Unconventional Fermi surface spin textures in the Bi$_{x}$Pb$_{1-x}$/Ag(111) surface alloy

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The Fermi and Rashba energies of surface states in the Bi$_{x}$Pb$_{1-x}$/Ag(111) alloy can be tuned simultaneously by changing the composition parameter $x$. We report on unconventional Fermi surface spin textures observed by spin and angle-resolved photoemission spectroscopy that are correlated with a topological transition of the Fermi surface occurring at $x = 0.5$. We show that the surface states remain fully spin polarized upon alloying and that the spin polarization vectors are approximately tangential to the constant energy contours. We discuss the implications of the topological transition for the transport of spin.

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Controlling the spin degree of freedom of the electron lies at the heart of spintronics [1]. One possibility to manipulate the electron spin without the need of any external magnetic field is found in the Rashba-Bychkov (RB) effect [2]. It appears in (quasi) two-dimensional electron or hole systems with a lack of inversion symmetry and plays a prominent role for a proposed spin field-effect transistor [3]. For most systems, the RB effect is small. Therefore, many of the related intriguing effects, such as a renormalization of the Fermi liquid parameters [4], changes in the electron-phonon coupling [5], enhanced superconductivity transition temperatures [6], and real space spin accumulation [7, 8, 9] remain for the most part experimentally unobservable.

Recently, it has been shown that the RB effect is dramatically enhanced in the Bi/Ag(111)($\sqrt{3}\times\sqrt{3})R30^\circ$ and Pb/Ag(111)($\sqrt{3}\times\sqrt{3})R30^\circ$ surface alloys due to an additional in-plane inversion asymmetry [10, 11, 12, 13, 14]. Furthermore, the band structure can be continuously tuned between these two systems by substituting Bi with Pb [15], as schematically illustrated in Fig. 1(a). The large RB effect combined with the tunability of the Fermi and the RB energies make Bi$_x$Pb$_{1-x}$/Ag(111)($\sqrt{3}\times\sqrt{3})R30^\circ$, henceforth Bi$_x$Pb$_{1-x}$/Ag(111), an ideal model RB system to study the geometrical and the topological changes in the Fermi surface of its surface states [5, 10]. It is clear that the large conductivity of the Ag substrate short-circuits possible spin currents at the surface of Bi$_x$Pb$_{1-x}$/Ag(111), but RB semiconductors [17] or thin metallic films [18] might be found that are equally tunable and suited for technological applications.

We present in this work spin and angle resolved photoemission spectroscopy (SARPES) data on surface states of Bi$_x$Pb$_{1-x}$/Ag(111) to resolve the changes in their Fermi surface spin textures (FSST) as a function of composition $x$. We will argue that the spin transport is strongly affected by a topological transition of the Fermi surface taking place at the critical value $x_c = 0.5$.

The RB effect occurs at interfaces or surfaces whenever the absence of the space inversion symmetry lifts the spin degeneracy due to the spin-orbit coupling. The simplest example of a RB Hamiltonian is given by [19],

$$H = H_0 + H_{RB},$$

where the kinetic energy of the two-dimensional hole gas with negative effective mass $m^*$ is

$$H_0 = \sigma_0 \left( E_F - \frac{\hbar^2}{2m^*} \nabla^2 \right),$$

FIG. 1: (color online) (a) Qualitative plot of the surface state band structure of Bi/Ag(111) ($x = 1$) along the direction $\bar{\Gamma}K$ in momentum space (adapted from Ref. [13]) showing the two Kramer’s pairs K1 and K2. As $x$ is decreased, the Fermi level (dashed lines) lowers continuously and the spin splitting becomes smaller (not shown). (b) Schematic picture of the Rashba effect for a two-dimensional hole gas around $\bar{\Gamma}$ and illustration of the relevant parameters. The yellow (light gray) arrows are the spin expectation values of the eigenspinors. (c) Density of states for the outer (o) and inner (i) constant energy contours. (d) Hole Fermi seas (gray regions) and Fermi surfaces (thick lines) when $E_0 > E_F$ and $E_0 > E_F > E_\bar{\Gamma}$.
while the RB term is
\[ H_{RB} = -\alpha_{RB} \left( i\sigma_y \frac{\partial}{\partial x} - i\sigma_x \frac{\partial}{\partial y} \right). \tag{1c} \]

The positive coupling constant \( \alpha_{RB} \) reflects the RB coupling. The unit \( 2 \times 2 \) matrix is denoted by \( \sigma_i \), while \( \sigma_x \) and \( \sigma_y \) are the standard Pauli matrices in the basis in which the quantization axis is along the \( z \) direction. The eigenenergies of \( H \) yield the upper (+) and lower (−) RB branches
\[ E_{\pm}(k) = E_{\bar{\Gamma}} + \frac{\hbar^2 |k|^2}{2m^*} \pm \alpha_{RB} |k| , \tag{2a} \]
with the corresponding eigenspinors
\[ \langle \mathbf{k}, \pm | = \left( e^{i(\varphi \pm \pi/2)} , 1 \right) / \sqrt{2} . \tag{2b} \]

where \( \varphi = \arctan(k_y/k_x) \) and the two-dimensional momentum \( \mathbf{k} \) is measured relative to the \( \bar{\Gamma} \) point. Although \( H_{RB} \) breaks the spin-rotation symmetry of \( H_0 \), it preserves time-reversal symmetry. The mechanism of the enhanced spin splitting in the \( \text{Bi/Pb}_{1-x}/\text{Ag}(111) \) surface alloy goes beyond this simple model. Nevertheless, many of the fundamental properties of this system are captured by the simple nearly free electron RB (NFERB) model described above.

We plot in Fig. 1(b) the dispersion of the NFERB model. The dispersion along any cut passing through the \( \bar{\Gamma} \) point can be assigned two distinct colors that distinguish the anti-parallel alignments of the spin expectation values for the eigenspinors. This gives two spin-split bands colored in blue and red in Fig. 1(b). They are offset by two opposite wave vectors of magnitude \( k_0 \) when measured from \( \bar{\Gamma} \). The Rashba energy \( E_R = \frac{\hbar^2 k_0^2}{2m^*} \) characterizes the strength of the RB effect. The spin polarization vectors \( S_{\pm}(\mathbf{k}) \), defined as the spin expectation values of the eigenspinors \( |\mathbf{k}, \pm \rangle \), are parallel to the basal plane of Fig. 1 and are orthogonal to \( \mathbf{k} \), as depicted by the yellow arrows in Fig. 1(b). Below \( E_F \), the spin polarization rotates counterclockwise alongside the outer constant energy contour and clockwise for the inner contour. Above \( E_F \), the spin polarization rotates counterclockwise along both contours. The experimentally determined spin polarization vectors will be denoted by \( \mathbf{P} = (P_x, P_y, P_z) \) and will be shown to obey this simple rule.

The density of states (DOS) \( \nu(E_F) \) of the NFERB Hamiltonian is also sensitive to the change in the geometry and topology of the Fermi surface upon tuning of the Fermi energy \( E_F \). The DOS \( \nu_{i,o}(E_F) \) of the outer (o) and the inner (i) Fermi contour shown in Fig. 1(c) are given by
\[ \nu_{i,o}(E_F) = \Theta(E_0 - E_F) \nu_{2D} \left| \frac{E_0 - E_F}{E_0 - E_F} \right| , \tag{3} \]

whereby \( \Theta \) is the Heaviside function and \( \nu_{2D} = \left| m^* / (2\pi\hbar^2) \right| \). The + refers to the outer Fermi contour, the − to the inner one. The sum \( \nu_{i,o}(E_F) \) reduces to the constant DOS \( 2\nu_{2D} \) of a spin degenerate two-dimensional hole gas with parabolic dispersion when \( E_F > E_F \), has a singular derivative when \( E_F = E_F \), while it displays the one-dimensional Van Hove singularity \( \nu(E_F) \sim (E_0 - E_F)^{-1/2} \) in the limit \( E_F \rightarrow E_F \).

The \( \text{Bi/Pb}_{1-x}/\text{Ag}(111) \) sample preparation was carried out \emph{in situ} under ultrahigh vacuum conditions with a base pressure better than \( 2 \times 10^{-10} \) mbar. The \( \text{Ag}(111) \) crystal was cleaned by multiple cycles of Ar+ sputtering and annealing. Bi and Pb were simultaneously deposited from a calibrated evaporator at a pressure below \( 4 \times 10^{-10} \) mbar, with the total amount corresponding to 1/3 of a mono-layer. The sample quality was affirmed by low-energy electron diffraction, which showed sharp (\( \sqrt{3} \times \sqrt{3} \))R30° spots and no further superstructure, and angle-resolved photoemission spectroscopy (ARPES), which showed a continuous tuning of the band structure and no superposition of the Bi/Ag(111) and the Pb/Ag(111) band structures. These are both strong indications that, although the surface is well ordered, Bi and Pb are randomly substituted.

The experiments were performed at room temperature at the Surface and Interface Spectroscopy beamline at the Swiss Light Source of the Paul Scherrer Institute using the COPHEE spectrometer [20]. The data were obtained using horizontally polarized light with a photon energy of 24 eV. Because of the inherently low efficiency of Mott detectors the energy and angular resolution were sacrificed in the spin-resolved measurements up to 80 meV and 1.5 degree, respectively. The coordinate system for the measurements is such that a momentum distribution curve (MDC) is taken along the \( k_x \)-axis. This means that a spin polarization vector \( \mathbf{P} \) is expected to point in the \( \pm \) direction if the NFERB model holds qualitatively.

A detailed description of the band structure of Bi/Ag(111) and Pb/Ag(111) can be found in Refs. [10, 11, 12, 13]. There are two Kramer’s pairs K1 and K2 of bands that are qualitatively drawn in Fig. 1(a). The inner one (K1) is mostly of \( sp_z \) symmetry. The outer one (K2) is mostly of \( p_{x,y} \) symmetry. For Pb/Ag(111) (\( x = 0 \)), both K1 and K2 are only partially occupied. For Bi/Ag(111) (\( x = 1 \)), K1 is fully occupied, while K2 is only partially occupied. Irrespective of \( x = 0 \) or \( x = 1 \), the spin polarization vectors for K1 are nearly parallel to the surface plane and are approximately perpendicular to the momenta, in agreement with the NFERB model. In contrast, the spin polarization vectors for K2 feature significant out-of-plane components depending on the crystallographic direction. This is a consequence of the stronger coupling to in-plane potential gradients [10–12]. We will only consider K1 from now on.

Fig. 2 shows the experimental band structure of the \( \text{Bi/Pb}_{1-x}/\text{Ag}(111) \) surface alloys along \( \bar{\Gamma}K \) for \( x = (0.5), (0.6) \) and (1) measured with (spin integrated) ARPES. Second derivative data are also shown to enhance the contrast. The band K1 is fully occupied for \( x = 1 \) and, as \( x \) is decreased, the Fermi level shifts down.
We show in Fig. 3 the experimental spin-resolved MDCs for $x = 0.5$ (left column) and $x = 0.6$ (right column) providing us with the FSST for $E_F > E_F$ and $E_F > E_F$, respectively. The extraction of the spin polarization vectors $\mathbf{P}$ was done by applying a two-step fitting routine [12] on the data of Fig. 3. For both compositions, we find that the surface states $K_1$ remain fully spin polarized with spin polarization vectors similar to those of the surface states of Bi/Ag(111) or Pb/Ag(111) found in [12]. The spin polarization vectors lie mainly in the surface plane perpendicular to $\mathbf{k}$ and both the out-of-plane and radial spin polarization components are comparatively small. This finding is corroborated by several similar measurements in different crystallographic directions and at different binding energies. We thus conclude that the spin polarization of the surface states $K_1$ is robust against the mixing of Bi and Pb.

For $x = 0.5$, the measurement is performed slightly below the crossing point of $K_1$. We observe the conventional situation, i.e., a straight cut from $\bar{\Gamma}$ to the surface Brillouin zone (SBZ) boundary crosses two bands with opposite spin polarization vectors. This can be seen in the spin-resolved spectra of Fig. 3(c), which are obtained from the fits of the corresponding spin polarization data shown in Fig. 3(c). The spin polarization vectors of the bands are opposite for all adjacent bands. The corresponding qualitative FSST are drawn in the inset of Fig. 3(c).

For $x = 0.6$, an unconventional FSST is observed. Fitting the spin polarization data of Fig. 3(d) clearly shows that, for positive and negative $k_x$, both bands crossing the Fermi energy have nearly parallel spin polarization vectors. The corresponding spin-resolved spectra are displayed in Fig. 3(b). Due to strong transition matrix element effects, the inner band on the left side of normal emission is only visible as a weak shoulder of the $I_{y,dn}$ curve. When $E_F > E_F > E_F$, the FSST match qualitatively those shown in the inset of Fig. 3(d). A cut from $\bar{\Gamma}$ to the SBZ boundary crosses two bands with parallel spin polarization vectors.

We have thus established that varying $x$ between 0.5 and 0.6 induces a topological transition in the shape of the Fermi surface of $K_1$ surface states with an impact on their spin texture and on their DOS that is qualitatively captured by the NFERB model. Intuitively, one could expect a spin filtering effect due to unconventional FSST, since states with parallel $k$-vectors possess identical spin polarization vectors. However, it is the group velocity which determines electronic transport and this remains the same for anti-parallel spin directions. We will now argue that the transport of spins across an ideal onedimensional boundary separating a spin-degenerate two-dimensional electron gas from a RB hole gas is sensitive to this topological transition.

In principle, a two-dimensional scattering geometry, as depicted in Fig. 4(a), could be realized by the deposition of Bi$_x$Pb$_{1-x}$ on Ag(111) through a shadow mask. We denote with $x$ and $y$ the coordinates of the two-
FIG. 4: (color online) (a) Incoming and outgoing surface plane waves from the interface at \( x = 0 \) between a spin isotropic \((x < 0)\) and a RB \((x > 0)\) metal. (b) Dispersions of the Ag(111) and RB surface states. (c) \( P_{Ag\rightarrow x} \) as a function of \( E_F - E_{\Gamma} \) as defined in the text. (d) \( P_{Ag\rightarrow x} \) as a function of \( E_F - E_{\Gamma} \) as defined in the text.

dimensional Ag(111) surface. A spin-degenerate electron gas with an effective mass corresponding to that of Ag(111) surface states meets the states from the K1 band of Bi\(_x\)Pb\(_{1-x}\)/Ag(111) at the ideal one-dimensional boundary \( x = 0 \). We imagine driving a small charge current through the boundary by applying an infinitesimal voltage difference across the interface. The polarity of this applied voltage defines whether the charge current is from the left to the right, i.e., from the Ag(111) to the RB side, or from the right to the left, i.e., from the RB to the Ag(111) side. In the Drude limit, the charge current can be calculated from the reflection coefficients \( R_x \) for an incoming surface state of energy \( E_F \) with spin quantum number \( \sigma \) along some quantization axis, which is here chosen to be the \( y \)-axis.

We denote the spin current by \( P_{Ag\rightarrow x} \) when the current is from the Ag(111) to the RB side or by \( P_{Ag\rightarrow x} \) otherwise. To quantify the transport of spin across the boundary, we divide, on the Ag(111) side, the spin current normal to the boundary (the difference between the spin up and spin down current) by the particle current normal to the boundary, i.e., \( P_{Ag\rightarrow x} = (j_{up} - j_{down})/j_{tot} \). We use the parameters \( m^*_A/m_e = 0.397, E_{F,Ag} = -63\text{ meV} \) on the Ag(111) side \([21]\) and \( m^*_e/m_e = -0.25, E_{0x} = 94\text{ meV}, E_{F,x} = 0 \), on the RB side \([15]\). The Ag(111) and RB dispersions are shown in Fig. 4(b).

We plot in Fig. 4(c) \( P_{Ag\rightarrow x} \), and in Fig. 4(d) \( P_{Ag\rightarrow x} \) for different band fillings as described by the value of \( E_F \) (see Ref. [22] for computational details). In the absence of RB coupling, the spin current across the boundary vanishes. The breaking of the spin-rotation symmetry by the RB coupling induces a spin current on the Ag(111) side in Figs. 4(c) and 4(d). This induced spin current is strongly enhanced by the onset of an unconventional FSST when the Fermi level triggers a topological transition of the RB Fermi surface and \( \nu_i \) vanishes. Thus, the RB metal acts as a spin injector or a spin acceptor depending on the polarity of the applied voltage difference across the boundary. Finally, even for non-ideal systems, such spin currents might lead to local spin accumulation that could be detected with magnetic STM.

To conclude, we have shown that substitutional alloying does not alter the spin polarization vectors of the mixed Bi\(_x\)Pb\(_{1-x}\)/Ag(111) surface alloys. Furthermore, unconventional Fermi surface spin textures were realized through an adequate choice of the composition and were measured. Systems with strong RB type spin-orbit splitting and \( E_F \approx E_{\Gamma} \) are suggested to function as a spin filter. One could also envisage using materials with similar properties as spin injectors for a “classical” RB system. This could reduce the problems encountered at interfaces to ferromagnets.

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