Electron capture decay of $^{81}Kr^m$

M. M. Lowry, R. T. Kouzes, F. Loeser, A. B. McDonald, and R. A. Naumann
Department of Physics, Princeton University, Princeton, New Jersey 08544
(Received 12 November 1986)

The $^{81}Kr^m$ electron capture branching ratio to ^{81}Br has been remeasured and found to be $(2.26\pm0.32)\times10^{-5}$ with a $\log ft$ value of 4.93 ± 0.06. This result is 2.5 times smaller than a previous measurement and implies a similar reduction in the rate for solar neutrino capture in an ^{81}Br-based detector.

This paper describes an improved measurement of the electron capture branching ratio for the decay of the 190 keV, 13-sec isomeric state of ^{81}Kr to the ground state of ^{81}Br (see Fig. 1). This Gamow-Teller transition dominates the capture cross section for low energy solar neutrinos on ^{81}Br. Recent proposals for large-scale solar neutrino experiments using ^{81}Br have emphasized the need for accurate ($\sim15\%$) determinations of this transition strength. We therefore undertook the present measurement, using techniques similar to those applied in a previous measurement[1] in our laboratory, but with substantially improved counting rates. With these improved rates, we were able to identify a large background contamination which invalidated the previous result. The $^{81}Kr^m$ production technique used in the present experiment reduces this background to a negligible level and enables an improved measurement with an accuracy $\sim15\%$.

The branching ratio for electron capture of $^{81}Kr^m$ is measured by comparing the number of Br $K\alpha$ x rays observed from a pure sample of $^{81}Kr^m$ with the far more abundant Kr $K\alpha$ x rays produced in the internal conversion of the isomeric state. As in the previous measurement, the Princeton Isotope Separator was used to implant $^{81}Kr^m$ in a Mylar tape which was then moved to a detection region for counting. To reduce the background from the tail of the Kr $K\alpha$ x-ray peak, a plastic scintillator detector with 98% efficiency for internal conversion electrons was used to veto such events. X-ray spectra were accumulated for five successive time intervals following the tape movement to confirm that the Br K x rays exhibited the 13-sec half-life characteristic of $^{81}Kr^m$.

In the earlier experiment, 22 MeV protons were used to produce $^{81}Kr^m$ via the (p,n) reaction on a solid potassium bromide target. In our initial attempt to improve this measurement, we used a boiling liquid target of bromoform. This target provided a larger count rate and much longer target lifetime than the original experiment. With this improved target system, we were able to examine a broad mass region near mass 81 and thus discovered a defocused, long-lived activity which produced a background of Br x rays. This contaminant is probably due to ^{79}Kr (34.9 h) which is also prolifically produced in (p,n) reactions on the natural bromine targets used. The contaminant accounted for a large fraction of the Br x rays observed at mass 81 and was undoubtedly also present in the original measurement. This background could be reduced by improving the mass resolution of the isotope separator, but it was decided to also change the method of producing $^{81}Kr^m$ to reduce the background to a negligible level.

$^{81}Kr^m$ was obtained from the decay of ^{81}Rb (4.58 h) which selectively populates the 190 keV isomeric state. Millicurie sources of ^{81}Rb were obtained from Medi-Physics Inc., South Plainfield, NJ. The ^{81}Rb was extracted with HCl solution from the ion exchange resin on which it was supplied, dried, and placed in the ion source region of the Princeton Isotope Separator. The emanating gas containing $^{81}Kr^m$ was ionized, accelerated to 60 keV, mass separated for 81, and implanted in an aluminized Mylar tape (25 μm thickness). A collimator allowed only the central portion of the isotope separator beam to reach the tape. After accumulating for 30 sec, the tape containing the $^{81}Kr^m$ was pulled into the detection region shown in Fig. 2 and a new section of tape began accumulating.

In the detection region the tape-borne source was viewed by a lithium drifted silicon detector [Si(Li)] and two vanes of 0.5 mm Pilot B scintillator coupled to an RCA 8575 photomultiplier. A major improvement was obtained in this experiment by mounting the scintillator within a slot in the light pipe, producing much better light collection and improving the detection efficiency from 79% to 98%. As shown in Fig. 2, a reentrant cavity for the Si(Li) was provided. For x rays in the region of the Br $K\alpha$ peak at 11.94 keV, the Si(Li) detector had a resolution of approximately 240 eV and a maximum total rate of about 1500 counts/sec. The scintillator was primarily sensitive to internal conversion electrons and operated at a maximum rate of about 150 000 counts/sec. A time-to-pulse-height converter (TPHC) operating on a 100 nsec scale was started by the Si(Li) and stopped either by an event in the scintillator or by the start signal delayed by 60 nsec. The coincidence peak width was 14 nsec FWHM. A valid TPHC pulse was required for every event so that dead time in the TPHC could not distort the Br to Kr ratio. Windows on the coincidence peak and the anticoincidence peak were used to sort the x-ray events from the Si(Li) detector into two spectra. Spectra from five successive 6 sec intervals following tap movement were separately accumulated in order to allow a measurement of the half-life for the Br events. The data taking sequencing was provided by a crystal-controlled microprocessor.

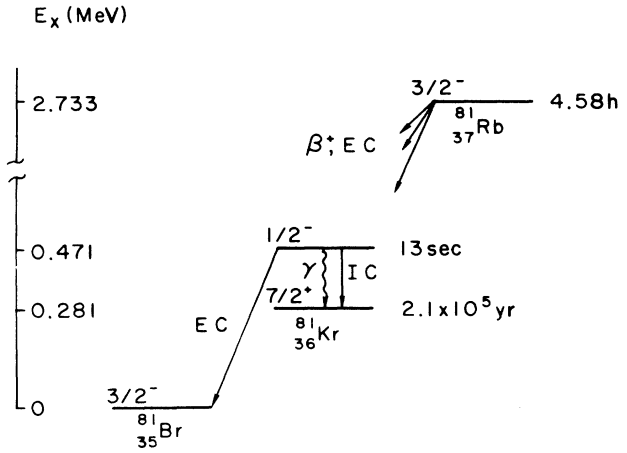

FIG. 1. Decay scheme of $^{81}Kr^m$, showing population of the isomer by ^{81}Rb as well as subsequent gamma ray, internal conversion, and electron capture decay branches.

A single ^{81}Rb generator provided an adequate count rate for about 15 h of data taking. Ten generators were used during two running periods at a rate of one per day. Long-term observation of a ^{81}Rb generator with a lithium drifted germanium gamma ray detector revealed a small ^{83}Rb ($t_{1/2}=86.2$ d) contamination. ^{83}Rb decays to $^{83}Kr^m$ ($t_{1/2}=1.83$ h). The isomer decays exclusively to the stable ^{83}K ground state, therefore producing no Br $K\alpha$ x rays. Internal conversion from this state could, in principle, contribute to the number of Kr $K\alpha$ x rays, but the low activity, the isotope separation, and the short residence time in front of the counters make this contribution negligible.

Figure 3(a) shows the sum of all our data for the x rays in anticoincidence with internal conversion (IC) electrons in the scintillator. Those in the coincidence are shown in Fig. 3(b). The much greater statistical accuracy of the krypton K x-ray peaks in Fig. 3(b) allows that spectrum to be scaled and subtracted from the anticoincidence one to produce Fig. 3(c), in which 80% of the Kr $K\alpha$ x-ray peak has been subtracted away. With this subtraction, a significant peak at the energy of the Br $K\alpha$ x ray can be seen in Fig. 3(c). In order to check the consistency and stability of this peak, spectra for individual days were subtracted in this way and analyzed as follows. The data were grouped in two different ways and areas determined with the peak-fitting program FITEK (Ref. 2) to produce the results in Table I. The upper part of the table shows the result of summing over the five (6 sec) decay intervals and grouping into three roughly equivalent data collection periods. The lower part of the table gives the result for each of the decay intervals summed over all the data collected.

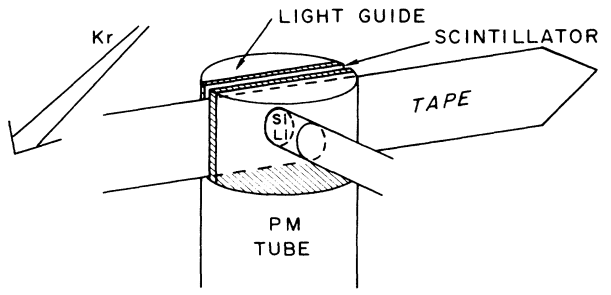

FIG. 2. Schematic overview of the source collection, tape transport, and detection regions of the experiment, showing the high geometric solid angle of the scintillator, improved light coupling to the photomultiplier (PM) and the reentrant cavity for the lithium-drifted silicon detector [Si(Li)].

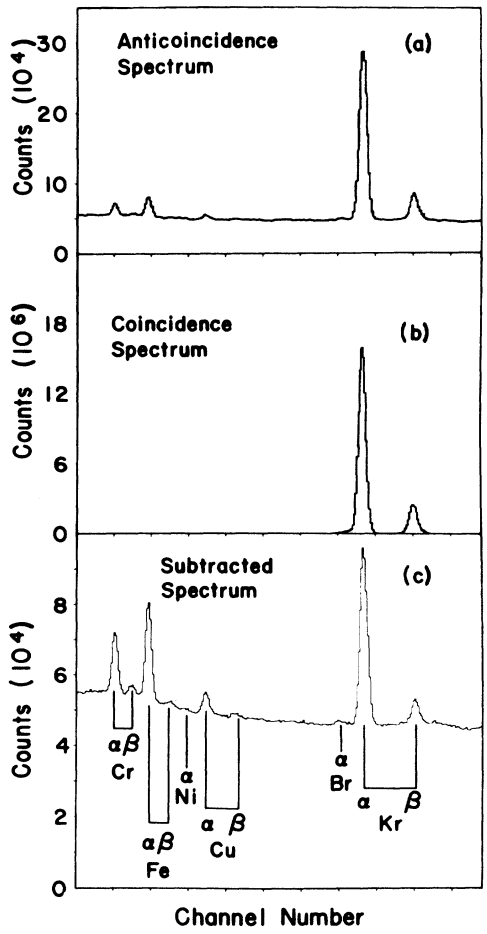

FIG. 3. X-ray spectra taken by the lithium-drifted silicon detector summed over all runs and all decay intervals. (a) X rays not in coincidence with internal conversion electrons detected in the scintillator. The flat background arises from Compton scattering of the 190 keV gamma branch; the krypton x rays from incomplete vetoing by the scintillator; and the Cr, Fe, Ni, and Cu x rays from fluorescing of the detector components by the 190 keV gamma rays. (b) X rays in coincidence with interconversion electrons. Fluorescence of detector constituents and the silicon escape peak are not visible on this scale. (c) Anticoincidence spectrum minus a scaled version of the coincidence one so as to remove 80% of the Kr $K\alpha$ x rays. The small bromine $K\alpha$ x-ray peak is clearly visible on the low energy side of the krypton $K\alpha$ peak.

TABLE I. Peak areas and ratios[a] for the x-ray spectra grouped by run and by decay interval.

	Chronological intervals			
	Kr $K\alpha$ coincidences	Kr $K\alpha$ anticoincidences	Br $K\alpha$	Observed ratio ($\times 10^{-5}$)[a]
April 1986	37 434 352	616 558	2760±505	7.25 (18.3%)
July 1986-1	33 030 192	454 031	2393±424	7.15 (17.7%)
July 1986-2	23 999 040	357 734	1635±402	6.71 (24.6%)
	94 463 584	1 428 323	6788±772	7.08 (11.4%)
	Decay intervals			
Time after source arrival (sec)	Kr $K\alpha$ coincidences	Kr $K\alpha$ anticoincidences	Br $K\alpha$	Observed ratio ($\times 10^{-5}$)[a]
0–6	30 952 480	519 609	2355±403	7.48 (17.1%)
6–12	23 790 928	351 740	1956±480	8.10 (24.5%)
12–18	17 996 304	251 826	1200±378	6.57 (31.5%)
18–24	13 521 351	183 782	614±305	4.48 (49.7%)
24–30	10 091 307	134 251	566±265	5.53 (46.8%)
	96 352 370	1 441 208	6690±836	6.83 (12.5%)

from fit Br $K\alpha = 6660.5 \pm 806.2 \Rightarrow 6.81 \times 10^{-5}$ (12.1%).

[a]Ratio=(Br $K\alpha$)/($\sum$ Kr $K\alpha$).

As can be seen, the groupings all give a ratio which is consistent with our adopted value of (6.9×10^{-5}) (±13%) for the ratio of Br $K\alpha$ x rays to Kr $K\alpha$ x rays. The Br x-ray peak areas in the lower part of the table can be fitted to the exponential decay of $^{81}Kr^m$ ($t_{1/2} = 13.10 \pm 0.02$ sec) (Ref. 3) with no background contribution. The one-parameter fit gives a reduced χ^2 of 0.3 and implies a total of 6661±806 Br $K\alpha$ x rays in the five intervals, as given at the bottom of Table I. (See Fig. 4.) A two-parameter fit, including a possible flat background such as from long-lived activities, still results in zero background with an upper limit for the background (90% confidence) of 1000 counts or 15% of the observed total.

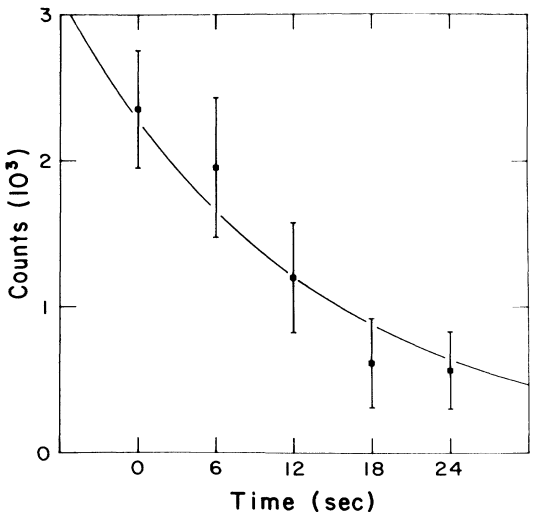

FIG. 4. Decay of the bromine $K\alpha$ peak. Peak areas for sequential 6 sec time bins are plotted at the time after source arrival that accumulation began in that bin. The curve is a one-parameter fit to the data with the half-life fixed at 13.1 sec and with a reduced χ^2 of 0.3.

More stringent limits can be placed by considering the possible Br x-ray producing contaminants. Most can be ruled out by the requirements that they still be present at least 4 h after production and survive being dissolved in HCl and then dried, a process which removes any gaseous components. The only remaining possibilities are ^{79}Rb (23 min) and $^{80}Br^m$ (4.42 h). Data were recorded at these two mass settings of the Isotope Separator for approximately an hour each. Small amounts of Kr x rays were seen and upper limits were established on possible Br x rays. Assuming the Kr x rays arise from incomplete mass separation of the $^{81}Kr^m$ gives mass separation efficiencies of 3.2×10^{-4} and 8×10^{-5} for 1 and 2 mass number spacings. Applying these to the observed Br x-ray upper limits implies upper limits (90% confidence) for mass 79 and 80 contributions to the total number of mass 81 Br $K\alpha$ x rays of 0.1 and 0.6 counts, respectively. These values are negligibly small, and therefore no additional uncertainty arises from the possibility of contaminants in the Br x rays.

The high separation efficiencies observed and lack of any large amount of other Kr x-ray producing activities means that the only source for contamination of that peak arises from the possibility of ^{81}Rb being vaporized, included in the isotope separator beam, and deposited on the tape. The Rb electron capture decay branch would lead to an x ray with no electron and so would be present in the anticoincidence spectrum only. Since this component would have a nearly constant rate over the 30 sec observation period, the number of anticoincident Kr $K\alpha$ x rays in bin 5 sets an upper limit of $<0.8\%$ for the fraction of Kr $K\alpha$ x rays from ^{81}Rb decay itself. This is a negligible

TABLE II. Corrections to observed $K\alpha$ x-ray ratio to obtain total branching ratio.

Corrections			Value	Refs.
(1) Relative efficiency	$\frac{1/\epsilon(11.92\ \text{keV})}{1/\epsilon(12.65\ \text{keV})}$	=	1.018 (1%)	4
(2) Total K x ray to $K\alpha$	$\frac{K/K\alpha\ (\text{Br})}{K/K\alpha\ (\text{Kr})}$	=	0.988 (3%)	5
(3) Fluorescence yield	$\frac{1/\omega_K\ (\text{Br})}{1/\omega_K\ (\text{Kr})}$	=	1.039 (3%)	5
(4) Total to K captures or conversions	$\frac{1+\text{EC}(L+M+\cdots)/\text{EC}(K)}{1+\text{IC}(L+M+\cdots)/\text{IC}(K)}$	$=\frac{1+0.118}{1+1/4.53}$	=0.916 (3%)	5 6
(5) Total decays to conversions	$\frac{1}{1+\gamma/\text{IC}}$	$=\frac{1}{1+1/0.49}$	=0.329 (1.5%)	5
			Total=0.315 (5.5%)	

contribution to the uncertainty.

The ratio of observed $K\alpha$ x rays must be multiplied by a number of corrections in order to obtain the total $^{81}Kr^m$ electron capture (EC) branching ratio. These corrections and their assumed values are listed in Table II and give a net correction factor of 0.315 ($\pm 5.5\%$). A measured correction of 1.042 ($\pm 1\%$) must be applied to account for Br x rays lost due to random coincidences with the scintillator. This yields a value of $(2.26\pm 0.32)\times 10^{-5}$ for the branching ratio. Combining this with a lifetime of 13.1 sec and a Q value of 0.471 MeV,[7] which implies a log$f$ of -0.83,[8] gives a logft of 4.93 ± 0.06 for the Kr ($\frac{1}{2}^-$) to Br ($\frac{3}{2}^-$) decay. This implies a logft for the inverse Br ($\frac{3}{2}^-$) to Kr ($\frac{1}{2}^-$) neutrino capture of 5.23 ± 0.06. It is important to note that because of the strongly suspected contamination in the original measurement, this redetermination should replace the previous result.

Another recent measurement[9] also gives a result significantly smaller than the original measurement and not inconsistent with our new determination.

This reduction in the expected yield of a bromine-based neutrino detector by a factor of approximately 2.5 is significant in terms of the detailed plans for such a detector, but does not affect the basic conclusion that such a detector is practical, provided a method of single atom counting of krypton can be developed. Moreover, the improved precision of the present measurement will allow the results from such a detector to be clearly interpreted in terms of a solar neutrino flux.

We thank Cary Davids for helpful suggestions and discussions. This work was supported by a grant from the National Science Foundation.

[1]C. L. Bennett, M. M. Lowry, R. A. Naumann, F. Loeser, and W. H. Moore, Phys. Rev. C **22**, 2245 (1980).

[2]W. Stoeffl (unpublished).

[3]M. M. Lowry, R. T. Kouzes, A. B. McDonald, and F. P. Calaprice, Bull. Am. Phys. Soc. **31**, 835 (1986), and (unpublished).

[4]E. C. Montenegro, G. B. Baptista, and P. W. E. P. Duarte, At. Data Nucl. Data Tables **22**, 131 (1978).

[5]*Table of the Isotopes*, 7th ed., edited by C. M. Lederer and V. S. Shirley (Wiley, New York, 1978).

[6]S. Vaisala, T. Raunemaa, A. Fontell, G. Graeffe, and A. Siivola, Phys. Fenn. **10**, 133 (1975).

[7]R. T. Kouzes, M. M. Lowry, and C. L. Bennett, Phys. Rev. C **25**, 1076 (1982).

[8]N. B. Gove and M. J. Martin, Nucl. Data Tables **10**, 205 (1971).

[9]C. N. Davids, T. F. Wang, I. Ahmad, R. Holzmann, and R. V. F. Janssens, Phys. Rev. C **35**, 1114 (1987).