EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Collinear resonance ionization of neutron-deficient indium: closing up on $N = 50$

October 2, 2024

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CERN-INTC-2024-072 / INTC-P-724
02/10/2024

Abstract: We propose to study the nuclear ground-state properties of the neutron-deficient indium isotopes ^{99,100}In, in the direct vicinity of ^{100}Sn, using Collinear Resonance Ionization Spectroscopy (CRIS). These measurements will yield results on magnetic dipole and electric quadrupole moments, the nuclear spins, as well as changes in the mean-square charge radii. This allows the study of the evolution of nuclear structure with a single proton hole in the $Z = 50$ shell closure and probes the doubly magic character at $N = 50$. Such experimental results are vital to benchmark state-of-the-art nuclear models and will yield new input for nuclear theory to improve their predictive power in this key region of the nuclear chart.

Summary of requested shifts: 21 (+3 stable) shifts in one run

1 Physics motivation and previous experiments

Numerous experimental and theoretical efforts have recently focused on investigating the region around ^{100}Sn, the heaviest self-conjugate nucleus featuring a magic number of $Z = 50$ protons and $N = 50$ neutrons [1, 2]. Nuclei in this region form an excellent laboratory to test effects of the neutron-proton interaction and the evolution of nuclear structure around shell closures.
To date, theoretical calculations remain challenging due to the large configuration space. Recently, in part due to an enhanced computing power and developments in the models, frameworks such as *ab initio*-theory, large-scale shell model (LSSM) and nuclear density functional theory (DFT) have been able to provide predictions of such complex nuclei [3–7]. Probing the single-particle behaviour and correlation effects experimentally allows benchmarking nuclear models to improve their predictive power towards yet more complex systems presently not in reach.
Experimental studies applying decay spectroscopy and Coulomb excitation of light Sn isotopes previously focused on confirming the double magicity and probing the changes in nuclear configuration in this region [8, 9]. Recent efforts, with a wide focus from experiments at ISOLDE, allowed for high-precision mass measurements [10–15] and laser spectroscopy measurements [3, 16, 17] (further studies in publication process) in this region. However, as of today, production yields hamper such experimental studies leading to scarce information on nuclear ground-state properties.
Studies in the neighboring indium isotopic chain ($Z = 49$), with an open proton hole compared to Sn, provide an ideal framework to study structural effects while approaching the $N = 50$ and $N = 82$ shell closures. From a simple shell-model picture, the ground state of the even-odd In isotopes should be dominated by the proton hole in the $\pi_{9/2}$-orbital. In addition, low-lying isomeric states are present due to proton excitation from the $\pi_{1/2}$-orbital into the $\pi_{9/2}$-orbital leaving a hole in the former. Both, the ground and isomeric states were recently studied along the isotopic chain [3, 18]. In addition, long-lived high-spin isomers in neutron-rich In were identified early on [19] and probed by laser spectroscopy at CRIS.
Using laser spectroscopy, the In isotopic chain was investigated from ^{101}In [3], only two neutrons away from the $N = 50$ shell closure, to ^{131}In at the $N = 82$ closure [18]. These studies allowed the extraction of nuclear moments μ, Q_s, changes in mean-square charge radii $\delta\langle r^2\rangle$, and an unambiguous identification of previously unknown nuclear spins I [3, 18]. Results on the nuclear moments around $N = 82$ challenged the common agreement of the indium chain forming a textbook example for single-particle behavior, dominated by the single unpaired proton. The sudden increase in the magnetic dipole moment at the shell closure, in comparison to the smooth stable trend observed along the isotopic chain as shown in Fig. 1, indicates a pronounced single-particle nature close to the Schmidt single-particle value [18]. On the contrary, the electric quadrupole moments were found to gradually decrease from mid-shell to the shell closures showing the reduction in polarization towards the single proton-hole value.
Accompanying nuclear model developments were made, using DFT (Hartree–Fock (HF) and Hartree–Fock–Bogoliubov (HFB)) and *ab-initio* (VS-IMSRG) frameworks. The new experimental results challenged the different DFT calculations to be tested, considering the inclusion of pairing correlations and time-odd fields. With different combinations of these effects, it was shown that a HF based mean-field description with the inclusion of time-odd fields is required to reproduce the data on magnetic moments of the ground state. The *ab-initio* results fail to

describe the absolute variations observed in electromagnetic properties, which is expected to be improved by the incorporation of two-body currents [20] but requires more experimental data to constrain new developments in the model.

The previously unexplained trend in decreasing magnetic moments of the $1/2^-$ isomeric states towards $N = 82$ was well captured by the new *ab-initio* calculations, indicating a mixed configuration for these states. These findings showed the need for further developments of the DFT calculations, which did not describe the observed trend in the isomeric states, towards the inclusion of other core configurations [18].

Mass measurements venturing into the region around ^{99}In [12] and measuring its $1/2^-$ isomer ^{99m}In [15], created another test case for nuclear theory models, in which an inconsistent description of excitation energies and nuclear moments was found. While further DFT-HF calculations predict a similar kink in the trend of magnetic dipole moments at $N = 50$, VS-IMSRG *ab-initio* and LSSM calculations result in a much smoother trend and no sudden increase at the shell closure, see Fig. 1. This is contrary to the results of a similar increase at $N = 82$ as for the DFT predictions. The LSSM predictions agree with the DFT results for quadrupole moments in lighter In nuclei [15], for which the *ab-initio* framework underestimates the trend and absolute variations down to ^{101}In [3]. This discrepancy in the available model predictions of nuclear moments in the vicinity of $N = 50$ requires new experimental data to test the models and to investigate the single-particle behavior around this shell closure.

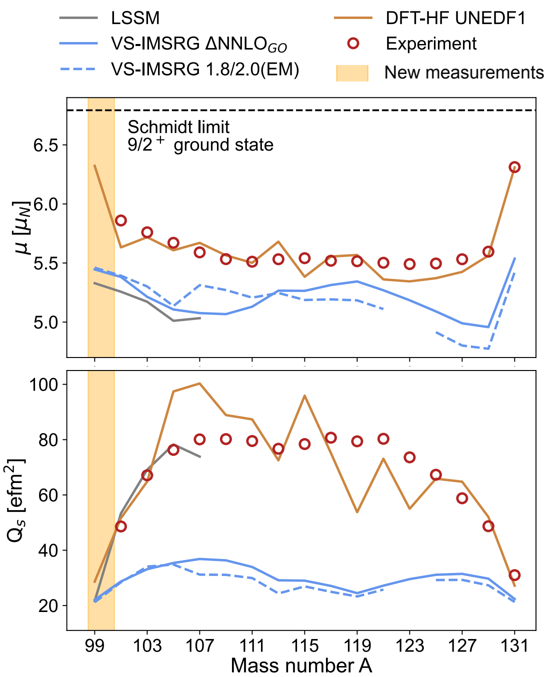

Figure 1: Experimental nuclear moments between $^{101-131}$In and predictions of nuclear models using DFT, *ab-initio*, and LSSM frameworks. The range of isotopes targeted in this proposal are highlighted [3, 15, 18].

Furthermore, the odd-odd-isotope ^{100}In, with a single neutron and one-proton hole from ^{100}Sn and a predicted spin of 6^+, is an ideal proxy for nuclear theory and available calculations in this region. The nuclear moments as observables are very sensitive to proton-neutron correlations. Close to both the shell closures, the purity of the states can be investigated, and a change in correlation be understood as a probe for the magicity of $N = 50$ and $Z = 50$. A similar study at CRIS on ^{78}Cu, near ^{78}Ni, in combination with nuclear model predictions, showed strong evidence for the doubly magic character of $N = 50$ for $Z = 28$ [21].

Further information on trends in mean-square charge radii around the shell closure will additionally allow the testing of predictions made by several DFT frameworks in Ref. [3], in which a sudden change in slope around $N = 54$ was calculated, contrary to a smooth trend from *ab-initio* results. The present experimental data does not allow this discrepancy to be investigated yet.

In summary, we propose to study neutron-deficient indium isotopes down to the $N = 50$ shell closure. The measurements of the hyperfine structures of ^{100}In and ^{99}In will offer an experimental insight into nuclear magnetic dipole and electric quadrupole moments, which are important to benchmark new available calculations from state-of-the-art nuclear models. Pinning down the

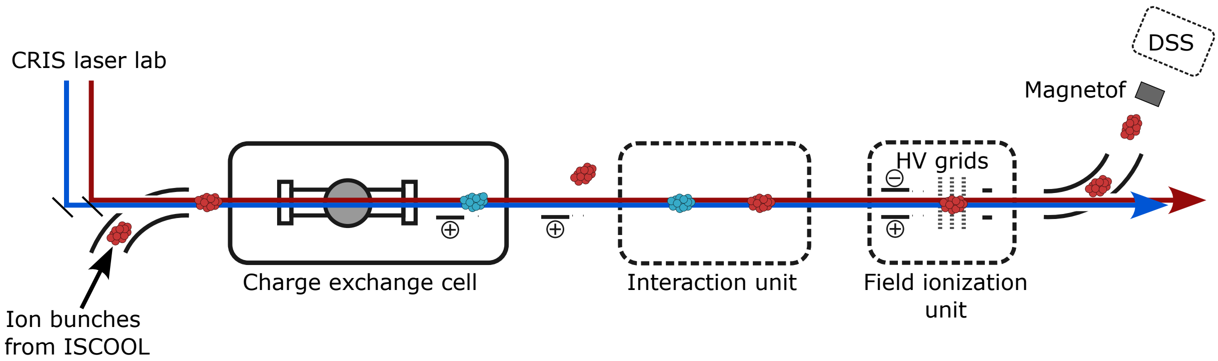

Figure 2: Schematic diagram of the CRIS experiment that will be used to measure the neutron-deficient In isotopes. The newly commissioned field ionization unit and upgraded DSS are integrated and available for upcoming experiments.

change in mean-square charge radius, by extracting the centroid of observed hyperfine structures, furthermore allows to investigate the decreasing trend in nuclear size to compare to model calculations with a sensitivity to changes in deformation close to $N = 50$ [3].

2 Experimental method

The proposed experiment will be performed at the CRIS setup which is schematically shown in Fig. 2. The CRIS technique allows for high-precision laser spectroscopy with a resolution close to the natural linewidth of the transition and high selectivity via multi-step resonance ionization. The ion beam of the required isotope, with a kinetic energy of about 30 keV, is mass separated by HRS, cooled and bunched in ISCOOL, and guided into the CRIS beamline. Positively charged ions are neutralized upstream in the charge exchange cell via interaction with a hot alkali vapor, in this case sodium for an enhanced charge exchange cross section. Remaining ions after the charge exchange cell are deflected and the neutral atoms proceed towards the interaction region, in which an ultra-high vacuum of around 10^{-10} mbar is maintained to reduce collisional ionization processes with the remaining gas atoms.
The neutral atom bunches are overlapped with pulsed laser beams following the respective RIS scheme to allow for resonant re-ionization, probing the first-step atomic transition. Laser-created ions follow the 34 °-bender into the detection section downstream of the beamline. Here, various detection methods are available, such as single-ion detection using a MagneTOF detector or beta-decay tagging in the CRIS DSS (decay spectroscopy station).

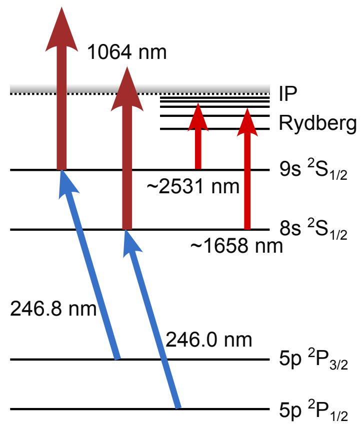

Figure 3: Two-step resonant ionization schemes to be used for the extraction of nuclear moments in indium [3, 18, 22]. For ionization, either a non-resonant 1064 nm step or a resonant excitation to Rydberg states followed by field ionization can be applied.

Key for the successful application is a RIS scheme which allows sensitive probing of nuclear ground-state properties and a reduction of laser-induced background, resulting from interactions with intense, isobaric contaminants. To minimize the background associated with a high-power non-resonant step, a scheme featuring a 1064 nm non-resonant step is typically chosen. The schemes for the proposed measurements are shown in Fig. 3. Two close-lying first-step transitions will be probed, which start from the 5p $^2P_{1/2}$ ground state and the metastable state $^2P_{3/2}$. Both states are populated efficiently in the charge exchange process to 37 % and 57 %, respectively [23]. The transition from the 5p $^2P_{1/2}$ to the 8s $^2S_{1/2}$ atomic level at 246.0 nm allows sensitive probing of the magnetic dipole moment, while a second transition from 5p $^2P_{3/2}$ to the 9s $^2S_{1/2}$ level at 246.8 nm will be studied for the extraction of the electric quadrupole moment. With the existing laser setup at CRIS, these two schemes were previously realized for measurements in indium [3, 18] and a change between both schemes can be performed within minutes.

Recent upgrades in the CRIS setup were made for the study of more exotic species, for which previous studies were hampered by orders of magnitude higher amounts of contamination. These methodological advances include the extension of the CRIS DSS towards an increased solid angle and higher efficiency, comparable to single ion counting using the MagneTOF, and the integration of a field ionization unit [22]. Field ionization in resonance ionization laser spectroscopy was shown early on to be a versatile tool for the suppression of collisional background by orders of magnitude [24]. In the design of the new CRIS field ionization unit, the final interaction region is greatly reduced to a few cm, which reduces the collisional background contribution. By deflecting collisional ions produced before the field ionization volume, a background suppression of more than a one order of magnitude can be achieved, considering the remaining post-ionization section and vacuum in the beamline sections [22]. With an additional post-acceleration and energy selection in the second CRIS bender, another order of magnitude in background suppression is possible. Field ionization at CRIS was previously demonstrated in successful off-line studies on indium [22], proving indium to be a suitable case for field ionization, and during on-line studies on stable potassium beam this year.

3 Yields and shift request

In total, 21 shifts with protons are requested, using a LaC_x target in combination with the RILIS ion source. In previous mass measurements of indium isotopes down to ^{99}In and its isomeric state [15], the production of these isotopes around $N = 50$ at ISOLDE was successfully demonstrated and yield measurements by the ISOLTRAP experiment were performed in comparison to expected yields. Over the duration of the experiment, no drop in yield for the exotic cases was observed. A summary of the measured yields and requested shifts per isotope is shown in Tab. 1.

With the CRIS technique, a sensitivity down to 20 ions/s was demonstrated with the setup in 2017 which allowed measurements on ^{78}Cu in about one shift [25]. Since then, the sensitivity of the setup was enhanced by a re-design of the end of the CRIS beamline in 2022 for a better ion transport and an improved vacuum in the interaction region to 10^{-10} mbar, reducing contributions of collisional background. In addition, the new DSS and the field ionization unit in the CRIS beamline will allow for an enhanced reduction of contaminant species and background from stable or longer-lived isobars.

Previous studies with the CRIS technique have shown successful experiments with more than 4 orders of magnitude higher beam contamination, due to the high selectivity of the technique. During an earlier run for indium studies near this region, a mistaken molybdenum contamination, stemming from a contained sample for target tests prior to the run, mainly hampered the study of ^{100}In [26]. This contamination can be avoided in the target production process. The main contaminants identified by ISOLTRAP for the ^{99,100}In beams are ^{80,81}SrF with a 1-2.5 orders of magnitude increased production rate compared to the isotopes of interest [15]. The half-lives of these contaminants are about 3 orders of magnitude increased (106.3 m and 22.3 m), which allows for a sensitive detection via decay of the isotope of interest with a 2-3 orders of magnitude suppressed contribution from the long-lived contaminants.

It was identified in studies on ^{101}In that resonant background in CRIS from molecular species, in similar order as the signal, was arising from the two-step RIS scheme using the high-power 1064 nm step. By applying field ionization, the high-power laser becomes obsolete. No further laser-induced background was previously identified as originating from the single, first-step laser. Furthermore, the remaining background from collisional ionization can be reduced further by an order of magnitude using the field ionization technique. The DSS tape station additionally allows to reduce build up of long-lived radioactivity on the detector, which was found at the end of a previous experiment to a rate comparable to the ^{101}In signal rate [26]. This combination of both, field ionization (suppressing laser related background and reducing collisional background) and DSS (reducing by 3 orders of magnitude the longer-lived ^{80}SrF background) yields a decisive advancement which will enable the successful measurement of ^{99}In.

The requested shifts are estimated from previous CRIS measurements on ^{78}Cu [21, 25], ^{52}K [27] and ^{131}In [18], assuming a common overall transport efficiency of 30 % and laser efficiency of about 10 %. The neutralization efficiencies of 37 % and 57 %, respectively, are taken into account. A detailed description of efficiencies during previous indium laser spectroscopy campaigns can be found in Ref. [26]. Frequent reference measurements, included with 3 shifts, have to be performed to account for any systematic effects and drifts of the ISCOOL platform potential. Additionally, 3 shifts of stable beam are requested for stable beam tuning and initial setup of the experiment.

Summary of requested shifts: 21 (+3 stable) shifts in one run

Isotope	$T_{1/2}$	I^π	Ions/2 μC	Shifts	New measurements
$^{112-122}$In	> 1 s	-	$\geq 10^4$	3	Reference
^{100}In	5.65(6) s	(6^+)	320	3	I, μ, Q and $\delta\langle r^2\rangle$
^{99}In	3.1(2) s	$(9/2^+)$	5	15	I, μ, Q and $\delta\langle r^2\rangle$
Total				21	
Setup				3	

Table 1: Proposed isotopes to be studied, known half-lives, spins and planned measurements. The given yields were measured and numbers extracted by ISOLTRAP [15]. The calculated shifts required per isotope with a LaC_x target in combination with the RILIS ion source are given for an average proton beam current of 2 μA.

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4 Details for the Technical Advisory Committee

4.1 General information

Describe the setup which will be used for the measurement. If necessary, copy the list for each setup used.

☒ Permanent ISOLDE setup: CRIS

- ☒ To be used without any modification
- ☐ To be modified: *Short description of required modifications.*

☐ Travelling setup (*Contact the ISOLDE physics coordinator with details.*)

- ☐ Existing setup, used previously at ISOLDE: *Specify name and IS-number(s)*
- ☐ Existing setup, not yet used at ISOLDE: *Short description*
- ☐ New setup: *Short description*

4.2 Beam production

For any inquiries related to this matter, reach out to the target team and/or RILIS (please do not wait until the last minute!). For Letters of Intent focusing on element (or isotope) specific beam development, this section can be filled in more loosely.

- Requested beams:

Isotope	Production yield in focal point of the separator (/μC)	Minimum required rate at experiment (pps)	$t_{1/2}$
$^{112-122}$In	$\geq 10^4$	$\geq 10^4$	$> 1\,$s
^{100}In	160	200	5.65(6) s
^{99}In	3	3	3.1(2) s

- Full reference of yield information (Yields from ISOLTRAP collaboration, see Ref. [15], experiment IS661)
- Target - ion source combination: LaC_x target with RILIS ion source
- RILIS? (*Yes for element In*)
  - ☒ Special requirements: Possibly a second first step laser to increase yield by an additional 20-30 %
- Additional features?
  - ☐ Neutron converter: No
  - ☐ Other: No
- Expected contaminants: ^{80}SrF, ^{81}SrF with yields of 500 ions /μC and 1000 ions /μC, respectively (before CRIS).
- Acceptable level of contaminants: limited by ISCOOL overfilling
- Can the experiment accept molecular beams? No, atomic beams are required.

- Are there any potential synergies (same element/isotope) with other proposals and LOIs that you are aware of? CRIS is having another proposal already accepted for n-rich In (IS723), which is planned to be requested for the running period of 2025. RILIS would use the same scheme, but the target would be different (UC_x).

4.3 HIE-ISOLDE

For any inquiries related to this matter, reach out to the ISOLDE machine supervisors (please do not wait until the last minute!).

- HIE ISOLDE Energy: *(MeV/u); (exact energy or acceptable energy range)*
  - ☐ Precise energy determination required
  - ☐ Requires stable beam from REX-EBIS for calibration/setup? *Isotope?*
- REX-EBIS timing
  - ☐ Slow extraction
  - ☐ Other timing requests
- Which beam diagnostics are available in the setup?
- What is the vacuum level achievable in your setup?

4.4 Shift breakdown

The beam request only includes the shifts requiring radioactive beam, but, for practical purposes, an overview of all the shifts is requested here. Don't forget to include:

- Isotopes/isomers for which the yield need to be determined
- Shifts requiring stable beam (indicate which isotopes, if important) for setup, calibration, etc. Also include if stable beam from the REX-EBIS is required.

An example can be found below, please adapt to your needs. Copy the table if the beam time request is split over several runs.

Summary of requested shifts:

With protons	Requested shifts
Reference scans $^{112-122}$In	3
Data taking ^{100}In	3
Data taking ^{99}In	15
Without protons	Requested shifts
CRIS setup with Stable In beam	3

4.5 Health, Safety and Environmental aspects

4.5.1 Radiation Protection

- If radioactive sources are required:
  - Purpose? Initial tests, efficiency calibration
  - Isotopic composition? Closed beta sources, for instance ^{90}Sr
  - Activity? few kBq
  - Sealed/unsealed? Sealed