Absolute γ-ray intensities in the decay of ^{79}Kr and ^{127}Xe†

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Absolute γ-ray intensities in the decay of ^{79}Kr and ^{127}Xe have been determined from sources which were produced by (p,n) reactions on encapsulated potassium-halide targets. Absolute γ-ray emission rates of these sources were first measured with several calibrated Ge(Li)-detector systems. Then, following separation of the ^{79}Kr or ^{127}Xe gas, absolute disintegration rates of the sources were obtained by counting internally in gas proportional counters. Comparisons of the present results with previous work are made and although there are no major discrepancies, several inconsistencies in the existing decay schemes are resolved and the uncertainties in the absolute γ-ray intensities are substantially reduced. The half-lives for ^{79}Kr and ^{127}Xe were measured by both gas proportional counting and γ-ray spectrometry and are 35.04± 0.10 h and 36.3± 0.3 day, respectively.

[RADIOACTIVITY ^{79}Kr, ^{127}Xe; measured $T_{1/2}$, E_γ, absolute I_γ.]

I. INTRODUCTION

The present investigation of absolute γ-ray intensities in the decay of ^{79}Kr and ^{127}Xe was undertaken to resolve some of the discrepancies and to reduce some of the uncertainties in existing decay schemes. Furthermore, these absolute γ-ray intensities are needed to assay accurately ^{79}Kr and ^{127}Xe by γ-ray spectrometry for various applications in nuclear medicine and health physics[1] and in nuclear chemistry and physics.[2]

The level structure of ^{79}Br is considerably documented from investigations of the decay of ^{79}Kr[3-8] as well as from nuclear-reaction studies.[4,7-10] The known level scheme consists of 12 excited states accommodating approximately 40 γ-ray transitions.[8,11,12] The placement of γ rays within this level scheme (as shown in Fig. 1) was previously determined from balancing γ-ray energies, from $\gamma\gamma$- and $e\gamma$-coincidence measurements[3,5,6,8] and the results of resonance fluorescence[4,8,9] and Coulomb excitation.[7] Absolute intensities for the transitions were primarily determined by balancing the decay scheme in the usual manner using only relative γ-ray intensities[6-8] and positron-to-electron-capture ratios.[11,12] Inasmuch as none of the previous measurements were based on absolute disintegration rates and since the decay to the ground state cannot be accounted for by just the relative measurements, the intensities were not known to better than ±(25–30)%.[12]

The properties of the ground-state decay of ^{127}Xe are known to a higher accuracy. A level scheme (Fig. 2) which consists of only three excited states accounts for over 99.9% of the decay.[13] The five transitions between the four lowest levels have been extensively studied by $\gamma\gamma$,[14-21] $e\gamma$[17,21-24] and $X\gamma$ coincidences,[14,21-24] and $\gamma\gamma$ and $e\gamma$ angular correlations.[15-20] As a result, definite level energies, lifetimes, spins, and parities for each of the lowest four levels in ^{127}I appear to be well established.[13] Absolute intensities for these five transitions in the decay of ^{127}Xe were derived mainly from relative γ-ray intensities,[14,25,26] relative internal-conversion-electron intensities,[17,25,27,28] and electron-capture ratios[25,26,29,30] requiring the usual intensity balances. These intensities were deduced

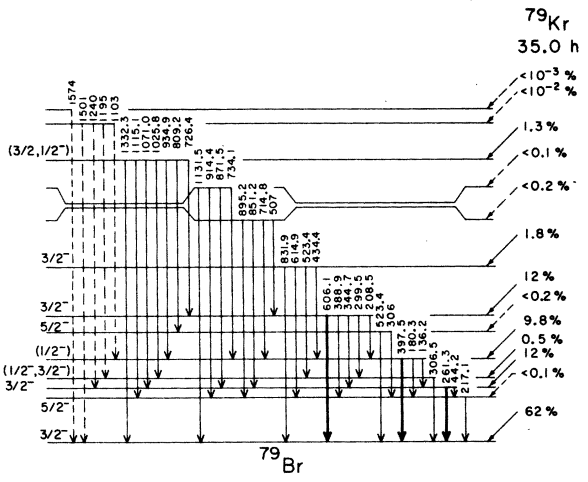

FIG. 1. Decay scheme of ^{79}Kr adopted from Ref. 8. The labeled energies of the transitions, with the exception of those from the 1574- and 1501-keV levels, are those obtained in this work. Total branching ratios for β decay (positron emission plus electron capture) to each level fed in the decay were deduced from balancing the presently measured absolute γ-ray intensities (see text).

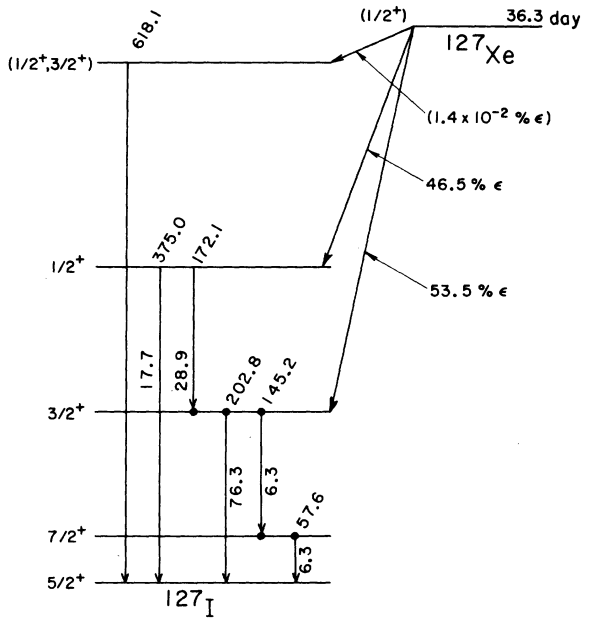

FIG. 2. Decay scheme of ^{127}Xe adopted from the compilation of Ref. 13. With the exception of the weak transition to the 618.1-keV level (Ref. 26), the labeled transition intensities (per 100 disintegrations) are those deduced from the absolute γ-ray intensities measured in this work (see text).

from experimentally determined multipolarity mixing ratios[28] and theoretical internal-conversion coefficients.[31] Although the transition intensities deduced from these previous measurements appear to be well known,[13] no absolute intensity (either γ-ray or internal-conversion-electron) measurements have ever been made. Therefore, a direct comparison of the reported intensities with values obtained from absolute γ-ray measurements should be of value.

II. EXPERIMENTAL TECHNIQUES

Sources of ^{79}Kr and ^{127}Xe were produced from (p,n) reactions by irradiating encapsulated potassium-halide targets with protons from the Brookhaven tandem Van de Graaff accelerators. The targets consisted of $\approx$2 mg/cm^2 of either KBr or KI vacuum evaporated over a 2$\times$2-cm area on $\approx$30-mg/cm^2 high-purity aluminum foil. The source to collector distance used for the evaporations was $\approx$45 cm and as a result the depositions over the 4-cm^2 area were extremely uniform. The targets were then encapsulated in vacuum between two sheets of $\approx$12-mg/cm^2 heat-sealable Mylar. It was found that without such encapsulation nearly 90% of ^{79}Kr produced in a target would escape by diffusion within one day after the irradiation. The Mylar-encapsulated targets, however, were shown to quantitatively retain both the ^{79}Kr and ^{127}Xe gas over the entire measurement period. This was established by comparison with targets completely enclosed in Ag foil packets which had been soldered together in vacuum. The sources were prepared by irradiations with 7- to 13-MeV protons. Each irradiation was of 30-min duration and was performed with a rather uniformly diffuse beam collimated to 1-cm diameter. The average beam current for the irradiations was typically 100-300 nA. Larger beam currents were not used since these would have produced severe radiation damage to the Mylar covers and possible loss of the radioactive gases.

To determine the absolute γ-ray intensities, the sources were first assayed with calibrated Ge(Li) detectors. γ-ray spectra for each of the irradiated targets were obtained at a variety of counting geometries with two large volume ($\approx$40- and $\approx$65-cm^3) high-resolution (full width at half maximum of $\approx$2 keV at 1332 keV) Ge(Li) coaxial detectors and 4096-channel pulse-height analyzers. The dead-time losses of the analyzers were always kept at less than 2%. A total of five different combinations of counting geometries and detectors were used. Each source was assayed in several of these combinations to minimize the possibility of errors in a single-detector calibration. The detectors were previously calibrated for efficiency (at the fixed counting geometries) with absolute International Atomic Energy Agency (IAEA) and National Bureau of Standard (NBS) standard sources.[32] Copper absorbers (369 mg/cm^2) were placed over the sources during counting in order to annihilate the β^+ radiation at the sources and prevent the positrons from entering the detector, and to attenuate the x rays to minimize coincident summing with γ rays. Analysis of the γ-ray spectra was performed by means of a modified version of the computer code BRUTAL.[33] After applying the efficiencies to obtain the γ-ray intensities, corrections were also made for absorption in both the source and the Cu absorber, and for summation of coincident γ rays, x-ray, and annihilation radiation. Energy measurements of the more intense γ rays were obtained from internal energy calibrations by simultaneously counting the sources with IAEA standards. Once the energies of these more intense γ rays were established, they in turn were used as secondary standards to determine the energies of the weaker lines. To account for any nonlinearity in the system during these calibrations, the energies were calculated by fitting the standard energies in the spectrum to a cubic polynomial in the computer analysis.[33]

Following the γ-ray measurements, the targets were processed to separate and collect quantitatively the gaseous activity for the absolute disintegration measurements. The targets were placed in glass ampules, evacuated, and a measured amount of either Kr or Xe carrier was added. The ampules were then heated to decompose the Mylar and release the radioactive gases from the packet. Water was then admitted to the evacuated system to completely dissolve the salt and to insure complete mixing of the radioactive and carrier gases. The samples were then transferred to a high-vacuum system through a trap cooled in a dry-ice/acetone bath and purified by gettering with hot

TABLE I. γ-ray transitions and intensities in the decay of ^{79}Kr. Comparison of the absolute γ-ray intensities measured in this work with the compilations of Refs. 11 and 12 and with the relative intensities of Ref. 8. Intensities are given as the number of photons per 1000 disintegrations.

Present work		Artna (Ref. 11)	Martin and Blichert-Toft (Ref. 12)	Weiss, Langhoff, and Schumacher (Ref. 8)	
E_γ (keV)	I_γ (abs.)	I_γ (abs.)	I_γ (abs.)	E_γ (keV)	I_γ (rel.)
44.2 ± 0.4	2.1 ± 0.2		1.2 ± 0.4	44.3	2.0
136.2 ± 0.1	10. ± 1.	7.	6. ± 2.	136.2 ± 0.3	8.3
180.3 ± 0.2	1.0 ± 0.5	0.8	<0.9	180.3 ± 0.5	0.14
208.5 ± 0.1	7.8 ± 0.4	7.	7. ± 2.	208.5 ± 0.2	7.0
217.1 ± 0.1	24. ± 1.	20.	18. ± 5.	217.1 ± 0.2	25.
261.3 ± 0.1	127. ± 4.	115.	110. ± 30.	261.4 ± 0.2	(127.)[a]
[b]				299.2	<0.4
299.5 ± 0.2	15.7 ± 0.7	13.	13. ± 4.	299.6 ± 0.2	15.
[b]				305.7	0.09
306.5 ± 0.1	26. ± 1.	25.	22. ± 6.	306.6 ± 0.2	24.
344.7 ± 0.2	2.4 ± 0.1	1.	1.3 ± 0.5	344.9 ± 0.5	1.9
388.9 ± 0.1	15.2 ± 0.7	16.	14. ± 4.	389.1 ± 0.2	16.5
397.5 ± 0.1	95. ± 3.	101.	80. ± 30.	397.6 ± 0.2	83.
434.4 ± 0.3	0.40 ± 0.05			434.7 ± 0.5	0.38
[b]				506.6 ± 1.0	0.89
523.4 ± 0.2	2.5 ± 0.1	3.	<3.	522.8	0.9
525.4 ± 0.2	4.3 ± 0.2	2.	3.0 ± 0.9	525.4	3.3
606.1 ± 0.1	81. ± 2.	97.	80. ± 30.	606.3 ± 0.3	72.
614.9 ± 0.3	0.96 ± 0.06	1.	1.1 ± 0.5	614.9 ± 0.8	1.7
650.8 ± 0.4	0.14 ± 0.02			651.3 ± 0.5	0.11
714.8 ± 0.4	0.06 ± 0.02			715.4 ± 0.5	0.18
726.4 ± 0.4	0.19 ± 0.03	0.5	0.4 ± 0.2	726.5 ± 0.5	0.14
734.1 ± 0.4	0.14 ± 0.03			734.2 ± 0.5	0.08
809.2 ± 0.3	0.92 ± 0.06	1.4	1.3 ± 0.4	809.1 ± 0.3	1.0
831.9 ± 0.1	12.6 ± 0.6	17.	12. ± 4.	832.0 ± 0.3	12.4
851.2 ± 0.4	0.38 ± 0.04			851.2 ± 0.5	0.38
871.5 ± 0.5	<0.10 ± 0.05			871.0 ± 0.5	0.05
895.2 ± 0.4	0.11 ± 0.02			895.6 ± 0.5	0.10
914.4 ± 0.5	0.07 ± 0.03			915.1 ± 0.5	0.11
934.9 ± 0.3	1.26 ± 0.07	1.0	1.1 ± 0.3	934.8 ± 0.3	1.1
1025.8 ± 0.3	1.56 ± 0.09	1.3	1.3 ± 0.4	1025.9 ± 0.3	1.52
1071.0 ± 0.3	0.65 ± 0.08	0.9	0.8 ± 0.2	1071.0 ± 0.3	0.64
1075.7 ± 0.4	0.09 ± 0.02			1076.7 ± 0.5	0.05
[c]				1103.8 ± 0.5	0.025
1115.2 ± 0.3	3.7 ± 0.2	4.	3.3 ± 0.9	1114.9 ± 0.3	3.8
1131.5 ± 0.4	0.42 ± 0.04			1131.6 ± 0.4	0.38
[c]				1195.4 ± 0.5	0.025
[c]				1240.2 ± 0.5	0.010
1332.3 ± 0.1	4.4 ± 0.3	5.	3.4 ± 1.1	1332.2 ± 0.2	4.2
[c]				1501.6 ± 0.5	0.001
[c]				1573.6 ± 0.5	0.005

[a] Normalized to this value.
[b] Could not be resolved, see text.
[c] Not observed, intensity must be <0.04, see text.

titanium at ≈900°C for about 20 min. The gas samples were transferred into cylindrical (1.9-cm-i.d., 30-cm-long) Bernstein-Ballentine[34] proportional counters. Experimental measurements indicated no loss (<1%) in this over-all processing. The counters were then filled with P-10 counting gas (90% Ar, 10% CH_4) to slighlty above 1 atm. Counting rate versus applied voltage of these counters exhibited both the onset of β^+ emission and distinct levels corresponding to the K- and L-shell electron capture. The counters were operated near the higher-voltage ends of the total sum plateaus. It has been shown[35] that under these conditions counters quantitatively detect L-capture events in ^{37}Ar(≃183 eV); therefore, virtually every disintegration of ^{79}Kr and ^{127}Xe either by electron capture or β^+ emission within the sensitive volume will be detected. The total counter efficiencies are given by the ratios of the sensitive volume to total volume. For these counters it has been shown[34,35] that the end effects are less than 1% and that within the over-all counting errors the sensitive volumes can be taken as the cathode volumes. These volume ratios (cathode to total) were previously measured to be (82–84)%. The dead time for these counters is 2 μsec. In obtaining decay curves for the gas samples, each count was of sufficient duration that the statistical uncertainty on every data point was less than 0.3%. The resultant decay curves were analyzed by a least-squares procedure using the CLSQ[36] program. The least-squares fits were used to obtain both the half-lives (see Appendix) and the absolute disintegration rates at the end of irradiation after applying the counter efficiencies. The absolute γ-ray intensities were then determined from the γ-ray emission rates (extrapolated to the end of irradiation) and these disintegration rates.

III. RESULTS AND DISCUSSION

The γ-ray energies and absolute intensities in the decay of ^{79}Kr which were obtained in this work are provided in Table I. The intensities are given as the number of photons per 1000 disintegrations. These results are based on absolute-disintegration-rate measurements from three ^{79}Kr sources. In addition, however, several other ^{79}Kr sources were used to obtain relative γ-ray intensities for the weaker transitions which were then normalized to the absolute intensities of the three strongest transitions. The error limits on the tabulated values of the absolute γ-ray intensities are 3 times the standard deviations of the means.

Of the 41 transitions observed by Weiss, Langhoff, and Schumacher,[8] 8 were not seen in this work. The 299.2-, 305.7-, and 506.6-keV γ rays which they[8] observed could not be resolved from the much more intense adjacent lines in our spectra. All five high-energy transitions resulting from deexcitation of the levels at 1574 and 1501 keV (see Fig. 1) were also not seen. Based upon counting a ≈1-μCi ^{79}Kr source for about 3 days, an upper limit of 4×10^{-5} photons per disintegration can be placed on the intensity of these five transitions. The previously reported[8] intensities are lower than this limit. Although there are no major discrepancies between the present results and the relative intensities of Weiss, Langhoff, and Schumacher[8] (normalized to the 261.3-keV γ ray), there are differences of as much as (10–15)% for some of the more intense transitions. In comparison to earlier less complete relative γ-ray intensity measurements for the major transitions, the results of Robinson *et al.*[7] (again normalized to the 261.3-keV γ ray) are in slightly better agreement with the present work, while the agreement with Langhoff *et al.*[6] and Bonacalza[5] is considerably poorer. The absolute intensities given in the compilations of Artna[11] and Martin and Blichert-Toft,[12] which were obtained by balancing the ^{79}Kr decay scheme, are also given in Table I. The comparison with Artna's[11] compilation indicates that while the intensities for the 261.3- and 397.5-keV transitions are in agreement to better than 10%, the intensities for the 606.1-keV transition differ by 20%. In contradistinction, the comparison with the compilation of Martin and Blichert-Toft[12] indicates that although the intensities for the 606.1-keV γ ray are in excellent agreement, their values for the 261.3- and 397.5-keV γ rays are low by (15–20)%. It was mainly this incongruence between the two sets and the large uncertainties[12] associated with each which motivated this study.

The percentage of β decay (positron emission

TABLE II. γ-ray transitions and intensities in the decay of ^{127}Xe. Comparison of the absolute γ-ray intensities measured in this work with the relative γ-ray intensities of Forrest *et al.* and the relative γ-ray intensities calculated by Geiger from his relative internal-conversion-electron intensities. Intensities are given as the number of photons per 100 disintegrations.

Present work		Forrest and Easterday (Ref. 25)	Geiger (Ref. 28)
E_γ (keV)	I_γ (abs.)	I_γ (rel.)	I_γ (rel.)
57.6 ± 0.2	1.31 ± 0.08	1.43 ± 0.14	1.33
145.2 ± 0.1	4.24 ± 0.21	4.36 ± 0.68	4.13
172.1 ± 0.1	24.7 ± 1.0	23.3 ± 1.2	25.7
202.8 ± 0.1	68.1 ± 1.3	(68.1) [a] ± 2.7	(68.1) [a]
375.0 ± 0.1	17.4 ± 1.0	20.5 ± 0.8	17.9

[a] Normalized to this value.

plus electron capture) to each level in ^{79}Br is provided in Fig. 1. They were obtained by balancing the decay scheme with absolute transition intensities. These were deduced by combining the absolute γ-ray intensities of the present work with theoretical total conversion coefficients[31] for their reported multipolarities.[8,11] For the weak transitions the multipolarities were not always known, but any reasonable assignment will not affect the intensity balance. These β- decay percentages are in good agreement with those given in the compilations of Artna[11] and Martin and Blichert-Toft.[12]

The present results for the γ-ray energies and absolute intensities (per 100 disintegrations) in the decay of ^{127}Xe are given in Table II. The tabulated intensities are the averages of the results from three ^{127}Xe sources. The quoted errors are 3 times the standard deviations of the means. A comparison to the relative γ-ray intensity measurements of Forrest and Easterday[25] and to those calculated by Geiger[28] from his relative internal-conversion-electron intensities is also made in Table II by normalizing their results to the intensity of the 202.8-keV γ ray. With the exception of the relative intensity for the 375.0-keV γ ray reported by Forrest and Easterday,[25] all of their[25,28] other values are in excellent agreement with the present work. No attempt was made to observe the very weak ($\approx 1.4\times10^{-4}$ photons per disintegration) 618.1-keV γ ray observed by Schmidt-Ott *et al.*[26] An apparent discrepancy in the intensity of the 375.0-keV transition is evident in the recent Nuclear Data compilation of Auble.[13] The reported γ-ray intensity for this transition which was obtained from the γ-ray measurements[25] is larger than the total transition intensity deduced from the internal-conversion-electron measurements[28] (when both are normalized to the 202.8-keV transition). The total transition intensities (Table III) were obtained by the use of theoretical total internal-conversion coefficients[31] and the adopted[13] multipolarity mixing ratios obtained from Geiger's[28] study. The intensities calculated by Geiger[28] from his relative internal-conversion-electron measurements and the values taken from the compilation of Auble[13] (which were deduced from balancing the ^{127}Xe decay scheme) are also given in Table III. As indicated, all these values are in agreement to well within the stated uncertainties. This work, however, resolves the apparent discrepancy in the intensity of the 375.0-keV γ ray. The decay scheme incorporating the revised intensities is given in Fig. 2.

Note added: In a recent publication Ludington, Gardulski, and Wiedenbeck have reported[37] a decay scheme for ^{79}Kr which is in excellent agreement with the conclusions of the present work.

TABLE III. Total transition intensities in the decay of ^{127}Xe calculated from the absolute γ-ray intensities measured in this work using theoretical total internal-conversion coefficients (Ref. 31) with the adopted multipolarity mixing ratios (Ref. 13) and compared to the relative intensities calculated by Geiger from his internal-conversion-electron measurements and to the values in the Nuclear Data compilation of Auble. Intensities are given as the number of transitions per 100 disintegrations.

Transition energy (keV)	α	Present work I (abs.) = $(1+\alpha)I_\gamma$ (abs.)	Geiger (Ref. 28) I (rel.) = I_{ce} (rel.)/$(1+1/\alpha)$	Auble' (Ref. 13) I (abs.)
57.6	3.78	6.26 ± 0.38	6.2	6.20
145.2	0.48	6.28 ± 0.31	6.1	6.09
172.1	0.17	28.9 ± 1.2	29.8	29.7
202.8	0.12	76.3 ± 1.4	(76.0) [a]	75.5
375.0	0.020	17.7 ± 1.0	18.1	18.3

[a] Normalized to this value.

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APPENDIX

As an outgrowth of the present work and concurrent measurements of the excitation functions for ^{79}Br(p,n) and ^{127}I(p,n) reactions,[2] half-lives for ^{79}Kr and ^{127}Xe were determined from numerous decay curves. The target irradiations, counting techniques, and decay-curve analyses are described in Sec. II of this paper.

The half-life of ^{79}Kr was obtained from the analyzed decay curves of the gas samples. For these decay curves, each sample was counted approximately 40–45 times over a period of eight to nine half-lives. For the three samples the least-squares-fitted[36] half-lives with their statistical uncertainties were 35.005 ± 0.015 h, 35.042 ± 0.016 h, and 35.108 ± 0.019 h. The weighted (inverse square of standard deviations) mean of these three values is 35.04 ± 0.01 h. However, the internal agreement between the three values implies the above standard error of the mean should be increased by a factor of 2.66 to give 0.026. This error limit is only 1 standard deviation (σ) statistical uncertainty. To account for possible small systematic errors, such as in the timing and in the proportional counter dead times, and to increase the error to

greater than the 99.9% confidence level ($\simeq 4\sigma$), the value

$$T_{1/2}(^{79}\mathrm{Kr}) = 35.04 \pm 0.10 \text{ h}$$

is adopted. This value is in reasonable agreement with the only other high-precision measurement 34.92 ± 0.05 h.[5] Other previously reported values for this half-life are 34.5 ± 1.0,[38] 36 ± 1,[27] 34.5 ± 0.2,[39] 34.3,[40] and 34 h.[41]

The ^{127}Xe half-life was determined from both the gas proportional counting and γ-ray spectrometry data. As a result of an instrumental failure, the ^{127}Xe gas samples were assayed for only approximately one half-life. The fitted half-lives for the two most reliable determinations were 36.16 ± 0.16 and 36.59 ± 0.31 day which have a weighted average of 36.25 ± 0.14 day. The half-life could not be determined to a high precision from these measurements because of the short decay-time interval and the instrumental problems. Therefore, it was decided to include the results from γ-ray counting. A total of 48 γ-ray decay curves obtained from counting the 145.2-, 172.1-, 202.8-, and 375.0-keV γ rays from 12 ^{127}Xe targets were used. These decay curves were followed for 2–4 half-lives. The unweighted mean of these 48 fitted half-lives was 36.14 ± 0.10 day. The weighted mean was 36.25 ± 0.09 day. On combining the weighted means of the results from both the gas and γ-ray counting, and again increasing the error limit to $\simeq 4\sigma$, the adopted value for the half-life is

$$T_{1/2}(^{127}\mathrm{Xe}) = 36.3 \pm 0.3 \text{ day}.$$

Previously reported values for this half-life include: 34 ± 2,[38] 32 ± 2,[42] 36.0 ± 0.5,[25] 36.4 ± 0.1,[29] 36.5 ± 0.1,[30] and 36.406 ± 0.016 day.[43] The last value, a mass spectrometric determination by Balestrini[43] with its reported high precision, has unduly influenced the adopted value.[13] If one considers that this result is based on a single determination and that his random reproducibility is only about 0.5% for the mass ratios, the reported uncertainty is probably underestimated by an order of magnitude. On increasing the error limit to reflect this, the uncertainty would be comparable to that of this work and in Refs. 25, 29, and 30.

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