Tests of the Exponential Decay Law at Short and Long Times

Eric B. Norman, Stuart B. Gazes,[(a)] Stephanie G. Crane,[(b)] and Dianne A. Bennett
Nuclear Science Division, Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720
(Received 29 December 1987)

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

PACS numbers: 23.40.−s, 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 nonexponential. 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 quasistationary state will exhibit deviations from exponential decay at sufficiently long times if the decay spectrum is cut off at a finite energy level (because of, for example, the nonzero 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 α decay of ^{222}Rn and found no deviations from exponential behavior out to the limit of his sensitivity at 27 half-lives. Butt and Wilson[24] extended such studies of radon out to 40 half-lives. Winter[25] and Gopych *et al.*[26] have performed searches out to 34 half-lives for the β decay of ^{56}Mn and to 33 half-lives for the β decay of ^{116}In^m. The decay curves of the μ^+, π^+, K^+, K_S^0, K_L^0, 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.[27]

One of the reasons for this dearth of experimental data is that the nonexponential effects discussed by Khalfin may be appreciable only in experimentally inaccessible time domains. 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, nonexponential 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 β 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 yr and 2.5785 h, respectively, and the energies of the γ rays emitted following their β 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 Lederer and Shirley.[28]

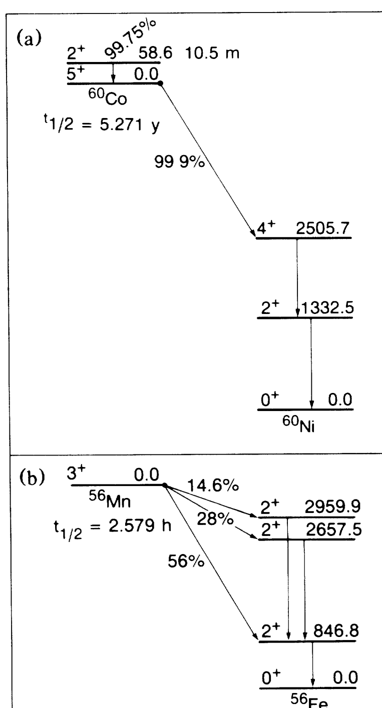

FIG. 1. Decay schemes of ^{60}Co and ^{56}Mn.

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 know accurately 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 University of California at Berkeley TRIGA Mark III reactor. A 1-g 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 ms. Immediately after this activation, the sample contained approximately 4 Ci of the $t_{1/2}=10.5$ min ^{60}Co^m. After we allowed this activity to decay for 3.5 h, the sample contained 15 μCi 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 $8\times10^{-5}t_{1/2}(^{60}\mathrm{Co})$ after the activation. γ-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. Because of 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 with the assumption of purely exponential decay with the known ^{60}Co half-life. One way to parametrize such decays has been given by Gopych *et al.*[26] as

$$N(t)+N_0\exp(-t/\tau)[1+\alpha\sin(\beta t/\tau)], \quad (1)$$

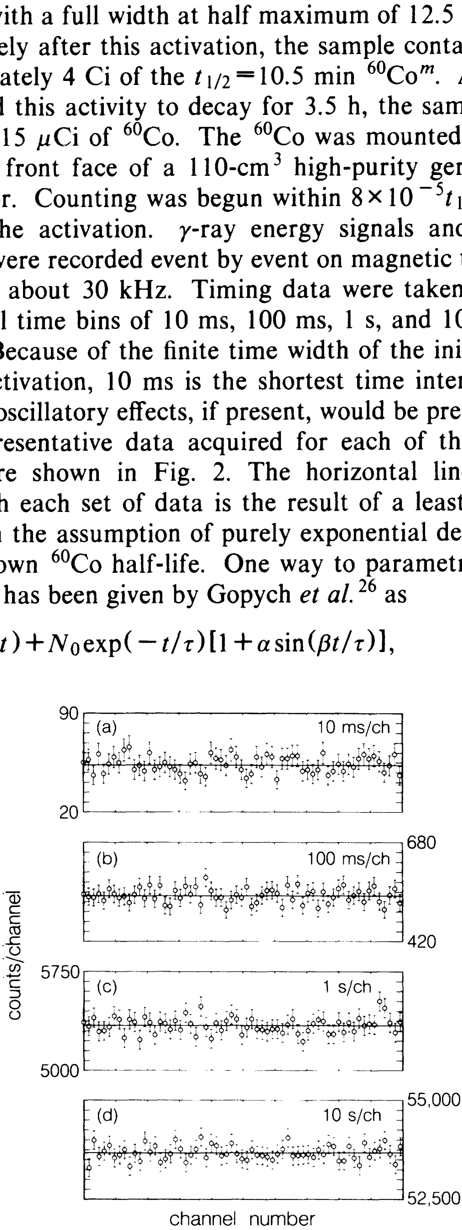

FIG. 2. Numbers of γ rays observed from the ^{60}Co source in time bins of (a) 10 ms, (b) 100 ms, (c) 1 s, and (d) 10 s. The straight line drawn through each data set is the result of a least-squares fit with the assumption of purely exponential decay with the known ^{60}Co half-life.

where τ is the lifetime of the decaying system, and α is the amplitude and β is the frequency of the nonexponential 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 with use of the fast-Fourier-transform algorithm. Such an analysis allows us to scan simultaneously 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 our taking the time spectra, adding a sine wave of known amplitude, and then performing the fast Fourier transform. Half of the ^{60}Co activity that we observed was produced initially as the $t_{1/2}=10.5$ min ^{60}Co^m. Thus, the amplitude limits derived from these fits were multiplied by a factor of 2 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 exposure of 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 microcuries 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. γ-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,[25] it was necessary to purify the fourth manganese sample chemically after activation to

TABLE I. Results of the searches for oscillatory behavior in the short-time decay of ^{60}Co. The limits on the amplitude α as functions of the assumed frequency β were derived from least-squares fits of our data by Eq. (1).

Time bin width	Frequency range	α
10 ms	$7.5\times10^{10}<\beta<5.9\times10^{8}$	<0.056
100 ms	$7.5\times10^{9}<\beta<5.9\times10^{7}$	<0.020
1 s	$7.5\times10^{8}<\beta<5.9\times10^{6}$	<0.0068
10 s	$7.5\times10^{7}<\beta<2.4\times10^{6}$	<0.0032

TABLE II. Results of the searches for nonexponential behavior in the long-time decay of ^{56}Mn. The limits on A were derived from least-square fits of our 1811-keV data by Eq. (2).

Sample	Initial activity	Counting interval [$t_{1/2}$ (^{56}Mn)]	$t_{1/2}$ (h)	A	$A(t/\tau)^2\|_{t=t_{max}}$
1	1 μCi	0.3–13	2.583 ± 0.008	$<4.0\times10^{-4}$	<0.035
2	1 mCi	11–22	2.585 ± 0.005	$<3.4\times10^{-5}$	<0.011
3	1 Ci	20–33	2.576 ± 0.005	$<1.0\times10^{-5}$	<0.0068
4	800 Ci	34–45	2.573 ± 0.019	$<1.3\times10^{-5}$	<0.013

remove ^{24}Na, ^{59}Fe, and ^{60}Co produced by neutron captures on parts-per-million impurities in the manganese sample. At the end of the irradiation, this sample contained approximately 800 Ci of ^{56}Mn. After we allowed 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 approximately 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 γ-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 γ rays shown in Fig. 3. The straight line drawn through each set of data is the result of a least-squares fit with the assumption of purely exponential decay with the known ^{56}Mn half-life. It has been suggested[26] that decay curves may actually be described by

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

Our data were also analyzed to search for nonexponential 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 with use of the β decay of ^{60}Co. Analysis of our data indicates that oscillatory behavior in the range of periods of $(10^{-10}$ to $3\times10^{-6})t_{1/2}$ contributes no more than 5.6% to the decay rate of ^{60}Co. We have also extended the limits

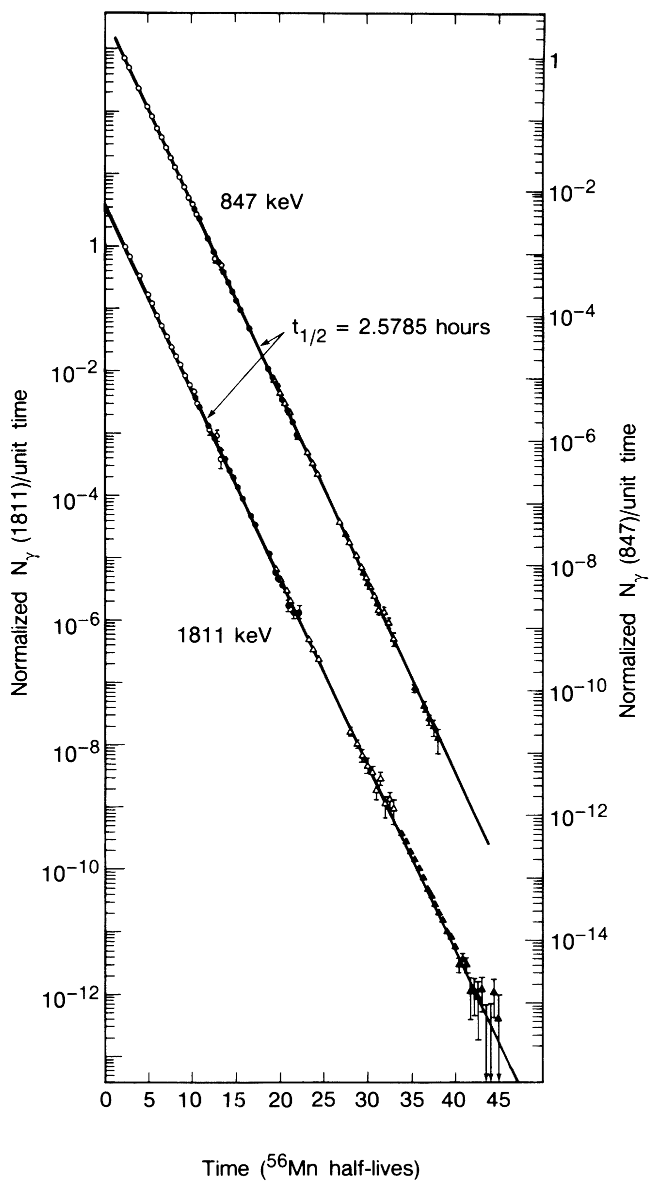

FIG. 3. Composite decay curves for the 847- and 1811-keV γ rays observed from the decay of ^{56}Mn. The straight lines are the results of least-squares fits with the assumption of purely exponential decay with the known ^{56}Mn half-life.

on the long-time searches out to approximately 45 half-lives using the β decay of ^{56}Mn. Our data show that nonexponential behavior of the type indicated in Eq. (2) contributes less than 1.3% to the decay rate of ^{56}Mn at $45t_{1/2}$. Chiu, Sudarshan, and Misra,[10] among others, have estimated that in order for the nonexponential effects to be comparable to the ordinary decay rate, one may have to begin counting at roughly $10^{-29}t_{1/2}$ in the case of ^{60}Co. Winter[25] estimated that for the decay of ^{56}Mn, the nonexponential terms may become as large as the exponential one at about $200t_{1/2}$. Extension of measurements such as ours out to these extremes seems to be beyond the range of present technology.

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 No. DE-AC03-76SF00098.

---

[a]Present address: Department of Physics and Astronomy, University of Rochester, Rochester, NY 14627.

[b]Present address: Department of Astronomy, Columbia University, New York, NY 10027.

[1]L. A. Khalfin, Zh. Eksp. Teor. Fiz. **33**, 1371 (1958) [Sov. Phys. JETP **6**, 1053 (1958)].

[2]P. T. Matthews and A. Salam, Phys. Rev. **115**, 1079 (1959).

[3]R. G. Winter, Phys. Rev. **123**, 1503 (1961).

[4]M. L. Goldberger and K. M. Watson, Phys. Rev. **136**, B1472 (1964).

[5]L. A. Khalfin, Pis'ma Zh. Eksp. Teor. Fiz. **8**, 106 (1968) [JETP Lett. **8**, 65 (1968)].

[6]H. Ekstein and A. J. F. Siegert, Ann. Phys. (N.Y.) **68**, 509 (1971).

[7]M. I. Shirokov, Yad. Fiz. **21**, 674 (1975) [Sov. J. Nucl. Phys. **21**, 347 (1975)].

[8]P. L. Knight, Phys. Lett. **56A**, 11 (1976).

[9]J. Dolejši and P. Exner, Czech. J. Phys. B **27**, 855 (1977).

[10]C. B. Chiu, E. C. G. Sudarshan, and B. Misra, Phys. Rev. D **16**, 520 (1977).

[11]M. D. Arthur *et al.*, Nuovo Cimento **63B**, 565 (1981).

[12]P. I. Margvelashvili and A. Baker, Phys. Lett. **131B**, 390 (1983).

[13]E. J. Robinson, Phys. Rev. Lett. **52**, 2309 (1984).

[14]M. H. Mittleman and A. Tip, J. Phys. B **17**, 571 (1984).

[15]E. J. Robinson, Phys. Rev. A **33**, 1461 (1986).

[16]E. J. Robinson, Phys. Rev. Lett. **57**, 1281 (1986).

[17]L. A. Khalfin, Phys. Lett. **112B**, 223 (1982).

[18]C. B. Chiu, B. Misra, and E. C. G. Sudarshan, Phys. Lett. **117B**, 34 (1982).

[19]G. N. Fleming, Phys. Lett. **125B**, 287 (1983).

[20]A. M. Lane, Phys. Lett. **99A**, 359 (1983).

[21]F. Miglietta and A. Rimini, Phys. Lett. **139B**, 353 (1984).

[22]K. Grotz and H. V. Klapdor, Phys. Rev. C **30**, 2098 (1984).

[23]E. Rutherford, Stizungsber. Akad. Wiss. Wien, Math.-Naturwiss. Kl., Abt. 2A **120**, 303 (1911).

[24]D. K. Butt and A. R. Wilson, J. Phys. A **5**, 1248 (1972).

[25]R. G. Winter, Phys. Rev. **126**, 1152 (1962).

[26]P. M. Gopych *et al.*, Yad. Fiz. **39**, 257 (1984) [Sov. J. Nucl. Phys. **39**, 159 (1984)].

[27]N. N. Nikolaev, Usp. Fiz. Nauk **95**, 506 (1968) [Sov. Phys. Usp. **11**, 522 (1968)].

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