Fluctuations in radioactive decays. I. Nonequilibrium effects and noise

Roberto Boscaino and Giorgio Concas
Dipartimento di Scienze Fisiche, Università di Cagliari, via Ospedale 72, I-09124 Cagliari, Italy

Marcello Lissia
Istituto Nazionale di Fisica Nucleare, via Negri 18, I-09127 Cagliari, Italy

Sergio Serci
Dipartimento di Scienze Fisiche, Università di Cagliari, via Ospedale 72, I-09124 Cagliari, Italy and Istituto Nazionale di Fisica Nucleare, via Negri 18, I-09127 Cagliari, Italy
(Received 2 August 1993)

Radioactive decay is a nonequilibrium process. For most practical purposes it can be modeled as a steady-state process, but there are instances where counting statistics should not be assumed constant; this remark is particularly relevant for long counting intervals. We give explicit formulas for the probability distribution of counts, the expected values of the mean, the variance, and the Allan variance. These formulas, rather than the Poissonian ones, should be used as benchmark for experiments aimed at revealing exotic effects. Formulas are compared to numerical simulations and actual experiments.

PACS number(s): 05.40.+j, 02.50.−r, 23.90.+w

I. INTRODUCTION

The essential presence of $1/f$ noise in electronic devices, and in other diverse physical systems [1], has inspired several studies of fluctuations in radioactive decays with the aim of detecting the same phenomenon, or other departures from Poissonian statistics. At present existing experimental data are not easily reconciled: several authors confirm the Poissonian behavior [2–7], while others find variances higher than the Poisson value by more than one order of magnitude [8–13]. Following the original papers by Handel [14–17], several mechanisms have been considered to explain the excess variance [1,11,18], but a unique interpretation of the experimental data is missing at present [1,19].

The importance of such studies is best understood by reminding the reader of the basic assumptions that determine the Poissonian nature of the counting statistics: the probability per unit time for the decay of a nucleus is a constant, and the decay of each nucleus of the ensemble is independent of the decay of its neighbors. The first assumption, which is equivalent to exponentially decaying amplitudes for unstable states, agrees with available experimental data from unstable nuclei and particles. However, one cannot avoid noticing that the amount of data available is not large, perhaps because a precise determination of the time dependence of the decay probability over long intervals is by no means trivial. The second assumption might also fail. For instance, coherent effects among the nuclei in the lattice might introduce correlations among decays. In this regard, even if outside the scope of the present work, we suggest the interesting possibility of studying counting statistics in radioactive decays as a means of getting information on such coherent effects.

The main purpose of this paper is to spell out what we like to call the standard picture for decay fluctuations: deviations from this picture would indicate interesting new phenomena. We show that the time dependence of the decay statistics produces sizable deviations from a pure time independent Poisson distribution for definite ranges of parameters; these deviations are well understood and should not be interpreted as novel effects. This no-new-physics result is then compared to several experimental studies of fluctuations in radioactive decays which we find particularly instructive.

By not taking correctly into account the time dependence of the statistics, one could be misled into interpreting as $1/f$ noise, or more exotic physics, deviations from the Poissonian statistics, which are mere consequences of the exponential drift. Moreover, the understanding of the dependence of the phenomenon on the experimental parameters (basically mean count per unit time and mean life of the nucleus) is essential in comparing experiments, which are in general run at different counting rates: our analysis yields formulas that depend on the counting rate, while in the framework of Poissonian distribution results are independent of the counting rates.

The main conclusions of this paper have already been anticipated in a previous work [7] where we reported our preliminary experimental results for the γ decay of ${}^{119m}_{50}\mathrm{Sn}$. Here we wish to give a more careful derivation of the formulas, discuss some of their consequences, and compare them to several published results. In a companion paper [20], which we shall refer to as II, we shall report our complete experimental data for the γ decay of ${}^{119m}_{50}\mathrm{Sn}$.

In Sec. II we give an intuitive description of the effect, estimate the leading corrections to the Poissonian expected values of the the mean counting, and give the

Allan variance [21,22] over n consecutive intervals. In Sec. III we report numerical simulations that estimate statistical uncertainties of measurements and systematic errors due to data treatment and compare them to published data. Finally, we draw our conclusions in Sec. IV. In the Appendix we derive our formulas within a more rigorous framework: we find that, for all practical purposes, results do not differ from the simpler calculation in Sec. II. In the Appendix we also report formulas for the probability distributions of counts and for the usual variance over n consecutive intervals.

II. TIME DEPENDENT STATISTICS

When decay statistics are time dependent, we expect sizable deviations from pure time independent Poisson distributions for definite ranges of parameters. We shall illustrate the reason of these deviations and describe the two physical effects present in these kind of processes: the statistical fluctuation and the exponential decay. In the Appendix we report a more rigorous approach that yields results identical to the leading order to the ones obtained in this section.

Let us consider an ensemble of identical nuclei with mean life τ and call M_k the number of decays recorded in the time interval between kT and $(k+1)T$ for $k = 0, 1, \ldots, n-1$. The statistics we shall presently consider are the average count over n consecutive intervals:

$$\overline{M}_n \equiv \frac{1}{n}\sum_{k=0}^{n-1} M_k \tag{1}$$

and the corresponding Allan variance [21,22]

$$A_n^2 \equiv \frac{1}{2(n-1)}\sum_{k=0}^{n-2}[M_k - M_{k+1}]^2 \quad . \tag{2}$$

We also define a new variance

$$G_n^2(j) \equiv \frac{1}{2(n-j)}\sum_{k=0}^{n-1-j}[M_k - M_{k+j}]^2 \tag{3}$$

which we call generalized Allan variance since it includes Eq. (2) as a particular case for $j = 1$. We introduce this generalized Allan variance because we find it a very sensitive tool to study drifts in the counting statistics. Moreover, the dependence on j^2 of the generalized Allan variance, which we shall show in Eq. (7), provides an additional test of our calculation. We shall come back to these remarks in II, where we present our experimental data [7,20]. Notice that in Eqs. (1)–(3) we should write $\overline{M}_n(T)$, $A_n^2(T)$, and $G_n^2(T,j)$, but we prefer not to show explicitly that these quantities depend on T to make the notation less cumbersome.

The heuristic derivation we present in this section is based on the physically motivated knowledge of the exact results in the two opposite limits: small and large T. For small counting periods T the difference of the average countings of two contiguous periods is of order T, the leading term when we expand the exponential that describes the decay, while the fluctuations around the average values are of order $\sqrt{T}$: therefore fluctuations dominate at small T. It follows that the expectation value of M_k is constant

$$\mu_n \equiv E[\overline{M}_n] = E[M_k] = E[M_0] \quad .$$

Moreover for Poissonian fluctuations, i.e., $E[M_j M_k] = \delta_{jk}E[M_k] = \mu_n$, the expectation value $\alpha_n^2(j)$ of the generalized Allan variance, defined by Eq. (3), is

$$\begin{aligned}\alpha_n^2(j) &= \frac{1}{2(n-j)}\sum_{k=0}^{n-1-j}\{E[(M_k - M_{k+j})^2]\}\\ &= \frac{1}{2(n-j)}\sum_{k=0}^{n-1-j}[\mu_n + \mu_n] = \mu_n \quad ,\end{aligned}$$

which yields the Poissonian result for the reduced Allan variance:

$$R \equiv \frac{\alpha_n^2(1)}{\mu_n^2} = \frac{1}{\mu_n} \quad . \tag{4}$$

Conversely, differences between contiguous periods are dominated by the exponential decay at long enough T, for the same reason for which Poissonian fluctuations dominate at short T: fluctuations grow like $\sqrt{T}$ while the exponential decay produces differences of order T. In this limit, then, each measure gives the same result, which is also the expected value, and we need only to take into account that the number of nuclei is decaying exponentially:

$$E[M_k] = M_0 \exp[-k\lambda T] = M_0(1-x)^k \quad ,$$

where $x \equiv 1 - \exp[-\lambda T] \approx \lambda T$ has been defined, with $\lambda = 1/\tau$. Therefore, our statistics are simply obtained by summing up geometric series and keeping the leading terms in the small parameter x:

$$\begin{aligned}\mu_n &= M_0\frac{1}{n}\sum_{k=0}^{n-1}(1-x)^k = M_0\frac{1-(1-x)^n}{nx}\\ &= M_0\left[1 - \frac{n-1}{2}x + O(x^2)\right] \quad ,\end{aligned} \tag{5}$$

$$\begin{aligned}\alpha_n^2(j) &= \frac{M_0^2}{2(n-j)}\sum_{k=0}^{n-1-j}[(1-x)^k - (1-x)^{k+j}]^2\\ &= \frac{M_0^2}{2(n-j)}[1-(1-x)^j]^2\frac{1-(1-x)^{2(n-j)}}{1-(1-x)^2}\\ &= \frac{M_0^2}{2}(jx)^2[1-(n-2)x + O(x^2)]\\ &= \frac{\mu_n^2}{2}(jx)^2[1 + x + O(x^2)] \ .\end{aligned} \tag{6}$$

Finally, we add the two contributions, the one due to fluctuations and the one due to the exponential decay; to leading order in x we find

$$\alpha_n^2(j) = \mu_n + \frac{j^2}{2(\rho_n\tau)^2}\mu_n^4 = \rho_n\tau x + j^2\frac{(\rho_n\tau)^2}{2}x^4 \quad , \quad (7)$$

$$R_G(j) \equiv \frac{\alpha_n^2(j)}{\mu_n^2} = \frac{1}{\mu_n} + \frac{j^2}{2(\rho_n\tau)^2}\mu_n^2 = \frac{1}{\rho_n\tau x} + \frac{j^2}{2}x^2 \quad ; \quad (8)$$

in particular the reduced Allan variance is

$$R = \frac{1}{\mu_n} + \frac{1}{2(\rho_n\tau)^2}\mu_n^2 = \frac{1}{\rho_n\tau x} + \frac{1}{2}x^2 \quad . \quad (9)$$

Since it is common practice to plot fluctuations against the average count $\overline{M}_n$ or the time interval T, we presented Eqs. (7)–(9) as function of μ_n, the expectation value of $\overline{M}_n$, and of $x \approx \lambda T$; to this end, we introduced $\rho_n \equiv \mu_n/T \approx \mu_n/(\tau x)$, the expected value of the average rate $r_n \equiv \overline{M}_n/T$. Note that interchanging any or all of the measured values $\overline{M}_n$, $A_n^2(j)$, and r_n with the corresponding expected value μ_n, $\alpha_n^2(j)$, or ρ_n does not modify results to leading order in x.

At this point we wish to remark that, while Eqs. (7)–(9) have the correct behavior for small and large T, they could have in principle correction terms at intermediate T. The incoherent sum of the decay and Poissonian contributions could miss, when the sizes of the two terms are comparable, some cooperative effect. Nonetheless, the most interesting qualitative feature of Eqs. (7)–(9), i.e., the presence of a minimum, is already guaranteed by our knowing the exact limiting behaviors: the variance grows both at small and large T. It turns out that the more rigorous and lengthy treatment reported in the Appendix does not add any new term to Eqs. (7)–(9), i.e., which therefore remain valid for every T, even outside those limits within which they were originally derived. The second contribution due to the exponential drift of the mean count can often be dropped compared to the first one because of the smallness of $x \approx \lambda T$; however, if we take T large enough, it will eventually become dominant, even for times T much shorter than the mean life τ. This is a major point of this paper, as this effect has often been underestimated. In this regard, we contrast the expression of R given by Eq. (9) with the one reported in Ref. [3]

$$R = \frac{1}{\mu_n}(1 + \tfrac{1}{2}x)$$

and with the one reported in Ref. [10]

$$R = \frac{1}{\mu_n}(1 + \tfrac{1}{2}x^2) \quad .$$

In both these expressions, it is always possible to drop the correction term for times T much shorter than the mean life τ. In our Eq. (9) for the Allan variance the two terms become comparable for $T \approx T_{\min}$, which is the value of T for which Eq. (9) has its minimum:

$$R_{\min} = \frac{3}{2}(\rho_n\tau)^{-\frac{2}{3}} \quad \text{at} \quad T_{\min} = \tau(\rho_n\tau)^{-\frac{1}{3}} \quad . \quad (10)$$

We wish to point out that the crossover from one regime, the one dominated by Poissonian fluctuations, to the other one, the one dominated by the exponential decay, depends on the counting rate. Experiments at different rates will see the crossover at different periods or even fail to see it depending on the range of periods considered. We suggest two possible experimental tests of the results reported in this section. One is to verify the dependence of the Allan variances on the counting rate, and in particular Eq. (10), by taking data for several rates. The other is to plot the the measured generalized Allan variance as function of j^2 and verify Eqs. (7) and (8); we report such tests in II.

III. MC SIMULATIONS AND EXISTING DATA

In this section we discuss the results of the Monte Carlo (MC) simulation of the decay of a closed system of emitter centers. MC simulations will be used here not only to confirm the analytical results but also to estimate statistical uncertainties. We point out that analytical expressions of the uncertainties could have been obtained by methods similar to the ones reported in the Appendix, even if with greater effort; nevertheless numerical methods seem more appropriate to our present purpose of evidencing general trends and getting order-of-magnitude estimates. Moreover, simulating the specific conditions in which experiments reported in the literature were carried out will help us to discuss the interpretation of the experimental results. In particular, we shall examine here a few representative experiments on (i) the γ decay of $^{119m}_{50}$Sn, investigated in II; (ii) the α decay of $^{210}_{84}$Po [5]; (iii) the β decay of $^{137}_{55}$Cs [6]; and (iv) the α decay of $^{241}_{95}$Am [4].

First, we illustrate the effect of the decreasing number of nuclei on the decay statistics and, for the sake of concreteness, we refer to the physical situation experimentally investigated in II: a source of $^{119m}_{50}$Sn with an average counting rate of 5500 counts per second, having a half-life of 293.0 days [23]. Our simulation reproduces strictly the experimental procedure. We consider N_t time intervals, multiple of a basic interval: $T_i = 2^{i-1}T_0$ with $i = 1, \ldots, N_t$. For each T_i, the statistics are calculated using the decays recorded in n consecutive intervals of duration T_i. The same value of $n = 64$ is used for every interval; we count the decay events in $n(2^{N_t} - 1)$ consecutive intervals of length T_0 and we group the counting results as follows: the first n are taken as the n measures at period $t = T_0$, the successive $2n$ are used to give the n measures at $t = 2T_0$, and so on. Therefore, our simulation consists in generating $n(2^{N_t} - 1)$ numbers from the appropriate distribution. Operatively, we start up with N nuclei and generate the number of decays in the first interval of length T_0 according to the binomial probability distribution of independent events with probability p out of a sample of N events; p is chosen so that Np is the expected number of decays. In the Appendix we argue that this is the correct distribution when no novel effects are expected. As a second step, we subtract the decayed nuclei from N and generate the number of decays in the next interval according to the new binomial probability distribution (N has changed). The iteration of the second step provides us with a set of data which corresponds to a complete experiment; parameters of the simulation, N, p, and T_0, are obviously chosen to match the rate, length of the time interval, and decay constant in the real experiment. We then repeat the simulation a given number of times (typically 64) to estimate statistical errors.

In Fig. 1 we report the results of the simulation, where data and error bars are averages and standard deviations over 64 simulations. For comparison we also show the expected result for a stationary Poissonian process (dashed line), as given by Eq. (4), and the theoretical behavior predicted by our Eq. (9), which takes into account the exponential decay (full curve). In agreement with the latter, the simulation results show the extent at which the decreasing number of nuclei is expected to affect the dependence of the counting variance at long periods: the variance decreases on increasing T, as expected for a Poisson process, until a minimum is reached at $T^{-1} = 1.6\times10^{-4}$ s^{-1} and it increases for longer values of T. The agreement between the experimental results and the simulation ones will be discussed in II. Here we emphasize that once the Allan variance is dominated by the decay, say for $T > T_{\rm min}$ with $T_{\rm min}$ given by Eq. (10), a strong reduction of its standard deviation from the average value occurs, as shown in Fig. 1.

In Figs. 2 and 3 we compare our simulations with the results reported by Azhar and Gopala [5,6]. These authors have investigated experimentally the statistics of the the α decay in $^{210}_{84}$Po [5] and of the β decay in $^{137}_{55}$Cs [6], searching for traces of $1/f$ noise in the dependence of the relative Allan variance on the counting period T. At variance to their expectations, they found that the decay of both sources obeys the simple Poisson statistics up to periods as long as 10 000 and 3300 min, respectively. Here we show that, according to the theory presented in Sec. II, a departure from the Poissonian behavior should be clearly observed in both cases.

In Fig. 2, open circles are experimental data taken from Ref. [5] on the α-particle decay of $^{210}_{84}$Po, the dashed line plots Eq. (4), valid for a stationary Poissonian process, and the full line plots Eq. (9) which takes into account the exponential decay of nuclei. We find puzzling that experimental data fail to show the effect of the decay, as this effect should dominate the statistical fluctuations at the longest period explored.

One might relate the disagreement to the circumstance that *add-up* procedures were used in data treatment in Ref. [5]. We recall that the so called *add-up* or *summing* procedures are used in most of the experimental investigations on the statistics of decay processes and aim at saving data acquisition time. They consist in using the same n counting measures taken with a period T_0 to give not only n data points at time T_0 but also $n/2$ data points at period $2T_0$ by grouping pairs of consecutive measures, $n/4$ data points at period $4T_0$ by grouping four consecutive measures, and so on. An immediate consequence of this procedure is that measures at longer and longer periods have lower and lower statistics; moreover, measures at different time intervals are not fully independent: the correlation between measures could, at least in principle, introduce systematic errors. To make clear this point, we also carried out MC simulations that make use of these procedures. The results of our simulations are reported as full circles in Fig. 2. Following Ref. [5], the add-up method was applied for periods longer than 1000 min; we used 138.367 days as the $^{210}_{84}$Po half-life [23] and estimated from their figures, since it is not explicitly reported

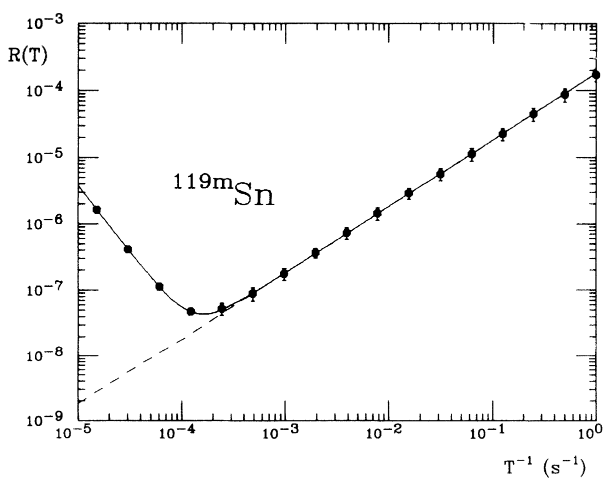

FIG. 1. Relative Allan variance vs $1/T$ for $^{119m}_{50}$Sn decay. Filled circles are averages over 64 MC simulations; error bars are the corresponding standard deviations. The dashed line corresponds to a stationary Poissonian process, described by Eq. (4), with an average counting rate of 5500 counts per second. The full line reproduces our Eq. (9) which takes into account the exponential decay with a 293.0 day half-life [23].

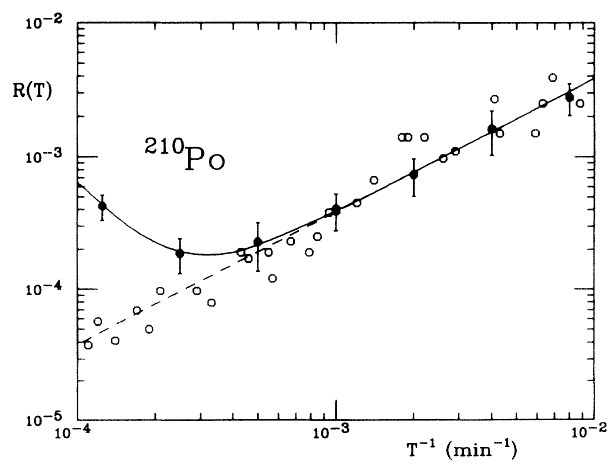

FIG. 2. Relative Allan variance vs $1/T$ for $^{210}_{84}$Po decay. Filled circles are averages over 64 MC simulations; error bars are the corresponding standard deviations. Open circles are data from Fig. 1 of Ref. [5]. The dashed line corresponds to a stationary Poissonian process, described by Eq. (4), with an average counting rate of 2.6 counts per minute. The full line reproduces our Eq. (9) that takes into account the exponential decay with a 138.367 day half-life [23].

in Ref. [5], an average rate of 2.6 counts per minute. The number of measurements for each time interval is also not reported by Ref. [5]: we made 32 measurements for periods up to 1000 min, a number estimated from the dispersion of the data, and we halved that number at each successive period so that only four measurements were made for the last time period, which was 8000 min long. We also performed 64 Monte Carlo simulations of the experiment to verify whether data were compatible with a statistical fluctuation away from the full curve or whether systematic effects due to the add-up technique might shift the data towards the dashed line; as evident in Fig. 2, the results of the simulations indicate that neither statistical fluctuations nor such a systematic effect gives any significant contribution.

In Fig. 3, we show the same kind of information shown in Fig. 2 but for the β decay of $^{137}_{55}$Cs: now open circles are experimental data taken from Ref. [6], the dashed line plots again Eq. (4), and the full line plots Eq. (9). We are again surprised that data are better fitted by Eq. (4) than by Eq. (9); MC simulations also fail to explain this result in terms of statistical fluctuations. For the sake of completeness, we report the parameters of the simulation: we estimated them from the figures of Ref. [6], since none of them are given there. We did not use any add-up method, since it is not mentioned in Ref. [6], and we made 32 measurements for each period, that number estimated from the dispersion of the data. We used 30.0 years as the $^{137}_{55}$Cs half-life [23] and an average rate of 159 600 counts per minute.

We analyze in detail one last case. The reason of its interest is that it is the only case we were able to find in the literature where the authors [4] realize that is possible to get sizable contributions to the variance from the exponential drift; they choose to take it into account by correcting the data. Figure 4 compares two runs from Ref. [4] to our simulation of the α decay of a sample of $^{241}_{95}$Am, whose half-life is 432.7 years [23], with an average counting rate of 600 000 decays per minute. Apart from the three shortest time intervals, data are taken using the add-up technique in such a way that the longest time interval has only four measures, the second longest eight measures, and so on. Correction was done by dividing out the exponential decay from the data. Averages over 64 iterations seem unaffected by systematic errors and have a Poissonian behavior, as expected after the decay contribution to the variance has been eliminated; dispersion around the average increases at longer time intervals, corresponding to the decreasing of the number of measures. Both runs from Ref. [4] are statistically compatible with a pure Poissonian behavior (dashed line).

Finally, we wish to point out that, in spite of our stressing the importance of contributions to the variance coming from the decay, there are instances when these contributions can be neglected. One of these cases is when the experimental settings are such that deviations would only appear for time much longer than the maximum interval considered: given the lifetime of the nucleus, the lower the rate the longer the maximum time interval, according to Eq. (10). We find one such case in Fig. 8 of Ref. [10]: application of Eq. (9) to the $^{241}_{95}$Am α decay with a rate of 18 000 counts per minute yields that the decay becomes important for times of the order of or longer than 18 000 min; deviations from a pure Poissonian behavior in the considered range between 1 and 10^4 min cannot be explained by the exponential drift. We can also fail to observe the contribution of the decay to the variance when the variance exceeds not only the Poissonian formula Eq. (4) but Eq. (9) as well. Nonethe-

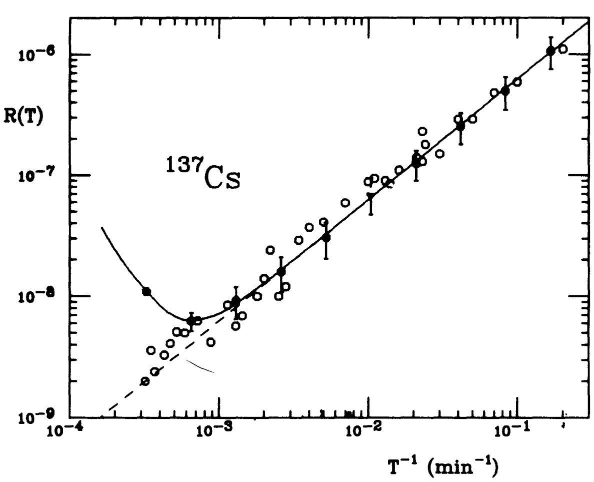

FIG. 3. Relative Allan variance vs $1/T$ for $^{137}_{55}$Cs decay. Filled circles are averages over 64 MC simulations; error bars are the corresponding standard deviations. Open circles are data from Fig. 2 of Ref. [6]. The dashed line corresponds to a stationary Poissonian process, described by Eq. (4), with an average counting rate of 159 600 counts per minute. The full line reproduces our Eq. (9) that takes into account the exponential decay with a 30.0 year half-life [23].

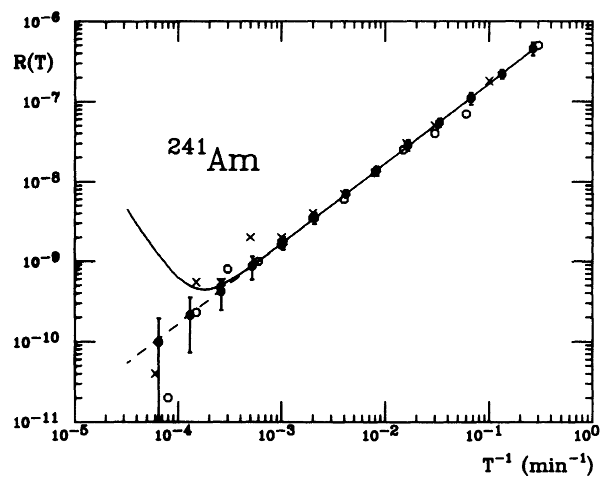

FIG. 4. Relative Allan variance vs $1/T$ for $^{241}_{95}$Am decay. Filled circles are averages over 64 MC simulations; error bars are the corresponding standard deviations. Open circles and crosses are data of, respectively, run 1 and run 2 from Fig. 5 of Ref. [4]. The dashed line corresponds to a stationary Poissonian process, described by Eq. (4), with an average counting rate of 600 000 counts per minute. The full line reproduces our Eq. (9) that takes into account the exponential decay with a 432.7 year half-life [23].

less we think it more correct to measure the excess of variance over the latter. For example, Ref. [12] reports a study of the $^{204}_{81}$Tl β decay; the rate reported implies that the decay drift becomes important at times larger than 2000 min. They observe a relative Allan variance that is, for their longer times, about twice the one given by Eq. (9), but almost an order of magnitude larger than the Poissonian result. The evidence for excess noise is only marginal, once errors are taken into account, if we correctly use Eq. (9) as our reference—evidence that would have appeared more convincing had we used Eq. (4) as reference.

IV. CONCLUSIONS

The experimental study of fluctuations of the number of radioactive decays appears to be an interesting way of gaining information about correlations in the sample or about other coherent effects that might contribute to the decay process [1]. However, fluctuations are also as sensitive to spurious correlations as the ones introduced by hardware drifts, making this kind of experiment rather delicate. In particular, we claim that the apparently small drift of the averages due to the exponential decay cannot be disregarded and, actually, such small drift can give sizable contributions to the variance. Small time intervals are dominated by Poissonian fluctuations, but long enough time intervals are dominated by the decay [7]. In this paper, we support this claim with explicit calculations and numerical simulations. We have already reported preliminary experimental evidence of this result in a previous work [7]. Our complete data analysis for the γ decay of $^{119m}_{50}$Sn shall appear in II. Since this contribution to the variance can be exactly taken into account, it does not, even when it is large, prohibit the study of smaller, but perhaps more interesting, contributions. We report a formula that includes the effect of the radioactive decay and we suggest measuring deviations from that formula. Finally, we find a few suspicious cases [5,6] in the literature that fail to see the effect of the exponential decay in spite of the fact that no correction was reported.

ACKNOWLEDGMENTS

We wish to thank A. Devoto for a critical reading of the manuscript. Financial support has been given by Italian Ministry of University and Research and by Istituto Nazionale di Fisica Nucleare.

APPENDIX

In this appendix we treat more carefully the statistics of radioactive decay. Our basic assumptions shall be that the probability of a nucleus decaying in the infinitesimal interval dt is a constant times the interval: λdt; the decay of each nucleus of the ensemble is independent of the decay of its neighbors.

From the first assumption, we can derive the probability that a given nucleus present at time t be still present at a later time $t+T$: it is the product of the probabilities that the nucleus has not decayed in any of N subinterval of length $\frac{T}{N}$, in the limit of large N

$$p(T) = \lim_{N\to\infty}\left(1-\lambda\frac{T}{N}\right)^N = \exp[-\lambda T] ,$$

which implies that the mean life of the nucleus τ is related to λ by $\tau = \frac{1}{\lambda}$. Therefore, the probability that a nucleus present at time $t=0$ decays in the time interval between t and $t+T$ is given by the product of the probabilities that the nucleus has not decayed between 0 and t and that it decays between t and $t+T$:

$$p(t,t+T) = \exp[-\lambda t]\,(1-\exp[-\lambda T]) .$$

Since we shall consider times t multiple of T, it is useful to define the probability of a nucleus decaying between kT and $(k+1)T$:

$$p_k \equiv \exp[-\lambda kT](1-\exp[-\lambda T]) = x(1-x)^k \quad , \quad \text{(A1)}$$

where we have introduced $x \equiv 1-\exp[-\lambda T]$, which is typically a small parameter $x \approx \lambda T = T/\tau$; we should keep in mind that p_k is a function of T, even if it is not shown explicitly.

Given a sample with N nuclei, our second assumption, that the decays are independent, yields that the probability of M_k of them decaying in the kth time interval is given by the binomial distribution corresponding to the possible combinations of M_k nuclei decaying and $N-M_k$ nuclei surviving:

$$\begin{aligned} P(M_k;T,N) &= \binom{N}{M_k} p_k^{M_k}[1-p_k]^{(N-M_k)}\Theta(N-M_k) \\ &= \frac{p_k^{M_k}}{M_k!}\left[\frac{d^{M_k}}{dp^{M_k}}(p+q)^N\right]_{\substack{p=0\\ q=1-p_k}} . \end{aligned}$$

Similarly, the combined probability of M_j nuclei decaying in the jth time interval and M_k of them decaying in the kth time interval is

$$P(M_j,M_k;T,N) = \frac{p_j^{M_j}}{M_j!}\frac{p_k^{M_k}}{M_k!}\left[\frac{d^{M_j}}{dp_1^{M_j}}\frac{d^{M_k}}{dp_2^{M_k}}(p_1+p_2+q)^N\right]_{\substack{p_1=0,\,p_2=0\\ q=1-p_j-p_k}} ;$$

it should now be obvious how to generalize the result to combined probabilities in more than two intervals. We should remark that, even if we correctly use the binomial distribution, it is often a very good approximation, being the number of nuclei very large, to use a Poissonian distribution if the number of expected decays in the interval is less than or of order of one or a Gaussian distribution if we expect a large number of decays.

The expected value of a stochastic variable $f_1(M_k)$, defined as a function of the number of decays M_k in the kth time interval, is

$$E[f_1(M_k)] \equiv \sum_{M_k=0}^{N} P(M_k;T,N) f_1(M_k) = \left[f_1\left(p\frac{d}{dp}\right)(p+q)^N \right]_{\substack{p=p_k\\ q=1-p_k}} . \quad \text{(A2)}$$

The expected value of a stochastic variable $f_2(M_j, M_k)$, defined as function of the numbers of decays M_j and M_k in the jth and kth time interval, is

$$E[f_2(M_j,M_k)] \equiv \sum_{\substack{M_j=0\\ M_k=0}}^{N} P(M_j,M_k;T,N) f_2(M_j,M_k) = \left[f_2\left(p_1\frac{d}{dp_1}, p_2\frac{d}{dp_2}\right)(p_1+p_2+q)^N \right]_{\substack{p_1=p_j,\, p_2=p_k\\ q=1-p_j-p_k}} . \quad \text{(A3)}$$

Analogous definitions for functions of the number of decays in more than two intervals are not needed presently.

At this point we have the tools to calculate the expected value of any stochastic variable that is a function of the number of decays in the intervals; in this paper we shall need only two such variables:

$$E[M_k] = \left[p\frac{d}{dp}(p+q)^N \right]_{\substack{p=p_k\\ q=1-p_k}} = Np_k = Nx(1-x)^k \quad \text{(A4)}$$

and

$$\begin{aligned} E[M_jM_k] &= \left[p_1\frac{d}{dp_1}p_2\frac{d}{dp_2}(p_1+p_2+q)^N \right]_{\substack{p_1=p_j,\, p_2=p_k\\ q=1-p_j-p_k}} \\ &= N(N-1)p_jp_k + \delta_{jk}Np_k \\ &= N(N-1)x^2(1-x)^{j+k} + \delta_{jk}Nx(1-x)^k , \end{aligned} \quad \text{(A5)}$$

from which we can calculate the expectations of the statistics we are presently interested in.

The expected value of the average count over n consecutive intervals, from the definition (1) and using Eq. (A4), is

$$\begin{aligned} \mu_n \equiv E\left[\frac{1}{n}\sum_{k=0}^{n-1} M_k\right] &= \frac{1}{n}\sum_{k=0}^{n-1} E[M_k] \\ &= Nx\frac{1}{n}\sum_{k=0}^{n-1}(1-x)^k = Nx\frac{1-(1-x)^n}{nx} \\ &= Nx\left[1-\frac{n-1}{2}x + O(x^2)\right] , \end{aligned} \quad \text{(A6)}$$

which is exactly the same as Eq. (5), if one identifies M_0 and Nx.

The expected value of the generalized Allan variance of the counts over n consecutive intervals, from definition (3) and by means of Eqs. (A4) and (A5), is

$$\begin{aligned} \alpha_n^2(j) &= E\left[\frac{1}{2(n-j)}\sum_{k=0}^{n-1-j}[M_k - M_{k+j}]^2\right] \\ &= \frac{1}{2(n-j)}\sum_{k=0}^{n-1-j}\{E[M_k^2] + E[M_{k+j}^2] - 2E[M_kM_{k+j}]\} \\ &= \frac{N}{2(n-j)}x\sum_{k=0}^{n-1-j}\{(1-x)^k + (1-x)^{k+j}\} + \frac{N(N-1)}{2(n-j)}x^2\sum_{k=0}^{n-1-j}\{(1-x)^{2k} + (1-x)^{2k+2j} - 2(1-x)^{2k+j}\} . \end{aligned}$$

Then we sum up the geometric series

$$\begin{aligned} \alpha_n^2(j) &= Nx[1+(1-x)^j]\frac{1-(1-x)^{(n-j)}}{2(n-j)x} + N(N-1)x^2[1-(1-x)^j]^2\frac{1-(1-x)^{2(n-j)}}{2(n-j)[1-(1-x)^2]} \\ &= \mu_{n-j}\left[\frac{1+(1-x)^j}{2} + \frac{(N-1)x}{2}[1-(1-x)^j]^2\frac{1+(1-x)^{(n-j)}}{2-x}\right] . \end{aligned} \quad \text{(A7)}$$

If we expand in the small parameter x, we get

$$\alpha_n^2(j) = \mu_n \left[1 + \frac{\mu_n}{2}(jx)^2 \left(1 - \frac{1}{N}\right)[1 + x + O(x^2)]\right] , \tag{A8}$$

where N is the total number of decays we would detect if all nuclei decayed, which is less than the total number of nuclei unless the whole solid angle is covered with efficiency of 100%, but it is still a large number; therefore, we can always neglect $\frac{1}{N}$ compared to 1. In conclusion, once we introduce the expected value of the average rate $\rho_n \equiv \mu_n/T$, the expectation for the generalized Allan variance to leading order becomes

$$\alpha_n^2(j) = \mu_n^2 \left[\frac{1}{\mu_n} + \frac{j^2}{2(\rho_n\tau)^2}\mu_n^2\right] = \mu_n^2 \left[\frac{1}{\rho_n\tau x} + \frac{j^2}{2}x^2\right] , \tag{A9}$$

which we can compare with the identical results, see Eq. (7), obtained with considerable less effort.

Notice that we shall be able to drop compared to 1 the small parameter $x \approx T/\tau$, when the time interval T is much smaller than the mean life of the nucleus, which is always the case for the experiment to make sense. However, as discussed in the main text, we cannot generally drop $\rho_n\tau x^3$ compared to 1: this term can be larger than 1, in spite of x being smaller than 1; it is enough that $\rho_n\tau$, the number of decays we would see if the rate stayed constant for a time equal to the lifetime of the nucleus, is much greater than one.

For the sake of completeness, we also report the result analogous to Eq. (A8) for the variance:

$$\sigma_n^2 \equiv E\left[\frac{1}{n}\sum_{k=0}^{n-1} M_k^2 - \left(\frac{1}{n}\sum_{k=0}^{n-1} M_k\right)^2\right]$$

$$\approx \mu_n^2 \left[\left(1 - \frac{1}{n}\right)\frac{1}{\mu_n} + \frac{n^2-1}{6}\frac{j^2}{2(\rho_n\tau)^2}\mu_n^2\right] \tag{A10}$$

$$= \mu_n^2 \left[\left(1 - \frac{1}{n}\right)\frac{1}{\rho_n\tau x} + \frac{n^2-1}{6}\frac{j^2}{2}x^2\right] . \tag{A11}$$

We notice that the variance, contrary to the Allan variance, has a dependence on n already in its leading correction; the correction is also a factor n^2 larger than in the Allan variance. The larger n the earlier (smaller T) this statistics deviates from the Poissonian behavior.

[1] C. M. V. Vliet, Solid State Electron. **34**, 1 (1991).
[2] W. V. Prestwich, T. J. Kennett, and G. T. Pepper, Phys. Rev. A **34**, 5132 (1986).
[3] G. T. Pepper, T. J. Kennett, and W. V. Prestwich, Can. J. Phys. **67**, 468 (1989).
[4] T. J. Kennett and W. V. Prestwich, Phys. Rev. A **40**, 4630 (1989).
[5] M. A. Azhar and K. Gopala, Phys. Rev. A **39**, 5311 (1989).
[6] K. Gopala and M. A. Azhar, Phys. Rev. A **37**, 2173 (1988).
[7] R. Boscaino, G. Concas, M. Lissia, and S. Serci, INFN Cagliari Report No. INFNCA93-BCLS1, 1993 (unpublished).
[8] J. Gong *et al.*, in *Noise in Physical Systems and 1/f Noise*, edited by M. Savelli, G. Lecoy, and J. P. Nougier (Elsevier, New York, 1983), p. 381.
[9] G. S. Kousik *et al.*, in *Noise in Physical Systems and 1/f Noise*, edited by A. D'Amico and P. Mazzetti (Elsevier, New York, 1986), p. 469.
[10] G. S. Kousik *et al.*, Can. J. Phys. **65**, 365 (1987).
[11] V. D. Rusov *et al.*, Nucl. Tracks Radiat. Meas. **20**, 305 (1992).
[12] M. A. Azhar and K. Gopala, Phys. Rev. A **39**, 4137 (1989).
[13] M. A. Azhar and K. Gopala, Phys. Rev. A **44**, 1044 (1991).
[14] P. H. Handel, Phys. Rev. Lett. **34**, 1492 (1975).
[15] P. H. Handel, Phys. Rev. Lett. **34**, 1495 (1975).
[16] P. H. Handel, Phys. Rev. A **22**, 745 (1980).
[17] C. M. V. Vliet and P. H. Handel, Physica A **113**, 261 (1982).
[18] C. M. V. Vliet, P. H. Handel, and A. V. der Ziel, Physica A **108**, 511 (1981).
[19] W. V. Prestwich, T. J. Kennett, and G. T. Pepper, Can. J. Phys. **66**, 100 (1988).
[20] R. Boscaino, G. Concas, M. Lissia, and S. Serci, following paper, Phys. Rev. E **49**, 341 (1994).
[21] J. A. Barnes, Proc. IEEE **54**, 207 (1966).
[22] D. W. Allan, Proc. IEEE **54**, 221 (1966).
[23] E. Browne and R. B. Firestone, *Table of Radioactive Isotopes* (Wiley, New York, 1986).