k-resolved electronic structure by soft-X-ray ARPES: From threedimensional systems to buried heterostructures

V.N. Strocov

Swiss Light Source, Paul Scherrer Institute, CH-5232 Villigen-PSI, Switzerland

A fundamental benefit of pushing the ARPES experiment into the soft-X-ray energy range is the photoelectron escape depth increasing by a factor of 3-5 compared to the conventional VUV-ARPES. This gives a boost to bulk sensitivity as well as enables access to buried heterostructures. However, until recently the spead of soft-X-ray ARPES (SX-ARPES) has been severely impeded by a loss of the valence band photoexcitation cross-section by a few orders of magnitude compared to the VUV energy range. This problem has recently been solved at Swiss Light Source with the new SX-ARPES facility at the ADRESS beamline [1] which delivers X-ray radiation with variable polarizations in a photon energy range from 300 to 1600 eV with routine energy resolving power $E/\Delta E$ up to 20000. High photon flux topping up 10¹³ photons/s/0.01%BW combined with small spot size and grazing X-ray incidence angle experimental geometry has not only overpowered the notorious cross-section problem but also enabled access to buried heterostructures despite the photoelectron attenuation in a few monolayers thick overlayers.

Applications of SX-ARPES to 3-dimensional (3D) materials are based on the fact that the increase of the

photoelectron escape depth, by the Heisenberg uncertainty principle, improves intrinsic definition of surface-perpendicular electron momentum. One of our first application examples was the paradigm transition metal dichalcogenide VSe₂ [2]. The experimental 3D band structure and Fermi surface (FS) of this material taken in a photon energy range from 885 to 945 eV (Figure) demonstrate a textbook clarity achieved by virtue of freeelectron final states, their sharp definition in 3D momentum and smooth atomic-like photoemission matrix elements delivered in the soft-X-ray energy range. An intriguing feature of VSe₂ is that despite its layered quasi-2D structure this material develops 3D charge density waves (CDWs). Autocorrelation analysis of the experimental FS has revealed its pronounced out-of-plane nesting which acts as the precursor of these exotic CDWs [2]. Other examples of applications of SX-ARPES to 3D materials include polarization dependence and alternating FS shapes in pnictide



HTSCs, demonstrating their 3D character and intra-cell interference effects, bulk Rashba spin splittings in noncentrosymmetric topological insulator BiTeI [3], etc.

The high brilliance of the ADRESS beamline has enabled the move from bulk materials to buried heterostructures. We illustrate this by resonant SX-ARPES of 2D electron gas at buried LaAlO₃/SrTiO₃ interfaces, which unveils different subbands and FS sheets formed by the interface states. Increase of temperature suppresses the momentum selectivity and allows depth profiling of the 2D electron gas from variations of the SX-ARPES signal with emission angle [4]. Another example is resonant spectroscopy of Mn magnetic impurities in the paradigm magnetic semiconductor GaMnAs, which reveals the energy position of the ferromagnetic Mn impurity band and mechanism of its hybridization with the 3D host GaAs bands [5]. These results have immediate implications for the origin of ferromagnetism in GaMnAs. The whole body of our unfolding results demonstrates an immense potential of SX-ARPES to deliver a clear **k**-resolved picture of electronic structure from 3D materials to buried heterostructures and impurities.

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