingly broad range of application in different scientific areas are ranging from electrochemistry to medicine [4]. Ellipsometry is the general name for the family of optical techniques, based on exploiting the polarisation transformation that occurs as a beam of polarized light is reflected from or transmitted through an optical component. The application of optical techniques to surface science is complicated in terms of optical response and surface sensitivity.

An impressive way to obtain surface sensitivity is offered by reflectance anisotropy spectroscopy (RAS), a young member of the old family of Ellipsometry techniques. The standard RAS was introduced in 1985 by Aspnes [5-6]. One of the surface science techniques is the RAS, a rapidly developing new technique which has already made itself a name in controlling semiconductor growth at monolayer level both under ultra-high vacuum (UHV) and non-UHV conditions [7-8].

The quantity obtained by RAS is the so-called reflectance anisotropy (RA). RA can be described as in Eq 1:

$$\Delta r / r_i = 2\left(r_x - r_y\right) / \left(r_x + r_y\right) \qquad (1)$$

where r_i is the Fresnel complex-amplitude reflection coefficient for light polarised along the ith axes. RAS takes advantage of the fact that most semiconductors for instance GaAs, Si and Ge possess an isotropic bulk (cube lattice) and anisotropic (reconstructed) surface [9]. Regarding the Fresnel equation, polarisation dependent differences in the reflection of a light beam are inevitable because of anisotropy somewhere in the sample [10]. Since only the surface is anisotropic, the measured reflectance anisotropy is connected only with the atomic composition of the surface, not of the bulk. For this class of semiconductors RAS can gain surface sensitivity.

When measuring the RA, the main problem is the small difference between r_x and r_y (~1% for semiconductors). This is due to the fact that exclusively the surface layers, which provide only an insignificant influence to the total reflectivity, cause the anisotropy in the reflection. Small drifts and fluctuations in the output of the lamp used would make measurements meaningless, unless a high averaging process is employed.

For (110) metal surfaces, RAS can be described as in Eq 2:

$$\frac{\Delta r}{r}=\frac{2\left(r_{[1\bar{1}0]}-r_{[001]}\right)}{r_{[1\bar{1}0]}+r_{[001]}} \tag{2}$$

In equation 1, the x = $[1\bar{1}0]$ and y = [0 0 1] directions are the two symmetry-directions of the surface.

EXPERIMENTAL APPARATUS IN RAS

The RAS equipment is shown schematically in figure 1. A 75 W Xenon (Xe) lamp is employed as a photon source. The energy range of typical RAS spectrometers is 1.5 - 5.0 eV, 830 - 225 nm, from visible to ultraviolet. Plane-polarized incident light is directed on the sample surface. The axes are focused on 45° to the plane of polarization of the incident beam. The RAS is planned to produce data on the complex Fresnel reflection amplitudes r_x and r_y related with two orthogonal symmetry surface directions $\vec{x}$ and $\vec{y}$ respectively. $\vec{x}$ and $\vec{y}$ are taken along $[1\bar{1}0]$ and [001] directions respectively.

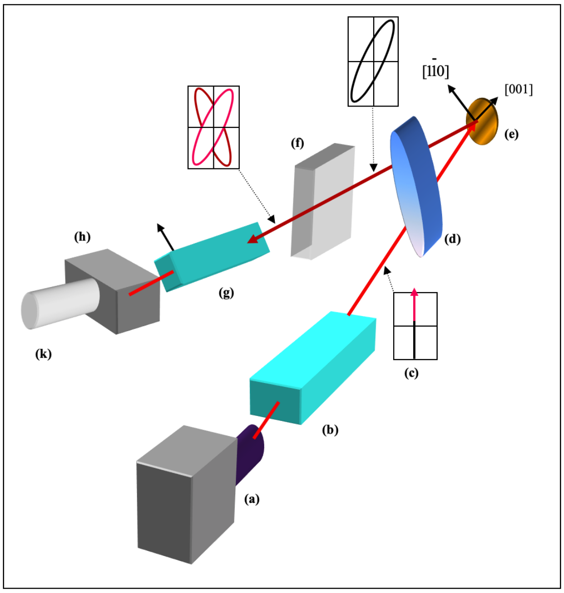

Figure 1. The RAS spectrometer: (a) Xe lamp (b) Polariser, (c) State of polarization, (d) Strain free RAS Window, (e) Sample in UHV environment, (f) photoelastic modulator, (g) Analyser, (h) Monochromator, (k) photomultiplier detector

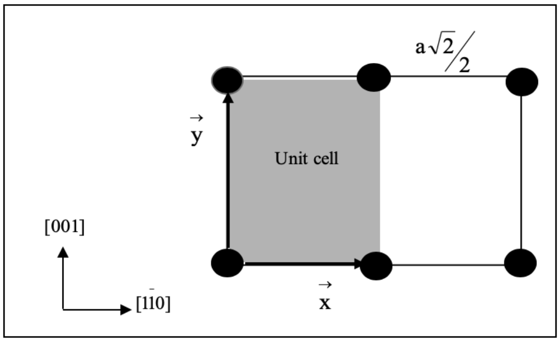

Figure 2. The geometry of the (110) surface in real space

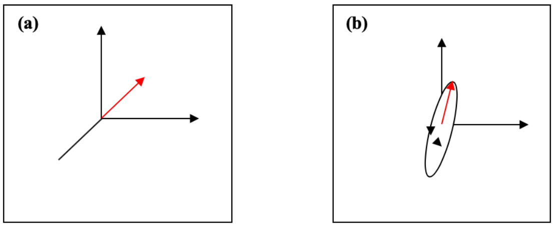

Figure 3. The polarisation of the (a) incident and the (b) emerging beam (for an arbitrary Δr/r) with respect to the principal axes of the sample.

In this review, for (110) metal surface can be taken into account. Figure 2 displays (110) metal surface and a unit cell of the real space lattice which is rectangular. The surface lattice vectors $\vec{x}$ and $\vec{y}$ are given by:

$$\vec{x}=\frac{a\sqrt{2}}{2}\hat{x} \tag{3}$$

$$\vec{y}=a\hat{y} \tag{4}$$

where $\hat{x}$ and $\hat{y}$ are unit vectors along $[1\bar{1}0]$ and [001] directions respectively.

Apparatus of RAS can be explained as follows:

Xenon-lamp:

The 75W high-pressure Xenon-lamp provides continuous and smooth spectrum with a sufficient output for light energies from 1.5 eV (830 nm) to 5.5 eV (225 nm). Due to the importance of a stable output, the lamp operates from a stabilised power supply. The light emerging from the lamp is linearly polarised before it strikes the sample at near normal incidence.

The polarisation-direction of the incident light beam is chosen so that we have equal amounts of polarised light along the 2 symmetry-directions as shown in Fig. 3(a). If the reflectivity for light polarised along these two directions were the same, the emerging light beam would obviously be linearly polarised as well. Since it is not, we get elliptically polarised light as shown in Fig. 3(b).

The tilt of the ellipse is related with the real part of the reflectance anisotropy i.e. Re (Δr/r), its breadth with the imaginary part of the reflectance anisotropy Im (Δr/r).

Photoelastic Modulator (PEM):

The subsequent arrangement of PEM and analyser generate an intensity-modulated signal from the light-beam, which contains the information about the reflectance anisotropy. The PEM modulates the phase of the component of the light beam linearly polarised parallel to its so-called modulation axis (frequency ω=50 kHz), while leaving the other component unaffected as presented in Fig. 4. As a

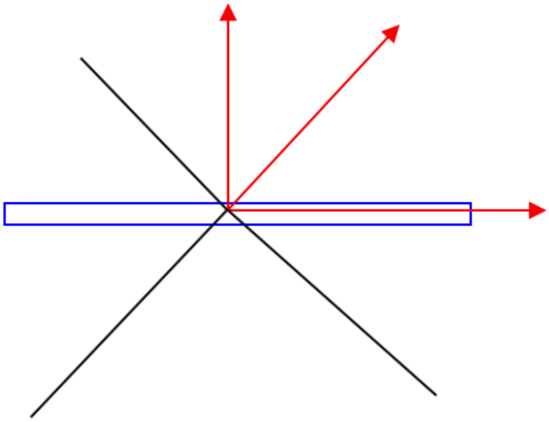

Figure 4. PEM modulates the phase of the component of the light beam

consequence of this modulation, the orientation and the form of the polarisation ellipse changes periodically [11-12]. The photomultiplier placed behind the analyser detects an intensity-modulated signal.

The Jones-analysis deals with the propagation of polarized light through polarization-modifying optical systems. Basically a light wave is represented by a vector and each optical component by a matrix. Modification of the state of the light wave by an optical component is obtained by multiplication of the vector of the wave with the matrix of the optical component.

It can be considered a monochromatic light beam with arbitrary polarization propagating along the z-axis of a coordination-fram (x,y,z). This can be described as [1]:

$$\begin{aligned} E(z,t) &= E_x \cos(\omega t - 2\pi z / \lambda + \delta_x) e_x \\ &+ E_y \cos(\omega t - 2\pi z / \lambda + \delta_y) e_y \end{aligned} \tag{5}$$

With E_i representing the amplitude of the linear oscillations of the electric field components along the i-axes, and δ_i the respective phases of these oscilations. In considering the polarization of the wave and its modification by an optical device, we do not need the full expression given by the wave above. The Jones-vector of the light wave contains complete information about amplitudes and phases of the field components, and hence, about the polarization of the above. Temporal and spatial information of the wave are supressed. The Jones-vector for the wave above is [1]:

$$E = \begin{pmatrix} E_x \\ E_y \end{pmatrix} \; with \; E_x = |E_x| exp(i\delta_x) \; ; \; E_y = |E_y| exp(i\delta_y) \tag{6}$$

The intensity of the light wave described by the Jones-vector is given by:

$$I = E^* E \; with \; E^* = \begin{bmatrix} E_x^* & E_y^* \end{bmatrix} \tag{7}$$

The description of the polarization-modifying effect of an optical device on an incident beam is given by a (2 x 2) matrix. It cab be assumed E_i to be the Jones-vector of the incident light beam with reference to a coordination frame (x,y,z) and E_o the Jones-vector of the emerging light-beam with reference to a coordination frame (x',y',z'), with the directions of the z- and the z'-axes parallel to the wave vectors k and k' respectively (z=0 and z' =0 are arbitrary) [1]:

$$E_i = \begin{pmatrix} E_{ix} \\ E_{iy} \end{pmatrix} \quad E_o = \begin{pmatrix} E_{ox'} \\ E_{oy'} \end{pmatrix} \tag{8}$$

In the absence of non-linearity and other frequency-changing processes, the pair of oscillations $E_{0x`}$ and $E_{0y`}$ at the output of the optical system are related to the pair of oscillations E_{ix} and E_{iy} at the input of the optical system by the following equations [1]:

$$E_{ox`} = T_{11}E_{ix} + T_{12}E_{iy} \tag{9}$$

$$E_{oy`} = T_{21}E_{ix} + T_{22}E_{iy} \tag{10}$$

$$\begin{pmatrix} E_{ox'} \\ E_{oy'} \end{pmatrix} = \begin{pmatrix} E_{ix} \\ E_{iy} \end{pmatrix} \begin{pmatrix} T_{11} & T_{12} \\ T_{21} & T_{22} \end{pmatrix} = TE_I \tag{11}$$

This expresses the law of interaction between the incident wave and the optical component as a simple linear matrix-transformation of the Jones-vector representing the wave. T is called the Jones-matrix of the optical componet, it is in general complex. The Jones-matrix T depends on the chosen coordination frames (x,y,z) and (x`,y`,z`).

Polarizer (Rochon-prism):

The lamp emits unpolarized light. It cannot be described the unpolarized light with a Jones-vector. Thus we start the analysis with the linear polarised light beam emerging from the polarizer. The Jones vector is [1]:

$$E_{PO^{xy}} = \begin{pmatrix} 1/\sqrt{2} \\ A \\ 1/\sqrt{2} \end{pmatrix} \tag{12}$$

where A takes account of the intensity. The first letter in the subscript stands for the optical component, the second for input (I) or output (O). The superscript xy means with reference to this frame.

Window:

We consider an ideal window that has negligible influence on the light beam. In this case we do not have to consider the windows in Jones-analysis.

Sample:

The Jones matrix of the sample is [1]:

$$T_{S^{xy}} = \begin{pmatrix} r_x & 0 \\ r_y & r_i \end{pmatrix} \tag{13}$$

where r_i is the Fresnel complex-amplitude reflection coefficient for light polarised along the i-axis.

The PEM modulates the phase of one component of the light beam periodically. We claim this is the only influence on the light beam by an ideal PEM. The principal axes of the analyser coincide with the xy-frame. The properties of an ideal analyser are analogous to those of an ideal polarizer [1].

$$T_{A^{xy}} = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \tag{14}$$

With the Jones matrix T_{RAS} of the whole arrangement it can be calculated the Jones' vector E_{AO} of the light beam arriving at the detector as:

$$E_{AO}{}^{xy} = T_{RAS}{}^{xy} E_{Pi}{}^{xy} \tag{15}$$

Regarding the matrix representing the analyser, we come to conclusion that:

$$T_{RAS}{}^{xy}{}_{(21)} = T_{RAS}{}^{xy}{}_{(22)} = 0 \tag{16}$$

Since the output of the analyser should be 0 for light polarised along the extinction axis we find that only: $T_{RAS}{}^{xy}{}_{(11)}$ is required to calculate the detected signal.

The reflectance anisotropy is described as [1]:

$$\Delta r \ / \ r \ = \ 2\left(r_x - r_y \right) / \ \left(r_x + r_y \right) \tag{17}$$

$$= \ 2 \left(a^2 + b^2\right) \ - \ 2\left(c^2 + d^2\right) \ + \ \frac{4i\left(ad \ - \ bc\right)}{\left[\left(a^2 + b^2\right) \ + \ \left(c^2 + d^2\right) \ + \ 2\left(ac \ + \ bd\right)\right]} \tag{18}$$

and if we use for the denominator a~c; b~d (as the difference is at best about 1 percent)

$$\begin{aligned} \Delta r / r \ &= \ 2 \left(a^2 + b^2\right) \ - \ 2\left(c^2 + d^2\right) \\ &+ 4i\left(ad \ - \ bc\right) / 4 \ \left(a^2 + b^2\right) \end{aligned} \tag{19}$$

which means:

$$Re \ \left(\Delta r / r\right) \ = \ \left[\left(a^2 + b^2\right) \ - \ \left(c^2 + d^2\right)\right] / \ 2\left(a^2 + b^2\right) \tag{20}$$

$$Im \ \left(\Delta r / r\right) \ = \ \left(ad \ - \ bc\right) / \ \left(a^2 + b^2\right) \tag{21}$$

Based on these two equations, Im (Δr/r) and Re (Δr/r) can be measured separately with the help of a Lock-In-amplifier.

THEORETICAL APPROACHES OF RAS

The propagation of the polarised light can be described through the optical apparatuses of the RAS by the Jones calculus [1]. The outcome of each optical factor upon the incident light is described by a (2 ´ 2) Jones matrix expressed in terms of fixed optical axes specific to the device of figure 1 and denoted T^{xy}_i where i shows an optical device. The effect is termed by a differential retardation as below [1]:

$$e^{i\delta_W} = \left(e^{i\delta_{Wf}}\right)\left(e^{i\delta_{Win}}\right) = \cos\delta_W + i\sin\delta_W \cong 1 + i\delta_W \tag{22}$$

The Jones matrixes and vectors have been summarized for each effective component in Table (1).

The pair of oscillations $E_{tx'}$ and $E_{ty'}$ at the output of the optical system are related to the pair of oscillations E_{ix} and E_{iy} at the input of the optical system in the absence of nonlinearity and other frequency-changing processes. Interaction between the incident wave and the optical component

Table 1. List of the Jones matrixes/vectors for each effective component

Device/Beam	*Jones Matrix*	*Jones Vector*	*Symbol/Remark*
Xe lamp	*N/A*	*N/A*	*Emits unpolarized light*
Incident light	*N/A*	$\begin{bmatrix} E_{ix} \\ E_{iy} \end{bmatrix}$	*E_i/Coordination frame (x,y)*
Transmission beam	*N/A*	$\begin{bmatrix} E_{tx'} \\ E_{ty'} \end{bmatrix}$	*E_t/Coordination frame (x′,y′)*
Linear polarised light	*N/A*	$K\begin{bmatrix} 1/\sqrt{2} \\ 1/\sqrt{2} \end{bmatrix}$	*$E_{Pt}{}^{xy}$/K as a constant of the detector*
Windows	$\begin{bmatrix} 1 & 0 \\ 0 & e^{i\delta_W} \end{bmatrix}$	*N/A*	*T_W/an ideal component*
Sample	$\begin{bmatrix} r_x & 0 \\ 0 & r_y \end{bmatrix}$	*N/A*	*$T_S{}^{xy}$/r_x and r_y are Fresnel complex amplitude reflection coefficient for light polarised along the $\vec{x}$ and $\vec{y}$ axes*
PEM	$\begin{bmatrix} 1 & 0 \\ 0 & e^{i\delta_M} \end{bmatrix}$	*N/A*	*$T_{PEM}{}^{t\,m}$ /t and m standing for transmission and modulation axes respectively, δ is B sin (ω t), so B is the amplitude and ω is the frequency of the modulation*
Analyser	$\begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix}$	*N/A*	*$T_A{}^{te}$/transmission axes of the analyser coincide with the xy-frame*
Polarizer	$\begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix}$	*N/A*	*$T_P{}^{te}$/The properties of an ideal analyser are analogous to those of an ideal polarizer*

can be given by a simple linear matrix-transformation of the Jones-vector as [1]:

$$\begin{bmatrix} E_{tx'} \\ E_{ty'} \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{bmatrix} \begin{bmatrix} E_{ix} \\ E_{iy} \end{bmatrix}, \quad E_{At}{}^{xy} = S_{RAS}{}^{xy} E_{Pi}{}^{xy} \tag{23}$$

where $S_{RAS}{}^{xy}$ is the Jones-matrix of the whole optical system. $S_{RAS}{}^{xy}$ can be written as:

$$S_{RAS}{}^{xy} = T_A{}^{te} R(\theta_M - \theta_A) T_M{}^{ts} R(-\theta_M) T_{Wf}{}^{xy} T_S{}^{xy} T_{Wi}{}^{xy} R(\theta_P) \tag{24}$$

The Jones vector $E_{At}{}^{xy}$ of the light arriving at the detector can be calculated by maintaining the Jones matrix $S_{RAS}{}^{xy}$ of the whole arrangement in figure 1 from the table 1. All matrices must operate in the correct order because they do not commute. Calculating $E_{At}{}^{xy}$ makes a final vector with one non-zero element, because the matrix representing the analyser $S_{RAS}{}^{xy}{}_{(21)} = S_{RAS}{}^{xy}{}_{(22)} = 0$. Since the output of the analyser should be zero for light polarised along the extinction axis, it was found that only $S_{RAS}{}^{xy}{}_{(11)}$ is required to calculate the detected signal. After matrix-multiplication the equation (23) gives:

$$E_{At}{}^{xy} = \left(1/2\sqrt{2}\right) K \begin{bmatrix} \left(r_x - r_y\right) + e^{i\delta}\left(r_x - r_y\right) \\ 0 \end{bmatrix} \tag{25}$$

Using Equation (22) and by substituting the azimuth angles in Equation (24), $E_{At}{}^{xy}$ is given by:

$$\begin{aligned} &E_{At}{}^{xy} = \\ &\left(1/2\sqrt{2}\right)\left[\left(r_x - r_y\right) + \left(r_x - r_y\right)e^{i\delta_M} - i\delta_W r_y \left(1 - e^{i\delta_M}\right)\right] \end{aligned} \tag{26}$$

The measured time-dependent light intensity, I, at the detector is given by:

$$I \infty E_{At} E_{At}{}^{*} \tag{27}$$

Following simple algebra, with $e^{i\delta} = \cos\delta + i\sin\delta$, $r_x = a - ib$ and $r_y = c - id$ where a, b, c and d are integer. Substituting these into Equation (27) results in an expression that can be separated into real and imaginary parts.

$$I \infty \left|E_{At}\right|^2 = \left\{\mathrm{Re}\left(E_{At}\right)\right\}^2 + \left\{\mathrm{Im}\left(E_{At}\right)\right\}^2 \tag{28}$$

After long calculations, Equation (27) becomes as below:

$$\begin{aligned} &I \infty 1/4\left[\left(a^2 + b^2\right) + \left(c^2 + d^2\right) + \left(c^2 + d^2\right)\delta_W{}^2\right] \\ &+1/4\left[\left(a^2 + b^2\right) - \left(c^2 + d^2\right) - \left(c^2 + d^2\right)\delta_W{}^2\right]\cos\delta_M \\ &1/2\left[\left(ab - cd\right) - \left(ac - bd\right)\delta_W\right]\sin\delta_M \end{aligned} \tag{28}$$

These expressions may be written in the form of:

$$I = I_{dc} + I_{\omega}\sin\delta_M + I_{2\omega}\cos\delta_M \tag{29}$$

The retardation δ_M induced by the PEM varies sinusoidally [12], following the expression

$$\delta_M = \alpha(\lambda)\sin(\omega t) \tag{30}$$

where a(l) and w are the amplitude of the modulation and the resonant angular frequency respectively. The frequency components of the signal are determined by the Fourier expansions of the terms $\cos\delta_M$ and $\sin\delta_M$ of Equation (29), introducing Bessel functions J of argument a (l) of order 'm' [11].

$$\begin{aligned} \cos\delta_M &= \cos\left(\alpha\sin\left(\omega t\right)\right) \\ &= J_0\left(\alpha\right) + 2\sum_{m=1}^{\infty} J_{2m}\left(\alpha\right)\cos\left(2m\omega t\right) \end{aligned} \tag{31}$$

$$\begin{aligned} \sin\delta_M &= \sin\left(\alpha\sin\left(\omega t\right)\right) \\ &= 2\sum_{m=0}^{\infty} J_{2m+1}\left(\alpha\right)\sin\left[\left(2m+1\right)\omega t\right] \end{aligned} \tag{32}$$

For the case of $J_0(\alpha) = 0$, I is achieved by adjusting the voltage applied to the PEM

$$I = I_{dc} + I_{\omega} 2J_1\left(\alpha\right)\sin\omega t + I_{2\omega} 2J_2\left(\alpha\right)\cos\left(2\omega t\right) + \cdots \tag{33}$$

By comparing equation (33) with equation (28) the intensity coefficients are defined. The normalized frequency terms then are established to be

$$I_{dc} \sim \left(\left|r_x\right|^2 + \left|r_y\right|^2\right)/2 \tag{34}$$

$$I_{\omega}/I_{dc} \sim \mathrm{Im}\left(\Delta r/r\right) - \delta_W \tag{35}$$

$$I_{2\omega}/I_{dc} \sim \mathrm{Re}\left(\Delta r/r\right) \tag{36}$$

Therefore, I_{dc} processes the reflectivity. The imaginary part of $\Delta r/r$ is dignified at frequency ω and the intensity is found to be sensitive to the first-order window, δ_W, strain.

CONCLUSION

The optical surface probe of RAS is a non-destructive technique for the study of metal-on-metal and semiconductor growth. RAS is an experimental optical method using visible light and is particularly attractive since it is not restricted to vacuum environments. RAS uses the basic components of ellipsometry. This review presents an overview of the RAS technique.

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