IR beamline at the Swiss Light Source

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Abstract. The infrared beamline at the Swiss light source uses dipole radiation and is designed to transport light to four experimental stations, A, B, C, D. Branch A is dedicated to far IR work in vacuum; branch B is a micro-spectrometer; branch C is dedicated to high resolution spectroscopy in the gas phase; branch D is a pump and probe set-up. This contribution describes the optical layout and provides a brief survey of currently available experimental stations. The beamline is in regular user operation since 2009.

1. Source and optical layout
The SLS storage ring (2.4 GeV, 400 mA) operates in top-up mode. Dipole radiation (39 mrad x 61 mrad) is extracted from a bending magnet (ρ = 5.75 m, B = 1.4 Tesla) using M1, a plane, slotted, water cooled Al mirror. If not in use, M1 is protected by a movable absorber. The dipole chamber was modified to accept a vacuum chamber that includes M1, as well as FM2 the first focussing element. This allows the distance between "source center" and M1 to be 820 mm. Radiation is transported along three segments (with focii F1, F2, F3) of 1:1 metal coated optics, f/D ≈ 3, at the end of which we find three equivalent focii, named ports A,B,C. From F2, a transfer line leads to port D. We perform experiments at one port at the time. Near F1, the first intermediate focal point, a wedged diamond or silicon vacuum tight window separates the UHV machine vacuum from the beamline vacuum. These windows can remotely be swapped at all times. Details about the window changer and a sketch of the optical layout are available on-line [1]. The slope error of each mirror has been specified to allow imaging in the visible part of the spectrum. This minimizes optical aberrations along the beamline. The quadratic transverse size of the light distribution at F3 (a replicate of the extended source emitted within the bending magnet) σ_T^2 = σ_x^2 + σ_y^2 + σ_φ^2 includes contributions from the e-beam source, diffraction and geometrical projection, respectively [2, 3, 4]. σ_T has been estimated using the SRW code [5]. σ_y has usually little practical importance, whereas σ_x dominates in the NIR range [4]. IR spectroscopy using the Fourier transform technique is utmost sensitive to light source instabilities that are sampled into the interferograms and appear as spectral artifacts in the data. We performed a detailed noise study of two comparable IR ports (Soleil and SLS) [6].

2. FIR-MIR vacuum spectrometers at branch A
At branch A a mirror coupling box (see section 3 for details) feeds light to a Bruker IFS66v Fourier transform spectrometer dedicated to far IR work. Liquid nitrogen (LN) cooled MCT
and Si detectors cooled to 4.2 K or 2.2 K are available. Based on a PSI prototype, the Group of Dirk van der Marel at the University of Geneva built a vacuum microscope using a pair of X15 Schwarzschild objectives. This unit fits the sample chamber of the spectrometer. At the focus of this optical system, there is a diamond anvil cell (DAC) cooled by a He$_4$ flow cryostat. This set-up allows IR investigations in transmission for samples kept at elevated pressure (few kbar < $P$ < 200 kbar) and reduced temperature (22 K < $T$ < 300 K). Preliminary tests using SR light have demonstrated the principle of operation. Fig. 1 shows the IR spectrometer in which the system that holds the cryostat, the DAC, steering and the focusing optics is mounted. The sample is positioned with an XYZ manipulator that can be actuated under vacuum, whereas the stages holding the Schwarzschild objectives can be actuated at room pressure only. The optical system is being extended to allow investigations on reflective samples. A movable dichroic mirror is used to steer laser light and collect fluorescence light used to monitor the pressure in the DAC.

![Vacuum microscope setup](image)

**Figure 1.** Vacuum micro-focus, DAC, and He$_4$ flow cryostat mounted on a IFS66v spectrometer.

Branch A also accepts a home-built Martin-Puplett-Interferometer (MPI), which can be operated in vacuum to avoid water absorption and has been designed to be directly flanged to the beam line output port. The sizes of the optical elements (focusing mirrors, roof mirror and wire grid beam splitters) allow beam diameters of 80 mm, which suits the MPI well for measurements in the long wavelength range (5 to 200 cm$^{-1}$ depending on the wire grid beam splitters). The MPI control is fully integrated in the SLS EPICS control system and provides in the present configuration step-scan capability. An upgrade of the moveable roof mirror for continuous rapid scan is foreseen for the near future.

The long wavelength transmission of the IR BL port A has been determined with the MPI [7]. A liquid He$_4$-cooled indium antimonide (InSb) hot electron bolometer with a frequency response up to 2 THz (67 cm$^{-1}$) has been used as an IR detector. Figure 2 shows the interferogram (left) and the spectral distribution (right) of the incoherent synchrotron radiation. As long wavelengths (< 10 cm$^{-1}$) are suppressed by the SLS IR BL (port A) the roll off at higher
wavenumbers (> 30 cm\(^{-1}\)) can be attributed to the bolometer sensitivity and the bandwidth of the free standing wire grid beam splitters. Interferograms have been taken in air (blue curves) and at pre-vacuum pressures of < 10\(^{-4}\) mbar (green curves) to show the influence of water absorption lines in this spectral range. A transfer function of the SLS IR BL (port A) has been fitted to the data measured by the MPI (green curve in fig. 3), agreeing quite well with SRW [5, 7] beam transport simulations of the beam line (red curve in fig. 3). It is worth emphasizing, that the intensity of the coherent FIR radiation extracted at port A when the storage ring is operated in the so-called "slice energy modulation" mode [8, 9] is significantly larger than in the regular operation mode. Using an electro-optical detection scheme, the "sliced" coherent FIR camshaft pulse length at port A is \(\approx\) 850 fs FWHM (not shown).

![Figure 2](image)

**Figure 2.** Left: step-scan interferogram (incoherent radiation) and right: spectrum measured with a home made Martin-Pupplet spectrometer at the SLS.

3. **Mid IR spectro-microscopy at branch B**
At branch B, a coupling box feeds focused light into a Bruker Vertex 70 vacuum Fourier transform spectrometer, itself coupled to a Bruker Hyperion 3000 IR microscope (\(m_m\) in table 1). The coupling box houses an elliptical mirror that reduces (\(m_c\) in table 1) the size of the light spot available at F3. From there, a \(f_p = 100\) mm parabolic mirror collimates light into the interferometer unit. Modulated light is sent directly [10] to the Hyperion microscope equipped with the usual palette of X15, X36, X20 (Ge)ATR, and X15 GIR objectives. To estimate the actual demagnification factors \(m_i\) expected with this set-up we consider \(m_i = m_c \cdot m_m\), where \(m_c\) and \(m_m\) are the contributions due to the coupling and microscope optics, respectively. The factor \(m_c\) is straightforward and depends of the focii ratio of the chosen ellipse. The factor \(m_m\) is approximated by the ratio \(f_p / f_m\), where \(f_m\) is the actual focal length of the Schwarzschild objectives. Table 1 summarizes the situation.

The estimated and observed demagnification factors differ; \(m_r > m_i\). Two comments are here in order. First, the X15 objective should actually produce a demagnification factor of 15. However, the ratio of focal distances for this optics matched to standard tube length of 160 mm gives 160 mm / 13.1 mm \(\approx\) 12. Second, because this microscope does not use infinity corrected objectives, there is (by design) one additional intermediate focus in the light path between the interferometer and the entrance pupil of the Schwarzschild. Thus, the assumption that \(m_m\) is solely given by the focii ratio between the collimating parabolic and Schwarzschild
Table 1. The focal length of the Schwarzschild optics $f_m = D/2NA$, $NA$ the numerical aperture, $D$ the size of the entrance pupil of the objective. $m_m = f_p/f_m = 100$ mm / $f_m$ assumes collimated transport to the X15 or X36 optics. The ratio of the focii of the elliptical coupling mirror is $m_c$. The expected demagnification factor is $m_i = m_m \cdot m_c$ whereas $m_r$ is the observed value. Dimensions are given in mm. Differences explained in the text.

| $f_m$ | $NA$ | $D$ | $m_m$ | $m_c$ | $m_i$ | $m_r$ |
|-------|------|-----|-------|-------|-------|-------|
| X 15  | 13.1 | 0.40| 10.5  | 7.6   | 6     | 45    | 60    |
| X 15  | 13.1 | 0.40| 10.5  | 7.6   | 3.4   | 25    | 34    |
| X 36  | 5.4  | 0.52| 5.6   | 18.5  | 6     | 110   | 143   |
| X 36  | 5.4  | 0.52| 5.6   | 18.5  | 3.4   | 63    | 81    |

secondary optics is not accurate for this optical system. We determined experimentally that the demagnification factor between the spectrometer input focus and the sample focal plane to be $m_{m, eff} = 10$ and 24, for the X15 and X36 optics, respectively. For all practical purposes, we used $m_{m, eff} \times m_c$ to estimate $m_r$ in Table 1. The left panel of Fig. 4 shows the wavelength dependence of the spot size, extracted from the intensity profiles (right panel) measured in transmission (X36, $NA = 0.52$, $m_c = 3.4$) by scanning a 5 $\mu$m diameter pin-hole across the beam. The slope of the experimental data is well described by $FWHH \propto 0.48 \cdot \lambda$ and agrees reasonably well with the expected diffraction limit [2, 11, 12]. The offset of the exp. data is $\approx 4.8 \mu$m and includes contributions from the electron source size (visible at higher photon energy [4]) as well as the finite size of the pinhole that was used to scan the spot profiles. Strictly speaking, in order to extract precise information on the source size we should perform a 2D
deconvolution of the measured light profiles taking into account the point spread function of the optics and the pinhole. This shall be discussed in detail elsewhere.

This IR spectro-microscope offers a diffraction limited spot size and serves the broad community of investigators that require focused light from 400 to 4000 cm$^{-1}$. The instrument was further improved with a custom manufactured XYZ manipulator for positioning of the condenser Schwarzschild objective, to allow spectro-microscopy in transmission with enhanced stability. Fig. 5 illustrates this unit. The micrometric XY stage used to manipulate samples in the focal plane has been removed for clarity. The stability of the condenser unit is affected by the power dissipated by the tungsten lamps illumination system. More precisely, it is a change in illumination that causes long term drifts, as the system reaches a new thermal equilibrium sitation. A LED-based illuminator, or a thermal decoupling of the lamps’ power supply from the optical assembly is likely to limit long term drifts. A liquid He$_4$ flow cryostat can be placed in the focus, for transmission and/or reflection work.

For work at elevated pressure using a diamond anvil cell (DAC), the pressure determination is often based on a shift of a fluorescence line in a crystal like ruby [13]. The excitation of the crystal and the collection of the fluorescence light is achieved ”in-situ”, with the DAC remaining in the IR focus. The change from ”IR work” to ”P-monitoring” is obtained by exploiting all the capability of the IR microscope. Our scheme does not require any modification (followed by re-alignment) of the microscope but only the placing of a tiny 2 mm diameter mirror to steers the excitation laser light along the IR path. This mirror can be positioned to avoid any IR light loss. Fluorescence light is collected through one of the ocular tubes [14].

Investigators active in cell biology, get access to a cell culture lab on-site allowing cell growth and providing Level II containment for samples of biomedical interest. A variety of enclosed sample holders allowing a controlled environment is available for spectromicroscopy experiments on solutions and contained samples, including living cells and solution processes.
4. **Ultra high resolution at branch C**

From F3C, radiation is transported through a dedicated vacuum transferline coupled to a ultra high resolution Fourier transform spectrometer Bruker HR 125. This instrument has 11 m of optical path difference and is operated by the group ”Molecular Kinetics and Spectroscopy” of Martin Quack at ETH-Zurich. The transfer line between F3 and the spectrometer input was designed to re-shape the spot using cylindrical optics. Details are given in Ref [15].

5. **Pump and probe at branch D**

Synchrotron-based infrared pump and probe (IRPP) experiments offer a unique experimental niche in the sense that they combine focusing capability together with extended spectral and dynamic bandwidth. The available time resolution lies in the 100 ps range is well suited to study dynamics occurring in condensed matter (phase change materials, carrier diffusion, electron-hole or quasiparticle recombination) or chemical processes in which the identification of intermediate reaction species (i.e. photo catalyzes) is required.

For these experiments at branch D, light is taken from F2, and steered along a 10 m long vacuum transfer line (focussed optics) into a purged Vertex 70 Fourier transform interferometer. The coupling scheme is similar to the one used at branches A and B. The side exit of the interferometer feeds collimated light to an optical system that allows broadband pump (laser) and probe (IR synchrotron) experiments with a time resolution $\approx$ 100 ps. Proof of principle as well as a technical description of this set-up is given in Ref. [16]. We aim at developing this IRPP set-up to the level of a user instrument.

A micro-focus setup allows the investigation of samples of sizes $\approx$ 10 $\mu$m using a pair of 15X Schwarzschild optics. Simultaneous measurements in both transmission and reflection are possible. The optical path is illustrated in Fig. 6 for the standard setup (focus length of 100 mm, $\approx$ 100 $\mu$m sized beam) with parabolic mirrors instead of Schwarzschild objectives. The data acquisition system is programmed to acquire the camshaft as well as fractions of the quasi continuous part of the synchrotron emission. With this method, the response of the sample
upon laser excitation gets recorded at later time stamps (> 30 ns) with a resolution given by the used detector (typically 10 ns) while maintaining the 100 ps time resolution at short delay times. Compared to a traditional time resolved step scan FTIR system using a thermal source, the (signal \times bandwidth) advantage of this synchrotron based system is > 2000 when using the 100 ps long camshaft pulse. In the quasi-continuous mode, the advantage reduces to > 50. The latter number reflects the brilliance advantage of the synchrotron which remain in spite of the significant beam losses likely to take place along a complex transfer line.

![Figure 6](image-url)

**Figure 6.** View of the focus region at the IRPP set-up. A similar beam path arrangement is obtained when the micro-focus unit based on a pair of Schwarzschild objectives is used.

Fig. 7 illustrates the spectral bandwidth as well as the timing capability of the entire system using the micro-focus unit. The Nd:YAG excitation pulse at 1.06 \( \mu \text{m} \) generates excess charge carriers in InGaAs, which lead to an excess absorption due to valence-interband and conduction-intraband transitions. We observe optical gain at energies above the InGaAs bandgap energy at 0.72 eV (see unpumped spectrum). The time evolution of this electron-hole population is probed dynamically by varying the time delay between the pump and the probe. Control of the time delay is achieved with a phase-lock loop system that synchronizes a master oscillator running on the pump laser to the SLS clock. Details are given in Ref. [16]

6. **Summary**

In summary, we described a versatile facility that allows at branch A to perform FIR spectroscopy in vacuum at moderate resolution and offers an easy to use experimental platform. In one such case, the high-pressure and low temperature set-up developed by the Geneva team is attached to the existing interferometer. Branch C allows FIR as well as MIR spectroscopy in vacuum at very high resolution. Branch B is the most popular platform, since it offers, in addition to a diffraction limited spot, the possibility to couple a large variety of experiments like XY sample stage, (heated) high pressure cells, micro-fluidic cells, a slim He4 flow cryostat, to name just a few. Branch D is dedicated to pump and probe experiments in the MIR range, and is still in development. A micro focus unit using a pair of X15 Schwarzschild optics (46 mm space along the optical axis) is available. Currently, we operate with synchrotron light one branch at the time.
Figure 7. Normalized MIR/NIR Transmission of a 4 µm thick InGaAs layer on InP upon optical pumping with a 1.06 µm laser excitation at ≈ 145 MW/cm² for different pump/probe delays. The microfocus unit allows spotsizes of 16 µm (probe) and 35 µm (pump).

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