2014

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Published in Europhysics Letters Vol 105, No 2 (2014) 22001 (1-5)

Theory of nuclear half-life determination by statistical sampling

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PACS 21.10.Tg Nuclear structure: Lifetimes, widths
PACS 29.85.Fj Nuclear data analysis
PACS 29.87.+g Nuclear data compilation

ABSTRACT

A remarkable method for measuring half-lives of radioactive nuclei was proposed several years ago that entailed statistical sampling of the source activity. A histogram of half-life estimates, calculated from *pairs* of activity measurements separated in time, took the unexpected form of a nearly perfect Cauchy distribution, the midpoint of which corresponded very closely to the true value of the half-life. No theoretical justification of the method was given. In this article I derive the exact probability density function (pdf) of the two-point half-life estimates, show how (and under what conditions) a Cauchy distribution emerges from the exact pdf—which, mathematically, shows no resemblance to a Cauchy function—and discuss the utility of the statistical sampling method. The analysis shows that the exact pdf, under the conditions leading to an empirical Cauchy lineshape, is an unbiased estimator of the true half-life.

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1. Introduction. –

The transition rate λ or half-life τ is one of the most frequently sought characteristics of an unstable system of which examples abound in the transmutation of radioactive nuclei, the de-excitation of atoms and molecules, the decay of elementary particles, and other stochastic processes statistically referred to as "pure birth" processes [1]. With applications to nuclear physics in mind, I shall refer to the transition as a decay. For systems exhibiting exponential decay, the transition rate and half-life are related by

$$\tau = \ln 2/\lambda \,. \tag{1}$$

In exponential decay, the theoretical rate of population loss is proportional to the instantaneous population size $N(t)$

$$\frac{dN(t)}{dt} = -\lambda N(t) \tag{2}$$

with λ the constant of proportionality. The solution to Eq (2) takes the well-known form

$$N(t) = N_0 e^{-\lambda t} \tag{3}$$

in which N_0 is the initial population. The half-life τ is the time at which the population has decreased to half its size and, by this definition, Eq (1) follows directly from Eq (3).

A standard experimental procedure for determining λ (and therefore τ) is to record over a sufficiently long period of time $t_n = n\Delta t$ the number of decays occurring within a succession of much smaller counting intervals Δt. The theoretical number of counts C_n recorded in the n^{th} time interval, assuming an ideal detector, is given by the relation

$$C_n = -\frac{dN(t_n)}{dt}\Delta t = \left(\lambda N_0 e^{-n\lambda\Delta t}\right)\Delta t \equiv A_n \Delta t \tag{4}$$

in which A_n is termed the activity of the source. One usually estimates the parameter λ (e.g. by maximum likelihood or least squares [2]) from linear regression of a plot of $\ln C_n$ or a non-linear (exponential) fit to a plot of C_n as a function of t_n. As a subsidiary point, it is worth noting that the two fits can lead to different values of λ because $\ln C_n$ attributes relatively greater weight to points in the tail of the decay curve.

Empirically, the number of counts C_n, and therefore the activity A_n, recorded in the n^{th} interval Δt is well represented by a Poisson random variable [3] $Poi(\mu_n)$ of mean

$$\mu_n = \mu_0 e^{-\lambda t_n} = (A_0\,\Delta t)e^{-n\lambda\Delta t} \;. \tag{5}$$

In all rigor, C_n is a time-varying binomial random variable $Bin(N,p)$ in which the probability p of a single decay within a short sampling interval Δt is $p = \lambda\Delta t$. Under conditions $p << 1$, $N >> 1$, $C_n << N$—i.e. a large number of nuclei observed for a time short compared to their half-life—the distribution $Bin(N,p)$ is very well approximated by $Poi(\mu)$ with $\mu = Np$. Under the additional condition $\mu >> 1$ of a high mean count per Δt, the distribution $Poi(\mu)$ is well approximated by a normal (i.e. Gaussian) distribution $N(\mu,\sigma^2)$ with variance equal to the mean: $\sigma^2 = \mu$.

In contrast to the standard procedure outlined above, a novel—if not mystifying—method based on statistical sampling was described in a 2007 conference proceedings [4].

The steps of the procedure, applied to a time series of n sequential activity measurements, $A_i \ (i = 1 \ldots n)$, may be summarized as follows:

- For each pair of activities (A_i, A_j) measured at times $t_j > t_i$ where $(i = 1 \ldots n-1;\ j = i+1 \ldots n)$, calculate the two-point half-life estimate derived from Eqs (1) and (4)

$$\tau_{ij} = \frac{t_{ij} \ln 2}{\ln\left(A_i / A_j\right)} \qquad \left(t_{ij} \equiv t_j - t_i > 0\right). \tag{6}$$

- Make a histogram of the $n_t = n(n-1)/2$ two-point estimates given by Eq (6).[2]
- Find the location of the center of the resulting distribution; this yields an empirical estimate of the true half-life.

The mystifying feature of the procedure is that the resulting histogram resembled very closely a Cauchy distribution. I have simulated by Poisson random number generator (RNG) the conditions of an experiment to measure the half-life of $^{55}\mathrm{Fe}$ [5] and found the fit of the histogram to a Cauchy distribution so good that it nearly failed a chi-square test because the chi-square value was considerably *smaller* than the number of degrees of freedom. Yet an examination of expression (6) which depends on the reciprocal of the logarithm of the ratio of two Poisson variates as expressed by (4), does not in any obvious way suggest that the random variable T (upper-case tau) whose realization is the set of values τ_{ij} follows a Cauchy distribution.

The author of Ref. [4] identified the Cauchy form of his histogram visually [6], but provided no theoretical analysis of its origin. In this paper, I derive the exact probability density function (pdf) of the two-point half-life estimate T, show how the Cauchy distribution emerges from the exact theory, and discuss the applicability of the method for measuring half-lives of radionuclides or other unstable quantum states.

2. Theoretical basis of the statistical sampling method –

Derivation of the pdf $p_{\mathrm{T}}(\tau)$ of the random variable T entails finding the pdf of a succession of transformed random variables of the form

$$\left.\begin{aligned} &Z_1\left(X(\tau), Y(\tau)\right) = X(\tau)/Y(\tau) \\ &Z_2(\tau) = \ln\left(Z_1(\tau)\right) \\ &Z_3(\tau) = Z_2^{-1}(\tau) \end{aligned}\right\} \tag{7}$$

where

$$\left.\begin{aligned} X &= Poi(\mu_X) \approx N(\mu_X, \mu_X) \\ Y &= Poi(\mu_Y) \approx N(\mu_Y, \mu_Y) \end{aligned}\right\} \tag{8}$$

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[2] Since (A_i, A_j) are Poisson random variables, there may occur instances, contrary to Eq (4), in which $A_j > A_i$ even though $t_j > t_i$. In such cases, the pair (A_i, A_j) is not included because it would lead to a negative two-point estimate of the half-life.

are independent Poisson variates subject to the experimental condition of high mean count per Δt. Applications of the theory of distributions of composite measurements [7][8] to variates (7) and (8), in which successive transformations of a pdf $p_{Z_\alpha}(z_\alpha)$ to pdf $p_{Z_\beta}(z_\beta)$ with transformation relations $z_\alpha = f(z_\beta)$ and $z_\beta = f^{-1}(z_\alpha)$, take the general form

$$p_{Z_\beta}(z_\beta) = p_{Z_\alpha}(z_\alpha)\left|\frac{dz_\alpha}{dz_\beta}\right| = p_{Z_\alpha}\left(f(z_\beta)\right)\left|\frac{df(z_\beta)}{dz_\beta}\right|, \qquad (9)$$

lead to the exact expression

$$p_T(\tau) = \frac{1}{n(n-1)/2}\frac{\ln 2}{\sqrt{2\pi}}\sum_{i=1}^{n-1}\sum_{j=i+1}^{n}\frac{\left(\frac{t_{ij}}{\tau^2}\right)\exp\left(\frac{t_{ij}\ln 2}{\tau}\right)}{\sigma_{ij}}\exp\left\{-\left(e^{\left(\frac{t_{ij}\ln 2}{\tau}\right)} - \frac{\mu_i}{\mu_j}\right)^2 \Big/ 2\sigma_{ij}^2\right\} \qquad (10)$$

with

$$\sigma_{ij}^2 = \frac{\mu_i}{\mu_j^2}\left(1 + \frac{\mu_i}{\mu_j}\right). \qquad (11)$$

Substitution of Eq (5) yields the specific time dependence of the ratio

$$\mu_i/\mu_j = \exp\left(t_{ij}\ln 2/\tau\right) \qquad (12)$$

and variance

$$\sigma_{ij}^2 = \mu_0^{-1}\exp\left(\frac{t_j \ln 2}{\tau}\right)\exp\left(\frac{t_{ij}\ln 2}{\tau}\right)\left(1 + \exp\left(\frac{t_{ij}\ln 2}{\tau}\right)\right) \qquad (13)$$

where μ_0 is the initial mean number of counts per unit sampling interval.

The basic structure of Eq (10) can be understood as follows. The first factor normalizes the pdf to unit area when integrated over τ. The numerator of the second factor is the constant relating decay rate and half-life; the denominator comes from a Gaussian normalization constant. The sums are over all pairs of observations such that $t_{ij} > 0$ and $\tau_{ij} > 0$. The exponential form $\exp\left(t_{ij}\ln 2/\tau\right)$ is the functional relation $Z_1(\tau)$ applied to the $(i\,j)^{th}$ variate

$$Z_1(A_i, A_j) \equiv A_i(\tau)/A_j(\tau) = N\left(\mu_i/\mu_j, \sigma_{ij}^2\right). \qquad (14)$$

The numerator of the rational expression within the sums comprises factors from the Jacobian function $\left|dZ_1(A_i, A_j)/d\tau\right|$. The final exponential factor comes from the Gaussian distribution signified by relation (14).

Equation (10) bears no resemblance to a Cauchy distribution. To see how this extraordinary evolution unfolds, I will remove all inessential factors from Eq (10) and express time in units $t_i = i\Delta t$ with $\Delta t = 1$ and $\hat{\tau} \propto \lambda^{-1}$ the sought-for true value of the half-life. The following conditions are then imposed:

(a) τ and $\hat{\tau}$ are long compared to the intervals $(j - i)$.

(b) The source is strong: $\mu_0 >> 1$.

(c) Numerous measurements are made: $n >> 1$.

The immediate consequence of condition (1) is that $\sigma_{ij}^2 \approx 2\mu_0^{-1}$ for all pairs of activity measurements, whereupon Eq (10) takes the basic form (apart from constant factors)

$$f(\tau)=\sum_{i=1}^{n-1}\sum_{j=i+1}^{n}\left(\frac{j-i}{\tau^2}\right)e^{\left(\frac{j-i}{\tau}\right)}\exp\left\{-\mu_0\left(e^{\left(\frac{j-i}{\tau}\right)}-e^{\left(\frac{j-i}{\hat{\tau}}\right)}\right)^2\right\}. \tag{15}$$

Next, condition (1) permits a Taylor-series expansion of exponentials in Eq (15) to first order, leading to the approximation

$$e^{(j-i)/\tau}-e^{(j-i)/\hat{\tau}}\approx(j-i)\left(\tau^{-1}-\hat{\tau}^{-1}\right). \tag{16}$$

The better conditions (2) and (3) are met, the narrower is the resulting lineshape, in which case the difference of reciprocals in relation (16) can be approximated by

$$\left(\tau^{-1}-\hat{\tau}^{-1}\right)=\frac{\hat{\tau}-\tau}{\tau\hat{\tau}}\approx\frac{\hat{\tau}-\tau}{\hat{\tau}^2}, \tag{17}$$

and one can also replace the variable τ by the constant $\hat{\tau}$ in the denominator of pre-factors. At this point, Eq. (15) has been transformed into a sum of Gaussians

$$f(\tau)\approx\sum_{i=1}^{n-1}\sum_{j=i+1}^{n}\left(\frac{j-i}{\hat{\tau}^2}\right)e^{\left(\frac{j-i}{\hat{\tau}}\right)}\exp\left\{-\frac{\mu_0}{\hat{\tau}^4}(j-i)^2(\tau-\hat{\tau})^2\right\}. \tag{18}$$

Because the exponential falls off rapidly outside a narrow interval centered on $\hat{\tau}$ and has an argument smaller than 1 close to $\hat{\tau}$, one can further approximate Eq (18) by a Taylor series expansion to first order

$$\exp\left\{-\frac{\mu_0}{\hat{\tau}^4}(j-i)^2(\tau-\hat{\tau})^2\right\}=\frac{1}{\exp\left\{\frac{\mu_0}{\hat{\tau}^4}(j-i)^2(\tau-\hat{\tau})^2\right\}}\approx\frac{1}{1+\frac{\mu_0}{\hat{\tau}^4}(j-i)^2(\tau-\hat{\tau})^2}, \tag{19}$$

which, apart from a normalization factor, has the form of a Cauchy function

$$f_C(\tau)=\frac{1}{\pi\gamma\left(1+\left((\tau-\hat{\tau})/\gamma\right)^2\right)} \tag{20}$$

with location parameter $\hat{\tau}$ and scale parameter γ.

Thus, to this point the exact Eq (10) has been transformed into a sum of Cauchy functions of different scale parameters all centered on the true half-life

$$f(\tau)\approx\sum_{i=1}^{n-1}\sum_{j=i+1}^{n}\frac{\left(\frac{j-i}{\hat{\tau}^2}\right)e^{\left(\frac{j-i}{\hat{\tau}}\right)}}{1+\frac{\mu_0}{\hat{\tau}^4}(j-i)^2(\tau-\hat{\tau})^2}\ . \tag{21}$$

Thus, $f(\tau)$ is an unbiased estimator of $\hat{\tau}$.

If the time-dependent, but non-resonant, exponential in the numerator can be ignored—a step that computer analysis confirmed to have little consequence—one can approximate the variable quantities $(j-i)$, $(j-i)^2$ in some judicious way (e.g. by their means) to collapse the double sum of relation (21) into a single Cauchy function (20) centered on $\hat{\tau}$ with approximate scale parameter

$$\hat{\gamma}=\frac{6\sqrt{\pi}\,\hat{\tau}^2}{\pi\,n\ln 2\sqrt{\mu_0}} \tag{22}$$

upon restoration of the original physical constants.

3. Simulated tests of the statistical sampling theory –

The greater the number n of samples (which, by virtue of the relation $t_i = i\,\Delta t\ (i = 1\ldots n)$, also measures the total duration of sampling) and the more active the source, the narrower is the width (22) of the distribution, and the better the empirical Cauchy pdf matches the theoretically exact pdf (10). As a test and illustration of these features, I used a Poisson RNG to simulate various time sequences of activities of a hypothetical nucleus of true half-life $\hat{\tau} = 1000\ \Delta t$. A nuclear system corresponding to the illustration would be $^{55}\mathrm{Fe}$, which decays to $^{55}\mathrm{Mn}$ by orbital electron capture with a half-life of approximately 1000 days. (In the following examples, therefore, one can set Δt = 1 day.)

Figure 1 shows the normalized frequency distribution (points) of two-point half-life estimates resulting from computer simulation of 300 sequential measurements of activity from a source with initial activity $10^6/\Delta t$. Superposed on the empirical distribution is the corresponding exact theoretical lineshape (solid line) calculated from Eq (10). Agreement between experiment and theory is virtually perfect. At this stage, no approximations have been made, apart from relations (8).

Figure 2A shows the variation in theoretical lineshape for fixed sample size of 100 activity measurements as a function of initial activity. The greater the activity of the source, the more precisely the center of the distribution can be located and therefore the true half-life be estimated. Figure 2B illustrates the variation in lineshape for fixed initial activity $10^6/\Delta t$ as a function of sample size (and therefore total duration of counting). The larger the sample size, the narrower and more bilaterally symmetric the lineshape becomes and the more closely centered it is on the true half-life $\hat{\tau}$.

Figure 3 shows a histogram of 200 simulated measurements of activity from a source with initial activity $10^6/\Delta t$ and compares the exact theoretical distribution (solid line) with a least-squares-fit (LS) Cauchy distribution (dashed line). The LS location parameter $\tau_0 = 998.7\,\Delta t$ estimates the true half-life $\hat{\tau} = 1000\ \Delta t$ to within 0.13%. The LS scale parameter $\gamma_0 = 25.7\,\Delta t$ corresponds closely to the theoretical estimate (22) $\hat{\gamma} = 24.1\,\Delta t$. (The uncertainty of LS estimates can be deduced by standard statistical analysis and is a routine matter outside the scope of this paper.) A chi-square test of the fit to the histogram yielded $\chi^2_{996} = 984.2$ with P-value $\Pr\left(\chi^2_{996} > 984.2\right) = 0.599$.

Thus, for statistical purposes the empirical and theoretical distributions of the two-point half-life estimates are seen to be well represented by a Cauchy distribution under the conditions assumed in the analysis leading from Eq (10) to Eq (21).

4. Conclusions –

The method of statistical sampling provides an operationally simple means to measure half-lives of unstable quantum systems such as radioactive nuclei. The essential content of this paper was (1) a derivation of the theoretical pdf of two-point half-life estimates based on pairs of activity measurements, and (2) the demonstration that this pdf becomes statistically equivalent to a Cauchy distribution when the following experimental conditions are met: (a) the number of decays per sampling interval is sufficiently high; (b) the number of sequential activity measurements is sufficiently large ($n > 100$ suffices); (c) the half-life is sufficiently long compared to time intervals between pairs of samples.

In experiments on long-lived radionuclides, statistical sampling can have a significant advantage in that sampling need not be done at regular time intervals because a histogram displays only frequencies of occurrence of events and not their time-ordering. In contrast to performing regression analysis on a decay curve, the statistical sampling method yields an estimate of the half-life directly from the center point of the histogram or location parameter of the best-fit Cauchy function. Moreover, as shown in Figure 3, accurate estimates of the true half-life can be obtained even when the total duration of sampling is a relatively small fraction (20% in Figure 3) of the half-life.

In recent experiments that refuted controversial published claims of correlations in nuclear decay [9], the lifetime of β^+ emitter $^{22}\mathrm{Na}$ with half-life ~ 950 d and activity $A \sim 400\ \mathrm{s}^{-1}$ was determined (together with other statistical quantities) from activities sampled in time windows of approximately 0.5 s, extending uninterruptedly for 167 h. Disruption in continuous sampling (e.g. by power failure) would significantly impact an experiment of this kind. Computer simulated statistical sampling with LS fit to a Cauchy profile led to estimates of the $^{22}\mathrm{Na}$ half-life accurate to within 0.5% from activity samples taken just 2 h/day over a period of only 60 d (i.e. 6.3% of the half-life).

The uncertainties encountered in the method of statistical sampling are complementary to those involved in regression of a decay curve. In matters where different measurements of half-life by the standard method do not agree, statistical sampling, which generates an unbiased estimator of the true half-life under the conditions analyzed in this paper, provides an independent means of resolving the ambiguity.

An extension, currently in progress, of the present research is to ascertain whether half-life measurement by statistical sampling is useful when the observed activity does not follow an exponential decay law. This situation can arise if (A) more than one exponential decay process with different decay rates contribute to the activity, or (B) if an elementary decay process itself deviates from strict exponential decay, as predicted by quantum mechanics [10] for time periods very short or very long relative to the half-life. Recent studies have either predicted or searched for oscillatory decay over intermediate periods [9], [11], [12]. In case (A) preliminary results of computer simulation showed that the histogram of two-point half-life estimates deviates significantly in location and shape from a uniform-decay Cauchy distribution even when one period greatly exceeds the other. Simulations of case (B) in which λ manifests a weak oscillatory time dependence $\lambda/\ln 2 = \tau_0^{-1}\left[1+\alpha\cos\left(2\pi\tau_1^{-1}\right)\right]$ led to detectably asymmetric histograms for $\alpha \ll 1$, $n > 100$, and τ_1 either larger or smaller than τ_0. The feasibility of statistical sampling to search for non-exponential nuclear decay is still under investigation and will be reported in detail when completed.

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FIGURE CAPTIONS

Figure 1: Plot of theoretical probability $p(\tau)d\tau$ (solid line) as a function of half-life τ for a series of 300 measurements of the activity of a hypothetical radionuclide of true half-life $\hat{\tau} = 1000\,\Delta t$ and initial activity $A_0 = 10^6$ per interval Δt. Superposed points mark the normalized frequencies of half-lives obtained from 300 measurements of activities simulated by a Poisson RNG set for initial activity A_0. The number of two-point samples is 44850. Bin width is $d\tau = 2\ \Delta t$.

Figure 2: Variation in half-life probability functions with activity A and sample size n for a hypothetical radionuclide with true half-life $\hat{\tau} = 1000\,\Delta t$. (A) Fixed sample size of 100 measurements with initial activities (a) 10^5, (b) 10^6, (c) 10^7 per Δt. The number of two-point samples is 4950. (B) Fixed initial activity of 10^6 per Δt for sample sizes of (a) 100, (b) 200, (c) 300 measurements. Respective numbers of two-point samples are (a) 4950, (b) 19900, (c) 44850. Bin width for both plots is $2\ \Delta t$.

Figure 3: Histogram of the distribution of half-life values obtained from 200 activity measurements of a hypothetical radionuclide of theoretical half-life $\hat{\tau} = 1000\ \Delta t$ and initial activity $A_0 = 10^6$ per Δt. The number of two-point samples is 19900. Superposed on the histogram are the exact theoretical probability function (solid line) and empirical Cauchy distribution (dashed line) with least-squares parameters $\tau_0 = 998.7\ \Delta t$, $\gamma_0 = 25.7\ \Delta t$. A chi-square test of the fit yielded $\chi^2_{996} = 984.2$ with $P = 0.599$. Bin width is $4\,\Delta t$.

Figure 1

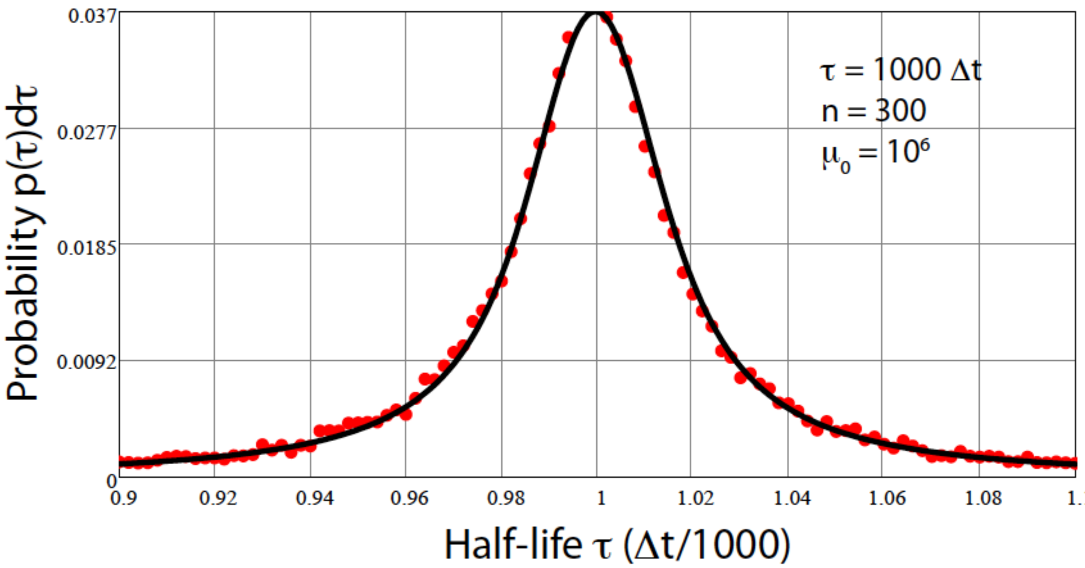

τ = 1000 Δt
n = 300
μ0 = 10^6
Probability p(τ)dτ
Half-life τ (Δt/1000)
0.037
0.0277
0.0185
0.0092
0
0.9
0.92
0.94
0.96
0.98
1
1.02
1.04
1.06
1.08
1.1

Figure 2

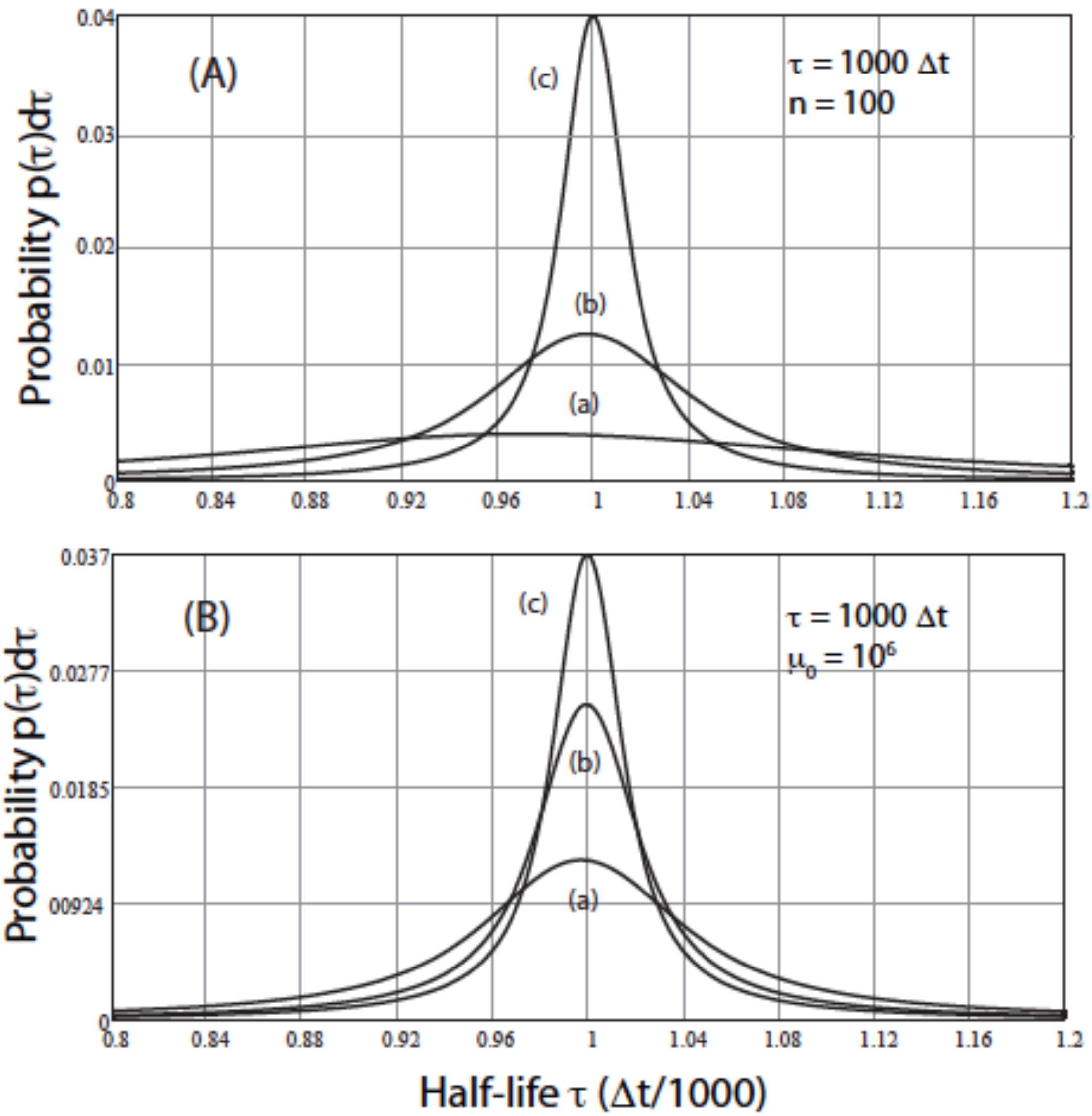
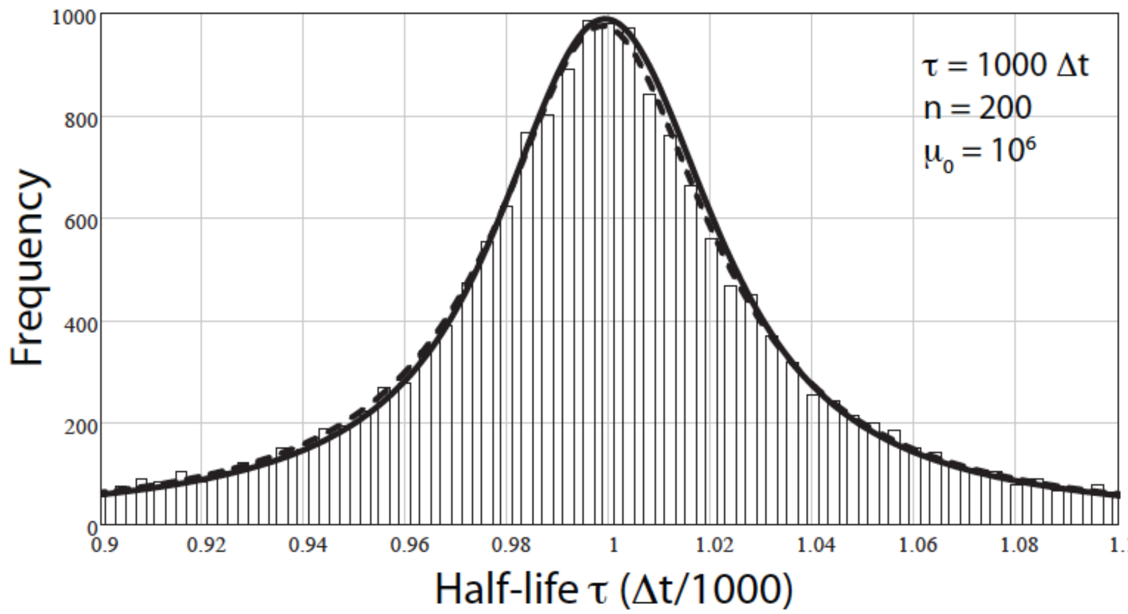

(A)
τ = 1000 Δt
n = 100
(a)
(b)
(c)
Probability p(τ)dτ
0
0.01
0.02
0.03
0.04
(B)
τ = 1000 Δt
μ0 = 10^6
(a)
(b)
(c)
Probability p(τ)dτ
0
00924
0.0185
0.0277
0.037
0.8
0.84
0.88
0.92
0.96
1
1.04
1.08
1.12
1.16
1.2
Half-life τ (Δt/1000)

Figure 3


τ = 1000 Δt
n = 200
$\mu_0 = 10^6$
Frequency
Half-life τ (Δt/1000)
1000
800
600
400
200
0
0.9
0.92
0.94
0.96
0.98
1
1.02
1.04
1.06
1.08
1.1