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Photoelectron anticorrelations and sub-Poisson statistics in scintillation detectors

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Abstract

The performance of scintillation detectors for x rays and gamma rays is limited fundamentally by the statistics of the scintillation light and the resulting photoelectrons. This paper presents a new experimental approach to studying these statistics by observing correlations in the signals from two photodetectors. It is shown that the Fano factors (ratios of variance to mean), both for the number the photoelectrons produced on the photocathode of the photomultiplier and for the underlying number of scintillation photons, can be deduced from these correlations. For $\text{LaBr}_3(\text{Ce})$ and 662 keV gamma rays, the photopeak signals obtained by photomultipliers on opposite faces of a thin sample are negatively correlated, and the Fano factor for the photoelectrons is significantly less than one. The inferred Fano factor for the optical photons is very small, indistinguishable from zero within experimental error.

Keywords

Fano factor; Photoelectron anticorrelation; sub-Poisson statistics

Scintillation detectors are used for spectroscopy and imaging of x rays and gamma rays in many applications, including high-energy physics, medical imaging and homeland security[1]. In any scintillation detector, each x-ray or gamma-ray photon produces many optical photons, which are converted to photoelectrons in one or more optical photodetectors such as photomultipliers (PMTs) or photodiodes, and statistics of the photoelectrons set the ultimate limit on detector performance[2,3].

These statistics are usually quantified by the Fano factor[4], which is defined for any integer-valued random variable as the ratio of the variance of the variable to its mean. For a Poisson random variable, the Fano factor is one. For a scintillation detector, we can define separate Fano factors for the optical photons and for the resulting photoelectrons. The Fano factor for the optical photons, denoted F_N in this paper, is not directly observed, but measurements of energy resolution in crystalline scintillation detectors have always shown that the Fano factor for the observable photoelectrons, denoted F_n , is greater than one[5]. For semiconductor detectors, however, the gamma-ray photon directly produces observable electrons and holes, and it is always found that the Fano factor for the number of electron-

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hole pairs, F_{eh} , is much less than one. Values of F_{eh} in the range 0.05 – 0.10 are routinely observed for Si and Ge detectors.

To understand why the Fano factor is less than one for semiconductor detectors, and why the observed Fano factor F_n for scintillation detectors is greater than one, we briefly review the physics of gamma-ray detectors.

When a gamma-ray photon of energy E is absorbed in a scintillator or semiconductor by a photoelectric interaction in an inner electronic shell, it initiates a complex cascade of random events[1,3]. The high-energy photoelectron can produce multiple lower-energy secondary electrons, and the inner-shell vacancy can be filled by a transition that results in either an x-ray photon or an Auger electron. The x-ray photon can again be absorbed photoelectrically, and that photoelectron as well as the Auger electron can also produce secondary electrons. If there is no escape of any of these energetic photons or particles from the scintillator volume, then all of the deposited energy E is converted either to lattice vibrations (primarily optical phonons) or electron-hole pairs. The holes and electrons are separated by an applied electric field in a semiconductor detector and collected on electrodes, but in a scintillator the holes and electrons may combine to form excitons. The excitons can then transport energy to luminescence centers in the case of doped crystals, or they can directly recombine radiatively or nonradiatively. In either case, the resulting optical photons produce the signals in PMTs or other photodetectors.

Events in which there is no escape of secondary x-ray photons or high-energy particles from the material are termed photopeak events. For photopeak events in a semiconductor, the constraint that the total energy E ends up as either electron-hole pairs or as phonons leads to sub-Poisson statistics for the observable signals. Consider the limiting case of a hypothetical semiconductor detector where all of the energy deposited by a gamma ray produces electron-hole pairs and none of it is diverted to phonons or other competing mechanisms. In this limit the number of electron-hole pairs is just the gamma-ray energy divided by the bandgap energy, and the variance of this number – and hence the Fano factor – approaches zero. More generally, semiconductors in which the mean energy per electron-hole pair is small have small Fano factors.

Similar arguments would lead us to expect sub-Poisson statistics for the optical photons in a scintillator, especially for newly developed scintillators with high conversion efficiency (gamma-ray energy to optical photons). On the other hand, the inefficiency of the conversion from optical photons to photoelectrons, determined by the quantum efficiency (QE) of the photocathode in a PMT, drives the Fano factor for the photoelectrons towards one. It is well known that binomial selection (random deletion) of events in any discrete random process tends to produce a Poisson distribution for the events that remain[2]. This does not explain, however, why $F_n > 1$ is observed even with new PMTs with very high QE, now of order 50%.

Effects that can increase the variance of the signals in a PMT-based scintillation detector include variation in light collection with interaction position, randomness in the gain of the PMT and noise in the external electronics. In many inorganic crystalline scintillators, there is also an increased variance from the nonproportional response of the material[6,7,8,5,9]. The cascade process described above produces multiple energetic secondary electrons, each of which eventually produces light, but the amount of light produced by each electron is not precisely proportional to its energy. The electron energies in the cascade are stochastic, so the overall process is doubly stochastic, and the net effect of the nonproportionality is an increase in the variance of the total number of optical photons produced and hence of the photodetector signals[2].

In this paper, we describe an alternative method[10] of studying the statistics of scintillation light, not based on the width of spectral lines, which yields direct evidence of a sub-Poisson distribution of the number of optical photons produced in a scintillation event. The method is based on observing the scintillation light with two PMTs, each of which collects approximately half the light, and observing the correlations of the two output signals. If the photoelectrons were described by Poisson statistics, there would be no correlations because Poisson-distributed events are intrinsically independent.

Consider an interaction event in which an absorbed gamma ray deposits its full energy E in a scintillation crystal. This interaction produces a random number N of optical photons, with mean (expectation) $\bar{N}$ and variance σ_N^2 . The Fano factor for the optical photons is

$$F_N \equiv \sigma_N^2 / \bar{N}. \quad (1)$$

If a photodetector receives light from the scintillation event, it produces n photoelectrons, where n is random both because N is random and also because of the randomness in which optical photons reach the photodetector and produce photoelectrons. Thus the mean of n can be written as [11]

$$\bar{n} = \sum_{N=0}^{\infty} \sum_{n=0}^N n \Pr(n|N) \Pr(N|\bar{N}). \quad (2)$$

If we assume that the photoelectrons are created independently, the conditional probability $\Pr(n|N)$ is a binomial, and the inner sum in (2) has the effect of replacing n with ηN , where η is the probability that an optical photon will reach the photodetector and produce a photoelectron. The outer sum in (2) then has the effect of replacing N with $\bar{N}$, so $\bar{n} = \eta \bar{N}$.

Similarly, the variance of n is given by

$$\begin{aligned} \sigma_n^2 &= \sum_{N=0}^{\infty} \sum_{n=0}^N n^2 \Pr(n|N) \Pr(N|\bar{N}) - (\eta \bar{N})^2 \\ &= \sum_{N=0}^{\infty} \left[\eta^2 N^2 + \eta(1-\eta)N \right] \Pr(N|\bar{N}) - (\eta \bar{N})^2 = \eta \bar{N} + \eta^2 (F_N - 1) \bar{N}, \end{aligned} \quad (3)$$

where in the second line we have used the fact that the conditional variance of n under the binomial probability $\Pr(n|N)$ is $\eta(1-\eta)N$. We have also used the elementary result that the second moment of a random variable is the variance plus the square of the mean. It follows from (3) that the Fano factor for the photoelectrons, F_n , is related to the Fano factor for the optical photons by

$$F_n \equiv \frac{\sigma_n^2}{\eta \bar{N}} = 1 + \eta(F_N - 1). \quad (4)$$

Note that $F_n = 1$ if the number of optical photons is Poisson distributed ($F_N = 1$) and that $F_n \rightarrow 1$ as $\eta \rightarrow 0$, regardless of F_N . In a scintillation detector with a PMT, η is the product of

the QE of the photocathode and the probability of the photon reaching the photocathode. If this product is small, the observed F_n will be near unity.

Now suppose that there are two photodetectors receiving light from each scintillation event, as in Fig. 1, and that the j th detector ($j = 1$ or 2) produces a random number n_j of photoelectrons. The means and variances of these random variables can be computed as above from the binomial probabilities $Pr(n_1|N)$ and $Pr(n_2|N)$, with probabilities η_1 and η_2 for producing photoelectrons in detector 1 or 2, respectively. To compute the covariance, however, we need the trinomial probability $Pr(n_1, n_2|N)$, where the probability of producing no photoelectron in either detector is $\eta_0 = 1 - \eta_1 - \eta_2$. The calculation is analogous to (3) but with properties of trinomials rather than binomials. The resulting covariance matrix for n_1 and n_2 is [2,11]

$$\mathbf{K}^{(n)} = \overline{N} \begin{bmatrix} \eta_1 + \eta_1^2(F_N - 1) & \eta_1\eta_2(F_N - 1) \\ \eta_1\eta_2(F_N - 1) & \eta_2 + \eta_2^2(F_N - 1) \end{bmatrix}, \quad (5)$$

where the elements of $\mathbf{K}^{(n)}$ are defined by $K_{ij}^{(n)} \equiv \langle (n_i - \bar{n}_i)(n_j - \bar{n}_j) \rangle$. Note that the off-diagonal elements vanish if $F_N = 1$ (Poisson light) and are negative if $F_N < 1$ (sub-Poisson light).

The observed signal s_j from the j th PMT is related to the number of photoelectrons by $s_j = n_j G_j$, with G_j being the gain of the j th PMT and its electronics. The covariance matrix of these signals, denoted $\mathbf{K}^{(s)}$, has elements defined by $K_{ij}^{(s)} \equiv \langle (s_i - \bar{s}_i)(s_j - \bar{s}_j) \rangle$. In the absence of PMT gain fluctuations, $K_{ij}^{(s)} = G_i G_j K_{ij}^{(n)}$, but we can account for the gain noise by use of the Burgess variance theorem [2]. The results are

$$K_{jj}^{(s)} = \frac{\bar{s}_j^2}{\bar{N}\eta_j} \left[1 + \alpha_j + \eta_j(F_N - 1) \right], \quad (6a)$$

$$K_{ij}^{(s)} = \frac{\bar{s}_i \bar{s}_j}{\bar{N}} (F_N - 1) \quad (i \neq j), \quad (6b)$$

where $\bar{G}_j$ is the mean PMT gain, $\alpha_j \equiv \text{Var}(G_j)/\bar{G}_j^2$, and $\bar{s}_j \equiv \eta_j \bar{G}_j \bar{N}$ is the mean signal on the j th PMT ($j = 1, 2$). Because the PMT gains are statistically independent, the off-diagonal elements of $\mathbf{K}^{(s)}$ do not depend on the noise factor α . The PMT gain noise broadens the photopeak, however, making it harder to observe sub-Poisson statistics from the energy resolution.

If the PMTs are identical, so that the subscripts on α and η can be dropped, the normalized correlation coefficient, denoted r_{12} , is given by

$$r_{12} \equiv \frac{K_{12}^{(s)}}{\sqrt{K_{11}^{(s)} K_{22}^{(s)}}} = \frac{\eta(F_N - 1)}{1 + \alpha + \eta(F_N - 1)}. \quad (7)$$

To investigate these effects experimentally, ten different samples of cerium-doped lanthanum bromide [LaBr₃(Ce)] were used. All were in the form of thin slabs, and a typical sample size was 8mm×8mm×1mm. Because LaBr₃ is hygroscopic, it was received from the manufacturer in mineral oil. Each sample was lightly abraded with a fine emery paper to remove surface precipitates and then coupled to the PMTs with an optical coupling grease (Visilox Systems V-788). The whole assembly was wrapped tightly in black electrical tape to keep out light and moisture, and all measurements were made within a day of sample preparation and sealing.

PMTs used were either Hamamatsu R6233-100 with super-bialkali (SBA) photocathodes, which has a typical peak QE of 35% as reported by the manufacturer, or Hamamatsu R7600U-200 with ultra-bialkali (UBA) photocathodes and typical QE of 43%.

Short 50 Ω coaxial cables from the PMTs were terminated with a 50 Ω resistor at the inputs of a LeCroy WAVEPRO 725ZI digital oscilloscope (6 GHz bandwidth, 20 GSamples/sec maximum), and the scintillator was irradiated with a Cs-137 source (primary gamma-ray energy 662 keV) as shown in Fig. 1. Both flood irradiation and collimated beams were used in different experiments, and data were collected with the full oscilloscope bandwidth and with internal low-pass filters of 20 and 200 MHz bandwidth. Each PMT signal was recorded as either the pulse height or the integral under the pulse waveform.

The data can be displayed as scatter plots, where each signal pair, (s_1, s_2), serves as coordinates for a point corresponding to that event. Alternatively, we can construct an energy spectrum from the sum of the two signals.

Data from a typical experimental run are shown in Fig. 2. In this case, we used a 7mm×6mm×1mm slab of LaBr₃(Ce), un-collimated radiation and SBA PMTs. The photopeak is the elliptical grouping of points in the upper right portion of the scatter plot, corresponding to the higher-energy peak in the spectrum in this figure. The scatter plot shows that the photopeak signals for the two PMTs are negatively correlated, because the elliptical cluster of photopeak events has its major axis approximately perpendicular to the diagonal where $s_1 = s_2$; a positive fluctuation in one signal tends to be associated with a negative fluctuation in the other signal. The photopeak in this spectrum shows an excellent energy resolution of 2.3% and other data runs have given as low as 2.1% (compared to 2.7 – 3.2% reported in the literature[12,13]). Below the photopeak, a subsidiary peak is seen at 629 keV where a 33 keV fluorescent x-ray photon escapes from the crystal.

To investigate the correlations quantitatively, we isolated the photopeaks by windowing the scatter plots parallel and perpendicular to the $s_1 = s_2$ diagonal. The covariance matrix $\mathbf{K}(s)$ for the windowed photopeak signals was estimated either with a conventional sample covariance or by performing a least-squares fit of the photopeak data to a correlated 2D Gaussian function, windowed to the same region as the isolated photopeak. The normalized correlation coefficient r_{12} was then calculated from the covariance matrix of the best-fit Gaussian.

With this estimate of r_{12} , we can use (4) and (7) to estimate the Fano factor for the photoelectrons as

$$\hat{F}_n = \frac{1 + \alpha \hat{r}_{12}}{1 - \hat{r}_{12}}, \quad (8)$$

where the caret denotes an estimate. To apply (8), we estimated α from the known dynode gain and simple formulas for PMT gain noise[11] and from observed single-electron spectra, obtaining $\hat{\alpha} = 0.1 \pm 0.05$ for the SBA PMTs and 0.2 ± 0.1 for UBA.

If η is known or estimated independently, we can also use $\hat{r}_{12}$ to estimate the underlying Fano factor for the optical photons. From (7), we find:

$$\hat{F}_N = 1 + \frac{\hat{r}_{12}}{1 - \hat{r}_{12}} \cdot \frac{1 + \alpha}{\eta}. \quad (9)$$

To estimate η , we corrected the manufacturer's quoted QE, presumably measured at normal incidence in air, for the reflectivity of the photocathode[14,15] and for an expected increase in QE away from normal incidence[16,17], and we also assumed that the geometrical collection efficiency of each PMT was 0.5. The resulting estimates of η were 0.28 for SBA and 0.35 for UBA.

The results obtained with these procedures are summarized in Table 1. The stated errors reflect variations among samples, different experimental procedures, different ways of isolating the photopeak, and the uncertainty in α , but they do not incorporate the uncertainty in η , which affects only $\hat{F}_N$.

The table shows that the observed electron Fano factor F_n is significantly less than one and that the inferred optical Fano factor F_N is very small; the error bars do not rule out $F_N = 0$. The latter conclusion holds no matter what we assume for η .

In summary, we have presented a method[10] for estimating the Fano factors of the optical photons and resulting photoelectrons produced by gamma rays in scintillation crystals. The method is based on correlations between the signals from two photodetectors on opposite faces of a thin slab of the material rather than on the observed signal variances. It was found that the signals from LaBr₃(Ce) showed strong negative correlations and that the Fano factors for both the optical photons and the photoelectrons were significantly less than one, indicative of sub-Poisson statistics.

Finally, we remark on the connection between this work and the quantum optics literature, where sub-Poisson statistics and anticorrelation are hallmarks of nonclassical light. We have been discussing only conditional statistics, conditional on one gamma-ray photon of one specific energy interacting with the scintillator. There is no implication that the overall light field from a random stream of gamma rays is nonclassical. On the other hand, isolating the photopeak events by signal processing is analogous to gating an optical photon stream by including only certain events, so it is not surprising that the statistics are similar. The key difference is that we have not preserved the light produced by the gamma rays, so we have not created a source of sub-Poisson optical radiation for other uses.

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Figure 1.
System for collecting light from opposite sides of a thin scintillator.

Figure 2. Scatter plot (left) and spectrum (right) for $\text{LaBr}_3(\text{Ce})$ with SBA PMTs and uncollimated radiation from Cs-137.

Table 1Results for LaBr₃(Ce) at 662 keV

Photocathode	$\hat{r}_{12}$	$\hat{F}_n$	$\hat{F}_N$
SBA	-0.32 ± 0.17	0.72 ± 0.06	0.10 ± 0.16
UBA	-0.36 ± 0.17	0.68 ± 0.07	0.09 ± 0.20