Title

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Publication Date

1987-12-01

LBL-24562
Preprint

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

Submitted to Physical Review Letters

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December 1987

NUCLEAR SCIENCE DIVISION

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

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Tests of the Exponential Decay Law at Short and Long Times

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Abstract

We have studied the beta decays of ^{60}Co at times $\lesssim 10^{-4}\ t_{1/2}$ and those of ^{56}Mn over the interval $0.3t_{1/2} \lesssim t \lesssim 45t_{1/2}$ to search for proposed deviations from the exponential decay law at short and long times, respectively. Over all time periods examined, our data is consistent with purely exponential behavior. From these measurements, stringent limits are derived on the amplitudes of possible deviations from the exponential decay law.

PACS # 23.90.+w, 03.65.Bz, 27.40.+z, 27.50.+e

Over the years, numerous authors have pointed out that the exponential nature of the radioactive decay law is only an approximation.[1-16] Deviations from purely exponential behavior are, in fact, expected at very short and very long times compared with the lifetime of the decaying system. The work of Khalfin[5] and others on the time evolution of unstable quantum states showed that the decay rate should approach zero for $t \rightarrow 0$. Therefore, there must exist a regime between $t=0$ and the known exponential domain when the decay rate is non-exponential. The existence of such a region has been suggested as a possible explanation for the null results of proton-decay experiments.[17-22] Khalfin[1] and others have also shown that the decay of a quasi-stationary state will exhibit deviations from exponential decay at sufficiently long times if the decay spectrum is cut off at a finite energy level (due to, for example, the non-zero rest masses of the decay products).

In spite of the considerable theoretical effort on this subject, we are not aware of any experimental searches at short times, and there have been relatively few searches at long times for deviations from the exponential decay law. Rutherford[23] studied the alpha decay of ^{222}Rn and found no deviations from exponential behavior out to the limit of his sensitivity at 27 half lives. Winter[24] and, more recently, Gopych et al.[25] have extended such searches out to 34 half lives for the beta decay of ^{56}Mn and to 33 half lives for the beta decay of $^{116}\mathrm{In}^{m}$, respectively. The decay curves of the μ^+ , π^+ , K^+ , K^0_S , K^0_L , Λ^0 , Σ^- , and Ξ^- have been studied out to times which range from 2.3 to 21.6 half lives. The results of these searches have been summarized by Nikolaev.[26]

Some of the reasons for this dearth of experimental data are the indications that the non-exponential effects discussed by Khalfin occur only in experimentally inaccessible regions of time. However, as pointed out by Goldberger and Watson[4] in their study of the decay of unstable particles in S-matrix theory, "the exponential decay law is only one of a discrete set of possible decay laws." Thus, non-exponential effects of this type could occur at any time. As a result of this analysis, these authors went on to suggest that "the time-honored study of decay curves (rather than the simple determination of mean lifetimes) might be worthwhile."

In this Letter, we present the results of our studies of the beta decays of ^{60}Co and ^{56}Mn in which searches were made for deviations from purely exponential behavior at short and long times, respectively. ^{60}Co and ^{56}Mn are good candidates for such studies for several reasons. Both cobalt and manganese are monoisotopic, have large thermal-neutron-capture cross sections, and are available in very high chemical purities. The half-lives of these two isotopes, 5.271 years and 2.5785 hours, respectively, and the energies of the gamma-rays emitted following their beta decays, also lend them to convenient study. The relevant portions of the decay schemes of ^{60}Co and ^{56}Mn are illustrated in Fig. 1. All of the energies, spins, parities, and half-lives are taken from Ref. 27.

It has been suggested[3,7] that at sufficiently early times, decay curves may exhibit oscillatory behavior. In order to search for such effects at short times compared to the lifetime of the decaying system, it is necessary to accurately know when each radioactive nucleus is produced. One way to achieve this is to produce all of the required activity in a very short time interval. For our studies, we produced ^{60}Co using the pulsed-mode operation of the U. C. Berkeley TRIGA Mark III reactor. A 1-gram sample of 99.997% pure cobalt was irradiated with approximately 4×10^{14} neutrons/cm^2 in a burst with a full width at half maximum of 12.5 milliseconds. Immediately after this activation, the sample contained approximately 4 Curies of the $t_{1/2}$ = 10.5 minute ^{60}Co^m . After allowing this activity to decay for 3.5 hours, the sample contained 15 micro-Curies of ^{60}Co. The ^{60}Co was mounted directly on the front face of a 110 cm^3 high-purity germanium detector. Counting was begun within $8\text{x}10^{-5} t_{1/2}(^{60}\text{Co})$ after the activation. Gamma-ray energy signals and arrival times were recorded event-by-event on magnetic tape at a rate of about 30 kHz. Timing data were taken in 512 channel time bins of 10-ms, 100-ms, 1-s, and 10-s duration. Due to the finite time width of the initial neutron activation, 10 ms is the shortest time interval over which oscillatory effects, if present, would be preserved.

Representative data acquired for each of these time bins are shown in Fig. 2. The horizontal line drawn through each set of data is the result of a least-squares fit assuming purely exponential decay with the known ^{60}Co half life. One way to parameterize such decays has been given by Gopych et al.[25] as

$$N(t) = N_0 \exp(-t/\tau) \left[1 + \alpha \sin(\beta t/\tau)\right] \qquad (1)$$

where τ is the lifetime of the decaying system, α is the amplitude, and β is the frequency of the non-exponential contribution to the decay curve. In order to search for such oscillations in our time spectra, we performed a discrete Fourier transform upon the data using the fast Fourier transform (FFT) algorithm. Such an analysis allows us to simultaneously scan over a large range of frequencies without making any assumptions about the phase. For each time spectrum, the transformed spectrum covers the frequency range $1/(2\Delta t) < f < (1/T)$, where Δt is the size of the individual time bins and T is the total duration of the time spectrum. In all four cases, none of the transformed spectra showed any structures other than the fluctuations associated with "noisy" time series. In order to extract upper bounds on the presence of oscillations in our data, the magnitude of the frequency components must be calibrated. This was done by taking the time spectra, adding a sine wave of known amplitude, and then performing the FFT. Half of the ^{60}Co activity that we observed was produced initially as the $t_{1/2}$ = 10.5 minute ^{60}Co^{m} . Thus, the amplitude limits derived from these fits were multiplied by a factor of two to obtain the limits on the amplitudes for oscillatory behavior as a function of the assumed frequency shown in Table I.

In order to search for deviations from exponential behavior at long times, one needs to produce as much initial activity as possible. For our studies, ^{56}Mn was produced by exposing 99.995% pure manganese samples to neutrons from the above-mentioned reactor. In order to examine the possible dependence of the half life on the age of the source, four different samples of ^{56}Mn were produced. Each sample was allowed to cool until it contained a few micro-Curies of ^{56}Mn. At this point it was mounted in close geometry to a 110 cm^{3} high-purity germanium detector shielded with 10 cm of lead. Gamma-ray energy spectra were recorded in 4000-s long time bins. System dead-time was monitored with a pulser. The various sample sizes, initial activities, and counting intervals are shown in Table II.

In order to extend the present measurements further out in time than had been achieved in the previous ^{56}Mn experiment of this type[24] , it was necessary to chemically purify the fourth manganese sample after activation to remove ^{24}Na, ^{59}Fe, and ^{60}Co activities produced by neutron captures on parts-per-million impurities in the manganese sample. At the end of the irradiation, this sample contained approximately 800 Curies of ^{56}Mn. After allowing this initial activity to decay for three days, the manganese was dissolved in concentrated HCl plus concentrated HNO_3 and then passed through columns of hydrated antimony pentoxide and AG1-X8 anion-exchange resin. Lanthanum carrier was then added to the sample followed by concentrated HF. Unwanted rare-earth activities such as ^{140}La and ^{160}Tb were precipitated as fluorides, then centrifuged and discarded. The manganese solution was then boiled down to approximatley 100 ml for counting.

The data obtained from each ^{56}Mn source were first analyzed separately. A ^{56}Mn half life was determined from each data set by a least-squares fit to the observed 847- and 1811-keV gamma-ray intensities in which the half life was a free parameter. In all four cases the half-life determined from our data agreed with the known value, thus indicating no dependence on the age of the source. The results of these fits for the 1811-keV transition are shown in Table II. After this was established, the individual decay curves were normalized to one another and then combined to obtain the composite decay curves for the 847- and 1811-keV gamma rays shown in Fig. 3. The straight line drawn through each set of data is the result of a least squares fit assuming purely exponential decay with the known ^{56}Mn half life. It has been suggested[25] that decay curves may actually be described by

$$N(t) = N_0 \exp(-t/\tau)\left[1+A(t/\tau)^2\right] \qquad (2).$$

Our data was also analyzed to search for non-exponential effects of this type. No indications of such deviations were found in our data and the limits derived on their amplitudes are shown in Table II.

In conclusion, we have performed the first search for deviations from the exponential decay law at short times compared with the lifetime of the decaying system using the beta decay of ^{60}Co. We have also extended the limits on long-time searches out to approximately 43 half lives using the beta decay of ^{56}Mn.

All of our data are consistent with purely exponential behavior out to the limits of our sensitivity.

We wish to thank T. Lim and M. Denton for performing the many neutron irradiations required in the present investigations. We also wish to thank Dr. B. G. Harvey for his comments and suggestions regarding this manuscript. This work is 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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Figure Captions

1. Decay schemes of ^{60}Co and ^{56}Mn.

2. Numbers of gamma rays observed from the ^{60}Co source in time bins of (a) 10 ms , (b) 100 ms, (c) 1 s , and (d) 10 s. The straight line drawn through each data set is the result of a least-squares fit assuming purely exponential decay with the known ^{60}Co half life.

3. Composite decay curves for the 847- and 1811-keV gamma rays observed from the decay of ^{56}Mn. The straight lines are the results of least-squares fits assuming purely exponential decay with the known ^{56}Mn half life.

TABLE I. Results of the searches for oscillatory behavior in the short-time decay of ^{60}Co. The limits on the amplitude, α, as a function of the assumed frequency, β, were derived from least-squares fits of our data to Equation 1.

Time Bin Width	Frequency Range (s^{-1})	α
10 ms	$7.5 \times 10^{10} < \beta < 5.9 \times 10^{8}$	< 0.056
100 ms	$7.5 \times 10^{9} < \beta < 5.9 \times 10^{7}$	< 0.020
1 s	$7.5 \times 10^{8} < \beta < 5.9 \times 10^{6}$	< 0.0068
10 s	$7.5 \times 10^{7} < \beta < 2.4 \times 10^{6}$	< 0.0032

TABLE II. Results of the searches for non-exponential behavior in the long-time decay of ^{56}Mn. The limits on A were derived from least-squares fits of our 1811-keV data to Equation 2.

Sample	Initial Activity	Counting Interval ($t_{1/2}$(^{56}Mn))	$t_{1/2}$ (hours)	A	$A(t/\tau)^2\big\vert_{t=t_{max}}$
1	1 μCi	$0.3\ t_{1/2} \longrightarrow 13\ t_{1/2}$	2.583 ± 0.008	$< 4.0\text{x}10^{-4}$	< 0.035
2	1 mCi	$11\ t_{1/2} \longrightarrow 22\ t_{1/2}$	2.585 ± 0.005	$< 3.4\text{x}10^{-5}$	< 0.011
3	1 Ci	$20\ t_{1/2} \longrightarrow 33\ t_{1/2}$	2.576 ± 0.005	$< 1.0\text{x}10^{-5}$	< 0.0068
4	800 Ci	$34\ t_{1/2} \longrightarrow 45\ t_{1/2}$	2.573 ± 0.019	$< 1.3\text{x}10^{-5}$	< 0.013

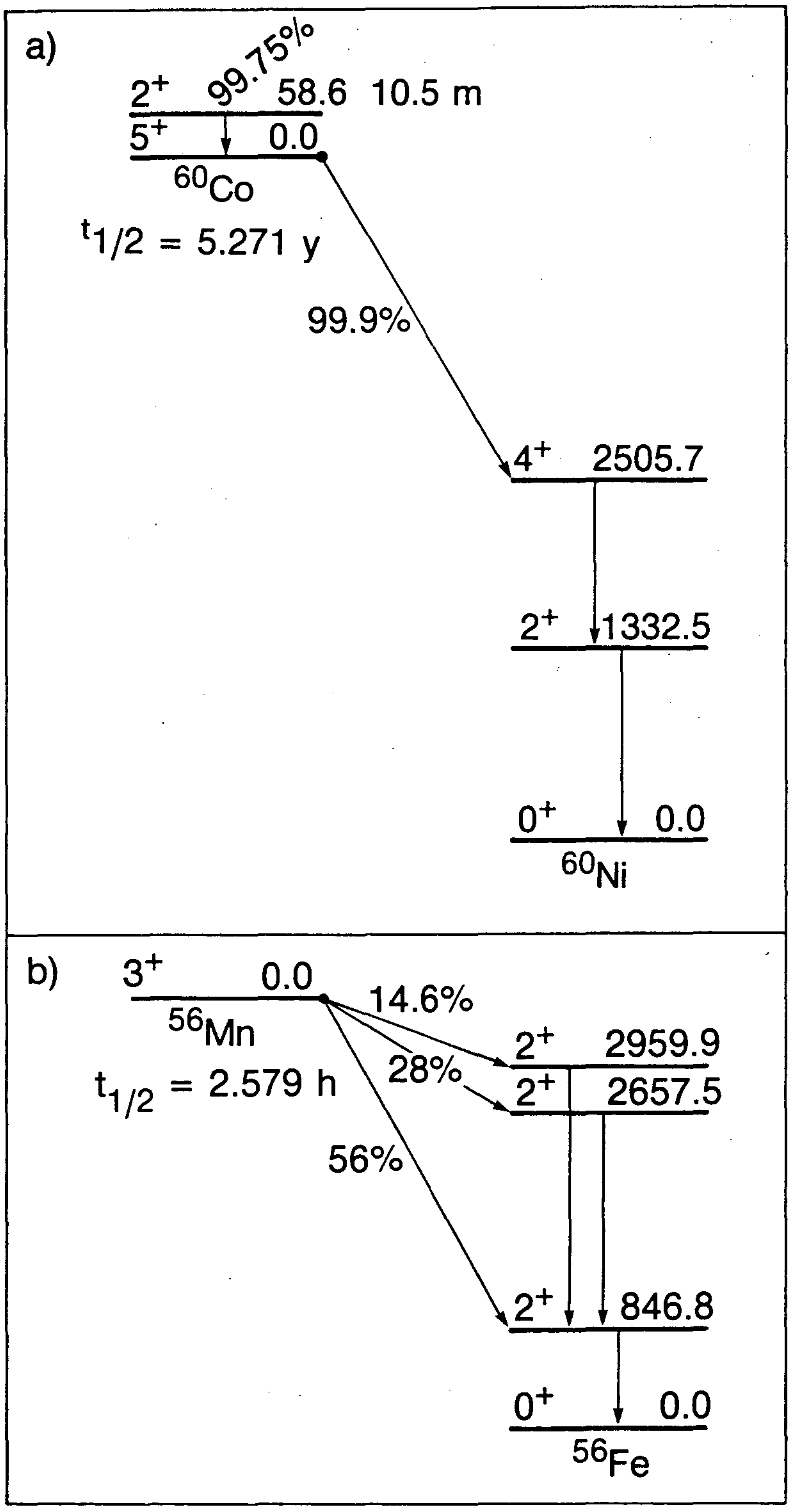

a)
2+
99.75%
58.6
10.5 m
5+
0.0
60Co
t1/2 = 5.271 y
99.9%
4+
2505.7
2+
1332.5
0+
0.0
60Ni
b)
3+
0.0
56Mn
t1/2 = 2.579 h
14.6%
28%
56%
2+
2959.9
2+
2657.5
2+
846.8
0+
0.0
56Fe
XBL 868-3135

Fig. 1

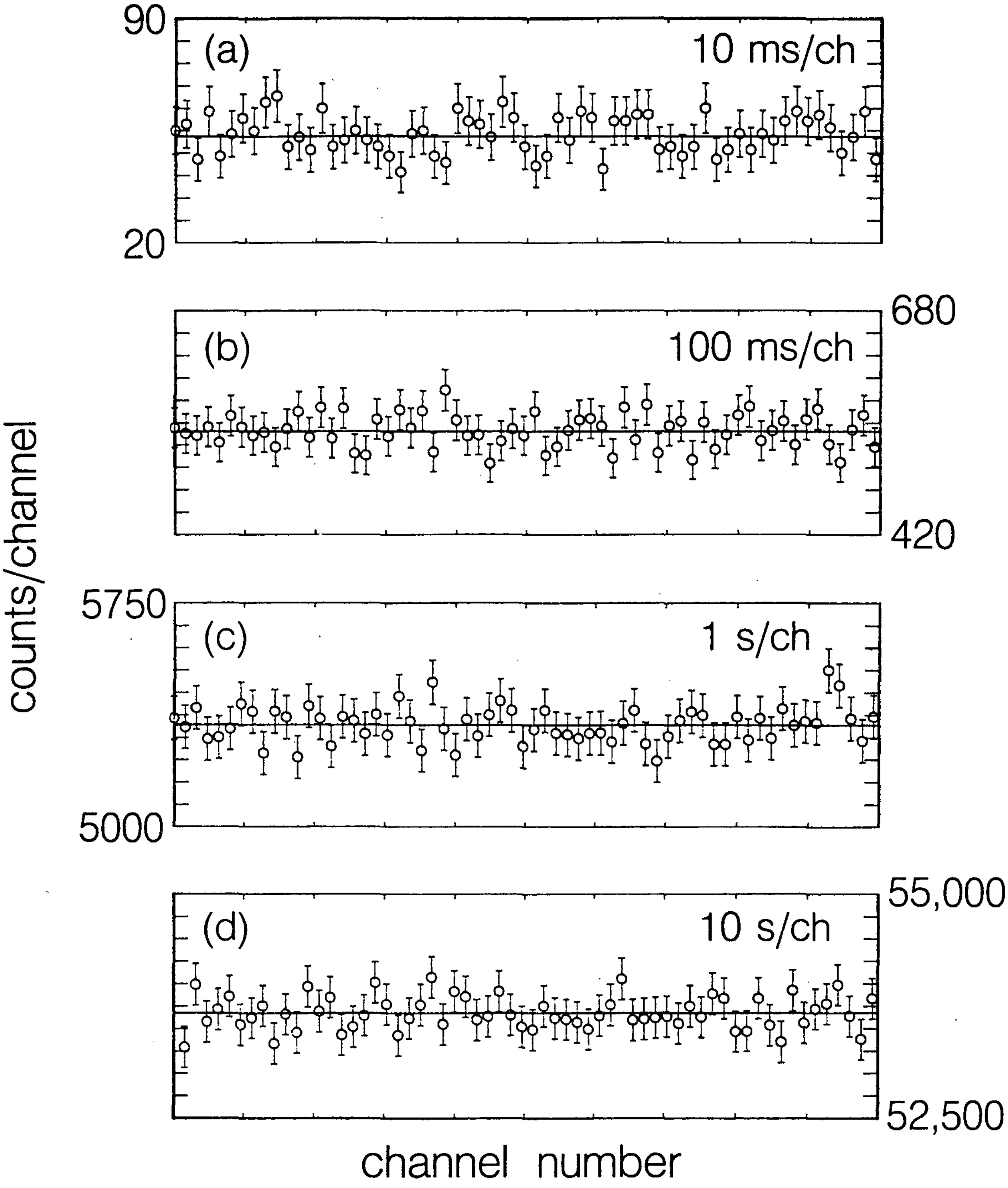
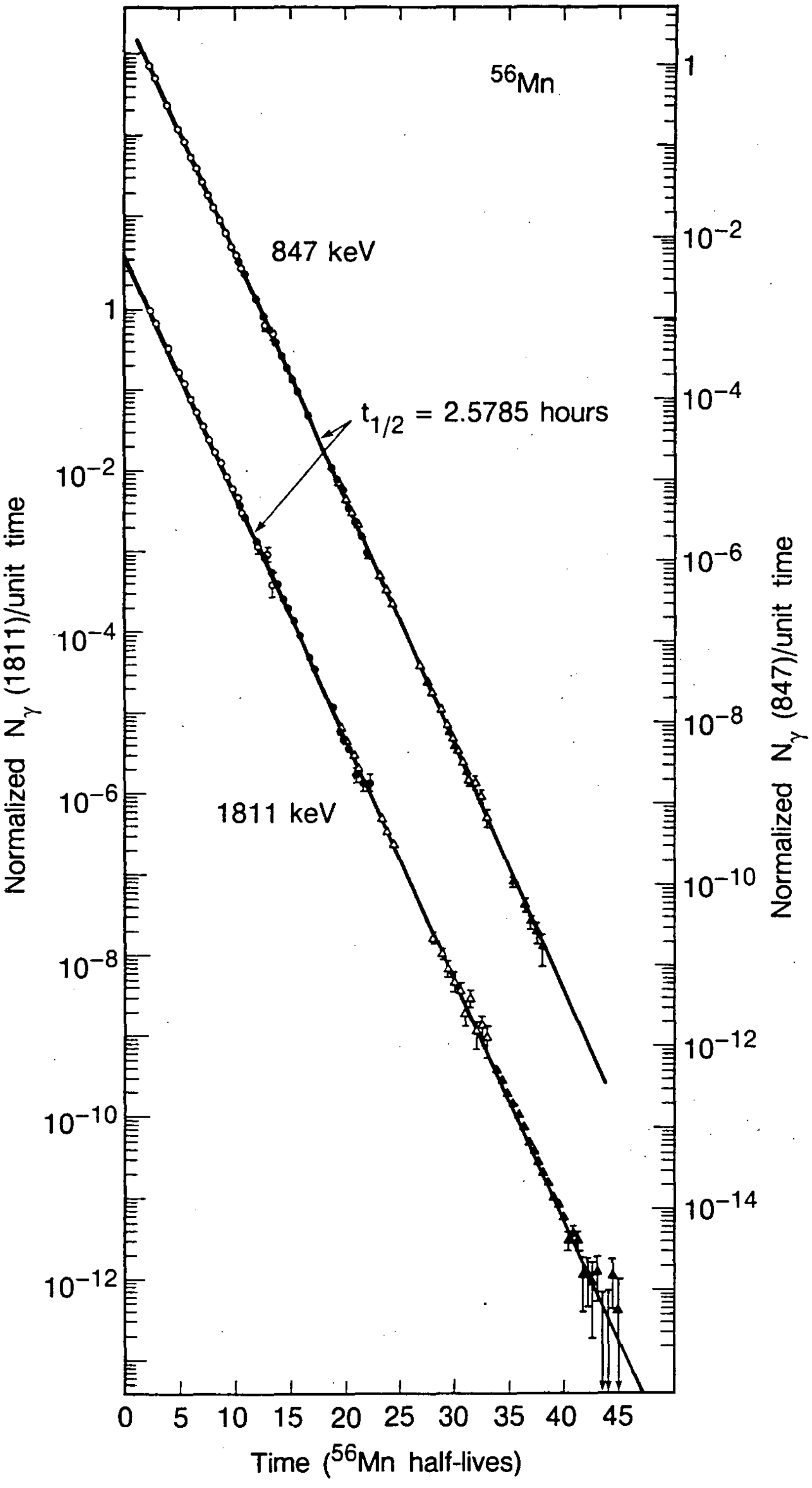

counts/channel
90
20
(a)
10 ms/ch
680
420
(b)
100 ms/ch
5750
5000
(c)
1 s/ch
55,000
52,500
(d)
10 s/ch
channel number
XBL 8711-9099

Fig. 2



XBL 8510-9584

Fig. 3

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