Spectroscopic Ellipsometry Focus Topic
Room 104C - Session EL+AS+EM+MI+TF-FrM

Spectroscopic Ellipsometry: Novel Applications and Theoretical Approaches

Moderators: Morten Kildemo, Norwegian University of Science and Technology, Nikolas Podraza, University of Toledo

8:20am **EL+AS+EM+MI+TF-FrM1 Magnetoopticalproperties of Metals, Half-Metals, and Garnets Probed by Vector-Magneto-Optical Generalized Ellipsometry**, ***Heidemarie Schmidt***, Technische Universität Chemnitz, Nano-Spintronics Group, Germany **INVITED**

Magnetotransport measurements are a standard technique for the electrical characterization of single layers on insulating substrates. However, magnetotransport measurements require electrical contacts and known current paths, which excludes application to multilayer stacks. Motivated by the recent development of fast Mueller matrix ellipsometers, we have set-up a vector magnetooptical generalized ellipsometer (VMOGE) with an 0.4 T octupole magnet [1] and have investigated magnetooptical response of a single layers and multilayer stacks in a magnetic field of arbitrary orientation and magnitude up to 0.4 T at room temperature. We assume that the off-diagonal element of the magnetooptical dielectric tensor of every magnetizable layer in the multilayer stack is a product of the magnetic field independent and wavelength dependent complex magnetooptical coupling constant and the magnetic field dependent and wavelength independent magnetization of the layer. As an example, the complex magnetooptical coupling constant of nominally 10, 20, and 30 nm thick ferromagnetic Ni films obtained from modelling corresponding VMOGE data is discussed. It was challenging to identify the magnetization direction of Ni films from different sets of magnetic field dependent Mueller matrix elements [2]. In the future knowledge of complex magnetooptical coupling constant of all magnetizable materials in a multilayer stack will allow for modelling and optimizing the magnetooptical response of given stack. As a second example, the modelled complex magnetooptical coupling constant of capped, ferromagnetic Fe, Ni20Fe80, Co, Ni80Fe20, and Ni thin films on ZnO substrates is discussed and related with the spin-dependent electronic bandstructure of given weakly correlated, magnetizable materials [3]. For this comparison the experimental complex off-diagonal elements of the magnetooptical dielectric tensor have been converted into theoretical complex off-diagonal elements of magnetooptical conductivity tensor. Finally, the experimental magnetooptical response of strongly correlated, magnetizable materials [4], e.g. half-metals and garnets, is presented and as an outlook development of new theoretical frameworks for calculating the bandstructure of such strongly correlated, magnetizable materials for a comparison with experiment is motivated. [1] K. M. Mok, N. Du, H. Schmidt, Rev. Sci. Instrum. 82 (2011) 033112; [2] K.M. Mok, C. Scarlat, G. J. Kovács, L. Li, V. Zviagin, J. McCord, M. Helm, H. Schmidt, J. Appl. Phys. 110 (2011)123110; [3] K.M. Mok, G. J. Kovács, J. McCord, L. Li, M. Helm, H. Schmidt, Phys. Rev. B 84 (2011) 094413; [4] G. Kotliar and D. Vollhardt, Physics Today 57 (2004) 53

9:00am **EL+AS+EM+MI+TF-FrM3 *In Situ* Terahertz Optical Hall Effect Measurements of Ambient Doping Effects in Epitaxial Graphene**, *S. Knight,* University of Nebraska-Lincoln*; C. Bouhafs, N. Armakavicius, P. Kühne, V. Stanishev, R. Yakimova,* Linköping University, Sweden*; S. Wimer, M. Schubert,* University of Nebraska-Lincoln*; V. Darakchieva,* Linköping University, Sweden*;* ***Tino Hofmann,*** University of North Carolina at Charlotte

Recently, the cavity-enhanced THz optical Hall effect (THz-OHE) has been demonstrated as non-contact method to obtain free charge carrier properties using low-field permanent magnets [1,2]. A tunable, externally-coupled cavity is used to enhance the THz-OHE signal which allows the accurate determination of a sample's free charge carrier properties even at low magnetic fields. In this work we take advantage of this approach by integrating the permanent magnet into a gas flow cell. We demonstrate for the first time the application of the cavity-enhanced THz-OHE for the *in-situ* characterization of free charge carrier properties of monolayer graphene on Si-face 4H-SiC as a function of ambient conditions. The experiments were performed using a new rotating-analyzer THz ellipsometer at Linköping University. Upon changing the CO_2, H_2O, and O_2 concentration in the cell, large variations in both free charge carrier sheet density N_S and mobility μ are observed for the *n*-type graphene. The lowest N_S was found for the as-grown sample with N_S =5.9(1) × 10^{11} cm^{-2} where μ =2507(57) cm^2/Vs. The highest N_S was found after purging the sample with nitrogen for 6 hours with N_S =2.43(4) × 10^{12} cm^{-2} where μ =1604(23) cm^2/Vs. These significant changes are attributed to a redox-reaction of oxygen and water at the graphene surface which results in the extraction of electrons from graphene [3]. This will be discussed in detail in our presentation. We further observe that this doping mechanism is only partially reversible at room temperature upon removal of oxygen, carbon dioxide, and water by purging the cell with nitrogen. In conclusion, we demonstrate *in-situ* THz-OHE as a new and powerful technique to determine ambient-dependent doping mechanisms which is illustrated here using monolayer epitaxial graphene on Si-face 4H-SiC.

[1] S. Knight, S. Schöche, V. Darakchieva, P. Kühne, J.-F. Carlin, N. Grandjean, C. M. Herzinger, M. Schubert, and T. Hofmann, Opt. Lett. **40**, 2688 (2015).

[2] P. Kühne, C.M. Herzinger, M. Schubert, J.A. Woollam, and T. Hofmann, Rev. Sci. Instrum. **85**, 071301 (2014).

[3] A.N. Sidorov, K. Gaskill, M.B. Nardelli, J.L. Tedesco, R.L. Myers-Ward, C.R. Eddy Jr., T. Jayasekera, K.W. Kim, R. Jayasingha, A. Sherehiy, R. Stallard, and G.U. Sumanasekera, J. Appl. Phys. **111**, 113706 (2012).

9:20am **EL+AS+EM+MI+TF-FrM4 Excitons at Interfaces in Ellipsometric Spectra**, ***Nuwanjula Samarasingha****, C. Rodriguez, J.M. Moya, N.S. Fernando, S. Zollner,* New Mexico State University*; P. Ponath, K. Kormondy, A. Demkov,* University of Texas at Austin*; D. Pal, A. Mathur, A. Singh, S. Dutta, J. Singhal, S. Chattopadhyay,* Indian Institute of Technology Indore, India

The presence of excitonic features in the optical constants and ellipsometry spectra of bulk semiconductors and insulators has been known for many years. In Si, Ge, and GaAs, the E_1 critical points are strongly enhanced by two-dimensional excitons, even at room temperature. Three-dimensional excitons have been seen in ellipsometry spectra for GaP and Ge. Excitons also influence the dielectric function of $SrTiO_3$. An exciton is an electron-hole pair bound by the Coulomb interaction, with properties similar to a hydrogen atom. The influence of excitonic absorption on the dielectric function was described by Tanguy.

In a thin epitaxial layer (with a thickness below or near the Bohr radius) on a substrate with a different band gap, the wave functions of the electron and hole are strongly modified. In a thin type-I quantum well, consisting of a narrow-gap semiconductor grown on a large-gap substrate, both the electron and the hole are confined, which leads to an increase in the dipole overlap matrix element. Therefore, the dominant absorption peak at 4.2 eV is larger in a 20 nm thick $SrTiO_3$ layer on a $LaAlO_3$ substrate than in bulk $SrTiO_3$. (The band gap of $LaAlO_3$ is larger than that of $SrTiO_3$.)

On the other hand, in a staggered type-II quantum well, either the electron is confined, or the hole, but not both. Therefore, the overlap dipole matrix element (and thus the excitonic absorption) is strongly reduced, because one quasiparticle resides in the quantum well and the other one in the substrate. If a $SrTiO_3$ layer is grown on Si or Ge, the valence band maximum occurs in the substrate, while the conduction band offset is very small. Therefore, the exciton wave function is delocalized (deconfined), which reduces the dipole overlap matrix element. Therefore, the real and imaginary part of ε of thin $SrTiO_3$ layers on Si or Ge are much smaller than in the bulk and decrease monotonically with decreasing thickness. A similar effect can be seen for thin ZnO layers on Si as a function of thickness.

The dielectric function of $SrTiO_3$ is not only affected by layer thickness. A very thick polycrystalline $SrTiO_3$ layer on Si has a much lower dielectric function than a single-crystalline $SrTiO_3$ substrate. In this case, we speculate that the magnitude of the dielectric function is related to other Tanguy parameters, perhaps the excitonic binding energy or the exciton decay rate (broadening). To investigate this further, we will perform temperature-dependent ellipsometry measurements on bulk zinc blende GaP, which has a much simpler band structure than wurtzite ZnO or the correlated metal oxide $SrTiO_3$, but shows similar excitonic effects.

9:40am **EL+AS+EM+MI+TF-FrM5 Infrared and Visible Dielectric Properties of $(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.35}$**, ***Jacqueline Cooke****, N.T. Nunley, T. Willett-Gies, S. Zollner,* New Mexico State University

Using spectroscopic ellipsometry, we determined the dielectric function of LSAT, from the mid-IR to the deep UV (0.03 to 6.5 eV). LSAT is an acronym for the chemical formula $(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.35}$, equivalent to $(La_{0.3}Sr_{0.7})(Al_{0.65}Ta_{0.35})O_3$. LSAT is a common substrate for epitaxial growth of complex metal oxides. Precise knowledge of the optical constants is useful to investigate the properties of epitaxial films grown on LSAT. We also investigated the band gap and the infrared-active phonons. Czochralski-grown LSAT wafers with (001) surface orientation were obtained commercially (MTI Corp., Richmond, CA). Single-side polished wafers were used for spectroscopic ellipsometry and two-side polished wafers with 0.5 mm thickness for transmission. Between 0.8 and 6.5 eV, we measured the normal-incidence transmission and the ellipsometric angles from 60° to 80° incidence in 2° steps on a J.A. Woollam variable angle of incidence ellipsometer with a computer-controlled Berek waveplate compensator. We also measured in the mid-IR on a rotating compensator FTIR ellipsometer. Transmission measurements show a steep rise of the absorption coefficient (α) between 4.6 and 4.8 eV, where LSAT becomes opaque. Fitting the ellipsometry data with a model containing two Tauc-Lorentz oscillators and 19 Å surface roughness thickness yields an excellent fit to the data. The Tauc gap is 4.9 eV and the high-frequency dielectric constant ε_∞= 4.0. Plotting α^2 versus photon energy yields a direct band gap of 5.8 eV. An Urbach tail extends towards lower energies. The resulting dielectric function is in agreement with previous ellipsometry and minimum-deviation prism measurements. The mid-IR dielectric function shows four ε_2 peaks due to TO phonon absorption. The loss function shows four LO peaks. A fifth TO phonon was seen at 155 cm^{-1} in far-IR ellipsometry. An ideal ABO3 perovskite has only three IR-active TO phonons. FCC ordering on the B-site as in (Sr_2AlTaO_3) adds a fourth phonon. We argue that the TO phonons at 155 and 283 cm^{-1} are vibrations of the tetrahedra against the La/Sr sublattices, respectively (mode splitting due to disorder). On the other hand, the 397 and 442 cm^{-1} modes are B-O bending modes due ordering in the Al/Ta sublattice. Finally, a B-O stretch mode at 664 cm^{-1} and broad two-phonon absorption at 765 cm^{-1} are also found. Fitting the spectra with a factorized TO/LO model yields better results than a sum of Lorentzians, because the individual TO/LO pairs are not well separated. The presence of FCC ordering was also confirmed with x-ray diffraction. We will also discuss temperature dependent ellipsometry and transmission measurements.

10:00am **EL+AS+EM+MI+TF-FrM6 A New Constant of Product of Electronic Scattering Time and Resistivity in Thin Silver Refractive Index Calculation from Ellipsometry and Resistivity Measurements***, **Guowen Ding**, C. Clavero, D. Schweigert, M. Le,* Intermolecular, Inc.

The optical and electrical response of metal thin films is highly affected by electronic scattering with the interfaces and defects. We are able to successfully model the electrical resistivity and near infrared (IR) optical response using a thickness dependent electronic scattering time. We investigated Ag films thickness in the range of 3 nm to 74 nm and determined that the product of electronic scattering time (τ) and resistivity (ρ) remains constant regardless of the thickness ($\tau \times \rho = C$), with a value of 59±2 μΩ cm·fs for Ag films. As a result, determining the constant C for a given thin film will allow to calculate the propreties of the film over a large range of wavelengths while limiting the number of measurements.Our findings enable us to develop a theoretical framework to determine the optical response of metal thin films in the near IR by using single wavelength ellipsometer measurements. An excellent agreement is found between experimental measurements and predicted values. We first reported this constantτ $\tau \times \rho =C$ for silver, and we posit that such constant concept could be applied for other conducting films. Application of the model presented here will allow rapid characterization of the IR optical response of metal thin films, with important application in a broad spectrum of fundamental and industrial applications, including optical coatings, low-emissivity windows and semiconductor industry.

10:20am **EL+AS+EM+MI+TF-FrM7 Realization of an *In Situ* Mueller-matrix Imaging Ellipsometer for the Real Time Observation of Surface Properties in an Ultra-high Vacuum EUV Facility***, **Pim Muilwijk**, N.B. Koster, F.T. Molkenboer, E. Sligte, te, A.F. Deutz, P. Walle, van der,* TNO Technical Sciences, Netherlands

TNO is realizing EUV Beamline 2 (EBL2), a facility to investigate the effects of Extreme Ultra-Violet (EUV) radiation on surfaces to enable future EUV High Volume Manufacturing (HVM) production. In this facility, samples with sizes up to 152x152x20 mm (6" EUV reticles) can be exposed to EUV radiation of up to 500W equivalent at intermediate focus (IF) under realistic environmental conditions and analyzed by in-situ ellipsometry and XPS. EBL2 consists of EUV source, automated handling system, beam line and an exposure chamber with an in-situ dual wavelength Mueller-matrix imaging ellipsometer.

Light from the dual wavelength light source (405 & 640nm) enters the exposure chamber through a polarizer, configurable retarder and a vacuum window producing a defined polarization state. After reflecting off of the sample, the light exits the exposure chamber through a vacuum window, configurable retarder and polarizer. The sample position is imaged on two camera's, one for each wavelength. By combining all combinations of 4 polarization illumination states with 4 analyser states the full Mueller matrix of the sample can be recovered.

Calibration is performed in-situ with two insertable polarizers and two different calibration samples. The calibration procedure does not require prior knowledge of the polarizer orientation nor of the calibration samples.

This presentation will focus on the design and realization of the ellipsometer and will also touch upon the process of interpreting the data.

EBL2 will be publicly accessible as a test facility for EUV lithography related research after qualification, which is expected to be finished end of Q1 2017.

10:40am **EL+AS+EM+MI+TF-FrM8 Conducting, Semi-Conducting and Insulating 2D-Materials Characterized by Spectroscopic Imaging Ellipsometry***, **Matthias Duwe**, S. Funke,* Accurion GmbH, Germany*; U. Wurstbauer,* Technical University of Munich, Germany*; A. Matkovic,* University of Belgrade, Serbia*; A. Green,* SUNY College of Nanoscale Science and Engineering*; A. Molina-Mendoza,* Universidad Autonoma de Madrid, Spain*; A. Castellanos-Gomez,* IMDEA Nanoscience, Spain*; P.H. Thiesen,* Accurion GmbH, Germany

Finding thin-film flakes of 2D-materials after the fabrication and identifying their layer thicknesses often is a challenging and time-consuming task. Here, we present various applications of spectroscopic imaging ellipsometry (SIE) to a variety of conducting, semi-conducting, and insulating 2D-Materials such as graphene, molybdenum disulfide (MoS_2), hexagonal boron nitride, and black phosphorus. As a combination of polarization-contrast microscopy and spectroscopic ellipsometry, SIE measurements localize microscopic flakes of the 2D-materials, yield the samples' optical dispersion functions, and determine the layer thicknesses.

Matkovic et al. [1] characterized monolayers of graphene by SIE, and they obtained the optical dispersion by Fano-resonance modelling. Using this dispersion, SIE offers a straightforward search for and identification of few-layer graphene flakes on various opaque or transparent substrates. As this flake search uses ellipsometric measurements, it depends far less on the used substrate compared to e.g. conventional light-microscopy. In a similar procedure, SIE identified monolayers of insulating hexagonal boron nitride, and it yielded the material's optical properties.

SIE measurements on MoS_2 revealed the repercussion of the used substrate [2]. Ellipsometric contrast micrographs showed the lateral variation of the optical parameters for a structured flake. Spectroscopic measurements of the ellipsometric values (Ψ & Δ) obtained from selected regions of interest on the flake yielded the optical dispersion for the in-plane and out-of-plane components of the dielectric function in the visible spectral range.

Finally, we will present imaging Mueller-matrix ellipsometry (IMME) for the characterization of thin-film flakes of the semi-conducting 2D-material black phosphorus. In contrast to MoS_2, black phosphorus also features an optical in-plane anisotropy. IMME-micrographs easily reveal this anisotropy as the Mueller matrix's off-diagonal blocks deviate from zero. By performing spectroscopic Mueller-Matrix mapping and rotational Mueller-matrix measurements combined with atomic force microscopy, we obtained the flake's layer thickness, the orientations of the optical axes, and the material's optical properties in the visible spectral range.

[1] A. Matković, A. Beltaos, M. Milićević, U. Ralević, B. Vasić, D. Jovanović, and R. Gajić, *Spectroscopic imaging ellipsometry and Fano resonance modeling of graphene*, *J. Appl. Phys.*, **112** 123523, (2012)

[2] S.Funke, E. Parzinger, B. Miller, P. H. Thiesen, A.W. Holleitner, U. Wurstbauer, *Imaging Ellipsometry of Mono- to Multilayer ofMoS_2 on Tranparent Sapphire Substrate*, Manuscript in preperation

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