

# Heavy Nuclei @ Dubna, JYFL and RIKEN

## Executive Summary

One of the major challenges of modern Nuclear Physics is to investigate the limits of nuclear existence. A simple and yet fundamental question is: what is the maximum number of protons and neutrons a nucleus can sustain? To answer such a question, the synthesis of new elements with an ever increasing number of protons ( $Z=114$  to  $122$ ) is carried out and motivated by the theoretical prediction of a new island of stability. Super heavy nuclei also provide a unique laboratory within which nuclear structure and dynamics under very intense Coulomb forces can be studied. While cross-sections to synthesize the heaviest elements are extremely low, the production rate of super heavy nuclei with proton numbers ranging from  $Z=100$  to  $Z=110$  is high enough to obtain significant information on their nuclear structure via spectroscopic studies.

Over the last 20 years, physicists at CSNSM and IPHC have developed research programs and solid collaborations to carry out studies of super heavy nuclei at leading facilities in the world. This is the case of synthesis reaction and spectroscopy studies at RIKEN (since 2014), decay spectroscopy studies at Dubna (since 2004) and prompt spectroscopy studies at JYFL (since 1999). These projects (milestones, budget & HR, achievements, perspectives & impact on other projects) will be detailed in the following sections.

## 1 Introduction

The synthesis and study of new elements and new isotopes is one of the most challenging prospects in nuclear structure research. However, searching for the limits of the Table of Mendeleev and probing the extension of the nuclear chart are not only a scientific challenge, they are also an experimental one. Indeed, the last discovered elements ( $Z=114-118$ ) were produced in fusion-evaporation reactions with  $^{48}\text{Ca}$  beams and actinide targets, with extremely small production cross-sections and an identification of only a handful of events in irradiations spanning many months<sup>1</sup>. Despite the low number of detected events, the decay modes, half-lives and alpha-decay energies of the newly discovered isotopes are a stringent test for nuclear models, while the measured production cross sections and excitation functions put reaction theories to the test.

Due to the very low reaction cross-sections these experiments represent the current observational limits. For example, only four elements of Oganesson ( $Z=118$ ) were observed with the DGFRS at Dubna<sup>2</sup> with a production cross section around  $300-550 \text{ fb}^3$ . Moreover, the direct discovery of Nihonium ( $Z=113$ ) was even more challenging. It required the equivalent of three full years of beam on target (over a period of nine years) to observe three  $^{278}\text{Nh}$  decays<sup>4</sup> ( $22 \text{ fb}$  reaction cross-section). Therefore the discovery of new elements requires a significant increase of beam intensity in order to achieve such goals with "only several months of beam". This triggered construction of new machines (the SHE-Factory at Dubna, the cw-LINAC at GSI and the LINAC for SPIRAL2) or upgrades of existing ones (new-RILAC at RIKEN). With these machines, beam intensities between 5 and 10  $\mu\text{A}$  are expected on target. Subsequently the development of new target backing materials is needed since current targets will not be able to withstand such beam intensities. Moreover, since Californium is the heaviest actinide material available in sufficient quantity to make targets, it is imperative to develop heavier beams such as  $^{50}\text{Ti}$ ,  $^{51}\text{V}$  and  $^{54}\text{Cr}$  to synthesize elements beyond  $Z=118$ . As it will be detailed later, developments of isotopic Metal Ions from Volatile Compounds (MIVOC) of these elements by the IPHC was a significant step for the availability of these beams for the synthesis of new elements.

Spectroscopic studies beyond Es ( $Z=99$ ) have made great progress in recent years due to the use of efficient detector arrays around the target position (prompt spectroscopy) and at the focal plane of

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<sup>1</sup> R.C. Barber et al., Pure Appl. Chem., 83 (2011) 1485, P.J. Karol et al., Pure Appl. Chem. **88** (2016) 139

<sup>2</sup> Y.T. Oganessian et al., Nucl. Phys. A734 (2004) 109

<sup>3</sup> Y.T. Oganessian et al., Phys. Rev. C 74 (2006) 044602

<sup>4</sup> K. Morita et al., Journal of the Physical Society of Japan 73 (2004) 2593

recoil separators (decay spectroscopy), as illustrated in figure 1. These have revealed that super heavy nuclei are very robust with respect to rotation, reaching spins of more than 20 hbar and revealing interesting alignment properties. Some of the most hindered decays in the whole of the nuclear chart have been observed from high-K isomers in the region around  $^{254}\text{No}$ .

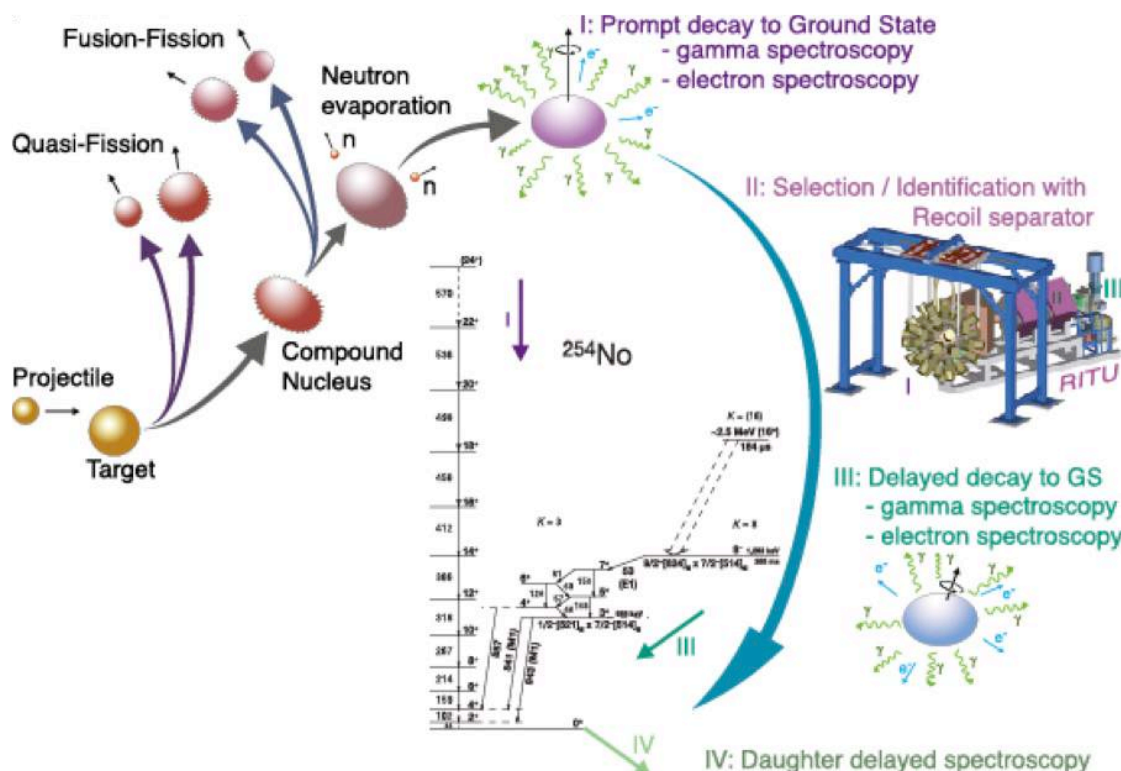


Figure 1: Illustration of the reaction process leading to an evaporation residue and the 2 possible spectroscopic studies: prompt spectroscopy around the target position and decay spectroscopy at the focal plane of a recoil separator. The observed excited states are shown in the case of  $^{254}\text{No}$  (taken from B. Gall & P.T. Greenlees, Nucl. Phys. News 23 #3 2013 27-31.).

Fine structure alpha decay has allowed trends in low-energy structures to be established as a function of N and Z. From this body of data, there exists now a rather clear disagreement between the predictions of single-particle sequences and gaps obtained from all existing effective interactions/energy density functionals and experiment. The data, however, remain scarce and clustered to nuclei reached by fusion evaporation reactions with Pb and Bi targets with large cross sections or transfer reactions on long-lived actinide targets, as shown in figure 2.

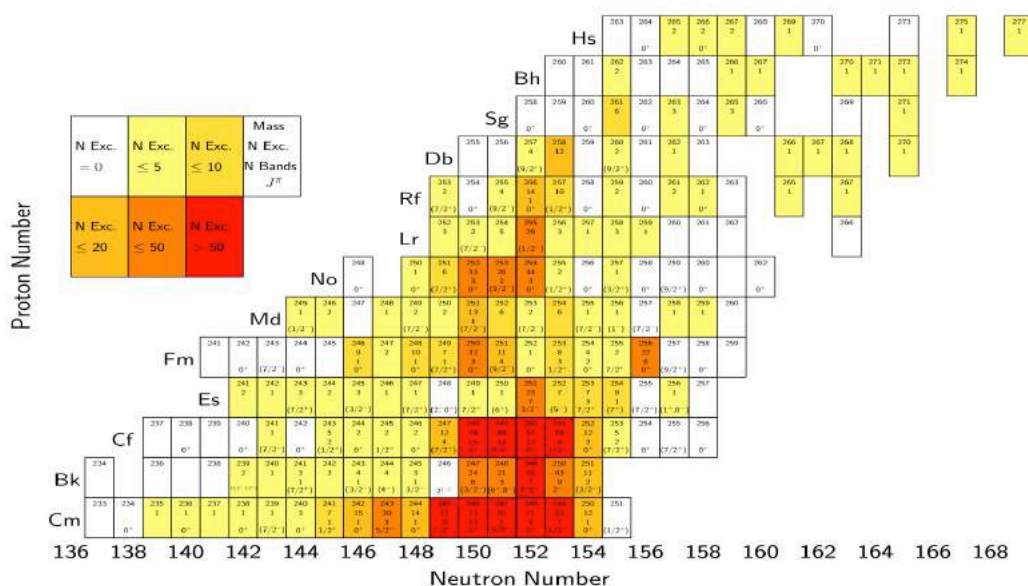


Figure 2: Summary of the experimental data available on nuclei from Cm to Db (taken from Ch. Theisen et al., Nuclear Physics A944 (2015) 33). The colour code gives the impression of the level of knowledge for a particular isotope.

## 2 State of the Art

The heaviest elements were first synthesized and observed at the focal plane of the DGFRS in Dubna ( $Z=114-118$ ). The production of Fl, Ms, Lv, Ts was confirmed in Berkeley and/or GSI. Hot fusion reactions of the very neutron-rich doubly magic  $^{48}\text{Ca}$  projectile with the heaviest actinides targets available ( $^{244}\text{Pu}$ ,  $^{243}\text{Am}$ ,  $^{248}\text{Cm}$ ,  $^{249}\text{Bk}$  and  $^{249-251}\text{Cf}$ ) were used. The direct production of element 113 was carried out at RIKEN using cold fusion and the recoil separator GARIS I associated to the RILAC linear accelerator. Synthesis experiments are now only performed at RIKEN using the separator GARIS II associated to the RRC Cyclotron. In the coming months two new machines should start campaigns devoted to synthesis studies: the newly inaugurated Super Heavy Element Factory (SHE-F) with the DC280 cyclotron associated to a new gas-filled separator in Dubna and the upgraded new-RILAC coupled to the new GARIS II gas filled separator. The SHE synthesis program at GSI awaits the new cw-LINAC. The program at Berkeley is mainly devoted to mass measurements with FIONA<sup>5</sup> while the chemical properties of the heaviest nuclei ( $Z=113$  and  $114$ ) are now the main focus at TASCA<sup>6</sup> at GSI and of the chemistry team in Dubna.

Decay spectroscopy of heavy elements is carried out only in a few places around the world (see figure 3): at the FLNR Dubna laboratory (Russia), at the University of Jyväskylä (Finland), at GSI (Germany), at JAEA (Japan), at Argonne National Laboratory (USA), in GANIL (France), in Lanzhou (China) and at the Lawrence Berkeley National Laboratory (USA). Each facility has its specificities (especially in terms of beam characteristics, availability of targets, recoil separation techniques, and available spectroscopic observables) but there is of course some overlap and the competition is strong. This is a healthy situation as it forces the physicists and engineers to improve their experimental setup in every way they can. Moreover, it is a guarantee that results are crosschecked, which is essential in this field of very rare events. The IN2P3 is strongly involved in the spectroscopy programme at Dubna with 1 to 4 months of beam time per year, as it will be detailed later.

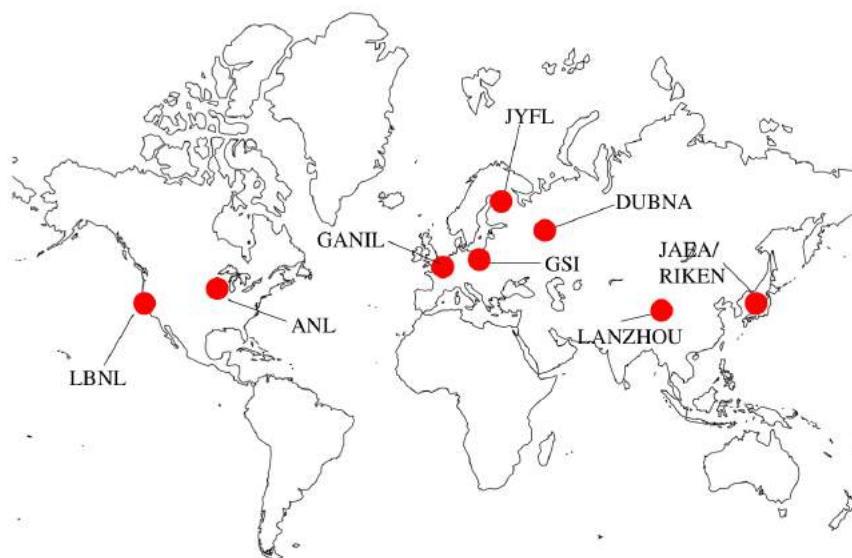


Figure 1: Map indicating facilities providing beam-time to perform spectroscopy of very heavy elements

The most recent decay spectroscopy study of heavy elements in GANIL was the study of  $^{257}\text{Db}$  performed at LISE. Since such experiments need long dedicated beam time, the studies of heavy elements at GANIL will resume when the Super Separator Spectrometer ( $S^3$ ) and SIRIUS detection system come online at the SPIRAL2 facility (at the earliest 2022/2023).

In Jyväskylä, numerous decay spectroscopy studies were performed using the RITU<sup>7</sup> gas filled spectrometer combined with the GREAT<sup>8</sup> focal plane detection system. Some of these experiments were performed with the SACRED<sup>9</sup> electron detector at the target position. Nowadays the beam intensities

<sup>5</sup> J.M. Gates et al., Phys. Rev. Lett. 121 (2018) 222501

<sup>6</sup> A. Semchenkov et al., Nucl. Instrum. Methods B 266 (2008) 415361

<sup>7</sup> M. Leino et al., Nucl. Instrum. Methods B 99 (1995) 653

<sup>8</sup> R.D. Page, et al., Nucl. Instrum. Methods Phys. Res. B 204 (2003) 634

<sup>9</sup> P.A. Butler et al., Nucl. Instrum. Methods Phys. Res. A 381 (1996) 433

available do not make it competitive for decay spectroscopy beyond Rf. This limitation should be overcome with the new HIISI 18 GHz ECR ion source.

At GSI, the spectroscopy program at SHIP has been replaced by a program of measurements of ground-state properties (masses, moments, spins...). Decay spectroscopy at GSI is now only performed at the focal plane of TASCA using the upgraded TASIPEC array<sup>10</sup>, with enhanced sensitivity to X-rays. In Berkeley, decay spectroscopy is now carried out at the focal plane of FIONA – which limits the species that can be studied to those with tens-of-millisecond lifetimes.

Prompt spectroscopy of heavy and super heavy nuclei is only carried out in 2 places worldwide; at JYFL with the JUROGAM<sup>11</sup> and SAGE<sup>12</sup> arrays and at ANL with GAMMASPHERE<sup>13</sup>. Only JYFL offers the possibility to measure prompt conversion electrons and photons in coincidence using SAGE. The IN2P3 was strongly involved in the JYFL spectroscopy programme, as it will be detailed later. GAMMASPHERE, on the other hand, is the only device that allows calorimetric measurements (i.e. one can remove the heavy metal absorbers in front of the BGO Compton suppression shields in order to use the BGO shields as active detectors and increase the total efficiency).

## 3 Research programmes

### 3.1 Synthesis of new elements

The synthesis of Oganesson was achieved by means of the  $^{249}\text{Cf}(^{48}\text{Ca},3n)^{294}\text{Og}$  fusion evaporation reaction. It corresponds to a limit in terms of target material and beam. Intense beam of  $^{48}\text{Ca}$  has enabled the synthesis of the heaviest elements using hot fusion evaporation reaction with heavy actinide targets. But, since the Californium is the heaviest element for which one can produce enough material for rotating targets suitable for beams at the 1  $\mu\text{A}$  and above intensity level, heavier beams are needed to produce new elements. To pursue these studies further with synthesis of elements 119-122, the availability of intense beams of  $^{50}\text{Ti}$ ,  $^{51}\text{V}$  and  $^{54}\text{Cr}$  as well as long irradiation times are obligatory.

Following the success of the  $^{50}\text{Ti}$  beam in JYFL, a collaboration started naturally with Dubna and later with RIKEN where this beam could also give outstanding results. Today, long beam times are available at RIKEN and will be available soon at the SHE-Factory in Dubna. The isotopically enriched chemical compound required to produce intense beams of  $^{50}\text{Ti}$  at both facilities is provided by the IPHC. As detailed in the section 6 of this document,  $^{51}\text{V}$  and  $^{54}\text{Cr}$  MIVOC compounds were also developed at the IPHC.

Since IPHC brought its first MIVOC compound to Dubna in 2012 many experiments were carried out with SHELS using  $^{50}\text{Ti}$ . Significant quantities of  $^{50}\text{Ti}$  and  $^{54}\text{Cr}$  were prepared for Dubna at the IPHC within this collaboration. Unfortunately, no synthesis experiments were run with Dubna gas-filled recoil separator (DGFRS) using  $^{50}\text{Ti}$  MIVOC compound from the IPHC even if it was several times envisaged. Now, the Synthesis program using  $^{50}\text{Ti}$ ,  $^{51}\text{V}$  and  $^{54}\text{Cr}$  will soon start at the SHE-Factory.

### **Milestones**

- First MIVOC beam tests with ECR ion source of Dubna (May 2012)
- First MIVOC beam tests with RILAC (July 2014)
- Study of SHE barrier distributions with GARIS I (2016-17)
- $^{295}\text{Og}$  experiment started in summer 2016 with RILAC and GARIS II (paused)
- E119 experiment started in Jan 2018 with RRC and GARIS II (ongoing)
- DC280 in Dubna and new RILAC in RIKEN should enter in operation S1 2020

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<sup>10</sup> L.L. Andersson et al., Nucl. Instr. Meth.A 622 (2010) 164

<sup>11</sup> S. Eeckhaudt et al., Eur. Phys. J. A 26 (2005) 227 and PhD thesis of Jyväskylä University

<sup>12</sup> J. Pakarinen et al., Eur. Phys. J. A50 (2014) 53

<sup>13</sup> I-Y Lee Nucl. Phys. A 520 (1990) c641

## RH & Budget

For the moment the synthesis program of new elements in Dubna has not implicated IN2P3 members. It will soon be the case when the SHE factory will enter in operation.

For the program in RIKEN, the origin of funds on the French side are two fold:

- AP IN2P3
- International associated lab (LIA)

In addition some of the travel expenses and subsistence were covered by RIKEN. One of us was awarded by JSPS a 20 days travel grant in 2019.

Considering human resources on the synthesis program, we had up to 4 months of beam time per year. We try to attend the experiments as much as reasonably possible. From IN2P3, mainly 4 permanent staff are presently involved in this heavy element synthesis project Z. Asfari, O. Dorvaux, M. Filliger and B. Gall. Note that two of them (Z.A. & M.F.) contribute here to the experiment through production of isotopic compounds for experiments. This is counted separately to R&D program detailed later. In addition, 3 PhD students of Strasbourg were involved in these programs (H. Faure, P. Brionnet and K. Kessaci).

This scientific case will be extended in short future when the SHE-Factory will start in Dubna where the  $^{50}\text{Ti}$  and  $^{54}\text{Cr}$  beams will be widely used for quest of elements.

## Achievements

As already stated, availability of intense beams of  $^{50}\text{Ti}$ ,  $^{51}\text{V}$  and  $^{54}\text{Cr}$  is one of the key points in the quest of the next new elements with proton number greater than 118. As detailed in a further section intense R&D programme was carried at IPHC Strasbourg in the framework of SHE synthesis collaborations. Two different vanadium MIVOC compounds were successfully tested in RIKEN in 2017. A chromocene compound was developed at the IPHC in 2018 and successfully tested in Dubna the same year. A highly enriched  $^{54}\text{Cr}$  compound was then used in 2019 in Dubna for a study of fission dynamics of element 120 ( $^{54}\text{Cr} + ^{248}\text{Cm}$ ).

Prior to a series of experiments dedicated to new element search, a campaign of barrier distribution measurements was performed in order to establish a model independent method to determine the optimal bombardment energy for synthesis. This study was performed using  $^{48}\text{Ca}$  and  $^{50}\text{Ti}$  beams on  $^{208}\text{Pb}$  and  $^{248}\text{Cm}$  targets leading to already known heavy elements. Results of the barrier distribution were then compared to known experimental neutron evaporation residue cross sections. This result was published in JSPS journal and selected as choice of the editor<sup>14</sup>. The method has since been applied to several reactions of interest in the quest for new elements (unpublished). On this basis we selected the bombarding energy for the synthesis of Oganesson with a titanium beam. In 2016, the synthesis of  $^{295}\text{Og}$  was started in RIKEN with at  $^{50}\text{Ti}$  beam provided by RILAC II impinging on a  $^{248}\text{Cm}$  target in front of GARIS II.

Due to the start of the upgrade of RILAC II to the new-RILAC the continuation of this campaign was postponed. The gas-filled separator GARIS II was moved to the beam lines of the RRC cyclotron S2 2017. After only a few months, we could commission GARIS II at this new position in December 2017. This enabled the start of the quest for element 119 with a  $^{51}\text{V}$  beam and a  $^{248}\text{Cm}$  target in front of GARIS II. Up to now, several months of beam time have been dedicated to this experiment, in which IPHC Strasbourg participates actively. The IPHC has developed dedicated analysis codes, which are used in parallel to the Japanese ones for independent analysis.

Using high intensity beam, this experiment also triggered high-temperature target tests where IPHC members could bring their expertise. This study is ongoing and confidential since it may lead to patents.

## Perspectives

The new accelerators DC280 at the Dubna SHE-Factory and new-RILAC at RIKEN should enter in action soon opening new opportunities of very intense beams. There will be some competition between these two sites for new elements discovery. Nevertheless, as it is the case for our American colleagues

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<sup>14</sup> T. Tanaka et al., Journal of the Physical Society of Japan 87, 014201 (2018)

who supply target material for both collaborations, we provide the beam material for the best scientific use of both these collaborations.

The perspective of discovery of new elements in the coming years is unique. Providing the beam and analysing the data, IN2P3 will bring a significant contribution to these outstanding experiments. These machines will also bring us some clues about the real nature of the chemistry at the limits of Mendeleev's table.

This synthesis programme is complementary to the programme foreseen in France with S<sup>3</sup> and will bring both strong expertise in high intensity facilities and student training for human resources for the Future of CNRS.

## SWOT

<p style="text-align: center;"><b>Strengths</b></p> <ul style="list-style-type: none"> <li>- high intensity metallic beams</li> <li>- beam time available on several sites</li> <li>- availability of a variety of targets</li> <li>- visibility</li> <li>- experience</li> <li>- collaboration built on complementary expertises</li> </ul>	<p style="text-align: center;"><b>Weaknesses</b></p> <ul style="list-style-type: none"> <li>- Uncertainty on the real intensity of beams with the new machines (SHE-Factory and new RILAC) so long no tests are performed.</li> <li>- Limited funds for travel</li> <li>- development of targets compatible with High beam intensity</li> </ul>
<p style="text-align: center;"><b>Opportunities</b></p> <ul style="list-style-type: none"> <li>- Start of SHE-Factory with new gas filled</li> <li>- Start of new RILAC + GARIS III</li> <li>- discovery potential</li> </ul>	<p style="text-align: center;"><b>Threats</b></p> <ul style="list-style-type: none"> <li>- Limited manpower</li> <li>- Reduced funding for missions</li> </ul>

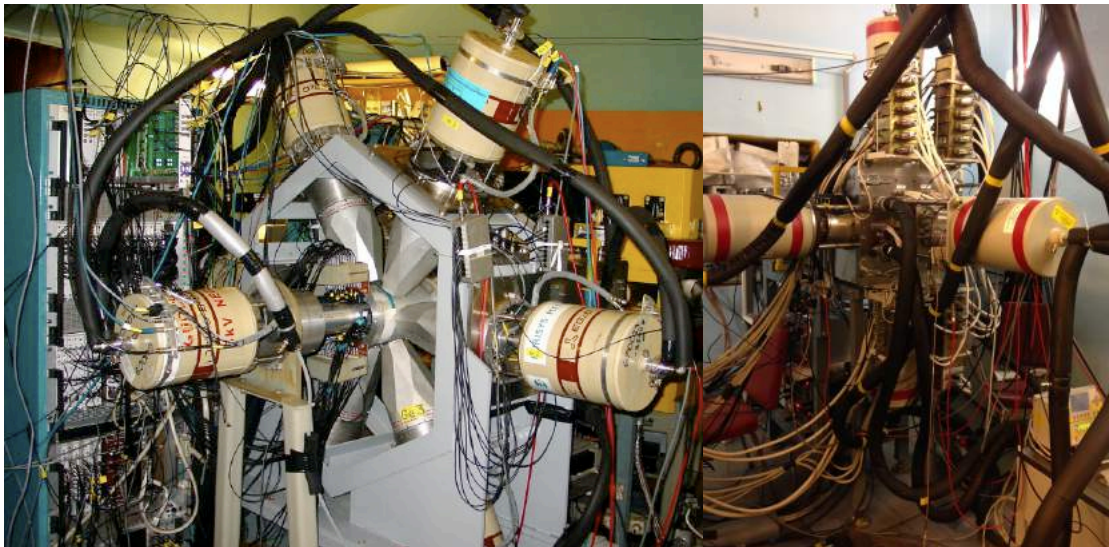
### 3.2 Decay Spectroscopy (GABRIELA)

To benefit from the beam time, the intense stable beams and the radioactive actinide targets uniquely available at the FLNR, an IN2P3-JINR (CSNSM - IPHC - FLNR) collaboration launched a project of electron and gamma-ray spectroscopic studies of heavy nuclei at the FLNR in 2003 (<https://www.csnsm.in2p3.fr/GABRIELA>).

At first, the plan was to perform prompt spectroscopy as well as decay spectroscopy of heavy elements, but the former project was abandoned after measuring the background conditions at the target position. A compact and efficient detector array was then jointly designed to be able to identify the fusion-evaporation residues at the focal plane of the recoil separator VASSILISSA and detect their subsequent radioactive decays involving the emission of alpha and beta particles, fission fragments, gamma and X rays and internal conversion electrons (ICE's).

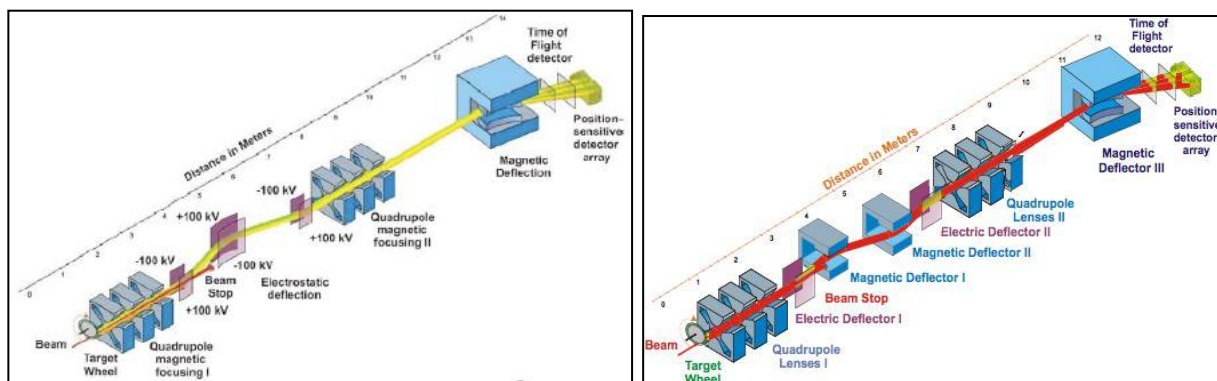
In the early spring of 2004, the name GABRIELA (**G**amma **A**lpha **B**eta **R**ecoil **I**nvestigations with the **E**lectromagnetic **A**nalyser) was imagined and by the summer of 2004, **GABRIELA** had been assembled, commissioned and the detector array fully characterized<sup>15</sup>. The first one month long experimental campaign occurred in September 2004. In figure 4 a photo of the original GABRIELA set-up (seven 70% Ge detectors from the France-UK loan pool) is shown beside the current set-up with the CLODETTE clover and 4 large volume Ge detectors.

<sup>15</sup> K. Hauschild et al. Nucl. Instr. Meth. A 560 (2006) 388



**Figure 4: Photography of the Compton-Suppressed Germanium-detector array GABRIELA (in 2004 on the left and 2016 on the right).**

The poor transmission of VASSILISSA, with which we had been battling since the beginning of the GABRIELA project, lead us to apply for funds to modernize the separator. A grant obtained in 2006 from the French funding agency ANR (ANR-06-BLAN-0034 project SHELS) and funds from JINR provided the necessary financial resources to upgrade the VASSILISSA separator to SHELS, which was commissioned in 2013<sup>16</sup> and highlighted in a Lettre électronique IN2P3<sup>17</sup>. A five-fold increase in the transmission of evaporation residues (ER) produced in asymmetric reactions with light beams has been measured. With the installation of a new large bore quadrupole lens at the entrance of SHELS in 2020 a further increase in transmission is expected.



**Figure 5: Schematic drawing of the VASSILISSA separator (left) and the upgrade to SHELS (right)**

In a second round of funding (ANR-12-BS05-0013 project CLODETTE) the focal plane detection system, which had been upgraded in 2006 and 2009, was vastly improved. State of the art germanium (Ge) detectors were bought from MIRION (ex-CANBERRA) increasing the photon detection efficiency by almost a factor of two<sup>18</sup>. Dedicated BGO shields, designed within the collaboration, were also bought. These shields improve the signal to background compared to a bare Ge detector and are indispensable when measuring rare gamma-ray quanta. A new vacuum chamber was also built specifically for the revised GABRIELA detection system, with special thin aluminium inserts to position the Ge detectors as close as possible to the source of radiation and to maximise detection efficiency for low energy photons. New digital electronics was also purchased to improve the timing resolution from ms to sub-ns. This allows to run an essentially zero dead time system granting the collaboration access to isomeric states with half-lives below 10 ms. While having excellent energy resolution for Si detectors (15-18 keV FWHM at 5499 keV and a full-scale of  $\sim 250$  MeV)<sup>19</sup> our current preamplifiers from TechInvest do not

<sup>16</sup> A. Yerebin, Physics of Particles and Nuclei Letters 12 (2015) 35, K. Rezykina, Acta Physica Polonica B46 (2015) 623

<sup>17</sup> Lettre électronique IN2P3 N° 135, mai 2013 “Premier faisceau de particules alpha pour le nouveau séparateur d’ions de recul Shels”

<sup>18</sup> K. Hauschild et al., in preparation

<sup>19</sup> A.V. Isaev, et al., Instruments and Experimental Techniques 54 (2011) 37

have a fast enough rise-time to achieve sub-ns timing. Therefore new preamplifiers are necessary in order to benefit from this digital revolution. The design and tests of new preamplifiers is currently on going. This new back end electronics will also allow for particle discrimination through Pulse Shape Analysis (PSA) techniques as was recently proven possible in an electronics test at GABRIELA. Using SHELS we transported  $^{233}\text{U}$  alpha particles from the target position to GABRIELA thus ensuring an electron free source of energy degraded alphas with energies overlapping the Internal Conversion Electrons (ICE) from a standard  $^{133}\text{Ba}$  source. In Figure 6(b) it can be clearly seen that ICE and low energy alpha particles can be distinguished from one another purely on the basis of the preamplifier rise-time. To our knowledge this is the first demonstration of ICE/ $\alpha$  discrimination at an energy range appropriate to decay spectroscopy. In principal this method could also be used for SIRIUS if the algorithm can be implemented within the limited FPGA resources of the NUMEXO2 digitiser. In addition, the discrete component charge sensitive preamplifiers developed for GABRIELA are pin compatible with those of SIRIUS. Their improved energy resolution (FWHM <6 keV at 320 keV and <12.5 keV at 5805 keV) and rise-time can therefore benefit the programme carried out at GANIL.

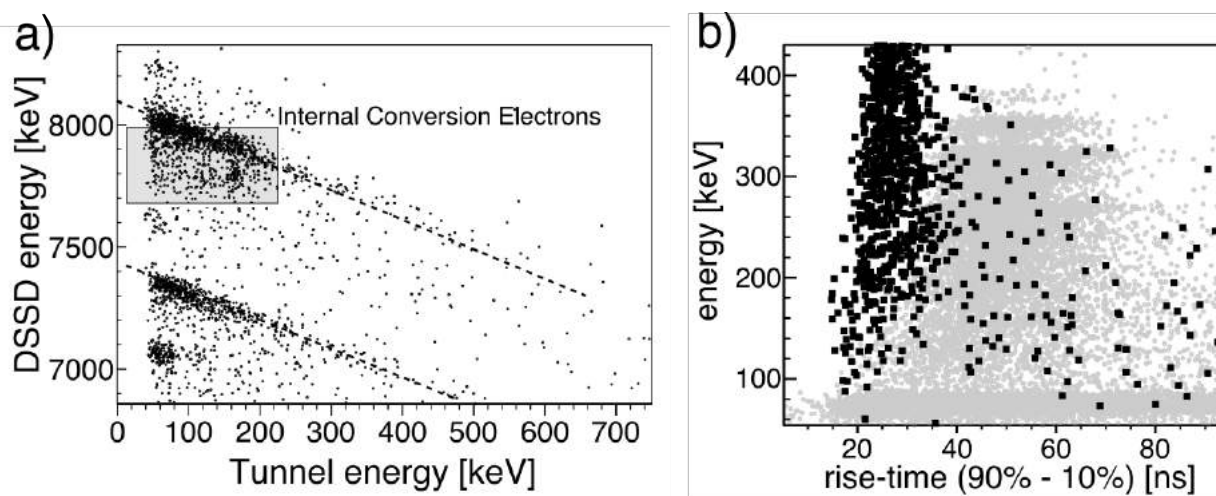


Figure 6: (a) Prompt coincident DSSD-tunnel energy matrix following the implantation of evaporation residues of the reaction  $^{208}\text{Pb}(^{48}\text{Ca}, xn)$ . Dashed lines indicate alpha particles that escape the DSSD and are detected in the tunnel. ICE in  $^{251}\text{Fm}$  from the alpha decay of  $^{255}\text{No}$  are indicated by the shaded box. (b) Particle energy as a function of preamplifier signal rise-time. Black squares:  $^{233}\text{U}$  alphas degraded in energy. Grey circles: conversion electrons from the decay of  $^{133}\text{Ba}$ .

## Milestones

- Approval by the Scientific Council of IN2P3 (December, 2003)
- Approval by the Scientific Council of JINR (January 2004)
- 2004: Start of the GABRIELA collaboration and IN2P3-JINR collaboration 04-63
- Construction of the GABRIELA detector array
- Scientific campaigns: 2004, 2005, 2006, 2008 and 2009
- ANR SHELS (2006-2011) & MoU on the Upgrade and exploitation of VASSILISSA (2008)
- Submission of the ANR DeSpeTran (2011)
- Commissioning of SHELS (2013-2015)
- ANR CLODETTE (2012-2017) & MoU on the Exploitation of VASSILISSA (2012)
- Commissioning of CLODETTE detectors (2016)
- Submission of the ANR PIONNER (2016)
- Scientific campaigns: 2016, 2017
- Submission of the ANR SHELDON (2017)
- Upgrade of GABRIELA electronics (on-going)
- Upgrade of SHELS (deflector plates: 2017-2018, entrance quadrupole: 2019-2020)
- Submission of SHEXI-Sensors, SHEXI-ASICs, SHEXI-BEE (ATTRACT 2018)
- Scientific campaigns: 2018, 2019



## Prizes & communication

- the JINR Instruments and Methods 1<sup>st</sup> prize for the construction of SHELS (2015)
- the JINR Instruments and Methods 2<sup>nd</sup> prize for GABRIELA (2007)
- In 2017, GABRIELA was mentioned in an article of Le Monde:  
 « Chez les chasseurs russes des nouveaux atomes », Le Monde, 12 juillet 2017

## Budget & Human resources

The origin of funds on the French side are many fold (see figure 7a):

- AP IN2P3
- Projet de Recherche Conjoint (PRC) n° 1944
- Laboratory private funds
- ANRs (SHELS – CLODETTE)
- IN2P3-JINR collaboration 04-63 (large part of necessary per diem and accommodation)

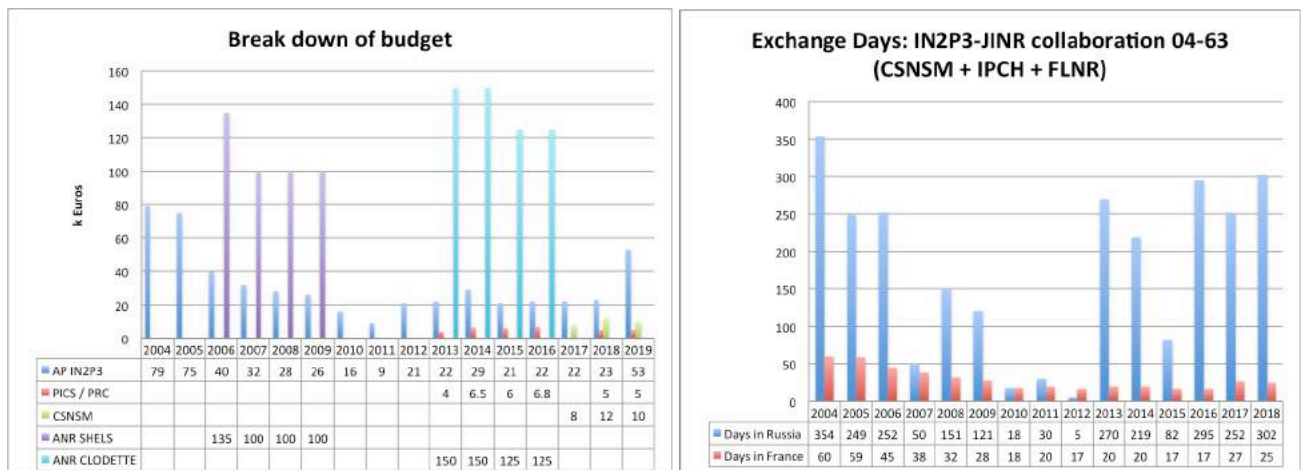


Figure 7: (a) French funds obtained for the decay spectroscopy program in Dubna since the start of the GABRIELA project. (b) Days spent in Russia and France up to now by Russian/French physicists and engineers.

On the Russian side, the project has benefited from many grants from the Russian Foundation for Basic Research. Furthermore, all running costs are paid by FLNR.

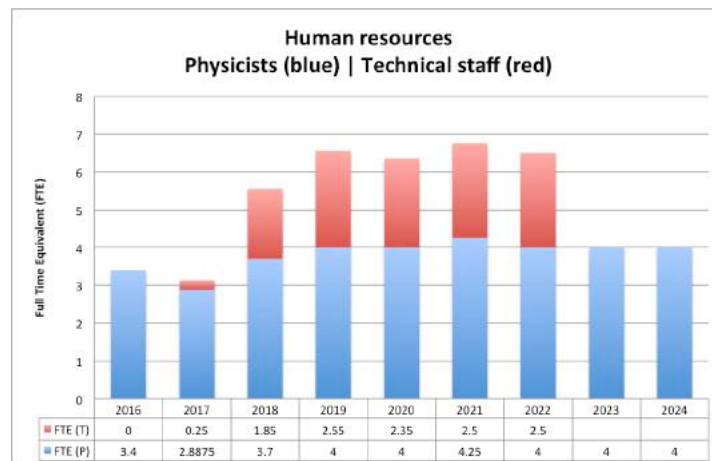
The setting up and carrying out of tests and commissioning runs and taking part in the long experimental campaigns (1-3 campaigns of 4-6 weeks/year) require considerable travel on the part of French scientists. The days spent in Dubna by IN2P3 personnel is shown in figure 7b together with the days spent by our Russian colleagues in France.

The beginning of the project saw the involvement of IN2P3 engineers for the design and construction of electronics for Ge detectors. This led to the development of TNT2-D cards<sup>20</sup>. While these cards have only been used for numerous electronics tests at Dubna they were successfully exploited in experimental campaigns at JYFL (see section 3.3) and form the basis of the SIRIUS electronics and detector tests at the CSNSM and IPHC. The current upgrade of both the front- and back-end (digital) electronics of GABRIELA also sees a renewed involvement of IN2P3 engineers.

Since the upgrade of SHELS (2013), the collaboration has been able to obtain PhD grants for 4 PhD students: K. Rezynkina (Université Paris Saclay, 2016), P. Brionnet (Université de Strasbourg, 2017), R. Chakma (Université Paris Saclay, 2020), K. Kessaci (Université de Strasbourg, 2021).

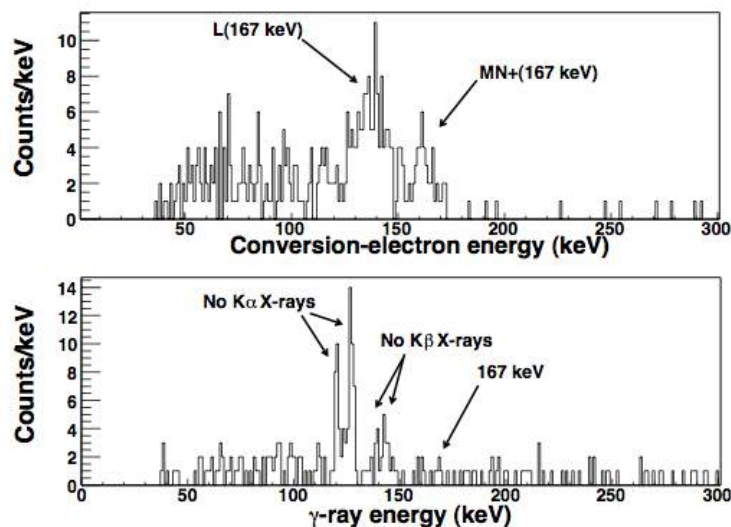
<sup>20</sup> L. Arnold et al., IEEE Trans. Nucl. Sci. 53 (2006) 723

The following graph shows the manpower (in full time equivalent) involved in the project since 2016 and foreseen up to 2024.



## Achievements

In the fall of 2004, the first very heavy nuclei were studied with GABRIELA using  $^{48}\text{Ca}$ -induced fusion-evaporation reactions on  $^{208}\text{Pb}$  and  $^{209}\text{Bi}$  targets. A new excited state was observed in  $^{249}\text{Fm}$  and internal conversion coefficients extracted for all the observed transitions<sup>21</sup>. A long-lived isomer in  $^{255}\text{Lr}$  was observed for the first time via decay gamma and ICE spectroscopy<sup>22</sup>. In  $^{253}\text{No}$ , the nature of the low-lying  $31 \mu\text{s}$  isomer<sup>23</sup> was firmly established through combined gamma and ICE spectroscopy. The presence of a high-K isomer could also be inferred<sup>24</sup>.



**Figure 9: Spectrum of ICE's emitted by the  $31 \mu\text{s}$  isomer of  $^{253}\text{No}$  populated directly in the reaction  $^{207}\text{Pb}(^{48}\text{Ca},2n)^{253}\text{No}$ . Bottom : Corresponding gamma-ray spectrum.**

In 2009, with an upgraded GABRIELA array, the decay of the high-K isomer in  $^{253}\text{No}$  was re-investigated (see figure 10) and a tentative decay scheme was established<sup>25</sup>.

The real breakthrough, in terms of production and detection of super heavy nuclei, came with SHELS and the commissioning of the new focal plane detection system in 2016. In one of the commissioning runs, the decay of the  $5/2^+$  isomer in  $^{251}\text{Fm}$  could be studied. A new technique to extract

<sup>21</sup> A. Lopez-Martens et al., Phys. Rev. C 74 (2006) 044303

<sup>22</sup> K. Hauschild et al., Phys. Rev. C 78 (2008) 021302(R)

<sup>23</sup> C.E. Bemis et al., Phys. Rev. Lett. 31, 6

<sup>24</sup> A. Lopez-Martens et al., Eur. Phys. J. A 32 (2007) 245

<sup>25</sup> A. Lopez-Martens et al., Nuclear Physics A852 (2011) 15, T. Wiborg-Hagen et al., Acta Physica Polonica B42 (2011) 605

mixing ratios was devised<sup>26</sup> and the collectivity of the transition de-exciting the isomer was measured<sup>27</sup>, thereby confirming the vibrational character of the metastable state.

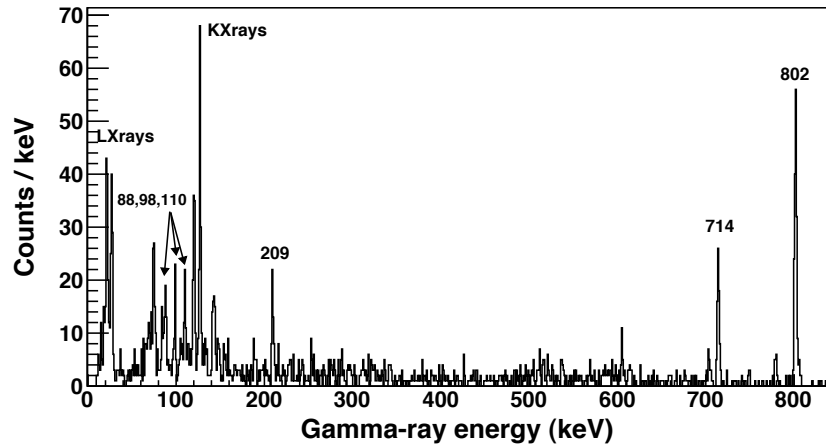


Figure 10: Energy spectrum of the gamma rays emitted in the decay of the high-K isomer in <sup>253</sup>No

The properties of <sup>257</sup>Db and <sup>255</sup>Rf were investigated<sup>28</sup> in 2016 and 2017 using an intense <sup>50</sup>Ti developed by the IPHC (see section 3.4) and <sup>209</sup>Bi and <sup>207</sup>Pb targets. In the experiment with Bi targets, evidence for pxn evaporation channels was observed<sup>29</sup>.

In 2018, the isotopes <sup>256,257</sup>Rf were studied. Thanks to an enhanced efficiency at low energy, the decay cascade from the lowest isomer in <sup>256</sup>Rf<sup>30</sup> could be determined, pinning down its excitation energy, spin and parity<sup>31</sup>. The analysis of the fine structure decay from the ground state and first excited state of <sup>257</sup>Rf has allowed the excitation energy of the 11/2<sup>-</sup> state in <sup>257</sup>Rf but also in <sup>253</sup>No to be established, providing information on the splitting of the j<sub>15/2</sub> shell from which the 9/2<sup>-</sup>[734] ground state and 11/2<sup>-</sup>[725] level emanate<sup>32</sup>.

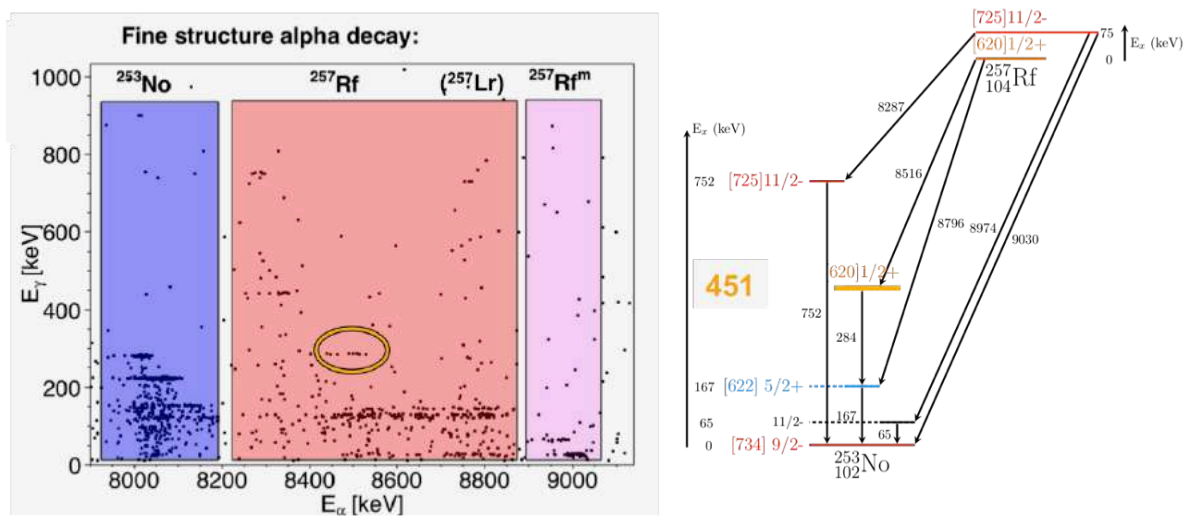


Figure 11: Matrix of the gamma rays de-exciting states in <sup>253</sup>No vs the coincident alpha-decay energies of <sup>257</sup>Rf, and corresponding established decay scheme of <sup>257</sup>Rf.

In 2019, the first successful experiment using a light <sup>22</sup>Ne beam and an actinide target (<sup>238</sup>U) was carried out. The alpha decay of <sup>256</sup>No was observed. The decay of short-lived isomeric states in <sup>255</sup>No and <sup>256</sup>No could also be observed for the first time<sup>33</sup>.

<sup>26</sup> K. Rezyunkina et al., Nucl. Instr. Meth. A 844 (2017) 96 and PhD, Université Paris Saclay, 2016

<sup>27</sup> K. Rezyunkina et al., Phys. Rev. C 97 (2018) 054332 and PhD, Université Paris Saclay, 2016

<sup>28</sup> P. Brionnet, PhD (Université de Strasbourg, 2017), R. Chakma, PhD (Université Paris Saclay, 2020)

<sup>29</sup> A. Lopez-Martens et al., submitted to Phys. Lett. B

<sup>30</sup> H.B Jeppesen et al., Phys. Rev. C 79 (2009) 031303(R)

<sup>31</sup> K. Hauschild et al., to be published

<sup>32</sup> K. Hauschild et al., to be published

<sup>33</sup> K. Kessaci, PhD (Université de Strasbourg, 2021)

## Perspectives

The GABRIELA collaboration has built a state-of-the-art setup at the focal plane of a modern recoil separator. Since the commissioning phase of GABRIELA at SHELS, many new results have been obtained on the structure of super heavy nuclei as well as on reaction mechanisms used to produce them. In the coming years, the program will continue with special emphasis on:

- spectroscopy of heavy nuclei produced in asymmetric reactions using actinide targets. The new entrance quadrupole lens to SHELS should further improve the acceptance of SHELS for the evaporation residues produced in such reactions.
- spectroscopy of heavy nuclei produced with newly developed heavy beams such as  $^{54}\text{Cr}$
- Weak decay studies, such as the fission from high-K isomers in already well-studied cases
- X-ray fingerprinting of the heaviest nuclei: determination of the atomic numbers of the superheavy elements to unequivocally pin down the isolated hot fusion region

This last axis of research requires an upgrade of the Si detectors and front-end electronics of GABRIELA. Indeed, though the vacuum chamber inserts have been especially thinned to increase their transparency, the photon detection efficiency of GABRIELA, decreases dramatically at low energies approaching 0 below 20-30 keV. It has become clear that highly efficiency and low background detection systems sensitive to L and K X-ray quanta are a necessary equipment for the Z-identification of the heaviest elements<sup>34</sup> whether to determine the Z of the daughter when the alpha decay feeds levels which emit internal conversion electrons (ICE) or to check for the presence of electron capture (EC) within a decay chain. The race is now on for the glory of unambiguously assigning the Z of these superheavy nuclei. Last year the project SHEXI (SuperHeavy Element X-ray Identification) was submitted to the IN2P3. ASIC technology from the field of X-ray fluorescence spectroscopy and high resistivity thick silicon wafers are the key to success. The JINR will ensure the procurement of the silicon wafers this year. This is the only French led project with a chance of competing with the Lund-GSI collaboration. It is critical to start the ASIC design immediately. From T0 the delivery of the ASICs can take up to one year. The ASIC design and prototype delivery can be broken down into 15k+40k+40k parts to allow an immediate (2019) start to ensure measurements in 2020. The United Nations has proclaimed 2019 the "International Year of the Periodic Table of the Chemical Elements". Measuring X-rays from the heaviest elements in the recently filled seventh row and resolving the current dispute would provide excellent publicity for the IN2P3.

The total estimated cost for the detectors, front- and back-end electronics and densimet shielding is 735k. Currently the FLNR has agreed to pledge 240k. If France does indeed become an associated member of the JINR we hope part of this upgrade could be funded as a payment in kind to the JINR. But we must again stress that the ASIC design must start immediately – one year has already been lost.

To pursue this research activity an annual operations and maintenance budget of ~70k will also be required in order to cover approximately 300 days presence at the FLNR, conference attendance, computer equipment, ensure a reserve of critical equipment (such as back-end chassis and digitisers) and replace aging high- and low-voltage supplies. A continuation of the Projet de Recherche Conjoint beyond 2020 could reduce this by up to 7k. Opportunity: Associate member – annual purchase of critical reserves (20k).

## Projected timeline

It is foreseen to install and validate the instrumentation in full digital electronics of the GABRIELA setup by mid-2020. A dedicated experimental setup for L X-ray measurements can be achieved within 18 months of T0.

## Impact on other projects

An important aspect of our programme at Dubna is that it provides training in the complex art of performing position- and time-correlation analysis of rare events. This is an essential skill for successful

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<sup>34</sup> U. Forsberg et al., Phys. Lett. B 760 (2016) 293–296 and D. Rudolph et al., Phys. Rev. Lett. 111 (2013) 112502

campaigns with SIRIUS at S<sup>3</sup>. With the SHE-Factory coming online and the beam-time available at SHELS it would be advantageous to extend this training to post-docs, or, for the longer-term benefit of this field, a new CR recruitment.

The CSNSM and IPHC groups have been active in the developments and scientific program of the GABRIELA setup at the focal plane of SHELS for 15 years now. This expertise and experience has been an asset in the development of the SIRIUS focal plane detection system of the S<sup>3</sup> spectrometer. Moreover, members of the GABRIELA collaboration are WP leaders of the SIRIUS project and also part of the SIRIUS collaboration board.

## SWOT

<p style="text-align: center;"><b>Strengths</b></p> <ul style="list-style-type: none"> <li>- state of the art detection system</li> <li>- beam time &amp; intensity</li> <li>- availability of a variety of targets</li> <li>- visibility</li> <li>- experience</li> <li>- long-lasting collaboration built on complementary expertises</li> </ul>	<p style="text-align: center;"><b>Weaknesses</b></p> <ul style="list-style-type: none"> <li>- beam energy (limitations and instabilities)</li> <li>- limited funds for R&amp;D</li> </ul>
<p style="text-align: center;"><b>Opportunities</b></p> <ul style="list-style-type: none"> <li>- spectroscopy at the SHF</li> <li>- discovery potential</li> <li>- France becoming an associate member of the JINR</li> </ul>	<p style="text-align: center;"><b>Threats</b></p> <ul style="list-style-type: none"> <li>- U400 cyclotron upgrade will interrupt - data taking for some time</li> <li>- Reduced funding for missions</li> <li>- Proposed “open” PAC could potentially limit the role and the data obtained by IN2P3 scientists on their own equipment</li> <li>- Uncertainty of access to technical staff after the 5 lab fusion in Orsay</li> </ul>

### 3.3 Prompt Spectroscopy

As mentioned before, prompt spectroscopy (nuclear states with sub-nanosecond lifetimes) of heavy nuclei is carried out systematically only in 2 laboratories: Jyväskylä (SF) and Argonne (USA). These studies started in the 90s at JYFL and were made possible thanks to the combination of state-of-the-art detection setups with the efficient gas-filled recoil separator RITU. Among the first experiments performed by the collaboration was the spectroscopy of <sup>223</sup>Pa<sup>35</sup> and <sup>216</sup>Th<sup>36</sup> with JUROSPHERE. The setups have included over time the following items:

- A focal plane detection system (including over time efficient planar and Ge detectors for low energy photons, a DSSD for high-resolution alpha detection and PIN diodes for ICE measurements) for decay spectroscopy,
- Powerful Ge arrays arrays such as JUROSPHERE, JUROGAM phase 1, then 2 and now 3, around the target position for prompt spectroscopy.
- Unique detection systems for prompt ICE spectroscopy (SACRED<sup>37</sup> and now SAGE<sup>38</sup>), which enable in particular the detailed study of odd nuclei.

Born in GSI, the so-called Recoil Decay Tagging method based on genetic correlations can be used to shed light on the properties of isomeric states and collective phenomena in (super/very) heavy nuclei. One of the first highlights was the spectroscopy of <sup>254</sup>No<sup>39</sup>.

<sup>35</sup> F. Hoellinger et al., Phys. Rev. C **60** (1999) 57301

<sup>36</sup> K. Hauschild et al., Phys. Lett. **87** (2001) 072501

<sup>37</sup> P. A. Butler et al., Nucl. Inst. Meth. **A381** (1996) 433

<sup>38</sup> P Papadakis et al., 2011 J. Phys.: Conf. Ser. **312** 052017

The French community, mainly the CSNSM and IPHC laboratories for the IN2P3 and the IRFU for the CEA, have been strongly represented and very active since the beginning with some French physicists staying in JYFL for long periods of time (postdocs, “CNRS mise à disposition”, PhD students).

## Prizes & communication

- First international Szymanski prize awarded at ENAM’08 for:  
“outstanding contribution to experimental in-beam studies of superdeformed, octupole deformed and heavy nuclei

## Achievements

The French community has been spokesperson/co-spokesperson of many experiments. They have also been the leaders or instigators of new technical developments for the instrumentation dedicated to the spectroscopy of very heavy nuclei such as:

- The design at the IPHC and the installation of a rotating target<sup>40</sup> permitting to accept higher beam intensities
- The transition from JUROGAM I to JUROGAM II, where IPHC physicists had a significant contribution to the design and test phases
- The installation of a triggerless digital electronic data-acquisition system TNT2 developed at IPHC for the GABRIELA project (see section 3.2), which led to the spectroscopic studies of <sup>255</sup>Lr<sup>41</sup> and <sup>246</sup>Fm nucleus<sup>42</sup> with the lowest cross section ever reached at that time of 11 nb. This study was highlighted in a “Lettre électronique de l’IN2P3” in May 2010 and the subject of J.Piot’s PhD thesis of Strasbourg University.
- The start of a collaboration of new material components for beam developments called MIVOC (cf. section 3.4). The development of MIVOC <sup>50</sup>Ti beam<sup>43</sup> gave access to a major result: the first prompt and decay spectroscopy study of a superheavy element, <sup>256</sup>Rf<sup>44</sup>. This study was the subject of J. Rubert’s PhD thesis of the Strasbourg University and gave a Viewpoint in the Physics revue of the American Physical Society in 2012. The moment of inertia of the band (seen in the figure below), compared to those of other ground state bands in the region, has confirmed that there is no deformed shell gap at Z=104.

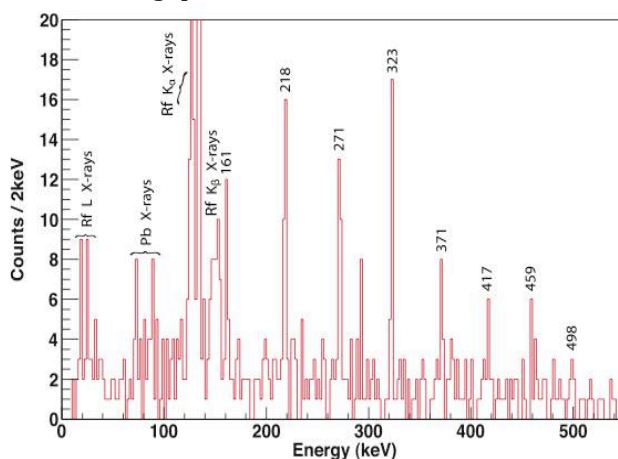


Figure 12: Spectrum of prompt gamma rays emitted by <sup>256</sup>Rf (taken from Phys. Rev. Lett. 109 (2012) 012501 ).

- The digitization of the planar detector to enhance the detection efficiency at low energy, especially important for isomer-decay spectroscopy

<sup>39</sup> M. Leino et al., Eur. Phys. J A (1999) 63

<sup>40</sup> Annual Report, JYFL, 2004

<sup>41</sup> S. Ketelhut Phys. Rev. Lett. 102 (2009) 212501

<sup>42</sup> J. Piot et al., Phys. Rev. C 85 (2012) 85

<sup>43</sup> J. Rubert et al., Nucl. Instr. Meth. Phys. Res. B276 (2012) 33

<sup>44</sup> P.T. Greenlees et al., Phys. Rev. Lett., 109 (2012) 012501, J. Rubert’s PhD thesis, Strasbourg University

The most recent activities at JYFL have been performed with the SAGE array, which was commissioned in 2009-2010, to which CSNSM and IPHC participated. The first experimental campaign dedicated to heavy elements with SAGE started in February 2012 with an 11-day run on  $^{251}\text{Md}$ , in which CSNSM, IPHC and JYFL physicists participated. The results, which have revealed an identical band phenomenon in this mass region, will be submitted to *Phys. Rev. Lett.*<sup>45</sup>. The nucleus  $^{253}\text{No}$  was revisited with SAGE, and the analysis has confirmed the assignment of  $9/2^-$  for the spin and parity of the ground state<sup>46</sup>. This was then confirmed by laser spectroscopy at SHIP in GSI<sup>47</sup>. An experiment was also proposed by the CSNSM and IPHC to study the nucleus  $^{255}\text{Lr}$ . It was also carried out in 2012, however, problems with the  $^{48}\text{Ca}$  beam intensity and neutron damage in the detectors did not allow the goals of the experiment to be reached. The experiment will be re-scheduled when beam developments have been carried out. In 2015, an experiment proposed by CSNSM and the University of Oslo was performed to look for an M1 scissors resonance in  $^{254}\text{No}$ . Such resonances have been observed in deformed actinide nuclei<sup>48</sup> and are predicted to exist in the transfermium and have a strong impact on astrophysical rates. The range of detectable photons in SAGE was increased and an excess intensity was observed, but with too few statistics to firmly establish and determine its nature.

## Perspectives

The activities of the French community in JYFL have decreased in these last years, mainly due to the fact that the gamma-ray detection system at the target of RITU was removed for the nuball campaign at ALTO (Orsay) but also because the field has reached its limit in terms of detection sensitivity. This last factor should drastically change by the end of the 2020s', with the foreseen installation of the AGATA array at JYFL. The envisaged program has been addressed in the AGATA white book and involves detailed spectroscopy of odd nuclei, the search for resonances and the measurement of fission barriers, such as has been done by CSNSM and IPHC physicist in  $^{254}\text{No}$ <sup>49</sup>.

## Budget & HR

The budget for prompt spectroscopy studies at JYFL has been obtained from IN2P3 and has mainly been for travel to and from Finland and living expenses on site. Regular funds from ENSAR2 have also been available, which has reduced the actual cost of missions.

Two IN2P3 physicists from CSNSM took a "mise à disposition" at JYFL for the commissioning phase of SAGE and the first experimental campaigns from 2009-2011.

The list of French PhD theses obtained on the subject of very/super heavy nuclei with data from JYFL are the following: F. Hoellinger, Strasbourg University (1999), A. Chatillon, Lyon University (2005), F. Khalfallah, Strasbourg University (2007), J. Piot, Strasbourg University (2010), F. Déchery, Paris 7 University (2012), J. Rubert, Strasbourg University (2013) and R. Briselet, Université Paris Saclay (2016).

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<sup>45</sup> R. Briselet et al., to be submitted to *Phys. Rev. Lett.*

<sup>46</sup> A. K. Mistry et al., *Eur. Phys. J. A* (2017) **53**: 24

<sup>47</sup> S. Raeder et al., *Phys. Rev. Lett.* 120 (2018) 232503

<sup>48</sup> M. Guttormsen et al., *Phys. Rev. Lett.* 109 (2012) 162503

<sup>49</sup> G. Henning et al., *Phys. Rev. Lett.* 113 (2014) 262505

## SWOT

<b>Strengths</b> <ul style="list-style-type: none"><li>- A powerful and proven recoil separator and its focal plane detection</li><li>- Prompt and decay spectroscopy simultaneously</li><li>- -user-friendly facility</li><li>- state of the art detection system for prompt spectroscopy</li><li>- experience</li><li>- long-lasting collaboration built on complementary expertises</li><li>-</li></ul>	<b>Weaknesses</b> <ul style="list-style-type: none"><li>- Rather low beam intensity (limitations to reach very low cross sections)</li></ul>
<b>Opportunities</b> <ul style="list-style-type: none"><li>- Prompt spectroscopy of SHE</li><li>- AGATA installation foreseen</li><li>- Odd nuclei spectroscopy</li><li>- Fission barrier measurements</li></ul>	<b>Threats</b> <ul style="list-style-type: none"><li>- Reduced funding for missions</li></ul>

### 3.4 Development of intense metallic beams

Started in 2006, the development of new intense titanium beams was driven by the need for a  $^{50}\text{Ti}$  beam in order to achieve the first prompt gamma-ray spectroscopy of the superheavy nucleus  $^{256}\text{Rf}$ . IPHC was co-spokesperson for this experiment at JYFL. Titanium has a high melting temperature and is rather difficult to ionize. It is thus rather difficult to produce intense beams. The investigation of possible techniques led to two solutions for the  $^{256}\text{Rf}$  experiment: the development of a new MIVOC compound and the use of metallic or titanium oxide pellets in an inductive oven. Both were developed. In this document, this development is accounted for separately from production of rare isotopic compound for the experiments.

#### MIVOC

The MIVOC Method was invented by M. Nurmia from JYFL. It is based on the fact that a volatile compound can efficiently bring a metal in the plasma of an ECR ion source and gives very low material consumption. At the start of the project, one titanium MIVOC compound was already identified in JYFL but could neither provide the needed intensity ( $^{50}\text{Ti}$  being only 5% of the natural Ti) nor be bought with isotopic material. Thanks to a very efficient multi-disciplinary collaboration between the DSA and DRS departments of IPHC, all the possible MIVOC compounds of titanium were synthesized and tested. Only two of them met the requirements:  $\text{Cp}_2\text{TiMe}_2$  and  $\text{Cp}^*\text{TiMe}_3$ . Unfortunately, both were quite difficult to handle.

In September 2011, after several years of intensive chemical synthesis program with natural material, a first isotopic  $\text{Cp}^*\text{TiMe}_3$  MIVOC compound gave almost  $20\ \mu\text{A}$  of  $^{50}\text{Ti}^{11+}$  with an ECRIS2 ion source for a three week-steady operation<sup>50</sup>. This experiment was the first in-beam spectroscopy of a superheavy nucleus,  $^{256}\text{Rf}$ , in which a rotational band was observed (see section 5). The conflict between previous focal-plane studies in Berkeley and Argonne was settled through the observation of a cascade of K isomers<sup>51</sup>. The same compound was tested in GANIL where a  $28\ \mu\text{A}$   $^{50}\text{Ti}^{10+}$  beam intensity was extracted and tested several hours under stable operation. Complementary tests were performed with a ECR4M source in Dubna reaching  $67\ \mu\text{A}$  for  $^{50}\text{Ti}^{10+}$  as well  $80\ \mu\text{A}$  for  $^{50}\text{Ti}^{5+}$  and a little later a  $15\ \mu\text{A}$   $^{48}\text{Ti}^{11+}$  beam was produced at RIKEN. Finally due to acceleration and transport efficiencies,  $\sim 0.5\ \mu\text{A}$  on target were achieved in all 3 installations.

<sup>50</sup> J. Rubert, J. Piot et al., Nucl. Instr. Meth. Phys. Res B **276** (2012) 33

<sup>51</sup> J. Rubert, B. Gall et al., to be submitted to Phys. Rev. C or Phys. Lett. B.



Further development of the synthesis process enabled starting from titanium oxide ( $\text{TiO}_2$ ) instead of tetrachloride ( $\text{TiCl}_4$ ). From then on,  $^{46}\text{Ti}$  and  $^{47}\text{Ti}$  (that one can only find as oxides on the market) could be accelerated for a SHE experiment in Dubna and for a neutron deficient polonium ( $^{188}\text{Po}$ ) study in JYFL in March 2017. This enabled also the reprocess of the remaining material saving the equivalent of 30 k€ of isotope for the collaboration.

Counting all the MIVOC beam time with  $^{46-50}\text{Ti}$ , one gets quite close to months of cumulative operation in JYFL, GANIL, DUBNA and RIKEN.

Vanadium MIVOC compound was developed in a few months. Two compounds were successfully tested and accelerated at RIKEN:  $\text{Cp}_2\text{V}$  (vanadocene) and  $\text{Cp}_2\text{VCl}_2$ . Both were working well reaching  $15 \mu\text{A } ^{51}\text{V}^{11+}$  out of the source, but since for this element the  $^{51}\text{V}$  isotope nearly exhausts the natural abundance, consumption is not an issue. This beam was finally accelerated at RIKEN with a high temperature oven technique.

As for  $^{46,47}\text{Ti}$ , enriched Chromium can only be found as oxide or metal and the development of a MIVOC compound needed a high-temperature chlorination step. Know-how developed for titanium enabled a rather rapid success of this synthesis. For Chromium, several compounds were tested with natural material and finally chromocene  $\text{Cp}_2\text{Cr}$  gave the best results. First tests with natural chromocene produced at IPHC Strasbourg were performed in Dubna early 2018 and first tests with highly enriched  $^{54}\text{Cr}$  were made a few months later reaching  $95 \mu\text{A } ^{54}\text{Cr}^{5+}$  out of the source ( $19 \mu\text{A}$ ). The first physics experiment (study of fission dynamics for element 120) ran in May 2019.

We plan to have a first spectroscopy run using  $\text{Cp}_2^{54}\text{Cr}$  with SHELS this autumn. Indeed since Coulomb barriers of reactions are higher, this beam needs higher bombarding energy or lower energy loss in the target backing. Both of these points need developments in Dubna. The situation is similar for the synthesis of new elements in Dubna where one plans to use this beam when the DC280 is fully operational.

These 3 MIVOC beams represent the key beams for the synthesis of elements 119-122. One major question is now whether we have the needed intensity. Up to now intensity on target with these MIVOC beams have been between 0,5 and  $1 \mu\text{A}$ . A factor of 10 more is needed. Improvements of the MIVOC injection is under study, but one absolutely needs test bench time for this. Another option is to develop in parallel new high temperature ovens.

## INDUCTIVE OVEN

Early developments of an inductive oven were made in 2006 in collaboration with JYFL where a high temperature inductive oven was in test. For these first tests, titanium oxide was reduced at IPHC and metallic pellets were produced as well as oxide ones. Rather promising results were obtained with metallic pellets around  $1600^\circ\text{C}$  where with oxide pellets one needed temperatures  $100^\circ\text{C}$  higher and where there were issues with Oxygen present in the plasma that was acting as a getter for the titanium. Finally these tests were stopped abruptly after a thermal induced demagnetizing of one of the permanent magnets of the ECR ion source.



Figure 13: Pellet preparation from reduced  $\text{TiO}_2$  in 2008

This development was started again at IPHC in mid 2017. Our plan was to have an inductive oven ready for integration in an ion source in 3 years of time. With R&D budgets for metallic beams at IPHC one could almost gather all the needed equipment in two years. A prototype of high temperature inductive oven should be operational this summer at IPHC with a size 2 oven. On the basis of the expertise gained with this test bench, one foresees to develop a version that will be integrated in an ECR ion source.

Dubna is candidate to host and finance a test version, but we will also discuss in June possibility of tests in France (GANIL or Grenoble). Feedback of this R&D program should be linked to the new A/Q = 7 ECR ion source for SPIRAL2/S3/SIRIUS.

## Milestones

- First tests in JYFL (April 2008)
- 2018 Tests of inductive circuits in air. Selection of dedicated power supply
- S1 2019 design of the test bench. Ordering pieces.
- Summer 2019 set-up of test bench
- S2 2019 test in vacuum and several inductors systems
- 2020 integration in a ion source first real operation tests
- Uranocene

## Prizes & communication

- JINR Instruments and Methods 2<sup>nd</sup> prize for the production of intense beams using the MIVOC method (2015)
- In 2017, MIVOC beams were mentioned in an article of Le Monde: « Chez les chasseurs russes des nouveaux atomes », Le Monde, 12 juillet 2017

## Budget & RH

A budget of around 9 k€ per year is allocated to intense beam development R&D in the framework of the IN2P3 master project "Stables beam developments". Production of isotopic compound is performed on a real cost basis where "selling prices" includes isotope furniture, chemistry efficiency, solvents and reactants and necessary labware replacement. In case some money is left, it is included in the R&D programme.

The development of MIVOC compounds in Strasbourg is the oldest pluridisciplinary collaboration of IPHC. The R&D programme is carried by two researchers (Z. Asfari - Chemist, and B. Gall - Physicist) and one assistant engineer (M. Filliger - 66% of his time). Three PhD students and post-docs also part-time contributed to the project: J. Piot (PhD 2011), J. Rubert (PhD 2013), H. Faure (PhD 2015).

This technical development also strongly depends on collaboration with major installation and drove strong and fruitful international collaboration with:

- JYFL (Jyväskylä, Finlande) : P. Greenlees, J. Ärje, H Koivisto and R. Seppälä,
- GANIL (Caen, France) : J. Piot, F. Lemagnen and his team of ion source R&D and operation,
- FLNR (Dubna, Russie) : A. Yeremin, S. Bogomolov, V. Loginov, B. Bondarchenko
- RIKEN (Tokyo, Japon) : K. Morita, K. Morimoto, H. Haba & M. Kidera.

## Perspectives

Uranium is also one key beam for SHE studies. R&D programme is on-going for the development of this beam through both MIVOC compounds and high temperature oven technique.. When the new DC280 cyclotron and the new-RILAC will enter in action beginning 2020, one will be able to test high intensity metallic beam production with methods developed at IPHC.

These developments driven by this VHE and SHE programme drove a renewal of the MIVOC method. With the skill developed, one can also imagine <sup>50</sup>Cr beams, isotopic Cp2Mg beams and many others that can now be prepared at IPHC.

## SWOT

<p style="text-align: center;"><b>Strengths</b></p> <ul style="list-style-type: none"> <li>- unique collaboration between chemist and physicists</li> <li>- Know-how worldwide recognized</li> <li>- Ability to develop new MIVOCs</li> <li>- international level of collaboration</li> <li>- Scientific Niche =&gt; Low risk</li> <li>-</li> </ul>	<p style="text-align: center;"><b>Weaknesses</b></p> <ul style="list-style-type: none"> <li>- Program relies on 3 people working part time on the project</li> <li>- low priority of pluridisciplinarity at CNRS</li> </ul>
<p style="text-align: center;"><b>Opportunities</b></p> <ul style="list-style-type: none"> <li>- Collaboration with JYFL (other "standard MIVOC's (Cp<sub>2</sub>Fe, Cp<sub>2</sub>Ni et Cp<sub>2</sub>Mg)</li> <li>- discovery potential:             <ul style="list-style-type: none"> <li>--&gt; <sup>295</sup>Uuo (<sup>248</sup>Cm + <sup>50</sup>Ti --&gt; <sup>295</sup>118 +3n, a heaviest 118)</li> <li>--&gt; <sup>296</sup>Uun (<sup>249</sup>Bk + <sup>50</sup>Ti --&gt; <sup>296</sup>119 +3n)</li> <li>--&gt; <sup>296</sup>Uun (<sup>248</sup>Cm + <sup>51</sup>V --&gt; <sup>296</sup>119 +3n)</li> <li>--&gt; <sup>296</sup>Ubn (<sup>249</sup>Cf + <sup>50</sup>Ti --&gt; <sup>296</sup>120 +3n)</li> <li>--&gt; <sup>296</sup>Ubn (<sup>248</sup>Cm + <sup>54</sup>Cr --&gt; <sup>299</sup>120 +3n)</li> </ul> </li> <li>- New machines (SHE Factory at Dubna, S<sup>3</sup> at GANIL and GARIS II at RIKEN)</li> <li>- Valorization possible for many systems developed within the project</li> </ul>	<p style="text-align: center;"><b>Threats</b></p> <ul style="list-style-type: none"> <li>- Some compounds are toxic</li> <li>- retirement of Z. Asfari (between 2020 and 2022)</li> <li>- Results of R&amp;D without guarantee</li> <li>- Need for high intensity ?</li> </ul>

## 4. Conclusion

The synthesis of new elements and spectroscopic studies of very and super heavy nuclei are of major interest for our knowledge of physics and chemistry at the upper mass limit of the Mendeleev's table. As demonstrated in this document, IN2P3 researchers have been pulling for new and outstanding results in the field. Their contribution to enable what was considered before as impossible experiments, led to an innovative rotating target, new beam production methods two digital electronics revolutions, first with the TNT2 boards and a second on-going one with our latest projects in Dubna. Forefront projects such as JUROGAM widely benefited from this expertise and provided, at the same time, the opportunity for IN2P3 researchers to run major experiments such as the first prompt spectroscopy of a superheavy nucleus <sup>256</sup>Rf.

Two ANR grants and an efficient and exemplar international collaboration between the IN2P3 and JINR researchers enabled the construction of GABRIELA and SHELS, which are forefront instruments for heavy elements spectroscopic studies. An excellent physics programme associated to numerous months of beam time generated an impressive list of achievements. This project has not only pushed the limits of the state of the art, but also created favourable conditions for the start of one of the major programme in France in heavy element studies, namely the S3 project. The commissioning of S<sup>3</sup> will widely benefit from the expertise gained during the commissioning and running of SHELS and the new GARIS II

High intensity beam developments not only opened the possibility to attempt the synthesis of new elements 119-122, but it also opened a wide research program with SHELS and soon S3/SIRIUS when the new A/Q = 7 injector is made able. It is the perfect illustration that these projects are complementary. With beam intensity increasing drastically targets able to operate under these extremely severe conditions need to be developed. This will certainly induce new methods and maybe even patents.

This programme is a great chance for the IN2P3 researchers, PhD students and associated engineers. There is no doubt that it will lead to major results in the coming 4 years with the new machines DC280 in Dubna, the RILAC in Riken and the LINAC of SPIRAL2 coming on line.

## Appendix: Publication list

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