# EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

## Letter of Intent to the ISOLDE and Neutron Time-of-Flight Committee

Research plans for the laser-polarization beamline VITO at ISOLDE

8 January 2020

A. N. Azaryan<sup>1,2</sup>, A. Antusek<sup>3</sup>, M. Baranowski<sup>4</sup>, M. L. Bissell<sup>5</sup>, L. Cerato<sup>6</sup>, J. Croese<sup>1,6</sup>, W. Gins<sup>7</sup>, K. Dziubinska-Kuehn<sup>1,8</sup>, A. Fijałkowska<sup>9</sup>, B. Karg<sup>6,1</sup>, M. Kowalska<sup>1</sup>, A. Korgul<sup>9</sup>, M. Kozak<sup>4</sup>, K. Kulesz<sup>6,1</sup>, M. Madurga<sup>10</sup>, Ch. Mazzocchi<sup>9</sup>, K. Miernik<sup>9</sup>, G. Neyens<sup>1,11</sup>, S. Pallada<sup>12</sup>, L. Popescu<sup>13</sup>, L. M. C. Pereira<sup>11</sup>, M. Piersa<sup>9</sup>, Z. Salman<sup>14</sup>, N. Severijns<sup>11</sup>, J. Van de Walle<sup>13</sup>, Ph. Wagenknecht<sup>10</sup>, S. Warren<sup>6,1</sup>, J. Wolak<sup>4</sup>, Z. Xu<sup>10</sup>, D. Zakoucky<sup>15</sup>

- 1. CERN, Geneva, Switzerland
- 2. JINR, Dubna, Russia
- 3. Slovak University of Technology, Bratislava, Slovakia
- 4. Adam Mickiewicz University, Poznan, Poland
- 5. University of Manchester, Manchester, Great Britain
- 6. University of Geneva, Geneva, Switzerland
- 7. University of Jyväskylä, Jyväskylä, Finland
- 8. University of Leipzig, Leipzig, Germany
- 9. University of Warsaw, Warsaw, Poland
- 10. University of Tennessee, Knoxville, United States
- 11. KU Leuven, Leuven, Belgium
- 12. HEPIA and HEdS, Geneva, Switzerland
- 13. SCK-CEN, Mol, Belgium
- 14. PSI, Villingen, Switzerland
- 15. Czech Academy of Sciences, Rez, Czech Republic

Spokesperson: Magdalena Kowalska (kowalska@cern.ch) Local contact: Magdalena Kowalska (kowalska@cern.ch)

#### Abstract

The new VITO beamline at ISOLDE is devoted to versatile studies with laser-polarised radioactive beams. The beamline has been already used for biological  $\beta$ -detected NMR and to prepare for the  $\beta$ asymmetry measurements related to the determination of the Vud matrix element from mirror transitions, for testing the Standard Model. In the future we plan to add to this program also −−neutron correlation studies to determine spins of excited nuclear states for nuclear structure and astrophysics related research,  $\beta$ -detected NMR to measure electromagnetic moments of short-lived nuclei for nuclear structure, and  $\beta$ -detected NMR for condensed matter studies.

Ongoing and planned VITO upgrades, which should allow for the above studies, include the implementation of a superconducting magnet, a more efficient liquid-vacuum interface, a reionisation cell, a  $\beta$ - $\gamma$  correlations setup, a three-way beam switchyard, and UHV chambers for energy-controlled implantations.

This document gives a brief overview of the above VITO projects and planned upgrades, along with the required space in the ISOLDE hall.

#### Application of spin-polarised radioactive nuclei:

The common point of studies using spin-polarised radioactive nuclei is the fact that the  $\beta$ particles are emitted asymmetrically in space. The angular distribution of  $\beta$  radiation  $W(\theta)$ relative to an orientation axis depends on the degree of polarization  $\langle I \rangle / I = P$ , the  $\beta$ -decay asymmetry parameter  $A$ , and the velocity of the emitted  $\beta$ -particle  $\nu$ :

$$
W(\theta) \sim 1 + A \frac{\langle I \rangle p_e c}{I E_e} \cos(\theta) = 1 + AP \frac{v}{c} \cos(\theta)
$$
 (1),

The experimental  $\beta$ -decay asymmetry for a given transition, measured usually for detectors placed at 0 and 180 degrees is thus given by (ignoring the solid angle of the detectors and other setup-related factors):

$$
A_{exp} = P A \tag{2}
$$

Furthermore, for allowed  $\beta$  transitions A depends in the following way on the spin of the decaying and the populated state,  $I_i$  and  $I_f = I_i + \Delta I$  respectively:

$$
-1 \quad \text{for} \quad \Delta I = -1
$$
\n
$$
I_i/(I_i + 1) \quad \text{for} \quad \Delta I = +1
$$
\n
$$
-1/(I_i + 1) \quad \text{for} \quad \Delta I = 0 \quad \text{(Gamow-Teller transition)}
$$
\n
$$
0 \quad \text{for} \quad \Delta I = 0 \quad \text{(Fermi transition)}, \tag{3}
$$

If no distinction is made between different decay channels, then the observed asymmetry is a weighted average of the asymmetries corresponding to different transitions. However, if the experimental asymmetry can be measured individually for different transitions (e.g. by making coincidences with subsequent  $\gamma$  rays of known energy), then *A* for a given transition can be derived and thus one can determine both  $I_i$  and  $I_f$ , if one of them is known. If one can measure *A* very precisely, then one can even test the details of the weak interaction governing the  $\beta$  decays. Furthermore, the disappearance of the asymmetry when the polarisation is destroyed with resonant radiofrequency can be used to perform sensitive studies using Nuclear Magnetic Resonance (NMR). This  $\beta$ -NMR approach can use a billion times fewer nuclei than in conventional NMR, because it relies on a high degree of polarisation and efficient detection of  $\beta$  particles. It can be used in nuclear structure to determine magnetic and quadrupole moments of short-lived nuclei [Ney05, Neu08], as well as in material science, and since more recently also chemistry and biology to determine the local environment of the probe nuclei [Jan17]. All the above applications of polarised radioactive nuclei are summarised in a graphical form in Fig. 1 below.



Fig 1. Selected research topics with spin-polarised short-lived nuclei

## Ongoing and planned research with spin-polarised nuclei at the VITO beamline:

In the following we will discuss the ongoing and planned research at the VITO beamline:

-NMR in chemistry and biology (ongoing), performed within M. Kowalska's ERC Starting Grant (running until Sept 2021). It is motivated by the study of the local electronic/chemical environment of metal ions in biologically-relevant materials. Since these are very dilute in nature it is hard to perform such studies using conventional NMR and therefore B-NMR presents great potential for such applications, due to the high sensitivity and element specificity. In this project, we are interested in determining the chemical environment of the metal, based on the NMR resonance frequency, line shape and the spin-lattice relaxation time  $T_1$ , with which the probe's polarization reaches equilibrium.

Our first scientific case, studied within the experiment IS645 [Kow17, Kow18], is the interaction of sodium with G-quadruplex DNA structures, which require alkali metals to be formed. Before Long Shutdown 2 (LS2) we recorded NMR resonances and spin-lattice relaxation time curves from 26-28Na in several solvents in absence and in presence of selected G-quadruplex DNA strands. Relaxation-time data is still under analysis with the aim to compare the measured  $T_1$  with those based on calculations performed by our theory colleagues (group of T. Wesolowski, UNIGE). NMR spectra will be more difficult to interpret, because the most suitable solvent to fold DNA into G-quadruplex structures (a mixture of glycerol and choline chlorine) gave very broad resonances, which cover the range of free sodium and sodium bound inside a G-quadruplex.

One clear physics result of our studies is the determination with part-per-million accuracy of the magnetic moment of  $^{26}$ Na [Har20]. This was possible thanks to recording narrow resonances in liquid hosts and quantum chemistry calculations which corrected the magnetic moment of the stable  $^{23}$ Na reference (performed by our theory collaborator A. Antusek, Slovak Academy of Sciences). The resulting magnetic moment is two orders of magnitude more accurate than achievable with other approaches. This will be important when comparing  $\beta$ -NMR chemical shifts to those in conventional NMR and might be interesting also for nuclear structure studies.

During LS2 we are working on different experimental upgrades. Firstly, a more efficient liquid-vacuum interface which will allow us working with biologically more suitable solvents (e.g. glycerol or even water). Secondly, a higher magnetic field provided by a superconducting magnet which will increase the NMR resolution. Finally, the use of potassium isotopes (which were polarised over 20 years ago at the COLLAPS setup [Neu18]) will allow us to look at G-quadruplexes that bind the metal ion even stronger than in the case of sodium. After LS2, we aim at looking at the interaction of Na and K with Gquadruplex structures using the upgraded setup. In the longer term, we want to expand the biochemistry applications to other biologically relevant metals: magnesium (already polarised at ISOLDE), copper, and zinc, and study their interaction with different metalloproteins (interesting e.g. for the group of M. Kozak, AMU, Poznan).

Determination of the  $V_{ud}$  matrix element from mirror decays and tests of the CKM-matrix unitarity (preliminary studies on <sup>35</sup>Ar, led by G. Neyens and N. Severijns were performed in 2017) [Vel14], [Gin19]. To determine  $V_{ud}$  from a mirror decay, in addition to the mass difference, lifetime, and branching ratio for the mirror transition, one needs also the ratio  $\rho$ between the total Fermi and Gamow-Teller strengths in the mirror transition. Since  $\rho$ determines the value of the transitions' asymmetry parameter *A*, (see equations 1-3) it can be determined from the measured  $\beta$ -decay asymmetry  $A_{\text{exo}}$ . Experiment IS601 aimed at establishing an acceptable level of polarisation for <sup>35</sup>Ar, high enough to measure *A* with 0.5% precision, such that  $V_{ud}$  can be determined with an acceptable precision after a 2-week beamtime with several  $10^8$  ions/s. We employed for the first time laser optical pumping with 3 laser frequencies simultaneously to enhance the induced spin polarization in the <sup>35</sup>Ar atom beam. Simulations and observed relative peak intensities in the hyperfine structure hint at close to 100 % polarisation of atomic spins in the relevant state. Calculations of the charge exchange process predict that about 30% of the total beam is this state after the neutralisation process. Unfortunately, the degree of  $\beta$ -decay asymmetry points to a nuclear polarisation that is of an order of magnitude lower [Gin19]. The polarisation loss occurs either as the spins are rotated into the strong magnetic field or upon implantation into the target due to the interaction of the spin of <sup>35</sup>Ar with the host.

Several technical improvements should make it possible to increase the decay asymmetry about 5-fold, which is needed to perform the final experiment of *A* of the mirror transition in <sup>35</sup>Ar. For this final measurement, one will need to know very well the degree of spin polarisation. This can be done by using as a reference another  $\beta$  branch with known  $\ddot{A}$ , which can be distinguished from the interesting transition, e.g. by a coincidence with a subsequent  $\gamma$  ray. <sup>35</sup>Ar possesses such a decay channel, with  $A = 1$  and 1% branching ratio, which is followed by a prompt  $\gamma$  ray of 1219 keV. The final setup will therefore need to combine a compact magnet with  $\beta$  and  $\gamma$  detection, allowing to tag  $\beta$  particles with the 1219 keV  $\gamma$  rays. A very similar setup is under construction by M. Madurga for his VITO project on spins of excited nuclear levels (see below), which brings good synergies within VITO.

Spins of nuclear excited levels in short-lived nuclei for nuclear-structure and astrophysics studies (planned by M. Madurga from U. Tennessee). The method relies on observing the different  $\beta$  emission asymmetries in coincidence with  $\gamma$  rays or in the future also neutrons depopulating a particular state to determine its spin-parity. This method was used by the Osaka group to determine the nuclear structure of daughters in a variety of experiments involving delayed  $\gamma$  [Nis17] and neutron emission [Miy03,Hir05]. The experimental setup is in preparation, a Letter of Intent was already submitted in 2017 to the INTC [Mad17] and – as requested by the INTC – a full scientific proposal will be submitted before the first physics studies are performed. The first regions of interest are neutron-rich Na, K, and Zn isotopes. The polarisation of these elements is also interesting for the biochemical studies with  $\beta$ -NMR, so we can profit from a clear synergy between these VITO sub-projects.

One of the motivations of this project are the recent advances in nuclear shell models, which can now reproduce shape coexistence from microscopic foundations [Tog16]. However, experimental information needed to validate these models is limited to cases close to stability [Kre16]. In many nuclei for which models predict shape coexistence (the so-called fifth-island of inversion at  $Z \sim 30$ , N $\sim 50$  [Now16]), only the half-life, the spin-parity of the ground state and the energies of the excited states are known. Using  $\gamma$  and neutron tagging it is possible to determine the spin-parity of excited states populated in  $\beta$  decay from their  $\beta$ asymmetry [Miy03]. This is the missing piece of the puzzle needed to firmly establish shape coexistence in the predicted regions, thus providing data with which we can validate the aforementioned calculations.

Magnetic dipole and electric quadrupole moments of short-lived nuclei with  $\beta$ -NMR for nuclear structure studies (planned by M. Kowalska, M. L. Bissell, G. Neyens). These observables are sensitive probes of nucleon configurations inside atomic nuclei, as shown e.g. in [Ney05] or [Neu08]. The studies can be performed without any modifications to the existing experimental setup. We will use  $\beta$ -NMR in solid and – if required – also liquid samples (which will give higher precision due to resonance narrowing in liquids [Har20]). One of the first studies will concern the quadrupole moments of neutron-rich K isotopes, in the vicinity of the new neutron closed shells N=32 and 34 discussed in the Ca chain. Potassium laser polarisation is interesting also for the biological  $\beta$ -NMR studies.

#### Depth controlled **β**-NMR for materials science (planned by L. Pereira and Z. Salman)

The size of magnetic and electronic systems studied by condensed matter research has decreased dramatically in the past two decades. Research in nanostructures opens up new opportunities for the design and optimization of materials properties for specific technological applications, such as electronics, optoelectronics and memory devices [Oht02, Gat06]. Material research has produced systems with reduced dimensionality such as thin films, interfaces, nano-particles, quantum dots and single-molecule magnets. In these nanostructures proximity to the surface or interface, finite size and quantum mechanical effects alter their bulk properties. Here we plan to develop a research program aimed at investigating the modification of bulk properties in systems with reduced dimensionality. Understanding how and why these properties are affected is of paramount interest both from a fundamental point of view as well as for technological applications, including nanoelectronics, spintronics, quantum technologies and energy materials. Depth controlled β-NMR is particularly powerful in this context [Kie03, Mor04, Sal06, Sal07, Sal07b, Sal12, Sug19]. While many techniques can be used to investigate the local properties of the top few monolayers in condensed matter systems (or alternatively the bulk), few are capable of probing these properties in buried interfaces or in a depth resolved manner. Depth controlled β-NMR [Mor04, Sal06], as well as low-energy µSR (LE- µSR, at PSI) [Mor03, Pro08] are two such novel techniques. These techniques are similar to conventional  $\mu$ SR and NMR, but the main difference is the ability to tune the implantation energy of the radioactive probe, and therefore its stopping depth, allowing a depth resolved measurement in the range 0-300 nm. While for LE-µSR one uses positive muons as a spin probe, spinpolarized Li ions (or other suitable radioisotopes) are used for β-NMR. Isotopes of light elements such as Li and Be are extremely versatile, as they can be used as probes in basically any solid (i.e. any research context), as their perturbation of to the host material is minimal. Isotopes of heavier elements (e.g. Na, Mg, Al, Cu) are better suited to study systems that contain such elements, for example, Na or Mg based battery materials (in addition to Li), or functional electronic materials containing Al or Cu (e.g. III-V semiconductors and magnetic semiconductors). The polarization of the spin probe is monitored through its asymmetric  $\beta$ decay. This detection method, the tunable low implantation energy, coupled with a high degree of spin polarization which is achieved before implantation, makes these techniques  $\sim$ 10<sup>13</sup> times more sensitive than conventional NMR [Kie03].

## Status and upgrades of the VITO beamline:

The VITO beamline can be currently used to laser-polarise beams of neutral atoms delivered from ISOLDE targets, to observe the resulting asymmetry in their  $\beta$  decay [Kow17b, Gin19b], and to perform  $\beta$ -NMR studies in solid and liquid hosts (at pressures between  $10^{-7}$ and  $10^{-1}$  mbar) [Kow18, Har20, Har20b]. At present the magnetic field at which the  $\beta$ -decay asymmetry and  $\beta$ -NMR resonances are recorded, is provided by an electromagnet which can reach 1.2 T (see Fig 1, top) ), and long-term stability and control of the static field are guaranteed by a stable <sup>1</sup>H-NMR probe with feedback loop [Har20].

The existing beamline has been designed, installed, and commissioned with the funding and workforce of M. Kowalska (CERN), G. Neyens and N. Severijns (KU Leuven), M. Madurga (U Tennessee), M. Kozak and M. Baranowski (Poznan).

In the near future we intend to implement the following upgrades to the VITO beamline (see Fig. 2 bottom for a preliminary layout), which will allow the studies described in the previous section and which will increase the VITO footprint in the ISOLDE hall:

-Horizontal 4.7 T superconducting magnet with 15 cm diameter bore. The magnet was given to M. Kowalska by ETHZ and will be transported to a storage at CERN on January  $13<sup>th</sup>$  2020. It will be crucial for higher resolution of biological  $\beta$ -NMR and will be useful for other  $\beta$ -NMR studies, concerning nuclear structure and material science. Responsible: M. Kowalska.

-Improved differential pumping system allowing us to maintain liquid samples at higher pressure, and therefore, letting us use different solvents for biological  $\beta$ -NMR studies. Responsible: M. Kowalska.

-Compact magnet with combined  $\beta$ -,  $\gamma$ -, and neutron detector arrays, allowing for nuclear structure studies and later also  $V_{ud}$  high precision determination. Responsible: M. Madurga.

-Reionization cell, based on the  $\beta$ -NMR design from TRIUMF, to ionise beams polarised as neutral atoms. This will allow deflection of the polarized beams to one of the experimental stations, and crucially, better control of the beam optics near the target. Responsible: M. Kowalska.

-Switchyard enabling to direct the ion beam to one of three experimental stations. We will consider implementing fast switching between the 3 setups, which will enhance the efficiency of beam-use at VITO. Responsible: M. Kowalska.

When funds become available for material-science activities, later on we plan to implement an UHV vacuum chamber with temperature control for both magnets and a system to control the energy of the implanted beam between 60 keV down to some eV.

The planned preliminary layout foresees the superconducting magnet in the straight line, the presently-used electromagnet towards REX and  $\beta$ -  $\gamma$ -nautron correlation setup towards the central beamline (Fig 2, bottom) or alternatively parallel to the superconducting magnet.





Fig 2. VITO beamline as used just before LS2 (top) and planned (preliminary) after LS2 (below).

# References:

[Hir05] Y. Hirayama et al., Phys. Lett. B 611, 239 (2005) [Gat06] Gatteschi, D.; Sessoli, R.; Villain, J. Molecular Nanomagnets; Oxford University Press, USA, 2006 [Gin19] W. Gins, KU Leuven, PhD thesis, CERN-THESIS-2018-324 [Gin19b] W. Gins, R. Harding et al, Nucl. Instrum. Methods A 925, 24 (2019) [Har20] R. Harding, J. Croese, S. Pallada et al, to be submitted to Phys Rev X 2020 (available on request) [Har20b] R. Harding, PhD Thesis, U. of York (2020), to be submitted to CDS (available on request) [Jan17] A. Jancso et al., J. Phys. G: Nucl. Part. Phys.44 064003 (2017) [Kie03] Kiefl, R. F.; MacFarlane, et al, Nucl. Instrum. Methods Phys. Res. Sect. B 2003, 204, 682–688. [Kow17] M. Kowalska et al, CERN-INTC-2017-071 ; INTC-P-521 (2017) [Kow17b] M. Kowalska et al, J. Phys. G: Nucl. Part. Phys.44 084005 (2017) [Kow18] M. Kowalska et al., CERN-INTC-2018-019 ; INTC-P-521-ADD-1 (2018) [Kre16] C. Kremer et al., Phys. Rev. Lett. 117, 172503 (2016) [Mad17] M. Madurga et al, CERN-INTC-2017-089 ; INTC-I-204. 2017 [Miy03] H. Miyatake et al., Phys. Rev. C 67, 014306 (2003) [Mor15] Morris, G. D.; MacFarlane, W. A.; Chow, K. H.; Salman, et al, Phys. Rev. Lett. 2004, 93 (15), 157601 [Mor03] Morenzoni, E.; et al., Phys. B 2003, 326 (1–4), 196–204 [Neu08] R. Neugart et al., Phys. Rev. Lett. 101, 132502 (2008) [Neu19] R. Neugart, private communication, unpublished [Ney05] G. Neyens, M. Kowalska, D. Yordanov, et al, Phys. Rev. Lett. 94, 022501 (2005) [Nis17] H. Nishibata et al., Phys.Lett. B 767, 81 (2017) [Now16] F. Nowacki, A. Poves, E. Caurier, and B. Bounthong, Phys. Rev. Lett. 117, 272501 (2016) [Oho02] Ohtomo, et al, H. Y. Nature 2002, 419 (6905), 378–380. [Pro08] Prokscha, T.; Morenzoni, E.; et al., Nucl. Instrum. Methods Phys. Res. Sect. A 2008, 595 (2), 317 [Sal06] Salman, Z.; Kiefl, et al, Phys. Rev. Lett. 2006, 96 (14), 147601–147604. [Sal07] Salman, et al, Nano Lett. 2007, 7 (6), 1551–1555. [Sal07b] Salman, Z.; Wang, et al, Phys. Rev. Lett. 2007, 98 (16), 167001–167004. [Sal12] Salman, Z.; Ofer, O.; Radovic, M.; Hao, H.; et al., Phys. Rev. Lett. 2012, 109 (25), 257207. [Sug19] Sugiyama, J. Hyperfine Interact. 2019, 240 (1), 17 [Tog16] T. Togashi et al., Phys. Rev. Lett. 117, 172502 (2016) [Vel14] P. Velten et al, CERN-INTC-2014-062; P-426, 2014; CERN-INTC-2015-016; CLL-020, 2015

# Appendix

#### **DESCRIPTION OF THE PROPOSED EXPERIMENT**

The experimental setup comprises: *VITO experimental beamline.* Existing parts are covered by VITO safety file. New elements will be added to the safety file before their arrival in the ISOLDE hall.



## **HAZARDS GENERATED BY THE EXPERIMENT**

Hazards named in the document relevant for the fixed VITO installation.

Additional hazards:





### **0**.1 Hazard identification

3.2 Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): *(make a rough estimate of the total power consumption of the additional equipment used in the experiment)*