ELLIPSOMETRY AT THZ FREQUENCIES: NEW APPROACHES FOR METROLOGY AND METAMATERIAL-BASED SENSING

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The precise measurement of electromagnetic material properties at THz frequencies is essential for the development of increasingly advanced THz optical systems and a prerequisite for the design and manufacturing of optical elements for this spectral range. The exploration of novel physical phenomena observed in artificially structured metamaterials and the application thereof is of interest due to its relevance for the design and fabrication of novel THz optical elements and sensors. Metamaterials have attracted continued interest for almost two decades due to their unique electromagnetic properties, which can differ substantially from their constituents and often do not even exist in naturally occurring materials.

We have demonstrated that although being orders of magnitude smaller than the probing wavelength, metamaterials composed of highly-ordered 3-dimensional metal nanostructures exhibit a strong anisotropic optical response at THz frequencies. I will discuss how these interesting optical properties can be used for sensing of minute target material quantities in the THz spectral range. In addition, I will focus on a novel avenue for the fabrication of THz metamaterials using stereolithographic techniques. For the THz spectral range, where spatial resolutions in the range of several um are sufficient to create subwavelength metamaterial building blocks, stereolithography-based additive fabrication might offer a readily accessible approach which is so far unrealized. Here, metamaterials composed of spatially coherent methacrylates wires will be shown as an example. Our observations demonstrate that stereolithography may provide an alternative avenue to the fabrication of metamaterials for the terahertz spectral range and may allow tailoring of the polarizability and anisotropy of the host material by design.

Keywords: THz ellipsometry, Metamaterials, THz sensing

Ellipsometric study of the TiO_2-based decorative coatings produced using magnetron sputtering technique at industrial conditions

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Titanium dioxide is a non-toxic, biocompatible and chemically stable material and is used as white pigment in paints and some high-tech applications including electrochromic devices, dye-sensitized solar cells (DSSC), photocatalysis, optical filters and antireflection coatings.

Moreover, due to the expressive colors of titanium oxides and oxynitrides deposited/formed on metal or metal-coated glass, these compounds can be applied as a decorative material for architecture, automotive industry, electronics and jewelry. In general, titanium dioxide is a non-absorbing material in the visible spectral range, thus the color effect is caused by the optical interference of light in the thin dielectric film.

In the last few years, the different TiO_2-based decorative coatings were produced using a industrial scale magnetron sputtering line and were analyzed at Institute of Mathematics and Physics UTP. The decorative coatings were deposited on glass, aluminum, Ti-coated aluminum, stainless steel, Ti-coated stainless steel, ceramic tiles as well as on polymer substrates. The presented results will be a summary of the last few years of research. These results show that for the opaque systems the color of sample can be related to thickness of the TiO_2 layer. The thickness of Ti film in TiO_2/Ti/glass samples is associated with their transparency, whereas in TiO_2/Ti/Al specimens - with saturation of color.

Keywords: TiO_2, decorative coatings, intereference, thin films

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High Speed Spectroscopic Ellipsometry Technique for On-Line Monitoring in Large Area Thin Layer Production

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Macro imaging spectroscopic ellipsometers has been developed for high speed mapping of large area thin layer coated substrates. Non-contact and non-destructive characterization techniques based on spectroscopic ellipsometry are widely used by the photovoltaic and semiconductor industry for process or quality control in production. The commercialization of large area thin film technologies and the related increasing surfaces lead to many key problems such as reduced efficiency caused by multiple non-uniformities of the layer properties over the entire panel or wafer resulting from the technological steps of individual layer components. For these reasons, thin layer properties must be mapped by an „in-line" or „in-situ" method avoiding mistakes resulting from layer inhomogeneities. Scanning ellipsometry methods based on the conventional narrow beam spectroscopic measurements provide high accuracy but suffer from long mapping times as the polarization state of the reflected beam must be detected. **Expanded beam ellipsometry** was developed to measure rapidly the polarization state changes after reflection from bigger surfaces. Our instruments use non-collimated illumination with a special light source and an optical arrangement allowing multiple angles of incidence [1]. New prototypes have been prepared for spectroscopic measurements that provide a **line image of spectroscopic ellipsometry** data with a lateral resolution of ~10 mm over the range of 350-1000 nm [2-4]. Ellipsometric information of large areas can be collected a **couple of 10 times faster** compared to scanning methods. Prototypes have been built for structures with nominal widths of 300-450-600-900 mm (SiO_2, ZnO/Mo, NiSi) **on rigid substrates**. Thin layers (ZnO/a-Si:H/Ag) on plastic foil substrates were also investigated **in roll to roll operation**. Measurements and results of different structures are presented.

We can demonstrate spectroscopic ellipsometry mapping measurements over 1800 points in ~1 min traverse of a 300-450 mm diameter Si-wafer (demonstration in clean room environment, see Fig. 1) or 600x1200 mm solar panel.

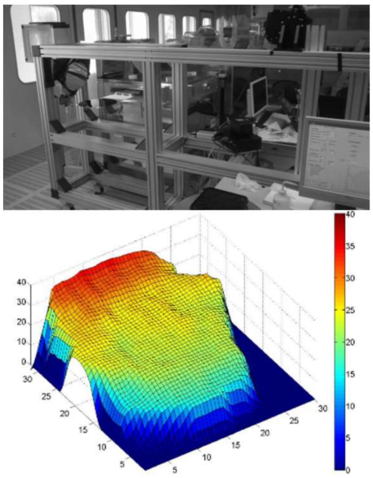

Fig. 1. Installed mapping device and thickness-map of a NiSi covered 300 mm diam. Si-wafer (complete annealing at 350° C).

Acknowledgements: Support from OTKA K115852, M-ERA-NET 117847, and NVKP_16-1-2016-0014 projects is gratefully acknowledged.

Keywords: Mapping; ellipsometry, thin film

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Characterization of dielectric films on glass for the semiconductor 3D packaging process

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Characterization of film homogeneity and mechanical strength of the film itself are important aspects in engineering materials for back-end of line (BEOL) semiconductor processes [1]. It is a challenge to optimize such materials both for low density and for a mechanically strong structure. In terms of device integration, thickness and composition change must be minimized during high temperature process steps [2]. These factors are especially pronounced in case of bonded wafers for 3D device structures. Such stacked plates consist of Si and glass wafers molded to each other.

The bow of the constituing glass wafer can be engineered by depositing stressful SiO2 which can be modified depending on the conditions of the deposition steps, type of precursors, or type of deposition tool. Since glass exhibits half of the modulus of Si, it can easily respond to a stressful film deposited on it. With the SiO2 stress as well as variation in thickness, the amount of bow can be calculated to be engineered on the glass that would even out the total bow when these are used as carriers.

Such characteristic samples are measured, tendencies and conclusions are drawn and will be presented.

Keywords: Si wafer; porous low-k; spectroscopic ellipsometry, ellipsometric porosimetry

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HIGH-SPEED SPRECTROSCOPIC ELIIPSOMETRY AND ITS APPLICATIONS

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As a comprehensive manufacturer of metrology tools and deposition tools, ULVAC developed an innovative high-speed spectroscopic ellipsometer for many deposition applications, such as PVD, CVD, ALD and others.

This novel spectroscopic ellipsometry can measure the thickness and optical constants of thin films at a dramatically fast speed. Its data acquisition time is as short as 10ms. It does not require any active components for polarization-control, such as a rotating compensator or an electro-optical modulator.

It created great opportunities for new applications of the spectroscopic ellipsometry in which the compactness, the simplicity and the rapid response are extremely important. It can be integrated into the deposition tool and successfully measure thin films in-situ and ex-situ. Obviously, those from PVD, CVD and ALD are some promising applications for this novel spectroscopic ellipsometry.

This presentation describes the principle, system configuration and creative efforts on developing a series of high-speed spectroscopic ellipsometers. Some of the novel applications will be also introduced, such as the PVD, CVD, ALD, EUV, OLED, MEMS and some measurement data of thin films from the semiconductor, flat panel display, and other industries.

Keywords: Spectroscopic Ellipsometry; High-order Retarder; High-Speed

FS-TIME-RESOLVED SPECTROSCOPIC ELLIPSOMETRY

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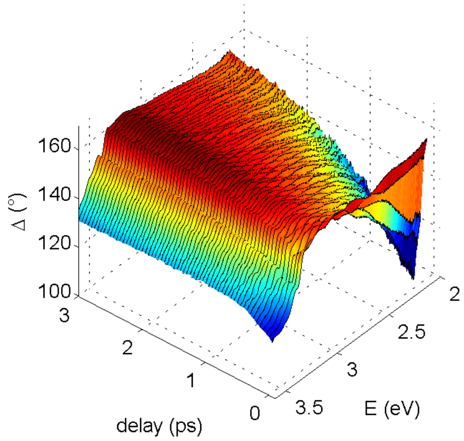

Fig. 1. Transient ellipsometric angle Δ for a *c*-plane oriented ZnO single crystal showing the response of plasma absorption as well the damping of the excitonic and higher transitions.

Implementing time-resolved ellipsometry to study effects of free charge carriers induced by pulsed laser excitation has been tried first in the 1970's [1]. However, for a long time, experimental limitations and single wavelength probes limited the ellipsometric approaches to indirect investigations of pump-induced charge carriers in the near-infrared. Today's possibilities to generate continuum white light pulses allow even spectroscopic ellipsometry with time resolution in the fs-range. Still, a number of experimental challenges as spectral fluctuations and group velocity delay remain. We demonstrate a new setup based on an amplified Ti:sapphire laser which is used to generate continuum white light in a CaF_2 crystal as probe while its fundamental, doubled or tripled frequency are applied as pump. Time-resolved spectroscopic ellipsometry measurements have been carried out in *polarizer-sample-compensator-analyzer* configuration by obtaining transient reflectance-difference spectra at different compensator azimuth angles.

We demonstrate first measurements of ZnO single crystals, films and resonators as well as Ge single crystals. In Ge, depending on the pump wavelength, band filling effects or renormalization are observed at the E1 and E1+Δ1 transitions which take place at the L point in the momentum space. In ZnO, the dynamics of UV-excited hot charge carriers can be monitored by their induced plasma absorption as well as damping of optical transitions of excitons and above the band gap. Carrier thermalization, recombination and lattice heating can be distinguished as processes on different spectral ranges and time scales ranging from fs to ns. A particular challenge to model transient ellipsometry data arises from the depth gradient of the density of excited charge carriers due to the pump laser absorption.

Keywords: dynamics; pump-probe; hot charge carriers

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ULTRAFAST IN-SITU ELLIPSOMETRY FOR STUDYING INTERACTIONS OF LASER PULSES AND MATERIAL SURFACES

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Monitoring the changes in material properties during pulsed laser excitation opens a pathway towards better understanding of the fundamental nature of laser induced processes. On one hand ultrafast single wavelength ellipsometry is an appropriate candidate for following such changes, since it provides more information than a conventional pump and probe reflection measurement. On the other hand the retrieval of the data is model based which necessitates the involvement of a broad spectral range.

Very recently, we demonstrated a unique, ultrafast pump-probe null-ellipsometry method, capable of assessing rapid changes of temperature and charge density upon femtosecond laser pulse irradiation [1]. To overcome the limitation due to the single wavelength of the probe pulse we rebuilt our measurement setup. Now it uses synchronized white light generated by femtosecond laser pulse (part of the pump) focused into a bulk glass plate, and operates as a quasi-rotating compensator ellipsometer.

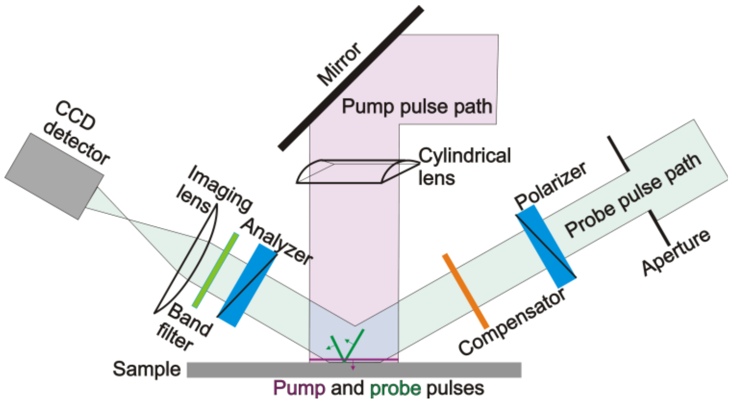

Fig. 1. Experimental setup

In this presentation, details of both experimental setups will be given. Advantages and disadvantages will be discussed through the example of silicon irradiated with fs laser pulses.

We believe that our results are not only interesting for the ellipsometric community but also represent an important contribution for applications in material science where ultrafast excitation of charge carriers is involved.

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

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ULTRAFAST ELLIPSOMETRY OF LASER-INDUCED PHASE TRANSITIONS IN MATTER

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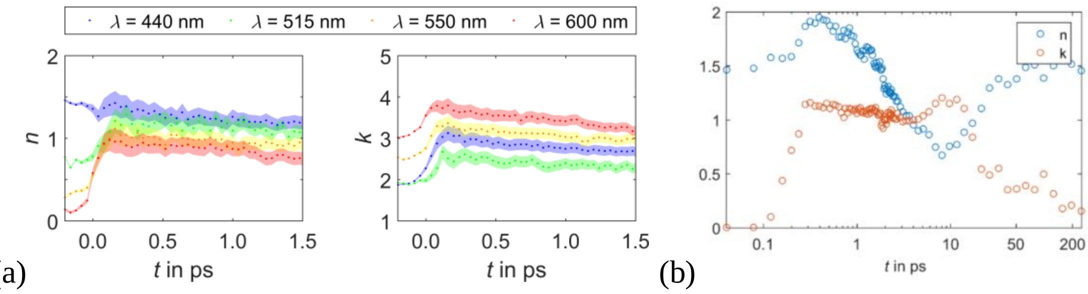
Irradiating matter with ultrafast laser radiation induces transient states with physical and chemical properties far away from any equilibrium. Thin gold film on a glass substrate (thickness d_{Au} = 200 nm, roughness R_a =3 nm) and bulk PMMA have been excited by single pulsed femtosecond laser radiation (τ_{H} = 40 fs) and the temporal evolution of the complex refractive index was detected spatially- and spectroscopically-resolved by a self-developed pump-probe ellipsometer. The gold film excited at the pump wavelength λ = 800 nm exhibits for a fluence of 1 J/cm² (below gentle ablation [1]) and a probe wavelength λ = 440, 515, 550, and 600 nm a complex dynamics of the refractive index n and the extinction coefficient k (Fig.1 a). Apparently, the refractive index converges for all investigated wavelengths about 100 fs after excitation to a value close to 1 describing the heating of the electron system, in which the refractive index for λ = 440 nm demonstrates a slight decrease and for λ = 515, 550 and 600 nm the refractive index increases strongly compared to the value at rest. Thereafter, a slight decrease of the refractive index is obtained, depicting the beginning of the cooling of the electrons [2].

PMMA, an organic dielectric material, is transparent for radiation at small intensities in the VIS and IR spectral range, but at large intensities (I =3·10^{13} W/cm²) the radiation induces field ionization generating free electrons [3, 4]. Exciting above ablation threshold with a single laser pulse (λ = 800 nm) a strong increase in the refractive index and extinction coefficient is induced within 100 fs probing at the wavelength λ = 440 nm. The extinction coefficient lasts about 20 ps, then decreasing again, depicting the lifetime of the laser-induced free electrons.

Keywords: ultrafast; transient state; ablation



(a) (b)

Fig. 1. Refractive index and extinction coefficient as function of time after excitation with single pulsed laser radiation (τ_{H} = 40 fs, λ = 800 nm) (a) of gold for different probe wavelengths, and (b) of PMMA for the probe wavelength λ = 440 nm.

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ELLIPSOMETRY IN THE ERA OF INDUSTRY 4.0

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In times of Internet of Things and Industry 4.0 production lines require more automation than ever. The operators influence on the resulting product is pushed back to minimize failure. Multiple diagnostic tools are used, spinning a tight monitoring network throughout the production cycle. The vast amount of data produced by the variety of diagnostic tools must be organized, crosslinked and evaluated to maximize yield and product quality.

The data are typically processed by a host software specifically designed for the production line. The communication between the host and the large variety of equipment is performed using SECS (SEMI Equipment Communications Standard) or more specifically SECS/GEM (General Model for Communications and Control of Manufacturing Equipment). The SECS/GEM defines standard commands, replies and events allowing the host to control and monitor the manufacturing equipment.

Being a standard in microelectronics for a while now, the SECS/GEM also becomes more and more popular in other industrial fields like photovoltaics and optoelectronics. An ellipsometer designed to be placed in such a demanding environment must match these requirements as well. In addition, precision, repeatability and measurement speed are traditional challenges for production control.

Typical requirements for equipment performance, software compatibility and maintenance are presented on actual cases of major optoelectronic manufactures. Special emphasis is put on the SECS/GEM compliance. Some examples are shown of how SECS/GEM can be used either as local remote control of the measurement software or even by complete remote access via the company network. The later even allows to perform measurements with a standard tablet.

Keywords: SECS/GEM; Production;

References

USE OF P-SPLINES FOR THE DIELECTRIC FUNCTION REPRESENTATION IN THE INTERPRETATION OF SPECTROSCOPIC ELLIPSOMETRY DATA

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About ten years ago Johs and Hale developed the Kramers–Kronig consistent B-spline formulation for the dielectric function modeling in spectroscopic ellipsometry data analysis [1]. But the actual performance of B-spline parameterization strongly depends on the number of knots (and, in general, their locations) used to describe the dielectric function. It is intuitively easy to conceive that fewer knots in the considered wavelength range may not be enough to fit all essential spectral features (underfitting). On the contrary, increasing the number of knots beyond the optimal value can result in significant overfitting of the experimental data, i.e., leads to an unrealistic result with measured noise reproduction. In previous papers [2,3], we used well-established information criteria approach for choosing a suitable B-spline model with optimal number of knots to balance fidelity to the measured data and complexity of the fit.

An alternative method to avoid overfitting by B-spline parameterization is to use one of the variants of smoothing splines called “penalized splines” or “P-splines” which include a penalty on B-spline coefficients. The amount of penalty, which based on the second- (or higher-) order finite differences of the coefficients of adjacent B-splines, is easily controlled by non-negative smoothing (or penalty) parameter α. Selection of $\alpha = 0$ leads to ordinary least-squares fit of the ellipsometric data with the B-spline model for the dielectric function. The larger the value of the smoothing parameter α, the smoother the result one can get. We will focus on application of the P-spline approach to dielectric function representation in spectroscopic ellipsometry data analysis.

The P-spline approach offers a number of advantages over well-established B-spline parameterization. First of all, it typically uses an equidistant knot arrangement which simplifies the construction of the roughness penalties and makes it computationally efficient. Since P-splines possess the “power of the penalty” property [4], the number and location of knots are no longer crucial, as long as there is a minimum knot number to capture all significant spatial variability of the data curves. We demonstrate the proposed approach by real-data case studies.

Keywords: Spectroscopic ellipsometry; Data analysis; Optical modeling; Dielectric function; Parameterization; Penalized splines; Optical metrology

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