4D-imaging of drip-line radioactivity by detecting proton emission from $^{54m}$Ni pictured with ACTAR TPC

Proton radioactivity was discovered exactly 50 years ago. First, this nuclear decay mode sets the limit of existence on the nuclear landscape on the neutron-deficient side. Second, it comprises fundamental aspects of both quantum tunnelling as well as the coupling of (quasi) bound quantum states with the continuum in mesoscopic systems such as the atomic nucleus. Theoretical approaches can start either from bound-state nuclear shell-model theory or from resonance scattering. Thus, proton-radioactivity guides merging these types of theoretical approaches, which is of broader relevance for any few-body quantum system. Here, we report experimental measurements of proton-emission branches from an isomeric state in $^{54m}$Ni, which were visualized in four dimensions in a newly developed detector. We show that these decays, which carry an unusually high angular momentum, $\ell = 5$ and $\ell = 7$, respectively, can be approximated theoretically with a potential model for the proton barrier penetration and a shell-model calculation for the overlap of the initial and final wave functions.
Nuclear stability is governed by the underlying nuclear shell structure. Closed nuclear shells, so-called magic numbers of protons ($Z$) and neutrons ($N$), are a central concept of nuclear structure. They confer to the nuclei a particular stability with respect to their neighbours. The study of nuclei in the vicinity of those with a magic proton number and a magic neutron number, i.e., doubly magic nuclei, is of prime interest: they allow us to adjust parameters of the nuclear shell model, a prime model at hand for the description of the structure of the atomic nucleus.

For the case of doubly magic $N=Z$ nuclei, another fundamental concept of nuclear structure physics comes into play: isospin. Isospin was introduced by Heisenberg to treat the proton and the neutron as two different quantum states of a single particle, the nucleon, to acknowledge the fact that protons and neutrons have similar properties: The nucleon–nucleon interaction is to a large extent charge independent and charge symmetric. However, a closer look revealed that the Coulomb interaction and parts of the strong force violate this isospin symmetry.

Nuclear physics research continues to thrive on identifying nuclei at the limits of nuclear existence in terms of $Z$, $N$, and mass number, $A$. Here, nuclear structure phenomena can often be filtered out more purely. Like a decay, proton emission is typically described as a quantum tunnelling process from a quasi-bound quantum state, confined for a finite time by the Coulomb and centrifugal barrier. Thus, nuclei or nuclear states beyond the proton drip-line are ideal candidates to study the influence of the nuclear continuum. Furthermore, the time-reversed process of proton-dripline are ideal candidates to study the influence of the nuclear continuum. Additionally, parts of the strong force violate this isospin symmetry.

Proton radioactivity (for the latest reviews see8,9) was discovered 50 years ago10, interestingly from an excited isomeric state, namely $^{53}\text{Co}$, located at 3.19 MeV excitation energy with a half-life of $19/2^+$. In this decay, the proton has to carry away impressive 9h units of angular momentum, $\ell = 9$, to decay into $^{52}\text{Fe}$, with only a 1.5% branch to its $0^+$ ground state known. In the same region of the nuclear chart, two-proton radioactivity was discovered from the ground states of $^{45}\text{Fe}$11,12, $^{48}\text{Ni}$13,14 and $^{54}\text{Zn}$15,16.

As just mentioned, these dripline phenomena can also arise from excited states of nuclei closer to stability17,18, in particular, if these states are relatively long-lived, i.e., isomeric like in $^{53}\text{mCo}$10. A second case of proton radioactivity from an isomer near $^{56}\text{Ni}$ was discovered in $^{54}\text{mNi}$19, the mirror partner of the well-studied $10^+_1$ isomer $^{54}\text{mFe}$20. For these so-called “mirror nuclei”, proton and neutron numbers are inverted. At and around the doubly magic, $N = Z = 28$ nucleus $^{56}\text{Ni}$, all these aspects can be observed and studied simultaneously.

The nuclear structure in the region of $^{56}\text{Ni}$ is very well described by the nuclear shell model (e.g.18). For the decay of the $10^+_1$ isomer in $^{54}\text{Fe}$, experimental findings and theoretical description match remarkably well. Surprisingly, this was not the case for its mirror decay from $^{54}\text{mNi}$19. In fact, in addition to its decay by electromagnetic transitions, an unexpected and significant proton-emission branch from the $10^+_2$ isomeric state to the first excited state of $^{53}\text{Co}$ could be inferred from the $\gamma$-ray study. But despite the addition of this decay branch, shell-model theory and experiment still did not match. The discrepancy could, however, be understood by assuming that an additional proton branch of similar strength to the ground state of $^{53}\text{Co}$ occurs, to which the previous experiment was insensitive to. Indeed, simple barrier-penetration calculations for each proton-emission branch show that the emission of an $E_p = 1.20$ MeV, $\ell = 5$ proton to the first excited state of $^{53}\text{Co}$ is approximately as likely as the emission of an $E_p = 2.50$ MeV, $\ell = 7$ proton to its ground state21–23. The situation is illustrated in Fig. 1.

The present paper describes an experiment, which allowed for a four-dimensional (4D) visualization of both proton-emission branches and to derive their precise branching ratio, thus completing the picture of the decay of the $10^+_2$ isomer in $^{54}\text{Ni}$. The experimental results allow for the anticipated improved understanding of isospin symmetry, and provide a precision test of proton-emission theory.

**Results**

The experiment was performed at the LISE3 beam line of GANIL. The $^{54}\text{Ni}$ ions were produced by fragmentation of a $^{58}\text{Ni}$ beam at 75 MeV/nucleon on a beryllium target. The fragments were selected by the LISE3 spectrometer and implanted in the active volume of the ACTAR TPC device25,26, a gas detector with 16384 read-out pads and an active volume of 25 cm $\times$ 25 cm $\times$ 20 cm working as a time projection chamber. Approximately 0.4% of the $^{54}\text{Ni}$ ions were implanted in the $10^+_2$ isomeric state ($E^* = 6457$ keV; $T_{1/2} \sim 155$ ns). About half of these isomers decay by emission of a proton that can be detected in ACTAR TPC.

Due to the short half-life of the isomeric state, the ionization signal from the emitted proton is registered together with the huge signal of the ion implantation (three orders of magnitude larger), making the detection impossible with standard techniques such as silicon detectors (e.g.11,15). Using ACTAR TPC, the proton signal is clearly visible when the particle track projection on the collection plane creates a signal on different pads of the detector (Fig. 2-left). For each pad of the $(X,Y)$ collection plane, the drift time of the signal, which is proportional to the $Z$ coordinate, is measured and allows for a 3D representation of the signal distribution. The proton is emitted a short time after the implantation ($T_{1/2} \sim 155$ ns), while the ionization signal of the ion has already started to drift towards the collection plane: the time offset between the end of the ion track and the beginning of the proton track (Fig. 2-right) is then a direct measurement of the decay time of the isomeric state. The analysis of events with the

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**Fig. 1 Decay scheme of $^{54}\text{mNi}$.** The scheme is based on previous19 and present results. Level energies are in keV and measured relative to the ground state of $^{54}\text{Ni}$. The $1^+$ proton to the first excited state of $^{53}\text{Co}$ was indirectly evidenced previously; the proton emission to the ground state ($p2$) is observed in the present work. The two-proton emission branch ($pp$) is also energetically possible, but is very unlikely because of its small energy.
implanted ion and the emitted proton thus provides 4D information about the decay: the 3D track with the related energy and the decay time of the isomer and the direct observation of the proton track length and decay time (Fig. 3 and Supplementary Information).

During 17 h of data taking, about two million $^{54}$Ni ions were implanted in the detector. About 3000 events with proton emission were identified. The analysis of these events (ion and proton track fit) was used to build the experimental distributions of the proton track length and decay time (Fig. 3 and Supplementary Movie 2). Due to challenges associated with the charge collection plane used during the experiment, some pads regions were blind. Hence, although the detector also measures the charge collected, which is proportional to the particle energy loss (5th dimension), the determination of the proton energy from fits of the proton track length had a better resolution.

The full proton-radioactivity scheme of the isomeric state could be extracted from the analysis, with the first observation of the $\ell = 7$ proton emission at $E_p = 2.5002(43)$ MeV to the ground state of $^{52}$Co and the direct observation of the $\ell = 5$ transition at $E_p = 1.1979(44)$ MeV to the first excited state, with respectively 1411 ± 40 and 1459 ± 40 counts. The decay energies and their uncertainties were derived from refs. 19,23. The detection efficiency was estimated from a dedicated Monte-Carlo simulation to be $(58.8 \pm 3.8)\%$ for the $E_p = 1.20$ MeV proton (p1) and $(81.7 \pm 2.2)\%$ for the $E_p = 2.50$ MeV proton (p2). As a result, a branching ratio of $(57.3 \pm 1.9)\%$ is deduced for the $E_p = 1.20$ MeV proton with respect to the total proton emission. Combining the proton decay measured in this experiment with the previous results from $\gamma$ spectroscopy15 where the relative intensities of $\gamma$-ray emission and the $E_p = 1.20$ MeV proton branch were determined, the absolute branching ratios of the decay of the isomeric state are $b_\gamma = (50.5 \pm 2.3)\%$ and $b_p = (49.5 \pm 2.3)\%$.

As indicated in Fig. 1, a two-proton (2p) emission branch from the $^{54m}$Ni isomer into the $^{52}$Fe ground state is energetically possible. We do not have any indication of its observation in our data. This is in-line with expectations, because the barrier-penetration half-life for two-proton emission is about a factor of 10$^6$ longer than for one-proton emission.

Discussion

The combined branching ratio of both proton-emission branches, $b_p = (49.5 \pm 2.3)\%$, confirms the isospin symmetry aspects discussed in ref. 19. The present precise result allows for an in-depth study of electromagnetic decays from the “mirror” isomers in $^{54}$Ni and $^{54}$Fe.

For the theoretical description of the high-$\ell$ proton-emission probabilities, we follow common procedures and assumptions used to calculate proton decay widths. The decay width can be factorized into a many-body nuclear structure part that gives the spectroscopic factors, $C^2S$, and a potential-barrier penetration part that gives the single-particle decay widths, $T_{2p}$:

$$\Gamma = (C^2S)\Gamma_{2p}$$

The results are expressed in terms of the half-life $T_{1/2} = h \ln 2/\Gamma$.

We start with the $f_p$ model space and then allow one proton to be excited into one of the high-$\ell$ orbitals $0h_{11/2}$ ($\ell = 5$), $0j_{13/2}$, and $0j_{15/2}$ ($\ell = 7$). The dominant proton configuration for the $^{10+}$ state in $^{54}$Ni is $(0f_{7/2})^2(0f_{5/2}1p_{3/2}1p_{1/2})^1$. The proton partitions are truncated to allow for the $(0f_{7/2})^2(0f_{5/2}1p_{3/2}1p_{1/2})^1, (0f_{7/2})^1$,
The standard Woods–Saxon potential, the single-particle energy (SPE) for the proton 0\(\ell\) orbital is below the Coulomb plus centrifugal barrier and comes 18.7 MeV above the SPE of the 0\(f_{7/2}\) orbital. The wave functions of the proton \(\ell = 7\) (i) orbitals are in the continuum. Based on extrapolated energies obtained by including the central well depth, we estimate the effective SPE of the 0\(j_{15/2}\) (0\(j_{13/2}\)) orbitals to be 32 (50) MeV above the SPE of the 0\(f_{7/2}\) orbital. The NuShelliX code\(^{38}\) is used to obtain the wave functions for \(54\)Ni and \(53\)Co and the proton-emission spectroscopic factors, \(C_p^2\). The single-particle decay widths \(\Gamma_{ap}\) were obtained from proton scattering from a Woods–Saxon potential. We start with the standard Woods–Saxon parameters used by Bohr and Mottelson\(^{34}\). The Coulomb potential was obtained from a uniform charge density distribution with radius \(r_c = 1.22\) fm and \(A = 53\). The parameter \(r_c = 1.22\) fm was chosen to reproduce the experimental displacement energy between \(52\)Fe and \(53\)Co of 9.07 MeV. With the Bohr–Mottelson potential diffuseness of parameter of 0.67 fm, the potential radius \(r_0 = 1.26\) fm was chosen to reproduce the experimental rms charge radius of \(52\)Fe of 3.73 fm\(^{35}\). The magnitude of the 0\(f_{7/2}\) proton SPE of 7.15 MeV is close to the experimental proton separation energy of \(52\)Fe, \(S_p = 7.38\) MeV. This potential was then used to calculate proton scattering from \(53\)Co with the code WSPOT. The potential depth was adjusted to give the \(Q_p\) value for each of the high-\(\ell\) orbitals.

The results of calculated proton-emission probabilities are summarized in Table 1. The results with the wave functions for the GXFPIA and KB3G Hamiltonians are similar. The calculated partial half-life for the decay to the 7/2\(^{-}\) state of (0.34/0.52) \(\mu\)s (GXFPIA/KB3G) is in reasonably good agreement with the experimental value of 0.73 \(\pm\) 0.06 \(\mu\)s. The experimental partial half-life for the decay to the 9/2\(^{-}\) state of 0.55 \(\pm\) 0.03 \(\mu\)s is much smaller than those calculated. With the calculated single-particle proton decay half-life, the spectroscopic factor deduced from the experiment would be \(C_p^2 = 4.6 \times 10^{-6}\).

The spectroscopic factor for the decay to the 9/2\(^{-}\) state from another 10\(^{+}\) state, 2 MeV higher in energy, has a spectroscopic factor of the order of 100 \(\times\) 10\(^{-6}\). The decay to the 7/2\(^{-}\) state, the spectroscopic factors for these two 10\(^{+}\) states are similar. Thus, a small mixing between these two 10\(^{+}\) states would bring the theory into good agreement with experiment for the decays to both the 7/2\(^{-}\) and 9/2\(^{-}\) states. This mixing may come from a calculation in a less truncated \(fp\) shell-model space, or it may reflect uncertainties in the \(fp\)-shell Hamiltonians.

Table 1 Calculated properties of the proton-emission branches from the 10\(^{+}\) isomer in \(54\)Ni. They are calculated with the GXFPIA and KB3G Hamiltonians.

| \(J_i\) | \(n\) | \(\ell\) | \(\epsilon\) | \(Q_p\) (MeV) | \(T_{1/2,sp}\) (\(\mu\)s) | \(C_p^2\) | \(T_{1/2}\) (\(\mu\)s) | \(C_p^2\) | \(T_{1/2}\) (\(\mu\)s) |
|---|---|---|---|---|---|---|---|---|---|
| 9/2\(^{-}\) | 0\(f_{11/2}\) | 1.22 | 2.5 | 0.008 \(\times\) 10\(^{-6}\) | 200 | 0.096 \(\times\) 10\(^{-6}\) | 26 |
| 7/2\(^{-}\) | 0\(f_{9/2}\) | 2.55 | 1.3 | 2.6 \(\times\) 10\(^{-6}\) | 0.50 | 1.4 \(\times\) 10\(^{-6}\) | 0.93 |
| 7/2\(^{-}\) | 0\(f_{7/2}\) | 2.55 | 0.57 | 0.55 \(\times\) 10\(^{-6}\) | 1.04 | 0.49 \(\times\) 10\(^{-6}\) | 1.16 |

The columns give the spins of the orbitals contributing, their quantum numbers, the proton-emission \(Q\) values for the emission to the ground and first excited states, the partial potential-barrier penetration half-lives, the spectroscopic factors, and the partial proton-emission half-lives for the respective orbitals.
of the pad signals (see Fig. 2). For a 3D analysis of the proton tracks, the drift time (maximum and total amplitude of the track pads, number of pads hit, track length), and the 2.50 MeV (red) protons.

The drift time difference \( \Delta t \) between the proton track stop and start points. The drift velocity \( v_d \) is estimated for each proton group with a linear regression defined from the total length (see Fig. 4): \( L^2 = (X_d)^2 + (Y_d - \Delta Y)^2 \), resulting in an average value of \( v_d = (5.3 \pm 0.4) \text{ mm } \mu \text{s}^{-1} \), in fair agreement with a GARFIELD calculation.

**Track fit.** The beam direction is along the X axis. The signal from pads corresponding to the ion implantation is fitted to get the ion direction angles in the (X, Y) and the (X, T) vertical planes. The stopping position is then defined as the last pad along X with a signal, corrected with an average offset computed from the intersection of proton tracks and the beam axis.

The final analysis of the proton tracks was performed with a full 3D fit of the proton signal distribution: a Bragg peak model (built from a Geant4 simulation for protons, as explained in §4.4.3 with \( \alpha \) particles) is used to compute the ionization signal along the trajectory and the 3D signal distribution is then calculated by the 3D signal dispersion along the drift towards the collection plane. Since the beginning of the proton track is hidden by the ion signal, the starting point of the proton track is the closest point of the track extrapolation in the (X, Y) plane to the ion stopping position (Fig. 5).

The proton track length is then computed from the 3D fit parameters defining the trajectory. The decay time is the time difference between the ion and proton tracks at the proton start point (Fig. 2).

**Detection efficiency.** A proton emitted after the implantation of a \(^{54}\text{Ni}\) may not be detected because its signal is hidden by the ion-track signal in the attenuation zone of the pad plane. In addition, energetic protons may escape from the active volume of ACTAR TPC. To obtain the branching ratio, \( b_{p} \), of each proton line, it is necessary to determine the detection efficiency for each proton energy. This is achieved with a dedicated simulation built from: (1) an event generator for ion implantation and random proton-emission direction with chosen energy, (2) the simulation of the proton energy loss along its trajectory in ACTAR TPC using Geant4 with the gas pressure adjusted to reproduce the measured tracks length, (3) the drift of the ionization signal, with dispersion and amplification on the collection plane, (4) the processing of the signal collected on the pads.

The implantation profile of the stopping point and the ion direction distribution are defined from the ion tracks determined experimentally for all \(^{54}\text{Ni}\) events. The proton emission is then generated with an isotropic distribution for tracking and signal processing. Applying the same selection criteria as for the experimental data allows the determination, for each proton energy, of the global detection efficiency that combines the selection of the events as a function of the observed proton signal and the detection volume escape probability. As a large number of events can be generated, the efficiency uncertainties are dominated by systematic effects due to uncertainties of the simulation parameters (Supplementary Fig. 1).

**Data availability**

The data that support the findings of this study are available from the corresponding author on reasonable request (https://doi.org/10.26143/GANIL-2019-E690).

**Code availability**

The WSPOT code is available at https://people.nscl.msu.edu/~brown/reaction-codes/. The analysis codes used for the experimental data analysis are available from the corresponding author on reasonable request.

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Author contributions
B.B., D.R. and J.-C.T. prepared the proposal for the experiment, J.G., T.R., J. Pa. set up the instrumentation, B.B, J.-C.T., L.C., O.K., O.S., C.J., Piot, prepared the radioactive $^{54}$Ni beam, J.G., T.R., B.B., D.R., H.A.-P., A.A.R., P.A., M.C., L.C., D.M.C., B.F., J.I.F., M.G., S.G., G.F.G., B.M., A.M., J, Pa., J.Pib, J.Piot, prepared the detector, data acquisition, and radioactive beam systems. J.G., T.R., B.B., D.R., and B.A.B. carried out the data analysis and interpretation of the data and prepared the manuscript.

Competing interests
The authors declare no competing interests.

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