Large β-Delayed Neutron Emission Probabilities in the ^{78}Ni Region

J. A. Winger,[1,*] S. V. Ilyushkin,[1] K. P. Rykaczewski,[2] C. J. Gross,[2] J. C. Batchelder,[3] C. Goodin,[4] R. Grzywacz,[5,2] J. H. Hamilton,[4] A. Korgul,[6,5,4,7] W. Królas,[8,7] S. N. Liddick,[3,5] C. Mazzocchi,[5,9] S. Padgett,[5] A. Piechaczek,[10] M. M. Rajabali,[5] D. Shapira,[2] E. F. Zganjar,[10] and I. N. Borzov[11]

[1]Department of Physics and Astronomy, Mississippi State University, Mississippi State, Mississippi 39762, USA
[2]Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA
[3]UNIRIB, Oak Ridge Associated Universities, Oak Ridge, Tennessee 37831, USA
[4]Department of Physics and Astronomy, Vanderbilt University, Nashville, Tennessee 37235, USA
[5]Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee 37996, USA
[6]Institute of Experimental Physics, Warsaw University, Warszawa, PL 00-681, Poland
[7]Joint Institute for Heavy-Ion Reactions, Oak Ridge, Tennessee 37831 USA
[8]Institute of Nuclear Physics, Polish Academy of Sciences, Kraków, PL 31-342, Poland
[9]IFGA, University of Milan and INFN, Milano, I-20133, Italy
[10]Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803, USA
[11]GSI, D-64291 Darmstadt, Germany

(Received 21 January 2009; published 8 April 2009)

The β-delayed neutron branching ratios ($P_{\beta n}$) for nuclei near doubly magic ^{78}Ni have been directly measured using a new method combining high-resolution mass separation, reacceleration, and digital β-γ spectroscopy of ^{238}U fission products. The $P_{\beta n}$ values for the very neutron-rich isotopes $^{76-78}$Cu and ^{83}Ga were found to be much higher than previously reported and predicted. Revised calculations of the βn process, accounting for new mass measurements and an inversion of the $\pi 2p_{3/2}$ and $\pi 1f_{5/2}$ orbitals, are in better agreement with these new experimental results.

DOI: 10.1103/PhysRevLett.102.142502 PACS numbers: 21.10.−k, 23.40.−s, 27.50.+e

The β-delayed neutron emission (βn) process may occur in the decays of very neutron-rich nuclei when the β-decay energy Q_β exceeds the neutron separation energy S_n in the daughter nucleus. The properties of βn emission are important in understanding the evolution of nuclear structure in neutron-rich nuclei and have important consequences in our understanding of many physical processes where βn emitters are produced in large quantities. Particularly relevant are environments such as the rapid-neutron capture (r process) sites of nucleosynthesis [1,2] and fission reactors; see, e.g., Refs. [3,4].

However, absolute βn emission probabilities ($P_{\beta n}$) are notoriously difficult to measure because of the experimental problems associated with neutron counting and the contamination of the samples. Using our new experimental end station and techniques at the Holifield Radioactive Ion Beam Facility (HRIBF), we demonstrate that existing data might be unreliable. Imprecise data may contribute to inaccuracies in theoretical modeling and extrapolations which can lead to unnecessary or erroneous requirements in the operation of fission reactors [3] as well as nuclear waste handling and transmutation [4]. The βn emission modifies the duration of the nuclear fuel cycle during shutdown by altering the isotopic mix of the remaining fuel as well as providing some additional neutrons. These changes can affect the energy (decay heat) stored in the spent fuels and the time required before postprocessing can begin.

The βn emission process also affects nucleosynthesis during supernova events altering the expected isobaric distribution of the ashes created in the r process. A number of waiting-point nuclei, the lowest-Z member of each isobaric chain when the neutron density has lowered resulting in the end (or freeze-out) of the r process, are expected to exist in the region just above $N = 50$ [1,2]. Kratz *et al.* have shown the r process passes through a bottleneck at the large nuclear-shell-energy gap (closed nuclear shell) $N = 50$ and then follows various paths dependent upon the neutron flux [1]. The observed natural abundances are then determined by the properties of waiting-point nuclei, in particular the various decay branches, including β, βn, and $\beta 2n$, as the isotopes decay back to stability. Detailed network calculations of the fission and β-decay processes require accurate data and the results from our experiment suggest that verification of previously measured data as well as new measurements and theoretical modeling may be necessary.

The βn emission probability ($P_{\beta n}$) increases for neutron-rich nuclei away from the line of β stability primarily due to the increase in the energy window for β decay (Q_β) and the reduction of the neutron separation energy (S_n) in the daughter activity. The properties of βn emission are also affected by details of the structure of both the parent and daughter nuclei which influence Q_β and S_n values as well as the energies of the levels primarily populated in the β decay. For example, for neutron-rich nuclei with a single neutron above the $N = 50$ nuclear-shell gap, the odd neutron will be weakly bound resulting in a low S_n value. Hence a large βn branching ratio can be expected in the decay of neutron-rich $N = 52$ isotones. The position of the proton and neutron orbitals involved in the β decay, i.e., their single-particle energies (SPE), can result in preferential feeding to states above the neutron separation energy which further enhances βn emission. This effect is particularly important when considering the decays of neutron-rich nuclei near closed shells where the allowed Gamow-Teller β transitions involve deeply bound states. Therefore, the SPE and the locations and size of shell gaps for neutron-rich nuclei are very important. Otsuka *et al.* have shown that the tensor interaction between the $\nu 1g_{9/2}$ state with the proton fp shell states reduces the spin-orbit splitting resulting in the $\pi 1f_{5/2}$ orbital dropping below the $\pi 2p_{3/2}$ orbital near ^{75}Cu [5,6]. These shell-model predictions are complemented by the mean-field calculations of Dobaczewski *et al.* which indicate the additional importance of many-body correlations and the inclusion of coupling to continuum states to understand the structure of very neutron-rich nuclei [7]. The nuclear-shell gaps known for stable nuclei can be reduced while new nuclear-shell closures may appear in very neutron-rich nuclei which can significantly affect the β-delayed neutron emission probability.

To address the difficult task of making reliable $P_{\beta n}$ measurements, a new technique using γ spectroscopy of isotopically purified beams of neutron-rich nuclei has been developed at the HRIBF at Oak Ridge National Laboratory. Beams of Cu and Ga ions were produced by proton-induced fission of a ^{238}UC$_x$ target inside a hot ion source. The beams were mass separated, passed through a charge exchange cell to produce negative ions, and sent through a high-resolution mass separator prior to acceleration in the HRIBF Tandem accelerator. The ions were tagged in time using a microchannel plate detector [8], passed through a six-segment ionization chamber (IC) [9] filled with CF$_4$, and implanted onto the tape of a moving tape collector (MTC). The atomic number Z of the ions passing through the IC could be clearly identified by energy loss. This permitted easy tuning of the high-resolution isobar separator to optimize the beam for rate and purity. For example, when tuning the mass 76 isobars the rate for ^{76}Ga was reduced by a factor of 60 compared to ^{76}Cu while reducing the maximum achieved ^{76}Cu rate by only $\sim$30%. The collected sources were observed by four Ge clover detectors and two plastic β detectors which surrounded the beam pipe. All detector signals were processed using digital pulse processing [10]. For ^{76,77}Cu and ^{83}Ga the γ-ray singles spectra were analyzed to obtain βn probabilities, while for ^{78}Cu the β-gated γ-ray spectra were used and the appropriate β detection efficiency determined. Representative spectra for the decays of $^{76-78}$Cu are shown in Figs. 1(a)–1(c).

Two modes of operation were used in the experiment: the ''ranging-out'' (RO) mode [9] with $\sim$200 torr gas pressure in the IC and the ''pass-through'' (PT) mode with $\sim$100 torr gas pressure in the IC. The RO mode takes advantage of the different stopping powers for the components of the beam. This was further enhanced in the case of

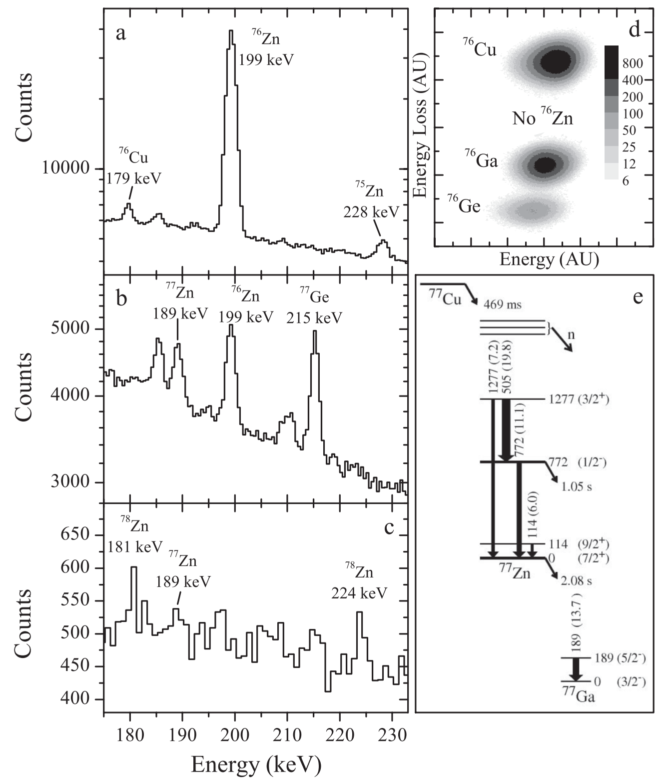

FIG. 1. The γ-ray spectra recorded for the βn precursors ^{76}Cu (a), ^{77}Cu (b), and ^{78}Cu (c) showing the decays of respective Zn isotopes used to determine the $P_{\beta n}$ values. For ^{76,77}Cu the γ-ray singles spectra taken in saturation mode are shown. For ^{78}Cu a β-gated spectrum taken using a 5.2 s MTC cycle is shown. For ^{76}Cu, the corresponding particle identification plot for the mass 76 isobars in the RO mode is shown in (d). Zinc ions are removed by the charge exchange cell while the higher-Z components of the beam (^{76}Ga and ^{76}Ge) are stopped by the ion chamber resulting in a pure ^{76}Cu beam. The partial decay scheme for ^{77}Cu indicating the significance of the 772-keV isomer in ^{77}Zn to the $P_{\beta n}$ value analysis is shown in (e). The number in parentheses following the γ-ray energy is the measured absolute γ-ray intensity from comparison of the γ-ray peak areas to the number of implanted ^{77}Cu ions.

Cu ions since negative Zn ions do not form in the charge exchange cell and are removed from the beam before postacceleration while the remaining $Z > 30$ contaminant isobars could be completely removed. The particle identification plot for the mass 76 isobars in this mode is shown in Fig. 1(d).

For the RO mode, the MTC collection point was placed within 1 cm of the exit window of the IC and periodically moved to a position at the center of the detector array. With Cu isotopes as the only component of the beam, the timing cycle was designed to maximize the detection rates for γ rays from the Zn daughters, thus precluding its use to study the short-lived Cu isotopes. In PT mode, the ions are implanted on the MTC in the center of the detector array with identification of individual ions in the IC allowing for absolute measurements. Our results are shown in Table I together with calculations of Ref. [11] and of this work.

The $P_{\beta n}$ value for ^{76}Cu has been previously reported as 3(2)% [12] and 2.4(5)% [13]. A recent work on ^{76}Cu decay [14] could not find any evidence for βn emission. However, the decay of ^{76}Cu can be considered as a good test case for βn studies since the decays of the daughter nuclei (^{75,76}Zn) are well characterized with known absolute branching ratios [15]. Here, the determination of $P_{\beta n}$ was made by comparing the relative intensities of the 228-keV (^{75}Zn), 199-keV (^{76}Zn) transitions [Fig. 1(a)], and 563-keV (^{76}Ga) γ ray, as well as comparing the absolute intensities of these γ rays to the measured number of ^{76}Cu ions in the IC in order to directly measure $P_{\beta n}$ and P_{β}. Since there is disagreement over the possibility of a β-decaying isomer for ^{76}Cu [14,16], we present here the result obtained from a saturation measurement using the PT mode with 68% beam purity and no ^{76}Zn present in the beam. For the relative intensity measurement, a $P_{\beta n}$ value of 7.3(6)% was obtained. For the absolute measurement, we obtained values of 7.0(6)% and 88(3)% for $P_{\beta n}$ and P_{β}, respectively. Although we slightly underestimate the total feeding [95(3)%], the overall agreement is quite good. We use a weighted average of the two results for our adopted value of 7.2(5)%.

The $P_{\beta n}$ value for ^{77}Cu was previously reported as 15^{+10}_{-5}% [13]. The only information on states in ^{77}Zn prior to this experiment was a 1.05 s $(1/2^-)$ $\nu p_{1/2}$ isomer decaying to the $(7/2^+)$ $\nu g_{9/2}$ ground state via a 772.4-keV $E3$ transition [17] and states at 114.9 and 803.6 keV observed in the βn branch of ^{78}Cu decay [14]. We have observed over 40 γ rays associated with the β decay of ^{77}Cu. A relevant subset of the decay scheme (to be published in a subsequent paper) is shown in Fig. 1(e). The assignment of spins and parities is based on systematics and the measured relative γ-ray intensities of the 505- and 1277-keV γ rays which are presumed to be $E1$ and $E2$, respectively. A significant amount of difficulty in the analysis is brought about by the presence of the $1/2^-$ ^{77}Zn isomer. The observed intensity of the 772-keV γ ray indicates a 66(5)% β-decay branch from the isomer for which nearly 100% will directly feed the ground state of ^{77}Ga bypassing the 189-keV level. Including this in our analysis, we could determine the appropriate branching through the 189-keV level as well as the effects of the different half-lives for each MTC cycle. A summary of these consistent results is presented in Table I. For the absolute measurement we also obtain a P_{β} value of 70(7)% indicating that we have correctly accounted for all the β decay intensity. We adopt a $P_{\beta n}$ value of 30.0(27)% which is taken from a weighted average of the values from the absolute measurement.

TABLE I. β-delayed neutron emission probabilities $P_{\beta n}$ (%).

			Experiment			Theory	
Nuclide	Mode[a]	MTC[b]	Rel.[c]	Abs.[d]	Adopt	[11]	Current
^{76}Cu	PT	Sat	7.3_6	7.0_6	7.2_5	4.2	5.6
^{77}Cu	RO	10.2 s	29_3	29_3	30.0_{27}	19.7	40.2
		20.2 s	28_4	28.7_{25}			
	PT	7.2 s	31.5_{24}	34.0_{23}			
		Sat.	32.1_{21}	28.5_{19}			
^{78}Cu	PT	5.2 s		65_8	65_8	42.6	53.1
^{83}Ga	PT	20.2 s		64_3	62.8_{25}	15.7	50.7
		Sat.		61_4			

[a]Ranging-out (RO) or pass-through (PT) mode.
[b]MTC cycle beam time on or saturation (Sat.).
[c]Comparison of the relative intensities of the most intense γ rays from nuclides in β and βn branches.
[d]Comparison of absolute intensities of the most intense γ rays from nuclei in the βn branch to the number of ions deposited.

The β decay of ^{78}Cu was measured using the PT mode (see Table I). Determination of the $P_{\beta n}$ value for ^{78}Cu was again hampered by the presence of the isomeric state in ^{77}Zn. Although the 114-keV γ ray is clearly seen in the data, the 189-keV γ ray (^{77gs}Zn $\rightarrow$ ^{77}Ga*) is surprisingly weak [see Fig. 1(c)]. However, it is consistent with the primary βn branch going by way of the $1/2^-$ isomeric state. The β branching probability is based on the 181- and 224-keV γ rays from ^{78}Zn decay which give a P_{β} of 35(8)%, yielding a value of 65(8)% for the βn probability which is consistent with the *lower limit* of $P_{\beta n} \geq 65(20)$% reported by Van Roosbroeck *et al.* [14] but significantly higher than the value of 15^{+10}_{-5}% listed in Ref. [13]. Van Roosbroeck *et al.* based their limit estimate on the relative intensities of the 730- and 114-keV γ rays fed in the β and βn branches of ^{78}Cu decay. We observe that the full intensity within the βn decay branch does not go through the 114-keV transition, so any agreement is coincidental.

The βn branching ratio for ^{83}Ga has been measured several times yielding discrepant data from 62.8(63)% to 14.9(18)%; see Ref. [12]. In our study, the determination of the βn branching ratio required a measured absolute branching ratio in the $A = 82$ decay chain since ^{83}Ge was not completely removed from the beam. The strongest transition observed in a saturation measurement is the 1092-keV γ ray from ^{82}Ge β decay indicating that the βn branch must dominate in the decay. The most logical explanation for this observation requires the assumption that the decay of ^{82}Ge is primarily to a single 1^+ excited state at 1092 keV with no feeding to the 2^- ground state in ^{82}As [18,19]. This sets the absolute branching ratios within the $A = 82$ decay chain. Using this value, we obtained 62.8(25)% for the βn branching ratio for ^{83}Ga, a value which is a factor of 5 larger than the presumably previous best measurement [12] and almost twice the recommended value of 37(17)% listed at the NNDC [15].

The measured $P_{\beta n}$ values are given in Table I and are shown in Fig. 2 plotted as a function of the βn energy window $Q_{\beta} - S_n$. The comparison of previous experimental results, shown in Fig. 2, to our $P_{\beta n}$ values obtained with

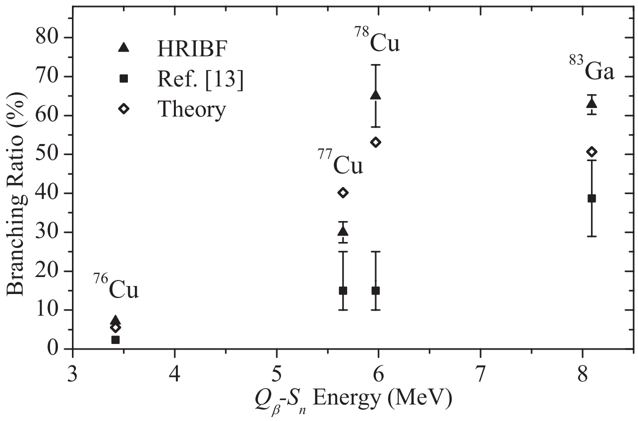

FIG. 2. The probability of β-delayed neutron emission as a function of the energy window for this decay mode. Results of this experiment (▲) are compared to those listed in Ref. [13] (■) and to our new theoretical estimates (◇); see text.

purified radioactive beams of known intensity clearly calls for a verification of earlier experiments. These experimental results also prompted new theoretical analysis of the βn emission process, with the results given in Table I and Fig. 2. The earlier calculations of $P_{\beta n}$ [11] within the continuum quasiparticle random-phase approximation underestimated the previously reported as well as our new, much larger, experimental $P_{\beta n}$ values. The new modeling of β-decay and β-delayed neutron emission followed the approach of Ref. [11]; however, updated Q_β and S_n values were taken from new mass measurements [20] together with data from Ref. [21]. Additionally, an inversion of the $2p_{3/2}$ and $1f_{5/2}$ proton orbitals for $N > 44$ isotones was assumed. The latter assumption follows indications from recent studies, e.g., [6,22] and our decay data, that this inversion occurs. By applying the blocking approximation within the standard density functional DF3 framework [23], the ground-state proton orbital was set to be $1f_{5/2}$ state for the ^{76,77,78}Cu and ^{83}Ga βn precursors. Compared to previous calculations [11], we observe a steeper increase of the βn probabilities towards the $N = 50$ shell closure. The new calculations agree much better with our data, see Table I and Fig. 2, and illustrate the importance of using the correct SPE sequence and masses for the modeling of β-decay properties.

In summary, the use of a high-resolution mass separator and postacceleration of radioactive beams allowed us to develop a new beam purification method, thus enabling $\beta\gamma$-decay spectroscopy with pure samples of individual neutron-rich isotopes. We measured the total β-delayed neutron probabilities for several nuclei near ^{78}Ni: the $Z = 29$ isotopes $^{76-78}$Cu and the $N = 52$ nuclide ^{83}Ga. Our more precisely measured $P_{\beta n}$ values for these decays are 2–5 times higher than previously reported. The large discrepancies with the previous results indicate the necessity of radioactive beam purification for these difficult measurements. These differences led to new calculations which also included updated isotope masses and changes in the ground-state proton configuration. This new modeling of the decay processes yielded much better agreement for the $^{76-78}$Cu and ^{83}Ga βn precursors. This work demonstrates a strong need for verification of existing experimental data which can lead to improved results in the theoretical modeling of βn emission that may be important to our understanding of the evolution of nuclear structure, r process nucleosynthesis, and the decay processes occurring in fission reactors.

The authors gratefully acknowledge the work done by HRIBF staff in producing such high quality radioactive ion beams. This work was supported under U.S. DOE Grants No. DE-FG02-96ER41006, No. DE-AC05-00OR22725, No. DE-FG02-96ER40983, No. DE-AC05-06OR23100, No. DE-FG02-96ER40978, and No. DE-FG05-88ER40407, through NNSA Grant No. DEFC03-03NA00143, through the Foundation for Polish Science, and through the DFG-436 RUS 113907/0-1 grant.

*j.a.winger@msstate.edu

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