MAGNETO-OPTICAL KERR EFFECT SPECTROSCOPY OF MAGNETIC OXIDES AND ORGANIC/OXIDE HETEROSTRUCTURES

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Recent progress in multiferroic materials and spintronic devices has renewed interest in metal oxide ferromagnetic and ferrimagnetic materials. We recently showed, on the example of CoFe₂O₄ (CFO) that the preparation of high-quality thin films of nanocrystalline ferrimagnetic can be achieven by means of an

environmentally benign aqueous solution processing route [1]. In addition to the ability to make high quality ferrite films, the aqueous solution processing strategy offers great flexibility for tuning film properties by incorporating or substituting additional transition metal ions. Furthermore, it can also be applied for the preparation of $La_{1-x}Sr_xMnO_3$ films [2], which is in discussion as material with high potential for spintronic applications as such or in combination with organic molecules.

The evolution of the structural, optical, and magnetic properties as a function of post-deposition annealing temperature was performed by HR-TEM and (magneto-)optical techniques: spectroscopic

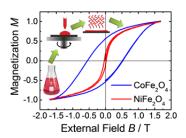


Fig. 1. Magnetization loops obtained by MOKE magnetometry for high-quality ferrite films fabricated by aqueous solution processing

ellipsometry and magneto-optical Kerr effect spectroscopy. The latter methods were also employed to study the effect of the substrate magnetization on the molecular orientation of phthalocyanine molecules impozed during the growth on the ferrite substrates.

Keywords: magnetic oxides, organic molecules, MOKE spectroscopy, spectroscopic ellipsometry

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VUV MAGNETO-OPTICAL TRANSIENT ELLIPSOMETER: ELIPS

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The ELIps instrument being built at the European Extreme Light Infrastructure Beamlines (ELI Beamlines) will combine three advanced techniques of ellipsometry: VUV ellipsometry, transient (Pump-probe) ellipsometry, and magneto-optical ellipsometry [1] The working range of energies in the VUV is between 12 eV and 40 eV, this VUV radiation will be provided by a High Harmonics Generation (HHG) source driven by a high-power femtosecond-laser.

The instrument complements already established VUV ellipsometers at synchrotron light sources [2] by using pulsed laser light sources that can be synchronized allowing the measurement of processes with a time-resolution of a few picoseconds. The pump pulse from a laser hits the sample first, triggering e.g. charge transfer processes; these processes can be observed and quantified by measuring the changes on the optical properties of the material by a probe pulse. The pump beam is a single wavelength beam that can be chosen from 180 nm – 20 um. A Helmholtz coil is also installed in the instrument, which can deliver a field of up to 1.5 T at a rate of up to 1 kHz. It would be possible to obtain the transverse magneto-optical Kerr Effect and probe e.g. the excitation of spin-polarized states.

All the components are contained within a single UHV chamber (with a target pressure lower than 10⁻⁸ mbar) designed with several additional ports to support future upgrades such as a sample preparation chamber. Furthermore, a cryostat allows temperature dependent studies.

Additional to the VUV ellipsometer, at ELI Beamlines, there is table top system for time-resolved ellipsometry utilizing super-continuum white-light pulses ranging from 350 nm - 750 nm that might be combined with the ELIps instrument.

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Keywords: VUV ellipsometry, Pump-probe, Transient, Ultrafast phenomena, UHV

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MAGNETO-OPTICAL SPECTROSCOPY AND SPECTROSCOPIC ELLIPSOMETRY OF Co60Fe₂₀B₂₀ THIN FILMS

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With the increasing interest in CoFeB for the realization of spintronic devices, the magneto-optical and optical transitions in spin-polarized electronic states are of significant importance for the characterization of such devices and further understanding of spin-dependent phenomena. We present the characterization of Co₆₀Fe₂₀B₂₀ thin films with combined magneto-optical Kerr effect (MOKE) spectroscopy and spectroscopic ellipsometry (SE).

Co₆₀Fe₂₀B₂₀ films with thicknesses ranging from 5 nm to 20 nm and a 3 nm Au capping layer were prepared by magnetron sputtering on glass and on thermally oxidized Si substrates. After a full characterization of the as-deposited samples, these were annealed in vacuum at 350°C in order to study the changes in the magneto-optical and optical spectra of the samples upon the crystallization of CoFeB.

Variable angle spectroscopic ellipsometry (VASE) measurements were performed in the spectral range from 0.73 eV to 5 eV, in transmission and reflection modes, the latter including

measurements at five angles of incidence (Φ = 50°, 55°, 60°, 65° and 70°). This allowed the relevant optical constants and the diagonal component of the dielectric tensor to be derived for CoFeB. The spectral dependence of the dielectric tensor component and the absorption coefficient were deduced from the experimental data for the samples prior and after annealing of CoFeB thin films, allowing further assessing the changes in the spectra due to the crystallization of the layer.

The MOKE investigations were performed in polar geometry in the spectral range of 1.5 eV to 5.0 eV, under magnetic saturation conditions. The Kerr rotation angle (θ_{Kerr}) and the Kerr ellipticity (η_{Kerr}) were measured as a function of the photon energy for the samples before and after annealing. The features of the θ_{Kerr} and η_{Kerr} —spectra at ~3.25 eV and ~2.5 eV are observed to

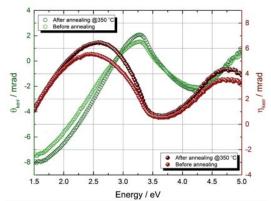


Figure 1: MOKE spectra measured at RT for Si/SiO₂(100nm)/Co₆₀F₂₀eB₂₀(20nm)/Au(3nm) before and after annealing the sample at 350 °C. The hollow circles show the real part (rotation) and solid sphere are the imaginary part (ellipticity) of the polar MOKE spectra.

become narrower after annealing, these spectral features are directly related to the magnetooptical transitions between the spin-polarized electronic states and the narrowing can be ascribed to the crystallization of CoFeB.

Finally, the present study provides access to the magneto-optical and optical characteristics of CoFeB thin films which can be determinant for spintronic device applications.

Keyword—CoFeB, Magneto-Optics, Ellipsometry, and Spintronic.

PLASMONIC PROPERTIES OF DEGENERATELY GE-DOPED CUBIC GAN

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The optical properties of highly-doped zincblende GaN (c-GaN) are investigated. Using germanium, free electron concentrations (n) exceeding 10²⁰cm⁻³ can be achieved while maintaining high structural sample quality [1]. Thin films were deposited by plasma-assisted molecular beam epitaxy on 3C-SiC quasi-substrates. Similar to the case of wurtzite GaN [2], they were studied comprehensively by emission and absorption related optical techniques. Spectroscopic ellipsometry yields the complex dielectric function (DF) of c-GaN from the mid-infrared into the deep ultraviolet spectral region. The transverse optical phonon mode and free carrier concentration dependent plasma frequencies are obtained from the IR-DF. Combined with Hall-effect data, we find a pronounced increase of the effective electron mass with *n* mirroring the non-parabolicity of the conduction band. The onset energy of interband absorption is determined by the fundamental band gap for lower *n* and blue-shifts due to phase-space-filling for increased electron density. Quantification of this so-called effective Burstein-Moss shift is possible when taking into account the counteracting band gap renormalization effect and the momentum dependence of the effective electron mass. Photoluminescence spectra reveal a blue-shift of the main recombination feature consisting of a donor-acceptor-pair band at doping levels below the degeneracy limit and a free-electron recombination band above. The lineshape fitting yields parameters emphasizing the values for gap renormalization and band filling obtained from DF.

Keywords: cubic GaN; effective mass; non-parabolicity; transition energy

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TEMPERATURE DEPENDENT DIELECTRIC FUCTION OF CUI THIN FILMS

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We present optical and structural properties of Cul thin films deposited by DC-sputtering at temperatures varying from 55°C to 310°C on c-sapphire substrate.

Scanning electron microscopy scans reveal a smooth surface morphology for films grown at temperatures above 60°C and large thickness inhomogeneity for films grown at lower temperatures. X-ray diffraction reveals good crystal quality for growth temperatures above 200°C.

The main features in the DF (see Fig. 1.) were assigned to exciton-related optical transitions at various critical points in the Brillouin zone. The observed energy separation of the split-off band is around 630 meV and is consistent with previous investigations [2].

The observed transitions reveal non-monotonic temperature dependence of the energy [1] as well as strong screening for excitons related to higher critical points.

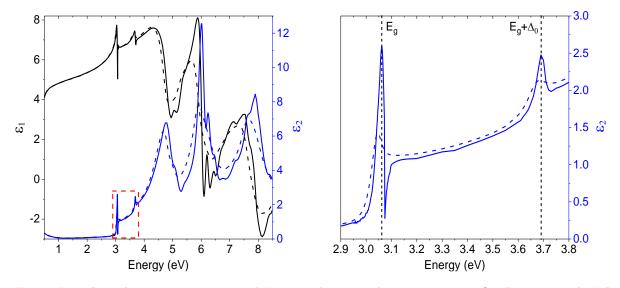


Fig. 1. Real (black) and imaginary part of dielectric function of the investigated Cul film at 10 K (solid) and at 300 K (dashed).

Keywords: Dielectric function; Cul;

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ELLIPSOMETRY OF TRANSPARENT CONDUCTING OXIDES FROM MID-INFRARED INTO VACUUM-ULTRAVIOLET

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Semiconducting metal oxides such as cubic In₂O₃, the various polytypes of Ga₂O₃, or rutile SnO₂ have attracted much interest in recent years. High-quality bulk crystals and single-crystalline heteroepitaxial films, covering a wide range of electron concentrations, became available allowing the determination of intrinsic optical properties as well as related fundamental band-structure parameters. This talk summarizes recent achievements.

Spectroscopic ellipsometry from the infrared (IR) into the vacuum-ultraviolet (VUV) spectral region is applied for determining the components of the dielectric tensor. The analysis of the IR dielectric function yields the phonon frequencies and the coupled phonon-plasmon modes from which electron effective mass as a function of carrier density (non-parabolicity of the conduction band) is obtained. Many-body effects such as exciton screening, band-gap renormalization, and band filling have a strong impact on the behavior around the fundamental band gaps, a quantitative description of these properties will be presented. Finally, synchrotron-based studies in VUV provide the transition energies related to critical points of the band structure.

Keywords: (In,Ga,Al)₂O₃; Band structure; effective mass, many-body effects

MAGNETRON SPUTTERED TiO_X LAYERS: STRUCTURAL, ELECTRICAL, OPTICAL AND THERMOCHROMIC ASPECTS

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Titanium oxide layers were prepared by sputter deposition with plasma emission monitoring in the whole stoichiometry range between Ti and TiO_2 without and with substrate heating to 240 °C. The layers were characterized with regard to their crystal structure and specific resistance. Optical constants were determined in the spectral range between 240 nm and 38 μ m by minds of spectral ellipsometry. The thermochromic behavior of a prepared Ti_2O_3 layer was measured and compared to calculations for bulk material.

Sub-stoichiometric titanium oxides and oxynitrides are used as absorber materials for solar thermal collectors [1]. But although a variety of thin film deposition techniques have been reported for the titanium-oxide system, infrared optical properties for these coatings were rarely determined. In recent years additional interest in thermochromic absorber layers has risen, especially for the infrared region as thermochromic absorbers are used to lower the collector stagnation temperature [2]. The Ti_2O_3 material comprising a switching temperature between 130 °C and 200 °C is a potentially interesting candidate for this application [3]. So the scope of this paper is to link the deposition parameters for sputtered titanium oxide layers to their morphology and optical constants, with special interest in the thermochromic phase Ti_2O_3 [4].

Keywords: solar thermal collector; titanium oxide; thermochromic absorber

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DIELECTRIC FUNCTION OF ε-(In,Ga)₂O₃ THIN FILMS

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The large band gap energy of about 4.8eV makes Ga_2O_3 interesting as transparent conductive oxide (TCO) since even in the presence of impurities a high transmissivity is sustained in the visible and even in the UV-A/B spectral range. The dielectric function for the β -phase of Ga_2O_3 was recently presented and discussed [1–3]. However, the dielectric function of the ϵ -phase, being interesting due to its spontaneous polarization, and its alloys with In and AI, which are used for tuning the band-gap energy, is not explored in great detail so far.

Here, we present a detailed analysis of the dielectric function of an ϵ -phase (In,Ga)₂O₃ thin film with lateral In composition spread for In concentrations between 1% and 35% in the infrared (350cm⁻¹ – 1500cm⁻¹) and NIR-VUV (0.5eV – 8.5eV) spectral range. By means of a parametric model dielectric function approach, we derive the refractive index dispersion and the properties of the electronic band-band transitions and phonons. We observe a red shift of the fundamental band-gap energy with increasing In concentration which results in an refractive index increase in the transparent spectral range.

The thin film samples have been grown by means of pulsed laser deposition (PLD), with the use of segmented PLD targets (consisting of half-segments of binary indium oxide respective indium oxide and binary gallium oxide). This resulted in films with a continuous composition spread [4].

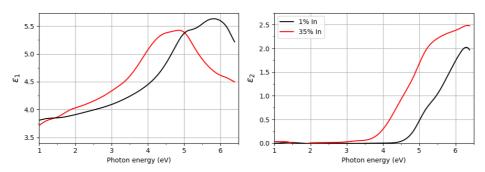


Fig.1. Real (left) and imaginary (right) part of the dielectric function of ε -Ga₂O₃:In for an In concentration of 1% (red line) and 35% (black line).

Keywords: dielectric function; Raman tensor; Ga₂O₃; wide band-gap oxide

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SPECTROSCOPIC INVESTIGATION OF CATION CONFIGURATION STATE OF SPINEL FERRITE THIN FILMS

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We present a systematic study of cation configurations of spinel type ZnFe₂O₄ (ZFO) and Zn_xFe_{3-x}O₄ thin films fabricated by pulsed laser deposition. Electronic transitions assigned in the model dielectric function (MDF) correspond to Fe²⁺ interband d-d band and O²⁻ anion 2p to Fe³⁺ cation 4s and 3d band optical transitions [1]. Taking into account the contribution of each optical transition to the MDF, we have estimated the cation concentration ratio corresponding to the disordered state of the normal spinel configuration, and have compared it to the room temperature magnetization saturation of ZFO films before and after annealing in oxygen and argon atmospheres at temperatures to 375°C, (see Fig.1 a). Surface sensitive XPS Fe 2p and 3p core level analysis generally follows the trend, but shows also a deviation in the divalent and trivalent cation ratio as obtained from the bulk MDF for the Zn_xFe_{3-x}O₄ films, (Fig. 1 b). Two competing exchange interactions, AFM oxygen mediated superexchange and FM double-exchange, were determined as a function of annealing temperature in ZFO and of the Zn/Fe ratio in Zn_xFe_{3-x}O₄ films. The results clarify their role in weakening the total ferrimagnetic response.

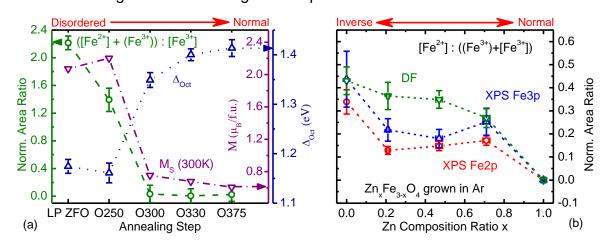


Fig. 1. (a) Area ratio of transitions involving divalent and trivalent Fe cations, representing disorder in a normal spinel structure, octahedral crystal field parameter and room temperature magnetization saturation as a function of annealing step in oxygen atmosphere. (b) Normalized area ratio of divalent and trivalent Fe cation contributions obtained from transitions in the DF and XPS Fe 2p and 3p core levels as a function of Zn composition ratio.

Keywords: Dielectric Function; Ferrimagnetic Material; Spinel Ferrites

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OPTICAL AND ELECTRONIC PROPERTIES OF LOW-SYMMETRY MATERIALS

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We discuss analysis of the dielectric function tensor for monoclinic metal-oxides. Generalized ellipsometry using an eigenpolarization vector summation approach [1] from the terahertz to the vacuum-ultra-violet spectral regions, and as a function of temperature (Fig. 1) [2], along with optical Hall effect, permit us to unravel all infrared active transverse and longitudinal optical phonon modes [3], free carrier excitations [3], band-to-band transitions [4], optical constants [4], exciton properties [4], effective mass parameters [5] and directional dependencies in single crystalline β -Ga₂O₃. We compare our findings with results from density functional theory calculations [4], and we revise and augment previous incomplete assignments [6,7].

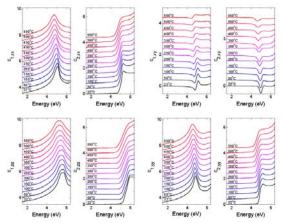


Figure 1: Real and imaginary components of the dielectric tensor elements, ϵ_{xx} , ϵ_{yy} , ϵ_{xy} , and ϵ_{zz} for room temperature (black) to 550°C (red) determined by generalized ellipsometry. The functions were shifted vertically by increments of 0.4 with respect to each other for convenience. Ref. [2]

We apply this same approach to other monoclinic oxides such as CdWO₄ [8] (Fig. 2) and Y_2SiO_5 [9] as well as to triclinic materials.

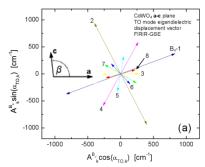


Figure 2: Schematic presentation of the Bu symmetry TO mode eigendielectric displacement unit vectors within the **a-c** plane according to TO mode amplitude parameters $A^{\text{Bu}}{}_{k}$ and orientation angles $\alpha_{\text{TO},k}$ with respect to axis a obtained from generalized ellipsometry analysis. Ref. [8]

Keywords: Low-symmetry, monoclinic, Ga₂O₃

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