Diamond-II
Proposal for flagship project
New nanoARPES Beamline

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Outline proposal for a flagship beamline project on Diamond-II

1. Summary/Impact statement

Over the last 5 years, Diamond Light Source (DLS) has become a centre of excellence for research on Quantum Materials, by developing world-leading instrumentation and expertise in angle resolved photoelectron spectroscopy (ARPES) from solids. This proposal builds on this success and will enable DLS to remain at the forefront of ARPES research for the next 15 years. It will help to enable a 2D revolution for industry, where new devices such as topological transistors will provide novel, fast and energy efficient electronic devices. The highly productive I05-ARPES beamline is currently offering high-resolution ARPES (HR-ARPES) and spatially resolved ARPES (nanoARPES) with an oversubscription of over a factor 4. We propose to transfer its state-of-the-art nanoARPES activity to a dedicated long straight section beamline. The higher brightness of Diamond-II together with a truly optimised beamline design will result in a higher flux delivered to the sample, opening up the possibility to explore and discover new electronic phenomena at the nanometre scale (<100nm). The proposed beamline design will have two branches, one of which will provide the best spatial resolution with high flux. The second sub µ-ARPES branch will enhance the existing demands of the user community. While this project will expand the user base and enable new science at DLS, it will also allow the UK and international communities to exploit additional capabilities such as laser-ARPES and spin-ARPES, to maintain a world leading position. The beamlines (nanoARPES and I05) will be highly complementary but with truly optimised layouts and end stations that will enable cutting edge science from the world's leading research groups, including those from the growing UK user base.

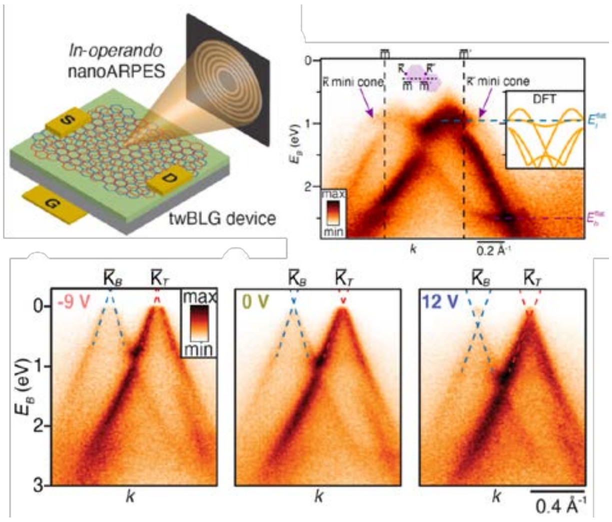
The vision for ARPES at Diamond including a new nanoARPES beamline was presented in May 2019 to the SAC Beamline Review Panel, which was very supportive. "The review panel strongly supports the suggestion to split the existing I05 facility over two beamlines … The above development will vastly increase the capacity and open up new scientific opportunities"

2. Scientific Case

The Diamond-II science case includes key research topics such as **2D materials** and **topological matter** with an emphasis on the experimental electronic structure measurements that will grow in importance to unlock the science in both fields. The proposed beamline is targeted at these two fields, but it will also benefit other research areas where the spatial resolution will either give access to new physics in known materials or enable the first measurements of samples at the nanometre length scale. The concomitant developments on beamline I05 will also establish some spin-resolved and laser based ARPES activity at DLS where we will make use of the unique links that we have with the Central Laser Facility. These will ensure that DLS remains one of the best facilities at which to carry out Synchrotron based ARPES; our ambition is to ensure it is the best.

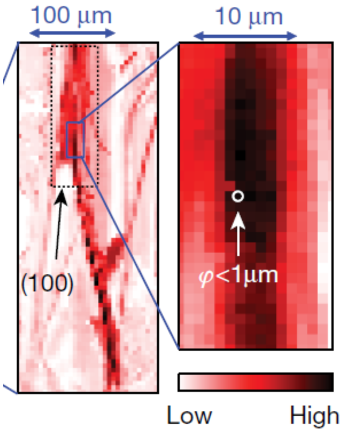
2D Materials: Since the Nobel prize-winning discovery of graphene kickstarted the field in the mid-2000s, "2D materials" have risen to a high prominence [1]. The research topic has expanded to include a vast array of atomically-thin materials, such as the transition metal dichalcogenide (TMDC) family, which hosts an unbelievable variety of electronic phenomena. Micro- and nano-ARPES has made significant contributions to measuring the properties of individual monolayers of such materials, but nowadays the central effort is on the novel physics that emerges from stacking 2D materials on top of each other. In 2018, a group in MIT reported superconductivity in **twisted bilayer graphene** [2]. Hybridisation between the famous "Dirac cone" dispersions of the two layers can lead to the formations of extremely flat bands, which are then susceptible to form novel exotic electronic ground states [3]. Nano-ARPES is unique in its ability to directly measure these flat bands [4]. When considering the possibility of heterobilayers involving different 2D materials [5], and the widely varying properties predicted as a function of twist angles, not even to mention structures involving more than two layers, spatially-resolved ARPES will be uniquely placed to contribute to this growing and exciting field.

While ARPES measurements are traditionally performed on a well-grounded sample, increasing efforts are being made to perform nano-ARPES measurements *in operando*. This research is highly complementary to transport experiments and device design. A conceptually simple, yet only recently feasible [6], example of this 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 and inaccessible conduction band states in semiconducting 2D materials, such as the TMDCs MoS_2 and WSe_2. Other *operando* parameters can also modify the electronic structure, such as mechanical strain that can be applied by, for example, piezoelectric sample stages.



Nano-ARPES spectra of a back-gated twisted bilayer graphene sample (I05, [7]). The energy of the Dirac points shifts with the gate voltage.

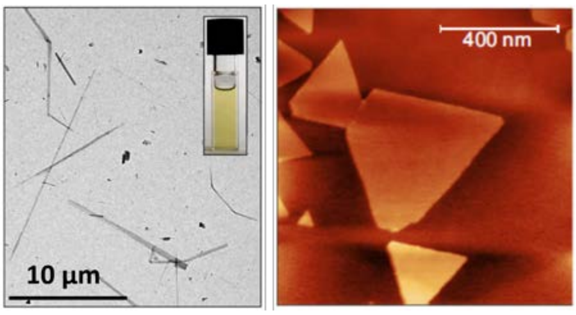
Topological Matter: ARPES measurements have played a critical role in the development of this field, but it is increasingly necessary to use spatially resolved ARPES to remain at the forefront of research. One particular example is the “weak topological insulator”, predicted to host topologically-protected surface states only on specific surfaces of the material. Spatially-resolved photoemission allows access to minority regions that host the desired cleavage plane, and nano-ARPES at I05 was used to demonstrate this for the first verified weak topological insulator, Bi_4I_4 [8]. There is also a more general application to the many topological materials where the samples are very small – certain materials simply don’t form as large single crystals – or when the sample cleave direction is not well-controlled. The discovery of novel electronic states in new materials tends to occur in small samples initially, due to the difficulty in optimising the growth parameters to create larger samples.



Nano-ARPES map from specific cleave orientations in a weak topological insulator (I05,[8]).

Correlated electron systems: Until recently, there has usually been a trade-off between spatial resolution, flux, and energy resolution, making it challenging to explore the small energy scales that are characteristic of correlated electron systems. However, nano-ARPES measurements have already been used to show how the photoemission spectrum of YBCO shows a strong **variation with surface termination** [9]. Similar measurements could be envisaged on delafossites [10] and many other materials that consist of alternate stacks of layers with differing electronic properties. Correlated electron systems nearly always form **structural or magnetic domains** when undergoing phase transitions. Nano-ARPES, where the spot is sufficiently focused means that in many materials, such as FeSe, the domains can be individually resolved, and their spectral functions individually measured [11].

<u>**Opening the door to ARPES on nano-scale structures:**</u> The new nanoARPES facility, with a target beam spot size of <100nm, will allow ARPES measurements on nanostructured materials. These include nanowires and nano-ribbons, which are often investigated for their combination of excellent transport and mechanical properties. For instance, phosporene – the single-layer variant of black phosphorous – was recently isolated in nanoribbon morphology [12]. In some cases, nanoribbons or nanosheets are the primary form of a certain material, which will become accessible to ARPES for the first time. Another unique capability will be accessing the electronic structure of individual islands of monolayer samples grown epitaxially [13]. Furthermore, with ultimate spatial resolution, one can consider attempting the direct detection of edge states in topological materials or Majorana fermions.



Left: phosphorene nano-ribbons. Right: individual islands of a monolayer TMDC.

3. Benefit to the Diamond research community

The development of brighter synchrotron sources has enabled ARPES to undergo a major transformation, where the electronic structure of quantum materials is correlated to the surface topology i.e. spatially resolved ARPES. Pushing the limits of technology, nano-ARPES on a sub 100nm scale will be a unique capability available only in few world leading synchrotron facilities. The proposed beamline will enable the discovery of novel phenomena in the domain of confined electronic systems, edge effects and further extend innovative scientific experiments, establishing links with the field of nanotechnology.

In addition, the proposed sub µ-ARPES endstation will have an immediate impact on the large 2D materials community, since the expected high performance of the instrument will enable a highly productive user program. This will also provide an improved platform for half of the proposals currently submitted to I05 and so will significantly increase the Diamond-II ARPES capacity. The spectral resolution will be comparable to the current HR branch performance, while users will enjoy the additional benefit of the smaller beam spot.

An indirect benefit of the current proposal is to enable a new development plan to keep I05 in a world leading position. The transfer of the current nano-ARPES activity will give space to I05 for spin-resolved ARPES and laser-ARPES, a separate project with massive demand from the user community. The complementarity of the fully optimised beamlines will double the ARPES capacity and address the current oversubscription.

The presence of two dedicated beamlines, along with soft X-ray ARPES on I09, will develop the ARPES portfolio and expand the world leading hub of ARPES-based research and expertise at Diamond and on the Harwell Campus (CLF). This increased capability and internal expertise, through enhancing the already-strong in-house research activity, will maintain DLS as one of the leading centres for ARPES research.

4. Outline Specification

The proposed nanoARPES beamline (on long straight 17) would feature a 5m long APPLE-II undulator source, a plane grating monochromator (PGM) and two end-stations as illustrated in Fig 4.1a). A shorter undulator is not the preferred option as it would considerably compromise the low energy activity where the novel science is expected. The current I05 undulator, to be replaced by an Apple-Knot device, could be transferred to the new beamline. As required for the science case, the nanoARPES beamline would operate in the energy region ~50eV – 240eV. Switching mirrors after the PGM would allow the beam to be delivered to either branch. The Ultra µ-ARPES end-station will have a similar but enhanced design to the highly productive I05 HR-ARPES. The ARPES performance is expected to be as good as the existing beamline, but the beam delivery will be improved to achieve a sub-µm spot size on the sample. Independent control of the spot size will be achieved by working out of focus of the capillary mirror. The second branch will be dedicated to ARPES experiments with sub 100nm spatial resolution. The home-designed instrument will be version II of the current I05 nanoARPES chamber, with improved stability and drift compensation, through the implementation of new schemes such as fast image referencing. The energy resolution of the nanoARPES is predicted to be 20meV at 100 eV photon energy (currently 35meV on I05 nanoARPES). Both end-stations will offer *operando* capability, which is highly demanded by the user community.

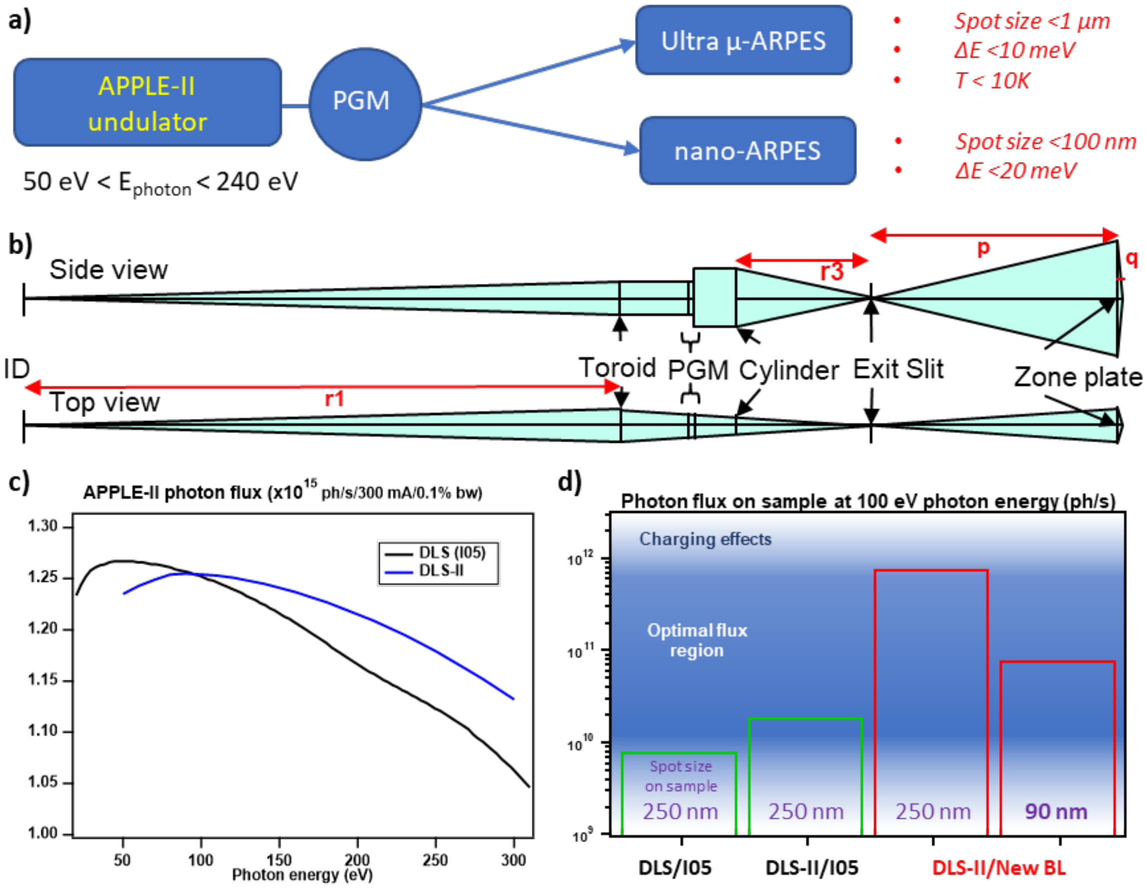


Figure 4.1: a) Layout of the new beamline showing both endstations with their performances. b) sketch of the optical path with the PGM demagnification (r3/r1) and ZP demagnification (q/p). c) The APPLE-II photon flux on DLS and Diamond-II will be similar (linear scale). d) nanoARPES branch photon flux on the sample at 100eV photon energy. The proposed BL on Diamond-II (red line) will considerably increase the usable photon flux.

The optical design of the nanoARPES branch will be optimised to achieve sub-100nm spatial resolution, while maintaining good energy resolution and speed of data acquisition (Fig 4.1 b). This requires increasing the photon flux on the sample, reducing the achromatic aberrations and improving the diffraction limit introduced by the Zone Plate (ZP). At 100eV photon energy, Diamond-II is expected to give the same flux as DLS (Fig 4.1 c). However, the reduced horizontal size of the Diamond-II photon source will lead to a smaller horizontal spot size on the exit slit, resulting in a 2.4 times increase of flux on the sample compared to I05.

Furthermore, the low divergence of Diamond-II will improve the flux illuminating the ZP. In a new beamline design where the photon source is further demagnified at the PGM exit slit and the beam divergence is matched better to the ZP size we predict an increase of the photon flux on the sample (Fig 4.1d) by a factor **98.2** compared to I05. This increase of photon flux can now be traded to improve the spatial resolution down to 90 nm and maintain a photon flux of nearly 10^{11}ph/s, enough to perform exciting ARPES studies at the nanometre scale.

5. State of the art benchmark

The new nanoARPES beamline is aiming for very high spatial resolution (90 nm) with excellent photon flux, to be able to perform high-quality measurements. Currently, the best spatial resolution achieved is at the ALS with 120 nm and they are expecting to reach 50 nm in the future (Table 5.1). Our analysis of the field indicates that the key probe size to study many interesting effects is ~100nm leading to the proposed beamline design. It could, however, deliver smaller focus at the cost of the photon flux giving us flexibility in the final design to adjust the balance between spot size and photon flux.

Synchrotron	High resolution ARPES beamline	Spatially resolved ARPES Beamline
DLS (UK)	I05	New nanoARPES BL **90 nm**
ALS (USA)	Beamline 4.0.3 (MERLIN), Beamline 10.0.1 (HR-ARPES + spin-ARPES)	Maestro **120 nm**
Soleil (France)	CASSIOPEE (HR-ARPES, spin-ARPES)	ANTARES **> 120 nm**
ASTRID2 (Denmark)	SGM3	SGM4 **1 µm**
Elettra (Italy)	APE, BaDElPh	Spectromicroscopy **3 µm**

Table 5.1: List of the high energy resolution or spatially resolved ARPES beamlines in synchrotron facilities.

In order to be able to push the instrumental limits, it is necessary to develop dedicated beamlines as realised at other synchrotrons sources (Table 5.1). The new beamline will provide world leading capability with high performance and high productivity, with a sample environment that is optimised for usability by the scientists. The new beamline will be user friendly with a very efficient data acquisition system, based on the I05 experience.

6. Community engagement

The UK has a very active community in the domain of material science and surface science, with world leading experts. We are developing our ARPES user community by facilitating beamtime access and providing training to new groups. New collaborations with the leading UK 2D materials institutes - e.g. National Graphene Institute, University of Bath - nanoESCA facility, National Physical Laboratory – is a way forward to reach the community. The nanoARPES beamline will offer a complementary approach for groups already working with microscopic techniques.

References

[1] P. Miró, M. Audiffred, and T. Heine, Chem. Soc. Rev. **43**, 6537 (2014)
[2] Y. Cao, …, and P. Jarillo-Herrero, Nature **556**, 43 (2018)
[3] Y. Cao, …, and P. Jarillo-Herrero, Nature **556**, 80 (2018)
[4] S. Lisi, …, and F. Baumberger, arXiv:2002.02289 (2020)
[5] S. Ulstrup, …, and J. A. Miwa, Sci. Adv. **6**, eaay6104 (2020)
[6] P. V. Nguyen, …, and N. R. Wilson, Nature **572**, 220 (2019)
[7] Alfred J. H. Jones, …, and Soren Ulstrup, submitted to Adv. Mater. (2020)
[8] R. Noguchi, …, and T. Kondo, Nature **566**, 518 (2019)
[9] H. Iwasawa, …, and M. Hoesch, Phys. Rev. B **99**, 140510 (2019)
[10] V. Sunko, …, and P. D. C. King, Nature **549**, 492 (2017)
[11] L. C. Rhodes, …, and T. K. Kim, arXiv:2004.04660 (2020)
[12] M. C. Watts, …, and C. A. Howard, Nature **568**, 216 (2019)
[13] A. Rajan, …, and P. D. C. King, Phys. Rev. Mater. **4**, 014003 (2020)