

# **Diamond-II Flagship Beamline Proposal New nano-ARPES beamline**

**Prepared for Diamond SAC/DISCO  
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## 1. Acknowledgements

This document has been written by the following members of the User Working Group and Diamond Staff, with input from the broader community. We are grateful to the whole team for their time and effort to complete this proposal.

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User Working Group Meetings – 4<sup>th</sup> September 2020 and 5<sup>th</sup> November 2020

User Engagement Webinar (118 participants) – 9<sup>th</sup> November 2020

## 2. Executive Summary

Diamond Light Source (DLS) has built a reputation as a centre of excellence for research on Quantum Materials, by developing world-leading instrumentation and expertise in angle-resolved photoemission spectroscopy (ARPES). This proposal builds on that success and will enable DLS to remain at the forefront of ARPES research for the next 15 years, in turn helping drive a materials revolution for industry, with new schemes such as 2D topological transistors providing fast, energy efficient electronic devices.

Whilst the current I05 beamline has a nano-ARPES capability, it is compromised by the design of the main high-resolution (HR) branch. The newly-available long straight section I17 provides the only opportunity for the UK to develop a world-leading nano-ARPES beamline. The higher brightness of Diamond-II, with an optimised two-branch beamline design, will deliver increased flux to the sample, opening possibilities to explore and discover new electronic phenomena at the nanometre scale:  *$\mu$ -ARPES* will deliver high-performance ARPES with spectral resolution approaching that of the world-leading I05-HR branch, but with probing area **>7500×** smaller ( $\approx 800$  nm beam); *nano-ARPES* maintains the flux used for typical operation on the existing I05 nano-ARPES, but with the beam size reduced to 100x84nm (**45×** smaller than at present), with an ultimate spatial resolution of  $\approx 50$  nm. The transformative impact is highlighted schematically in Fig. 1, section 3.

By dramatically expanding the applicability and operation regimes of ARPES, this project will develop the user base and enable new science. It will also address significant oversubscription for beamline I05 (average 3.9 $\times$  - last five allocation periods), and provide possibilities for developing spin-ARPES and laser-ARPES at DLS, to maintain a world leading position. The nano-ARPES and I05 beamlines will be highly complementary, with optimised layouts and end stations to enable cutting edge science from the world's leading research groups, including the growing UK user base.

### 3. Scientific case

#### 3.1 Introduction

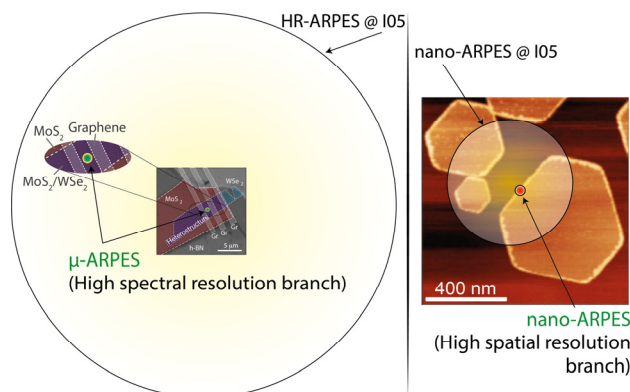
The electronic band structure of a solid underpins its optical, electrical, and thermodynamic properties. As such, electronic structure measurements are of crucial importance to spur the discovery, optimisation, and design of new materials for a myriad of applications. Angle-resolved photoemission spectroscopy (ARPES) provides the most direct way to measure the momentum-resolved band dispersions in solids, and offers key insights on the quantum many-body interactions that are at the heart of many of their useful physical properties. ARPES has consistently proven itself to be one of the most influential experimental techniques in solid state physics over the past decades: from pioneering studies establishing unconventional order parameter symmetries and a myriad of complex ordered phases in high-temperature superconductors,<sup>1</sup> through the observation of the famous relativistic Dirac electronic structure of graphene,<sup>2</sup> to the discovery of a host of topologically non-trivial states of matter that have revolutionised modern solid state physics.<sup>3</sup> However, the beam size available for traditional synchrotron-based ARPES has made measurements reliant on the preparation of atomically-clean mm-scale uniform surfaces, substantially limiting the range of materials systems which can be investigated. Recent advances in optics for focussing vacuum ultraviolet (VUV) light to sub-micron length scales, coupled with the increased coherence and brightness enabled by the Diamond-II upgrade, open a unique opportunity to develop a state-of-the-art beamline dedicated to spatially-resolved ARPES.

We propose the creation of a beamline optimised for spatially-resolved ARPES use in the core energy range of 50-250 eV, and extending up to  $\approx 800$  eV to enable bulk-sensitive and elementally-resolved photoemission studies, with two interconnected end-stations.  $\mu$ -ARPES will be optimised for spatially-resolved ARPES with a sub-micron probing region and good spectral resolution. Capillary optics will deliver high photon flux, while an optimised sample environment will enable high-performance ARPES on small samples and domains. *nano*-ARPES will exploit zone-plate x-ray nano-optics for a  $< 100$  nm beam size, and will be optimised for novel spectro-microscopic studies. Together with Diamond's world-leading I05 beamline, this facility will create a unique world-wide capability covering a broad range of spatial (see Fig. 1) and energy scales, dramatically expanding the applicability of ARPES, and impacting on multiple fields of solid state physics and materials research in which the UK currently has internationally-recognised strengths.

#### 3.2 Science enabled by project

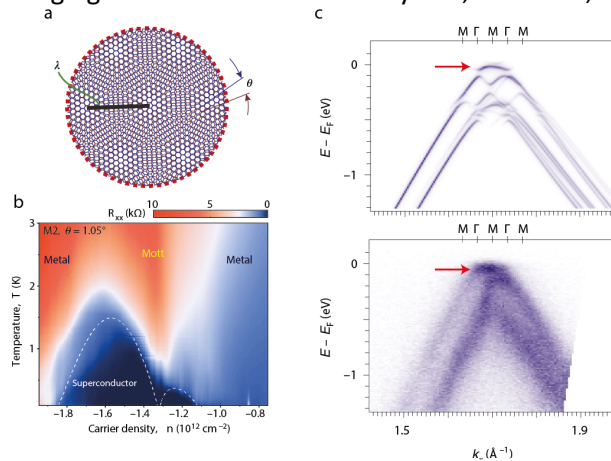
##### 3.2.1 2D Materials and Devices

Mechanical exfoliation, brought to prominence by the isolation of graphene (Geim/Novoselov, Manchester, Nobel Prize 2010) and now utilised by numerous groups in the UK and worldwide, has created new opportunities for fabricating novel electronic "meta-materials" by stacking single atomic layers of disparate materials systems together into heterostructure geometries, with entirely new tuning parameters such as the twist angle between layers.<sup>4</sup> A wide variety of 2D materials are now being explored: transition-metal dichalcogenides, for example, host a diverse set of novel charge-ordered, superconducting, and topological states, while their semiconducting phases show enormous promise for atomically-thin transistors and optoelectronic devices. Magnetic order has been shown to persist to the monolayer limit in several halides and phosphides, adding magnetism to the 2D materials toolbox, paving the way for atomically-thin spintronics,<sup>5</sup> while stacking 2D materials together offers almost limitless opportunity to control their electronic structures, optical properties, and quantum many-body states.<sup>6</sup>



**Fig. 1:** Comparison of beam size at the proposed nano-ARPES facility and I05, overlaid on sample images (left) MoS<sub>2</sub>/WSe<sub>2</sub> heterostructures<sup>44</sup>; (right) TiSe<sub>2</sub> islands<sup>28</sup>.

ARPES – as a surface sensitive technique – is in principle ideally placed to have a massive impact on the study of 2D materials. However, mechanical exfoliation typically results in samples only a few micrometres in size, necessitating measurements using focussed light spots. Existing spatially-resolved ARPES instruments have had several major early successes in this field, mainly focussed on graphene and the semiconducting transition-metal dichalcogenides, revealing carrier-driven band renormalizations,<sup>7</sup> characterising effective masses and band offsets,<sup>8</sup> and imaging band hybridisation from moiré superlattice formation.<sup>9</sup> Many current studies are, however, limited by the available spectral resolution. The pressing need for improvements is clearly demonstrated by pioneering results on twisted bilayer graphene (TBG). Hybridisation between the Dirac dispersions of the two layers has been predicted to form extremely flat bands, and exciting recent results have demonstrated the emergence of a host of correlated insulating, superconducting, and magnetic states at low twist angles,<sup>10,11</sup> marking this as one of the most exciting systems of the last decades of research in quantum materials (*e.g.* award of the Buckley Prize 2020, Wolf Prize 2020). ARPES is unique in its ability to directly measure the electronic structure underpinning the phenomenology of these systems, and initial studies<sup>12,13</sup> have already revealed the presence of flat bands emerging at the Fermi level. They are, however, limited by available spectral resolution. Instrumental



**Fig. 2:** (a) Moiré superlattice in twisted bilayer graphene. (b) Rich doping-dependent phase diagram in ‘magic-angle’ graphene.<sup>11</sup> (c) Calculated (top) and measured (bottom) electronic structure from nano-ARPES.<sup>13</sup>

improvements are required to enable studying the correlated states on their relevant energy scales, and more generally to enable distinguishing band features in a whole host of multi-band 2D materials, to search for the influence of spin-orbit coupling and interlayer interactions, *e.g.*, in hybridising states, and to open the door to studies of low-energy physics in charge density wave and superconducting 2D materials.

The proposed  $\mu$ -ARPES beamline will unlock enormous potential for electronic structure studies in this field. Combined high flux and spectral resolution and an optimised sample environment will enable not only the study of gross band features of semiconducting systems, but also probing electronic states on lower energy scales, at lower temperatures, and to make better links with transport and optical probes of 2D material heterostructures. Given the wide parameter space available (*e.g.*, constituent materials, stacking sequence, twist angle), rapid feedback on the resulting dependencies in the electronic structure is urgently required. The use of capillary mirrors, combined with extensive sample preparation capability, will dramatically increase throughput in  $\mu$ -ARPES, whilst providing a spot size of relevant dimensions for typical exfoliated samples. True nano-ARPES capability in the zone plate branch will in turn enable studies of spatial inhomogeneity within the flake, study of individual magnetic domains in 2D magnets, optimised performance for *operando* measurements (*see* 3.2.2), and open up possibilities to study the newest materials that are typically isolated as only small-scale flakes.

**Key new science enabled:** Characterising electronic states in designer 2D material heterostructures; probing single magnetic domains in 2D magnets; studying correlated states in 2D quantum materials. **Key technical drivers:**  $\mu$ -ARPES: high flux for high throughput and improved energy resolution, large photon energy range and controllable linear polarisations allowing elemental and orbital-specific studies of electronic structure. Nano-ARPES: <100 nm spatial resolution for spatial studies within a flake, to study intrinsic inhomogeneity and to find optimal clean sample patches, and to probe the smallest flakes. **Sample preparation:** *In situ* annealing and plasma treatments, integrated glove box.

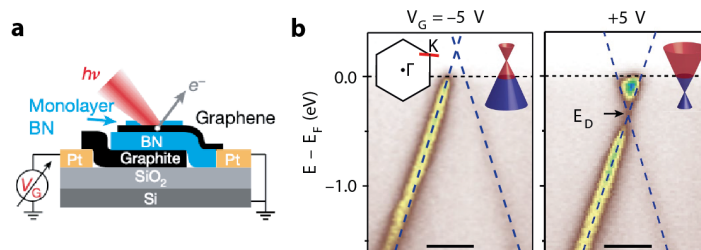
### 3.2.2 Operando measurements

Whilst ARPES is traditionally carried out using a well-grounded sample, increasing efforts are being made to perform ARPES measurements out of equilibrium. A conceptually simple, yet only recently feasible,<sup>14</sup> example is in the use of a gate voltage to bias a sample of graphene, changing its chemical potential and thereby the carrier density. Gating also allows tuneable access to the usually unoccupied conduction band states in semiconductors, ultimately probing the states underpinning electronic device operation and providing information highly complementary to that obtained from transport and optical experiments. Performing ARPES while passing a current through the sample provides another route to manipulate the underlying electronic system, and is of interest, for example, in controlling metal-insulator transitions,<sup>15</sup> suppressing superconductivity,<sup>16</sup> and for electrical switching of magnetic moment orientation.<sup>17</sup> Small probing areas ensure the measured spectra are less affected by broadening due to voltage drop across the light spot and inhomogeneous fringe fields. The ability to probe a small sample area makes possible the study of new device architectures by *in operando* ARPES, including those fabricated from exfoliated crystals of 2D materials operated using ARPES-compatible back-gate geometries, lithographically-defined thin film devices,<sup>18</sup> and bulk samples structured into device geometries using focused ion beam techniques,<sup>19</sup> enabling combined ARPES and transport measurements.

Additional *operando* parameters can also modify the electronic structure. ARPES studies with static strain applied mechanically indicate that this can drive metal-insulator transitions,<sup>20</sup> mediate Fermi surface topological Lifshitz transitions,<sup>21</sup> detwin multi-domain samples,<sup>22</sup> and manipulate topological states.<sup>23</sup>  $\mu$ -ARPES from small samples under tuneable *in situ* strain could be enabled via piezo-based technology, as has proved instrumental in uniaxial strain studies with transport and scattering probes.<sup>24,25</sup> The smaller sampling area of nano-ARPES would enable, amongst many others, probing of spatially-inhomogeneous strain fields induced by sample bending, probing of local regions where strain-induced gauge fields can mimic magnetic fields  $> 300$  T,<sup>26</sup> and investigations of how local strain variations influence electronic properties in materials which have been plastically-deformed. Such studies are of major fundamental and applied interest.

**Key new science enabled:** *Operando* ARPES of devices; combining ARPES and transport; ARPES of metastable and non-equilibrium phases; realising novel strain tuning of electronic states.

**Key technical drivers:** Sample manipulators with integrated contacts; glove box with wire bonding capability; offline electrical testing rigs.

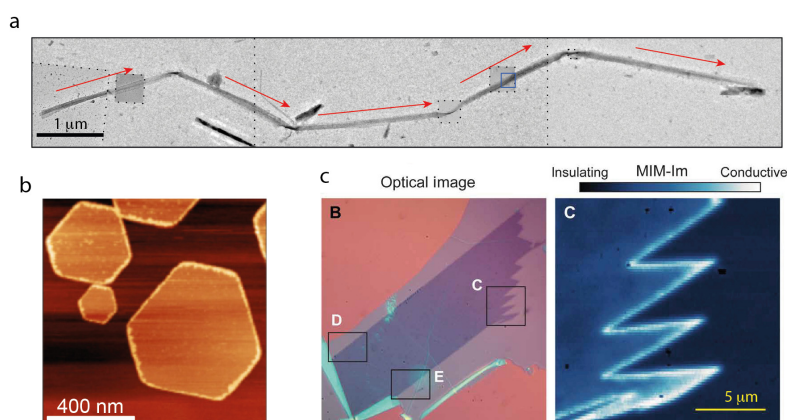


**Fig. 3:** Electrostatic gating in ARPES. (a) Schematic of a graphene heterostructure device. (b) ARPES measurements show evolution of the band filling as a function of applied gate voltage.<sup>14</sup>

### 3.2.3 Nanomaterials & nanoscience

The new nano-ARPES capability, combined with efficient electron detection schemes, will allow valence band mapping of truly nanostructured materials. These include nanowires and nanoribbons, which are often investigated for their combination of excellent transport and mechanical properties, and will become measurable by ARPES for the first time, as well as arrays of organic molecules which form central elements for organic electronics. Varying dimensionality, periodicity, and feature size can have a dramatic influence on electronic structure.<sup>27</sup> Nano-ARPES will provide a powerful probe of how the nanostructured materials' band alignments, substrate interactions, and many-body effects are influenced by their changing geometry. Another unique capability will be the ability to investigate the electronic structure of individual islands of monolayer samples grown epitaxially via techniques such as molecular-beam epitaxy or chemical vapour





**Fig. 4:** (a) Individual phosphorene nano-ribbons<sup>27</sup>; (b) epitaxial islands of TiSe<sub>2</sub> monolayers<sup>28</sup>; (c) optical image (left) and scanning microwave impedance mapping (right) of WTe<sub>2</sub> exfoliated flakes<sup>29</sup>; signatures of the edge states of the quantum spin Hall insulator are apparent.

deposition. Typically, the as grown samples contain domains of sub-micron size,<sup>28</sup> which can often be misoriented relative to one another, necessitating methods to probe inside a single island. Samples deliberately patterned at the nanoscale, *e.g.* for optimised optical or plasmonic performance, or which contain catalytically-active clusters, will become accessible for study by ARPES. Direct detection of edge states in topological materials<sup>29</sup> will also move into reach, enabled by ultimate spatial resolution. While some features of interest will inevitably remain below even this spatial resolution, specially designed samples – *e.g.*, where multiple interfaces, edges or boundaries are located within the probing light spot – can be used to boost the signals.<sup>30</sup> Moving ARPES into the realm of spot sizes of a similar order to the relevant spatial scale of interest in the material, as proposed here, will make it possible to obtain sufficient signal from individual nanowires, epitaxial interfaces (studied “side-on”) and edge boundaries, in turn opening up entirely new fields of research in electronic structure studies on the nanoscale.

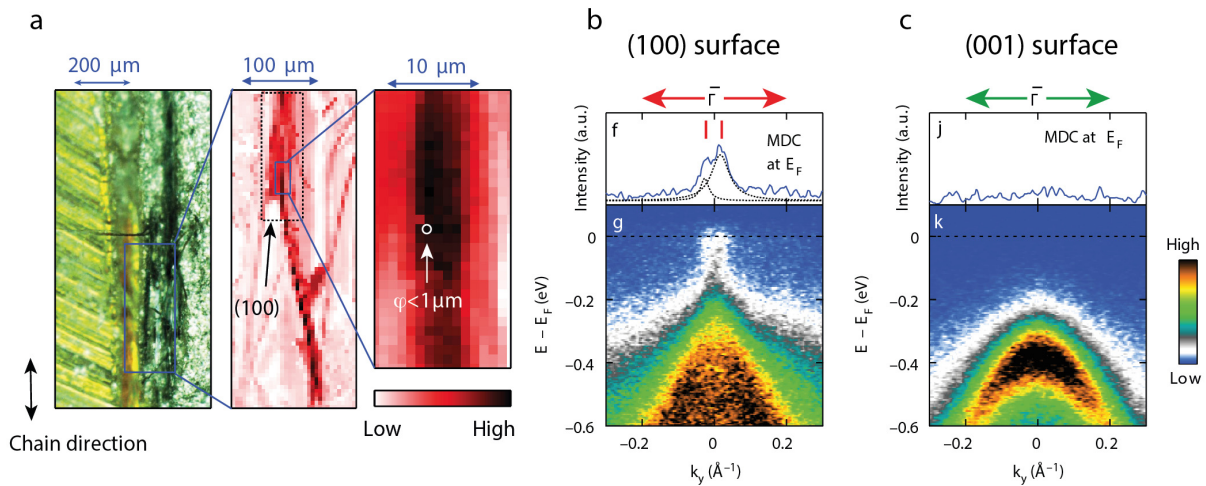
**Key new science enabled:** Imaging electronic structure of individual nanotubes; studying surface molecular systems; probing interface states in heterostructures and edge states of topological materials.

**Key technical drivers:** Nano-ARPES: Ultimate spatial resolution; efficient photoelectron detection schemes for fast parallel measurements (band and Fermi surface mapping); stable sample manipulation.

### 3.2.4 Quantum Materials Exploration and Understanding

ARPES measurements of large-area single crystal samples have played a critical role in the development of the fields of **topological** and **strongly-correlated electronic matter**, but it is increasingly necessary to move beyond this established approach to remain at the forefront of research in these fields. The discovery of novel electronic materials tends to occur in polycrystalline or powder samples, with volume probes such as resistivity or magnetisation employed to search for signatures of interesting electronic or magnetic states. Single-crystal samples are difficult to synthesise while some materials simply do not form as large single crystals. Many interesting materials do not cleave well (the de facto standard surface preparation method); and mixed surface terminations often complicate interpretation of the measured data. These challenges represent a strong roadblock to the application of ARPES to an active materials discovery economy.

The proposed beamline, probing much smaller regions of interest, would enable selective measurements from polycrystals or small single crystal samples as they first become available, or for which the desired cleavage plane is not naturally obtained. A recent success in this regard is the first verification of the weak topological insulator using the existing nano-ARPES setup at Diamond’s I05 beamline.<sup>31</sup> Bi<sub>4</sub>I<sub>4</sub> was predicted to host topologically-protected surface states only for specific surface orientations which are not natural cleavage planes. Spatially-resolved photoemission allowed access to minority surface regions that host the desired orientation, verifying the surface orientation-dependent formation of topological surface states. However, improved energy and spatial resolution are clearly required to study the topological states in detail. We expect higher throughput  $\mu$ -ARPES measurements to strongly advance the active materials



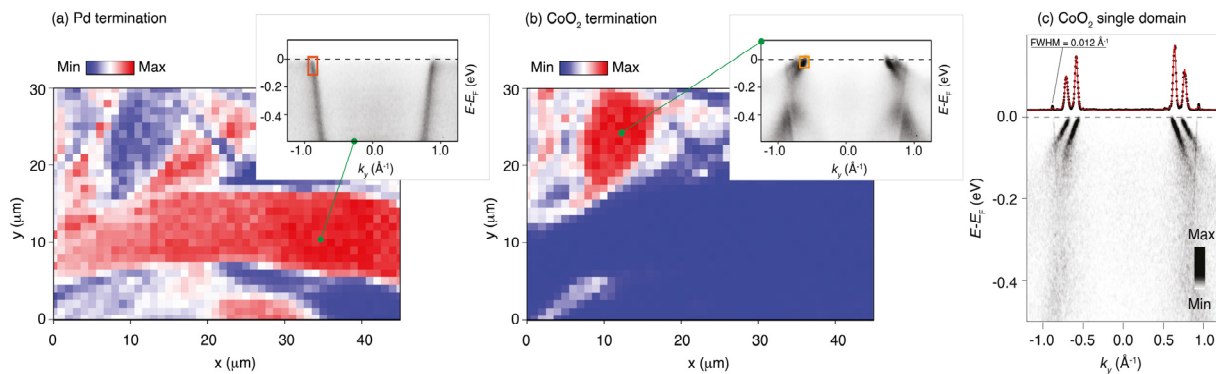
**Fig. 5:** (a) Optical micrograph of a cleaved sample of  $\beta$ -Bi<sub>4</sub>I<sub>4</sub>, and corresponding photoemission intensity map.<sup>31</sup> (b) Micro-ARPES measurements taken from different surface terminations exposed at different spatial positions of a cleaved surface, enabling observation of surface orientation-dependent formation of topological surface states.

discovery effort in topological solids, including in materials not previously amenable to ARPES, while nano-ARPES would allow much sought-after identification of higher order topological phases and enable the highest-quality measurements from small patches of desired surface orientation to be obtained, together opening up a wide array of new systems for experimental electronic structure study.

Existing spatially-resolved ARPES instruments have until recently been hampered by a severe trade-off between achievable spatial resolution, flux, and energy resolution, making it challenging to explore the small energy scales that are characteristic of correlated electron systems and largely restricting its use to the study of weakly-interacting solids. Despite this, pioneering nano-ARPES measurements have shown how the electronic structure of the high- $T_c$  superconductor YBCO shows a strong variation with surface termination,<sup>32</sup> while preliminary measurements from PdCoO<sub>2</sub> surfaces (see Fig. 6) indicate how improved linewidths can be obtained from single-domain regions when imaged using spatially-resolved ARPES.<sup>33</sup> This points to the potential of spatially-resolved ARPES for quantitative studies of quasiparticle dynamics, if energy resolution and sample temperature can be improved. The combined high flux and good spectral resolution, in combination with optimised sample environment, of the proposed  $\mu$ -ARPES end station promises transformative advances in this respect for the study of correlated materials.

An optimised spatially-resolved ARPES capability will also yield countless opportunities to probe intrinsically spatially-inhomogeneous electronic structures. Correlated electron systems nearly always form structural or magnetic domains when undergoing phase transitions. Micro and nano-ARPES measurements will enable resolving the spectral function individually from within a domain,<sup>34</sup> as well as mapping local electronic inhomogeneity,<sup>35</sup> with high energy resolution enabling gap measurements and quantitative treatment of many-body interactions. Furthermore, by sculpting samples with focussed ion beam approaches, it will be possible to cleave small patches of the sample along unfavourable planes, or to develop a cleaved surface in a polycrystalline sample, opening up new possibilities for probing the electronic structure of three-dimensional correlated and topological solids. The highest proposed photon energies available at the beamline will enable tighter focussing, allowing greater spatial resolution for studies of such electronic inhomogeneity, as well as enhanced bulk sensitivity. This promises to enable bulk-relevant photoemission studies on the nanoscale, ensuring its relevance well beyond surface phases. An interesting possibility in this respect is in the analysis of “combinatorial” materials, whereby a material’s composition gradually evolves across a wafer, as well as spatial profiling of band bending potentials in semiconducting nanowires, of interest, for example, as hosts of Majorana qubits.<sup>36</sup> Spatial-dependent studies using micro- and nano-





**Fig. 6:** ARPES of PdCoO<sub>2</sub> (@105-nano)<sup>33</sup>. (a,b) Spectro-microscopy of Pd- and CoO<sub>2</sub>-terminated surfaces indicate a rich spatial-dependent electronic structure. (c) A single CoO<sub>2</sub>-terminated domain yields extremely sharp linewidths.

ARPES would allow rapid screening of how materials properties vary with composition and position, enabling access to a wide phase space of electronic materials, with relevance extending far beyond the study of correlated and topological materials, from energy systems to photocatalysts, and cementing spatially-resolved ARPES as a core technique within the materials discovery pipeline.

**Key new science enabled:** Accelerating “guided” materials discovery and design; characterising new states of topological matter; new insights on the high- $T_c$  and quantum many-body problem.

**Key technical drivers:**  $\mu$ -ARPES: <10 meV energy resolution @ 100 eV, sample temperature < 10 K for spectral function measurements; high flux for high throughput; photon energies from 50 to 800 eV to allow high momentum resolution and increased depth sensitivity, respectively. **Nano-ARPES:** < 20 meV energy resolution @ 100 eV to resolve low-energy states and topological states in moderate band gaps; <100 nm spatial resolution for probing electronic inhomogeneity and small domains/cleave orientations. **Sample preparation:** *In situ* cleavage; *in situ* annealing; transfer to growth tools in Diamond and external user labs for studying *e.g.* oxide heterostructures.

### 3.2.5 ARPES beyond the dipole approximation

The combination of an improved coherent fraction of the Diamond-II beam and advances in x-ray zone plate optics opens up the possibility to exploit wavefront shaping – long since used in conventional optics<sup>37</sup> – for novel applications in photoemission. An obvious application is the generation of probing light with finite orbital angular momentum ( $\pm\hbar, \pm 2\hbar, \dots$ ), to allow optical transitions in the photoexcitation process beyond those allowed within the dipole approximation. This would open new possibilities for resonant ARPES and x-ray absorption spectroscopy (*e.g.* allowing *s-d* or *p-f* transitions) with application in the study of transition metal oxides and heavy fermion systems, as well potentially providing new routes to probe spin and orbital angular momenta of the initial state wavefunctions in ARPES. Additional more complex schemes could be envisaged, for example generating higher-order Hermite-Gauss modes with multiple closely-spaced intensity maxima in the beam profile, opening up the possibility for spatially-resolved phase-sensitive experiments in ARPES. Ultimately, developing a state-of-the-art zone-plate based nano-ARPES system at Diamond-II will ensure that it is well placed to profit from continuing advances in x-ray nano-optics, and the novel capabilities that these will provide for future photoemission studies of complex materials.

**Key new science enabled:** Modified transition-matrix elements in ARPES, allowing new insights into atomic, spin and orbital character of states; exploiting interference in the photoemission process.

**Key technical drivers:** nano-ARPES: Advanced zone-plate optics, benefitting from the improved coherence of the Diamond-II storage ring; high spatial resolution and stable sample handling.

### 3.2.6 Core benefits from Diamond-II

The improved horizontal emittance of Diamond-II will be instrumental to achieving tighter focus as required for a state-of-the-art nano-ARPES setup, while the greater coherent fraction will enable new possibilities for x-ray nano-optics for photoemission. The increased brightness, in particular at higher photon energies, will enable the beamline to operate from the VUV into the soft x-ray regime, enhancing the range of materials systems that can be studied, enabling greater depth sensitivity, and allowing for spatially-resolved resonant photoemission, gaining elemental sensitivity. Moderate transmission through zone plates makes this an extremely photon-hungry technique. The major science gain from the proposed setup comes as much via improved flux (enabling higher spectral resolution) as improved spatial resolution. Moreover, decreasing *spectral* resolution to gain flux simultaneously results in loss of *spatial* resolution due to a dominant contribution of achromatic aberrations. Installation on a mid-straight would lead to 5.6x lower flux. For nano-ARPES, this would lead to an ultimate spot size at least 4x larger than proposed here, strongly compromising the science case for the study of nanomaterials, edge states, and spatial inhomogeneity. Similarly, the loss of flux would lead to a reduction in achievable spectral resolution for  $\mu$ -ARPES, compromising the high-performance ARPES at the heart of the new science enabled by this branch line. Thus, ***installation on the long straight section is strongly desired.***

### 3.2.7 Complementarity

A dedicated spatially-resolved ARPES beamline within Diamond-II will open numerous opportunities for synergistic developments across the Harwell campus. ***Across Diamond***, it will promote combined real-space probing of materials and their electronic structure using spatially-resolved ARPES, with: chemical/magnetic real-space imaging using photoemission electron microscopy (PEEM); spatially-resolved structural characterisation via micro- (I18) and nano- (I14) x-ray diffraction, well matched to the spatial scales of micro- and nano-ARPES, respectively; and local electronic and valence mapping in micro-XAS (I06, I18), ultimately enabling a holistic approach to elucidating structure-property relations. A particularly appealing prospect is in combined studies with the planned spatially-resolved RIXS upgrade (I21). RIXS has emerged as a powerful tool for studying collective excitations of energy and quantum materials, and is an ideal complement to information on the one-electron removal spectral function obtained by ARPES. Correlating spatially-resolved ARPES and RIXS promises new insights on emergent electronic inhomogeneity and on the interplay of competing phases in quantum materials – one of the most important questions in modern solid-state physics. Capabilities for sample transfer between the beamlines, using vacuum suitcases where needed and ensuring compatible sample handling requirements, will be carefully discussed during the technical design phase, to enable the goal of gaining complementary insight from multiple techniques on a single sample.

***Across campus:*** The unique contrast mechanism in spatially-resolved ARPES motivates combined studies with well-established imaging and microscopy techniques. There is a significant opportunity for combined studies using the ePSIC electron microscopy facility. Further, the additional capabilities for high-resolution ARPES at I05 will ensure a wide coverage of length and energy scales for ARPES, while the extra capacity that will be realised on I05 by establishing spatially-resolved ARPES on its own beamline (see Section 3.6) will provide further possibilities for developing cross-campus links, for example through the realisation of capabilities for spin-resolved ARPES – with links to magnetic characterisation (*e.g.* I06/I10/I16/I21 and ISIS) – and for strengthening links with the Central Laser Facility for non-equilibrium studies.

There are also numerous opportunities for collaborative ***technical developments*** with other beamlines, which in many cases share similar sample handling constraints and some common detection requirements for spatially-resolved experiments. For example, I05, I09, and I21 all currently share a common low-temperature sample manipulator, designed as part of the development of the high-resolution branch of I05; there is potential for co-development of new sample manipulation schemes with *e.g.* I21, dedicated for low-temperature nano-imaging spectroscopy experiments such as nano-ARPES/RIXS. In both cases, the final

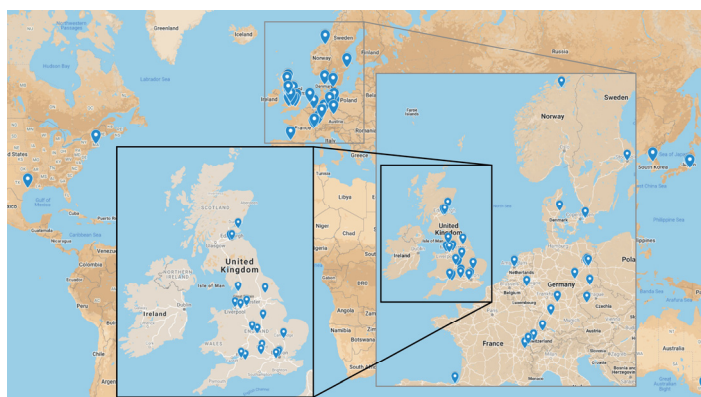
measurement can be time consuming, raising potential for co-development of secondary detection schemes (e.g. fluorescence mapping) for initial spatial mapping/focussing, as well as the incorporation of complementary probes (e.g. Raman or PL spectroscopy). Collaborations on scan time optimisation (e.g. synchronous data recoding with sample movement) can be envisaged with all nano-imaging beamlines.

### 3.3 Academic user community and beneficiaries

The research enabled by spatially-resolved ARPES is core to several thematic areas outlined in the ‘Diamond-II Science Case: Advancing Science’, namely *Quantum Materials*, *Energy* and *Chemistry and Catalysis*, and lies at the heart of numerous UKRI and UK government strategic priorities. It is thus squarely aligned with the research objectives of the UK community. Studies using spatially-resolved ARPES will advance understanding across a broad spectrum of *Advanced Materials* with potential application from efficient electronic devices to solar cells, underpinning *key studies to deliver a productive, connected, and resilient nation* in line with the EPSRC Delivery Plan. It will enable research on 2D materials, materials for energy-efficient ICT, and will facilitate understanding from Atoms to Devices, all core themes of the UK’s Henry Royce materials institute, and will in turn help to drive *energy security and efficiency* in line with the UK Industrial Strategy. Studies of a broad range of materials, including combinatorial studies, will provide core feedback to enable “dialling up a desired property using new principles” in EPSRC’s *Nanoscale design of functional materials* grand challenge, to develop *Materials of the future*, while probing edge states of topological materials promises critical insight for developing spin-based *quantum technologies*.

The international community in ARPES is huge, and growing rapidly, with groups at many of the major universities in Europe, the US, and Asia. Within the UK, there is a strong community of researchers whose activity is focussed, almost entirely or in part, on studying the electronic structure of solids. This includes core users of high-resolution ARPES, those who use high-resolution ARPES alongside other materials probes (e.g. quantum oscillations, crystal growth, structural and magnetic measurements), and early pioneers of nano-ARPES and *operando* studies. With the new possibilities that arise from performing ARPES with improved spatial resolution, the existing UK community and other international leaders in the field wish to make heavy use of the new beamline (see Section 7). The success of I05 has undoubtedly played a major role in growing the UK community, and further natural expansion of this can be expected in the coming years as a crop of talented and experienced early career researchers find independent positions at UK universities (see support letters). A similar catalysing effect on a related but partially-distinct community can be expected through the development of a dedicated nano-ARPES beamline. The UK, for example, has a particularly large and active community in the science and fabrication of 2D materials (e.g., the National Graphene Institute and National Physical Laboratory have both expressed their support for the proposed beamline), and in microscopic characterisation of materials, who will naturally be attracted by the spatial resolutions that will be achieved. The possibility to perform measurements on a much broader array of materials than the “perfect” single crystals that have been the mainstay of ARPES up to now will further makes the beamline attractive to solid state chemists and materials scientists.

The breadth of support from both existing and new future users is evident in supporting statements for the proposal (Fig. 7 and Section 7). It presents a timely opportunity to establish the UK as a leader in the nascent technique of nano-ARPES. Diamond has a strong in-house research effort in ARPES;



**Fig. 7:** UK and world-wide distribution of support statements from current and prospective future users of nano-ARPES.

active collaborations with this group will be an effective way to encourage users whose traditional expertise is in *e.g.* materials physics/chemistry, device properties, or microscopy to start using nano-ARPES, and to benefit from the unique insights which it can deliver. To this end, the beamline would plan to appoint joint PhD students with groups not traditionally part of the core ARPES community. There are several lab-based photoemission facilities available for this user community to access across the UK, including the nano-ESCA facility in Bristol and a laser-spin-ARPES facility being developed in St Andrews. These offer complementary capabilities to those proposed here and routes to pre-beamtime sample study, and with ARPES@Diamond help to provide a strong community hub for UK-ARPES. We will organise joint workshops across these facilities, advertising nano-ARPES to a broad potential user base, and providing training to new users.

### 3.4 Industrial user community and beneficiaries - impact on UK PLC

ARPES is an enormously powerful tool for fundamental studies with a focus on the discovery of new phenomena in novel materials. Whilst there has been huge scientific development and general maturity of the technique over the last decades, the majority of studies would be classified as Technology Readiness Level 0 – 1. It is clear, however, that excellent fundamental studies of new materials by ARPES provides foundational information underpinning industrial development of electronic and spintronic schemes of the future. Indeed, we are heading for a looming catastrophe in silicon-based electronics: conventional size-scaling is reaching its limits, clock frequency is no longer increasing, and projections put ICT demands at more than 20% of global electricity consumption by 2030.<sup>38</sup> Given the timelines involved in development of entirely new mechanisms for technology applications, the fundamental science that will underpin these must be prioritised now. In this respect, the spatially-resolved ARPES studies that will be enabled by the new beamline can be expected to have long-term benefit also to the industrial community. At the same time, the ability to move away from the traditional “perfect” crystals, as well as the possibility to study the electronic structure evolution of simple devices (*e.g.* transistors, capacitors) under their conditions of operation (see Section 3.2.2) moves the enabled ARPES studies closer to practical regimes.

### 3.5 Comparison to other facilities, current and planned

A realistic aim of the proposed beamline is to create the leading facility for spatially-resolved ARPES measurements in Europe, and a system with unique capabilities not found anywhere worldwide. The few existing and planned spatially-resolved ARPES capabilities are summarised in Table 1. Existing nano-ARPES facilities in Europe use zone plate or Schwarzschild optics, delivering moderate spatial resolution (sub-micron) but with low flux and thus strongly compromised spectral resolution. A new beamline currently under development at

Beamline	Spot size (nm)	Energy range (eV)	Instrumental/Sample environment
Proposed nanoARPES I17 @ Diamond	<100 (ult. 50)	50 - 800 (opt. 50 - 250)	6 d.o.f., <25K (zone plate), <10K (high flux capillary)
I05-nano @ Diamond (UK)	620	50 - 100	~35 K, 5 d.o.f.
Spectro-microscopy @ Elettra (Italy)	1000	Discrete 27 or 74	>40 K, small rotatable analyser
ANTARES @ SOLEIL (France)	>120	95 - 1000	>50K, 5 d.o.f, fixed position analyser
MAESTRO @ ALS (USA)	120	100	Capillary-mirror + Zone-Plate, room temp
ASTRID2 (Denmark, in development)	1000	opt. 15-70	Capillary-mirror, >30 K

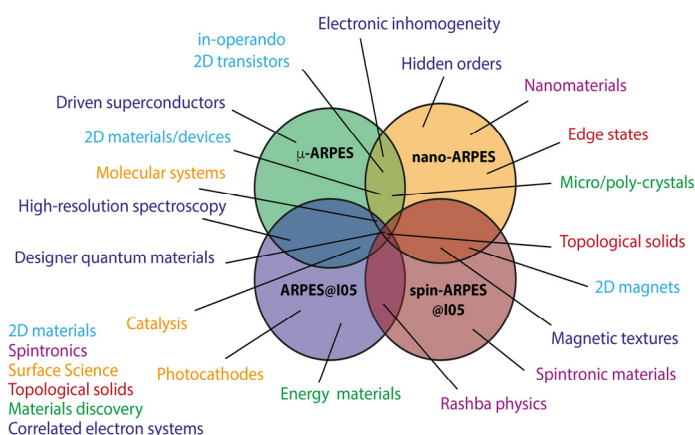
**Table 1:** Typical operating parameters of nano-ARPES facilities.

ASTRID, Denmark, will use capillary-optics, but operating on a small ring will not be able to deliver the flux or photon energy range to be achieved here. No existing spatially-resolved ARPES capabilities are set up with an optimised sample environment for low-temperature (<10 K) measurements and high energy



resolution  $<10$  meV @ 100 eV. Thus, the  $\mu$ -ARPES capability proposed here will be the first of its kind, a true next-generation instrument offering high-quality ARPES from a sub-micron spot. There is also the possibility to set a new standard in Europe for spatial resolution with nano-ARPES. The ALS currently delivers the world-leading capability in this respect, with a spot size of 120 nm, with an aspiration to achieve  $<10$  nm at ALS-U. The extremely low planned emittance there makes it possible to achieve tighter focussing than is possible here. Nonetheless, the system proposed here will still deliver an extremely competitive spatial range for nano-ARPES, while the slightly less tight constraints on beam focus (and hence end station stability, thermal drift etc.) will enable placing a higher priority on sample environment – the new beamline will thus be world leading for nano-ARPES at cryogenic temperatures. As ARPES is sensitive to quasiparticle lifetimes, both small spot and excellent sample conditions (including low temperature) are essential to many of the innovative science areas outlined above. Such measurements are only practical in a few special use cases with the current generation of nano-ARPES beamlines: ***the status quo simply cannot deliver the science advances outlined in Section 3.2.*** Further, the possibility for *in situ* transfer between the micro and nano-ARPES branches, and to connected sample preparation capabilities, will increase efficiency and create a unique combined infrastructure for the large and active UK community focussed on studies of electronic materials, including via ARPES (see Section 3.3). The alternative route, via application to other facilities in Europe and further afield, cannot meet this demand, nor will it open the new science opportunities envisaged here. The result will be missed scientific opportunities, at the detriment to UK competitiveness.

### 3.6 Combined impact of project and added value in relation to other on-site activities or facilities



**Fig. 8:** Science areas and complementarity of an upgraded ARPES infrastructure at Diamond-II.

As well as creating a dedicated state-of-the-art capability for spatially-resolved ARPES, there is substantial added value of the proposed developments for the broader photoemission and materials capabilities of Diamond (see Fig. 8). The heavily ( $\approx 4\times$ ) oversubscribed high-resolution I05 beamline will no longer require a first-generation nano-ARPES end station, increasing its capacity and opening a new branch-line slot on a beamline optimised for high-resolution ARPES. An exciting possibility would be to transform this into a spin-resolved ARPES branch-line. Spin-resolving detectors have undergone substantial advances in the last few years, to the point where high-

resolution studies will become possible if coupled with a suitable source, but none of these currently occupy a slot on a high-performance synchrotron beamline offering the energy range, resolving power, and flux of I05. There would thus be an exciting opportunity to deliver a state-of-the-art capability for spin-resolved imaging of the electronic structure of solids, of enormous topical interest. Insights gained from developing high-flux  $\mu$ -ARPES on the new beamline would enable realising a small ( $<5$ - $10$   $\mu\text{m}$ ) spot using capillary optics on I05 to develop spin-resolved  $\mu$ -ARPES, bringing spin-ARPES of *e.g.* topological and magnetic materials into a new regime. The other branch-line on I05 would remain optimised for highest energy resolution and lowest sample temperatures, as is still strongly required by the community. Further opportunities include developing capability for complementary laser-based ARPES on I05. Combined with soft x-ray ARPES and hard x-ray photoemission on I09, and the dedicated capabilities for spatially-resolved ARPES proposed here, this would create one of the most powerful worldwide setups for photoemission investigations of materials and their surfaces, covering the full gambit of energy and spatial scales. Combined with materials synthesis and preparation capabilities, this would be a key driver of an active materials programme at Diamond.



#### 4. Beamline performance specification and requirements

The specification to deliver the science case outlined in section 3 requires:

- **Core energy range:** 50 eV - 250 eV (secondary range extending to  $\approx 800$  eV to achieve smaller spots and for complementary measurements, *e.g.* core-level spectroscopy, resonant photoemission)
- **Sample spot:** <100 nm at 100 eV regular operating conditions (<50 nm ultimate spatial resolution)
- **Resolving power:**  $E/\Delta E$  of at least 10,000 (nano-ARPES) and 15,000 ( $\mu$ -ARPES)
- Nano branch **sample flux:**  $\geq 1.7 \times 10^{11}$  ph/s (experience on existing I05 beamline shows that this sample flux is required for typical user operation)
- **Zone plate to sample working distance:**  $\approx 3$ -4 mm – nano-ARPES branch
- **Scanning capability** for mapping with  $\approx 10$  nm resolution – nano-ARPES branch
- **Integrated sample preparation and distribution** system between end stations
- **Sample manipulator:** temperatures down to 10 K ( $\mu$ -ARPES) and 25 K (nano-ARPES) and good stability (<100 nm drift over 5 hours). All translation and angular degrees of freedom for sample motion. Connections to enable sample biasing or current flow.
- Collection of 4d data sets (two spatial dimensions, energy and angle of photoelectrons) will require **fast data pipelines** to reduce and visualise data; modest increase in computing power required
- Opportunity for **automatic data processing**, *e.g.* using machine learning, to accelerate experiments; possibilities to model data using approaches including DFT

Our simulations of optimised beamline designs yield key and transformational gains over existing capability:

**Nano-ARPES** – For typical user operation flux, the area probed will be reduced by a factor **45 $\times$**  (better than  $100 \times 100$  nm). In ultimate conditions, **50 nm** will be available with usable photon flux.

**$\mu$ -ARPES** – For a similar spot size as the current I05 nano-ARPES there will be a **16 $\times$  increase in flux**. The area probed will be reduced by a factor **>7500 $\times$**  compared to the current I05 HR branch, while offering similar ARPES spectral resolution.

In addition to the major improvements in flux and spot size, modest gains (2 – 3 $\times$ ) in the energy range, resolving power, energy resolution and sample temperatures will be achieved, as summarised below.

**Table 2:** Summary of the proposed beamline performance compared to the current I05 nano-ARPES operation.

	Parameters	Values	Gain factor compared to I05 nano-ARPES
<b>Nano-ARPES</b>	Spot size @ 100 eV [nm]	<100 *	<b>45</b>
	Energy region [eV]	50 – 250 †	3
	Photon flux on sample @ 100 eV [ph/s]	> $10^{11}$	1
	Photon resolving power	>10,000	
	Total energy resolution @ 100 eV [meV]	20	1-1.5
	Sample temperature [K]	< 25	2
<b><math>\mu</math>-ARPES</b>	Spot size @ 100 eV [nm]	<1000	<b>&gt;7500</b> ( <i>c.f.</i> HR-ARPES)‡
	Energy region [eV]	50 – 250 †	1
	Photon flux on sample [ph/s]	> $10^{12}$	<b>16</b>
	Photon resolving power @ 100 eV	>15,000	2
	Total energy resolution @ 100 eV [meV]	< 10	3
	Sample temperature [K]	< 10	3

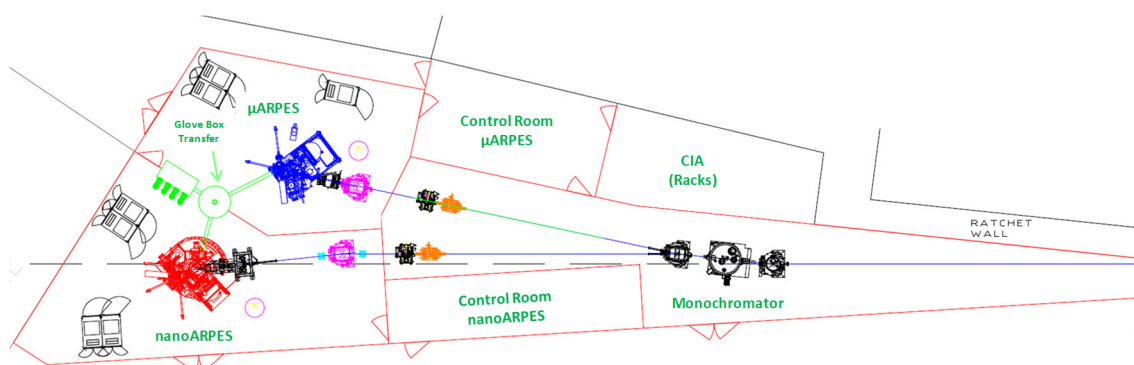
\* Ultimate spot size  $\approx 50$  nm in highest spatial resolution mode, with photon flux  $\approx 0.2 \times 10^{11}$  ph/s, comparable to flux achieved in highest-resolution user experiments performed on I05 nano-ARPES to date

† Energy range optimised for 50 – 250 eV, with extended operation up to 800 eV desired

‡ Gain factor in area probed >7500 as compared to HR-ARPES with similar ARPES performance and sample environment

## 5. Schematic outline of beamline or project

We describe below the outline design of a dedicated beamline for spatially-resolved ARPES in the VUV photon energy region, to enable the science case discussed above (Fig. 9). A 5m long APPLE-II (or APPLE-Knot) undulator combined with a Plane Grating Monochromator is the optimum configuration to deliver light to two state of the art end-stations, developed for nano-ARPES and  $\mu$ -ARPES, respectively. Each experimental cabin will have an independent control room for the user groups, while both *in situ* and offline (located in a nearby peripheral laboratory) sample preparation capabilities will further expand the beamline capabilities.



**Fig. 9:** Layout of the proposed nano-ARPES beamline on the I17 wedge.

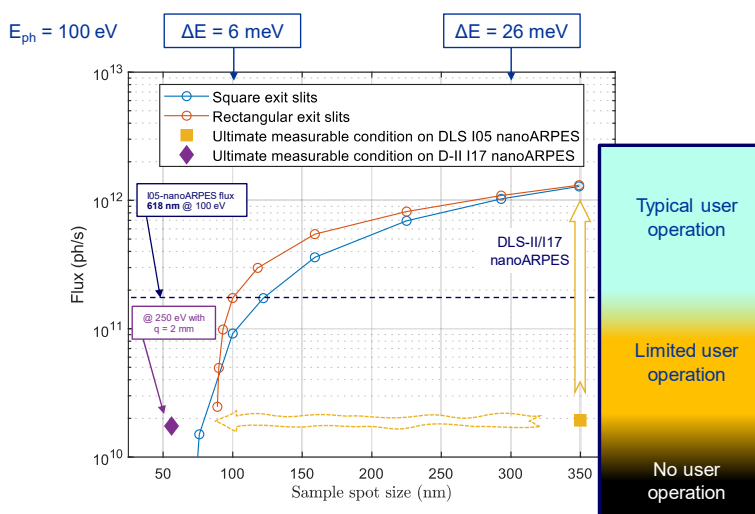
### 5.1 Source

High resolution spatially-resolved ARPES is a photon hungry technique as only a small portion of the light generated can be focused into a small spot size. The APPLE-II undulator of I05 is currently delivering  $12 \times 10^{14}$  ph/s for energies below 100 eV. On Diamond-II with an electron beam energy of 3.5 GeV, the spectrum is shifted to higher energies and the minimum energy accessible is increased from 18 eV to 50 eV. The flux predicted above 50 eV is well suited to the requirements for  $\mu$ -ARPES. At higher photon energies, important for nano-ARPES, the flux remains high. Consequently, we propose to use a 5 m long APPLE-II undulator for the new nano-ARPES beamline providing an operational regime above 50 eV. An upgrade of the magnet array with shorter period would be an efficient way to optimise the flux at high photon energy relevant for the nano-ARPES activity. Currently DLS is developing a new APPLE-knot undulator for I05 that will, in principle, enable operation down to 10 eV on Diamond-II. This new undulator development will also be considered as an upgrade path for the new nano-ARPES beamline.

### 5.2 Optics

A key deliverable of the new nano-ARPES beamline is to provide the smallest spot size possible in the VUV photon energy range, as well as sufficient flux for real experiments. The optical scheme investigated combines a Plane Grating Monochromator (PGM) with Exit Slits (ES) and the use of two different refocusing optics, a Fresnel Zone Plate (ZP) or a capillary mirror (CM), which are located close to the sample. The main optimisation parameters are the positions of the optics along the beam path. These were calculated by minimising the sample spot size at 250 eV with a ZP-sample distance of 3 mm. The analytical expression of the spot size includes the diffraction,<sup>39,40</sup> geometric and achromatic effects, whereas the fluxes were obtained using the ray-tracing package ShadowOui<sup>41</sup> together with the results of REFLEC.<sup>42</sup>

The achieved spot size and photon flux delivered onto the sample are strongly dependent on the ES size as shown in Fig. 10. For spot sizes >100 nm, this largely reflects a dominant contribution of achromatic aberrations (the photon energy resolution simultaneously varies between 6 and 26 meV over this range). A rectangular exit slit aperture provides slightly higher flux, while the lowest achievable spot size is slightly



**Fig. 10:** Calculated photon flux at 100 eV versus the spot size for the nano-ARPES branch when ES size is varied from 5  $\mu\text{m}$  to 1000  $\mu\text{m}$ .

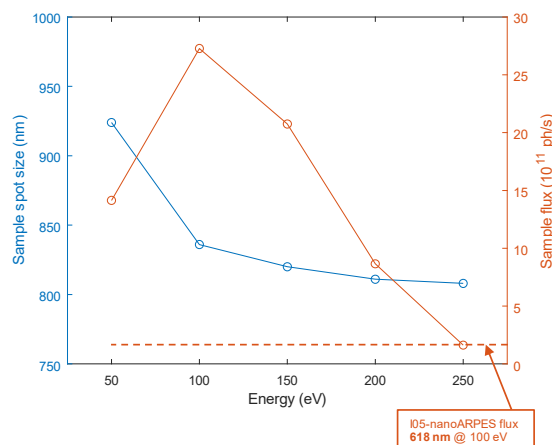
energy of 250 eV and  $q = 2$  mm (purple diamond in Fig. 10) indicates that a spot size on the order of 50 nm can be achieved for a photon flux of  $2 \times 10^{10}$  ph/s. This is comparable to the flux used for the highest-resolution user measurements performed on the I05 nano-ARPES instrument (orange square in Fig. 10, see also Fig. 6c). While this flux will not be suitable for all measurements, it will be possible to utilise it for individual measurements where the highest spatial resolution is required. Below 50 nm, our simulations indicate that the flux would be too low to carry out practical user experiments. For the  $\mu$ -ARPES branch, we employ a reflective optic (capillary mirror) to focus the light on the sample. The flux delivered is thus expected to be significantly higher (one to two orders of magnitude higher than for the nano-ARPES branch), while the spot size becomes limited by the optic's surface quality (Fig. 11): we thus expect to achieve a sub-micron spot size with very high resolving power (yielding 6.5 meV photon beam resolution at 100 eV) giving access to high spectral resolution ARPES on the micrometric scale.

### 5.3 End-station(s)

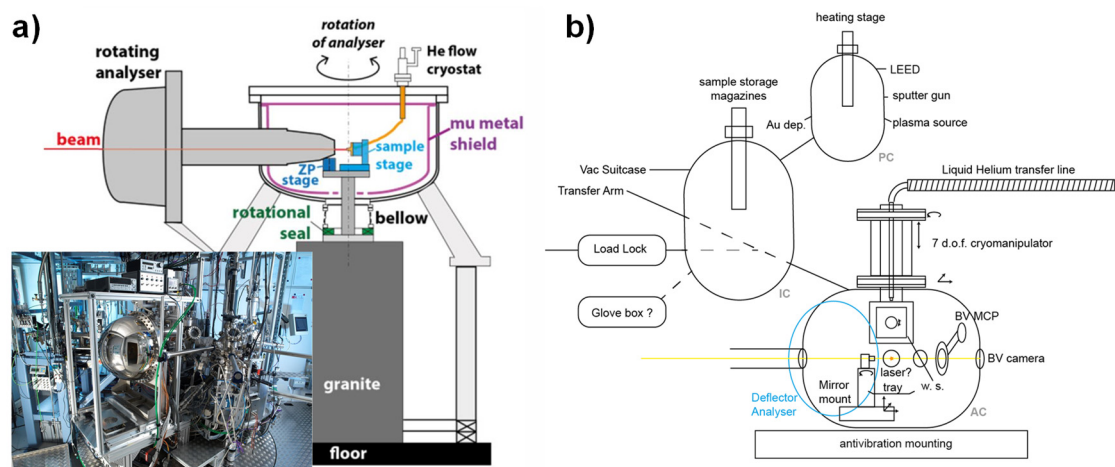
Nano-ARPES and  $\mu$ -ARPES experiments have different instrumental requirements; the end-stations will be carefully designed to optimise either their spatial (nano-ARPES) or their spectral ( $\mu$ -ARPES) performance.

**Nano-ARPES:** We propose a phased installation plan. In phase I, we will transfer the existing I05 nano-ARPES end-station (Fig. 12a), with minor upgrades. This makes good use of existing assets and experience gained to date as well as freeing up some of the staff time to work on the brand new  $\mu$ -ARPES branch. The key features of the end-station are the optics and sample positioning with piezo motors (10 nm precision), while the analyser can be rotated around the sample to expand the momentum range accessible without moving the light spot on the sample. The end-station performance is already well-tested for stability and reproducibility, while an improved cooling mechanism and redesigned top flange will minimise the

reduced (from 90 nm to 75 nm at 100 eV) for a square slit array. For all spot sizes above 100 nm, the predicted flux is higher than that typically employed for user experiments on I05 (horizontal dotted line), indicating conditions well suited for typical user operation but with a probing area 45x smaller than that delivered on I05-nano. Below 100 nm the diffraction effect becomes critical. To access smaller spot sizes requires an increased photon energy, while further reduction can be achieved through increasing the numerical aperture by reducing the zone plate-sample distance. An example calculation for a photon



**Fig. 11:** Calculated spot size (blue) and photon flux (red) vs. photon energy for the  $\mu$ -ARPES branch with capillary mirror. The resolving power is fixed at (15000).



**Fig. 12:** (a) Schematic and picture of existing I05 nano-ARPES end-station. (b) Conceptual design of the  $\mu$ -ARPES system. Acronyms: BV MCP = beam visualiser multichannel plate. W.S. = wobble stick. Au dep. = gold evaporator.

stabilisation time. In phase II, a new endstation will be developed for optimised performance and to work routinely with a sub 100 nm spot size, with a focus on minimising mechanical vibrations and optimising the setup for short zone-plate working distances. At the same time, new electron spectrometers with efficient detection schemes (e.g. utilising recently-developed schemes for simultaneous 2D momentum-space mapping) will be considered, possibly alongside a conventional analyser.

**$\mu$ -ARPES:** This instrument will be designed with a focus on achieving the best possible spectroscopic resolution without compromising the microscopy performance, in order to deliver a user-friendly and highly productive system. The optics of the  $\mu$ -ARPES branch will allow an energy resolution down to 10 meV with a spot size below 1  $\mu\text{m}$ , matched by a sample environment with cryogenic cooling ( $<10\text{ K}$  on the sample) based on an upgraded version of the 6-axis I05 cryo-manipulator, with contacts for *operando* experiments. The end station will be designed for low vibrations and long-term stability, with an in-vacuum hexapod to position the focussing optics close to the sample (Fig. 12b). Based on a strong user demand, the endstations will be linked together and to sample preparation facilities, allowing combined characterisation of the sample by  $\mu$ -ARPES for spectral resolution and nano-ARPES for measurements on the smallest length scales.

#### 5.4 Sample preparation facilities

Instruments including optical microscopes with micromanipulators, a wire bonder, and electrical testing rigs will be provided for preparation of heterostructures and devices. All of this equipment will be housed in a large dedicated glove-box with an argon atmosphere, important for working with 2D materials. *In situ* capabilities for sample (re-)preparation, as well as integrated transfer systems via vacuum suitcases will be implemented, as well as more conventional methods for mounting samples *ex situ*. A peripheral lab will be required for additional sample preparation instrumentation and general lab work related to operations.

#### 5.5 Computing infrastructure and support

The data collection at the beamline will be done according to Diamond standards and the data format will be based on the I05 NEXUS implementation. It is anticipated that a significantly higher amount of data will be generated at the new beamline compared with the existing I05 system, due to the frequent and rapid collection of 4D data sets (two spatial dimensions, vs energy and angle of photoelectrons), with potential for quite high traffic on the beamline network. One of the challenges of spatially-resolved ARPES is the need to efficiently process and visualise such 4D data sets. We anticipate having a high-performance computer in the control room dedicated for on-the-fly analysis, with fast network connection to the data storage server, and will integrate or develop software solutions for data visualisation and reduction. We will investigate approaches, developed for other spatial mapping techniques and starting to be employed for ARPES, where machine learning techniques can allow efficient sampling for spatial mapping using only a fraction of the pixels of a traditional grid scan (e.g. 10-20%),<sup>43</sup> thus further improving beamline throughput.

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## 7. Expressions of interest & support from the community

The proposed beamline is strongly desired by the community, as evidenced by the 105 supporting statements and significant engagement (118 participants) at the nano-ARPES webinar. The depth and breadth of interest in the UK is evident from 58 supporting statements by researchers based in 18 universities/research institutions. This includes established members of the ARPES community, and researchers who have not traditionally used ARPES, but who see the capabilities of next-generation nano-ARPES as transformative to their research. Nobel Laureate Prof. Sir Kostya Novoselov (Manchester & NUS), for example, remarked *'My group would be certainly very interested in doing research on upgraded Light Source, as the present flux cannot guarantee the sufficient spatial resolution.'*, while Prof. Claudia Felser, Director, Department of Solid State Chemistry at the Max-Planck Institute for Chemical Physics of Solids stated *'the electronic structure, space resolved can significantly impact materials science far beyond quantum and energy materials'*. Strong institutional-level support has been received (12 statements), including from the National Graphene Institute (via its director Prof. Vladimir Falco), the National Physical Laboratory (via Chief Scientist, Jan-Theodoor Janssen), numerous University Research Centres, the EPSRC-funded nanoESCA and Harwell XPS facilities, and from relevant Centres for Doctoral Training (*Condensed Matter Physics, Modelling of Heterogeneous Systems, Quantum Materials, Graphene and Related Nanomaterials* and *Advanced Characterisation of Materials*). Each represents a substantial user base that would benefit from access to this proposed state-of-the art nano-ARPES facility.

Individual statements have been submitted by group leaders at the Universities of Bath, Birmingham, Bristol, Cambridge, Edinburgh, Heriot-Watt, Imperial, Lancaster, Liverpool, Manchester, Nottingham, Oxford, St Andrews, UCL, Warwick, and York, the National Physics Laboratory, the Cockcroft Institute, and the Central Laser Facility. The research areas outlined cover the breadth of the proposed science case, including 2D materials, devices, nanomaterials, energy materials, magnetic materials, catalysts, molecular systems and quantum materials. Prof. Moriarty (Nottingham Physics), for example, indicates *'... the proposed ARPES facility will be of particular interest with regard to our research programmes on molecular self-assembly, spin structure and control in a variety of samples, including the 2D systems ... and nanostructured semiconductor surfaces'* whilst Prof. Speller (Oxford Materials) states *'... really understand local variations in the electronic properties (... like Fe-based superconductors that often phase separate on the micron length scale)'*. We are pleased to see many supporting statements from early-career researchers. Dr. Grubisic-Cabo (KTH), for example, comments *'... looking to develop my own group, the establishment of this user resource could be a game-changer for me'* and Dr. Rhodes (1851 Fellow) writes *'I would be able to directly compare STM and ARPES measurements on a spatial scale which has never before been achieved'*.

There is strong international support from Europe, the USA, and Asia, endorsing a UK nano-ARPES capability as both timely and critical: Prof. Baumberger (Geneva), for example, states *'This beamline will be transformative.'*; *'will unquestionably trigger substantial growth of the ARPES community in the UK and abroad.'* and *'The impact ... on quantum matter research and future quantum technologies can hardly be overestimated'*. Prof. Parkin (Director, Max-Planck Institute of Microstructure Physics, Halle) writes *'... future devices are typically of sub-micron spatial dimensions ... a great potential for the proposed nano-ARPES beamline to deliver many new and vital insights to meet the demands of this industry-facing research community, and it should be a cornerstone of the science case for the Diamond-II synchrotron upgrade.'*

Finally, we note that the proposed beamline specification has been particularly welcomed: Prof. Golden (Amsterdam) states *'The\_real\_killer\_app\_for\_ARPES\_on\_quantum\_materials\_is\_low\_T\_nanoARPES\_and\_1\_micron-ARPES\_without\_compromise\_on\_the\_energy\_and\_k-resolution.This\_is\_exactly\_what\_is\_being\_proposed.'*; Prof Da Como (Bath Physics) comments *'The\_proposed\_spatial\_resolution\_combined\_with\_the\_optimal\_photon\_flux\_and\_photon\_energy\_range\_are\_certainly\_offering\_a\_unique\_end\_station\_worldwide.'*; Prof. Brian Gerardot (Heriot-Watt Physics) writes *'The\_ability\_for\_nano-ARPES\_with\_~100\_nm\_spatial\_resolution\_would\_be\_a\_game-changer'*.

## 8. Expressions of interest & support from the community (statistics)

### Statements of support summary

Total number of submissions: 105

Key for statements in Appendix A	Respondent's primary field of research	Percentage of respondents
	Physics	61.0%
	Materials Sciences	29.5%
	Chemistry	6.7%
	Energy	2.9%

Respondent location	Percentage of respondents
UK	66.7%
International	33.3%

Type of organisation supporting	Percentage of respondents
Academic	91.4%
Government	5.7%
Other	2.9%

Diamond user status	Percentage of respondents
Not currently a user at Diamond	33.3%
Currently a user at Diamond	66.7%

### Engagement webinar summary

Date of webinar: Monday 9<sup>th</sup> November 2020

Total number of attendees: 118

Attendee location	Percentage of attendees
UK	44%
International	56%