

Rashba-type spin splitting and spin interference of the Cu(111) surface state at room temperature

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Abstract

We report on the measurement of the Rashba-type spin splitting of the Shockley surface state on Cu(111) by spin- and angle-resolved photoemission at room temperature. Along the spatial direction expected for a Rashba-type effect the measured spin splitting corresponds to what has previously been reported by first principle calculations which were verified by high resolution ARPES using low temperatures and perfect crystals. Furthermore it is found that structural defects cause a spin-interference in the photoemission process and as a result the main measured spin signal is in the plane orthogonal to the typical Rashba orientation. Although the determination of the exact origin of this signal requires further investigations, the main results can be used as a bench mark for future spin-resolved photoemission set-ups.

Lifting the spin degeneracy of states at the Fermi level is one of the main prerequisites for the control of the electron spin in solids[1]. Besides using the exchange interaction in ferromagnetic materials another promising pathway is to use materials with large spin-orbit interaction (SOI). In combination with a breaking of the global inversion symmetry, either on a surface or due to stacking order, the SOI lifts the spin degeneracy according to the Rashba-Bychkov effect [2]. Although described for semiconductor heterostructures, and also observed therein by transport [3–5], the clearest observations have been made by (spin- and) angle-resolved photoemission ((S)ARPES).

Early results were on heavy metal single crystals such as Au(111) [6, 7], W(110) [8, 9], and the different faces of Bi [10, 11]. Later it was realised that not only pure crystals host Rashba-type spin split surface states, but that even larger splittings can be achieved in mixed surface structures on Ag(111) surfaces [12, 13], which are controllable through further mixing [14–16]. Also in thin films the inversion symmetry is broken as on one side is the substrate and on the other the vacuum. As a result Rashba-type spin splittings have been observed in quantum well states (QWS) in thin metal films on heavy metal substrates [17, 18]. A control of the size of the spin splitting can even be achieved for Pb QWS on semiconductor substrates [19–21]. In a 3D electronic structure a Rashba-type spin splitting can be obtained by breaking the inversion symmetry in the unit cell of a layered crystal. A prime example of this is BiTeI [22], where the 3D Fermi surface can be described by a spin polarized spindle torus [23].

A common characteristic of all the above-mentioned examples is that they involve high Z materials, typically from the 6th row of the periodic table. This has led to the assumption that such materials are a prerequisite for the observation of the Rashba effect and that the size of the splitting depends solely on the atomic spin-orbit interaction of the material involved. However, this is a strongly simplified picture as exemplified by the fact that the Rashba-type spin splitting of the Sb(111) surface state is larger as the one found on Au(111) [6, 24, 25], and that the spin splitting for the Pb surface alloy on Ag(111) is much smaller than for the Bi induced one [26]. This can be understood by considering the fact that the Rashba effect is not as much an atomic effect as a result of the complete electronic structure. An adequate picture can only be obtained if one considers the exact crystal structure, the orbital mixing, local dipoles, and their influence on the shape of the wave function around an atom core [27, 28].

Here we will show that also in the relatively low Z material Cu the Shockley surface state on the (111) surface is spin split according to the Rashba effect. The size of the splitting is found to be less than 4 times smaller as for the Au(111) surface state and corresponds well to recent laser ARPES results for a carefully prepared Cu(111) surface at low temperatures [29]. In contrast, our results are obtained at room temperature on surfaces with a large number of defects, which again shows the power of SARPEs for the study of such phenomena [30]. This is especially the case for systems where perfect defect-free crystals can't be obtained.

Without going in the detailed background of the Rashba effect we review the resulting band structure for a simple parabolic band in Figure 1 (a). The SOI splits the parabolae by a value $\Delta k = 2k_0$ resulting in an energy splitting that increases linearly with in-plane momentum. For a perfectly isotropic dispersion in the absence of a strong coupling of the electronic structure to the momentum (or so called warping) the spin polarization vector lies in the surface plane perpendicular to the momentum vector. The resulting Fermi surface consists of two concentric circles with a helical spin texture. Based on the measured spin splitting and effective mass (m^*) the Rashba parameter can be obtained as follows: $\alpha_{RB} = \hbar^2 k_0 / m^*$.

The (S)ARPES measurements were performed at the COPHEE end station at the Swiss Light Source [31]. The data was obtained in 2009 before the installation of a six axis manipulator and hence the measured momentum range (k_x) is limited to the main scattering plane as defined by the incoming light and detected electrons. As a result the spin polarization signal expected for a Rashba-type spin splitting is oriented along the y -axis. The Cu(111) single crystal was cleaned using several cycles of Ar^+ sputtering and subsequent annealing at 300°C. The surface structure was determined from low energy electron diffraction (LEED) as shown in Figure 1 (b). The SARPEs measurements were performed after allowing the sample to cool down to room temperature (300K) using p-polarized light at photon energies ranging from 21 to 27 eV.

Figure 1 (c) and (d) show the measured Fermi surface and band dispersion along the $\bar{\Gamma} - \bar{M}$ direction of the Cu(111) surface state. A comparison to the results obtained in ref. [32] leads to the conclusion that in our case the binding energy of the band apex is lower and the diameter of the Fermi surface is slightly smaller. Here it should be noted that the Fermi surface is obtained by rotating the sample around the surface normal and the band map by rotating around an axis along the sample y direction. The smaller Fermi surface of

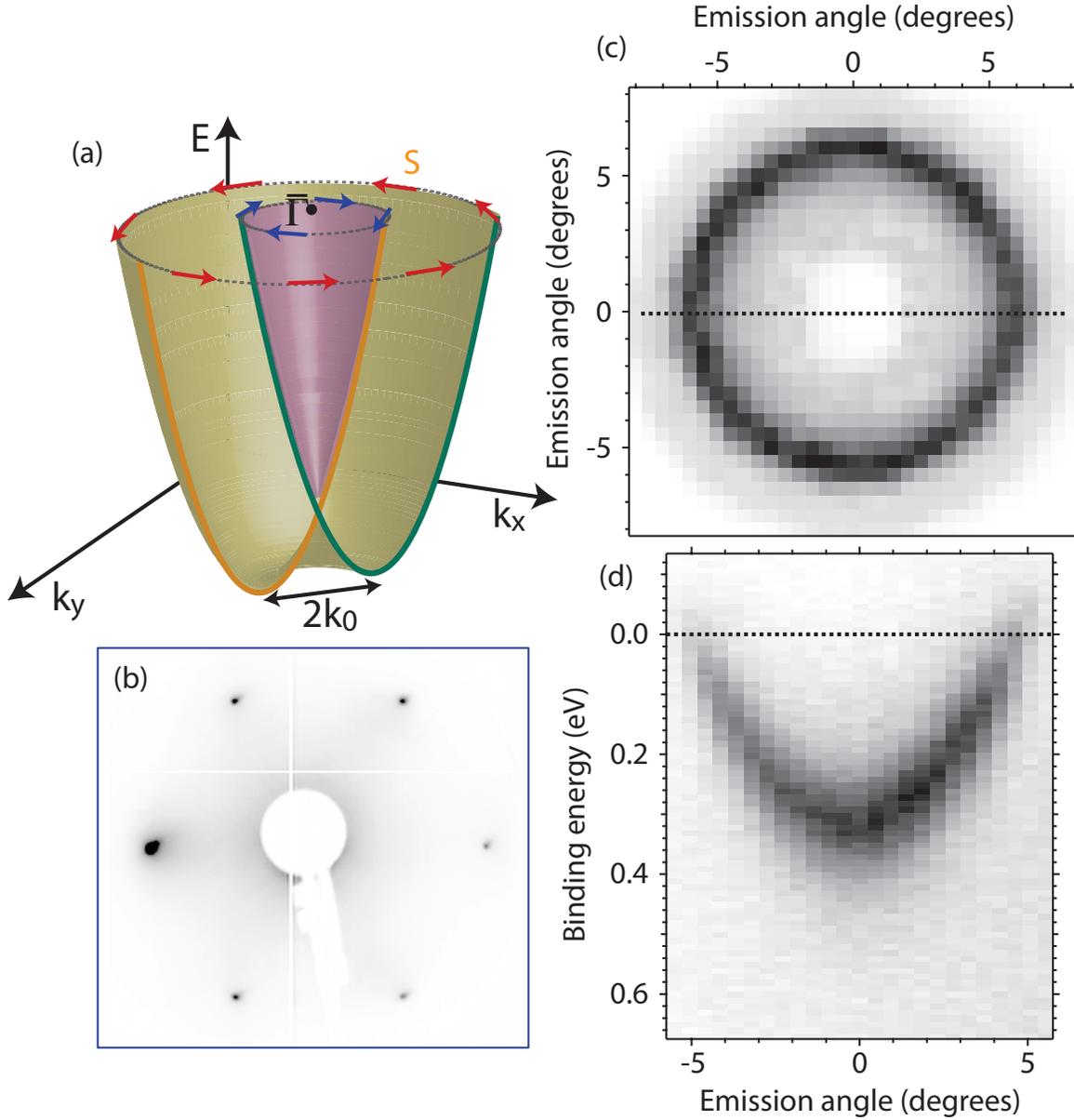


FIG. 1. (a) Illustration of Rashba effect. (b) LEED image of Cu(111). (c) Fermi surface and (d) ARPES band map of Cu(111) surface state along the Γ -M direction measured at a photon energy of 21 eV.

the surface state is an indication of a larger number of defects compared to previous studies [29, 32].

In Figure 2 we present the spin-resolved momentum distribution curve (MDC) obtained along the dashed line in Fig. 1 (c) and (d) for a photon energy of 21eV. The main spin signal appears along the x- and z-directions with an amplitude close to 30%. On the other

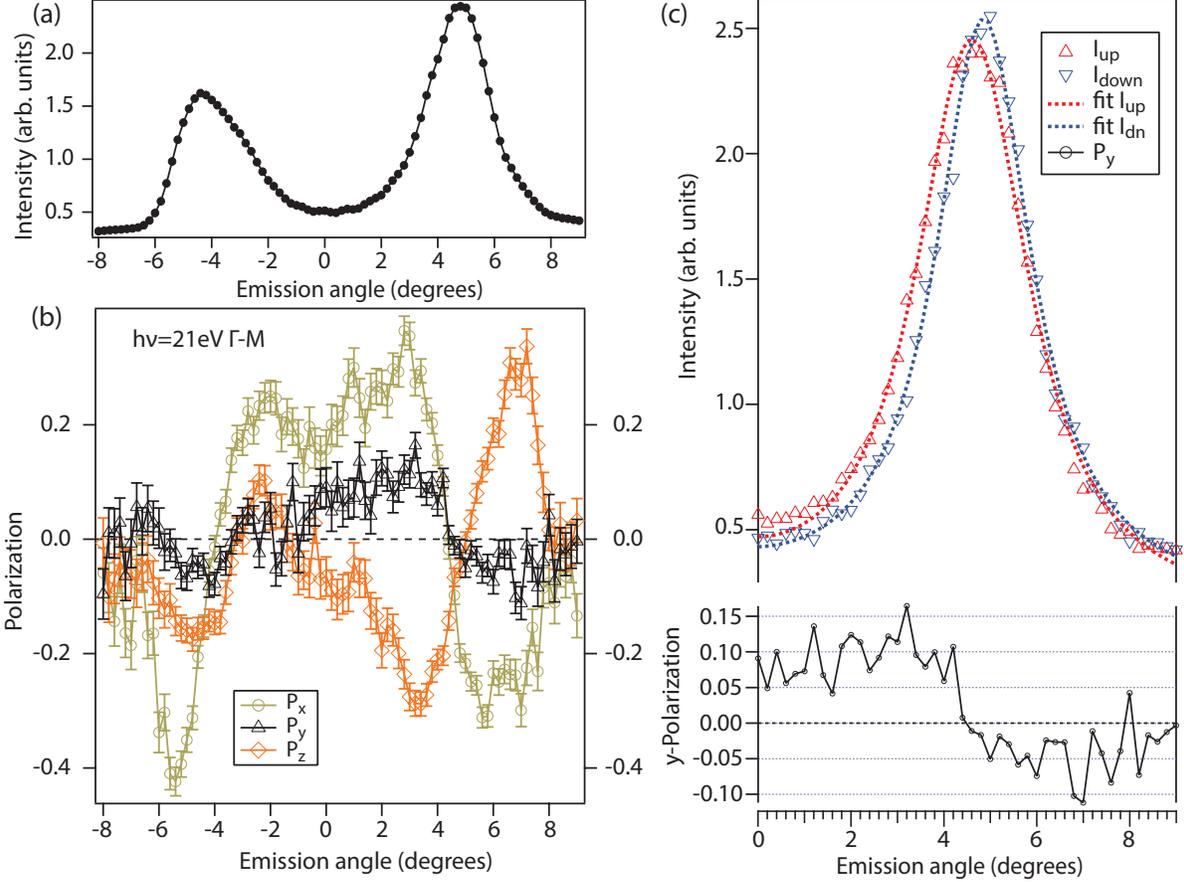


FIG. 2. (a) Total intensity of MDC measured with the Mott detectors at a photon energy of 21 eV. (b) Spin polarization along the three spatial directions. (c) Zoom in on the Fermi level crossing at positive angles for the y-polarization and the spin up and spin down intensities projected on this direction. The fits yield a splitting of 0.24° or 0.008 \AA^{-1} .

hand the spin signal along the y-direction, along which one would expect the Rashba signal for this geometry, is only about 5%. Furthermore, the spin signal along the x-direction is clearly symmetric with respect to $\bar{\Gamma}$. These unexpected P_x and P_z signals closely resemble observations for the Sb induced surface reconstruction on Ag(111) [33], where it was found that spin-interference in the photoemission process can induce a spin signal perpendicular to the direction expected for a Rashba signal [34]. This effect is part of a wide range of photoemission processes that can induce a spin signal many of which are well understood and others which remain unclear at the moment [35]. The measurements were repeated along the $\bar{\Gamma} - \bar{K}$ direction (Fig. 3 (a)) and with photon energies of 24 and 27eV (Fig. 3 (b)

and (c)) and apart from a change in the amplitudes no qualitative differences are observed. The measurements were obtained in the following order with intermediate sample cleaning after every experiment; 24 eV, 21 eV ($\bar{\Gamma} - \bar{M}$), 27 eV, 21 eV ($\bar{\Gamma} - \bar{K}$). Whether the change in signal amplitude is a result of the reduced surface contamination after every sputter cycle or that the photon energy has an intrinsic influence on the interference signal requires further systematic measurements. The expected reduction of the spin signal from a change in the peak-to-background ratio is visible along the Rashba (y-)direction in Figure 3 (d).

The main requirement for the observation of such an interference between two Rashba-split bands is that atomic defects induce a broadening of the bands of a similar magnitude as the splitting between the bands [34]. When this is the case the quasiparticle states with opposite spin can overlap and the spinors should be summed coherently. The resulting spin polarization will be in the plane perpendicular to the Rashba direction whereby the exact orientation is determined by the phase with which the two spinors are summed. In contrast to ref. [34] the interference spin signal is here not maximal at the peak position, but passes through zero there. This implies a change of phase by π (or sign reversal) when passing the middle between the Rashba-split states. The origin of this phase change is unclear at the moment and requires further theoretical and experimental investigations. A possible explanation of this node in the phase could be based on the relative size of the splitting with respect to the broadening of the quasiparticle states or on the exact orbital composition of the initial and final state. Such considerations might also help explain the unusual spin signal which was observed for epitaxial graphene [36] and of which it was later shown that it could not be explained by a simple Rashba scenario [37, 38].

It should be noted that a similar two level spin interference process was recently also used to explain variations in the spin signal observed for the surface state of the topological insulator Bi_2Se_3 [39]. In this case the interference does not occur between two separate states, but between the orbital components and their opposite spin helicities [40].

In the following we restrict ourselves to the signal along the y-direction as this will give an indication of the Rashba-type spin splitting. The MDC measurements were obtained starting at positive angles with a measurement time of approximately 7 minutes per data point because of the low spin signal. This means that the surface has degraded by the time the measurement reaches the bands at negative angles and the spin signal there is even lower. Therefore to obtain an accurate measure for the Rashba splitting we fit the spin-split peaks

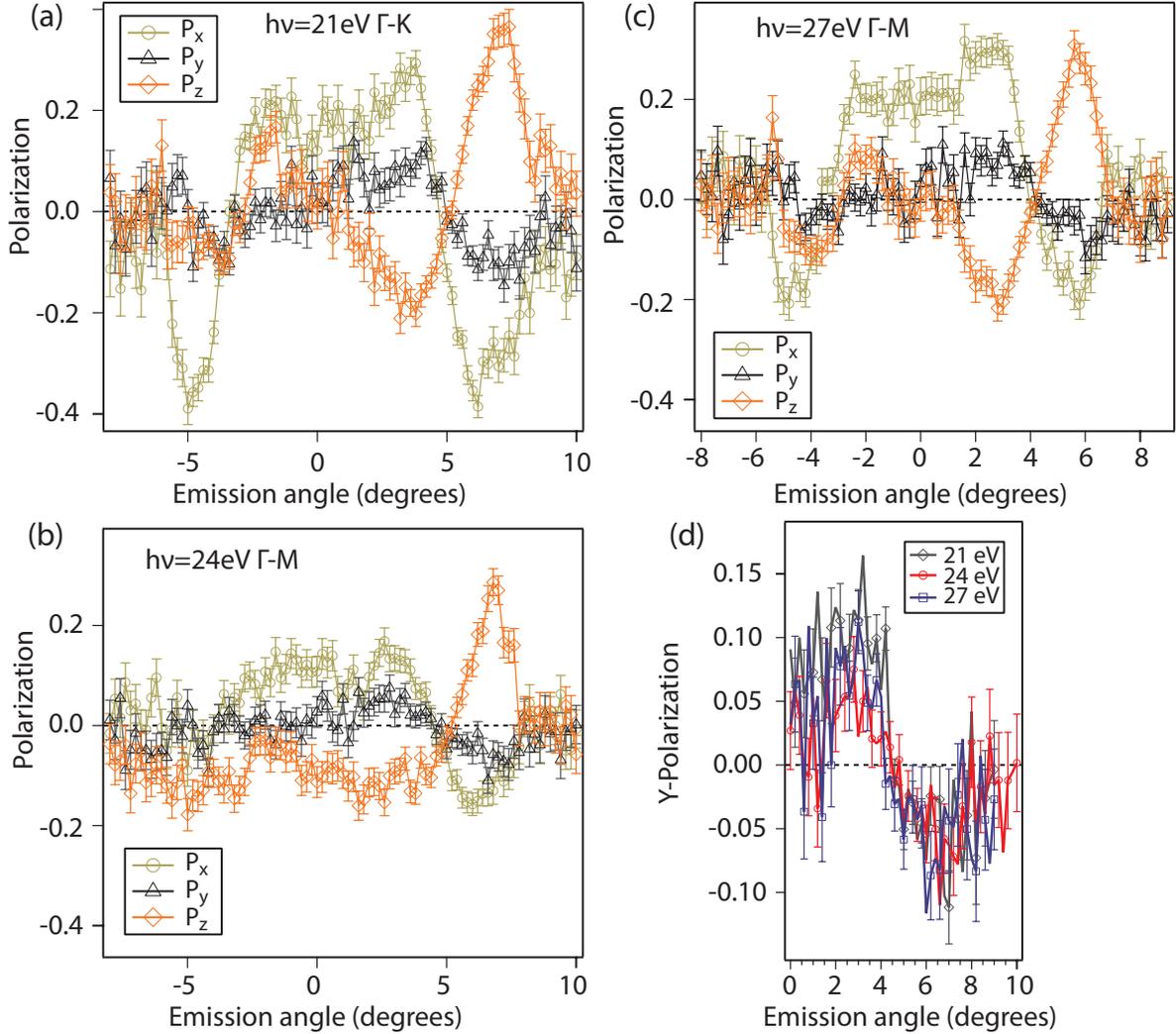


FIG. 3. Spin polarization measured along the three spatial directions for a photon energy of (a) 21eV along $\bar{\Gamma} - \bar{K}$, (b) 24eV, and (c) 27eV. (c) Zoom in on the Fermi level crossing at positive angles for the y-polarization for the three photon energies used along $\bar{\Gamma} - \bar{M}$.

at positive angles with voigt functions. The obtained splitting is 0.24° which corresponds to $\Delta k = 0.008 \text{ \AA}^{-1}$ and $\alpha_{\text{RB}} = 0.06 \text{ eV\AA}$. This is slightly larger than the value obtained with spin-integrated ARPES at low temperatures, but corresponds nicely to expectations from ab-initio calculations [29]. Given the fact that both the SARPES and ARPES measurements are at the limit of current technical possibilities it is difficult to judge whether this small discrepancy lies within the respective error bars, is due to a slight miscalibration in either set-up, or due to the difference in sample quality or temperature. Both in this respect and

to obtain a better understanding of the interference effects it would be interesting to study the same system with a further state-of-the art SARPES set-up [41].

To conclude, we have directly determined the Rashba-type spin splitting of the Cu(111) surface state by SARPES at room temperature. The splitting is found to be at the limit of what is currently measurable and can provide a bench mark for other SARPES experiments. The main spin signal is found to originate from an interference effect in the photoemission process which rotates the spin polarization vector to the plane perpendicular to the typical Rashba orientation.

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