DESIRS : a state-of-the-art VUV beamline featuring high resolution and variable polarization for spectroscopy and dichroism at SOLEIL

L Nahon, N de Oliveira, G A Garcia, J-F Gil, D Joyeux, B Lagarde and F Polack

Synchrotron SOLEIL, l’Orme des Merisiers, St Aubin, BP 48, 91192 Gif sur Yvette Cedex, France

E-mail: laurent.nahon@synchrotron-soleil.fr

Abstract. DESIRS is a new undulator-based VUV beamline at SOLEIL (France) optimized for the study of gas phase matter in the 5-40 eV range. It is equipped with two dedicated endstations: a VUV Fourier-Transform Spectrometer (FTS) for ultra-high resolution absorption spectroscopy (resolving power up to $10^6$) and an electron/ion imaging coincidence spectrometer. The photon source is a 10 m-long pure electromagnetic variable polarization undulator providing, at the sample location, fully calibrated quasi-perfect horizontal, vertical and circular polarizations. The optical design includes a beam waist allowing the implementation of a gas filter to suppress the undulator higher harmonics. The 6.65 m Eagle off-plane Normal Incidence Monochromator equipped with four gratings allows the tuning of the flux-to-resolution trade-off. Measured ultimate instrumental resolving powers are 124000 (174 µeV) around 21 eV and 250000 (54 µeV) around 13 eV, while the typical measured flux is in the $10^{10}$-$10^{11}$ ph/sec range in a 1/50000 bandwidth and $10^{12}$-$10^{13}$ ph/sec in a 1/1000 bandwidth.

1. Introduction
In the last two decades several undulator-based VUV beamlines have been constructed providing an optimal flux-to-resolution compromise [1-6] and, in some cases, variable polarization [7-10]. Here we present the concept and performance of DESIRS, a VUV undulator-based beamline operational at SOLEIL since 2008, covering the 5-40 eV range for the study of dilute matter (cold molecules, radicals, biological species, chiral systems, clusters and nanoparticles) as well as of condensed matter. DESIRS is designed for high resolution, spectral purity, variable polarization and high flux.

2. Optical conception
The conception, construction and performances of the HU640 undulator, allowing the production of controlled tailored ellipses of polarization, to adjust for the neither purely grazing nor normal incidence on the optics, has been described elsewhere [11; 12].
DESIRS is designed to provide harmonic-free photons, owing to the presence of a gas filter [13], to three independent endstations: (i) a white beam branch feeding a novel wave-front division-based interferometer Fourier-Transform Spectrometer (FTS) [14]. This unique instrument for VUV absorption spectroscopy, offers unprecedented resolving power in the $10^6$ range [15] over a wide spectral range corresponding to the undulator envelope (see figure 1). Located downstream of a dedicated versatile sample environment chamber, the FTS is the core of the permanent absorption facility of the beamline. (ii) Two monochromatized branches A and B. On branch A, a molecular beam endstation equipped with an imaging electron/ion coincidence spectrometer [16] is permanently installed, while branch B is designed to accommodate different experimental chambers from external users.

The general optical layout is presented in figure 2 and the main characteristics of the optics as well as the detailed description of the actual constituting elements of the beamline can be found elsewhere [12]. Briefly, the SR emitted by the undulator impinges onto a first pair of mirrors, M1 and M2, deflecting the beam in the horizontal plane. M1 is planar and takes most of the heat load (up to 120 W), while M2 is toroidal in order to achieve a tight focus (~ 0.3 mm diameter) at the gas filter center. The incidence angle (70°) and reflecting material (Si) of M1 and M2 have been chosen so that virtually no photons with energies above 60 eV are transmitted downstream. After the beam-waist created in the middle of the gas filter, the SR can be steered and refocused towards the FTS endstation by inserting the toroidal MFT mirror, for absorption spectroscopy experiments. For all other types of experiments, this mirror is removed from the beam to let it continue towards the pre-focusing elements (M3 and M4) of the 6.65 m Normal Incidence Monochromator (NIM). M3 and M4 deflect the beam in the vertical plane with the same incidence (70°) and reflecting material (Si) as M1M2. We have chosen to keep the toroid M4 and apply the shape correction to the otherwise flat mirror M3. This corrector plate includes a meridional S shape (coma correction). M3+M4 realize a strong demagnification (factor ~18) of the beam in the vertical plane to maximize the flux throughput via the entrance slit (EnS) - typically a measured throughput of 60 % (resp. 90 %) in 10 µm (resp. 20 µm) EnS - and to ensure a correct illumination of a maximum number of grating lines.

After M4 the SR enters the Eagle off-plane 6.65 m NIM which has been transferred from the former SU5 beamline at Super ACO, described in detail elsewhere [2; 17; 18]. In addition to the two
highly-dispersive gratings with 2400 and 4300 grooves/mm, two extra low dispersion gratings with 200 and 400 grooves/mm have been added for the performance of flux-hungry experiments on DESIRS, providing tunable resolving powers (RP=\(\lambda/\Delta\lambda\)) from the 100000 range down to about 100.

![Figure 2](image.png)

**Figure 2.** Optical layout of the beamline: (top) side view; (bottom) bird’s view. Red rays (resp. green rays) correspond to the undispersed white beam (resp. monochromatized beam). 4QPD stands for 4-quadrants Photodiode, EnS (resp. ExS) for entrance (resp. exit) slit. Reproduced with permission of the International Union of Crystallography.

After the Exit slit (ExS), the monochromatized SR is refocused and steered towards the experimental points A or B by the toroidal mirror M5. An in vacuo, insertable 2 x 3 reflections-based VUV polarimeter transferred and adapted from the SU5 beamline [7], has been installed in the branch A post-focusing arm, after the last optic, providing, without any assumption, the whole set of Stokes parameters defining the polarization ellipse of the incoming photons.

### 3. Performances

#### 3.1. Spectral resolution

This section describes the high spectral resolution that can be achieved by the 6.65 m NIM equipped with its two highly-dispersive gratings: G2, the 2400 line/mm grating (uncoated SiC) optimized for the 4-20 eV range and G4, the 4300 line/mm grating (Pt-coated silica) optimized for the 15-40 eV range. The general capabilities of this NIM equipped with the same two gratings, installed on the former SU5 beamline, have been described in great detail elsewhere [17]. We focus here only on the ultimate resolution capabilities brought by its implementation on the DESIRS beamline at SOLEIL.

The 4300 line/mm grating (G4) was tested on Ne. In figure 3, we show an autoionization spectrum recorded in between the two 2p\(^t\) thresholds with 5 \(\mu\)m/10 \(\mu\)m (EnS/ExS). This spectrum shows excellent long term-stability over the ~3 hours-long acquisition. A Gaussian fitting profile of the 18s’ line, as shown in the insert of figure 3, leads to an instrumental bandwidth of 174 \(\mu\)eV, corresponding to a resolving power of 124000. This is a slightly better performance than that obtained on SU5 with 8 \(\mu\)m slits (184 \(\mu\)eV), showing that the ultimate resolution with this grating is mainly limited by the slope errors (0.8 \(\mu\)rad) below 15 \(\mu\)m slits. Nevertheless to our knowledge, this resolving power is the highest ever published in this energy range for a scanning monochromator. At lower photon energies,
the limitations induced by the slope errors of the 4300 line/mm grating are less severe so that on the 16s’ line of Xe around 13.34 eV it has been possible by closing the slits down to 5 µm, to reach an instrumental state-of-the-art resolving power of 249000 (53 µeV).

![Figure 3](image.png)

**Figure 3.** Autoionization spectrum of Ne (total ion yield) recorded in between the two 2p-1 thresholds with 5 µm entrance slit / 10 µm exit slit and the 4300 lines/mm grating. The insert shows a blow-up of the 18s’ resonance and an instrumental Gaussian fit showing a 174 µeV linewidth (resolving power 124000). Reproduced with permission of the International Union of Crystallography.

The 2400 lines/mm grating (G2) was tested on several rare gases. The analysis of the autoionization spectrum of the Xe 16s’ line, obtained with slits of 5 µm provides an instrumental Gaussian linewidth of 56 µeV, corresponding to a resolving power of 238000, instead of an ultimate resolving power of 155000 on SU5 with 10 µm slits. The corresponding spectral width (3.9 mÅ) has been dramatically reduced as compared to SU5 (6 mÅ) by closing the slits from 10 to 5 µm, showing that in the case of G2, featuring much smaller slope errors (0.47 µrad), the ideal pure slit-limited bandwidth behavior is maintained well below 10 µm slits. This slit closing was possible on DESIRS because of the low emittance of the source and the performance of the pre-focusing optical system.

### 3.2. Absolute flux

A summary of the flux performances of DESIRS is presented in figure 4, showing for the 4 gratings the absolute available flux measured at the sample location with a calibrated Si photodiode (AXUV100, IRD). The data correspond to a 1/50000 bandwidth for the highly dispersive gratings G2 and G4, and to a 1/1000 bandwidth for the low dispersion gratings G1 and G3.

Over the 5-32 eV range, the typical flux is $10^{10}$-$10^{11}$ photons/sec in a 1/50000 bandwidth and $10^{12}$-$10^{13}$ photons/sec in a 1/1000 bandwidth before falling to lower values above 32 eV, because of the low reflectivity on the normal-incidence gratings at high energy. The comparison with optical simulations is quite interesting. At low photon energies, below 10 eV, the agreement between the measurements and the simulation is quite satisfactory while, with increasing energy, the measured data are about an order of magnitude below the simulations. This trend cannot be explained by some limiting angular apertures on the beamline, which would favor high photon energies. The main cause of discrepancy probably lies in an overestimation of the Si reflectivity from the literature data, possibly due to the fact that the crystalline state considered in these data may not be fully representative of the actual surface layer of our polished silicon mirror. Even a small discrepancy can have a dramatic effect, considering that 5 Si-mirrors have to be taken into account. However, despite a quite successful handling of the carbon contamination problem [12; 19] it is likely that residual contamination is probably still present.
on some of the optics, which may decrease the measured flux. Despite the lower than modeled flux, the measured flux level is very satisfactory, allowing the achievement of the whole initial scientific program, and represent the state of the art.

Figure 4. Absolute harmonic-free photon flux for a 500 mA current within the full beamline aperture (0.65 x 0.65 mrad²), in the linear vertical polarization mode, for the 4 gratings of DESIRS with different resolving powers (RP). Symbols correspond to the measured data (Si photodiode) and lines correspond to optical simulations. Reproduced with permission of the International Union of Cristallotherapy.

3.3. Polarimetry

One of the features of DESIRS is the variable polarization capability of the undulator associated with the ability of measuring in vacuo at any time, within typically 30 minutes, the polarization ellipse, just upstream the sample, with a dedicated VUV polarimeter [7]. To our knowledge, DESIRS is the only variable polarization beamline in the world equipped with such a permanently-available device. This allows full disentanglement of the polarization as given by its Stokes parameters decomposition:

\[ \sqrt{S_1^2 + S_2^2 + S_3^2 + S_4^2} = 1 \]  

where \( S_1 \) (resp. \( S_2 \)) is the normal (resp. 45° tilted) linear component, \( S_3 \) is the normalized circular component and where \( S_4 \) is the normalized unpolarized component. Such a measurement is critical in order to build up the calibration table in the Circularly Polarized Light (CPL) mode of operation, to check for the effect of Carbon contamination onto \( S_4 \), as well as to know the absolute circular polarization rate \( S_3 \), needed to normalize CD signals [20]. We have published [12] 3 examples of polarization ellipses recorded at 20 eV. The ellipses “LV” and “LH” have been recorded respectively in the pure vertical and horizontal linear polarization modes. These measurements are extremely satisfactory since they do correspond to quasi-perfect linear polarization with an absolute \( S_1 \sim \pm 0.99 \).

In the more challenging case of the CPL mode of operation, one has to determine which polarization ellipse of undulator emission leads, at the sample location, to a pure left or right-handed CPL (l-CPL and r-CPL). Within a single iteration, it is possible to minimize the residue of linear component (\( S_1 \) and \( S_2 \)) to a negligible amount (below 1 or 2 %), leading to a quasi-perfect CPL as shown in [12], where \( S_1 = 0.99 \). This shows that the insertion device behaves as expected from the helical undulator algebra [21; 22]. We checked also that the helicity switching of light is simply achieved by switching the polarity of the current driving the coils producing the horizontal magnetic field, leading to the same absolute value of \( S_3 \). Note that this switching is currently done within ~ 20 sec (DC mode), a figure that should be reduced to ~ 1 sec in the near future (AC mode).
Presently, the absolute circular polarization rates $S_3$ in the CPL mode, which are the actual figures of merit for the different type of CD experiments performed on the beamline, are known to be above 97% over the whole VUV range, reaching 99% on most of the VUV range. They are mainly limited by a spurious $S_4$ contribution probably due to a remaining slight C-contamination on the optics. As a result DESIRS is today fully calibrated for the 4 “normal” modes of operation (LV, LH, r-CPL and l-CPL) in the 4.5 to 40 eV range, with an undulator operation totally transparent for the users.

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