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Progress in Physics (88)

Photoemission from Solids

Moritz Hoesch [1], Claude Monney [2], and Felix Baumberger [3]
[1] Photon Science, Deutsches Elektronen-Synchrotron (DESY), Notkestrasse 85, DE-22607 Hamburg
[2] Department of Physics & Fribourg Center for Nanomaterials, University of Fribourg, Ch. du Musée 3, CH-1700 Fribourg
[3] Department of Quantum Matter Physics, University of Geneva, 24 Quai Ernest-Ansermet, CH-1211 Geneva 4, and Swiss Light Source, Paul Scherrer Institut, CH-5232 Villigen PSI

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I. Introduction

Photoemission spectroscopy analyses the energy and momentum distribution of photoelectrons from samples under illumination by ionising radiation. The theoretical description, involving quanta of energy $h\nu$ famously gave rise to Einstein's nobel prize (see the historical summary by H. R. Ott [1]). In modern notation, this energy conservation is written as

$$E_k = h\nu - \Phi - E_B, \quad (1)$$

where E_k is the kinetic energy of the photoelectron, ν is the frequency of the radiation, Φ is the work function of the surface under study and E_B is the binding energy of the electron's initial state. A spectrum of photocurrent as a function of E_k thus readily reveals the occupied states in a material. In addition to this energy conservation, also the momentum is conserved, although effects of the inevitably broken symmetry of the surface allow for strict momentum conservation only for the component parallel to the surface. The electron spin is conserved, too. The combination of energy and momentum conservation is the basis for plotting the data as maps against energy and momentum or against two momentum components. These intensity maps correspond to the band structure diagrams used in the theoretical description the electronic structure of solids. If a momentum map of the photoelectrons with the highest E_k is plotted, its high intensity lines directly correspond to the Fermi surface of the material [2], the fundamentally important part of the electronic structure, where transport occurs, thermal excitations are possible and magnetic as well as charge order phenomena can be linked to the movement patterns of the electrons in the material.

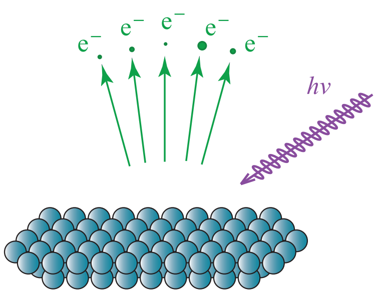

FIG. 1: Schematic view of photoemission. Photoelectrons are ejected from the sample upon irradiation with monochromatic ionising radiation. Different directions of emission with respect to the crystal lattice as well as to the polarisation and propagation of the light are recorded as spectra.

The technique goes by the name angle-resolved photoemission spectroscopy (ARPES) when such combined data sets are analysed. The potential of ARPES for the study of quantum materials has been recognised early on. Discussing Cuprate superconductors, P. W. Anderson famously wrote in 1992 in Physics Today "*Angle resolved photoemission is, for this problem, the experiment that will play the role that tunneling played for BCS superconductivity*". Capitalizing on this potential, however, required numerous instrumental advances to push the energy resolution of ARPES to the natural energy scale k_BT of superconductivity and other low-temperature quantum phases of matter.

II. High Resolution ARPES

Early key contributions to the understanding of the electronic structure of solids were realised solely due to high energy resolution [3]. These allowed for a distinction between Fermi liquid like electronic structures, with a characteristic high energy Fermi cut off and unusual non-Fermi-liquid-like metallic states. Also the direct measurement of the excitation gap of superconductors became possible in this way [4, 5] (see Fig. 2). When the momentum of the photoelectron excitation is analysed, too, the coupling of the itinerant electronic states to all other degrees of freedom of the solid, vibrational, magnetic and correlation-driven excitations becomes accessible [6]. In addition to mapping the Fermi surface and occupied bands of the electronic structure, the photoemission experiment can thus measure the full spectral function of the electronic system, including all excitations that interact with the charges, and it can do this at any temperature and on any crystalline material, provided the conductivity is high enough that the overall removal of electrons does not distort the spectra due to macroscopic charging of the sample.

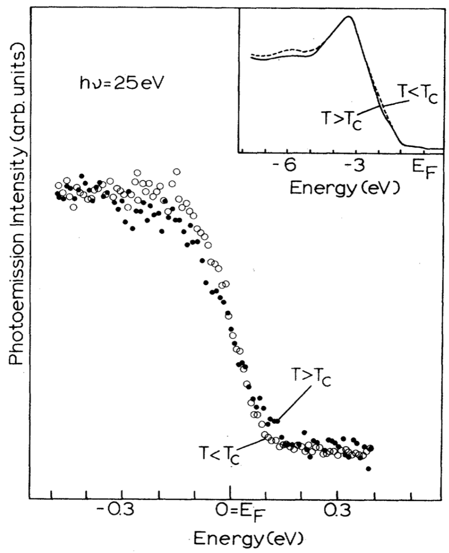

FIG. 2: Measurement of the superconducting gap of $Bi_4Ca_3Sr_3Cu_4O_{16+x}$ in 1989. From this data, Chang et al. [4] estimated a gap of 15 – 45 meV. The inset shows spectra over a larger energy range (reprinted from Ref. [4] with permission by the publisher).

On the technical side, every photoemission instrument consists of three components: a light source, a sample holder for positioning and orientation, and an electron spectrometer that analyses or selects the photoemission by kinetic energy and angle. These components are shown in Fig. 3. The technology of all three has developed over the years. The key light source of the turn of the century was the

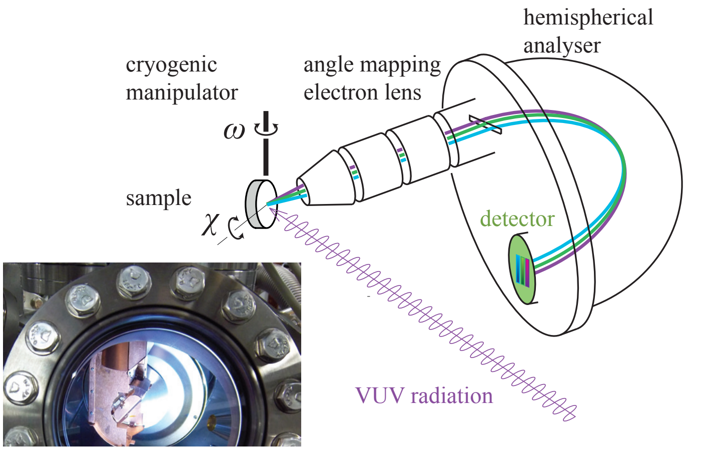

FIG. 3: Components of a photoemission instrument: the hemispherical analyser with pre-lens selects angles in one direction and projects the other, as well as a segment of kinetic energy onto the detector. The manipulator aligns the sample and is used to scan an angle. The light source provides monochromatic, polarised radiation.

He discharge lamp. When operated at optimised He gas pressure this yields spectroscopically sharp radiation at $h\nu$ = 21.221 eV among a few other emission lines. When combined with a monochromator to select this line, radiation of the highest purity at high intensity is obtained. The sample environment of photoemission is ultrahigh vacuum (UHV) with a low magnetic field background. Specialised cryogenic sample manipulators with temperatures down to few K nowadays allow for easy temperature changes and accurate positioning, all with minimised degassing. Equal progress has been made in the technology of photoemission spectrometers. The standard instrument is an electrostatic hemispherical analyser with a pre-lens that can either select a specific sampling area or, as shown in Fig. 3, project the angular distribution of photoelectrons onto the entrance slit area of the energy analyser and thus on the detector in one direction.

The use of synchrotron radiation for photoemission is particularly attractive due to its tuneable photon energy and polarisation. The technology of monochromators has also evolved, and an energy resolution that is quite comparable to the He lamp is routinely available, at any photon energy. This is particularly important for the use of the momentum resolution in photoemission. The momentum component parallel to the surface is fully conserved in photoemission. The only way to vary all components and map all of momentum space is by varying the excitation energy $h\nu$. A dedicated photoemission beamline is thus found in the instrument portfolio of almost every synchrotron radiation facility, including the Swiss Light Source.

These instruments are available to provide insight into the electronic structure of materials and electronically driven phenomena of solid state physics. Recent reviews summarise the most significant successes [7, 8], and here below we cite a few scientific cases that illustrate well recent progress. The largest number of publications of the last years relates to materials with nontrivial topological symmetries. Their rich phenomenology of momentum structures is routinely verified in relation to theoretical band structure calculations [9]. Among all iron based unconventional superconductors, FeSe has received the strongest attention from the ARPES community. It can be considerered the simplest, due to its simple stoichiometry, yet it shows the catching electronic stucture features that are common to all members of this family. A refined, although still controversial view on the mechanism of the 90 K phase transition, termed nematic order, has been derived from such data [10]. The rather low superconducting temperature and corresponding low gap value then made full use of the high performance of modern instruments [11] and important conclusions were drawn on the relationship with the orbital character in the nematic state and the gap isotropy [12]. Also the continuously intriguing properties of doped Cuprate high T_c superconductors continue to be further clarified by numerous ARPES studies [13]. Apart from superconductivity, the study of electron phonon coupling has enabled the distinction of fundamentally important model cases, including the unambiguous visualisation of the spectral signatures of the Fröhlich model in a 2D electron gas in $SrTiO_3$ [14]. The intricate temperature dependent re-arrangements of electronic states in Kondo lattice materials have been clarified in combinations of careful ARPES studies with other techniques [15, 16].

III. ARPES Using Lasers

The introduction of deep UV laser sources, pioneered by the group of S. Shin at the University of Tokyo marked an important step to improving the energy resolution of ARPES. As for other spectroscopies, high energy resolution in ARPES comes at the expense of low count rates. Light sources for ARPES should thus combine a narrow bandwidth with a high photon flux. This is natural strength of lasers. What proved more difficult is to maintain this combination deep into the UV range, which is reached by successive frequency conversion in non-linear crystals or gas cells. Efficient frequency conversion requires high peak power, which competes directly with the necessity of long pulses ($\gtrsim$ 10 ps) to maintain a narrow line width and a high repetition rate to avoid a loss of resolution from space charge, the inelastic scattering of photoelectrons in vacuum. Initially, narrow bandwidth laser-sources could only be achieved with non-linear crystals for $\lambda > 177$ nm ($h\nu < 7$ eV). This is sufficient to overcome the work function barrier but the low kinetic energy of photoelectrons emitted with such light sources restricts the accessible momentum range. Recent developments have now expanded the photon energy range of high-resolution laser-ARPES

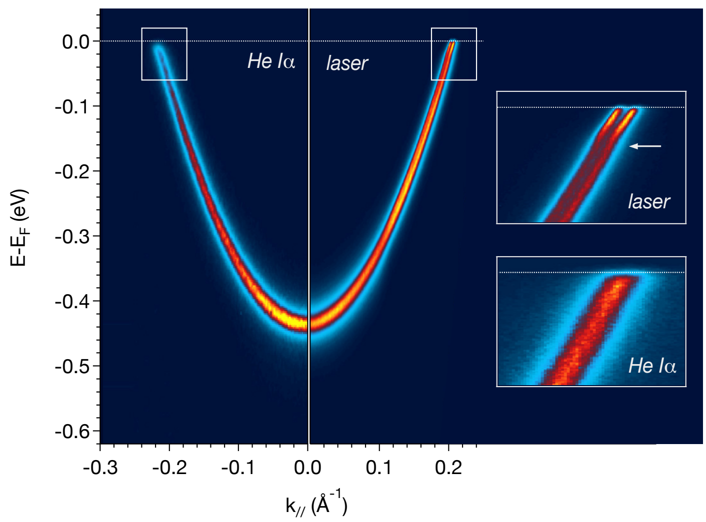

FIG. 4: Laser ARPES data from the Cu(111) surface state detecting a previously unresolved minute Rashba-type spin-splitting (adapted from Ref. [17] with permission by the publisher).

up to 11 eV - sufficient to map the entire Brillouin zone of most materials - while maintaining a bandwidth and photon flux that is superior to even the very best synchrotron beamlines [18, 19].

The improvement in resolution and cleanliness of ARPES data enabled by UV laser systems is illustrated in Fig. 4. Reinvestigating a well-known surface state on Cu by laser-ARPES, Tamai *et al.* reported a minute, previously unresolved Rashba-type spin splitting. The same data also shows a faint 'kink' in the dispersion near the chemical potential. This arises from the mass enhancement of low-energy quasiparticles interacting with phonons. The clear evidence for such a kink in Cu, which has an exceptionally low electron-phonon coupling strength, illustrates the remarkable sensitivity of modern ARPES experiments.

Laser-ARPES also evolved into a powerful tool for the study of topological insulators and semimetals. A particularly remarkable example is the detection of a Dirac cone topological surface state on the iron based superconductor $FeTe_{0.55}Se_{0.45}$ [20]. This surface state connects across a small spin-orbit induced band gap of a few 10 meV and its detection would have been out of reach only a few years earlier.

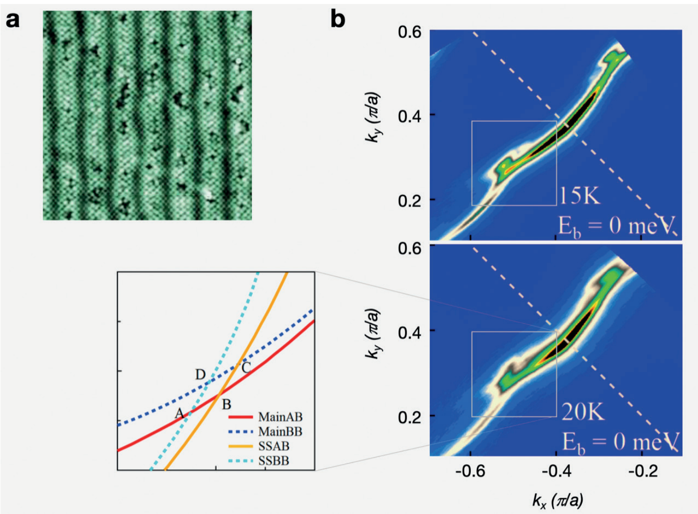

FIG. 5: Laser-ARPES data from Bi2212. (a) shows an STM topography with the characteristic superstructure (adapted from Ref. [21]). (b) laser-ARPES Fermi surfaces of optimally doped and overdoped Bi2212. The bottom right panel shows a schematic of the main and backfolded bands (adapted from Ref. [22] with permission by the publisher).

Fig. 5 shows an example for the application of laser-ARPES to cuprate superconductors. $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212), the most widely studied cuprate in ARPES has a periodic unidirectional superstructure, well known from diffraction and STM experiments. This additional periodicity leads to diffraction replicas of the electronic bands, evident in numerous conventional ARPES studies. Using a 7 eV laser source, Gao *et al.* could now show for the first time that these diffraction replicas open small hybridization gaps at the (avoided) crossings with the main bands [22]. This identifies the replica bands as an initial state effect modifying the intrinsic electronic structure of Bi2212 (as opposed to diffraction of the outgoing photoelectron final state).

IV. Time-Resolved ARPES

By using the pump-probe technique, ARPES can be extended to the time-domain to study the out-of-equilibrium dynamics of materials. In this approach, an intense pulse of low photon energy, the pump, excites the material under investigation and a subsequent weak pulse of light probes it by ejecting a photoelectron. By precisely tuning the time delay between the two pulses, one acquires a series of spectra to reconstruct the time-evolution of particular spectral features typically on the sub-picosecond timescale.

First pioneering time-resolved photoemission experiments were performed well before the year 2000, as for instance in the work of Fann and coworkers setting up the basis for the study of the thermalization of excited electrons in metals [25]. However, it was mainly after the turn of the century, notably thanks to technological advances made possible by commercial Ti:sapphire lasers, that time-resolved PES was used to look at the ultrafast dynamics of complex materials. The goal was typically to suppress an ordered phase and to identify the mechanism of its origin by comparing the relevant timescales for its suppression and subsequent recovery. One of the prominent cases was the study of the Mott-Insulator transition in the transition metal dichalcogenides (TMDC) TaS_2 by Perfetti and coworkers in 2006 [26] (see Fig. 6(A)).

Following this early success on correlated material, time-resolved ARPES studies looking at complex electronic dispersions came soon after with a study of the collapse of the charge density wave (CDW) phase in the tritelluride $TbTe_3$ [23]. A direct correlation between the collapse of the CDW band gap and the coherent oscillation of the related phonon mode allowed to identify the electron-phonon interaction as the origin of the CDW phase.

At that time, the probe photon energy was about 6 eV, close to the highest possible photon energy reachable using non-linear crystals. However, such low photon energies allow only to probe band dispersions close to the center of Brillouin zones. A breakthrough beyond this limitation occurred a few years later thanks to the process of high-harmonics generation (HHG) in gases. In 2011, Rohwer and coworkers made the tour de force of using 43 eV probe photon pulses generated by HHG to probe the CDW phase in the TMDC $TiSe_2$ [27]. By looking at the ultrafast collapse of the CDW band occurring in this material at high momenta (at the border of the Brillouin zone), they evidenced a surprisingly fast response that was attributed to the disputed excitonic insulator phase.

However, during the time the pump pulse is present and in interaction with the material, coherent interaction between the field of the pulse and the electronic system can lead to a new state of matter that can be optically manipulated. The work of Mahmood and coworkers on $Bi2Se_3$ opens the way to the observation of so-called Floquet-Bloch states, i.e. hybrid electronic states dressed by the photon field, with ARPES in 2016 [28].

The photoexcitation generated by the pump pulse can also be tuned to bring a material in an out-of-equilibrium state that displays an electronic structure or physical properties

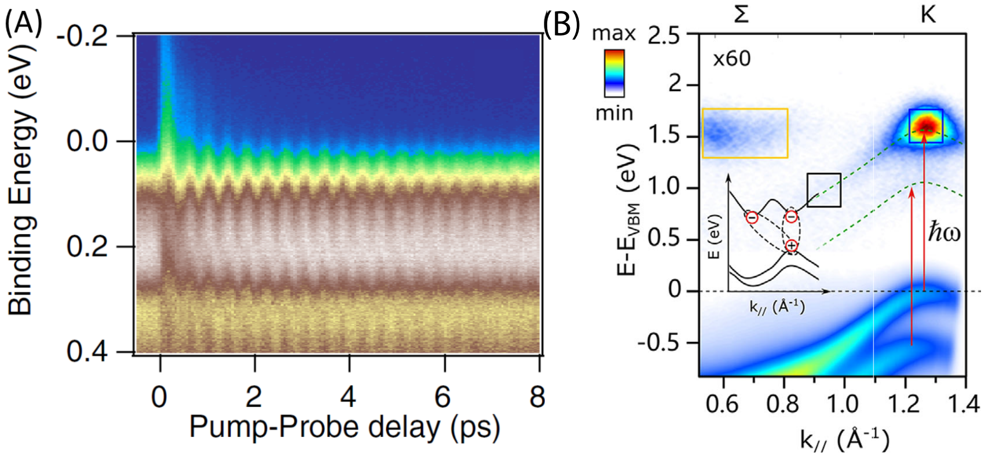

FIG. 6: (A) Time-resolved ARPES measurements on TaS_2 on a few ps timescale, showing the quasi-instantaneous closure of the Mott gap, concomitant with a coherent phonon oscillation (adapted from Ref. [26] with permission by the publisher). (B) Transient photoemission signal of the resonantly excited exciton appearing in the band gap of the semiconductor WSe_2 (adapted from Ref. [24] with permission by Prof. Ralph Ernstorfer).

that are never encountered at equilibrium. Optical manipulation of a semiconductor band gap was for instance evidenced in the layered material Ta_2NiSe_5 in 2017 [29].

Recently, time-resolved ARPES was taken to the next level with the realization of high repetition rate HHG sources. Indeed, previously, sources were typically operating at a few kHz, limiting the achievable quality of the signal, especially for states above the Fermi level and compromising on the energy resolution. In 2018, using HHG driven at 500 kHz as a pulsed photon source, Nicholson and coworkers uncovered the mechanism of the photoinduced phase transition in In nanowires by mapping their transient electronic structure [30]. More recently, Dong and coworkers presented a remarkable mapping of the unoccupied states in the semiconductor WSe_2, highlighting the ultrafast dynamics of excitons with time-resolved ARPES (see Fig. 6 (B)) [24].

V. Experiments in Operando

ARPES with few-micrometer or even sub-micrometer spatial resolution opens the way to study in-operando electronic devices. In micro- or nano-ARPES experiments, a conventional electron spectrometer is combined with a scanning sample manipulator and a tightly focused UV beam defining the spatial resolution. The main application of this variant of ARPES to date are 2D materials and in particular mechanically exfoliated van der Waals crystals and heterostructures thereof. This is a very active field of quantum matter physics where remarkable progress was achieved within a few years only. By now dozens of 2D materials including wide gap insulators, direct gap semiconductors, superconductors, magnets and topological materials have been exfoliated to monolayer or few-layer thickness. Combining such crystals into vertical heterostructures offers virtually unlimited possibilities to design artificial quantum matter with tailored properties. However, the typical lateral dimensions of a few microns of such van der Waals heterostructures pose a challenge for many traditional experimental methods of condensed matter physics. In ARPES this challenge has by now largely been overcome and successful experiments on exfoliated 2D crystals were performed at multiple specialized synchrotron nano-ARPES beamlines as well as with focused UV laser sources. Recent examples including different 2D semiconductors, the 2D topological insulator WTe_2 or magic angle twisted bilayer graphene are reviewed in Ref. [32].

The properties of 2D materials can often be tuned by electrostatic gating. Nano-ARPES experiments allow to image the effects of gating on the band structure. One of the first successful such experiments is shown in Fig. 7. Using a van der Waals heterostructure consisting of monolayer WSe_2 with a graphene contact, a graphite back gate and a boron nitride gate dielectric, Nguyen *et al.* showed that they can tune the chemical potential across the band gap of WSe_2 and populate the conduction band [31]. These experiments confirmed the direct band gap at the K-point in monolayer WSe_2 and showed that the Q-valley of the conduction band is only populated at slightly higher gate voltage corresponding to carrier densities around 10^{13} cm^{-2}. Recent proof-of-principle experiments of the Hofmann group on monolayer graphene also demonstrated local ARPES experiments on a current carrying device [33]. Extending such measurements to imaging the low-energy manybody physics of more complex systems such as magic angle twisted bilayer graphene will be a formidable challenge for the next decade.

VI. Summary and Outlook

ARPES, in its many flavours shown above will continue to be a key player in the exploration of solids. The photon energies used for these experiments span from the barely

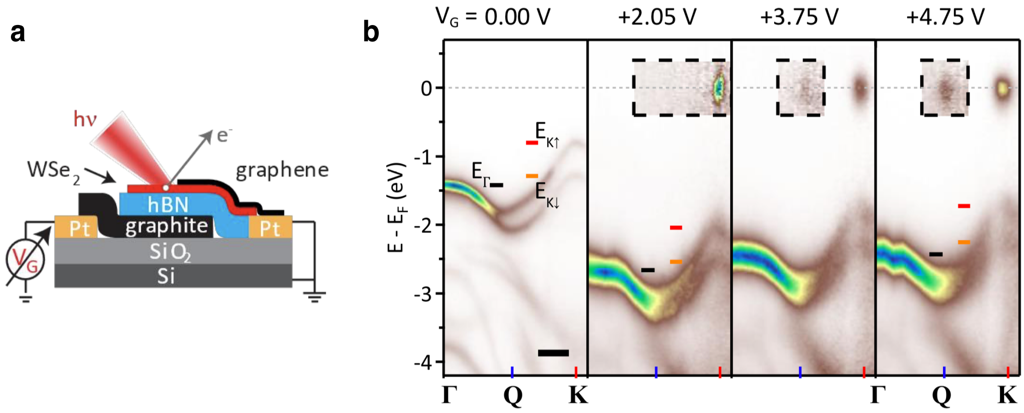

FIG. 7: ARPES measurements on gated monolayer WSe_2. (a) shows a schematic of the heterostructure with a graphite bottom gate, a hexagonal boron nitride dielectric and the monolayer WSe_2 contacted by monolayer graphene. (b) Gate voltage dependent ARPES data showing the bottom of the conduction band at the K point. At high gate voltage a second valley at Q becomes populated (adapted from http://arxiv.org with kind permission of Prof. Neil R. Wilson).

ionising 6 eV to approx. 0.6 keV where declining photoionisation yield marks the border beyond which the photon-hungry experiment starts to be unfeasible. ARPES requires a narrow photon bandwidth, but also gains hugely in precision and reliability when the photon energy is tunable. In this way the key development prospect continues to be on the side of photon sources, with interesting challenges for laser developers, as well as further optimisations of synchrotron radiation beam lines, the latter also on ultrashort pulse free electron lasers. All these highly coherent beams allow for ultimately diffraction limited focussing, thus the instruments can be equipped for in operando studies that will continue to yield entirely novel insight. Even the regime of fully coherent photoemission, where electron's coherence length of the state under study is larger than illumination beam spot will become accessible in the foreseeable future. The key benefit of ARPES to the progress of research is, however, its accessibility by researchers who focus on the questions and have the right materials for the studies. This benefit is best tapped into by further developments into reliable and robust detectors, sample manipulators and efficient protocols in their use.

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Moritz Hoesch is staff scientist at Deutsches Elektronen Synchrotron (DESY) in Germany and scientist in charge of the soft x-ray beamline P04 at the PETRA III storage ring. Before joining DESY he was in charge of construction and operation of the beamline I05-ARPES at Diamond Light Source in the UK where he directed the inhouse research program on ARPES, too.

Claude Monney is professor of physics at the University of Fribourg since 2018. His main research efforts are geared towards understanding and manipulating correlated materials using high energy resolution, as well as time-resolved spectroscopies.

Felix Baumberger is professor of physics at the University of Geneva with a joint affiliation at the Swiss Light Source. His group uses laser-based angle resolved photoemission to study correlated metallic states, oxide interfaces and van der Waals heterostructures.