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Exotic Decays at the Proton Drip Line

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Abstract:
In this paper we review the general physics learned at the proton drip line. The extraordinary experimental problems which must be overcome are discussed in addition to several novel solutions. Finally, results of several recent experiments to look for ground state one- and two-proton emission are discussed.

I. Introduction

Studies of light proton-rich nuclei have attempted to answer many fundamental nuclear physics questions. Many of these questions can be answered only because the proton drip line is accessible to experimental probes. This has permitted rigorous testing of nuclear models which derive quantities such as atomic masses, level structures and half-lives. Experiments in these light proton rich nuclei have progressed from observations of standard beta and gamma decay to the now commonplace beta-delayed proton emission. This first observation of beta-delayed particle decay has now been extended to beta-delayed multi-particle decay. Major goals of these studies are to understand the decay mechanisms of these more exotic decays and to utilize their relative uniqueness to probe the underlying nuclear structure of nuclides which otherwise would be impossible to resolve due to the simultaneous and copious production of nuclei which only exhibit standard beta-gamma decay. Although these standard beta-gamma measurements are still extremely interesting, the frontier is now at the proton drip line where tests of the predictive powers of these nuclear models is most rigorous. Ground state proton decay defines the drip line. Concerted searches for this rare decay mode have, however, yielded only four examples, $^{151}$Lu, $^{147}$Tm, $^{113}$Cs, and $^{109}$I; these discoveries were made more than ten years after the original discovery of proton decay from a high-spin isomer in $^{53}$Co. Many experiments are currently searching for new examples of this decay mode and some of these will be discussed herein.
The experimental difficulties associated with ground state proton decay studies are magnified in searches for another extremely exotic and rare decay mode, ground state two-proton decay; it has never been seen, only predicted. To obtain adequate mass predictions to search for such a decay requires not only an extraordinary knowledge of the mass surface, but also some insight into the decay mechanism. However, this latter item is extremely difficult to predict because of the lack of any prior examples of a similar nature. In this paper we explore the methodology necessary to search for these exotic decays and we review the status of experiments at the proton drip line.

II. Exotic Decays At/Near the Drip Line

Exotic decays of proton-rich nuclei can easily be divided into beta-delayed particle and direct particle decays. Although recent reviews of beta-delayed proton decay and general properties of proton-rich nuclei cover these topics in much greater detail, we will give a general overview for completeness. Beta-delayed particle emission can be viewed simply as a process where beta decay proceeds to states in the daughter nucleus which are unbound to emission of that particle. In its simplest format, emission of this particle is governed solely by the Coulomb and angular momentum barriers which must be traversed.

Figure 1 depicts the generic energetics of a beta-delayed proton emitter. Additionally, Fig. 1 shows the isobaric analog state unbound to proton emission. When this criterion is met, we say that that nuclide is a strong beta-delayed proton emitter; otherwise it would be considered a weak delayed proton emitter. The first heavily investigated sequence of strong beta-delayed proton emitters was the $A = 4n+1, T_z = -3/2$ series beginning with $^9\text{C}$; the decays of this series have been reported extensively elsewhere. More recently, this series has been extended to $^{61}\text{Ge}$. However, attempts to observe the next member of this series, $^{65}\text{Se}$, failed. Although examples of weak beta-delayed proton emission abound in light nuclei, heavy delayed proton emitters necessarily are weak because the isobaric analog state is energetically inaccessible via beta decay. Although very useful information can be obtained from delayed proton studies in heavy nuclei, the large density of states makes the spectroscopic information obtained less definitive unlike the general information often obtained for specific nuclear states in light nuclei. Beta-delayed proton emission has also served to provide much insight into the structure of medium mass nuclei near the drip line such as $^{65}\text{Ge}$, $^{73}\text{Kr}$ and $^{77}\text{Sr}$. Of course beta-delayed proton emission is not the only possibility for beta-delayed particle decay in proton-rich nuclei.
Beta-delayed alpha decay has been known for some time in nuclei such as $^8$B and $^{20}$Na. However, there are few examples due to the much larger alpha particle barriers; also since alpha decay is generally well understood, we will not discuss this decay mode. One can also envisage such rare decays as delayed $^3$He emission, but an even more exotic decay mode would be one where beta decay is followed by the emission of more than one particle. The emission of two protons was first postulated by Gol'danskii $^{13}$; fig. 1 graphically depicts the energetics necessary for this decay mode. Close examination of the mass surface and the energies of the isobaric analog states showed that the $T_z = -2$ nucleus $^{22}$Al would be an excellent candidate to exhibit this new decay mode. Since beta-delayed single proton emission had been used to discover $^{22}$Al $^{14}$ the mass of the isobaric analog state in $^{22}$Mg was already known.

Following the development of suitable detectors, beta-delayed two-proton decay was discovered $^{15}$ in $^{22}$Al. Unlike the relatively well-understood weak decay followed by tunneling of a charged particle through a barrier problem, this new decay mode posed many new questions regarding the exact mechanism. Were the two protons emitted simultaneously as either a correlated or uncorrelated pair or sequentially via an intervening nuclear state? If the latter mechanism prevailed, then the additional kinematical complication of emission from a moving source would occur. Figure 2 shows two superimposed two-proton sum spectra taken with experimental setups designed to look at relative proton angles of 5-70° (small angle) and 70-170° (large angle). The observed kinematical shift in conjunction with the corresponding breakup spectra is consistent with a sequential decay process. More details and the exact kinematical formulae are given elsewhere $^{16}$. The extremely interesting case of the emission of two correlated protons emitted in a $^1S_0$ state ($^2$He) was also investigated in $^{22}$Al decay $^{17}$; results from this angular correlation measurement were consistent with sequential emission, but a 10% $^2$He branch could not, however, be excluded.

This new and unique decay mode provided a way to study spectroscopically and identify nuclei even further from beta stability. Searching for beta-delayed two-proton branches permitted the discovery of several new isotopes or the first decay studies of these species. These included $^{26}$P $^{18}$, $^{35}$Ca $^{19}$, the first stable $T_z = -5/2$ nuclide, and $^{31}$Ar $^{20}$. This last nuclide, along with several other $T_z = -5/2$ nuclides, were first identified $^{21}$ with the recoil product separator LISE $^{22}$ at GANIL. Additionally, the $Bp$ decay of $^{31}$Ar has also been studied $^{23}$ at GANIL. The ability to utilize a coincidence measurement to observe
nuclei produced with very low cross sections has proven to be very effective. (A more complete review of beta-delayed two-proton decay is given elsewhere.) However, two problems make these observations even more difficult: the small detector solid angles necessary for adequate resolution and the fact that often only a few MeV of available energy must be shared between the two protons. These problems become even more severe as one attempts to study nuclei across the drip line, but both of these problems can, in principle, be solved simultaneously.

Nuclear decays at the proton drip line now become very short lived because of the lack of the slow weak decay process. Only the Coulomb and angular momentum barriers serve to impede the decay rate. (For a more complete description of the decay rates associated with proton decay, see ref. 25.) A complete review of ground state proton emission has recently been completed. Since all ground state proton emitters discovered to date are in the medium to heavy mass region where the Coulomb barriers are much larger, searches for light mass proton emitters are hampered by the much shorter lifetimes associated with the smaller Coulomb barriers and the generally smaller angular momentum barriers. Two prime candidates in the lighter masses, which can be produced in relatively large yield in heavy ion reactions on calcium targets, are $^{65}$As and $^{69}$Br. The predicted proton separation energies for these nuclides, though, suggested that techniques were not available for observing very short-lived nuclei and very low-energy protons. This detection of low energy protons is even more important and more difficult in the search for ground state two-proton radioactivity.

Ground state two-proton radioactivity was first proposed by Gol'danskii more than 20 years ago. This decay mode, however, requires a very unique situation to occur on the atomic mass surface, namely that the nuclide be unbound to two-proton emission but bound to single proton emission. This requirement necessarily dictates that any search for this decay mode utilize reliable mass predictions. Unfortunately, the severe exponential dependence of the half-life on the two-proton separation energy makes all existing mass predictions too inaccurate. One must therefore use a composite mass estimation system based upon many current predictions plus general experience regarding certain types of mass formulae. For example, in very proton-rich light nuclei, recursive formulae seem to give consistently the best results. We commonly use the Kelson-Garvey mass relation to obtain mass estimates and one- and two-proton separation energies. Figure 3 (see ref. 8)
graphically depicts the use of the Kelson-Garvey mass relation to predict these separation energies for a) $T_z = -2$, b) $T_z = -5/2$, and c) $T_z = -3$ nuclei.

A promising candidate for observing ground state 2p radioactivity is $^{39}$Ti. Its predicted two-proton separation energy is $-780 \pm 300$ keV. The two protons must be emitted simultaneously in either a correlated or uncorrelated manner; normal phase space considerations would generally preclude the latter from happening. Thus, we need only consider the correlated case, i.e., $^2$He emission. In reaction studies, the final state interaction and the large kinetic energies confine the $^2$He breakup cone to $-40^\circ$ 27. Unfortunately, the kinetic energy of an emitted $^2$He arising from two-proton decay could be very small, which could make the relative angle between the two protons nearly $180^\circ$. Thus any experiment must not only cope with possibly a very short-lived species which emits two very low energy protons, but it must also be capable of covering almost all relative angles simultaneously. In the next section we will examine general approaches to these problems and a few specific examples that we have chosen to use.

III. Experimental Approaches

The helium-jet recoil transport method 28 is probably one of the oldest and simplest methods for removing nuclei away from the intense radiations associated with various production techniques to a lower background area for radioactive assay. Dependent upon the capillary length, gas flow and active collection volume, the helium-jet transport time can vary anywhere from a few milliseconds to a few seconds. Its chemical universality for non-gaseous products is both boon and bane. It is in general use for the study of exotic nuclear decays; a setup is depicted schematically in Fig. 4. This single 70 cm long capillary system has a transit time of approximately 25 ms. Twenty-five milliseconds, however, is still too long to search for ground state one- and two-proton emission, and thus other faster techniques need to be employed.

One very successful method used to study exotic nuclei is by using recoil product separators. These devices can operate anywhere from a few MeV/nucleon all the way up to several hundred MeV/nucleon. Examples of these types of devices are SHIP 29 at GSI and LISE 23 at GANIL. The primary advantage of this type of device is the rapid (typically a few hundred nanoseconds) physical separation of the products of interest. A disadvantage of such systems is that for decay studies the primary beam must be turned off to await the decay of an identified nucleus. If the half-life is very short, this poses little problem, but if the half-life exceeds a few tens of milliseconds, then the overall yield can be significantly
reduced. Moreover, implantation of the products (even in a detector) makes the detection and identification of quite low energy protons difficult.

Another method involves catching recoils in foils for subsequent observations of their radioactive decay, typically under low duty factor accelerator conditions. Because there is no physical separation of the products, large backgrounds due to similar decays from competing reaction products could easily mask any signal. This technique is best suited, therefore, for proton searches in regions of the nuclidic surface where few beta-delayed proton emitters could be formed that have low-energy proton groups. Proton decays from $^{113}$Cs and $^{109}$I$^{25}$ were discovered using this general technique. It is important to note that beta-delayed proton emitters in this mass region generally exhibit no protons with energies below ~1 MeV. Thus any single, low-energy peaks are more easily identified.

On-line isotope separators (ISOL) are widely used to separate nuclei of interest from products of competing nuclear reactions. Many ion source techniques have been utilized with ISOL systems, but most of these techniques involve significant sublimation or diffusion times. Although rapid release techniques have been developed for some elements, in general fast ISOL systems are helium-jet based. These include helium-jet coupled ion source systems such as the Berkeley-88 RAMA system and the ion guide system originally developed at Jyvaskyla. The primary holdup time for the RAMA system is due to the capillary transit time (~200 ms); the ion source holdup time is short because of the rapid but low efficiency use of charge exchange with He$^{1+}$ ions. The ion guide relies on the high first ionization potential of helium; in principle, product nuclei remain in the +1 charge state once the recoils have thermalized. The helium is skimmed off and any charged atoms are accelerated to a final energy suitable for mass separation (typically 40-60 kV). Both of these techniques have little or no chemical selectivity. The generally lower efficiencies, though, make studies at the drip line nearly impossible given the typically infinitesimally small production cross sections.

Although the above techniques are very useful for many experiments, the beam structure of the LBL 88-Inch Cyclotron and the desire for rapid removal of recoil products on the sub-millisecond time scale led to the development of the fast rotating wheel system depicted schematically in Fig. 5. The general idea is that some recoils from the target are caught in the aluminum catcher foils (the percentage is dependent upon the aluminum foil thickness). These catcher foils are rotated (continuously) between pairs of detector telescopes suitable for the appropriate decay measurements. The arrival of the radioactivity
at the detector location coincides with the time when the beam is turned off during a 50% on/50% off cycle and is independent of the wheel speed; the wheel speed can be varied from 20-5000 rpm, corresponding to 250-1 ms cycle times. The entire system is rotated 70° from normal to permit a threefold increase in stopping material for a unit traverse by the emitted decay particles. Details are given in ref. 20. This fast rotating wheel partially solved the short lifetime problem by permitting studies of nuclides with half-lives down to 100 μs. The difficult experimental problem of detecting low-energy protons remained, however. A review of detectors which could be used for this purpose is the subject of the next section.

Most prior studies of low-energy proton emission (< 1 MeV) have been performed with single silicon counters. Particle identification was generally accomplished on a peak-by-peak basis by comparing the measured energies with and without a thin degrader foil. This technique does not work, however, for very low yield experiments. One must identify on an event-by-event basis all emitted particles; this requires a telescope.

Three general types of telescopes can be envisioned: all-silicon, gas-silicon hybrid, and all-gas telescopes. The first type could only be realized because of the recent development of epitaxially grown silicon crystals 1-3 μm thick. Unfortunately, the large area (for large solid angles) wafers needed for low count rate experiments have such large capacitances that the resolution is sufficiently poor to preclude their use. The last type of detector telescope can encompass both Bragg curve spectrometers (see ref. 3, for example) and proportional counters. These types of systems generally have higher thresholds (0.5 MeV) due to the thick window needed to withstand the high gas pressures necessary to stop the low-energy protons.

Gas-silicon hybrid detectors thus became an attractive option for experimental development. All gas counters generally suffer from very slow charge collection times. This problem was overcome by designing the gas-silicon detector depicted in the upper part of Fig. 6. The small active gas volume has charge collected from the center of the detector. When combined with the use of CF₄ gas, the majority of the charge can be collected in 1 μs; this timescale is also typical for silicon counters and is thus ideal for a hybrid system. Figure 7 shows a two-dimensional spectrum obtained with a gas-silicon detector telescope arising from products of the 40 MeV ³He + Mg reaction. The proton peaks clearly evident in this spectrum are all attributable to the beta-delayed proton decay of ²⁵Si. This detector has essentially unit efficiency for protons with energies of 250-6000 keV. The lower part of Fig. 6 shows the six-telescope systems constructed for use with the fast rotating wheel. A more complete description of these detectors is in preparation 34.
IV. Recent Results and Future Studies

Using these new experimental systems, several searches for examples of ground state one- and two-proton emission have been started. $^{28}$Si and $^{32}$S bombardments of calcium targets have yielded no evidence so far for the ground state proton decay of either $^{65}$As or $^{69}$Br in both our fast wheel measurements and in velocity-filter-separated product measurements (with a single silicon counter) at Daresbury. Successful searches for light mass proton emitters will depend on a very small proton separation energy to classify the nuclide as radioactive rather than unbound; the proton separation energy must be large enough, though, to compete with beta decay. The general requirements which are necessary have been covered in greater detail elsewhere. These searches are further complicated by the well known Thomas-Ehrman shift which frequently adds several hundred keV of stability to nuclides at the drip line.

We have also searched extensively for two-proton radioactivity from $^{39}$Ti. To date, we have found no evidence for this decay mode. Additionally, recent results from GANIL have shown $^{39}$Ti to have a half-life of $28 \pm 9$ ms, so that it is a beta-emitter. $^{39}$Ti was one of the best candidates in which to observe ground state two-proton decay. We believe that its non-observation is more probably due to an as yet not understood decay mechanism rather than a mispredicted mass surface. Although on general systematics (or on a weak Thomas-Ehrman shift in this higher $I$-value nuclide) one might expect $^{39}$Ti to be ~150-200 keV better bound than its predicted 780 keV unbound, a weak ground state two-proton decay branch should still be present. Further studies of $^{39}$Ti or other candidates will hopefully yield the discovery of this tantalizing decay mode.

References

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Figure Captions

Fig. 1  Generic decay scheme showing conditions necessary for beta-delayed one- and two-proton emission.

Fig. 2  Superimposed $^{22}$Al beta-delayed two-proton sum spectra taken at average angles of 420° and 120° dramatically showing the kinematic shift associated with sequential two-proton decay.

Fig. 3  One- and two-proton separation energies for a) $T_z = -2$, b) $T_z = -5/2$, and c) $T_z = -3$ nuclides.

Fig. 4  Schematic diagram of a helium-jet apparatus. A standard telescope arrangement for detecting beta-delayed protons is shown.

Fig. 5  Schematic diagram of the fast rotating wheel system.

Fig. 6  Top-cross section of a single gas-silicon telescope developed to detect protons down to 250 keV. Bottom-external view of one of the six-telescope arrays constructed for use with the fast rotating wheel.

Fig. 7  Two-dimensional plot of the silicon energy versus the differential energy loss in the gas counter. The proton band is clearly visible. (A small zero suppression has been used in this figure- the beta tail can interfere with the observed low-energy protons.)
$Z - 3, N + 1 + 2p$

Two-Proton Daughter

$(Z - 2, N + 1) + p$

Proton Daughter

$(Z - 1, N + 1)$

Emitter

**Figure 1**

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Figure 1
Two-proton Energy (MeV)

Counts

$^{22}\text{Al}$ (Small angle)

$^{22}\text{Al}$ (Large angle)

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Figure 2
Figure 3(a)
Figure 3(b)
Figure 3(c)
Rapidly-Rotating Recoil Catcher Wheel

200 \mu g/cm^2 Al recoil catcher foils

To Faraday cup

Target ladder

Telescope holder

Beam

Collimator

Wheel

Beam

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Figure 5
Cross Section of One Low-Energy Proton Telescope

- 30 μg/cm² polypropylene window
- Wire grid at ground
- 70 μg/cm² Ni foil at 520 V
- Wire grid at ground
- 300 μm Si detector

Top Detector Face
Figure 7

40 MeV $^3$He + Mg

Alpha Region

Proton Gate

Gas Energy (arbitrary scale)

Proton Energy (MeV)