

Chapter 2

Experiment

In the center of this thesis lies the unoccupied surface state of $\text{Ti/Si}(111)-(1 \times 1)$ and its spin texture. Experimental access to the unoccupied surface state is obtained via spin- and angle-resolved inverse photoemission. To shed light onto the spin texture, which forms complex patterns in momentum space, one of the project's main objectives has been to implement 3D spin analysis in the SR-IPE experiment to get access to all components of the spin polarization.

SR-IPE (Donath 1994, 1999) is the complementary method to spin- and angle-resolved photoelectron spectroscopy (SARPES) (Johnson 1997; Heinzmann and Dil 2012; Okuda and Kimura 2013). In SARPES, spin resolution is obtained in the detection channel with the help of spin-polarization detectors based on spin-orbit interaction, e.g., Mott scattering (Burnett et al. 1994) and spin-polarized low-energy electron diffraction (SPLEED) (Kirschner and Feder 1979; Yu et al. 2007), or exchange interaction, e.g., in very-low-energy electron diffraction detectors (Winkelmann et al. 2008; Okuda et al. 2008; Escher et al. 2011). In SR-IPE, spin resolution is achieved in the excitation channel by the use of spin-polarized electrons. Thus, to implement 3D spin analysis, spin-polarized electrons with variable polarization direction are needed.

In analogy to SARPES, where 3D spin detection is realized by using two spin-polarization detectors (Hoesch et al. 2002; Okuda et al. 2011), one could think of installing an additional electron source, but this requires a complicated setup with two lasers and two photocathodes. An alternative approach for spin manipulation has been realized before. Magnetic fields are used to alter the electron beam polarization direction, e.g., in spin-polarized electron sources for appearance potential spectroscopy (Rangelov et al. 2000) or spin-polarized low-energy electron microscopy experiments (Duden and Bauer 1995). This, however, is disadvantageous, in that magnetic stray fields affect the angular distribution of the electron beam and, as a consequence, limit the momentum resolution. Hence, spin-polarized electron sources

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for inverse photoemission have been restricted to one spin-polarization direction, so far. In this work, a **RO**tatable **S**pin-polarized **E**lectron source (ROSE) has been developed, which enables 3D spin analysis by embedding a variable rotation of the source chamber. It produces a transversally spin-polarized electron beam, whose polarization direction can be chosen freely. This allows investigations of the spin-dependent unoccupied electronic structure with sensitivity to two in-plane spin-polarization directions and, for nonnormal electron incidence, with sensitivity to the out-of-plane spin-polarization component. Importantly, as the pressure in the source chamber during rotation stays below 5×10^{-10} mbar, measurements with different sensitivities can be conducted at one and the same sample preparation.

In the following, an introduction to the basic concept of SR-IPE is given in Sect. 2.1. General aspects of the experimental setup and the most relevant techniques employed in this thesis will be briefly addressed in Sect. 2.2. The ROSE is presented in Sect. 2.3. Constructional details of the ROSE are described in Sect. 2.3.1, whereas the experimental characterization of the ROSE is part of Sect. 2.3.2. Finally, the performance of the SR-IPE experiment with the ROSE is demonstrated on the basis of two examples in Sect. 2.4: SR-IPE of ferromagnetic Ni/W(110) (Sect. 2.4.1) and SR-IPE of the prototypical Rashba-system Au(111) (Sect. 2.4.2).

2.1 Spin- and Angle-Resolved Inverse Photoemission

Inverse photoemission (IPE) can be viewed as Bremsstrahlung spectroscopy, where a solid is bombarded with low-energy electrons (Dose 1977, 1985; Smith 1988). IPE is a form of spontaneous emission and is often regarded as the time-reversed process to photoemission (PE). An electron in an excited state of the system decays into a state with lower energy and emits a quantum of energy of light. In IPE experiments, this is utilized to investigate the unoccupied electronic structure $E(\mathbf{k})$ of solids (Pendry 1980). In SR-IPE experiments, information on the spin-dependent surface electronic structure is obtained by resolving the energy E_{kin} , the angle of electron incidence θ , which is related to the momentum $\mathbf{k}$, and the spin polarization $\mathbf{P}$ of the electrons. SR-IPE becomes surface sensitive by the use of electrons with kinetic energies in the electron-volt range ($E_{\text{kin}} = 7 - 20$ eV), because of the shortness (≈ 10 Å) of the corresponding electron mean free path (Powell 1988). A detailed description of IPE and SR-IPE can be found in Dose (1985), Smith (1988), Himpsel (1990), Donath (1994) and Donath (1999). The most relevant aspects of IPE and SR-IPE for this thesis will be briefly described in the following:

The IPE process is schematically shown in Fig. 2.1a. An impinging electron with defined energy E_{kin} and momentum $\mathbf{k}$ couples to an unoccupied state of the sample, also referred to as initial state $|i\rangle$. Coupling is efficient, if the overlap of the free-electron plane wave and the wave function of the initial state is large. This explicitly depends on the symmetry of the initial state and is strongest for bands with nearly-free electron character (Dose 1985). The electron may then radiatively decay into a lower lying state, the final state $|f\rangle$. An IPE transition can be described within the

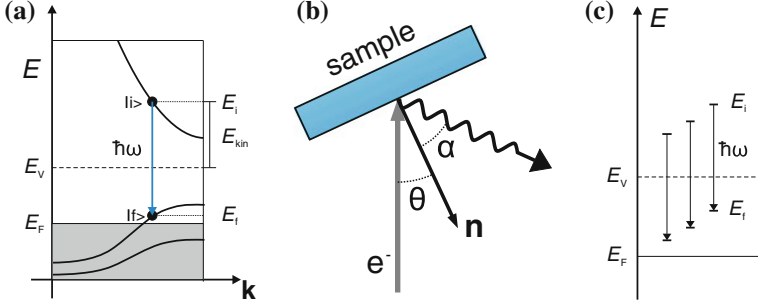


Fig. 2.1 **a** Illustration of the IPE transition in the reduced zone scheme. A free electron couples to an unoccupied state of the crystal, the initial state $|i\rangle$ and radiatively decays into a lower lying unoccupied state, the final state $|f\rangle$. The emitted photon has an energy of $\hbar\omega = E_i - E_f$. **b** Definition of the angle of electron incidence θ and the photon emission angle α with respect to the surface normal $\mathbf{n}$. **c** Visualization of the isochromat mode of the IPE experiment. The initial state energy E_i is swept, while the photon detection energy $\hbar\omega$ is kept constant. As a result, the experiment scans the final state energy E_f

dipole approximation. A specific dipole transition relates to a specific dipole axis, which leads to a distinct angular distribution of the photon emission (Eberhardt and Himpfel 1980; Desinger et al. 1984; Donath et al. 1986; Fauster et al. 1989). The transition probability from an initial state $|i\rangle$ to a final state $|f\rangle$ can be expressed via a transition matrix $\langle f | \mathbf{A} \cdot \mathbf{p} | i \rangle$, where $\mathbf{A}$ is the vector potential of the electromagnetic wave and $\mathbf{p}$ is the momentum operator (Eberhardt and Himpfel 1980).

Considering the conditions of conservation of energy, the energy E_i of the initial state is in a fixed relation to the energy of the emitted photon $E_{ph} = \hbar\omega$ and the final state energy E_f :

$$\hbar\omega = E_i - E_f = E_{kin} + E_V - E_f, \quad (2.1)$$

where E_i can be expressed by the kinetic energy E_{kin} of the free electron and the vacuum level E_V of the sample ($E_V - E_F$ is also referred to as the work function of the sample, where E_F is the Fermi level). Regarding the conservation of momentum, the momentum parallel to the surface plane $\mathbf{k}_{\parallel}$ is conserved but for reciprocal lattice vectors of the surface $\mathbf{g}_{\parallel}$. For $\mathbf{g}_{\parallel} = 0$, the absolute value of $\mathbf{k}_{\parallel}$ can be expressed as

$$k_{\parallel} = \sqrt{\frac{2m_e}{\hbar^2}} \cdot \sqrt{E_{kin}} \cdot \sin \theta = \sqrt{\frac{2m_e}{\hbar^2}} \cdot \sqrt{\hbar\omega + E_f - E_V} \cdot \sin \theta, \quad (2.2)$$

where m_e denotes the mass of a free electron, $\hbar$ the reduced Planck constant and θ the angle of electron incidence (see Fig. 2.1b). The momentum perpendicular to the surface $\mathbf{k}_{\perp}$ is affected by the vacuum-solid boundary and is not conserved.

Two-dimensional states such as surface states are independent of $\mathbf{k}_{\perp}$ and are completely characterized by the $E(\mathbf{k}_{\parallel})$ dispersion. This is directly accessible by the IPE experiment and can be obtained as follows. In this work, the IPE experiments are

operated in the isochromat mode (see Fig. 2.1c). The kinetic energy of the incident electrons and, thereby, E_i is swept, while the emitted photons are detected with a fixed mean detection energy. Thereby, the final state energy E_f is scanned. An IPE transition may be observed as a peak-like feature in photon yield (intensity) *versus* kinetic energy curves, in analogy to energy distribution curves in photoelectron spectroscopy experiments. By analyzing the energetic position E_f of the surface-state emission for different angles of electron incidence, $E(\mathbf{k}_{\parallel})$ is obtained with the help of Eq. 2.2. Here, three independent methods are used to obtain reliable values for E_f . (i) The positions E_f are determined by fitting a function $I = W * [(\sum_i L_i + B)(1 - f(E))]$ to the SR-IPE spectra, where W is the apparatus function, $f(E)$ the Fermi distribution, L_i the considered Lorentzian-shaped state emissions and B the background intensity (see also Stolwijk et al. 2010). The fit parameters are the energetic positions and the widths of the state emissions. Appropriate backgrounds are chosen taking also into account states at higher/lower energies. Variations of the background are considered to get a measure of the confidence interval. (ii) The minima of the second derivatives of the SR-IPE spectra are analyzed. Ideally, their energetic positions correspond to the energetic positions of the state emissions (see also Appendix A.2). Note that this procedure only leads to reasonable results as long as the background intensity can be treated as a polynomial up to the order E^2 . (iii) The positions E_f are determined by an estimation based on the experience of the experimentalist.

In systems where the spin degeneracy of the states is removed, e.g., due to ferromagnetism or SOI (see Sect. 1.2), the respective spin states $\mathbf{S}$ have to be considered. The spin-polarization expectation value $\bar{P} = \langle \mathbf{S} \rangle$ of $|f\rangle$ and $|i\rangle$ is generally less than unity (Henk et al. 1996), whereas $\bar{P}$ of the free electron is unity. Here, $\bar{P}$ is not to be mistaken as the spin-polarization value of the electron beam P , which is defined for the electron ensemble and is, typically, less than 100%. The spin-polarization direction of the impinging electrons determines the spin sensitivity of the SR-IPE experiment. In this context, an important aspect in SR-IPE measurements deals with the presentation of the gathered data. To account for an incomplete electron beam polarization P , the raw data have to be normalized to 100% beam polarization. Additionally, one has to consider the angle Φ between the quantization axis of the electron spin polarization and the spin quantization axis of the sample. The raw data are then normalized to 100% effective beam polarization, see for instance in Donath (1994). If $N_{\uparrow}$ and $N_{\downarrow}$ are the raw data of the two spin channels, the normalized data $I_{\uparrow}$ and $I_{\downarrow}$ are determined by

$$I_{\uparrow,\downarrow} = \frac{N_{\uparrow} + N_{\downarrow}}{2} \cdot \left(1 \pm \frac{N_{\uparrow} - N_{\downarrow}}{N_{\uparrow} + N_{\downarrow}} \cdot \frac{1}{P \cos \Phi} \right).$$

Normalization is not possible, if Φ is not known. In the case that the measurement is simultaneously sensitive to two spin-polarization directions, additional information is mandatory. Additional information from other experiments or symmetry arguments can help to exclude the existence of one of the possible polarization directions. Note

that for SARPES experiments the same considerations apply. There, the raw data is normalized to the effective Sherman function S of the spin-polarization detector.

Conclusively, 3D spin analysis in SR-IPE has to be achieved in the excitation channel by the use of electron beams with variable spin-polarization direction. The following sections are concerned with the experimental realization.

2.2 Experimental Setup

The employed SR-IPE experiment is part of a multi-chamber ultra-high vacuum system, which is schematically shown in Fig. 2.2. Several publications give a detailed description and demonstrate the performance of this setup (Yu et al. 2007; Budke et al. 2007a,b; Budke and Donath 2008, 2009; Allmers and Donath 2009; Stolwijk et al. 2010; Allmers and Donath 2010; Allmers et al. 2011). Basically, it consists of three units (see Fig. 2.2a): the preparation chamber (P), the microscopy chamber (M) and the analysis chamber (A). The chambers are connected via a sample transfer system.

The preparation chamber is equipped with standard tools of surface science for film growth and film characterization. It provides several evaporators and an ion sputter gun. The manipulator accepts Omicron flag-style sample plates and allows sample

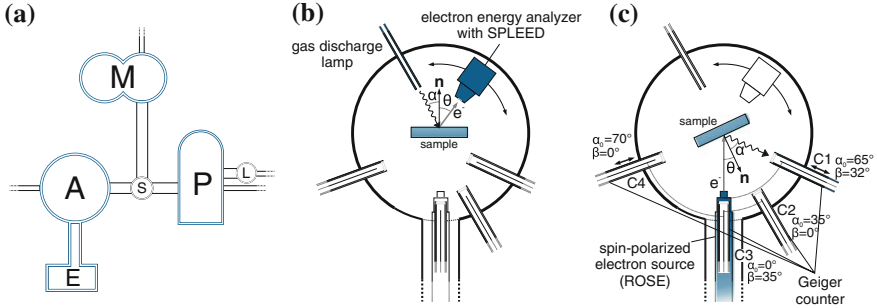


Fig. 2.2 **a** Schematic drawing (not to scale) of the multi-chamber ultra-high vacuum system with the preparation chamber (P), the microscopy chamber (M), the analysis chamber (A) and the electron source chamber (E). The chambers are connected via a sample transfer system. Samples can be inserted into the system via a load lock (L) and stored in UHV (S). **b** (S)ARPES equipment (not to scale). The (S)ARPES experiment consists of a gas discharge lamp and an electron-energy analyzer/SPLEED combination. The polar (azimuthal) angle of photon incidence with respect to the surface normal $\mathbf{n}$ is $\alpha = 30^\circ$ ($\beta = 35^\circ$). The electron-energy analyzer is mounted on a goniometer to vary the angle of electron emission θ . **c** The SR-IPE setup (not to scale) includes the spin-polarized electron source (shown is only the last part of the electron-transfer optics) and four Geiger counters (C1, C2, C3 and C4). The photon detectors are fixed, while the sample is rotated to vary the angle of electron incidence θ . The polar angle of photon detection α of the counters depends on θ . For $\theta = 0^\circ$, the photon detection angles are denoted as α_0 . β denotes the azimuthal angle of photon detection. Data shown in this thesis were mainly detected with counter C1

cooling to 115 K with liquid nitrogen and indirect heating up to 1200 K. To enable fast heating to high temperatures (flashing) up to 2500 K, the chamber is equipped with an electron bombardment station (Stolwijk 2009). Moreover, the preparation chamber includes sample characterization techniques such as low-energy electron diffraction (LEED) (Kittel 1967; Kopitzki 1989; Henzler and Goepel 1994), medium-energy electron diffraction (MEED),¹ Auger electron spectroscopy (AES) (Auger 1925; Linsmeier 1994) and an in-situ magneto-optical Kerr-effect (MOKE) setup (Kerr 1877; Polisetty et al. 2008; Allmers and Donath 2010).

The microscopy chamber features a scanning tunneling microscope from Omicron NanoTechnology GmbH. A detailed introduction to STM can be found in Binnig and Rohrer (2000) and Chen (2008). In this work, STM is used to characterize the topography of the studied films.

At the heart of the experiment lies the analysis chamber with a unique combined setup for direct and inverse spin- and angle-resolved photoemission (Budke et al. 2007a). In the course of this work, the analysis chamber has been equipped with a 5-axis manipulator from VG Scienta (Cryoax 5 with Omniax 200 mm Z slide module and ± 25 mm XY stage) with nonmagnetic Omicron flag-style sample holder (SH2). It enables cooling of the sample to 60 K or to 120 K by the use of liquid helium or liquid nitrogen, respectively. Indirect heating to 1200 K is possible. A key feature of the manipulator is the possibility to rotate the sample about two axes. Rotation about the manipulator axis is denoted as primary rotation θ . Rotation about the surface normal varies the azimuth φ of the sample. The equipment for the (i) SARPES and (ii) SR-IPE experiment is schematically illustrated in Fig. 2.2b, c, respectively.

- (i) The SARPES setup consists of a HIS 13 gas discharge lamp from Focus GmbH (Schönhense and Heinzmann 1983) and an electron-energy analyzer (50 mm hemispherical sector analyzer SHA50 from Focus GmbH). The analyzer is mounted on a goniometer to change the electron-emission angle without changing the incidence angle of light. The experiment can be operated in a spin-resolved (SARPES) and a spin-integrated mode (ARPES). Spin resolution is achieved with a SPLEED detector (Focus GmbH) (Kirschner and Feder 1979; Yu et al. 2007) attached to the analyzer. The Sherman function S of the SPLEED detector is about $S = 0.24$ (Budke et al. 2007a). The energy resolution ΔE_{PE} of the ARPES/SARPES experiment depends on the pass energy of the electron analyzer and ranges from 50 to 160 meV full width at half maximum (FWHM). The angular resolution $\Delta\theta_{PE}$ is approximately $\Delta\theta = 2^\circ$ (FWHM) (Budke et al. 2007a; Allmers 2009).
- (ii) The SR-IPE experiment includes four bandpass photon detectors (C1-C4), i.e., Geiger counters using acetone as counting gas and CaF_2 entrance windows (Budke et al. 2007b; Thiede 2010). This combination creates an optical bandpass, which depends on the temperature T_{CaF_2} of the CaF_2 entrance window. At room temperature, a bandpass of about 320 meV (FWHM) with a mean detection energy of 9.9 eV is obtained. Two detectors (C2 and C4) possess heatable

¹ Similar to high-energy electron diffraction (Peng et al. 2011).

CaF₂ entrance windows to optimize the total energy resolution of the SR-IPE experiment. A characterization of the bandpass and the energy resolution will be shown in Sect. 2.3.2. Additionally, two detectors (*C1* and *C4*) are mounted on linear drives to allow a close approach to the sample. Thereby, the solid angle of photon collection is enlarged and the photon yield increased. To measure angular-dependent photon yields, the detectors are positioned at various angles with respect to the surface normal as denoted in Fig. 2.2c. The spin-polarized electron source for the SR-IPE experiment, the ROSE, is located in the source chamber (E), which is connected with the analysis chamber via electron transfer optics. A detailed discussion of the ROSE is presented in the following section.

2.3 The Rotatable Spin-Polarized Electron Source

2.3.1 Constructional Details and Quantization Axes

Briefly, the principle of the ROSE is as follows. A circularly polarized laser beam excites spin-polarized electrons at an activated GaAs photocathode as described in Pierce and Meier (1976) and Pierce et al. (1980). These are extracted by an emission lens and guided through a toroidal electrostatic deflector with a slit aperture at the focal point of the deflector exit. The deflector and slit form the energy-selective element. Additionally, the spin polarization of the electron beam is changed from initially longitudinal to transversal. After passing the slit aperture, the electrons enter the transfer part of the electron optics and are guided through a gate valve into the analysis chamber onto the sample. The gate valve is located in the middle of the transfer optics and can separate the source chamber from the analysis chamber. This is necessary to avoid contamination of the sample during the preparation procedure of the GaAs photoemitter and vice versa.

Up to now, spin-polarized electron sources for inverse-photoemission experiments provided only one fixed polarization direction, with the polarization vector being either perpendicular (Wissing et al. 2013) or parallel (Donath 1994) to the plane of incidence, as shown in Fig. 2.4c, d, respectively. Here, a straightforward way to establish an in-situ variation of the transverse polarization of the electron beam is realized by introducing a rotatable source chamber. For this purpose, two major modifications have been applied to the previous electron source (Budke et al. 2007b).

(i) To implement the gate valve within the electron transfer optics, in previous electron sources a removable screening tube at lens potential was used to bridge the gap between the two parts of the lens element E_2 (see left hand side of Fig. 2.3a and Kolac et al. 1988). In the course of the electron source reconstruction, this mechanical system was replaced by two graphite-coated circular condenser plates at the ends of the lens element E_2 , as shown in Fig. 2.3a (right-hand side). This has two advantages: First, the mounting of a removable screening tube and its supporting guide rail in combination with rotatable optic parts is not a trivial task. Second, the

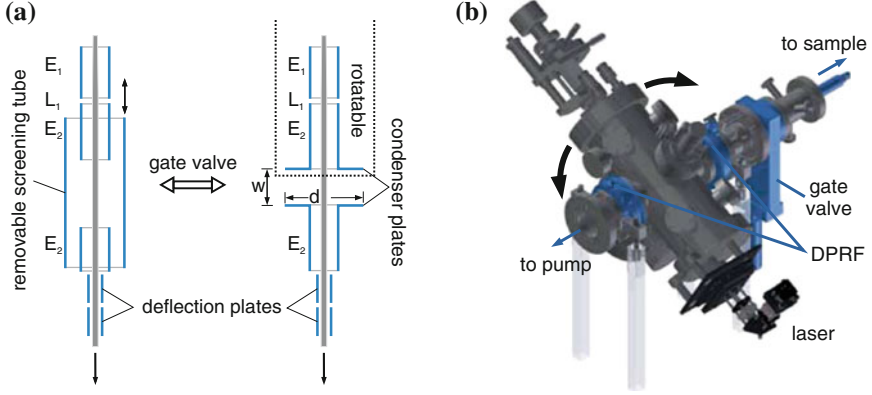


Fig. 2.3 **a** Schematic drawing of the transfer optics of the ROSE in the vicinity of the gate valve. Former (*left-hand side*) and new design (*right-hand side*) with screening tube and condenser plates, respectively. The upper part of the electron optics (indicated by the *dotted box*) is rotated along with the source chamber. **b** Technical drawing of the ROSE. The differentially pumped rotary feedthroughs (DPRF), the gate valve and the electron optics are colored in *blue*. The source chamber can be rotated along the transfer-optics axis as illustrated by the *black arrows*

new design abstains from moving mechanical parts, which may become a source of failure. To ensure a homogeneous electrical field in the region of the electron beam, the diameter of the condenser plates is maximized ($d = 60 \text{ mm}$) and the distance between the condenser plates is as small as possible ($w = 27 \text{ mm}$), while still leaving enough room for the gate-valve plate. The beam properties were left unchanged by this modification. At the same time, the gap in the transfer optics constitutes a constructional separation and, therefore, a rotation of the two parts relative to each other is unproblematic. In the setup presented in the scope of this thesis, the optics part attached to the source chamber is rotated with the source chamber, while the other part remains fixed.

(ii) The source chamber is attached between two differentially pumped rotary feedthroughs (DPRF) as shown in Fig. 2.3b, which carry the load of the source chamber. It should be noted that this setup demands for a highly accurate alignment of the source-chamber flanges and rotary drives, as irregular axial stress would likely break the sealing of the differentially pumped rotary drives. Therefore, prior to mounting, the source chamber flanges have been mechanically aligned to be parallel with an accuracy of below 0.1° . In addition, two compensation elements in the form of bellows are embedded to equalize small tilts. As a result, one is able to perform a continuous rotation of the source chamber around the transfer optics axis. Since the part of the transfer optics up to the gate valve is attached to the rotary drive, it is rotated as well. This motivated the modification described in (i). The rotary drives are attached to a turbo-molecular pump/rotary pump combination to ensure UHV conditions in the source chamber. The base pressure of the electron source is less than $1 \times 10^{-10} \text{ mbar}$ and stays below $5 \times 10^{-10} \text{ mbar}$ during rotation. Hence, the GaAs

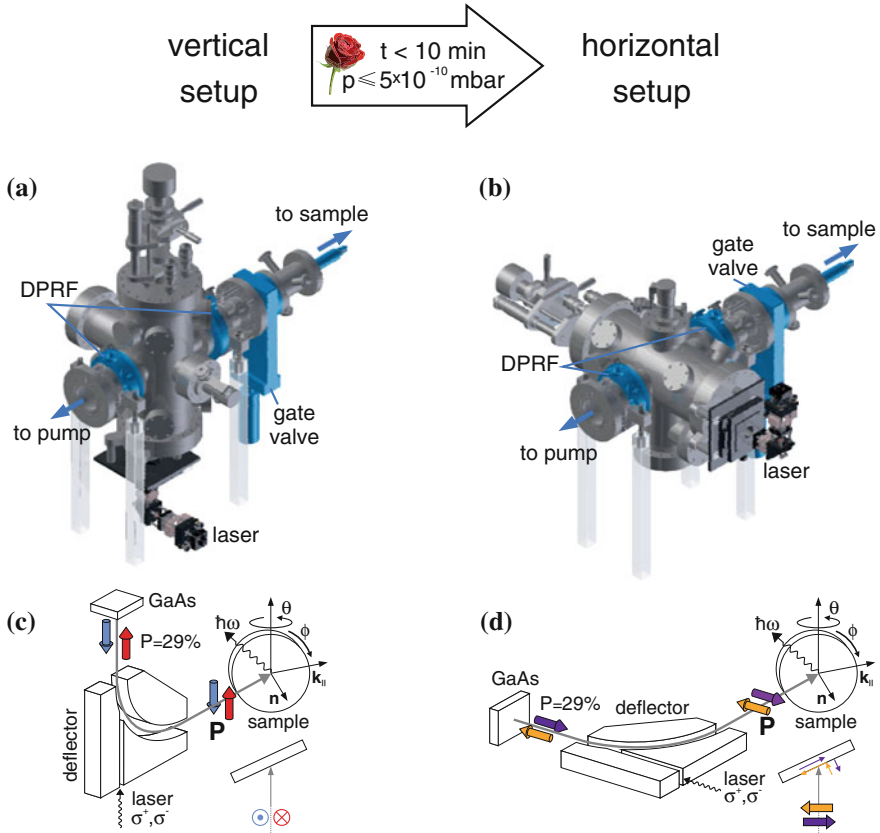


Fig. 2.4 **a, b** Technical drawing and **c, d** schematic view of the ROSE in **(a, c)** vertical and **(b, d)** horizontal setup. The schematic representation of the **c** vertical and **d** horizontal setup of the ROSE illustrates the spin-polarization direction of the electron beam, which is perpendicular (vertical setup) or parallel (horizontal setup) to the plane of incidence. Sensitivity to the in-plane spin-polarization direction, i.e., parallel to the surface and perpendicular (vertical setup) or parallel (horizontal setup) to the plane of incidence is obtained. Additionally, in the horizontal setup for $\theta \neq 0^\circ$, out-of-plane sensitivity is realized

photocathode and, for the case that the gate valve is open, the sample condition remain unaffected.

By selecting the vertical and horizontal orientation of the electron source as shown in Fig. 2.4a, b, respectively, it is possible to analyze two orthogonal in-plane spin-polarization directions. This becomes clear in the schematic view of the vertical and horizontal setup (see Fig. 2.4c, d). In the vertical (horizontal) setup, the SR-IPE experiment is sensitive to the in-plane spin-polarization direction perpendicular (parallel) to the plane of incidence, which is denoted as in-plane_⊥ (in-plane_∥) direction. In the case of horizontal orientation and nonnormal electron incidence, i.e., $\theta \neq 0^\circ$, the experiment is simultaneously sensitive to the in-plane_∥ and the out-of-plane

spin-polarization component (see polarization components in Fig. 2.4d). Switching between horizontal and vertical position takes about five to ten minutes time. After rotation, only minor corrections have to be made to the electron beam alignment.

2.3.2 Characterization

In this section the inverse-photoemission experiment using the ROSE is put to the test and the essential parameters of the experiment are determined: (i) the spin polarization P , (ii) the width of the electron-energy distribution ΔE_{el} , (iii) the angular resolution $\Delta\theta$, (iv) the spot size d_{el} and (v) the total energy resolution ΔE of the SR-IPE experiment. It is emphasized that a rotation of the ROSE does not vary these values.

(i) The spin polarization P of the electron beam is determined with the SPLEED detector of the photoemission setup as sketched in Fig. 2.5a, b. The SPLEED detector is sensitive to the spin-polarization direction of the electron beam parallel to the plane of incidence (in-plane_{||}) according to Fig. 2.4d. Figure 2.5c shows the spin-resolved intensity distribution (the SPLEED channeltron intensities N_1 and N_2) of the electron beam with arbitrarily chosen energy E_0 . The obtained spin asymmetry $A = SP = 0.071$ is composed of P and the Sherman function S of the detector. With the analysis employed in Budke et al. (2007a) a spin polarization of $P = 29\%$ is found, which is in agreement with typical values of GaAs photocathodes.

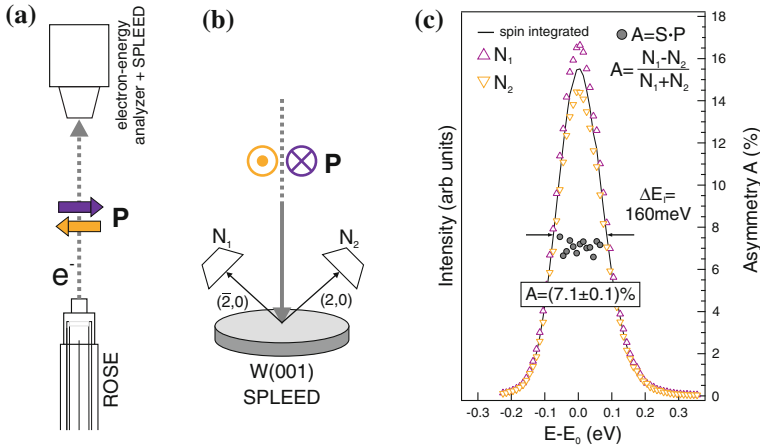


Fig. 2.5 **a** Calibration of the electron beam spin polarization with the SARPES setup. The electron beam from the ROSE is guided to the entrance of the electron-energy analyzer. **b** Subsequent to the analysis of the electron energy, the electron beam is guided onto the W(001) crystal of the SPLEED detector. **c** Spin-resolved intensity distribution (purple up- and orange down-pointing triangles) and asymmetry A (gray markers) of the electron beam measured with the SPLEED detector

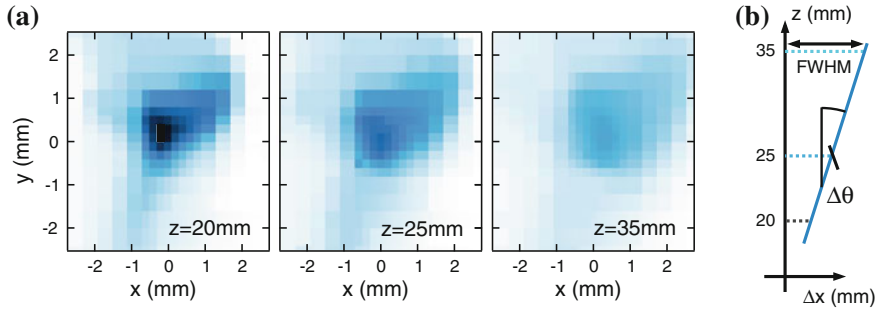


Fig. 2.6 a Intensity maps of the electron beam for different positions z behind the last lens element of the ROSE. To determine the angular resolution $\Delta\theta$, the width w (FWHM) of the electron beam is analyzed. The slope a of $w(z)$ relates to the angular resolution $\Delta\theta = \arctan(a)$

(ii) The width of the electron-energy distribution ΔE_{el} is determined by analyzing the width ΔE_I of the intensity distribution from the measurement in Fig. 2.5c. ΔE_I is a convolution of ΔE_{el} with the energy resolution of the electron analyzer ΔE_A : $\Delta E_I^2 = \Delta E_{\text{el}}^2 + \Delta E_A^2$. With $\Delta E_I \approx 160\text{ meV}$ (FWHM) (see Fig. 2.5c) and $\Delta E_A \approx 95\text{ meV}$ (FWHM), one obtains $\Delta E_{\text{el}} \approx 129\text{ meV}$.

(iii) The momentum resolution $\Delta \mathbf{k}$ of the inverse-photoemission experiment solely depends on the beam divergence of the electron beam. In order to get a measure of the angular resolution $\Delta\theta$, the intensity distribution of the electron beam is mapped with a Faraday cup at three different distances z behind the last lens element (see Fig. 2.6a). For each position, the width of the electron beam w (FWHM) is extracted. $\Delta\theta$ is derived as schematically shown in Fig. 2.6b. Deviations from a Gaussian-shaped profile are observed and interpreted as a consequence of astigmatism of the electron transfer optics. The data reveal a beam divergence of $\pm 1.5^\circ$, i.e., a total angular resolution of $\Delta\theta = 3^\circ$, which corresponds to a resolution of the wave vector parallel to the surface at the Fermi level and normal electron incidence of $\Delta k_{\parallel, \text{F}} \approx 0.06\text{ \AA}^{-1}$.

(iv) An ideal electron source would be a point source, followed by an electron optics without lens aberrations. In this case, a truly parallel electron beam can be formed. A real electron source, however, suffers from a finite emission spot (here given by the size of the laser spot), lens aberrations and electron-electron Coulomb repulsion, which becomes increasingly important for low beam energies (here 7–20 eV) and high current densities. Therefore, the beam is optimized with respect to parallelism, i.e., high momentum resolution, at the cost of a small beam size. A beam size d_{el} of less than 2 mm (FWHM) is achieved at the sample located at $z = 25\text{ mm}$ with beam currents of a few μA (see Fig. 2.6a). For smaller samples, apertures can be used at the last lens element to restrict the beam diameter.

(v) The total energy resolution ΔE of the employed SR-IPE experiment is given by the energy distribution of the electron beam and the width of the bandpass photon detector, the Geiger counter. Experimentally, ΔE is determined by a measurement of the Fermi edge at a polycrystalline Ta sample plate. The energy resolution is derived

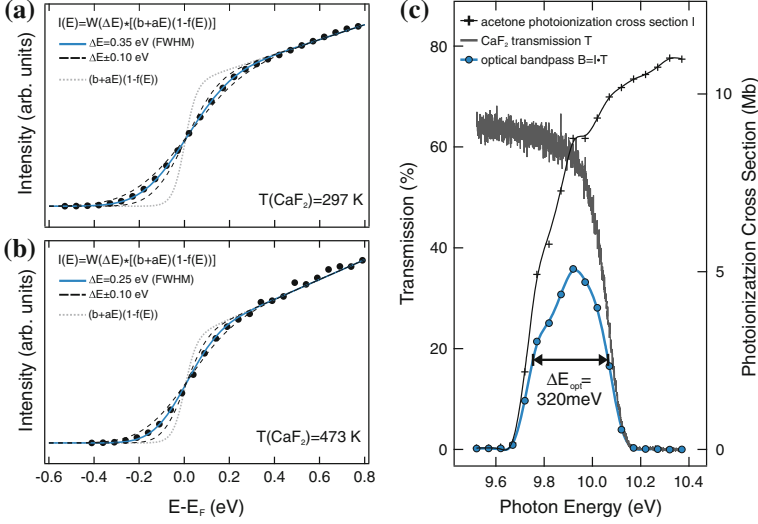


Fig. 2.7 Measurements of the Fermi edge at a polycrystalline Ta sample for **a** $T_{\text{CaF}_2} = 297 \text{ K}$ and **b** $T_{\text{CaF}_2} = 473 \text{ K}$ of the entrance window of the Geiger counter. *Solid lines* represent simulated spectra $I(E)$ to deduce the total energy resolution ΔE of the SR-IPE experiment. *Dashed lines* show simulated spectra with higher or lower energy resolution to represent confidence intervals. **c** Determination of the optical bandpass with the help of the acetone photoionization cross section I (black markers and line) from Cool et al. (2005) and the transmission cut-off T of the CaF_2 entrance window (gray line) at room temperature. The bandpass (blue markers and line) is obtained via $B = I \cdot T$ and possesses a width (FWHM) of about $\Delta E_{\text{opt}} = 320 \text{ meV}$

by simulating the obtained spectra as discussed in Budke et al. (2007b). It is assumed that the observed spectral intensity $I(E)$ is composed of a constant background b plus a linear slope $a(E)$, multiplied by 1 minus the Fermi function $f(E)$ and convoluted with the respective apparatus function W : $I(E) = W * [(b + aE)(1 - f(E))]$. The full width at half maximum of W represents the energy resolution ΔE of the experiment and is varied to fit the spectra. In the presented setup, to improve the total energy resolution, the optical bandpass of the Geiger counter can be reduced by heating its CaF_2 entrance window (Budke et al. 2007b). Figure 2.7a, b shows the Fermi edge of the polycrystalline Ta for $T_{\text{CaF}_2} = 297 \text{ K}$ and $T_{\text{CaF}_2} = 473 \text{ K}$, respectively. A total energy resolution of 350 and 250 meV is obtained, respectively, in agreement with values found prior to the applied modifications discussed in Sect. 2.3.1. The optical bandpass is determined by the ionization threshold of the acetone gas (see, e.g., Cool et al. 2005) and the transmission cut-off of the employed CaF_2 window.² The multiplication of the threshold with the transmission cut-off represents the optical bandpass (see Fig. 2.7c). At $T_{\text{CaF}_2} = 297 \text{ K}$, ΔE_{opt} amounts to $\Delta E_{\text{opt}} \approx 320 \text{ meV}$

²The transmission cut-off of the CaF_2 window used in this counter was measured in a separate setup (Thiede 2010).

(FWHM). Accordingly, the width of the electron energy distribution is determined to $\Delta E_{el} = (140 \pm 20)$ meV, in agreement with ΔE_{el} found in (ii).

2.4 Performance

Experiments on Ni/W(110) and Au(111) demonstrate the performance of the SR-IPE experiment with the ROSE.

2.4.1 Ni/W(110)

The variable spin sensitivity of the ROSE is demonstrated by means of SR-IPE measurements on the spin-dependent surface electronic structure of a Ni(111) film on W(110). A Ni(111) film with a thickness of 20 monolayers was epitaxially grown on a W(110) single crystal, which had been cleaned by several cycles of heating to 1500 K for 10 min at an oxygen partial pressure of 5×10^{-8} mbar and subsequent flashing to 2300 K. Annealing of the Ni film up to 600 K for 10 min produced a well ordered surface with a sharp diffraction pattern (see Fig. 2.8a). The Ni(111) film is magnetized along the in-plane easy axis $[\bar{1}10]$ ($\bar{\Gamma}\bar{K}$ direction) (Farle et al. 1990) between two coils with a total magnetic field strength of 500 G. This leads to a remanently magnetized film as indicated by the in-situ magneto-optical Kerr-effect measurement (see Fig. 2.8b).

With the state of the film magnetization exactly known, i.e., fixed quantization axis along $\bar{\Gamma}\bar{K}$, the spin-polarization directions of the electron beam can be determined with the help of SR-IPE measurements. The basic approach is as follows. A SR-IPE spectrum is measured at a fixed angle of electron incidence $\theta = 40^\circ$ under four different conditions. For the first two measurements, the ROSE is in the vertical position, i.e., the spin polarization is perpendicular to the plane of incidence. In this

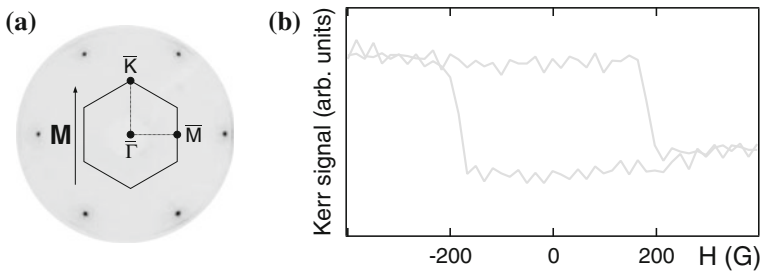


Fig. 2.8 **a** Low-energy electron diffraction image of the Ni(111) film and **b** magneto-optical Kerr-effect measurement (MOKE) with the magnetization direction along $\bar{\Gamma}\bar{K}$

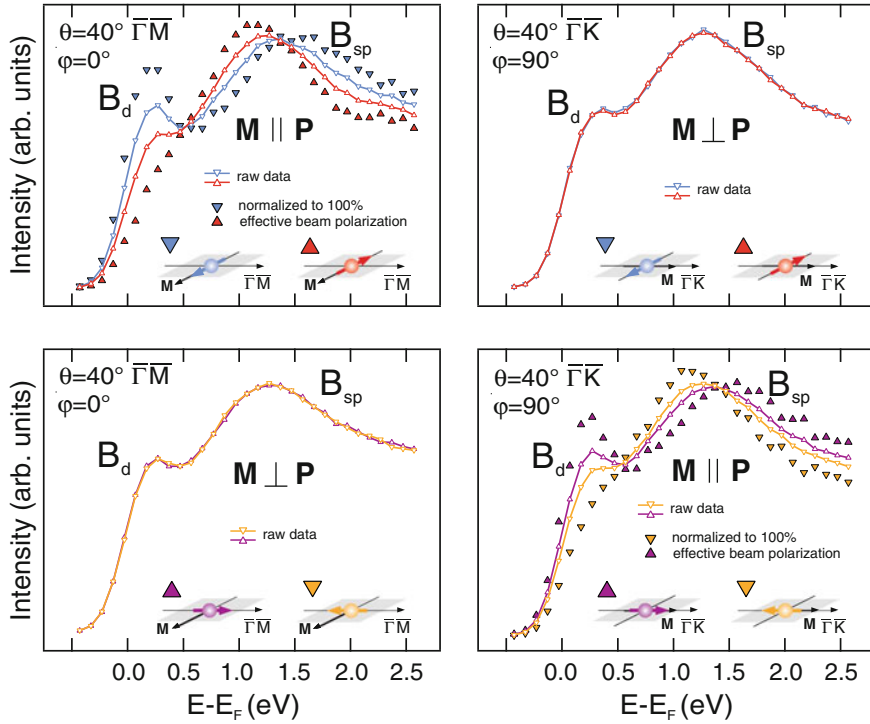


Fig. 2.9 SR-IPE spectra at $\theta = 40^\circ$ on Ni/W(110) magnetized along $\bar{\Gamma}\bar{K}$ with the magnetization direction $\mathbf{M}$ pointing perpendicular (*left spectra*) or parallel (*right spectra*) to the plane of incidence. Measurements have been conducted with the ROSE in vertical (*upper spectra*) and horizontal setup (*lower spectra*) realizing conditions, where the effective beam polarization $\mathbf{P}_{\text{eff}}$ is parallel or perpendicular to $\mathbf{M}$

geometry, spectra are taken with the magnetization direction $\mathbf{M}$ perpendicular and parallel to the plane of incidence, simply by varying the azimuth φ of the sample. Corresponding measurements are conducted with the ROSE in the horizontal position. Note that in the latter case the effective beam polarization is reduced by $\cos 40^\circ$. For the electron beam spin polarization $\mathbf{P}$ being parallel to the magnetization direction ($\mathbf{M} \parallel \mathbf{P}$), spin-dependent features are expected, whereas in the case of perpendicular alignment ($\mathbf{M} \perp \mathbf{P}$) no polarization should be observed.

The spectra are shown in Fig. 2.9. Small red up- and blue down-pointing triangles and small purple up- and orange down-pointing triangles represent the raw data obtained with the ROSE in vertical and horizontal position, respectively. The normalized data are shown as large colored triangles. In each spectrum, a bulk d -like state B_d and a bulk sp -like state B_{sp} is observed, in agreement with SR-IPE data of Ni(111) bulk single-crystals (Donath 1994).

With the magnetization direction and electron spin polarization pointing perpendicular to the plane of incidence ($\mathbf{M} \perp \mathbf{P}$, upper left spectrum), clearly, the minority

character of B_d and the exchange splitting of B_{sp} are identified. By changing the azimuth of the sample, so that the magnetization direction lies now in the plane of incidence ($\mathbf{M} \perp \mathbf{P}$, upper right spectrum), spin sensitivity is lost. With a rotation of the ROSE to the horizontal position spin sensitivity is regained ($\mathbf{M} \parallel \mathbf{P}$, lower right spectrum) and lost again if the magnetization direction is set to be perpendicular to the plane of incidence ($\mathbf{M} \perp \mathbf{P}$, lower left spectrum). Notably, the spin-integrated spectra along corresponding high-symmetry lines are almost identical. In particular, considering the steep dispersion of B_{sp} (Donath 1994), a rotation of the ROSE, therefore, does not significantly alter the angle of electron incidence. Also the sample condition is not affected. Evidently, the ROSE gives access to two orthogonal in-plane spin-polarization directions, the in-plane $_{\perp}$ and the in-plane $_{\parallel}$ spin-polarization direction.

2.4.2 *Au(111)*

The study of the Au(111) surface highlights the importance of a correct electron beam and sample alignment, when investigating samples with spin textures that are more complex than in ferromagnetic systems, such as Ni/W(110), with a single spin quantization axis along the magnetization direction.

The surface electronic structure of Au(111) is discussed in several theoretical and experimental studies and is the prototypical Rashba system (Paniago et al. 1995; LaShell et al. 1996; Nicolay et al. 2001; Henk et al. 2003; Reinert 2003; Hoesch et al. 2004; Henk et al. 2004a, b). It features a nearly-free-electron-like dispersing spin-orbit-split Shockley surface state, which exhibits a spin texture as predicted by the Rashba-Bychkov model. Recently, SR-IPE investigations confirmed the Rashba-type spin texture of the unoccupied part of the Au(111) surface state (Wissing et al. 2013). Deviations from a nearly-free-electron-like dispersion occur as soon as the surface state approaches the gap boundary and becomes a surface-resonant state. Figure 2.10a shows a sketch of the $E(\mathbf{k}_{\parallel})$ dispersion of the Au(111) surface state in the vicinity of $\bar{\Gamma}$, where only minor deviations from a parabolic dispersion occur.

As a first step, the spin-orbit-split surface state is identified with the help of SR-IPE measurements for three angles of electron incidence θ along $\bar{\Gamma}\bar{M}$ as illustrated in Fig. 2.10a: $\theta = 14^\circ$, 7° and 0° . Prior to the experiment, the Au(111) surface was cleaned by several cycles of sputtering and annealing, whereas the cleanliness and crystalline quality of the surface were checked with AES, LEED, and STM. Figure 2.10b presents the SR-IPE data. The angle of electron incidence θ has been calibrated with the help of additional SR-IPE measurements (see Fig. 2.10c). The sensitivity of the experiment is set to the in-plane $_{\perp}$ spin-polarization direction as denoted by the red up- and blue down-pointing triangles. Spectra have been normalized to 100 % in-plane $_{\perp}$ spin polarization of the electron beam.

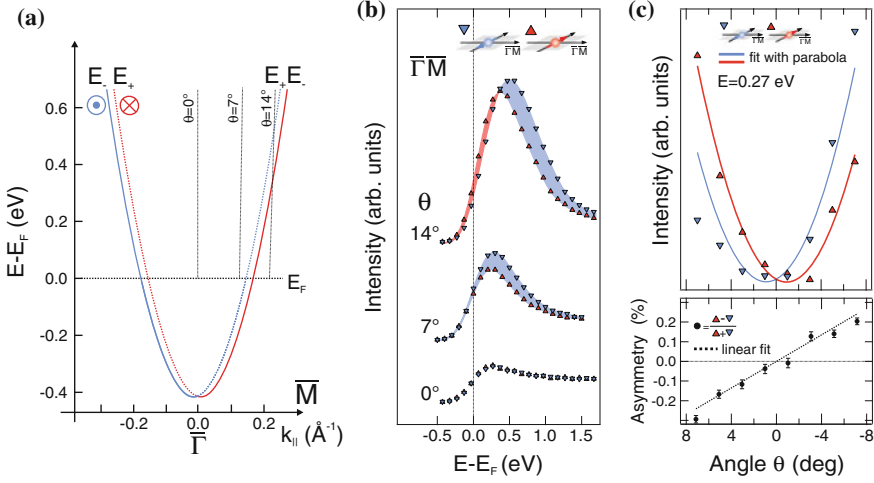


Fig. 2.10 **a** Sketch of the Au(111) Shockley surface state in the vicinity of the $\bar{\Gamma}$ point along $\bar{\Gamma}\bar{M}$. The spin-polarization direction of the two spin-orbit-split surface-state components is in-plane and perpendicular to $\mathbf{k}_{||}$ (in-plane $_{\perp}$). SR-IPE measurements at constant angles of electron incidence θ scan the surface state as illustrated by the *dotted lines*. **b** SR-IPE measurements of the Au(111) surface with sensitivity to the in-plane $_{\perp}$ spin-polarization direction along $\bar{\Gamma}\bar{M}$ for $\theta = 0^\circ, 7^\circ$ and 14° . **c** (*upper graph*) Spin-resolved intensity at $E = 0.27$ eV for different angles of electron incidence θ and (*lower graph*) according to spin-asymmetry data

- (i) At $\theta = 14^\circ$, both spin-orbit-split surface-state components E_+ and E_- are completely unoccupied. These can be clearly observed in the SR-IPE spectra. The energy splitting is approximately $\Delta E \approx 150$ meV.
- (ii) The spin-orbit-split surface-state components become unoccupied for momenta parallel to the surface of about $k_{||,+} \approx 0.15 \text{ \AA}^{-1}$ and $k_{||,-} \approx 0.18 \text{ \AA}^{-1}$, respectively. At $\theta = 7^\circ$ ($k_{||,+,-} \approx 0.13 \text{ \AA}^{-1}$), both surface-state components are located below E_F . However, spectral intensity close to E_F is observed. This is a consequence of the limited momentum resolution in combination with the Fermi-level cut-off and the finite lifetime of the surface state of the SR-IPE experiment (Wissing et al. 2013). In addition, the observed peak positions close to E_F are influenced by the limited energy resolution and do not necessarily represent the final-state energies (Donath 1989; Budke et al. 2007a; Stolwijk et al. 2010). The observed intensity difference between both spin channels is owed to the fact that one spin component is located closer to E_F than the other.
- (iii) At $\theta = 0^\circ$ ($\bar{\Gamma}$), the surface-state components are degenerate and completely occupied. In the SR-IPE spectra, a small spectral intensity close to the Fermi level remains. In agreement with the Rashba-Bychkov model, no in-plane $_{\perp}$ spin polarization is detected. Note that the observation of spin asymmetry close to the Fermi level is highly sensitive to slight variations of θ , e.g., for -2° and 3° intensity asymmetries are detected, due to the same reasons as discussed for $\theta = 7^\circ$ (Wissing et al. 2013). In return, this can be used to calibrate θ . Figure 2.10c

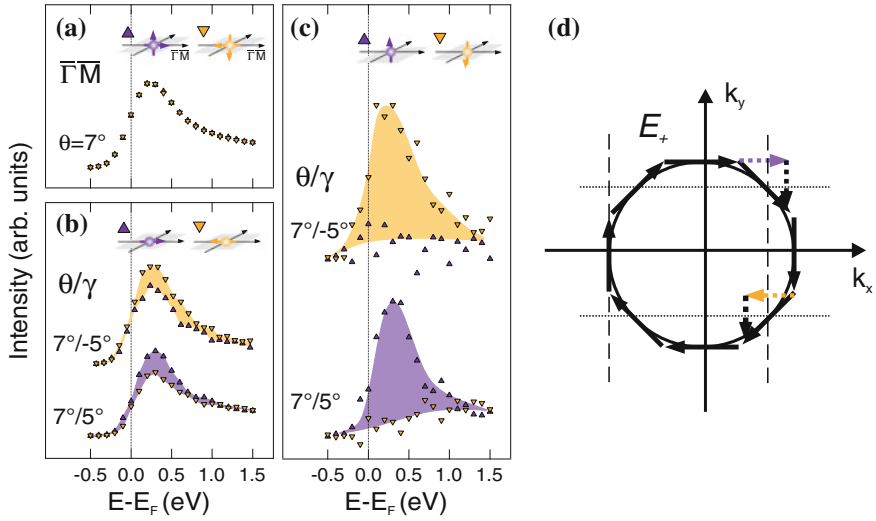


Fig. 2.11 **a** SR-IPE measurement for $\theta = 7^\circ$ with sensitivity to the out-of-plane/in-plane $_{\parallel}$ spin-polarization direction. **b, c** The same measurement as in **(a)** with intentionally tilted electron beam by $\gamma = \pm 5^\circ$. Spectra in **(b)** and **(c)** are obtained from the same raw data. In **(b)**, a normalization to 100 % in-plane $_{\parallel}$ beam polarization is applied, in **(c)**, the raw data are normalized to 100 % out-of-plane beam polarization. **d** Illustration of the dispersion and spin-polarization direction (arrows) of the Au(111) surface-state component E_+ in the k_x - k_y plane. The dashed (dotted) lines correspond to measurements with $k_x \neq 0^\circ$ ($k_y \neq 0^\circ$). For $k_x \neq 0^\circ$ and $k_y \neq 0^\circ$ (crossings of the dashed and dotted lines), the spin-polarization vector features components along k_x (orange and purple dotted arrows) and k_y (black dotted arrows)

presents a measurement of the spin-resolved intensity at $E = 0.27$ eV as a function of θ . The spin-asymmetry data ($A = (I_{\uparrow} - I_{\downarrow}) / (I_{\uparrow} + I_{\downarrow})$) illustrate the reversal of the spin-polarization direction between positive and negative angles. Spin asymmetry is only absent at $\theta = 0^\circ$.

Clearly, the measurements detect the Au(111) spin-orbit-split Shockley surface state. As predicted by the Rashba-Bychkov model the states are in-plane $_{\perp}$ spin polarized.

The $\bar{\Gamma}\bar{M}$ direction lies in the mirror plane of the system and out-of-plane spin polarization is neither expected theoretically (Henk et al. 2004b) nor measured by SARPES experiments (Hoesch et al. 2004). In Fig. 2.11a, the SR-IPE measurement for $\theta = 7^\circ$ along $\bar{\Gamma}\bar{M}$ with the spin sensitivity set to the out-of-plane/in-plane $_{\parallel}$ spin-polarization direction (purple up- and orange down-pointing triangles) is able to confirm this finding. Intriguingly, spin asymmetries are observed, when the electron beam is intentionally tilted by γ along the direction orthogonal to $\bar{\Gamma}\bar{M}$. Figure 2.11b presents SR-IPE spectra taken at $\theta = 7^\circ$ with a tilt of about $\gamma = 5^\circ$ and $\gamma = -5^\circ$. Obviously, the spin quantization axis of the surface state has changed so that out-of-plane and/or in-plane $_{\parallel}$ spin-polarization components emerge. This would be impossible for systems with fixed spin quantization axis such as ferromagnetic

surfaces. Note that a normalization to 100 % in-plane_{||} beam polarization has been applied, without knowing the exact spin quantization axis.

By way of trial, it is assumed that the observed spin asymmetry is a result of a pure out-of-plane spin polarization. In this case, a normalization to 100 % out-of-plane beam polarization has to be applied. The resulting spectra are shown in Fig. 2.11c. The SR-IPE measurements would suggest completely out-of-plane spin-polarized surface-state components. However, theory predicts an out-of-plane spin-polarization component of below 5 % (Henk et al. 2004b). An interpretation in terms of large out-of-plane spin-polarization components is, therefore, unlikely. Instead, the results are interpreted as follows. A rotation θ of the sample leads to a momentum $\mathbf{k}_x$ along $\bar{\Gamma}\bar{M}$. A tilt γ effectively produces a momentum $\mathbf{k}_y$ orthogonal to $\mathbf{k}_x$ and parallel to $\bar{\Gamma}\bar{K}$. According to the Rashba-Bychkov model, the quantization axis of the surface-state spin polarization is perpendicular to $\mathbf{k}_{||} = (k_x, k_y)$. A rotation θ , thus, leads to in-plane_⊥ spin polarization and a tilt γ gives rise to in-plane_{||} spin polarization. Figure 2.11d illustrates the surface-state component E_+ in the k_x - k_y plane. For $\theta \neq 0^\circ$ ($k_x \neq 0$) and $\gamma \neq 0^\circ$ ($k_y \neq 0$), E_+ exhibits a spin-polarization component parallel to $\mathbf{k}_x$. The value of the in-plane_{||} spin polarization P_x depends on the ratio $\frac{k_y}{|k_{||}|}$, see, e.g., Bentmann et al. (2011). In the case of the Au(111) surface state, for $\theta = 7^\circ$ and $\gamma = 5^\circ$, P_x is large enough to be detected. The spin polarization reversal between $\gamma = 5^\circ$ and $\gamma = -5^\circ$ affirms this argumentation.

Ultimately, the strength of the SR-IPE experiment with the ROSE is to analyze unoccupied states with respect to more than just one spin-polarization direction and, thereby, uncover complex spin textures. The Au(111) spin-orbit-split Shockley surface state is identified. It is demonstrated that the spin texture of the surface-state components is in agreement with the Rashba-Bychkov model. Along $\bar{\Gamma}\bar{M}$, only in-plane_⊥ spin polarization is detected. Notably, it is shown that in systems, where the quantization axis of the spin polarization is more complex than in single axis systems, a correct sample and electron beam alignment is indispensable. For the Au(111) surface state, a tilt of the electron beam of $\gamma = 5^\circ$ already leads to an observation of in-plane_{||} spin polarization, which can be easily misinterpreted as out-of-plane spin polarization, if the alignment is not exactly known.

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