Spectroscopic Ellipsometry Focus Topic
Room A212 - Session EL+EM-WeA

Spectroscopic Ellipsometry: Novel Applications and Theoretical Approaches

Moderators: Vanya Darakchieva, Linkoping University, Sweden, Nikolas Podraza, University of Toledo

2:20pm EL+EM-WeA-1 Optical Hall Effect in the Multi-valley Semiconductor Te-doped GaSb, *Farzin Abadizaman, C Emminger,* New Mexico State University*; S Knight,* University of Nebraska-Lincoln*; M Schubert,* University of Nebraska-Lincoln, Linköping University, Sweden, Leibniz Institute of Polymer Research Dresden, Germany*; S Zollner,* New Mexico State University

The authors conducted optical Hall effect (OHE) measurements on Te-doped GaSb (n-type) at room temperature in the far-infrared between 30 cm^{-1} and 700 cm^{-1} at magnetic fields of ±7 T and 0 T. The measurements were performed at an angle of incidence of 45 and a resolution of 2 cm^{-1}. The complex dielectric functions and Mueller Matrix (MM) elements were determined from spectroscopic ellipsometry at 0 T in the range of 300 cm^{-1} to 8000 cm^{-1} using an FTIR-VASE ellipsometer and from 30 cm^{-1} to 700 cm^{-1} using the FIR ellipsometer. Using a sum of a Lorentzian oscillator and two Drude terms, the experimental data at zero magnetic field were modeled. From the Lorentzian term, we found the transverse optical (TO) phonon energy at 226 cm^{-1} and the longitudinal optical (LO) phonon energy at 237 cm^{-1}.

Although GaSb is a direct band gap semiconductor, a calculation of the electron concentration indicates that at T = 300 K and a total electron density below 10^{18} cm^{-3}, the majority of carriers are located at the L-valley (67%) while the Γ-valley contains only 33% of the carriers. This implies that in the absence of the magnetic field, two Drude terms are needed to model the data. The surfaces of constant energy at the L-point in the Brillouin zone form eight half-ellipsoids at L, which are characterized by their anisotropic masses. However, due to the symmetry, the valleys at this point are two by two equivalent, which leads to the total number of four valleys. In the absence of a magnetic field, the contribution of all eight half-ellipsoids in the L-valley is reduced to only one Drude term, where the effective mass is the harmonic average of the transverse and longitudinal masses. As the magnetic field is turned on, each ellipsoid contributes to the anisotropic dielectric tensor, which, depending on the effective mass tensor, contributes differently to the total dielectric tensor. Therefore, in the presence of a magnetic field, the data is modeled by the sum of a Lorentzian, a Drude tensor at the Γ-valley and four Drude tensors at the L-valley.

2:40pm EL+EM-WeA-2 Study of the Temperature-dependent Optical Constants of Noble Metals based on High Temperature Spectroscopic Ellipsometry, *Jiamin Liu, H Jiang, S Liu,* Huazhong University of Science and Technology, China

Noble metals have been widely used in thermo-plasmonics field, such as thermo- photovoltaics, heat-assisted magnetic recording and photothermal therapy, thus studying the temperature-dependent optical constants of these metals are crucial for both understanding the temperature effects on optical properties and providing essential data for the plasmonic simulations.

In this work, a high temperature spectroscopic ellipsometry covering the spectral range of 200-1000nm has been built, which is able to measure the ellipsometric parameters of samples when temperature is varying from 300K to 1200K. The noble metallic samples are heated at a mixing atmosphere of 5% H_2 and 95% Ar to avoid the possible thermal oxidation. An oscillator- parametrization regression method based on the Drude-dual-TaucLorentz-Lorentz model and the B-spline model has been proposed to determine the optical constants and the roughness of the heated noble metals. Both the optical constants and the electronics parameters of noble metals heated below 900K have a temperature-dependency similar to the recently reported results. Taking the gold film as an example, the DC resistivity is increasing from 2.273×10^{-6} to 2.414×10^{-6} Ω·cm with the temperature increasing from 300K to 800K, while the electron relaxation time is decreasing from 20.787 to 9.021fs. Additionally, it has been noticed the first absorption peak near 2.7eV first increases and then decreases, while the second absorption peak near 3.7eV shows the opposite characteristics with the temperature increasing from 300K to 800K. Besides, the optical constants of Au film heated above 900K has some

similarities to that of SnTe, which might be caused by the combined effect from the possible formation of Au-Si binary phase and the possible transition of vertical columnar grains to granular grains.

3:00pm EL+EM-WeA-3 Optical Monitor for the Attitude Tracking using Polarimetry, *Song Zhang, H Gu, H Jiang, S Liu,* Huazhong University of Science and Technology, China

The attitude angles are important parameters describing the motion of the object. In the fields of precision manufacturing, robotics control, navigation of the aircraft, the accurate and real-time measurements of the attitude angles (yaw angle, pitch angle and roll angle) are very important. Due the advantages of non-contact, low cost, non-destructive and high precision, the optical methods have been popular used for measuring the attitude of the object.

In our work, we present a novel optical monitor for the attitude tracking. The proposed method utilizes the principle that polarized light incident in different directions into the birefringent crystals can produce different phase modulations. Then, the attitude angle of the object attached with a birefringent crystal can be obtained by measuring the phase change. The optical monitor is based on the division-of-amplitude polarimetry with a time resolution of several nanoseconds, which is capable of monitoring the changes in all the attitude angles simultaneously. In order to verify the correctness and the performance of the optical monitor, we performed real-time measurement experiments on the attitude angles of a zero-order quarter-wave plate and a multi-order half-wave plate. The roll angle is continuously changed within the range of 0 ~ 360°, while the pitch angle and yaw angle are varied within ±7° and ±40° respectively. The results show that not only the attitude angles, but also the angular velocities and the accelerations of the roll angle, can be extracted, and the errors of all attitude angles is less than 0.5°.

3:20pm EL+EM-WeA-4 New Progress on the Channeled Spectroscopic Ellipsometry and its Applications, *Gai Chin,* ULVAC Inc., Japan

This presentation describes a novel method for the spectroscopic measurement of the state of polarization of light. A pair of thick birefringent retarders is incorporated into the spectroscopic polarimeter, so the generated channeled spectrum is composed of three quasi-cosinusoidal components carrying the information about the state of polarization of the light that is being measured. Fourier inversion of the channeled spectrum provides the significant parameters for determination of the spectrally resolved Stokes parameters of light. No mechanical movable components for polarization control or active devices for polarization modulation are used, and all the Stokes parameters can be determined at once from only the single spectrum.

The channeled spectroscopic ellipsometry is a snapshot method for the spectrally-resolved polarization analysis. A pair of high-order retarders are utilized to generate a channeled spectrum carrying information about the wavelength-dependent multiple parameters of polarization of light. This method has a feature that it requires no mechanical or active components for polarization-control, such as a rotating compensator and electro-optic modulator.

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 measured 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 our new progress on some key technologies for enhancing the performance of this channeled spectroscopic ellipsometry by system configuration, data analysis and other creative efforts on developing a series of new high-speed spectroscopic ellipsometers. Some novel applications will be also introduced, such as the PVD, CVD, ALD, EUV, OLED, MEMS and some excellent measurement data of thin films from the semiconductor, flat panel display and other industries.

4:20pm EL+EM-WeA-7 The Physics of Low Symmetry Metal Oxides: Applications of Ellipsometry, *Alyssa Mock,* U.S. Naval Research Laboratory; *S Knight, M Hilfiker,* University of Nebraska-Lincoln; *V Darakchieva, A Papamichail,* Linkoping University, Sweden; *R Korlacki,* University of Nebraska-Lincoln; *M Tadjer,* U.S. Naval Research Laboratory; *Z Galazka, G Wagner,* Leibniz-Institut für Kristallzüchtung, Germany; *N Blumenschein,* North Carolina State University; *A Kuramata,* Novel Crystal Technology, Inc., Japan; *K Goto, H Murakami, Y Kumagai,* Tokyo University of Agriculture and Technology, Japan; *M Higashiwaki,* National Institute of Information and Communications Technology, Japan; *A Mauze, Y Zhang, J Speck,* University of California Santa Barbara; *M Schubert,* University of Nebraska-Lincoln, Linköping University, Sweden, Leibniz Institute of Polymer Research Dresden, Germany **INVITED**

We discuss the analysis of the dielectric function tensor for monoclinic metal oxides obtained from generalized spectroscopic ellipsometry. We investigate the potential high-power device material gallium oxide and derive dispersions of transverse, longitudinal and plasmon coupled modes [M. Schubert *et al.*, Phys. Rev. B 93, 125209 (1-18) (2016); Editors' Suggestion] and the band-to-band transitions and excitons along with their eigenvectors [A. Mock *et al.*, Phys. Rev. B 96, 245205 (1-12) (2017)]. Additionally, we show that this technique can fully explain the unusual ordering of optical phonon mode pairs which is observed in beta-Ga2O3 [M. Schubert, A. Mock et al. Phys. Rev. B 99, 041201(R) (2019)] as well as their dependency on free charge carrier concentrations. [M. Schubert, A. Mock et al. Appl. Phys. Lett. 114, 102102 (2019) – Editor's Pick]. We apply this technique also for the identification of transverse and longitudinal phonons in scintillator material cadmium tungstate [A. Mock *et al.*, Phys. Rev. B 95, 165202 (1-15) (2017)], and then further extend our methodology for analysis of the dielectric and inverse dielectric tensor for transverse and longitudinal phonon mode dispersion characterization in high-power laser material yttrium orthosilicate [A. Mock *et al.*, Phys. Rev. B, 97 165203 (1-17) (2018)].

We apply our technique to investigate the effective electron mass tensor using optical Hall effect measurements [S. Knight, A. Mock *et al.*, Appl. Phys. Lett. 112, 012103 (2018); Editors' Pick], the temperature dependence of band-to-band transitions energies [A. Mock *et al.*, Appl. Phys. Lett. 112, 041905 (2018)], and the effects of aluminum alloying concentration onto the band-to-band transition energies [M. Hilfiker, A. Mock *et al.* Appl. Phys. Lett. (Under Review)]. We further apply our technique to epitaxial layers of beta-phase gallium oxide and discuss the relationship between the X-ray diffraction measured strains with respect to the optically determined shifts in transverse optical phonon modes as compared to the bulk material. Understanding of the stress and strain relationship to properties in monoclinic materials will help facilitate better control of material properties for engineering next generation power devices based on beta-Ga2O3.

5:00pm EL+EM-WeA-9 Terahertz Dielectric Anisotropy in Randomly Distributed, Spatially Coherent Polymethacrylate Microwire Arrays Fabricated by Stereolithography, *Serang Park,* University of North Carolina at Charlotte; *Y Li,* University Of North Carolina at Charlotte; *S Lee,* Harris Corp.; *S Schöche, C Herzinger,* J.A. Woollam Co., Inc.; *T Hofmann,* University Of North Carolina at Charlotte

Fabricating terahertz (THz) optical components with tailored dielectric properties including scalable anisotropies via additive manufacturing is drawing substantial interest as it potentially offers a rapid, low-cost pathway for THz optical system development. Metamaterials composed of slanted columnar structures have been reported to exhibit anisotropic behaviors at THz frequencies, which may allow the design of novel optical components including filters and sensors for the THz frequency range. Here, we report on the anisotropic THz-optical response of stereolithographically fabricated polymethacrylate slanted columnar layers. The samples are composed of randomly distributed, spatially coherent polymethacrylate wires with a diameter of 100 μm and a length of 700 μm, which are tilted by 45° with respect to the surface normal of the substrate. Generalized spectroscopic ellipsometry is employed to obtain Mueller matrix spectra of these samples in the range from 210 to 350 GHz. A simple biaxial (orthorhombic) layer homogenization approach is used to analyze the THz Mueller matrix data obtained at different azimuthal orientations. Our observations confirm that randomly distributed, spatially coherent polymethacrylate wire arrays exhibit a strong anisotropic response. In conclusion, stereolithographic fabrication is introduced as an effective tool for fabricating metamaterials with anisotropic THz-optical properties.

5:20pm EL+EM-WeA-10 Ultrafast Dynamics of Ge, InP and Si Proved by Time-Resolved Ellipsometry, *Shirly Espinoza, S Richter, M Rebarz,* Institute of Physics, Academy of Sciences of the Czech Republic, Czechia; *O Herrfurth, R Schmidt,* Universität Leipzig, Felix-Bloch-Institut für Festkörperphysik, Germany; *J Andreasson,* Institute of Physics, Academy of Sciences of the Czech Republic, Czechia; *S Zollner,* New Mexico State University

Recent developments in time-resolved ellipsometry allow us to study the ultrafast behavior of single crystals of undoped Ge, InP and Si at room temperature after carriers have been excited by an ultrashort laser pulse of 1.55 eV. Information about the dynamic processes such as scattering mechanisms of the hot charge carriers and electron-phonon coupling was obtained.

With a resolution of 120 fs, and a time scale from femtoseconds to nanoseconds, the observed changes are bigger in Ge than in the other materials. Our results are in agreement with theoretical and experimental work done some years ago on the dynamics of germanium studied by time-resolved ellipsometry [1,2]. The result of our experiments could go deeper into the details of the dynamics thanks to the development of the time-resolved experimental setup using state of the art technology in the fields of ultrafast lasers, electronics, and optics.

Our current spectral range is from 1.7 to 3.5 eV. The generated carrier density is on the order of $10^{20}\,cm^{-3}$, which allows us to compare the results with published data on doped materials [3].

References

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[2] Zollner S., Myers K.D., Jensen K.G., Dolan J.M., Bailey D.W., Stanton C.J. Femtosecond interband hole scattering in Ge studied by pump-probe reflectivity. *Solid State Commun.* 104 (1), 51-55 (1997)

[3] Xu C., Fernando N.S., Zollner S., Kouvetaki J., Menendez J. Observation of Phase-Filling Singularities in the Optical Dielectric Function of Highly Doped n-Type Ge. *Phys. Rev. Let.* 118, 267402 (2017)

5:40pm EL+EM-WeA-11 Optical Properties of Organic-Inorganic Lead Halide Perovskite Thin Films for Photovoltaics, *Biwas Subedi, M Junda, K Ghimire, N Podraza,* University of Toledo

Organic-inorganic lead halide perovskite based photovoltaics (PV) exhibit high initial efficiency, can be solution processed with potentially low material costs, and material band gaps can be tuned by composition. Unfortunately, these perovskites exhibit degradation upon exposure to atmosphere, light, and heat. Spectroscopic ellipsometry over the near infrared to ultraviolet range (0.73-5.9 eV) has been applied to characterize the complex optical response of solution processed ABX_3 (*A*: methylammonium—MA, formamidinium—FA, Cs; *B*: Pb, Sn; *X*: I, Br, Cl) perovskite thin films of different compositions. A parametric optical property model has been developed which includes contributions from electronic transitions above the band gap, the direct band gap, an exponentially decaying Urbach tail, and sub-gap absorption due to defect states. Using this model, above gap critical points, band gap energies, and sub-gap absorption are compared primarily as functions of *A*- and *B*-cation compositions for thin films. In situ, real time spectroscopic ellipsometry (RTSE) of perovskite films undergoing degradation induced by controlled relative humidity is used to track optical properties changes, particularly with respect to sub-gap absorption, and morphology changes occurring at the substrate / film and film / ambient interfaces. These optical property and morphology changes are tracked by RTSE for perovskite thin films of different compositions. Optical properties characterized by spectroscopic ellipsometry are used as input for external quantum efficiency (EQE) simulations of perovskite based PV devices. Comparisons between simulated and measured EQE spectra are used to identify differences in perovskite characteristics arising from the complete solar cell device fabrication process.

6:00pm EL+EM-WeA-12 Optical Constants of Ni at 300 K from 0.03 to 6.0 eV, *Stefan Zollner, F Abadizaman,* New Mexico State University

The optical constants of single-crystalline, polycrystalline, and thin films of Ni from 0.06 to 6 eV are determined from spectroscopic ellipsometry at an angle of incidence of 70˚. The experimental data are analyzed using three alternative methods. In the first method, the dielectric function is written as a sum of Lorentz and Drude oscillators. The second method writes the dielectric function as a product of these oscillators (Kukharskii product). In the third method, a Drude model with frequency dependent scattering rate

and plasma frequency is used. We used two Drude terms in the sum model to account for d- and s-electrons. The plasma frequencies were found to be 11.9 eV and 4.86 eV for d- and s-electrons, respectively, leading to a DC conductivity of about 80,000 (1/Ωcm) at 300 K, compared to the electrical DC conductivity of 143,000 (1/Ωcm) reported previously. Furthermore, the model reveals a very large free-electron contribution to the optical constants of Ni, which disproves earlier claims about their insignificance. We also employ graphical techniques to find the plasma frequencies and free-electron scattering rates, which agree well with the parameters found from the first and the second methods.

To prepare clean samples and reduce the thickness of the overlayers, the samples were maintained in ultrahigh vacuum at a temperature of 750 K for 6 hours and then cooled down overnight. A surface roughness thickness of 1-3 nm was found using atomic force microscopy and x-ray reflectivity.

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