## Demonstration of phase-resolved spin-ARPES on topological surface states in Bi<sub>2</sub>Te<sub>3</sub>

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Optical transition such as photoemission takes place in attosecond scale (1 as  $= 10^{-18}$  s). It is very interesting to phenomena in the attosecond region from point of view of the pursuit of ultimate understanding of quantum mechanics and pioneering electronics that can be controlled on as-time-scale. Great development of ultrashort pulse laser technology over the past few decades has permitted to track the ultrafast dynamics as the attosecond scale in the time domain [1-4]. However, the ultrashort pulse nature of the attosecond pulse diminished energy resolution because of a trade-off relationship between the pulse width in time and frequency domains. This means that it is difficult to experimentally trace the ultrafast photoemission dynamics together with resolving valance band structures.

In this work, to overcome the above difficulty, we attempted phase-resolved spin-ARPES [5-7] by laserbased angle-resolved photoelectron spectroscopy with 3D spin detection. Using this technique, we accessed phase information in the transition matrix of the photoelectron states and its kinetic energy dependence that can be converted to the time scale of photoemission dynamics in as-time-scale [8]. For this measurement, a picosecond laser was used to achieve a 20-meV energy resolution for spin-ARPES and 3D spin detection was used for the delay phase determination[9]. In addition, we built a polarization rotation control system that allows us to perform the automatic spin-ARPES measurements collecting a number of data points.

We studied the spin polarization of the photoelectrons emitted from the topological surface state of the typical topological insulator  $Bi_2Te_3$ . The topological surface state is ideal for this experiment because they form fully spin-polarized electronic states that are locked to be a helical texture in momentum space. Let us also note that the spin-polarization of the photoelectron is collected on the mirror plane [Fig.1(a)], and therefore the orbital selection rule is allowed in the photoemission process. Owing to these simple experimental geometry, one can selectivity excite either *even* or *odd* orbital function that is coupled to different spin wavefunction [10] and superimposes these two dipole matrixes with the different spin in photoelectron final state, resulting in the rotation of the spin polarization axis [5-7]. This process is schematically summarized in Fig1.(b).

Though this process, the spin-orbit coupled wavefunction in the topological surface states allows us to selectively excite the spin and control its direction by photon polarization. Furthermore, the spin direction as a function of the photon polarization sensitivity depends on the relative phase between the two matrix elements, which therefore allows us to extract the phase from our spin-ARPES data. The extracted relative phase  $\phi$  between two transition is used for time conversion according to the Eisenbud-Wigner-Smith (EWS) model [1,11]:

$$\tau_{EWS} = \hbar \frac{\partial \phi}{\partial E_k} \tag{1}$$

where  $\tau_{EWS}$  is a time delay obtained in the EWS-model, and  $E_k$  is kinetic energy of the photoelectron.

Fig2.(a) represents a Fermi surface and ARPES of the topological surface states in Bi<sub>2</sub>Te<sub>3</sub>. Fig2.(b) represents the results of photoelectron spin interference performed at each of wavenumbers k<sub>1</sub>~k<sub>4</sub> in Fig2.(a). Also, we just give a brief description here, a slight correlation is observed between each phase and kinetic energy, indicating that phase decomposition is possible. Using its data and Eq.(1), we obtained the result that the delay time  $\tau_{EWS} = 1.29 \times 10^{-18}$  s.



[Fig1.] (a) Electron orbitals and spins of topological surface states on the mirror plane. (b) Concept of photoelectron spin interference. (c) Concepts that tie our work to EWS model [5-7,11]. The wave packets of two transition are scattered differently as they pass thorough the potential V, therefore  $\phi$  occurs between the two transitions. We noted that  $\phi$  is extracted for each  $E_k$  and  $\tau_{EWS}$  is calculated from Eq.(1).



**[Fig2.]** (a) ARPES of topological surface states in Bi<sub>2</sub>Te<sub>3</sub>. Temperature is 40~50K. Photon energy is 6.3eV. Photon is liner polarization.  $k_1 \sim k_4$  are wavenumber of each spin detection. Clean surface was obtained by cleaving in  $7.3 \times 10^{-8}$  Pa. The mirror plane ( $\Gamma$  - M direction), the detection plane and the slit direction are all parallel. (b) Spin detection with polarization rotation at binding energies corresponding to each wave number.  $S_{\text{seff}} = 0.27$ . The dots are experimental values, and the solid lines are model fittings [5-7].

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