Diamond-II
Outline proposal for a flagship project – I21 hν^2-RIXS upgrade

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1. Impact statement

Beamline I21 is devoted to the study of advanced materials using high-resolution resonant inelastic soft X-ray scattering (RIXS). The instrument is best in class and transforming the way we probe the collective excitations in quantum matter such as cuprate superconductors, or the way we study vibrational spectra and electronic transitions in technologically relevant materials such as metal-ion batteries. Soft X-ray RIXS has been revolutionized in the last few years by the step change in the energy resolution to well below 100 meV. However, the low fluorescence yield in the soft X-ray regime limits its detection efficiency in applications, for instance, mapping the fine structure of the collective excitations in the full reciprocal space or speedy measurements of materials under various conditions (temperature, doping, voltage *etc.*). Here, we propose the novel hv^2-RIXS scheme as a major upgrade to the I21 Beamline. The new spectrometer will enable either (a) a factor of 5 - 10 times gain in photon throughput while maintaining or potentially improving the energy resolution with respect to the current RIXS setup or (b) imaging-RIXS with a few hundred nanometres spatial resolution to establish the study of collective excitations in phase separated advanced materials. The benefit in photon throughput and the imaging capability will put I21 in a unique position in the condensed matter physics and material science communities.

2. Scientific Case

Quantum matter exhibits emergent phenomena which only appear through the collective behaviour of many individual particles. The intimate coupling of spin, charge, orbital and lattice degrees of freedom often yields novel properties such as the metal-insulator transition, antiferromagnetic order, charge density waves (CDWs), multiferroicity, spin chirality, and superconductivity.

The collective modes of electronic ordering typically reach the sub-hundred meV energy range and is too weak to be detected by conventional spectroscopic techniques. RIXS, on the other hand, is highly sensitive to many relevant collective excitations with element and polarization selectivity. In the past decade, RIXS has shed new light on the physics of complex systems. For example, studies in cuprates have revealed that short-range spin fluctuations survive in a large area of the composition phase diagram [1]. These observations support spin fluctuations as one potential pairing mechanism for superconductivity. However, electron-phonon coupling (EPC), CDW correlations, and plasmons may also be crucial for the unconventional properties of cuprates. It is now widely known, mostly through RIXS, that short-range CDW fluctuations are ubiquitous in all underdoped cuprate superconductors (Figure 1A) [2-3]. RIXS studies on Bi-based cuprates also identified dispersive CDW excitations strongly interacting with the EPC indicating that the interplay between CDWs and the EPC is an important and potentially generic effect [4]. However, the exact form of CDW excitations and their interplay with phonons at the low energy (< 50 meV) in the full reciprocal space remains largely unexplored due to the low efficiency of high-energy resolution RIXS.

Cuprates are typical 3*d* transition metal oxides characterized with a strong on-site Coulomb interaction energy (U) and negligible spin-orbit coupling (λ). Moving towards 4*d* and 5*d* transition metal oxides, U becomes smaller and λ starts to influence the ground state properties. Theoretical investigations on materials containing 4*d* and 5*d* ions have uncovered a large number of unconventional electronic states arising from the competition between U and λ, including spin-orbit Mott insulators, axion insulators, Weyl semi-metals and other topologically non-trivial electronic states. Recently RIXS has been demonstrated as an ideal tool to probe the excitonic magnetism and the spin-orbit coupling physics in ruthenates [5]. Implementing RIXS up to 3 keV at the I21 is unique as no other RIXS facilities cover most 4*d* and 5*d* elements, however, the low diffraction efficiency of gratings at 2-3 keV may severely limit the throughput of this area of research.

The competition between various forms of electronic ordering often leads to phase separation or inhomogeneity. In the cuprate superconductor, $HgBa_2CuO_{4+y}$, charge-density-wave puddles, like steam bubbles in boiling water, have been found to have a highly inhomogeneous distribution (Figure 1B and 1C). Surprisingly, the quenched disorder, which arises from oxygen interstitials in spacer layers, has a distribution that is spatially anticorrelated with the charge-density-wave domains in the CuO_2 planes, because higher doping does not favour the stripy charge-density-wave puddles, leading to a complex emergent geometry of the spatial landscape for superconductivity [6]. Here, mapping the collective charge excitations and their competition with superconductivity over the sub-micron landscape will uncover the effects of competing orders far more accurately than current RIXS studies which average over a large area.

Interestingly inhomogeneity can sometimes lead to unexpected effects. $Re_{0.5}Ba_{0.5}MnO_3$ (where Re is a rare earth element) can be prepared both in ordered and disordered forms with respect to the Re-Ba distribution [7]. Remarkably, only the latter was found to exhibit colossal magnetoresistance. This suggests that when phases compete, the effect of (typically small amounts of) quenched disorder results in dramatic properties that are very different from those of a slightly impure material. Disorder in the regime of phase competition is not a mere perturbation; it alters qualitatively the properties of the material. Imaging the dynamical properties of the electronic phases in real space with sub-micron spatial resolution would be highly desirable, however, it remains almost an unexplored territory due to the limited availability of suitable probes.

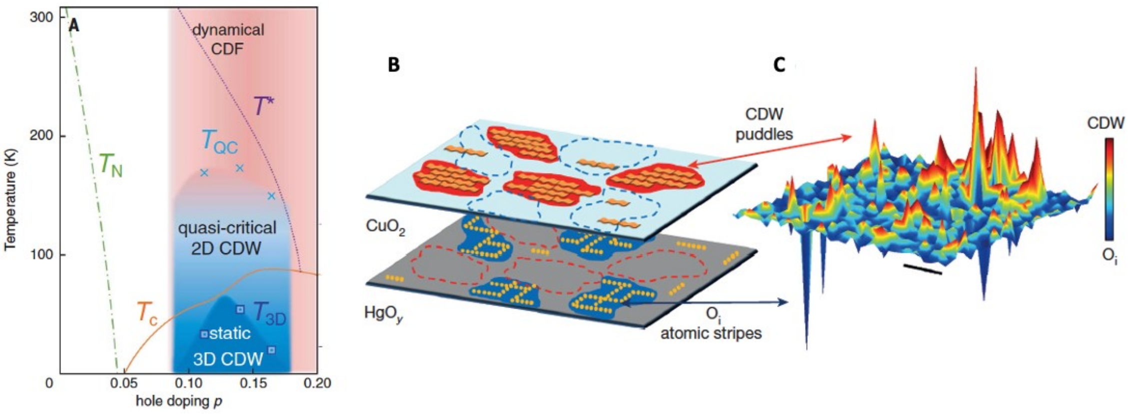

Figure 1 (A) Static and dynamic CDWs in the phase diagram of cuprates. Adapted from Ref. 2. (B) Schematic view of the anticorrelation between the CDW puddles and the Oi interstitial stripes in $HgBa_2CuO_{4+y}$. (C) Difference map of CDW and Oi regions generated using X-ray microdiffraction. Adapted from Ref. 7.

Going beyond quantum complex matter, RIXS has also facilitated a better understanding of energy materials. In a very recent study on the intercalation cathodes $Na_{\square}Li_xMn_{1-x}O_2$ (NLMO), it was found that the first-cycle voltage hysteresis is determined by the local ordering of the lithium and manganese ions in the transition metal layers. Specifically, the honeycomb superstructure of NLMO is lost on charging and partially driven by the formation of weakly-bounded molecular O_2 inside the solid. Here, owing to high resolution RIXS, the cathodes show characteristic vibrational spectra whose frequency unambiguously identified the presence of free molecular O_2 and explained the structural deformation on discharge [8]. As energy-mapping RIXS is particularly useful for these materials, improvement of the photon throughput by enlarging the incident energy bandwidth will represent a revolutionary change in high-throughput measurements of energy materials under various operational conditions.

There are no doubts that the ultra-high energy resolution (< 30 meV) RIXS will uncover more interesting phenomena in the aforementioned science areas. However, we face a big challenge by reducing the energy bandwidth in that the flux limitations do not allow us to probe the intrinsically low scattering signal with enough statistics. With an upgrade to Diamond-II, the reduction of the horizontal beam size and divergence will enable a new RIXS scheme with parallel detection of incoming and outgoing photon energies, the so-called hv^2-RIXS [9], which can (a) substantially increase the photon throughput while maintaining or improving the energy resolution for homogeneous materials, (b) deliver spatial resolution on the hundreds of nanometre scale enabling mapping of the collective excitations of the phase separations in advanced materials in real space.

3. Benefit to the Diamond research community

The upgrade to the hv^2-RIXS will have several implications for beamline I21. First, the direct benefit is the gain of the photon throughput (in the order of a factor of 5-10) compared to the current optical scheme. While improving the energy resolution, the relative gain will be even higher in comparison to the conventional RIXS setup with the same energy resolution. Second, by implementing the hv^2-RIXS concept, one could also start developing imaging RIXS capabilities with reasonable spatial resolution (hundreds of nanometre). High-energy resolution and spatially resolved RIXS is currently unavailable in any synchrotron facilities in the world. Implementation of such a scheme will certainly bring a major breakthrough for the condensed matter community. Third, imaging RIXS will be a complementary tool to other imaging techniques such as PEEM, STXM, nano-ARPES in studying advanced materials on Diamond-II. For the UK research community, the high photon throughput will enable I21 to contribute to energy materials research (*e.g.* transition-metal based Li/Na batteries, catalysts) in connection with the latest investments made in the Henry Royce Institute for Advance Materials, Maxwell Centre, Bragg Centre for Materials Research and the Faraday Institution.

4. Outline Specification

We propose to upgrade the I21 Beamline to accommodate the hv^2-RIXS spectrometer. More specifically, the current beamline optics and mechanics will largely remain the same apart from the upgrade of two mirrors to deliver a different refocusing scheme. The sample end station can be possibly recycled however major modifications need to be made to accommodate the new optics. The entire RIXS spectrometer has to be upgraded to the hv^2 concept with a completely new design, construction and delivery. *In addition, a compact hv^2-RIXS spectrometer with fixed scattering geometry may be considered for energy materials research in the future.*

Figure 2 illustrates the key difference between the conventional RIXS and the novel hv^2-RIXS optical schemes. Fundamentally, the focused X-ray spot at the sample acts as the secondary source of the spectrometer contributing to the total energy resolution. For the conventional RIXS scheme, since both the beamline and the spectrometer have the energy dispersive plane lying vertically (Figure 2A), the attempt to increase the photon throughput by opening up the exit slit will immediately deteriorate the energy resolution due to the enlarged vertical beam spot. In the hv^2-RIXS scheme, the spectrometer dispersive plane lies horizontally and orthogonal to that of the beamline (Figure 2B). Note that the hv^2 concept is impossible for the current Diamond electron optics and can only be achieved owing to the reduced horizontal beam size and divergence at Diamond-II. Under this configuration, the source for the spectrometer (*i.e.*, the horizontal beam spot) is maintained while increasing the exit slit opening.

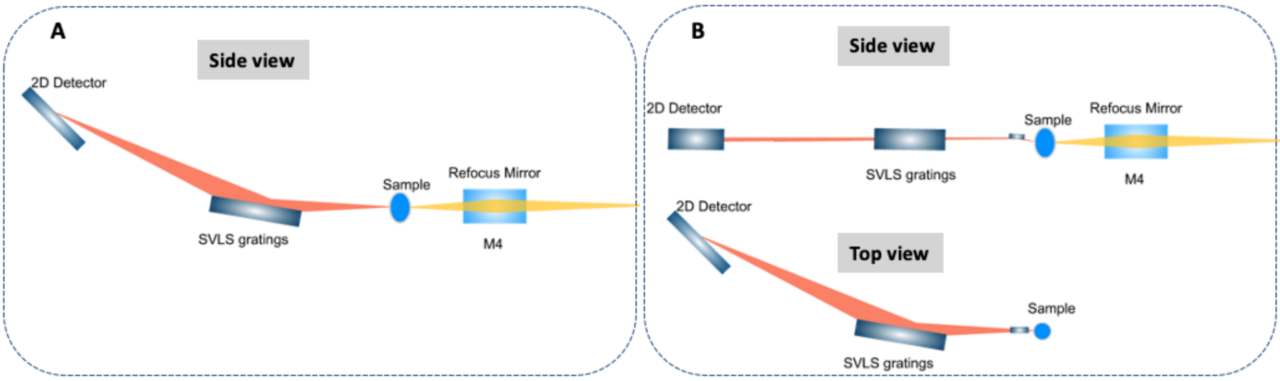

Figure 2 (A) The conventional RIXS optical scheme adopted at the I21-RIXS Beamline. (B) The hv2-RIXS concept to be employed on Diamond-II.

RIXS experiments demonstrated that the spectral profiles are invariable as long as the incoming photon bandwidth (*i.e.*, vertically staggered stripes) is kept within the full width at half maximum of the core-hole lifetime of the *L*-edges transition metal elements. For most 3*d* transition metal oxides, the throughput increase (the summation of the RIXS spectra excited by each X-ray stripe) is in the order of a factor of 5-10 for the current energy resolution at the I21 (*e.g.*, 33 meV at the Co *L*-edge). The relative gain of the throughput will be even higher if the energy resolution can be improved to 10-20 meV. For inhomogeneous samples, RIXS spectra resulting from each stripe of X-rays are magnified by the focusing optics and spatially resolved by the area detector. For a 1μm vertical beam size, such an optical scheme can deliver at least a 500 nm vertical spatial resolution with the current detector. Future improvements in detector technology will then very likely improve the spatial resolution of the imaging RIXS.

The outline specification is listed below:

- Source: APPLEII
- Energy range: 280 - 3000 eV;
- Energy resolution (E/ΔE): $3 \times 10^4 – 6 \times 10^4$ @ 1000 eV;
- Photon flux: 10^{11} - 10^{13} phs/s;
- Beam-size at sample spot: 0.5 (H) × 1.0 (V) μm^2
- Sample manipulator: Cryo-cooled six-axes manipulator
- Sample environment: low temperature, electric/magnetic field
- hv^2-RIXS spectrometer: ~ 15 m, VLS scheme, continuous rotation
- Enhancement of throughput: ×5 ~ ×10 compared to the current
- Spatial resolution: 0.5 (H) × 0.5 (V) μm^2

5. State of the art benchmark

Below we compare the proposed hv^2-RIXS beamline to the operational RIXS facilities worldwide.

Beamline	Diamond-II	Diamond	ESRF	NSLS-II	TPS
	I21-hν^2-RIXS	I21-RIXS	ID32	SIX	BL41A
Source	APPLE-II	APPLE-II	APPLE-II	EPU	APPLE-II
Energy range (eV)	280 - 3000	280 - 3000	450 - 1500	250 - 1500	400 - 1200
Energy resolution (meV) ΔE/E @ 930 eV	15 – 30	35 – 50	35 – 50	25 – 50	50
Photon flux (phs/s)	10^{11} - 10^{13}	10^{11} - 10^{13}	10^{11} - 10^{13}	10^{11} - 10^{13}	10^{11} - 10^{13}
Focused beam size at the sample H × V (μm^2)	0.5 × 1.0	35 × 2.5	50 × 3.5	6 × 1	3 × 3
Spatial resolution H × V (μm^2)	0.5 × 0.5	-	-	-	-

6. Community engagement

The Magnetic Materials Group and the I21 beamline team will organise science workshops to engage with the user community to refine the key specifications. A large number of international leading groups in condensed matter physics are already accessing I21 for their experiments and the magnetic materials group has strong links with other quantum materials groups. The full proposal will be developed in close connection with these UK and international communities.

7. References

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