Comparative performance of an integrator and a photon counter in luminescence measurements

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To measure luminescence yield an integrator of pulses by photomultiplier tube-amplifier-discriminator system serving as the light detector is often used. In the present work the performance of an integrator is compared with that of a photon counting (PC) system which is also used for the same purpose. A PC system is found to be more suitable in measurement of luminescence yield and decay time. Furthermore, the values of yield and decay time is seen to be best obtained when PC is achieved by using a multichannel scaler (MCS). However, before adapting an MCS based PC for these measurements it is necessary to establish the single photon response of that system. The performances of these systems have been evaluated from experiments done on lyoluminescence of mannose irradiated by γ-ray to 550 Gy.

1 Introduction

Light yield and its time rate of emission are two important characteristics which are required to be measured in a luminescence. True light yield is determined by the total number of photons emitted in the process and the time rate of decrease in the number of these photons, which has been found to follow the well known exponential decay law, corresponds to the decay time. These two quantities have different values for different luminophors. Our present interest lies in studies on lyoluminescence (LL) and in that connection aim at measuring these two quantities. Incidentally, certain solids, upon irradiation with X- or γ-rays, charged particles or neutrons, when dissolved in suitable solvents, emit light. This phenomenon is known as lyoluminescence. However, the following discussions also apply to the case of some other types of luminescence.

Light emitted during LL are most conveniently detected by a suitable photomultiplier tube (PMT). An avalanche photodiode could also be used particularly when the emission occurs in the IR region. In detecting low intensity light corresponding to small LL emission, pulsed mode of operation of a PMT is preferred to the dc mode to achieve higher sensitivity. This is because a small light intensity represented by a small number of pulses can be measured with a higher degree of confidence by recording the corresponding number of counts than by recording statistically fluctuating and exponentially decreasing low dc current. This is why pulsed mode of operation is generally favoured over dc mode. For measuring very large yields, however, dc mode can also be employed successfully as an alternative. We will consider only pulsed mode of operation in the present discussion.

In the pulsed mode, an alternative to direct counting of pulses is integration of these pulses with an integrator of suitable time constant. While the total number of counts recorded in a scaler after background subtraction should directly measure yield, the corresponding integrated output has to be recorded in a chart recorder and the yield extracted therefrom. A survey of literature shows that, in view of the lower cost and simplicity, use of integrators have been quite popular in luminescence experiments as long as measurement of only yield is desired.

An alternative arrangement, termed as photon counting (PC) has also been used. In this case the photon incident on the PMT are individually counted. Different forms of PC arrangements have been used by Ettinger, Kannan and Banerji et al. for the purpose. Banerji et al. have indicated how PC enables measurement of yield as well as the time dependence of emission (i.e., decay time) in the LL process.

In the present work we propose to make a comparative assessment of the performances of integrator based (IB) and PC based arrangements as applied to LL studies with a view to ascertaining which of the two should be
considered more suitable for application in experiments conducted to determine both LL yield and decay time.

2 Pulse Counting and IB Systems

Typical pulse counting and IB systems are schematically shown in Fig.1. A photon incident on the PMT window produces a small pulse at its anode. Such single-photon pulses have a distribution in amplitude. These pulses are then amplified by a linear pulse amplifier and passed on to the input of a pulse amplitude discriminator. At its output, a discriminator provides rectangular pulses of identical shape and amplitude in disregard to the amplitude and shape of the pulses at its input. If now a scaler is used to count them, the system is rendered a pulse counting system. If, on the other hand, they are integrated over a certain period of time to produce a dc voltage, it becomes an IB system in our description.

An integrator of pulses arriving from a discriminator forms the most important part of the IB system. Such an integrator is generally constructed with a suitable operational amplifier having a very high input impedance as in a FET. The output of the integrator delivering a dc voltage after a time \( t \) represents the number of pulses it has integrated in that time. This output is then recorded with a chart recorder as a function of time. The initial part of such a chart record certainly hints to the existence of decay of LL emission (cf. Fig.2). A simple pulse counting cannot provide this picture.

When any one of these two modes of data acquisition is resorted to, one does not get any clear information about (i) whether each LL photon incident on the PMT has been individually counted and (ii) the characteristic value of decay time of the LL phosphor. Remembering that, besides yield, knowledge of decay time is also important in understanding of the LL process, information on yield should be supplemented by that on decay time.

In considering about (i), we are to bear in mind that LL yield is determined by the total number of photons emitted in the process. If now, say, two photons are incident simultaneously or in quick succession on the PMT, a single pulse is generated and only one count in each such case is recorded leaving no way to recognize that only one in place of two photons has been recorded. In detecting LL emission, the PMT delivers output pulses which are generated upon incidence of single-photon as well as multiple photons. Particularly two-
photon pulses are not rare. The discriminator is incapable of distinguishing between single- and multi-photon pulses. In fact, therefore, one discriminator output pulse is recognized by the scaler or the integrator as equivalent to only one single-photon event. Such a situation is likely to become important at the initial stage of data acquisition where the rate of emission is normally very high. As a result, we record a somewhat smaller than the actual number of photons; hence, a smaller value for the yield. On the whole, with a simple pulse counting arrangement one may unknowingly record counts which may consist at least partly of multi-photon events regarded as singles. The same argument applies to an IB record of the data. Possibility of occurrence of multi-photon peaks along with the prominent single-photon one has been demonstrated by Houtermans and Gangopadhyay et al.8.

While considering (ii) we find that by adopting simple pulse counting it is not possible to throw light on the time-dependence of emission. Accordingly, we will not deal upon simple pulse counting any further. From the experimental and calculated IB-curves of Fig.2(b), on the other hand, existence of time-dependence can be inferred but an attempt to quantitatively estimate it by adopting some suitable fitting procedure will not deliver a sufficiently reliable result. This is because it is not possible to generate a dead time corrected IB-curve on which the fitting procedure is to be applied. We will come across this point once again in section 5.

3 Computation of Integrator Output Voltage

Let the discriminator output positive pulse corresponding to each LL photon (ideally single-photon) have a rectangular shape with an amplitude $V_m$ and duration $T$ seconds (Fig.1). If the PMT under use is maintained stable during use it will deliver a time independent statistically steady dark pulse rate $m$ per second. Since most of the PMT noise and the single-photon LL pulses from the PMT have identical pulse amplitude distribution9, the discriminator output pulse rate is determined by the sum of both. Then the net integrator output voltage $V_0(t)$ at time $t$ is:

![Diagram](https://example.com/diagram.png)

Fig. 2 — (a) The experimental IB curve showing contributions of $p$ and $m$ along with that due to LL. The $V_0(t \rightarrow \infty)$ is also shown. The dotted line gives the net background. (b) The background subtracted experimental IB curve A shows the nature of time dependence of $V_0(t)$ due to LL only. The $V_0(t \rightarrow \infty)$ is also shown. Curve B is computed by using the $m_1$ and $\tau$-values extracted from PC-MCS curve A of Fig. 3. Curve C is a similar one computed by using the $m_1$ and $\tau$-values extracted from the dead time corrected curve B of Fig. 3. The corrected value $V_0(t \rightarrow \infty)$ is seen to be 1.45 times higher than that obtained directly from the IB curve.
Fig. 3 — Curve A: The PC-MCS spectrum recorded in parallel to the IB curve of Fig. 2(a) to ensure data acquisition under identical conditions. Curve B: The counts accumulated in each channel of curve A is corrected for dead time (adjusted to 10 μsec) and replotted. The $n_0$- and $\tau$-values have been extracted from both A and B to compute curves B and C of Fig. 2(b) respectively.

\[ V_0(t) = -\frac{V_{oT}}{CR} \left[ m t + n_0 \tau \left( 1 - e^{-p/t} \right) \right] \quad \ldots (1) \]

where $n_0$ is the single-photon LL pulse rate at the output of the PMT at $t = 0$ and $n(t) = n_0 e^{-p/t}$ which gives the LL pulse rate at time $t$, expresses the exponential LL decay. Here, $\tau$ is the decay time constant and $CR >> \tau$. In the above formulation LL decay has been assumed to proceed via only one channel.

Drift due to input offset current is inherent in an integrator constructed by using an operational amplifier. Such a drift is a source of error. Since LL data acquisition time is generally several seconds the net integrator output may drift substantially within this period causing concern. Moreover, the amount of drift generally tends to change with time. Drift can be minimised by selecting a suitable operational amplifier but cannot be totally eliminated.

Introduction of the contribution of drift in the above expression for $V_0(t)$ changes Eq. (1) to

\[ V_0(t) = -\frac{V_{oT}}{CR} \left[ m t + p t + n_0 \tau \left( 1 - e^{-p/t} \right) \right] \quad \ldots (2) \]

where $p$, the output drift rate assumed constant, may add to $m$ in the case of one integrator chip but subtract from it in another. The operational amplifier we have used shows a positive going drift. The situation is shown in the experimental curve of Fig. 2(a). The region PO in this figure arises from drift only, region QR indicates that $m$ slightly overcompensates $p$ to show a small net linear decrease on the negative side of the curve and the region RS originates from drift, PMT background and LL all taken together.

When the decay proceeds through more than one channel as is common in LL [Ref. 5] Eq. (2) takes the form

\[ V_0(t) = -\frac{V_{oT}}{CR} \left[ m t + p t + \sum \frac{n_0 \tau \left( 1 - e^{-p/t} \right)}{} \right] \quad \ldots (3) \]

To compute $V_0(t)$ for LL, knowledge of $n_0$, $\tau$, and $p$, besides that of $V_{oT}$, $T$ and $CR$ is necessary. For this purpose the experimental curve of Fig. 3 is drawn by using a photon counter in conjunction with a multichannel scaler (PC-MCS) in parallel with an IB system. The three $n_0$- and $\tau$-values obtained from this curve is used to compute $V_0(t)$ from Eq. (3). The IB curve of Fig. 2(a) is redrawn as curve A in Fig. 2(b) after subtraction of the contributions of $m$ and $p$. The curve B is computed by using $n_0$- and $\tau$-values extracted from the corresponding PC-MCS curve A of Fig. 3 which is not corrected for dead time loss of counts. The curve C of Fig. 2(b) is drawn on the basis of curve B of Fig. 3 which has been
drawn after applying correction for dead time of the PC-MCS system (adjusted to 10μs) to the curve A of Fig.3. It is interesting to note the congruity in the nature of the experimental and the computed curves of Fig. 2(b). However, as expected, the LL yield given by the respective values of \( P(t \to \infty) \) as shown in Fig. 2(b) are not the same. We find that the values of yield obtained from curves A and B of Fig. 2(b) are not much different because the IB and the PC-MCS systems have been adjusted to have nearly same dead time. The value of yield obtained from curve C is larger than those obtained from curves A and B. This value represents the actual yield.

4 Photon Counting System

As stated earlier PC refers to counting of all the photons emitted in LL individually. To implement this unambiguously the pulse height distribution which originates when photons are incident singly on a PMT, has to be measured. In this case the PMT pulses are amplified by a linear pulse amplifier and the distribution in pulse height pertaining to single-photons is measured by a multichannel analyser (MCA). The standard pulse height spectroscopy assembly is labelled as PC-MCA system in Fig. 1. A typical single-photon response (SPR) measured with such a setup is shown in Fig. 4.

The photon counter devised by Kannan is a fast device which can measure \( 6.7 \times 10^6 \) statistically distributed counts per second at a counting loss of 1%. Although fast pulses help reduce the dead time of the counting system the rather large size of the system and the related cost are unfavourable elements that make it disadvantageous for use. This is particularly true since
most often we are concerned with such yields for the determination of which data acquisition using very fast pulses is not indispensable. As we will see in section 5 below that a simpler PC-MCS system, even if it is made somewhat slower should very effectively replace this fast system since in this case it is possible to obtain proper results after correction of the acquired data for dead time channel by channel as is demonstrated by curve B of Fig.3. Moreover, Kannan\(^4\) has not supplied a pulse height spectrum which alone does bear a mark of credence on the SPR of the system\(^5\). Therefore, without establishing this for any system one cannot be sure about the multi-photon content which is likely to become large at yields giving counts as large as 10\(^6\) counts per second. This remark also applies while measuring decay time. A discriminator-scaler can provide means to measure an integral bias-curve but it is unsuitable for a straightforward measurement of a pulse height distribution. Kann has presented a few integral bias curves none of which, on differentiation, yields a peaked SPR of the type presented in Fig.4. Besides, one is thrown into confusion by the way the MCA in his arrangement is shown connected for data acquisition. This fast system is, in fact, incapable of measuring a peaked single-photon pulse height distribution but permits of only MCS mode of recording. Ettinger\(^3\) has described a somewhat elaborate photon counter assembly. But he has also not supplied any characteristic peaked SPR curve.

On the contrary, a simple PMT-linear amplifier-MCA combination (PC-MCA) which is the least embellished yet an elegant and effective pulse height spectroscopy system and which can be assembled quite easily, can produce a peaked SPR\(^6\)\(^7\). This system is capable of handling a reasonably large input pulse rate. The spectrum of Fig.4 recorded with a PC-MCA system shows a peaked SPR. It is seen that at a small intensity level, the multi-photons do not contribute much to the SPR but do so at a large yield. Their presence may be too distinct to be ignored when large yields are to be measured\(^7\)\(^8\). Since we aim at determining the total number of photons, it is necessary to have a knowledge of the SPR because that alone can make possible estimation of single- and multi-photon contents in the pulse height spectrum. The SPR curves of Fig.4 show a large peak corresponding to single-photon events but only an unresolved continuum beyond the ULD setting (Fig.4) pertaining to multi-photon events. To resolve the peaks pertaining to one, two and higher number of photons, it is best to use a PMT having a high gain fast dynode (viz., RCA 8850).

A PC-MCA type photon counting system of the above description, though capable of establishing a SPR, suffers from the same limitations as in pulse counting and IB systems in determining yield and decay time. Not only are these limitations obviated but also the necessity of recording the PMT dark background in a separate experiment is avoided if a PC-MCS based assembly is used.

5 Evaluation of Yield and Decay Times

For reasons stated above a simple pulse counting system does not warrant further discussion. We will therefore consider IB, PC-MCA and PC-MCS based measurements in the following.

A recorded \(V_o(T)\) versus time curve (Fig.2) can supply relative yield, knowledge of which suffices in many cases. But it does not immediately give the correct yield. The correct yield is the total number of photons emitted in the process which, from eq. (3), is

\[
\sum n_o, V_o = \frac{V_o(t \rightarrow \infty) CR}{V_m T}
\]

The quantities on the right hand side of Eq.(4) are circuit parameters except \(V_o(t \rightarrow \infty)\). The circuit parameters can be measured quite accurately. The value of \(V_o(t \rightarrow \infty)\) can be obtained from the curve as indicated in Fig. 2(b) with an accuracy sufficiently high, if corrected for dead time loss, for relative yield measurement.

From Fig. 2(a) it is seen that after the LL emission has decayed down to a very small level, \(V_o(t)\) continues to grow linearly due to continued integration of the constant PMT dark pulses and integrator drift taken together. \(V_o(t \rightarrow \infty)\) can be measured from this part of the curve as indicated in Fig. 2(b).

From this experimental value of \(\Sigma_{m1}\), we can neither extract the values of \(n_o\) nor those of \(t\). The same consideration applies to a PC-MCA based data. To get these values explicitly some other supplementary source of information is needed and only other such source is a simultaneously measured decay time curve (cf. Fig. 4).

As the measured value of \(V_o(t \rightarrow \infty)\) arises from integration of the PMT pulses it should be corrected. if true yield is sought, for factors such as (a) light collection efficiency including light source-PMT geometry in the LL reader, (b) quantum efficiency of the PMT photocathode which is dependent on the wavelength band of the incident light, (c) dead time of the system and (d) fraction of the multi-photon events.
Corrections in respect of (a) can be applied with only limited accuracy because dissolution takes place in a glass vial which serves as an extended LL-source. This light source may not become homogenous during the period of data acquisition. Unequal reflectivity of the vial walls and the back cover adds to the inhomogeneity and causes the amount of light to somewhat vary while reaching the PMT. Applying correction in respect of (b) requires adoption of a rather lengthy procedure. The LL emission wavelength band arising from the same luminophor is known to be different under different experimental conditions. Therefore, emission spectrum at each such set of conditions of measurement has to be drawn first before attempting correction. These remarks equally apply to all the three modes of measurement and the values of yields are rendered relative to each other regardless of the data acquisition system used.

Correction factors in respect of (c) and (d) have serious implications and should not be ignored except at very small yields. This is true even if we are ready to accept relative values for yields. Any one system of Fig. 1 introduces a finite dead time in it. The recorded $V_d(t)$ versus $t$ and the SPR curves fail to accommodate some of the pulses due to system dead time particularly at the initial stage of data acquisition during which the pulse rate remains at a high level. The matter becomes more serious as measurement of large yield is intended. In any case, it has been found that dead time correction must be duly applied. Unfortunately, point to point dead time correction on the IB curve is difficult to apply.

Applying dead time corrections to the total counts in the IB or PC-MCS based data could be straightforward had the count rate been maintained at a statistically steady level during the period of data acquisition. Since the intensity of LL emission decreases with time the count rate remains high at the initial stage and decreases as data acquisition proceeds, finally reaching the steady background level. This situation leads to a correction factor that becomes time-dependent. Hence, before we propose to apply it, it is essential to obtain a count vs time curve. Such a curve can be generated only by using a PC-MCS system.

The difference between results with and without dead time correction can be seen in the curves of Fig. 2(b) and Fig. 3. In order to make a direct comparison, the dead time of the PC-MCS system has been adjusted close to that of the IB system. As a result, the $V_d(t)$ versus $t$ curve computed with the help of $m_r$ and $t$ values extracted from the corresponding PC-MCS curve A of Fig. 3 is also close to the IB-curve A of Fig. 2(b). The curve computed with $m_r$ and $t$ values extracted from curve B of Fig. 3, i.e., after dead time correction, reveals the steeper initial part and larger $V_d(t \rightarrow \infty)$ value which indicate how the IB curve should have been had there been no dead time in that system.

The $V_d(t)$ vs $t$ curve can not provide any information on the multi-photon content in the total counts. Multi-photon content can be ascertained only from the SPR curve. In recording all pulses above a threshold set in the discriminator, we would tacitly assume the absence of multi-photon events. Accordingly, a PC-MCA based measurement of SPR should precede all measurements. Once the SPR curve is drawn, it is possible to set the single channel analyser (SCA) window to select the single-photon peak and estimate the multi-photon content that is being left out (Fig. 4). Subsequently, either an IB or a PC-MCS assembly may be used. Such a choice therefore makes possible a measurement in which only single-photon events have been taken part, a condition that has to be served when measurement of decay time is intended. In generating curves of Fig. 2 and Fig. 3 such a selection has been made. As stated earlier, the values of $m_r$ and $t$ cannot be explicitly obtained from an IB record of data.

6 Conclusion

The correct values of yield and decay time are not obtained unless dead time correction is made on the recorded data. Correction for multi-photon content in the data should also be made except when its contribution is known from the corresponding SPR to be negligible. This implies that drawing a SPR is essential under the given experimental condition prior to making actual measurement.

Data acquired by using any of simple pulse counting, IB and PC-MCS systems, even if only single-photon pulses are allowed to actuate the data acquisition process, cannot provide knowledge of correct yield. Applying dead time correction to these data requires knowledge of pulse rate as a function of time. Unfortunately, the rate of arrival of pulses are not recorded in these cases. Therefore, both dead time correction as well as decay time measurement are not possible from these data.

PC-MCS is the proper way to acquire data where the above mentioned time dependence is recorded. But before PC-MCS is applied it is essential to ascertain if a SPR has been established and the SCA window has been selected to record single-photons. From this SPR the
multi-photon content can also be determined and correction on account of this can be applied. Since counts are acquired in each of the MCS channels for a small dwell time (typically 20-40 msec for measurement on LL) they may be safely considered to have originated from a steady pulse source during that small time interval, the dead time correction can then be easily applied to the counts accumulated in each of the MCS channels individually. In view of the above considerations PC-MCS should be considered as the best choice for both yield and decay time measurements.

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