Search for ground state proton emission from ^{65}As and ^{69}Br

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The ground state proton decays of ^{65}As and ^{69}Br have been searched for in ^{28}Si and ^{32}S bombardments of a natural calcium target. These studies employed a newly developed rapidly rotating recoil-catcher wheel and a low-energy particle-identification telescope. No proton groups that could be assigned to either of these nuclides were observed. The minimum detectable limits indicate that ^{65}As and ^{69}Br either decay predominantly by beta emission or have half-lives less than 100 μs. The overall evidence strongly indicates that ^{65}As predominantly beta decays.

INTRODUCTION

Nuclear decay by the emission of a proton from the ground state defines the proton drip line. Although it is expected to be the dominant decay mode for the most neutron-deficient odd-Z nuclei, intensive searches in several mass regions[1] have, to date, identified only four ground state (g.s.) proton emitters; ^{109}I,[2,3] ^{113}Cs,[2,3] ^{147}Tm,[4] and ^{151}Lu.[5] This interesting decay mode is rarely identified because g.s. proton decay can be observed only in very special cases on the nuclear mass surface in which the available proton decay energy combines with the angular momentum and Coulomb barriers in such a way that the nucleus lives long enough to be observed in the laboratory and yet decays quickly enough to compete with beta decay.

In order to further characterize this unique decay mode, we undertook a search for the g.s. proton decays of the $T_z=-\frac{1}{2}$ nuclei ^{65}As and ^{69}Br. These are the lightest members of the $T_z=-\frac{1}{2}$, $A=4n+1$ chain which are most likely unbound to proton decay. The predicted proton separation energies of ^{57}Cu, ^{61}Ga, ^{65}As, and ^{69}Br from several appropriate mass models are given in Table I.[6] All seven models listed in Table I are in agreement with the closest measured mass value in this region in that they predict ^{57}Cu to be stable towards proton decay. Moreover, the predicted proton separation energies from these models are in good agreement with the observed proton decay energies for the two lightest known g.s. proton emitters, ^{109}I and ^{113}Cs. All of the models listed in Table I indicate that the next member of the $T_z=-\frac{1}{2}$, $A=4n+1$ chain, ^{61}Ga, is bound towards proton emission ($S_p=-1$ keV is, for all practical purposes, bound). On the other hand, four of the seven mass models predict that ^{65}As is unbound towards proton decay and five of the seven models indicate that ^{69}Br is unbound towards proton decay.

In addition to delineating the proton drip line in this mass region, determining the proton stability of these nuclei is important for astrophysical r-p process calculations.[7] Briefly, the relatively long half-lives of ^{64}Ge (64 s) and ^{68}Se (34 s) compared to the time scale of the r-p reaction chain that leads to higher masses means that the r-p chain must go through the ^{64}Ge$(p,\gamma)^{65}$As and ^{68}Se$(p,\gamma)^{69}$Br reactions. If, however, either ^{65}As or ^{69}Br is a proton emitter, the (p,γ) reactions cannot occur and the chain is effectively broken.

The partial width for g.s. proton decay, Γ, can be written as

$$\Gamma=2P_l\gamma^2 ,$$

where P_l is the penetrability of a proton with angular momentum l through the Coulomb and angular momentum barriers and γ^2 is the reduced width for the proton decay channel. The Bohr approximation[8] can be used as an estimate of the proton reduced width along with the known or estimated available proton decay energy to predict the proton partial half-life. For the known g.s. proton emitters near $A=150$ this approximation underestimates the proton partial half-lives by a factor of 4 and for the two cases near $A=110$ this approach underestimates the observed values by factors of 11 and 35 for ^{109}I and ^{113}Cs, respectively. A plot of the predicted proton partial half-life values for ^{65}As and ^{69}Br from such a calculation is given in Fig. 1. The angular momentum of $l=1$ for the emitted proton used in the calculations shown in Fig. 1 is based upon a proposed spin and parity of $\frac{3}{2}^-$ for the g.s. of both ^{65}As and ^{69}Br. The g.s. spin and parity of the $T_z=+\frac{1}{2}$ mirror nuclei ^{65}Ge (Ref. 9) and ^{69}Se (Ref. 10) are $\frac{3}{2}^-$.

It is clear from the curves given in Fig. 1 that the available proton decay energy must fall within a rather narrow energy window in order for g.s. proton emission to be observed. Assuming that ^{65}As is unbound towards proton emission by 200 to 300 keV (Table I), then our calculations indicate that its proton partial half-life could range from 3 min to 1 ms. Similarly, if ^{69}Br is unbound towards proton emission by 200 to 700 keV (Table I), its proton partial half-life could fall anywhere between 3 h and 10 ps. With this in mind, we set out to search for the g.s. proton decays of these two nuclides in the half-life range of 100 μs to 100 ms. It is expected that above 100

TABLE I. Proton separation energies deduced from the updated list of atomic mass predictions (in MeV). (See Ref. 6.)

Nuclide	Experimental	Moller, Nix	Moller *et al.*	Comay, Kelson, Zidon	Tachibana *et al.*	Janecke, Masson	Masson, Janecke	Wapstra, Audi, Hoekstra,
^{113}Cs	-0.967 ± 0.004	-0.971	-0.891	-0.781	-0.871	-0.801	-0.741	-0.961
^{109}I	-0.819 ± 0.005	-1.251	-0.981	-0.571	-0.461	-0.691	-0.611	-0.821
^{69}Br		-0.111	-0.131	-0.531	0.029	-0.661	-0.631	0.009
^{65}As		0.039	0.079	-0.181	-0.011	-0.261	-0.321	0.369
^{61}Ga		-0.001	0.009	0.299	0.449	0.269	0.339	0.644
^{57}Cu		0.738	0.429	0.429	0.889	0.819	0.789	1.289

ms positron decay would be the dominant decay mode. During the course of our investigation, two other unsuccessful searches for the g.s. proton decays of these two nuclides were reported.[11,12]

EXPERIMENTAL PROCEDURE

Our searches for the g.s. proton decays of ^{65}As and ^{69}Br were performed using beams produced by the ECR-injected 88-Inch Cyclotron at Lawrence Berkeley Laboratory. These experiments were initiated with the development of (1) a rapidly rotating recoil-catcher wheel and (2) a low-energy particle-identification telescope. The recoil-catcher wheel provides the means for rapidly collecting and transporting short-lived nuclear reaction products and the new particle-identification telescope can identify low-energy protons in a high radiation environment.

The rapidly rotating recoil-catcher wheel is illustrated in Fig. 2 and described in detail in Ref. 13. Briefly, a fraction of the reaction products which recoil out of the target were stopped in the 200 μg/cm^2 aluminum catcher foils which were fastened to the circumference of the wheel shown in Fig. 2. The target ladder and wheel are inclined at an angle of 20° with respect to the beam in order to maximize the catcher efficiency of the foils while minimizing the recoil range effects on the proton energy resolution. Because the detector electronics must be disabled while the beam is on target to eliminate the observation of beam-related events, the beam is pulsed by turning on deflection plates in the injection beam line between the ECR and the cyclotron; the beam-on time was set equal to the beam-off time. The wheel can be rotated at speeds ranging from 20 to 5000 rpm yielding beam cycle times (beam-on time plus beam-off time) that range from 250 to 1 ms, respectively.

The detector telescopes used in these experiments were developed to enable us to measure low-energy protons in a high radiation environment. As can be seen from Fig. 3, the new design consists of a gas ΔE detector and a 300 μm Si E detector. The active volume of the gas ΔE is defined by two wire grids 3.0 mm on either side of a 70 μg/cm^2 thick nickel foil. The wire grids are grounded

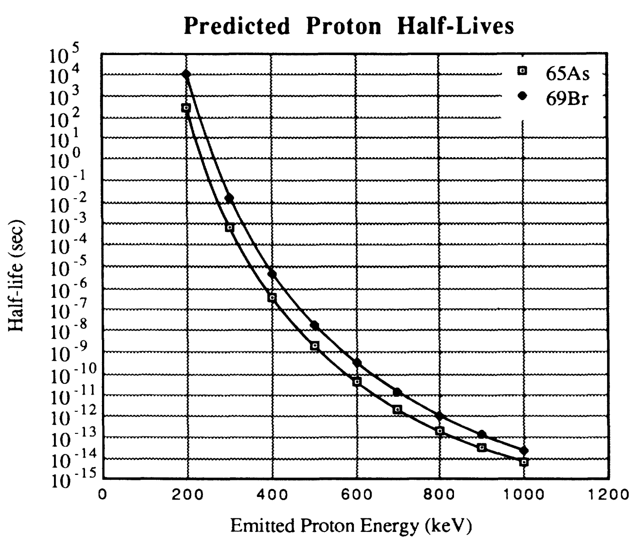

FIG. 1. The proton half-life for ^{65}As and ^{69}Br as a function of emitted proton energy. These curves were calculated using the Bohr approximation for the proton reduced width and assuming that the protons are emitted with an angular momentum of $l=1$.

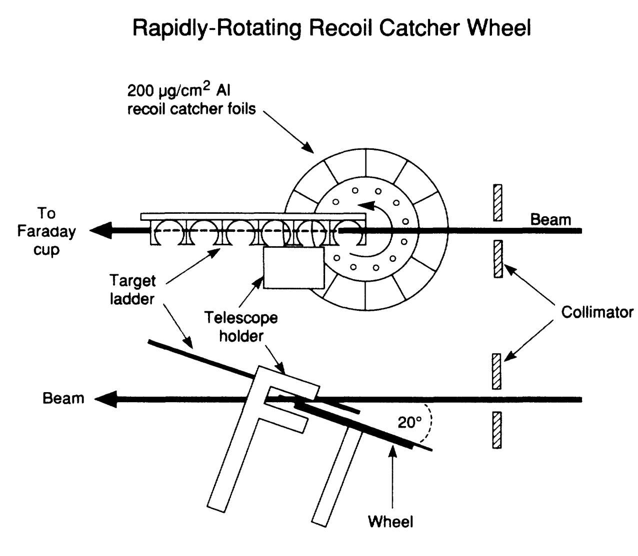

FIG. 2. A schematic diagram of the rapidly rotating recoil-catcher wheel used to search for the g.s. proton decay of ^{65}As and ^{69}Br (see text).

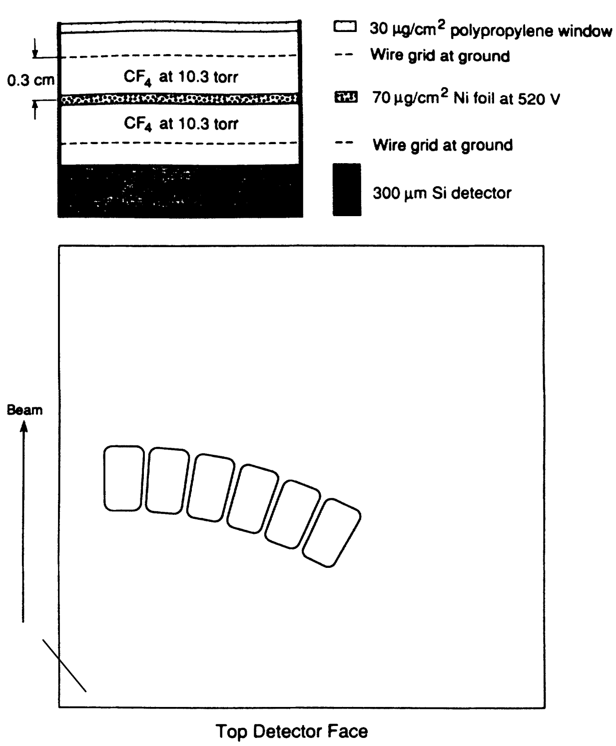

FIG. 3. A cross section of a single low-energy particle-identification telescope and the top view of one telescope array used to search for the g.s. proton decay of ^{65}As and ^{69}Br.

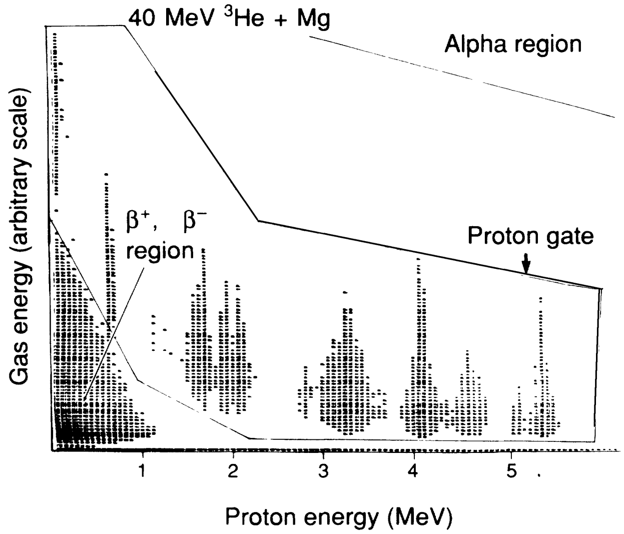

FIG. 4. A two-dimensional ΔE-E spectrum from one of the particle-identification telescopes showing the proton band. This spectrum was acquired from the beta-delayed proton decay of ^{25}Si produced by the reaction of a 40 MeV ^{3}He beam with a natural magnesium target.

and the nickel electrode is maintained at a potential of 520 volts. This high electric field (≈ 1600 V/cm) places the gas detector just below the avalanche region and provides the gas amplification necessary for particle identification. Isobutane, Freon-14, propane, and an argon-methane mixture were tested as gases for the ΔE counter and it was found that Freon-14 yielded the best gas amplification for protons, was the least sensitive to high-energy positrons and electrons, and gave the best shaped signal. As can be seen from the representative two-dimensional ΔE-E spectrum shown in Fig. 4, the gas gain achieved with this telescope design clearly separates the proton band from the β^+/secondary e^- band.

With the new low-energy particle-identification telescopes, the signals from the ΔE counters are used for particle identification but the final energy signal for a proton is taken solely from the silicon E counter as the protons lose so little energy in the gas ΔE counter. For example, a 300 keV proton loses approximately 15 keV in the gas of the ΔE counter and a 4 MeV proton loses approximately 8 keV of energy in the gas. This telescope design has been used to successfully measure protons whose energies range from 200 keV to 5.5 MeV in a β^+ background of $\approx 10^5$ cps. The new design has the advantage over the solid-state particle-identification telescopes used in previous low-energy proton measurements, e.g., Refs. 14 and 15, in that it allows us to measure protons with energies less than 700 keV on an event-by-event basis.

For these searches, two arrays, each containing six of the new low-energy particle-identification telescopes, were constructed. These arrays (Fig. 3) were placed 3 mm above and below the circumference of the catcher wheel (Fig. 2). The distance between the beam and the detector arrays was adjusted such that the leading edge of the activated portion of the catcher foils reached the middle of the first telescope of each array when the beam was turned off. Use of the six-detector arrays served to (1) increase the overall detection efficiency, and (2) permit us to make a half-life determination of an observed proton group at a single wheel speed.

EXPERIMENTAL RESULTS AND DISCUSSION

The overall performance of the experimental system was established by measuring the beta-delayed proton decay of ^{25}Si ($t_{1/2}=220$ ms).[16–18] This nuclide was produced via the ^{24}Mg(^{3}He,$2n$)^{25}Si reaction at 40 MeV on a 1.5 mg/cm^2 natural magnesium target. The activity was measured at a wheel speed of 20 rpm which corresponds to a 250 ms beam cycle (125 ms beam on–125 ms beam off). The ^{25}Si spectrum presented in Fig. 5 was generated simply by gating on the proton band in the two-dimensional ΔE-E spectrum shown in Fig. 4. The results of these β^+-p measurements were used to determine that the product of the collection efficiency for the ^{25}Si recoil nuclei and the detection efficiency of the telescopes for protons whose energies range from 370 keV to 5.5 MeV was $(6\pm2)\%$. The "overall system" efficiency was calculated using the measured cross section of 150 μb for the (^{3}He,$2n$) reaction on magnesium[17] in conjunction with the absolute proton intensities reported in Refs. 16 and 17. The calculation took into account (1) an effective target thickness (2) the decay of the ^{25}Si in transport, and (3)

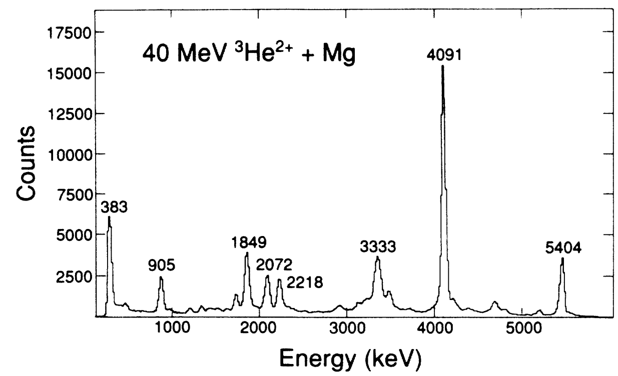

FIG. 5. The beta-delayed proton spectrum resulting from a projection of the proton band shown in Fig. 4.

the solid angle subtended by the detector telescopes. The observation that the detector efficiencies remained constant over this proton energy range is consistent with previous β^+-p measurements of this telescope design.[16]

In order to determine whether or not the overall system efficiency changed when heavier bombarding beams were used, the β^+-delayed proton spectrum resulting from the bombardment of a 1.75 mg/cm^2 natural calcium target with an 180 MeV ^{14}N beam utilizing a wheel speed of 30 rpm (170 ms beam cycle) is shown in Fig. 6. Proton groups from the decays of two $T_z=-\frac{3}{2}$, $A=4n+1$ β^+-p emitters ^{37}Ca ($t_{1/2}=170$ ms) and ^{41}Ti ($t_{1/2}=80$ ms) are identified in the spectrum. By using the production cross sections of 0.5 and 3 μb for ^{37}Ca and ^{41}Ti, respectively, from the statistical model fusion-evaporation code ALICE,[19] in conjunction with the absolute proton intensities reported in Ref. 20, we determined that the overall system efficiency with the heavier bombarding beam was again $(6\pm2)\%$. Although these calculations rely upon the absolute value of predicted cross sections (the production cross sections for these reactions have not been measured), it has been observed that, in this region, ALICE predictions are good to, at worst, an order of magnitude. Moreover, the fact that both ^{37}Ca and ^{41}Ti gave the same result as ^{25}Si suggests that the predicted cross sections are consistent.

After the system performance tests and calibrations were completed, a 1.75 mg/cm^2 natural calcium target was bombarded with a 200 MeV ^{32}S beam in order to search for the g.s. proton decays of the $T_z=-\frac{1}{2}$ nuclides ^{69}Br and ^{65}As via the ^{40}Ca(^{32}S,$p2n$)^{69}Br and ^{40}Ca(^{32}S,$\alpha p2n$)^{65}As reactions. At this beam energy, ALICE predicts that the cross sections for the production of ^{69}Br and ^{65}As via these reactions are 150 and 110 μb, respectively. The bombardments were carried out at wheel speeds of 5000 rpm (1 ms beam cycle) and 1250 rpm (4 ms beam cycle) with beam currents ranging from 130 p nA to 150 p nA. The total integrated beam on target in the 5000 and 1250 rpm bombardments was 3.4 and 4.8 mC, respectively. After this, a cross bombardment of the calcium target with a 175 MeV ^{28}Si beam was performed in order to search for ^{65}As via the ^{40}Ca(^{28}Si,$p2n$)^{65}As reaction. The predicted cross section

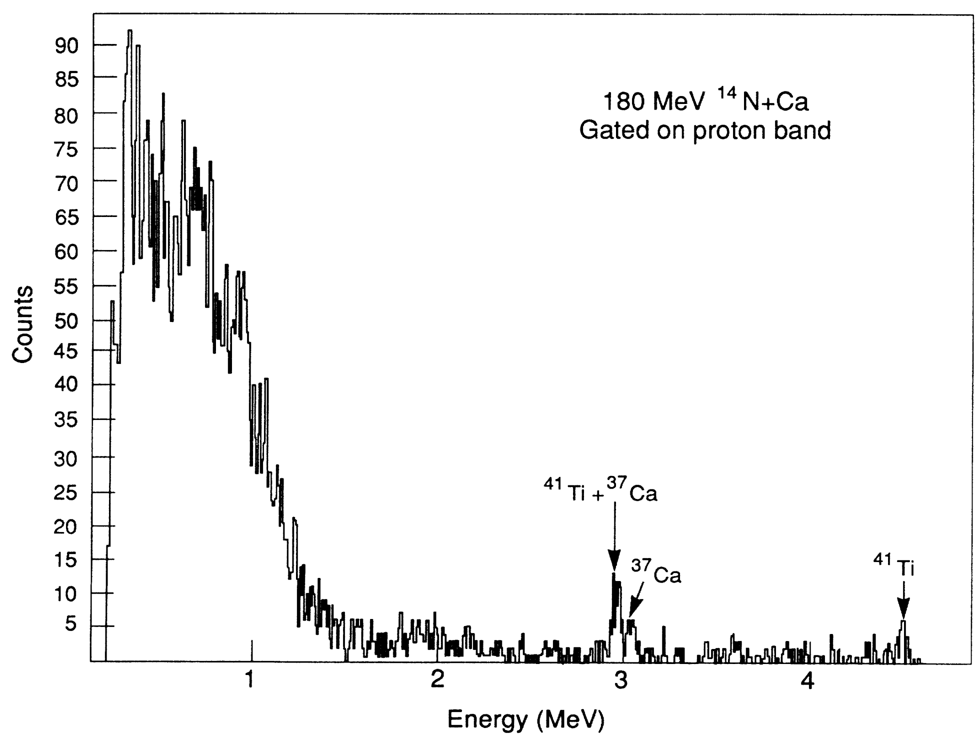

FIG. 6. The beta-delayed proton spectrum arising from the bombardment of a 1.75 mg/cm^2 natural calcium target with a 400 p nA 180 MeV ^{14}N beam.

for this reaction at 175 MeV is 120 μb. The ^{28}Si beam current ranged from 80 to 100 p nA and the amount of integrated beam at each wheel speed was 1.4 mC at 5000 rpm and 460 μC at 1250 rpm.

The ^{32}S+Ca bombardments at 5000 and 1250 rpm revealed no proton groups in any telescope which could be assigned to either ^{65}As or ^{69}Br g.s. proton decay. An example of the proton spectrum obtained in the ^{32}S bombardments is shown in Fig. 7. From the overall system efficiency determined in the two calibration experiments, the number of counts that would have been seen given a specific cross section and half-life has been estimated and is shown in Fig. 7 as a dashed peak centered at 350 keV. The total number of counts expected in the first three telescopes of an array from the 5000 and 1250 rpm experiments are plotted in Figs. 8(a) and 8(b) as a function of reaction product half-life. These curves were calculated assuming a 100% proton decay branch and the reaction cross sections given in the figures. In the half-life range of 10 μs to 1 ms, a 100% g.s. proton branch is a good estimate as beta decay half-lives are expected to be at least 10 ms.

From the expected count rate values, upper and lower limits can be set for the ^{65}As and ^{69}Br g.s., proton partial half-lives for a given production cross section. The minimum number of counts, N_x, that must be observed in a peak to meet a 95% confidence level is typically taken as

$$N_x = 3(N_B)^{1/2} ,$$

where N_B is the number of background counts under the peak.[21] In both the 5000 and 1250 rpm experiments, the backgrounds observed in the sum spectra obtained by adding the data from the first three telescopes in an array yields a minimum detectable limit on the order of 100 counts for a proton peak in the energy range of 200–700 keV. The width of a proton peak in this energy range was taken as the width of the 383 keV proton peak observed in the beta-delayed proton decay of ^{25}Si. Application of this minimum detectable limit to the expected number of counts in the three-telescope sum spectrum from the 1250 rpm bombardment indicates that, if the production cross section for either ^{65}As or ^{69}Br is on the order of 150 μb as predicted by ALICE, then ^{65}As and ^{69}Br must have proton half-lives shorter than 300 μs or longer than 30 ms (again assuming 100% g.s. proton decay

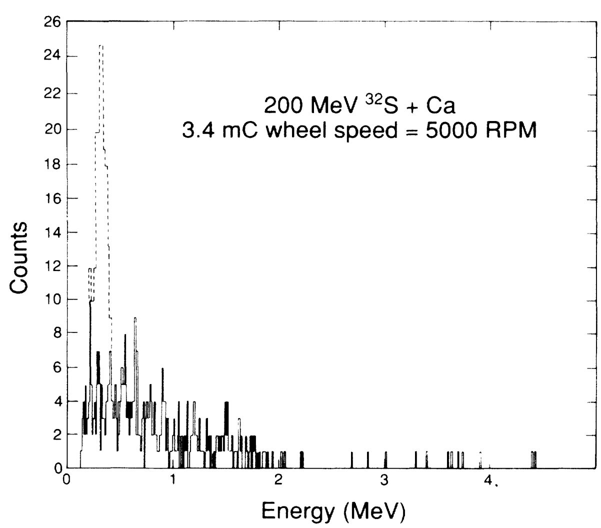

FIG. 7. A typical single telescope spectrum obtained from the ^{32}S+Ca bombardment with a wheel speed of 5000 rpm. The dashed peak illustrates the minimum requirement for an observable proton peak (see text).

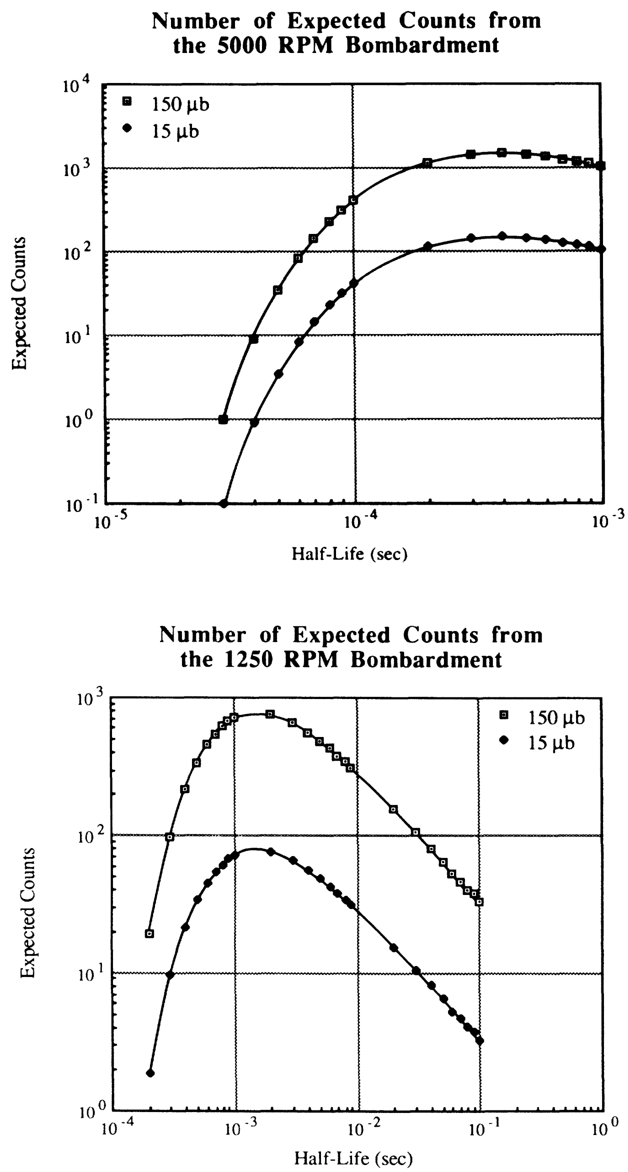

FIG. 8. (a) A plot of the number of counts expected in the sum of the first three telescopes of a detector array as a function of half-life for a 100% g.s. proton emitter and a wheel speed of 5000 rpm. Approximately 100 counts would have had to be seen in order to identify a peak at the 95% confidence level. (b) Same plot as in (a), but for a wheel speed of 1250 rpm. Again, approximately 100 counts would have had to be seen in order to identify a peak at the 95% confidence level.

branch). Furthermore, a proton peak would have been observed in the 5000 rpm experiment if either ^{65}As or ^{69}Br had half-lives in the range of 60 μs to >1 ms. Although the absence of proton peaks in these experiments could be a result of production cross sections much below that predicted [see Figs. 8(a) and 8(b)], we believe that the absence of any ^{65}As or ^{69}Br proton activity is due to the fact that their half-lives lie outside the above ranges. The reaction cross sections predicted by ALICE are within an order of magnitude of the known experimental (HI,$p2n$) cross section values in this mass region.

The ^{28}Si+Ca bombardments at 5000 and 1250 rpm also revealed no proton groups in any telescope which could be attributed to the g.s. proton decay of ^{65}As. By making a similar set of statistical arguments to those made for the sulfur bombardments, the absence of a proton peak in the silicon bombardments at 5000 and 1250 rpm can again be used to assign either a lower half-life limit of 100 μs or an upper half-life limit of 40 ms.

In order to investigate the possibility that ^{65}As decays predominantly by beta emission but also with a weak proton branch, the ^{28}Si+Ca bombardment was performed again at a wheel speed of 50 rpm (a total integrated beam of 5.4 mC). This wheel speed, corresponding to a 100 ms beam cycle, permitted the detection of a g.s. proton decay branch in nuclei whose half-lives were consistent with the beta decay process (≥ 10 ms). No low-energy proton peaks which could be attributed to the decay of ^{65}As were observed. The number of protons from ^{65}As which were expected to be observed with our system was calculated in the same manner as for the 5000 and 1250 rpm experiments. This time, however, various direct proton decay branches were used. In these calculations, if a 50 ms β^+ half-life was assumed along with the predicted ALICE cross section of 120 μb, a g.s. proton decay branch would be less than 0.5%.

CONCLUSION

From our search for the g.s. proton decays of ^{65}As and ^{69}Br, we have concluded that ^{65}As and ^{69}Br either decay predominantly by beta emission, decay by proton emission with half-lives less than 100 μs, or have production cross sections mush less than predicted by ALICE calculations. These data refer to protons in the energy range of 200–700 keV (though the detectors go to several MeV).

During the preparation of this paper, two other searches for the proton decays of these nuclei were reported.[11,12] Both of these investigations also employed the ^{32}S+Ca and ^{28}Si+Ca reactions. The measurements of Hourani *et al.*[12] were similar to ours in that they covered the proton energy range of 250–600 keV and a half-life range of 10 μs to 100 ms, but had a lower production cross section limit of 1 μb. Similar half-life and production cross section limits were also reported by Hotchkis *et al.*[11] In these latter measurements, however, the proton energy cutoff was higher (≈ 500 keV) due to the large beta background seen in the particle detector. The results of these two investigations are consistent with our observations in that no proton peaks which could be assigned to the g.s. proton decay of ^{65}As and ^{69}Br were observed.

Taking these composite negative results for ^{65}As, along with (1) the theoretical mass data of Table I which predict either a bound nuclide or a *maximum* proton separation energy of 321 keV, and (2) the barrier penetration calculations of Fig. 1 which show a minimum half-life for an $l=1$, 321 keV proton of ~ 100 μs (for the Bohr approximation reduced width,[8] which is much higher than that observed for the other known g.s. proton emitters), it would seem that these results strongly indicate ^{65}As is predominantly a beta emitter, which should be taken into account in astrophysical *r-p* process calculations. Given the much greater spread in predicted S_p values for ^{69}Br, no similar conclusion can be drawn.

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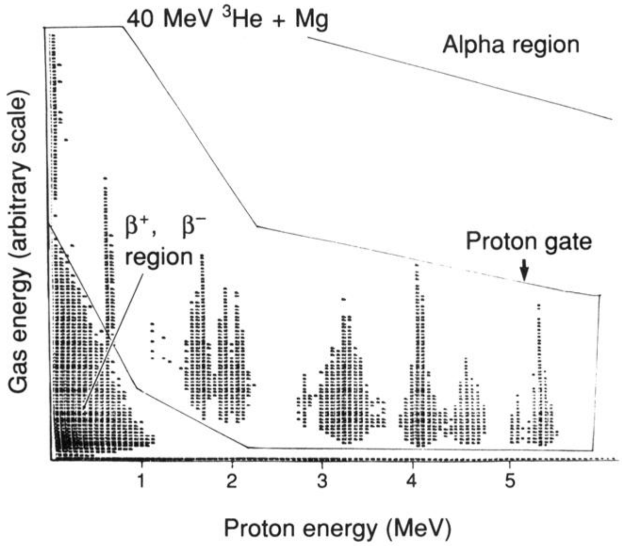

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