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Abrupt Rotation of the Rashba Spin to the Direction Perpendicular to the Surface

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The polarization vector of the Rashba spin, which must be parallel to the two-dimensional (2D) plane in an *ideal* system, is found to change abruptly and definitely to the direction perpendicular to the surface at the $\bar{K}$ point of the Brillouin zone of a *real* hexagonal system, the Tl/Si(111)-(1 × 1) surface. This finding obtained experimentally by angle-resolved and spin-resolved photoemission measurements is fully confirmed by a first-principles theoretical calculation. We found that the abrupt rotation of the Rashba spin is simply understood by the 2D symmetry of the hexagonal structure.

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The decrease in size of a crystal to nanometer scale leads to a reduction from a three-dimensional periodical structure to a lower-dimensional one that is often accompanied with the appearance of novel exotic solid-state properties [1–3]. In the case of a reduction into a two-dimensional (2D) system, a spin-polarized 2D electron gas (2DEG) that originates from the so-called Rashba-Bychkov (RB) (or simply Rashba) effect [4] is formed even for nonmagnetic materials. Generally, the electronic states of opposite spin orientation are considered to be degenerate in nonmagnetic materials, i.e., the Kramers degeneracy, due to the presence of both time-reversal and space-inversion symmetries. However, this degeneracy will be lifted by the spin-orbit interaction (SOI) when the latter symmetry is broken due to structural inversion asymmetry in a 2D system such as a crystal surface, and the SOI leads to a pair of split bands in the momentum ($\vec{k}$) space. In the case of an *ideal* 2DEG in the (x, y) plane, the split originates from the RB Hamiltonian

$$H_{\mathrm{RB}} = \alpha_R(|\epsilon|)\vec{\sigma} \cdot (\vec{k}_{\parallel} \times \hat{e}_z), \tag{1}$$

where $\vec{k}_{\parallel} = (k_x, k_y, 0)$ is the in-plane momentum of a surface electron ($k_{\parallel} = \sqrt{k_x^2 + k_y^2}$), $\hat{e}_z = (0, 0, 1)$, α_R is the Rashba parameter ($\alpha_R = \hbar^2 k_0/m^*$; m^* is the effective mass and k_0 is the offset by which the $E(\vec{k}_{\parallel})$ parabola is shifted away from the $\bar{\Gamma}$ point), and ϵ is an electric field determined by the potential gradient normal to the surface. The split band is a completely polarized electronic state with the "spin polarization vector ($\vec{P}$) in the surface plane", and the directions of the $\vec{P}$ of the two bands are opposite, i.e., $\vec{P}(\vec{k}_{\parallel}) = -\vec{P}(-\vec{k}_{\parallel})$.

This spin-splitting effect, which has been observed on clean surfaces of noble metals [5–10] and heavy group V elements [11–13], has recently been reported to be enhanced in systems in which heavy element atoms are adsorbed on light element substrates, such as Bi or Pb on a Ag(111) surface [14,15]. The spin-split bands observed in these studies show similar behaviors as those of the simple RB effect, i.e., they show pairs of split bands in the $\vec{k}$ space and the $\vec{P}$ almost lies in the surface plane. Up to now, all measurements on spin-splitting have been performed on metal and semimetal surfaces only, though the same phenomenon on semiconductor surfaces has a technological importance in spintronic devices like spin transistors [16,17]. A surface of a light element, such as Si, with adsorbed Tl as the heavy element is a prime candidate for a semiconducting system with enhanced RB splitting. The adsorption of one monolayer (ML) of Tl on Si(111) leads to the (1 × 1) structure shown in Fig. 1(a) [18,19].

In this Letter, we report the properties of the RB spin in a *real* 2D system, the Tl/Si(111)-(1 × 1) surface. We found that the surface-state band shows a "curious splitting" at

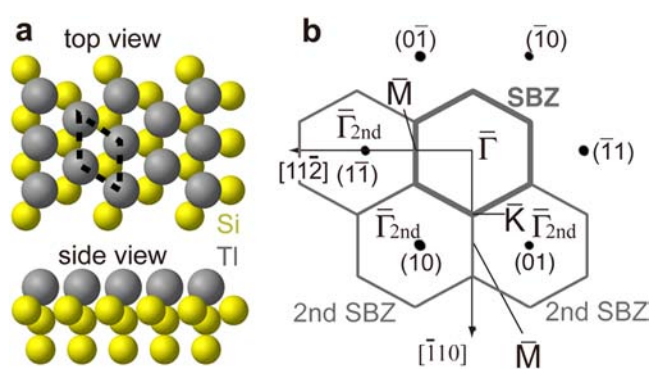

FIG. 1 (color online). (a) Schematic illustrations of the Si(111)-(1 × 1) surface formed by the adsorption of 1 ML of Tl. The thick dashed lines indicate the unit cell of this surface. (b) SBZs and LEED pattern of the Tl/Si(111)-(1 × 1) surface. $\bar{\Gamma}$, $\bar{M}$, and $\bar{K}$ are the symmetry points of the (1 × 1) SBZ.

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the $\bar{K}$ point of the surface Brillouin zone (SBZ), and that the $\vec{P}$s of these spin-split bands point completely along the direction perpendicular to the surface at this symmetry point. These novel quantum phenomena, which have not been observed so far, are corroborated by the combination of high-resolution angle-resolved photoelectron spectroscopy (ARPES) and spin-resolved (SR)-ARPES measurements, and a state-of-the-art theoretical calculation. The abrupt rotation of the RB spin, which is simply understood by the 2D symmetry of the system, will not only add a new concept in spintronics, but also provide a deeper understanding to the physical properties of 2D materials, such as graphene. (Massless Dirac fermions have been observed with the Dirac point at the $\bar{K}$ point [2,20,21] and the magnetic properties of graphene originating from the RB effect are reported to affect the electronic structure at $\bar{K}$ [22,23].)

The ARPES measurements have been performed at beamline 33 of MAX-lab, Lund, Sweden, using synchrotron radiation light with a photon energy of 21.2 eV, and the SR-ARPES measurements have been performed in a spin-resolved photoemission system of the Hiroshima Synchrotron Radiation Center, Hiroshima University, Japan, using He I radiation (21.22 eV) and a Mott detector operating at 25 keV. The energy resolution was approximately 50 meV in the ARPES measurements and 200 meV in the SR-ARPES ones, and the momentum resolutions were 0.06 Å^{-1} and 0.02 Å^{-1} at the $\bar{\Gamma}$ and $\bar{K}$ point, respectively, in both measurements. Tl was deposited on a Si(111)-(7 × 7) clean surface, which was obtained by annealing at 1520 K, from a Knudsen cell at a substrate temperature of 570 K. The base pressure was below 4×10^{-11} Torr during the measurements, and below 1×10^{-10} Torr during the Tl evaporation. The effective Sherman function in the SR-ARPES measurement, 0.128 ± 0.05, was obtained by a self-calibration method in the maximum figure of merit (1.8×10^{-4}) [24].

A first-principles electronic structure calculation based on the Kohn-Sham theory [25], which employs ultrasoft pseudopotentials [26] and plane waves, was used in the theoretical study. This calculation has taken into account the fully relativistic effect that includes the SOI [27] and the self-consistent treatment [28] for two-component-spinor wave functions [29]. We used the generalized gradient approximation [30], the energy cutoffs of 25 and 300 Ry for wave functions and densities [31], and a repeated slab model, in which each slab contains a Tl monolayer (ML), 24 Si ML and a H ML. Each slab was separated by a vacuum space of 9.7 Å. The atomic positions except those of the H atoms and Si atoms bonded to H were fully optimized to an assumed criterion of atomic force (less than 0.01 eV/Å). In order to exclude the artificial dipole electric field imposed on the surface from the image cells in the repeated slab, the scheme of effective screening medium [32] was employed.

Figure 1(b) displays the SBZ of a Tl/Si(111)-(1 × 1) surface together with the low-energy electron diffraction (LEED) pattern of the sample shown by the black spots. The intensities of the (10), $(\bar{1}1)$, and $(0\bar{1})$ spots are the same, but the intensities of the (01), $(\bar{1}0)$, and $(1\bar{1})$ spots are weaker than those of the three other spots. This indicates that instead of the sixfold symmetry of the 2D Tl layer, this surface has actually a three-fold symmetry resulting from the quasi-2D structure that includes the first Si layer.

The electronic band dispersion measured using ARPES along the $\bar{\Gamma}$-$\bar{M}$-$\bar{\Gamma}$ direction is shown in Fig. 2(a). Of the two observed bands, only the lower binding energy (E_B) one is located in the gap of the projected bulk bands (hatched area [33]). This means that there is only one surface-state band on the Tl/Si(111)-(1 × 1) surface. Since the $6s^2$ electrons of the Tl atoms act as an inert pair and are inactive in the Tl-Si bonding, which results in a monovalent character of the Tl atoms [18,19], the observed surface-state band originates from the hybridization between the single Tl $6p$ electron and one electron from the surface Si atom. Thus, there is no orbital degeneracy in the observed surface-state band. Further, no spin-split band is observable in Fig. 2(a), indicating that the Rashba energy (E_R) of this system is small and cannot be resolved in the present ARPES study [the definition of E_R and its value obtained in our theoretical calculation described below are shown in Fig. 4(b)]. Along the $\bar{\Gamma}$-$\bar{K}$-$\bar{M}$ direction, the surface-state

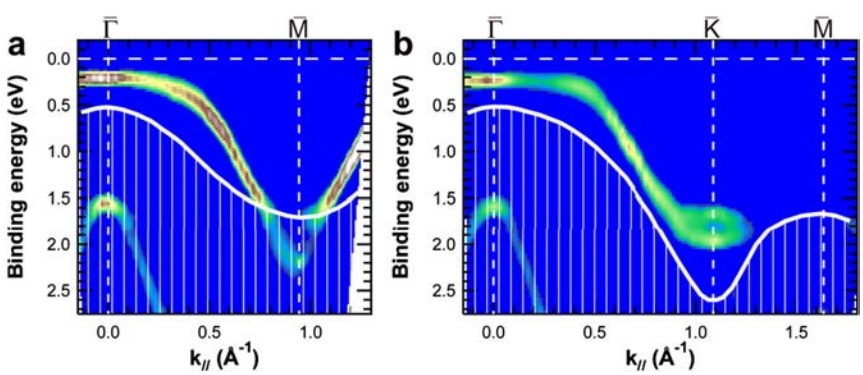

FIG. 2 (color online). Band structures measured along the (a) $\bar{\Gamma}$-$\bar{M}$-$\bar{\Gamma}$ and (b) $\bar{\Gamma}$-$\bar{K}$-$\bar{M}$ directions. The hatched areas show the bulk band projection taken from Ref. [33]. The horizontal dashed line at $E_B = 0$ eV indicates the Fermi level.

band shows a "curious splitting" around the $\bar{K}$ point that has not been observed on any other 2D or 3D system so far. Since there is no orbital degeneracy in the surface-state band, one of the most possible explanations is a spin-splitting. Note that the splitting appears along the energy axis at around $\bar{K}$ only, while if the split is due to a simple RB effect it would be observed in the whole SBZ except at high symmetry points such as $\bar{\Gamma}$.

Figure 3 shows the SR-ARPES spectra measured along the $\bar{\Gamma}$-$\bar{K}$-$\bar{M}$ direction with two different experimental setups that allow us to obtain separate spectra for different $\vec{P}$ in order to confirm the spin states of the electrons. In Fig. 3(a), the spin states with $\vec{P}$ that are parallel to the surface and perpendicular to the $\bar{\Gamma}$-$\bar{K}$ direction are degenerate at an emission angle (θ_e) of 0°, which corresponds to the $\bar{\Gamma}$ point. However, although this degeneracy agrees well with the ARPES results shown in Fig. 2, spin-split bands with in-plane spin polarization are observed at higher θ_e. That is, the peak positions of the red and blue spectra are the same at $\theta_e = 0°$, but their E_B become different at $\theta_e = 10°$. This means that SR-ARPES further resolves the spin of the "single" band observed in ARPES. The small intensity of the higher E_B peak (the red one for positive θ_e and blue one for negative θ_e), which results from a final state effect for relativistic electrons [34], might hide the split feature in the spin-integrated ARPES spectra of Fig. 2.

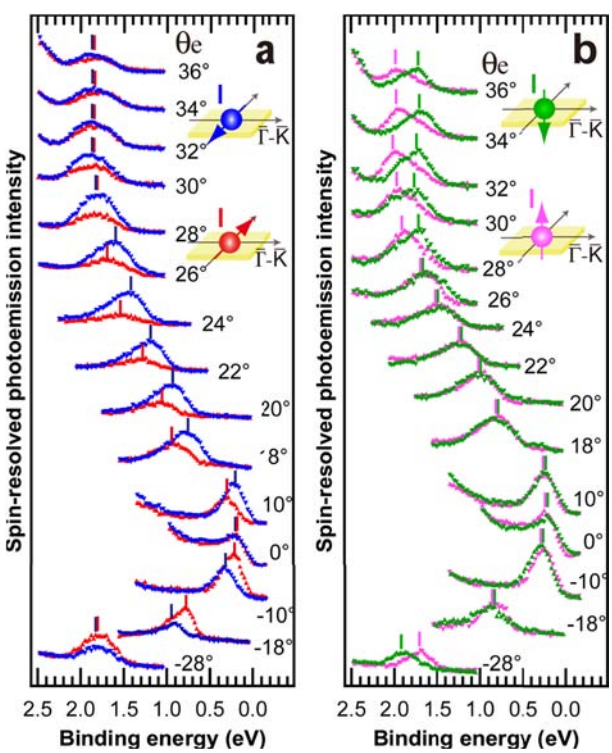

FIG. 3 (color online). (a) SR-ARPES spectra of the electrons with $\vec{P}$ parallel to the surface and perpendicular to the $\bar{\Gamma}$-$\bar{K}$ direction, and (b) spectra of the electrons with $\vec{P}$ perpendicular to the surface. $\theta_e = 0°$ corresponds to $\bar{\Gamma}$, and $\bar{K}$ is between $\theta_e = 32°$ and 34°. The insets indicate the $\vec{P}$ of the detected electrons.

The spin-split bands are observable till $\theta_e = 26°$, but at $\theta_e = 28°$ and higher θ_e, the difference in E_B of the two bands becomes negligible.

The $\vec{P}$ perpendicular to the surface shows a completely different behavior. That is, the two spectra do not show any spin-splitting till $\theta_e = 26°$, but a clear split is observable at higher θ_e. These results indicate that the $\vec{P}$ of the surface-state band, which lies in the surface, "*rotates abruptly*" and points along the direction perpendicular to the surface around the $\bar{K}$ point ($\theta_e \sim 34°$). Note that in contrast to the 10% spin polarization aligned perpendicular to the surface plane, which results from the slight continuous rotation of $\vec{P}$ in the direction out from the surface plane on Bi/Ag(111) and Pb/Ag(111) surfaces [14,15], the spin polarization is 100% along the direction perpendicular to the surface around the $\bar{K}$ point only in the present case. In order to understand the origins of the abrupt rotation of the RB spin and the curious splitting around $\bar{K}$, that cannot be explained by the simple RB effect described in Eq. (1), we first discuss the RB effect in a real 2D system. Using Bloch wave functions $\varphi_{n\vec{k}}(\vec{r}) = (1/\sqrt{\Omega})\exp(i\vec{k}\cdot\vec{r})u_{n\vec{k}}(\vec{r})$, the effective SOI Hamiltonian of the "extended RB effect" can be described as

$$H_{\mathrm{SOI}}(\vec{k}) = \vec{\sigma}\cdot[\vec{\alpha}_n(\vec{k})\times\vec{k}] + \vec{\sigma}\cdot\vec{B}_n(\vec{k}), \qquad (2)$$

where $\vec{\alpha}_n = (\hbar^2 N/4m_e^2c^2\Omega)\int_{\mathrm{cell}} d\vec{r}|u_{n\vec{k}}(\vec{r})|^2\vec{\nabla}V(\vec{r})$ and $\vec{B}_n(\vec{k}) \approx (\hbar^2 N/4m_e^2c^2\Omega)\int_{\mathrm{cell}} d\vec{r}(1/r)(dV(\vec{r})/dr)u^*_{n\vec{k}}(\vec{r})\times(\vec{\ell})u_{n\vec{k}}(\vec{r})$. The second term of Eq. (2), which is not considered in the case of an ideal 2D system, is a Zeeman term that acts as an effective magnetic field to the band spin magnetization. The field $\vec{B}_n(\vec{k})$ is obtained as an averaged value on the angular momentum operator ($\vec{\ell}$), and is mainly caused by the atomic character of the wave function. When the wave function $u_{n\vec{k}}(\vec{r})$ is a good quantum state of the angular momentum operator, $\vec{B}_n(\vec{k})$ can have a large finite value and the electron spin tends to become parallel to the direction of $\vec{B}_n(\vec{k})$.

Figures 4(a) and 4(b) display the calculated spin-split band structure together with the experimental results. The good agreement between the dispersions of the calculated bands and the experimental ones corroborates the origin of the splitting at $\bar{K}$ to be spin-splitting. Further, the calculated band indicates that k_0 and E_R are 0.2 Å^{-1} and 20 meV, respectively. The direction of $\vec{P}$ of one of the split bands, as derived from the normal RB effect [Eq. (1) and the first term of Eq. (2)], is illustrated in Fig. 4(c). As shown in this figure, $\vec{P}$ rotates anticlockwise around $\bar{\Gamma}$ and clockwise around $\bar{M}$, whereas at $\bar{K}$, the rotations of the $\vec{P}$ along $\bar{\Gamma}$-$\bar{K}$ and those along $\bar{M}$-$\bar{K}$ are opposite. This opposite rotation implies that $\vec{P}$ of the normal RB spin is indeterminable at the $\bar{K}$ point. Indeed, the first term of Eq. (2) is found to vanish at $\bar{K}$ owing to its C_3 symmetry in the theoretical study. This means that only the second term contributes to the spin polarization at $\bar{K}$. The function

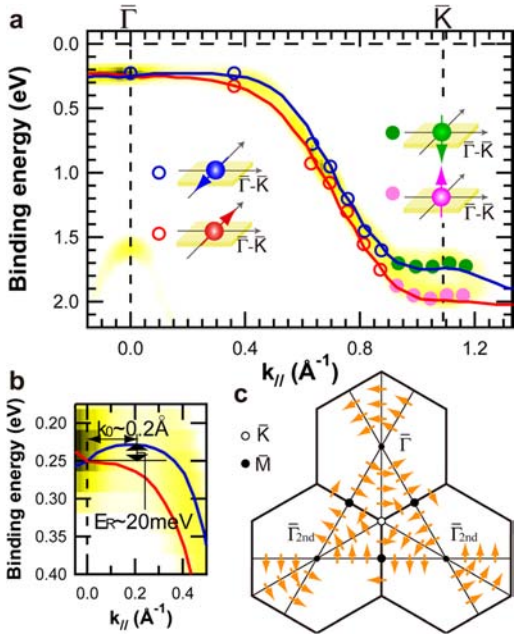

FIG. 4 (color online). (a) Spin-split surface band structures obtained experimentally (open and filled circles) and theoretically (solid lines) along the $\bar{\Gamma}$-$\bar{K}$ direction. (b) Band structures around the $\bar{\Gamma}$ point. $k_0 = 0.2$ Å^{-1} and $E_R = 20$ meV are obtained from the calculated spin-split bands. (c) The arrows show the $\vec{P}$ of one of the split bands, as derived from the normal RB effect. The alternating direction of $\vec{P}$ around $\bar{K}$ is discussed in the text.

$p_x - ip_y$ (or $p_x + ip_y$) forms an irreducible representation of the C_3 group when the SOI is neglected. When the SOI is on, the interaction lifts the spin degeneracy and, consequently, the effective field of $\vec{B}_n(\vec{k})$ becomes parallel to the z direction, leading the spin polarization of the surface-state band to be perpendicular to the surface. At the $\bar{\Gamma}$ point with C_{3v} symmetry and at the $\bar{M}$ point with C_{1h} symmetry, no spin-splitting has been observed neither experimentally nor theoretically. This is well explained by the fact that SOI cannot lift the spin degeneracy in these two symmetries.

In conclusion, our study reveals that a 2D system with hexagonal structure exhibits a novel quantum effect, the "abrupt rotation of the RB spin to the direction perpendicular to the surface", and that this effect is a general property of a reciprocal lattice point with C_3 symmetry. These results imply that the $\vec{P}$ of a real RB spin might be different from that of an ideal RB one depending on the symmetry of the sample. The present results will provide a deeper understanding on the physical properties of 2D materials, and they indicate that a proper knowledge of $\vec{P}$ and thus of the symmetry of the 2D band structure is indispensable to estimate the efficiency of the spin-current in a spintronic device. Moreover, the presence of rotated spins that are aligned with the surface normal suggests the possibility to double the functionality of spin-transistors by picking electrons with a certain wavelength or wave vector, i.e., one that corresponds to the $\bar{K}$ point.

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