

# Monday Afternoon, October 30, 2017

## Spectroscopic Ellipsometry Focus Topic

Room: 9 - Session EL+AS+EM-MoA

### Spectroscopic Ellipsometry: Novel Applications and Theoretical Approaches

Moderator: Maria Losurdo, CNR-NANOTEC

#### 1:40pm EL+AS+EM-MoA1 Temperature Dependent Mueller Matrix Measurements of Magnetised Ni near the Curie Temperature, *Farzin Abadizaman, S. Zollner*, New Mexico State University

The temperature dependence of the optical constants of the magnetized bulk Ni demonstrates an anomaly near the Curie temperature. We investigate this anomaly by taking a measurement of the temperature dependent Mueller Matrix (MM). Using spectroscopic ellipsometry at an energy 1.96 eV, the MM measurement was taken from 350 K to 500 K with 10 K steps, and from 500 K to 670 K with 1 K steps, and from 670 K to 730 K with 10 K steps.

In order to distinguish the anisotropic properties of the magnetized Ni from the windows effects, three samples (Ge, Ni, and SiO<sub>2</sub> on Si) were measured inside the cryostat in the energy range from 1 eV to 5 eV with 0.1 eV steps. The results show that the anisotropic elements of the MM behave in the same fashion for all samples, except for element M<sub>24</sub>, which depends on the sample.

The MM data of magnetized Ni indicate very small changes in the anisotropic portion of the MM compared to the windows effect. The authors believe that these changes are due to the magneto-optical Kerr effect. However, since a rotating-analyzer ellipsometer was used, the last row of the MM is absent and a complete MM measurement needs to be performed to find the magneto-optical Kerr effect in the other anisotropic elements too.

However, very large decreases in the isotropic MM elements were found near the Curie temperature. This means, the changes in optical constants near T<sub>c</sub> are due to the on-diagonal Drude part of the dielectric tensor, which can be explained by s- to d-band electron-phonon scattering above and below T<sub>c</sub>. These changes are absent when decreasing the temperature and for unmagnetized Ni.

#### 2:00pm EL+AS+EM-MoA2 Ellipsometry Based Observation of Material Ordering Process in Holography, *Hao Jiang, H. Peng, Y. Liao, S. Liu*, Huazhong University of Science and Technology, China

Ordered materials with superior performance have been constantly pursued. Nevertheless, the direct, precise and non-destructive observation of the ordering process, which is especially critical for continuous manufacturing, remains a formidable challenge. Herein, we introduce Mueller matrix ellipsometry (MME) as a nondestructive method to quantitatively observe the material ordering process during holography. This non-destructive observation directly offers the exact width, refractive index, nanoparticle weight fraction and volume fraction in each constructive (bright) or destructive (dark) interference area, which is impossible to be implemented using other existing techniques. Meanwhile, the width of dark region is observed to decrease while the width of bright region increases with an augmentation of the holographic recording time, distinct from previous assumption. More importantly, an apparent diffusion coefficient of  $1.3 \times 10^{-15} \text{ m}^2 \text{ s}^{-1}$  is determined on the basis of recording time-dependent grating structure observation, which is 3 orders of magnitude lower than the Stokes-Einstein prediction. The distinct diffusion coefficient is regarded to be the result of competition between the forth diffusion driven by the chemical reaction and backward diffusion arising from nanoparticle concentration gradient. This novel protocol is envisioned to pave the way for precisely and non-destructively understanding the ordered structure formation in electronics, photonics, photovoltaics, biomaterials and other disciplines.

#### 2:20pm EL+AS+EM-MoA3 Coherence in Polarimetry and Ellipsometry: Synthesizing Mueller Matrices in an Ellipsometer, *Oriol Arteaga*, Departament de Física Aplicada, Universitat de Barcelona, Spain **INVITED**

Interference phenomena are ubiquitous in optics and are the basis of the industry behind thin film optical coatings or thin film thickness measurements with ellipsometry. The coherence of polarized beams was experimentally understood in 1819 when Arago and Fresnel derived the four laws governing the interference of polarized light. While spectroscopic ellipsometry provides information based on the position and number of interference oscillations in thin films, typically one has no macroscopic control of the beams that coherently superpose.

In this work we present a polarimetric analysis of an analogue of Young's double slit experiment that allows merging beams in a well-controlled manner. The experiment is analyzed with a new formalism that is useful to

describe optical coherence and polarization and that shows that the superposition of two macroscopically distinguishable beams can be an effective method to experimentally synthesize Mueller matrices with on-demand polarization properties. This offers the opportunity of working with "synthesized" optical elements that behave just like "real" ones. We will discuss how this method can have a practical application in the construction of ellipsometers or polarimeters.

#### 3:00pm EL+AS+EM-MoA5 Femtosecond Spectroscopic Ellipsometry on Optoelectronic Materials and Photonic Structures, *Mateusz Rebarz, S.J. Espinoza*, ELI Beamlines - Czech Academy of Science, Czech Republic, *S. Richter, O. Herrfurth, R. Schmidt-Grund*, Universität Leipzig, Germany, *J. Andreasson*, Chalmers University of Technology, Sweden, *S. Zollner*, New Mexico State University

The ongoing progress in miniaturization and operational rates of electronic and optoelectronic devices obliges materials scientists to deeply understand the dynamics of the carriers upon external electromagnetic stimulus in very short time scale. Some phenomena such as scattering of electrons and phonons as well as recombination processes can be as short as tens of femtoseconds. All these processes affect the temporal and local dielectric constants and determine many operational parameters of the devices. In this work, we report on recent progress in developing a spectroscopic ellipsometer for characterization of ultrafast dynamic changes of dielectric properties in materials and photonic structures technologically relevant in optoelectronics. Our time-resolved ellipsometer is based on pump-probe technique and offers monitoring the time evolution of the dielectric properties in range 0-5 ns with time resolution ~100 fs in broadband spectral range (340-750 nm).

We present here the results of the measurements performed on Ge samples in comparison with theoretical predictions. Ultrafast phenomena such as excitation, relaxation and diffusion of charge carriers, band-gap renormalization and excitons screening will be discussed. In addition we report on the first data obtained from a ZnO-based planar microcavity especially on the temporal evolution of the microcavity modes. The time evolution of the ellipsometric parameters in the spectral range around the exciton-polariton mode will be discussed. We observed that such modes disappear upon the excitation, possibly due to screening of the excitons and re-appear after a few hundreds of femtoseconds as blueshifted modes. The investigation of the short-time dynamics of such modes can stimulate new theoretical approaches for the description of exciton-polariton systems.

#### 3:20pm EL+AS+EM-MoA6 Temperature Dependence of the Dielectric Function and Interband Critical Points of Bulk Germanium, *Carola Emminger, N. Samarasingha, F. Abadizaman, N.S. Fernando, S. Zollner*, New Mexico State University

Exploration of the optical properties of bulk germanium (Ge) is necessary for the advancement of Ge technology. In fact, many of the Ge applications depend on the dielectric function ( $\epsilon$ ), which is directly related to the electronic band structure. Here we investigate the effect of temperature on the optical properties and interband critical points (CPs), primarily the E<sub>0</sub> and E<sub>0</sub>+ $\Delta_0$  critical points of bulk Ge in the temperature range from 10 to 738 K using spectroscopic ellipsometry at 70° angle of incidence. The data was taken in two parts, in the near IR region and in the UV region, which provides data from 0.5 to 6.2 eV. The low temperature environment was created in a UHV cryostat with liquid helium and nitrogen as cryogens. To reduce the thickness of the native GeO<sub>2</sub> layer, the Ge sample was cleaned using ultra-pure water, isopropanol, an ultrasonic bath and ozone cleaning. It was possible to reduce the oxide thickness to about 11 Å at room temperature.

The authors used a two-phase model (GeO<sub>2</sub> layer/Ge substrate) and a parametric oscillator model with a set of adjustable parameters to extract the real and imaginary parts of the complex dielectric function of the bulk Ge for the whole temperature range. To investigate this temperature dependence of the CP parameters (threshold energy, broadening and phase angle) further, we also compared the second derivative  $d^2\epsilon/d\omega^2$  of the dielectric function with analytical line shapes.

The temperature has a significant influence on both the real and imaginary parts of the complex dielectric function of bulk Ge. This temperature dependent  $\epsilon$  can be explained by a Bose-Einstein occupation factor. Due to the electron-phonon interaction, we find a temperature dependent red shift (shift to lower energies) of the E<sub>0</sub> and E<sub>0</sub>+ $\Delta_0$  critical point energies. The temperature independent spin orbit splitting  $\Delta_0$  is found to be 286 meV. A similar effect has been seen in the E<sub>1</sub>, E<sub>1</sub>+ $\Delta_1$ , E<sub>0</sub>' and E<sub>2</sub> CP energies. These CPs are broadened and shifted to the lower energies with increasing temperature.

4:00pm **EL+AS+EM-MoA8 VUV Magneto-Optical Transient Ellipsometer**, *Shirly Espinoza, J. Andreasson*, Institute of Physics ASCR, Czech Republic

This talk is about the ELIps instrument, an instrument that combines three advanced techniques of ellipsometry: VUV ellipsometry, Transient (Pump-probe) ellipsometry, and Magneto-optical ellipsometry [1].

The ELIps instrument allows users to work in a very wide range of energies from 1 eV to 40 eV. For measurements in the 1 eV to 6.5 eV range, it uses a transmission polarizer and analyzer combination. Meanwhile, for measurements in the 6.5–40 eV range, it uses a triple-reflection polarizer and analyzer combination. All the components are contained within a single UHV chamber designed with several additional ports to support future upgrades. For time resolved measurements in the VUV range the instrument will be used together with a high intensity High Harmonics Generation (HHG) source.

Time-resolved transient measurements of the optical properties of materials can be performed in the range of a few femtoseconds to nanoseconds. Initial experiments with transient absorption and transient ellipsometry in the NIR-UV range is being performed on proton-conducting materials for solid oxide fuel cell applications.

Additionally, ELIps is equipped with a Helmholtz coil, which allows the studies of the transverse magneto-optical Kerr effect on the sample, and with a cryostat for measurements at different temperatures.

Where is this instrument located? ELIps is in Prague, Czech Republic, at the European Extreme Light Infrastructure Beamlines (ELI Beamlines), a user facility project that will hold some of the most intense lasers in the world. It will open to the public on January 2018.

#### References

[1] S. Espinoza, G. Neuber, C. D. Brooks, B. Besner, M. Hashemi, M. Rübhausen and J. Andreasson. 2017. User oriented end-station on VUV pump-probe magneto-optical ellipsometry at ELI beamlines. *Applied Surface Science*. Published online. doi:10.1016/j.apsusc.2017.02.005

#### Acknowledgements

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4:20pm **EL+AS+EM-MoA9 Infrared Ellipsometric Spectroscopy of Hg<sub>1-x</sub>Cd<sub>x</sub>Te Bulk Samples**, *Yanqing Gao*, Shanghai Institute of Technical Physics, Chinese Academy of Sciences, China

**The infrared spectroscopic ellipsometry of Hg<sub>1-x</sub>Cd<sub>x</sub>Te (x=0.195–0.37) bulk samples with different compositions were measured. The refractive index n and extinction coefficient k were obtained. An obvious refractive index enhancement effect was observed in the refractive index spectra for each composition. The energy position of the maximal refractive index value equals approximately that of the band gap. With the decrease of the component, the refractive index increased and the peak position shifted to the low energy direction, which consistented with the absorption edge. The refractive index n at E<sub>g</sub> changed linearly with the composition x.**

4:40pm **EL+AS+EM-MoA10 Infrared Ellipsometry Study of the Photo-generated Charge Carriers at the (001) and (110) Surfaces of SrTiO<sub>3</sub> Crystals and the Interface of Corresponding LaAlO<sub>3</sub>/SrTiO<sub>3</sub> Heterostructures**, *Meghdad Yazdi-Rizi, P. Marsik, B. Mallett*, University of Fribourg, Switzerland, *K. Sen, A. Cerreta*, University of Fribourg, A. Dubroka, Masaryk University, *M. Scigaj, F. Sánchez, G. Herranz*, Institut de Ciència de Materials de Barcelona, *C. Bernhard*, University of Fribourg, Switzerland

With infrared (IR) ellipsometry and DC resistance measurements we investigated the photo-doping at the (001) and (110) surfaces of SrTiO<sub>3</sub> (STO) single crystals and at the corresponding interfaces of LaAlO<sub>3</sub>/SrTiO<sub>3</sub> (LAO/STO) heterostructures. In the bare STO crystals we find that the photo-generated charge carriers, which accumulate near the (001) surface, have a similar depth profile and sheet carrier concentration as the confined electrons that were previously observed in LAO/STO (001) heterostructures. A large fraction of these photo-generated charge carriers persist at low temperature at the STO (001) surface even after the UV light has been switched off again. These persistent charge carriers seem to originate from oxygen vacancies that are trapped at the structural domain boundaries which develop below the so-called antiferrodistortive transition at  $T^* = 105$  K. This is most evident from a corresponding photo-doping study of the DC transport in STO (110) crystals for which the concentration of these domain boundaries can be modified by applying a weak uniaxial stress. The oxygen vacancies and their trapping by defects are also the source of the electrons that are confined to the interface of LAO/STO (110) heterostructures which likely do not have a

polar discontinuity as in LAO/STO (001). In the former, the trapping and clustering of the oxygen vacancies also has a strong influence on the anisotropy of the charge carrier mobility. We show that this anisotropy can be readily varied and even inverted by various means, such as a gentle thermal treatment, UV irradiation, or even a weak uniaxial stress. Our experiments suggest that extended defects, which develop over long time periods (of weeks to months), can strongly influence the response of the confined charge carriers at the LAO/STO (110) interface.

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