２． Status of SARPES

Spin and angle-resolved photoemission spectroscopy (SARPES) stands as a powerful experimental technique that reveals various information about the occupied electronic states in solids, including their energy, momentum, and spin. In recent years, the surge in interest around Rashba surface states and topological materials with spin-polarized electronic structures derived from strong spin-orbit interactions has elevated the significance of spin-resolved measurements. These spin-resolved experiments require high energy resolution and enough photoelectron yield rates to effectively detect the small energy scales of about several meV orders. Following these criteria, we have successfully developed a high-energy resolution SARPES setup utilizing a vacuum-ultraviolet (6.994-eV) laser and spin detectors with the very-low-energy-electron-diffraction (VLEED) at the Laser and Synchrotron Research Center (LASOR) in the Institute for Solid State Physics (ISSP) [1]. Since our initiative of developing the laser-SARPES in FY2014 and the facility commencement for collaborative research in FY2015, our SARPES station has been used to obtain precise spin-resolved electronic structures in proximity to 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 of these chambers are connected via UHV gate valves. The hemispherical electron analyzer is a custom-made ScientaOmicron DA30-L adapted to incorporate VLEED-type spin detector. The avairable photon sources to excite electrons in materials are the 6.994-eV laser, generated by 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, the surface evaluating and preparing instruments, such as evaporators, low energy electron diffraction, sputter-gun, and quartz microbalance, can be installed, and samples can be heated by direct current heating or electron bombardment. The carousel chamber offers strage for 16 samples within the UHV environment. Spin-polarized states were investigated in both the bulk and surface of various topological materials, including magnetic and superconducting ones, atomic layers, and ferromagnetic compounds.

Beginning in FY2018, we initiated an upgrade of our laser-SAPRES apparatus by integrating a pulsed laser to develop a pump-probe measurement setup. The newly installed pulsed 10.7-eV laser system [2,3] is ytterbium fiber-based, achieving a 270-fs, 1-MHz repetition rate, and high power by a chirped pulse amplification technique, developed by the Kobayashi group at LASOR in ISSP[4]. This newly developed system, as shown in Fig. 1, enables us to measure optically excited electron populations in the unoccupied bands in energy and momentum space and track the pump-induced ultrafast dynamics of both charge and spin. In addition to a great capability of the time-resolved measurements, the 10.7-eV laser system provides wider momentum information than that is possible with low photon energy sources such as 6.994-eV and gives better momentum resolution and polarization controls than the helium discharge lamp. In the developed system, while the light polarization of 10.7-eV probe laser, obtained as the third harmonic generation in Xe gas, can be selectively controlled by MgF_2 half-wave plate, pump photon is selectable from 1.19-eV or 2.38-eV for a wide variety of band-gap materials. The original 6.994-eV light source is still available by switching some mirrors and lens in the vacuum beamline.

Currently, we have been working on upgrading the 10.7-eV laser system aimed at stabilizing the pulsed light and installing an optical parametric amplifier (OPA). In addition to the development of an autocorrelation measurement setup, we have assembled laser evaluation systems such as Frequency-resolved optical gating (FROG) to improve the stability of the light. A new amplifier (rod fiber) has been introduced to the system, making higher output light available. These upgrades will enable us to combine our system with the OPA, where the wavelength (photon energy) of the pump light can be selectively tuned between 1350 nm (~0.92-eV) and 4200 nm (~0.3-eV). By leveraging such tunable low-energy pump lights, we will be able to access various exotic light-induced states in quantum materials, such as Floquet states with spin resolution.

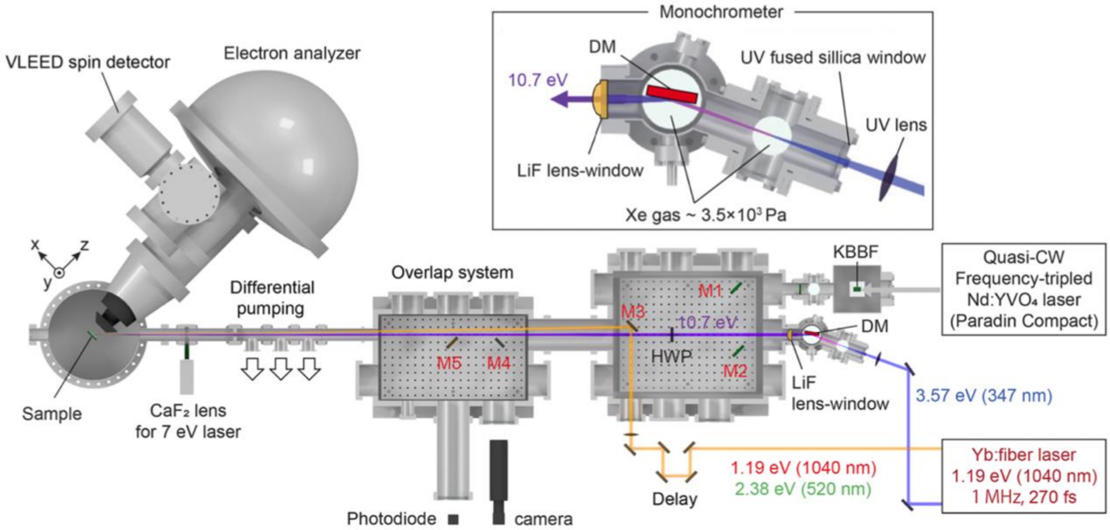

Fig. 1 Scaled layout of the 10.7-eV pulse laser beamline for tr-SARPES[4].

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)