

Precise absolute γ -ray and β^- -decay branching intensities in the decay of ^{67}Cu

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Absolute γ -ray emission probabilities in the β^- decay of ^{67}Cu were measured by means of γ -ray and β^- -decay singles and β^- - γ coincidences. The new results, together with the known decay scheme of ^{67}Cu , were used to determine absolute β^- -decay branching intensities. The present data differ significantly from previously published values. In addition, the half-life of the $I^\pi = \frac{1}{2}^-$ isomer in ^{67}Zn was measured as $T_{1/2} = 9.37(4) \mu\text{s}$, in a good agreement with earlier measurements. From the analysis of the Fermi–Kurie plots, $Q_{\beta^-}(\text{g.s.}) = 560.3(10) \text{ keV}$ was deduced, which differs from the previously measured value of $577(8) \text{ keV}$ but is in good agreement with $Q_{\beta^-}(\text{g.s.}) = 561.3(15) \text{ keV}$ recommended in the latest Atomic Mass Evaluation.

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I. INTRODUCTION

The neutron-rich ^{67}Cu ($N = 38$) nucleus decays by emission of β^- particles to the ground state and to the first three excited levels of the daughter ^{67}Zn nucleus, as indicated in the decay scheme of Fig. 1. Early work of Easterday [1] measured the β^- -decay branching intensities, with $I_{\beta_0} \approx 20\%$ reported for the ground state to ground state branch. By using this value, the absolute γ -ray emission probabilities were determined in the subsequent γ -ray spectroscopy studies of Raman *et al.* [2] and Meyer *et al.* [3]. The latter were adopted in the most recent nuclear data evaluation [4]. It should be noted, however, that although the absolute γ -ray intensities in Refs. [3,4] are rather precise, this is somewhat misleading since they are deduced by using the less accurate I_{β_0} value of Easterday [1].

There are several motivations for a precise knowledge of the absolute decay properties of ^{67}Cu . For example, the β^- decay to the ^{67}Zn ground state involves a $\pi(p_{3/2})^1 \rightarrow \nu[(f_{5/2})^5, (p_{3/2})^4] \ell$ -forbidden, Gamow–Teller (GT) transition and the precise branching intensity is needed to determine the corresponding $B(\text{GT})$ value that can be used to validate shell-model predictions in this region located near the $N = 40$ sub-shell closure. The β^- branching intensities are also of interest in measurements of the β -asymmetry parameter in the decay of ^{67}Cu , which can provide information on the search for physics beyond the standard model [5]. Lastly, ^{67}Cu is a promising radionuclide for cancer diagnostics and radio-immunotherapy (see, for example, Ref. [6] and references therein). Although it has favorable decay properties, its wide application is still hampered by difficulties in production and, as a consequence, the lack of reliable supply [7]. Thus, precise knowledge of the absolute γ -ray-emission probability of the strongest 184 keV γ ray is needed in order to accurately determine the resultant activity, and the corresponding production cross sections for this isotope. Other decay properties, such as the absolute

β^- -decay branching intensities, for example, are important in therapeutic applications and in quantifications of radiation doses.

In this paper, we report on precise measurements of absolute γ -ray emission probabilities in the β^- decay of ^{67}Cu . By using the new data and the adopted decay scheme of ^{67}Cu [4], β^- -decay branching intensities were also determined. Our results differ significantly from those reported by the previous measurements [1–4].

II. EXPERIMENTAL DETAILS

A. Preparation of sources

The ^{67}Cu nuclei were produced via the $^{68}\text{Zn}(\gamma, p)$ reaction using a bremsstrahlung beam with 36 MeV endpoint energy at the Argonne Low Energy Accelerator Facility and were chemically isolated from the bulk zinc matrix by utilizing a dry sublimation technique [8]. The copper-rich residue was digested with hydrochloric and nitric acids under gentle heating. The resulting solution was allowed to cool and was then passed through a 15 mL gravity-fed anion exchange column (Dowex® AG 1 \times 8 HCl) to separate the copper fraction. From this solution, thin and open sources of ~ 2 mm diameter and activity of ~ 0.5 to $1.0 \mu\text{Ci}$ were prepared on plastic backings. These sources were used in a series of γ -ray and β^- -decay singles, and β^- - γ coincidence measurements. Thin, sealed sources were also produced and were used to monitor for possible impurities by means of γ -ray-spectroscopy measurements with a Low Energy Photon Spectrometer (LEPS) and a HPGe detector, but none were found at a level of $>0.01\%$ of the ^{67}Cu activity.

B. γ and β^- singles, and β^- - γ coincidence measurements

Singles γ -ray spectra were measured with a $2 \text{ cm}^2 \times 1 \text{ cm}$ planar LEPS [full width at half maximum (FWHM) of 0.5 keV at 122.1 keV] and a 25% coaxial HPGe (FWHM = 1.8 keV at 1332.5 keV) detector. The sources were positioned in plastic holders and were placed 7 cm (LEPS) and 10 cm (Ge) from the detectors, thus minimizing possible coincidence summing

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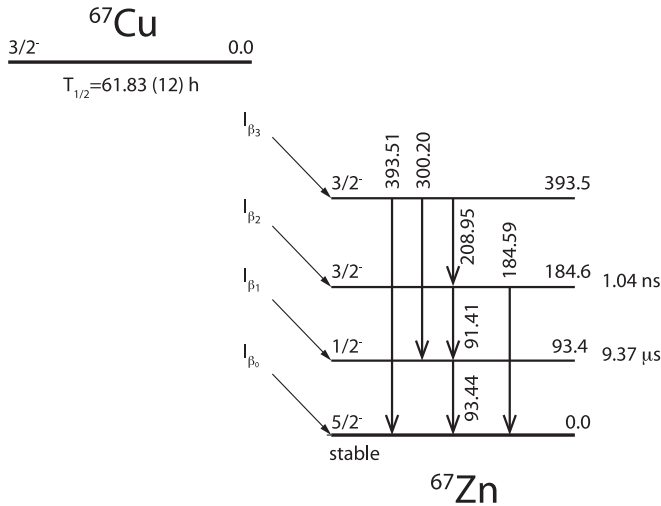


FIG. 1. Decay scheme of ^{67}Cu [4]. The γ -ray energies and the half-life of the $I^\pi = \frac{1}{2}^-$ level shown here are from the present work.

effects. The signals from the detectors were connected to ORTEC572 spectroscopy amplifiers and digitized by using a PC-based, single-parameter analog data-acquisition system, operated by the GENIE2000 software [9].

Singles electron (β^- particles and conversion electrons) spectra were measured in a small chamber that was kept under a vacuum of $\sim 2 \times 10^{-3}$ torr with a 25-mm²-area \times 500- μ m-thick passivated implanted planar silicon (PIPS) detector. The source-to-detector distance was 3.5 cm. The signals from the PIPS detector were routed into an AMPTEC A250CF cooled pre-amplifier and the output signals were connected to an ORTEC572 spectroscopy amplifier and digitized by using the PC-based, single-parameter analog data-acquisition system.

Singles and β^- - γ coincidence data were also collected by using the LEPS and a 150-mm²-area \times 1-mm-thick PIPS detector. The source was placed in the vacuum chamber with one side facing the PIPS detector, located inside the chamber at 3.5 cm above the source, while the other faced the LEPS that was 4.5 cm away from the source. A multiparameter

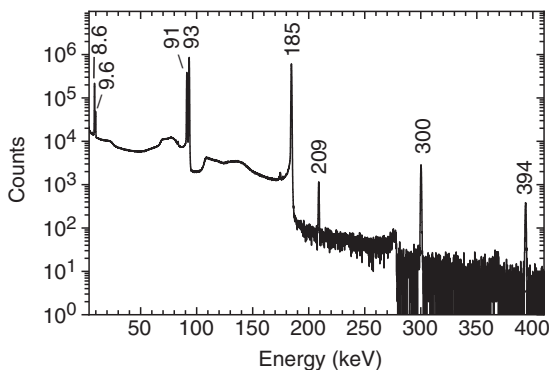


FIG. 2. γ -ray singles spectrum of the open ^{67}Cu source measured with the LEPS. The γ rays are labeled with their rounded-off energies in keV. The low-energy 8.6 and 9.6 keV peaks correspond to the K_α and K_β Zn x-rays, respectively.

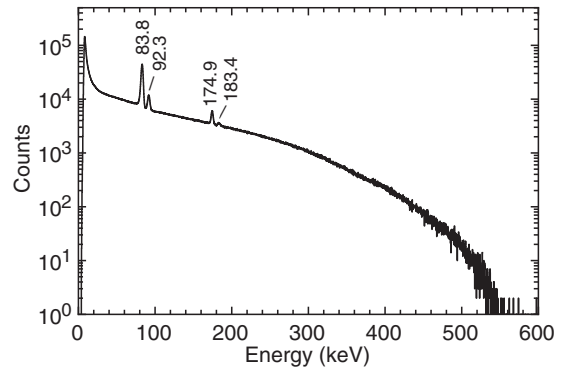


FIG. 3. Electron (β^- and conversion electrons) singles spectrum of the ^{67}Cu open source measured with the 25-mm²-area \times 500- μ m-thick PIPS detector. The K and L conversion electron peaks associated with the 91.4, 93.4, and 184.6 keV γ rays are labeled with their energies in keV.

digital data-acquisition system [10,11], based on 100 MHz, 14-bit digitizers, was used in these measurements. Pre-amplifier signals from the LEPS and PIPS detector were digitized and pulse-shape analysis was performed in order to determine their energies, while the timing information was obtained from a global time stamp. β^- - γ coincidences were established in the offline analysis by examining time differences between events from the PIPS detector and the LEPS.

Background singles and β^- - γ coincidence spectra were recorded shortly after each set of measurements and were appropriately subtracted from the sources spectra. Sample γ -ray and electron spectra measured with the LEPS and PIPS detector are presented in Figs. 2 and 3, respectively.

III. RESULTS AND DISCUSSIONS

The analyses of the γ -ray and electron spectra were performed by using the gf3 program of the RADWARE package [12]. The peak shape parameters, including the energy-dependent FWHM, were determined from a number of calibration sources (see below) and were kept fixed in the analysis of all spectra.

A. Energy and efficiency calibrations

The γ -ray energy and efficiency calibrations of the LEPS and the HPGe detector were performed by using a multi-nuclide source (produced and calibrated by the Eckert and Ziegler Company [13]) that contained the $^{57,60}\text{Co}$, ^{85}Sr , ^{88}Y , ^{109}Cd , ^{113}Sn , ^{137}Cs , ^{139}Ce , ^{203}Hg , and ^{241}Am radionuclides. Detector efficiencies were also measured by using mono-isotopic sources of ^{152}Eu , ^{182}Tl (Ge detector), and ^{243}Am (LEPS), and those were combined, after appropriate normalization, with the absolute values from the calibrated source. These calibration measurements were carried out in the same geometry as that used for the ^{67}Cu samples. The uncertainty in the absolute efficiency was dominated by the certified uncertainty of the calibrated source ($\sim 2\%$ [13]), while that for the relative efficiency was $\sim 0.5\%$. The energy calibration of the PIPS detectors was carried out by using open, mass-separated ^{137}Cs

TABLE I. γ -ray energies and relative intensities in β^- decay of ^{67}Cu measured in the present work and comparison with results from previous studies.

E_γ (keV)	Relative intensities (I_γ^{rel})			Absolute intensities (I_γ^{abs})	
	Present	Ref. [2]	Ref. [3]	Present ^a	Ref. [3] ^b
91.41(5)	14.23(14)	15.5(15)	14.37(22)	6.29(11)	7.0(1)
93.44(5)	33.5(3)	36.0(36)	33.1(5)	14.81(24)	16.1(2)
184.59(5)	100	100	100	44.2(6)	48.7(3)
208.95(5)	0.243(5)	0.24(4)	0.236(10)	0.107(3)	0.115(5)
300.20(5)	1.68(3)	1.57(16)	1.64(3)	0.743(17)	0.797(11)
393.51(6)	0.448(11)	0.43(5)	0.452(17)	0.198(6)	0.220(8)

^aFrom I_γ^{rel} and $I_\gamma^{\text{abs}}(185\gamma) = 44.2(6)\%$ in the present work.

^bBased on $I_{\beta_0} \approx 20\%$ [1].

and ^{243}Cm sources, which have a number of well-known discrete conversion electron lines [14–16]. A thin, open source (~ 2 mm diameter and activity of ~ 0.5 μCi) of a mass-separated ^{141}Ce radionuclides was also prepared on the same backing material that was used for the ^{67}Cu sources in order to determine the average β^- singles and β^- - γ coincidence efficiencies, and their ratios (see below).

B. γ -ray-emission probabilities and β -decay branching ratios

The present work confirmed the previously known [2,3] decay scheme of ^{67}Cu . The relative γ -ray intensities, normalized to 100 for the strongest 185 keV γ ray, determined from singles measurements by using the LEPS, are listed in Table I. The quoted uncertainties were obtained from the quadratic sum of the statistical uncertainties of the full-energy peaks and the uncertainties in the relative LEPS efficiencies. Because of the long lifetime of the 93 keV level (Fig. 1), no coincidence summing is expected to affect the intensities of the main γ rays. Measurements of the relative γ -ray intensities were also performed with the HPGe detector and the results were found to be consistent (within 1σ) with those from the LEPS. Values for the relative γ -ray intensities deduced from the data of Raman *et al.* [2] and Meyer *et al.* [3] are also listed in Table I and these were found to be in good agreement with our results.

The absolute γ -ray-emission probability of the 185 keV γ ray, $I_\gamma^{\text{abs}}(185\gamma)$, was obtained from β^- singles and β^- - γ coincidence data as

$$I_\gamma^{\text{abs}}(185\gamma) = \frac{N_{\beta\gamma}(185\gamma)}{N_\beta(^{67}\text{Cu})\epsilon(185\gamma)}, \quad (1)$$

where $N_{\beta\gamma}(185\gamma)$ is the total number of β^- - γ coincidences between β^- particles and the 185 keV γ ray, $N_\beta(^{67}\text{Cu})$ is the total number of β^- singles corresponding to decay of ^{67}Cu . In Eq. (1), $\epsilon(185\gamma)$ is the ratio of the average β^- - γ coincidence efficiency, $\epsilon_{\beta\gamma}(185\gamma)$, and the average PIPS efficiency for detecting β^- particles from all ^{67}Cu decays, $\epsilon_\beta(^{67}\text{Cu})$. Thus,

$$\epsilon(185\gamma) = \frac{\epsilon_{\beta\gamma}(185\gamma)}{\epsilon_\beta(^{67}\text{Cu})} = \frac{\epsilon_\beta(185\gamma)}{\epsilon_\beta(^{67}\text{Cu})} \epsilon_\gamma^{\text{abs}}(185\gamma), \quad (2)$$

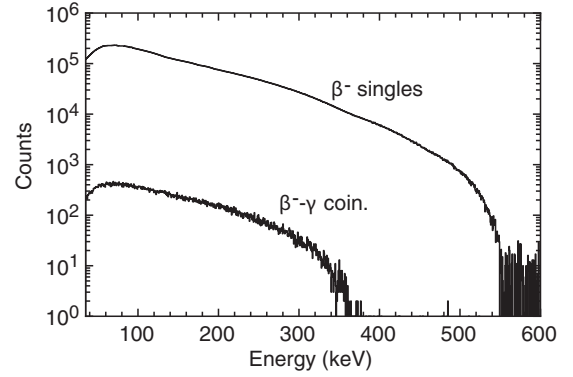


FIG. 4. β^- spectra of the ^{67}Cu open source measured with the 150-mm²-area \times 1-mm-thick PIPS detector. The top one is a β^- singles spectrum with contributions from room background and conversion electrons subtracted, while the bottom one is a β^- - γ coincidence spectrum produced by gating on the 185 keV γ ray.

where $\epsilon_\gamma^{\text{abs}}(185\gamma)$ is the absolute γ -ray efficiency of the LEPS for detecting the 185 keV γ ray and $\epsilon_\beta(185\gamma)$ is the average PIPS efficiency for detecting β^- particles from ^{67}Cu decays populating the 185 keV level.

A two-dimensional histogram of β^- -particle energies from the PIPS detector versus γ -ray energies from the LEPS ($E_\beta - E_\gamma$) was created in the off-line analysis by imposing a 40 ns-wide gate on the prompt-coincidence time peak between β^- -particle and γ -ray signals. A second $E_\beta - E_\gamma$ histogram was also created by gating in the time spectrum on the flat, random-coincidence background. The latter histogram was subtracted, after proper normalization, from the prompt-coincidence one. From the resulting histogram, a β^- spectrum was produced by gating on the 185 keV γ ray, shown in Fig. 4, from where the $N_{\beta\gamma}(185\gamma)$ value was obtained, after subtracting contributions from the background under the 185 keV γ -ray peak.

The $N_\beta(^{67}\text{Cu})$ value was obtained from the singles spectrum collected by the PIPS detector, also shown in Fig. 4, after subtracting contributions from the room background and the conversion electrons emitted in the decay of ^{67}Cu .

In the present work, $\epsilon(185\gamma)$ was determined relative to $\epsilon(145\gamma)$ from measurements with the ^{141}Ce source. The ^{141}Ce radionuclide decays via β^- emissions to the ground state (30.3%) and to the first-excited state at 145 keV (69.7%) of the daughter nucleus ^{141}Pr [17]. The latter deexcites via a single 145 keV γ ray to the ground state whose absolute emission probability is well established, $I_\gamma^{\text{abs}}(145\gamma) = 48.29(20)\%$ [17]. [Note that in the most recent evaluation by Nica [18] it is incorrectly given as 48.4(3)%.] Using Eq. (1), we determined $\epsilon(145\gamma)$ as

$$\begin{aligned} \epsilon(145\gamma) &= \frac{N_{\beta\gamma}(145\gamma)}{N_\beta(^{141}\text{Ce})I_\gamma^{\text{abs}}(145\gamma)} \\ &= \frac{\epsilon_\beta(145\gamma)}{\epsilon_\beta(^{141}\text{Ce})} \epsilon_\gamma^{\text{abs}}(145\gamma) = 0.879(13)\%, \end{aligned} \quad (3)$$

with $N_{\beta\gamma}(145\gamma)$ and $N_\beta(^{141}\text{Ce})$ deduced identically to the values obtained for the ^{67}Cu decay, as described earlier.

Since the PIPS detector was sufficiently thick to stop all β^- particles with energies below ~ 600 keV and given the nearly identical Q_{β^-} (g.s.) = 580.4(11) keV (^{141}Ce) and 561.3(15) keV (^{67}Cu) [21] and β^- -decay endpoint energies to the 145 and 185 keV levels in ^{141}Ce and ^{67}Cu , respectively, the ratio of average PIPS efficiencies is practically unity:

$$\frac{\epsilon_{\beta}(185\gamma)}{\epsilon_{\beta}(^{67}\text{Cu})} \bigg/ \frac{\epsilon_{\beta}(145\gamma)}{\epsilon_{\beta}(^{141}\text{Ce})} \simeq 1.00. \quad (4)$$

This was confirmed by Monte Carlo simulations of the PIPS detector efficiency as a function of the β^- particle energy using the MCNPX code [19].

Thus, the $\epsilon(185\gamma)$ value was obtained in the present work from the experimentally determined $\epsilon(145\gamma)$ value [Eq. (3)] and the relative LEPS efficiencies for the 145 and 185 keV γ rays as

$$\epsilon(185\gamma) = \epsilon(145\gamma) \frac{\epsilon_{\gamma}^{\text{rel}}(185\gamma)}{\epsilon_{\gamma}^{\text{rel}}(145\gamma)} = 0.539(9)\%. \quad (5)$$

Finally, the absolute γ -ray-emission probability of the 185 keV γ ray was deduced as $I_{\gamma}^{\text{abs}}(185\gamma) = 44.2(6)\%$. The quoted uncertainty was determined by taking in quadratures the statistical uncertainties for $N_{\beta}(^{67}\text{Cu})$, $N_{\beta\gamma}(185\gamma)$, and $\epsilon(185\gamma)$. It should be noted that, since ratios of $N_{\beta}(^{67}\text{Cu})/N_{\beta\gamma}(185\gamma)$ and $N_{\beta}(^{141}\text{Ce})/N_{\beta\gamma}(145\gamma)$ are presented in Eqs. (1) and (3), respectively, dead-time corrections and other systematic uncertainties in the β^- detection system cancel in the final result. It is also worth noting that the present $I_{\gamma}^{\text{abs}}(185\gamma)$ value is 9.2% smaller than that reported by Meyer *et al.* [3], which was adopted in Ref. [4]. By using the relative γ -ray emission probabilities of Table I and the present $I_{\gamma}^{\text{abs}}(185\gamma)$ value, absolute intensities for all γ rays in decay of ^{67}Cu were also determined, and these are also provided in Table I.

Since the β^- -decay radiation is not discrete, the β^- branching intensity to the ground state of the daughter nucleus cannot be measured directly. Instead, one can determine the absolute γ -ray-emission probabilities of the discrete γ rays emitted in the decay of the parent nuclide and then, by using the known decay scheme and intensity-balance considerations, determine the absolute β^- -decay branching ratios. By using this approach, the β^- branching intensities were determined in the present work and the results are presented in Table II,

TABLE II. β^- -decay branching ratios and $\log ft$ values in the decay of ^{67}Cu .

i	E_{level} (keV)	$I_{\beta_i^-}$ (%)		
		Present ^a	Ref. [1]	Ref. [4]
0	0.0	27.4(5)	≈ 20	≈ 20
1	93.4	19.8(5)	≈ 35	≈ 22
2	184.6	51.7(2)	≈ 45	≈ 57
3	393.5	1.0(1)		≈ 1.1

^aFrom I_{γ}^{abs} and intensity balances at each level. The total conversion coefficients were taken from Ref. [16], while the $M1 + E2$ mixing ratios for the 91, 185, 209, and 300 keV γ rays were adopted from Ref. [4].

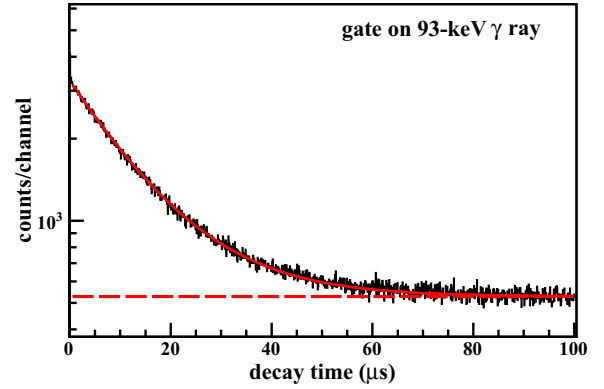


FIG. 5. (Color online) Time spectrum used to determine the half-life of the $I^{\pi} = \frac{1}{2}^-$ isomer in ^{67}Zn . The solid line corresponds to a least-squares fit to the data with a single-exponential decay and a constant background (shown as the dashed line).

together with the values reported in Refs. [1,4]. The present value of $I_{\beta_0} = 27.4(5)\%$ is significantly different from $I_{\beta_0} \approx 20\%$ reported by Easterday [1]. In addition, our β^- -decay branching ratios to the excited levels of ^{67}Zn also differ significantly from those measured in Ref. [1]. The latter are also different from values recommended in Ref. [4], even though $I_{\beta_0} = 20\%$ was used in both works.

C. Half-life of $I^{\pi} = \frac{1}{2}^-$ isomer in ^{67}Zn and β^- -decay endpoint energies

From the two-dimensional histogram of γ -ray energies versus the time differences between the signals from the LEPS and PIPS detector, a time spectrum was produced by gating on the 93 keV γ ray after subtracting the background contribution located under this peak. The resultant spectrum can be found in Fig. 5. A least-squares fit by means of a single-exponential decay and a constant background gave a value of $T_{1/2} = 9.37(4) \mu\text{s}$ for the half-life of the 93 keV level, which is comparable to results from previous studies [4].

From the $E_{\beta^-} - E_{\gamma}$ histogram, β^- spectra were produced by gating on the 91, 93, and 185 keV γ rays and converted to Fermi–Kurie plots [20], together with the spectrum from the singles measurement. By using linear least-squares fits to the tails of the Fermi–Kurie histograms, the endpoint energies were deduced and converted to Q_{β^-} (g.s.) values, as listed in Table III. A value of Q_{β^-} (g.s.) = 560.3(10) keV was determined as a weighted average of all measurements. It is worth

TABLE III. Measured endpoint energies, $E_{\beta_{\text{max}}}$, and corresponding Q_{β^-} (g.s.) values in the ^{67}Cu β^- decay.

E_{γ} gate (keV)	$E_{\beta_{\text{max}}}$ (keV)	Level energy (keV)	Q_{β^-} (g.s.) (keV)
Singles	559(2)	0.0	559(2)
93	468(2)	93.4	561(2)
91	374(2)	184.6	559(2)
185	377(2)	184.6	562(2)
Weighted average			560.3(10)

noting that, although our result is smaller than the Q_{β^-} (g.s.) = 577(8) keV value reported earlier by Easterday [1], it is in good agreement with Q_{β^-} (g.s.) = 561.3(15) keV, recommended in the latest mass evaluation table, AME2012 [21].

IV. CONCLUSIONS

The β^- decay of ^{67}Cu was studied in a series of γ -ray and β^- -decay singles, and β^- - γ coincidence measurements by using chemically purified sources. The absolute γ -ray emission probabilities and β^- -decay branching intensities were precisely determined and were found to differ significantly from previously reported values. In addition, the half-life of the $I^\pi = \frac{1}{2}^-$ isomer in ^{67}Zn was measured as $T_{1/2} = 9.37(4) \mu\text{s}$, in a good agreement with earlier results. From the analysis of the Fermi–Kurie plots, the value of Q_{β^-} (g.s.) = 560.3(10) keV was determined, which differs from the previously measured result of 577(8) keV but is in good agreement with the

recommended value of 561.3(15) keV from the latest Atomic Mass Evaluation. The new results will impact applications where accurate decay data on ^{67}Cu are required.

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