DS 21: Optical Analysis of Thin Films

Time: Friday 9:30–12:15 Location: A 053

Invited Talk DS 21.1 Fri 9:30 A 053

Spotlight on Crystalline Textured Anilino-Squaraine Thin Films featuring Multiple Davydov Splitting and Charge Transfer Excitons — •Manuela Schiek — JKU Linz, Austria

Quadrupolar anilino squaraine dyes exhibit distinct excitonic signatures in their visible absorption spectra due to strong intermolecular interactions. These excitons are a result of the spatial arrangement of the molecular backbones, which is steered by non-chromophoric terminal functionalization patterns. They can be of predominantly Frenkel-excitonic nature [1] or are hybridized with intermolecular charge transfer [2] and may feature an extraordinary strong excitonic circular dichroism [3]. Polarized spectro-microscopy and imaging ellipsometry are implemented to study the linear dichroism of crystalline domains, which is a direct probe for the excitonic transitions. Combined with structural analysis the orientation of molecules is traced and the excitonic bands are assigned by modelling. Especially imaging ellipsometry allows determination of the complete dielectric tensor [4], from which a multiple Davydov splitting including a dark state can be deduced for the case of an orthorhombic polymorph containing multiple non-equivalent molecules within the primitive unit cell.

[1] Balzer, Breuer, Witte, Schiek. Langmuir 38 (2022) 9266. [2] Balzer, Hestand, Zablocki, Schnakenburg, Lützen, Schiek. J. Phys. Chem. C 126 (2022) 13802. [3] Gavazzi, Schumacher, Grisanti, Anzola, Di Maiola, Zablocki, Lützen, Schiek, Painelli. J. Mater. Chem. C 11 (2023) 8307. [4] Funke, Duwe, Balzer, Thiesen, Hingerl, Schiek J. Phys. Chem. Lett. 12 (2021) 3053.

DS 21.2 Fri 10:00 A 053

Theory of X-ray excitation of two-dimensional materials — Dominik Christiansen[1], Ivan Maliyov[2], •Joris Sturm[1], Malte Selig[1], Marco Bernardi[2], and Andreas Knorr[1] — [1]Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, Hardenbergstr. 36, 10623 Berlin, Germany — [2]Department of Applied Physics and Materials Science, California Institute of Technology, Pasadena, CA 91125, USA

In this contribution, we present a combined theory of X-ray Bloch - and Maxwell's equations to describe X-ray absorption experiments like XANES and EXAFS. Particularly crucial in the calculation of EXAFS spectra is a detailed knwoledge about the electronic structure and the electron - X-ray interaction matrix elements. In contrast to prior work, where a semi-empirical tight-binding approach was employed [1], we have improved our methodology by including *ab initio* electronic structure calculations. As a proof of principle we calculated the absorption spectrum of graphene which recovers now spectral signals missing in the previous approach [1].

[1] Christiansen, Dominik, et al., Physical Review Research 5.2 (2023): 023002.

[2] Christiansen, Dominik, et al., "Joint ab initio and Maxwell-Bloch approaches for the description of X-ray excitations of two-dimensional materials" **(in preparation)**

DS 21.3 Fri 10:15 A 053

Probing ultrafast dynamics employing a laser-driven broadband soft X-ray reflectometer — •Jasmin Jarecki[1], Martin Hennecke[1], Lutz Ehrentraut[1], Matthias Schnürer[1], Stefan Eisebitt[1,2], and Daniel Schick[1] — [1]Max-Born-Institut für Nichtlineare Optik & Kurzzeitspektroskopie, Berlin, Germany — [2]Institut für Optik & Atomare Physik, TU Berlin, Germany

Soft X-ray reflectometry (XRR) combines elemental and chemical sensitivity when hitting core-to-valence resonances, few-nanometer spatial resolution due to short X-ray wavelengths, and femtosecond temporal resolution when employing ultrashort sources. This allows probing of ultrafast dynamics of buried layers and across interfaces of stratified nanostructures. Here, we present a laboratory-based setup for time-resolved XRR, employing broadband soft X-ray pulses with energies ranging from 200 to 600 eV and pulse durations of 26 fs provided by a high harmonics generation (HHG) source. In contrast to synchrotron radiation or FEL sources, the quasi-continuous spectrum of the HHG offers efficient probing of broad spectra, e.g. across L-edges of transition metals, and of a large volume in reciprocal space in only a single acquisition. We benchmark our setup by probing laser-induced structural dynamics of a Mo/Si superlattice (SL). In very good agreement with simulations, we observe a shift of the 1^{st} SL Bragg peak in reciprocal space probed at photon energies around 500 eV. Our results are an important step towards accessing also ultrafast electronic and magnetic dynamics in similar scattering experiments at such high photon energies outside of large-scale facilities.

DS 21.4 Fri 10:30 A 053

Synthesis, structure, and optical properties of Multi-Au capped Si nanoislands as a novel saturable absorber with a high modulation depth — Ali Karatutlu[1], Umut Taylan[1,2], •Zehra Gizem Mutlay[1], and Bülend Ortaç[1] — [1]UNAM–Institute of Materials Science and Nanotechnology, Bilkent University, Ankara, 06800-Turkey — [2]Empa, Swiss Federal Laboratories for Materials Science & Technology, Laboratory for Advanced Materials Processing, Feuerwerkerstrasse 39, CH-3602 Thun, Switzerland

This study demonstrates the first-time synthesis of multi-Au capped nanoislands and its potential as a new non-linear hybrid material. The synthesis was conducted by a vapor-liquid-solid mechanism using a subsequent laser ablation and plasma-enhanced vapor deposition. The results are promising for the use of this material in non-linear optics as a saturable absorber and potential mode-locker due to a relatively high-modulation depth reaching around 15% measured using Ti-sapphire femtosecond (fs) laser light with a wavelength at 800 nm. The hybrid saturable absorber consisting of metallic and semiconductor structures was initially characterized using UV-Vis-NIR absorption and reflection spectroscopy, scanning electron microscopy, and transmission electron microscopy studies followed by a power-dependent transmission measurement of an fs light.

15 min. break

DS 21.5 Fri 11:00 A 053

Exploring the Tunability of Phonon Properties in Ultra-Thin Bismuth Films — •Felix Hoff[1], Timo Veslin[1], Abdur Rehman Jalil[2], Peter Kerres[2], Jonathan Frank[1], Thomas Schmidt[1], Yazhi Xu[3], Dasol Kim[1], Riccardo Mazzarello[3], and Matthias Wuttig[1,2] — [1]I. Institute of Physics (IA), RWTH Aachen University, Germany — [2]Peter Grünberg Institute - JARA-Institute Energy Efficient Information Technology (PGI-10), Germany — [3]Dipartimento di Fisica, Sapienza University of Rome, Italy

In recent years, ultra-thin bismuth films have gathered attention for applications in thermoelectrics, ferroelectrics, and recently for topological applications, too. Our study focuses on the systematic exploration of ultra-thin bismuth films, utilizing advanced characterization techniques to unveil the intriguing changes in phonon properties as the layer thickness is reduced. Raman spectroscopy and optical fs pump probe spectroscopy were employed to measure phonon frequencies and lifetimes, exposing a complex interplay of factors influencing the observed hardening and softening of phonon modes. Additional investigations employ XRD, revealing structural modifications and distortions. DFT calculations unravel the intricate relationship between structural changes, chemical bonding, and optical properties. We determine the layer-thickness-dependent dielectric function, which show remarkable confinement effects. Epitaxially grown samples, detailed in a prior publication, ensure reproducibility. Motivated by achieving high-quality thin films, our work contributes to understanding the underlying mechanisms governing phonon properties in ultra-thin bismuth.

DS 21.6 Fri 11:15 A 053

Surface-Sensitive and Bulk-Suppressed Raman Scattering by Transferable Nanoporous Plasmonic Membranes — •Pietro Marabotti[1], Roman M. Wyss[1], Günter Kewes[1], Martin Frimmer[2], Karl-Philipp Schlichting[2], Markus Parzefall[2], Eric Bonvin[2], Martin F. Sarott[2], Morgan Trassin[2], Lala Habibova[1], Giorgia Marcelli[1], Marcela Giraldo[2], Jan Vermant[2], Lukas Novotny[2], Mads C. Weber[3], and Sebastian Heeg[1] — [1]Humboldt-Universität zu Berlin, Germany — [2]ETH Zürich, Switzerland — [3]Le Mans Université, France

The Raman response of surfaces or thin films is often too weak to be detected and obscured by dominant bulk signals. Here we overcome this limitation by placing a transferable porous Au membrane (PAuM) on top of the surface of interest. Slot-like nanopores in the membrane act as plasmonic slot antennas and enhance the Raman response of the surface underneath. The PAuM also suppresses the penetration of the excitation laser into the bulk, efficiently blocking the bulk Raman signal. Using graphene as a model surface, we show that these two effects increase the surface-to-bulk Raman signal ratio by 3 orders of magnitude. We find that 90% of the Raman enhancement occurs within the top 2.5 nm of the material, demonstrating truly surface-sensitive Raman scattering. To validate our approach, we analyze the surface of a $LaNiO_3$ thin film, observing a Raman mode splitting from the surface-layer, which is evidence that the surface structure differs from bulk. This shows that PAuM give direct access to Raman signals of surfaces and their structural properties.

DS 21.7 Fri 11:30 A 053

Thermal conductivitiy of hard-metal thin films microscopically resolved by Brillouin scattering — •Nils Denkmann[1], Donita Delijaj[1], Lasse Leukefeld[1], Nelson Filipe Lopes Dias[2], Finn Ontrup[2], Wolfgang Tillmann[2], and Jörg Debus[1] — [1]Department of Physics, TU Dortmund — [2]Institute of Materials Engineering, TU Dortmund

Common procedures to measure the thermal conductivity of thin films are, e.g., the differential scanning calorimetry or the transient thermoreflectance. These methods predominantly provide macroscopic values and are not applicable to samples with complex geometries. To determine the thermal conductivity with micrometer spatial resolution and independently of the sample geometry as well as to understand the nanoscopic mechanisms alternative methods are needed. Here, we show how acoustic surface waves inside hard-metal thin films (e.g., TiAlN) are examined directly using Brillouin laser-light scattering. This method allows us to determine the thermal conductivity as function of the waves' propagation direction and the shear/Young's modulus. Spatial scanning is moreover exploited to correlate our microscopic acquisition with complementary macroscopic data. The results will provide a thorough insight into the effects of thermo-mechanical properties of hard-metal thin films.

DS 21.8 Fri 11:45 A 053

Scanning Reflectance Anisotropy Microscopy: Strain Mapping of Metasurfaces and Beyond — •Fabian Haake, Joan Sendra, Henning Galinski, and Ralph Spolenak — ETH Zurich, Zurich, Switzerland

A common failure mode of flexible electronic devices is catastrophic failure or device degradation due to mechanical strain, making accurate strain control a critical aspect in their design and characterization. Here, we introduce scanning reflectance microscopy (SRAM) as a broadband multi-material platform for strain mapping on the microscale. This multi-material optical platform serves not only to access microscopic strain distributions but also shows high phase sensitivity.

This technique provides a practical approach to mechanical characterization, yielding valuable insights into the mechanical behavior of diverse materials, including metamaterials, amorphous and crystalline semiconductors, and metals. The microscope's capability to generate diffraction-limited strain and phase maps enables a thorough analysis of materials properties without causing damage.

The capabilities of the technique are discussed on specific examples, such as strained metals and semiconductors as well as metasurfaces. Special emphasis is laid on the chance to apply this technique for insitu studies and post-mortem analysis of fractured materials.

[1] J. Sendra, F. Haake, M. Calvo, H. Galinski, and R. Spolenak, Multi-Material Strain Mapping with Scanning Reflectance Anisotropy Microscopy, Adv Funct Materials, p. 2302179, Jun. 2023, doi: 10.1002/adfm.202302179.

DS 21.9 Fri 12:00 A 053

Exploring the optical constants of ruthenium (Ru) through EUV metrology:a study on thin films — •Samira Naghdi and Victor Soltwisch — Abbestraße 2-12, 10587 Berlin, Germany

This study presents a comprehensive investigation into the optical constants of ruthenium (Ru), focusing on thin films with a thickness of 30 nm. Employing Extreme Ultraviolet (EUV) metrology, our study investigates the spectral response of Ru across a wavelength range of 10 to 20 nm. The experimental setup involved measurements at varying angles, ranging from 2 to 88 degrees, to provide a thorough understanding of the material's optical behavior. The utilization of EUV metrology in this research not only ensures precision in wavelength selection but also offers a unique insight into the optical properties of Ru within the EUV spectrum. The study aims to contribute valuable data to the broader understanding of Ru's behavior, particularly in thin film applications, with potential implications for advancements in nanotechnology, optoelectronics, and other fields where ruthenium plays a crucial role. The results of this study not only contribute to the fundamental understanding of ruthenium's optical properties but also open avenues for further exploration and application in cutting-edge technologies.