The SIRIUS facility : a powerful tool for studying radiation effects in materials

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SIRIUS geographical location

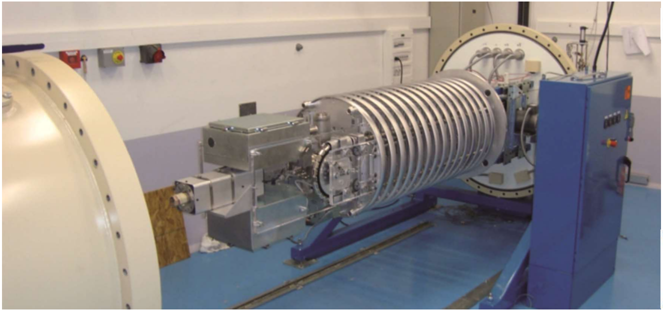
- The SIRIUS[a] facility is located at Laboratoire des solides irradiés (LSI[b])
- The LSI is a tripartite laboratory

[a] Irradiation platform for innovation and scientific uses
[a] Irradiated solids laboratory

The SIRIUS facility - *Production of radiation defects*

SIRIUS hosts a 2.5 MeV NEC Pelletron accelerator :

- Continuous electron beam
  - 150 keV $<$ Energy $<$ 2.5 MeV
  - 200 nA $<$ Current beam $< 40\ \mu$A
- Few mm to few cm beam size
- New installation (2013) working 24h/24, 7d/7 (300 days a year)



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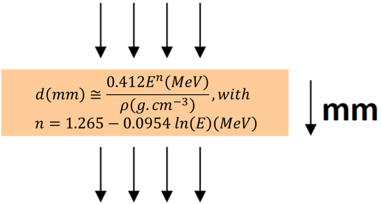
Electron irradiation damages in materials

2 components of damage under MeV electron irradiation

Electronic component

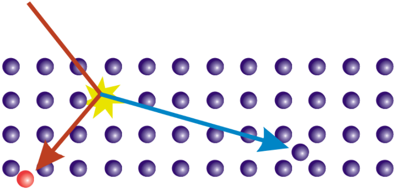
(*Electron excitation/ionization*)



Homogeneous effects in the target volume (few mm thick materials)

Nuclear component

(*Ballistic or displacement damage*)



Single or double atom displacements (knock-on mechanism)

Main research topics at SIRIUS facility

- **Influence of electronic excitation in insulating materials**
  - Nuclear fuel cycle (glass, concrete and clays)
  - Transient events under irradiation in components (quartz USO in space)
  - High density in optical materials (optical fiber performs)
- **New materials for nuclear applications**
  - Threshold displacement and migration energies from ballistic events (semiconductors, ceramics, metals)
  - Long term behavior of nuclear materials
- **Irradiation as a tool for improving material properties**
  - Doping effect (topological insulators, semiconductors, superconductors)
  - Radiografting (polymers, nanowires)
  - Nanostructuration induced by irradiation (plasmonic nanoparticles)

2 beamlines hosting numerous equipment for online measurements

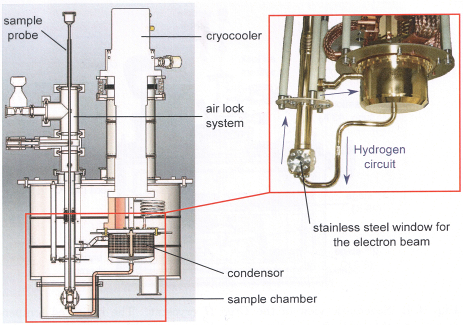
Beamline	Setup available	Irradiation specificities			Experiment topics examples
		Temperature (K)	Current max (µA)	Sample types	
Line 1	**CRYO1**	20	20	Bulks	**Conductivity measurements** in superconductors and topological insulators (Bi_2Te_3, BI_2Se_3)
	CRYO2	4 ≤ T ≤ 300	2	Bulks, powders	• **Resistivity and Hall effect measurements** in topological insulators (Bi_2Te_3) • **Electron paramagnetic resonance** *
	IRRAPLAST	300	10	Polymers, nanowires	• Irradiation in PVDF for piezoelectrical applications • Radiografting of polymers and nanowires
	LSIC *	100 ≤ T ≤ 300	40	Large flat samples (25 cm² max)	**Electrical measurements** (IV characteristics) in real-size triple junction solar cells
	***in situ* spectroscopy**	300	40	Bulks	• **Photoluminescence** in Sm-doped glasses, UO_2 discs • **Time-resolved photoluminescence** in halide perovskites for photovoltaic applications • **Cathodoluminescence** • **Raman** spectroscopy
	users experiments	*Depend on setup*	40	Bulks, powders, liquids	• **I-V measurements** of solar cells under illumination • Gas emitted during polymers irradiations analyzed by **mass spectrometry**
Line 2	**CIRANO**	300 ≤ T ≤ 450 *(670 K possible)*	40	Bulks (7 cm² max), powders	• Defects production in nuclear glasses, Zy, Titania, UO2, SiC (*ex situ* characterization of induced point defects, threshold displacement energy calculations) • **Absorption** in quartz crystal for space applications

** Under development*

SIRIUS equipments (1/6) : CRYO1

Set-up properties

- H_2 recondenser (sample chamber filled with liquid H_2)
- $T_{irr.,meas.} = 20$ K
- Cooling power >25 W at 20 K
- Beam current max ∼20 μA
- *in situ* electrical measurements



Relevant recent works using CRYO1

Yamashita *et al.*, 2017, *Fully gapped superconductivity with no sign change in the prototypical heavy-fermion $CeCu_2Si_2$*, Sci. Adv., **3**:e1601667

Zhao L. *et al.*, 2016, *Stable topological insulators achieved using high energy electron beams*, Nat. Comm., **7**:10957

Cho K. *et al.*, 2017, *Energy gap evolution across the superconductivity dome in single crystals of $(Ba_{1-x}K_x)Fe_2As_2$*, Sci. Adv., **2**:e1600807

Irradiation induced doping of topological insulators

New class of materials → insulating in the bulk, but with gapless edge or surface states

Applications in spintronic and optoelectronic

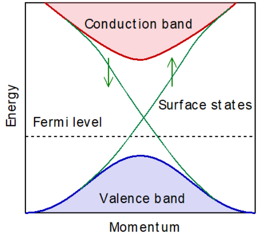


Figure: Idealized band structure for a topological insulator. The Fermi level falls within the bulk band gap which is traversed by topologically-protected surface states.

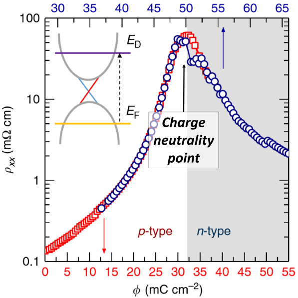
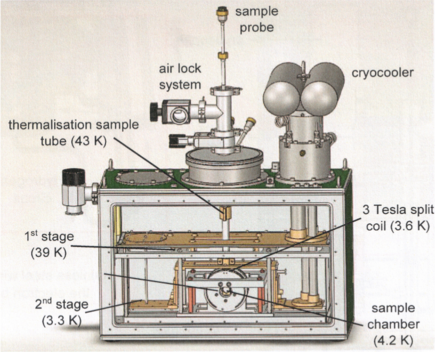


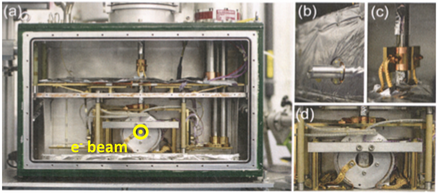
Figure: Conductivity type inversion by irradiation with 2.5 MeV electrons in Bi_2Te_3 measured at 20 K.

Electron irradiation offers a path to large scale access to topological states

Set-up properties

- Two stage 4 K pulse tube cryocooler (1.5 W at 4 K)
- 3 T superconducting magnet system
- $4\ K \leq T_{irr.,meas.} \leq 300\ K$
- *in situ* electrical measurements (resistivity, Hall effect)
- *EPR under development*





Recent work using CRYO2

RISCHAU C.W. *et al.*, 2013, *Doping of Bi_2Te_3 using electron irradiation*, Phys. Rev. B, **88**:205207

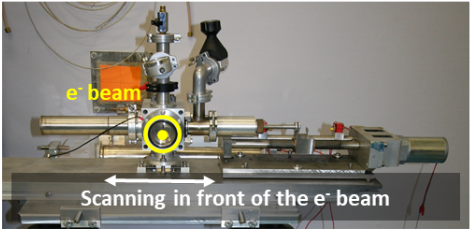
SIRIUS equipments (3/6) : IRRAPLAST

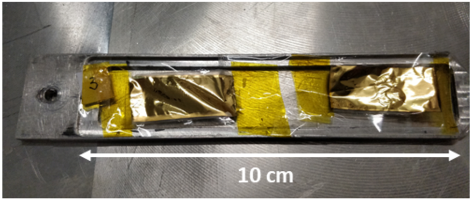
Set-up properties

- $T_{irradiation} = 300$ K
- Air or inert atmosphere (He, Ar)
- Current beam up to sev μA
- Translation along one axis (15 cm amplitude)

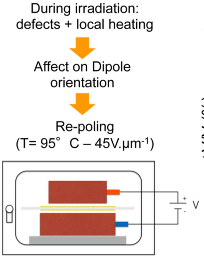
Applications

- Effect of electron irradiation on PVDF piezoelectric properties
- Radiografting of polymer films and nanowires

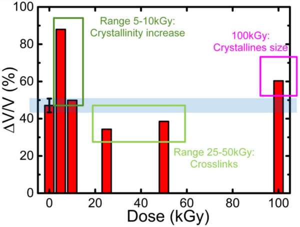


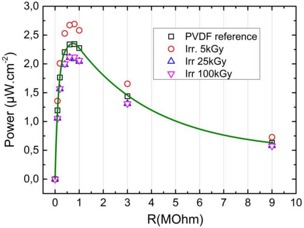


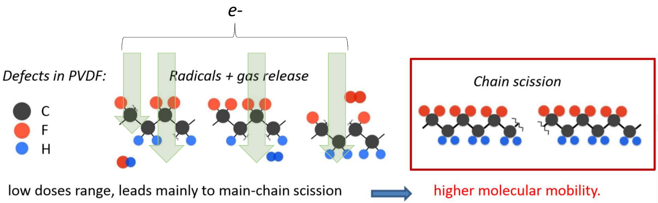
Irradiation of PVDF piezoelectric polymers



$$\frac{\Delta V}{V} = \frac{V_{after\ re-poling} - V_{initial}}{V_{initial}}$$







Enhancement of PVDF piezoelectrical response due to radiation-induced point defects

Melili G. et al., 2016 *How swift-heavy ions and/or e-beam irradiations act on the piezoelectric response of nanostructured polarized PVDF films*, GANIL-SPIRAL2 Week

SIRIUS equipments (4/6) : LSIC

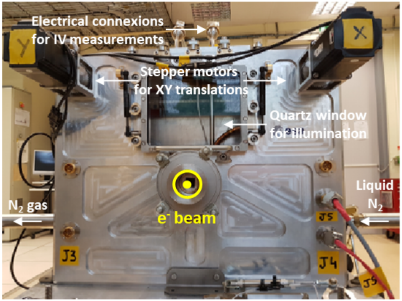
Set-up properties

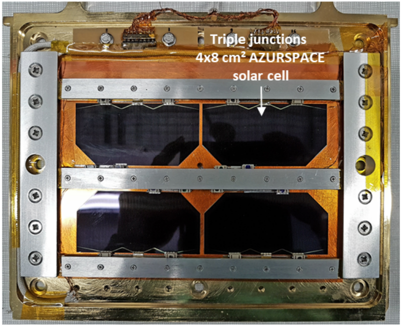
- Large irradiation surface area : 180×130 mm^2
- 100 K $\leq T_{\text{irr.,meas.}} \leq 300$ K
- *in situ* electrical measurements
- *LED solar simulator to be implemented*

Application

Irradiation of large surface plane samples (real-size TJ solar cells)

LEFÈVRE J. *et al.*, 2017, *Dedicated tool for irradiation and electrical measurement of large surface samples on the beamline of a 2.5 MeV Pelletron electron accelerator*, E3S Web of Conferences **16**:16003





Degradation of TJ solar cells under irradiation at low T

Loss of power generation of solar cells due to radiation-induced defects at low T (minority carriers trapping + tunneling effect)

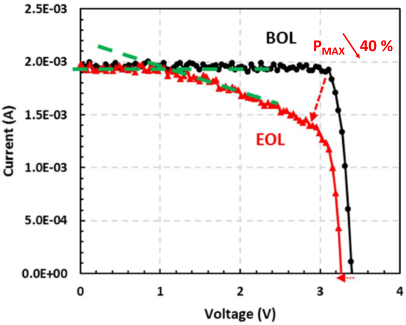


Figure: **Light I-V characteristics** of a TJ solar cell at 123 K before and after 1 MeV electron irradiation (1.5 x $10^{15}/cm^2$)

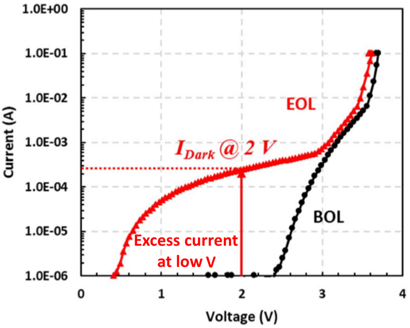


Figure: **Dark I-V characteristics** (log scale) of a TJ solar cell at 123 K before and after 1 MeV electron irradiation (1.5 x $10^{15}/cm^2$)

Park S. *et al.*, 2017 *Origin of the degradation of triple junction solar cells at low Temperature*, E3S Web Conf., **16**, 11th European Space Power Conference

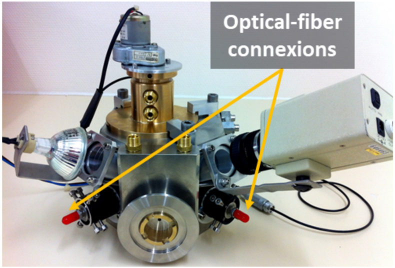
SIRIUS equipments (5/6) : *in situ* spectroscopy

Set-up properties

- $T_{irr.,meas.} = 300$ K
- Water-cooled sample holder
- Photoluminescence (PL), time-resolved PL, cathodoluminescence & Raman spectroscopy
- *Pulse-tube cryocooler to be implemented*

Applications

- Influence of damage sublattice on PL & Raman in sintered UO_2 discs
- Effect of temperature on radiation-induced point defects in silica
- Mechanisms responsible for the radiodarkening of optical fiber preforms



Time-resolved photoluminescence (PL)	
Excitation wavelength (nm)	266, 355, 532
Pulse duration (ns)	10 (at 10 Hz)
Detection	SR-303i Shamrock spectrometer & ICCD Istar Andor camera
Raman spectroscopy	
Detection	Jobin Yvon HR800 spectrometer

Radiodarkening of optical fiber preforms

Darkening of rare-Earth doped optical fibers used for laser applications or in harsh environments

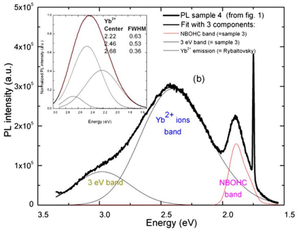


Figure: Emission of electron-irradiated Yb-doped fiber preforms samples under a 355 nm pulse laser excitation

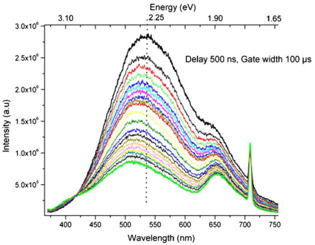


Figure: Recovery of the emission under the 355 nm laser beam during 2 hours

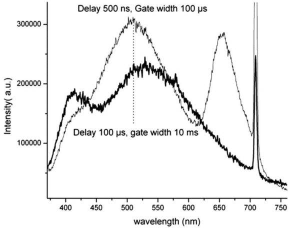


Figure: Emission under 355 nm excitation with pulse gate delay and width (500 ns, 100 μs) and (100 μs, 10 ms)

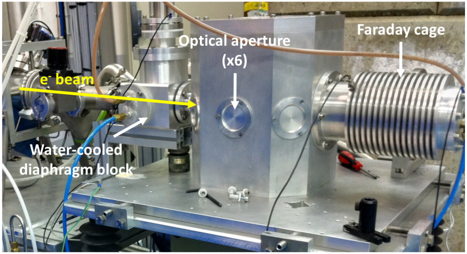
Radiodarkening process of Yb-doped optical fiber preforms based on a pair association of Yb^{2+} with the so-called AlOHC point defects

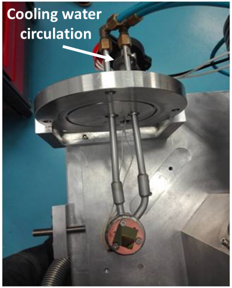
Ollier N. *et al.*, 2016 *in situ observation of the Yb^{2+} emission in the radiodarkening process of Yb-doped optical preform*, Optics Letters, **41**, pp. 2025-2028

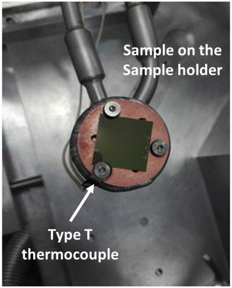
SIRIUS equipments (6/6) : CIRANO

Set-up properties

- Current beam up to 40 μA
- 300 K $\leq$ $T_{irr.}$ $\leq$ 450 K (670 K possible)
- Irradiation in air, inert atm., or under primary vacuum
- Powder or bulk samples (few cm^2 max)
- Water-cooled sample holder
- Optical apertures for *in situ* UV-VIS absorption measurements







Radiation sensitivity of ultra stable quartz oscillators (USO)

Frequency drifts of quartz USO used in spacecrafts

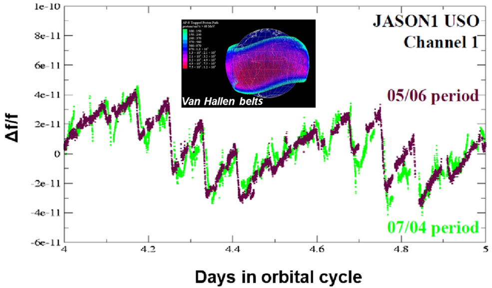


Figure: Frequency variations of the Channel 1 USO observed on-board JASON1 satellite

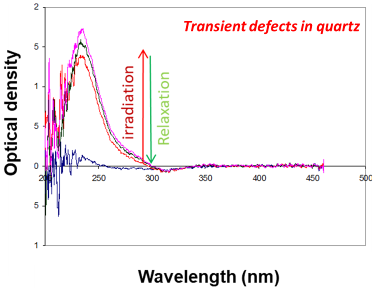


Figure: *in situ* absorption spectrum of an electron-irradiated quartz crystal type USO

Radiation-induced transient defects in the quartz crystal structure responsible for the radiation sensitivity of quartz USO

Cibiel G. *et al.*, 2006 *Ultra stable oscillators dedicated for space applications: oscillator and quartz material behaviors vs radiation*, E3S Web Conf., **16**, International Frequency Control Symposium and Exposition, IEEE

SIRIUS :

- is a platform dedicated to the production and study of electron-irradiation induced defects in materials
- hosts numerous equipment for online measurements
- welcomes an ever increasing number of international users
- is involved in a wide range of R&D topics

SIRIUS is a part of EMIR, the french network of accelerators dedicated to material irradiation...

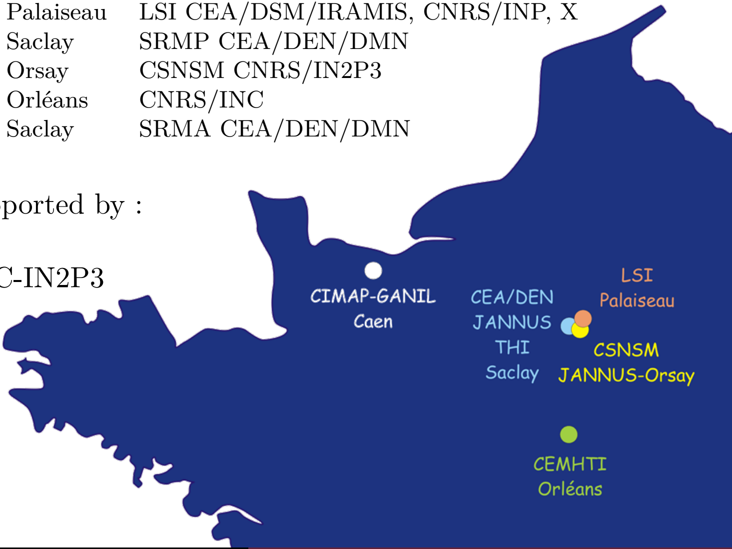
The EMIR federation

Five complementary platforms :

GANIL	Caen	CIMAP CEA/DSM/IRAMIS, CNRS/INP
SIRIUS	Palaiseau	LSI CEA/DSM/IRAMIS, CNRS/INP, X
JANNUS	Saclay	SRMP CEA/DEN/DMN
	Orsay	CSNSM CNRS/IN2P3
CEMTHI	Orléans	CNRS/INC
HVTEM	Saclay	SRMA CEA/DEN/DMN

EMIR is supported by :

- CNRS
- INP-INC-IN2P3



The EMIR platforms

Main specificities of the EMIR platforms (*http://emir.in2p3.fr*)

Platfom	Specificities		*in situ* characterizations
	IONS		
CEMHTI *(Orléans)*	*HIGH-ENERGY LIGHT IONS* Cyclotron H→He 10 – 45 MeV VdG H→He 0.5 – 3 MeV	• Large range of ions • 4 beam lines	• RBS • Variable temperature Raman • Mechanical stress
GANIL *(CIMAP Caen)*	*SWIFT HEAVY IONS* IRRSUD C→U 10 – 90 MeV SME H→Bi 0.5 – 570 keV	• Large range of ions • Effects dominated by electronic excitations	• X-ray diffraction • IR and UV-visible spectroscopies • Gas emission
JANNUS *(CSNSM Orsay)*	*DOUBLE BEAMS* ARAMIS 2.0 MV H→Bi 0.5 – 15 MeV IRMA 170 kV H→Bi 0.5 – 570 keV	• Irradiation or implantation and TEM observation simultaneously • 100 K < $T_{irradiation}$ < 1200 K	• Transmission electron microscopy • Raman and RBS spectroscopy, ERDA, NRA
JANNUS (*SRMP Saclay*)	*TRIPLE BEAMS* EPIMETHEE 3.0 MV H→Bi 0.5 – 30 MeV JAPET 2.5 MV H→Bi 0.5 – 12 MeV PANDORE 3.0 MV H→He 0.5 – 3 MeV	• Irradiation and implantation simultaneously • 77 < $T_{irradiation}$ < 800 K	• Raman spectroscopy • Rutherford backscattering spectrometry (RBS) • Elastic recoil detection analysis (ERDA) • Nuclear reaction analysis (NRA)
	ELECTRONS		
HVTEM *(SRMA Saclay)*	1.0 MeV	• Irradiation inside a high voltage TEM • 300 K < $T_{irradiation}$ < 1000 K	Imaging of structural X-ray diffraction, environmental electron microscope
SIRIUS *(LSI Palaiseau)*	*CONTINUOUS ELECTRON BEAM* 2.5 MeV	• Large irradiated area • Large energy range • 20 K < $T_{irradiation}$ < 670 K	• Electrical measurements (Hall effect, resisitivity, IV characterization) • Raman spectroscopy, absorption, time-resolved PL, cathodoluminescence • Corrosion under irradiation • *Electron paramagnetic resonance being developed*