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# Anomalous *d*-like surface resonances on Mo(110) analyzed by time-of-flight momentum microscopy

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ABSTRACT

The electronic surface states on Mo(110) have been investigated using time-of-flight momentum microscopy with synchrotron radiation ($h\nu = 35$ eV). This novel angle-resolved photoemission approach yields a simultaneous acquisition of the $E$-vs-$\boldsymbol{k}$ spectral function in the full surface Brillouin zone and several eV energy interval. ($k_x$,$k_y$,$E_B$)-maps with 3.4 Å$^{-1}$ diameter reveal a rich structure of *d*-like surface resonances in the spin–orbit induced partial band gap. Calculations using the one-step model in its density matrix formulation predict an anomalous state with Dirac-like signature and Rashba spin texture crossing the bandgap at $\bar{\Gamma}$ and $E_B = 1.2$ eV. The experiment shows that the linear dispersion persists away from the $\bar{\Gamma}$-point in an extended energy- and $k_{\parallel}$-range. Analogously to a similar state previously found on W(110) the dispersion is linear along $\bar{H}-\bar{\Gamma}-\bar{H}$ and almost zero along $\bar{N}-\bar{\Gamma}-\bar{N}$. The similarity is surprising since the spin–orbit interaction is 5 times smaller in Mo. A second point with unusual topology is found midway between $\bar{\Gamma}$ and $\bar{N}$. Band symmetries are probed by linear dichroism.



## 1. Introduction

The spin–orbit interaction of electronic surface bands has recently attracted much interest because it induces novel emerging phenomena. The Rashba effect [1] in metals couples the electron spin to its momentum leading to large magnetotransport effects. In topological superconductors the non-abelian statistics of Majorana fermion zero modes [2] may be used as a building block for a topological quantum computer. In insulators a strong spin–orbit coupling may bridge the energy gap forming spin-polarized Dirac-type surface states. Prominent examples of these topological insulators are $Bi_2Te_3$ and other Bi-chalcogenides [3].

The amount of spin–orbit splitting observable in a corresponding surface state is controlled by the strength of the reflection properties of the semi-infinite bulk, namely by the so-called bulk reflection matrix. This means that the multiple scattering between bulk and surface is responsible for the absolute value of the Rashba splitting of a surface state [4]. Another important aspect is the connection of the topological surface state to the bulk valence band [5] where hybridization gaps explain unexpected weak surface-bulk mixing.

The criteria for the appearance of Dirac-states at surfaces of topological insulators are well-understood and many materials exhibit these special, topologically-protected states. They show insulating behavior in the bulk and a metallic surface state. Therefore it was a surprise that recently a spin-polarized state with linear dispersion was discovered in a spin–orbit-induced symmetry band gap of W(110) [6]. Unlike the Bi chalcogenides with $C_{3v}$ symmetry, the anomalous surface resonance on W(110) has *d*-character and the surface has $C_{2v}$ symmetry. Due to its linear dispersion in an energy range of 220 meV and Rashba-type spin signature the state on W(110) is reminiscent of a topological edge state and has been referred to as “Dirac-like” or massless [6]. Its special nature was confirmed by two ab-initio calculations [7,8] arriving at identical conclusions.

Although W has a large spin–orbit interaction and this state arises in a spin–orbit induced partial bandgap, the existence of a state with Dirac-type signature on a metal surface was unexpected. The question, whether its existence is connected with a non-trivial (local) topology is still under debate. Experiments exploring other heavy bcc metals like Ta are being performed, in order to shed more light into this finding. Recently, inverse

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photoemission revealed an analogous state for Ta above the Fermi level $E_F$ [9] but excluded a state below $E_F$.

In the present work we address the question whether a state of this type exists on the isoelectronic surface of Mo(110). This is particularly interesting because spin–orbit interaction in Mo ($Z$=42) is about a factor of 5 weaker than in W ($Z$=74) [10] and thus maybe insufficient for the appearance of an anomalous state. On the other hand it is well known that W and Mo show very similar surface band structures [11–16]. However, the question of anomalous, "Dirac-like" surface resonances was not addressed in literature prior to the work of Miyamoto et al. [6]. Below we present a detailed experimental and theoretical study of the *d*-like surface resonances on Mo(110). The novel technique of time-of-flight (ToF) momentum microscopy, here for the first time using synchrotron radiation at BESSY II ($h\nu$=35 eV), yields the 3D representation of the $E$-vs-$\boldsymbol{k}$ relation in the full surface Brillouin zone (SBZ) for an energy interval of several eV. The 3D ($k_x$,$k_y$,$E_B$)-data matrix of $10^6$ voxels allowed to analyze the topology of bands in much detail. Non-relativistic band symmetries were probed by linear dichroism in the angular distribution (LDAD). The analysis revealed an exceptional state with linear dispersion in the spin–orbit band gap at the $\bar{\Gamma}$-point, 1.2 eV below $E_F$. In full analogy to the finding for W(110) [6], we observe a Dirac-like signature, Rashba spin texture (calculated) and high anisotropy. The band shows linear dispersion along $\bar{\Gamma}-\bar{S}$ and $\bar{\Gamma}-\bar{H}$ but weak dispersion in $\bar{\Gamma}-\bar{N}$. The linear dispersion persists in sections parallel to the $\bar{\Gamma}-\bar{H}$ direction along the $\bar{\Gamma}-\bar{N}$ line. A second special point, characterized by a linear dispersion in an energy range of 0.8 eV, occurs midway between $\bar{\Gamma}$and $\bar{N}$.

## 2. Experimental technique

In momentum microscopy the *reciprocal* image is detected with ultimate *k*-resolution; Tusche et al. [17] reported a resolution value of $<5\times10^{-3}$ Å$^{-1}$. Owing to $k_\parallel$-conservation in the photoemission process, the reciprocal image directly yields the surface-projected band structure inside the crystal. This image occurs in the back-focal plane of the cathode lens [18]. The intriguing advantage is that this novel approach to angular-resolved photoelectron spectroscopy gives access to the $k_\parallel$-distribution of the full half-space above the solid at kinetic start energies up to 80 eV, and still a solid angle of $\pi$ at start energies as high as 5000 eV [19]. Corresponding regions in *k*-space exceed the first *Brillouin* zone.

The time-of-flight (ToF) momentum microscope was jointly developed by University of Mainz and Max–Planck-Institute of Microstructure Physics, Halle. It combines the full-field *k*-imaging properties of a cathode-lens optimized for best *k*-resolution with the superior energy resolution and parallel acquisition capability of imaging ToF electron microscopy [20] that is principally capable of an energy resolution in the few-meV range [21]. It requires pulsed photon sources like synchrotron radiation, lasers or high-harmonic sources. Fig. 1(a) shows the experimental geometry and (b) illustrates the method of data acquisition: all counting events in the $E$–$k_\parallel$ paraboloid confined by the Fermi-energy cut at $E_{kin}^{max}$ and the photoemission horizon (condition $k_\perp$=0) are registered and accumulated in the ($k_x$,$k_y$,$E_{kin}$)-voxels of the data matrix, using a 3D image detector of the delay-line type [22,23]. Since the full half space above the sample and an energy interval of several eV is detected simultaneously, this data matrix is obtained at a fixed setting of the momentum microscope. The time resolution of the present detector was 180 ps, yielding an energy dispersion of $\Delta E$=0.255 meV $(E_d/\text{eV})^{3/2}$ (for 900 mm drift section), cf. [20]. Typical drift energies between $E_d$=40 eV and 4 eV thus lead to theoretical energy resolutions between $\Delta E$=60 meV and 2 meV, respectively. The delay-line detector (active area 40 mm diameter, spatial resolution 80 µm) resolves 500 points along the image diagonal at a maximum integral count rate of 5 Mcps. ToF momentum microscopy thus yields 3D "*k*-space objects" without any sweeping of lenses or rotation of sample or detector.

Energy and momentum resolution of the present instrument were determined as 19 meV and $10^{-2}$ Å$^{-1}$, respectively (not in the synchrotron experiment but using a picosecond laser). This yields $10^4$–$10^5$ resolved *k*-points, depending on the diameter of the momentum disk. In the synchrotron experiment the total energy bandwidth (containing contributions of photon bandwidth, thermal broadening, electronic noise in the total setup and intrinsic ToF resolution) was 80 meV, measured at a drift energy of 20 eV and a sample temperature of 140 K. The depth of focus was 2–3 eV, yielding 30–40 simultaneously resolved energy surfaces. A size-selectable and adjustable field aperture located in the first Gaussian image plane defines the source spot on the sample surface (in the present experiment 80 µm diameter), independent from the focusing quality of the photon source. For sufficiently high photon flux density on the sample, a field of view down to the micrometer range can be selected. Details of the instrument will be published elsewhere.

The measurements were performed using Synchrotron radiation of BESSY II at beamline *U125-2 SGM* [24] during single-bunch operation (pulse period 800 ns). A spherical grating monochromator (500 l/mm) provides photons in the energy range 29–138 eV. Given the work function of Mo(110) of 4.95 eV [25] this results in a kinetic energy range of 30 eV with the lowest 10 eV being cut off by the transfer lens to the ToF section. During single-bunch operation the beam current was below 10 mA, hence the photon flux was of the order of $10^{10}$–$10^{11}$ photons per second.

Initially, the Mo(110) crystal was cleaned by many repeated cycles of heating in oxygen at 1500 K followed by sublimation of

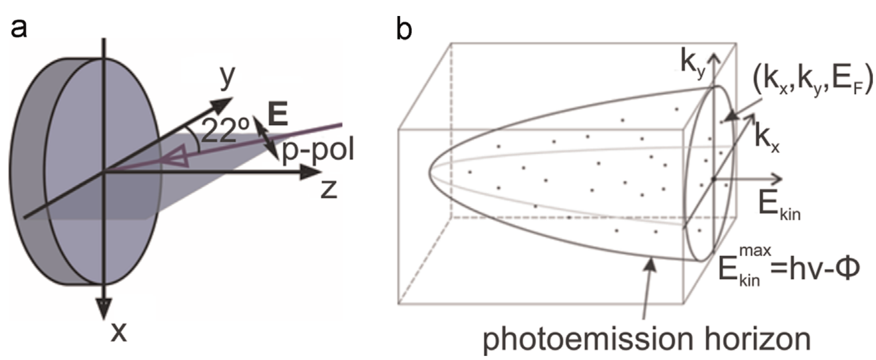


**Fig. 1.** Geometry of the experiment (a) and scheme of the 3D data acquisition of the ToF momentum microscope (b). The angle of incidence was 68° with respect to the surface normal; the photon beam was p-polarized with electric field vector $\boldsymbol{E}$ in the $y$–$z$ plane. The 3D data matrix in (b) is confined by the paraboloid-shaped photoemission horizon and the cut through the Fermi energy, visible as $E_{kin}^{max}$.

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