Electron-capture branching ratio of $^{81}Kr^m$

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The branching ratio of the electron-capture decay of $^{81}Kr^m$ (13 s) has been measured in order to obtain the rate of the neutrino capture reaction $^{81}Br(\nu,e^-)^{81}Kr(190.6\ keV)$. This quantity is needed to determine the properties of an ^{81}Br solar neutrino detector. A branching ratio of $(3.14\pm0.58)\times10^{-5}$ was obtained, leading to a logft of $5.10^{+0.06}_{-0.07}$ for this transition.

I. INTRODUCTION

One of the long-standing puzzles in modern-day astrophysics is the solar neutrino problem. The observed rate of solar neutrino capture in the only experiment designed to measure the flux of neutrinos from the sun (the ^{37}Cl experiment[1]) is about a factor of 3 lower than current calculations predict.[2] As a consequence, a number of other experiments have been proposed to help find the source of this discrepancy.

The neutrino capture process is endoergic, with a cross section proportional to $(E_\nu-E_0)^2$. Here E_ν is the incident neutrino energy and E_0 is the threshold neutrino energy for capture by the particular detector under consideration. This dependence makes each proposed experiment sensitive to a particular energy region of the calculated neutrino spectrum from the sun. The ^{71}Ga experiment has a threshold energy of 233 keV, and receives most of its signal from the p-p neutrinos. The ^{37}Cl experiment has a threshold of 814 keV, and is primarily sensitive to the neutrinos from the decay of ^8B.

Scott[3] originally proposed a radiochemical solar neutrino experiment based on ^{81}Br. He noted that neutrino capture would predominantly take place through the 13 s, $J^\pi=\frac{1}{2}^-$ isomer in ^{81}Kr at 190.6 keV (see Fig. 1). This gives the ^{81}Br detector a threshold energy of 471.2 keV, making it primarily sensitive to neutrinos from the decay of ^7Be to the ^7Li ground state. At that time, no measurement existed of the logft value for the inverse process of electron-capture (EC) decay for the isomeric state. Scott used similar transitions between neighboring nuclei to estimate a logft of 5.3, and he suggested that a measurement of the EC branching ratio for $^{81}Kr^m$ be attempted.

Bennett *et al.*[4] made the first measurement of the $^{81}Kr^m$ EC branching ratio. These authors used the Princeton AVF cyclotron to produce $^{81}Kr^m$ via the $^{nat}Br(p,n)^{81}Kr$ reaction. After separating out the more copiously produced ^{79}Kr in an isotope separator, they implanted the ^{81}Kr on a tape. This in turn was periodically moved to bring the source spot in front of a Si(Li) x-ray detector, where the Br K x rays following EC decay were observed. Placed on each side of the tape at the detector position was a thin plastic scintillator coupled via a light pipe to a photomultiplier tube. The purpose of the scintillator was to detect the electron emitted in coincidence with a Kr K x ray when the ^{81}Kr isomer decayed by internal conversion. In this way the large (10^4:1) background of Kr x rays would appear as coincidence events between the two detectors, while the Br K x ray following the EC decay would be unaccompanied by other radiation, and would therefore be detected in anticoincidence.

Because the efficiency for detecting coincidences between the Kr K x rays and their associated conversion electrons was only 79% in the experiment of Ref. 4, the anticoincidence spectrum still contained a very large Kr x-ray component. Since the Br and Kr K x rays are separated in energy by only 725 eV, this necessitated a subtraction of the remaining Kr x-ray component using the line shape obtained from the coincidence spectrum. After this was done there still remained a substantial uniform background in the anticoincidence spectrum, resulting from Compton scattering of the 190.6-keV γ ray in the Si(Li) detector. These events, unaccompanied by any signal in the scintillators, could not be removed. A value of 4.88 ± 0.11 for the logft of the $^{81}Br\rightarrow{}^{81}Kr^m$ transition was reported in Ref. 4. This was derived from their measured $^{81}Kr^m$ EC branching ratio of $(5.64\pm1.38)\times10^{-5}$.

Because of the importance of obtaining an accurate value for the EC branching ratio of $^{81}Kr^m$, as outlined above, it was felt that the measurement should be confirmed. In addition, a number of improvements to the technique pioneered by Bennett *et al.*[4] have been incorporated in the present experiment. Roughly 100-fold better rejection of the Kr K x rays has been accomplished by surrounding the Kr source completely with an electron detector. The uniform background found in the anticoincidence spectrum was reduced by arranging to detect the residual photons after Compton scattering in the Si(Li) detector. Those 190.6-keV photons undergoing forward (30°—40°) scattering in this detector deposit 10—15 keV of energy, the amount that creates unwanted background near the Br K x-ray energy of 11.91 keV. Finally, to prevent the possibility of contamination by Br x rays from ^{79}Kr decay, a different source of $^{81}Kr^m$ has been used.

II. EXPERIMENTAL PROCEDURE

Gindler *et al.*[5] have developed, at Argonne, a technique for making a ^{81}Rb-$^{81}Kr^m$ generator for use in pulmonary studies. It was decided to use such a generator as the source of the $^{81}Kr^m$. The Kr isomer is continuously pro-

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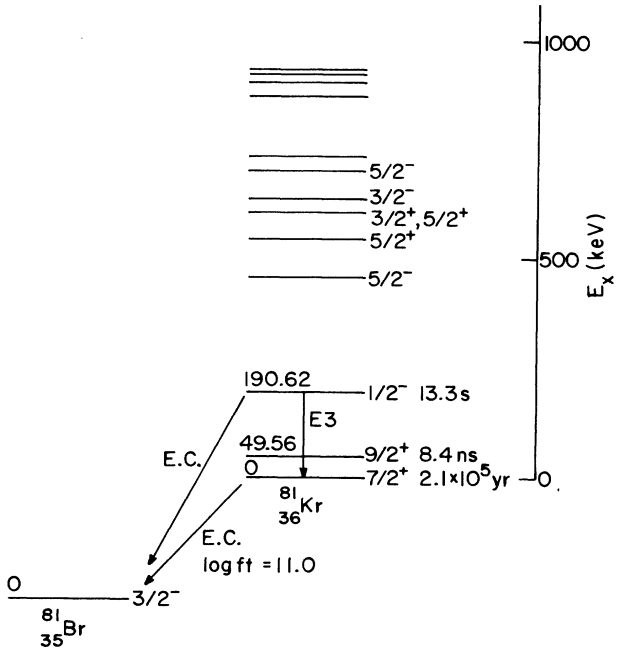

FIG. 1. Level scheme of the ^{81}Kr-^{81}Br system.

duced by the decay of 4.6-h ^{81}Rb, and can be swept out of the generator by a gas flow. This source is now available commercially.[6] The eluted Kr gas has a very high radiochemical purity (>99.9%). The remaining impurities are ^{81}Rb, $^{82}Rb^m$, and $^{83}Kr^m$, none of which lead to the emission of a Br K x ray.

For the present experiment, the $^{81}Kr^m$ was periodically swept by helium gas into a small counting cell located in front of a 200 mm^2×5 mm Si(Li) x-ray detector, with the cell itself being made of plastic scintillator. Figure 2 shows a schematic layout of the geometry. The counting cell, highly polished on all surfaces, was optically coupled to a 5 cm diam. XP2230 photomultiplier. Two layers of 17.8 μm Al foil were wrapped around it as a light shield. The actual counting volume of the cell formed a right circular cylinder, 12.7 mm in diameter and 12.7 mm high. A 1 mm diam. hole drilled in one side served as the gas inlet and outlet, and the end wall through which the x rays passed on their way to the Si(Li) detector was 1.78 mm thick. Control of the gas flow was accomplished by the use of subminiature dc solenoid valves[7] A, B, C, and D (see Fig. 2) driven by a crystal-controlled sequence timer.[8] All volumes were kept as small as possible in the gas transfer system. This ensured that only fresh isomer was delivered when gas was swept through. Operation of the entire system was checked using a source of $^{83}Kr^m$ (260 d).

Referring to Fig. 2, the counting cell was first evacuated by opening valves A and B for 2 s, with valves C and D closed. Then the cell was filled with $^{81}Kr^m$ by closing valve B and opening valve C. The cell was brought to a pressure of one atmosphere of helium by closing valve C and momentarily opening valve D. Finally, valve A was closed, and counting proceeded for 20 s. This cycle was repeated for 38 h, a period determined by the strength of the ^{81}Rb source. To prevent the escape into the room of any Kr activity, a liquid N_2-cooled tube containing a molecular sieve was placed in the vacuum line.

Since the ^{81}Rb has a half-life of 4.6 h, the detector count rates would vary widely throughout the course of the experiment if a constant cell filling time were used. To prevent pulse pileup and loss of energy resolution, the initial count rate in the Si(Li) detector was kept at about 1 kHz throughout the entire experiment. Coarse control of the $^{81}K^m$ fill rate was accomplished by restricting the gas flow in the line between the helium supply and the ^{81}Rb source. For fine control of the amount of isomer delivered to the counting cell, the output of a ratemeter measuring the count rate in the plastic scintillator was used to control the $^{81}Kr^m$ fill valve C (Fig. 2). This scheme worked well in keeping the average count rate constant throughout the experiment, during which time the source intensity dropped by a factor of 250.

To provide Compton suppression of the Si(Li) detector, four 5 cm×5 cm NaI(Tl) detectors were mounted in an array as shown in Fig. 3. After matching the gains, their fast outputs were combined to provide a signal for coincidence measurements.

Conventional fast and slow electronic modules were employed. Constant-fraction discriminators were used for

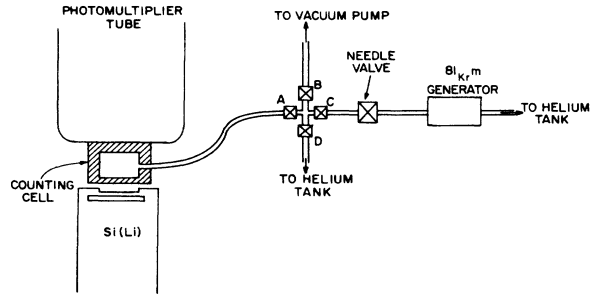

FIG. 2. Schematic drawing of the counting apparatus. The ^{81}Rb source is encased in a lead shield. Connecting tubing between the valve manifold and counting cell is also shielded by copper tubing. Not shown here is the array of four NaI(Tl) detectors used for Compton suppression.

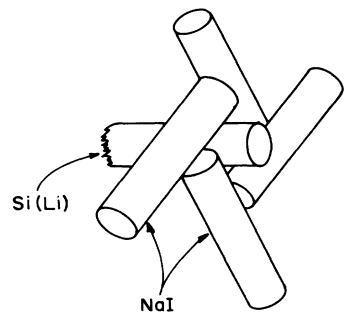

FIG. 3. Array of four NaI(Tl) detectors used for Compton suppression of the Si(Li) detector.

the fast timing, and a shaping time of 8 μs was employed for Si(Li) energy signal. All coincidence-anticoincidence relations were determined by a time-to-amplitude converter (TAC), which was started by a signal from the Si(Li) detector. The TAC was stopped either by signals from the NaI(Tl) or plastic detectors, with appropriate delays inserted in the different channels. The shortest delay occurred for the NaI(Tl)-Si(Li) coincidence, followed by the plastic-Si(Li) coincidence with a longer delay. If the Si(Li) event was not accompanied by a signal from either of the other two detectors, the TAC was stopped by a self-delayed signal from the Si(Li) discriminator. The use of this technique assured that coincidence and anticoincident events were processed in the same way.

Unaccompanied events in the Si(Li) detector such as the Br K x rays were subject to loss because of random coincidences with the high rate of plastic scintillator events ($\sim$1 MHz at the beginning of a counting cycle). The losses due to random coincidences with the NaI(Tl) detectors were negligible. To minimize these losses, events from the NaI(Tl) and plastic detectors were allowed to stop the TAC only if they fell within a 50 ns time window centered around the Si(Li) pulses. Conventional overlap timing was used for these Si(Li)-plastic and Si(Li)-NaI(Tl) coincidences. The narrow window, coupled with observed slewing in the Si(Li) detector timing, resulted in a small energy-dependent leakage from the coincidence to the anticoincidence spectrum. This manifested itself through a slight differential coincidence efficiency between the $K\alpha$ and $K\beta$ components of the Kr K x rays. However, the slightly reduced efficiency thereby resulting was more than compensated for by the benefit of having to make only a small correction for random coincidence losses, since a 99.8% overall coincidence efficiency for the Kr K x rays (see below) was achieved in the experiment.

The 20 s counting period was divided into eight 2.5 s time bins, and the bin number was digitized and stored on magnetic tape along with the other two parameters, Si(Li) energy and the time information from the TAC.

III. RESULTS

Figure 4 shows projected energy spectra in the Si(Li) detector for TAC slices corresponding to the various coincidence or anticoincidence conditions. The first panel, Fig. 4(a), gives a logarithmic plot of the events in coincidence with the plastic scintillator, mainly the Kr K x rays. Figure 4(b) shows the energy spectrum corresponding to coincidences in the NaI(Tl) gate. In the region of the spectrum near the Br K x ray, about one third of the total background is contained in the NaI(Tl) gate. Figure 4(c) shows the energy spectrum associated with the unaccompanied radiation, i.e., the anticoincidences. The data in this spectrum have been binned by combining adjacent channel pairs, but have been plotted on the same energy scale. It contains a small Kr K x-ray component, a 9.39-keV γ ray (from the decay of the ^{83}Kr^m contaminant), and some small peaks near 6 and 11 keV, associated with fluorescence peaks seen in the total energy spectrum, Fig. 4(a). In addition, there is a small peak at 11.92$\pm$0.02 keV which we identify as the sought-after Br K x ray from the EC decay of ^{81}Kr^m. Figure 4(d) shows the result of subtracting 1/450th of the plastic scintillator coincidence spectrum, Fig. 4(a), from the spectrum of Fig. 4(c). This has been done in order to decrease the contribution of the Kr K x rays. The dip at the position of the Kr $K\beta$ x ray in Fig. 4(d) results from an over-subtraction due to the

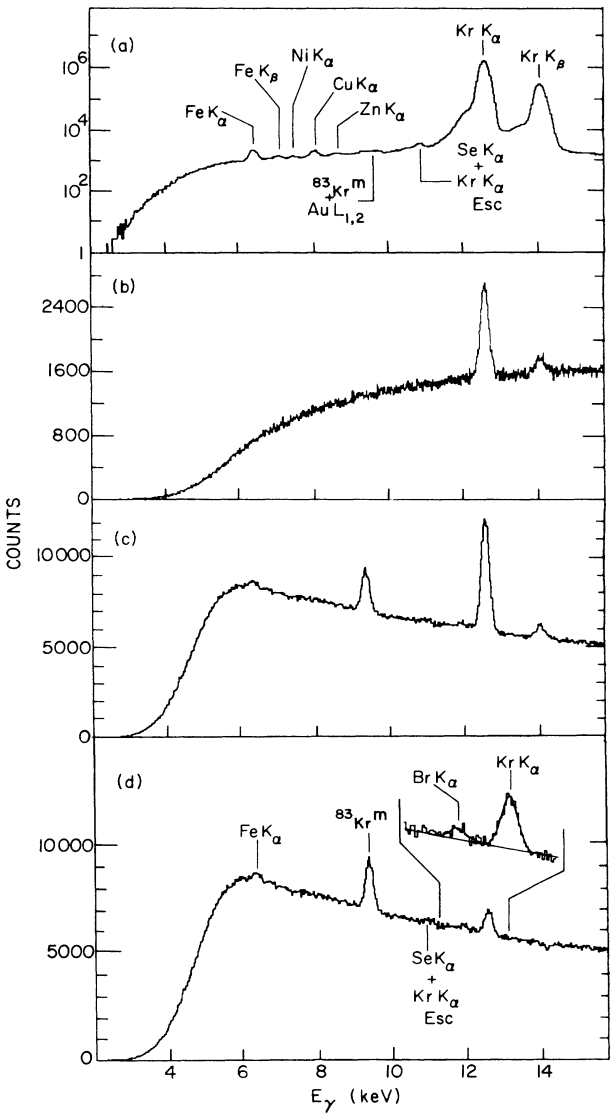

FIG. 4. Energy spectra in the Si(Li) detector corresponding to various cuts in the TAC parameter. (a) Events in coincidence with the plastic scintillator, plotted on a logarithmic scale. (b) Events in coincidence with the NaI(Tl) detectors, primarily due to Compton scattering of 190.6-keV γ rays in the Si(Li) detector. The Kr K x-rays are from random coincidences. (c) Events in the Si(Li) detector unaccompanied by signals in either the plastic scintillator or NaI(Tl) detectors. (d) Same as (c) except that 1/450th of the plastic scintillator coincidence spectrum (a) has been subtracted. This procedure decreased the amplitudes of the Kr K x rays. The inset shows the region near 12 keV; the solid line is a portion of the fit to the data. In the analysis the data between 9 and 13 keV were fitted using Gaussian peak shapes and a quadratic background function.

TABLE I. Corrections applied to Eq. (1) to obtain the total EC branching ratio.

Effect corrected	Size of correction
Br-Kr differential x-ray absorption (plastic, aluminum, Be window)	+5.8%
Loss of Br x rays due to random coincidences (integrated over counting period)	+3.5%
Using $K\alpha$ instead of all K x rays	−0.6%
L, M, and higher shell capture (Ref. 10)	+11.9%

differential coincidence efficiency mentioned in the preceding section. The Br x rays are unaffected by these considerations, since they are not in coincidence with any other radiation. The time dependence of the Br K x-ray yield is also consistent with the 13 s half-life of $^{81}Kr^m$.

The coincidence efficiency of the apparatus for the Kr K x rays and their associated electrons was determined by measuring the total number of Kr K x rays detected and the number of Kr K x rays appearing in the anticoincidence spectrum [Fig. 4(c)]. This yielded a coincidence efficiency of 99.8%, using the $K\alpha$ component.

The branching ratio (BR) for K capture is given by

$$\text{BR}\ (K\ \text{capture}) = \frac{\alpha_K(\omega_K)_{\text{Kr}}(\text{Br}\ K\ \text{x-ray yield})}{(1+\alpha_T)(\omega_K)_{\text{Br}}(\text{Kr}\ K\ \text{x-ray yield})} = 0.281\frac{(\text{Br}\ K\ \text{x-ray yield})}{(\text{Kr}\ K\ \text{x-ray yield})}\ , \quad (1)$$

where the α_K and α_T are, respectively, the K and total conversion coefficients of the 190.6-keV $E3$ isomeric transition, and the ω_K's are the respective K-fluorescence yields. The following values were used: $\alpha_K = 0.40$, $\alpha_T = 0.48$ (Ref. 9), $(\omega_K)_{\text{Kr}} = 0.646 \pm 3\%$, and $(\omega_K)_{\text{Br}} = 0.622 \pm 3\%$ (Ref. 10).

It should be noted that only the $K\alpha$ x rays from Br are observable in the spectra of Figs. 4(c) or (d). The predicted $K\beta$ intensity is less than 15% of that for the $K\alpha$ peak, and is thus too weak to be seen in the present experiment. Therefore the $K\alpha$ components were used to determine the K-capture branching ratio, with the appropriate intensity correction being made (see Table I). In the experiment, a total of $(2.2451 \pm 0.0005) \times 10^7$ Kr $K\alpha$ x rays were accumulated (a linear background was assumed; the uncertainties quoted here are purely statistical), along with 2060 ± 300 Br $K\alpha$ x rays.

To obtain the total EC branching ratio, a number of effects must be taken into account. One is the differential attenuation of the 11.91-keV Br and 12.63-keV Kr $K\alpha$ x rays as they pass through the plastic scintillator, the aluminum light shield, and the 50 μm Be window of the Si(Li) detector. A further correction is needed for the loss of Br x rays due to random coincidences with events in the plastic scintillator. In addition, it should be noted that electron capture can take place from the L and higher shells. Since these events cannot be observed in the present experiment, a correction must be applied. Table I gives a summary of the corrections used in obtaining the branching ratio.

The resulting value for the total EC branching ratio of $^{81}Kr^m$ is $(3.14 \pm 0.58) \times 10^{-5}$. Using a half-life[11] for $^{81}Kr^m$ of 13.32 ± 0.15 s, and a Q_{EC} for the decay to the ^{81}Br ground state of 471.2 ± 6.0 keV,[12] we obtain a logft for the decay $^{81}Kr^m(J\pi = \frac{1}{2}^-) \rightarrow {}^{81}\text{Br}(J^\pi = \frac{3}{2}^-)$ of $4.80^{+0.06}_{-0.07}$. An uncertainty of 6.0 keV has been used for the Q_{EC} value because of the small inconsistency remaining in the ^{81}Kr-^{81}Br mass difference.[12] The logft for the inverse neutrino capture transition $^{81}\text{Br}(J^\pi = \frac{3}{2}^-) \rightarrow {}^{81}\text{Kr}(J^\pi = \frac{1}{2}^-)$ is thus $5.10^{+0.06}_{-0.07}$. Haxton[13] has calculated a value of 4.99 for this transition, in good agreement with the present result.

IV. DISCUSSION

We have determined the logft for neutrino capture from the ^{81}Br ground state to the 190.6-keV $J^\pi = \frac{1}{2}^-$ state in ^{81}Kr to be $5.10^{+0.06}_{-0.07}$. In order to compare this value to that obtained in Ref. 4 (log$ft = 4.88 \pm 0.11$), two facts should be noted. The first is that the decay energy Q_{EC} used in Ref. 4 was 512 keV, instead of the more recently determined[12] 471.2 keV. If the newer decay energy is used to recalculate the logft measured in the experiment of Ref. 4, a value of 4.81 ± 0.11 is obtained. Secondly, it is not clear from Ref. 4 whether a correction for L-electron capture was made in going from the K-capture branching ratio to the logft for EC decay. This is an 11.9% effect,[10] and corresponds to a further reduction in the logft value of 0.05.

The effect of the logft value obtained in the present experiment on an ^{81}Br solar neutrino detector is to reduce the previously calculated neutrino capture rate to the 190.6-keV $J^\pi = \frac{1}{2}^-$ state in ^{81}Kr by a factor of 1.66. This means that the rates calculated[14] for other ^{81}Kr states based on the results of charge-exchange reactions and the previous 190.6-keV rate must also be reduced accordingly. Assuming a standard solar model neutrino flux and that capture to the 190.6-keV state is dominant, a ^{81}Br solar neutrino detector will therefore have a capture rate of about 10 solar neutrino units (SNU's).[15] Here one SNU is one capture per 10^{36} target atoms per second. This translates to ~ 3 ^{81}Kr atoms produced/day in a detector containing 10^3 tons of $CHBr_3$, about six times the capture rate observed in the ^{37}Cl experiment.

Hurst and collaborators[16] have proposed a ^{81}Br experiment which differs from that originally proposed by Scott.[3] They suggest using a tank of Br-rich fluid situated

underground, as in the ^{37}Cl experiment. The resulting ^{81}Kr atoms produced would be periodically swept out of the tank by helium gas. Because of the long half-life ($t_{1/2}=2.1\times10^5$ yr) of the ^{81}Kr ground state, direct decay counting is difficult. However, Hurst *et al.*[16] have recently demonstrated the ability to count fewer than 1000 atoms of ^{81}Kr using resonance ionization spectroscopy combined with mass separation. These developments considerably increase the attractiveness of ^{81}Br as a solar neutrino detector.

Further work must be done in order to clarify the contribution to neutrino capture of higher excited states in ^{81}Kr. This can be done by determining the Gamow-Teller matrix elements for states with potential involvement in neutrino capture, using the (p,n) reaction at medium energies. Eligible states that have been identified[17] are located at 456.7 keV ($\frac{5}{2}^-$), 636.7 keV ($\frac{3}{2}^-$), 700.8 keV ($\frac{5}{2}^-$ or $\frac{3}{2}^-$), and the isobaric analog state at 9.675 MeV. Of these, only the 456.7-keV state can participate in capturing neutrinos from the decay of ^{7}Be.

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