# STUDY ON THE HELICAL SPIN TEXTURE OF TOPOLOGICAL SURFACE STATE IN SUPERCONDUCTOR 2M-$WS_2$

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Layered transition metal dichalcogenides (TMDs) of $MX_2$ (M = Mo, W; X = S, Se, Te) have been studied intensively because of a great interest in both fundamental and applied research. Among the stable phases of TMDs, Monolayer 1T′-$MX_2$ (M = Mo, W; X = S, Se, Te) were suggested as a promising candidate to realize quantum spin Hall (QSH) effect in strictly two-dimensional systems [1-4]. QSH insulators are expected to have two conducting edge channels with quantized electric conductance $2e^2/h$ and opposite spin polarizations, which are robustly protected by the time-reversal symmetry. It could provide a possible alternative to quantum electronic devices with low dissipation. However, because of the limitation of monolayer, the TSS of QSH insulators in TMDs have been not addressed by the experimental tools.

Recently, new stable structure phase 2M-$MX_2$ have been found with the topological surface state (TSS) and the superconductivity with the highest transition temperature 8.8 K among TMDs [5-10]. Because the crystal structure of bulk 2M phase is identical with monolayer 1T′-$MX_2$, 2M-$MX_2$ have a similar band structure and TSS that is similar dispersion with TSS of monolayer 1T′-$MX_2$. The observation of TSS in 2M-$MX_2$ can provide the information of topological feature in monolayer 1T′-$MX_2$ as well as own great interesting feature such as a possible candidate for the QSH insulator and the Majorana bound state together with the superconductivity [11].

We investigated the electronic and spin structure of the TSS in 2M-$WS_2$ utilizing high-resolution angle- and spin- resolved photoemission spectroscopy. The low-energy photoemission spectra taken from the laser-based ARPES with 6.994 eV photons clearly describe the fine dispersion of the topological surface state with the higher momentum resolution in Fig. 1(a). The hole-like band dispersion near the $k_y = 0$ is corresponding to the lower part of the Dirac cone of topological surface state, and the crossing point is expected to be located above the Fermi level. To show the helical spin structure of the topological surface state, we exploit the spin-resolved ARPES. Figure 1(b) show the spin-resolved EDCs of $s_y$ with red and blue colour code with spin up and spin down, respectively. The result indicates

that the sign of $s_y$ is reversed between $k_L$ and $k_R$. Moreover, we obtain that the in-plane spin-polarization $P_y$ is reversed between $k_L$ and $k_R$ near the Fermi level, but another spin-polarization $P_x$ and $P_z$ are negligible. It was expected that the topological surface state of 2M-$WS_2$ show the helical spin texture, thus allowing the opposite spin direction between left and right momentum at the Fermi level. In the view of the Fermi surface as the schematic picture based on our experimental results, the lower Dirac cone branch of the topological surface state in 2M-$WS_2$ has the helical spin texture with a clockwise rotation as shown in Fig. 1(d). Given the crystal and electronic structural similarity between 2M-$WS_2$ and monolayer 1T′-$MX_2$, our results can provide the general knowledge about the edge conductance and spin-based device as manufactured with quantum spin Hall insulator, monolayer 1T′-$MX_2$.

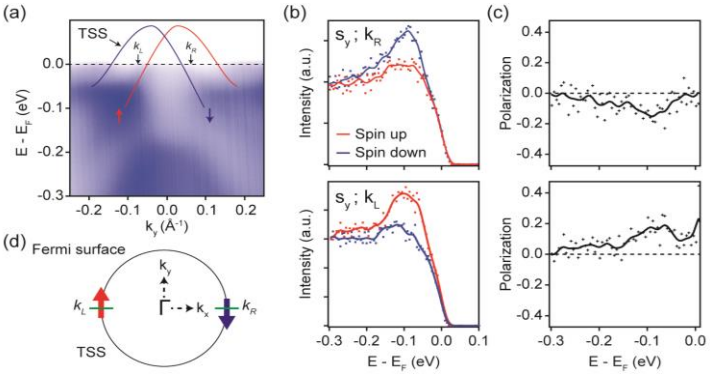


Figure 1(a) ARPES spectra of 2M-$WS_2$ with the TSS obtained from the band calculation and the possible direction of the spin momentum based on the experimental results. The red (blue) arrow indicates the spin up (down). (b) Spin-resolved EDCs of the TSS at the left and right momentum cuts as marked in (a). The spin-resolved EDCs at $k_L$ and $k_R$ shows spin texture along the $S_y$ direction. (c) Representative spin-polarization magnitude for the TSS at $k_L$ and $k_R$, whose obtained from the spin-resolved EDCs in (b). (d) Schematic picture of the helical spin texture for the TSS at Fermi surface based on the spin-resolved EDC and polarization. in (b) and (c)

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# ENHANCED RASHBA-SPLITTING OF AU(111) SURFACE STATES BY THE MELAMINE HONEYCOMB LAYERS

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In quasi-two-dimensional electron systems with broken spatial inversion symmetry, such as metal surfaces and semiconductor interfaces, Rashba-type spin-orbit interactions (Rashba effect) are known to occur through atomic spin-orbit interactions. The Rashba effect has been actually demonstrated in quantum wells and thin films based on III-V semiconductors, as well as in two-dimensional electron gases in the metal surface, and is expected to play an important role as an elemental technology in spintronics.

Many studies have shown that the Rashba coupling can be modified by the adsorption of atoms and molecules on metal surfaces and by distortions at the semiconductor interface. In recent years, it has become possible to intentionally control the strength of the Rashba coupling and design systems for various applications. For example, adsorption of noble gases or organic molecules on noble metal (111) changes the strength of the Rashba coupling[1].

Organic materials such as metal-organic frameworks (MOFs) and covalent organic frameworks have been extensively studied over the past few decades and are potentially attractive for a variety of applications, including gas storage and separation, catalysis, chemical sensors, and drug delivery[2]. On the other hand, new porous materials called hydrogen-bonded organic frameworks (HOFs), which are constructed from pure organic or metal-containing organic building blocks by hydrogen bonds, have recently attracted much attention. Although many pioneering studies have been conducted on HOFs, their progress has lagged behind that of MOFs and others. The main reason for the lag is that the structure of many HOFs collapses when solvent molecules are removed from the pores formed by weak hydrogen bonds. On the other hand, the instability of HOFs is caused by weak hydrogen bonds, but on the contrary, they are rather flexible compared to strong covalent or coordination bonds. The use of this soft hydrogen bond is also beneficial for the flexibility of the framework of porous materials. Carbon nitride molecules such as melamine (1,3,5-Triazine-2,4,6-triamine) and melem (2,5,8-triamino-heptazine) are typical supramolecular species that form HOFs through highly electronegative nitrogen-mediated hydrogen bonds. The studies in these HOFs are rather well known for cyanuric acid and melamine complexes, which have been studied for many years, where cyanuric acid and melamine are combined by N-H···O and N-H···N hydrogen bonds to form rosette-shaped superstructure[3]. On the other hand, melamine and melem are known to readily form highly oriented monolayers consisting of two-dimensional ordered supramolecular structures by adsorption on metal surfaces (surface HOF: SHOF), without the help of solvent molecules. This is probably due in large part to the flexibility of hydrogen bonding. Surfaces modified by such supramolecular structures are becoming powerful tools in applications such as chemical sensors and nanoelectronics, thanks to their ability to exhibit molecular recognition features. However, melamine is one of the most basic units in the systems that create such SHOFs, but even for the structure of the SHOFs of melamine, the determination of their structures remains arbitrary so far, although there are a few reported studies using STM[4]. Furthermore, there are no reported studies that have conducted detailed investigations of the formation process of melamine SHOFs, their electronic structure, or their interfacial electronic structure with metal surfaces. Therefore, even now, many problems remain in understanding the basic physical properties of melamine SHOFs as a prototype of SHOF.

In this study, we determine the formation process and the structure of melamine SHOFs on Au(111) by X-ray photoemission spectroscopy (XPS), low-energy electron diffraction (LEED) and their effect on the Au(111) surface electronic structure by angle-resolved photoemission spectroscopy (ARPES) and spin-resolved and angle-resolved photoemission spectroscopy (SARPES). The SHOF formed when melamine adsorbs on Au(111) is a simple but most typical system, so clarifying its formation is inevitable for understanding the formation of other SHOFs. From the LEED results shown in Figure 1, melamine, upon adsorption on Au(111), immediately forms a unique hexagonal SHOF through cooperative intermolecular hydrogen bonds. Many other organic molecules, whose main cohesive force is dispersion force, do not form such cooperative superstructures and usually involve molecular deformation upon adsorption to a surface, but in the case of melamine SHOFs, molecular deformation is minimized by multiple hydrogen bonds between the molecules. The SHOF retains its two-dimensionality, like graphene adsorbed on Au(111). Theoretical calculations also show that the SHOF induces a slight, but obvious, change in the position of nuclei in the outermost layer of Au(111) in the surface vertical direction. This indicates that there is strong orbital mixing between SHOF and Au(111), meaning that SHOF forms a new periodic potential on the surface. Such a strong coupling between the SHOF and the metal surface electron system has a significant effect on the electronic structure of the surface electron system, leading to a reconfiguration of the surface charge density and a modulation of the electrostatic potential in the outermost layers of the surface. As a result, it is shown to significantly enhance Rashba coupling in Shockley state (SS) of Au(111). The conclusions drawn here indicate that SHOF may provide new guidelines in the manifestation of the spin-galvanic effect, the Edelstein effect, and the inverse Edelstein effect, which are attracting attention as elemental technologies in spintronics.

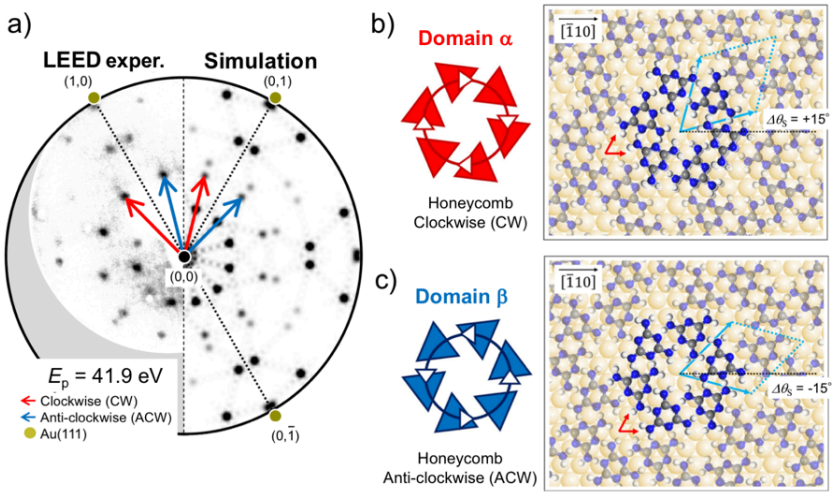


Figure 1. a) Experimental and simulated LEED results for melamine/Au(111). The left half of the circle shows the LEED pattern observed at primary energy of $E_p$ = 41.9 eV; the right half of the circle shows the simulated LEED pattern. Red and blue arrows indicate diffraction spots from two domains α and β with different pseudo-chirality, respectively. b) Schematic drawing of the adsorption structure of domains α and β of melamine/Au(111).

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# ELECTRONIC STATES OF TOPOLOGICAL SEMIMETALS STUDIED BY SPIN- AND ANGLE-RESOLVED PHOTOEMISSION SPECTROSCOPY

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## INTRODUCTION

By breaking the time reversal symmetry in Dirac semimetals, one would obtain topological Weyl semimetals or nodal line semimetals [1]. However, usually only extremely large external magnetic field can realize such topological phase transition, which is challenging in the laboratory. A layered phosphide, $EuP_3$, has been reported to archive topological phase transition when external magnetic field is applied: it would be a magnetic Weyl semimetal in perpendicular magnetic field, but nodal-line semimetal in in-plane magnetic field [2]. This material has drawn increasing attention for it can be an ideal platform to study the topological phase transition, as well as its promising application in engineering.

We have observed of the band structure in α-$EuP_3$ by angle-resolved photoemission spectroscopy (ARPES) with synchrotron lightsource. We used resonant photoemission to directly confirm the *f-p* coupling. Further, with the high-resolution ARPES, we discovered an unexpected band splitting in the valence band. This band splitting occurs only along $k_y$ direction without any global symmetry breaking. To compare with the results, we also measured an isostructural material $SrP_3$, and found similar splitting. This result suggests that the crystal structure contributes to the band splitting. We need to confirm whether this kind of band splitting is intrinsic by measure their spin texture with SARPES.

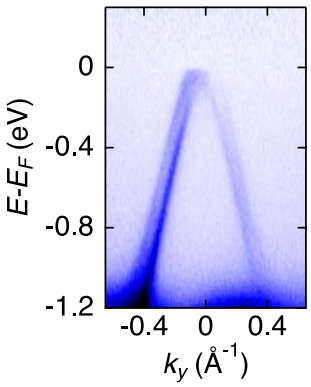


Fig.1 Band dispersion of $EuP_3$, showing an unconventional splitting.

## EXPERIMENTS

During the experiment, all $EuP_3$ and $SrP_3$ samples were cleaved *in situ* with top posts, and the *ab* plane [(001) surface] was exposed and measured. The *ab* plane is flat and shiny, suggesting that the compounds is metallic. It should be noted that there is another cleavage except (001) surface but no band dispersion can be observed from that.

The experiment was carried out with the spin-resolved laser ARPES (SARPES) at ISSP, the

University of Tokyo. The light source is 7 eV laser from KBBF crystal.

## RESULTS

We measured the in-plane spin texture of $EuP_3$ and $SrP_3$. Although due to the matrix element effect, we cannot directly observe the two splitting band dispersions, we found that the valence bands are spin-polarized. The spin textures of both materials are Dresselhaus-like, suggesting the space reversion symmetry is broken by unknown mechanism. However, we need further study to confirm whether the splitting is a combination of Dresselhaus and Zeeman effects because we have not excluded the out-of-plane spin polarization. μSR can directly demonstrate direct evidence of the spin fluctuation [3]. We also need theoretical study to explain why Dresselhaus effect occurs in the centrosymmetric $EuP_3$ and $SrP_3$ crystals.

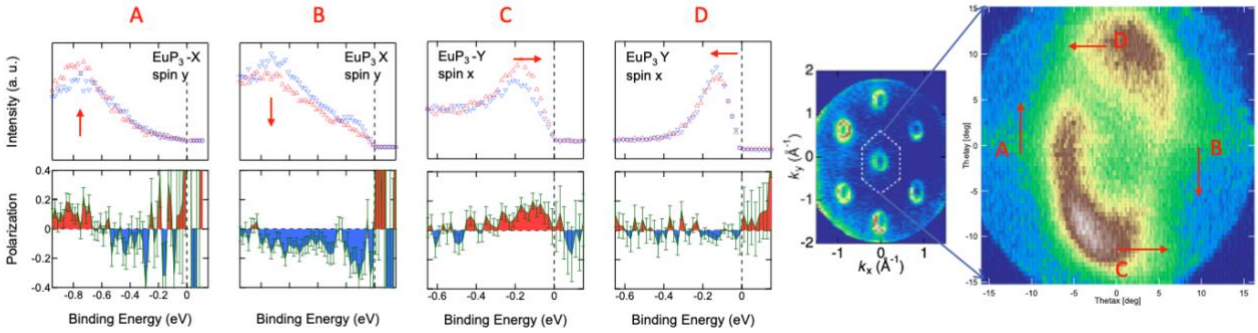


Fig. 2 Spin texture of $EuP_3$. The valence band is spin-polarized along both *x* and *y* directions, suggesting a Dresselhaus-like band splitting.

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# LASER-SARPES STUDY OF SPIN-POLARIZED ELECTRONIC STATES FOR $PB(BI_{1-X}SB_X)_2TE_4$

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The formation mechanism of a topological surface state (TSS) is well established, where the bulk band inversion between the top of the valence band and the bottom of the conduction band is required. The phase transition from a trivial insulator to a topological insulator is known as a topological phase transition. The topological phase transition can be controlled by tuning the composition, the lattice constant, and the film thickness of the system [1-4]. Understanding the modification in an electronic structure induced by topological phase transitions is essential not only for fundamental interests but also for applications.

A family of Pb-based ternary compounds, $Pb(Bi_{1-x}Sbx)_2Te_4$, has been revealed to be topological insulators [5-8]. In the previous angle-resolved photoemission spectroscopy (ARPES) studies [7,8], the TSSs above a Dirac point (DP) have been clearly observed, where the TSSs exist in a projected bulk band gap. On the other hand, the dispersive feature is unclear below DP, which might be due to the existence of the bulk electronic states in the close vicinity of the TSSs. In addition, no spin-resolved ARPES (SARPES) study has been reported so far.

In the present study, we have investigated spin-polarized electronic states of $Pb(Bi_{1-x}Sb_x)_2Te_4$ (x = 0.70, 0.79) by SARPES using a laser (laser-SARPES). Our laser-SARPES reveals Rashba-type occupied surface resonances near the TSSs. Besides, the TSSs continuously connects to a branch of the Rashba-type surface resonances. Our results provide direct evidence that the present system undergoes the topological phase transition.

Laser-ARPES and -SARPES measurements have been performed at the Institute for Solid State Physics, The University of Tokyo [9]. The samples were cleaved with scotch tape in an ultra-high vacuum chamber below the base pressure of 2.0 x $10^{-8}$ Pa. The photoelectrons were excited by a quasi-continuous-wave laser with a photon energy of 6.994 eV. The energy resolutions for laser-ARPES and -SARPES were set to 5 and 10 meV, respectively. The sample temperature was kept at 30 K during the measurements.

Figure 1 displays an ARPES intensity map for x = 0.70 taken along the $\bar{\Gamma}\bar{M}$ direction. Overall dispersive features of the TSS and the bulk conduction bands agree with the previous studies [7,8]. The precise band structure near the Dirac point has been demonstrated thanks to the high-resolution laser-ARPES. The Dirac

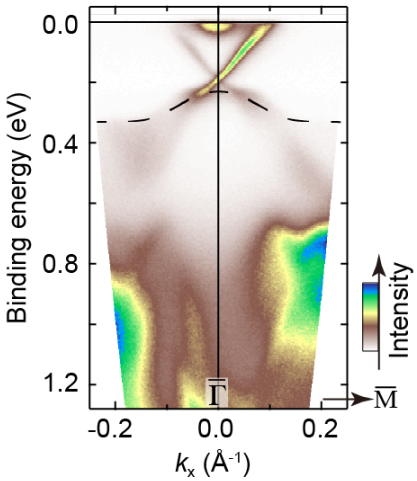


Fig. 1 ARPES intensity map for x = 0.70 along $\bar{\Gamma}\bar{M}$. A dashed curve represents the projected bulk band edge determined by a leading edge of the corresponding photoelectron intensity.

point (DP) is observed at the binding energy of 190 meV. The photoelectron intensity from the TSS in the bulk band gap is prominent. The TSS penetrates into the projected bulk bands at $k$ = ±0.05 Å$^{-1}$, indicating that the surface states change to surface resonances. The surface resonances disperse in the binding energy range of 0.2-0.5 eV. One can notice here that the spectrum widths of the surface resonances are broadened compared with those of the TSSs. We also find several bands below $E_B \sim 0.7$ eV.

Laser-SARPES measurements have been performed for x = 0.70 and x = 0.79 samples [Fig. 2(a,b)] in the $\bar{\Gamma}\bar{M}$ direction with a p-polarized light. The dispersive feature for x=0.79 is quite similar to that for x = 0.70. Here, the DP for x=0.79 is located above $E_F$. The observed spin direction is inverted with respect to the $\bar{\Gamma}$ point, meaning the TSSs possess a helical spin texture. More interestingly, spin components opposite to the TSS are observed below DP, corresponding to the spin textures of the surface resonances.

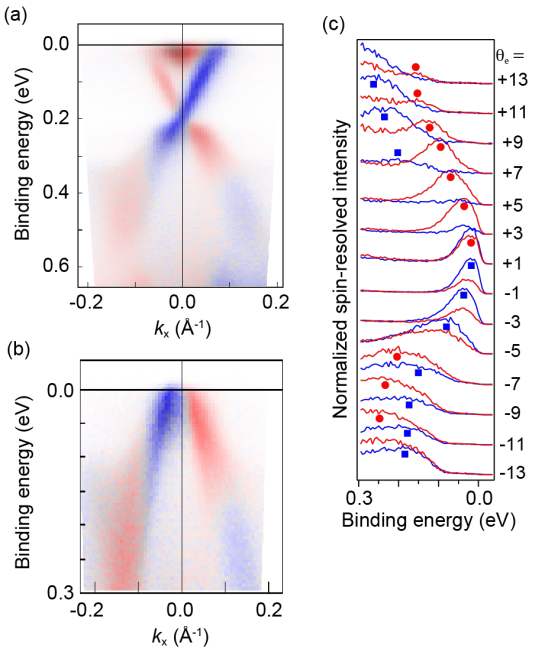


Fig. 2 SARPES images for (a) x = 0.70 and (b) x = 0.79 along $\bar{\Gamma}\bar{M}$. (c) Spin- and angle-resolved spectra for x = 0.79.

The band structures of the TSS and the surface resonances are identified from spin-resolved energy distribution curves [Fig. 2(c)]. The surface resonances are spilt into two in the energy direction in |k| = 0.1–0.2 Å$^{-1}$. The surface resonance that appears in the lower binding energy side possesses the same spin orientation as the TSS. Another surface resonance dispersing in the energy range of 0.2–0.3 eV possesses the spin direction opposite to that of the counterpart.

In addition, the spin polarizations of the surface resonances are inverted with respect to the $\bar{\Gamma}$ point. Thus, we conclude that the surface resonances exhibit the Rashba-type spin splitting. More importantly, the upper branch of the surface resonances is continuously connected to the TSS. Our result provides direct evidence that the system undergoes the topological phase transition.

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# ELECTRONIC STRUCTURES OF HALF-METALLIC FERROMAGNET $La_{1-x}Sr_xMnO_3$ BY HIGH-RESOLUTION SPIN-RESOLVED PHOTOEMISSION SPECTROSCOPY

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Half metallic ferromagnets have a unique spin dependent electronic structure near the Fermi level ($E_F$), where only one of the spin states cross $E_F$ [1]. The unique electronic structure gives rise to many body effects different from other systems. Theoretically, the electronic states induced by many-body effects in half metals has been proposed as non quasiparticle (NQP) states [2]. The experimental verification for NQP states has been made recently for a half metallic ferromagnet $CrO_2$ by using high-resolution (HR) spin-resolved photoemission spectroscopy (SRPES) [3]. In the present research, we have performed HRSRPES of another half metallic ferromagnet $La_{1-x}Sr_xMnO_3$ to experimentally explore the characteristics and universality of NQP states. $La_{1-x}Sr_xMnO_3$ is one of the famous perovskite materials, exhibiting colossal magnet resistance around Sr concentration x of 0.3. The previous SRPES study has provided evidence for half metallic electronic structure of $La_{1-x}Sr_xMnO_3$ at 40 K[4]

Homoepitaxial films of $La_{1-x}Sr_xMnO_3$ were grown on the atomically-flat (001) surface of Nb-doped $SrTiO_3$ substrates using a laser MBE method. Laser-based SRPES experiments were performed at the Institute for Solid State Physics, The University of Tokyo [7]. The *p*-polarized light with $h\nu = 6.994$ eV was used to excite the photoelectrons. Photoelectrons were analyzed with a combination of a ScientaOmicron DA30L analyzer and a very-low-energy-electron-diffraction (VLEED) type spin detector. The energy and angular resolutions were set to 6 meV and 0.3˚ (corresponding to ~0.005 Å) for spin-integrated ARPES and 10 meV and 1˚ (corresponding to ~0.02 Å) for SARPES, respectively. During the measurement, the instrumental energy resolution was set to 20 meV and the base pressure was kept below $1 \times 10^{-8}$ Pa. Calibration of $E_F$ for the samples was achieved using a gold reference. The data were taken at $T = 20$, 50, 100 and 150 K. Clean surfaces of the samples for all measurements were obtained by *in situ* annealing at 200℃ for 20 minutes. We magnetized the LSMO(001) sample along the

magnetic easy axis ([110] direction) by bringing the sample close to a magnet . The approximate magnitude of the magnetic field at the sample position was 700 Oe.

We obtained HRSRPES data at 20, 50, 100, and 150K. Compared to the minority spin states showing negligible intensity around the near-$E_F$ region, the minority spin states have enhanced intensity with a Fermi edge at all measured temperatures. These results are in line with the previous SRPES studies reporting that LSMO is a half metallic ferromagnet. However, we found that polarization is slightly smaller than 100%. We tentatively attribute the reduced polarization to detecting the information of the substrate. We plan to measure HRARPES with thicker films in the future experiment.

## ACKNOWLEDGEMENT

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