EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Laser spectroscopy of neutron-rich and neutron-deficient Cadmium isotopes using MIRACLS

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S. Malbrunot-Ettenauer[1,2], S. Lechner[3], F. Buchinger[4], L. Croquette[1,4], E. Leistenschneider[5], F. Köhler[6], K. König[6], B. Maaß[6], F.M. Maier[7], K. Mohr[8], L. Nies[9], W. Nörtershäuser[6], P. Plattner[9], J. Palmes[6], L.V. Rodríguez[10], A. Roitman[4], L. Schweikhard[11], D.T. Yordanov[12]

[1] *TRIUMF 4004 Wesbrook Mall, Vancouver BC V6T 2A3, Canada*
[2] *Department of Physics, University of Toronto, Toronto, Ontario, M5S 1A7, Canada*
[3] *CENBG, UMR 5797 Universite de Bordeaux CNRS/IN2P3*
[4] *McGill University, Montréal, Québec, H3A 2T8, Canada*
[5] *Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*
[6] *Institut für Kernphysik, TU Darmstadt, D-64289 Darmstadt, Germany*
[7] *Facility for Rare Isotope Beams, East Lansing, Michigan 48824, USA*
[8] *GSI Helmholtzzentrum für Schwerionenforschung GmbH, 64291 Darmstadt, Germany*
[9] *Experimental Physics Department, CERN, CH-1211 Geneva 23, Switzerland*
[10] *Max-Planck-Institut für Kernphysik, 69117 Heidelberg, Germany*
[11] *Institut für Physik, Universität Greifswald, D-17487 Greifswald, Germany*
[12] *Institut de Physique Nucléaire, CNRS-IN2P3, Université Paris-Sud, 91406 Orsay, France*

Spokespersons
S. Malbrunot-Ettenauer sette@triumf.ca and S. Lechner

Contact persons :
A. Roitman anthony.moshe.roitman@cern.ch and L. Nies lukas.nies@cern.ch

Abstract: We propose to perform laser-spectroscopic studies on cadmium (Cd) isotopes to access unexplored Cd nuclides at and beyond the $N = 50$ and $N = 82$ neutron shell closures. The necessary improvement in experimental sensitivity is achieved by using MIRACLS, where ions are stored in a Multi-Reflection Time-of-Flight (MR-ToF) device while being repeatedly probed by the spectroscopy laser. In addition to spin assignments and electromagnetic moments in ^{99,131}Cd, the measurements will reveal the nuclear charge radii of ^{98,99,131,132}Cd. Fayans-based nuclear density functional theory has successfully predicted charge radii along several isotopic chains ranging from

calcium all the way to tin, thus, approaching a global description of this observable. Hence, Cd charge radii across $N = 82$ and of ^{98}Cd (^{100}Sn minus 2 protons) represent formidable benchmarks for these latest advances in nuclear theory. This proposal completes the MIRACLS project before its MR-ToF device is reconfigured for beam purification at PUMA.

Requested shifts: 24 shifts of radioactive and 6 shifts of stable beam (split into 2 runs)

1 Physics motivation

Nuclear charge radii are sensitive probes for a wide range of nuclear physics phenomena such as shell evolution, pairing, or deformation and shape coexistence. Hence, their accurate description all across the Segrè chart within one global theoretical framework constitutes an ambitious long-term goal of nuclear theory. A promising approach is nuclear density functional theory (DFT) [1], which has shown success across a wide range of nuclei. In recent years, the Fayans functional Fy(Δr) has emerged as particularly effective, resolving long-standing issues such as the odd-even staggering observed in charge radii of calcium (Ca) isotopes. This staggering, especially between the doubly magic ^{40}Ca and ^{48}Ca, constituted a demanding test for nuclear models. An even more evolved version of the functional, Fy(Δr,HFB), extends its applicability to more exotic isotopes like ^{52}Ca, accurately reproducing even their unexpectedly large charge radii [2, 3].

In recent years, the Fayans functionals have also been successfully applied to other elements, from Ca to tin (Sn) isotopes, including Fe [4], Ni [5], Cu [6], Ge [7], Cd [8], In [9], and Sn [10], covering proton numbers $Z = 20$ to $Z = 50$ and mass numbers $A = 36$ to $A = 134$. In [11], a Fayans functional also reproduces fermium charge radii ($Z = 100$) fairly well. The Fayans functionals have thus proven their versatility across this wide range of masses and predict well the odd-even staggering in charge radii as well as their characteristic kinks at shell closures.

In contrast, the Skyrme functional SV-min fails to reproduce these features, for example, neither showing odd-even staggering in Cd isotopes, see Fig. 1(a), nor a kink at $N = 82$ in Sn [10], see Fig. 1(b). Notably, Fy(Δr,HFB) accurately reproduces the odd-even staggering visible in the experimental Cd charge radii as well as the kink at the $N = 82$ shell closure in Sn. Moreover, it predicts a similar kink at $N = 82$ for Cd isotopes, though this has yet to be confirmed due to a lack of data for neutron-rich Cd isotopes beyond $N = 82$. New measurements of ^{131,132}Cd will thus be rewarding for further testing the predictive power of the Fayans functional.

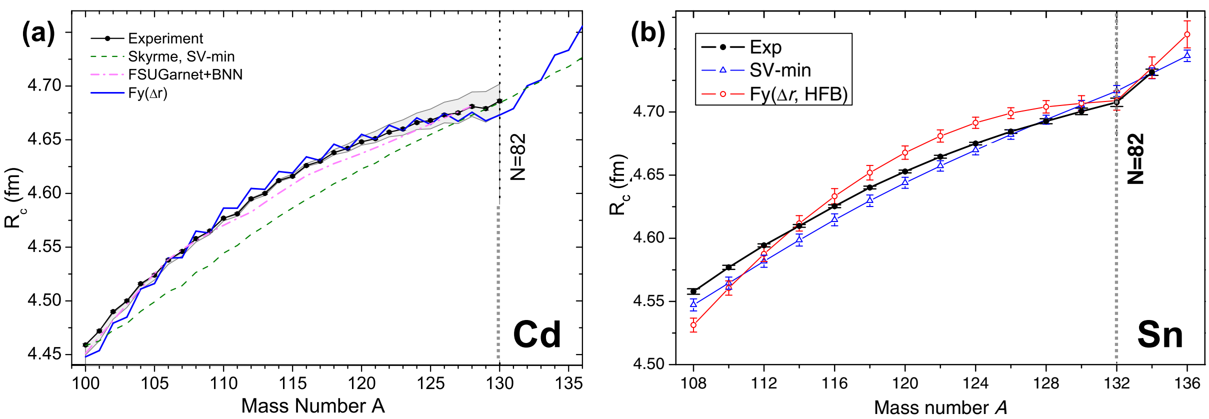

Figure 1: Rms nuclear charge radii R_c for Cd and Sn isotopes. The experimental values obtained at COLLAPS are compared to the results of density functional theory, employing the Skyrme functionals SV-min and the Fayans functional Fy(Δr). Figures from [8, 10].

Despite the successes of Fy(Δr,HFB), recent work [5] indicated a potential deficiency in its description of mid-shell region at which the slope and curvature of differential radii turned out to be overestimated. Hence, a new Fayans functional was recently introduced which added an isovector component in the Fayans pairing functional [9, 7]. The proposed measurement of the unexplored ^{98,99}Cd isotopes will be ideally suited to benchmark this new Fayans functional since the relative decrease in the charge radii at the shell closure $N = 50$ defines the lower-mass end of the parabolic trend, see the neutron-deficient end of Fig. 1(a).

In addition to the successful DFT application on nuclear charge radii, ab-initio nuclear theory has now also reached the mass region of this proposal [12, 13]. It employs nuclear potentials which are linked through chiral effective field theory to the underlying forces described in quantum chromodynamics. Recent progress in ab-initio machinery allows the calculation of nuclear charge radii of the proposed Cd isotopes. They will serve as additional benchmarks for ab-inito calculations, for instance, for ongoing work in coupled cluster theory [14] as well as in-medium similarity renormalization group [15].

Experimentally, root-mean-square (rms) nuclear charge radii can be obtained with high precision and accuracy from collinear laser spectroscopy (CLS) [16, 17, 18, 19]. Conventional CLS has been successfully employed by COLLAPS to obtain the Cd and Sn data in Fig. 1 [8, 10]. The low production yields of the proposed ^{98,99,131,132}Cd isotopes demand a new approach. We therefore intend to take advantage of the Multi Ion Reflection Apparatus for CLS (MIRACLS) for increased experimental sensitivity.

Spins and electromagnetic moments are additional key nuclear observables which are directly accessible via CLS. For ^{99,131}Cd, only tentative spin assignments of $I = 5/2$ and $7/2$, respectively, are available. Nothing is experimentally known about their nuclear magnetic dipole moments μ or electric quadrupole moments Q. The results for the neutron-deficient $^{101-109}$Cd isotopes [20] suggests a simple linear trend over mass number in the electromagnetic moments for their 5/2 ground states, similar to the observation for the 11/2 states in heavier Cd isotopes [21]. Such a linear mass dependence can be explained by the unique-parity states $h_{11/2}$ and following a strict interpretation of the nuclear shell model. However, for the ground states in $^{101-109}$Cd with $I = 5/2$, an involvement of the $d_{5/2}$ orbital has to be expected. Hence, nothing prevents the unpaired neutron to migrate to neighbouring $g_{7/2}$, $s_{1/2}$, and $d_{3/2}$ orbits of identical parity. A linear trend in the electromagnetic moments as suggested by the data for these Cd isotopes is hence a priori not expected. This simplistic interpretation would predict a quadruple moment of $Q = -600$ mb for ^{99}Cd. On the other hand, a large shell model calculation results in $Q \approx -240$ mb [20]. Consequently, a first CLS measurement of ^{99}Cd is of importance to find out whether the linear trend in the quadrupole moments is continued in ^{99}Cd in contrast to the shell-model calculation. More globally, as a nuclide resembling a closed-shell nucleus plus a single neutron, ^{99}Cd plays a central role in the understanding of the nuclear structure in close vicinity of ^{100}Sn. Analogously, the electromagnetic moments of ^{131}Cd, again closed-shell plus one neutron, will shed new light on the nuclear structure beyond the $N = 82$ shell closure.

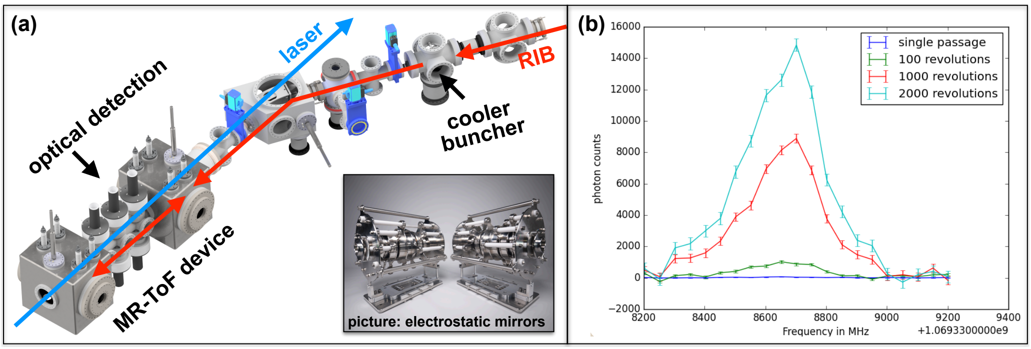

Figure 2: (a) Schematics of the MIRACLS setup (b) CLS resonance spectra of $^{24}Mg^+$ ions for different number of revolutions in the MR-ToF device. See text for details.

2 Experimental technique

This experiment will make use of the high-resolution MIRACLS setup which is coupled to ISOLDE at the LA2 beamline. This apparatus was recently commissioned and successfully performed online measurements of exotic magnesium (Mg) isotopes, including the determination of the previously unknown charge radii of $^{33,34}Mg$.
A schematic of the setup is shown in Fig. 2(a). The incoming RIB is stopped, cooled, and accumulated in a cooler buncher filled with Helium buffer gas. The ions are subsequently released in bunches with a typical temporal width of a few 100s of nanoseconds. After passing a transfer beamline including a 30° electrostatic bender, the ions are injected into the MR-ToF device where they are confined at a beam energy of presently 10.5 keV. A spectroscopy laser is longitudinally overlapped with the ion beam. Fluorescence emitted from laser-excited ions is collected in an optical detection region with currently 3 detection units, each consisting of a pair of aspherical lenses that guide photons onto a photomultiplier tube. The number of detected photons as a function of laser frequency results in a resonance spectrum from which the nuclear properties of interest are extracted.

After a proof-of-principle experiment in a low-energy MR-ToF device (2017-2020) as well as extensive preparation work in simulation and mechanical designs, the construction of the new MIRACLS apparatus begun in late 2021. Ions delivered from ISOLDE were first trapped in the MR-ToF device in August 2023. The online commissioning of the apparatus was achieved in July 2024 during a test beamtime, and its successful scientific exploitation begun with the measurement of neutron-rich Mg isotopes in November 2024.
To illustrate the MIRACLS principle, Fig. 2(b) shows background-subtracted resonance spectra of $^{24}Mg^+$ ions accumulated after different numbers of revolutions in the MR-ToF device. A massive increase in CLS signal is observed when trapping the Mg^+ ions for thousands of revolutions, translating to the increased CLS sensitivity.
As a sensitivity benchmark, the average number of Mg^+ ions injected into MIRACLS' MR-ToF device per measurement cycle was reduced in offline measurements to as little as 0.75 ions. Despite this reduction, a resonance spectrum remained distinguishable above background. Relating the used number of cycles to the timing sequence of ISOLDE proton pulses, such a measurement would last about 7.5 h for radionuclides with short half-lives or target release times. The efficiency of the MIRACLS cooler-buncher has been measured

with ISOLDE beam to be typically 15-25 %, accross mass ranges from $A = 20$ to 238. The 0.75 Mg^+ ions injected into the MR-ToF device hence corresponds to 3-5 ions delivered to MIRACLS. Thus, the sensitivity needed to study the most challenging target radionuclide in the this proposal, namely ^{132}Cd with a yield of 5 ions/s, was already demonstrated.

Another advantage of the MIRACLS technique is its ability to perform collinear and anticollinear laser spectroscopy simultaneously as ions are reflected within the electrostatic mirrors: Two laser beams are applied from the same side of the MR-ToF device, where one is tuned to match the Doppler-shifted resonance frequency for ions moving along the laser-beam direction and the other against it. This approach has been successfully introduced in the recent measurements of neutron-rich Mg isotopes.

The MIRACLS concept works best for closed two level systems in which a laser-excited ion decays back into its initial state and can thus be probed 'indefinitely'. Mg has been ideal as a first MIRACLS science case since its ionic D1 and D2 lines at ~ 280 nm represent such closed 2-level systems at the fine-structure scale. Since even-mass Mg isotopes, i.e. with nuclear spin $I = 0$, exhibit no hyperfine splitting, the ions can be probed for as long as they are trapped without the need of repumping. Even-mass Cd^+ ions have a similar closed ionic system for the $^2S_{1/2} \rightarrow {}^2P_{3/2}$ transition at 215 nm. Its similarity to Mg makes Cd the ideal next science case for MIRACLS. The proposed measurements of $^{98,132}Cd^+$ thus benefit from MIRACLS maximal sensitivity and, given the recent advances discussed above, are within the reach of our apparatus. Their data taking will be conducted in simultaneous collinear and anticollinear laser spectroscopy.

For the proposed measurements of odd-mass $^{99,131}Cd^+$ ions, closed 2-level fine-structure transitions break up in a manifold of hyperfine levels and laser transitions between those tend to transfer population to other states such that the CLS signal would diminish over time. Despite this effect, the MIRACLS approach results in an increase in signal compared the conventional CLS. For example, in MIRACLS' low-energy, proof-of-principle setup, ^{25}Mg ($I = 5/2$) has been studied as a test case using the D1 transition line. Working with a single laser beam, an increase in CLS signal by more than a factor of 5 has been achieved compared to single passage CLS. Thus, a similar enhancement would also be expected for $^{99,131}Cd^+$ ions in the most basic, single-laser application of MIRACLS.

As an additional sensitivity enhancement, a second laser beam can be utilized to repump from the dark hyperfine structure state back into the initial state such that even odd-mass Cd^+ ions can be probed during the entire ion storage in the MR-ToF device. This approach was recently applied at MIRACLS in its successful measurement of the exotic magnesium isotope ^{33}Mg ($I = 3/2$) and stable ^{25}Mg ($I = 5/2$) which also exhibit a hyperfine splitting due to their non-vanishing nuclear spins.

Odd-mass Cd isotopes with $I > 1/2$ feature a transition very similar to the cases of $^{25,33}Mg^+$. Compared to the D2 line in Mg^+ ions, however, the individual 6 transitions in Cd^+ ions tend to be much better resolved [20, 21]. Among these 6 transitions, there are two which represent fully closed 2-level systems, hence, no optical pumping to other hyperfine states takes place. For $^{99,131}Cd$, we thus intend to take advantage of the maximal MIRACLS signal gain for theses 2 closed transitions to find the position of the two hyperfine multiplets. Once located, the remaining 4 transitions can be closely probed by using the reduction of brightness in the closed-transition once the second laser is in resonance and removes population out of the two-level system.

3 Beamtime request

We base our beam-time request on the sensitivities achieved in our recent online and offline measurements with Mg isotopes. The Cd yields that are used in this estimation are summarized in Tab. 1. For neutron-deficient Cd isotopes, ISOLTRAP has recently obtained beam from a LaC_x target, benefiting from laser ionization [23]. All yields are comfortably in MIRACLS' sensitivity range. Contamination in form of SrF molecules will be present in the Cd beams. The 'worst' ratio of contaminant to ion of interest will be found in the $A = 98$ beam with at most a factor of 10 larger than ^{98}Cd. This amount of contamination is easily manageable by MIRACLS.

For the neutron-rich isotopes, a UC_x target is required. Here, the presence of both cesium and barium as surface ionized, isobaric contaminants has been observed [13]. Using a tungsten neutron converter, it is possible to reduce the fraction of these contaminants. Additionally, their yield can be decreased by employing a quartz transfer line. To avoid saturation of the quartz line, we will schedule the most exotic isotope ^{132}Cd close to the start of the experimental run. Even with these methods of mitigating the isobaric contamination in place, the yield of ^{132}Cs and ^{132}Ba will be higher than ^{132}Cd [13]. As the yield of these contaminants is not expected to cause significant space-charge limitations in the MIRACLS buncher, an additional drop in count-rate for ^{132}Cd is not likely.

For all odd-mass cases, we will use the closed hyperfine transitions (see Sect 2) to locate both hyperfine triplets at first. Then, an extended scanning of these regions is necessary to measure the remaining 4 transitions. This extended laser scanning results in a requested beamtime similar to some of the even-mass cases that have lower yields. Our beamtime request for a division in 2 runs is as follows: A neutron-deficient run with 2 shifts for ^{98}Cd, 3 shifts for ^{99}Cd, 4 shifts for ^{100}Cd and systematics. A neutron-rich run with 7.5 shifts for ^{132}Cd, 4 shifts for ^{131}Cd and 3 shifts for ^{130}Cd and systematics. Both runs will also require 0.25 shifts for yield measurements as well as 3 shifts of stable beam for setup and measurement of stable Cd for a King plot analysis.

Summary of requested shifts: 24 shifts of radioactive beam time and 6 shifts of stable beam spread over 2 runs.

Table 1: Cd information and yields according to the ISOLDE yield database [22].

Isotope	half life	Spin I^π	LaC_x Yield (ions/ μC) surface	LaC_x Yield (ions/ μC) RILIS	UC_x Yield (ions/ μC) RILIS
^{98}Cd	9.3(1) s	0	1.0×10^1	$\approx 5 \times 10^1$ [23]	x
^{99}Cd	17(1) s	$(5/2)^+$	4.5×10^2	$\approx 2 \times 10^3$ [23]	x
^{100}Cd	49.1(5) s	0	8.5×10^3	$\approx 8 \times 10^4$ [23]	x
^{130}Cd	129(3) ms	0	x	x	1.0×10^4
^{131}Cd	98(2) ms	$(7/2)^-$	x	x	3×10^2 (ions/s)* [13]
^{132}Cd	84(5) ms	0	x	x	5×10^0 (ions/s)* [13]

∗ Recent values at CA0 in identical target-ion-source combination as required for this proposal. These values differ from the ISOLDE database with protons directly on target [24] which results in overwhelming contamination

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4 Details for the Technical Advisory Committee

4.1 General information

☒ Permanent ISOLDE setup: *MIRACLS*

- ☒ 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

- Requested beams:

Isotope	Production yield in focal point of the separator (/μC)	Minimum required rate at experiment (pps)	$t_{1/2}$
^{98}Cd	≈50	20	9.3 s
^{99}Cd	≈2,000	200	17 s
^{100}Cd	≈80,000	200	49 s
^{130}Cd	10,000	200	129 ms
^{131}Cd	300 (per second)	200	98 ms
^{132}Cd	5 (per second)	3 (per second)	84 ms

- Full reference of yield information: ISOLDE yield database as well as references [23] and [13].
- Target - ion source combination:
  Neutron-deficient Cd: LaC_x
  Neutron-rich Cd: UC_x
- RILIS? *Yes*
  - ☐ Special requirements: (*isomer selectivity, LIST, PI-LIST, laser scanning, laser shutter access, etc.*)
- Additional features?
  - ☒ Neutron converter: *for neutron rich Cd isotopes*
  - ☒ Other: *mass markers with stable Cd isotopes, quartz transfer line for neutron rich Cd isotopes*

- Expected contaminants: *Isotopes and yields*

  Neutron-deficient Cd: SrF molecules. At ^{98}Cd the contaminants are expected to be at most a factor of 10 more intense than the ions of interest.

  Neutron-rich Cd: cesium and barium as surface ionized, isobaric contaminants. To reduce these contaminants to acceptable levels, we request a neutron converter and a quartz transfer line, analogously to what has been done for the ISOLTRAP measurements described in [13].

- Acceptable level of contaminants: Our experiment is sensitive to stable and radioactive contamination. The biggest risk of contaminants is to saturate our cooler-buncher such that we cannot accumulate sufficient amount of Cd ions prior to the laser-spectroscopic measurements in the MR-ToF device. The anticipated ratios of contamination to ion of interest fall within the acceptable range for MIRACLS.

- Can the experiment accept molecular beams? No, our experiment requires atomic Cd beams.

- Are there any potential synergies (same element/isotope) with other proposals and LOIs that you are aware of?

4.3 Shift breakdown

Summary of requested shifts:

Part 1: Neutron-deficient Cd isotopes

With protons	Requested shifts
Yield measurement of ^{98,99,100}Cd	0.25
Data taking, ^{98}Cd	2
Data taking, ^{99}Cd	3
Data taking, ^{100}Cd	1
Systematics	3
Without protons	Requested shifts
Optimization of experimental setup using stable Cd	1.5
CLS measurements of stable Cd isotopes for King plot analysis	1.5

Part 2: Neutron-rich Cd isotopes

With protons	Requested shifts
Yield measurement of ^{130,131,132}Cd	0.25
Data taking, ^{132}Cd	7.5
Data taking, ^{131}Cd	4
Data taking, ^{130}Cd	1
Systematics	2
Without protons	Requested shifts
Optimization of experimental setup using stable Cd	1.5
CLS measurements of stable Cd isotopes for King plot analysis	1.5

4.4 Health, Safety and Environmental aspects

All hazards and safety protocols involving the MIRACLS installation at LA2 are documented, discussed with, and approved by the CERN safety team.