EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

A new approach for nuclear spin determination via in-source resonance ionisation spectroscopy and yield measurements of neutron deficient radium isotopes

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Abstract: This Letter of Intent requests on-line beam-time to validate a new approach for extracting nuclear spins from in-source resonance ionisation spectroscopy by harnessing optical pumping effects within the laser-atom interaction region. This approach should be applicable for multiple elements, of these Ra is the ideal test case as optical pumping effects have already been observed and there is an opportunity for a broader in-source spectroscopy campaign on Ra. This study would extract isotope shifts, nuclear spins and electromagnetic moments of both neutron rich and neutron deficient Ra.

Summary of requested shifts: [6] shifts, (split into [1] run over [1] year)

1 Introduction

In-Source Resonance Ionisation Spectroscopy (IS-RIS) has been applied at ISOLDE across a broad range of isotopic chains in the heavy region of the nuclear chart. These experiments have determined charge radii and electromagnetic moments of exotic isotopes of Cu, Ag, Au, Hg, Tl, Pb, Bi, Po and At. Despite this success, conclusively characterising the nuclear spins of these isotopes has proven to be more challenging, either due to the number of hyperfine structure (hfs) components of the atomic line under study, a lack of resolution, or a combination of the two. The observation of optical pumping within the hot cavity environment [1] presents an opportunity to access favourable atomic transitions that enhance the applicability of IS-RIS for the determination of nuclear spins. This approach could potentially be employed to determine the nuclear spins of Cd, Hg, Te, Yb, Po and Ra isotopes.

Ra is the ideal test case to validate the technique due to the previous incorporation of optical pumping into a Ra ionisation scheme [1] and the potential to apply it within a future IS-RIS campaign to study exotic Ra isotopes. In addition to determining the nuclear spins, such an experiment would enable the extraction of magnetic dipole moments, electric quadrupole moments and isotope shifts. These results would provide information on the nuclear configurations of the odd isotopes and the point at which Ra isotopes depart from sphericity below N=126. Previous attempts to investigate the isotopes below ^{208}Ra by the CRIS experiment suffered from lower than anticipated yields at the neutron deficient end of the Ra isotopic chain. IS-RIS offers comparatively enhanced sensitivity, managing rates as low as 0.01 ions/s [2], but an assessment of the yields is still required to estimate the accessible isotopes below ^{208}Ra. In addition, the upcoming long shutdown offers an opportunity for development to address issues related to the extraction of these isotopes, should it be required.

2 A new approach for nuclear spin determination via IS-RIS

In the absence of significant admixtures, the hfs visible in P_0 to S_1 transitions is solely the result of the nuclear magnetic dipole moment μ as the spherical symmetry of the S_1 state does not result in an electric field gradient at the nucleus. The positions of the three hfs components are determined by the atomic angular momentum J, the nuclear spin I and the hyperfine coupling constant A. This principle was harnessed to successfully determine the spin of ^{193}Hg using pre-laser techniques [3]. For certain elements, such states can be accessed via optical pumping. An illustrative hfs for such a transition is shown in Fig.1, with the three components labelled r_{F1}, r_{F2} and r_{F3}.

In order to unambiguously extract I, A and its associated uncertainty can be eliminated by dividing the distance between r_{F1} and r_{F2} by the distance between r_{F2} and r_{F3}. This simplifies to an equation dependent only on the known J and the unknown I. For S_1

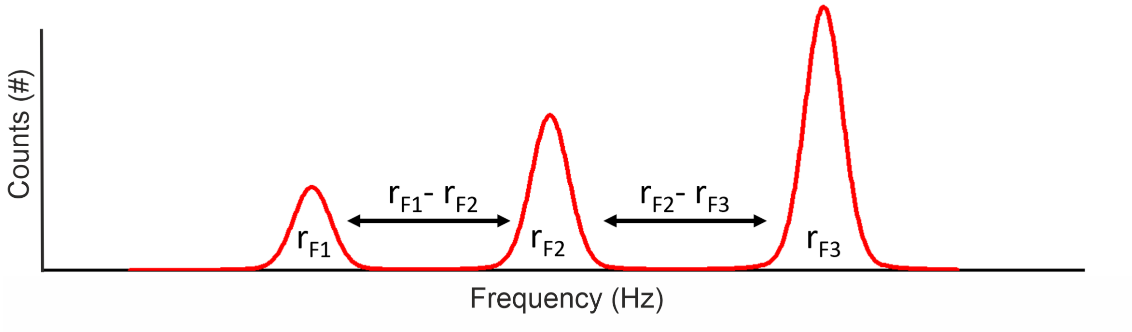

Figure 1: Illustrative hyperfine structure of a P_0 to S_1 transition.

(J=1) states, this equates to

$$\frac{r_{F1} - r_{F2}}{r_{F2} - r_{F3}} = \frac{I}{I+1} = x, \tag{1}$$

where there is a unique x value for each I. The uncertainty on an experimentally determined x value is related only to that associated with the distance between individual hfs peaks and the magnitude of the hfs splitting. The repeatability of the frequency difference between peaks in individual laser scans was analysed for a recent IS-RIS experiment studying Bi and reported to have a standard deviation of 20 MHz [4].

3 Motivation for studying the Ra isotopic chain

3.1 Validating a new approach for determining nuclear spins via IS-RIS

Ra is well suited for characterising optical pumping in a hot cavity surface ion source.

- The existing {714 nm|784 nm|993 nm^{decay}|615 nm|615 nm} ionisation scheme was found to have the highest efficiency of those tested on-line at ISOLDE [1]. This ionisation scheme is depicted in Figure 2b), together with the {714 nm|784 nm|731 nm^{decay}|731 nm|558 nm} ionisation scheme proposed for this study.

- The A-factor associated with the 7s8s $^3S_1$ state corresponds to a sufficiently large hfs to enable the extraction of nuclear spins for Ra isotopes. This is depicted in Figure 2a), where the shaded areas represent the regions within three standard deviations of x considering the 20 MHz standard deviation for determining the distance between individual hfs components and the magnitude of the hyperfine splitting based on magnetic moments from [5] and [6], and the calculated hyperfine coupling constants for ^{223}Ra from [7]. The x values and the associated shaded regions are shown for multiple spins to highlight the theoretical capability of the approach to discriminate between them. This LOI seeks to validate these calculations.

- The 34(15) s [8] lifetime of the 7s7p $^3P^\circ_0$ atomic level is effectively infinite compared to the 100 μs duty cycle of the RILIS lasers. Consequently, it is an ideal candidate for investigating the impact of target and ion source parameters on the efficiency of ionisation schemes incorporating optical pumping. Any depopulation within the duty cycle will be the result of collisional de-excitation or effusion from the laser atom interaction region.

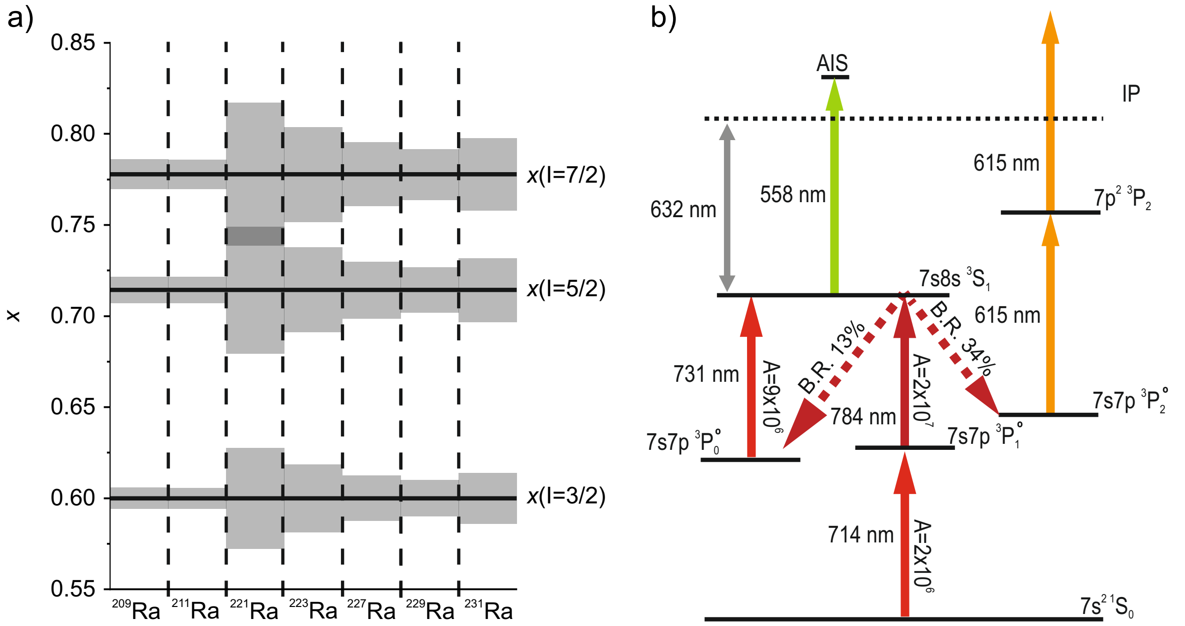

Figure 2: a) x values for the hfs of the 7s8s $^3S_1$ state of atomic Ra, with $I = 3/2$, $5/2$ and $7/2$. The shaded areas represent the regions estimated to be within three standard deviations of experimentally determined x values, based on a σ=20 MHz for determining the distance between hfs peaks and the magnitude of the hyperfine splitting. Further details in the text. b) Laser ionisation schemes employed for the study of Ra isotopes. Further details in the text.

The variation in the size of the shaded areas in Figure 2a) is driven by differences in the magnitude of the magnetic moments of the isotopes listed. The smaller the magnetic moment, the less pronounced the hyperfine splitting and consequently the larger the uncertainty.

3.2 Proposed programme for studying exotic Ra isotopes via IS-RIS and existing data

A successful validation of the technique will open the door to its application across multiple chains of isotopes including Ra. Atomic Ra isotopes were last studied at the CRIS experiment in 2016. An initial campaign to study neutron deficient isotopes [9] was diverted due to lower than anticipated yields. The target performed well at the neutron rich end of the isotope chain, enabling the measurement of mean square charge radii up to ^{233}Ra and magnetic dipole and electric quadrupole moments up to ^{231}Ra [6]. Following the successful completion of the proposed work, we plan on extending the measurements at both the neutron deficient and neutron rich ends of the isotopic chain. The scope of the proposed programme is listed in Table 1.

Table 1: Proposed programme of measurements subject to the successful completion of this LOI. Ticks represent new measurements, $*$ indicates a measurement with improved resolution. The extraction of Q_s for ^{233}Ra will not be possible in the case of $I = 1/2$.

Isotope	$\delta\nu$	I	μ	Q_s
^{205}Ra	✓	✓	✓	✓
^{206}Ra	✓	-	-	-
^{207}Ra	✓	✓	✓	✓
^{231}Ra	Known	✓	Known	Known
^{233}Ra	$*$	✓	✓	(✓)
^{234}Ra	✓	-	-	-

On the neutron deficient side, the motivations of the successful INTC-P-413 proposal [9] are still valid. Isotope shift measurements would probe the point at which neutron deficient Ra isotopes depart from sphericity below N=126, with a significant increase in deformation predicted between $^{206-205}$Ra [10] in contradiction to other theoretical nuclear models in the region [11, 12, 13, 14, 15]. The proposed ionisation scheme offers the possibility of isomer selectivity for ^{205,207}Ra via both the 714 nm and the 784 nm transitions. This would enable the ground state or the high spin isomer to be independently studied via the 731 nm transition. Extraction of the magnetic moments from the 714 nm and the 784 nm transitions would provide information on the nuclear configurations of odd-A Ra isotopes. While the 714 nm transition would also provide information on the spectroscopic quadrupole moments.

For the neutron rich isotopes, the isotope shift of ^{233}Ra could be measured with increased resolution and the isotope shift of ^{234}Ra measured for the first time. The nuclear spins of ^{231,233}Ra could be unambiguously determined, and the magnetic moment of ^{233}Ra extracted. Extracting Q_s from ^{233}Ra would enable the trend of the deformation with increasing N to be established. The spin of ^{233}Ra was tentatively assigned as $I = 1/2$ based on systematics in the region [16] and theoretical calculations [17]. If confirmed, this would prevent the extraction of Q_s. The predicted increase in deformation with increasing N may be inferred though from an increase in the magnitude of the odd-even staggering in the mean square charge radii [6, 18]. This could be identified from a higher resolution measurement of the isotope shift of ^{233}Ra and a measurement of the isotope shift of ^{234}Ra.

4 Additional applications of optical pumping assisted resonance ionisation spectroscopy

4.1 Atomic lifetime measurements

Optical pumping in the hot cavity environment enables the population of metastable atomic states, the lifetimes of which can be measured by delaying the laser exciting from this state with respect to the previous excitation steps. The range of atomic lifetimes that this approach can be employed to measure will be determined as a by-product of the planned investigations. Delaying the 731 nm and 558 nm transitions with respect to the 714 nm and 784 nm transitions will reveal the loss rates of the $7s7p^3P^\circ_0$ state due to non-radiative de-excitation mechanisms such as collisions with the walls of the hot cavity. The lifetime of this state makes it the perfect candidate for such studies.

4.2 Enhanced isotope/isomer selectivity

The incorporation of optical pumping into RILIS ionisation schemes offers the possibility of enhanced isomer selectivity. This can be seen in Figure 2b), where the addition of the 731 nm step offers a third transition that is sensitive to both isomer and isotope shifts and a simple three component hfs enabling isotope/isomer specific ionisation. This capability has proven to be useful beyond IS-RIS experiments where a specific isomer is of interest [19, 20].

4.3 Off-line studies during LS3

The work will guide related development efforts during the upcoming long shutdown. If the technique is validated on-line, then the focus will be on developing RILIS ionisation schemes to broaden the applicability of the technique to other elements. Should issues be identified, the shutdown would also provide the time for them to be addressed.

5 Beam time request and specific plans for this LOI

We request 6 shifts to validate the new approach for determining nuclear spins via IS-RIS using Ra isotopes, and to assess the yields at the neutron deficient end of the chain. The proposed measurements and shift plan for the LOI are listed in Table 2.

After establishing a baseline for optical pumping performance with the known {714 nm|784 nm|993 nm^{decay}|615 nm|615 nm} ionisation scheme and ^{226}Ra, the new {714 nm|784 nm|731 nm^{decay}|731 nm|558 nm} ionisation scheme will be established and optimised. The temperature of the ion source will also be optimised at this point if required. ^{223}Ra will be used to optimise the resolution as it represents the most challenging of the easily available Ra isotopes for resolving the hfs of the 7s7p $^3P^\circ_0$ to 7s8s $^3S_1$, 731 nm, transition. Following this, there will be scans of both the 731 nm and 784 nm transitions for ^{227,231}Ra to test the approach with broader hfs and establish the

Table 2: Proposed programme of measurements subject to the successful completion of this LOI.

Time required	Measurement plan
0.5 shifts	Establish a baseline for optical pumping performance with ^{226}Ra and the known {714 nm|784 nm|993 nm^{decay}|615 nm|615 nm} ionisation scheme.
1.5 shifts	Verify and optimise the {714 nm|784 nm|731 nm^{decay}|731 nm|558 nm} ionisation scheme with ^{223}Ra.
2 shifts	Two scans of the 784 nm transition and three scans of the 731 nm transition for ^{227,231}Ra. ^{226}Ra used as a reference.
1 shift	Investigations of the sensitivity of optical pumping to varying operational conditions using ^{226}Ra.
1 shift	Yield checks for ^{206}Ra with IDS.

trend of the isotope shifts for the transitions in preparation for the full experiment. Next we will explore the sensitivity of optical pumping in the hot cavity to varying operational conditions. Measurements will be taken with and without the protons impinging on the target and at a range of target and ion source temperatures. The losses versus time delay of the 784 nm and 731 nm transitions and the relative efficiencies of the {714 nm|784 nm|558 nm} and {714 nm|784 nm|731 nm^{decay}|731 nm|558 nm} ionisation schemes will be recorded.

Finally, the yield of ^{206}Ra will be measured at the ISOLDE Decay Station with α-detection to provide a data point for the extrapolation of the yields of neutron deficient Ra isotopes. Ra has an effective ionisation potential of 5.14 eV at 2000°C, consequently a portion of the Ra atoms will be surface ionised [1]. This will enable the yield measurement to take place without the prior determination of the isotope shifts for ^{206}Ra. The expected laser ionised yield will be determined using laser on-off measurements with the other Ra isotopes under study. The surface ionised background did not impede the previous laser spectroscopy studies in hot cavity surface ion sources at ISOLDE as the surface ions represented a stable background, consequently this is not expected to hamper the investigations of optical pumping on the hot cavity environment. The planned investigations employing a narrow band RILIS laser will confirm the applicability of the approach to the study of the more exotic Ra isotopes.

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6 Details for the Technical Advisory Committee

6.1 General information

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

- ☒ Permanent ISOLDE setup: *IDS*
  - ☒ 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*

6.2 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:
  Note: all yields assume surface ionised radium from a tantalum hot cavity ion source at 2000°C.

Isotope	Production yield in focal point of the separator (/μC)	Minimum required rate at experiment (pps)	$t_{1/2}$
^{206}Ra	2x10^{4}	The experiment is the yield measurement	240 (2) ms
^{223}Ra	3x10^{7}	6x10^{6}	11.43 (5) d
^{226}Ra	8x10^{6}	6x10^{6}	1600 (7) y
^{227}Ra	3x10^{6}	6x10^{6}	42.2 (5) min
^{231}Ra	4x10^{2}	1x10^{2}	103.9 (14) s

- Full reference of yield information:
  ^{206}Ra: Yield to be measured. Production yield at the focal point of the mass separator estimated based on the FLUKA in-target production rates (https://isoyields2.web.cern.ch/InTargetProductionChart.aspx) with an extraction efficiency based on https://doi.org/10.1038/s41598-024-60331-z.
  ^{223}Ra: https://doi.org/10.1038/s41598-024-60331-z
  ^{226}Ra: Production yield at the focal point of the mass separator estimated based on the FLUKA in-target production rates (https://isoyields2.web.cern.ch/InTargetProductionChart.aspx) with an extraction efficiency based on https://doi.org/10.1038/s41598-024-60331-z.

^{227}Ra: Production yield at the focal point of the mass separator estimated based on the FLUKA in-target production rates (https://isoyields2.web.cern.ch/InTargetProductionChart.aspx) with an extraction efficiency based on https://doi.org/10.1038/s41598-024-60331-z.
^{231}Ra: https://doi.org/10.1038/s41598-024-60331-z

- Target - ion source combination: UC_x + Ta surface ion source
- RILIS? (*Yes*)
  - ☒ Special requirements: (*narrow band Ti:Sa for in-source spectroscopy, laser scanning*)
- Additional features?
  - ☐
- Expected contaminants: Fr contamination is expected on all of the masses studied.
- Acceptable level of contaminants: For A=206, the capability of suppressing the Fr signal using timing and energy gates at IDS is part of the experiment. 1×10^6 pps of ^{206}Fr could be expected based on https://doi.org/10.1103/PhysRevX.4.011055. For A=223, 226 and 227, a surface ionised background in the region of 6×10^7 is acceptable. On A=231, Ac contamination is also anticipated, if problematic, this will be managed by reducing the temperature of the target and ion source.
- Can the experiment accept molecular beams? No.
- Are there any potential synergies (same element/isotope) with other proposals and LOIs that you are aware of? No.

6.3 Shift breakdown

Summary of requested shifts:
* It should be noted that this the time required for RILIS set-up with Ra available from the target. Prior to this, 3 days of RILIS set-up will be required without beam.

With protons or with a pre-irradiated target	Requested shifts
RILIS set-up with Ra*	1
With protons	Requested shifts
Establish a baseline for optical pumping performance with ^{226}Ra and the known {714 nm|784 nm|993 nm^{decay}|615 nm|615 nm} ionisation scheme.	0.5
Verify and optimise the {714 nm|784 nm|731 nm^{decay}|731 nm|558 nm} ionisation scheme with ^{223}Ra.	1.5
Two scans of the 784 nm transition and three scans of the 731 nm transition for ^{227,231}Ra. ^{226}Ra used as a reference.	2
Investigations of the sensitivity of optical pumping to varying operational conditions using ^{226}Ra.	1
Yield checks for ^{206}Ra.	1