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Towards 10 meV resolution: The design of an ultrahigh resolution soft X-ray RIXS spectrometer

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We present the optical design of the Centurion soft X-ray resonant inelastic X-ray scattering (RIXS) spectrometer to be located on the SIX beamline at NSLS-II. The spectrometer is designed to reach a resolving power of 100 000 at 1000 eV at its best resolution. It is also designed to have continuously variable 2θ motion over a range of 112° using a custom triple rotating flange. We have analyzed several possible spectrometer designs capable of reaching the target resolution. After careful analysis, we have adopted a Hettrick-Underwood spectrometer design, with an additional plane mirror to maintain a fixed direction for the outgoing beam. The spectrometer can cancel defocus and coma aberrations at all energies, has an erect focal plane, and minimizes mechanical motions of the detector. When the beamline resolution is accounted for, the net spectral resolution will be 14 meV at 1000 eV. This will open up many low energy excitations to study and will expand greatly the power of soft X-ray RIXS. *Published by AIP Publishing.* [<http://dx.doi.org/10.1063/1.4964847>]

I. INTRODUCTION

Resonant inelastic X-ray scattering (RIXS) in the soft X-ray range (nominally 200–2000 eV) has become an important scientific tool to study electronic and other low energy excitations in matter. RIXS allows one to study the energy, momentum, and polarization dependence of the photon scattering process, and hence the charge, spin, orbital, and lattice excitations of matter.¹ Furthermore, the soft X-ray energy range is home to the $1s \rightarrow 2p$ core level transitions of the second row elements, the $2p \rightarrow 3d$ transitions of the 3d transition metals, and the $3d \rightarrow 4f$ transitions of the rare earths, thereby offering direct access to the valence electronic states. The soft X-ray RIXS is thus a particularly powerful tool for condensed matter physics.

Following pioneering studies in the early 1990s,² soft X-ray RIXS instrumentation has seen a factor of 10 improvement in energy resolution during the last fifteen years, from ~ 1.5 eV in 1996 to ~ 100 meV in 2008 at 931 eV, the Cu L edge.¹ Until recently, the best operating RIXS instrument in the world was the 5 m long SAXES spectrometer at the Swiss Light Source, boasting an energy resolution of ~ 100 meV at 1000 eV.^{3,4} With this resolution, you can resolve for instance charge transfer and d-d excitations,^{5,6} spin excitations in cuprates^{7,8} and iron pnictides,⁹ high energy phonons,⁷ and vibrations in simple molecules.^{10,11} However, better energy resolution will open up RIXS to new science, some of which can be anticipated. Better resolution will allow the vibrational spectrum, and hence potential energy surfaces of more complex molecules to be studied. It will open up the study of spin dynamics in a large range of material with weaker magnetic coupling. It will be possible to observe superconducting gaps and pseudogaps in condensed matter systems. There are also expected applications to catalytic,

heterogeneous, nanostructured, and thin film materials.^{1,12} These are some of the foreseeable applications. But beyond that, improvement in resolution always leads to the discovery of new, unanticipated details about the excitation spectrum of materials.¹ The pursuit for higher resolution RIXS instruments in the soft X-ray energy range is currently underway at several major light sources, with different design choices being implemented. There are advanced RIXS beamlines at various stages of development such as ID32 ESRF (spherical grating spectrometer,¹³ which recently became operational¹⁴), I21 at Diamond (spherical grating spectrometer, under construction¹⁵), the Veritas beamline at MAX IV (Rowland circle spherical grating spectrometer, planned¹⁶), the high throughput instrument currently operational at TPS (AGM-AGS matched monochromator and spectrometer¹⁷). At NSLS-II, we are currently involved in an ambitious project to build a world leading RIXS beamline/spectrometer, with a goal of reaching 10 meV resolution on both the beamline and spectrometer, for a combined resolution of 14 meV at 1000 eV. The name of the beamline is SIX, for soft inelastic X-ray scattering, and the name of the spectrometer is Centurion, to represent the resolving power of 100k.

The inelastic scattering process which leads to a RIXS signal is inherently a low probability event; in fact, it is the resonant enhancement of the inelastic cross section by orders of magnitude near an atomic core level transition that makes this a viable technique at all in the soft X-ray energy range.¹ RIXS requires a high power undulator beamline, with the associated problem of thermally induced deformation of the pink beam optics. It also requires careful thought to the spectrometer design to achieve as high a throughput as possible. The quest for higher resolution presents many challenges. Optics quality must be the best available. In the limit of perfect optics and mechanical stability, the resolution of both the beamline and the spectrometer scale inversely with the length, which implies large instruments. Of course, nothing is perfect, and mechanical stabilities must be

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sub-micron in position and sub-100 nrad in angle to approach 10 meV resolution. We present here our spectrometer design to address these challenges.

II. ENERGY RESOLUTION OF A GRATING INSTRUMENT

We first set out the equations required to determine the resolution of a grating spectrometer. The grating equation states the diffraction condition (where k is the grating line density, α is the incident angle, β is the diffracted angle, m is the diffraction order, λ the wavelength, and E is the energy¹⁸)

$$\sin \alpha + \sin \beta = mk\lambda, \quad E = \frac{hcmk}{(\sin \alpha + \sin \beta)}. \quad (1)$$

From the perspective of the grating, a finite source size results in a finite spread ($d\alpha = \sigma_{src}/r_a$) of incident angles at a given point on the grating, where σ_{src} is the source size and r_a is the distance from the source to the grating. Differentiating Equation (1) with respect to α yields the source term. The source size σ_{src} is taken as the variance of the source distribution, which corresponds to the standard deviation if the distribution is Gaussian. We define the resolution as^{3,4,19}

$$|dE|_{src} = \frac{E^2}{hcmk} \cos \alpha d\alpha = \frac{E^2 \cos \alpha}{hcmk} \left(\frac{2.64\sigma_{src}}{r_a} \right). \quad (2)$$

From Equation (2), it is clear that grazing incidence favors better resolution, as well as a small source size and a large instrument. Working in higher order and high line density favors better resolution as well, but the loss in grating efficiency must be acceptable.

From the point of view of the grating, in a monochromator, the exit slit allows a finite range of diffracted angles ($d\beta = s_{exit}/r_b$) and hence energies to leave from the instrument, where s_{exit} is the exit slit size and r_b is the distance from the grating to the exit slit. Differentiating Equation (1) with respect to β yields the exit term^{3,4}

$$|dE|_{exit} = \frac{E^2}{hcmk} \cos \beta d\beta = \frac{E^2 \cos \beta}{hcmk} \left(\frac{s_{exit}}{r_b} \right). \quad (3)$$

The same general conclusions can be reached from this equation: small slit size, grazing angles, and a large instrument improve the resolution. For a monochromator, one has the option of reducing s_{exit} and shortening the output arm of the monochromator in such a way as to keep the ratio $\frac{s_{exit}}{r_b}$ constant; this maintains the resolution. Care must be taken that the aberrations in the focus at the exit slit are tolerable. However, this has non-trivial consequences for the design of the refocusing stage of the monochromator. The divergence increases proportionately, requiring either longer refocusing mirrors or a shorter refocusing stage with a reduced working distance and higher curvatures on the refocusing mirrors.

Equation (3) applies to a monochromator; in a spectrometer, the exit slit is typically replaced by a position sensitive detector, and the resolution is determined by a Gaussian like spatial resolution rather than the hard cutoff of an exit slit. If σ_{det} is the variance of the detector spatial resolution, then

$$|dE|_{exit} = \frac{E^2}{hcmk} \cos \beta d\beta = \frac{E^2 \cos \beta}{hcmk} \left(\frac{2.64\sigma_{det}}{r_b} \right). \quad (4)$$

In a high resolution grating instrument, the surface quality of the optics is a significant factor limiting the resolution, and cannot be ignored. For a grating, the slope error can be viewed as locally changing both α and β , and the contribution to the resolution can be expressed as²⁰ (where Δ_{gr} is the rms slope error on the grating),

$$|dE|_{gr} = 2.64\Delta_{gr} \frac{E^2}{hcmk} (\cos \alpha + \cos \beta). \quad (5)$$

If the optical design has additional optics, the slope error on these must also be considered. If the mirror is upstream of the grating, then slope errors act to broaden the range of incident angles on the grating, and the effects can be found from Equation (1) considering the effect on α (where Δ_{pm} is the slope error on the premirror),

$$|dE|_{gr} = 2.64(2\Delta_{pm}) \frac{E^2 \cos \alpha}{hcmk}. \quad (6)$$

The total resolution is found by adding the various components in quadrature. It is common to take the full width half-maximum of an assumed Gaussian distribution as the resolution criterion; we use the criterion proposed by Namioka¹⁹ which is $\sim 12\%$ more stringent.

Another contribution to the resolution is optical aberrations, which are specific to a given optical design and must be analyzed on a case by case basis. We use analytic expressions to obtain the vertical line space (VLS) coefficients, and evaluate the effect of aberrations by raytracing. Spectrometers typically are designed to have as large an angular acceptance as possible and their aberrations at large acceptance cannot be ignored.

III. CHOICE OF THE SPECTROMETER OPTICAL DESIGN

The resolution target for the RIXS spectrometer is to reach a resolving power of 100 000 at 1000 eV with enough flux for reasonable throughput. There has been a significant amount of work in the development of soft X-ray RIXS spectrometers, which leaves a variety of choices for the designer, and as always, the correct choice depends to a large extent on what goal is to be met. We have studied five designs with the potential to reach our resolution goals with respect to achieving the desired resolution, maximizing throughput, and minimizing mechanical motions to maximize stability. As a starting basis for reference, we have analyzed a Rowland circle (RC) design. In addition, we have analyzed a variable line spacing plane grating (VLS-PG) design, a VLS spherical grating design such as that used for the SAXES spectrometer at the Swiss Light Source (VLS-SG)⁴ under two possible operating constraints, and a VLS Hettrick-Underwood design (VLS-HU).^{21,22} A schematic drawing of the different designs is shown in Figure 1.

We analyze the designs using the analytical expressions for the successive terms in the path function expansion for the optical aberrations, assuming an on-axis point source. We consider the defocus, coma, and spherical aberration terms. We define the tuning curve as the mechanical motions required

FIG. 1. Schematic of the five spectrometer optical schemes, along with the required motions to zero all aberration terms up to coma. For the VLS-spherical grating design B, the addition of a horizontal translation to the grating allows one to keep the overall optical length constant. The bottom panel shows the tuning curves for the five spectrometer designs. The curves show the detector position as a parametric function of the energy, along with the angle of the focal plane relative to the optical axis. An angle of 90° therefore means that focal plane is exactly normal to the optical axis. A negative value for the angle implies that the detector must be facing downwards to match the focal plane, if the grating is reflecting upwards. For the VLS-SG:B design, the grating position which varies for this design is also shown. The curves are slightly displaced for clarity where required.

to zero the defocus and coma terms at all relevant energies. As will be shown, the tuning behavior varies significantly for the different spectrometer designs. Harada *et al.*²³ lists the expressions for the VLS-SG design. The expressions for a VLS-PG design can be found from the VLS-SG expressions by setting the radius of curvature to infinity. The expressions for a RC design can be found from the VLS-SG expressions by setting to zero all but the constant term for the grating line density expansion. When constrained to the Rowland circle, the defocus and coma term vanish. Amemiya *et al.*^{24,25} give the expressions for the VLS-HU design.²⁶

In the discussion here, for the sake of clarity, we only consider the optics needed for the vertical dispersion direction. A horizontal deflection parabolic collection mirror can be added to any of the designs and in principle does not affect the energy resolution. The energy range taken is from 180-2300 eV. The maximum overall length is taken as ~ 14.5 m. For designs involving a VLS grating, the line density k is varied

as a function of the longitudinal position along the grating x according to the following, where the positive x direction is taken as away from the source

$$k(x) = a_0 + a_1x + a_2x^2 + a_3x^3. \quad (7)$$

The VLS parameters are chosen to zero all aberration terms up to a_3 at 1000 eV, and the grating line density is chosen such that the spectrometer resolution is 10 meV at 1000 eV. The angle of incidence on the grating is taken as 88.3° for the Rowland circle spectrometer, and 88.3° at 1000 eV for all other designs. This angle is chosen as it represents a decent compromise between resolution and efficiency. A detailed analysis shows that the grating throughput as a function of the incident angle α , when the line density is adjusted to maintain constant resolution and weighted for the acceptance, varies little over an $\sim 1^\circ$ range, so the exact choice of α is not critical for the comparison. The vertical source size is taken as $0.6 \mu\text{m}$ FWHM, representative of the target beamline focus for the SIX beamline. The grating works in positive first order diffraction (inside order).

For the sake of resolution calculations, we assume a CCD detector based on an electron multiplying CCD chip,²⁷ run in photon counting mode utilizing charge centroiding for sub-pixel spatial resolution. Such a detector has a real spatial resolution of $<5 \mu\text{m}$ FWHM down to at least 530 eV.²⁸ The effective spatial resolution can be increased by tilting the detector. At 25° the effective spatial resolution is $2 \mu\text{m}$ FWHM, which is the value we will assume for these comparisons.

We have also considered a double dispersing design,²⁹ in which the exit slit of the beamline monochromator is imaged to create a dispersed polychromatic image at the sample. The scattered photons are then dispersed and imaged by a grating in the horizontal and reimaged onto a 2 dimensional detector in the vertical direction. Thus, you get the incident energy imaged along the vertical, and the energy loss spectrum dispersed along the horizontal. Depending on the design details,³⁰ one can collect in principle a substantial part of the 2 dimensional RIXS plane simultaneously, with an increased throughput. However, the optics required are nontrivial and must be near perfect in both the horizontal and vertical directions to obtain 10 meV resolution. This is not true for single dispersing designs, where the optical and mechanical tolerances are much less in the direction orthogonal to the spectrometer energy dispersion. Based on these considerations, we do not think it is possible at this time to reach 10 meV resolution with a double dispersing spectrometer. It is our assessment that this design is best suited for a lower resolving power spectrometer.

A. Rowland circle (RC) spectrometer

The RC spectrometer uses a spherical constant line density grating. The source, the grating pole, and the focus of the dispersed beam are constrained to lie on the Rowland circle. The design features several nice optical properties, in that the defocus and coma aberration terms are zero. However, this comes at a price: the focal plane is tangent to the Rowland circle and hence is near grazing. Furthermore, focusing at the grazing incidences required for soft X-rays requires a large

Rowland circle (~ 100 m for this design), and hence a very large translation of the detector to stay on the Rowland circle. The magnification of the RC design is exactly 1, and thus it is difficult to tailor the size of the image on the detector to the detector spatial resolution.

Figure 1 shows the tuning curve for a RC spectrometer that meets the resolution and energy range goals. Only r_2 and β need to be variable since coma is cancelled at all energies. The parameters are: $k = 3200 \text{ mm}^{-1}$, α fixed at 88.3° , $R = 117\,474 \text{ mm}$, and $r_1 = 3485 \text{ mm}$. These parameters give an overall spectrometer length of $\sim 14.5 \text{ m}$ at 1000 eV . Apart from the glancing focal plane, the problem with working on the Rowland circle is immediately apparent. A single grating requires an unreasonable large translation of the detector to cover a wide energy range. To go from 180 to 2300 eV requires a horizontal translation of over 16 m and a vertical translation to almost 6 m above beam height. This is clearly unworkable.

The mechanical difficulties of working on the Rowland circle are usually met by either limiting the photon energy range or providing multiple gratings to cover a given energy range. It is also possible to vary the incident angle by allowing r_1 to vary. These solutions are certainly workable for smaller instruments, but they do not address the grazing focal plane or the requirement for large motions inherent in the design, and become less and less palatable the larger the instrument. There is an additional limitation. Due to the constant line spacing of the grating, spherical aberrations are not canceled. This leads to a limited vertical acceptance, 1.6 mrad for this design.

B. VLS-plane grating spectrometer (VLS-PG)

The first step in the right direction is to consider a VLS-PG design. Using a plane figure for the grating allows one to achieve figure errors of $\sim 50 \text{ nrad}$, and thus to reach the highest resolution for a given line density. The focusing property and aberration correction afforded by the VLS grating allows one to work off of the "Rowland circle" and tune the spectrometer magnification to suit the detector, and at the same time achieve excellent resolution over a very wide energy range. The parameters are: $k = 1700 \text{ mm}^{-1}$ and $r_1 = 2920 \text{ mm}$. In this design, α , β , and r_2 must vary. The tuning curve is shown in Figure 1.

This curve has a remarkable feature. It is very well behaved. Over this large energy range, r_2 varies by only 75 mm . When considered in the laboratory frame, this requires a horizontal translation of the detector by only 375 mm . The vertical travel above beam height required is now a much more reasonable 2.7 m .

The design has one serious flaw. The angle of the focal plane is extremely grazing. In light of the fact that we prefer to use a soft X-ray CCD detector, we consider this essentially a fatal flaw of this design. We would be required to operate the detector at a reasonable angle of incidence ($> 15^\circ$). At such high angles relative to the focal plane, the resolution rapidly degrades as a function of position from the ideal focal point. Incremental improvements can be made in the angle of the focal plane by increasing the line density and increasing r_1 ; however, both of these options decrease the throughput.

C. VLS-spherical grating spectrometer (VLS-SG)

In this design, you have one additional optical parameter to utilize, the radius of curvature of the grating. By apportioning some of the focusing power to the substrate curvature and some to the focusing term of the VLS law, it is known that the angle of the focal plane can be raised.^{4,31,32} So we use this additional parameter to set the focal plane angle to 25° at 1000 eV . Under these conditions, the parameters are: $k = 3100 \text{ mm}^{-1}$, $R = 165\,000 \text{ mm}$, and $r_1 = 2500 \text{ mm}$.

1. VLS-SG:A

The minimal motions needed to zero the defocus and coma over the entire energy range are α , β , and r_2 . The tuning curve is shown in Figure 1. The problem of a grazing incidence focal plane is mitigated. If a single grating is used, the angle of the focal plane varies over a wide range and even inverts; however, it stays above 10° over the entire energy range. In principle, if a rotation is added to the detector, the detector face can be matched to the focal plane. Practically, however, since the detector needs to be cooled, it will be challenging mechanically to implement this rotation over such a large angular range. In addition, one finds that the travel required on the detector becomes prohibitive. The use of a single grating requires a horizontal throw of over 11 m and a vertical travel of nearly 5 m above the beam height. The usual solution of using several gratings to cover the entire energy range is possible, but cumbersome for so large an instrument. For a reasonable number of gratings, one will be left still with several meters of horizontal travel required.

From the perspective of the vertical acceptance, the performance is quite good. At 1000 eV , the instrument can in principle accept up to 3.4 mrad with little loss in resolution due to aberrations; in practice, with a 200 mm long grating this would be limited to 2.4 mrad .

2. VLS-SG:B

The VLS-SGA design has good vertical acceptance, an acceptable focal plane angle, and requires only one optic. However, it has poor tuning properties. It is possible to add an additional degree of freedom. One could allow the grating to translate in the horizontal, as is done for SAXES.⁴ Optically, this allows both r_1 and r_2 to be varied, in addition to α and β . This allows the defocus and coma aberrations to be zeroed with an additional constraint, which can be chosen to keep the spectrometer length constant ($r_1 + r_2 = \text{constant}$). We have solved the equations to zero defocus and coma with this constraint, taking the spectrometer total length to be 14.5 m . The parameters at 1000 eV are identical to the VLS-SG:A design.

The results are shown in Figure 1. By design, the detector behavior is very good and the total optical length of the spectrometer remains constant. In addition, the behavior of the focal plane is very good and varies by less than 10° over the entire energy range. The height of the detector stays below 3 m above beam height as well.

However, to achieve this good behavior at the end of the spectrometer, a single grating must travel 3.8 m to cover

the entire energy range. At the low end of the energy range, the grating is unreasonably close to the sample position. As stated previously, this difficulty can be addressed by limiting the energy range or using several gratings. However, the energy resolution is highly sensitive to the stability of the grating, and therefore we dislike complicating the grating mechanics by requiring a long horizontal travel on the grating stage.

D. VLS-Hettrick Underwood spectrometer (VLS-HU)

The VLS-HU design has excellent tuning properties. Three optical degrees of freedom are required for aberration cancellation: these are α , β , and r_2 . It is not necessary to vary r_1 . In addition, this design has a focusing mirror (we take this as a sphere) upstream of the grating, to cause the light to converge gently on the grating. This can be viewed as creating a virtual source for the grating, and hence r_1 is negative. We place this mirror at 1500 mm from the source, with an incident angle of 88.5° , and an image distance such that $r_1 = -r_2$ at 1000 eV.

The tuning properties are shown in Figure 1 and have nearly ideal properties. The motion required by the detector is very similar to the VLS-PG design. The change in r_2 is only 114 mm over the entire energy range, which when considered in the laboratory frame requires a horizontal translation of the detector by only 409 mm. The vertical travel above beam height required is only 2.8 m. But unlike the VLS-PG design, the problem of the grazing incidence focal plane has been solved. The focal plane stands nearly erect, within 10° of normal to the optical axis over the entire energy range. This is a well-known feature of the VLS-HU design.³³

E. Discussion

From a mechanical point of view, the tuning properties and the orientation of the focal plane of the VLS-HU design are clearly superior. It has the excellent tuning properties of a VLS-PG and a nearly upright focal plane over the entire energy range. The cost is one additional optic. Since RIXS is inherently a low count rate experiment, an important question is how does the throughput compare for the various designs.

Table I lists the optical parameters required for each design to reach 10 meV resolution at 1000 eV. Once the optical parameters are known, the resolution as a function of the vertical acceptance can be determined via ray tracing. For grazing incidence optical designs, geometrical aberrations are often the limiting factor in the V acceptance. It is possible to cancel the lower order terms with a VLS grating, but as the acceptance is increased the higher order terms kick in. We define the aberration free vertical acceptance as the point where the resolution is degraded by 10% relative to the ideal resolution.

Considering the aberration free vertical acceptance, we see that the RC design is the poorest performer. In principle, the VLS-PG design can accept 1.75 times more light in the vertical. The acceptance of the VLS-SG and VLS-HU designs is similar, ~ 2.2 times larger than a RC design. But this assumes that the grating is as large as it needs to be. If one considers

a grating of 200 mm length, for the VLS-PG and VLS-SG design, the actual acceptance is considerably less than what the aberration limit would be. In contrast, the VLS-HU design can physically accept more light than the aberration free limit, which gives one the option of running at higher throughput for general experiments and limiting the acceptance only when the highest resolution is required. This is due to the focusing mirror which puts converging light on the grating.

One must also consider the grating efficiency, which decreases with higher line density. The line density required to reach 10 meV resolution varies considerably among the various designs. It is lowest for the designs using a plane grating due to the lower figure error obtainable on a planar substrate. Of these, the central line density is higher for the VLS-HU design due to the need to compensate for the additional figure error on the premirror. The grating efficiencies were calculated at 1000 eV for a blazed grating with an ideal profile and are listed in Table I.

The relative throughput is then determined by the product of the premirror reflectivity, the vertical acceptance, and the grating efficiency relative to the poorest performer, which is the RC design. If one is not limited by the grating length, the VLS-plane design is significantly better than the rest. This conclusion is somewhat simplified, as it ignores the losses in detector efficiency that would result in operating the detector at a low incidence angle. However, this is very difficult to quantify and we do not attempt to do so. If one considers a grating whose length is limited to 200 mm, then the VLS-HU design is the best performer. Thus, even with the additional mirror the HVLS-HU design has the highest throughput. Considering everything, we find that designs based on a spherical grating require very large motions, which is incompatible with the very stringent mechanical tolerances required to reach 10 meV resolution. Another way of saying this is that spherical grating designs, which may be appropriate for smaller instruments, scale very poorly with size. The VLS-HU design overcomes these problems, and at the same time has a better vertical acceptance at the cost of increased optical complexity.

IV. CENTURION OPTICAL DESIGN

With the Hettrick-Underwood design being chosen, we now present the final design of the spectrometer and its expected performance. The final design must of course take into account real world geometric limitations, such as sample chamber dimensions, clearances required for other components, and mechanical stabilities of available mechanics. In addition, we incorporate several additional optics to increase the throughput and mechanical stability. A pair of horizontal parabolic collimating mirrors is added close to the sample to increase the horizontal acceptance. We also include a planar premirror, on a mount similar to that commonly used in a plane grating monochromator, to keep the output beam direction fixed and nearly horizontal. While this decreases the throughput, it eliminates the need to translate the detector several meters vertically, thus removing a major risk for mechanical instability.

TABLE I. Optical parameters and performance of the various spectrometer designs.

Design	Grating figure ^a			VLS law				Spectrometer parameters at 1000 eV						
	VFM	R (mm)	rms slope error (μrad)	a ₀ (mm ⁻¹)	a ₁ (mm ⁻²)	a ₂ (mm ⁻³)	a ₃ (mm ⁻⁴)	r ₁ (mm)	Focal plane γ (deg)	mag ^b	V acc ^c (mrad)	Image size 1σ ^d (μm)	Grating efficiency ^e	Relative throughput ^f
RC	...	117 474	0.1	3200	...	...	...	3485	5.4	1	1.6[1.7]	1.48[0.26]	0.067	1[1]
VLS-PG	...		0.05	1700	0.5976	-7.90 × 10 ⁻⁵	6.2 × 10 ⁻⁸	2920	2.0	1.65	2.8[2.0]	0.93[0.43]	0.125	3.26[2.2]
VLS-SG:A/B	...	165 000	0.1	3100	0.2616	-6.81 × 10 ⁻⁵	5.3 × 10 ⁻⁸	2500	25.3	1.54	3.5[2.4]	1.63[0.40]	0.069	2.25[1.5]
VLS-HU	Sphere ^g	...	0.05	2300	0.3913	-3.05 × 10 ⁻⁴	-2.8 × 10 ⁻⁷	2730	-83.8	3.23	3.7[4.4]	1.49[0.84]	0.092	2.19[2.5]

^aAll spherical optics can be replaced by the related circular cylinder.

^bMagnification = $\frac{\cos \theta}{\cos \theta_1}$. For the VLS-HU design, the magnification of the VFM must also be taken into account.

^cThe point at which the monochromatic image size increases by 10% at 1000 eV. The values in [] give the acceptance of a 200 mm long grating. In this case of the HU-plane design, this takes into account the effect of the focusing of the premirror.

^dValue in [] is the geometric size, given by the source size times the magnification.

^eGrating efficiency is calculated by GSolver V5.2 code for a blazed grating with an ideal profile and blaze angle.

^fThe relative throughput is given by the premirror reflectivity $\times$ vertical acceptance $\times$ grating efficiency, normalized to the RC design. The first value assumes that the grating can accept enough light to maintain the resolution to below 11 meV at 1000 eV; the second value in brackets gives the value limited by a 200 mm long grating.

^gR = 102934 mm, $\theta_{inc} = 88.5^\circ$, 0.1 μ rad rms slope error. The mirror is placed 1500 mm from the source. The reflectivity of Au is 0.69 at 88.5° incidence and 1000 eV.

FIG. 2. Conceptual optical scheme of the Centurion spectrometer.

A conceptual overview of the spectrometer is shown in Figure 2. An ellipsoidal refocusing mirror will focus the beam at the sample with a submicron vertical size, which acts as the source for the spectrometer. The sample will be manipulated by an in-vacuum diffractometer. To achieve the target resolving power with reasonable line densities on the grating, a ~ 15 m long spectrometer is required. It will be on a long beamline and housed in a dedicated satellite building. The spectrometer is a Hettrick-Underwood design, with variable line space (VLS) gratings, dispersing in the vertical. A pair of parabolic mirrors placed close to the sample will collimate the scattered light in the horizontal. A vertical focusing mirror focuses the beam beyond the grating, forming a virtual source point for the grating. A plane mirror upstream of the grating will allow the output direction from the grating to remain constant, eliminating the need to raise the detector as the energy is varied. The grating disperses and focuses the beam vertically onto the detectors. The spectrometer will have sufficient degrees of freedom to cancel the defocus and coma aberrations at all energies from 180-2300 eV. The ability to continuously vary the 2θ angle will be done by a novel combination of rotary flanges that we term the “triple rotating flange.”

A more detailed layout of the Centurion optical design is shown in Figure 3. Details of the optics are given in Table II. We will discuss the various optical components sequentially.

A. Source

The source for the spectrometer is an image of the monochromator exit slit demagnified by a factor of 6. The vertical size is therefore a function of the exit slit width. We envision three main resolution modes for the beamline, and in combination with the two spectrometer gratings this gives a range of achievable resolutions shown in Table III. To achieve the highest resolution, the exit slit on the beamline monochromator must be set to 5 μ m vertical, achieving a net resolution of 14 eV. The flux on the sample can be doubled and the net resolution increased by only 20% in the ultrahigh resolution mode by opening the monochromator exit slit to 10 μ m. The horizontal focus size is 2.2 μ m 1σ , however, what the spectrometer sees depends on both the angle of incidence of the beam on the sample and the scattering angle of the spectrometer.

B. Dual parabolic collimating mirrors (CM)

A pair of horizontal deflection collimating mirrors is placed close to the sample position to increase the horizontal

FIG. 3. Schematic diagram of the Centurion optics. Dimensions are in mm.

TABLE II. Optical design of the Centurion spectrometer.

Component	Distance from source (mm)	Optical dims. tan $\times$ sag (mm)	Incident angle ^a (deg)	rms slope error tan $\times$ sag (μ rad)	Figure	Comments
Source	0	---	---	---	---	Image of monochromator exit slit demagnified by 6
CM: collimating Mirror pair	275	250 $\times$ 10	88.5	0.5 $\times$ 1	Tangential parabolic cylinder H deflection	Collimates beam in horizontal
VFM: V focusing Mirror	1500	300 $\times$ 40	88.5	0.1 $\times$ 2	Tangential circular cylinder R = 102 700 mm, V deflection	Provides converging light onto grating
PM: plane Premirror	2375	525 $\times$ 40	varies	0.05 $\times$ 0.5	plane, V deflection	Redirection mirror to maintain constant Output direction
Gratings	2500	200 $\times$ 40	varies	0.05 $\times$ 0.2	VLS-plane, V deflection $a_0 = 2500 \text{ mm}^{-1}$ $a_1 = +0.415 80 \text{ mm}^{-2}$ $a_2 = +0.000 382 \text{ mm}^{-3}$ $a_3 = +4 \times 10^{-8} \text{ mm}^{-4}$	Disperses and focuses light onto detector Ultrahigh resolution, blaze angle = 1.6°
				0.1 $\times$ 0.4	$a_0 = 1250 \text{ mm}^{-1}$ $a_1 = +0.208 27 \text{ mm}^{-2}$ $a_2 = +0.000 356 \text{ mm}^{-3}$ $a_3 = +5 \times 10^{-9} \text{ mm}^{-4}$	High resolution, blaze angle = 1.0°
Detector	14 500	25H $\times$ 25V	0 to 65	---	---	2 side by side detector chips, ~5 mm central gap.

^aRelative to normal.

TABLE III. Resolution modes.

Mode	Beamline				Spectrometer		
	Grating (mm^{-1})	Mono exit slit (μm)	Focus size at sample H $\times$ V 1σ , (μm)	Res. (meV)	Grating (mm^{-1})	Res. (meV)	Net res. (meV)
Ultrahigh1	1800	5	2.2 $\times$ 0.26	10	2500	10	14
Ultrahigh2	1800	5	2.2 $\times$ 0.26	10	1250	23	25
High1	1200	17	2.2 $\times$ 0.82	23.5	2500	25 ^a	34
High2	1200	17	2.2 $\times$ 0.82	23.5	1250	49 ^a	54
Medium	500	30	2.2 $\times$ 1.45	60	1250	87 ^a	92

^aLimited by number of coherently illuminated grooves on grating.

acceptance of the spectrometer. They result in a pair of beams whose centers are 28.8 mm apart. This is close enough that if the optical width of the VFM, PM, and grating are 40 mm wide, then both beams can be captured with a single optic with no loss. Due to the fact that the mirrors are close to the sample, the collimation degrades

as the sample theta is rotated to more grazing angles. This affects only the horizontal width of the beam at the detector and does not degrade the energy resolution.

From one point of view, the design can be considered poor. The proximity of the mirrors to the source magnifies greatly the horizontal sensitivity of the spectrometer to the

lateral position of the source, and the pitch and lateral position sensitivity of the CM. In principle, however, this affects only the horizontal position of the beam on the detector and not the energy resolution. It will require careful alignment of the mirrors to achieve good collimation. In this case, since we are necessarily reducing the flux on the sample considerably by aiming for 10 meV incident bandwidth, we needed to maximize the horizontal acceptance. The reflectivity weighted angular acceptance at 1000 eV for a single CM in its current location is 18 mrad, versus ~ 12 mrad for a 1 m mirror placed outside of the sample chamber, a 50% increase in acceptance. Another option is to forgo the CM and simply place a row of detectors on the far end of the spectrometer; this may be an option for a smaller instrument. In our case, however, it would require a 90 mm wide grating and ~ 21 one inch wide detectors to have the same acceptance of the dual mirrors.

C. Vertical focusing mirror (VFM)

The VFM provides converging light onto the grating with the proper focusing power that the entrance arm length for the grating(s) is virtual and equal to r_2 in magnitude. There are two choices for the figure: a tangential circle and a tangential ellipse. In the arrangement here, the mirror is strongly magnifying ($\times 8.7$), so an ellipse focuses the beam with much less aberration. However, the tangential slope error is a very important contributor to the energy resolution. For this reason, we have opted for the tangential circular cylinder, which can be manufactured with better figure errors. The VLS grating can correct for the aberrations in the virtual source and raytracing shows that with the proper VLS law, the resulting image at the detector is identical with either figure.

D. Plane premirror (PM)

The PM redirects the beam to keep a constant output direction while allowing the included angle on the grating to change. This eliminates the need to translate the detector vertically. However, it results in a somewhat painful loss in throughput due to the extra reflection. The decision to go with this design was motivated solely by the primary motivation of the spectrometer, to reach a resolving power of 100 000, which requires very stringent mechanical stability.

The mirror will be mounted using the well-developed Reimer-Torge mechanism.³⁴ It is usual for monochromators to choose the output direction as parallel to the input direction. However, this is not required. If the deviation angle is chosen to deflect the beam further away from the grating, the incident angle required on the PM is more grazing (maximizing the reflectivity) but requires a longer PM. We have determined that we can keep the PM length reasonable with a 2° upward deviation relative to parallel output, resulting in a net 1° downward angle of the beam relative to horizontal after the grating. The required energy range can be covered for the two gratings with a vertical beam offset of 20 mm and a PM optical length of 525 mm.

The reflectivity loss on the PM for several constant deviation angles is shown for both gratings in Figure 4. The

FIG. 4. Premirror reflectivity for various constant deflection angles relative to parallel output of the premirror/grating pair.

ultrahigh resolution grating (2500 mm^{-1}) shows the larger loss due to the higher dispersion of the grating, which results in a smaller included angle. For a good part of the energy range, the reflectivity is above 0.5 for a deviation angle of 2° . The performance with a 2° deviation is considerably better than the parallel output, especially at higher energies.

Since we are working in positive first order (inside order), $|\beta| < \alpha$. Therefore, the effect of PM slope error is less, if the mirror is placed before the grating. Also, when working in positive first order the output beam is larger than the input beam by a factor of $\frac{\cos \beta}{\cos \alpha}$, resulting in a significantly longer mirror being required if it is placed after the grating.

The addition of a PM is not as innocent as it may first appear; it results in an inversion of both the image and the aberrations of the virtual focus of the VFM. This must be taken into account in determining the VLS law of the gratings.

E. Gratings

The spectrometer will be built initially with two VLS gratings, one for ultrahigh energy resolution (2500 mm^{-1}) and one for high and medium resolution but increased throughput (1250 mm^{-1}). The constraints imposed by the finite length of the PM will limit the energy range of the ultrahigh resolution grating to between ~ 250 and 2300 eV and the high resolution grating to ~ 180 - 1500 eV. The VLS laws are optimized at 1000 eV to focus the beam at 12000 mm from the grating and zero for both the coma and spherical aberrations. Because of the insertion of the PM, the equations given in Refs. 24 and 25 no longer work to determine the coma and spherical terms of the VLS law (a_2 and a_3) but the focusing term (a_1) remains identical. The a_2 and a_3 terms are therefore determined via raytracing with SHADOW.³⁵ As a further consequence, the tuning curve cannot be computed analytically. We have determined via raytracing, however, that the same tuning curve used for the spectrometer without the PM performs essentially the same.

In order to achieve the desired resolution, the number of coherently illuminated grooves on the grating must be equal to or greater than the desired resolving power. To estimate this, we determine the transverse coherent length at the grating, which is given by the van-Cittert Zernicke formula, $L_t = \frac{\lambda}{2} \frac{r}{s}$, where the source size s is given by the FWHM of the focus at the sample, and for this design, r is approximately given by the distance from the source to

the VFM, since this mirror nearly collimates the beam. The focus size at the sample, and hence the transverse coherent length at the sample depends on the size of the beamline exit slits. The number of coherently illuminated grooves is then given by the length of the projection of the coherent length onto the grating times the groove density. The values are given in Panel (c) of Figure 5 for the five resolution modes listed in Table III. The number of coherently illuminated lines is sufficient to reach the target resolving powers.

The harmonic rejection ratio of the spectrometer is shown in Panel (d) of Figure 5. This accounts only for the spectrometer optics, and does not take into account the beamline optics, slit settings, or undulator energy spectrum. The design is not optimized for harmonic rejection because RIXS is a resonant phenomenon, and for most RIXS experiments, harmonic

contamination is not a major concern. In the event that it is, such as when measuring an elastically scattered feature, using the higher line density grating will help in this regard.

F. Detector

The detector will be a CCD detector based on an electron multiplying CCD chip, run in the photon counting mode utilizing charge centroiding for sub-pixel spatial resolution. Such a detector has a real spatial resolution of $<5 \mu\text{m}$ FWHM down to at least 530 eV.²⁸ The advantage of using an electron multiplying CCD is that the effective read noise can be sub-electron per pixel. In the event that the photon incident rate is too high for centroiding, the camera can be operated in integrating mode, although with a high readout noise (25 e/pixel). If the centroiding works as anticipated, it should be possible to operate the camera normal to the incident beam to match the focal plane of the spectrometer; it will, however, be possible to rotate the camera down to 20° grazing incidence to increase the effective spatial resolution.

The image size on the detector in the ultrahigh resolution mode is $1\sigma_i = 1.63 \mu\text{m}$ at full V acceptance as determined by raytracing at 1000 eV. The detector spatial resolution in centroiding mode is $\sigma_D = 0.94 \mu\text{m}$ at 1000 eV.³⁶ With the detector at normal incidence, the ratio $\sigma_i/\sigma_D = 1.7$, close to meeting the Nyquist criterion, and this mode of operation should be sufficient for many situations.

To achieve the highest resolution, it will be necessary to tilt the detector. If the detector is operated at 25° grazing angle, then the ratio $\sigma_i/\sigma_D \sin(25^\circ) = 4$, enough to satisfy the Nyquist criterion. Operating this way adds the complication that the detector face no longer lies in the focal plane of the spectrometer, so the resolution degrades as you move away from the elastic peak. This resolution loss is very tolerable; for instance, at 1000 eV the resolution degrades by $<10\%$ over a 10 eV window.

G. Performance

The expected resolution is shown in Figure 5. All resolution calculations are done with the slope errors listed in Table II. The spectrometer meets the design goals in the ultrahigh resolution mode of 10 meV at 1000 eV. As is the nature of grating instruments, the resolution improves as you go to lower energies; the spectrometer will achieve ~ 3 meV resolution at the Ti L edge and ~ 1 meV at the C K edge. With similar resolution on the beamline monochromator, the resolving power will be significantly greater than 100 000 for some important absorption edges.

To estimate the throughput of the spectrometer, we compare the relevant design parameters with the successful 5 m SAXES spectrometer. We take into account the calculated beamline flux, the horizontal and vertical acceptance, and the grating efficiency. The parameters for the ADDRESS beamline/SAXES spectrometer are calculated from the parameters given in Refs. 3 and 4. The results at 1000 eV are shown in Table IV. In the medium resolution mode, Centurion is expected to have over 10 times the throughput at similar

FIG. 5. Expected spectrometer performance. The calculations are for a source size of $0.26 \mu\text{m} \times 5 \mu\text{m}$ H 1σ . Slope error is included for all the optics. Panel (a) left shows the analytically calculated spectrometer resolution versus energy. Panel (a) right shows the raytraced image at 1000 eV for the 2500 mm^{-1} grating, at the full vertical acceptance of 5 mrad. In addition to the source, detector, and slope error terms, the raytracing also accounts for any optical aberrations. The image formed is nearly Gaussian with a σ of $1.6 \mu\text{m}$, indicating the lack of significant aberrations. The horizontal collimation results in a straight line on the detector of ~ 7 mm wide, dependent upon the sample and detector θ , 2θ values. Panel (b) shows the number of coherently illuminated grooves on the gratings for the five resolution modes listed in Table III. Panel (c) shows the calculated 1st order diffraction efficiency for the two gratings. The calculations are done with GSolver, and assume a realistic antiblaze angle which is three times the blaze angle. Panel (d) shows the harmonic rejection ratio of the spectrometer, including the efficiency of all the spectrometer optics.

TABLE IV. Relative throughput of Centurion at 1000 eV.

	BL grating (mm ⁻¹)	Band-width (meV)	Flux at sample (ph/sec)	H acc. ^a (mrad)	V acc. (mrad)	VFM refl. ...	PM refl. ...	Spectr. grating (mm ⁻¹)	Grating efficiency ...	Net res. (meV)	Relative thruput ...
SAXES	800	~100	1×10^{13}	5.1	4	...	...	3200, laminar	0.05	~100	1
Centurion medium	500	60	9×10^{12}	36	5	0.7	0.66	1250, blazed	0.15	86	11
High	1200	23.5	2×10^{12}	36	5	0.7	0.54	2500, blazed	0.06	31	0.8
Ultrahigh	1800	10	1.5×10^{11}	36	5	0.7	0.54	2500, blazed	0.06	14	0.06
		12	3.0×10^{11}							16	0.12

^aReflectivity weighted acceptance.

resolution. In the high resolution mode, the performance is expected to be very similar to SAXES but with three times better resolution. In the ultrahigh resolution mode, the decrease in flux at the sample, an unavoidable consequence of reduced bandwidth, results in a throughput 8–17 times less than SAXES, albeit at up to 10x better resolution, depending on how hard you push for the minimal resolution. This reduced bandwidth will make measurements at this resolution challenging. However, another factor must be considered. The ultrahigh resolution mode will be used for measuring low energy excitations near the elastic peak. These features are expected to have resolution limited linewidths. In the case of a resolution limited linewidth, 10 times fewer photons measured at ten times better resolution results in the same peak height. The extraction of small features in a spectrum is very dependent upon the nature of the background, so we do not attempt to quantify this effect exactly. However, we expect that this “resolution advantage” will allow us to make ultrahigh resolution measurements in a reasonable amount of time at SIX.

H. Mechanical tolerances

Eventually the optical design will meet the real world and its vibrational disturbances from natural and cultural sources, temperature drifts, limitations in mechanical performance of real motion stages, etc. In addition, if one cannot achieve a given stability, then one must at least be able to measure the mechanical drift to apply some type of correction either with active feedback or post-correction of the data. The mechanical tolerances required for the most critical motions are listed in Table V. These have been determined by raytracing. Only the most sensitive motions are listed in the table. For the motions that affect the energy resolution, the value that degrades the resolution independently by 1 meV σ is given. If these motions are random, and the specified values are achieved, then the spectrometer resolution would degrade at most to 10.5 meV. Many of these tolerances are stringent, requiring submicron positional stability and sub-100 nrad angular stability. Some motions affect only the horizontal position of the beam on the detector, and therefore in principle do not affect the energy resolution.

The required mechanical tolerances led us to consider the adoption of a planar premirror mounted on a Riemer-Torge mechanism to keep the output direction fixed and eliminate the need to elevate the detector. The detector vertical sensitivity is less than 0.5 μ m. We considered this to be difficult to achieve for a height needing to vary up to 3.6 m above

ground level. Furthermore, the ability to measure to this level of positional accuracy (1 part in 10×10^6) is completely nontrivial on a stand that has to both rotate in 2θ and change height over meters. There is an additional subtlety not captured in the table. Any practical detector mount to achieve the detector height and longitudinal position (r_2) motions is likely to do so by a combination of horizontal and vertical translations. This means the longitudinal position of the detector then becomes critical as well, 1.5 μ m. With a fixed output direction design, the detector is at a fixed height and the optical axis is nearly horizontal. The longitudinal position sensitivity of the detector is then greatly relaxed, to 2.5 mm, due in part to the focal depth of the spectrometer (4 mm).

Nothing comes for free. First, you have the complication and reflectivity losses of an additional optic. The reflectivity losses can be minimized by careful design. The height and pitch stability of the premirror are stringent. In addition, they decrease the height tolerance of the VFM by a factor of 2. This is due to the action of the premirror on the virtual source formed by the VFM. However, we have had excellent experience with the current state of the art rotational stability achievable. Bestec, the supplier for the beamline monochromator, has achieved <15 nrad rms stability for periods up to 3 h for uncooled gratings in the beamline monochromator. In addition, with high resolution rotary encoders, it is possible to achieve <10 nrad resolution on the pitch measurement.

TABLE V. Mechanical stabilities.

Element	Motion	Tolerance	Comment
Focus at sample	Height	0.014 μ m	Note 1
	Lateral	10 μ m	Note 2
Dual parabolic mirrs.	Lateral	8 μ m	Note 2
	Pitch	16 μ rad	Note 2
	Roll	10 μ rad	Note 1
V focusing mirr.	height	0.17 μ m	Note 1
	Longitude	5 μ m	Note 1
	Pitch	0.05 μ rad	Note 1
plane premirr.	height	0.32 μ m	Note 1
	Pitch	0.07 μ rad	Note 1
grating	Height	0.72 μ m	Note 1
	Longitude	6 μ m	Note 1
	Pitch	0.03 μ rad	Note 1
Detector	Height	0.47 μ m	Note 1
	Lateral	25 μ m	Note 1 ^a

Note 1: affects energy resolution. Note 2: results in 500 μ m H shift of beam on detector.^aIf pixel rows are tilted.

FIG. 6. Triple rotating flange which allows access to a 2θ range of 112° while maintaining ultrahigh vacuum conditions.

Therefore, we are replacing the unknown and likely unmeasurable risks of large mechanical motions of the detector with known and measurable mechanical risks of a premirror.

We estimate that it will require ~ 1 h to obtain acceptable spectra in an ultrahigh resolution mode, and we therefore aim to achieve the specified tolerances for an hour. We do not envision any active feedback to compensate for mechanical motions and drifts. For short term (> 1 Hz) vibrational and temperature disturbances, we will attempt to compensate for by making the sample mechanics stand, optics and detector 2θ tracks as massive as possible, in order to minimize any amplification of the inherent floor vibrations and increase thermal damping as much as possible. The building itself has a massive air handler and is well insulated to maintain the temperature stability. We currently have achieved a temperature stability of 0.01 C/hr in the spectrometer area. In terms of long term drift, we consider temperature stability to be the most critical factor. It is interesting to note that the height of the sample plays no role in the energy resolution, if the sample face is in the vertical plane, and a little role otherwise.

I. Triple rotating flange

One of the goals of this spectrometer is to be able to collect data over a wide range of q space while maintaining ultrahigh vacuum in the spectrometer. The soft X-ray energy requires high vacuum conditions to limit the absorption of the X-rays. However, the requirement to keep the sample clean, and to cool down to temperatures of ~ 20 K requires ultrahigh vacuum conditions. This leads to the completely

nontrivial design problem of rotating a flange over a wide range of 2θ while maintaining ultrahigh vacuum. Our solution is to couple three differentially pumped rotary mechanisms as shown in Figure 6. When 2θ changes from a larger to a smaller value, Flange A rotates counterclockwise, Flange B rotates clockwise, and Flange C rotates counterclockwise (when viewed from the outside) in a coordinated manner such that the beam out port traverses in the horizontal plane. The full 2θ range allowed is 112° . There is not a unique solution to this problem; indeed other major spectrometer projects have adopted different approaches.³⁷ However, this approach has certain advantages. First, differentially pumped rotary mechanisms are a mature technology and have been used successfully in many ultrahigh vacuum applications. Second, the sample chamber remains stationary, which in practice is required to provide a stable base for the in-vacuum sample diffractometer, and for implementing other ancillary equipment such as a sample transfer system, cameras, and a wavefront analyzer. And finally, no new surfaces are exposed to vacuum during movement to a new 2θ value. This will limit the time required to return to base vacuum levels.

V. SUMMARY

The design of a high resolution soft X-ray RIXS spectrometer presents numerous design challenges. After careful analysis, we have adopted a Hettrick-Underwood spectrometer design. This design utilized a vertical focusing mirror to condense the beam gently onto the grating. Its use leads to a flat focal plane and greatly minimizes the mechanical

motions needed to cancel aberrations over the entire operating range. Despite the additional optical element, the throughput of the Hettrick-Underwood design is actually greater than the other designs. We have added an additional planar redirection mirror to keep the output beam direction fixed, eliminating the need for a large vertical translation of the detector. It is also designed to have continuously variable 2θ motion over a range of 112° using a custom triple rotating flange. The spectrometer is designed to reach a resolving power of 100 000 at 1000 eV at its best resolution. The success of this project will rest on achieving state of the art optics and maintaining very stringent mechanical tolerances for key motions. When the beamline resolution is accounted for, the net spectral resolution will be 14 meV at 1000 eV. This will open up many low energy excitations to study and will expand greatly the power of soft X-ray RIXS.

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¹L. J. P. Ament, M. van Veenendaal, T. P. Devereaux, J. P. Hill, and J. van den Brink, *Rev. Mod. Phys.* **83**, 705 (2011).

²Y. Ma, N. Wassdahl, P. Skytt, J. Guo, J. Nordgren, P. D. Johnson, J.-E. Rubensson, T. Boske, W. Eberhardt, and S. D. Kevan, *Phys. Rev. Lett.* **69**, 2598 (1992).

³V. N. Strocov, T. Schmitt, U. Flechsig, T. Schmidt, A. Imhof, Q. Chen, J. Raabe, R. Betemps, D. Zimoch, J. Krempasky, X. Wang, M. Grioni, A. Piazzalunga, and L. Patthey, *J. Synchrotron Radiat.* **17**, 631 (2010).

⁴G. Ghiringhelli, A. Piazzalunga, C. Dallera, G. Trezzi, L. Braicovich, T. Schmitt, V. N. Strocov, R. Betemps, L. Patthey, X. Wang, and M. Grioni, *Rev. Sci. Instrum.* **77**, 113108 (2006).

⁵P. Kuiper, H.-H. Guo, L.-C. D. C. Sthe, J. Nordgren, J. J. M. Poethuizen, F. M. F. de Groot, and G. A. Sawatzky, *Phys. Rev. Lett.* **80**, 5204 (1998).

⁶Y. Harada, T. Kinugasa, R. Eguchi, M. Matsubara, A. Kotani, M. Watanabe, A. Yagishita, and S. Shin, *Phys. Rev. B* **61**, 12854 (2000).

⁷L. Braicovich, J. van den Brink, V. Bisogni, M. M. Sala, L. J. Ament, N. B. Brookes, G. M. D. Luca, M. Salluzzo, T. Schmitt, V. N. Strocov, and G. Ghiringhelli, *Phys. Rev. Lett.* **104**, 077002 (2010).

⁸M. Guarise, B. D. Piazza, M. M. Sala, G. Ghiringhelli, L. Braicovich, H. Berger, D. v. d. Marel, J. N. Hancock, T. Schmitt, V. N. Strocov, L. J. Ament, J. van den Brink, P. H. Lin, P. Xu, H. M. Rnow, and M. Grioni, *Phys. Rev. Lett.* **105**, 157006 (2010).

⁹K. J. Zhou, Y. B. Huang, C. Monney, X. Dai, V. N. Strocov, N. L. Wang, Z. G. Chen, C. Zhang, P. Dai, L. Patthey, J. van den Brink, H. Ding, and T. Schmitt, *Nat. Commun.* **4**, 1470 (2013).

¹⁰F. Hennies, A. Pietzsch, M. Berglund, A. Fhlisch, T. Schmitt, V. Strocov, H. O. Karlsson, J. Andersson, and J.-E. Rubensson, *Phys. Rev. Lett.* **104**, 193002 (2010).

¹¹A. Pietzsch, Y. P. Sun, F. Hennies, Z. Rinkevicius, H. O. Karlsson, T. Schmitt, V. N. Strocov, J. Andersson, B. Kennedy, J. Schlappa, A. Fhlisch, J. E. Rubensson, and F. Gel'mukhanov, *Phys. Rev. Lett.* **106**, 153004 (2011).

¹²T. Schmitt, F. M. F. de Groot, and J.-E. Rubensson, *J. Synchrotron Radiat.* **21**, 1065 (2014).

¹³See <http://www.esrf.eu/UsersAndScience/Experiments/ElectStructMagn/ID32> for information on the new RIXS beamline at ESRF.

¹⁴N. Brookes, Invited presentation at 12th International Conference on Synchrotron Radiation International, New York, USA, 6–10 July 2015.

¹⁵See <http://www.diamond.ac.uk/Beamlines/Spectroscopy/I21.html> for information on the RIXS beamline under construction at the Diamond Light Source.

¹⁶See <https://www.maxlab.lu.se/veritas> for information about the RIXS beamline under construction at MAX IV.

¹⁷C. H. Lai, H. S. Fung, W. B. Wu, H. Y. Huang, H. W. Fu, S. W. Lin, S. W. Huang, C. C. Chiu, D. J. Wang, L. J. Huang, T. C. Tseng, S. C. Chung, C. T. Chen, and D. J. Huang, *J. Synchrotron Radiat.* **21**, 325 (2014).

¹⁸We use the convention that the angles are measured relative to the surface normal; α and β have opposite signs if they are on opposite sides of the normal; it follows that positive orders are the inside orders.

¹⁹T. Namioka, T. Haga, and H. Kinoshita, in *JSPE Proceedings on Soft X-ray Optics* (Japan Society for Precision Engineering, Tokyo, 1997), pp. 185–198.

²⁰M. R. Howells, "X-ray data booklet," in *Center for X-ray Optics and Advanced Light Source*, 3rd ed. (Lawrence Berkeley National Laboratory, CA, USA, 2009), Chap. 4.3.

²¹M. C. Hettrick, J. H. Underwood, P. J. Batson, and M. J. Eckart, *Appl. Opt.* **27**, 200 (1988).

²²T. A. Callcott, W. L. O'Brien, J. J. Jia, Q. Y. Dong, D. L. Ederer, R. N. Watts, and D. R. Mueller, *Nucl. Instrum. Methods Phys. Res. A* **319**, 128 (1992).

²³T. Harada, K. Takahashi, H. Sakuma, and A. Osyczka, *Appl. Opt.* **38**, 2743 (1999).

²⁴K. Amemiya, Y. Kitajima, T. Ohta, and K. Ito, *J. Synchrotron Radiat.* **3**, 282 (1996).

²⁵K. Amemiya and T. Ohta, *J. Synchrotron Radiat.* **11**, 171 (2004).

²⁶We have found empirically that the expressions in the above two references work if the incident angle on the vertical focusing mirror, θ_A , is taken to be negative.

²⁷The CCD sensor is model CCD207-40 back illuminated 2-phase IMO Series electron multiplying CCD sensor manufactured by e2v.

²⁸M. R. Soman, D. J. Hall, J. H. Tutt, N. J. Murray, A. D. Holland, T. Schmitt, J. Raabe, and B. Schmitt, *Nucl. Instrum. Methods Phys. Res. A* **731**, 47 (2013).

²⁹V. N. Strocov, *J. Synchrotron Radiat.* **17**, 103 (2010).

³⁰T. Warwick, Y.-D. Chuang, D. L. Voronov, and H. A. Padmore, *J. Synchrotron Radiat.* **21**, 736 (2014).

³¹C. Dallera, E. Puppini, G. Trezza, N. Incorvaia, A. Fasana, L. Braicovich, N. B. Brookes, and J. B. Geodkoop, *J. Synchrotron Radiat.* **3**, 231 (1996).

³²V. N. Strocov, T. Schmitt, U. Flechsig, L. Patthey, and G. S. Chiuzaian, *J. Synchrotron Radiat.* **18**, 134 (2011).

³³M. C. Hettrick, *Nucl. Instrum. Methods Phys. Res. A* **266**, 404 (1988).

³⁴F. Riemer and R. Torge, *Nucl. Instrum. Methods* **208**, 313 (1983).

³⁵B. Lai and F. Cerrina, *Nucl. Instrum. Methods Phys. Res. A* **246**, 337 (1986).

³⁶M. R. Soman, D. J. Hall, J. H. Tutt, N. J. Murray, A. D. Holland, T. Schmitt, J. Raabe, and B. Schmitt, *J. Instrum.* **8**, C01046 (2013).

³⁷C.-J. Englund, M. Agker, P. Fredriksson, A. Olsson, N. Johansson, J.-E. Rubensson, and J. Nordgren, *Rev. Sci. Instrum.* **86**, 095110 (2015).