Reinvestigation of the Direct Two-Proton Decay of the Long-Lived Isomer ^{94}Ag^m [0.4 s, 6.7 MeV, (21+)]

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An attempt to confirm the reported direct one-proton and two-proton decays of the (21+) isomer at 6.7(5) MeV in ^{94}Ag has been made. The 0.39(4) s half-life of the isomer permitted use of a helium-jet system to transport reaction products from the ^{40}Ca + $^{\mathrm{nat}}$Ni reaction at 197 MeV to a low-background area; 24 gas ΔE-(Si)E detector telescopes were used to identify emitted protons down to 0.4 MeV. No evidence was obtained for two-proton radioactivity with a summed energy of 1.9(1) MeV and a branching ratio of 0.5(3)%. Two groups of one-proton radioactivity from this isomer had also been reported; our data confirm the lower energy group at 0.79(3) MeV with its branching ratio of 1.9(5)%.

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Sometimes proton-neutron coupling preferences near doubly closed nuclear shells cause nuclear states to be caught in ‘‘spin-traps’’—isomeric states of high angular momentum in which rapid gamma ray decay is forbidden—so that other, slower decay processes such as beta-decay or direct low-energy proton emission can then compete. In fact, proton radioactivity [1] was discovered in 1970 as a 1.5% branch in the dominant beta decay of a spin-trap isomer in ^{53}Co (0.25 s, 19/2−). In a series of experiments by the online mass separator group at Gesellschaft für Schwerionenforschung (GSI) in Darmstadt [2–9], the odd $Z =$ odd N nuclide ^{94}Ag has been shown to have such a spin-trap—a long-lived (21+) state at 6.7(5) MeV excitation with a half-life of 0.39(4) s which decays primarily by beta decay and by beta-delayed proton emission. In the more recent publications, Mukha and collaborators have reported additional decay modes for this ^{94}Ag isomer: direct proton emission in 2005 [6] and—most surprisingly—direct two-proton emission in 2006 [8]. Observing both of these latter types of decay from the same nuclear state is unique. [Though single-proton decay from an odd Z nuclide can be expected under suitable conditions, two-proton decay is expected to occur from even Z, very proton-rich nuclides (and as such has been recently observed in ^{45}Fe, e.g., see [10]).] Further, the quantum-mechanical conditions necessary to explain this two-proton decay from an odd Z parent state require a highly deformed prolate—(cigar)—shaped isomer; the two protons must then be emitted simultaneously with a relatively large angular momentum ‘‘either from the same or from opposite ends of the ‘cigar’ [8].’’ Though follow-up measurements related to a more accurate excitation energy of the ^{94}Ag isomer and to the gamma decay scheme in the ^{92}Rh daughter have been reported (and will be discussed later), no experiment to date has confirmed either the direct proton or the direct two-proton emission from this isomer.

The reported two-proton decay branch (19 observed decays) from ^{94}Ag^m [8,9] produces two protons with a total energy of 1.9(1) MeV and a branching ratio of 0.5(3)% (corresponding to a fusion-evaporation cross section of 350 pb). The GSI experiment collected mass-separated data from the ^{58}Ni(^{40}Ca, $p3n$)^{94}Ag reaction at 192 MeV on a tape positioned in the center of a large array of silicon and germanium detectors. Fourfold coincidences between two gamma rays known to lie in the spectrum of the daughter nucleus ^{92}Rh with two charged particles in the silicon detectors (with a lower energy limit of 400 keV) were acquired as the primary data.

Given the quite high beta-particle background in this difficult, low-yield experiment, we wanted to repeat it and *identify* as protons the two charged particles that had been in coincidence with the two triggering gamma rays. We employed a ^{40}Ca beam from the 88-inch cyclotron at the Lawrence Berkeley National Laboratory on a natural nickel target to produce the same activities that were made at GSI. Because of the relatively long half-life of the isomer, we could utilize our helium-jet system [11] to transport the produced radioactivities (with no mass separation) from the bombardment area to a low-background counting chamber.

Our setup is shown in Fig. 1. The 197 MeV ^{40}Ca beam enters an isolated, water-cooled target chamber filled with 1.3 atm of helium plus ethylene glycol as an additive. The presence of the beam creates an aerosol from the additive to which the nuclides recoiling from the target stick with a 50(20)% efficiency for heavy ion bombardments. These aerosols are collected by four capillaries uniformly distributed over the recoil range which feed a single main capillary (1.3 mm i.d., ~4 m long). The activities are deposited in 0.20(5) s on a slowly rotating catcher wheel to remove long-lived beta emitters. This collection spot is viewed by an array of 24 gas $\Delta E1$-gas $\Delta E2$-(Si)E detectors [12]

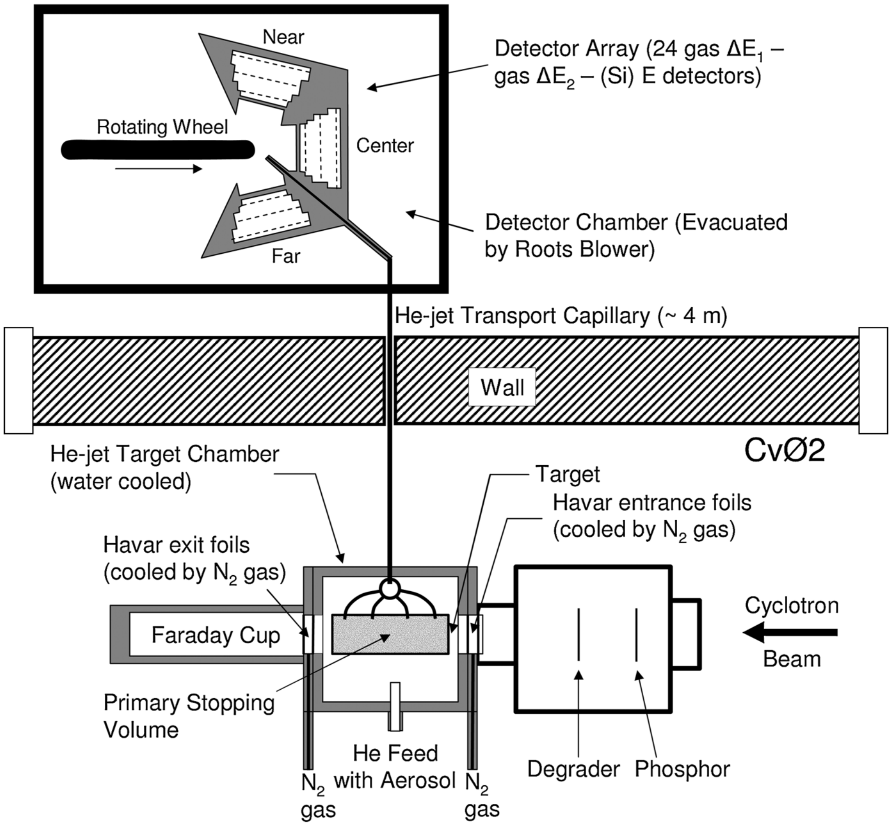

FIG. 1. A schematic diagram of the experimental setup.

capable of identifying protons down to 400 keV. These detector telescopes are arranged in three vertical blocks (denoted near, center, and far) each having two identical "modules" denoted "top" and "bottom"; each module consists of a 300 μm silicon wafer divided like a ladder into four E detectors, all of which share the same gas $\Delta E1$ and gas $\Delta E2$ detectors. To illustrate the physical dimensions of the detector setup, each E detector is 20 mm horizontal by 7 mm vertical and the minimum distance between the collection spot and the closest E detector(s) is 28 mm. Individual telescopes are then identified numerically beginning from the center line between the modules. (An example would be that near top 1 and near bottom 1 lie closest to the center line.)

These 24 detector telescopes were then separately calibrated by using beta-delayed protons from the decay of 0.22 s ^{25}Si, produced via the reaction ^{24}Mg(^{3}He, $2n$)^{25}Si using a 2 μA beam of 40 MeV ^{3}He on a natural Mg target. The well-known proton groups [13] from 387 keV to 5.4 MeV were observed with low background in all the telescopes using gas $\Delta E2$-(Si)E coincidence techniques. Though two separate gas ΔE-E proton identifications had been planned for each event of interest to further reduce the beta-particle background [12], these ^{3}He measurements showed that there were problems with the gas gains of the $\Delta E1$ detectors, so that only a single identification of protons using the gas $\Delta E2$-(Si)E coincidences was utilized. (However, the quality of the gas $\Delta E2$-(Si)E identifications alone was quite good and was similar to that shown in Fig. 3(d) of [14] for many of the telescopes.)

Data were recorded from a 63.6 h run with 100 pnA of 197 MeV ^{40}Ca incident on a thick (4 mg/cm^2) natural nickel target (68.1% ^{58}Ni). Overall, our anticipated two-proton coincidence yield—based on the total ^{40}Ca beam on ^{58}Ni in the target, a 50% transport efficiency, and the solid angle of our telescope array—was expected to be comparable to that in the GSI experiment.

Figure 2 shows the identified proton energy spectrum for telescope near top 2 for the entire run. This spectrum is dominated by the beta-delayed protons from 2.0 s ^{95}Ag from the ^{58}Ni(^{40}Ca, $p2n$) reaction which peaks near 2.4 MeV [2] (since we do not have mass separation). There is a small peak near 0.8 MeV in Fig. 2 which also appears in telescopes near bottom 4, center bottom 4, far top 2 and far top 3. [Our ability to observe weak branches of direct low-energy protons above beta-delayed proton background in an intense flux of beta particles which extends to above 2 MeV (see [4]) varies with the behavior of each particular telescope. This does not affect our ability to see proton-proton coincidences at these low energies.] Figure 3 then shows the sum of the data from these five telescopes. The peak marked as (I) in Fig. 3 lies at 0.79(2) MeV and has a yield corresponding to 1.6(7) nb. This (within errors) is exactly the energy [0.79(3) MeV]

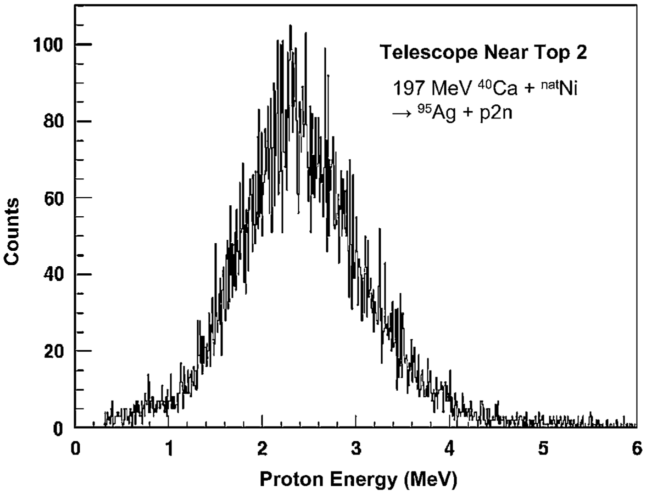

FIG. 2. The identified proton spectrum from 197 MeV ^{40}Ca + $^{\mathrm{nat}}$Ni for the detector near top 2.

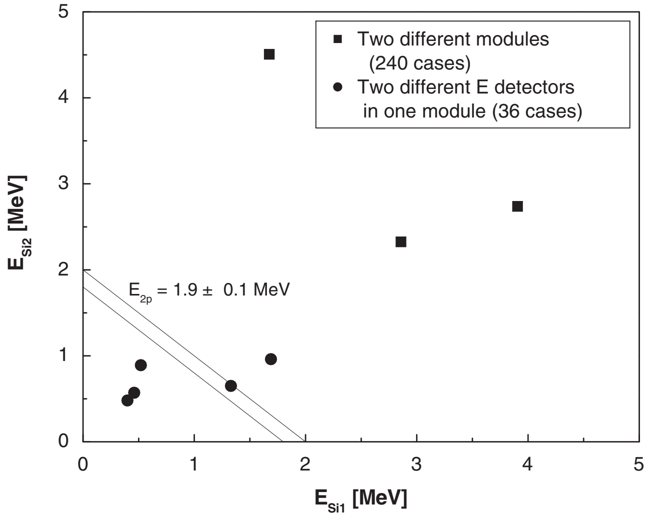

FIG. 4. The proton-proton coincidence spectrum with events from the two different coincidence categories indicated. See text.

and the cross section [~1.3 nb] of the lower energy of the two direct one-proton decay branches from the ^{94}Ag(21+) isomer [6] produced by the ^{58}Ni(^{40}Ca, $p3n$)^{94}Ag^m reaction. Interestingly, Fig. 3 shows no clear indication above the beta-delayed proton background of the higher energy, direct one-proton decay branch at 1.01(3) MeV, marked as (II), which has a slightly higher reported yield.

All the proton-proton coincidence events from the run are displayed in Fig. 4. Given the nature of our setup, the data analysis for the 276 possible coincidence combinations for the 24 telescope array is broken down into two categories: (a) coincidences from events in *different* modules (240 cases) which favor larger angles between the protons and (b) coincidences from events from two different E detectors in the *same* module (36 cases) which are restricted to small angles. (Since proton-proton coincidences within the same module have separate E signals but a common, summed $\Delta E2$ signal, a higher threshold to discriminate against beta particles can be set on the $\Delta E2$ signals when performing the analysis in this category.) Only one two-proton coincidence event is observed with a summed energy in the range of 1.8–1.95 MeV reported by Mukha *et al.* [8] for the decay of ^{94}Ag^m. Overall, the observed coincidences are reasonably consistent with predictions of chance coincidence rates.

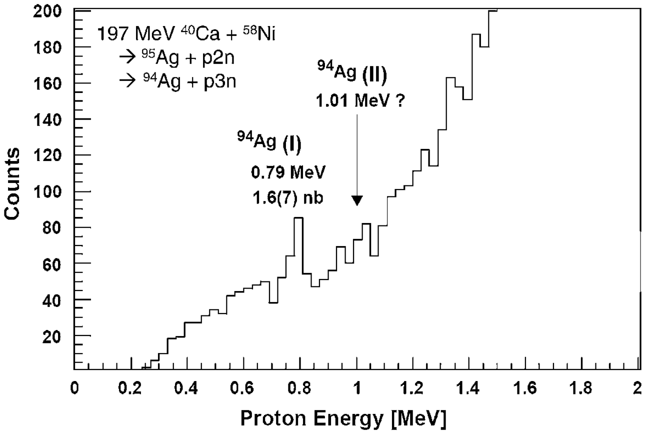

FIG. 3. Summed energy spectra up to ~1.5 MeV from the five telescopes specified in the text.

Figure 5 then shows the results of a GEANT 4 Monte Carlo simulation of the detection efficiency versus angle for two possible emitted protons for the two categories, as well as their sum. In order to estimate the expected number of coincidences that we should have seen based on the GSI partial fusion-evaporation cross-section of 350 (210) pb for this two-proton decay, we will consider (a) isotropic emission from the isomer and (b) emission at a 10° relative angle [about 65% of the two-proton decays are

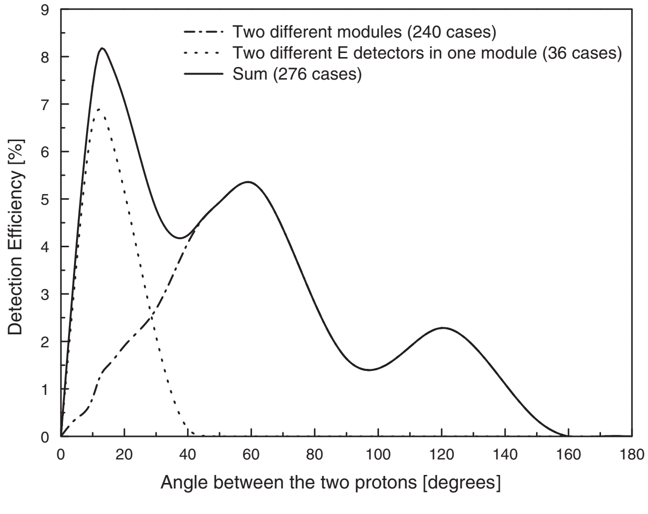

FIG. 5. The results of a GEANT 4 Monte Carlo simulation of the detection efficiency versus angle for two possible emitted protons for the two different coincidence categories. See text.

postulated to occur from the same end of a cigar-shaped ^{94}Ag isomeric state, see Figs. 3(a) and 4(b) in Ref. [8]]. For isotropic emission with our overall efficiency of 2.9%, we should have observed $13+/-8$ two-proton decays; for emission at a small angle (10°) we should have seen $22+/-13$ decays. [Reference [7] states that *beta-delayed* two-proton emission from this isomer was also observed by gating on low-lying transitions in ^{92}Ru: the reported cross section was 140(140) pb. Any such protons would probably be emitted isotropically and could perhaps be present in Fig. 4; however, no details on the energies of the emitted protons are known to us.]

Even higher expected numbers of two-proton coincidences in our data could arise when we take into account recent results on the level structure of the two-proton daughter ^{92}Rh by Pechenaya *et al.* [15–18]. The GSI data were taken by requiring two of eight possible γ rays from excited states in ^{92}Rh to be in coincidence with the two charged particles. However, of the original eight triggering γ rays, the three at 307, 565, and 833 keV are either not observed as excited states in the ^{92}Rh daughter or lie too high in excitation energy to be relevant. If these three γ rays used by GSI were actually random, then, for isotropic emission of the two protons, we would have expected to observe $37+/-22$ events and for emission at a small angle, $62+/-37$ events.

A recent report by Kankainen *et al.* [19] using mass data from a Penning trap spectrometer to determine the excitation of the ^{94}Ag isomer finds that the direct one-proton decay data (isomer around 7.0 MeV) and the two-proton decay data (isomer around 8.4 MeV) disagree with one another. As noted above, we confirmed the energy of one of the two reported direct proton groups based on the (21+) isomer lying near 6.7(5) MeV [6]. Should there somehow be two isomers at high excitation in ^{94}Ag, we would have produced both of them since our experimental conditions directly parallel those at GSI.

In conclusion, we find no evidence to support the decay [8] of the long-lived isomer ^{94}Ag^{m} [0.4 s, 6.7 MeV, (21+)] by direct emission of two protons with a summed energy of 1.9(1) MeV and a branching ratio of 0.5(3)%. This isomer was also reported [6] as being single-proton radioactive with two such decay branches—one with a proton energy of 0.79(3) MeV and a branching ratio of 1.9(5)% and the other with an energy of 1.01(3) MeV and a branching ratio of 2.2(4)%. Our data confirm the energy and branching ratio of the lower energy group; we do not observe the higher energy group, which may be due to our lack of mass separation and the beta-delayed proton background in the singles data.

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