EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Laser spectroscopy of neutron-rich Ni with PI-LIST

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Abstract: We propose to perform in-source laser spectroscopy of the neutron-rich nickel isotopes $^{69-74}$Ni ($Z = 28$, $N = 41$-46) using the laser ion source and trap (LIST) apparatus and its perpendicular-illumination (PI-LIST) variant. The ground-state and isomer properties of these isotopes are key to understand the role of multiparticle-multihole cross-shell excitations in nuclear states at low energy between ^{68}Ni and ^{78}Ni, where magicity and shape coexistence coincide. Moreover, our experiment will mark the lightest element studied online with in-source laser spectroscopy to date.

Requested shifts: 24 shifts delivered in 1 run using a UC_x target with LIST.

1 Scientific motivation

The region around neutron-rich Ni is of great interest for nuclear structure and astrophysics research. The Ni chain spans two neutron shell closures ($N = 28, 50$) and one sub-shell closure ($N = 40$), while the protons occupy orbitals in a closed shell at the magic number $Z = 28$. As a result, systematic studies of nuclear spins, quadrupole transition strengths, magnetic dipole and electric quadrupole moments, two-neutron separation energies, and other observables in Ni isotopes and neighboring elements provide deep insight into the evolution of shell structure and resulting magic numbers across a large range of isospin values. Magic numbers are central in the study of nuclear structure [1], and their evolution far from stability [2, 3] provides a stringent test of modern nuclear theories and our understanding of the nucleon-nucleon interaction, including multi-nucleon forces [4, 5, 6, 7].

Significant research efforts have been dedicated to understanding nuclear structure in the vicinity of $Z = 28$, $N = 50$, following the discovery of well-deformed intruder configurations in multiple nuclei [8, 9, 10, 11] that question the doubly magic character of ^{78}Ni. The large excitation energy of the first 2^+ state in ^{78}Ni [12] and the ground- and excited-state systematics around ^{79}Cu [13, 14] provide strong evidence for the double magicity of ^{78}Ni. However, the experimental indication of a well-deformed ($\beta \approx 0.3$) intruder configuration in ^{78}Ni [12], interpreted with large-scale and Monte Carlo shell model calculations [12] and recently reproduced by *ab initio* nuclear theory [15], suggests shape coexistence even at $Z = 28$, $N = 50$ due to residual interactions that reduce the energy gap between the filled $\nu 0g_{9/2}$ orbital and the $\nu 1d_{5/2}$, $\nu 2s_{1/2}$ orbitals in the next shell, permitting nucleon excitations across the shell gap [16]. This is supported by shell model calculations of the intruder configuration in ^{78}Ni with the PFSDG-U interaction, which reveal a multi-particle-hole (np-nh) nature driven by, on average, 4 neutrons across $N = 50$ and 2 protons across $Z = 28$ [17].

Coinciding signatures of a sub-shell closure and shape coexistence are also observed in ^{68}Ni at $N = 40$. While the energy of the first 2^+ state in ^{68}Ni is approximately 0.5 MeV higher than in the neighboring isotopes and more than 1 MeV than in neighboring isotones (see Fig. 1), the existence of three 0^+ states, including the ground state, at low excitation energy is a remarkable example of triple shape coexistence [18]. This is consistent with the formation of an island of inversion (IoI) at $N = 40$ centered around ^{64}Cr. Together with the intruder configuration in ^{78}Ni, these observations have led to arguments about the emergence of a fifth IoI at $N = 50$ [17] and the merging of the $N = 40$ and $N = 50$ IoIs in the neighboring Cr and Fe chains. The ground states of Ni isotopes between $N = 40$ and $N = 50$ are commonly assumed to be spherical, and the evidence of $N = 40, 50$ (sub-)shell closures in Ni, as compared to their expected disappearance in Cr and Fe by PFSDG-U calculations, indicates that the IoI merging might not manifest in Ni [17].

Laser spectroscopy can provide insightful information in neutron-rich Ni that can elucidate shape evolution in light of the interplay of shell closure and shape coexistence.

Measurements of the magnetic dipole moments for odd-A isotopes and comparisons with theory evaluate the purity of the ground-state wavefunctions [19] and the presence of np-nh cross-shell excitations [20]. Moreover, the ground-state nuclear spins of neutron-rich even-odd Ni isotopes have not been firmly assigned, and laser spectroscopy offers a model-independent way to do so.

Additionally, $1/2^-$ isomers with a neutron hole in the normally fully occupied $1p_{1/2}$ orbital of the fp subshell have been observed in ^{69,71}Ni, whose charge radii and magnetic dipole moments remain unknown. Measurements of their moments and the isomer shift in the nuclear charge radii between isomeric and ground states will provide key information about the intruder wavefunction, and they will enable state-of-the-art nuclear calculations that aim to describe deformation in this region to compare against experimental data. Above $N = 40$, neutrons occupy the $\nu 0g_{9/2}$

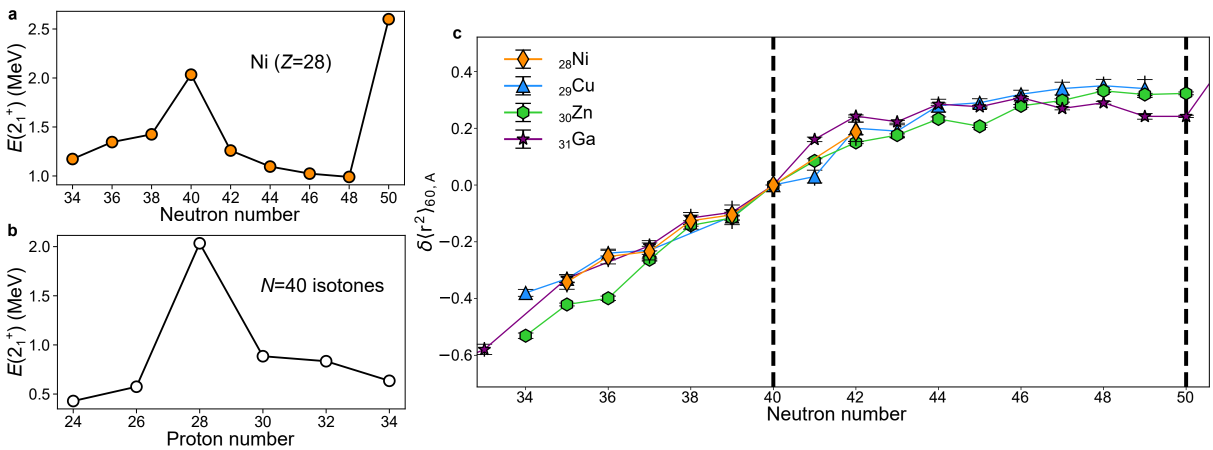

Figure 1: (a) Energy of the first excited 2^+ state across Ni isotopes. (b) Energy of the first excited 2^+ state across $N = 40$ isotones. (c) Comparison of the changes in mean-squared nuclear charge radii $\delta\langle r^2\rangle$ in Ni ($Z = 28$) [21], Cu ($Z = 29$) [22], Zn ($Z = 30$) [23], and Ga ($Z = 31$) [24].

orbital until it is filled at $N = 50$. Measurements of the electric quadrupole moments will allow extracting the quadrupole deformation parameter β_2 of the even-odd ground states, searching for complementary evidence for the previously suggested mid-shell increase in intruder content in the even-even isotopes [25]. Such information is important for efforts to trace the shape evolution and deviation from sphericity as the orbital is filled.

Lastly, measurements of the changes in mean-squared nuclear charge radii are highly sensitive to the underlying nuclear structure in this region, including the weakening of odd-even staggering above $N = 45$ in Zn ($Z = 30$) [23] and its disappearance in Cu ($Z = 29$) [22], shown in Fig. 1. Extending radii measurements to Ni for the same neutron numbers would be key for isolating the role of unpaired protons above $Z = 28$ in the strength of odd-even staggering, as previously suggested [22]. Measurements of the isomer shift with respect to the ground states in ^{69,71}Ni can provide critical information for understanding deformation around $N = 40$, as demonstrated with ^{79g,m}Zn around

$N = 50$ [8]. Laser spectroscopy provides a precise means for a comparison in nuclear size between isomers and ground states. Lastly, the observation or lack of a kink in nuclear charge radii is often interpreted as evidence of a (sub-)shell closure, and completing the knowledge of charge radii around $N = 40$ will provide additional information for an evaluation of the doubly magic nature of ^{68}Ni.

So far, laser spectroscopy has been performed on neutron-rich Ni up to ^{70}Ni [26, 21], excluding ^{69}Ni, with the results highlighting the role of two-body current contributions in the magnetic dipole moments. Due to the refractory nature of the Ni atom and the presence of strong contamination, extending laser spectroscopy beyond ^{70}Ni at ISOLDE is favorable via the in-source approach, where highly sensitive detection via decay spectroscopy at the ISOLDE Decay Station (IDS) and mass spectrometry at ISOLTRAP can be utilized. In this experiment, we propose to perform laser spectroscopy on neutron-rich Ni isotopes using the laser ion source and trap (LIST) and its higher-resolution PI-LIST variant based on perpendicular illumination, aiming to extend the current measurements of nuclear spins, charge radii, and electromagnetic moments beyond $N = 40$. Using fast-timing spectroscopy at IDS for the detection needs of our experiment, level-scheme information of neutron-rich Cu isotopes can also be gathered concurrently, enhancing the scientific reach of the proposal. For instance, as very little is known about the level scheme of ^{74}Cu, fast-timing measurements of the β-decay of ^{74}Ni at IDS will provide new information even considering the low production yield of ^{74}Ni.

In addition to the direct nuclear structure motivation, our proposed experiment will mark the lightest element studied online with in-source spectroscopy to date. The linewidth compression that can be achieved using PI-LIST will be investigated in this experiment as a means to expand the range of elements that are compatible with in-source spectroscopy for nuclear-structure studies, even if the resultant resolution in a medium-mass atom (thus with large Doppler broadening) is lower than previously achieved with PI-LIST with heavy elements at ISOLDE [27]. The results of this experiment would also inform the potential of PI-LIST for nuclear-state-selective ionization for medium-mass isotopes with low yields and in the presence of isobaric contamination, as a way to provide radioactive beams of only ground or isomeric states for experimental study with other techniques.

2 Method

The aim of this proposal is to perform laser spectroscopy of neutron-rich isotopes of Ni using the Laser Ion Source and Trap (LIST) apparatus combined with its Perpendicular Illumination (PI-LIST) mode, as highlighted in Ref. [28]. This technique has been demonstrated at ISOLDE in experiments with Ac [27], Po [29], Tl [30], and Lu [31]. To produce the neutron-rich Ni isotopes, a UC_x target is requested. Modifications to the target or ion source to enhance the extraction of Ni isotopes such as target or ion source linings or back of the line heating would be advantageous.

In-source laser spectroscopy can typically reach higher sensitivity compared to collinear techniques, and it can be employed at most online facilities. However, the high temperatures of the ion source (in excess of 2000 °C) result in large Doppler broadening, typically limiting the spectroscopic resolution to the GHz regime. For the heavy-mass region, where this technique has been regularly implemented, the dipole hyperfine splitting tends to be significantly larger than the Doppler-limited linewidth, and isotope shifts are also of the order of 1 GHz.

In the medium-mass region, where the hyperfine splitting is typically smaller and isotope shifts are of the order of tens or hundreds of MHz, a sub-GHz linewidth is required in order to extract nuclear-structure observables from the hyperfine structure. To achieve this, perpendicular illumination of the radioactive atom cloud in the PI-LIST configuration of the LIST apparatus is critical. In this mode of LIST operation, the conventional counter-propagating illumination is substituted with illumination at 90°, whereby an intersected region of the velocity distribution present in the hot atom cone is probed, overcoming the large Doppler broadening and leading to a significantly improved resolution. This has been demonstrated online with Ac at ISOLDE, where a resolution of 200 MHz was achieved [27], and offline with Tc with a resolution of 100 MHz [32] and Cf with a resolution of 55 MHz [33]. The use of LIST induces losses in the total yield, typically by a factor of ∼100 compared to RILIS operation, with PI-LIST inducing a further reduction by a factor of ∼3; this has been accounted for in the shift request of this proposal. Moreover, for the even-A isotopes ($I^{\pi} = 0^{+}$), a factor of ∼3 can be regained when switching to standard collinear LIST mode, as there is no hyperfine structure that has to be resolved.

Collinear laser spectroscopy was previously performed on neutron-rich Ni at ISOLDE, as presented in Refs. [21, 26]. The $3d^{9}4s\ ^{3}D_{3} \rightarrow 3d^{9}4p\ ^{3}P_{2}$ electronic transition used in those studies is identified as a suitable candidate for this proposal as well, as it is sensitive to the changes in mean-squared charge radii and electromagnetic moments. The proposed laser ionization scheme using the $3d^{9}4s\ ^{3}D_{3} \rightarrow 3d^{9}4p\ ^{3}P_{2}$ transition is illustrated in Fig. 2a, using an autoionizing state. Furthermore, using the hyperfine constants reported in Ref. [26], the expected hyperfine structure of ^{65}Ni and ^{67}Ni with a variety of resolutions from the in-source techniques is given in Figs. 2b,c. Presented alongside the suggested ionization scheme in Fig. 2a is the typical production scheme employed at RILIS for reference. The narrowband laser light required to probe the spectroscopy transitions will be supplied by an injection-seeded ring cavity. A key benefit of in-source laser spectroscopy is the different detection methods that can be employed, varying between the available experimental setups. For this proposal, we aim to use a combination of single-ion counting with a MagneToF detector in the central beamline and decay tagging at IDS, with the potential of also using the ISOLTRAP multi-reflection time-of-flight mass spectrometer for isobaric separation, if contamination proves to be overwhelming during the experiment. For low-yield cases, such as $^{69m,71-74}$Ni, IDS will be used to detect the decay of the Ni isotopes. In this combined RILIS-IDS configuration, a sensitivity below 1 ion/s can be expected.

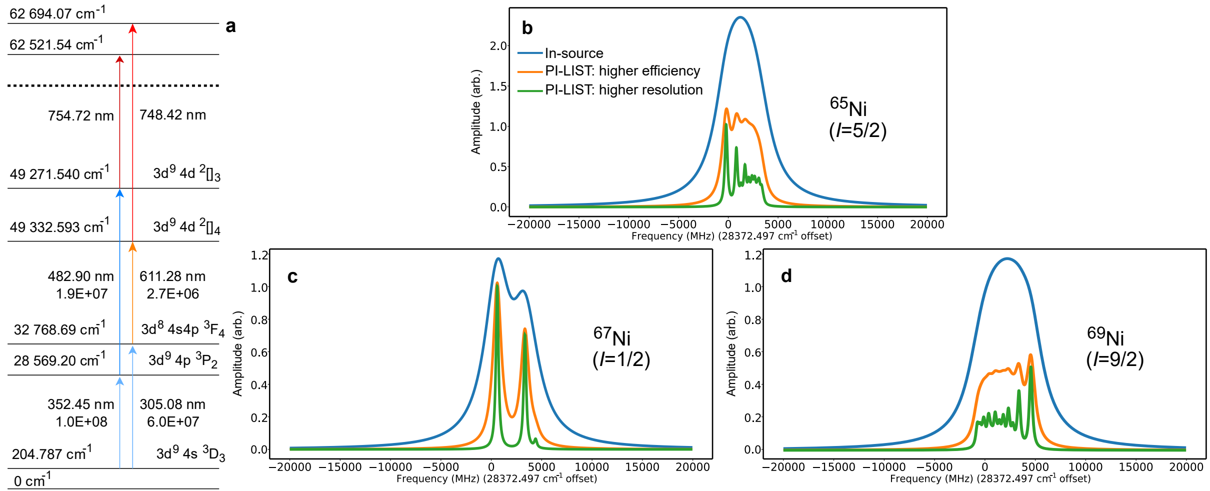

Figure 2: (a) Proposed laser ionization schemes. Left: scheme based on spectroscopic transition used in past collinear experiments. Right: production scheme used by RILIS. (b) Simulated hyperfine structure for ^{65}Ni ($I^\pi = 5/2^-$) using the 352-nm transition, at different projected spectroscopic resolutions in RILIS mode ($\delta f = 3.5$ GHz), PI-LIST mode tuned for optimal efficiency using a dual-etalon laser ($\delta f = 1$ GHz), and PI-LIST mode tuned for optimal resolution ($\delta f = 350$ MHz). (c) Similar simulations for ^{67}Ni ($I^\pi = 1/2^-$), and (d) for ^{69}Ni ($I^\pi = 9/2^+$) using the Schmidt value for the g-factor.

A major challenge in the study of neutron-rich Ni is the overwhelming surface-

Isotope	I^π	$T_{1/2}$	RILIS	LIST	PI-LIST	Detection
^{66}Ni	0^+	54.6 hr	1×10^8	1×10^6	3×10^5	Ion
^{69}Ni	$(9/2^+)$	11.4 s	2×10^4	2×10^2	7×10^1	β
^{69m}Ni	$(1/2^-)$	3.5 s	2×10^3	2×10^1	7	β
^{70}Ni	0^+	6.0 s	1×10^4	1×10^2	3×10^1	β
^{71}Ni	$(9/2^+)$	2.6 s	4×10^3	4×10^1	1×10^1	β
^{71m}Ni	$(1/2^-)$	2.3 s	4×10^2	4	1	β
^{72}Ni	0^+	1.8 s	1×10^3	1×10^1	3	β
^{73}Ni	$(9/2^+)$	0.8 s	5×10^1	0.5	0.1	β
^{74}Ni	0^+	0.5 s	1×10^1	0.1		β

Table 1: Predicted production yields per μC of neutron-rich Ni from a UC_x target for RILIS, LIST, and PI-LIST modes of in-source spectroscopy.

ionised isobaric contamination, previously observing a ratio of 1:10^4 ^{70}Ni:^{70}Ga [21]. With LIST, the contamination can be significantly suppressed using the ion repellers positioned at the entrance aperture of the LIST laser-atom interaction region. This design allows for the majority of surface ions created in the hot-cavity ion source to be repelled while the laser ions produced within the interaction region are extracted. Furthermore, the well-defined laser ion time of flight (ToF) can be gated and used in conjunction with the recently implemented 10-kHz fast-switching beam gate to isolate the part of the ToF profile with the highest laser-to-surface ion ratio.

Table 2: Summary of requested shifts.

	$T_{1/2}$	Shifts	New Measurements
^{66}Ni	54.6 hr	3	*Reference measurements*
^{69}Ni / ^{69m}Ni	11.4 s / 3.5 s	2	$I, \mu, Q_s, \delta\langle r^2\rangle$
^{70}Ni	6.0 s	1	
^{71}Ni / ^{71m}Ni	2.6 s / 2.3 s	4	$I, \mu, Q_s, \delta\langle r^2\rangle$
^{72}Ni	1.8 s	2	$\delta\langle r^2\rangle$
^{73}Ni	0.8 s	6	$I, \mu, Q_s, \delta\langle r^2\rangle$
^{74}Ni	0.5 s	4	$\delta\langle r^2\rangle$
Tuning/Optimization		2	
Total:		**24**	

3 Request

The yields and expected loss factors induced by transitioning between standard RILIS operation, LIST, and PI-LIST modes are summarized in Table 2. Current yields of neutron-rich Ni available in the ISOLDE yield database are only given up to ^{70}Ni, where these values have been given in the RILIS column of Table 2. To give an approximation of the expected yields beyond ^{70}Ni, the known yields were fitted with a diffusion-effusion model that takes half-lives into consideration, and the unknown yields were extrapolated. These numbers are consistent with past measurements and upper limits at ISOLDE from a UC_x target [34]. For the ^{69m,71m}Ni isomers, an order-of-magnitude lower yield compared to the ground state is considered. The number of shifts requested is given in Table 2.

It is noted that in worst-case scenarios, the yield loss factor when going from RILIS to LIST can be as high as $\times 1000$. A realistic loss factor of $\times 100$ is considered here. A loss factor of $\times 500$ would still allow for the majority of objectives to be achieved within the shift request.

Importantly, we note that the yields in Table 2 are based on past yield checks using the standard RILIS production scheme (Fig. 2a, right). During the preparation of this proposal, the scheme we propose based on the 352-nm transition that has high sensitivity to nuclear moments and radii (Fig. 2a, left) was tested and demonstrated more than five times higher efficiency than the standard RILIS scheme.

Summary of requested shifts: 24 shifts delivered in 1 run using a UC_x target with LIST.

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DESCRIPTION OF THE PROPOSED EXPERIMENT

Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing
LIST and PI-LIST	☒ To be used without any modification ☐ To be modified
If relevant, describe here the name of the flexible/transported equipment you will bring to CERN from your Institute [Part 1 of experiment/ equipment]	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
[Part 2 of experiment/ equipment]	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
[insert lines if needed]	

HAZARDS GENERATED BY THE EXPERIMENT

Additional hazard from flexible or transported equipment to the CERN site:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	☐	[pressure] [bar], [volume][l]
	Vacuum	☐	
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	[fluid] [m3]
Electrical Safety	Electrical equipment and installations	☐	[voltage] [V], [current] [A]
	High Voltage equipment	☐	[voltage] [V]
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	☐	[fluid], [quantity]
	Toxic/Irritant	☐	[fluid], [quantity]
	Corrosive	☐	[fluid], [quantity]
	Oxidizing	☐	[fluid], [quantity]
	Flammable/Potentially explosive atmospheres	☐	[fluid], [quantity]
	Dangerous for the environment	☐	[fluid], [quantity]
Non-ionizing radiation Safety	Laser	☐	[laser], [class]
	UV light	☐	
	Magnetic field	☐	[magnetic field] [T]
Workplace	Excessive noise	☐	
	Working outside normal working hours	☐	

	Working at height (climbing platforms, etc.)	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards			