

2. Status of SARPES

Spin and angle-resolved photoemission spectroscopy (SARPES) is a powerful experimental technique to reveal various information about the occupied electronic states in solids, including their energy, momentum, and spin. In recent years, the demand for spin-resolved measurements has been getting high because of much interest in Rashba surface states and topological materials with spin-polarized electronic structures derived from strong spin-orbit interactions. These experiments require high energy resolution and enough photoelectron yield rates to detect their small energy scales (~several meV). Following these demands and requirements, we have developed a high-energy resolution SARPES apparatus using a vacuum-ultraviolet (6.994-eV) laser and very-low-energy-electron-diffraction (VLEED) type spin detectors at the Laser and Synchrotron Research Center (LASOR) in the Institute for Solid State Physics (ISSP) [1]. Our SARPES apparatus is currently utilized to obtain precise information on fine distributed spin-dependent electronic structures near the Fermi level in solids. We started the project to construct the laser-SARPES apparatus in FY 2014 and joint research at this station started in FY2015.

Our laser-SARPES apparatus consists of an analysis chamber, a carousel sample-bank chamber connected to a load-lock chamber, and a molecular beam epitaxy (MBE) chamber. All of these chambers are connected via UHV gate valves. Figure 1 represents the hemispherical electron analyzer, which is a custom-made ScientaOmicron DA30-L, modified to attach the VLEED type spin detectors. The electrons are currently excited by 6.994-eV photons, yielded by the 6th harmonic of a high-power Nd:YVO₄ quasi-continuous wave laser. A helium discharge lamp (VG Scienta, VUV5000) is also available as a photon source. At the MBE chamber, samples can be heated by a direct current heating or electron bombardment. The surface evaluating and preparing instruments, such as evaporators, low energy electron diffraction, sputter-gun, and quartz microbalance can be installed. At the carousel chamber, 16-samples can be stocked in the UHV environment. Spin-polarized states were investigated in both bulk and surface of various topological materials including magnetic and superconducting ones, atomic layers, and ferromagnetic compounds.

Spin-resolved technique with highly efficient photoelectron yield rates has advantages not only in high-resolution measurements but also in time-resolved

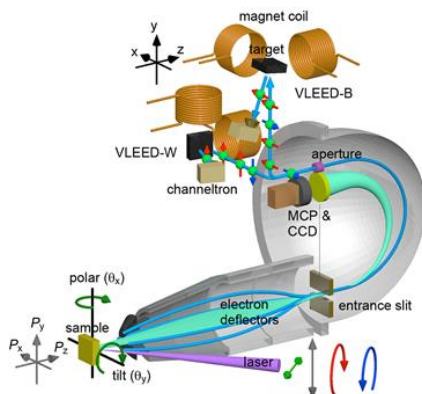


Figure 1: The laser-SARPES apparatus at LASOR in ISSP [1]. The double VLEED spin detectors are attached to a hemispherical analyzer (DA30-L, ScientaOmicron). Currently, 7-eV laser for high energy resolution measurement and 10.7eV pulsed laser for pump- probe measurements are available.

measurements based on pump-probe methods. A combination of SARPES with pump-probe laser techniques makes it possible to image the optically excited spin-polarized electron population in the unoccupied bands in energy-momentum space and track the dynamics of charge and spin in the ultrafast time-domain, which is useful for studying novel optical responses of many materials such as topological and spin-valley materials. Since FY2018, we have started to upgrade our laser-SAPRES apparatus by combining a pump-probe laser, setting up the system named time-resolved SARPES (tr-SARPES). This project requires a short pulse laser, and thus we newly installed a pulsed 10.7-eV laser system [2,3] developed by the Kobayashi group at ISSP's LASOR. This fundamental laser system is ytterbium fiber-based, achieving a 270-fs, 1-MHz repetition rate, and high power by a chirped pulse amplification technique, which allows the pump-probe measurements with high photoelectron yield rates. In addition to a great capability of the time-resolution, our 10.7-eV laser gives more wide momentum information in contrast to the low photon energy sources such as 7-eV.

The development progressed, and now tr-SARPES measurements have been realized. The 10.7-eV laser as the probe photon is obtained as the third harmonic generation in Xe gas, and a unique Xe gas chamber has been developed for the system up to the monochromatization and the focusing system. In addition, the light polarization of the 10.7-eV laser can be selectively controlled by MgF₂ half-wave plate. The pump photon is selectable from 1.19-eV or 2.38-eV for a wide variety of band-gap materials. The beamline is designed as a vacuum chamber after 10.7-eV generation, and it is connected to the UHV analyzer chamber through a differential pumping system without a vacuum window. Furthermore, some mirror and lens in the vacuum beamline are movable, allowing the system to switch between 7-eV and 10.7-eV beamlines. In this way, this system has made tr-SARPES possible, which is demonstrated in Fig. 2. This state-of-the-art tr-SARPES apparatus can be widely used for studying unoccupied electronic states in spintronics materials and their ultrafast carrier/spin dynamics.

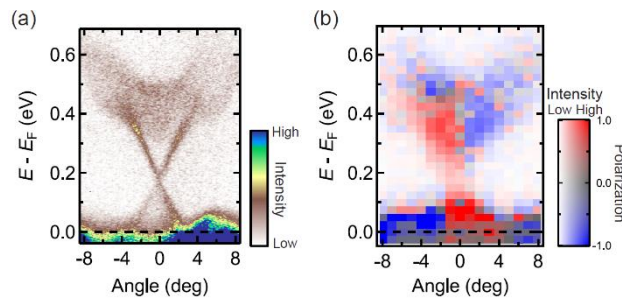


Figure 2: Time-resolved ARPES (a) and tr-SARPES mapping (b) of unoccupied topological surface state in Sb₂Te₃, observed by 10.7-eV laser after 1.19-eV laser pumping.

References:

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