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PHOSPHOR LIFETIME MEASUREMENT EMPLOYING THE TIME BETWEEN PHOTONS METHOD*

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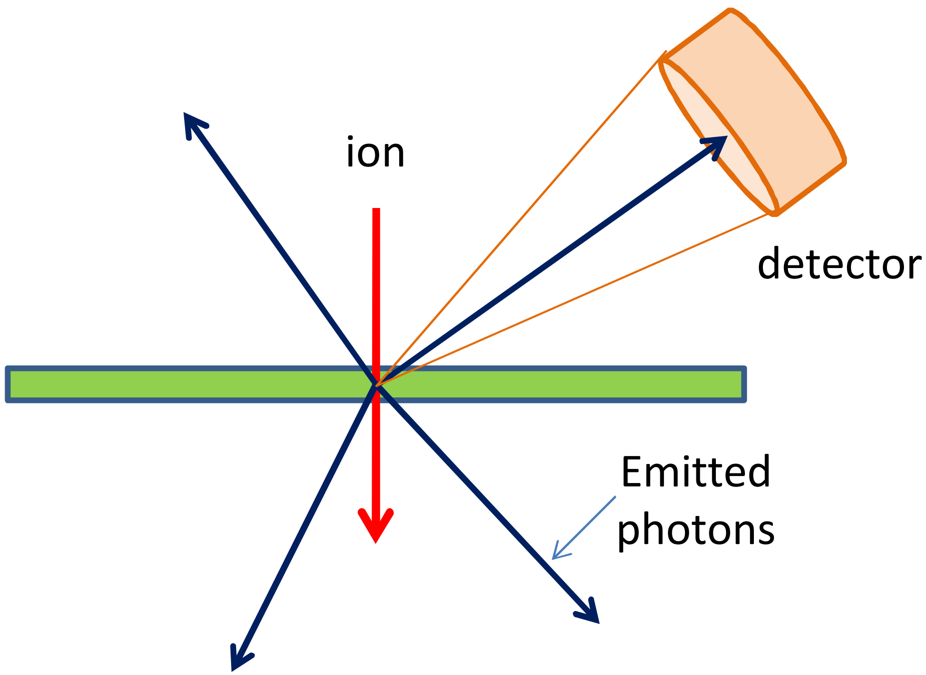
Phosphors, emitted photons and their detection



N, the number of atoms, excited by a single ion, that decay radiatively, may range up to several thousand.

S, the detection efficiency, including the absorption in the traversed materials, the detector solid angle and QE , is a small number (10^{-2} to 10^{-5}).

N·S, the number of detected photons, may range from nearly zero to a few units

The TBP ("Time Between Photons") theory concerns the DETECTED photons emitted from a phosphor that is traversed by a SINGLE ionizing particle.

Supposing we measure one of these two quantities:
1) the time distribution of the 1st detected photons; or
2) the distribution of the time between two sequentially detected photons;
This theory allows the evaluation of the LIFETIME of phosphor.

The TBD Theory

This theory disregards complex situations such as:
-the stimulated emission, i.e. a laser-like effect, if the concentration is high enough; or,
-the presence of co-operative effects, if the excited states are sufficiently extended to allow overlap of the wave functions.

The theory relies upon two assumptions (similar to those of the nuclear radioactivity):

1)the individual decays are independent from each other
2)the excited atoms, no matter how long they were in the excited state, have the same probability of decay

Basic difference: here we handle "only" thousands (N) of "excited" atoms (those corresponding to a single ion), the time between photons arriving to the detector is reasonable, and the number of detected photons range from zero to a few units.
So we can consider the detection time of single photons, while in nuclear radioactivity we measure how many gammas per unit time we have as a f(t)

The Basic Differential Equation

$P_L(t)$: Probability that an atom is still excited ("Living") at the time t.

From the assumption we made (same of nuclear deacays):
1)Indipendence of atoms from each other
2) Probability of "decay" indipendent of time,
it follows that the relative change of probability $dP_L/P_L(t)$ in the time interval dt for the atom to be still in a excited state is proportional to dt according to the differential equation:

$$\frac{dP_L(t)}{P_L(t)} = -\frac{dt}{\tau}$$

Where τ is a positive constant

Basic formulas

The solution of the basic differential equation

$$\frac{dP_L(t)}{P_L(t)} = -\frac{dt}{\tau}$$

Is:

If $P_L(t=0) = 1$, *then:* $P_L = e^{-t/\tau}$

$$P_D = 1 - e^{-t/\tau}$$

Probability that the atom decays in [0, t]

$$dP_D = dt\,\frac{1}{\tau}e^{-t/\tau}$$

Probability that the atom decays in [t, t+dt]
= nuclear "activity"

Definitions

dP : probability that something happens in [t, t+dt]

dP_D : probability that an atom decay in [t, t+dt]

$dP_D^{(1)}$; probability that the first decay happens in [t, t+dt]

dQ : probability of "detection" in [t, t+dt]

$dQ_D^{(1)}$: probability of first detected photon in [t, t+dt]

$dQ_D^{(k)}$: probability of k^{th} detected photon in [t, t+dt]

$d^2R(t,T)$: combined probability that a photon is detected in [t, t+dt] and the following detected photon is detected in [t+T, t+T+dT]

$dR(T)$: probability that two consecutive detected photons are detected with a time difference T in [T, T+dT]

Probability of the 1st decay

The probability $dP_D^{(1)}$ that the "first" decay happens in [t, t+dt] 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.

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

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**

Detection

S = Detection efficiency

U = 1 -S

The probability that a photon is detected in [0,t]

$$P_S(t) = SP_D(t) = S(1 - e^{-t/\tau})$$

The probability that a photon is detected in [t, t+dt]

$$dP_S(t) = dt(1/\tau)Se^{-t/\tau}$$

The probability of being undetected in [0, t] is:

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

1^{st} detection

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 (multiplying by N):

$$dQ_D^{(1)}(t) = N \cdot dP_S(t) \cdot P_U(t)^{N-1} = dt \cdot \frac{1}{\tau} \cdot (NS) \cdot e^{-(t/\tau)} \cdot (U + Se^{-t/\tau})^{N-1}$$

REMARK: if NS<<1, N is several thousands, also S<<1 and U~1, so the last formula becomes simple exponential with τ as time constant:

$$dQ_D^{(1)} = dt\ NS\ (1/\tau)\ e^{-t/\tau},$$

This suggests a way to measure τ

Time Probability of kth detected Photon

The probability $dQ_D^{(k)}$ that the "kth" detected decay occurs in [t, t+dt] is

for each atom, equal to the probability that its photon is detected multiplied 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:

$$kB(N,k) = k\frac{N!}{k!(N-k)!}$$

Or, combining:

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

$$= kB(N,k) \cdot dt \cdot \frac{1}{\tau} \cdot e^{-(t/\tau)} \cdot S^k \cdot (1 - Se^{-t/\tau})^{k-1} \cdot (P_U(t))^{N-k}$$

Time Probability of k[th] detected photon - 2

The probability of un-detection may be expressed as:

$$P_U(t)^{N-k} = (UP_U(t) + P_L(t))^{N-k} = \sum_{m=0}^{N-k} B(N-k,m)\cdot(UP_D(t))^{N-k-m}\cdot P_L(t)^m$$

Hence:

$$dQ_D^{(k)}(t) = kB(N,k)\cdot dt\cdot\frac{1}{\tau}\cdot e^{-(t/\tau)}\cdot S^k\cdot(1-Se^{-t/\tau})^{k-1}\bullet$$

$$\sum_{m=0}^{N-k} B(N-k,m)\cdot(UP_D(t))^{N-k-m}\cdot P_L(t)^m$$

$P_U(t)^{N\text{-}k}$

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) the combinatorial binomial coefficient.

Time between photons

If m atoms are excited at t, the probability that the first detected decay of this group occurs in [t+T, t+T+dT] is

$$dQ_D^{(1)}(m,T) = dT \cdot \frac{1}{\tau}(mS) \cdot e^{-(t/\tau)} \cdot (U + Se^{-t/\tau})^{m-1}$$

hence multiplying each term $(P_L(t))^m$, representing the probability that m atoms stay excited, of the sum in ✡ 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:

$$d^2R_D^{(k+1)}(t,T) = kB(N,k) \cdot dt \cdot \frac{1}{\tau} \cdot e^{-(t/\tau)} \cdot S^k \cdot (1 - Se^{-t/\tau})^{k-1} \bullet$$

$$\sum_{m=0}^{N-k} B(N-k,m) \cdot (UP_D(t))^{N-k-m} \cdot P_L(t)^m \cdot dQ_D^{(1)}(m,t)$$

Time between photons - 2

Integrating this expression over t from 0 to ∞, with a complex procedure that we do not report here, we obtain that

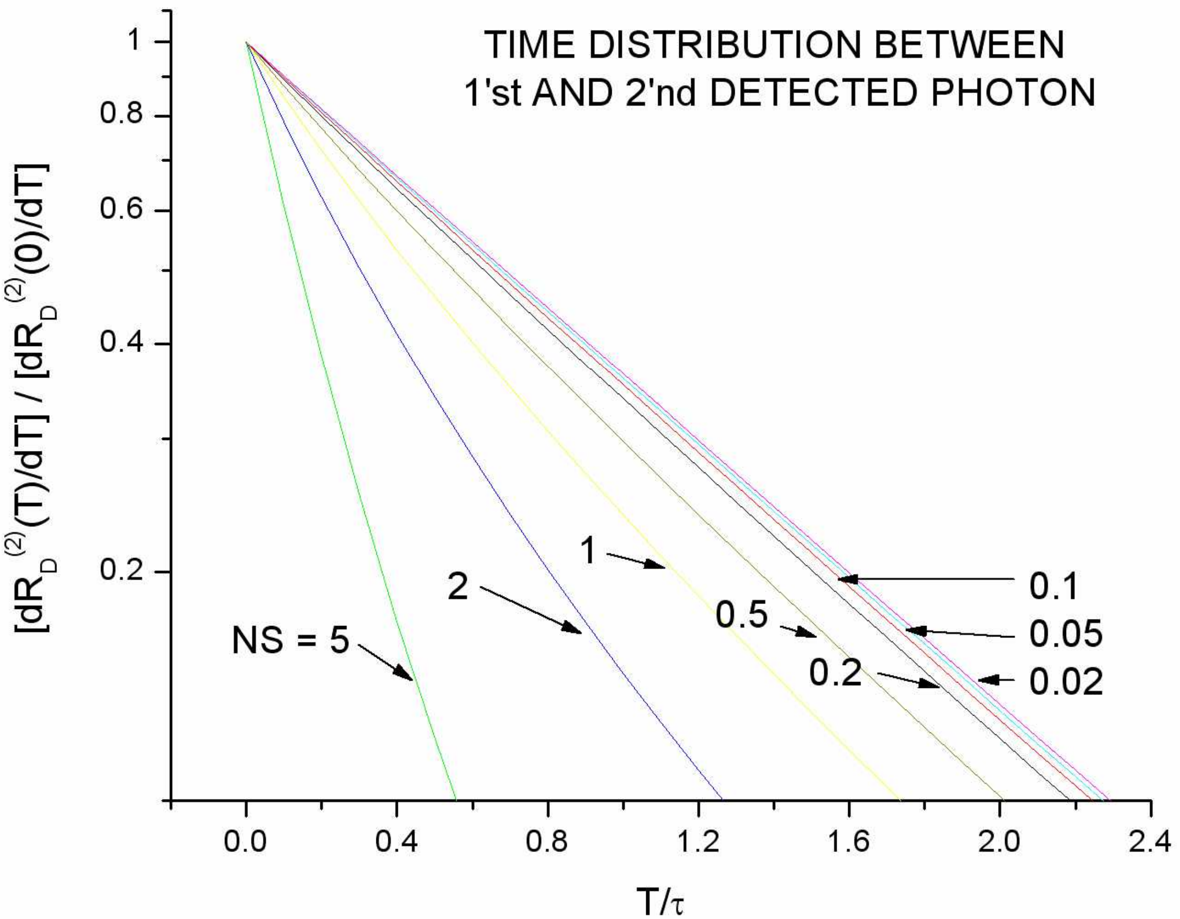
the probability $dR_D(t, T)^{k+1}$ that the time between the k and k+1 photon is measured in the time interval [T,T+dT] is:

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

being T the "time between photons". Although this is a summation over many thousands of terms, we may take $(S\ e^{-T/t})$ to be small and truncate the summation, thereby allowing an efficient computation.

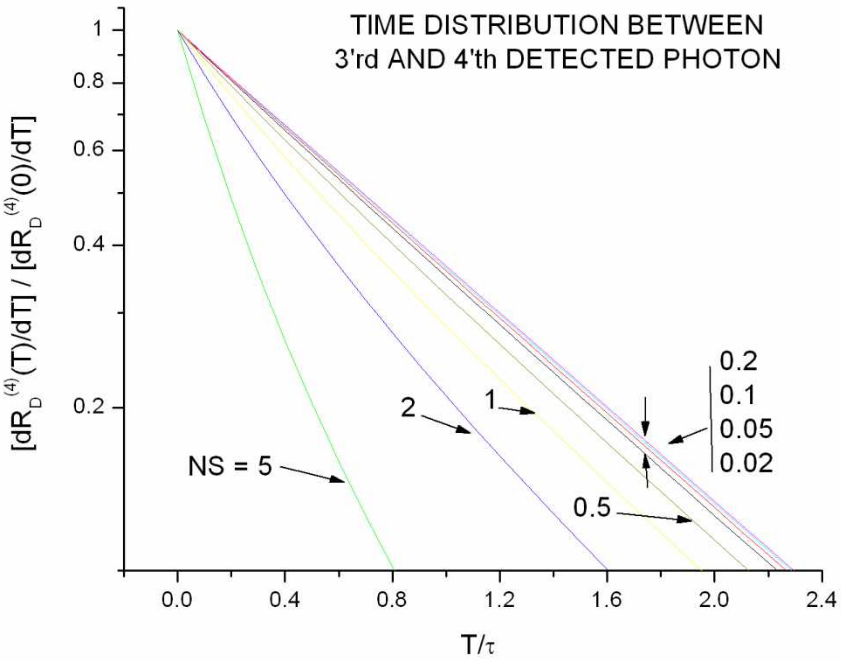
REMARK: for N>>n+k, this expression depends only on SN product

TBP

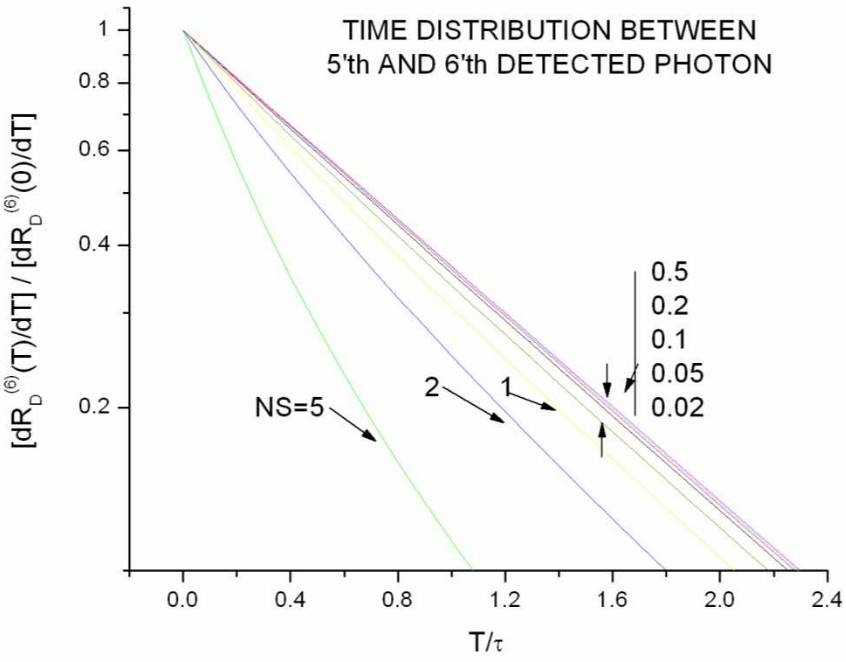


TIME DISTRIBUTION BETWEEN
1'st AND 2'nd DETECTED PHOTON
[dR_D^(2)(T)/dT] / [dR_D^(2)(0)/dT]
T/τ
1
0.8
0.6
0.4
0.2
0.0
0.4
0.8
1.2
1.6
2.0
2.4
NS = 5
2
1
0.5
0.2
0.1
0.05
0.02

TBP -2



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



TIME DISTRIBUTION BETWEEN
5'th AND 6'th DETECTED PHOTON
[dR_D^(6)(T)/dT] / [dR_D^(6)(0)/dT]
T/τ
NS=5
2
1
0.5
0.2
0.1
0.05
0.02

TBP for NS << 1

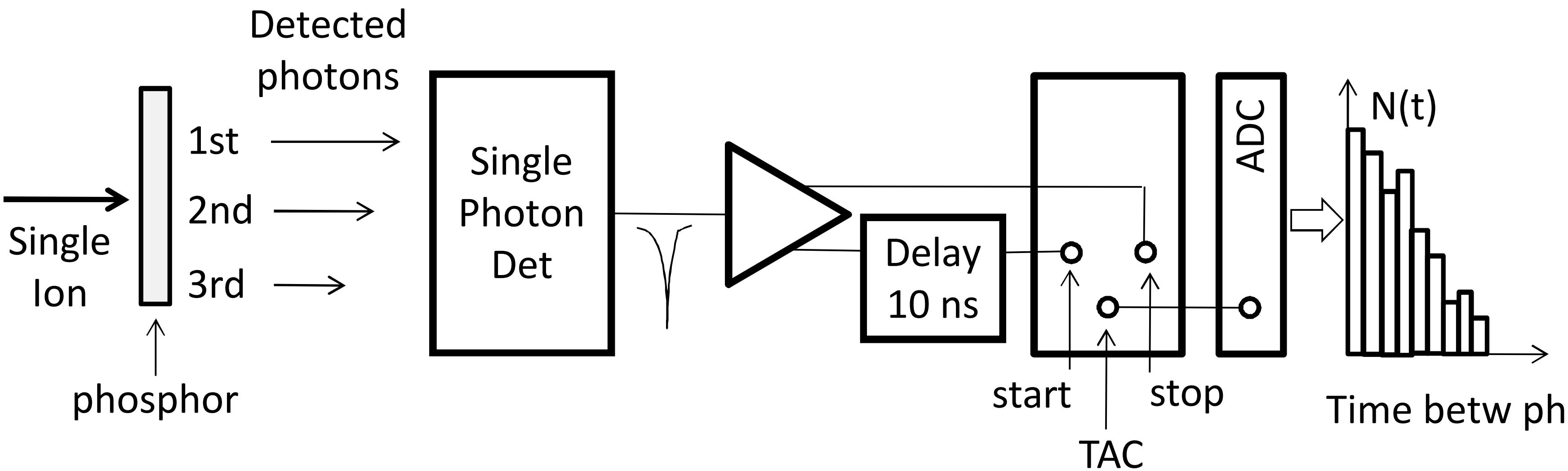
As suggested from the behavior shown in these figures, one can demonstrate that:

when NS<<1, the TBP follows a simple exponential decay curve with a slope of $-1/\tau$ in logarithmic scale

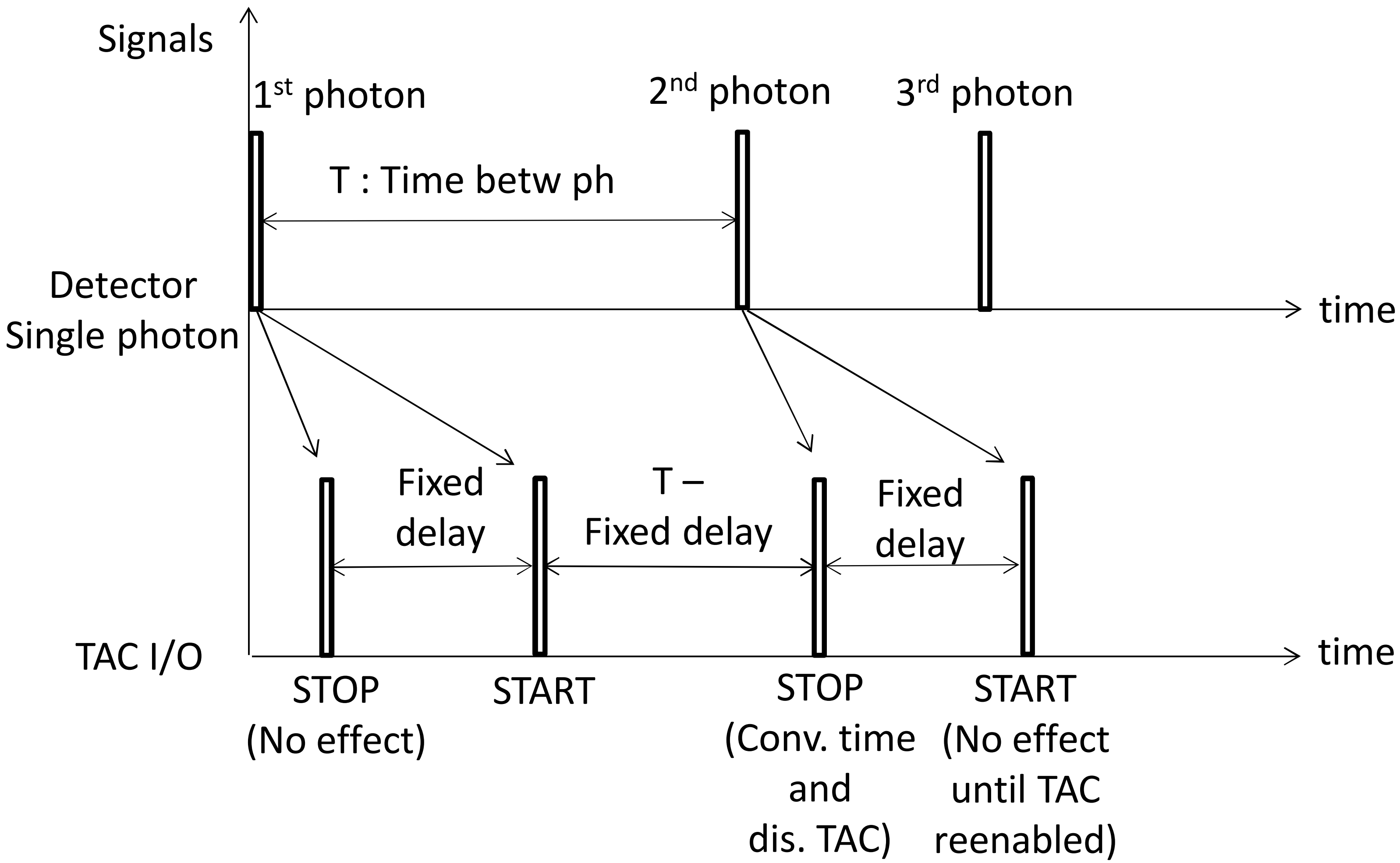
So the analysis of TBP data is considerably simplified by reducing N (e.g. by using very thin films) or S (e.g. by using filters or high thresholds on the timing electronics) so that NS<<1. When this case is satisfied the logarithmic slope of the TBP curve provides $1/\tau$.

The fact that NS<<1 is not really an issue, because while NS<<1 there still is a small probability of producing two or even more photons

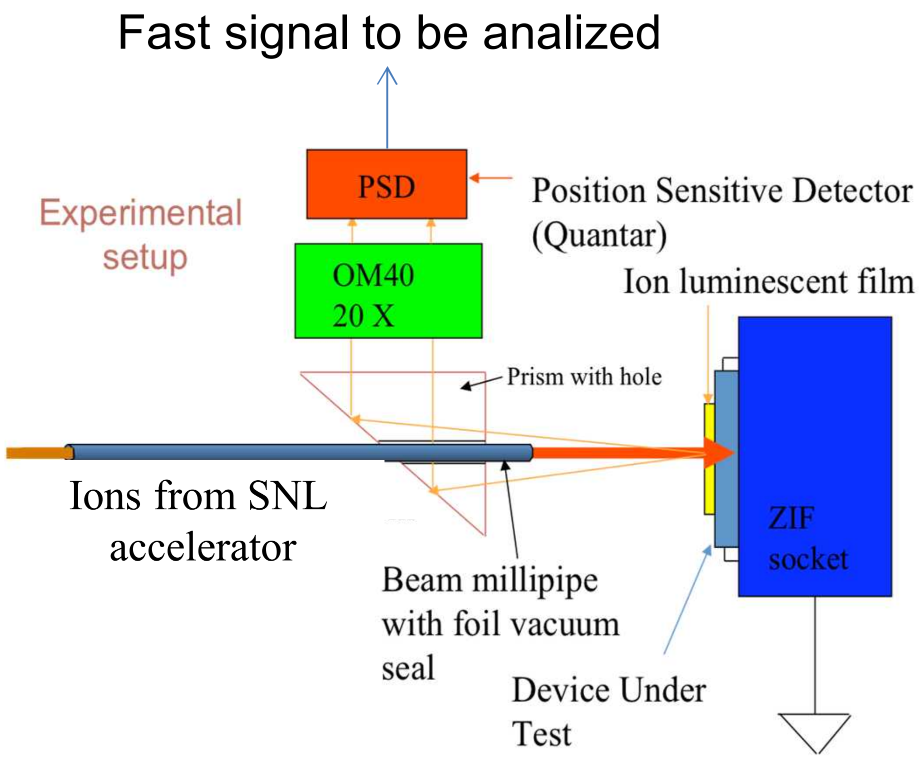
"Time between photons" measurement setup



Time Diagram



PHOTONS MEASUREMENT THROUGH IPEM QUANTAR-PSD



Accelerator IPEM

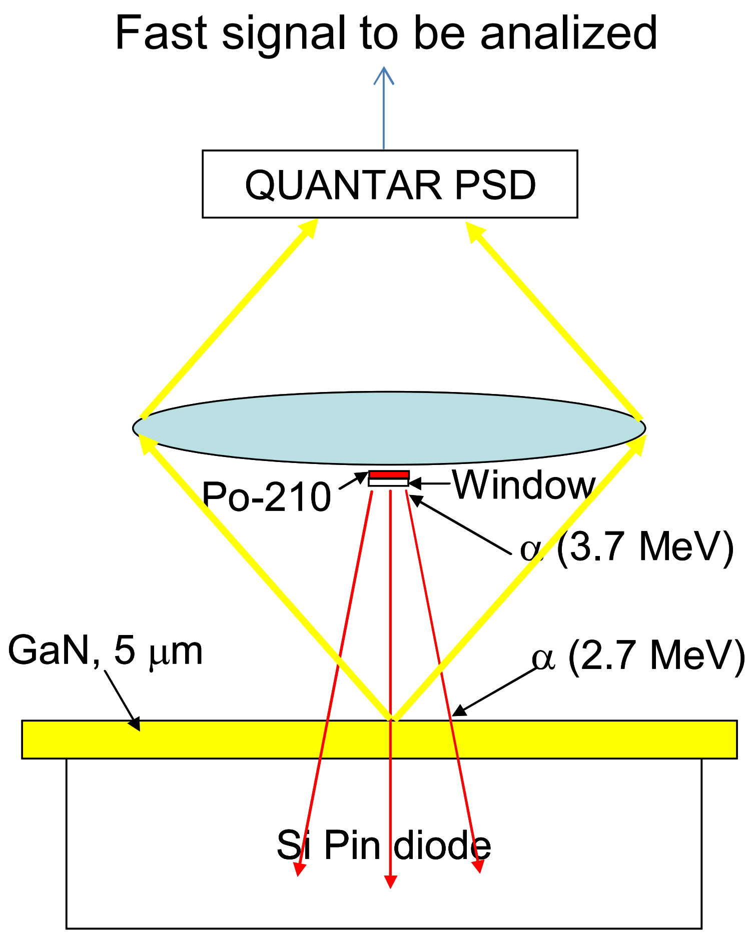
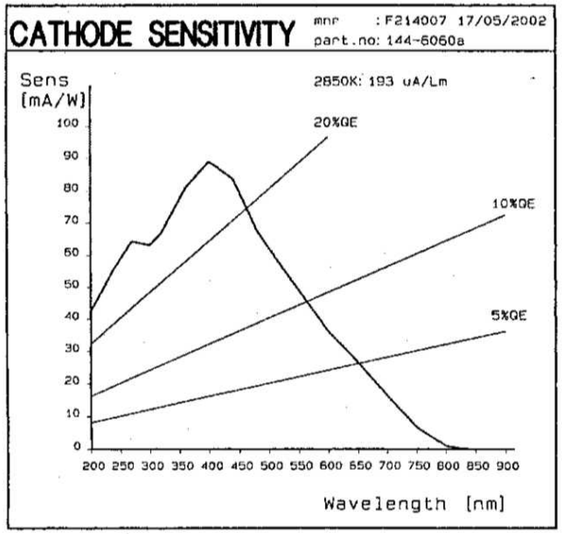
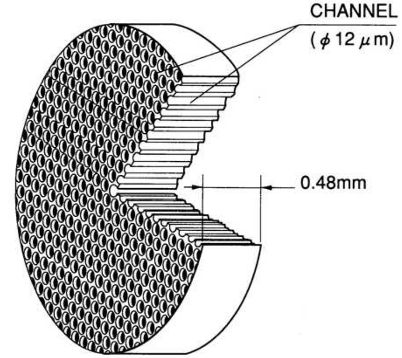


Table-Top IPEM

QUANTAR MEPSICRON

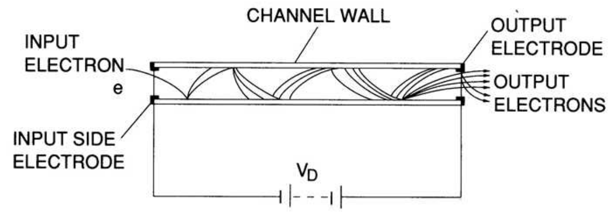
High Sensitivity Detection System With Position Detection

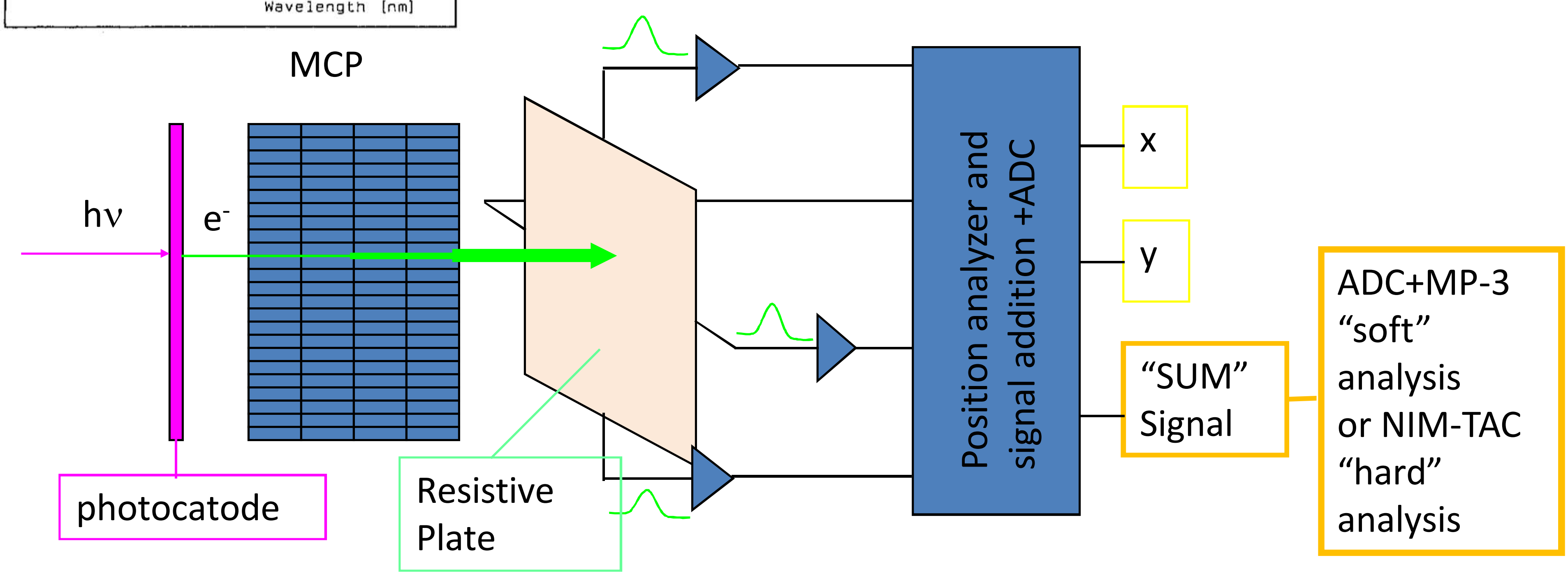




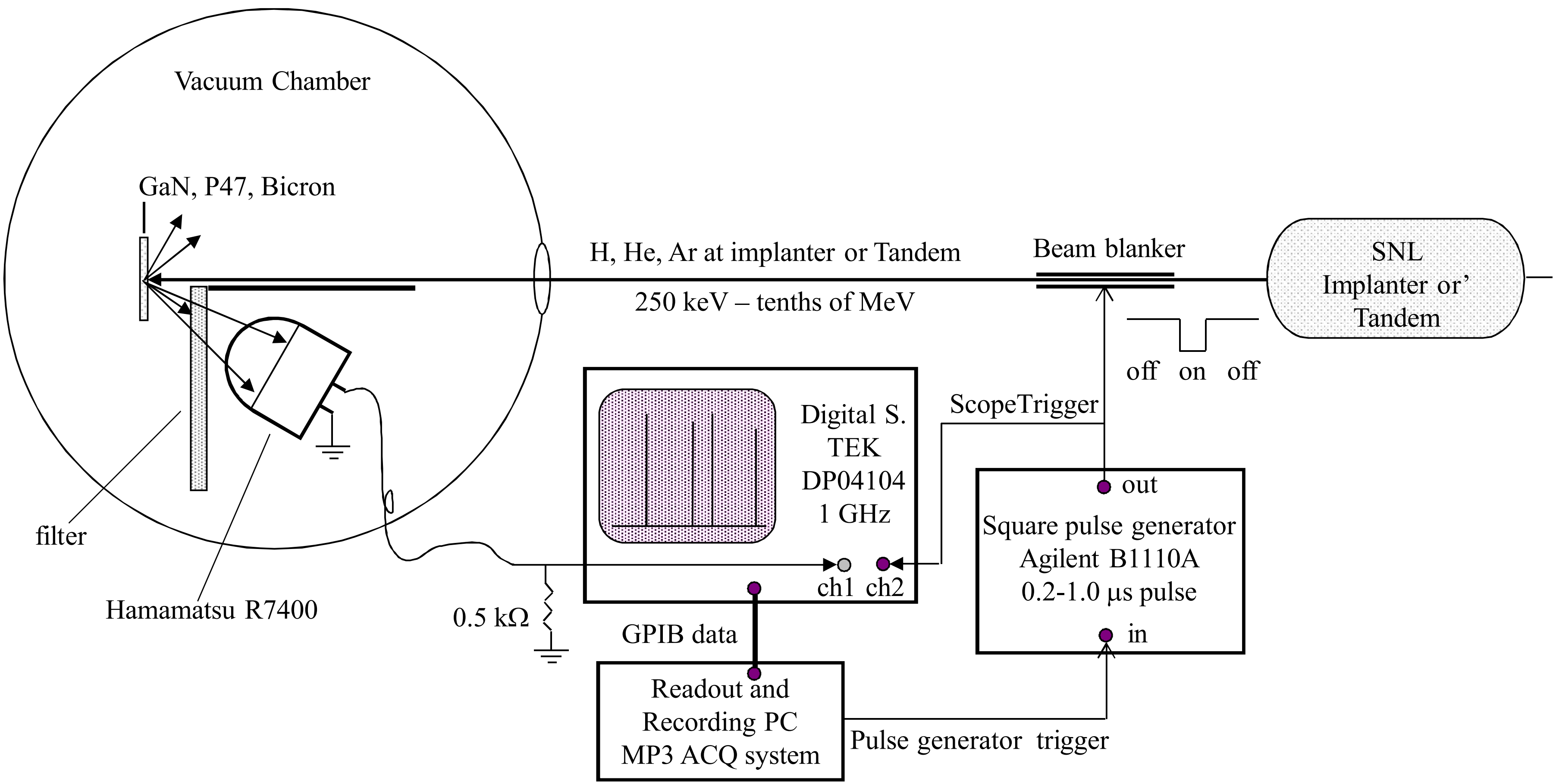
Micro Chan. Plate

1-stage gain 10^2 - 10^4



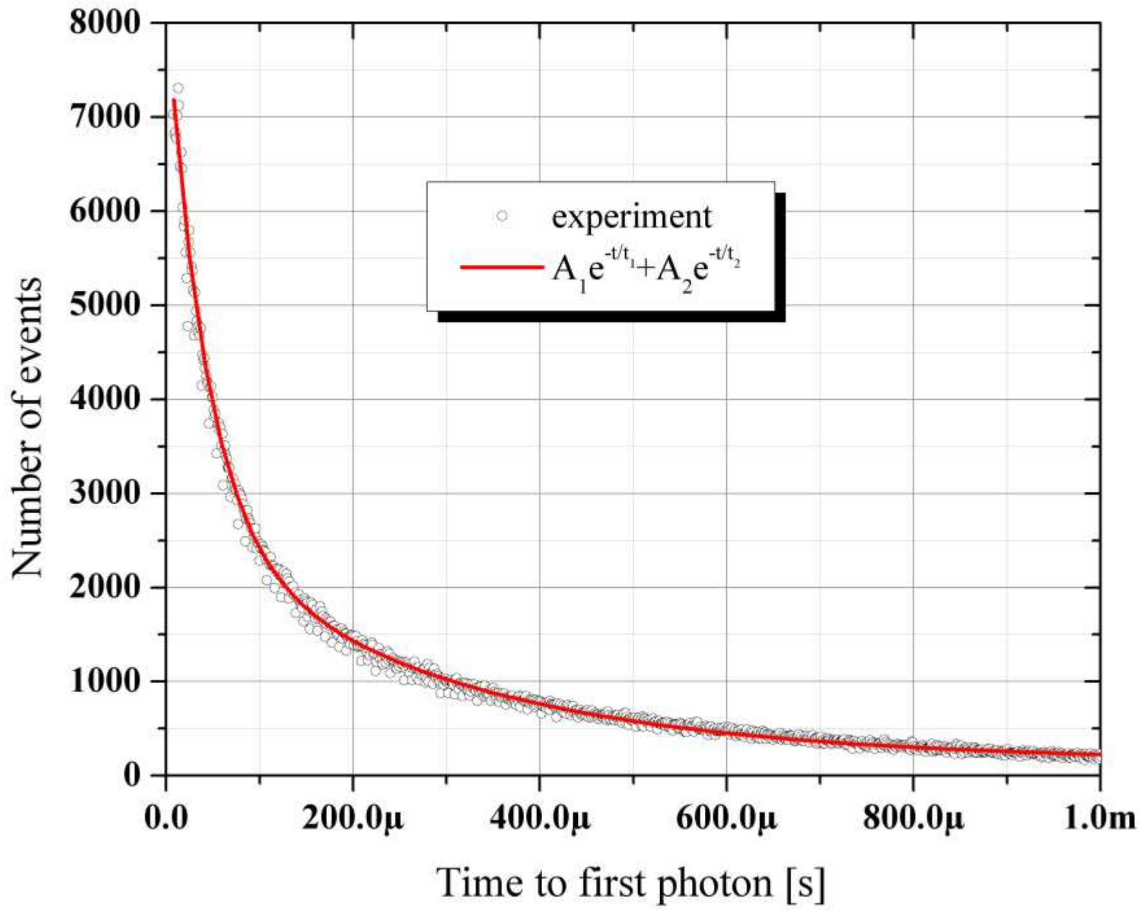


Life Time measurements with the TBP method and the fast R7400P PM



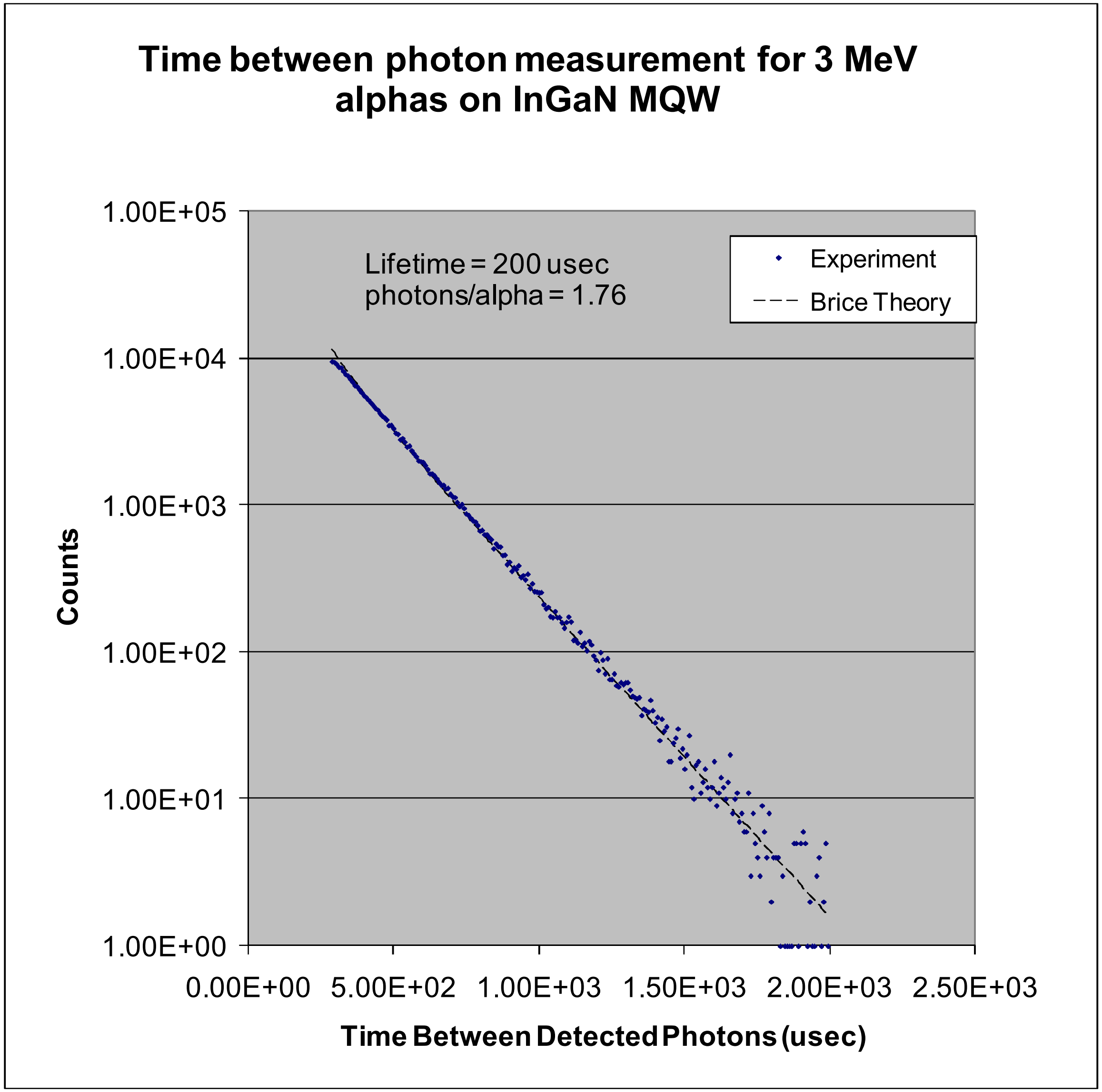
Lifetime measurements(Apr 08)on GaN wafers

Single ion strikes (7.5 MeV alpha) a GaN wafer, single photon detected



Data colleted by IFAST MP3 system, reporting time of every event in 50 ns time slices.
Analysis according to theTBP theory of "time to first photon" gives 280μs and 40μs decay

phosphor "InGaN Multiple Quantum Wells"



NS (measured)= 1.76

τ (fitted)= 200 μs

CONCLUSION

The Time Between Photons method (TBP), although introduced to assess the properties of IPEM phosphors, proves to be a general method to measure phosphors lifetimes in ion-luminescence.

The TBP resolution depends on the time resolution of the photon detector, when energy measurement is not required, and may easily range down to a few nanoseconds and even fraction of nanosecond.

The TBP method relies upon the capability to disentangle the photons coming from a single ion, so the ion beam current has to be extremely low. For this reason, the sample is not damaged and the results are more reproducible