O 60: Focus Session: Unoccupied States by Inverse Photoemission I

The Other Half of the Picture: 50 Years of Direct Access to Unoccupied States by Inverse Photoemission

In 1981, J. Pendry evaluated the experimental access to electron states in solids: "Currently only half of the picture can be seen with photoemission. Inverse photoemission provides the other half." A complete picture of electron states needs both occupied and unoccupied states in order to describe, understand, and finally tailor macroscopic material properties. In 1976, V. Dose had submitted a first paper on Bremsstrahlung Isochromat Spectroscopy in the VUV range: "The physics involved may be most simply described as an inverse photoelectric effect." The first experiments provided surface-sensitive information on the density of unoccupied states. Later, momentum and spin resolution were added to investigate the spin-dependent E(k) dispersion of unoccupied electron states. During five decades, the technique was further developed by several groups worldwide to enhance the intensity and improve the resolution in energy, momentum, and spin. A wealth of information was gained about metals, semiconductors, ultrathin films as well as adsorbate systems. The experimental studies were accompanied by several theoretical approaches, which are able to accurately describe the unoccupied electronic structure and model the inverse photoemission process. In 2012, H. Yoshida extended the energy range to the near-UV range (low-energy inverse photoemission), which is especially suited to study organic samples due to a lower damage risk caused by the exciting electron beam.

Current research fields for inverse photoemission are, e.g., spin textures of exchange- and/or spin-orbit-induced influenced systems and topological insulators, gap structures in transition metal dichalcogenides, LUMO levels in semiconductors for photovoltaic applications, electronic structure of atomic-layer and quantum materials. This focus session will highlight recent advances obtained by inverse photoemission in different fields and material systems. Also, it will bring together researchers from different areas for addressing current trends and future applications of inverse photoemission from experimental as well as theoretical perspective.

Organized by Markus Donath, Fabian Schöttke and Peter Krüger (U Münster).

Time: Wednesday 10:30–12:30 Location: WILL/A317

Invited Talk O 60.1 Wed 10:30 WILL/A317

Disentangling the origins of spin polarization in (inverse) photoemission from solid surfaces — •Jürgen Henk — Martin Luther University Halle-Wittenberg, Halle, Germany

The spin polarization of electronic states in solids can be probed by various photoemission-based techniques. We discuss how optical excitation modifies the spin polarization of the initial electronic states. Illustrative examples include Rashba-type surface states observed in photoemission (PE) from Au(111), the Dirac-like surface state in inverse photoemission (IPE) from W(110), and magnetic contrast in photoemission electron microscopy (PEEM) from Fe(001). Instead of employing a full group-theoretical analysis, we introduce a simplified symmetry-based approach that enables a clear separation of the different contributions to the photoelectron spin polarization—arising from light polarization, spin-orbit coupling, and intrinsic spin textures. This framework facilitates the interpretation of spin-resolved measurements at surfaces without relying on fully relativistic photoemission calculations.

O 60.2 Wed 11:00 WILL/A317

Inverse photoemission - just the inverse of photoemission? — •Markus Donath — University of Münster, Germany

Photoemission (PE) is known as the most powerful experimental technique to investigate the occupied energy levels of electrons in matter. The photoelectric effect, where electrons are detected after excitation with light, was first observed by H. Hertz and W. Hallwachs in 1887/1888. The reverse effect, the emission of Bremsstrahlung after irradiation of electrons was discovered by W.C. Röntgen in 1895. Bremsstrahlung spectroscopy provides access to unoccupied states. The first Bremsstrahlung spectroscopy experiment in the vacuum ultraviolet (VUV) range was performed by V. Dose fifty years ago [1]. In 1980, J.B. Pendry named this technique inverse photoemission (IPE) [2]. By using light in the VUV spectral range, PE and IPE became valuable complementary tools for studying the energy vs momentum dispersion of electron states below and above the Fermi level, even with spin resolution. After some historical remarks, this talk will review strengths and challenges as well as future perspectives of IPE.

[1] V. Dose, Appl. Phys. **14**, 117 (1977).
[2] J.B. Pendry, Phys. Rev. Lett. **45**, 1356 (1980).

O 60.3 Wed 11:15 WILL/A317

A Retrospective on Spin-Resolved IPES in Milan — •Alberto Calloni, Gianlorenzo Bussetti, and Franco Ciccacci — Dipartimento di Fisica, Politecnico di Milano, Piazza Leonardo da Vinci 32, 20133 Milano, Italy

For over thirty years, the Milan group led by Franco Ciccacci has been a key contributor to the development of spin-resolved Inverse Photoemission Spectroscopy (SR-IPES), advancing both the experimental methods and the study of empty electronic states in solids and interfaces. The activity began in the early '90s, when a dedicated system for spin- and angle-resolved measurements was conceived and built in Milan, drawing on the experience gained earlier with one of the first IPES setups using a UV grating. The new apparatus, featuring a spin-polarized GaAs electron source and a solid-state photon detector that ensured efficient detection of the weak IPES signal, enabled the direct investigation of spin-dependent unoccupied states at a time when such studies were still in their infancy. Building on this capability, the group has carried out a broad range of SR-IPES studies on metallic surfaces, magnetic thin films and multilayers, addressing phenomena such as spin-polarized quantum-well states and surface- or interface-induced spin polarization. More recently, together with studies on systems in which spin-orbit effects define the spin texture of the unoccupied states, the IPES setup has also been applied to hybrid organic/inorganic and molecule/ferromagnet interfaces, offering insight into the stability of the unoccupied states and their possible modifications upon interaction with the surrounding environment.

O 60.4 Wed 11:30 WILL/A317

Spin-Orbit Effects in Atomic-Layer Materials: Tl on Ag(111) — •Sven Schemmelmann[1], Peter Krüger[2], Patrick Härtl[3], Matthias Bode[3], Yuichiro Toichi[4], Kazuyuki Sakamoto[4], and Markus Donath[1] — [1]Physikalisches Institut, Universität Münster, Germany — [2]Institut für Festkörpertheorie, Universität Münster, Germany — [3]Experimentelle Physik II, Universität Würzburg, Germany — [4]Department of Applied Physics, The University of Osaka, Japan

Growing Tl on Ag(111) results in several structures of the Tl adlayers. For low coverage a $TlAg_2$ surface alloy is formed [1]. The Tl monolayer and bilayer exhibits moiré superstuctures with a continuous rotation of the Tl adlayer. Interestingly, all these structures exhibit characteristic spin-orbit-induced effects in their unoccupied electronic structure revealed by spin- and angle-resolved inverse photoemission. The surface alloy is predicted to exhibit states with a Rashba-type spin splitting.

In experiment, the intensity of these states is too low for a spin analysis [1]. In the Tl monolayer, spin-dependent hybridization of different overlayer states is observed [2]. Surprisingly, also an image-potential state located in front of the surface is found to hybridize with Tl states. The Tl bilayer, in contrast, exhibits a non-Rashba-like spin-polarized Fermi surface whose origin is found in the dispersion of the unoccupied states.

[1] Härtl *et al.*, Phys. Rev. B **107**, 205144 (2023)
[2] Schemmelmann *et al.*, Phys. Rev. B **109**, 165417 (2024)

O 60.5 Wed 11:45 WILL/A317

Surface state on Au(111): Shockley vs topology — •Patrick Geers, Fabian Schöttke, and Markus Donath — University of Münster, Germany

The L-gap surface state on Au(111) is the famous prototype for a Rashba-split Shockley surface state (SS). In 2015, Yan *et al.* posed the question whether Shockley-type SSs may be interpreted as variation of SSs in topological insulators, as both states exist in inverted band gaps [1]. The behaviour of the SS away from $\overline{\Gamma}$ is important to answer this question. It is crucial to know whether the SS potentially closes the inverted band gap between $\overline{\Gamma}$ and $\overline{\mathrm{M}}$. An analysis of not only the existence of SSs but also their spin textures is necessary to show a potential topologically nontrivial character of such SSs.
Inverse photoemission (IPE) as a technique to study the unoccupied electronic states is capable to measure these SSs with spin resolution at expected energies of about 4 eV above the Fermi level. In this talk, we present spin-resolved IPE results for Au(111) showing the energy vs momentum range between $\overline{\Gamma}$ and $\overline{\mathrm{M}}$, where the SS merges into the bulk bands.

[1] Yan *et al.*, Nat. Commun. **6**, 10167 (2015).

O 60.6 Wed 12:00 WILL/A317

Combining Spin Polarized Inverse Photoemission Spectroscopy with Angle-resolved X-ray Magnetic Circular Dichroism — •Takashi Komesu[1], Arjun Subedi[1], Bharat Giri[1], Ather Mahmood[1], Will Echtenkamp[1], Mike Street[1], Alpha N'Diaye[2], Xiaoshan Xu[1], Christian Binek[1], and Peter Dowben[1] — [1]University of Nebraska-Lincoln, Lincoln, NE, U.S.A — [2]Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, CA, U.S.A

While angle-resolved magnetometry and angle-resolved X-ray Magnetic Circular dichroism (XMCD) might provide hints of spin or moment canting at a surface, even XMCD taken in the total electron yield (TEY) mode cannot convincingly implicate the surface. Spin Polarized Inverse Photoemission Spectroscopy (SPIPES) is hugely surface sensitive, which is an advantage for characterizing boundary polarization of materials. With in-plane SPIPES, we have convincing proof of deviation from the expected perpendicular anisotropy for boron (B) doped Cr_2O_3(0001) and at the surface of $NiCo_2O_4$(001). For $NiCo_2O_4$(001), the Ni magnetic moments are canted slightly off the surface normal, as evident in elemental specific angle-resolved XMCD and consistent with the very surface sensitive SPIPES. In this presentation, we will show the great value of combining SPIPES with XMCD and magnetometry. Furthermore, we can provide an example where an exponential decay of spin polarization is not evidence of a finite paramagnetic correlation length, as in the case of thickness dependent Pd over layers on Cr_2O_3(0001), where Pd is no longer paramagnetic.

O 60.7 Wed 12:15 WILL/A317

On the sign of the Rashba parameter in image-potential states — •Fabian Schöttke[1], Kaishu Kawaguchi[2], Kenta Kuroda[3], Peter Krüger[1], Thorsten Deilmann[1], Sven Schemmelmann[1], Ayumi Harasawa[2], Shuntaro Tani[2], Yohei Kobayashi[2], Takeshi Kondo[2], and Markus Donath[1] — [1]Universität Münster, Germany — [2]Institute for Solid State Physics, The University of Tokyo, Japan — [3]Hiroshima University, Hiroshima, Japan

Image-potential surface states are simple model systems, where spin-dependent effects can be studied in view of spintronic applications. The Rashba effect in surface states at high-Z materials is related to both the strong spin-orbit interaction in heavy atoms and the orbital angular momentum arising from the inversion-symmetry breaking at the surface. Our studies with spin- and angle-resolved inverse photoemission and three-photon photoemission on image-potential states at Re(0001), Bi_2Se_3 and Bi_2Te_3 show a clear correlation between orbital angular momentum and spin directions. A key factor of our analysis is spin resolution, which enhances the effective resolution to separate spectral features considerably, provided they have different spin character.