Observation of Proton Radioactivity of the (21^+) High-Spin Isomer in ^{94}Ag

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We have observed direct one-proton decay of the (21^+) isomer in the $N = Z$ nuclide ^{94}Ag into high-spin states in ^{93}Pd by detecting protons in coincidence with γ-γ correlations and applying γ gates based on known ^{93}Pd levels. Two decay branches have been identified, with proton energies of 0.79(3) and 1.01(3) MeV and branching ratios of 1.9(5)% and 2.2(4)%, respectively. The corresponding partial half-life values are 21(6) and 18(4) s. The Q value of the direct proton decay of the (21^+) isomer was found to be 5.78(3) MeV. The very small reduced widths of the observed proton decays might reflect dominating collective configurations in the (21^+) isomer, and the fine structure of the proton spectrum might indicate a strong deformation of this state.

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Unstable atomic nuclei transmute into more stable ones by means of the weak, electromagnetic, or strong interaction. The corresponding disintegration modes are β decay, γ-ray deexcitation, or the spontaneous emission of protons, α particles, or heavier nuclei. Proton radioactivity represents the direct spontaneous emission of protons from a long-lived nuclear state. The term "direct" indicates that one does not deal with protons emitted promptly in a nuclear reaction or following β decay; the term "long-lived" characterizes a nuclear lifetime long enough to establish the atomic structure [1]. Although these particular states are proton unbound to the strong nuclear force, Coulomb and centrifugal barriers together with structure effects can considerably slow down the decay. Proton radioactivity was discovered in the decay of the $(19/2^-)$ isomer in ^{53}Co to the 0^+ ground state of ^{52}Fe [2]. Meanwhile, a number of proton-emitting ground and isomeric states has been investigated for $50 < Z < 84$ nuclei, the resulting data becoming a versatile spectroscopic tool [3,4]. In a few cases, even the fine structure was identified, i.e., the population of more than one level of the daughter nucleus (e.g., [5]). Moreover, the long-expected two-proton radioactivity was recently discovered (see [6] for a review).

In this Letter, we report on the one-proton decay of a (21^+) isomer in ^{94}Ag ($N = Z = 47$), whose properties are unique in the entire chart of nuclides. This high-spin isomer combines a high excitation energy of 6.7(5) MeV with a remarkably long half-life of 0.39(4) s, and is open to at least five decay modes, i.e., β-delayed γ ray, proton, and two-proton, as well as direct proton and two-proton radioactivity. The β-delayed γ-ray and proton emissions from the (21^+) isomer have been reported in [7–10], whereas its β-delayed two-proton as well as direct proton and two-proton decay has been communicated only as a conference contribution [11].

The measurements were performed with the GSI online mass separator [12]. The ^{94}Ag nuclei were produced by ^{58}Ni(^{40}Ca, $p3n$) fusion-evaporation reactions and ionized by an ion source [13], which provided a beam intensity of about 2 s^{-1} for the (7^+) and (21^+) isomers of ^{94}Ag while suppressing the ^{94}Pd contamination. The 29 ms ground state of ^{94}Ag(0^+) is too short-lived to be extracted from the ion source. The mass-separated $A = 94$ beam was implanted into a tape surrounded by an array of charged-particle and γ-ray detectors. Three large-area silicon (Si) multistrip detectors, equivalent to 6 single detectors, were used to measure charged particles and 17 germanium (Ge) crystals for recording γ rays. The total photopeak efficiency was 3.2% for 1.33 MeV γ rays. The energy dependence of the detection efficiency was determined by using standard calibration sources. The Si detectors were arranged around the collection point covering 65% of 4π solid angle. More details about the experiment are given in [9].

The spectrum of charged particles following the β decay of the ^{94}Ag isomers is shown by the solid curve in Fig. 1. It has a pronounced peak around 0.5 MeV due to energy loss of positrons from β decay. The spectrum falls off exponentially with increasing energy up to 2 MeV. At higher energies (not shown here), the spectrum has a broad bump around 3 MeV due to β-delayed protons [9]. In searching for a presumably small branch of direct proton decay, we employed multiple γ-γ coincidences to "tag" this disintegration mode. Direct proton decay of the (21^+) isomer was identified by demanding coincidences between single-hit events recorded in the Si and γ-γ coincidence events

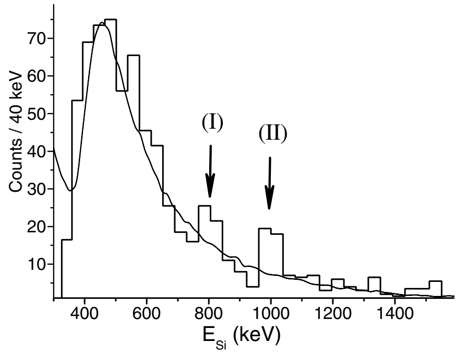

FIG. 1. Spectrum measured in the Si detectors for the decay of ^{94}Ag isomers (histogram), using the Si-γ-γ coincidence with double γ gates on any pair of ^{93}Pd transitions (listed in Table I). The peaks (I) and (II) indicate the direct proton decay of the (21^+) isomer. The singles spectrum of the Si detectors (solid line) is scaled down by a factor of 240.

registered in the Ge detectors. Only by blind γ-gate setting based on the recently established ^{93}Pd level scheme [14] do the direct proton branches become evident. Such a correlation technique has been used in studies of γ-delayed charged-particle emission; e.g., see [15]. The combination of detector arrays with large granularity and high-multiplicity γ decays of high-spin daughter states enabled stringent gating conditions in order to pickup weak decay branches.

The charged-particle spectrum obtained in the above-mentioned way is shown by the histogram in Fig. 1. It has the following two features. First, two weak but statistically significant peaks occur at 0.79(3) and 1.01(3) MeV, which are marked as (I) and (II), respectively. The peaks are interpreted as being due to direct proton emission from the (21^+) isomer in ^{94}Ag. This conclusion is based on the observation that the proton events (I) and (II) are coincident with several γ rays following deexcitation of the known states in ^{93}Pd [14]. Second, the broad bump around 0.5 MeV stems from β^+ particles which are coincident with Compton-scattered γ rays (from the β decay of the ^{94}Ag isomers) forming background in the γ gates. Both conclusions are based on the γ-ray data shown in Fig. 2. The γ spectrum displayed in the upper panel of Fig. 2 reveals 10 known γ transitions in ^{93}Pd [14], whose properties, together with those of the respective ^{93}Pd levels, are listed in Table I. These data yield evidence that the proton line (I) is coincident with all γ rays following the deexcitation of the known $(33/2^+)$ state in ^{93}Pd. In a similar way, the experimental results shown in the middle panel of Fig. 2 and the corresponding data listed in Table I prove that the proton line (II) is coincident with the same ^{93}Pd γ rays except the 1132 keV one. In addition, new γ rays (614, 403, and 276 keV) are observed, the latter is identified in coincidence with the known 275 keV γ ray. The spectrum shown in the lower panel of Fig. 2 is dominated by the

TABLE I. Level energies and spin-parities of the ^{93}Pd states together with respective γ-ray energies and Si-γ coincidence relations, observed in direct proton decay of ^{94m}Ag (21^+).

E_{level} (keV)	I^π	E_γ (keV)	Coincident E_γ, Si-gate (I)	Coincident E_γ, Si-gate (II)
0.0	$(9/2^+)$	—	—	—
984[a]	$(13/2^+)$	984	196, 208	167, 196, 361
2079[a]	$(17/2^+)$	1096	167, 196, 275, 349	167, 275
2079[a]	$(17/2^+)$	208	153, 167, 196, 275, 887, 984, 1132	167, 275, 403, 614, 887
2232[a]	$(17/2_2^+)$	361	167, 196	167, 275, 403, 514, 984
2232[a]	$(17/2_2^+)$	153	167, 196, 208, 275, 887, 1132	167, 349, 403, 614, 887
2429[a]	$(19/2^+)$	349	167, 275, 991, 1096	153, 167, 275, 349, 426, 514, 614
2429[a]	$(19/2^+)$	196	153, 167, 208, 275, 361, 984, 1096	275, 403, 887, 984, 991
2596[a]	$(21/2^+)$	167	153, 196, 208, 275, 361, 887, 991, 1096, 1132	153, 208, 275, 349, 361, 403, 426, 514, 614, 984, 991, 1096
2871[a]	$(25/2^+)$	275	153, 167, 196, 208, 1096	167, 196, 208, 276, 349, 361, 403, 514, 614, 991, 1096
3862[a]	$(29/2^+)$	991	167, 349	275
4994[a]	$(33/2^+)$	1132	153, 167, 208	—
3734[b]	$(29/2^-, 31/2^-)$	349	—	153, 167, 275, 349, 361, 426, 514, 614
4137[b]	$(29/2^-, 31/2^-)$	403	—	153, 167, 196, 275, 361, 614
4137[b]	$(29/2^-, 31/2^-)$	276	—	196, 208, 275, 349, 361, 403, 514, 614, 991, 1096
4751[b]	$(33/2^-, 35/2^-)$	614	—	153, 167, 275, 349, 403

[a]Level energies and spin-parities are adopted from [14], where the more accurate energies of respective γ transitions are given.
[b]Tentative assignment made on the basis of coincidence relations with known ^{93}Pd transitions.

known β-delayed γ rays from the two ^{94}Ag isomers, being in coincidence with Compton-scattered γ rays in the applied γ gates. The fact that some intense ^{94}Pd γ transitions also occur in the spectra displayed in the upper and middle panels of Fig. 2 are then due to β background under the proton lines.

To inspect a possible mock up of direct proton emission by contributions from more intense decay branches, several cross-checks were made. In particular, a Si spectrum was generated by shifting the above-mentioned γ-energy gates away from the nominal position of ^{93}Pd transitions by 3 keV. The resulting spectrum does not show any indication of the previously observed peaks (I) and (II). An analogue energy-shift procedure, now applied to the peaks (I) and (II), resulted in a γ spectrum without ^{93}Pd* transitions. One more cross-check was done for events with two Si detectors fired, demanding Si-Si-γ-γ coincidence with the same conditions as in Fig. 1. The resulting Si spectrum does not show any peak, only the low-energy bump around 0.5 MeV. Moreover, no ^{93}Pd γ rays were observed in the corresponding γ spectra selected with the same conditions as in Fig. 2.

The scheme of the direct proton decay of the (21^+) isomer in ^{94}Ag, obtained by combining the results from this work with the ^{93}Pd level scheme [14], is shown in Fig. 3. It is based on the following considerations. The proton-decay branching ratio depends strongly upon the proton energy. A WKB calculation [16] shows, e.g., that

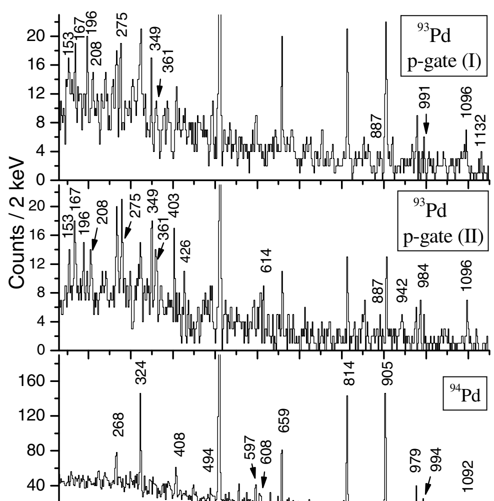

FIG. 2. Energy spectra of γ rays from the decay of the ^{94}Ag isomers, measured in Si-γ-γ coincidence with different conditions. The labels "p-gate (I)" and "p-gate (II)" indicate that protons were selected in the energy range 0.75–0.85 MeV and 0.95–1.05 MeV (upper and middle panels, respectively), and the sum of single-γ gates on ^{93}Pd transitions (listed in Table I) was chosen. The γ spectrum in the lower panel was obtained by selecting charged particles of 0.4–0.7 MeV and the same ^{93}Pd γ gates as those in p-gate (II). Transitions in ^{93}Pd and ^{94}Pd are marked by their energy in keV.

the branching ratio increases by a factor of 3–5 per each 50 keV energy increase above a proton energy of 0.8 MeV. The proton branching ratio is also very sensitive to orbital angular momentum carried away by the proton (ℓ_p), e.g., changing by a factor of 700 per two units shift from $\ell_p = 4$. The yrast line in ^{93}Pd puts a lower limit on the excitation energy of a state with given spin and parity. Combining these effects with the high-spin character of the (21^+) isomer leads to a small window of opportunity for possible direct proton decay populating preferably the ^{93}Pd yrast states. Last but not least, we assumed that both proton branches are from the same (21^+) level, and the respective excitation energies should match. The placement of the proton peak (I) is based on the 1132 keV γ deexcitation of the highest observed $(33/2^+)$ state at 4994 keV [14]. The proton line (II) is placed by using the 349 keV γ ray from the known 3734 keV level [14] and involving the new γ lines (403, 276, and 614 keV) which are ordered to match the known ^{93}Pd level scheme. In such a case, the 349 keV γ peak fed in the proton decay (II) should have about a twice larger intensity in comparison with the branch (I) because it includes two 349 keV γ transitions. This is confirmed by the ratio of the intensities of the 196 and 349 keV γ lines derived from the spectra shown in the upper and middle panels of Fig. 2, which are 1.0(3) and 0.52(17), respectively. The branching ratios of the transitions (I) and (II) were derived, 1.9(5)% and 2.2(4)%, respectively. They were determined by comparing the proton-γ-γ coincidence intensities with the β-delayed proton branches of the ^{94}Ag isomers [9]. From the proton and γ data, compiled in Fig. 3, the excitation energy of the (21^+) isomer was found to be 6.7(5) MeV, assuming a proton separation energy of 0.89(50) MeV for ^{94}Ag [17]. This first directly measured value of excitation energy agrees with the respective estimates obtained in the β-decay study of the (21^+) isomer [9,10]. The levels grouped in the side band in Fig. 3 are possibly of odd parity. Indeed, the 4751 keV level in ^{93}Pd, being populated in direct proton decay with almost the same intensity as the 4994 keV, $(33/2^+)$ state, is unlikely to belong to an even-parity yrast band, and a possible odd-parity assignment may be considered. The

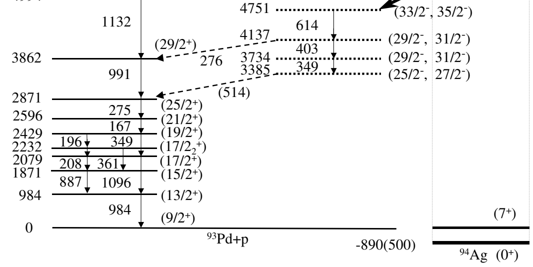

FIG. 3. Direct proton decay of ^{94}Ag (21^+) to excited states in ^{93}Pd. Energies of γ transitions and levels in ^{93}Pd are shown in keV and are given with respect to the ^{93}Pd + p threshold.

$N = 47$ isotones of ^{93}Pd, ^{89}Mo, and ^{91}Ru show odd-parity yrast states with spin-parities $(23/2^- – 33/2^-)$ at excitation energies of 3–5 MeV [18], and one may expect to find similar states in ^{93}Pd. With the odd-parity assumption, we checked possible spins of the 3385, 3734, 4137, and 4551 keV states. The 276 and 514 keV γ rays connecting even- and odd-parity states are likely to be $E1$ transitions. This leads to the very tentative spin ranges shown in Fig. 3.

On the basis of the tentative spin and parity assignments for the ^{93}Pd states populated in direct proton decay, orbital momenta $\ell_p(\mathrm{I}) = 4$ and $\ell_p(\mathrm{II}) = (3 \text{ or } 5)$ are ascribed to the two-proton-decay branches of the (21^+) isomer. We used this information, together with the experimental proton energies, to evaluate the reduced proton width or an "experimental spectroscopic factor" in two ways. First, the respective dimensionless reduced widths $\Theta_p^2 = \gamma_p^2/\gamma_W^2$ are estimated to be 1×10^{-6} and $(1 \times 10^{-11}, 2 \times 10^{-7})$, the reduced proton width γ_p^2 being obtained from the R-matrix partial width $\Gamma_p = 2\gamma_p^2 P_{\ell_p}(E_p)$, and $\gamma_W^2 = 3\ \hbar^2/2Mr^2$ being the respective Wigner estimate of a reduced width due to a single-proton ℓ_p configuration in the parent state [19]. Second, a simple version of the WKB approach [16] was used to estimate experimental spectroscopic factors as ratios between the respective calculated and experimental partial half-lives for proton emission. This estimate yields 1×10^{-6} and 3×10^{-7} for the $\ell_p = 4$ and 5 transitions, respectively. These widths or experimental spectroscopic factors are very small in comparison with those derived from shell-model calculations. For example, a spectroscopic factor of 0.044 is found for the $\ell_p = 4$ branch as an overlap of the single-proton $g_{9/2}$ configuration in the (21^+) isomer with the daughter ^{93}Pd $(33/2^+)$ state in spherical shell-model calculations. The calculations were performed with the code OXBASH [20] considering protons and neutrons within the $\pi, \nu(p_{1/2}, g_{9/2})$ model space and ^{76}Sr as an inert core, the single-particle energies being fixed on the ^{88}Sr states. The empirical residual interaction derived by Gross and Frenkel from nuclei with $N = 48–50$ [21] has been used. The strong quenching of single-proton components in the (21^+) parent state in comparison with those predicted by shell-model calculations of multiplet structures often reflects dominant collective configurations in the parent state (e.g., [22]). The observed fine structure of the proton spectrum might point to a deformation of the (21^+) isomer similar to that deduced for the proton decay of ^{141m}Ho [23]. The possible $\ell_p = 5$ transition might be connected with the occurrence of core excitations across the $N = 50$ shell gap, such as an admixture of the $h_{11/2}$ single-proton orbital in the (21^+) state. The existence of the odd-parity states in ^{93}Pd between 3 and 5 MeV should be checked through more in-beam studies. Further shell-model calculations including particle-hole excitations involving the $\pi, \nu(2p, 2d, 2f, 1g, 1h)$ orbitals are needed.

To summarize, we report direct one-proton decay of the (21^+) isomer in ^{94}Ag into two high-spin daughter states in ^{93}Pd with a strong quenching of the partial decay widths, indicating the importance of the parent state deformation. A refined theoretical analysis of the data may allow one to quantitatively probe the collective structure of the (21^+) and daughter states. The (21^+) isomer in ^{94}Ag has unique properties in the entire chart of nuclides. It features the highest spin that has been observed so far for β-decaying nuclei, and has at least five different decay modes involving high-spin states in the respective daughter nuclei. Finally, the method of multiple γ-ray coincidence, applied for purifying the charged-particle spectrum and yielding an unambiguous isotope assignment of proton-emitting states, has proven to be a promising tool for studies of proton emitters with high spin.

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