Experimental study of multi-photon contamination on the measurement of fluorescent decay time*

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Abstract In the measurement of fluorescent lifetime based on time correlation-single photon counting technique by means of TAC, due to the contamination of multi-photons a deviation of fluorescent lifetime measured from the expected value is experimentally studied. A correction function instead of a simple exponential function is used to fit the experiment data. The validation of the correction function is checked using the experimental data of several test samples: YAP, NaI(Tl) and LSO. The results show that the correction function well fits the data and the reasonable fluorescent lifetimes are obtained.

Key words time correlation-single photon counting, fluorescent lifetime, multi-photon contamination

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1 Introduction

Fluorescent lifetime is one of the most important parameters for all fluorescent materials. Usually the time correlation single photon counting technique[1—3] by means of time-to-amplitude converter(TAC) is used to measure the fluorescent decay times of scintillation material. The basic principle of this method is: a large number of samples of excited fluorescent events are generated by a special source for a test scintillation sample. Each event has a reference time T0 when the fluorescent centers are produced. The fluorescent centers emit their photons with a lifetime τ. The TAC (TDC) accepts the T0 pulse from the T0-detector as its start signal and a single photon pulse as its stop signal from the single photon sensitive detector which counts the fluorescent photons of this event according to their time order. TAC (TDC) accepts a start signal, it will wait for the FIRST signal to come to a stop within the time range of TAC, then output a given amplitude of a pulse (proportional to the time interval between Start and the FIRST Stop signals). Due to the limited function of TAC, for one cycle of converter, only the FIRST stop signal is active and the others are ignored. Apparently, if single photon from each excited fluorescent event is assured in a large volume of excited fluorescent events, the TAC (TDC) system will read out a decay time curve with a correct fluorescent lifetime; if some of the excited fluorescent events have more than one photon detected one after another by single photon detector, called 'multi-photon contamination', the biased sampling mentioned above makes TAC (TDC) system readout a distorted decay time curve with a false shorter fluorescent lifetime[4]. In order to lower the probability of multi-photon contamination, the mean number of measurable photons per event, m, in the single photon detector must be limited to the relative small values in the stop channel of TAC. According to the Poisson law, the probabilities for 0, 1 and more than one measurable photons per excited event depend on m, which is listed in Table 1. The second column of the table shows the probabilities of inefficiency of single photon detector; the third column shows the probabilities of one single photon events, 'gold events' for fluorescent lifetime measurement; and the forth column shows the ratio of multi-photon contamination events to gold events. Apparently, to make this ratio less than 1%, you have to limit m to less than 0.01 and the inefficiency of the stop channel will be up to 99%. Therefore multi-photon contamination always exists in the TAC sys-

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tem for fluorescent lifetime measurement. Authors[5], based on probability model, had educed a fluorescent time distribution function with a given average photon number m as the following

$$P_{\rm p}(t,k\leqslant n)=\frac{m}{1-\exp(m)}\exp[-mI(t)]i(t)=P_{\rm mp}(t,m)i(t), \quad (1)$$

$$i(t)=\frac{1}{\tau}{\rm e}^{-t/\tau} \quad I(t)=A{\rm e}^{-t/\tau}.$$

Table 1. Probabilities of single photon existing changed with mean photon number.

m	$P(0,m)$	$P(1,m)$	$[1-P(0)-P(1)]/P(1,m)$
1	0.3679	0.3679	0.72
0.1	0.9048	0.0905	0.052
0.01	0.9900	0.0099	0.010

For a TAC system with the average measurable photon number m the time spectrum of TAC response is not a simple exponential decay distribution $I(t)$ but a function (1) with multi-photon contamination. τ is the fluorescent lifetime of a test sample.

In this paper, using time correlation-single photon counting system based on TAC, the fluorescent decay time spectra of YAP-crystal samples are experimentally studied with various average measurable photon numbers, m.

2 Arrangement of experiment

The arrangement of experiment is shown in Fig. 1.

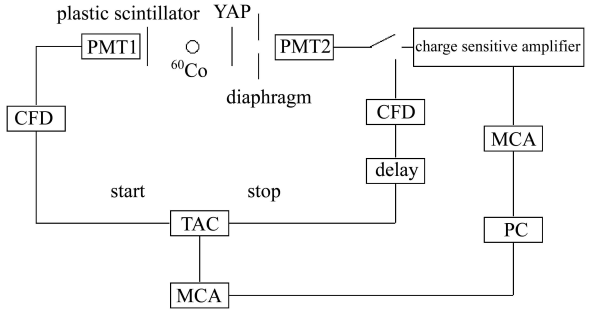

Fig. 1. Experiment configuration for time correlation-single photon counting method.

The cascade gamma 1(2) of the source (^{60}Co) hits the plastic scintillator which is tightly coupled with PMT1, and the pulse output from the PMT anode passes through a CFD with a T0 pulse output which has sub-ns time walk to start TAC. At the same time, the other gamma 2 (1) from the same cascade hits the test sample YAP and is absorbed by YAP, a time correlation fluorescent excited event is generated in the TAC system. The average measurable photon number m arrived at PMT2 from YAP crystal is controlled by diaphragm. The PMT2 which is sensitive to single photon-electron will produce an anode pulse which should pass CFD. The timing pulse from CFD after a proper delay becomes a stop pulse of TAC. For a time correlated fluorescent excited event, if TAC receives the start pulse from PMT1 and there is a stop pulse arriving inside its time range, the TAC and MCA will work together to read out the time interval between the start pulse and the FIRST arrived stop pulse. A mass of time correlated fluorescent excited events is sampling, a time spectrum of the test crystal will be stored and demonstrated in the PC.

The right loop in Fig. 1 is used to determine the average measurable photon number m detected by PMT2 coming from the test sample. The pulses come from the PMT2 anode switch to charge sensitive amplifier for taking the charge spectra first, then to CFD for taking the time spectra of the FIRST single photon arriving (the left loop of Fig. 1).

3 Data analysis and results

For YAP crystal, the charge spectra from PMT2 with various hole sizes of diaphragm are fitted using an amplitude response function $AmS_{\rm real}$ (Q_0, t_0, Q_1, t_1, w, a, u, x)[6] of PMT2, the Q_0 and t_0 are the mean charge and the variance of noise Gaussian distribution respectively, w, a are the weight and decay constant respectively of the exponential component in two-component noise; Q_1, t_1 are the mean and variance of the anode output charge for one photoelectron collected in the first dynode of PMT2; u is the average measurable photon number that arrived at the photo-cathode; x is the variable of amplitude response function, channels of MCA; Am is the amplitude of the response function and is decided by the total counts of the measured spectrum. Fig. 2 is a typical charge distribution spectra of PMT2 , the solid line is the fit result of Am $S_{\rm real}$, the fit parameters are at the top right in the figure. As soon as the average measurable photon number m is determined, The output pulses of PMT1 are switched to the left loop of Fig. 1. The fluorescent lifetime spectrum of YAP is accumulated. The time spectrum is fitted separately by single exponential decay function and correction function (1) with the known m value. Fig. 3 shows some of the fitting results and experimental data are quite well described by the fitting functions.

There are 5 runs with various average measurable photon numbers. All the fitting results for YAP crystal are listed in Table 2.

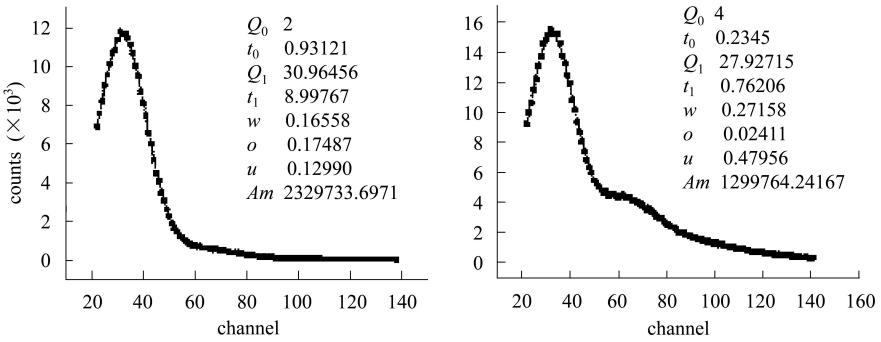

Fig. 2. MCA amplitude spectrum of mean photon number $m = u$ =0.13 (left figure) $m = u$ =0.48 (right figure).

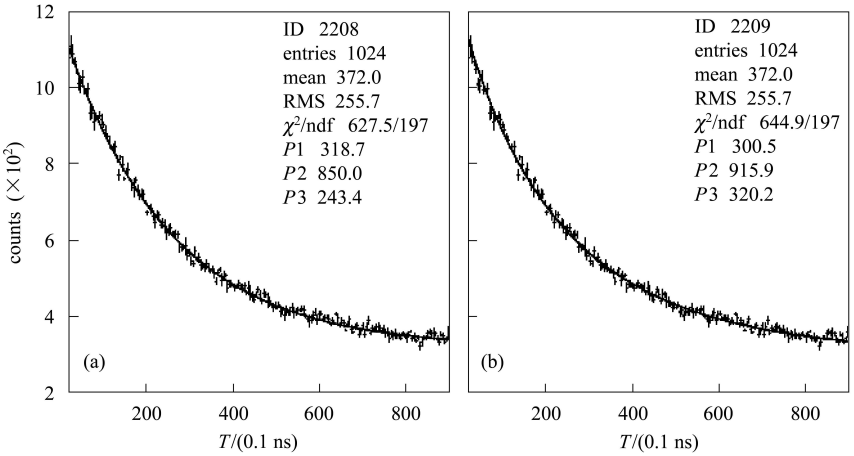

Fig. 3. The decay time spectra with $m = u$ =0.480. (a) Single exponential function fit; (b) The correction function fit.

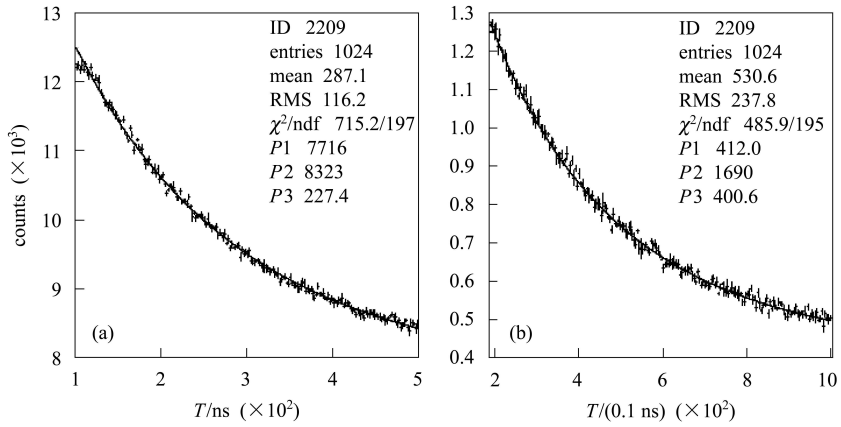

Fig. 4. (a) The correction function fits the time spectrum for NaI(Tl); (b) Correction function fits the time spectrum for LSO.

Table 2. Fitting results (parameters and their fitting errors) of YAP.

average number of photon		single exponential fit		Corr. function fit	
M	Δm	τ'/ns	$\Delta\tau$'/ns	τ/ns	$\Delta\tau$/ns
0.130	0.021	29.42	0.225	32.16	0.26
0.226	0.022	27.96	0.26	32.20	0.32
0.480	0.051	24.34	0.18	32.02	0.29
0.566	0.032	23.35	0.28	32.60	0.50
0.631	0.008	23.24	0.34	32.20	0.34

The simple exponential fits output the values of fluorescent lifetime which becomes smaller when the average measurable photon numbers getting larger (multi-photon contamination is getting severe). The function (1) fits output the values of fluorescent lifetime which is around an average value with a reasonable error.

For checking the validity of the function (1) with a given average photon number m, two test samples of NaI(Tl) and LSO are tested. The charge spectra and the decay time spectra are shown in Fig. 4.The function (1) with m=0.293 fits the distribution (a) giving output τ=227.4±2.03 ns for sample NaI(Tl). The function (1) with m=0.534 fits the distribution (b) giving output τ=40.06±0.44 ns for sample LSO.

4 Conclusion

The fluorescent lifetime of a YAP crystal sample is studied under various average measurable photon number m based on time correlation-single photon counting technique with TAC. The measured lifetimes from single exponential function fit severely deviate from the true value as the multi-photon contamination (m) increases. The corrected function (1) describes the decay time spectra with a single τ value within small errors. With the corrected function (1), the fluorescent lifetimes measured are: τ= 40.06±0.44 ns for LSO; τ = 32.22±0.13 ns for YAP(Ce) and τ = 227.4±2.03 ns for NaI(Tl), free of multi-photon contamination.

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