Dichroism and Spectroscopy through Interaction with Synchrotron Radiation

DESIRS (Dichroïsme Et Spectroscopie par Interaction avec le Rayonnement Synchrotron)

Valence shell excitations: atoms, molecules, clusters, solids


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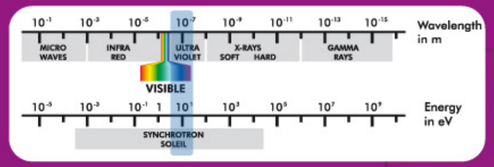
Energy range of DESIRS : 5 – 40 eV (range called « VUV » - Vacuum UltraVolet)



Light source: HU640 type OPHELIE2 electromagnetic undulator, specifically designed for DESIRS by SOLEIL, providing fully variable polarisation (including linear and circular)

Experimental techniques:

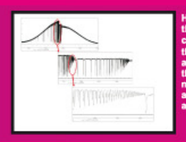
- Ultra high resolution Fourier transform VUV absorption spectroscopy
- Photoelectron imaging, mass spectroscopy, electron-ion coincidence
- Circular dichroism

DESIRS provides a high intensity, high resolution photon beam over the whole VUV range (valence shell excitation), with perfect spectral purity and variable polarisation.

Topics and applications

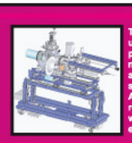
→ Ultra high resolution spectroscopy (consultant: M. Vervloet)

- Fourier transform spectroscopy: multiplex capability
- Small molecular systems of astrophysical or atmospheric Interest, cold molecules and radicals


High resolution absorption spectrum of Neon in the region of Rydberg states transitions converging towards the 2p^5 ($^2P_{3/2}$ and $^2P_{1/2}$) thresholds. Owing to a free jet expansion achieved in the "sample environment chamber", the Doppler width is reduced so that the raw measured linewidth is 42 meV corresponding to a RP of ~ 500000). The typical duration of acquisition is a few hours.

→ Molecular dynamics and reactivity

- State to state photochemistry (TPEPICO)
- Mass spectrometry
- Ion-molecule reactivity (Associated scientist: C. Alcaraz LCP, Orsay)

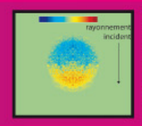

The molecular beam chamber SAPHIRS may be used with gaseous, liquid or solid samples, producing a cold and collimated beam of stable molecules or radicals species. It is equipped with a very versatile imaging electron/ion coincidence spectrometer, DELICIOUS2, able to perform AR-PEPICO on fast electron with 5 % energy resolution (up to 16 eV electron), or TPEPICO with a sub-meV resolution for threshold electrons.

→ Photoionisation dynamics

- Rydberg states dynamics
- Two-colour experiments : laser + synchrotron radiation
- Clusters

→ Alignments effects and chirality

- Electron- ion vectorial correlations
- Circular dichroism in photoionisation/photoabsorption
- Homochirality of life



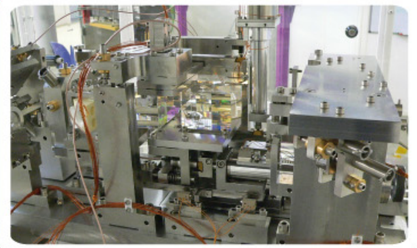
Photoionisation of the right enantiomer of camphor in the gas phase using circularly polarised light. The asymmetry of the emitted photoelectrons reveals the molecular chirality of camphor.


Asymmetric photo-fragmentation of Leucin amino-acid induced by circularly polarized light that may support the hypothesis of a spatial origin of Life's homochirality.

→ Excitation and relaxation in the condensed phase

- Luminescent solids
- Molecular liquids

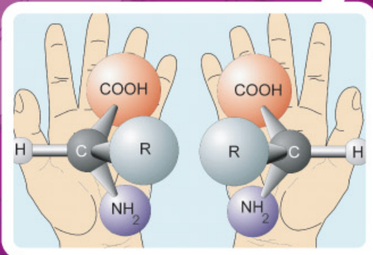
Zoom: VUV Fourier transform spectrometer



This Fourier transform spectrometer, based upon wave-front division (no beamsplitters), is able to achieve unprecedented spectral resolution over a wide bandwidth (typically 1 eV) on the 5 to 30 eV range, with full multiplex advantage. The measured resolving power (E/ΔE) of the instrument is close to 10^6 (ex: 27 μeV at 21 eV).

(Consultant: D. Joyeux)

Zoom: Interaction between chiral molecules and cirularly polarised light (CPL)



As our hands, a so-called "chiral" molecule comes in two forms, called enantiomers (left and right) that cannot be superimposed but which are mirror images of each other. Such an « asymmetry », chirality, will exhibit itself when these molecules are embedded into a chiral environment such as, for instance, an helical physical field as the one associated to Circularly Polarized Light (CPL): this is called circular dichroism (CD).

Two types of photon-induced asymmetric processes on chiral species are studies on DESIRS

- Asymmetric photochemistry induced by circularly polarized light. When a 50/50 mixture of left and right molecules is irradiated with UV-CPL, one of these forms is observed to dominate after a certain time, because of enantio-selective photo-destruction. This induced excess of one of the enantiomers might explain why, on Earth, amino-acids, the chiral building blocks of life, occur only in the left-form: this is the so-called "Life's homochirality", a clear signature of life, whose origin is still an open question. The irradiation results fit within a scenario in which amino-acids are assembled in interstellar/circumstellar space prior to reaching Earth. Samples can be solid films (micro-meteorites) or ice analogs (comets).

  Collaboration: Université de Nice /CNRS; IAS – Orsay

- Photophysics: When a given enantioner in the gas phase is photoionised by CPL, the angular distribution of photoelectrons shows a forward/backward asymmetry with respect to the direction of light propagation, reflecting the molecule chirality: this is Photoelectron Circular Dichroism (PECD). This spectacularly intense new type of CD (asymmetry up to several 10 %) depends on the photon energy and on the initial orbital, and appears to be very sensitive to the precise molecular structure: a fine probe of chemical substitution and conformer distribution. Photoelectron imaging measurements in coincidence with the photoion are carried out with the DELICIOUS 2 AR-Imaging PEPICO spectrometer.

  Collaboration: University of Nottingham (UK)

SOLEIL SYNCHROTRON