DESIRS beamline: world record resolution in wideband VUV absorption spectrometry

Since September 2007, the DESIRS beamline, run by Laurent Nahon, has a new instrument which is unique in all the world: a Fourier Transform (FT) spectrometer based on a wavefront division interferometer. It offers a far greater resolving power than the best grating interferometers used today. The first users can now have access to the ultra-high resolution of the FT branch of the beamline, and also to the high resolution spectroscopy techniques, associated to the monochromator branch.

In the early 1990s, Michel Vervloet, a research scientist at the Laboratoire de Photophysique Moléculaire (LPPM) at the University of Orsay, approached Denis Joyeux and François Polack of the Institut d'Optique (Optical Institute) with the following question: is it possible to design a Fourier Transform Spectrometer (FTS) in the high-energy ultraviolet range? Denis Joyeux and Nelson de Oliveira told us what happened next...

VUV Absorption Spectrometry[1]

Absorption spectrometry is a technique for studying the electronic energy levels of atoms and molecules, based on the way an atom or molecular structure absorb photons of a given energy. The presence of the different species contained in a sample can then be recognised and/or quantified by studying the recorded spectra.

The energy levels, however, also reveal the chemical behaviour of these species; and if the photons absorbed are VUV photons, spectroscopy can give access to the photochemistry[2] of media as different as planetary atmospheres or the interstellar medium, and even certain plasmas.

Several techniques can be used to record the absorption spectrum of a sample. Among all, Fourier transform spectroscopy has very attractive properties: high resolving power, precision of the intensity and position of the measured lines making up the spectrum. This technique would certainly bring valuable information in the VUV spectral range.

The weak point concerns the instrumentation; the set-up used for FTS in other wavelength ranges (IR, visible or lower-energy UV) are very difficult to 'export' in the VUV range. Indeed, the beamsplitter, one of the essential components of the amplitude division interferometers used (see inset) is very difficult, or even impossible, to manufacture for wavelengths below 140 nm.

'Amplitude division' vs. 'wavefront division'

Let us return to the 1990s and Michel Vervloet's question.

To answer it, the two optical physicists decided to overcome the beamsplitter problem by developing a scanning VUV wavefront division interferometer. It was a variation of the classical Fresnel bi-mirror interferometer, a set-up requiring no beamsplitter, but only planar mirrors (see inset). The specific feature of a FTS (scanning interferometer) requires a special design: the light beam (VUV in this case) is reflected by two similar roof-shaped right angled reflectors (see Figure 1). The two reflectors are separated by about 100 microns apart, one is fixed and the second is mobile. The incident beam undergoes two consecutive reflections on each of the reflectors; it is retroreflected. The upper reflector is slightly tilted, and the two reflected beams recombine and interfere. The interferometric signal is recorded for different positions of the mobile reflector, generating an interferogram. Finally, a Fourier Transform must be

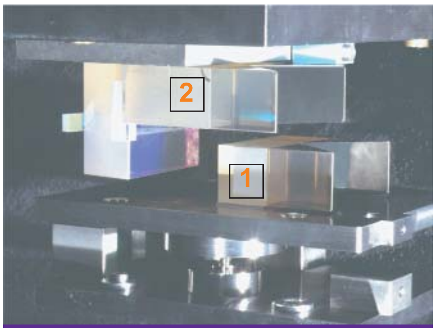
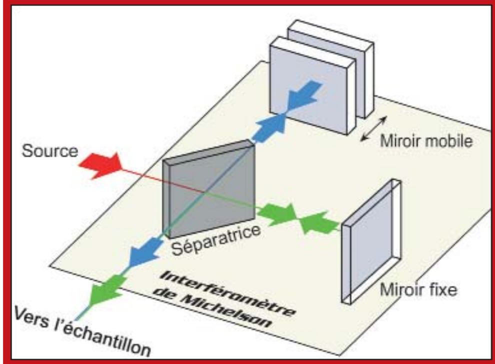


**Figure 1: Wavefront division interferometer with variable path-length difference.
1: Moving reflector. 2: Fixed reflector. The space separating 1 and 2 was enlarged for the photo. It is normally of the order of 1/10 mm.**

Michelson interferometer: amplitude division

This interferometer was invented by the American Albert Michelson in the late 19th century. It contains a beamsplitter and two mirrors: one fixed, and the other mobile. The beamsplitter is a semi-reflecting window which divides the light source into two independant beams of roughly equal intensity. After reflection on the fixed and mobile mirrors, the reflected beams again pass through the beamsplitter, where they are recombined with a phase shift which depends on the position of the mobile mirror. If coherence conditions are fulfilled, the two beams can interfere. In that case, the wavelength of the light (in the visible spectrum $\lambda \sim 0.5$ μm) gives the scale of sensitivity of the interferometer. The interferometric signal variations are directly linked to the relative displacement between the two interferometer arms.



The Michelson interferometer is a powerful metrological tool, used for example to measure the quality of optics.

Because it provides very precise measurements of 'optical lengths', Michelson and Edward Morley used it in the 1880s to show that the speed of light is isotropic and independent of the frame of reference. Their observations led to the ruling out of the theory of ether, a medium in which light was thought to propagate...

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[1] VUV: Vacuum Ultraviolet, high-energy UV, corresponding to the 30-200 nm (6-40 eV) range of wavelengths. Called 'VUV' because the light is absorbed by air at these wavelengths, and so the experiments must take place in a very high vacuum.

[2] Photochemistry: light-induced chemical reactions. In this case: by VUV photons.

computed onto this interferogram in order to extract the source spectrum.

In the late 1980s, Denis Joyeux and François Polack were developing VUV and EUV interferometers. This VUV FTS project became a subject for a PhD student in 1996, and Nelson de Oliveira joined the team.

They had a precise goal: to conceive a spectrometer that could work up to 60 nm (~ 20 eV), with very high resolution (i.e. resolving power $\lambda/\delta\lambda$ on the order of 500,000).

Optics, Mechanics and Electronics

This brings us to the early days of second-generation (1980s) and then third-generation (early 1990s) synchrotrons: extremely bright and partially coherent light sources, coherence being an essential property in order to make two beams interfere.

At Orsay, the LURE is in service, and SOLEIL is still only a project.

This is the context in which the work is beginning.

Even though a wavefront division interferometer only requires planar mirrors, they must be of very high quality: actually, a perfect flatness right to the edges is needed. By relying on the skills of the 'Institut d'Optique' personnel, Denis Joyeux and François Polack had already overcome most of the technical problems involved in manufacturing such mirrors and mounting them in an interferometer suitable for very short wavelengths. In fact, it was the dynamic aspect, specific to FTS, which made the challenge even more difficult to face. FTS requires a very precise measurement of the displacement of the mobile reflector with respect to the fixed one. Bearing in mind that the tolerances on this measurement are on the scale of the wavelengths of the incident light, we very quickly tend towards values on the order of a few nanometres.

The mechanical and electronic part which makes such precision obtainable was designed and built by a four-member team led by Denis Joyeux. It was truly a job for expert craftsmen: a very sensitive optical device to monitor and measure displacement, based on many reflections between two plane mirrors, was associated to the interferometer. It was placed behind the reflectors and had a dual function: to stabilise the orientation of the mobile reflector, and to measure the displacement of the mobile optical block with the greatest possible accuracy.

And it works!

The first experiments took place in July 2003. In order to use synchrotron radiation to validate this brand new spectrometer, measurements were performed in air on the SU5 beamline of Super-ACO. And the absorption spectrum of the oxygen (of the hall of Super-ACO) in the mid-UV range (180-200 nm) thus obtained turned out to comply with the reference experimental data (well established, since situated in the near UV).

This was not only a cause for celebration by the team; it was also a weighty argument, which persuaded the ANR to participate in funding the optimisation of the instrument. And so, on to Version 2!

Since that time, LURE has run out of operation, to be succeeded by SOLEIL, where Nelson de Oliveira is now one of the three scientist working on the DESIRS beamline. The spectrometer he and Denis Joyeux are currently testing, has been completely revamped since the experiments on SU5, and its performance has been considerably improved: resolving power of the order of 10^6 compared to the 500,000 initially expected, for a limit wavelength of 45 nm (compared to the anticipated value of 60 nm). The first spectra obtained show great promise for the future of high-resolution FTS on DESIRS! (See inset on next page.)

For which studies?

The scientific case is most notably in the field of universe and atmospheric sciences. Astrophysicists will be interested in studying the small molecular systems present in interstellar space (H_2, CO, C_2H_2, CH...) or in the atmospheres of planets (CO_2, N_2...).

Figure 2: The spectrometer installed on the DESIRS beamline. The 'interferometer' part is visible in the middle (mirrors).

A comparison of high-resolution spectroscopic data obtained in the laboratory (on the DESIRS FTS) to data collected by spectrometers on board satellites makes it possible to calculate the temperature, density and isotopic composition of the species being studied. An analysis of their photochemistry (lifetimes of the excited states) makes it possible to deduce the abundance of certain compounds and therefore to model the birth of stars and their associated planets.

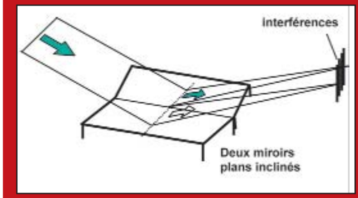
And the rest is happening now...

In early January 2008, a postdoc joined the DESIRS high-resolution unit at SOLEIL to work in particular on setting up and optimising the 'sample environment' placed upstream of the spectrometer. In January 2008, Denis Joyeux left the team, although Nelson intends to continue seeking his advice as a consultant.

As for the unique device, it should attract

The Fresnel interferometer: wavefront division

In the early 19th century, there was an ongoing dispute between the supporters of the purely wave theory of light and those, who were in the majority, who maintained Newton's viewpoint about a corpuscular theory of light. This was the context in which Augustin Fresnel carried out many experiments in optics. In particular, he studied light diffraction and interference, and defended a wave model of light propagation before the academy of sciences; this model was based on the so-called 'Huygens-Fresnel' principle.



Here is what Augustin Fresnel said about the bi-mirror interferometer which now bore his name: After placing the two mirrors beside each other so that their edges are in perfect contact, they are tilted until they are almost in the same plane but still form a slight angle so that they show two simultaneous images of the source [...] We then look for the fringes localized where the two reflected beams are superimposed [...] these fringes show a series of bright and dark bands which are parallel to each other and equally spaced.

SOLEIL users interested in very high resolution in this range of wavelengths.

Until perhaps copies appear in other synchrotrons in a few years' time?

Figure 3: Denis Joyeux and Nelson de Oliveira beside the spectrometer - here, placed in a vacuum chamber

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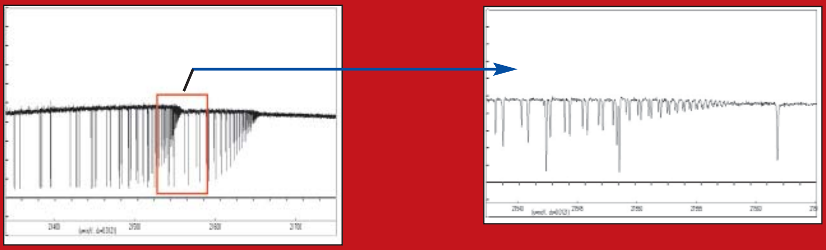
Initial results

Kenji Ito (Photon Factory, Japan), first expert user on the DESIRS beamline, “inaugurated” the FT spectrometer in December 2007. The idea was to revisit the absorption spectra of various noble gases (Ar, Xe, Kr, Ne) in the region of the Rydberg states converging towards the ionisation thresholds. Using the unprecedented resolving power, it is possible for example to analyse ray profiles far more precisely to extract information concerning the relaxation dynamic of these highly excited states. This can most notably be done by comparing data with very precise ab initio calculations which can be carried out on these relatively simple models: the noble gases.



Absorption spectrum of Ne in the region of the ionisation threshold (2p-1) at 21.6 eV. The measured resolution of the instrument δE is 29 µeV corresponding to a resolving power of 745,000.

This first campaign was a real success because, beyond this scientific aspect, these experiments clearly validate the instrument by checking the predicted resolving powers experimentally. Thus, a resolving power of the order of 700,000 has be measured over a spectral range extending from 12.5 to 28.6 eV! This is significantly higher than what is achieved by the best current grating spectrometers (< 250,000, at best, in this same range).

On the figures above, one can see the Rydberg series of neon converging towards the first and second ionisation thresholds around 21.6 eV. This is the region in which the so-called autoionisation effects of particular interest to Kenji Ito can be observed.