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Experimental Tests for Randomness of Quantum Decay Examined as a Markov Process

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Abstract

The number of decays from four distinct nuclear disintegration processes were recorded over a long succession of counting intervals, converted into sequences of binary outcomes based on parity, and examined as a discrete two-state Markov process. The difference in single-step transition probabilities was found to be null to within an uncertainty of order 10^{-3}, supporting the proposition that quantum particles decay at random unaffected by their past history.

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1. Introduction: Quantum Decay as a Stochastic Process

A fundamental prediction of quantum theory is that spontaneous quantum transitions are random events independent of the previous history of the particle (nucleus, atom, molecule, etc.) undergoing the transition. One cannot predict which particle of an ensemble of quantum particles will be the next to undergo a transition or precisely when such a transition will occur. Only the statistics of the aggregate transitions occurring within a system can be known (i.e. calculated).

Although the predictions of quantum theory have been tested in many ways over the past three quarters of a century, there have been surprisingly few investigations of quantum processes specifically for randomness, and these have generally focussed on measuring the distribution of time intervals between two sequential nuclear decays [[1]]. We have recently reported the results of a new series of tests of quantum behaviour based on the measurement of long temporal sequences of nuclear alpha, beta, and electron-capture decays which we analysed by means of the theory of runs [[2],[3]]. A run of length k is an uninterrupted sequence of k identical events in a series of binary outcomes (like heads H and tails T in a coin toss). Someone unfamiliar with the laws of probability might intuitively expect a random sequence to give rise to short runs {for example, sequences like ...HTH...HTTTH....HTTTH....), but an overabundance of such reversals between binary outcomes signifies a departure from statistical control. By contrast, the occurrence of long runs (for example ...THHHHHHT...) in a data sequence may seem to signify an underlying order or regularity, but in fact is a natural and calculable outcome of a perfectly random process and occurs with greater probability the larger the number of trials. In short, the intuitive perception of what is random can be greatly misleading, and one must rely on mathematically objective tests of randomness.

It is worth noting at the outset that investigations of nonlinear dynamics and algorithmic complexity theory have shown that no finite sequence can in principle be random [[4],[5]]. Thus, since every experiment having a beginning and an end must necessarily yield a finite number of data, one cannot with certainty demonstrate empirically that a particular stochastic process is random. Nevertheless, if a data sequence generated by a stochastic process is sufficiently long, it will appear for all practical purposes to be a random sequence. Nonrandom behaviour unrelated to sequence length, therefore, should signify that the underlying process is not random—or, in statistical parlance that there is an "assignable cause" to the results. Moreover, no matter how many tests of randomness a stochastic process passes, there may yet be one more test that it fails—and it takes only *one* unproblematic failure to demonstrate that a process is *not* random[6]. Hence the necessity for subjecting quantum processes to a variety of complementary tests.

In the present Letter we report a new series of tests of the randomness of quantum decay by examining the disintegration of nuclei as a two-state discrete Markov process [[7]]—i.e. a stochastic process in which the state of the system at one time is determined by its history through a chain of antecedent states. In contrast to our previous run tests, where the question at issue was basically "How many times in succession is the outcome of a Bernoulli trial the same?", the essential question in our Markov-chain test is this: Given that a system is initially in a particular state ε_i, what is the probability of finding the system n time intervals later in the same state ε_i (retention probability) or in another state ε_j (transition probability)? If the single-step retention and transition probabilities differ, then the probability of finding the system in a given state at time n will depend on n. According to quantum theory, which we elaborate below, this probability should be independent of n.

We have tested for Markoffian behaviour four distinct nuclear processes:

(a) alpha decay of ^{241}Am (half-life 432.2 years)

$$^{241}_{95}\mathrm{Am} \rightarrow {}^{237}_{93}\mathrm{Np} + {}^{4}_{2}\mathrm{He}\,, \tag{1a}$$

(b) beta decay of ^{137}Cs (half-life 30.4 years)

$$^{137}_{55}\mathrm{Cs} \rightarrow {}^{137}_{56}\mathrm{Ba} + \beta^{-}, \tag{1b}$$

(c) electron-capture decay of ^{54}Mn (half-life: 312.3 days)

$$^{54}_{25}\mathrm{Mn} + e^{-} \rightarrow {}^{54}_{24}\mathrm{Cr}\,, \tag{1c}$$

and (d) beta decay of ^{214}Bi (half-life 19.9 minutes) followed by alpha decay of ^{214}Po (half-life 164.3 μs)

$$
\begin{aligned}
{}^{214}_{83}\mathrm{Bi} &\rightarrow {}^{214}_{84}\mathrm{Po} + \beta^- \\
{}^{214}_{84}\mathrm{Po} &\rightarrow {}^{210}_{82}\mathrm{Pb} + {}^{4}_{2}\mathrm{He}
\end{aligned}
. \tag{1d}
$$

The transmutation of americium (1a) and polonium (1d) produce alpha particles of mean energies 5485.6 keV and 7687 keV respectively. The transmutation of cesium and manganese give rise to gamma rays of mean energies 661.7 keV and 834.8 keV respectively. Our experimental procedure is to count the numbers of alphas or gammas in a long sequence of time intervals, each interval (a "bin") of 100 ms duration. Details of the alpha and gamma spectrometers are given in References 2 and 3. In the course of our experiments the activity of each radioactive source was effectively constant, either because the lifetime of the source was much greater than the duration of the experiment (as in the case of $^{241}\mathrm{Am}$, $^{137}\mathrm{Cs}$, and $^{54}\mathrm{Mn}$) or as a result of the secular equilibrium [8] of a short-lived nuclide with a much longer-lived parent nuclide (as in the continuous regeneration of $^{214}\mathrm{Bi}$ and $^{214}\mathrm{Po}$ from $^{226}\mathrm{Ra}$ [half-life 1620 years]).

Nuclear counting was performed under experimental conditions of (a) high mean count per bin ($\mu >> 1$) and (b) low mean count per bin ($\mu << 1$), and the temporal sequence of digital counts $\{x_i,\ i=1...N\}$ for each disintegration process was converted to a sequence $\{\varepsilon_i,\ i=1...N\}$ of binary outcomes by replacing each x_i with $\varepsilon_i = 0$ for an even parity count and with $\varepsilon_i = 1$ for an odd parity count. In this way, the nuclear disintegration data were modeled as a 2-state Markov process of chain length N. For the low count rate configuration (b) there was never more than one count per bin, and therefore the two Markoffian states $\varepsilon_i(x_i = 0) = 0$ and $\varepsilon_i(x_i = 1) = 1$ correspond directly to the two nuclear basis states "no decay" and "decay" of a Schrödinger's cat experiment.

2. Markov Chain Model

Let ξ_n be a binary random variable with values 0 or 1. We denote the single-step transition probability p_{ij} as the probability that the system is in state ε_j at time n if it was in state ε_i at time $n-1$, i.e.

$$
p_{ij} = \Pr\{\xi_n = j \mid \xi_{n-1} = i\}, \tag{2a}
$$

from which follows the single-step transition matrix [9]

$$\boldsymbol{P} = \begin{pmatrix} p_{00} & p_{01} \\ p_{10} & p_{11} \end{pmatrix}. \tag{2b}$$

Conservation of probability requires that

$$p_{00} + p_{01} = p_{11} + p_{10} = 1\,. \tag{3}$$

The eigenvalues of $\boldsymbol{P}$ are 1 and d, in which

$$d = p_{00} - p_{10} = p_{11} - p_{01} = p_{11} + p_{00} - 1 = 1 - (p_{01} + p_{10})\,. \tag{4}$$

[The equivalence of the various expressions for d follow from use of Eq. (3).]

We further denote $p_{ij}(n)$ as the probability that the system is in state ε_j at time n if it was initially in state ε_i at time $n = 0$

$$p_{ij}(n) = \Pr\{\xi_n = j | \xi_0 = i\}\,, \tag{5a}$$

from which follows the two Markov chain recursion relations:

$$\begin{aligned} p_{i0}(n) &= p_{i0}(n-1)p_{00} + p_{i1}(n-1)p_{10} \\ p_{i1}(n) &= p_{i0}(n-1)p_{01} + p_{i1}(n-1)p_{11} \end{aligned} \tag{5b}$$

expressible succinctly as a single matrix relation

$$u_i(n) = u_i(n-1)\boldsymbol{P} \tag{5c}$$

in which

$$u_i(n) = \left(p_{i0}(n), p_{i1}(n)\right) \tag{5d}$$

is a row vector giving the state of the system at time n.

The solution to Eq. (5b) or (5c) is

$$u_i(n) = u_i(0)\,\boldsymbol{P}^n \tag{6a}$$

where the n^{th} power of $\boldsymbol{P}$ takes the explicit form [10]

$$\boldsymbol{P}^n = \frac{1}{1-d}\begin{pmatrix} p_{10} + p_{01}d^n & p_{01}\left(1-d^n\right) \\ p_{10}\left(1-d^n\right) & p_{01} + p_{10}d^n \end{pmatrix} = \begin{pmatrix} 1 - p_{01}\left(\dfrac{1-d^n}{1-d}\right) & p_{01}\left(\dfrac{1-d^n}{1-d}\right) \\ p_{10}\left(\dfrac{1-d^n}{1-d}\right) & 1 - p_{10}\left(\dfrac{1-d^n}{1-d}\right) \end{pmatrix}. \tag{6b}$$

Suppose, for example, that the initial state of the system is $\varepsilon_0 = 0$. It then follows from Eqs. (5d) and (6b) that

$$p_{00}(n) = \frac{p_{10} + p_{01}d^n}{p_{10} + p_{01}} \tag{7a}$$

$$p_{01}(n) = \frac{p_{01}\left(1 - d^n\right)}{p_{10} + p_{01}}. \tag{7b}$$

In the case of a symmetric transition matrix with $p_{00} = p_{11}$ and $p_{01} = p_{10}$, the eigenvalue d —which for this special case we designate Δ —is simply the difference between the single-step state retention and transition probabilities. Eqs. (7a) and (7b) then reduce to the expressions

$$p_{00}(n) = p_{11}(n) = \tfrac{1}{2}\left(1 + \Delta^n\right) \tag{8a}$$

$$p_{01}(n) = p_{10}(n) = \tfrac{1}{2}\left(1 - \Delta^n\right) \tag{8b}$$

in which

$$\Delta = p_{ii} - p_{ij} \qquad (i \neq j) \tag{8c}$$

reflects the extent to which the system, once in state 0 or state 1, tends to persist in that state rather than undergo a transition to the opposite state.

3. Quantum Predictions

According to quantum mechanics, the probability p that a radioactive nucleus decays within a time interval Δt is $p = \lambda \Delta t$, where λ is the decay constant; p is consequently independent of the past history of the nucleus. Under the conditions (ordinarily characteristic of a nuclear counting experiment) that $p << 1$ and that the number of nuclei $\mathcal{N}$ greatly exceeds the number k decaying within a specified time interval, the probability of k decays in Δt is given by a Poisson distribution [2]

$$p_\mu(k) = \frac{\mu^k e^{-\mu}}{k!} \tag{9}$$

where $\mu = \mathcal{N} \lambda \, \Delta t$ is the mean count.

(a) Consider first the case where $\mu >> 1$. It then follows from Eq. (9) that the probability $P_e(\mu)$ of obtaining an *even* number of counts within Δt is

$$P_e(\mu) = \sum_{k=0}^{\infty} p_{2k}(\mu) = e^{-\mu}\cosh\mu \ . \tag{10a}$$

Similarly, the probability $P_o(\mu)$ of obtaining an *odd* number of counts within Δt is

$$P_o(\mu) = \sum_{k=1}^{\infty} p_{2k-1}(\mu) = e^{-\mu}\sinh\mu \ . \tag{10b}$$

Since the number of nuclei decaying within the interval Δt is predicted to be independent of the number of disintegrations within an earlier interval, the elements of the single-step transition matrix become

$$\begin{aligned} p_{00} &= p_{10} = P_e(\mu) \\ p_{11} &= p_{01} = P_o(\mu) \end{aligned} \tag{11}$$

and it follows from Eq. (4) that $d = 0$. The n-step transition probabilities then reduce to

$$p_{00}(n) = p_{10}(n) = \frac{1}{1+\tanh\mu} \xrightarrow[\mu>>1]{} \frac{1}{2} \tag{12a}$$

$$p_{11}(n) = p_{01}(n) = \frac{\tanh\mu}{1+\tanh\mu} \xrightarrow[\mu>>1]{} \frac{1}{2} \ . \tag{12b}$$

Figure 1 shows the variation of $P_e(\mu)$ and $P_o(\mu)$ with μ. In the limiting case, in which the mean count per bin μ is well in excess of unity (μ ranged from about 60-125 counts depending on the source), the probability of obtaining an even or odd count is 0.5, and the n-step transition matrix reduces to

$$\boldsymbol{P}_{\text{QM}}^{n} = \boldsymbol{P}_{\text{QM}} = \begin{pmatrix} \frac{1}{2} & \frac{1}{2} \\ \frac{1}{2} & \frac{1}{2} \end{pmatrix} . \tag{12c}$$

(b) Consider next the case $\mu <<1$. One deduces from Eq. (9) that the single-step transition probabilities and the probabilities $P_0(\mu)$ and $P_1(\mu)$ of obtaining respectively 0 or 1 count within Δt are related by

$$p_{00} = p_{10} = P_0(\mu) = e^{-\mu} \tag{13a}$$

$$p_{11} = p_{01} = P_1(\mu) = \mu e^{-\mu} \tag{13b}$$

in which case $d = 0$ again and the n-step transition matrix becomes

$$\boldsymbol{P}_{\mathrm{QM}}^{n} = \boldsymbol{P}_{\mathrm{QM}} = \begin{pmatrix} \dfrac{1}{1+\mu} & \dfrac{\mu}{1+\mu} \\ \dfrac{1}{1+\mu} & \dfrac{\mu}{1+\mu} \end{pmatrix}. \qquad (14)$$

4. Experimental Analyses

We have experimentally determined the probabilities $p_{ij}(n)$ $(i = 0,1;\, j = 0,1)$ for each of the four nuclear decay processes described in Section 1 by decomposing each full data string of N bins into temporal units of length m which we call "m-bins" ($2 \le m \le 20$). For the configuration with $\mu >> 1$, the value of N for each nuclide was N_{Am} = 720,896, N_{Cs} = N_{Po} = 1,048,576, and N_{Mn} = 524,288. For the configuration with $\mu << 1$, we had N_{Cs} = 29,032,448 and N_{Mn} = 3,359,528. To determine $p_{0i}(n)$, for example, we counted the number $N_0(m)$ of m-bins ($m = n+1$) whose first element was 0, and of these counted the numbers $N_{00}(m)$, $N_{01}(m)$ of m-bins whose m^{th} element was 0, 1 respectively. By the strong form of the law of large numbers [6], the probabilities are then given by the respective quotients $p_{00}(m) = N_{00}(m)/N_0(m)$ and $p_{01}(m) = N_{01}(m)/N_0(m)$. In the same way the probabilities $p_{1i}(m)$ were determined. We consider respectively the results for $\mu >> 1$ and $\mu << 1$.

(a) <u>High count rate configuration</u>

Table 1 shows a sample of results for ^{137}Cs ($\mu \sim 60$) over the range $2 \le m \le 10$. (The full data set spanned $2 \le m \le 20$.) Figures 2, 3, 4, and 5 show plots of $p_{00}(m)$ and $p_{01}(m)$ vs. m for ^{241}Am ($\mu \sim 125$), ^{137}Cs, ^{54}Mn ($\mu \sim 100$), and ^{214}Po ($\mu \sim 70$), respectively. Corresponding plots of $p_{1i}(m)$ are graphically similar and statistically equivalent and are not presented here for economy of space.

The uncertainty (± one standard deviation σ_p) associated with each point p, which is a ratio of counts of the form $p = N_1/N_2$, is given by

$$\sigma_p = \sqrt{\frac{1}{N_1} + \frac{1}{N_2}}\, p. \qquad (15)$$

Eq. (15) follows from the general relation

$$\sigma_f = \sqrt{\sum_{i=1}^{N} \left(\frac{\partial f}{\partial x_i} \right)^2 \sigma_{x_i}{}^2} \ . \tag{16}$$

for the standard deviation of a function $f(x_1, x_2, \dots x_N)$ of independent random variables $\{x_i\}$ $(i = 1 \dots N)$.

Table 1: ^{137}Cs m-Bin Counts (μ >> 1)

m	N_0	N_{00}	N_{01}	N_1	N_{10}	N_{11}
2	262,010	131,230	130,780	262,278	130,795	131,483
3	174,642	87,523	87,119	174,862	87,101	87,761
4	131,017	65,459	65,558	131,127	65,573	65,554
5	105,079	52,727	52,352	104,617	52,337	52,280
6	87,311	43,612	43,699	87,441	43,681	43,760
7	74,961	37,446	37,515	74,831	37,529	37,302
8	65,454	32,718	32,736	65,618	32,751	32,867
9	58,240	29,190	29,050	58,240	29,066	29,174
10	52,510	26,309	26,201	52,322	26,217	26,105

The difference in single-step transition probabilities Δ was obtained by a least-squares fit of Eq. (8a) to the plots $p_{00}(m)$ and of Eq. (8b) to the plots $p_{01}(m)$. [Recall that $m = n - 1$.] Although Eqs. (8a, b) are not linear in Δ, it is possible to convert the nonlinear difference equations into a set of linear difference equations, and thereby obtain a closed-form analytical expression for Δ, in the following way. Define the functions F_m

$$F_m = \begin{cases} 2p_{00}(m) - 1 = \Delta^{m-1} & (m \neq 1) \\ 1 - 2p_{01}(m) = \Delta^{m-1} & \\ 1 & (m = 1) \end{cases} \tag{17}$$

It then follows from Eq. (15) that $F_2 = \Delta F_1$, $F_3 = \Delta F_2$, ... , $F_M = \Delta F_{M-1}$ where M is the length of the longest m-bin. Upon minimising the error $E = \sum_{m=2}^{M} (F_m - \Delta F_{m-1})^2$ by solving $\frac{dE}{d\Delta} = 0$, we obtain the least-square value

$$\Delta_{ls} = \sum_{m=2}^{M} F_m F_{m-1} \Big/ \sum_{m=2}^{M} \left(F_{m-1}\right)^2 . \tag{18}$$

The uncertainty in Δ_{ls} is obtained by applying Eq. (16) to Eq. (18) and is given approximately by $\sigma_\Delta \approx \frac{2\sigma_p}{F_2}\Delta_{ls}$ where σ_p (approximately the same for all $m \geq 2$) is given by Eq. (15). The exact expression, which is cumbersome and need not be given here, was used in reduction of all data.

Table 2 summarises the results of the four quantum decay processes.

Table 2: Least-Squares Determination of Δ ($\mu >> 1$)

Nuclide		p_{00}	p_{01}	p_{11}	p_{10}
^{241}Am	Δ	-1.90 (-03)	-1.90 (-03)	-6.06 (-04)	-6.06 (-04)
	σ_Δ	4.10 (-03)	4.11 (-03)	4.14 (-03)	4.15 (-03)
^{137}Cs	Δ	1.66 (-03)	1.66 (-03)	2.66 (-03)	2.66 (-03)
	σ_Δ	3.40 (-03)	3.40 (-03)	3.40 (-03)	3.39 (-03)
^{54}Mn	Δ	-2.97 (-04)	-2.97 (-04)	-2.69 (-03)	-2.69 (-03)
	σ_Δ	4.82 (-03)	4.82 (-03)	4.80 (-03)	4.82 (-03)
^{214}Po	Δ	2.70 (-04)	2.70 (-04)	3.52 (-04)	3.52 (-04)
	σ_Δ	3.39 (-03)	3.39 (-03)	3.39 (-03)	3.39 (-03)

(b) Low count rate configuration

For this configuration the single-step transition matrix is not symmetric ($p_{00} \neq p_{11}$, $p_{10} \neq p_{01}$), but we can reduce the data by the same procedure as that of part (a), without having to determine simultaneously any of the elements p_{ij}, by noting [from Eqs. (4) and (6b)] that $p_{01}(n) + p_{10}(n) = 1 - d^n$. In analogy, therefore, to relation (17), we define the function F_m $(n = m-1)$

$$F_m = \begin{cases} 1 - p_{01}(m) - p_{10}(m) \;=\; d^{m-1} & (m \neq 1) \\ 1 & (m = 1) \end{cases} \tag{19}$$

from which the least-squares solution d_{ls} follows from an equation of the same form as (18) with standard deviation $\sigma_d \sim \sqrt{\sigma^2_{p_{01}(2)} + \sigma^2_{p_{10}(2)}}$; $\sigma^2_{p_{ij}(2)}$ is given by Eq. (15).

Figure 6 shows plots of $p_{00}(m)$ and $p_{01}(m)$ vs. m for ^{137}Cs ($\mu \sim .05$); a corresponding plot for ^{54}Mn ($\mu \sim .013$) is graphically similar and not presented here. The plots suggest visually, and least-squares analysis confirms, that $d_{ls} = 0$ to within ±1 standard deviation for both beta decay processes. For $d = 0$, as predicted by quantum mechanics, Eqs. (7ab) [or Eq. (14)] and measurements of the elements of $\boldsymbol{P}^n$ directly yield the value of μ according to

$$\mu = \frac{1 - p_{00}(n)}{p_{00}(n)} = \frac{p_{01}(n)}{1 - p_{01}(n)} . \tag{20}$$

This result is interesting in its own right, for, together with the sample mass and counting interval Δt, it enables one to determine the nuclear decay constant or half-life under conditions where direct temporal measurement of a decay transient is not convenient or possible.

Table 3 summarises the low count rate measurements of d and μ.

Table 3: Least-Squares Determination of d ($\mu \ll 1$)

Nuclide	d_{ls}	σ_d	$\mu(p_{00})$	$\mu(p_{01})$
^{137}Cs	-1.57 (-04)	1.6 (-03)	.0486±.009	.0486±.002
^{54}Mn	-2.94 (-03)	9.5 (-03)	0.0132±.003	.0132±.0002

5. Conclusions

In keeping with the foundations of quantum theory (qt) which hold that the spontaneous decay of a particle occurs randomly and is uninfluenced by its past history, we find that the difference in Markoffian single-step retention and transition probabilities is zero to within an uncertainty of the order of 10^{-3} for all of the nuclear decay processes we have examined under both high count rate and low count rate conditions.

To our knowledge there is at present no viable alternative to qt to explain how correlations could come into play were qt actually to fail these tests. In the absence of theoretical guidance, the experimental examination of quantum decay under conditions of both high and low counting rates provides complementary information. For example, for $\mu >> 1$ it is conceivable (although our tests have shown this not to be the case within present experimental uncertainties) that a high rate of nuclear disintegration in one time interval may lead to a diminished or enhanced rate in the following interval if qt-violating correlations were somehow dependent on sample size (i.e. number of decaying particles). By contrast, for $\mu << 1$, the rare occurrence of a distintegration after a long period of nuclear quiescence might modify the decay probability of a subsequent particle if qt-violating correlations were somehow sensitive to proximity. In such a case, a violation of qt would be more noticeable within a counting interval containing at most one particle than a hundred particles.

We also stress that the results reported here (as well as those of Refs. 2 and 3) are distinct from—and provide a test more fundamental than—measurements of the exponential character of nuclear decay. The two features attributed to quantum transmutation processes (randomness and exponential decay) are frequently confounded, but that is incorrect. Although experimental tests of which we are aware confirm the exponential decay law for nuclei [11], in all rigour qt predicts variations from this law at very short and long times [12], and such variations have been seen in non-nuclear systems [13]. Furthermore, particles (e.g. atoms and molecules) that decay from a linear superposition of energy eigenstates give rise to a harmonically modulated exponential decay law [14] (the phenomenon of "quantum beats"); individual decays, however, are expected to occur at random.

Although the Markov chain analysis, in contrast to application of the theory of runs, provides a simple and direct way to determine d analytically, it is worth emphasising that the distribution of runs is also very sensitive to d. As a consistency check and as a way of gauging the sensitivity of our statistical procedures to any underlying regularity (or assignable cause) in nuclear decay, we simulated a string of 10^6 Bernoulli trials with a random number generator, assigning a "bias" Δ in the single-step transition probabilities in the following way. For a total range $1 \le r \le R$ of the random number r (with R = 50,000 in our simulations), we assigned single-step probabilities $p_{ii} = R_{ii}/R$ and $p_{ij} = R_{ij}/R$ where $R_{ii} + R_{ij} = R$. That is, if the state at time n was $\varepsilon_n = 0$ and the next trial yielded a random number in the range $1 \le r \le R_{ii}$, the state remained unchanged, $\varepsilon_{n+1} = 0$. If r fell within the range $(R_{ii}+1) \le r \le R$, the state underwent a transition, $\varepsilon_{n+1} = 1$. Thus, by adjusting the subrange limit R_{ii} with fixed R, we determined the distribution of runs of 0's or 1's for different values of the bias $\Delta = \left(R_{ii} - R_{ij}\right)/R$.

In Figure 7 is plotted $\Delta\, r_k = r_k^{\mathrm{expt}} - r_k^{theory}$, the difference between the experimental (i.e. simulated) and theoretical numbers of runs of 0's of length k obtained in 10^6 trials, as a function of k for a bias $\Delta = 0.005$. The theoretical relations for random runs (i.e. with no bias) are given in References 2 and 3. The positive bias signifies a higher probability for remaining in a state than for transition ($p_{ii} > p_{ij}$), in which case one would expect a smaller than random number of short runs. The pronounced negative deviation of $\Delta\, r_k$ from zero for k = 1 and 2 bears out this expectation. There is, as well, a visibly larger than random number of runs of length $k = 3$.

Of particular interest here is the fact that for a number of Bernoulli trials (10^6) of the same order as the number of counting intervals in our nuclear data the simulated distributions of runs showed a recognisably nonrandom pattern for Δ as small as approximately .001—i.e. in complete accord with the limits of detection of Δ inferred from the Markov chain analysis. To place a more stringent limit on any difference in single-step transition probabilities—and therefore on the history dependence of quantum decay—would require increasing the number of bins either by shortening the counting interval Δt or lengthening the total counting time. We are presently looking into both possibilities.

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[6]The question of whether the failure of a finite string of data to satisfy a test for randomness actually indicates that the underlying process is random is not a trivial one. The answer is not simply yes or no, but a confidence level dependent upon the length of the data string. We have established (e.g. by computer simulation of nonrandom processes) that with a string of the order of one million bits we could detect a bias of the order of one part in a thousand with virtual certainty. This is discussed in Section 5.

[7]For the specific characteristics of discrete and continuous Markov processes see Y. A. Rozanov, *Introductory Probability Theory* (Prentice-Hall, Englewood Cliffs NJ, 1969) 83-114.

[8]G. Friedlander and J. W. Kennedy, *Nuclear and Radiochemistry,* (Wiley, New York, 1955) pp. 132-134

[9]Although we have designated elements like p_{00} a "retention probability" and p_{01} a "transition probability", all the elements of the transition matrix $\boldsymbol{P}$ are technically regarded as transition probabilities.

[10]If $\boldsymbol{M}$ is the matrix that brings $\boldsymbol{P}$ into the diagonal form $\boldsymbol{D} = \begin{pmatrix} d & 0 \\ 0 & 1 \end{pmatrix}$, then $\boldsymbol{P}^k = \boldsymbol{M}^{-1}\boldsymbol{D}^k\boldsymbol{M}$.

The diagonalising matrix $\boldsymbol{M}$ is the matrix of eigenvectors. See M. P. Silverman, *More Than One*

Mystery: Explorations in Quantum Interference (Springer-Verlag, New York, 1995), pp 139-143.

[11] E. B. Norman, S. B. Gazes, S. G. Crane, and D. A. Bennett, "Tests of the Exponential Decay Law at Short and Long Times", *Physical Review Letters* **60** (1988) 2246-2249.

[12] M. P. Silverman, *Probing The Atom: Interactions of Coupled States, Fast Beams, and Loose Electrons,* (Princeton University Press, Princeton NJ, 2000), pp. 122-127. Exponential decay arises from poles in the lower half of the second Riemann sheet in the evaluation of the imaginary part of the forward scattering amplitude. Corrections to exponential decay arise from integration around a branch cut along the real axis.

[13] S. R. Wilkinson, C. F. Bharucha, M. C. Fischer, K. W. Madison, P. R. Morrow, Q. Niu, B. Sundaram, and M. G. Raizen, "Experimental evidence for non-exponential decay in quantum tunnelling", *Nature* **387** (5 June 1997) 575-578.

[14] M. P. Silverman, *More Than One Mystery: Expolorations in Quantum Interference* (Springer-Verlag, New York, 1995), pp. 100-128

Figures

Figure 1 Variation of probabilities $P_e(\mu)$ and $P_o(\mu)$ as a function of mean count μ for a stochastic process (nuclear decay) following a Poisson distribution.

Figure 2 Variation of single-step transition probabilities (a) $p_{00}(m)$ and (b) $p_{01}(m)$ as a function of m-bin length m for the alpha decay of ^{241}Am ($\mu \sim 125$).

Figure 3 Variation of single-step transition probabilities (a) $p_{00}(m)$ and (b) $p_{01}(m)$ for the beta decay of ^{137}Cs ($\mu \sim 60$).

Figure 4 Variation of single-step transition probabilities (a) $p_{00}(m)$ and (b) $p_{01}(m)$ for the electron capture decay of ^{54}Mn ($\mu \sim 100$).

Figure 5 Variation of single-step transition probabilities (a) $p_{00}(m)$ and (b) $p_{01}(m)$ for the beta decay of ^{214}Bi followed by alpha decay of ^{214}Po ($\mu \sim 70$).

Figure 6 Variation of single-step transition probabilities (a) $p_{00}(m)$ and (b) $p_{01}(m)$ for the beta decay of ^{137}Cs ($\mu \sim .05$)

Figure 7 Simulated distribution of runs of 0's for 10^6 Bernoulli trials (with binary outcome 0 or 1) with bias parameter $\Delta = .005$. The plot shows the difference between experimental (i.e. simulated) and theoretical ($\Delta = 0$) values of the numbers of runs of length k as a function of k.

Figure 1

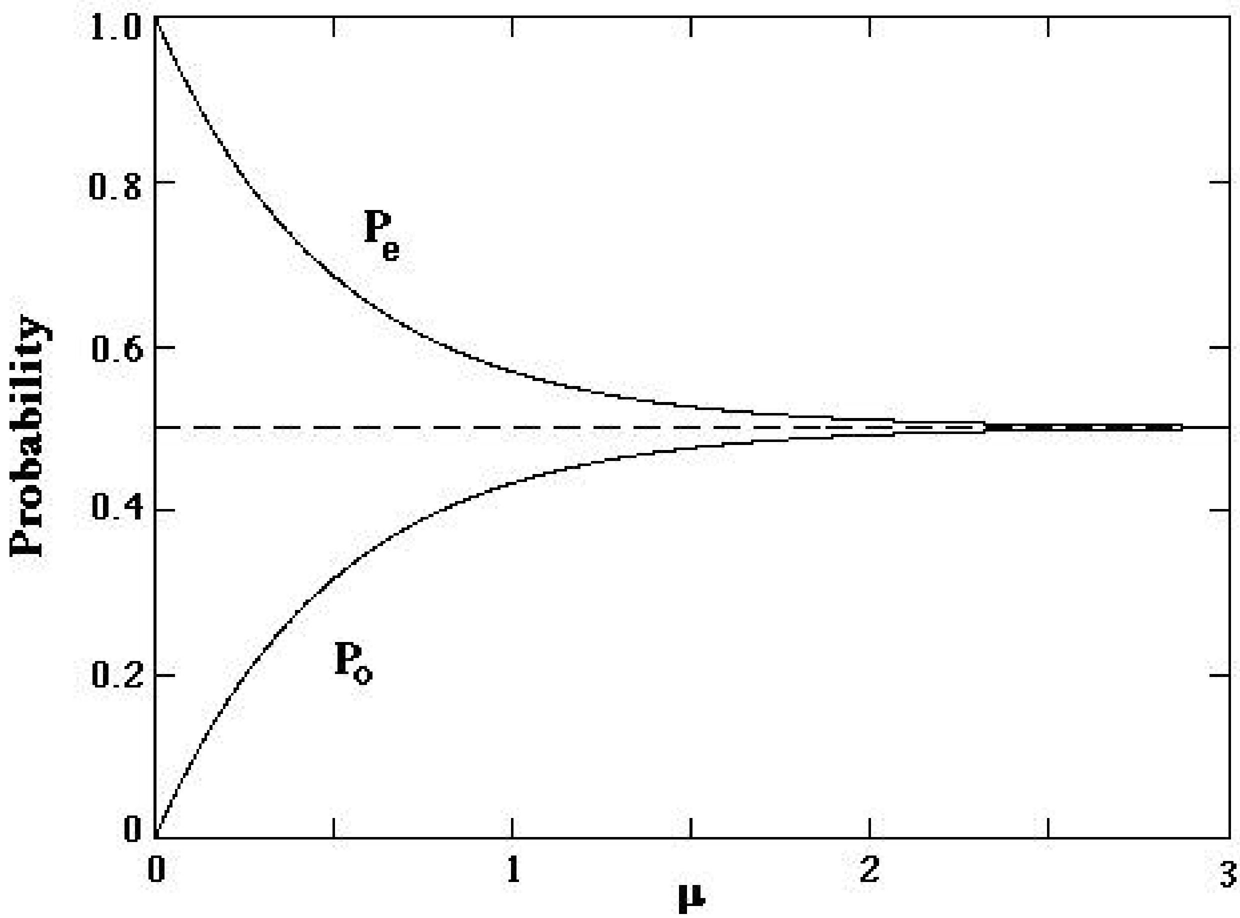
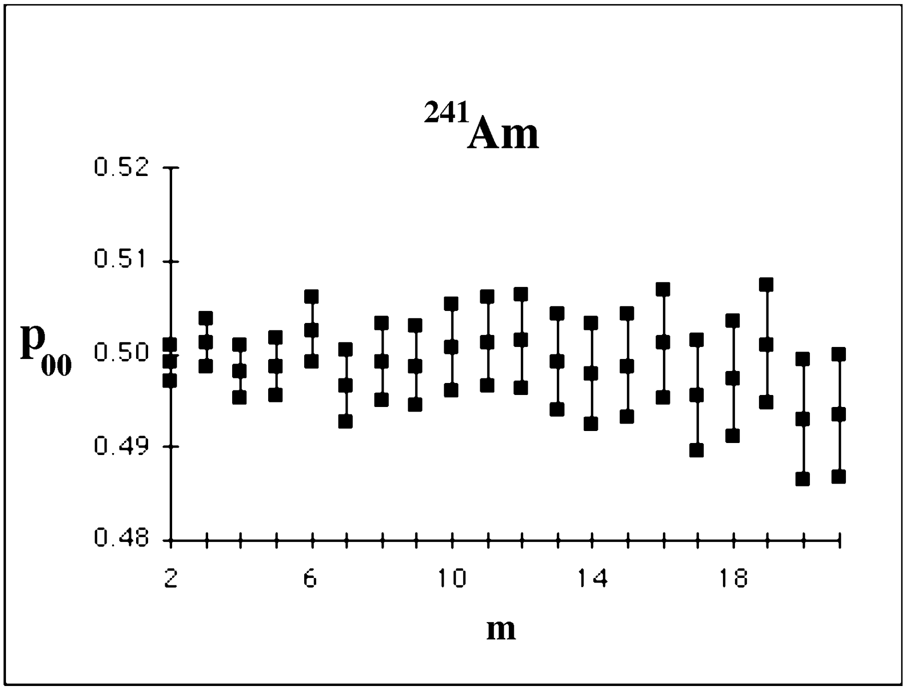


Figure 2


241Am
0.52
0.51
0.50
0.49
0.48
p00
2
6
10
14
18
m

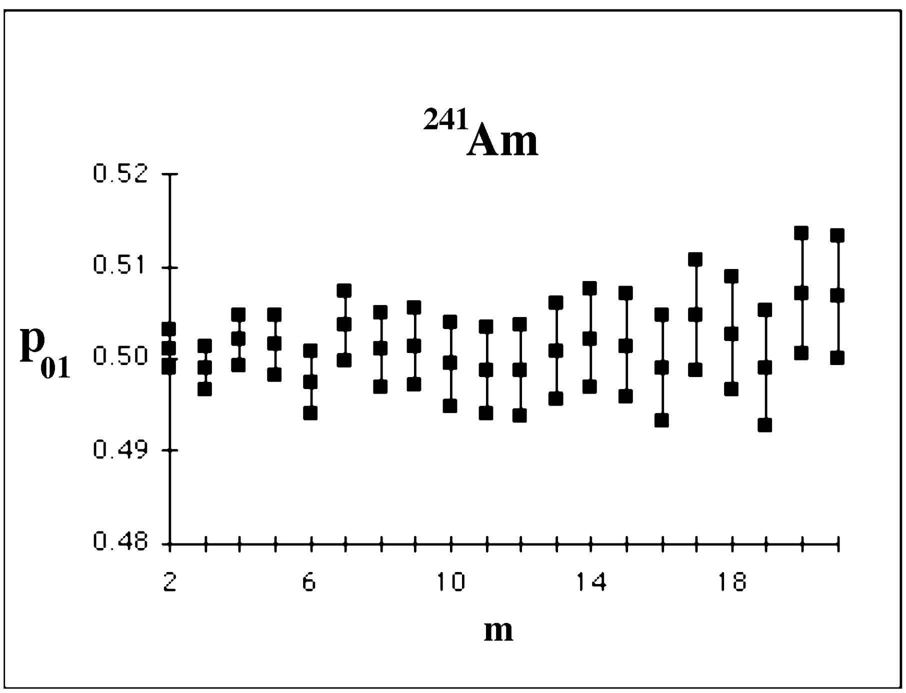
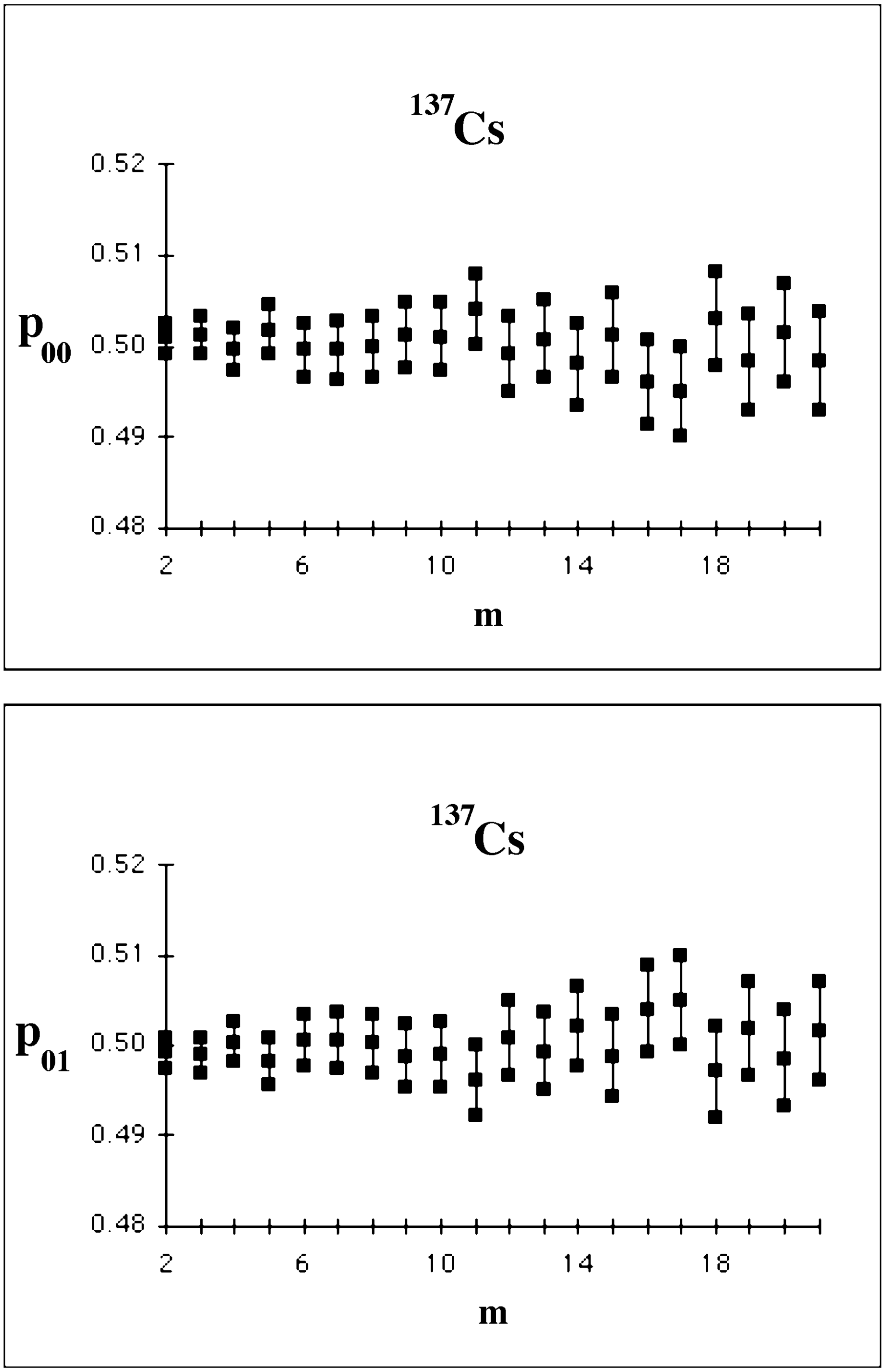
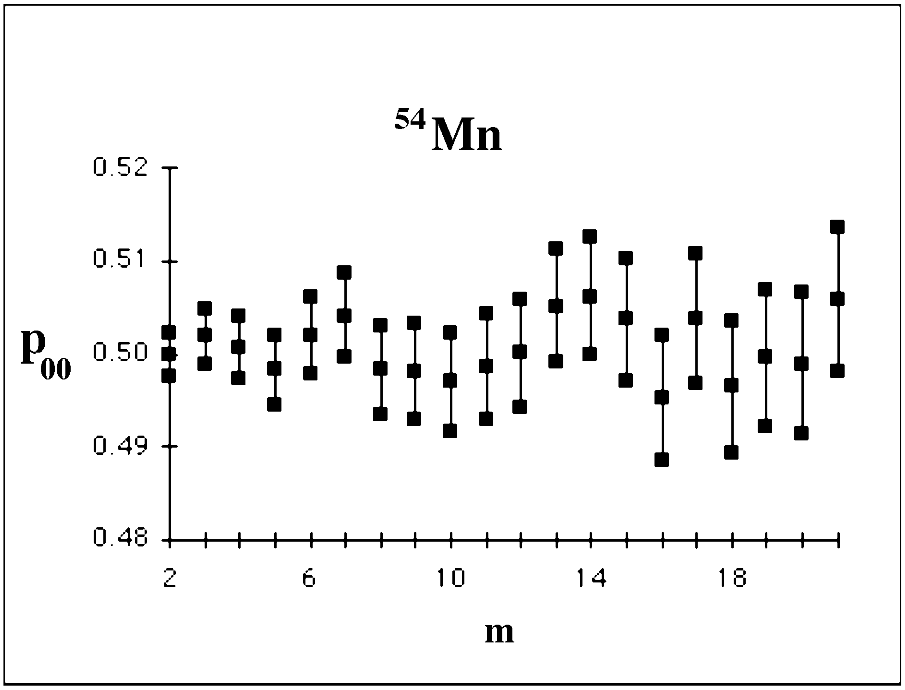

241Am
0.52
0.51
0.50
0.49
0.48
p01
2
6
10
14
18
m

Figure 3

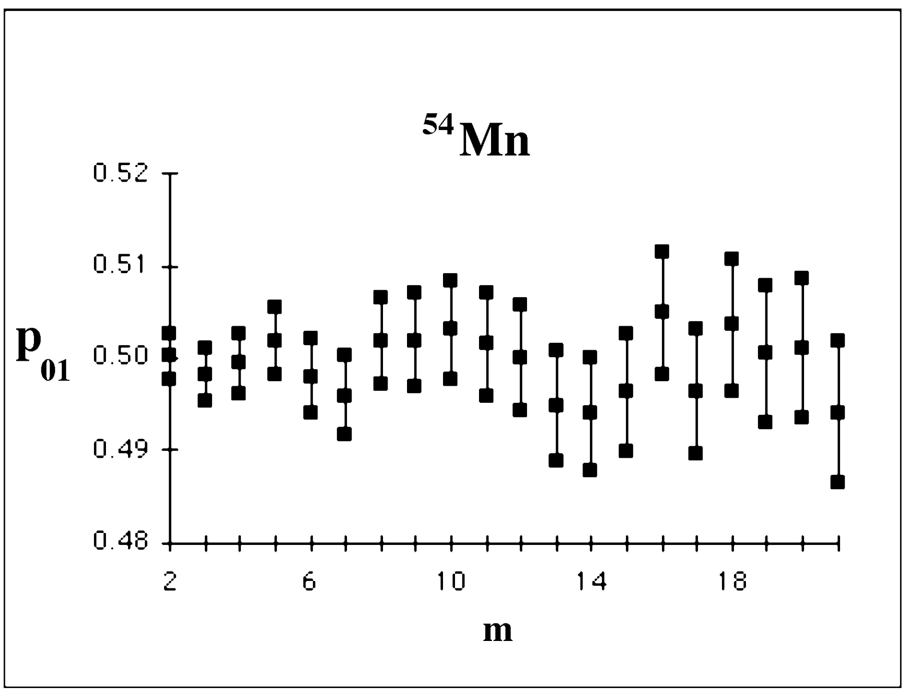


137Cs
0.52
0.51
0.50
0.49
0.48
p00
2
6
10
14
18
m
137Cs
0.52
0.51
0.50
0.49
0.48
p01
2
6
10
14
18
m

Figure 4

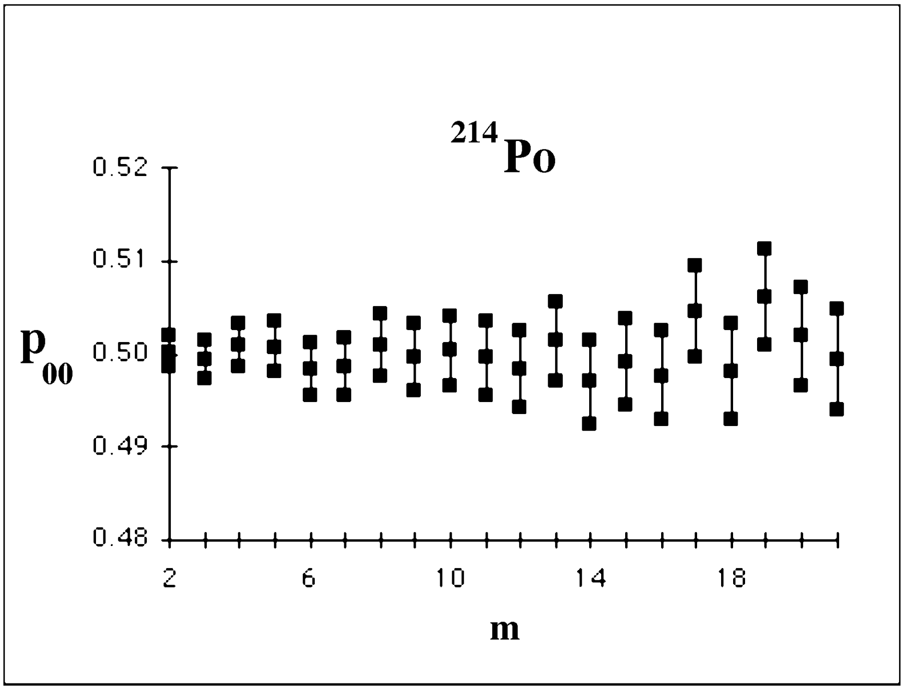


^{54}Mn
0.52
0.51
0.50
0.49
0.48
p_{00}
2
6
10
14
18
m

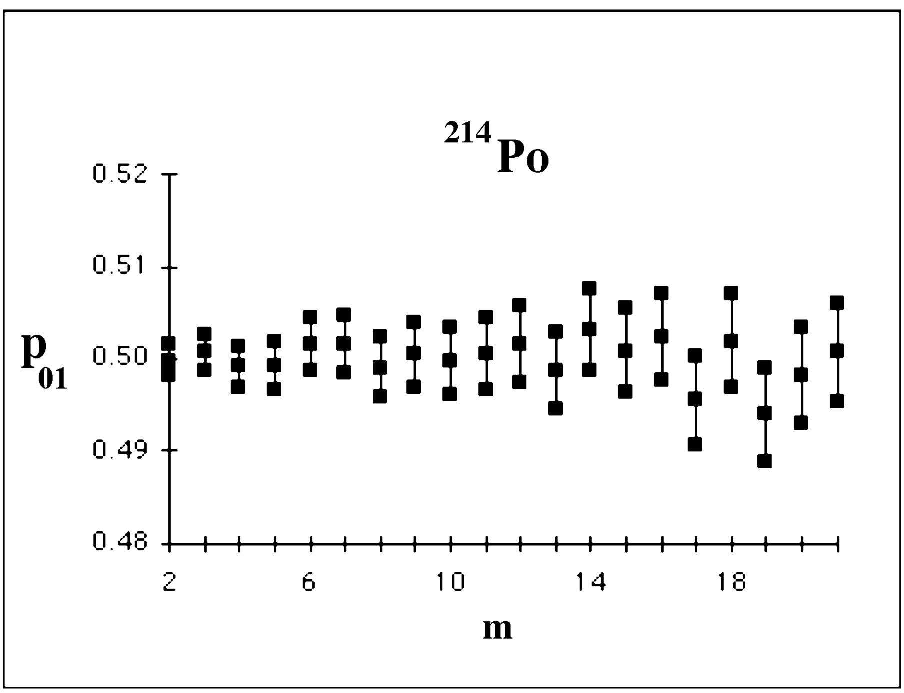


^{54}Mn
0.52
0.51
0.50
0.49
0.48
p_{01}
2
6
10
14
18
m

Figure 5

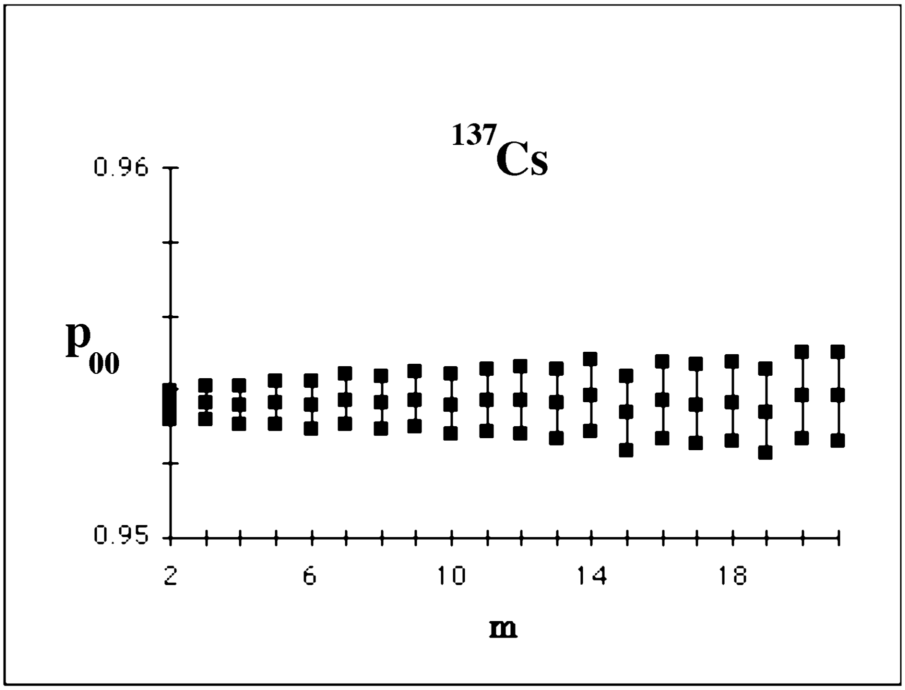


^{214}Po
0.52
0.51
0.50
0.49
0.48
p_{00}
2
6
10
14
18
m

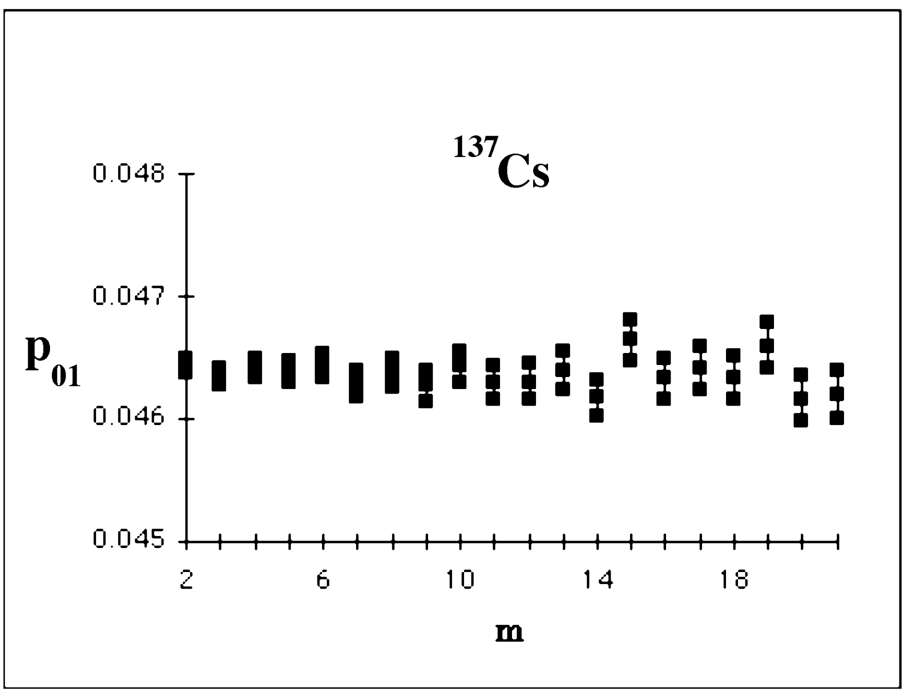


^{214}Po
0.52
0.51
0.50
0.49
0.48
p_{01}
2
6
10
14
18
m

Figure 6

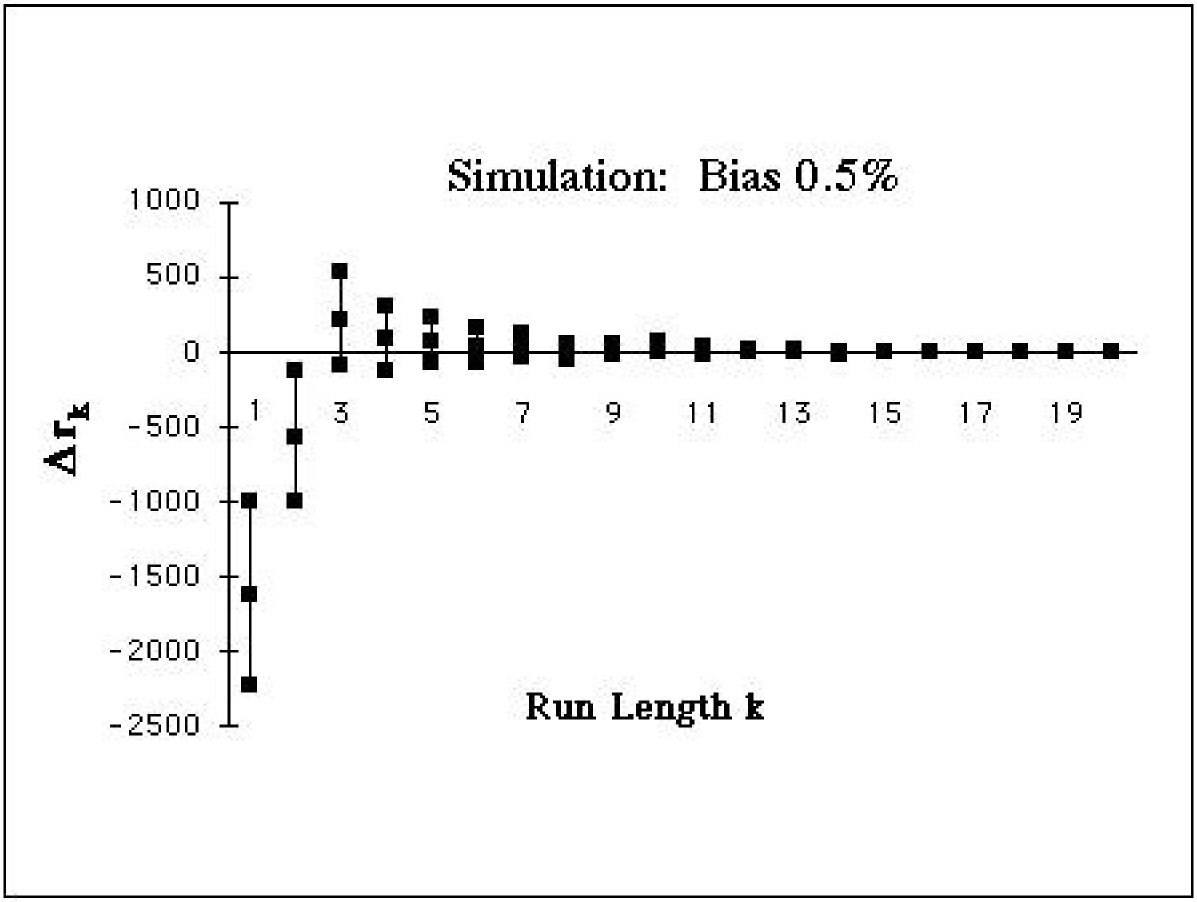


^{137}Cs
0.96
0.95
p_{00}
2
6
10
14
18
m



^{137}Cs
0.048
0.047
0.046
0.045
p_{01}
2
6
10
14
18
m

Figure 7



Simulation: Bias 0.5%
1000
500
0
-500
-1000
-1500
-2000
-2500
Δr_k
1
3
5
7
9
11
13
15
17
19
Run Length k