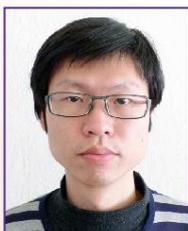


# 2022 Taiwan-AVS Symposium

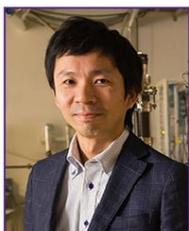
利用同步輻射能譜技術探索先進材料的新興特性  
Exploring the emergent properties of  
advanced materials with synchrotron-based spectroscopy

 **25 January, 2022, Taipei, Taiwan**



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**2022 TAIWAN-AMERICAN VACUUM SOCIETY (AVS) SYMPOSIUM:  
Exploring the emergent properties of advanced materials with synchrotron-based spectroscopy**

美國真空協會台灣分會:利用同步輻射能譜技術探索先進材料的新興特性

**Time: 25 January 2022 (Tuesday)**

**Location: General Hall 3F/International Conference Hall**

**地點: 綜合館 3F 國際會議廳**

***Session I***

**Time zone (UTC+8)**

<b>Time</b>	<b>Events</b>
08:50	<b>Opening Remarks</b>
	<i>Session Chair: Cheng-Maw Cheng</i>
09:00	<b>Multi-dimensional doping of topological Kondo insulator SmB<sub>6</sub> with ARPES spectromicroscopy</b> <i>L. Andrew Wray, New York University, USA</i>
09:30	<b>Micro-ARPES study of novel topological materials</b> <i>Takafumi Sato, Tohoku University, Japan</i>
10:00	<b>Electronic structure studies of atomically thin 4d/5d transition metal oxide films by angle resolved photoemission spectroscopy</b> <i>Changyoung Kim, Seoul National University, Korea</i>
10:30	<b>Evolutions of Dirac Fermions in Atomic Layers</b> <i>Iwao Matsuda, University of Tokyo, Japan</i>
11:00	<b>Plenary Talk &amp; Lunch</b>

***Session II***

**Time zone (UTC+8)**

	<i>Session Chair: Ashish Chainani</i>
13:00	<b>Present status of UVSOR photoelectron momentum microscope: a case study for the molecular film</b> <i>Satoshi Kera, UVSOR Synchrotron Facility, Institute for Molecular Science, National Institutes of Natural Sciences, Japan</i>
13:30	<b>Recent status of a soft X-ray photoemission electron microscope end-station at the Taiwan Photon Source</b> <i>Tzu-Hung Chuang, NSRRC, Taiwan</i>
13:50	<b>Weyl Fermions in chiral crystals</b> <i>Alberto Crepaldi, Politecnico di Milano, Italy</i>
14:20	<b>Optimizing high harmonic generation EUV source for Tr-ARPES by multiple-plate continuum technique</b> <i>Ping-Hui Lin, NSRRC, Taiwan</i>
14:40	<b>Coffee break</b>

### *Session III*

**Time zone (UTC+8)**

<b>Time</b>	<b>Events</b>
	<i>Session Chair: Ro-Ya Liu</i>
16:50	<b>Soft x-ray spectroscopy studies of n-type ferromagnetic semiconductor (In,Fe)As: A materials perspective for carrier-induced ferromagnetism</b> <i>Masaki Kobayashi, University of Tokyo, Japan</i>
17:20	<b>In operando electronic structure of 2D material devices</b> <i>Søren Ulstrup, Aarhus University, Denmark</i>
17:50	<b>Frontiers in spin-resolving momentum microscopy: from magnetism to topology</b> <i>Christian Tusche, Forschungszentrum Jülich, Germany</i>
18:20	<b>Closing Remarks</b>
18:30	<b>Banquet</b>

# Multi-dimensional doping of topological Kondo insulator $\text{SmB}_6$ with ARPES spectromicroscopy

L. Andrew Wray<sup>1</sup>, Erica Kotta<sup>1</sup>, Yishuai Xu<sup>1</sup>, J. W. Lee<sup>2</sup>, Shouzheng Liu<sup>1</sup>, Lin Miao<sup>3</sup>, Jonathan D. Denlinger<sup>4</sup>, Turgut Yilmaz<sup>5</sup>, Elio Vescovo<sup>5</sup>, B. K. Cho<sup>2</sup>

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Controlling the density of electrons inside an insulator via the chemical potential is a cornerstone of modern electronics, enabling the electrical conductivity of semiconductors and the emergence of fascinating new properties linked with electronic correlations. The compound  $\text{SmB}_6$  has drawn widespread attention in recent years as the first insulator to feature both strong electronic correlations and topological quantum order, potentially enabling a host of new phenomenologies as charge density is modified. However, chemical potential has not been experimentally controlled in studies to date of the electronic structure. I will present an angle-resolved photoemission spectromicroscopy ( $\mu$ -ARPES) study of  $\text{SmB}_6$  alloys, using the natural inhomogeneity of sample surfaces to create the analogue of a multi-dimensional doping series. The role of electronic strong correlations is observed in the interplay of the topologically ordered conducting states with one another and with the chemical potential. Higher impurity densities are found to result in the transformation of these interdependencies as the disorder from impurities erodes quantum coherence. These findings set the stage for a holistic understanding of the interplay between strong correlations and topological features in the electron system.

# Micro-ARPES study of novel topological materials

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Angle-resolved photoemission spectroscopy (ARPES) has established as a standard experimental technique to elucidate key electronic states of quantum materials such as high-temperature superconductors and topological insulators. Recent progress in the spin- and time-resolved ARPES techniques further enable us to clarify electron dynamics and spin-dependent band structure of various functional materials in great details. Spatially resolved ARPES also offers an excellent opportunity to access electronic states in even wider materials which are hard to be accessed by conventional ARPES.

We have recently developed a versatile micro-focused ARPES system with Kirkpatrick-Baez mirror optics in BL28-A, KEK-PF, Japan [1], and have achieved the beam spot size of 10  $\mu\text{m}$  (horizontal) $\times$ 12  $\mu\text{m}$  (vertical). By using micro-ARPES, we have studied various topological materials such as topological Dirac semimetals [2] and Kagome superconductors [3, 4], and clarified key electronic states associated with the mechanism of unconventional physical properties. We are currently developing a nano-spin-ARPES system which achieves sub-  $\mu\text{m}$  spatial resolution. In this talk, I will discuss the present status and future perspective of micro/nano-ARPES and its application to quantum materials.

This work was supported by JST-CREST, and was carried out in collaboration with T. Kato, T. Kawakami, T. Kawakami, D. Takane, C. X. Trang, S. Souma, K. Sugawara, K. Nakayama, T. Takahashi (Tohoku Univ.), M. Kitamura, K. Horiba, H. Kumigashira (KEK), K. Segawa (Kyoto Sango Univ.), K. Yamauchi (Osaka Univ.), Z. Wang (Beijing Inst. Tech.), and Yoichi Ando (Univ. Collogne).

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[2] D. Takane *et al.*, *Sci. Rep.* **11**, 21937 (2021).

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# Electronic structure studies of atomically thin 4d/5d transition metal oxide films by angle resolved photoemission spectroscopy

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In this presentation, I will discuss our recent results from ultrathin SrRuO<sub>3</sub> and SrIrO<sub>3</sub> films. 2D spin-polarized band structures of perovskite oxides generally support symmetry-protected nodal lines and points that govern both the sign and the magnitude of the AHE. To demonstrate this is the case for SrRuO<sub>3</sub> ultrathin films, we performed angle-resolved photoemission studies of ultrathin films of SrRuO<sub>3</sub>, a prototypical metallic ferromagnet with spin-orbit coupling (SOC). We found there are nodal lines and quadratic band crossing points in ultrathin SrRuO<sub>3</sub> films. These symmetry-protected nodal lines and quadratic band crossing points are sources of Berry curvature which causes the observed novel anomalous Hall effects.

We go further and studied 1 unit cell (uc) thick SrRuO<sub>3</sub> films. We observe that 1 uc films are not insulator but correlated metal. We can also use strain from various substrates and induce metal-insulator transition. This includes octahedron distortions as well as simple strain. We show that we can control the system from a good metal to a Mott insulator. Such control is also attempted for SrIrO<sub>3</sub> and we also find it metallic down to 1 uc film.

While these systems are ultimate 2D systems, we find it important to have the energy tunability and polarization control of Synchrotron radiation. In light of this aspect, I will discuss the importance of and *in situ* film growth system at the beam line.

# Evolutions of Dirac Fermions in Atomic Layers

Iwao Matsuda<sup>1</sup>

<sup>1</sup> *the Institute for Solid State Physics, the University of Tokyo, JAPAN*

Two-dimensional (2D) monatomic layers containing Dirac fermions have recently attracted intensive interests for pursuing fundamental science and developing quantum devices [1]. Research on free-standing graphene layers have developed the basic science of Dirac fermions and revolutionized our nanotechnology. This has motivated the further works to trace the carrier dynamics and to synthesize novel monatomic layers, such as “borophene” [2] and atomic sheets of the Dirac nodal semimetal [3,4]. Methods of photoemission spectroscopy have made significant contributions for detailed characterizations of the electronic structure. We have updated the techniques not only in momentum, spin, and time resolutions but also for *operando* measurements under the ambient conditions to reveal the functionalities [5-7].

In this presentation, I will introduce photoemission experiments in our institute and the related instrumentation, a nano-focusing mirror, that is developed recently through our experience with X-ray free electron lasers at SACLA [8]. As an example of the material science of atomic layers, comprehensive research of carrier dynamics in the epitaxial graphene will be described. We performed measurements of time- and angle-resolved photoemission spectroscopy (ARPES) on various systems of single and bilayer graphene on the Si-face SiC substrates [5]. In quasi-crystalline bilayer graphene, we confirmed temporal evolutions of photo-excited carriers in Dirac cones that were asymmetric between the upper and lower layers, which can be described by the transient doping from the interface. The quantitative discussion was made with the structural parameters, interlayer distance, determined by positron diffraction (PD) experiments [9]. The concerting usages of ARPES and PD reveal the transport mechanism that opens a way to regulate carrier dynamics in graphene by the environment effect for developing the next-generation optoelectronics.

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# **Present status of UVSOR photoelectron momentum microscope: a case study for the molecular film**

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<sup>1</sup> *UVSOR Synchrotron Facility, Institute for Molecular Science, National Institutes of Natural Sciences,  
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Functional organic molecule (FOM) has recently attracted considerable attention both for fundamental research and device applications because of peculiar properties not found in inorganics and small molecules. However, the mechanisms and the origin of various device characteristics are still under debate. Scientific discussions have been redundant because of long-standing beliefs that the electronic structure of FOM would be conserved as in an isolated molecule even for solid phases due to the weak van der Waals interaction. To reveal characteristics of FOM, it is essential to investigate precisely the electronic structure at various interfaces, including organic-organic and organic-inorganic (metal/semiconductor) contacts.

We have realized that the weak electronic interaction manifests itself as small intensity modulations of fine structures in photoelectron spectra, depending on the adsorption and aggregation conditions on the surface. Thanks to recent instrumentation improvements we can assess hidden fine features in the electronic states.

Recent results taken by the photoelectron momentum microscope (PMM) installed at UVSOR Synchrotron Facility, which allows giving a global view of an electron cloud distribution for weakly bounded electronic systems, will be shown to discuss. We have built a new PMM station for 3D momentum-resolved photoelectron spectroscopy with a microscope field of view at the soft X-ray beamline BL6U of UVSOR-III. The details of the specification evaluation result are described elsewhere [1-3]. In brief, the energy, spatial, and momentum resolutions of the analyzer were estimated to be 20 meV, 50 nm, and  $0.012 \text{ \AA}^{-1}$ , respectively. Samples can be cooled down to 8 K and heated up to 400 K. We will upgrade the current system in FY2022 for spin-resolved PMM.

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[2] S. Makita, et al., e-J. Surf. Sci. Nanotech. 19, 42 (2021).

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# Recent status of a soft X-ray photoemission electron microscope end-station at the Taiwan Photon Source

Tzu-Hung Chuang<sup>1</sup>

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The Photoelectron Related Image and Nano-Spectroscopy (PRINS) end-station at the Taiwan Photon Source (TPS) 27A2 will host a photoelectron microscope that aims to work on photoelectron-related imaging and nano-scale spectroscopy through the combination of an imaging-type electron column integrated with a hemispherical electron energy analyzer and an imaging spin filter. The microscope is able to conduct full-field imaging by collecting photoelectrons in either real-space or momentum-space with spin contrast.

Taking advantage of an elliptically polarized undulator (EPU) photon source and powering by an in-house designed active-mirror plane grating monochromator, which delivers soft x-rays in a range of 90-3000 eV [1], the PRINS microscope has multiple capabilities, such as (1) X-ray absorption spectroscopy (XAS)-based and X-ray magnetic circular dichroism (XMCD)-based real-space imaging to obtain element-resolved mapping or spin-texture information, (2) X-ray photoelectron spectroscopy (XPS)-based real- and momentum-space imaging to obtain spatially-resolved chemical state mapping and band-structure imaging, (3) spin-resolved band structure measurement when the imaging spin filter is introduced, and (4) micro-area spectroscopy (XAS, XPS, and angle-resolved photoelectron spectroscopy) extracted from a series of images measured at different kinetic energies.

The on-site testing of the system is expected to be performed by an UV light source in the first-half of 2022. A sample preparation chamber with standard surface science characterization capabilities, including the ion sputtering, low energy electron diffraction, Auger electron spectroscopy, and electron-assisted thin-film deposition, will be connected to the main microscope system to enable in-situ experiments. Technical capabilities and scientific opportunities of the end-station will be discussed in the talk, and the recent status will also be reported.

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# Weyl Fermions in chiral crystals

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In chiral crystals, the absence of inversion and mirror symmetry is responsible for unconventional spin properties and the emergence of exotic topological fermions [1]. Trigonal tellurium is one of the simplest chiral crystals, and by using spin-and-angle-resolved photoemission spectroscopy (srARPES) we have recently reported signatures of composite, accordionlike and Kramers-Weyl (KW) fermions in its band structure [2]. In contrast to conventional Weyl fermions that arise from band inversion, and are thereby degenerate in energy, KW points with opposite topological charges are pinned at different energies and at different time-reversal invariant momenta (TRIM) by the action of time-reversal symmetry.

In KW fermions the spin is predicted to lie parallel to the wavevector, thus realizing a hedgehog texture [1]. In our study we clarify that the radial spin texture is not a prerogative of the KW fermions, but it can be observed also at non-TRIM point [2], as confirmed by an independent srARPES study of the spin properties around the H point of Te [3]. This is made possible by the presence of multiple rotational axes, combined with the breaking of mirror symmetry. Our results illustrate how the arrangement of spin in the reciprocal space is a consequence of the local point group symmetry, and it does not reflect only global properties of the crystal. Spin texture more complex than hedgehog-like can be stabilized, sharing common features with the arrangement taken in Skyrmions by magnetic momenta in real space [3, 4]. These spin textures are important ingredient to explain the chiral induced spin selectivity (CISS) effect that might find application in spintronics devices [5]; hence a complete classification of the spin texture for all local point group symmetries is highly demanded.

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# Optimizing high harmonic generation EUV source for Tr-ARPES by multiple-plate continuum technique

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The advent of high power femtosecond lasers and high harmonic generation (HHG) processes are definitely the milestone for time-resolved spectroscopy technique. For the application of the angle-resolved photoemission spectroscopy HHG allows us access to a broader area in momentum space. However, the photon flux and the stability, especially for the probe beam, hold the key to the experiment execution. Sufficient photon flux is essential for acquiring decent spectrum within a reasonable acquisition time.

With limited average power of the laser source, one would have to balance between the high repetition rate, peak power in a single pulse and try to maximize the HHG yield. We employ the multi-plate continuum (MPC) technique [1] and compress the fundamental pulse to 30 fs before the gas cell. In cooperation of conical geometry grating, the final photon flux is in the order of  $10^{10}$  to  $10^{11}$  photons/sec at 10 kHz repetition rate.

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# Soft x-ray spectroscopy studies of ferromagnetic semiconductors: A materials perspective for carrier-induced ferromagnetism

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Spintronics is a research field aiming at manipulating and utilizing both the charge and spin degrees of freedom of carriers [1]. To apply these spintronics functionalities to the well-established semiconductor technology, materials having both the semiconducting and ferromagnetic properties are highly desirable. Ferromagnetic semiconductors (FMSs), in which the cation sites in a semiconductor crystal are partially replaced by magnetic atoms, bear a high promise for the applications in semiconductor spintronics because of their capability to manipulate both the charge and spin degrees of freedom of carriers. Novel Fe-doped III-V FMSs have recently attracted much attention because Fe-doped FMSs can accommodate both n- and p-type carriers and exhibit ferromagnetism with high Curie temperature  $T_C$  above room temperature [1,2]. Using the p-type or n-type Fe-doped FMSs, spintronics devices have already been demonstrated.

Fundamental understanding of the origin or mechanism of ferromagnetism in FMSs is important for the application of the FMS materials to spintronics devices. Soft x-ray angle-resolved photoemission spectroscopy (SX-ARPES) and resonant inelastic x-ray scattering (SX-RIXS) are a powerful tool to examine the mechanism of carrier-induced ferromagnetism in FMSs [3,4]. Our SX-ARPES and SX-RIXS studies on FMSs demonstrate that the IBs hybridized with the ligand bands of the host semiconductors are key to understand the carrier-induced ferromagnetism and the band structures are deformed depending on the carrier concentrations, which is unexpected from the rigid-band picture. These results suggest a perspective for the realization of carrier-induced ferromagnetism in FMS materials.

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# In operando electronic structure of 2D material devices

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Understanding electronic and structural mechanisms that define intrinsic and extrinsic performance limits of two-dimensional (2D) materials integrated in functional device architectures is essential for the realization of truly 2D technologies. Obtaining such insights demands a probe that is capable of visualizing electronic structure on mesoscopic length scales and during operating conditions of devices.

Here, I will demonstrate the ability to visualize the energy- and momentum- dependent quasiparticle dispersion in 2D devices composed of graphene supported on a hexagonal boron nitride (hBN) dielectric and a graphite back-gate. The charge carrier-dependent spectral function of graphene is resolved using angle-resolved photoemission spectroscopy with nanoscale spatial resolution (nanoARPES) while the back-gate voltage is tuned. I will discuss how such measurements directly reveal the doping-dependent renormalized Fermi velocity of Dirac quasiparticles and elucidate electron-phonon and electron-plasmon interactions in graphene [1]. Position-resolved measurements of the graphene Dirac cone in the presence of an electrical transport current provide a map of the local electrostatic potential, which is combined with the local doping to estimate the spatially-dependent carrier mobility of graphene on hBN [2]. The work demonstrates the powerful concept of unifying spectroscopic and transport measurements on 2D materials, which allows for a simultaneous noninvasive local measurement of composition, structure, many-body effects and carrier mobility in the presence of high current densities and tunable charge carrier concentrations.

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# Frontiers in spin-resolving momentum microscopy: from magnetism to topology

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The physical properties of condensed matter systems are largely determined by the details of their electronic structure. The concepts of symmetry and topology govern the novel field of quantum materials [1]. The interplay of competing mechanisms often results in unusual charge and spin transport phenomena in such materials. In order to understand the physical properties of quantum materials on a fundamental level, we need to explore the underlying electronic structure in detail and disentangle role of the various interactions.

Only recently, the comprehensive experimental access to the spin-resolved band structure became feasible by spin-resolved momentum microscopy [2]. This novel concept combines high resolution imaging of the spectral function in two-dimensional ( $k_x$ ,  $k_y$ ) maps of the valence electronic structure with an imaging spin filter [3].

With this comprehensive spin-resolved information of the electronic states we discuss the role of the individual interactions and symmetry-breaking mechanisms. Starting with single crystalline materials, our experimental results reveal intricate effects of non-local electron correlations in ferromagnets [4] and the mixing of different spin states in topological materials [5]. Moreover, combining strong spin-orbit coupling and magnetism gives rise to complex spin-orbital textures, and topological phase transitions in the Fermi surface [6].

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