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APSUSC-35562; No. of Pages 6
ARTICLE IN PRESS
Applied Surface Science xxx (2017) xxx–xxx
Contents lists available at ScienceDirect
Applied Surface Science
journal homepage: www.elsevier.com/locate/apsusc

Full Length Article

Ultrafast in-situ null-ellipsometry for studying pulsed laser – Silicon surface interactions

J. Csontos [a,b], Z. Toth [c], Z. Pápa [a,b], B. Gábor [a], M. Füle [b,d], B. Gilicze [d], J. Budai [a,b,*]

[a] University of Szeged, Department of Optics and Quantum Electronics, H-6720 Szeged, Dóm tér 9., Hungary
[b] ELI-HU Non-Profit Ltd, Szeged, Hungary
[c] University of Szeged, Department of Oral Biology and Experimental Dentistry, H-6720 Szeged, Tisza Lajos krt. 64-66, Hungary
[d] University of Szeged, Department of Experimental Physics, H-6720 Szeged, Dóm tér 9., Hungary

ARTICLE INFO

Article history:
Received 1 August 2016
Received in revised form 17 March 2017
Accepted 21 March 2017
Available online xxx

Keywords:
Null-ellipsometry
Pump and probe method
Laser excitation
Silicon
Two-temperature model

ABSTRACT

The measurement of transient optical properties due to pulsed laser excitation allows better understanding of the nature of laser induced processes. Conventional ellipsometry is not capable of following changes in the femto-, pico- or nanosecond timescale. In this work, the pump and probe technique is combined with a single wavelength null-ellipsometry. This enabled us to follow the optical changes of silicon due to sub-ps laser pulse irradiation with ps time resolution. The combination of the 496 nm probe pulses with a Polarizer – Compensator – Sample – Analyzer (PCSA) configuration imaging null-ellipsometer provided Ψ and Δ ellipsometric angles of silicon irradiated with 248 nm pump pulses. Different laser intensities and delay times between the probe and pump pulses are used in the experiments. It is shown that besides thermal effects, the in depth free charge carrier distribution and their electron-phonon relaxation time has to be taken into account in the frame of the two-temperature model for satisfactory interpretation of the experimental results.

1. Introduction

The investigation of ultrafast laser pulse – material interaction is essential from fundamental and practical aspects. Changes in the electron states due to femto- to picosecond laser pulses lead to changes both in reflection and absorption. So far, majority of the works presented in-situ measurement of transient reflection applying pump and probe methods [1–7]. However, absorption or imaginary part of dielectric function can not be measured with reflection experiments. There were a few attempts to introduce ellipsometric setups for femto- or picosecond pump and probe measurements. Firstly, Auston et al. investigated the time evolution of optically generated free carriers in intrinsic germanium with a pump-probe ellipsometric arrangement. The transmission changes of the sample due to picosecond pulses have been used to estimate the density of the plasma forming after the absorption of an intense excitation pulse [8]. Choo et al. studied Ge-Si alloys and determined changes in dielectric function due to low intensity pump pulse irradiations at 620 nm wavelength and 100 fs pulse duration [9]. More recently, Boschini et al. developed a spectroscopic pump-probe ellipsometric setup which utilizes supercontinuum generation with 60 fs duration. The possibility to measure dielectric tensor and magneto-optical properties in CrO_2 was demonstrated [10]. Rapp et al. used a pump-probe ellipsometer to determine the transient complex refractive index of molybdenum below and above ablation threshold with the wavelengths of 1056 nm and 528 nm, with pulse lengths of 680 fs and 540 fs, respectively [11]. Although silicon was widely used in the case of pump and probe experiments, only reflection and transmission measurements were performed in polycrystalline silicon films by Bergner [12].

For bulk single crystalline samples, transmission measurements can not be carried out in the visible range of light, therefore we aimed to use pump and probe ellipsometric setup in reflection mode to investigate transients of silicon's optical properties. Pump and probe ellipsometric investigation of silicon with ultrashort laser pulses below ablation threshold is challenging since the optical changes are minor compared e.g. to germanium [9]. In this work we present a pump and probe setup combined with single wavelength null-ellipsometry to monitor the excitation of Si surface. By measuring the polarization change, our pump and probe null-ellipsometer enables us to follow the optical changes due to relaxation processes after a sub-ps laser pulse irradiation with a ps time resolution. The pump energy density was set to such low values which allowed investigating the processes in the single crys-

* Corresponding author.
E-mail address: jbudai@titan.physx.u-szeged.hu (J. Budai).

http://dx.doi.org/10.1016/j.apsusc.2017.03.186
0169-4332/

Please cite this article in press as: J. Csontos, et al., Ultrafast in-situ null-ellipsometry for studying pulsed laser – Silicon surface interactions, Appl. Surf. Sci. (2017), http://dx.doi.org/10.1016/j.apsusc.2017.03.186

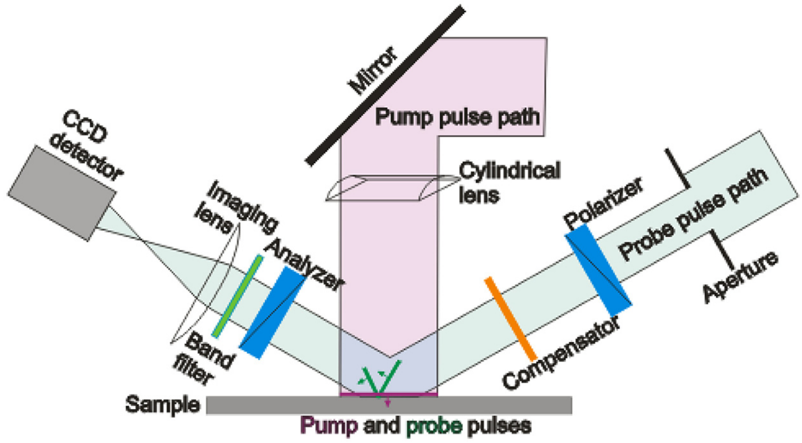

Fig. 1. Schematic drawing of the pump-probe null-ellipsometry setup. The moment is shown, when pump pulse hits the surface, while a part of the probe pulse has already been reflected from the untreated surface. The later reflecting parts probe the pump laser excited area.

talline phase, i.e. melting and ablation were excluded from our study.

2. Experimental methods

Laser pulses from a distributed feedback dye laser (wavelength: 496 nm) are used for probe pulses. A part from these pulses are frequency doubled and amplified in a KrF excimer laser system (wavelength: 248 nm, pulse length: 480 fs) [13]. The UV pulses are used as pump pulses, and focused to a line by a fused silica cylindrical lens onto a ⟨100⟩ orientation, boron-doped silicon surface. The spot size of the pump pulse on the sample is 35 mm × 0.7 mm. According to the measured pulse energies and spot size average fluence values applied in the experiments are 12 and 17 mJ/cm^2, which is more than one order of magnitude less than the melting threshold (170 mJ/cm^2) of the silicon [3].

Probe pulses leaving the laser are led through an optical delay line containing a reflecting prism mounted onto a translation stage. This element allows the fine adjustment of the delay between the pump and the probe pulse by moving the prism along the optical path. The accuracy of the delay setting is ~30 fs, which is determined by the accuracy of the micrometer screw of the translation stage. With this configuration 8 different time delays were set in the range of 2–146 ps.

An optical fiber (core diameter: 105 μm, NA: 0.22) is applied to transport the probe pulses from the delay line to the arm of a goniometer, where the probe beam is collimated. As shown in Fig. 1, the probe beam is directed onto the UV excited line region of the silicon surface. The angle of incidence is 67°. After reflection the probe pulse is imaged onto the sensor of a CCD camera (Thorlabs DCU224 M) which is synchronized with the laser pulses. In the detector arm, a band pass filter for 496 nm is set to eliminate the scattered UV light from the pump pulse.

By introducing polarizing elements into the probing beam line that corresponds to a Polarizer – Compensator – Sample – Analyzer (PCSA) configuration (Fig. 1), null-ellipsometry can be performed which can provide transient Ψ and Δ angle values. Crystal polarizers are used as the polarizer and analyzer (extinction ratio: 1:100 000 at 632 nm), while a quartz quarter wave plate is applied as a compensator. When the nulling condition is set for bare Si, the onset of the excitation by the UV pulse can be clearly seen by the appearance of the increasing intensity in the image. The zero delay is determined by this intensity increase. The null azimuth angle of the analyzer and polarizer is calibrated by measuring extinction of p-polarized probe light reflected from a glass plate close to its Brewster angle.

3. Data reduction

The RMS fluctuation of the pulse energy of the lasers is ~10%, which makes necessary the continuous monitoring of the pump and probe pulse energies. For this, parts of the UV and VIS laser pulses are coupled out with a fused silica and a glass plate, respectively. The UV light is focused onto a fluorescing glass plate, and the fluorescent light is coupled into an optical fiber. A second fiber was applied to transfer the out-coupled part of the probe beam. The transmitted reference signals are imaged onto the CCD detector with the help of a concave mirror. In this way, the image of the CCD contained not only the pump and probe images, but also their reference energy density signals (see Fig. 2). Calibration of pump energies was performed by measuring simultaneously the pulse energy above the silicon sample and the average pixel intensity within the reference signal.

Based on the CCD images the steps of the image processing are the followings: i) determination of the dark level of the image ii) subtraction of the dark level from all image elements (from the area marked with B in Fig. 2 a)) iii) determination of the pump energies using its reference signal (R UV in Fig. 2 a)) and iv) normalizing the probe image to the probe reference signal (R VIS in Fig. 2 a)). Step iii) allows selecting the images taken at the same pump energy densities and thus the pump energy density dependence of the ellipsometric data can be followed.

Nulling condition for the probe pulse can be set by finding the accurate polarizer and analyzer azimuth angles (A and P), that minimize the pixel intensities corresponding to a given sample area in the CCD image. Fig. 2 presents two images when the nulling condition is set to a) the pumped area and b) the intact area. To determine more accurately the Ψ and Δ angles, instead of direct nulling of the image, a set of A and P angle positions were chosen close to estimated nulling condition. For each A-P setting ~80 images were recorded in the vicinity of the values corresponding to the estimated nulling conditions in ±8° range. After recording the ~80 images intact sample surface was illuminated again. By analyzing the recorded images probe intensity maps as a function of A-P angles were plotted. The maps ensure accurate determination of the nulling condition for the UV laser pumped areas and of the corresponding Ψ and Δ values. Such maps are plotted in Fig. 3, where pixel intensity is mapped as a function of A-P a) for an untreated silicon wafer and b) for a laser treated one when time delay was set to be 25 ps.