O 11: Electronic Structure of Surfaces: Spectroscopy, Surface States I

Time: Monday 15:00–18:00 Location: H4

O 11.1 Mon 15:00 H4

Optimizing the photon detection in inverse photoemission — •Jan Willermann, Fabian Schöttke, and Markus Donath — Physikalisches Institut, Universität Münster, Münster, Germany

The detection of vacuum-ultraviolet photons ($\hbar\omega = 9.9\,\mathrm{eV}$) in inverse-photoemission experiments is usually carried out with gas-filled counting tubes. Commonly, iodine/argon gas fillings were used for Geiger-Müller type photon detection. Recently, the gas filling was replaced by acetone to increase stability and the operation was changed to proportional mode [1]. While doing this, the counting tube geometry must not necessarily be changed. We carried out systematic measurements to understand the photon detection process in acetone-filled counting tubes and optimize the geometry accordingly. In the range of optimal gas pressures, we found that the photon mean free path in acetone is in scale of a few millimeters instead of a few centimeters as in iodine/argon. Furthermore, we observed that the electron mean free path in proportional-type counting tubes is also reduced to few millimeters. In comparison, the mean free path of electrons in Geiger-Müller-type counting tubes is in the range of the radius of the counting tube. As a consequence, the position of the cathode wire close to the entrance window becomes a critical parameter for the detection efficiency.

[1] C. Thiede *et al.*, Meas. Sci. Technol. **29**, 065901 (2018).

O 11.2 Mon 15:15 H4

Excitation-Mediated Transport through Nano-scale Josephson Junctions in the Coulomb Blockade Regime — •Zhengyuan Liu, Sebastian Scherb, Werner M.J. van Weerdenburg, Daniel Wegner, Nadine Hauptmann, and Alexander A. Khajetoorians — IMM, Radboud University Nijmegen, the Netherlands

Josephson junctions (JJs) are essential for superconducting quantum computing and sensing technologies. It has been shown that superconductivity in elemental BCS superconductors, including those typically used for JJs [1], can persist down to the 2D limit. However, the effects of such quantum confinement on both the electronic structure of a JJ and its subsequent transport remains unclear, and fabricating conventional source-drain JJ devices at the nanometer scale is challenging. Here, we use low-temperature scanning tunneling microscopy and spectroscopy to study the electronic structure and transport of model nano-JJ stacks grown on Si(111) surface. We first characterize the quantum well states and superconducting gap of the underlying layer. Then, we explore how both the dielectric and overlying metallic layer affect the electronic and superconducting states. We study the tunneling transport in the Coulomb blockade limit for a number of JJ stacks, revealing that the conductance is dominated by an excitation mediated process. We also discuss the superconducting properties of these stacks, and the potential to detect superconductivity through their charge transport. [1] Werner M. J. van Weerdenburg et al. Sci. Adv. 9, eadf5500 (2023).

O 11.3 Mon 15:30 H4

Double Photoemission Spectroscopy of C_{60} on $SrTiO_3$ (001) with high efficiency high-order harmonic light source — •Kathrin Plass[1], Robin Kamrla[1], Frank O. Schumann[2], and Wolf Widdra[1] — [1]Institute of Physics, Martin-Luther-Universität Halle-Wittenberg, Halle (Saale), Germany — [2]Max Planck Institute of Microstructure Physics, Halle (Saale), Germany

The photoemission spectroscopy is one of the main tools for studying the electronic structure of solids. However, the effects of electron correlation can only be inferred indirectly. In contrast, double photoemission spectroscopy (DPE) enables the direct observation of such phenomena by detecting pairs of correlated photoelectrons ejected following the absorption of a single photon [1].

C_{60} is considered a strongly correlated material, exhibiting a highly structured valence band spectrum. Recently, we discovered orbital-resolved correlation-induced two-electron binding energy shifts in C_{60}. In this study, we analyzed DPE data from C_{60} thin films on $SrTiO_3$(001) obtained using a laboratory-based high-order harmonic generation (HHG) light source operating at MHz repetition rates [2]. By boosting the HHG light source to higher photon energies and repetition rates, we are now able to investigate plasmon-assisted double photoemission in C_{60}, predicted recently [3].

[1] J. Berakdar et al., Phys. Rev. Lett. **81**, 3535 (1998)
[2] C.-T. Chiang et al., ELSPEC **200**, 15 (2015)
[3] M. Schüler et al., Sci. Rep. **6**, 24396 (2016)

O 11.4 Mon 15:45 H4

Surface Orbitroscopy: Emergent Phenomena of Orbital Angular Momentum — •Maximilian Ünzelmann[1], Tim Figgemeier[1], Begmuhammet Geldiyev[1], Hendrik Bentmann[2], and Friedrich Reinert[1] — [1]Exp. Physik VII and Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg, Germany — [2]QuSpin, NTNU Trondheim, Norway

Quantum degrees of freedom in electronic states are a key facet of modern quantum materials. Recently, the orbital angular momentum (OAM) — an orbital analogue of electron spin — has attracted broad attention in condensed matter and surface physics. For instance, orbitronics has been predicted as a promising route towards new functionalities, such as low-dissipation orbital currents or magnetization switching by orbital torques. In this talk, I will present orbital-sensitive angle-resolved photoemission spectroscopy experiments that demonstrate three key features of the OAM: (i) it acts as a central mediator between lattice and spin in spin-orbit-coupled systems, such as Rashba-type surface states [1,2], (ii) it might be associated with orbital currents, and (iii) its momentum texture carries topological information, giving rise to intriguing paradigms like OAM monopoles [3] or momentum-space quantum vortices [4].

[1] M. Ünzelmann *et al.*, Phys. Rev. Lett. **124**, 176401 (2021), [2] B. Geldiyev, M.Ü., *et al.*, Phys. Rev. B **108**, L121107 (2023), [3] M. Ünzelmann *et al.*, Nat. Commun. **12**, 3650 (2021), [4] T. Figgemeier, M.Ü., *et al.*, arXiv:2402.10031 (2024)

O 11.5 Mon 16:00 H4

Computationally Efficient First-Principles Treatment of Scattering States in Photoemission: A Pseudo-potential Approach — •Gian Parusa[1,2,3] and Michael Schüler[1,2,3] — [1]PSI Center for Scientific Computing, Theory and Data, 5232 Villigen PSI, Switzerland — [2]National Centre for Computational Design and Discovery of Novel Materials (MARVEL), Paul Scherrer Institute, 5232 Villigen PSI, Switzerland — [3]Department of Physics, University of Fribourg, 1700 Fribourg, Switzerland

The calculation of photoemission matrix elements requires the consideration of several key factors, including Bloch states, light-matter coupling, and scattering states. The evaluation of scattering states in solids from first-principles, particularly within a plane-wave basis, is computationally demanding due to the large number of plane waves required at high energies. A well-established strategy for reducing the computational cost in the treatment of valence states involves the use of pseudo-potentials. In this work, we extend the concept of optimized norm-conserving pseudo-potentials to scattering states, utilizing a non-local Vanderbilt projector to restore the wave function properties at a specified target energy. This approach is applied to scattering states, specifically for energies above 1 Rydberg. The method is validated by simulating the photoemission spectrum of graphene and hexagonal boron nitride (h-BN) with the results compared to all-electron calculations, demonstrating both the accuracy and computational efficiency of the proposed technique.

O 11.6 Mon 16:15 H4

Electronic structure of ferromagnetic CrTe — •Chien-Wen Chuang[1], Muthu P.T. Masilamani[1], Hibiki Orio[1], Maximilian Ünzelmann[1], Chia-Nung Kuo[2,3], Chin-Shan Lue[2,3], Ashish Chainani[4], and Friedrich Reinert[1] — [1]Exp. Physik VII and Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg, Würzburg, Germany — [2]Department of Physics, National Cheng Kung University, Tainan, Taiwan — [3]Taiwan Consortium of Emergent Crystalline Materials, National Science and Technology Council, Taipei, Taiwan — [4]National Synchrotron Radiation Research Center, Hsinchu, Taiwan

Recent studies showed that CrTe and doped CrTe exhibit various emergent properties, such as a large magnetocaloric effect, spontaneous skyrmions and ferromagnetism with high Curie temperature $T_C \sim 340$ K. We carried out Cr L-edge ($2p$-$3d$) X-ray absorption spectroscopy

(XAS) and resonant photoemission spectroscopy (Res-PES) with right and left circularly polarized light on single crystal CrTe to investigate the role of Coulomb correlations in Cr 3*d* density of states(DOS). The Res-PES spectra showed a resonantly enhanced sharp peak at a binding energy of 1.5 eV, which corresponds to the Cr main 3*d* DOS, while states at the Fermi level do not show a clear resonance enhancement. A small circular dichroism of Cr 3*d* DOS was observed. In addition, we observed the Cr 3*d* two-hole L_3VV Auger feature which corresponds to a correlation satellite. Using the Cini-Sawatzky method, we estimate the on-site Coulomb interaction energy in Cr 3*d* states to be $U_{dd}\sim 3$ eV.

O 11.7 Mon 16:30 H4

Unveiling the Role of Inelastic Mean Free Path in Photoelectron Diffraction: A Computational Study by Multiple Scattering — •Trung-Phuc Vo[1,2], Olena Tkach[3], Sylvain Tricot[4], Didier Sébilleau[4], Aimo Winkelmann[5], Olena Fedchenko[3], Yaryna Lytvynenko[3,6], Dmitry Vasilyev[3], Hans-Joachim Elmers[3], Gerd Schönhense[3], and Ján Minár[1] — [1]Univ. West Bohemia, Czech Republic — [2]Institute of Physics, Czech Academy of Sciences, Czech Republic — [3]Univ. Mainz, Germany — [4]Univ. Rennes, IPR, France — [5]AGH Univ. Krakow, Poland — [6]Institute of Magnetism of the NAS of Ukraine and MES of Ukraine, Ukraine

The inelastic mean free path (IMFP) of electrons near solid surfaces describes the average distance an electron travels through a solid before losing its kinetic energy via inelastic collisions. In surface analysis techniques such as photoelectron diffraction (PED), a short IMFP makes photoelectrons highly surface sensitive, allowing precise structural determination of surfaces. Conversely, a longer IMFP allows access to deeper layers, facilitating the study of bulk properties. The influence of the IMFP on PED is particularly evident with the advent of advanced time-of-flight (ToF) measurements. Controlling this key parameter is crucial. In this work, we summarize the PED implementation within the SPRKKR package and systematically explore its application over a wide kinetic energy range (106-1036 eV) for Ge 3d core levels. Our computational efforts provide insight into both Kikuchi diffraction patterns and their relation to valence band mapping, thus bridging PED analysis with electronic structure studies.

O 11.8 Mon 16:45 H4

Towards accessing the initial state by dichroic photoemission — •Jakub Schusser[1,2], Hibiki Orio[1], Maximilian Ünzelmann[1], Johannes Hessdörfer[1], Muthu Prasath Thirugnanasambandam Masilamani Masilamani[1], Florian Diekmann[3,4], Kai Rossnagel[3,4], and Friedrich Reinert[1] — [1]Experimentelle Physik VII and Würzburg- Dresden Cluster of Excellence ct.qmat, Universität Würzburg, Würzburg, Germany — [2]University of West Bohemia, Pilsen, Czech Republic — [3]Ruprecht Haensel Laboratory, DESY, Hamburg, Germany — [4]Institute of Experimental and Applied Physics, Kiel University, Germany

Despite its many advantages, photoemission has so far not allowed direct experimental access to detailed information about the initial state of solids. Dichroism in angle-resolved photoemission spectroscopy arises inherently from the matrix-element effects that depend on the initial and final states as well as the light field perturbation. By comparing both experimental and theoretical soft X-ray data in bulk WSe_2 we show the robustness of the newly introduced dichroic technique against variation of photon energy, light polarization and angle of incidence. Such robustness of the matrix-element effect represents a leap towards accessing the initial state by this differential technique with high relevance in the field of topological materials, layered systems and other material classes.

O 11.9 Mon 17:00 H4

Surface electronic structure of Te chains on Au(100) via ARPES — •Begmuhammet Geldiyev[1], Maximilian Ünzelmann[1], Tim Figgemeier[1], Hendrik Bentmann[2], and Friedrich Reinert[1] — [1]Experimentelle Physik 7 and Würzburg-Dresden Cluster of Excellence ct.qmat, Universität Würzburg — [2]Center for Quantum Spintronics, Department of Physics, NTNU, Norway

In this talk, we will provide a comprehensive description of the surface electronic band structure in epitaxial Te chains grown in the submonolayer regime on a Au(100) substrate. First, the deposition of 0.25 ML Te results in an adlayer square lattice superstructure with $p(2 \times 2)$ periodicity. Particularly, the band structure of this system features an interface state – derived from hybrid Te-Au orbitals – that exhibits an anisotropic spin and orbital Rashba effect [1]. Second, by slightly increasing the Te coverage to 0.30 ML, the earlier square arrangement evolves into a chain structure with $c(10 \times 2)$ periodicity [2]. Here, we will address whether the latter system inherits an analogous Rashba effect scenario. Furthermore, we will unravel a flat electronic band with a bandwidth of ≈ 20 meV, indicating an almost perfect one-dimensional character.

[1] B. Geldiyev et al., Phys. Rev. B 108, L121107 (2023)
[2] L. Hammer et al., Surf. Sci. 750, 122589 (2024)

O 11.10 Mon 17:15 H4

Unoccupied electronic structure of the AgTe/Ag(111) surface alloy: A spin-resolved inverse photoemission study — •Marcel Holtmann, Carolin Benfer, and Markus Donath — Physikalisches Institut, Münster University, Germany

The AgTe/Ag(111) surface-alloy system has recently been investigated to understand the microscopic origin of the Rashba effect [1]. ARPES measurements suggest that tellurium p orbitals hybridize with states of the Ag(111) substrate resulting in two p_{xy} valence bands. For the unoccupied band structure, a third hybridization state with out-of-plane symmetry is detected using two-photon photoemission. Apart from this, the unoccupied electronic structure of AgTe/Ag(111) remains unexplored.

We employed spin- and angle-resolved inverse photoemission (IPE) to measure the unoccupied electronic structure with a broader scope. Using our rotatable spin-polarized electron source, we were able to measure the three-dimensional spin texture of the unoccupied states. As expected in [1], we found that the hybridization state shows a large Rashba-type spin splitting, which is untypical for non-heavy-metal systems. Using several photon detectors at different take-off angles to measure the angular distribution of the emitted photons, we gained access to the orbital symmetries of the involved electronic states. In addition, like the pristine Ag(111) surface, the AgTe/Ag(111) surface hosts an image-potential state.

[1] M. Ünzelmann *et al.,* Phys. Rev. Lett. **124**, 176401 (2020)

O 11.11 Mon 17:30 H4

Surface Sensitivity of the VLEED Scattering Process at Pt(111) — •Hannah Unterberg, Christoph Angrick, and Markus Donath — Universität Münster, Germany

The diffraction pattern of low-energy electrons (LEED) from Pt(111) shows a three-fold symmetry, although the surface layer exhibits a six-fold symmetry. This result proves the non-neglible influence of the second and deeper atomic layers resulting from the finite probing depth of the electrons. Here, we report on a spin-polarized very-low-energy electron diffraction (VLEED) [1,2] experiment on Pt(111). The reflected specular beam is measured at a fixed polar angle of incidence $\Theta = 45°$, while the azimuthal angle of incidence is varied over a wide range. This allows us to probe the surface sensitivity of the VLEED scattering process.

Our measurements at the non-reconstructed Pt(111) surface reveal distinct differences in the electron reflectivity and the spin-orbit-induced reflection asymmetry along the high symmetry directions $\overline{\Gamma}\,\overline{\mathrm{M}}$ and $\overline{\Gamma}\,\overline{\mathrm{M}}$'. These directions are equivalent (non-equivalent) in the case of six (three)-fold rotational symmetry. Our result indicates a substantial contribution from both the first and subsequent atomic layers to the VLEED scattering process. In contrast, results for the Au(111) surface, which hosts the prominent herringbone reconstruction, exhibit only minor differences between $\overline{\Gamma}\,\overline{\mathrm{M}}$ and $\overline{\Gamma}\,\overline{\mathrm{M}}$'. The different results for Pt(111) and Au(111) are discussed in view of the reconstruction.

[1] U. Burgbacher *et al.*, Phys. Rev. B **87**, 195411 (2013)
[2] C. Angrick *et al.*, J. Phys.: Condens. Matter **33**, 115001 (2020)

O 11.12 Mon 17:45 H4

Broadband THz Non-Linear Response In Topological Noble Metal Dichalcogenides — George de Coster[1,2], Lucas Lafeta[3], Stefan Heiserer[1], Zdeněk Sofer[4], Achim Hartschuh[3], Georg Duesberg[1], and •Paul Seifert[1] — [1]Institute of Physics, University of the Bundeswehr Munich, EIT, Werner-Heisenberg-Weg. 39, 85577 Neubiberg, Germany — [2]DEVCOM Army Research Laboratory, 2800 Powder Mill Road, Adelphi, Maryland, United States — [3]Department of Chemistry and Center for NanoScience (CeNS), Ludwig-Maximilians-Universität München, Butenandtstraße 5-13 (E), 81377 Munich, Germany — [4]Department of Inorganic Chemistry, University of Chemistry and Technology Prague, Technická 5, 166 28 Prague 6, Czech Republic

Noble metal dichalcogenides belong to the material class of layered 2D materials and were shown to host type-II Dirac semi-metallic behavior, as well as topological surface states and superconductivity. Intriguingly, noble metal dichalcogenides display strong second harmonic generation and second order photocurrent response despite their centrosymmetric crystal structure. We investigate the spectrally-resolved optical response and reveal second and third order non-linear response at both, optical frequencies as well as in the THz range. The latter is analyzed in polarization resolved spectroscopy, that points towards spin-polarized bands at the symmetry broken surface as origin of the non-linearities. Our results elucidate the spectral opto- electronic response at low energies and discuss its anisotropy in light of underlying symmetry constraints.