Report on the needs and recommendations for interdisciplinary research at GANIL for the Michel SPIRO Commission

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INTRODUCTION

Interdisciplinary research is in GANIL's DNA! It has been included in the scientific ambition of the facility since its design. With a dedicated experimental area for few-electron ion spectroscopy, known as LISE (Ligne d'Ions Super Epluchés), and a material irradiation chamber, interdisciplinary research was possible in the initial building. From the very beginning, 10% of the beam time was allocated for interdisciplinary research. The experiments were selected by a specific GANIL Program Advisory Committee for "Non-Nuclear Physics", known today as iPAC (Interdisciplinary Program Advisory Committee). The role of this committee has been of tremendous importance as it helped in structuring the scientific objectives of this new branch of science using swift heavy ions. The third important decision for GANIL was to create a new laboratory, CIRIL (now renamed CIMAP and originally attached to CNRS/INP and CEA/IRAMIS institutes) that is in charge of the GANIL interdisciplinary users and is embedded in the respective scientific communities. Several tasks have been assigned to CIRIL:

- beam time allocation and user support
- proposal management together with iPAC
- development of on line instrumentation for GANIL interdisciplinary users
- animation and expansion of the non-nuclear scientific community

From today's perspective, one can say that these first ingredients, combined with the strong engagement and synergy of the members of GANIL and CIMAP, are the recipe for scientific success. Indeed, during the first period in the eighties, the availability of high energy ions led to world class results in QED effects in atomic spectroscopy, in ion channeling in crystals and in the first description of the damage morphology of ion tracks soon followed by ion track technology. Also the first radiobiology experiments were performed at this time. This very active community soon started to suffer from the limited beam time as new scientific questions arose. Thus, at the end of the eighties, a new project, coming from a previous idea known as GANIL INTER, has been pushed by CIMAP and GANIL in order to increase the beam time. This project led to the SME (Sortie Moyenne Energie) that allowed an increase of the beam time available for the GANIL/CIRIL users by a factor of ten. This first step toward a multi-users facility was decisive for interdisciplinary research as it extended the access to GANIL beams in particular to an energy range at the maximum of the electronic stopping power. A consequence of the significantly expanded scientific community was the launching of a new conference series initiated by CIMAP (CIRIL) and GSI, the International Symposium on Swift Heavy Ions in Matter (SHIM). Today, SHIM is a well-established international conference with typically more than 150 participants. The 11th edition was to be organized in 2020 in Helsinki (postponed to 2022!).

The positive dynamics was on its track! After the successful installation of SME, several other beam lines were constructed (ARIBE, IRRSUD) opening an even wider and unique range of energy and ions to up to 4 simultaneous users. This multi-user configuration allows the different interdisciplinary research communities to meet and connect, leading to the emergence of new projects. This is only possible because of the very specific GANIL accelerator configuration of 3 cyclotrons in cascade. Due to the parallel operation for several users, interdisciplinary research has about the same amount of beam time as nuclear physics.

Moreover, GANIL is not only attractive because of the quality and variety of its ion beams. All along the years a continuous effort has been put in the development of *in-situ* material characterization techniques for the users: X-Ray diffraction, IR/Visible/UV emission/absorption photon spectroscopies, but also on-line gas analysis, low temperature irradiation, biology sample preparation laboratory, etc. This original and unique instrumentation (which nuclear physicists would call detectors) guarantees the quality of the scientific results with GANIL ions.

Beside this success in the attractiveness of GANIL for interdisciplinary research, great efforts have been devoted to its visibility. CIMAP coordinated the GDR PAMIR, the EMIR&A Federation and the European infrastructure networks LEIF and ITS-LEIF. CIMAP was a partner of the Labex EMC3, the INFRA France-Hadron and the European infrastructure networks SPIRIT and RADIATE. On the PIA front, CIMAP has proposed several Equipex in order to enrich the tools at disposal of the GANIL users:

- GENESIS: several instruments obtained for nanoscale analysis of radiation induced defects in materials (FIB/MEB and X-Ray diffractometer). Obtained in 2011
- DIAPASON: coordinated by EMIR&A. Strongly supported by IN2P3, INP and INC. Positively evaluated but not selected in 2020
- e-DIAMANT: coordinated by ENS Paris Saclay. Production of low energy nano-beams at GANIL for quantum magnetic sensors. Obtained in 2020.
- NEWGAIN: coordinated by GANIL. Future possibility of material irradiation at the second injector of SPIRAL2. Obtained in 2020.

The dynamism of this scientific community, which has never decreased since its birth, is ultimately evident in the wide variety of research performed at GANIL as highlighted in a special issue of the Journal of Physics on the occasion of the thirtieth anniversary of interdisciplinary research at GANIL (http://iopscience.iop.org/1742-6596/629/1), as well as in the number of RICL (1153), conference communications (914), PhD theses (191) and number of contributing authors (2788).

As requested by the Spiro commission, this report presents the achievements, the projects and the needs of the interdisciplinary community at GANIL. The following chapters focus on (i) Material Science: inorganic materials, (ii) Material Science: organic materials, (iii) Astrochemistry and (iv) Radiobiology. Two other important activities with GANIL beams are not presented in this report. Atomic and molecular physics research was already present with the first GANIL beams. However during the last decade, these activities moved more and more toward the low energy beams (ARIBE beam line) and only a few experiments are using the cyclotron beams (see appendix for an overview of this research sent as contributions to the future of GANIL in March 2020). The second activity not included explicitly herein concerns the tests of electronic components because they are managed by the industrial application department of GANIL.

This report has been written by the following group of international experts:

- Marika Schleberger, University of Duisburg-Essen, Germany, representative of the GANIL interdisciplinary research user in the GANIL Users Executive Committee (GUEC)
- Christina Trautmann, GSI Darmstadt, Germany
- Muriel Ferry, CEA/DES Saclay, France
- Claire Rodriguez-Lafrasse, CNRS/IN2P3/IP2I Lyon, PUPH Université Lyon I, France
- Eric Quirico, IPAG-Université Grenoble-Alpes, France

Coordinated by:

- Nathalie Moncoffre, CNRS/IN2P3/IP2I Lyon, France
- Amine Cassimi, CEA/IRAMIS/CIMAP Caen, France

Material science: Inorganic Materials

The **scientific topics** being addressed by materials science researchers at the GANIL facilities are directly related to the specific properties of the swift heavy ions delivered at the GANIL beamlines. These ions deposit enormous energy densities into solids and drive the local atomic and electronic structure far out of equilibrium. This results in ultrafast phase transitions and complex structural modifications, including transient and permanent radiation damage, which are otherwise unobtainable. The details of these processes depend strongly on the thermal, electronic, and structural properties of a given material. Materials science research is therefore dedicated to understanding fundamental mechanisms of ion-solid interaction and of ion-induced material modifications, as well as developing novel applications and ion beam analysis tools. Due to the complexity of the ion-solid interaction and subsequent processes, even seemingly well-established applications are still being explored and offer the opportunity for exciting research. The many challenges in this field can only be tackled with ion beams covering a wide mass, charge, and energy regime and on-site, state-of-the-art experimental equipment.

Interdisciplinary materials research topics include:

- Investigation of fundamental processes, i.e., ion-induced excitations/ionizations and the subsequent mechanisms in inorganic materials as well as understanding combined effects due to electronic and nuclear energy losses.
- Understanding radiation-induced damage processes as a key prerequisite for safe operation and reliable lifetime estimates of devices and components to be used in extreme dose environments and space applications.
- Tailoring structural, optical, electronic, magnetic, thermal, and other material properties through defect and Fermi level engineering.
- Applications in nanotechnology based on ion-track membranes as filters, sensors or templates for the synthesis of nanowires.
- Ion-induced effects in complex solids such as ceramics, highly refractory high entropy alloys, metallic glasses, mesoporous silica, metal-organic-frameworks, and 3D-printed materials.
- Ion-induced modifications radiation tests for spintronics, nanomagnetism, MRAM devices, piezogenerators, and sensors.

- Processes related to focused ion beam induced deposition; ion nanobeam development for defect engineering on the single-ion level.

New challenges and future topics

Future developments in materials research is strongly driven by recent advances in solid-state physics related to materials synthesis, time-resolved experimental techniques, and multiscale predictive modelling techniques based on massive computing power and machine learning. Further impetus comes from environmental and societal challenges that drive, for example, the optimization of materials for storage, conversion, or harvesting of energy. Highly attractive fields where ion beams can provide unique contributions concern tailoring quantum and energy materials such as Mott and topological insulators, magnets, superconductors, and van der Waals heterostructures at the nanoscale. Moreover, ion-beam induced non-equilibrium conditions may relax via so-called hidden states that cannot be reached within the equilibrium energy landscape. This provides access to novel phase transitions and unique states, as well as unprecedented possibilities of control. This leads to new challenges as characterization of the adiabatic final state is no longer sufficient. Pump-probe-type experiments combining short ion pulses of different type and laser triggered detection schemes are required to address e.g. ion-induced transient electronic states, formation of intermediate species, or lattice dynamics.

Uniqueness of GANIL

For materials science, GANIL´s versatile infrastructure is indispensable due to the unique combination of the four experimental stations (HE, SME, IRRSUD, and ARIBE). This combination is perfectly suited to select specific contributions of electronic and nuclear energy loss, thus exploiting or avoiding their synergy. Worldwide, there are only very few beamlines in the high-energy range (MeV-GeV) dedicated to materials science and of these, most are not ideally suited because they are limited in beam energies and/or intensities, or do not (yet) have the onsite expertise and infrastructure required for materials science. The specific advantages of the GANIL facilities are the large variety of ion species up to uranium and the broad energy range. The uniqueness also includes the sophisticated on-line equipment available for in-situ analysis, a capability that is of particular importance for materials science. Last but not least, it should be mentioned that the scientific expertise and support of the CIMAP team has an excellent reputation in the national and international user community.

Links to applications and industry

The opportunities for applications are manifold and are outlined in the research topics. Direct links exist to the nuclear power industry, to companies that rely on ion beam analysis tools, and to topics related to track-etched membranes.

Recommendations

Newly discovered classes of materials, progress in experimental techniques, and advances in modeling have led to novel topics in materials science and will certainly lead to an increasing demand for beam time and state-of-the-art experimental equipment in the future. In order for GANIL to maintain its excellent reputation, continue to expand its leading role in materials science, and focus its activities on future scientific topics in this field, the following conditions must be met:

- The unique selling point for materials science is the versatility of GANIL, i.e., the possibility to select the most suitable energy regime (left and right of the Bragg maximum), ion range, and charge state to address specific scientific questions. To maintain this unique selling point in the future, the four cyclotrons should be refurbished or even renewed. Their combined operation should be maintained as well as the in-parallel operation of the low-energy beamlines, which provides flexibility and improves the overall access to beam.
- This could be further improved by extending the available energy range towards very low energy (eV) ions and by providing more access to the high energy beam for materials science. The intermediate energy range, particularly targeting the energy regime between IRRSUD and ARIBE where the largely unexplored synergistic effects become important, could be significantly strengthened by the addition of an independently operating van-de Graff accelerator. Ideally, this set-up would enable dual-beam irradiations and target analysis via in-situ Rutherford Backscattering Spectroscopy (RBS).
- The specific needs of the community require that sufficient beam time be allocated on a regular basis. Some experiments require multiple beam times over several years, others multiple runs over the year, often with different beam parameters. This means that multiple beam time slots should be allocated, ideally in all three trimesters. The total amount of beam time should be increased, back to the level of ten years ago, when beam time was provided in four two-month runs during the entire year. This is also strongly recommended to attract and maintain international collaborations and ensure that PhD students can complete their theses in a timely manner.
- Recently, allocated beam times have been cancelled on several occasions due to technical failures. In order to provide sufficient beam time, the reliability of the cyclotrons must therefore be improved.
- To address the challenges related to emerging and future topics discussed above, the current experimental infrastructure should be further developed, focusing on (i) better defined target systems that can be studied in-situ (heating of samples, UHV, load-locks, inert conditions), (ii) the development and implementation of additional on-line and in-situ spectroscopy techniques (Raman, electron paramagnetic resonance, RBS), (iii) the development of time-resolved experiments on all time scales, i.e., slow (up to seconds), fast (10^{-8} s) and ultra-fast ($<10^{-12}$ s), requiring modifications of the accelerator (e.g. an ion bunch suppressor) and the development and implementation of appropriate detection techniques. New opportunities and synergies arising from SPIRAL2 in this respect should be exploited.
- It is imperative to complement the technical infrastructure with excellent user support, as currently provided by the CIMAP/CIRIL team with their in-house scientific expertise and highly qualified technicians and engineers.
- Continuous efforts should be made to keep the materials science community informed about the unique opportunities offered by GANIL.

Material science: Organic Materials

Radiolysable materials (RMs[1]) are decomposed under ionizing radiation and the decomposition mechanism is atmosphere- and temperature- dependent. At GANIL, the studies of RMs encompass ***basic research*** on Swift Heavy Ions (SHI) interactions with matter, ***upstream research*** mostly related to the ***nuclear power industry (NPI)*** and ***applied research*** as the ***Ion Track Technology (ITT)***; each of these domains allows improving the knowledge in the two others.
Whatever the research type, the forthcoming document demonstrates how GANIL is unique and vital for all these domains.

From basic research

Basic research with SHI on RMs at GANIL has the aim to understand the influence of high densities of electronic excitations induced by SHI on the radiation-induced modifications in RMs in terms of structure, type and concentrations. Studying the combined effects of the irradiation environment, the temperature and the energy loss (LET) has allowed, among many others, a better understanding of the main difference between SHI and γ or β irradiations. The ultimate goal of basic researches is to coin a comprehensive model enabling the prediction of the radiation-induced evolution of any RM, based on its chemical composition, its microstructure and the irradiation characteristics (LET, environment, temperature...). The ***presence of oxygen during irradiation*** induces one of the major aging processes leading to a severe loss of properties in organic materials. Predicting the oxidative radiation-induced evolution of these materials requires an excellent understanding of the oxidation chain reaction kinetics and the knowledge of the corresponding kinetic constants. Available data are based on conditions not taking into account the specific properties of the energy deposition induced by particles and some of the hypotheses have not experimentally been proven. Future experimental and theoretical works, ***with the help of the CIMAP staff,*** are planned with the aim of accessing reactive species involved in fast and ultra-fast reactions to directly access kinetic constants.

[1] For instance: organic molecules and macromolecules (polymers) and water containing geopolymers.

Through applied research

Although many challenges remain in basic research, the advances in this domain have laid the foundations for several ***upstream research domains***.
Knowledge on synthetic ***polymers under SHI can efficiently be applied on biological materials*** (e.g. triple helix structure of Collagen II) to evaluate their behavior under hadron therapy. The objective is to understand how the biomechanical properties of healthy tissues are altered under carbon ion irradiation during the hadron therapy treatment of inoperable, radiation and chemically resistant cancers. Besides, the efficiency of carbon irradiation on tumor cells is expected to be increased by combining nanoparticles targeting and pulse irradiations. However, prior to any application, the underlying mechanisms should be addressed through the use of ***pulse radiolysis techniques with different time resolutions***.
Provided ions and energies are chosen to meet the LET and the radial dose repartition induced by α particles, the evolution of RMs under SHI irradiation is also employed in the NPI domain to ***simulate the radiolysis of RMs in contact with actinides***. Understanding and modelling the polymers' ageing in the context of long-lived medium-activity nuclear waste management (long time, *i.e.* high doses) is of a ***crucial societal interest***. The innovative use of SHI at GANIL in the NPI framework allows homogeneous irradiation of materials over greater thicknesses and under increased safety, and has enabled the acquisition of essential data, mainly but non-exhaustively, on:

1) The safety of RM-rich (mainly polymers) nuclear waste packages under disposal, transport and storage conditions,
2) The safety and the integrity of the confinement matrices (bitumen and geopolymers),
3) The safety and the efficiency of the spent fuel reprocessing process at La Hague.

Results from basic research on radiation-induced oxidation will also bring new outcomes in the NPI for the simulation of long term behavior of polymers constituting cable sheaths, composing organic-rich nuclear waste packages or forming the confinement matrix. The ***deep knowledge of the CIMAP staff*** on RMs behavior under SHI irradiations is of a great importance as, in presence of new RMs, it is possible to quickly target the relevant aging conditions and to optimize the cost associated with the experiments. Therefore, reducing the delay necessary to answer specific requests of ANDRA, related to the CIGEO deep repository of RMs-rich nuclear waste packages, to providers such as ORANO, CEA and EDF.

To industrial research

The wealth of knowledge acquired through basic research on RMs under SHI at GANIL has set off several domains of ***industrial research***. SHI are widely used to structure polymers at the nanometric scale thanks to the formation of ion tracks. By chemical etching, these trackscan be converted into open channels and be used either as they are (membranes for filtration such as commercial Nucleopores membranes from Whatman™ or membranes for osmotic energy recovery) or functionalized. Nanostructured functional polymers have gained growing interest in high value-added applications such as sensors, analytical devices and/or diagnostics, especially for lab-on-chip applications. Pushed by the recent EU policy on environmental norms, important industrial groups in France such as VINCI Construction for soil leachate («Quick Soil Analysis» CEA project - norm NF-EN-12457-2-) and TOTAL for on-site water quality control in offshore oil production platforms (OSPAR regulation), have already financed R&D on these sensors; more is still to come. Based on their expertise in functionalizing nanopores

made by ITT in polymer thin films, PCnano team (CEA/DRF//LSI) is developing a portable early warning sensor for toxic metals that monitors the water quality. The future of membrane irradiations for ITT appears very bright with a growing number of industries interested in such sensors. A recent market study by Erdin Cabinet (2018) revealed that the Business to Business market represents around 135 M€ in France, 900 M€ in EU and 3.2 billion Euros in the world. Ongoing Business to Client market study is under evaluation revealing that customers are ready to pay for water tests at home. Sensors for lab-on-chip medical applications are on constant growth.
Within the framework of ITT, the device developed for single ion irradiation of membranes, used in lab-on-chip applications, enables an opening rate close to 90%; the highest among SHI irradiation facilities.

GANIL uniqueness and requirements

The uniqueness of ***GANIL in its current form, i.e. the two cyclotrons***, is given by:

1) The competence of the CIMAP staff, to understand the results and to conceptualize unique or near to unique set-ups.
2) The multidisciplinary and the versatility of ion beams and lines
    - Access to X-Y scanned beams on wide surfaces (6x5 cm^2)
    - Similarity of the dosimetry procedure across the lines
    - Unique variety of available ions and the wide energy range enabling access to a wide map of ion/energy couples with a wide range of LETs and radial dose distributions

The ***CIMAP staff has acquired a unique expertise*** in the development of specific set-ups for real-time and *in-situ* monitoring of radiation-induced alteration of RMs over a very broad temperature range (8K - 500K) but also under different atmospheres. The next developments are dedicated to *in-situ* and time-resolved analyses (ESR at first glance) to access reactive species in radiation-induced oxidation.
Studies on RMs use mainly three beam lines:

1) Basic research: ***IRRSUD*** (C to Kr, up to 1 MeV/A), ***IRASME*** (C to Xe, up to 13.7 MeV/A) and ***IRABAT*** (C to Ar, up to 95 MeV/A),
2) Applied research: ***IRASME*** (C to Ar, 6-13 MeV/A) and ***IRABAT*** (C to Ar, up to 95 MeV/A),
3) Industrial research: ***IRASME*** (Xe or Kr 10 MeV/A).

The ***annual beam time*** necessary for ***a dynamic and quality research on RMs***, is evaluated at ***1 to 2 weeks on IRRSUD, between 9 to 13 weeks on IRASME*** and ***at least 2.5 weeks of C and Ar beams on IRABAT***. For an agile research on RMs and provided the robustness of the beam, those beams should be ***scheduled*** not as a unique beam session for each beam type but ***as a two-session beam time on two trimesters***.
Finally, ***GANIL favors collaborations between different communities*** in many ways, paving the way to very innovative researches, which have merged thanks to:

- Sharing set-ups developed for one community has allowed another one to perform some innovative and pioneering experiments,
- Providing several beams at the same time in a unique place accelerated the communities meeting and led to the emergence of new projects with mixed competences, for instance:

- Organic materials / radiobiology
    - Polymers to mimic biological systems
- Organic materials / inorganic materials
    - MOF (Metal–Organic Frameworks): very radiation resistant materials
    - Inclusion of metal nanoparticles in polymers to increase in a controlled way the local dose deposition
- Organic materials / astrochemistry
    - Irradiation of glasses made of small organic molecules at cryogenic temperatures

One main evolution could ***impact positively and trigger new research for RMs*** at GANIL. The ***construction of a low energy*** (between ARIBE and IRRSUD energies) ***line in the D1 cave (IRASME) so to combine low and medium energy*** irradiations, leading to the combination of multiple LET and radial dose distributions. This new line would allow taking fully into account the complexity of irradiation in nuclear waste packages.

Astrochemistry

Achievements and new topics

At the crossroad of astrophysics and chemistry, *astrochemistry* is devoted to the chemical evolution of the Universe, in particular Interstellar medium (ISM), proto-planetary disks and Solar System Objects. Since the late 2000s, experimental astrochemistry has become a very active field of research at GANIL. The opening of the CASIMIR, and then the IGLIAS experiments to the community has offered unique opportunities to investigate various aspects of ion irradiation in the context of astrochemistry. Three fields of research have been mostly investigated:

(1) *Galactic Cosmic Rays (GCR) induced-chemistry of ices in interstellar-medium (ISM)*. Extensive studies have focused on ice radiolysis and sputtering, providing insights into molecular complexification, sputtering yields and branching ratio of sputtered species that feed the gas phase. By-products such as radiolytic cross-sections and sputtering yields have been extrapolated to the full range of GCR energy and ions, have been made available to feed complex chemical models, and are valuable help to interpret radio observations.

(2) *Ion irradiation at the surface of air-less small bodies in the Solar System*. Solar wind (SW), solar energetic particles (SEP), magnetospheric ions in the environment of giant planets (MI) and GCR are the main sources of ions that irradiate and modify to various extents the surface materials of air-less small bodies. This covers a broad range of energy between 1 keV/u to several GeV/u, resulting in ion implantation depth of a few tens of nm up to several meters and doses up to several hundreds of eV/atom in case of lack of recent resurfacing processes. The ion-surface interactions include both elastic and electronic regimes, and result in complex chemical and structural transformations of the surface materials. Flux, doses and nature of the ions in the local environment of planetary bodies are estimated from satellites and spacecraft measurements, but experiments are essential to stress the chemical and optical effects of irradiation. Experiments driven at GANIL have provided new insights about the main radiolytic by-products synthesized at/in the icy surface of Trans-Neptunian Objects, on their spectral properties (reddish surface) and new constraints into the origin of the Ctulhu organic belt of Pluto.

(3) *Radiolytic formation of carbonaceous compounds in primitive asteroids and comets*. This topic concerns two new scenarios proposed for the origin of N-rich ultracarbonaceous Antarctic Micro-Meteorites (UCAMMs) and Insoluble Organic Matter in primitive chondrites.

The activities on ISM chemistry and air-less small bodies will be continued and even amplified, due to the forthcoming observations of the JWST (James Web Space Telescope), which will be launched on October 2021.

Emerging topics for 2021-2031

The JUICE mission operated by ESA (European Space Agency) will orbit the Jovian moons Ganymede, Europa and Callisto in 2030. These satellites are exposed to magnetospheric ions and electrons that deliver huge doses and dramatic modifications of their surfaces. A large magnetospheric ions flux (H, S, O) covers the range of 1-10^5 keV, **hence GANIL will be essential to investigate the radiolytic chemistry and the formation of teneous exospheres by sputtering experiments (IRRSUD and ARIBE beamlines)**. Low-energy facilities usually used in planetary sciences will not be suitable to investigate the full energy range. The Hayabusa 2 and OSIRIS-ReX missions will also benefit from GANIL experiments, which will help **interpreting micro-analysis of the returned asteroidal samples**. A comprehensive investigation of SW implantation could be achieved, including the contribution of the suprathermal tail.

Uniqueness of GANIL:

Several low-energy accelerators (< 1 MeV/u) are worldwide dedicated to Astrochemistry studies: University of Catania, Italy; NASA Godard Space Center, Maryland USA; CSNSM University Paris-Saclay France. They usually accelerate light ions (H, He) and are not suitable to investigate processes at high energy and heavy ions. In addition, at low energy the irradiated zone in samples is a very thin layer, which critically limits *in situ* and *ex situ* characterization of the sample. More energetic ions circumvent this problem by delivering thicker irradiated samples (> 1 µm) and offer a broader analytical strategy. **GANIL offers unique opportunities because it operates across a broad range of energy (keV-GeV) with ions from carbon to uranium.**

Recommendations

1. The first recommendation is, needless to say, to maintain the interdisciplinary approach at GANIL. This is definitely a unique opportunity for European Astrochemistry. This action warrants very competitive original research, and a support to major space missions like ESA-JUICE.
2. An accelerator filling the energy gap between ARIBE and IRRSUD (0.3-1 MeV/u) would be well suited for planetary studies. This range is of interest for the suprathermal tail in SW and MIs. This accelerator would handle light ions as well, and its implementation along with existing cyclotron GANIL beams on the same experimental set-ups would allow studies of synergetic effects in mixed radiation fields.
3. The existing set-ups for users are based on FTIR spectroscopy. The IGLIAS experiment is in addition operating with a QMS spectrometer and FTIR/VUV spectrometers. A significant improvement would consist in the implementation of a dedicated TOF-SIMS

setup for a better characterization of sputtered by-products, possibly with a laser-desorption system. The implementation of a Raman microscope, as in Catania and GSI-Darmstadt, would allow characterizing IR-inactive species and carbonaceous materials.

Radiobiology

Context of heavy ions in the arsenal of innovative radiotherapies

Heavy ions exhibit a huge density of ionization within individual tracks resulting in very localized reactive oxygen species (ROS) production within cells, unlike X-ray irradiation where the ROS production is spread uniformly. This ROS distribution at the nanometric scale explains the higher relative biological effectiveness (RBE) of carbon ions, which results mainly from unrepaired complex DNA lesions, and consequently, leads to higher tumor control. Their therapeutic advantages also result from fewer adverse effects (cancer progression, metastasis, angiogenesis ...) due to the non-activation of survival and defence pathways which are under the dependence of a ROS threshold not reached in the cell. This phenomenon, which can be described as the "stealth bomber effect", places carbon therapy above any photon radiotherapy technique. In the field of innovative radiotherapies, it is therefore essential to pursue research on heavy ions, particularly by exploring those (He, …) of an atomic number between protons, whose RBE is little above 1, and carbon whose cost of production is very high. Studying the biological effects of heavier ions (O, N, Ne …) could also be of great interest for clinical transfer since they have a higher RBE and less lateral diffusion.

Current scientific topics:

The research in hadron biology, resulting from experiments of local or external teams with heavy ions beams at GANIL, can be classified according to three main axes:

- Acquisition of biological data for the improvement of the simulation models used for the calculation of the biological dose implemented in patient treatment planning
- Effects of carbon ion irradiation on cancer cells - as relevant for hadron therapy:
  - Specificities of the molecular response to heavy ions, radioresistance, cancer progression, and metastasis
  - Efficacy of combined treatments (nanoparticles, radiosensitizers (PARP inhibitors, cetuximab, immunotherapy ...))
- Effects of accelerated heavy ions on normal tissue - as relevant for hadron therapy and space missions:
  - Side effects of hadron therapy on normal tissues; bystander effect
  - Space radiation effects on different cell types

Future topics:

The three main axes described above should be maintained and further included in the coming year's experiments with:

- 3D cellular models or organoids closer to tumors, for testing simulation models and also correlating biological results with the clinical outcome;
- Animal models with xenografted or syngeneic tumors, to study the efficacy of hadron therapy combined with promising adjuvant treatments such as immunotherapy or inhibitors of DNA repair (DNA-PK, PolQ…); or metastasis/ secondary cancer formation;
- An expanded ion panel: Helium, Oxygen, Nitrogen… for development of innovative radiotherapies and new applications of space radiation research;
- Beam evolution to enable (i) very high dose-rate as 10 Gy/ms for Flash ion therapy; (ii) low doses (up to 1 mGy), low energy, and low-dose rates (up to 1cGy/min) for space radiation research.

Uniqueness of the GANIL facility in the context of radiobiology research:

GANIL has considerable strengths in the worldwide arsenal of ion accelerators:

- It is the only French platform enabling access to high energy ions:
  - between carbon and uranium
  - with variable energy of beams and precise dosimetry
  - with the assistance of CIMAP technicians and ARIA biologists for experiments
  - with free access after selection by an expert committee (iPAC)
- It also benefits from:
  - a fully equipped laboratory for cell biology experiments (cell culture with or without hypoxia, microscopy…), biochemistry, and molecular biology with the team hosting;
  - a nearby guest house to accommodate the teams.

Recommendations:

- Beam time:

Despite all these strengths, the time allocated to radiobiology at GANIL is extremely low compared to that devoted to physics experiments. For the 2016 - 2020 period, only 34.5 UTs (i.e. less than 7 UT/year) were dedicated to biology. Moreover, out of these 34.5 UTs, due to beam failure, only 24 UTs used by local, national, and international teams.
The beam time available for radiobiology is very insufficient, considering the international competition in the domain (Germany, China, Japan, Italy…), and therefore, the period for publishable scientific advances is too long. Consequently, teams have to look for beam times in other countries which costs more and is more difficult to organize. Besides, many radiobiology teams in France have turned away from experiments with ion beams due to the difficulty of carrying them out and the time required to be able to publish results, since research evaluation is lying on publication records.
Nevertheless, the dynamics of the teams carrying out heavy ion irradiation of cells at GANIL has led to at least 48 scientific publications with a good impact factor (IJROBP, Cancer Letters,

Front Oncol, Cancers...) over the period 2005 - 2020 and 38 oral presentations in international conferences.
It is essential to increase the beam time available at GANIL for radiobiology whether it is oriented towards clinical radiotherapy or space research. It is also mandatory not to wait for the deployment of CyclHad in 2024 to strengthen the radiobiology of ions in France and to prepare the transfer of discoveries to the clinic. The time allocated for radiobiology at GANIL must be extended beyond 2024 because the beam parameters (energy, dose rate, available ions) which will be proposed by CyclHad will not allow all the research programs to be carried out successfully.
Three independent repetitions of the experiments being requested before the publication of data, multiple (3-4 times) access to beam times (1 or 2 UT/session) per year/team would be necessary.

- Ion delivery:

- Irradiation at different positions in the Bragg peak
- Very high dose-rate (10 Gy/ms) for Flash ion therapy low doses (up to 1 mGy), low energy, and low-dose rates (up to 1cGy/min) for space radiation research
- For space radiation research, access to ions with Z numbers up to 26, that are abundant in the galactic cosmic ray spectrum (He, Be, C, O, Ne, Mg, Si, S, Ar, Ca, Ti, Cr, Fe)
- Robustness of the beam to not cancel, at the last moment, very expensive cell experiments that have been prepared for several weeks.

- New set-ups and techniques:

- Modification of the opening of the concrete door (entering and leaving the room requires 15 minutes) to be able to make irradiation kinetics of fewer than 30 minutes;
- Improvement of the online dosimetry:
    - reduction of time for initial dosimetry and for changing the dose-rate
    - use of some adapted ionization chambers and/or specific online software as Dosion
- Adaptations of sample holders and size of the irradiation field to enable:
    - the easy and fast transition from two-dimensional cell culture to three-dimensional culture and organoids
    - the control of the temperature and O_2 concentration
    - the irradiation at low energy
- Improvement of the reception capacities of ARIA by an increase in cell culture equipment (PSM, incubators with oxygen control, multiple centrifuges)
- Development of a biology room adjoining the irradiation line with an incubator and a PSM to maintain cell flasks between two room-accesses.

Position of GANIL facilities in the European and international context

Uniqueness of the GANIL/CIMAP facilities

The uniqueness of the GANIL/CIMAP facilities is based on the combination of the high- and medium-energy beamlines HE and SME as well as the low-energy beamlines IRRSUD and ARIBE.
The facilities provide excellent conditions for many disciplines including material science, solid state physics, surface science, crystallography, mineralogy, geosciences, nanotechnology, space science, astrochemistry, biology and many others. The research activities in these areas are extremely diverse and are subject to rapid scientific developments.
The GANIL/CIMAP platform goes far beyond accelerators or implanters available at universities and in the semiconductor industry. They offer unique possibilities and the following specific advantages:

- All ion species from carbon up to uranium are available; by the combination of the two cyclotrons, a broad variety of ion energy loss values is accessible.
- The users can select the most suitable energy regime (left and right of the Bragg maximum), ion range and charge state according to their specific project.
- Beams from low to extremely high energies (few keV/n – 100 MeV/n) cover energies across more than 6 orders of magnitude. The high energies are not provided by any other French facility and correspond to extreme ion ranges enabling the irradiation of macroscopic samples of up to mm-thickness.
- Sophisticated equipment installed at the different beamlines (e.g., x-ray diffraction, absorption spectroscopy, heating and cooling stage) allows the users to perform measurements and analysis directly on-line and/or in-situ.

It should be emphasized that (i) the large beam flexibility is particularly important for interdisciplinary and applied research and (ii) the low energy beamlines can partly be operated in parallel, giving flexibility and enhancing the overall access to beam.

Position of GANIL facilities in the European and international context

The GANIL/CIMAP facilities are rather unique in terms of beam parameters, instrumentation and expertise of the scientific staff. Existing and/or future facilities with such broad possibilities are rare but extremely important to the national and international interdisciplinary science community.

Within Europe the following ion beam facilities provide suitable conditions for application-oriented research activities. Together with GANIL, they present the current worldwide leadership of the European facilities:

- The UNILAC and SIS-18 of **GSI** in **Darmstadt** (Germany) covers the energy regime from 1.4 MeV/n up to 1 GeV/n. With the upcoming FAIR facility even higher energies and intensities will be available. GSI operates a rather similar platform as CIMAP/GANIL with beamlines specifically dedicated to material science and with in-situ and on-line analytical instrumentation.
- The JYFL accelerator laboratory **RADEF** in **Jyväskylä** (Finland) runs three large-scale accelerators: The K130 cyclotron, MCC30/15 cyclotron and the 1.7 MV Pelletron for light and heavy ions up to Au. In particular, for electronic device testing, cocktails of heavy ion are available with maximum energies of about 16 MeV/n.
- **Louvain La Neuve** (Belgium) operates a cyclotron for light and heavy ions up to Xe (8 MeV/n) with strong activities for electronic tests and for the commercial irradiation of polymer films.

Also worldwide, the number of large-scale high-energy ion accelerators providing beamlines and instrumentation dedicated to interdisciplinary research is rather limited:

- In **Lanzhou** (China), the **HIRFL** facility (IMP/CAS) consists of the Sector Focusing Cyclotron, the Separated Sector Cyclotron, the Cooler Storage Ring (CSR), and a number of experimental terminals. All ion species (protons up uranium) can be accelerated up to low (1 MeV/n) or high (1 GeV/n) energies. The laboratory has a large material science department and operates a facility for the production of ion-track membranes. They are also active in the field of biology and space radiation issues.
- For applied topics, the **JINR FLNR** cyclotrons (IC-100 and U-400) in **Dubna** (Russia) provide ion beams up to Bi in the energy regime between 1 and 3 MeV/n. The have a material research group and produce ion-track membranes for commercial application.
- Other large-scale facilities such as **Riken** (Japan) and **MSU** and **BNL** (both USA) could in principle partly provide high-energy beams, but at the present stage they do not have dedicated infrastructures required for experiments in material science or other applied fields.

The following upcoming facilities are currently under construction and will open up new opportunities:

- The **NICA** facilty in **Dubna** (Russia) plans applied physics research with ion beams of energy 250–800 MeV/n extracted from Nuclotron. Three new experimental areas are planned. Topics of interest are radiobiology, cosmic ray simulation, particle therapy, radiation hardness of electronic devices and novel technologies in materials science.

- The particle physics laboratory **RAON** under construction near **Daejeon** (South Korea) plans to provide beams from proton up to uranium ions (200 MeV/n). They also aim at exploiting the ion beams for material science, biomedical topics and other applications, but so far have little experience.
- The cyclotron facility **CyclHad** in **Caen** plans medical and radiobiology research with beams of up to 400 MeV/n carbon ions extracted from a cryo-cyclotron under construction. The building with three experimental areas is ready. The topics of interest are clinical research, radiobiology, radiation chemistry and physics applied to hadron therapy.

RECOMMENDATIONS

The report summarizes the final recommendations to the Spiro Commission compiled by experts from various non-nuclear scientific fields concerning the future of GANIL in the field of interdisciplinary research. To date, GANIL has provided excellent conditions for this interdisciplinary community and has enabled unique and most original research activities. The uniqueness of GANIL for interdisciplinary and applied research highlighted in this report relies on the following main points:

- The broad **range ion species and beam energies** provide a perfect match with energy loss values around the Bragg peak.

- The **cyclotron cascade** allows up to three users at a time (plus one on ARIBE beam line)

- State-of-the-art **instrumentation** is available for the users for in situ and on line measurements

- The unique **expertise of the CIMAP staff** provides optimal operation and the further development of the specific instrumentation

- The GANIL platform is an important **meeting place** for users. It allows this research community to share and strengthen interdisciplinarity and to promote the emergence of new topics.

To keep GANIL in this leading and unique position, the expert committee identified several recommendations, which are summarized below:

- The possibility to provide a broad range of energies, ion species and charge states has to be kept. In the last years, the users suffered from numerous technical failures that finally led to beam time cancellations. To improve the reliability of the facility, we recommend **refurbishing or** completely **renewing the 4 cyclotrons** (C01, C02, CSS1, CSS2)

- The **cyclotron cascade acceleration** is very essential not only because of the wide range of energies available, but **also because several users can simultaneously be provided with beams.**

- During the last years, the beam time available has been strongly reduced (more than a factor of two). It is important to **increase the allocated beam time for interdisciplinary research** (originally four runs of two months per year) in order to keep the national and international activities going, attract new collaborations and ensure PhD projects.

- High energy beam time presently allocated for radiobiology is obviously insufficient. Considering the international competition as well as the specific needs of this research field, we recommend extending **the high energy beam time for radiobiology and providing short but frequent beam time slots.**

- The expert committee supports the demand of **further extending the energy range at GANIL** by filling the gap between the ARIBE and IRRSUD beam lines. This would allow **unique double-beam experiments** as well as **new in-situ characterization techniques.**

- In-situ, online and operando instrumentation is a key point attracting new national and international users. The committee strongly encourages to continue the **development of new instrumentation** according to the needs and evolution of the different research fields (e.g., EPR, RBS, Raman spectroscopy, irradiation under controlled atmosphere, …).

- Time-resolved experiments would strongly benefit from **renewing the ion bunch suppressor** ("suppresseur de paquets") which could benefit from the recent development of such a device for SPIRAL 2

APPENDIX

Contributions to the future of GANIL
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https://indico.in2p3.fr/event/20534/overview

Atomic and molecular collision physics at GANIL

L. Adoui, A. Cassimi, J.-Y. Chesnel, A. Domaracka, B. A. Huber, A. Méry, J.-C. Poully, J. Rangama, P. Rousseau, and V. Vizcaino
Centre de Recherche sur les Ions, les Matériaux et la Photonique (CIMAP), 14000 Caen, France

in collaboration with groups from the following institutions:

Institute for Nuclear Research, Hungarian Academy of Sciences (Atomki), Debrecen, Hungary
CNR-Instituto di Struttura della Materia, Area della Ricerca di Roma 1, Monterotondo Scalo, Italy
Departamento de Química, Módulo 13, Universidad Autónoma de Madrid, Madrid, Spain
Department of Physics, Stockholm University, Stockholm, Sweden
Department of Particle Physics and Astrophysics, Weizmann Institute of Science, Rehovot, Israel
J. Heyrovský Institute of Physical Chemistry v.v.i., The Czech Academy of Sciences, Prague, Czech Republic
Faculty of Applied Physics and Mathematics, Gdansk University of Technology, Gdansk, Poland
Faculty of Sciences, Siedlce University of Natural Sciences and Humanities, Siedlce, Poland
Zernike Institute for Advanced Materials, Univ. of Groningen, 9747AG Groningen, Netherlands
Tokyo Metropolitan University, Tokyo, Japan
Institut für Kernphysik, Goethe Universität, Frankfurt, Germany
Inter-University Accelerator Center (IUAC), New Dehli, India
Instituto de Física - Universidade Federal do Rio de Janeiro, Cidade Universitária - Rio de Janeiro, Brazil

Since about 35 years, the community of atomic and molecular collision physics has widely used the GANIL facility for countless experiments performed within numerous national and international collaborations. Recent reviews of the advances achieved at GANIL in this field are available here: X. Fléchard et al JPCS 629 (2015) 012001 and H. Zettergren JPCS 629 (2015) 012003. These experiments have been focusing on the study of the interaction of ions with dilute matter ranging from isolated atoms and molecules to molecular clusters, as well as nanoparticles nowadays. Thanks to the wide range of projectile energies and species available on the different beam lines of the GANIL facility, elementary processes such as electron capture, ionization and excitation have been extensively studied. Since the last years, the relaxation processes of the collision partners after the collision have been another specific source of interest.

The community of atomic and collision physics aims to provide valuable upstream information relevant for both fundamental physics and societal issues. In the field of molecular collision physics, a major goal of the present and future short- and medium-term experiments at GANIL is the investigation of the stability and the fragmentation dynamics of multi-atomic systems after their excitation/ionization

by ion impact at velocities ranging from a few tenths to several atomic units. These studies provide new insights into radiation damage processes by describing – at the molecular level – the early physical stages of ion-induced excitation and ionization of molecular systems and of their subsequent fragmentation. Another emerging foremost goal is the investigation of the formation of new molecular species in excited and ionized molecular clusters, in order to gain insights into the molecular growth occurring in the interaction between solar/stellar wind ions and the interstellar medium (ISM) and planetary atmospheres.

The uniqueness of the GANIL facility is that it offers complementary state-of-the-art beamlines producing a variety of ion species ranging from light ions (down to protons) to heavy ions (up to uranium) with a large range of charge states and with energies ranging from a few keV per charge unit at ARIBE to 0.25-1 MeV/u at IRRSUD and even up to 95 MeV/u at the high energy beamline dedicated to interdisciplinary research. This makes GANIL the most versatile facility in Europe, and possibly in the world. This versatility is a vital feature for the community of atomic and molecular collision physics as this research field requires systematic investigations as a function of the energy and/or of the charge state of the incoming ion. For example, the balance between the energy transferred to the target electrons (electronic energy loss in soft collisions) and the energy transferred to the target nuclei (nuclear energy loss in hard binary collisions) depends on the ion velocity and to a certain extend on the initial charge state of the projectile.

ARIBE and IRRSUD are currently the most widely used beamlines for the experiments carried out at GANIL in the field of atomic and molecular collision physics. Indeed, both ARIBE and IRRSUD provide a large variety of ion species with charge states and energies that are of astrophysical and atmospheric interest, as they are relevant to mimic ions of stellar winds and cosmic rays in interaction with ISM and planetary atmospheres. In addition to protons and alpha particles, stellar winds contain trace amounts of heavy ions and atomic nuclei (such as C, N, O, Ne, Mg, Si, S, and Fe). Only quite recently, it has been shown that such heavy ions can play a key role in the chemistry of planetary atmospheres and ISM medium and up to now their role was in general neglected in astrophysical models. Based on GANIL know-hows on the production of metallic ion beams, the implantation of the associated methods on the ion source of ARIBE will offer an extended range of heavy ions of interest for stellar winds (e.g., Fe). Ion beams of ARIBE and IRRSUD are also of major interest for radiobiology since they are relevant for hadron therapy and radiation damage, especially ion energies at IRRSUD are in the range of Bragg peak energies (e.g., 1 MeV/u carbon ions) and those at ARIBE are useful to mimic "post-Bragg-peak" ions at the end of their path in matter. The wide relevance and versatility of these beamlines makes the GANIL installation very attractive for our community. This attractiveness results in the fact that each call for proposals issued by iPAC leads to numerous proposals for experiments in our field, a majority of them coming from external and often foreign teams. In view of the success of the calls for proposals, a general increase of the beam time allowance at GANIL would be very welcome.

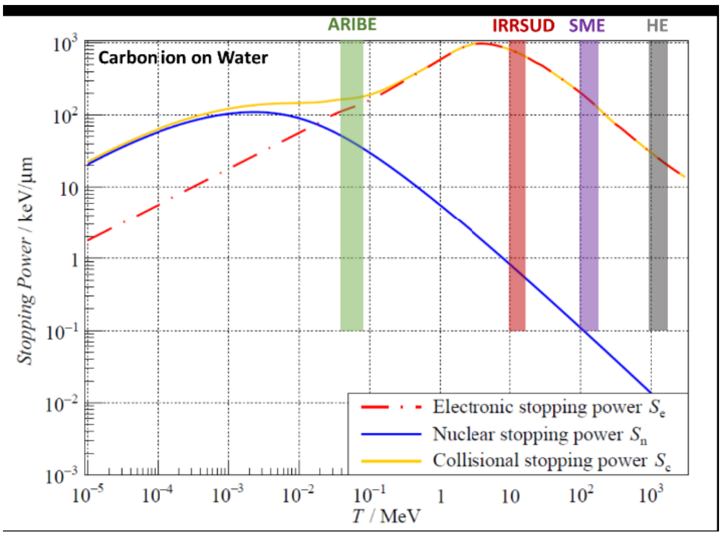
An emerging topic is the time-resolved investigation of collision-induced molecular fragmentation dynamics. While high harmonic generation sources (HHG) and free electron lasers (FEL) provide short temporal resolutions, ion storage devices give access to relaxation processes over very long time scales (up to seconds). Moreover it is possible to prepare well-defined target states with laser excitation and in a more general way to perform laser spectroscopy on ions. It is noteworthy that such laser spectroscopy in an electrostatic linear ion trap is under development as well in nuclear physics (MIRACLS project at CERN) and is foreseen for DESIR. Thus to extend the possibilities offered by the ARIBE facility, one can consider to convert the so-called "banc 2" into a molecular-ion beamline equipped with a linear electrostatic trap. This beamline is already coupled to a laser beamline and the existing laser lab can be upgraded with additional lasers. Thanks to the complementary skills of the CIMAP and GANIL researchers and engineers, we expect that the implementation of the linear trap at ARIBE can be carried out in the framework of an efficient cooperation offering new perspectives in molecular physics at GANIL and a sandbox to develop new instrumentation for nuclear physics.

As a last recommendation, we would like to emphasize that the construction of a Very Low Energy (range of a few eV) beamline at ARIBE would allow GANIL to reach the completeness of the range of energies that are relevant for experiments dedicated to stellar wind ions. We anticipate that this Very Low Energy beamline would open the way for new series of systematic studies of astrophysical and atmospheric interest, as well as of radiobiological interest.

The current projects in atomic and molecular collision physics are largely supported at regional, national and international scales. For example, in the local context in Caen, several projects leaded by CIMAP researchers are currently under progress at GANIL thanks to the financial support from the ANR, the CNRS and the Normandy Region: ANR IMAGERI, ANR FRAPA, RIN Emergence MAGIC and CNRS International Associated Laboratory LIA DYNAMO (CIMAP-Stockholm University-Universidad Autonoma de Madrid). Moreover, a large majority (> 80%) of the experiments performed at GANIL in atomic and molecular collision physics take place within the framework of international collaborations that are supported by various cooperation contracts between CNRS and foreign institutions. This strong support is encouraged by the large number of publications in peer review journals with high impact factor (> 20/year) and invited talks in international conferences like ICPEAC, HCI, ECAMP, etc, (> 10/year) reporting on our works carried out at GANIL. As shown by the success of the recent calls for proposals dedicated to interdisciplinary research, the GANIL facility is very attractive and it is essential to tackle urgent questions to understand the evolution of dilute matter exposed to ion impact in the context of e.g. astrophysics, atmospheric sciences, and radiation damage.

Collision physics: application to ion beam therapy

Ion beam therapy has a great potential for treating radio-resistant cancers, but intensive research is needed to minimize deleterious side effects while maximizing treatment efficiency. This requires efforts in clinical and biological research, but also chemistry and physics, which is the aim of the advanced research center for hadron therapy in Europe (ARCHADE) in Caen, in addition to state-of-the art proton therapy that started in 2018. ARCHADE is located next to GANIL, which has been providing beam time for hadron therapy related research since the LARIA radiobiology platform was launched.



Achieved and current work

At CIMAP, in recent years, we have increased the effort to better understand the biological, physical and chemical response of biological matter under irradiation, and thus the factors underlying the efficiency of hadron therapy. Notably, we have carried out the IRHEMME project aiming at probing the consequences of a hadron therapy of cartilage. This tissue being mostly composed of the collagen structural protein, we have applied collision physics experimental techniques coupled to mass spectrometry to unravel the molecular products of isolated collagen peptides after direct carbon ion irradiation, and found specific small molecules that might be toxic for cartilage cells and provide an explanation for the arthritis observed after cartilage radiotherapy. Furthermore, the specific triple helix structure of collagen is damaged by carbon ions. The IRRSUD beamline has been used because the energy of the ions (about 1 MeV/u) is very close to the maximum energy transfer in biological matter (, see Figure 1) which corresponds to the Bragg peak. This has been achieved thanks to an international collaboration involving the University of Groningen. Our collaboration is also investigating radiation-effects on DNA, and in particular the role of specific duplex or G-quadruplex structures on single- and double-strand breaks. The latter have been shown to play a crucial role in the death of cancer cells in radiotherapy. Therefore, much effort is nowadays dedicated to increasing the number of double-strand breaks during radio- and hadron therapy, notably by means of radiosensitizers such as nanoparticles (NPs). Indeed, the latter have been shown to be a source of electrons that induce double-strand breaks either directly or via the production of HO• radicals.

Recent theoretical work has demonstrated that electron emission from NPs results from different types of collective electron excitation. These processes are thought to be nanoparticle size dependent but also to vary quite significantly with the projectile ion energy. At CIMAP, to quantify these processes, the recent IMAGERI ANR project aims to measure absolute cross-sections of electron emission from isolated metallic nanoparticles, and compare the results from different GANIL beamlines.

Perspectives and needs

These last years, due to the lack of available beam time at GANIL, allocated beam time by the iPAC has drastically dropped compared to the increasing of the requested beam time***.*** This is a problem for keeping the interest of external users involved in our international collaborations, and regarding the amount of data that our PhD students can obtain in three years. Besides, although only scarcely used for the moment in our field, ion beams delivered by the SME and HE beamlines (around 10 and 100 MeV/u, respectively) are also of interest, because they corresponds to the energy of ions when they interact with healthy tissues in hadron therapy. Within the IMAGERI ANR project, measurement of absolute cross-section of electron emission from biomolecular building blocks and nanoparticles will be performed in a near future on the SME beamline. Moreover, our goal is to extend the work done at the IRRSUD beamline on proteins and DNA to these higher energy ranges. Therefore, a first need for our community is more beam time, especially at IRRSUD, a factor of two or three compared to what was scheduled recently. Second, carbon ion beams at different charge states are required to investigate the effect of this parameter on the processes induced, especially at the IRRSUD beamline, since the charge state decreases quickly with penetration depth in tissues around the Bragg peak.
One of the most active areas of research in hadron therapy nowadays is the investigation of the Flash effect. This is a very promising way because it has been shown that electrons and photons irradiation at very high dose rates (on the order of 100Gy/s) but usual doses induce less damage to healthy tissues than conventional treatment modalities, while being as effective on cancer cells. The first attempts to look for such a Flash effect for protons have been made only very recently, notably at the Institut Curie (Orsay, France). GANIL should be one of the leading facilities regarding the quest for a Flash effect for carbon ions, due to its

high potential in sparing healthy tissues while killing tumor cells. This would imply a technical update of the existing cyclotrons to deliver such high dose rates in very short and intense ion pulses. It would be highly interesting for radiobiologists, but also for our community, because of the possibility of time-resolved pump-probe experiments with ions and/or lasers, yet to be performed. Such experiments could shed light on dynamics of ion-induced denaturation of proteins and DNA, a totally unexplored field.