Spin-Filtered Time-of-Flight k -Space Microscopy of Ir

– Towards the “Complete” Photoemission Experiment

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Abstract

The combination of momentum microscopy (high resolution imaging of the Fourier plane) with an imaging spin filter has recently set a benchmark in k -resolution and spin-detection efficiency. Here we show that the degree of parallelization can be further increased by time-of-flight energy recording. On the quest towards maximum information (in earlier work termed “complete” photoemission experiment) we have studied the prototypical high- Z fcc metal iridium. Large partial bandgaps and strong spin-orbit interaction lead to a sequence of spin-polarized surface resonances. Soft X-rays give access to the 4D spectral density function $\rho(E_B, k_x, k_y, k_z)$ weighted by the photoemission cross section. The Fermi surface and all other energy isosurfaces, Fermi velocity distribution $v_F(\mathbf{k}_F)$, electron or hole conductivity, effective mass and inner potential can be obtained from the multi-dimensional array ρ by simple algorithms. Polarized light reveals the linear and circular dichroism texture in a simple manner and an imaging spin filter exposes the spin texture. One-step photoemission calculations are in fair agreement with experiment. Comparison of the Bloch spectral function with photoemission calculations uncovers that the observed high spin polarization of photoelectrons from bulk bands originates from the photoemission step and is not present in the initial state.

1. Introduction

The maximum information content of a photoemission experiment has been discussed for free atoms by Kessler [1]. The measurable quantities are the photoelectron intensity, its dependence on the polarization state of the photon beam (also termed dichroism in photoemission) and its spin orientation, in the general case comprising three vector components. As long as the photon momentum is negligible, a measurement of this set of quantities provides the maximal quantum-mechanical characterization (in terms of transition matrix elements and relative phases), termed “perfect” or “complete” experiment [1]. In the present paper we reconsider this concept and attempt to transfer it to solid-state photoemission. We have chosen the prototypical high-Z fcc metal iridium because its electronic structure is characterized by large partial bandgaps and strong spin-orbit interaction, leading to highly spin polarized surface resonances.

In complete analogy to the atomic-like concept, the measurable quantities in solid-state photoemission are the photoelectron intensity I (depending on the photon polarization) and spin-polarization vector $\mathbf{P}$, both being functions of emission direction and binding energy E_B . The main difference concerns the number of contributing partial waves in the initial state (symmetry-adapted eigenfunctions or Bloch waves). Moreover, the bulk symmetry is lifted at the surface giving rise to surface states and resonances. Owing to translational symmetry, the accessible information on the electronic band structure is contained in the first bulk and surface Brillouin zone (BZ and SBZ), the k -space unit cells for 3D or 2D states, respectively.

Due to the existence of bulk and surface states, a comprehensive determination of the electronic structure of a material requires angle-resolved photoelectron spectroscopy (ARPES) in two spectral ranges with different information depths. Photoemission in the vacuum ultraviolet (VUV) emphasizes the surface electronic structure since the escape depth comprises just the topmost atomic layers. This surface sensitivity made ARPES a superior method for the study of surfaces, adsorbates, chemical surface reactions (e.g. heterogeneous catalysis) and thin films [2]. Additional spin-polarization analysis yields the spin character for ferromagnets [3,4], spin-orbit effects in the band structure [5] and the spin texture of topological surface states [6]. With the advent of high-resolution synchrotron beamlines in the soft X-ray range, the new discipline of SX-ARPES was launched that opened the route towards *truly bulk-sensitive band mapping*. Work at several synchrotron sources has impressively demonstrated the feasibility of high-resolution bulk band mapping [e.g. 7,8]. SX-ARPES measurements probing bulk bands even below a protective capping layer have overcome the old paradigm that photoemission probes just the surface.

Electronic band mapping is mainly performed using hemispherical spectrometers that have reached an excellent technical standard with sub-meV resolution and $<0.1^\circ$ angular resolution [9]. However, remarkable resolutions in the few-meV range were reached as early as 1964 by Boersch et al. [10] in energy-filtered TEM. Later it became clear that cathode-lens microscopy combined with an imaging energy filter provides an alternative tool for gaining photoemission spectra [11,12]. Although being optimized for real-space imaging, a normal photoelectron microscope (PEEM) can image reciprocal space (k -PEEM). However, k -resolution is poor and the zoom-range of k -magnifications is limited. As dedicated tools for k -imaging, a new class of instruments has been developed that are optimized for high-resolution imaging of the Fourier plane, a wide range of k -magnifications and micro-area selection. A set of laterally adjustable apertures in an intermediate Gaussian image plane allows precise determination of size and position of the region of interest on the sample surface down to the micrometer (and possibly sub-micrometer) range without the need of a fine-focused photon beam. This field aperture is a key feature of momentum

microscopes since it acts as contrast aperture for the reciprocal image. An optimized microscope of this kind, developed at the Max Planck Institute in Halle, has recently proven to be competitive with the best conventional angle-resolving photoemission spectrometers. A benchmark experiment by Tusche et al. [13] revealed a 3-fold “warping” of the well-known Shockley state on the Au(111)-surface with a momentum sensitivity of $10^{-4} \text{ \AA}^{-1}$ (corresponding to an angular precision of 0.005°). Such precision in momentum space is presently unachievable by any other method, justifying the term *momentum microscope*.

In this work we performed VUV photoemission at the Synchrotron beamline U125-2 NIM at BESSY II (Berlin) and soft X-ray photoemission at beamline P04 of PETRA III (DESY, Hamburg). The k -microscope combines the imaging column of the Halle instrument with time-of-flight (ToF) parallel energy recording using a delay-line detector as developed for ToF-PEEM [14-16] and an imaging spin filter. Full-field imaging of a k -range exceeding the first BZ and parallel energy recording of the entire d -band complex establishes a high degree of data throughput, paving the way towards the “complete” experiment in solid state photoemission. Intensity information yields bulk-band and surface-state dispersions (measured in the soft X-ray and VUV range, respectively) as well as the Fermi surface and Fermi velocity distribution. The dependence on photon polarization is quantified by the linear and circular dichroism in the angular distribution (LDAD and CDAD), being easily obtained in the entire BZ by a full-field imaging technique. In addition the spin texture is measured using an imaging spin filter of the W(100)-type. In the present work, only the transversal component P_y of the spin-polarization vector has been measured (hence the criteria of the complete experiment are not yet fulfilled). A strategy of extending the imaging spin-filter technique to a full vectorial analysis has recently been developed by Schaefer et al. [17]. Three independent asymmetry measurements, involving azimuthal rotation of the spin-filter crystal, allow to determine P_x , P_y and P_z separately and thus the full spin texture.

The experimental results are compared with calculations using the one-step model in density matrix formulation [18]. The surprisingly complex band structure with many surface resonances is accompanied by a rich spin texture with high degrees of spin polarization and large dichroism asymmetries present in all bands. The agreement between experiment and theory is generally good, quantitative differences are discussed. Comparison of the spin-resolved Bloch spectral function with photoemission calculations and experiment uncovers that the high photoelectron polarization observed for photoelectrons from bulk bands is no ground-state effect but arises in the photoemission process.

2. Set-up and Data Acquisition

2.1 Experimental set-up

The electron-optical scheme of the time-of-flight momentum microscope with imaging spin filter is shown in Fig. 1. The lens system is optimized for maximum k -resolution and a large zoom range of the k -image (0.5 up to $> 3 \text{ \AA}^{-1}$ radius). Two lens groups transport the image to the entrance of the field-free low-energy ToF section (typical drift energies 10 - 30 eV). An array of 9 selectable and adjustable field apertures in the first Gaussian image plane allows the selection of size and lateral position of the region of interest (ROI) on the sample surface in a wide range between 1 and $200 \text{ }\mu\text{m}$. This real-space aperture serves as contrast aperture for the k -image.

In the (spin integral) lower branch the data array $I(\tau, k_x, k_y)$ (τ : time-of-flight) is recorded by the first delay-line detector (DLD 1). The DLD is characterized by spatial and temporal resolutions of 80 μm and 180 ps, respectively, at a maximum count rate of 8 Mcps. At such count rates, the intensity pattern is visible in real time with 1 s acquisition time. A data set like the one at the bottom of Fig. 1 typically takes 10 minutes of acquisition. The binding energy E_B is determined from time-of-flight τ by reference to the Fermi energy. Energy and momentum resolution of the microscope are presently 19 meV and 0.01 $\text{\AA}^{-1}$; at the synchrotron light sources the overall energy resolution was 55 meV.

For spin-resolved imaging the spin-filter crystal (“spin mirror”) is shifted into the beam in the field-free space behind the lens column. The specular beam is imaged in the upper ToF branch (DLD 2) yielding spin-filtered data arrays. The top left inset illustrates the principle of multichannel spin detection (resolving k_x , k_y and τ simultaneously). A sequence of constant-energy sections through such an array is shown on the top right of Fig. 1 (red, blue and grey encoding positive and negative polarization and unpolarized intensity, respectively). The ToF discrimination bears the additional advantage that inelastic electrons created in the spin-filter crystal are completely suppressed in the spin-filtered image (later arrival time τ).

Figure 1. Scheme of the spin-filtered ToF momentum microscope. The retractable spin mirror allows for a rapid change between spin-integral (straight branch, DLD1) and spin-filtered imaging (upper branch, DLD2). The electron-optical column consists of cathode lens (see detail of backfocal plane BFP region), 2 zoom lenses, and field-free drift space. Trajectories shown for 100eV initial energy (radial coordinate of the rays stretched by a factor of 6). Top left inset: Scheme of simultaneous detection of spin-polarization distribution $P_y(\tau, k_x, k_y)$. k -image sequences: spin-filtered (upper DLD) and spin-integral (lower DLD) for Ir(111) taken at $h\nu=16$ eV (with different fields of view).

In the present experiment we exploited the spin sensitivity of the (0,0)-beam scattered from a W(100)-surface [19-21]. The usable energy interval is limited by the chromatic aberration of the electron lenses (depending on the desired k -resolution) and by the characteristics of the W(100) spinfilter crystal. $P_y(\tau, k_x, k_y)$ data arrays were taken at the working point of 26 eV scattering energy

and 55° scattering angle, where the spin asymmetry shows a broad maximum of about 3 eV width. Further details on the present setup can be found in [22], the general development of the spin-filter technique is discussed in [23].

So far, only the transversal spin component P_y perpendicular to the scattering plane has been detected, cf. top left inset in Fig. 1. A full vectorial analysis is possible when the in-plane scattering asymmetry at a different azimuthal angle of the spinfilter crystal (off the mirror plane) is used and, for the third component, the transversal spin component is rotated by a longitudinal magnetic field. Three different geometrical conditions yield three asymmetry values which allow to determine all three spin polarization components (for details, see [17]).

The Ir(111) crystal was cleaned by repeated cycles of sputtering, heating in oxygen at 1100 K and an annealing flash at 1300 K. The surface quality was checked by LEED prior to UHV-transfer into the microscope. The sample was inserted into a He-cooled hexapod stage for motorized alignment with micrometer precision and azimuthal rotation. During the experiments the sample was cooled to 30 K. The spinfilter crystal was cleaned by hot oxygen treatment and flashes as described in [21].

The total count rate in the investigated energy region is limited by the space-charge effect. For photon energies in the VUV range less than 10 electrons per photon pulse are ejected from the region of interest, hence the Coulomb interaction in the beam is negligible. In the soft X-ray range the cross section for the valence bands is orders of magnitude smaller than in the VUV and the photoemission spectrum is dominated by intense core level signals and a large background of secondary electrons. The total electron yield in this range is of the order of 1, i.e. the total electron flux (including secondaries) equals the photon flux [24]. The large number of slow electrons exerts a significant repulsive interaction (i.e. acceleration) on the fast electrons from the valence bands. Due to the special properties of the cathode lens the amount of acceleration depends on the radial coordinate in the momentum patterns [24, 25]. The data sets of section 4 have been corrected for the space-charge acceleration (amount of correction < 0.5 eV, details will be discussed elsewhere). Since the main effect is an increase of the energies, the shapes of the bands are not significantly affected. For larger energy shifts of several eV, the band features become blurred due to the stochastic e-e interaction.

2.2 Exploitation of symmetry properties in linear and circular dichroism and spin texture

Full-field imaging allows to exploit symmetry properties of the setup in dichroism and spin measurements. If plotted as $I(E_B, k_x, k_y)$ the spectral distribution function fills a paraboloid, confined by the photoemission horizon (condition $k_\perp=0$) and by the cut through E_F (condition $E_{kin}^{max} = h\nu - \Phi$, Φ workfunction). Linear dichroism in the angular distribution (LDAD) occurs for p-polarized light when the $\mathbf{E}$ -vector lifts the symmetry of a mirror plane. In analogy to the optical definition, the term “dichroism” refers to the different response of a system to the two orthogonal polarization states of a photon beam. In the geometry of Fig. 2(a) this is the case for the vertical mirror plane (y-z plane). Then the mirror operation leaves the crystal lattice unchanged but the photon operator is changed from $(z + x \tan 22^\circ)$ to $(z - x \tan 22^\circ)$, equivalent to switching the in-plane component of $\mathbf{E}$ from positive to negative x. In this case the *linear dichroism asymmetry* A_{LDAD} can be determined from a single intensity array according to

$$A_{LDAD}(E_B, k_x, k_y) = \frac{I(E_B, k_x, k_y) - I(E_B, -k_x, k_y)}{I(E_B, k_x, k_y) + I(E_B, -k_x, k_y)}. \quad (1)$$

If the plane of incidence of p-polarized light coincides with a mirror plane of the sample, this mirror symmetry is retained. This is the case for the x-z plane (hatched) in Fig. 2(a). Then the intensity patterns above and below this plane are identical and there is no LDAD with respect to this plane. However, the *spin polarization* component parallel to this plane reverses its sign under the mirror operation, since the spin polarization is an axial vector. In turn, the spin-polarization patterns in the half-space above and below the hatched mirror plane show opposite signs as sketched for the spin component P_x in directions k_1 and k_2 . In the imaging spin filter this “up-down asymmetry” can be exploited in the same way as the left-right asymmetry of a spin-orbit type spin detector (like the Mott or SPLEED detector [26]). The 3D array of the *spin-polarization component* P_x is derived from a single measurement of the scattered intensity pattern $I_s(E_B, k_x, k_y)$ behind the spin filter as

$$P_x(E_B, k_x, k_y) = \frac{I_s(E_B, k_x, k_y) - I_s(E_B, k_x, -k_y)}{I_s(E_B, k_x, k_y) + I_s(E_B, k_x, -k_y)}. \quad (2)$$

P_x vanishes in the mirror plane. Since an axial vector perpendicular to a mirror plane is retained upon the mirror operation, the transversal component P_y can be different from zero in the mirror plane (momentum vector k_3 in Fig. 2(a)) and is identical above and below the plane ($P_y' = P_y$). When the y-z plane is also a mirror plane, the transformation property of P_y is analogous to P_x : The mirror operation would reverse the y-component, yielding $P_y'' = -P_y$ for direction k_4 . If the y-z plane is no mirror plane (like for Ir(111)), the symmetry is lifted and the latter relation does not hold. Finally, a longitudinal polarization component (not shown in the figure) transforms like P_x , i.e. it is reversed by the mirror operation.

Figure 2. Symmetry properties of dichroism and spin texture for linearly (a) and circularly polarized light (b). The photoelectrons are detected in the full half space in front of the sample surface, in the E -vs- $k_{\parallel}$ plot this corresponds to a paraboloid. (a) Geometry for p-polarized light, giving rise to LDAD. The electric vector E lies in the x-z plane (hatched, assumed to be a mirror plane of the sample). The axial vectors indicate the symmetry behavior of the transversal spin-polarization components P_x and P_y . E retains the mirror plane, i.e. the intensity patterns above and below the mirror plane are identical. (b) Geometry for circularly polarized light, giving rise to CDAD and the *Fano* spin component. The axial vector of the photon helicity S_y breaks the mirror symmetry, hence the upper and lower half-space show a CDAD asymmetry.

Due to the vector structure of the photon operator the photoemission process does not retain the initial spin orientation. For circular photon polarization the phenomenon of optical spin orientation is well known for free atoms (termed *Fano effect*) [26] and solids [27]. But even for excitation by linearly polarized and unpolarized light (which is not “isotropic”), the photoelectrons are spin polarized. Degrees of spin polarization up to 50% have been measured for free rare-gas atoms with

unpolarized [28] and linearly polarized light [29]. In a simplified picture photoemission can be considered as “half a scattering process”, i.e. the outgoing photoelectrons (having well-defined orbital quantum numbers, corresponding to impact parameters in scattering) experience the same spin-orbit forces as a scattered electron.

Circular dichroism in the angular distribution (CDAD) has different symmetry properties. In the geometry of Fig. 2(b) the photon beam lies in the same plane as in (a), but the helicity vector is not mirror invariant and thus lifts the mirror symmetry of the xz -plane (hatched). Reversal of the helicity vector S_γ from right (RCP) to left circularly polarized light (LCP) changes the photon operator from $(z + iy)$ to $(z - iy)$ (for grazing incidence). In turn, the final state partial waves excited by the “y-part” of the operator experience a phase shift of $\pm\pi/2$ (since $i=e^{i\pi/2}$) with respect to those excited by the “z-part”. This is the essence of the CDAD [30]. In this case the *circular dichroism asymmetry* A_{CDAD} can be determined from the two intensity arrays (measured with opposite photon helicities) according to

$$A_{CDAD}(E_B, k_x, k_y) = \frac{I^{RCP}(E_B, k_x, k_y) - I^{LCP}(E_B, k_x, k_y)}{I^{RCP}(E_B, k_x, k_y) + I^{LCP}(E_B, k_x, k_y)}. \quad (3)$$

If the plane of incidence coincides with a mirror plane of the sample, A_{CDAD} can be obtained from a single measurement without helicity reversal. For excitation by circularly polarized light the optical spin orientation leads to a polarization component (termed *Fano* component) along the helicity vector. As sketched in Fig. 2(b) this component reverses its sign when switching the helicity. The most prominent example of this effect is the GaAs-source for polarized electrons [27].

3. Theory

The self-consistent electronic structure calculations were performed within the *ab-initio* framework of spin-density functional theory. The Vosko, Wilk, and Nusair parameterization for the exchange and correlation potential was used [31]. The electronic structure was calculated in a fully relativistic mode by solving the corresponding Dirac equation. This was achieved using the relativistic multiple-scattering or KKR formalism [18,32,33]. The resulting electronic structure represented by single-site scattering matrices for the different layers and the corresponding wavefunctions for initial and final-state energies serve as input quantities for the corresponding photocurrent calculations.

Our spectroscopic analysis is based on the fully relativistic one-step model [34], in its spin-density matrix formulation [35,36]. This approach allows to describe properly the complete spin-polarization vector. For the CDAD calculations, in addition, we account for the surface itself by use of a Rundgren-Malmstrom-type surface potential [37]. This allows for the correct description of the energetics and dispersion of all surface-related features. Furthermore, the relative intensities of surface states and resonances are quantitatively accounted for by calculating the corresponding matrix elements in the surface region. This procedure is described in detail, for example, in Ref. [38]. The energy-dependent retarded KKR Greens function which represents the initial state has been calculated for a complex energy, with a constant imaginary part $V_i = 0.03$ eV, to account for hole life-time due to inelastic scattering events. Also in the final-state calculation many-body effects have been included phenomenologically to account for the inelastic mean free path (IMFP) of the high-energy photoelectron. In detail, a constant imaginary value of $V_h = 1.2$ eV was chosen according to the photon energy of $h\nu = 16$ eV used in the spectroscopic measurements. The calculations were performed with circularly polarized light, except for the 200 eV calculation in

Fig. 3 which was done for linearly p-polarized light. The polar angle of the incoming photon beam was fixed to 68° with respect to the surface normal for all calculations. As the incoming light is partially absorbed by a metal surface the index of refraction is complex in general. In the general case, the linearly polarized photon beam is transformed to elliptically polarized light, where the corresponding components of the vector potential are determined by the Fresnel equations [39]. If the plane of incidence coincides with a mirror plane of the crystal the beam stays linearly polarized. The corresponding dielectric response is described in a phenomenological way using a macroscopic dielectric constant. For the dielectric constant the value $\epsilon(16 \text{ eV})=(1.21;0.83)$ [40] has been chosen in our spectroscopic analysis.

The top row in Fig. 3 shows results of a ground state calculation, i.e. the Bloch spectral function (BSF) (a) and the three components of the spin polarization (b-d) in a E_B -vs- $k_{||}$ section along the $\bar{\Gamma}\bar{K}$ azimuth of Ir(111). The bottom row shows the corresponding photoemission calculation for a photon energy of 200eV. The bulk bands are labelled B1 – B6 with increasing energy; the *sp*-band B1 crosses the $k_{||}=0$ axis at about $E_B=11\text{eV}$, outside of this energy window. In the BSF the bulk bands show up as broadened regions due to the k_z -dispersion (projection of the bulk bands onto the surface BZ), as will be discussed in the next section. In addition to the bulk bands, a rich pattern of surface resonances is visible, labelled S1-S5. Since they show only small k_z -dispersion, S1-S5 appear sharp in these sections. The surface resonances exhibit large spin-polarization values in P_y and P_z (c and d; note the different color code in b). In the BSF the bulk bands are not significantly spin polarized.

Figure 3. Top row: Bloch spectral function (a) and its spin-polarization components (b, c, d) of iridium. All panels show a section along the $\bar{\Gamma}\bar{K}$ direction of Ir (111). B2-B6 denote bulk bands, S1-S5 are surface resonances. Significant degrees of spin polarization occur only in the surface resonances; the bulk bands are essentially unpolarized. Second row: Photoemission calculation for Ir(111) at $h\nu=200\text{eV}$; intensity (e) and corresponding spin-polarization texture (f, g, h). In photoemission the bulk bands appear with a significant spin polarization.

In the photoemission calculation (second row) the situation is different. At the photon energy of 200eV the bulk bands dominate due to the enhanced photoelectron escape depth. Since the photon energy selects a certain k_z -value, the bulk bands appear relatively sharp. Moreover, they are considerably spin polarized, see panels (f-h). The surface resonances are almost invisible. Obviously the bulk bands, being unpolarized in the initial state, attain a sizeable spin polarization in the photoexcitation step. The photon operator of the linearly polarized light induces a spin orientation in all three components, due to the angular momentum selection rules.

4. Mapping of Fermi Surface and Fermi Velocity Through the 4D Spectral Function $\rho(E_B, k_x, k_y, k_z)$

At the beginning of the experimental chapter we point out that the analogy to the concept introduced by Kessler [1] refers to the experimentally accessible quantities, but not to a “reverse” determination of matrix elements and phase differences. In solid-state photoemission the number of partial waves in the initial state is generally large and a “reverse” determination of matrix elements and phases from the measured quantities is practically impossible or supposes simplifying assumptions (e.g. a pure p_z -like initial state for the π -band of graphite [44a]). In solid-state photoemission the *ground-state electronic structure* (band dispersions, gaps, Fermi surface, Fermi velocity) is of utmost importance because it defines e.g. transport, optical and thermodynamical properties of the electron system. This information is contained in the spectral function ρ as discussed in this section. The spectral function is described in momentum space and due to the translational symmetry the 1st Brillouin zone contains the full information.

Photoelectron dichroism arises in the photoemission step, is governed by matrix elements and phase-shift differences, and gives valuable information on the initial-state orbital wavefunctions, cf. section 5.1. The spin-polarization texture contains an initial-state contribution (in the present case of Ir(111) from a Rashba-type polarization of surface resonances) and a final-state contribution that occurs due to the photoemission transition, cf. section 5.2.

Shape and topology of the Fermi surface and the velocity of the electrons at the Fermi energy $v_F(\mathbf{k}_F)$ are key features in materials research. De Haas van Alphen measurements provide precise values of the extremal-path cross sections, but strongly fragmented surfaces and complex topologies are not accessible. The alternative method of choice is photoemission. Looking directly into k -space, a momentum microscope is the ideal tool for Fermi surface mapping as we will demonstrate in this section. The Fermi surface of Ir (Fig. 4(a), calculated according to [41]) is particularly interesting since it consists of unconnected electron and hole pockets centered at the Γ - and X-points of the bulk BZ. The inner sheets (corresponding to bands B6 and B4, cf. Fig. 3(a)) are surrounded by the outer sheets (B5 and B3).

The basis for the determination of Fermi surface and velocity is the spectral density function ρ , a 4D data array. Two momentum coordinates (k_x, k_y) and binding energy E_B are measured simultaneously as was illustrated in Fig. 1. The fourth coordinate k_z is accessed by variation of the photon energy exploiting the concept of direct transitions as explained in Fig. 4(b). In the extended zone scheme the final states corresponding to a given energy lie on a sphere centered at the Γ -point of the first BZ (analogously to the Ewald sphere in diffraction). The photon momentum $\mathbf{k}_{h\nu}$ is transferred to the photoelectron, hence the sphere is shifted by the vector $\mathbf{k}_{h\nu}$. The perpendicular and in-plane momentum components are denoted as $k_f^\perp$ and $k_f^\parallel$. The momentum microscope

detects the intersection of this sphere with the periodic pattern of Brillouin zones, and the k_f distribution of this section is visualized on the screen. By tuning the photon energy k_z is varied across the full BZ. These measurements have been performed at beamline P04 [42] of PETRA III (DESY, Hamburg), which provides photons in a wide energy range (260-2000eV). The high parallelization of data acquisition and the high brilliance of this beamline compensate for the dramatic drop of the photoemission cross section with increasing photon energy.

Scanning of k_f via variation of $h\nu$ thus gives access to the coordinate k_z , in a tomographic-like mode. 3D data arrays have been taken at 14 different photon energies, mainly in the momentum range corresponding to the 4th repeated BZ along k_z ($h\nu=369$ to 616 eV, see Fig. 4(b)). The photon beam contained a significant contribution from second-order diffraction of the monochromator. As photoelectrons excited by second-order photons have much larger initial kinetic energies they fly essentially straight from the sample to the detector and form a narrow pencil-like beam. This unwanted electron background was completely suppressed by blocking the center of the beam with an eccentric k -aperture and by mapping of the first BZ in lateral direction as sketched in Fig. 4(b).

The sequence of 3D arrays is then concatenated along k_z yielding $I(E_B, k_x, k_y, k_z)$, which is the discretized (by the spatio-temporal resolution) and weighted (by the photoemission cross section) representation of the spectral function ρ . In order to eliminate dichroism effects the data array is fully symmetrized. Four momentum sections from the concatenated stack at $E=E_F$ are shown in Fig. 4(c-f). All four bands forming the Fermi surface are visible with good contrast. Section (c) crosses the Γ -point and shows a cut through sheets B5 and B6, centered at Γ . Section (d) is located between the Γ - and X-points and shows part of the repeated Fermi surfaces with bands B5' of the neighboring BZs. In the reciprocal lattice (bcc) the Fermi surfaces are indented. Section (e) runs through the X-points and crosses the centers of pockets B3 and B4. Finally, section (f) crosses the L-point. According to the calculated Fermi surface (see (a)) the plane crossing the L-point does not contain bulk bands at E_F . Hence the feature in the center (here intensified for better visibility) is ascribed to a surface resonance as will be discussed in the next section.

Sequence (g-k) shows the evolution of pockets B3 and B4 with binding energy. Both expand with increasing binding energy. From their band curvature we conclude that both are hole pockets. The outer pocket (B3) exhibits a pronounced warping in the shape of an elongated hexagon. In contrast, the inner pocket (B4) appears in a diamond shape reflecting the fourfold symmetry of the BZ face containing the X-point. In (k) B4 converts to a hexagon, rotated by 30° in comparison to the hexagon of B3 in (g-i).

The bottom row in Fig. 4 shows two exemplary patterns of the *circular dichroism in the angular distribution* (CDAD). Sketch (l) illustrates the geometry and the appearance of the CDAD for an atomic-like d_{z^2} -orbital lying in the crystal mirror plane m . The two components of the photon operator ($z \pm iy$) excite different final state (p and f) partial waves that are superimposed coherently. Reversal of the photon helicity leads to a phase shift in those partial waves excited by the y -component. In turn, this causes a rotation of the differential cross section pattern, clockwise for RCP (red) and counterclockwise for LCP light (blue). The essence of this effect is retained for band states composed of several initial partial waves causing CDAD intensity asymmetries such as shown in Fig. 4(m,n). Panel (m), marked as dashed rectangle in (c), reveals that the outer and inner Fermi-surface sheets centered at Γ exhibit opposite signs of the CDAD. Panel (n) displays the CDAD of intensity section (h), the full and dashed lines mark identical positions. The CDAD of hole pockets B3 and B4 shows a pronounced bipolar texture. The top – bottom dissymmetry

reflects an additional contribution of the LDAD due to the tilted direction of photon incidence (from top to bottom). Since the photon operator acts on the orbital parts of the wavefunctions, the CDAD asymmetries reflect the orbital-symmetry character of the initial states (for details, see [43,44]).

Figure 4. (a) Theoretical Fermi surface of iridium (data from [41]), the sheets are labelled according to the bands denoted in Fig. 3a. (b) Quantitative sketch of direct transitions in Ir(111) photoemission; k_f is the final-state electron momentum consisting of $k_f^\perp$ (along the k_z axis) and $k_f^\parallel$ (in the k_x - k_y plane). All final states lie on spheres centered at the Γ -point but displaced by the photon momentum $k_{h\nu}$ (red spherical sectors for three photon energies). (c-f) *Constant- k_z* sections, symmetrized according to the lattice symmetry to eliminate the dichroic photoemission effects, in the 4th repeated Brillouin zone at E_F through the Γ -point (c), further towards the X-point (d), through the X-point (e) and through the L-point (f). (g-k) Sections of the concentric hole pockets B3 and B4 centred at the X-point for different binding energies (region marked by dashed rectangle in (e)), as measured. (l) Geometry scheme of the circular dichroism CDAD and (m,n) CDAD patterns of the region marked by the dashed rectangle in (c) and full panel (h).

The *Fermi surface* is the boundary of the occupied part of the spectral function (termed support ρ). Borrowing notation from topology, the Fermi surface, a 2D object in 3D k -space, can be denoted as

$$\partial \text{supp}(\rho(E = E_F; \mathbf{k})). \quad (4)$$

This mathematically captures the process of isolating “slices” corresponding to the Fermi energy at different k_z , finding the boundary between zero and nonzero regions according to a threshold, and concatenating these slices along z . Perspective views of the resulting Fermi surface of iridium are shown in Fig. 5(a,b). The inner sheet B6 becomes visible in the cut-out region in (b) because it is completely surrounded by sheet B5.

The *Fermi velocity distribution* $v_F(\mathbf{k}_F)$ (group velocity of electrons with energy E_F) is given by

$$\mathbf{v}_F(\mathbf{k}_F) = \frac{1}{\hbar} \nabla_{\mathbf{k}} E(\mathbf{k})|_{E=E_F}. \quad (5)$$

The vector $\mathbf{v}_F(\mathbf{k}_F)$ is oriented perpendicular to the Fermi surface. The gradient can be numerically determined from the distance of adjacent energy isosurfaces (E_F , $E_F - \Delta E$). The results are color coded and superimposed on the Fermi surface, see Fig. 5(c-e). The velocity v_F reaches values of 10^6 m/s, in reasonable agreement with the expectation for a free electron gas with the lattice parameters of Ir (2×10^6 m/s). As shown above, band features B3 and B4 expand with increasing binding energy (hole pockets, color yellow to red). Conversely, features B5 and B6 contract and the corresponding curvature of $E(\mathbf{k})$ reveals that they are electron sheets (blue in Fig. 5c-e).

Figure 5. (a,b) Experimental Fermi surface of iridium in different views (arrows indicate the directions in k -space). (c-e) Superimposed Fermi-velocity distribution $\mathbf{v}_F(\mathbf{k}_F)$, velocities see color bar (blue and red correspond to electron and hole conductivity, respectively).

For the quantitative evaluation, we have to consider several effects that partly show up in terms of deformations (curvatures) in the k -images. Significant contributions originate from the physics of the photoemission process and from the Coulomb interaction in the beam.

(i) The physics of direct transitions induces a fundamental obstacle, namely the non-planarity of the sectional area of the *final-state sphere* with the periodic pattern of Brillouin zones. Moreover, the final-state sphere is displaced from the center of the 1st BZ by the photon-momentum vector as depicted in Fig. 4(b) by red spherical sectors. In the momentum microscope the intersection of the spherical sectors with the band features are projected onto the planar image detector, yielding directly the $k_{||}$ -distribution.

(ii) The Coulomb repulsion between the true valence photoelectrons (detected at a rate of typically one per photon pulse) and the large number of secondary electrons (space-charge effect) causes an acceleration of the photoelectrons that depends on the radial coordinate in the momentum image [24,25].

(iii) Unlike other ToF instruments, the time lag due to the curvature of the isochrones marked by dots on the trajectories in Fig.1 is negligible (less than 100 ps for a typical drift energy of 20 eV). Also, the image-field curvature [45] is negligible up to k -fields of $4 \text{ \AA}^{-1}$ diameter. At larger fields a barrel-type image distortion becomes significant which can be corrected, if necessary.

We briefly summarize this section: The 4D data body ρ contains all information on the (spin-integral) bulk electronic structure and allows extracting the relevant quantities by simple algorithms without assumptions or approximations. The *Fermi surface* (and all analogous isosurfaces at other energies) can be projected from ρ as the boundary of the occupied part of the spectral function, the Fermi velocity is the gradient of $E(\mathbf{k})$ in k -space. These quantities are of central importance for electronic transport and thermodynamical properties of the electron system.

5. Texture of Dichroism and Spin Polarization

5.1 Band structure and linear dichroism

In this section we discuss the measurements at low photon energies where the surface electronic structure is expected to dominate the spectra. In the VUV range the inelastic mean free path is of the order of 1-2 atomic layers. Fig. 6 shows measured and calculated intensities and linear dichroism patterns (quantified by a color bar). Experimental data have been taken at $h\nu=16$ eV (U125 NIM of BESSY II, Berlin). This normal-incidence monochromator provides linearly polarized VUV radiation between 5 and 40 eV with a photon flux of about 10^{10} s^{-1} in single bunch mode (at few meV bandwidth).

The $E=\text{constant}$ sections Fig. 6(a,b) show the well-known surface resonance S1 [46] closely below E_F . S1 is surrounded by bulk band B6, here appearing as a triangular feature (as visible also in the soft X-ray measurement in the center of Fig. 4(d)). The threefold symmetry is removed by the electric vector, cf. Fig. 2a. The hexagonal pattern at the periphery reminds of the hexagon B6 in Fig. 4c. However, here we see essentially surface resonances S2 and S3 because the photon energy does not reach the Γ -point of the bulk BZ.

The $E_B\text{-vs-}k_{\parallel}$ sections (c,d) show the dispersion of bands B6, B5, B4 and B3, partly overlapping with B2. In earlier spin-resolved work [47,46] the topmost bands have been labelled A-D. Along k_z (Γ -A-L) the corresponding irreducible representations are Λ^3_{4+5} , Λ^3_6 , Λ^3_6 and Λ^3_{4+5} , respectively, where the upper index denotes the orbital symmetry (all d , $l=2$) and the lower one the double-group symmetries. The sp -band B1 (symmetry Λ^1_6) starts at 10 eV binding energy and its second branch B2 intersects the d -band complex, showing a crossing point with B3 (different double-group symmetry) and an avoided crossing with B4 (same double group symmetry).

The bulk bands show pronounced dispersions with 3-fold symmetry. Note that the left panels of Fig. 6(c) and (d) show sections in two different planes, the left halves along $\bar{\Gamma}\bar{K}$ (fcc mirror plane) and the right halves along $\bar{\Gamma}\bar{M}$. The LDAD panels in (c,d) show only the cut along $\bar{\Gamma}\bar{K}$. With increasing $k_{\parallel}$ the topmost bands B6 and B5 disperse upwards and downwards, respectively. G1 and G2 correspond to local bandgaps in the bulk bandstructure (see also Fig. 3a).

The LDAD asymmetry patterns reveal fair agreement between experiment and theory. For both, the asymmetries of B6 and B5 reach about 50% and B6 and B5 exhibit opposite signs. The dispersion looks different because at $h\nu=16$ eV experiment and calculation lead to different positions for the final-state band (see next section). In addition, quantitative differences are obvious with respect to intensities and size of the dichroism. The downward-dispersing surface resonance S1 is visible in experiment and theory but its large Rashba splitting [46] appears less pronounced in theory than in experiment. Conversely, this band shows a huge LDAD asymmetry in theory (dominating the LDAD pattern), whereas it's LDAD is almost invisible in the experiment. The experimental LDAD pattern (c) is dominated by the mirror-asymmetric texture of B6 and B5 which should be repeated by B4 (showing the same texture as B6) and B3. However, the latter band hybridizes with sp -band B2, leading to a bipolar LDAD signature in the B2/B3 mixing regions.

Fig. 6(e-l) show $E=\text{constant}$ sections at increasing binding energies as marked by arrows on the right-hand side of panels (c,d). This sequence shows excellent agreement between experiment and theory. In sections (g,h), crossing the topmost bandgap, theoretical and experimental intensity patterns are practically identical and also the LDAD patterns have all essential features in common.

Figure 6. Sections through the array $I(E_B, k_x, k_y)$ and corresponding LDAD (see color bar) for $h\nu=16$ eV (rows 1 and 3, experiment; 2 and 4, theory). (a,b) Experimental and theoretical $E=\text{constant}$ sections at the onset of surface resonance S1 closely below E_F . (c,d) E_B -vs- $k_{\parallel}$ sections showing the dispersion of bulk bands B2-B6 and surface resonances S1-S3. The left and right halves of the intensity patterns in (c,d) correspond to different azimuthal orientations $\bar{\Gamma}\bar{K}$ (fcc mirror plane) and $\bar{\Gamma}\bar{M}$, respectively. G1 and G2 denote local band gaps. (e-l) Sequence of $E=\text{constant}$ sections at the positions marked by arrows in (c,d). (m-s) Constant- k_y sections in the hybridization region of B5 and B4, having identical double-group symmetry.

The bottom row of Fig. 6 shows a measured $k_y=\text{constant}$ sequence of the hybridization region of B5 and B4, having same double-group symmetry Λ_6 . The E_B -vs- k_x sections (m,n,o,p,q,r and s) correspond to $k_y = -0.2, 0.0, 0.1, 0.2, 0.4, 0.6$ and $0.8 \text{ \AA}^{-1}$, respectively. In section (m) the five topmost bands show pronounced LDAD with B2 and B3 being distinguishable by different signs of LDAD. In the sequence (m,n,o) the slope of B5 changes rapidly from upward to downward dispersion. In turn, an avoided crossing with B4 occurs (marked by circles), leading to pronounced changes in the LDAD texture. In particular the LDAD in the bottom part of B4 changes its sign (see arrows in bottom part of (n,o)). Due to its quantum-mechanical nature [43,44], the LDAD reflects changes in the orbital character of the initial wavefunctions in the hybridization region. In contrast, the intensity patterns do not show strong signatures of band hybridization. The calculation

in Fig. 3(d) shows a similar effect upon crossing of surface band S3 with S1, marked by a circle. The sign of spin polarization of the spin-orbit doublet S3 is reversed at the crossing point as reflected by the reversal of the color of the two sub-bands. The surface states have different symmetry as the bulk bands. Hence S1 crosses B6 and B5 without hybridization, see arrows in top part of (n). In sections (q,r) we see surface band S3 crossing the bandgap as predicted by theory (cf. Fig. 3c).

The combination of surface bands with bulk bands, involving several avoided crossings and hybridization regions makes the valence band structure of iridium quite complex (see also [48]). The full 4D and 3D spectral functions given in chapters 4 and 5, respectively, are necessary to disentangle the band features.

5.2 Photoelectron spin polarization

Spin-resolved measurements have been performed for the Ir(111) surface at several energies between 16 and 25 eV using the beamline U125-2 NIM at BESSY II. The photon beam was p-polarized and the spin detector was mounted so that the in-plane polarization component P_x along the direction parallel to the plane of photon incidence was recorded, cf. Fig. 2(a). The anti-symmetry of the axial spin-polarization vector under mirror operation in principle allows the measurement of the full spin-polarization data array $P_x(E_B, k_x, k_y)$ parallel to the mirror plane in a single measurement of the spin-filtered intensity I_s , according to Eq. (2). Actually, the spin polarization has been determined from two data sets acquired for two different scattering energies applying the procedure described in [22]. Acquisition of each data array comprising about 10^4 resolved data points took about 2 hours, with short flashes of the W(100) spin-filter crystal every half hour. The Ir(111) target crystal was prepared prior to each acquisition run.

Selected sections from the data array at $h\nu=16$ eV are shown in Fig. 7 together with the corresponding calculation. The panels show sections of the polarization array P_x , the values are quantified by the color bar: Red and blue denote positive and negative polarization (color scale from -50% to +50%) and the grey level quantifies the unpolarized intensity. In this representation a band with 50% polarization appears in saturated red (or blue) and an unpolarized band appears in grey, the level denoting its intensity. Rows 1 and 2 show measured and calculated $P_x(k_x, k_y)$ sections at constant energy (E_B values given in the top row are identical for experiment and theory). Rows 3 and 4 show E_B -vs- k_y sections at several values of k_x as marked in panels (a, g) as dashed lines.

Calculations have been performed for several photon energies. The best agreement with the 16 eV experiment was found for the calculation at $h\nu=13$ eV. For a qualitative understanding of this difference we refer to [49]. While lifetime effects are considered in the calculations by parameterizing the imaginary part of the self energy for initial and final states, the real part of the self energy has been set to zero. An ab-initio calculation of the real part for realistic systems, which would be necessary for considering the interaction of photoelectron with the photo hole, is extremely time consuming. In the present local density approximation (LDA) the final (free-electron like) state appears at a too low energy. This effect is similar to the fact that semiconducting band gaps are typically smaller in LDA calculations than experimental values. Thus, realistic final state bands have larger energies compared to LDA calculations and therefore larger photon energies are needed to reach these states.

Figure 7. Measured (rows 1 and 3) and calculated (rows 2 and 4) photoelectron spin texture P_x for photoemission from Ir(111) with p-polarized light in the geometry of Fig. 2 (experiment $h\nu=16\text{eV}$, calculation $h\nu=13\text{eV}$). (a-l) $P_x(k_x, k_y)$ sections at constant binding energy, values of E_B given in the top row. (m-v) $P_x(E_B, k_y)$ sections at constant k_x as denoted by dashed lines in (a, g). The spin polarization is quantified by the color scale (the scaling error is 20%).

The experimental spin-polarization patterns are dominated by the strongly-dispersing bulk bands B6 and B5, which are highly polarized (up to $\pm 50\%$). With increasing binding energy the blue and red branches of B6 approach each other (a-c) and merge at the band bottom at $\bar{\Gamma}$ (d). Bulk band B5 dispersing in the opposite direction shows the opposite spin polarization (e,f). The E_B -vs- k_y sections in the third row show the band dispersion in five planes ($k_x=0, \pm 0.4 \text{ \AA}^{-1}, \pm 0.8 \text{ \AA}^{-1}$). The separation of bulk band B6 and B5 at $\bar{\Gamma}$ near $E_B=1.1 \text{ eV}$ is visible in panel (o), i.e. the section through $\bar{\Gamma}$. Contrarily, the separation almost vanishes in the theoretical data (t).

The calculated spin-polarization patterns (rows 2 and 4) are dominated by the surface resonances S2 and S3, forming the hexagon in (g-i). The fact that the hexagon is warped and shows only threefold symmetry reflects the fact that S2 and S3 are surface resonances, i.e. these bands still “feel” the bulk symmetry O_h . Bands B6 and B5 show the same dispersion behavior as measured (cf. panels o and t). The calculated spin polarization of B6 and B5 is significantly smaller than the measured polarization. Contrarily, the calculated spin polarization of the surface resonances S2 and S3 are larger than the corresponding experimental values.

This discrepancy results partly from the fact that the observed photoemission intensities deviate from the calculated ones: The intensity of the bulk bands dominates the experimental spectra while they appear rather weak in the calculation. On the other hand all surface resonances being prominent in the calculation show rather weak intensities in the experimental data. Considering that the photoemission intensity comprises an unpolarized background, a small photoemission intensity leads to a reduction of spin polarization, too, and vice versa. The pronounced intensity of bulk states in the experimental data indicates that these bands exist already in the topmost layers, i.e. within the mean free path of the excited photoelectrons. Obviously, it is not necessary to probe many atomic layers in order to observe a bulk state and its 3D dispersion. The same has been observed in previous spin-resolved experiments for Ir(111) [48] and Pt(111) [50] in normal emission (i.e. along Γ - Λ -L), where the correct k_z -dispersion of the bulk bands could be measured in the VUV range, see also [53]. In the early experiments the *Fano-type* spin-polarization component for excitation by circularly polarized light was measured, which is strong in normal emission. A modification of the matrix elements due to many-body correlation effects (similar to the deviation in the excitation energy) could provide a second explanation for the discrepancy between experiment and theory in Fig. 7.

6. Summary and Conclusion

In this article we introduced spin-filtered time-of-flight (ToF) k -space microscopy as novel tool for a full characterization of the electronic structure by the example of Ir(111). Central part is an electron-optical column optimized for imaging of the Fourier plane of a cathode lens at maximum momentum resolution in a large zoom-range in k -space. Using this column, a k -sensitivity of $10^{-4} \text{ \AA}^{-1}$ (corresponding to an angular precision of 0.005°) was demonstrated by the Halle group [13]. Such values are presently unachievable by conventional photoemission techniques. The second achievement was the implementation of parallel energy recording via an imaging ToF-spectrometer with delay-line detector, a high-speed single event counting device [15,16]. Permanent integral count rates of up to 8 Mcps at spatial and temporal resolutions of $80 \text{ }\mu\text{m}$ and 150 ps, respectively, are key factors for high detection efficiency. This detector can record (k_x, k_y) -images with 0.2 (0.8) Mpixels for 40 mm (80 mm) diameter and – at the same time – more than 1000 time slices for photon-pulse periods of $\sim 200 \text{ ns}$ or more. Given these values, the detector is no longer the bottleneck in high-speed data acquisition, but electron-optical limitations (above all the chromatic aberration) and the space-charge problem pose practical limits. Third, the advent of multichannel spin polarimetry (via imaging spin filters [19,20]) established an enormous enhancement of spin-detection efficiency in comparison with present-days' single-channel spin detectors.

Employing this combination of novel techniques, we performed a comprehensive investigation of the electronic structure of iridium. Large partial bandgaps and strong spin-orbit interaction give rise to a sequence of highly spin-polarized surface resonances. The overlapped bulk and surface bands make the prototypical high-Z fcc metal Ir an ideal testing ground for validation of the new method. The central point of the paper is the most complete determination of the experimentally-accessible quantities, adopting a concept that was originally introduced by Kessler [1] in the framework of the “complete” experiment for atomic photoemission. The measurable quantities are the *intensity*, its dependence on photon polarization, quantified in terms of the *circular and linear*

dichroism in the angular distribution (CDAD, LDAD) and the *photoelectron spin polarization vector*.

The complete information on the electronic structure of a solid is contained in the spin-resolved spectral density function $\rho_{\uparrow\downarrow}(E_B, \mathbf{k})$ in the full BZ and in the energy range of interest (e.g. the d -band complex of a transition metal). ToF k -microscopy gives access to the spin-integral array ρ , weighted by the photoemission cross section. By using different photon polarizations, ρ gives access to the CDAD and LDAD texture (cf. Sections 2.2 and 5.1). An imaging spin filter allows to measure the spin-polarization vector $\mathbf{P}=(P_x, P_y, P_z)$, also depending on E_B and $\mathbf{k}$. The present set-up only allowed mapping of component P_y , the extension to vectorial detection will give access to P_x and P_z as well, as described in [17].

The bulk and surface electronic structure are accessible at high and low photon energies, exploiting the energy-dependence of the inelastic mean free path. For the bulk analysis, we have used the high-brilliance soft X-rays of beamline P04 at PETRA III (DESY, Hamburg). The perpendicular momentum k_z was varied stepwise across the 4th repeated BZ by using 14 selected values for the photon energy (Fig. 4(b)). All 3D arrays are concatenated yielding the complete 4D spectral function $\rho(E_B, \mathbf{k})$. The *Fermi surface* was derived as the boundary of the occupied part of ρ . The Iridium Fermi surface is fragmented into two concentric electron sheets around the Γ -point and two concentric hole pockets centered at the X-points. The *Fermi-velocity* distribution $\mathbf{v}_F(\mathbf{k}_F)$ ($\sim \nabla E(\mathbf{k})|_{E_F}$) was determined by numerical differentiation. It varies between extremal values of -10^6 m/s for the hole pockets and $+10^6$ m/s for the electron pockets (cf. color scale in Fig. 5). *Electron or hole character* are visible in the curvature of the bands near E_F and the *inner potential and effective mass* can be accessed via the identification of high-symmetry points in the k_z -dependence. The analogous analysis for tungsten revealed a smaller velocity for the hole pockets [51].

The surface resonances on the Ir(111) surface and their dichroism and spin-polarization textures have been measured at low photon energies at beamline U125-2 (normal-incidence monochromator) of BESSY II (Berlin). Surprisingly, the iridium bulk bands are well visible in the VUV range so that the spin-polarization texture of surface and bulk bands could be measured in a single measurement. Linear dichroism and spin polarization of the topmost two bulk bands reaches 50%. It seems to constitute a special case because bulk bands are still quite intense at low energies. In this photon-energy range bulk bands are completely invisible for Mo(110) [52] and W(110) [22]. This interesting result has been further investigated in a recent comparative study [53]. The dichroism patterns contain information about the orbital symmetries of the wave-functions involved in the photo transition. The LDAD of all bands shows a pronounced, uniform $\pm$ texture. However, in hybridization regions it varies rapidly (including changes of sign) due to the changes in the partial-wave composition of the initial-state wavefunctions. The CDAD texture in the soft X-ray range enables us to discriminate between the different orbital-wavefunction composition of the two concentric Fermi-surface sheets around Γ and X.

One-step photoemission calculations including dichroism and spin texture are in fair overall agreement with experiment. The *spin-resolved Bloch spectral function* exhibits high spin polarization for the surface resonances but practically zero polarization for the bulk bands. However, for the same bands the *photoemission calculation* yields sizeable degrees of spin polarization (Fig. 3(f-h)) and the experimental values are even higher (Fig. 7(o)). This proves that the high spin polarization (up to 50%) observed for bulk bands arises in the photoemission process.

It originates from the spatial dissymmetry of the photoemission process and requires significant spin-orbit interaction. The degree of polarization is determined by the matrix elements and phase differences of final-state partial waves. Electrons in surface states experience strong electric-field gradients, mostly the potential gradient of the first atomic layer. The resulting Rashba-Hamiltonian [54] causes a ground-state spin orientation in the near-surface region, showing up as highly-polarized surface bands in the Bloch spectral function (Fig. 3(c,d)). The different contributions to photoelectron spin polarization are also discussed in [55].

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Short version:

On the quest towards maximum information the prototypical high-Z fcc metal iridium has been investigated using spin-filtered time-of-flight momentum microscopy. Large partial bandgaps and strong spin-orbit interaction lead to a sequence of spin-polarized surface resonances. A combined study of vacuum ultraviolet and soft X-ray photoemission give access to the bulk and surface spectral functions. The Fermi surface and all other energy isosurfaces, Fermi velocity distribution $vF(kF)$, electron or hole conductivity, effective mass and inner potential can be obtained from the measured 4D data array by simple algorithms. The complete analysis of the electronic structure requires measurements of spectral function, dichroism, and spin polarization.