

TWO-PROTON RADIOACTIVITY: THE INTERESTING CASE OF ^{67}Kr AND FURTHER STUDIES*

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We report on the observation of ^{67}Kr that has been produced in an experiment performed at the RIKEN/BigRIPS facility. The two-proton decay of ^{67}Kr has been evidenced and this nucleus is thus the fourth observed long lived ground-state two-proton emitter, after ^{45}Fe , ^{48}Ni and ^{54}Zn . In addition, the decay of several isotopes in the mass region has been investigated. While for previous cases of two-proton radioactivity, the theoretical models could reproduce the measured data, this is not the case anymore for ^{67}Kr . Two interpretations have been proposed to explain this discrepancy: a transition between real two-proton and sequential decay or the influence of deformation. These hypotheses will be tested in future experiments by measuring the angular and energy correlations of the emitted protons.

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1. Introduction

The two-proton radioactivity is a very exotic decay mode that occurs for nuclei beyond the proton drip-line, with an even atomic number Z . Together with 1-proton radioactivity (for odd- Z nuclei), the phenomenon was suggested in the 1960s by theoretical predictions [1]. The process has been evidenced in the decay of ^{45}Fe in experiments at GANIL/LISE3 [2] and GSI/FRS [3] in 2002. For candidate nuclei, the last two protons are not bound by strong interaction, but the ground-state energy is below the Coulomb barrier, which gives the life-time of the emitter. In addition, due to pairing, sequential emission of the protons is energetically forbidden, and the correlated protons have to be emitted simultaneously. After the emission by tunnel effect through the barrier, the sub-system (p - p , or ^2He) is unbound.

Today, 4 long-lived emitters (in the ms range) are known: ^{45}Fe [2–5], ^{48}Ni [6, 7], ^{54}Zn [8] that have been produced and studied at GSI, GANIL and NSCL, and recently ^{67}Kr [9–11], in an experiment at the RIKEN/BigRIPS facility that is reported in this paper. These nuclei have been produced at fragmentation facilities, and the first experiments were based on the implantation-decay correlation technique in silicon detectors. This allowed for an indirect observation of the two-proton radioactivity based on global quantities: the half-life, the branching ratio and the energy of the decay. Calculations performed in a theoretical 3-body framework [12, 13] showed that the p - p correlation pattern is sensitive to the orbital configuration of the nuclei. New devices have been developed, in order to measure the tracks of the individual particles in time projection detectors (TPC) and deduce the angular and energy correlations of the emitted protons [4, 5, 14].

In this paper, we report some results from the experiment performed at RIKEN to produce nuclei at the proton drip-line in the ^{67}Kr mass region. The exotic nuclei have been produced in the fragmentation of a ^{78}Kr beam

(250 nA, 350 A MeV) on a beryllium target, and selected by the BigRIPS and the ZDS separators [15]. They were implanted in the WAS3ABi [16] device consisting of 3 DSSSDs (each detector is 1 mm thick, with 60 vertical and 40 horizontal strips of 1 mm width). The charged particles emitted during the radioactive decay of implanted nuclei were detected in the silicon detectors that are surrounded by the EURICA [17] array of germanium detectors for the coincident γ detection.

New isotopes have been identified, and the decay of several nuclei have been investigated. In the case of ^{67}Kr , we could conclude on two-proton emission, in competition with β^+ /electron capture decay. For previous cases of two-proton radioactivity, the experimental data could be fairly well understood in available theoretical frameworks. In the case of ^{67}Kr , these models could not reproduce the measured half-life, and this triggered new theoretical developments [18, 19] in two-proton radioactivity modeling. Further experimental studies should be carried out to confirm the proposed hypotheses, by measuring p - p correlations for this decay.

2. New isotopes

The fragments were identified with the BigRIPS standard detection, from energy loss, time-of-flight and magnetic rigidity determination [11]. The result is presented in Fig. 1. ^{63}Se , ^{68}Kr and ^{67}Kr have been observed for the first time [9], with respectively 348, 477 and 82 counts at the F7 focal plane of the separator. Only a fraction of these nuclei is transmitted through the ZDS spectrometer to the implantation/decay detectors. This fraction is about 40% for the most exotic fragments.

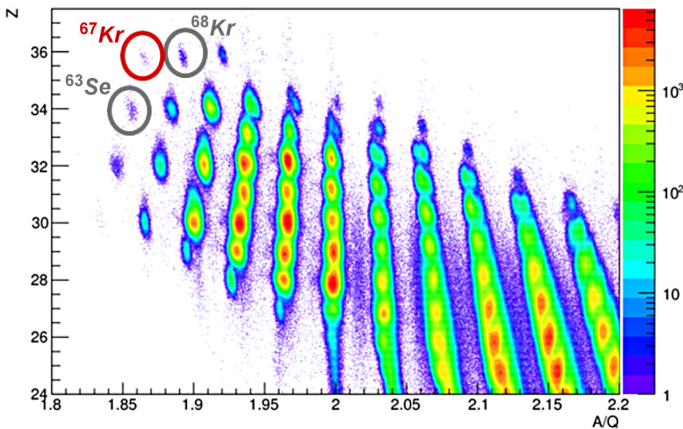


Fig. 1. Fragment identification matrix from the BigRIPS detection. The circles indicate the isotopes that have been observed for the first time.

3. Beta-decay results

For ions implanted in the WAS3ABi detectors, the correlation of subsequent decay events allows to determine the charged particles energy distribution (with coincident γ rays) and the time distribution between implantation and decay events from which the decay half-life is estimated. The details of the analysis can be found in Ref. [11].

TABLE I

Comparison of the half-lives and total beta-delayed proton emission ratios measured for the decay of several isotopes produced in the experiment. The results are taken from Ref. [11].

Isotope	Isospin T_Z	Half-life $T_{1/2}$ [ms]		Total β -proton branching [%]	
		Previous	Present	Previous	Present
^{63}Se ^{59}Ge	$-5/2$			13.2 ± 3.9 13.3 ± 1.7	89 ± 11 93 ± 7
^{68}Kr	-2			21.6 ± 3.3	89_{-10}^{+11}
^{69}Kr	$-3/2$	28 ± 1 [20]	27.8 ± 1.6		93_{-6}^{+7}
^{65}Se		27 ± 3 [21]		99_{-11}^{+1} [21]	
^{61}Ge		33 ± 4 [21]	34.2 ± 0.7	88_{-13}^{+12} [21]	
^{57}Zn		45 ± 6 [22]	40.7 ± 0.4	62 ± 4 [22]	87 ± 3
^{55}Cu		40 ± 15 [23]			
		48 ± 3 [22]	45.7 ± 0.6	78 ± 17 [22]	
		40 ± 10 [24]			
		57 ± 3 [25]	55.5 ± 1.8	0 [25]	
		27 ± 8 [26]		15.0 ± 4.3 [26]	
^{64}As	-1	72 ± 6 [27]	69.0 ± 1.4		
^{60}Ga		18_{-7}^{+43} [28]			
		76 ± 3 [29]	70.8 ± 2.0		
		70 ± 13 [28]			
		70 ± 15 [30]		1.6 ± 0.7 [30]	
^{56}Cu		80 ± 2 [29]	80.2 ± 0.7		
		82 ± 9 [28]			
		93 ± 3 [31]		0.40 ± 0.12 [31]	
		78 ± 15 [32]			
^{65}As	$-1/2$	126 ± 5 [27]	130.3 ± 0.6		
		128 ± 16 [33]			
^{63}Ge		156 ± 11 [29]	153.6 ± 1.1		
		149 ± 4 [27]			
		150 ± 9 [34]			

The decay of ^{59}Ge , ^{63}Se and $^{67,68}\text{Kr}$ has been measured for the first time. ^{59}Ge and ^{63}Se were candidates for two-proton radioactivity, but no evidence of such a decay branch could be observed in the data. In addition, the decay of several β -delayed proton or β emitters has been analyzed. The results are summarized in Table I.

When previous measurements are available, the half-lives are all in good agreement with literature values. For the only proton emission branching ratio that was previously measured (^{61}Ge decay), our value is 5 standard deviations away from the previous value. The reason for this discrepancy is that in previous measurement, the proton emission at an energy below the main peak at 3.2 MeV is not observed, resulting in an underestimated branching ratio. When considering only the proton emission from this peak, both measurements are in good agreement.

4. Two-proton radioactivity of ^{67}Kr

The charged particle energy and time distributions for the decay of ^{67}Kr are presented in Fig. 2. The peak at 1690 keV is assigned to the two-proton emission branch (details in Ref. [10]). We obtained a half-life of 7.4 ± 2.9 ms for the nucleus and a 2P branching ratio of $37 \pm 14\%$. The partial half-life is thus 10 ± 6 ms for the β decay, in agreement with the value from the *gross theory* [35], and 20 ± 11 ms for two-proton emission.

The two-proton half-life has been compared to a hybrid model [36] that combines the good dynamics of a 3-body model [37, 38] with the partial emission spectroscopic amplitudes from the configuration interaction model (details in Ref. [36]). While the calculated half-lives are in fairly good agreement (within a factor of 2) with the experimental values for the previously studied precursors (see also Refs. [14, 39]), in the case of ^{67}Kr , the difference is in the order of a factor 20. This result has triggered new theoretical calculations aiming to explain this large discrepancy.

A first hypothesis is related to the decay mechanism. According to a recent semi-analytical R -matrix calculation [18], depending on the position of the $^{66}\text{Br}+p$ resonance, the decay could take place in a transition region between a direct and a sequential two-proton emission. In such a case, the energy sharing between the emitted particles must be sensitive to the sequential component, with a contribution that is not centered on an equal sharing of the energy.

Another possible reason of the discrepancy is that ^{67}Kr is most likely located in a deformation region [40]. A calculation in the three-body Gamow coupled-channel (GCC) framework resulted in a half-life of 24_{-7}^{+10} ms [19], in agreement with the experiment, when considering a deformation of $\beta_2 \simeq -0.3$ due to the $g_{9/2}$ orbital. The GCC calculation also predicts proton-proton angular correlations with different distributions whether the $g_{9/2}$ orbital is considered or not.

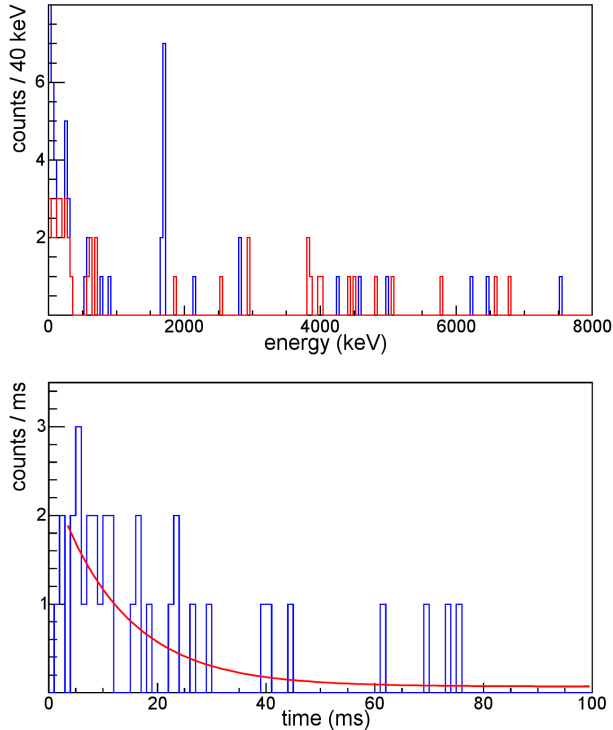


Fig. 2. (Color online) The upper plot shows the charged particle energy distribution in the decay of ^{67}Kr , measured in the detector where the implantation occurred. The black/blue histogram corresponds to the events with no coincident signal from a β particle in neighboring detectors. The peak at 1690 keV corresponds to the two-proton transition. The lower plot represents the time distribution of events after implantation. The fit (gray/red curve), taking into account the daughter decay of the different decay branches (two-proton and β), gives a half-life of 7.4 ± 2.9 ms.

5. Further studies

The experimental perspectives for ground-state two-proton radioactivity studies follow two main directions. The first one is to improve the knowledge on already identified emitters such as ^{48}Ni , ^{54}Zn and ^{67}Kr for which very little or nothing is known about the correlations of emitted protons. The second direction is to search for new candidates, which implies to produce and identify new isotopes and observe their decay modes. The proton drip-line between the known cases and the tin isotopes is of special interest because if other emitters are found, this would map a region between the closed shells $Z = 28$ and $Z = 50$, crossing a deformation zone. This region of the table of isotopes should be reachable in a near future with the SuperFRS at GSI/FAIR.

The GCC calculation is performed for ^{67}Kr , but also for the spherical case ^{48}Ni as a benchmark for the model. In both cases, the correlation pattern is not measured, but this can be achieved using ACTAR TPC [41] as tracking detector, in order to measure the angle and energy of individual protons. This device has been used in a recent experiment [42] at GANIL/LISE3 where the proton radioactivity of isomers in ^{53}Co and ^{54}Ni has been successfully observed. An experimental program for the correlation measurement is proposed at GANIL/LISE3 for ^{48}Ni [43] and at RIKEN/BigRIPS for ^{67}Kr [44].

In the decay of ^{67}Kr , the angular distribution will allow to discriminate between GCC predictions with or without the $g_{9/2}$ orbital, but also to test the sequential decay hypothesis by measuring the energy sharing of the protons. It is also necessary to measure the angular correlations of the protons in the decay of ^{48}Ni because the predictions from the GCC model differ from the one that we can extrapolate from the 3-body model, available only for ^{45}Fe [4, 12] and for ^{54}Zn [14].

On the experimental side, in addition to the measurements of the proton–proton correlation pattern that will be performed for ^{48}Ni and ^{67}Kr , a higher statistics would also be of great interest in the case of ^{54}Zn for a detailed comparison with available calculations. With the development of intense beams and new facilities (such as FAIR/SuperFRS), the search for new emitters should allow to reach the next closed shell at $Z = 50$.

6. Conclusion

During the experiment performed at the RIKEN/BigRIPS facility, we produced nuclei at the proton drip-line in the region $Z \leq 36$ and studied their radioactive decay. From the analysis of the ^{67}Kr decay data, we measured a 7.4 ± 2.9 ms half-life with a two-proton radioactivity branch of $37 \pm 14\%$. The experimental results are in disagreement with expectation from available calculations. This lead to new theoretical works suggesting either a transitional situation between direct and sequential two-proton emission or the influence of deformation in order to reproduce the measured half-life. These hypotheses will be addressed in further experiments foreseen to be carried out with the tracking detector ACTAR TPC to measure the angular correlation and the energy sharing of the emitted protons.

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REFERENCES

- [1] V. Goldansky, *Nucl. Phys.* **19**, 482 (1960).
- [2] J. Giovinazzo *et al.*, *Phys. Rev. Lett.* **89**, 102501 (2002).
- [3] M. Pfützner *et al.*, *Eur. Phys. J. A* **14**, 279 (2002).
- [4] K. Miernik *et al.*, *Phys. Rev. Lett.* **99**, 192501 (2007).
- [5] J. Giovinazzo *et al.*, *Phys. Rev. Lett.* **99**, 102501 (2007).
- [6] C. Dossat *et al.*, *Phys. Rev. C* **72**, 054315 (2005).
- [7] M. Pomorski *et al.*, *Phys. Rev. C* **83**, 061303(R) (2011).
- [8] B. Blank *et al.*, *Phys. Rev. Lett.* **94**, 232501 (2005).
- [9] B. Blank *et al.*, *Phys. Rev. C* **93**, 061301(R) (2016).
- [10] T. Goigoux *et al.*, *Phys. Rev. Lett.* **117**, 162501 (2016).
- [11] T. Goigoux, Ph.D. Thesis, Université de Bordeaux, 2017.
- [12] L. Grigorenko, I. Mukha, M. Zhukov, *Nucl. Phys. A* **714**, 425 (2003).
- [13] L.V. Grigorenko *et al.*, *Phys. Rev. C* **82**, 014615 (2010).
- [14] P. Ascher *et al.*, *Phys. Rev. Lett.* **107**, 102502 (2011).
- [15] T. Kubo *et al.*, *Prog. Theor. Exp. Phys.* **2012**, 03C003 (2012).
- [16] S. Nishimura *et al.*, *RIKEN Accel. Prog. Rep.* **46**, 182 (2013).
- [17] P.A. Söderström *et al.*, *Nucl. Instrum. Methods Phys. Res. B* **317**, 649 (2013).
- [18] L. Grigorenko, *Phys. Rev. C* **95**, 021601(R) (2017).
- [19] S.M. Wang, W. Nazarewicz, *Phys. Rev. Lett.* **120**, 212502 (2018).
- [20] M. Del Santo *et al.*, *Phys. Lett. B* **738**, 453 (2014).
- [21] A.M. Rogers *et al.*, *Phys. Rev. C* **84**, 051306 (2011).

- [22] B. Blank *et al.*, *Eur. Phys. J. A* **31**, 267 (2007).
- [23] M. Hotchkis, *Phys. Rev. C* **35**, 315 (1987).
- [24] D.J. Vieira *et al.*, *Phys. Lett. B* **60**, 261 (1976).
- [25] V. Tripathi *et al.*, *Phys. Rev. Lett.* **111**, 262501 (2013).
- [26] C. Dossat *et al.*, *Nucl. Phys. A* **792**, 18 (2007).
- [27] A.M. Rogers *et al.*, *Nucl. Data Sheets* **120**, 41 (2014).
- [28] M.J. López Jiménez *et al.*, *Phys. Rev. C* **66**, 025803 (2002).
- [29] L. Kucuk *et al.*, *Eur. Phys. J. A* **53**, 134 (2017).
- [30] C. Mazzocchi *et al.*, *Eur. Phys. J. A* **12**, 269 (2002).
- [31] R. Borcea *et al.*, *Nucl. Phys. A* **695**, 69 (2001).
- [32] M. Ramdhane *et al.*, *Phys. Lett. B* **432**, 22 (1998).
- [33] E. Browne, J. Tuli, *Nucl. Data Sheets* **111**, 2425 (2010).
- [34] B. Blank, *Eur. Phys. J. A* **15**, 121 (2002).
- [35] T. Tachibana, M. Yamada, WWW Chart of the Nuclides, <http://wwwndc.jaea.go.jp/CN14/index.html>, 2014.
- [36] B.A. Brown, B. Blank, J. Giovinazzo, *Phys. Rev. C* **100**, 054332 (2019).
- [37] L.V. Grigorenko *et al.*, *Phys. Rev. Lett.* **85**, 22 (2000).
- [38] L.V. Grigorenko *et al.*, *Phys. Rev. C* **64**, 054002 (2001).
- [39] L. Audirac *et al.*, *Eur. Phys. J. A* **48**, 179 (2012).
- [40] R. Bengtsson, P. Möller, *Nature* **449**, 411 (2007).
- [41] T. Roger *et al.*, *Nucl. Instrum. Methods Phys. Res. A* **895**, 126 (2017).
- [42] D. Rudolph *et al.*, GANIL experiment E690, 2016, 2019.
- [43] J. Giovinazzo *et al.*, GANIL proposal, E742, 2019.
- [44] J. Giovinazzo *et al.*, RIKEN/RIBF proposal RIBF138-R1, 2016.