Spin- and angle-resolved photoemission (PE) as well as inverse photoemission (IPE) provide us with detailed information about spin-orbit-influenced electronic states [1–5]. Despite the undoubtedly wealth of experimental results, the information content with respect to the spin polarization is still under debate [6,7]. Recently, spin- and angle-resolved PE experiments on the topological nontrivial surface state of Bi2Se3 suggested additional spin-polarization effects on top of the intrinsic spin polarization. The measured spin polarization of photoelectrons excited from the surface state could even be manipulated by the polarization of the incident light [8,9]. Furthermore, spin- and angle-resolved IPE from the paradigmatic Rashba system Bi/Ag(111) showed that the measured spin information does not always reflect the intrinsic spin texture of the surface states. For states with mixed orbital symmetries, the measured spin information may be ambiguous and has to be taken with skepticism [10]. The effects described so far are reported for surfaces with threefold rotational symmetry (point group C3v).

Beyond that, it was shown that spin-polarized photoelectrons, excited by unpolarized light, are emitted from unpolarized bulk-derived electronic states even in normal-emission geometry from Pt(110), Pt(100), Pt(111), and Au(111) [6,11–13]. In these experiments, the observed spin features of photoelectrons were found to be strongly influenced by the crystal symmetry. Similarly, a recently described Dirac-like surface state on W(110) shows spin polarization even for normal photoelectron emission, excited by unpolarized light [14,15]. Besides, W(110) is a prototype system for studying spin-orbit coupling effects with spin-resolved photoemission techniques, since it also hosts Rashba-type bands, spin split for off-normal electron emission [16].

Considering the above, the appearance of spin polarization in (I)PE may be regarded as usual and not surprising. This raises the question on specific features of the involved electronic states which produce or do not produce large spin signals in (I)PE and how they can be tuned. To answer this question, a well-understood surface system, such as W(110), lends itself to such an investigation.

In this paper, we report on a joint experimental and theoretical study. We show that W(110) hosts simultaneously electronic states which produce a vanishing and a large spin signal. Detailed spin-resolved IPE experiments are presented and then explained by model calculations as well as ab initio electronic-structure calculations, allowing one to pinpoint the origin of the effects.

We choose an experimental situation with high symmetry: (i) normal electron incidence on the W(110) surface, (ii) spin-polarization directions within mirror planes of the surface, and (iii) detection of unpolarized light. Importantly, we detect the emitted photons under several different take-off angles. Thereby, we deliberately break the symmetry of the setup and are able to tune sign and magnitude of the spin signals. This IPE approach with multiple photon detectors under different detection angles is advantageous with respect to typical PE setups with only one photon source at a fixed angle of incidence.

The spin- and angle-resolved IPE experiments have been performed with ROSE, our rotatable spin-polarized electron source [17]. A spin-polarized electron beam was employed to investigate the spin-dependent unoccupied electronic structure of the W(110) surface. The transversal spin polarization of the beam is rotatable. In particular, for normal electron incidence, the spin polarization can be aligned with the two mirror planes of the surface. Here, we mainly focus on two highly symmetric configurations with spin polarization either along $x$ or $y$, i.e., along $\Gamma N$ or $\Gamma H$ of the surface Brillouin zone, as defined in Fig. 1(a). The emitted photons with an energy of $h\omega = 9.9$ eV are detected by several Geiger-Müller counters with different detection angles. The total-energy resolution of the IPE experiment is about 350 meV [18].

The W(110) surface was cleaned by repeated cycles of heating in an oxygen atmosphere ($6 \times 10^{-8}$ mbar to $1 \times 10^{-8}$ mbar) at 1500 K and subsequent flashing to 2300 K.
During the final flash, the pressure did not exceed $6 \times 10^{-7}$ mbar. This cleaning procedure was successful to remove contaminants, such as carbon and oxygen, from the surface. The surface quality was confirmed by Auger electron spectra and by a sharp $(1 \times 1)$ low-energy-electron diffraction (LEED) pattern with low background intensity [Fig. 1(a)].

The electronic-structure calculations were performed within the framework of the local spin-density approximation to density functional theory. Our computations rely on the relativistic layer Korringa-Kohn-Rostoker method, in which spin-orbit coupling is accounted for by solving the Dirac equation [19,20]; for details, see Ref. [21]. The W(110) surface is treated as a semi-infinite system. The top layer is relaxed inward by $\Delta d = -3\%$ with respect to the bulk interlayer distance, in agreement with experiments [22,23].

From the layer-resolved Green function $G_{ll}$ of the entire system, we calculate the spectral density $n_{l}(E,k_{\parallel})$ for layer $l$ at energy $E$ and surface-parallel wave vector $k_{\parallel}$:

$$n_{l}(E,k_{\parallel}) = -\frac{1}{\pi} \text{Im Tr } G_{ll}(E,k_{\parallel}).$$

The spectral density is further decomposed with respect to angular momentum and spin projection. Spin textures are discussed by means of spin differences $n_{\uparrow\downarrow}(E,k_{\parallel}) - n_{\downarrow\uparrow}(E,k_{\parallel})$, in which $\uparrow$ and $\downarrow$ refer to a specified quantization axis.

Figure 1(c) presents a spin-integrated IPE spectrum for normal electron incidence on W(110), which exhibits four distinct features. W0 just above the Fermi energy $E_{F}$ is attributed to a surface state with $p_{z}$ character, and IS at 4.5 eV to an image-potential-induced surface state [24,25]. W1 and W2 at 2.3 and 3.2 eV, respectively, are assigned to transitions into $d$ states of tungsten [24].

Three photon detectors with different take-off angles $(\alpha,\beta)$ have been used: $(70^\circ,180^\circ)$ for counter C1, $(35^\circ,90^\circ)$ for C2, and $(35^\circ,0^\circ)$ for C3, as sketched in Fig. 2(a). The polar angle $\alpha$ and the azimuthal angle $\beta$ are defined with respect to the Cartesian coordinate system for the nonrotated sample $(\phi = 0^\circ)$, as shown in Fig. 1(b); the $z$ axis coincides with the electron incidence direction, i.e., the surface normal.

Spectra taken with an electron spin polarization along $y$ [Fig. 2(b)] show a remarkable result. While the spin asymmetry of W2 is zero for all counters, it exhibits values larger than 30% for W1: positive for C1, zero for C2, and negative for C3. This result is surprising because electronic states at $\Gamma$ have no intrinsic spin polarization due to time-reversal symmetry. Interestingly, the spin-asymmetry data with opposite sign were obtained by C1 and C3, which lie on opposite sides of the surface normal. Additional experiments with the electron spin polarization along $x$ (by rotation of the electron source) do not lead to any spin asymmetry for all counters [Fig. 2(c)]. This reflects the twofold symmetry of the bcc(110) surface. In a control experiment, we rotated both the electron spin polarization and the sample by 90° [Fig. 2(d)]. This geometry differs from the one used for the data in Fig. 2(b) in that C1 and C3 are now in equivalent positions as C2 was before, and C2 is now in a position like C1. As a consequence, the result of C2 is expected to resemble the former result of C3, while C1 and C3 should not show any spin asymmetry. Both expectations are convincingly met by the experimental results.

Our results show that for W1, we can deliberately produce a desired spin signal from an unpolarized state, i.e., positive, zero, or negative, by carefully choosing the experimental geometry. This is demonstrated by additional results, shown in Fig. 2(e), in which the spin asymmetry of W1 is shown as a function of the azimuthal rotation angle $\phi$ of the sample. In this experiment, the electron spin polarization $P$ of our ROSE was set along the $y$ axis of the nonrotated sample. By rotating the sample, the spin signal of W1 reveals a sinusoidal dependence. Its periodicity is 180° for both C1 and C3, while it is 90°, including a sign change, for C2.

For W2, independent of the experimental geometry, no spin signal is observed, in striking contrast to W1. Two questions arise: What is the origin of this effect? What distinguishes W1 from W2?

Figure 3 presents calculations of the electronic structure of W(110) along $\Gamma$N: spectral densities $n(E,k_{\parallel})$ for the bulk [Fig. 3(a)], for the surface layer [Fig. 3(b)], and the spin difference of the spectral densities for the surface layer [Fig. 3(c)]. At the energies of W1 and W2, high spectral density is found in the bulk and at the surface. A pure surface band at lower energies with downward dispersion is attributed to W0.

To gain deeper insight into the different spin behavior of W1 and W2, we analyzed their orbital and symmetry characters. In Fig. 3(d), the orbital decomposition of the surface spectral density is shown for symmetries for which the density is nonzero. While W0 appears with an almost pure $p_{z}$ orbital character and W2 with an almost pure $d_{xz}$ character, W1 consists of two orbital contributions, $d_{xz}$ and $d_{x^{2}−y^{2}}$. We can...
FIG. 2. (Color online) (a) Experimental geometries of counters C1, C2, and C3, with photon-detection angles specified. (b)–(d) Spin-resolved normal-incidence IPE spectra of W(110) and corresponding spin-asymmetry data for \( \phi = 0^\circ \) (sensitive to the spin-polarization components \( P_y \) and \( P_x \)) and \( \phi = 90^\circ \) (sensitive to \( P_x \)). Panels with nonzero spin asymmetry are highlighted by frames with thicker linewidth. (e) Spin asymmetry of W1 vs azimuthal angle \( \phi \) for spin polarization \( P \) perpendicular to the plane of incidence.

FIG. 3. (Color online) Calculated electronic structure of W(110) along \( \overline{\Gamma\Gamma} \). Spectral densities \( n(E,k_||) \) (a) for a bulk layer and (b) for the topmost surface layer, sharing a common color scale (dark is small values and bright is large values). (c) Spin difference \( n_\uparrow(E,k_||) - n_\downarrow(E,k_||) \) of the spin-projected surface spectral density, illustrated in red (blue) where spin-up (spin-down) intensity prevails (white denotes zero spin difference). (d) Orbital decomposition of the spectral density (surface and subsurface layer) with assigned symmetry representation.

We now discuss these findings in the framework of a group-theoretical analysis of photoemission [27]. Within this very general approach, we restrict our present analysis to dipole-transition effects for linearly polarized light and non-magnetic materials with \( C_{2v} \) symmetry. The spin asymmetries \( A_y \) \((A_x)\) are measured for excitation by electrons with spin polarization \( P_y \) \((P_x)\). The theoretical analysis predicts the assign the irreducible representation of the single group \( C_{2v} \) to the orbital contributions (neglecting spin-orbit interaction) [26]: \( \Sigma_1 \) to W0 and W2, \( \Sigma_1 \) and \( \Sigma_4 \) to W1. This mixed symmetry character of W1 has important consequences for its spin signal.
following: For detection of \( p \)-polarized light, a spin signal \( A_y \) is only generated by a mixing of \( \Sigma_1 \) and \( \Sigma_4 \) (\( \Sigma_1 \) and \( \Sigma_4 \)) symmetry, while for \( s \)-polarized light, no spin signal \( A_x, A_y \) is induced. By detecting unpolarized light (which is an incoherent superposition of \( s \)- and \( p \)-polarized light), the \( p \)-polarized part leads to a spin signal. Since \( W1 \) is a mixed state of \( \Sigma_1 \) and \( \Sigma_4 \), a spin signal is expected for \( A_y \) but not for \( A_x \). In contrast, no spin asymmetry is produced for \( W2 \) due to its pure representation. This is exactly what is observed in the high-symmetry point\( \Gamma \) sinusoidal model curves included in Fig. 2(e).

Away from it, Rashba-type spin asymmetry is induced. By detecting unpolarized light (which is an incoherent superposition of \( s \)- and \( p \)-polarized light), the \( p \)-polarized part leads to a spin signal. Since \( W1 \) is a mixed state of \( \Sigma_1 \) and \( \Sigma_4 \), a spin signal is expected for \( A_y \) but not for \( A_x \). In contrast, no spin asymmetry is produced for \( W2 \) due to its pure representation. This is exactly what is observed in Figs. 2(b)–2(d). From theory, it follows that for dominating \( \Sigma_1 \), the spin asymmetry \( A_y \) can be written as \( A_y \propto c(\alpha) \cos \beta \), with \( c(\alpha) \) being a polar-angle-dependent amplitude. In agreement with this prediction, \( A_y \) is positive for \( C1 \) (\( \beta = 0^\circ \)), 0 for \( C2 \) (\( \beta = 90^\circ \)), and negative for \( C3 \) (\( \beta = 180^\circ \)) in Fig. 2(b). The observed spin signals, reproduced in Figs. 2(b)–2(d), are well explained by this analysis.

Next, we address the spin-asymmetry signal observed as a function of azimuth \( \phi \) in Fig. 2(e). Due to the concomitant rotation of the photon-detection angles relative to the sample by \( \phi \), the above angular dependence becomes \( A_y \propto c(\alpha) \cos(\beta - \phi) \). The observed spin asymmetry \( A_y \) is expressed as \( A_y \propto \sin \phi + A_y \cos \phi \). Since \( A_y = 0 \), we arrive at

\[
A \propto \begin{cases} 
-c(\alpha) \cos^2(\phi) & \text{for } C1 \ (\beta = 180^\circ) \\
-c(\alpha) \cos(\phi) \cos(90^\circ - \phi) & \text{for } C2 \ (\beta = 90^\circ) \\
-c(\alpha) \cos^2(\phi) & \text{for } C3 \ (\beta = 0^\circ). 
\end{cases}
\]

Note that \( \alpha \) is not the same for all counters and, here, \( c(\alpha) \) is negative. These relations reproduce the periodicity of \( 90^\circ \) for \( C2 \) and \( 180^\circ \) for \( C1 \) and \( C3 \); they are used to derive the sinusoidal model curves included in Fig. 2(e).

So far, we have concentrated on the unexpected spin signal at the high-symmetry point \( \Gamma \). Away from it, Rashba-type spin dependence is expected for electronic states at surfaces of high-Z materials such as tungsten. Therefore, in Fig. 4, we show spin-resolved IPE spectra for \( W1 \) and \( W2 \) for off-normal electron incidence along \( \Gamma N \) (\( \phi = 0^\circ \)). The spin polarization was aligned to the Rashba component. The data for \( \beta = 0^\circ \), \( 90^\circ \), and \( 180^\circ \) (Fig. 2(b)) and for \( \theta = 0^\circ \) (Fig. 2(c)). The clear signature of a Rashba-type wave-vector-dependent spin polarization is supported by the spin difference of the surface spectral density \( n_\uparrow (E, k_z) - n_\downarrow (E, k_z) \), shown in Fig. 3(c). In the energy range of \( W2 \), the spectral density reverses sign, shown as faint blue for negative \( k \) and faint red for positive \( k \). We conclude that for \( W2 \), i.e., the state with pure symmetry, the measured spin asymmetry reflects the intrinsic spin texture of the electronic state. For \( W1 \), the calculations predict a Rashba spin splitting into two oppositely spin-polarized states. However, the splitting is too small to be resolved by our experiment. Therefore, we do not observe a spin dependence for \( W1 \) in \( C2 \). Apart from that, the strong experimental spin signal of \( W1 \) in \( C1 \) for normal electron incidence (discussed above) persists for off-normal electron incidence. It dominates any small Rashba-type spin signal, if experimentally detectable.

In conclusion, we showed that the spin signals of electronic states, observed by spin-resolved IPE, are not only influenced by their intrinsic spin polarization but also by the choice of symmetry-breaking experimental parameters in combination with the particular symmetry characters of the involved states. In our spin- and angle-resolved IPE study for normal electron incidence on \( W(110) \), we demonstrated how the spin signal can be deliberately tuned from negative to positive values in the absence of any intrinsic spin polarization. A group-theoretical analysis of the electronic structure revealed that this appears for states with mixed symmetry but not for pure states. The latter is convincingly confirmed by simultaneous measurements on a state of pure symmetry, which shows no spin signal. Our study has general impact in two ways: (i) the need to be careful with the interpretation of spin signals, which might not be intrinsic but offer insight into the orbital characters of the involved states, and (ii) the possibility of producing spin signals tunable in both sign and magnitude from unpolarized states.

It is a pleasure to thank P. Krüger for fruitful discussions. Financial support by the Deutsche Forschungsgemeinschaft (DFG) is gratefully acknowledged.


[26] Note that spin-orbit coupling results in a single irreducible representation ($\Sigma^3$) of the double group.