EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Development of new beams of spin-polarized radioactive nuclei using stable beams from ISOLDE

31.05.2017

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Abstract

We request access to stable beams of K, Cu, and Zn during CERN long shutdown 2 (LS2), in order to develop efficient laser-polarization schemes to be later applied to radioactive isotopes of the above chemical elements. The letter will be studied using the beta-gamma and beta-gamma-neutron correlations and β-NMR techniques. The development will take place at the laser-polarization beamline, where the degree of nuclear spin polarization of stable nuclei will be determined using the fluorescence polarization-checking setup, which is presently under development. The first motivation comes from the nuclear structure studies, where correlation studies will allow determining spins and parities of excited states in neutron-rich Cu and Zn isotopes, and the β-NMR studies will give access to quadrupole moments of neutron-rich K nuclei. The second goal is related to the interaction of metal ions such as K, Cu, or Zn with nucleic acids and proteins, whose investigation can largely profit from the ultra-high sensitivity of the β-NMR technique.
Introduction and motivation

Spin-polarized beams of radioactive nuclei can be of interest for studies in a range of fields, including nuclear structure, fundamental interactions, material science and life sciences. The studies rely on the fact that the emission of \( \beta \) particles from spin-polarised nuclei is anisotropic in space, with their angular distribution given by

\[
W(\theta) = 1 + a_\beta \frac{v}{c} P_1 \cos(\theta),
\]

where \( a_\beta \) is the beta-decay asymmetry parameter depending on the decay scheme of the probe nucleus, \( P_1 \) is the degree of nuclear spin polarization, \( \theta \) is the angle between the direction of particle emission and the magnetic field, and \( v \) is the velocity of the emitted \( \beta \) particle. What is observed directly in the experiment, is the \( \beta \)-decay asymmetry, which is defined as the normalised difference in the number of beta particles detected parallel and antiparallel to the magnetic field direction.

Experimental studies observe the direction of radiation emission (\( \beta \) particles, gamma rays) or apply radiofrequency signals and perform beta-detected Nuclear Magnetic Resonance (NMR) studies. In nuclear physics, angular correlations between the emitted radiation and particles provide information on spins and parities of excited states, as shown e.g. in [Shi14]. \( \beta \)-detected NMR allows measuring unknown electromagnetic moments of nuclei and in combination with hyperfine structure measurements, also deriving spins and parities of nuclear states, see e.g. [Ney05]. In fundamental interaction studies beta-gamma coincidence studies provide detailed information on beta-decay properties and contribute to the determination of the \( V_{ud} \) element of the CKM quark mixing matrix [Vel14]. In material science beta-NMR in solid samples allows studying interfaces, crystal lattices, or semiconductors. Finally, in chemistry and biology the ultrahigh sensitivity of beta-NMR in liquids might help in understanding better the interaction of metal ions with proteins, DNA, and RNA [Jan17].

Once the radioactive beam of interest is available, it is possible to verify and optimise the degree of spin polarization, by observing the \( \beta \)-decay asymmetry. However, this means that any optimisation and development is limited, since radioactive beam at ISOLDE is very competitive and it requires CERN protons. An ongoing development should allow us to develop new polarization schemes and to optimise the degree of achieved polarization by observing fluorescence from stable isotopes. These activities are ideally suited to take place during long breaks in proton availability, such as the upcoming Long Shutdown 2 (LS2). For this reason, we request access to several stable beams from ISOLDE during LS2.

Technique and experimental setup

The development of new spin-polarized beams will take place at the ISOLDE laser-polarization beamline, which was recently commissioned with \( ^{26,28}\text{Na} \) beams. Details of the experimental setup and the results are given in [Kow17]. The nuclei are polarized outside of the host material, in our case via the laser optical pumping. Spin polarization via optical pumping relies on multiple resonant excitations of the ion or atom by circularly polarized laser light, in order to polarize the atomic spins. Due to the hyperfine interaction between the electron spin and the spin of the nucleus in free atoms (or ions), polarization of the atomic spins results also in the polarization of the nuclear spins, with the polarization axis along the laser beam axis. The observed nuclear polarization is reached after the adiabatic decoupling of the spins in a gradually increasing static magnetic field.
The experimental setup devoted to the measurement of the degree of spin polarization $P_I$ is presently in the design phase, with the aim to be ready for first tests at the end of 2017. The technique was already used, e.g. in MPIK-Heilderberg or Florida State University, where nuclear reactions with stable polarized beams were performed, see e.g. [Mem93]. It relies on determining the population of different magnetic substates $m_I$ in a strong magnetic field by exciting them with a probe laser that can be tuned into resonance with one $m_I$ substate at the time. From the populations $n(m_I)$ the degree of spin polarization can be calculated as:

$$P_I = \frac{\langle m_I \rangle}{I} = \frac{\sum n(m_I) \cdot m_I}{\sum n(m_I)}$$

The probing light is circularly polarized, it is parallel to the strong magnetic field and thus parallel to the direction of the spin polarization $P_I$. It triggers $\sigma^+$ or $\sigma^-$ transitions where $m_I$ changes by +1 or -1, respectively, with no change in $m_i$.

The polarization checker which we are presently designing is shown schematically in Fig. 1. It consists of a small vacuum chamber, which can be placed instead of the measurement chamber in the middle of the magnet used for experiments. After passing a set of mirrors, the light from the pump laser propagates parallel to the magnetic field lines, and perpendicular to the ion/atom beam direction. A photomultiplier placed on top of the chamber collected the fluorescence light resulting from the excitation with the probe laser, and a set of mirrors and lenses helps to collect as much light as possible.

Fig. 1 Schematic representation of the fluorescence polarization-checking setup

Due to the strong magnetic field, the different $m_I$ substates are well separated in energy and so are the transitions between different $m_I$ of the ground-state and excited levels, as shown in Fig. 2 on the example of $^7$Li spectra reported in [Men93] (top), as well as in Fig. 3 on the example on our simulations for $\sigma^+$ and $\sigma^-$ probing on unpumped states of $^{23}$Na (therefore all peaks have comparable amplitudes).

The laser frequency is scanned, so that at different moments in time a different transition is in resonance and thus a population of a different $m_I$ state is probed. After performing two sets of measurements, one with $\sigma^+$ light and another with $\sigma^-$ probe light, populations of all ground state $m_I$ substates can be determined (see again Fig. 2 and 3). In this way, we will be able to optimise the degree of nuclear spins polarization for different excitations schemes, different arrangements of the transitional magnetic field, different strengths of the strong magnetic
field, and after inserting different elements into the path of the ion/atom beam, e.g. thin vacuum window, reionization cell, or bending elements.

Fig. 2 Left: Simultaneous optical pumping of $F=1/2$ and $3/2$ states in $^7$Li atomic ground state to $F=3/2$ substate of the first excited state with $\sigma^+ \text{laser polarization}$ (triggering $\Delta m_F=+1$), together with the increased $m_F$ populations of states 1 and 6 (in green). Right: subsequent signal from a fluorescence checker. On top – $\sigma^-$ laser probes states marked 1-3 (transitions with $\Delta m_J=-1$), bottom: $\sigma^+$ laser probes the population of 4-6 states ($\Delta m_J=+1$). Adapted from [Men93].

Fig. 3. Our first simulations of the fluorescence pattern resulting from probing unpumped $^{23}\text{Na}$ (I=3/2) in 1000 G probing field, with probe-laser frequency scanned in the range of +/- 5 GHz and using $\sigma^-$ and $\sigma^+$ transitions. Note that $\sigma^-$ transitions probe the population of the first 4 $m_I$ states whether $\sigma^+$ probes the population of the 4 states.

We are presently modifying the optical pumping code, used e.g. to explain $\beta$-asymmetry spectra of neutron-rich Mg isotopes [Kow08], so that we can predict what fluorescence patterns we will observe with the polarization checker for different degrees of the original polarization (see Fig. 3 for first simulations).
Envisaged studies

During LS2 we would like to produce spin polarization on stable beams of several elements, whose selected radioactive isotopes are interesting for nuclear physics or biological studies.

Beams interesting for nuclear structure are the neutron-rich nuclei of the following elements:

K with the aim to determine the quadrupole moments using $\beta$-NMR in non-cubic hosts ($\beta$-NQR). Several laser spectroscopy experimental campaigns have been dedicated to the measurement of ground state properties of exotic K isotopes with complementary experimental setups (COLLAPS@ISOLDE, BECOLA@MSU) [Pap14, Ros15]. These experiments have resulted in a large amount of information related to the shell evolution in K when neutrons start to fill $d_{3/2}$, $f_{5/2}$, $p_{3/2}$ orbits, due to the monopole interaction between protons and neutrons [Pap14]. Furthermore, investigations with the CRIS experimental setup at ISOLDE are ongoing to check the magicity of the proposed new magic number $N = 32, 34$ in K isotopic chain [Yan16]. The measured magnetic moments up to $^{51}$K ($N = 32$) have been well accounted for by the shell model calculation using SDPF-U and SDPF-NR interactions, except for a few cases where mixing configurations from the particle excitations appeared. Generally, spins, magnetic moments and charge radii measurements already provide a wealth of information to address the questions related to the magicity and single-particle states. However, quadrupole moments are valuable for a more sensitive probing of the cross-shell excitation, proton-neutron correlations, and deformations. In particularly, the quadrupole moment measurement of the $^{51}$K isotope, with a single proton-hole inside of the proposed doubly magic nuclei $^{52}$Ca, will allow the investigation of the magicity of $^{52}$Ca. This measurement could not be realized by laser spectroscopy techniques, because the available laser transition was not sensitive enough to this observable. This makes $\beta$-NQR on polarized K beams a very promising approach.

Cu and measurement of beta-delayed neutron correlations. Several Total Absorption Spectrometry (TAS) experiments in the region close to doubly magic $^{78}$Ni have observed substantial gamma branches from neutron unbound states [Tai15, Spy16]. This is not totally unexpected, as Gamow-Teller transitions in this region populate high spin-parity states, resulting in substantial angular momentum barriers delaying neutron emission. However, the complicated, multi-step process in delayed neutron emission makes it hard to disentangle the factor(s) resulting in gamma competition. The neutron branching ratio is determined by a combination of two matrix elements, the Gamow-Teller strength and the neutron emission spectroscopic factor, with the kinematical factor, the angular momentum barrier [Nym90]. Combining the measurement of neutron and gamma-ray intensities emitted from neutron unbound state allows us to determine the Gamow-Teller strength [Ras17]. However, typically, beta-delayed neutron spectroscopy experiments measure the intensity of neutrons emitted from all $3$ possible allowed spin-parities. The decay of nuclei oriented in a magnetic field will allow us for the first time to unequivocally determine the angular momentum distribution of the neutrons. This, in turn, combined with the Gamow-Teller strength can be used to extract the neutron spectroscopic factors, and thus elucidate the origin of the gamma competition. Copper isotopes, with large neutron branching ratios, and where neutron-gamma competition has already been observed [Ily09], offer a perfect proving ground to first study this phenomenon.

Zn isotopes with $N>50$ with the aim to measure beta-gamma correlations and beta-gamma-neutron correlations around the shell closure $N=50$. The region north east of doubly magic $^{78}$Ni has captured the attention of nuclear physics due to the rich nuclear structure effects
driven by the large neutron-proton imbalance [Ots05]. In the case of Ga isotopes, an inversion of the spin-parity of their ground state and first excited state was observed at COLLAPS [Che10]. However, the single-particle structure beyond the ground state has not been fully determined yet. Determining the spin-parity of excited states typically requires the use of well characterized nuclear reactions, which impose limits in the beam intensity, and therefore are not possible yet in this region of the nuclear chart. The use of beta-gamma coincidences in the decay of laser-polarized Zn isotopes will allow for the first characterization of the spin-parity of proton single-particle excited states in odd-even Ga nuclei, including semi-magic (N=50) $^{81}$Ga. In particular, we will track the evolution of the proton $fp$g orbitals as the neutron g and d orbitals are filled. The changes in their energies are expected to be fully determined by proton-neutron (tensor) part of the nuclear force [Ots10].

Beam interesting for $\beta$-NMR biophysics studies are:

**37K** to investigate the interaction of alkali metal cations with DNA G-quadruplex structures. These DNA structures are formed in nature in nucleic-acid sequences rich in guanine, such as near the ends of the chromosomes, and alkali metal ions are known to play important roles in their formation, stability, and structural polymorphism. NMR can be a powerful tool to investigate such biological systems, however the direct detection of metal-ion NMR signals at physiological concentrations in G-quadruplex liquid samples is difficult [Won05]. $\beta$-NMR can overcome this challenge since it is up to which a billion-times more sensitive than conventional NMR. For more details on the motivation and studies envisaged with the already available spin-polarized Na$^+$ beams, see proposal submitted to the same INTC session [Kow17b].

**58,74,75Cu**, because Cu is present in many enzymes involved in electron transfer and activation of oxygen. As one of the most abundant trace elements in living organisms Cu is essential for many metabolic pathways including cellular respiration, photosynthesis, or removal of reactive oxygen species. Conventional Cu liquid NMR studies are very challenging since both nuclei are quadrupolar (I>1/) with low gyromagnetic ratio, causing low sensitivity and broad NMR lines. Only a very few works on Cu solution-phase NMR are published due to the technical challenges of the experiments and the limited information obtained [Phi86].

$^{75m,77m}$Zn (I=1/2) since Zn is the most abundant trace element in human body and it plays catalytic and structural roles and is involved in regulation of genetic message transcription and translation. Proteins use the chemical properties of Zn$^{2+}$ in order to perform a variety of important cellular functions. Despite possessing a common geometry and similar binding pockets, Zn binding centers are found in proteins involved in very different biological activities [Cri12]. Unfortunately, $^{67}$Zn has a small abundance and large quadrupole moment, making it almost impossible to investigate it in liquid-NMR. Thus the hope in $^{75m,77m}$Zn $\beta$-NMR, since both of these nuclei have spin $\frac{1}{2}$ and thus will not suffer from quadrupolar broadening of the lines.

Summary of the requested beams and the laser excitations which have been already used to perform collinear laser spectroscopy is given below.
Table 1 Requested stable beams and laser wavelength for the pumping and probe lasers

<table>
<thead>
<tr>
<th>Stable beam</th>
<th>Transition</th>
<th>Laser wavelength</th>
<th>reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>39, 41K</td>
<td>Atom: 4s 2S1/2 → 4p 2P1/2, 3/2</td>
<td>766.49 nm</td>
<td>Pap14</td>
</tr>
<tr>
<td>63,65Cu</td>
<td>Atom: 4s 2S1/2 → 4p 2P1/2, 3/2</td>
<td>327.4 nm</td>
<td>Vin10</td>
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<tr>
<td>67Zn</td>
<td>Atom: 4s4p 3P2 -&gt; 4s5s 3S1</td>
<td>481.2 nm</td>
<td>Wra17</td>
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</tbody>
</table>

**Beamtime request**

Here we request several days with each of the above beams, produced with GPS or HRS targets and spaced by least with several days in between each study to allow the change of the laser system required for each laser frequency.

**References:**


[Kow17b] M. Kowalska et al., proposal to the INTC committee, CERN-INTC-2017-071 (INTC-P-521)


[Yan16] X.F.Yang et al, Proposal to INTC, CERN-INTC-2016-008 ; INTC-P-458(2016)


Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises:

<table>
<thead>
<tr>
<th>Part of the experiment</th>
<th>Availability</th>
<th>Design and manufacturing</th>
</tr>
</thead>
<tbody>
<tr>
<td>[permanent laser-polarization setup]</td>
<td>☒ Existing</td>
<td>☐ To be used without any modification ☒ To be modified</td>
</tr>
<tr>
<td>☐ New</td>
<td>☒ Standard equipment supplied by a manufacturer ☒ CERN/collaboration responsible for the design and/or manufacturing</td>
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<tr>
<td>[beta-NMR setup]</td>
<td>☒ Existing</td>
<td>☐ To be used without any modification ☒ To be modified</td>
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<td>☐ New</td>
<td>☒ Standard equipment supplied by a manufacturer ☒ CERN/collaboration responsible for the design and/or manufacturing</td>
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<tr>
<td>[COLLAPS laser system]</td>
<td>☒ Existing</td>
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<td>☒ Standard equipment supplied by a manufacturer ☒ CERN/collaboration responsible for the design and/or manufacturing</td>
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</table>

[insert lines if needed]

HAZARDS GENERATED BY THE EXPERIMENT

Hazards named in the document relevant for the fixed laser-polarization setup:

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<th>Hazards</th>
<th>Charge-exchange and post-acceleration chamber</th>
<th>Optical detection and optical pumping section</th>
<th>ISOLDE-standard focussing triplet</th>
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<td>[pressure][Bar], [volume][l]</td>
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<tr>
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<tr>
<td>Beam intensity</td>
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<td>Beam energy</td>
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<tr>
<td>Gases</td>
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</table>
### Calibration sources:
- Open source
- Sealed source
- Isotope
- Activity

### Use of activated material:
- Description
- Dose rate on contact and in 10 cm distance: [dose][mSV]
- Isotope
- Activity

### Non-ionizing radiation
- Laser
- UV light
- Microwaves (300MHz-30 GHz)
- Radiofrequency (1-300MHz)

### Chemical
- Toxic
- Harmful
- CMR (carcinogens, mutagens and substances toxic to reproduction)
- Corrosive
- Irritant
- Flammable
- Oxidizing
- Explosiveness
- Asphyxiant
- Dangerous for the environment

### Mechanical
- Physical impact or mechanical energy (moving parts)
- Mechanical properties (Sharp, rough, slippery)
- Vibration
- Vehicles and Means of Transport

### Noise
- Frequency
- Intensity

### Physical
- Confined spaces
- High workplaces
- Access to high workplaces
- Obstructions in passageways
- Manual handling
- Poor ergonomics

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**0.1 Hazard identification**

**3.2 Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above):**