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PHOSPHORS' LIFETIME MEASUREMENT EMPLOYING THE TIME BETWEEN PHOTONS METHOD*

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Abstract

The Time Between Photons theory (hereafter TBP) is applied to the evaluation of the lifetime of phosphors employed in the Ion Photon Emission Microscope (IPEM). IPEM allows Radiation Effects Microscopy (REM) without focused ion beams and appears to be the best tool for the radiation hardness assessment of modern integrated circuit at cyclotron energies. IPEM determines the impact point of a single ion unto the sample by measuring the light spot produced on a thin phosphor layer placed on the sample surface. The spot is imaged by an optical microscope and projected at high magnification onto a Position Sensitive Detector (PSD). Phosphors, when excited by an ion, emit photons with a particular lifetime, which is important to evaluate. We measured the statistical distribution of the Time Between consecutive detected Photons (TBP) for several phosphors and have been able to link it to their lifetime employing a

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theory that is derived in this paper. The single photon signals are provided by the IPEM-PSD, or faster photomultipliers when high-speed materials had to be assessed.

Keywords: Radiation Effects Microscopy, Ion beam induced luminescence, Ion beam analysis, phosphors

PACS codes: 61.85.JH; 85.30.TV; 78.60.Hk, 07.78.+s

1. Introduction

Phosphors emit light in different modes that depend on the exciting radiation, so that one speaks of photo-, cathode- or ion- luminescence. The usual way to measure the lifetime of phosphor's excited centers is to shine them with an intense short pulse of the radiation of interest and then measure the emitted light intensity as a function of time. The substantial power released to the phosphor might sometimes modify the material and give unpredictable results. We propose here an alternate way to measure the phosphor's lifetime that employs an extremely low exciting current and actually applies only to ion-luminescence. We have to disentangle the photons generated by a single ion, which can be several thousand, and employ a single-photon detector able to measure the photon arriving time with high timing resolution. The TBP theory [1], described in the next section, is able to connect this "arrival time" to the phosphor lifetime, which is what we want to know. We have applied this theory to study phosphors that are part of the Ion Photon Emission Microscope [2, 3, 4, 5, 6, 7], a technique invented at Sandia. Section 3 describes the setup employed to measure the time between consecutive detected photons and the results we have obtained. Conclusion follows in section 4.

2. The TBP theory

The theory concerns the detection of photons emitted from a phosphor that is traversed by a single ionizing particle. N, the number of excited atoms that decay radiatively, i.e. emitting photons, may range up to a few thousand for each ion. The photons, emitted isotropically, are detected according to an efficiency S that may be a small fraction of 1, like 10^{-2} to 10^{-5}. Moreover, the detector has a dead time, typically a few hundreds of nanoseconds, which usually prevents, in case of fast phosphors (e.g. light emission of plastic scintillators) the detection of all the photons. Certain detectors, such as photo-converters coupled to microchannel plates, are capable of detecting single photons, and we call the initial ion-induced photon detected "the first detected photon". If the phosphor is slow, e.g. emitting light for microseconds or even hundreds of microseconds after excitation, one may be also measure the detection time of the second, third, … photons and thereby determine the "time between two consecutive detected photons". The TBP theory offers a functional connection between the probability distributions of these two experimentally measurable quantities, i.e. the "first detected photon time" and "the time between detected photons", to the lifetime of the phosphor, which is what we want to know. We shall now show the guidelines of this theory and leave the description of the measurement methods to the next paragraph.

We restrict ourselves to a basically simple situation, where the phosphor material is homogenous, the individual decays are independent from each other and the excited atoms, no matter how long they were in the excited state, have the same probability of decay. Of course, if the concentration is high enough, there might be stimulated emission, i.e. a laser-like effect, or, if the excited states are sufficiently extended to allow overlap of the wave functions, there might be co-operative effects. But we here disregard these complex situations.

The "simple situation" we defined above is formally equivalent to the decay of radioactive isotopes in nuclear physics and may be treated in the same way. Of course, we handle here "only" thousands (N) of excited atoms (those corresponding to a single ion), the number of detected photons may range from nearly zero to a few units, and the time between detected photons may be large enough to be measured. Going back to the nuclear decay analogy, we postulate that the fraction of change, during the interval dt, of the probability $P_L(t)$ for the atom to be still in an excited state is proportional to dt according to the differential equation: $dP_L(t)/P_L(t) = -(dt/\tau)$, where τ is a positive constant. The subscript "L" of $P_L(t)$ indicates that an atom is still "Living" in its excited state at the time t. The well known solution, with the condition that the initial probability $P_L(t=0) = 1$, is $P_L(t)=e^{-t/\tau}$. Also, the probability that an atom decays in the time interval [0,t], is $P_D(t) = 1- e^{-t/\tau}$, or the differential probability that an atom decays in the interval [t, t+dt] is $dP_D(t) = dt\ (1/\tau)\ e^{-t/\tau}$, which is simply the analog of the differential "activity" in nuclear physics.

What is the probability $dP_D^{(1)}$ that the "first" decay happens in [t, t+dt]? For each atom, it is simply the product of the probability to decay and the probability that all the other atoms are still alive. Then we have to sum over all atoms, i.e. simply multiply by N.

$$dP_D^{(1)} = N\ dP_D(t)\ P_L(t)^{N-1} = N\ dt\ (1/\tau)\ e^{-t/\tau}\ (\ e^{-t/\tau}\ )^{N-1} = dt\ (1/(\tau/N)\ e^{-t/(\tau/N)} \qquad (1)$$

So, the probability for the "first" photon to be emitted is similar to the probability of a "generic" photon to be emitted; only τ has to be replaced by τ/N.

2.1 Detection. If S is the detection efficiency and U = 1- S, the probability that a photon is detected in [0,t] is $P_S(t)=S\,P_D(t) = S\,(1 - e^{-t/\tau})$, or is detected in [t, t+dt] is

$dP_S(t) = dt\,(1/\tau)\,S\,e^{-t/\tau}$; while the probability of being undetected in [0, t] is:

$P_U(t) = 1-P_S(t) = 1-S+S\,e^{-t/\tau} = U+S\,e^{-t/\tau}$.

The probability $dQ_D^{(1)}$ of the "first" detected decay in [t, t+dt] is, for each atom, equal to the probability that its emitted photon is detected in [t, t+dt] multiplied by the probability that the others N-1 atoms decays are being undetected in [0,t]. Then, one has to sum up over all atoms:

$$dQ_D^{(1)} = N\,dP_S(t)\,P_U(t)^{N-1} = dt\,NS\,(1/\tau)\,e^{-t/\tau}\,(U+S\,e^{-t/\tau})^{N-1} \qquad (2)$$

It should be noted that if NS <<1, being N several thousands, and S<<1 and U~1, so the last formula becomes simply $dQ_D^{(1)} = dt\,NS\,(1/\tau)\,e^{-t/\tau}$, and the 1st detected decay time distribution is a simple exponential with τ as time constant. This suggests a method to measure τ: measure the distribution of 1st photon detection times and determine the average number of detected photons (NS).

The probability $dQ_D^{(k)}$ that the "k^{th}" decay occurs in [t, t+dt] is, for each atom, is equal to the probability that its photon is detected multiplied by the probability that k-1 atoms have already been detected and N-k have not been detected. Then, one has to multiply the result by the combinatorial factor that gives the number of ways in which this may happen which is: $k\,B(N,k) = k\,N!/(k!\,(N-k)!)$, or combining:

$dQ_D^{(k)} = k\,B(N,k)\,dP_S(t)\,P_S(t)^{k-1}\,P_U(t)^{N-k}$.

Now $P_U(t)^{N-k} = (UP_D(t)+P_L(t))^{N-k} = \Sigma_{m=0,\,N-k}\,B(N-k), m)\,(UP_D(t))^{N-k-m}\,P_L(t)^m$, hence:

$$dQ_D^{(k)} = k\,B(N,k)\,dt(1/\tau)\,e^{-t/\tau}\;S^k\,(1 - e^{-t/\tau})^{k-1}\,\Sigma_{m=0,\,N-k}\,B(N-k, m)\,UP_D(t)^{N-k-m}\,P_L(t))^m \qquad (3)$$

The factors in the sum represent: 1) $UP_D(t)^{N-k-m}$, the probability that N-k-m atoms have decayed but stayed undetected; 2) the probability that m atoms stay excited $(P_L(t))^m$; 3) B is the combinatorial binomial coefficient.

2.2 Time between detected photons. If m atoms are excited at t, the probability that the first detected decay of this group occurs, after T, at [t+T, t+T+dT] is:

$dQ_D^{(1)}(m,T) = dT/(\tau/m)\ S\ e^{-T/\tau}\ (U+S\ e^{-T/\tau})^{m-1}$, hence multiplying each term $(P_L(t))^m$, representing the probability that m atoms stay excited, inside the sum in (3) by $dQ_D^{(1)}(m,T)$, we obtain the probability, $d^2R_D^{(k+1)}(t, T)$, that the k^{th} detected photon is observed in [t, t+dt] and the $(k+1)^{th}$ detected photon follows in [t+T, t+T+dT] is:

$$d^2R_D^{(k+1)}(t, T) = k\ B(N,k)\ dt(1/\tau)\ e^{-t/\tau}\ S^k\ (1 - e^{-t/\tau})^{k-1} \quad x$$

$$\Sigma_{m=0,\ N-k}\ [B(N-k), m)\ UP_D(t)^{N-k-m}\ P_L(t))^m\ dQ_D^{(1)}(m,T)] \qquad (4)$$

Evaluating the sum and integrating this expression over t from 0 to ∞, with a complex procedure that we do not report here, we obtain that the probability $dR_D(t, T)^{k+1}$ that the time between the k and k+1 photon measured in the time interval [T,T+dT] is simply:

$$dR_D(T)^{k+1} = dT\ (1/\tau)\ e^{kT/\tau}\ \sum_{n=1,\ N-k}\ n\ B(N, n+k)\ (S\ e^{-T/\tau})^{n+k}\ U^{N-n-k} \qquad (5)$$

where, as defined above, T is the "TBP". Although this is a summation over many thousands of terms, we may take $(S\ e^{-T/\tau})$ to be small and truncate the summation, thereby allowing an efficient computation. For this case, also the variables S and N always appear together as (SN) with excellent approximation provided N >> (k+n). Figure 1, 2 and 3 show distributions of the time between consecutive detected photons, respectively for k=1, 3, and 5. Different values of NS are considered. As suggested from the behavior shown in these figures, one can demonstrate that, when NS<<1, the TBP follows a simple exponential decay curve with a slope of $-1/\tau$. This

result at first seems non sequitur because NS<<1 implies that the number of photons detected for each projectile is much less than ONE, and yet the TBP theory applies to the measurement of the time difference between TWO consecutive detected photons. This is not really a problem however, because while NS<<1 there still is a small probability of producing two or even more photons. This is also a very important result because it indicates that the analysis of TBP data is considerably simplified by reducing N (e.g. by using very thin films) or S (e.g. by using filters or high thresholds on the timing electronics) so that NS<<1. When this case is satisfied the logarithmic slope of the TBP curve provides $1/\tau$.

3. TBP measurement setup and IPEM data

The arrival time of a photon, whenever its energy does not matter, can be easily determined with a precision of few nanoseconds. This is for example the time resolution of the leading edge of the "sum" signal of the Quantar PSD detector we usually employ for IPEM measurements. Compact Hamamatsu photon-counters like the R7400P may act even faster. Figure 4 show the schematics we employed at the Ion Beam Laboratory at Sandia National Laboratories. A low current (atoAmps) beam is provided by a 6.5 MV Tandem impinging on different phosphors under test. The emitted light is then measured by a single photon PSD (the Quantar Mepsicron) [8] and the fast "sum" signal is handled by a standard NIM chain, including also an ORTEC TAC-567. Figure 5 shows the time diagram of the single photon detector signals and the TAC I/O START and STOP signals.

Employing the IPEM device, we have measured several phosphors lifetime. We show in Figure 7 the TBP distribution of a "InGaN Multiple Quantum Well - MQW" sample excited by single 3

MeV alpha particles. NS~2 is simply the average number of detected photons. Data have been fitted to the formula (5) obtaining a $\tau \sim 200$ μs.

4. Conclusion

The TBP theory has been primarily developed as a powerful tool to assess properties of phosphors employed in the Ion Photon Emission Microscopy – IPEM, which is due to become the ideal instrument for Radiation Effects Microscopy of integrated circuits at the GeV-energy cyclotrons [9, 10]. The IPEM phosphors must be extremely bright, fast enough and workable into self-supporting blades. Many phosphors have been assessed by employing this simple experimental setup and the TBP. GaN or the new Quantum Wells InGaN showed to be the most promising phosphors in terms of high efficiency, low beam degradation, and robustness [11,12]. We note that in addition to our IPEM application, that that this new TBD technique that measures a phosphor lifetime to a few nanoseconds resolution without modifying the sample, may also find numerous applications in other IBA and nuclear microprobe studies that use ion-luminescence to characterize materials [13].

Acknowledgments

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Figures Captions

Figure 1. Theoretical distribution of elapsed time between 1st and 2nd detected photons.

Figure 2. Theoretical distribution of elapsed time between 3rd and 4th detected photons.

Figure 3. Theoretical distribution of elapsed time between 5th and 6th detected photons.

Figure 4. TBP measurement setup, see text for details.

Figure 5. Time diagram of photon detector signals and TAC I/O signals.

Figure 6. Distribution of TBP for the “InGaN MQW” phosphor initiated by 3 MeV alpha-particles

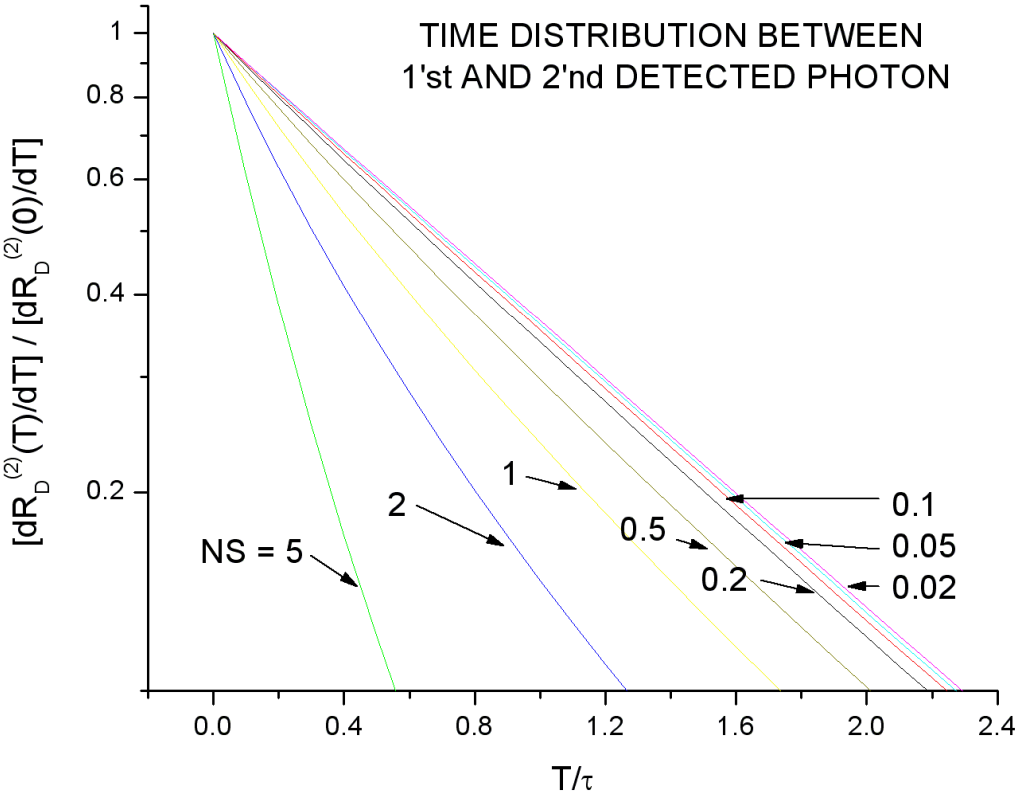
TIME DISTRIBUTION BETWEEN
1'st AND 2'nd DETECTED PHOTON
[dR_D^(2)(T)/dT] / [dR_D^(2)(0)/dT]
T/τ
1
0.8
0.6
0.4
0.2
0.0
0.4
0.8
1.2
1.6
2.0
2.4
NS = 5
2
1
0.5
0.2
0.1
0.05
0.02

Figure 1

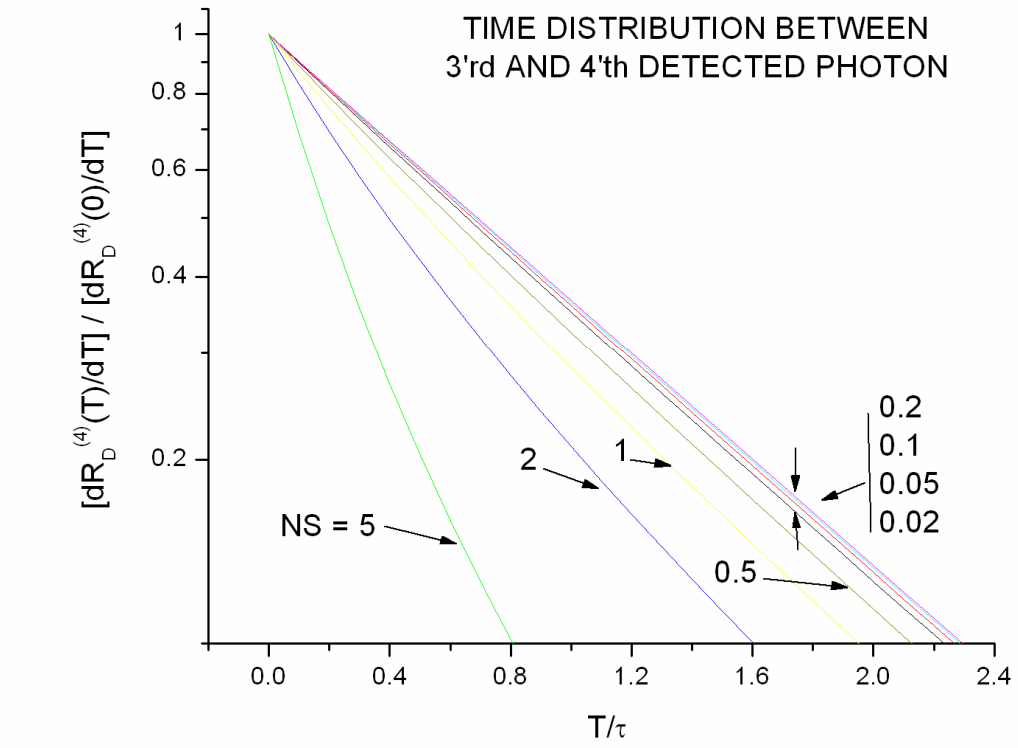
TIME DISTRIBUTION BETWEEN
3'rd AND 4'th DETECTED PHOTON
[dR_D^(4)(T)/dT] / [dR_D^(4)(0)/dT]
T/τ
1
0.8
0.6
0.4
0.2
0.0
0.4
0.8
1.2
1.6
2.0
2.4
NS = 5
2
1
0.5
0.2
0.1
0.05
0.02

Figure 2

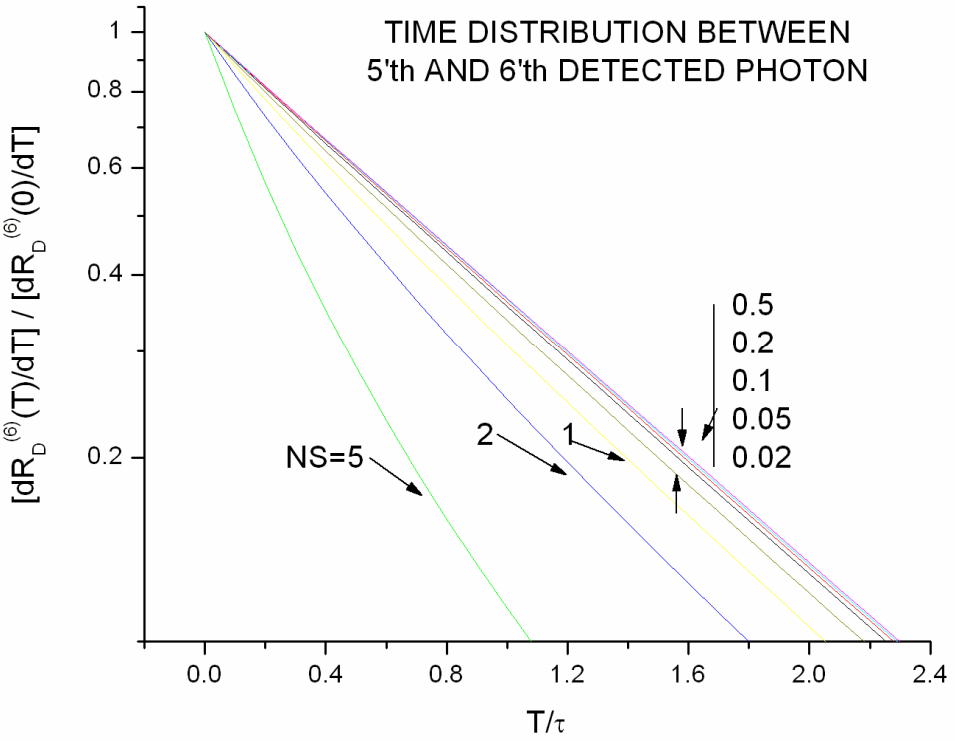
TIME DISTRIBUTION BETWEEN
5'th AND 6'th DETECTED PHOTON
[dR_D^(6)(T)/dT] / [dR_D^(6)(0)/dT]
T/τ
0.5
0.2
0.1
0.05
0.02
NS=5
2
1
0.0
0.4
0.8
1.2
1.6
2.0
2.4
0.2
0.4
0.6
0.8
1

Figure 3

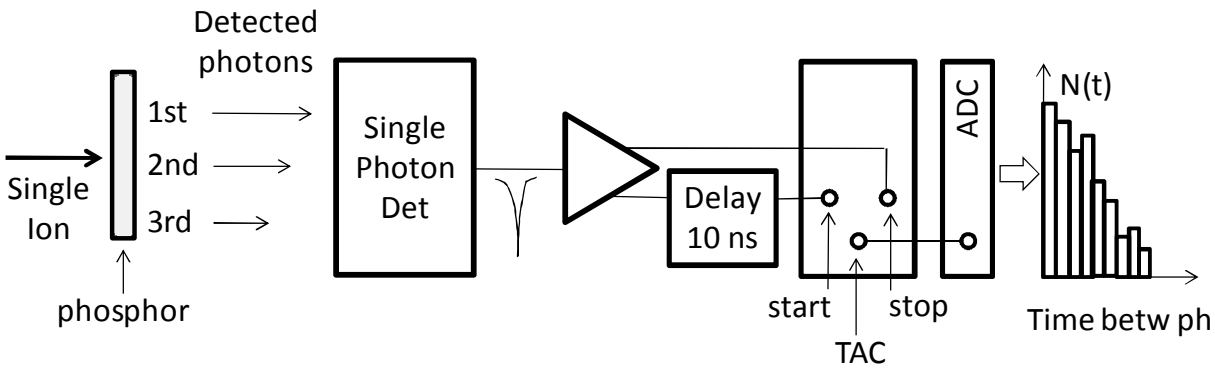
Detected
photons
1st
2nd
3rd
Single
Ion
phosphor
Single
Photon
Det
Delay
10 ns
start
stop
TAC
ADC
N(t)
Time betw ph

Figure 4

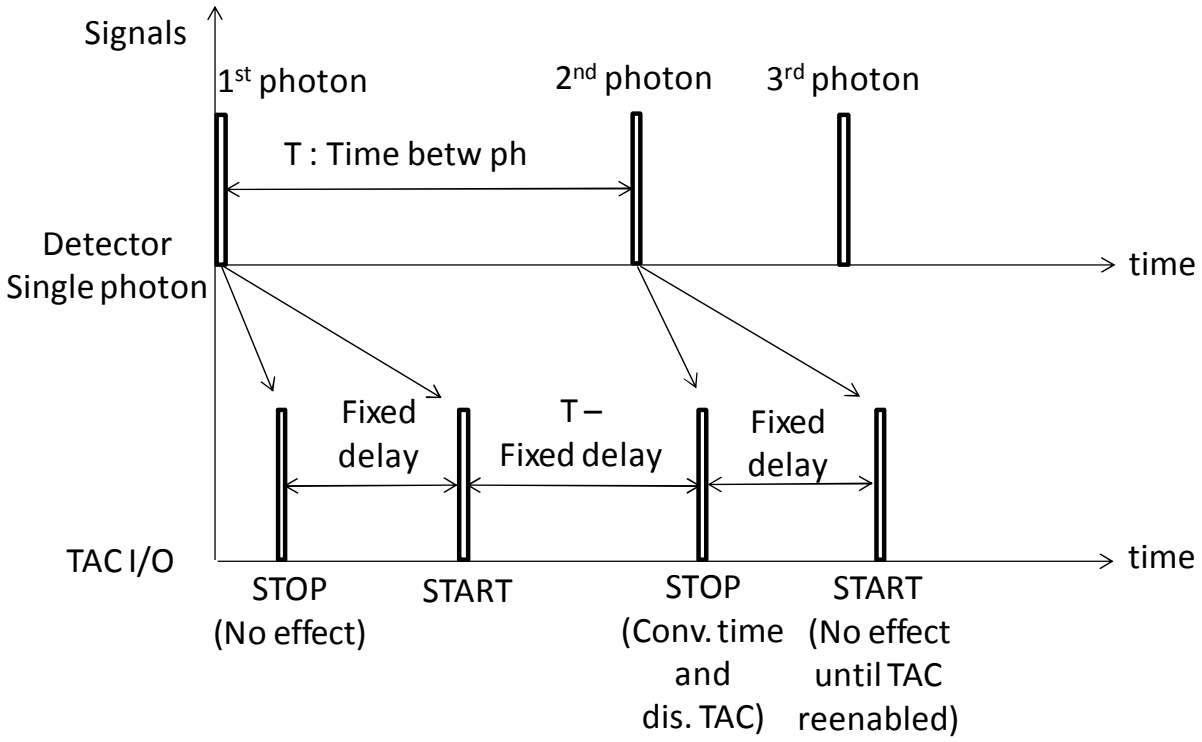
Signals
1st photon
2nd photon
3rd photon
T : Time betw ph
Detector
Single photon
time
Fixed
delay
T –
Fixed delay
Fixed
delay
TAC I/O
time
STOP
(No effect)
START
STOP
(Conv. time
and
dis. TAC)
START
(No effect
until TAC
reenabled)

Figure 5

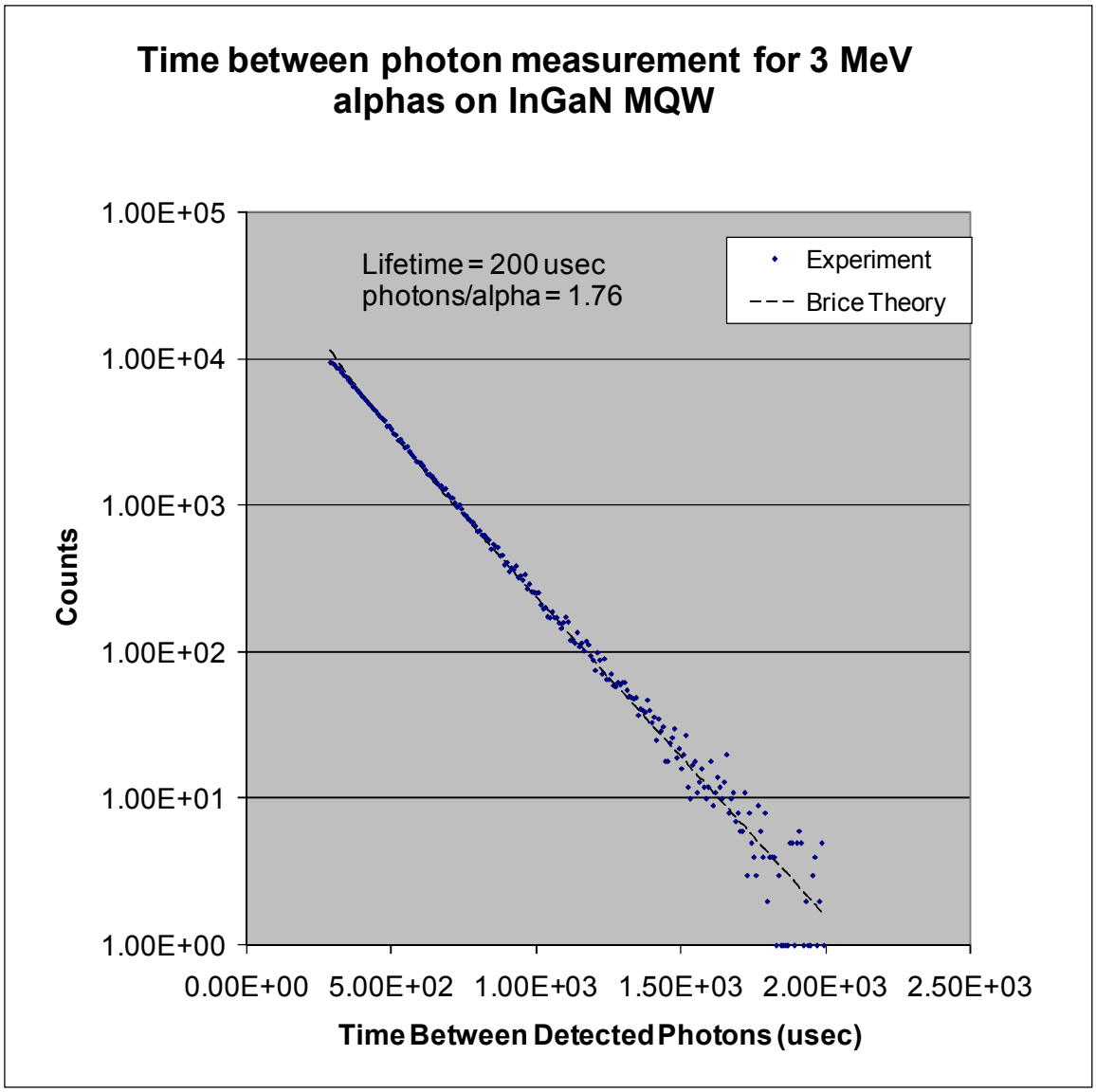
Time between photon measurement for 3 MeV alphas on InGaN MQW
Lifetime = 200 usec
photons/alpha = 1.76
Experiment
Brice Theory
Counts
1.00E+05
1.00E+04
1.00E+03
1.00E+02
1.00E+01
1.00E+00
0.00E+00
5.00E+02
1.00E+03
1.50E+03
2.00E+03
2.50E+03
Time Between Detected Photons (usec)

Figure 6