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Submitted to Nuclear Physics A

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August 1983

Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

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This work was supported by the Director, Office of Energy Research, Division of Nuclear Physics of the Office of High Energy and Nuclear Physics of the U.S. Department of Energy under Contract DE-AC03-76SF00098.

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Abstract

The decay of ^{251}Bk has been studied by radiochemical techniques. The half-life of (55.6 ± 1.1) minutes agrees with the literature. Relative intensities of gamma-rays, including previously unknown gamma-rays at 129.9 keV and 163.8 keV have been determined. The estimated absolute intensity of the gamma-ray at 177.7 keV is (5.2 ± 0.6) %. The maximum beta energy has been measured to be (915 ± 10) keV and was used to determine the mass of ^{251}Bk. A decay scheme based on these measurements is given.

Keywords

RADIOACTIVITY ^{251}Bk [from ^{248}Cm (^{18}O , X), E = 97 MeV], measured β^- end-point, half-life, deduced I_γ, ^{251}Cf levels.

1. Introduction

Berkelium-251 was first observed in the 8% alpha decay branch of ^{255}Es.[1] Recoiling atoms from isotopically enriched samples of ^{255}Es were collected and counted for gamma-rays and beta particles. The half-life of ^{251}Bk was measured to be (57.0 ± 1.7) minutes, and the beta endpoint energies of $\approx \frac{1}{2}$ and ≈ 1 MeV were estimated. Later experiments[2] established two gamma-rays at 152.8 keV and 177.7 keV with an intensity ratio of 0.38 to 1. From the decay scheme of its ^{251}Cf daughter it is evident that these gamma-rays arise both from the $\frac{3}{2}+$ state at 177.7 keV. ^{251}Bk has also been produced by beta decay of ^{251}Cm.[3] The beta endpoint energy of ^{251}Bk was determined to be ≈ 1 MeV.

In recent work by Lee et.al.[4,5] the yields of berkelium isotopes from transfer reactions of ^{18}O with ^{248}Cm were measured using radiochemical techniques. The relatively high cross section (≥ 24μb) for the production of ^{251}Bk in this reaction made it possible to investigate further the decay of this nuclide. In the present work we have measured the beta endpoint energy and relative intensities of photons and electrons emitted in its decay. Furthermore, we have estimated absolute gamma-ray intensities and its ground state mass defect.

2. Experimental

A uniform target of 560 $\mu g/cm^2$ of ^{248}Cm (97 mass %) was electroplated[6] in a 7 mm spot on a 2.5 mg/cm^2 thick beryllium backing. The target was irradiated[4] with 1 to 4 electrical microamperes of 111 MeV $^{18}O^{4+}$ at the 88-inch Cyclotron of the Lawrence Berkeley Laboratory for approximately 45 minutes. The beam energy in the curium target was roughly 97 MeV. Recoiling reaction products were collected in a 2 mg/cm^2 gold foil. The gold foil was dissolved in aqua regia, and berkelium was separated and purified using a procedure given by Liu et.al.[7] The final Bk fraction was evaporated and ignited on a platinum disk. The time between the end of the irradiation and the start of counting amounted to about 40 minutes. The chemical yield was only about 20 % but the samples were very clean and contained no activities other than the Bk isotopes. A somewhat faster procedure[8] did not produce clean enough samples.

The berkelium fraction was counted for photons and electrons. The sample was sandwiched between two pieces of 2 mg/cm^2 polyethylene foil and was placed between an intrinsic germanium gamma-ray detector and a beta detector. The activity on the plate was facing the beta detector in order to minimize degradation of the electrons. The germanium detector was calibrated with a mixed radionuclide standard, counted through each berkelium fraction after the reaction products had decayed. For the measurement of beta particles a gas-proportional counter with a thin window was used in the majority of the experiments. In a few cases it was replaced by a plastic scintillator in order to measure beta spectra. The efficiency and energy calibrations of these detectors were performed with a set of standard beta reference sources. The counting time during the decay of the berkelium fraction was partitioned into several measurements, each of them being much shorter than the half-life of ^{251}Bk. The data were stored on magnetic tape for further processing.

The areas of the gamma-ray peaks were computed and corrected for detector efficiencies using standard computer programs. Half-life and initial activity were determined with an iterative non-linear least-squares program.

A multiple component decay analysis was performed on the californium K X-rays and on the gross beta activity, including conversion electrons. The gross beta activity was determined with the proportional counter using a detector efficiency corresponding to the energy of the beta endpoint which was assessed as follows: Beta spectra were taken with the plastic scintillator at regular time intervals and were divided into energy windows (E_0); the areas of these windows were analysed as a function of time (figure 1). Multiple component decay curve analysis was performed on each window using known half-lives of the berkelium isotopes in order to obtain the beta spectra of the individual nuclides.

3. Results and discussion

3.1. Half-life of ^{251}Bk

Decay curves of the gamma-ray peaks at 152.8 keV and 177.7 keV are shown in figure 2. The errors include statistical uncertainties only. The weighted average of seven separate determinations of the half-life is (55.6 ± 1.1) minutes. This result is in agreement with the value of (57.0 ± 1.7) minutes.[1]

3.2. Relative intensities of photons and electrons

Two previously unknown gamma-rays in the decay of ^{251}Bk at 129.9 keV and 163.8 keV were deduced from the gamma-ray spectra and the decay analysis. The intensity ratios of the gamma-rays were obtained from a re-fit of the decay curves with a half-life for ^{251}Bk of 55.6 minutes. Relative K X-ray intensities were obtained from a three component (^{248}Bk, ^{250}Bk, and ^{251}Bk) decay analysis of the californium K X-rays. Since the $K_{\beta 1}$ X-ray and the 129.9 keV gamma-ray could not be resolved, the individual intensities were deduced from literature values for ^{251}Cf K X-rays.[9] (See Section 3.4). The gross beta activity was measured with the proportional counter, ignoring the presence of conversion electrons. Weighted averages of the results are presented in Table 1. The $\gamma_{153}/\gamma_{178}$ ratio of 0.385 ± 0.025 is consistent with the decay scheme of ^{251}Cf.[10]

3.3. Beta endpoint energy of ^{251}Bk

Beta spectra of ^{250}Bk and ^{251}Bk obtained by the procedure given in the experimental section are shown in figure 3. The background activity above the apparent endpoint energies of the spectra is an artifact of decay curve analysis with fixed half-lives, with contributions from Bremsstrahlung and gamma-rays.

The error bars are based on the fit of the decay data of each energy window (figure 1), and the data points are plotted at the energy central to the windows. The endpoint energies shown in figure 3 are the results of least-squares fits at the endpoint regions of Fermi-Kurie functions[11] after subtracting the background from higher energy regions.

A beta endpoint energy for ^{251}Bk of (915 ± 10) keV is found. A very weak component with an endpoint of (1130 ±35) keV is also observed, as seen in the Fermi-Kurie plot in figure 3. The Q_{β^-}-value predicted from systematics[12] is 1120 keV. Most of the ^{251}Bk decays populate the $\frac{3}{2}$[622] rotational band, with only ≈5 % of the decays going directly to the ground state.

In order to check the accuracy of our procedure we determined the energy of the two known beta endpoints of ^{250}Bk. The upper endpoint in our measurements is (1820 ± 35) keV, in agreement with the literature ((1760 ± 50) keV)[13], and the Q_{β^-}-value of 1780 keV.[12] Our value for the second β-decay branch of (705 ± 20) keV also compares well with the values of (725 ±15) keV[13] and 745 keV[12].

3.4. Estimation of the absolute γ-ray intensities of ^{251}Bk

The most important gamma-ray transitions in the decay scheme of ^{251}Cf have values of $\Delta J = 0,1$ with no parity change. They are therefore of mixed M1 and E2 multipolarity. From single-particle Weisskopf[14] estimates, the rate of E2 transitions in ^{251}Cf is given as

$$R_{E2}(^{251}Cf) = 1.5 \times 10^{11} E_\gamma^5 \ \ sec^{-1} ,$$

and from Moszkowski's estimates[15] for M1 transitions,

$$R_{M1}(^{251}Cf) = 3.1 \times 10^{12} E_\gamma^3 \ \ sec^{-1} ,$$

giving

$$\frac{R_{E2}}{R_{M1}} = 4.8 \times 10^{-2} E_\gamma^2 \quad .$$

The single particle estimate of the admixture of E2 radiation in low energy gamma transitions in ^{251}Cf is negligible. However, most E2 rates are faster than those predicted from the single particle model due to collective rotational and vibrational enhancement. Examination of the ^{251}Cf level scheme gives some indication that the E2 rates are small in spite of this enhancement.

The Bohr-Mottelson gamma transition rates for gamma decays to the different members of a rotational band[16] are related by

$$R(L) \;\; \alpha \;\; E_\gamma^{2L+1} \, [< J_f \; K_f \mid J_i \; L \; K_i \; \Delta K >]^2$$

where J_f and K_f describe the total angular momentum of the final state and that of its band head, J_i and K_i describe the initial state and $\Delta K = K_f - K_i$. Ignoring conversion of electrons for the moment, the E2 multipolarity intensity for the transition between the $\frac{5}{2}$+ level at 211.6 keV and the $\frac{1}{2}$+ ground state should be 26 times that arising from the transition between the same $\frac{5}{2}$+ level and the $\frac{3}{2}$+ rotational level at 24.8 keV. Though the latter photon is observed, the former is not, inconsistent with E2 multipolarity. In M1 transitions, the $\Delta J = 2$, $\frac{5}{2}$+ to $\frac{1}{2}$+ transition is forbidden. Similarly, the $\frac{3}{2}$+ level at 177.7 keV decays to both the $\frac{1}{2}$+ ground state and the $\frac{3}{2}$+ state at 24.8 keV. If the photons arise from transitions of E2 multipolarity, the intensity of the 177.7 keV gamma ray should be roughly half as intense as the 152.8 keV gamma ray, rather than 3 times as intense, which is what is observed. The gamma transitions in ^{251}Cf following ^{251}Bk decay will be treated as being of pure M1 multipolarity in the calculations which follow.

Conversion coefficients as a function of transition energy and multipolarity and the nuclear charge are tabulated[17,18] and interpolation gives the data in Table 2. The relatively small size of the total conversion coefficient for the 129.9 keV gamma transition indicates that the gamma ray intensity from this transition might be appreciable. Unfortunately, the K_{β_1} X-ray line (which contains the small K_{β_3} group) is at 130.8 keV[10], so no individual resolution was possible. However, the relative intensities of the K X-rays can be predicted to a 1% accuracy[9] for ^{251}Cf:

$$K_{\alpha_2} / K_{\alpha_1} / K_{\beta_1} + K_{\beta_3} / K_{\beta_2} = 64.2 / 100 / 34.3 / 13.0 .$$

The experimental ratios (Table 1) are

$$(63.3 \pm 4.4) / 100 / (52.7 \pm 3.6) / (11.3 \pm 1.5) .$$

Only the K_{β_1} (+ K_{β_3}) value deviates from the literature value. Subtraction of the predicted and measured values and using the data given in Table 1 yields a relative intensity of (0.58 ± 0.12) for the 129.9 keV gamma ray. Correcting observed gamma ray intensities for internal conversion leads to relative depopulation rates of the 177.7 keV level of

$$R(177.7\ keV) / R(152.8\ keV) / R(129.9\ keV) = 1 / (0.57 \pm 0.04) / (0.35 \pm 0.07) .$$

The rates expected for M1 transitions from the Bohr-Mottelson model are in the ratios

$$R(177.7\ keV) / R(152.8\ keV) / R(129.9\ keV) = 1 / 0.51 / 0.078 .$$

The experimental and calculated depopulation rates for the 177.7 keV and 152.8 keV gamma rays match very well, but the 129.9 keV gamma ray is about 4.5 times more intense than the calculated value. This is due to the properties of the $\frac{5}{2}+$ receiving state at 47.8 keV.

The 163.8 keV gamma ray also terminates at the 47.8 keV state and is anomalously intense. The predicted ratio of intensities of the 163.8 keV gamma ray to the 186.8 keV gamma ray should be about 0.55. What is actually observed[19] is a ratio of about 19. Another anomaly involving the $\frac{5}{2}+$ state at 47.8 keV is in the alpha hindrance factors from ^{255}Fm decay[19]. Decay to each member of the $K = \frac{1}{2}+$ rotational band from the $\frac{7}{2}+$ ^{255}Fm ground state must proceed by $l = 4$ alpha emission by the K selection rules and parity conservation[20,21]. It should be expected that the hindrance factor to each member of the rotational band should be roughly the same large number. The hindrance factors for alpha decay to the $\frac{1}{2}+$, $\frac{3}{2}+$ and $\frac{5}{2}+$ levels in the ground state rotational band are 4800, 2900 and 540, respectively[10]. Once again, decay to the $\frac{5}{2}+$ state is too fast. Coriolis coupling of some state with $K > \frac{1}{2}$ to this state is effectively increasing its K, increasing the Clebsch-Gordan factor for gamma decays resulting in this state and decreasing the l-hindrance to alpha decay.

The intensity of the 163.8 keV gamma ray to that of the 177.7 keV gamma ray is 0.060 ± 0.012. From data on the gamma rays accompanying ^{255}Fm alpha decay, the relative intensity of the 186.8 keV gamma live is roughly 0.003. Correction of both gamma rays for internal conversion gives a transition rate for the depopulation of the 211.6 keV level of 0.68 ± 0.14 relative to that depleting the 177.7 keV level of 17.2 ± 1.0 (from Table 1 data). Remembering that approximately 5% of the ^{251}Bk decays to the ground state band of ^{251}Cf, the population of the 211.6 keV level by ^{251}Bk decay is approximately 4% and that of the 177.7 keV level 91%. The ratio of the population of the 211.6 keV level to that of the 177.7 keV level is slightly higher than that arising from ^{251}Es decay[10], which is expected since the decay energy is higher. An assignment of a value of (90 ± 5)% for the fraction of ^{251}Bk decays populating the 177.7 keV level in ^{251}Cf seems to be justified.

The absolute intensity of the 177.7 keV gamma ray can be determined by taking into account the population of the initial state (0.90 ± 0.05), the fraction of gamma transitions going to the final state (0.521 ± 0.027, from Table 1 data), and the total conversion coefficient (7.95, from Table 2). This yields a value of

$$I_{\gamma,178} \;=\; (5.2 \pm 0.6)\ \% \quad .$$

Based on our results and the preceding discussion we propose the decay scheme shown in figure 4 for ^{251}Bk.

3.5. Mass excess of ^{251}Bk

The beta endpoint energy of (915 ± 10) keV, the 177.7 keV gamma-ray and the known mass excess of ^{251}Cf[10] can be summed to give the mass excess of ^{251}Bk. Its value of (75.220 ± 0.013) MeV disagrees only by roughly 30 keV from the literature value of 75.25 MeV which was deduced from systematics[12]. This is an independent indication that the beta decay with E_{max} = 915 keV of ^{251}Bk feeds the 177.7 keV level in ^{251}Cf.

Acknowledgements

We would like to thank the staff of the Lawrence Berkeley Laboratory's 88-Inch Cyclotron for their support, and J. Rasmussen, M. Nurmia and A. Ghiorso for assistance and suggestions. E. Calhoon and H. Harrington helped with the target preparation and handling, K. Gregorich in the detector calibrations, and R. Welch and Luo Cheng in part of the chemistry. The interest and support of D. C. Hoffman is acknowledged. H. R. von Gunten acknowledges support by the Swiss National Science Foundation.

This work was supported, in part, by the Director, Office of Energy

Research, Division of Nuclear Physics of the Office of High Energy and Nuclear Physics of the U.S. Department of Energy under Contract DE-AC03-76SF00098.

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Table 1. Relative intensities of the emitted radiations from ^{251}Bk decay

Cf K_{α_2} X-ray	2.02 ± 0.11
Cf K_{α_1} X-ray	3.19 ± 0.14
Cf K_{β_1} X-ray*	1.68 ± 0.09
Cf K_{β_2} X-ray	0.36 ± 0.05
153 keV γ-ray	0.385 ± 0.025
164 keV γ-ray	0.060 ± 0.012
178 keV γ-ray	1
gross β^- activity	≈ 33

* Including contribution from 129.9 keV γ-ray

Table 2. Internal conversion coefficients for M1 gamma transitions from ^{251}Bk decay. Interpolation of data from references 17 and 18.

Electron Origin	Transition Energy: Transitions depopulating the 3/2+ state at 177.7 keV			Transition Energy: Transitions depopulating the 5/2+ state at 211.6 keV	
	177.7 keV	152.8 keV	129.9 keV	186.6 keV	163.8 keV
K shell	6.20	9.45	-	5.40	7.80
L shell	1.33	2.05	3.30	1.16	1.68
M shell	0.32	0.50	0.80	0.29	0.41
N shell	0.10	0.15	0.25	0.08	0.12
Total	7.95	12.15	4.35	6.93	10.01

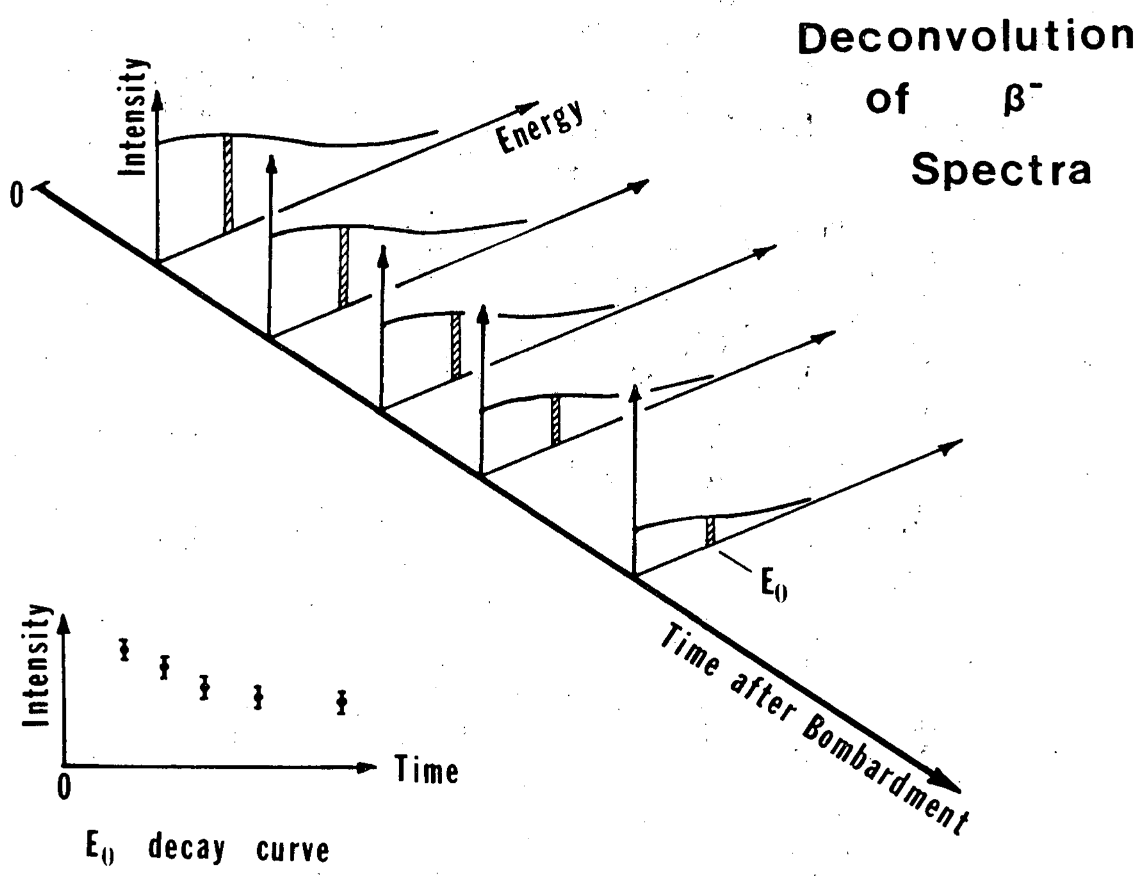
Figure Captions

1. Deconvolution of the beta energy spectra of the berkelium isotopes produced in the $^{18}O + ^{248}Cm$ bombardments. The intensities and spectra of ^{251}Bk are obtained by analysis of the complex decay curves of each E_0 energy window.

2. Decay curves of the 177.7 keV, 152.8 keV and 163.8 keV gamma rays of ^{251}Bk. The 163.8 keV data are fitted with a half-life of 55.6 minutes.

3. Beta particle spectra obtained from the deconvolution of the composite berkelium beta spectra (see fig. 1). The indicated beta endpoint energies result from Fermi-Kurie plots as shown for ^{251}Bk.

4. The decay scheme of ^{251}Bk. Gamma transitions are labelled with absolute photon intensities.



XBL 836-10254

Figure 1

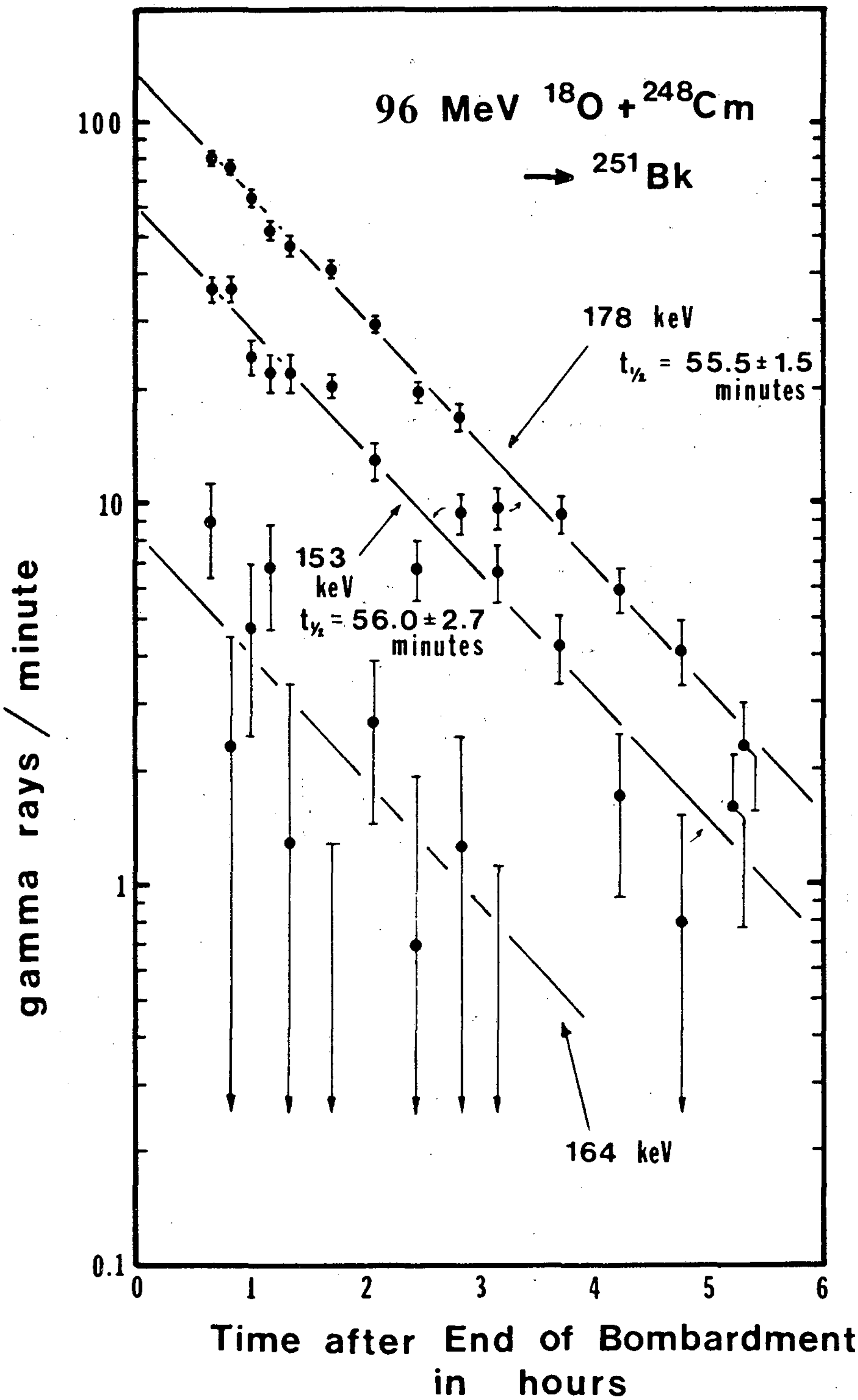


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Figure 2

Deconvolution of Bk beta spectra

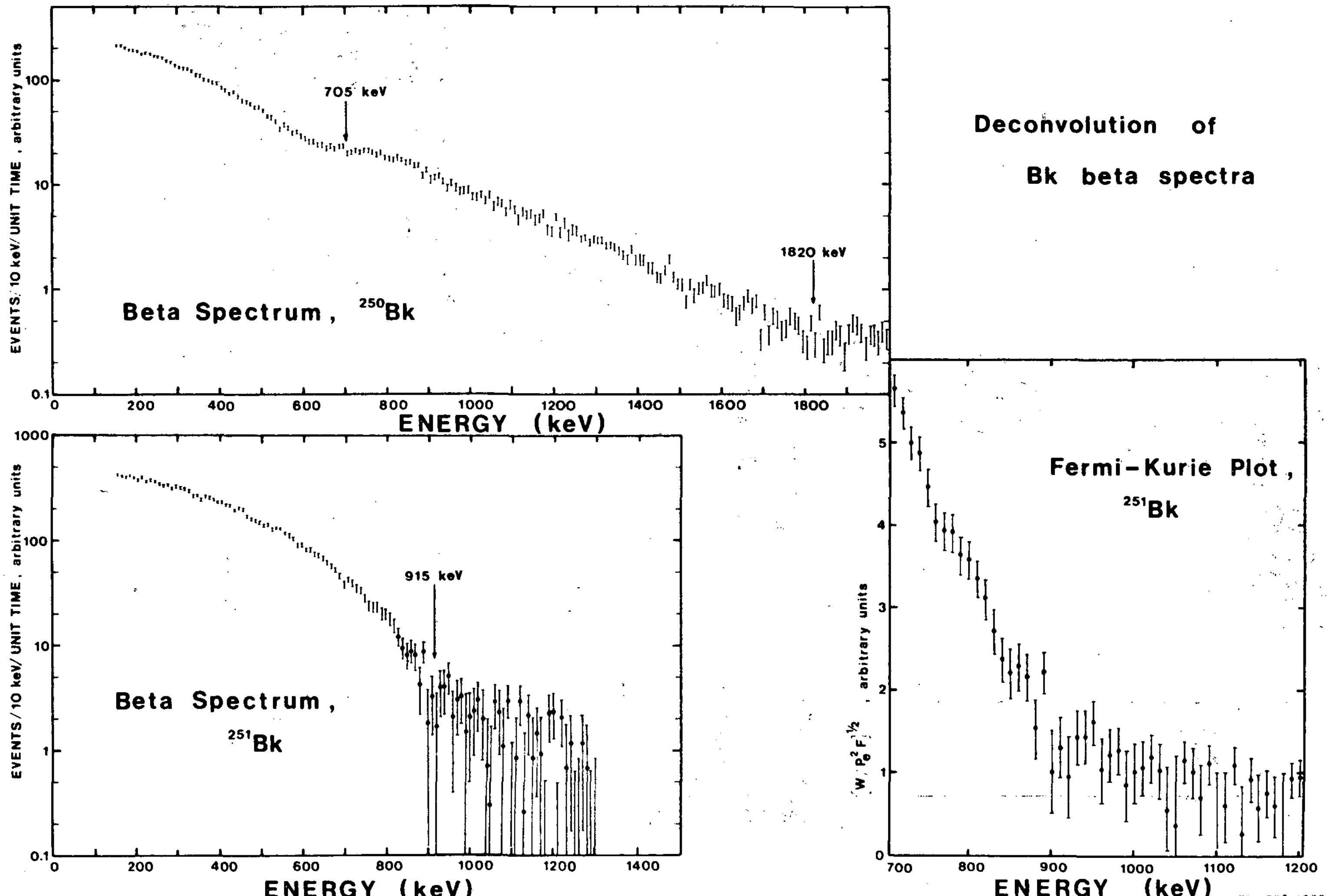

Figure 3

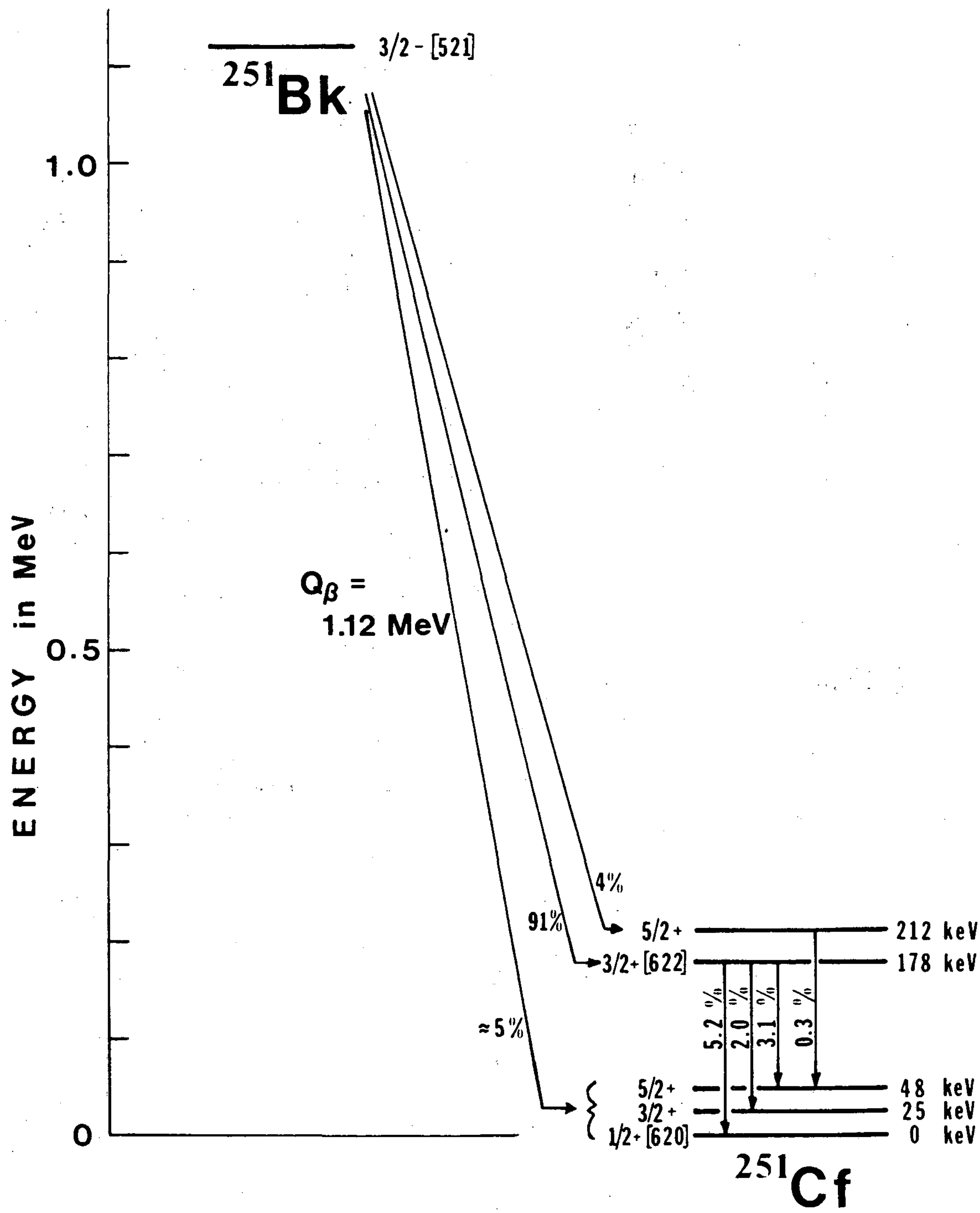

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Figure 4

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