# Diamond Beamline Proposal 051

# A High Resolution Angle-resolved Photoelectron Spectroscopy Beamline

**A proposal prepared for the SAC April 2008**

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Thanks to Emily Longhi (DLS) for calculations of potential insertion devices.

A High-Resolution Angle Resolved Photoelectron Spectroscopy Beamline for the Diamond Light Source

## 1. Summary

Angle resolved photoemission (ARPES) is the most direct probe of the electronic band dispersion in crystalline solids. It has played a key role in elucidating the properties of many frontier materials such as the high-temperature superconductors and continues to make a vast impact in correlated electron physics and surface/nano-science. The development of high resolution UV-beamlines at third generation synchrotrons and the advent of electron spectrometers with massive parallel detection have improved the relevant figure of merit in ARPES, which is given by the product of energy/momentum resolution and count rate, by at least 5 decades over the past 15 years. This has helped transforming ARPES from a niche spectroscopy into one of the most important techniques in experimental condensed matter physics.

Utilizing the full strength of ARPES as a many-body spectroscopy imposes stringent requirements on resolution, stability and sample environment. Often, the most interesting phases of new materials are found far below room temperature. Taking the mean-field Peierls gap of $2\Delta = 3.53\ k_BT$ as a reference of the relevant energy scales, 100 K [4 K] corresponds to approximately 15 meV [0.6 meV], which is also comparable to the thermal broadening of the Fermi function at these temperatures. Resolving spectral features of this magnitude requires sufficient flux at a millielectronvolt (or even *sub* – meV) resolution and stability. Tunability of the polarization is important to selectively excite states of a given symmetry. These requirements can be met by a low energy undulator beamline providing linear and circular polarized radiation between ≈ 8 – 80 eV with a resolving power > $10^4$ over the entire energy range. A further advantage of low photon energies is the steeply increasing bulk sensitivity near the work function cut-off. Although Diamond was designed for optimum performance in the soft x-ray range it can compete with the world-best sources at UV energies. It is a realistic goal to aim at providing Europe's leading ARPES facility.

The end-station and in particular the sample environment and handling are crucial for ARPES experiments. The restrictions arising from the common combination of a broad range of instrumentation in a single end-station often limit the performance of entire beamlines and are hardly compatible with the ambitious goal of providing a world leading instrument. Rather than compromising on performance in favour of increased versatility we propose to focus this beamline on the core requirements for high-resolution ARPES and to excel in a core technique of modern condensed matter physics. More specialized photoemission techniques or a combination with other spectroscopies may be implemented in an optional branch line at a later stage. A particularly promising development, which would ideally complement the capabilities of the main line, is "nano-ARPES", *i.e.* angle-resolved photoemission from a sub-micron focus of the radiation.

The UK has an internationally recognized strength in correlated electron science but little to no presence in modern ARPES. Leading experimental and theoretical condensed matter physicists throughout the UK have raised concerns about this rather unsatisfactory situation in their support letters and have stressed that ARPES is vital for the development of a number of frontier fields, such as neutron scattering, low-temperature physics or surface and nano-

science. The proposed beamline provides an ideal opportunity to turn this situation around and to boost scientific output across many fields.

# 2. Science Case

## *2.1 Introduction*

A central challenge in modern condensed matter physics is to further the understanding of quantum many-body systems in which strong interactions lead to the self-organization of valence electrons in novel ground states substantially different from those of simple metals or insulators. Examples include unconventional superconductivity, quantum magnetism, colossal magneto-resistance or non-Fermi liquid behaviour near quantum critical points, and have been observed in various complex materials, such as transition metal oxides, *f*-electron intermetallics or molecular crystals. ARPES has played a key-role in studying the above phenomena and materials and has helped to set the scientific agenda in many ways. The numerous publications in Science and Nature with at least six recent ARPES papers [1-6] appearing in the *"ten most cited physics papers"* periodically tracked by the Institute of Scientific Information give a measure of the insight and significance of results obtained by this technique. However, what has surfaced thus far is only the tip of the iceberg.

In the following we will identify some of the current frontier areas in the field of complex materials and illustrate the science cases, which can be investigated with a new high-resolution ARPES beamline. Finally, we will discuss future opportunities in nano-ARPES.

## *2.2 Science opportunities in ARPES*

### *2.2.1 Carbon based materials: graphite, graphene, nanotubes, fullerenes*

The recent discovery of graphene, a perfectly two-dimensional form of carbon, at the University of Manchester has triggered enormous interest for its peculiar electronic properties and because of its potential for applications in electronic devices [7, 8]. Many of its unusual properties such as a non-integer quantum Hall effect, which is observed up to room temperature or its cyclotron mass $m_c$ described by $E = m_c c^2$ are direct consequences of the peculiar band structure dominated by massless Dirac Fermions. Recently, ARPES has provided the first direct observation of Dirac Fermions in bulk graphite [9] and detailed studies of the band dispersion in epitaxial graphene near the Fermi level have provided a comprehensive understanding of the many-body interactions in the system [10]. Moreover, ARPES has been used to demonstrate that by controlling the carrier density in a bilayer of graphene, the occupation of electronic states near the Fermi level and the magnitude of the gap between valence and conduction band can be manipulated [11].

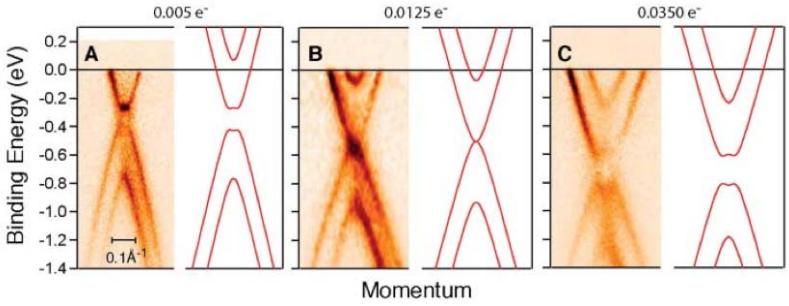


*Figure 1: Control of the bilayer electronic structure of graphene as monitored by ARPES (from [11]).*

The potential of ARPES for the study of correlated molecular solids has recently been demonstrated by the observation of a renormalized dispersion and coherent quasiparticle-like states in a monolayer of $C_{60}$ on Ag(111) (see Fig. 4) [12]. Interestingly, this is the only system to date where electron spectroscopy has shown coherent electronic states and strong many-body signatures as they are expected in a variety of metallic or superconducting organic systems. This remarkable lack of data is largely due to technical difficulties, most notably the small Brillouin zones and band-widths in molecular systems, rendering conventional ARPES studies challenging. We expect that the beamline proposed here will markedly improve this situation and will provide rich information on the many-body physics of various organic conductors. This could form the basis for a more fundamental understanding of the rich phase diagrams of molecular materials and may stimulate their exploitation in future electronic devices.

### *2.2.2 Transition metal oxides*

#### *a. High temperature superconductors*

The discovery of superconductivity with high transition temperature in oxides by Bednorz and Müller in 1986 [13] has changed the agenda in condensed matter physics. Within a few months after this break-through, new materials with $T_c$s above the temperature of liquid nitrogen have been discovered, generating enormous interest because of their potential for applications. By now high-temperature superconductors are used in diverse products ranging from MRI imaging systems, circuit breakers, power-lines to electrical motors and many more. The scientific importance of high-$T_c$s stems from the fact that they highlight an intellectual crisis, opening the opportunity for major advances in the quantum theory of solids.

ARPES has been very successful in establishing the basic electronic structure of the cuprate high-$T_c$s and has contributed landmark results elucidating the unconventional properties of these materials [14]. Some of most important results include the first direct evidence for an anisotropic order parameter in $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) [4], the detection and characterization of the pseudogap [2, 3] and the observation of strong electron-phonon coupling in different families of cuprates [15].

Despite the enormous success of ARPES in the study of high-$T_c$, many phenomena remain poorly understood. Below, we highlight some of the open issues.

***Symmetry of the order parameter*** Knowledge of the order parameter symmetry is crucially important in unconventional superconductors. In fact, the large gap anisotropy of cuprates, first observed by ARPES in Bi2212 is a corner stone in the experimental evidence distinguishing HTSCs from conventional superconductors, which conserve the lattice

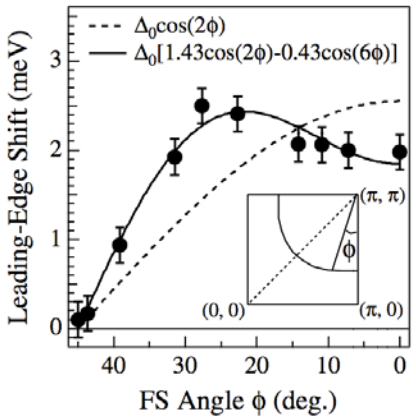


*Figure 2: Evidence for a non-monotonic d-wave superconducting gap in the electrons doped cuprate $Pr_{0.89}LaCe_{0.11}CuO_4$.*

symmetry and only break gauge symmetry. However, the precise shape of the gap remains an open issue. There is overwhelming evidence for non-*s*-wave superconductivity in cuprates, but existing ARPES data cannot exclude deviations from a pure *d*-wave gap. Given that most published photoemission data on cuprates was measured with an energy resolution of 10 – 15 meV, it is entirely possible that small non *d*-wave components exist, but eluded detection by ARPES. Recent μSR experiments, for instance, provided evidence for an s-wave component in LSCO of $\Delta_s \approx 1.6$ meV [16]. A gap of this order of magnitude is extremely challenging to resolve on existing ARPES beamlines. Clearly, the superior resolution and temperature control of the new beamline opens an opportunity for significant advances in this regard. Intriguing recent results giving a glimpse at possible discoveries include the observation of fully gapped single particle excitations in underdoped cuprates [17], or the evidence for a non-monotonic *d*-wave gap in the electron doped cuprate $Pr_{0.89}LaCe_{0.11}CuO_4$ [18]. Systematic experiments with improved resolution are highly desirable.

***Nodal liquid*** Overdoped cuprates behave in many ways like Fermi liquids. They have a well-defined Fermi surface, extensively characterized by ARPES, and quasiparticle like excitations. Upon underdoping, the Fermi surface appears to break up into disconnected "Fermi arcs". This phenomenon is intimately related to the opening of a momentum dependent pseudogap and to the suppression of coherent spectral weight near the anti-nodes, which has been connected to the charge ordering seen in STM and the stripes observed by neutron scattering. However, the interpretation and even the very existence of "Fermi arcs" is the subject of intense debate. The recent observation of quantum oscillations in YBCO questions the idea of disconnected arcs and points towards the existence of small closed Fermi contours. Improved ARPES experiments with higher sensitivity and resolution could shed light on these issues by mapping the momentum dependence of the superconducting gap and the pseudogap with higher precision and by searching for backfolded Fermi contours, as they may arise in a density wave state.

***Electron mode coupling and self-energy*** One of the most important recent papers in high-temperature superconductivity reported a "kink" in the dispersion of several families of cuprates [15]. Its interpretation in terms of strong electron-phonon coupling has been controversial, but has undoubtedly renewed the interest in lattice degrees of freedom and in the relevance of low-energy bosonic modes for the pairing mechanism. A key issue in the debate is the interpretation of the signatures in the single particle spectral function. ARPES data provided good evidence for coupling to multiple bosonic modes [19, 20]. However, the identification of these modes is non-trivial and can only be successful if the ARPES data is related to other techniques. In particular neutron scattering with its ability to directly measure the dispersion of lattice and magnetic modes has provided key-input into this debate. With ISIS, the UK has a leading neutron facility, which has boosted activity in this field. The

proposed beamline could propel electron spectroscopy in a similar way, opening up exciting perspectives for interdisciplinary research on complex materials.

### *b. Colossal magnetoresistive oxides*

Intense interest in doped compounds of the perovskite $LaMnO_3$ (LSMO) was stirred by the observation of *colossal magneto-resistance (CMR)* – an extremely large drop in resistivity induced by the application of a magnetic field near the Curie temperature [21]. The theoretical understanding of the CMR phenomenon is still incomplete but it is generally accepted that a combination of double-exchange, electron-phonon coupling and orbital ordering are important ingredients. Very recently, ARPES has for the first time observed well-defined quasiparticle excitations in manganites [22, 23]. This breakthrough has been enabled by the improved resolution and cleanliness of the experiment and by advances in sample preparation. These results open numerous possibilities for elucidating striking similarities to the pseudogap phase of high-temperature superconductors [22] and for future studies of the momentum, doping and temperature dependence of the quasiparticle dynamics, which defines the unique magnetic and transport properties of manganites.

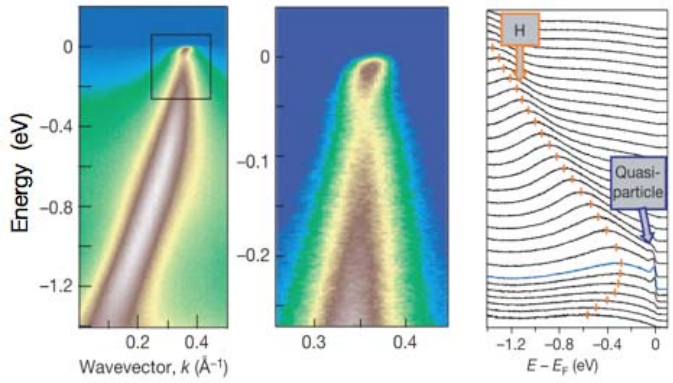


*Figure 3: Observation of strongly renormalized "nodal" quasiparticles and an "antinodal" pseudogap in a bilayer manganite.*

### *c. Ruthenates, rhodates, iridates*

Ruthenates, rhodates and iridates in many ways bridge the gap between the intensely studied 3*d* transition metal oxides and *f*-electron intermetallics and can provide new insight in the correlated electron problem. The most important family, the ruthenates of the Ruddlesden-Popper series $(Sr/Ca)_{n+1}Ru_nO_{(3n+1)}$, are structurally closely related to cuprates and manganites and show a broad range of interesting phases, including itinerant magnetism, metamagnetism, unconventional superconductivity and correlation induced insulating states.

***Fermiology*** The normal state electronic structure of the prototypical layered perovskite $Sr_2RuO_4$ has been determined in greater detail and with higher precision than that of any other transition metal oxide. Surprisingly, the situation is remarkably different for the other members of the Ruddlesden-Popper series, and even the most basic question concerning the orbital population remains essentially open in most materials. Clearly, high-resolution data on more compounds are required for advancing the understanding of this important material class. An interesting aspect of Fermi surface studies is the relation of the observed quasiparticle electronic structure to band structure calculations within the local density approximation (LDA). Ruthenates offer a unique opportunity to advance the understanding of the applicability of band-theory to strongly correlated materials because of the availability of

high-quality Fermi surface data on at least a few of the simpler compounds [24]. This is by no means trivial. In fact, $Sr_2RuO_4$ and $Sr_2RhO_4$ are the only correlated metals to date for which consistent bulk dHvA and surface sensitive ARPES data that fully determine the Fermi surface size and shape are available. Intriguingly, it appears that the low-energy electronic structure of $Sr_2RuO_4$ is well described within the LDA [24], whereas substantial quantitative differences have been observed for $Sr_2RhO_4$ [25].

***Many-body interactions*** Understanding how chemically closely related compounds can have so vastly different physical properties is a central theme in condensed matter physics. In the case of the strontium ruthenates, it has been argued that much of this diversity is controlled by structural distortions, which modify the band width and thus the relative importance of electron correlations [26]. However, there is little doubt that some of the more subtle properties are governed by the interaction of electrons with collective spin and lattice excitations. This is certainly true for the superconducting ground state of $Sr_2RuO_4$. The pairing of electrons can only be mediated by bosonic modes, such as spin fluctuations or phonons in conventional BCS superconductors. ARPES is a uniquely powerful tool to access electron-boson interaction directly. To date, only a few results focusing on high-energy optical phonons have been reported [27]. With the increased resolution of the new beamline, it becomes feasible to investigate the coupling to low-energy lattice modes associated with the rotation and tilting of entire octahedra and to the magnetic fluctuations, which profoundly influence the macroscopic properties of the layered ruthenates.

### *2.2.3 Heavy fermion systems*

Angle-integrated photoemission studies have been very successful in revealing single ion properties in the *k*-integrated *4f* spectrum of Ce materials [28]. However, more detailed studies of the Fermi surface and of the mechanisms of heavy mass formation have been hampered so far by technical barriers, mostly by the insufficient resolution of previous generation beamlines. Recent laser excited ultra-high resolution photoemission data on the heavy fermion compound $CeRu_2$ demonstrate the potential of ARPES to overcome these barriers and to resolve the subtle electronic structure effects on the ultra-low energy scale of *f*-electron systems [29].

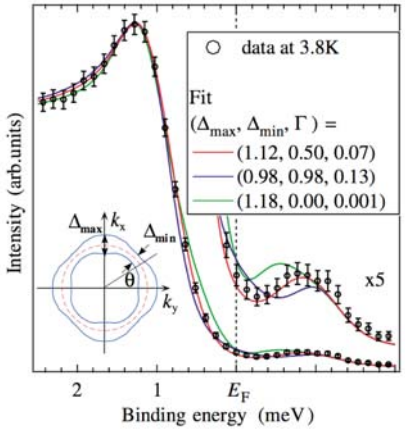


*Figure 4: Ultra high-resolution angle integrated photoemission spectrum from the heavy fermion superconductor $CeRu_2$ [28]. The data were taken with a combined energy resolution of analyzer and light source of 0.36 meV. A superconducting gap < 1 meV is clearly resolved. The detailed line shape gives evidence for a non-uniform gap.*

The proposed beamline will offer a resolution comparable to the above laser-ARPES data, while maintaining full tunability of the photon energy. This is crucially important for the investigation of *f*-electron systems where the hybridisation of localized and itinerant states leads to strongly energy and momentum dependent matrix elements.

### *2.2.4 New correlated electron systems*

In recent years, ARPES has driven advances in understanding across oxide physics, heavy fermion physics, and the physics of carbon based materials. Whilst not being a "smoking gun" technique delivering the first results on new materials, ARPES has provided rich information on the electron dynamics and the effects of many-body interactions required for an in-depth understanding. With the continuous advances in technology, ARPES begins pushing into new territory. The speed with which reliable photoemission results become available after a new material 'breaks' has greatly increased over the last decade helping to transform ARPES into a primary tool for materials research. In several recent cases including the correlated metal $Sr_2RhO_4$ [25], the new superconductor $Cu_xTiSe_2$ [30] or the triangular cobaltate $Na_xCoO_2$ [31], ARPES has provided crucial electronic structure information long before results from bulk sensitive probes such as the de Haas – van Alphen effect became available. There is no sign of slowing in the pace of improvement of ARPES and it is safe to predict that the importance of the technique in the study and optimization of new materials will continue to increase.

### *2.2.5 Electronic structure of surfaces and interfaces*

Surface and interface science underpins much of nano technology and contemporary material science and is widely recognized as fundamental for an extremely broad range of technologies. Angle resolved photoemission is intrinsically surface sensitive and is being applied to vastly different problems in surface science. Instead of striving for a comprehensive overview of such a broad field, we will restrict this paragraph to highlighting a few opportunities in surface science, which are directly relevant to the main science case.

The reduced dimensionality at surfaces provides a natural playground to study the physics of low dimensional electronic systems. Particularly well suited for such studies are a variety of highly ordered metal over-layers on semiconductor surfaces [32-34]. Because of the band gap in the substrate electronic structure, these systems are structurally and electronically ideally two-dimensional with the electronic states confined in a single atomic layer. Moreover, chain-like quasi one-dimensional metal on semiconductor systems may be grown on stepped templates, with a degree of inter-chain coupling, that can be controlled by the terrace width of the substrate [32, 34]. In particular the periodic arrays of monatomic Au chains, grown on vicinal Si surfaces have recently been the subject of an intense debate, because of the possible observation of spin-charge separation [34]. An exciting aspect of monatomic wires of metal atoms on semiconductor surfaces is their potential to exhibit a low-temperature metallic phase [35]. While the ideal 1D metal is unstable with respect to a Peierls distortion of the lattice, and thus becomes insulating at low temperatures, the MI transition may be avoided in supported wires by the strong bond of the metal atoms to the underlying 3D material [32]. Since this bond is entirely mediated by the low-lying valence states, it should be possible to study low energy excitations in a truly 1D system and link the results to the exotic properties of quasi-1D bulk systems [36, 37].

Recently, there has also been significant progress in the preparation of two-dimensional metallic systems on semiconductor surfaces. For the case of the ($\sqrt{7} \times \sqrt{3}$) In/Si(111) surface, it has been demonstrated that a surface quality where the mean free path of electrons is limited only by scattering at the scarce surface steps, may be achieved [33]. With such systems at hand, it becomes feasible to study the role disorder, correlations and electron-phonon interactions in a perfectly two-dimensional electron gas. Very recent results from an

Ag overlayer on Si indicate rich physics including a novel metal to non-metal transition [36]. These systems differ from the more thoroughly investigated one- and two-dimensional electron gases in semiconductor heterostructures by their several orders of magnitude higher charge density pushing into a largely unexplored field. Presently, the available resolution and sample temperature limit such studies, which therefore could greatly benefit from the ultrahigh resolution that will become available at the proposed beamline.

### *2.3 Opportunities in nano-ARPES*

Combining the energy and momentum resolution of ARPES with nanometer spatial resolution offers exciting perspectives for novel experiments. However, spatially resolved ARPES is technically challenging and requires a highly specialized setup. In order to optimize the performance and productivity of the entire beamline, we envision focussing the main line on non-spatially resolved studies at very high momentum and energy resolution. A nano-ARPES end-station may then be built on an optional separate branch-line without compromising the performance of the main line.

Using state-of-the-art zone plates in combination with a momentum resolving electron spectrometer can provide an ultimate spatial resolution below 100 nm. This offers a window to a range of new and interesting problems in nano technology and correlated electron physics, as discussed in the following.

***Nano technology*** Photoemission with sub – 100 nm spatial resolution offers numerous possibilities in the study of nanostructures with interesting properties. A central theme in nano-technology is the exploitation of organic molecules as functional elements in electronic circuits. Spatially resolved ARPES could be used to provide unique information on band-alignment, molecule-substrate interaction and many-body renormalization in individual molecular switches dispersed on a substrate. Other possible experiments include spectroscopy of the quantized states in catalytically active clusters as shown in figure 5, or of nano-scale islands. With a spatial resolution near 100 nm, it becomes feasible to zoom in on individual clusters or islands, while maintaining a high momentum resolution, only limited by the intrinsic momentum uncertainty $\Delta k = 1/\Delta x$ in nano-scale objects. This offers unique opportunities for studying electronic confinement and correlations in nano structures.

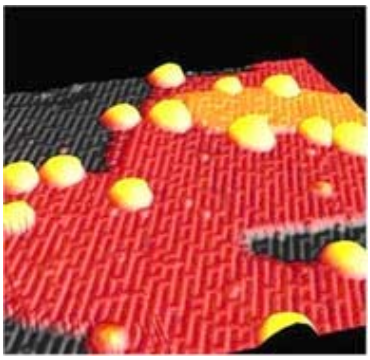

*Figure 5: Au cluster deposited on rutile $TiO_2$ (courtesy of R. Schaub).*

***Correlated electron systems*** ARPES has been very successful in determining the bulk electronic structure of correlated electron systems. However, as a *k*-space probe, it relies heavily on spatially homogeneous electronic properties. This is not always the case in correlated electron systems. Strong interactions can lead to a reorganization of the electronic system in different coexisting phases. An example of such a phase separated state near the metal-insulator transition in a perovskite manganite is shown on the right [38]. Spatially resolved ARPES could provide unique information on the properties of individual phases and their interplay.

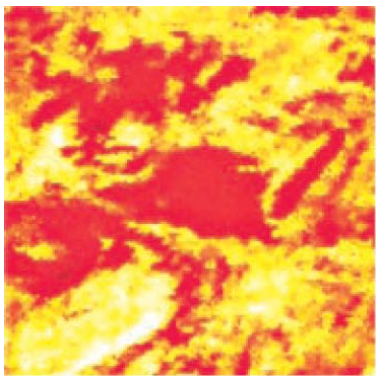

*Figure 6: Inhomogeneity of the metal-insulator transition in (La,Ca)$MnO_3$. Light (dark) colors show insulating (metallic) areas. Image size is 610 x 610 $nm^2$ [38].*

Moreover, nano-ARPES promises high-quality spectroscopic results on a much broader range of materials than possible today. The necessity of large and highly perfect surfaces for conventional ARPES studies has long limited the range of accessible samples. A sub – 100 nm spatial resolution would enable investigating materials and problems not previously accessible. Examples include systems with different cleavage planes, such as YBCO, the gold standard in most bulk experiments on high-temperature superconductors, compounds with surfaces that are rough on a macroscopic length-scale but well-ordered microscopically, or simply any material for which large crystals cannot be grown.

***Feasibility*** The combination of a zone-plate with aperture selection system focussing the radiation to a *sub* – 100 nm spot with a commercial momentum resolving electron analyzer is the most promising approach to nano-ARPES. The spatial resolution of such a setup is given by the spot size at the focus of the incoming radiation, which depends on the minimal zone spacing, the photon energy and the monochromaticity of the beam. State-of-the-art zone plates for a photon energy of 100 eV can provide a focus near 50 nm for a band width of the incoming radiation of $\Delta E/E = 1/5000$. Assuming an efficiency of 5% for the zone plate and a coherent fraction of 10%, the flux expected for such a setup is around 200 times lower than achievable with standard refractive optics for non-spatially resolved ARPES. Clearly, this requires compromising spectrometer resolution, sampling density and signal to noise ratio in the data sets, if measurements are to be performed in a useful time. However, it will still permit obtaining interesting data. For instance, if all of the above quantities are reduced by a factor of 3, a data set with *sub* – 100 nm resolution could be acquired in the same time as a standard, non-spatially resolved measurement requires.

### *2.4 Technique development*

One of the key-challenges in ARPES experiments is to manipulate atomically clean surfaces using a two- or three-axis goniometer at cryogenic temperatures. Inefficient cooling and degradation of samples due to adsorption of residual gas on the surface are severe problems and often the limiting factors in real experiments. The focus on a single technique for the main end-station offers an opportunity to go significantly beyond the state-of-the-art in this respect. We plan to improve cooling performance and to markedly increase surface lifetimes by enclosing the sample almost completely in a cryogenic radiation shield. This results in vacuum conditions not achievable by conventional UHV technology. Low temperature STM studies under similar conditions showed a reduction in surface contamination through adsorption from the gas phase by at least 3 orders of magnitude. Even with the necessary openings in the cryo-shield for photon beam and escaping electrons, it is feasible to reduce

surface contamination by about an order of magnitude, which would immediately lead to a marked increase in data quality and reliability.

### *2.5 Alternative beamlines*

Diamond Light source currently has no operational or planned beamlines on insertion devices that cover the UV range below 100 eV, which is vital for high-resolution ARPES experiments.

The main competition for the proposed high-resolution ARPES beamline comes from US synchrotron sources, in particular from the Advanced Light Source (ALS) in Berkeley and from the Stanford Synchrotron Radiation Laboratory (SSRL). Currently four operational beamlines at these rings (BL7, BL10, BL12 at ALS and BL V-4 at SSRL) focus on closely related scientific goals and provide ARPES infrastructure in the same energy range, although with slightly lower performance than possible today. Two more projects currently under commissioning will further strengthen US activities in this field.

The main competition within Europe comes from the SIS beamline at the Swiss Light Source (SLS), the "$1^3$" spectrometer on the UE112 PGM beamline at BESSY, Berlin, and from two ambitious projects currently under commissioning at Soleil (ANTARES, providing sub-micron spatial resolution and CASSIOPEE focusing on high energy resolution).

The proposed beamline at Diamond could compete with all the above existing or planned beamlines in terms of flux and resolution. It is realistic to aim at establishing Diamond as the premier facility for high-resolution ARPES in Europe.

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## 4. Requirements

The scientific case relies on achieving a combined energy resolution of beamline and electron spectrometer < 10 meV in routine operation, with an ultimate resolution < 1 meV for the study of heavy quasiparticles in f-electron intermetallics and transition metal oxides. With current technology, a sufficient photon flux in a band-width of 1 meV can only be achieved at photon energies below 100 eV. A beamline dedicated to high-resolution ARPES needs to cover a minimal energy range of 20 – 80 eV with an ultimate resolution < 1 meV matching the performance of state-of-the-art electron spectrometers. Lower photon energies (6 – 20 eV) provide enhanced bulk sensitivity, allow for the highest energy and momentum resolution and are useful for tuning matrix elements, which vary rapidly at low energy. Higher photon energies (80 – 200 eV) on the other hand would facilitate the implementation of a nano-ARPES branchline operating at slightly lower energy resolution and would give access to shallow core-levels. Polarization control is important for selectively exciting states of a given symmetry.

Achieving the highest possible flux in a given band-width is key for high-resolution ARPES. As with many other spectroscopies, resolution can be traded in for acquisition speed in photoemission. Practical experience shows that ARPES beamlines are rarely operated at the resolution limit, simply because data acquisition times increase to unacceptable levels, which can even surpass surface lifetimes in good ultrahigh vacuum.

The end-station design is crucially important in ARPES. Particular emphasis needs to be placed on rapid and reliable sample transfer, excellent temperature control down to < 8 K, extremely clean UHV and to the robustness and user friendliness of the entire setup. Basic surface-preparation and characterization facilities including low-energy electron diffraction (LEED) will be provided in the end-station UHV system. A range of evaporation sources and a variable temperature scanning tunnelling microscope for the preparation and characterization of artificially grown thin films and nanostructures will be available in a separate stand-alone system.

**Beamline and end-station performance targets:**

| | |
|---|---|
| Photon energy: | 8 – 150 eV |
| Energy resolution: | < 1 meV for hν < 50 eV, E/DE > $10^4$ over entire energy range |
| Higher harmonics contamination: | < 1 % |
| Polarization: | linear horizontal and vertical, left/right circular |
| Spectrometer resolution: | < 1 meV / 0.1° |
| Sample temperature range: | < 6 K to 450 K |
| Temperature stability: | < 20 mK over 12 hours |
| End-station vacuum: | < 5 $10^{-11}$ mbar, < 5 $10^{-12}$ mbar with cryo-shield |

# 5. Outline Beamline Specifications

### *5.1 Source*

It is important to realize that the resolution of practical ARPES experiments is limited by the available photon flux and not by the ultimate resolution of the monochromator or the electron spectrometer. Hence, only undulator sources are suitable for high performance experimental stations. The energy range is determined by the requirement of a high combined energy resolution (< 10 meV) in day-to-day operation. This can only be achieved at relatively low photon energy (8 – 80 eV), where the resolving power must be matched to the requirement of a high enough photon flux. In fact, the penalty for working at higher photon energies is two-fold: the resolving power of the monochromator needs to increase, while at the same time the cross section for photoemission decreases roughly proportional to hν-3. In order to explore selection rules from matrix elements or simply to optimizing the experimental signal it is advantageous to have full polarization control (linear horizontal, linear vertical, circular).

Based on these arguments a helical undulator would seem to be the most suitable device. The major issue is trading off the power output of such a device operating in linear mode with the requirement for high-photon flux. Table 1 shows the expected flux and power for several devices with different periods that can achieve energy of 8 eV. Clearly a 200 mm device delivering an energy of 8 eV would be a demanding source in terms of heat load. A useful trade-off could be a device using a 300 - 400 mm period. In addition harmonic rejection is crucial at low energies, which will require a quasi-periodic ID. It appears attractive to implement this device using electromagnetic technology but a decision to use permanent or electromagnetic technology would be taken after detailed considerations during implementation phase.

**Table I**: Photon flux at 8 eV and the total power emitted from different period undulators (DLS: 3 GeV, 300 mA)

| **Undulator Period** | **$B_{Max}$ [T]** | **No. of periods** | **$K_{Max}$** | **Flux at 8 eV [ph/s/0.1%BW]** | **Total Power [kW]** |
|---|---|---|---|---|---|
| 200 | 0.55 | 40 | 10.23 | $2.0 \times 10^{15}$ | 4.1 |
| 300 | 0.30 | 26 | 8.32 | $1.2 \times 10^{15}$ | 1.2 |
| 400 | 0.19 | 20 | 7.17 | $9.5 \times 10^{14}$ | 0.5 |
| 500 | 0.14 | 16 | 6.38 | $7.5 \times 10^{14}$ | 0.3 |

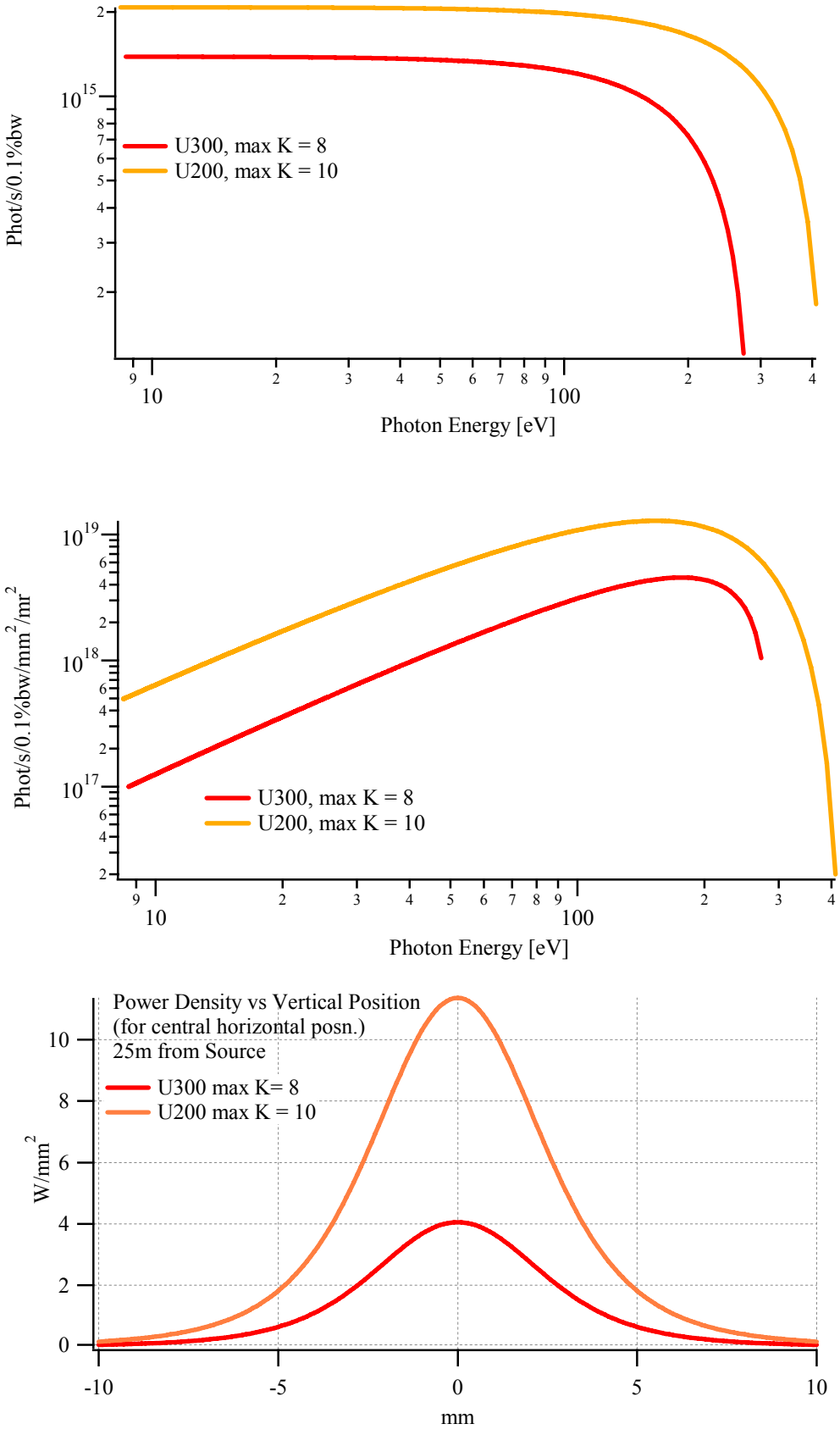


Fig 1.: Photon flux, photon brightness and power density for U200 and U300 (3GeV, 300 mA)

### *5.2 Beamline Optics*

The beamline optics is required to address a variety of competing requirements – the main ones being ability to provide a large energy range of 8-80 eV, to provide < 1 meV energy resolution over the core energy range and to provide adequate harmonic rejection.

The first optical element will have to absorb a large and variable power output from the ID. A possible solution is to use a side cooled SiC mirror at a relatively high grazing angle. This would of course need validation by thorough finite element analysis.

As regards the monochromator, a beamline design based on collimated plane grating monochromator (cPGM) has several merits and is also already being used at Diamond on two beamlines (I06 Nanoscience and I10 BLADE). It is therefore attractive to use a cPGM design for this beamline. The only limitation is that at very low energies (<30 eV) the desired harmonic rejection of <1 % would not be achievable with a cPGM even when using a quasi-periodic undulator. It is therefore essential to have another monochromator serving the lower part of the photon energy range of interest. For such low energies (8 < E < 30 eV) an off-Rowland circle normal incidence monochromator (NIM) is ideal. Making two monochromator branches would not only make the switching mechanism complex and expensive, it would also not offer straightforward and seamless scanning of the energy throughout the range. One possibility therefore is to combine the NIM and cPGM in to the same monochromator vessel as has been successfully demonstrated at the Surface and Interfaces Spectroscopy (SIS) beamline at the SLS. Minor changes to the well-established cPGM design can allow installation of an additional mirror and a normal incidence grating in to the cPGM vessel.

A preliminary beamline concept, inspired largely by the SLS design, is shown in Fig. 2. The first mirror collimates the beam and is used in the sagittal geometry to gain from the forgiveness to slope errors. The mono vessel contains two plane mirrors and two (or three) plane gratings. A horizontally deflecting focusing mirror follows the monochromator and focuses the beam on the exit slit. The distance between the focusing mirror and the exit slit would be set to a large value, subject to space constraints, to achieve high spectral resolution. The intermediate focus at the exit slit is then refocused by a toroidal or a KB-mirror pair to the sample position with a spot size of ~ 50 μm x 50 μm that is well matched to the requirements of the experiment.

For using the NIM monochromator, the plane mirror (M3) of the cPGM would be retracted from the beam path and a second plane mirror (M3) in the mono vessel would steer the beam on to the NIM grating. M4 would be intensely cooled to remove the high power density incident on it. The NIM grating would be very small in size – only about 10% in length of the cPGM grating and would be installed in the cPGM grating holder using an appropriate adaptor.

In addition, to provide for a branch line to facilitate nano-ARPES, a retractable mirror M6 can be brought in to the beam path to deflect the monochromatic beam to the branch line. Appropriate refocusing would have to be designed for the branch line to get < 1μm spot size.

Detailed ray tracing simulations would be done later for performance prediction of the beamline.

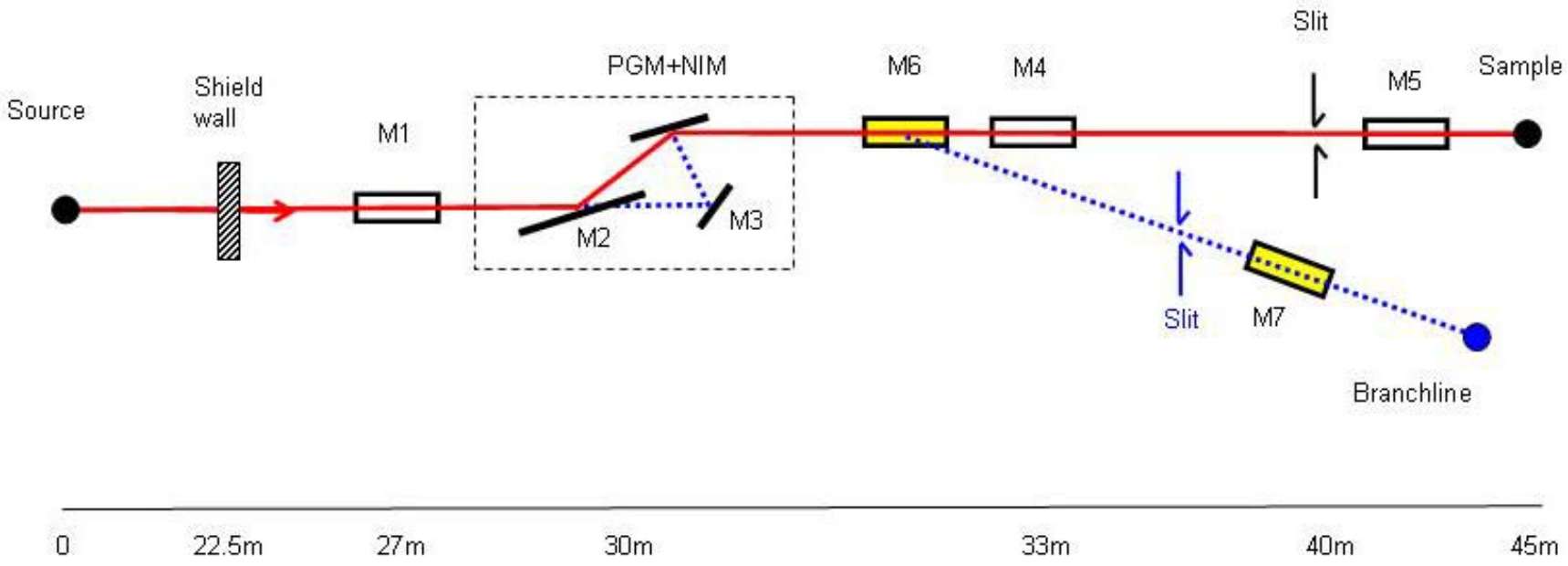


Fig 2: Beamline schematic