1. Status of SARPES

Spin and angle-resolved photoemission spectroscopy (SARPES) stands as a powerful experimental technique that provides detailed information about the occupied electronic states in solids, including their energy, momentum, and spin. Recetnly, the growing interest in Rashba surface states and topological materials with spin-polarized electronic structures due to strong spin-orbit interactions has highlighted the importance of spin-resolved measurements. These spin-resolved experiments require high energy resolution and sufficient photoelectron yield rates to effectively detect small energy scales on the order of several meV. In response to these criteria, we successfully developed a high-energy resolution SARPES setup at the Laser and Synchrotron Research Center (LASOR) in the Institute for Solid State Physics (ISSP), using a vacuum-ultraviolet (6.994-eV) laser and spin detectors with the very-low-energy-electron-diffraction (VLEED) [1]. Since our initiative of developing the laser-SARPES in FY2014 and the commencement oft he facility for collaborative research in FY2015, our SARPES station has been instrumental in obtaining precise spin-resolved electronic structures near the Fermi level in solids.

Our laser-SARPES setup consists of an analysis chamber, a carousel sample-bank chamber connected to a load-lock chamber, and a molecular beam epitaxy (MBE) chamber, all connected via ultra high vacuum (UHV) gate valves. The hemispherical electron analyzer, a custom-made ScientaOmicron DA30-L, is designed to incorporate a VLEED-type spin detector. The available photon sources for electron excitation include the 6.994-eV laser, generated as the 6th harmonic of a high-power Nd:YVO4 quasi-continuous wave laser, and a helium discharge lamp (VG Scienta, VUV5000). In the MBE chamber, various instruments for surface evaluation and preparation, such as evaporators, low-energy electron diffraction, a sputter gun, and a quartz microbalance, can be installed, with samples heated by direct current heating or electron bombardment. The carousel chamber offers UHV storage for up to 16 samples. Spin-polarized states have been studied in both the bulk and surface of various topological materials, including magnetic and superconducting ones, atomic layers, and ferromagnetic compounds.

In FY2018, we began upgrading our laser-SAPRES system by integrating a pulsed laser to establish a pump-probe measurement setup. The newly installed pulsed 10.7-eV laser system [2,3], based on ytterbium fiber, achieves a 270-fs pulse duration, 1-MHz repetition rate, and high power through chirped pulse amplification, developed by the Kobayashi group at LASOR in ISSP[4]. This advanced system, as shown in Fig. 1, allows us to measure optically excited electron populations in unoccupied bands across the energy and momentum space and to track pump-induced ultrafast dynamics of both charge and spin. In addition to offering significant capabilities for the time-resolved measurements, the 10.7-eV laser system provides wider momentum information than lower photon energy sources such as 6.994-eV laser and offers better momentum resolution and polarization controls than the helium discharge lamp. The polarization of the 10.7-eV probe laser, generated through third harmonic generation in Xe gas, can be selectively controlled using an MgF_2 half-wave plate, while the pump photon energy is selectable between 1.19-eV and 2.38-eV to accommodate a variety of band-gap materials. The original 6.994-eV light source remains available by adjusting mirrors and lenses in the vacuum beamline.

In FY2023, nine research proposals from external researchers were accepted and conducted using our SARPES setup, resulting in the publication of two research papers [4,5]. These proposed studies explored spin-polarized states across various platforms, including half-metallic ferromagnets, Dirac semimetals, topological insulators, two-dimensional materials, ferromagnetic oxides, and organic compounds. To support these advanced studies and further improve our experimental capabilities, we have also been focusing on upgrading the 10.7-eV laser system to enhance the stability of the pulsed light. This includes the development of laser evaluation systems, such as an autocorrelation measurement setup and Frequency-Resolved Optical Gating (FROG), as well as the integration of a new rod fiber amplifier into the system, which allows for higher output light.

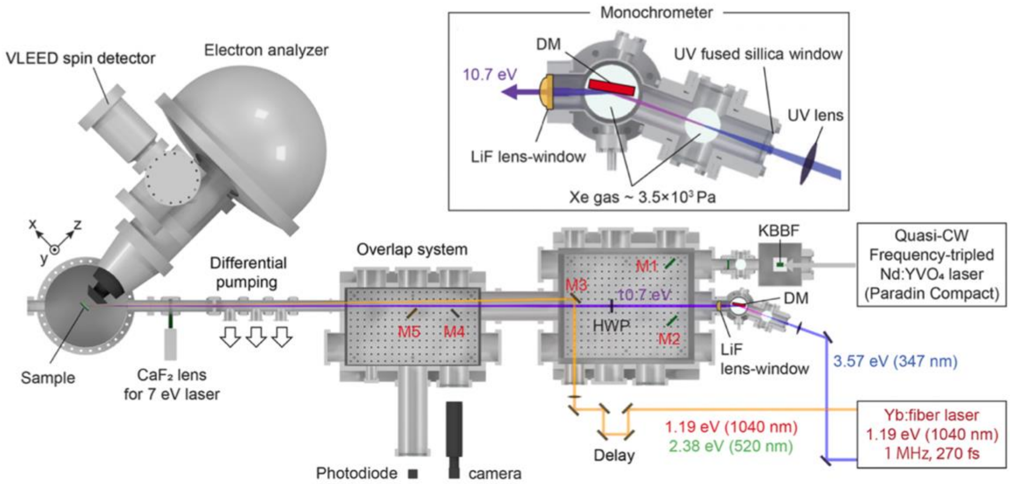

Fig. 1 Scaled layout of the 10.7-eV pulse laser beamline for tr-SARPES[4].

References:

[1] K. Yaji, A. Harasawa, K. Kuroda, S. Toyohisa, M. Nakayama, Y. Ishida, A. Fukushima, S. Watanabe, C.-T. Chen, F. Komori and S. Shin, Rev. Sci. Instrum. 87, 053111 (2016).

[2] Z. Zhao and Y. Kobayashi, Opt. Exp. 25, 13517 (2017).

[3] Z. Zhao, K. Kuroda, A. Harasawa, T. Kondo, S. Shin and Y. Kobayashi, Chin. Opt. Lett. 17, 051406 (2019).

[4] K. Kawaguchi, K. Kuroda, Z. Zhao, S. Tani, A. Harasawa, Y. Fukushima, H. Tanaka, R. Noguchi, T. Iimori, K. Yaji, M. Fujisawa, S. Shin, F. Komori, Y. Kobayashi, and T. Kondo, Rev. Sci. Instrum. 94, 083902 (2023)

[5] R. Moue, H. Yamazaki, T. Kitazawa, K. Yaji, H. Yaguchi, K. Kuroda, T. Kondo, A. Harasawa, T. Iwahashi, Y. Ouchi, S. Shin, and K. Kanai, Chem. Nano. Mat. 9, e202200538 (2023)