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Analysis of bulk sample photothermal response by time resolved ellipsometry

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Abstract: Ellipsometry is an optical technique where the polarisation of light beam reflected by a sample is analysed in order to determine its optical properties. Combining ellipsometry measurements with a pulsed excitation, we have been able to monitor in time the variation of the complex refractive index $n - ik$ of a strongly absorbing bulk sample and to reach the absolute surface temperature history subsequent to the excitation in sub-microsecond time-scale.

1. INTRODUCTION

Thermal-wave techniques are used for obtaining informations about the thermal and/or optical and/or electronic properties variations in semiconductors samples. The most used of these techniques is modulated (or pulsed) photorefectance which has been successfully applied to the study of doping, implantation dose, surface contamination, and carrier diffusion. However all the informations contained in the optical reflectivity are not collected from a measurement of reflected absolute intensity; information regarding the polarisation state of the reflected electric field is lost. This additional information can be obtained by an ellipsometry experiment.

Two years ago we presented ellipsometry under pulsed or modulated excitation as a new photothermal probe [1]. We have transformed a commercial spectroscopic ellipsometer SOPRA ES4G and developed an experimental methodology to realise the measurement of dynamical changes of the ellipsometric parameters induced by an external light excitation [1]. The potential and the sensitivity of the method have been illustrated by measurements performed in time and frequency domains for bulk samples [2] as well as multilayered samples [3].

In the present contribution, we report a quantitative analysis of experimental results obtained for a Ge bulk sample under pulsed excitation. We demonstrate that the absolute temperature time evolution can be directly determined from the photoinduced variations of the real and imaginary parts of the refractive index provided by pulsed ellipsometry experiments.

2. EXPERIMENTAL DETAILS

In a conventional ellipsometry experiment, a polarized probe beam is reflected at the sample surface and the change of the polarization state is measured. This measurement is performed through the determination of a couple of parameters (ψ , Δ) or ($\tan\psi$, $\cos\Delta$) defined by :

$$\frac{r_p}{r_s} = \tan\psi \cdot e^{i\Delta}$$

where r_p and r_s are the reflection coefficients for the electric vector components parallel and perpendicular to the plane of incidence.

The SOPRA ellipsometer is a rotating polarizer - sample - analyser system that we have adapted in order to perform measurements under unstationary excitation. The experimental procedure is first to fix the azimuth of the polarizer (usually 45°), then successively to move the azimuth of the analyser at four positions (with intervals of 45°) for which the static and the photoinduced signals are measured. From these data the ellipsometric parameters $\tan\psi$, $\cos\Delta$ and their variations $\delta\tan\psi$ and $\delta\cos\Delta$ are determined.

For pulsed excitation, a low power laser ($\approx 1\text{mW}$) is used as probe source to provide a convenient signal to noise ratio. The reflected beam is detected by a fast silicon photodiode (100MHz bandwidth). An amplifier and a digital oscilloscope working as a signal averager are used for signal processing. The sensitivity to the relative signal variation is of the order of 10^{-5} with 10MHz detection bandwidth and an average over 1000 shots. As excitation source, the second harmonic of a YAG laser at $0.53\mu\text{m}$ has been used (pulse of about 3mJ and 30ns). The spot diameters on the sample surface were 1.2mm for the pump beam and $50\mu\text{m}$ for the probe.

3. RESULTS AND DISCUSSION

We have studied a thick wafer of Ge covered by a native oxide layer. Complex refractive index of Ge, index and thickness ($\approx 5.3\text{nm}$) of the layer are determined by spectroellipsometry.

The photon energy of the excitation pulse (2.4eV) is three times greater than the value of the band gap of Ge (0.7eV), most of the energy is then converted into heat, and carrier effects on the optical properties can be neglected. Indeed as a result of photon absorption, the conduction (valence) band of the sample contains a high density of electrons (holes) with a wide temperature distribution. Electrons (holes) located high (low) in the conduction (valence) band rapidly relax within that band by phonon emission increasing the lattice temperature.

Fig. 1 : Time evolution of :

- (a), (b) the ellipsometric parameters variations induced by a pulsed light excitation ($0.53\mu\text{m}$) for the Ge sample, for two wavelengths : $0.63\mu\text{m}$ (a), $0.67\mu\text{m}$ (b), at the incidence angle of 75° , averaged over 1000 scans.
- (c), (d) the real and imaginary parts variations of the refractive index calculated from (a) and (b) respectively.

Fig. 1 shows the ellipsometric parameters variations measured with two probe sources: a $0.63\mu\text{m}$ HeNe laser and a $0.67\mu\text{m}$ diode laser. Let us notice that for $0.67\mu\text{m}$, $\delta\cos\Delta$ is lower than 10^{-4} at any time. Every curve exhibits a $t^{-1/2}$ behaviour (except at very short times) as expected for the transient thermal response of a bulk sample surface heated by the instantaneous absorption of a light pulse. Indeed the pump beam diameter on the sample surface is greater than the thermal penetration depth for the time scale of the measurements, hence heat diffusion can be assumed to be one dimensional. In this assumption, the surface temperature history of a thick homogeneous sample after absorption at time $t = 0$ of a pulsed energy density Q (Dirac heat flux) is [4]:

$$\delta T(t) = \frac{Q}{\rho c \sqrt{\pi D t}} \quad (1)$$

ρ is the density, c specific heat, and D the thermal diffusion coefficient. In actual configuration, the finite pulse duration affects the beginning of the temperature history. Balageas et al [4] show that formula (1) can be used with a negligible error for $t > 10\tau$ (τ duration of the pulse).

Using the same numerical treatment which leads to the refractive index $n - ik$ of a substrate under a known layer from $\text{tg}\Psi$ and $\cos\Delta$, and assuming that this layer is unaffected by the heating and that the substrate changes are uniform through the optical penetration depth ($0.8\mu\text{m}$) of the probe beam, the δn and δk changes have been calculated from the $\delta\text{tg}\Psi$ and $\delta\cos\Delta$ curves and are shown in Fig. 1(c) and (d).

In order to prove the temperature dependence of the photoinduced index variations, spectroscopic ellipsometric measurements of $\text{tg}\Psi$ and $\cos\Delta$ were achieved in the $0.4 - 0.8\mu\text{m}$ wavelength range at different temperatures from 15 to 108°C . The sample was heated with an electrical resistance plate and the temperature was measured by a thermocouple in contact with the front face of the sample. The incidence 75° near the Brewster angle of Ge was chosen in order to increase the measurement sensitivity. From these ellipsometry data, n and k of the substrate are calculated with the assumption that the oxide layer remains the same during the heating. The spectra are shown in Fig. 2.

Fig. 2 : Spectra of the real and imaginary parts of the refractive index for the Ge sample at different temperatures deduced from spectroscopic ellipsometry measurements : (-----) 15°C , (— · — · —) 38°C , (————) 66°C , (— · — · —) 108°C .

The real and imaginary parts n and k of the refractive index are found to vary linearly in the measured temperature range. From these results $\delta n/\delta T$ and $\delta k/\delta T$ for wavelengths $0.63\mu\text{m}$ and $0.67\mu\text{m}$ are deduced:

λ (μm)	$\frac{\delta n}{\delta T} \times 10^4 (^\circ\text{C}^{-1})$	$\frac{\delta k}{\delta T} \times 10^4 (^\circ\text{C}^{-1})$
0.63	14	33
0.67	18	16

Using these coefficients, we have been able from the four results of Fig. 1(c) and (d) (n and k for the two wavelengths) to deduce the surface temperature history subsequent to the excitation. The temperature values presented in Fig.3 are in a 20% range and in a rather good agreement with the theoretical temperature decay curve shown in the figure and calculated with the following conditions: a square wave pulse of 50ns duration and an energy density of $0.02\text{J}/\text{cm}^2$ (see ref [4]) which are close to the experimental ones.

Fig. 3 : Temperature time evolution of the sample surface calculated from the variation of the real and imaginary parts of the refractive index shown in Fig. 1 (c),(d) induced by a pulsed excitation for two probe wavelengths $0.63\mu\text{m}$ and $0.67\mu\text{m}$. Full curve : theoretical thermal decay of a bulk sample surface heated by a 50ns square-wave pulse.

4. CONCLUSION

Finally the absolute temperature decay history of an opaque bulk sample, even covered by a thin oxide transparent layer, irradiated by a pulsed light excitation can be monitored by a time resolved ellipsometry experiment. The temperature dependence of refractive index needed for the calculation are determined by conventional spectroscopic ellipsometry measurement. Reciprocally, if the temperature can be determined for a given probe wavelength, the temperature coefficient of the optical constants n and k can be reached at any other probe wavelengths for which the sample is opaque and the pulsed ellipsometry measurement can be performed.

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