Plasma diagnostics and simulations

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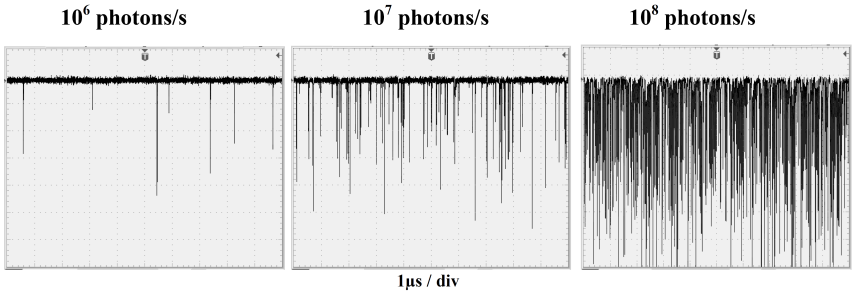
TCSPC

Why to use photon counting?

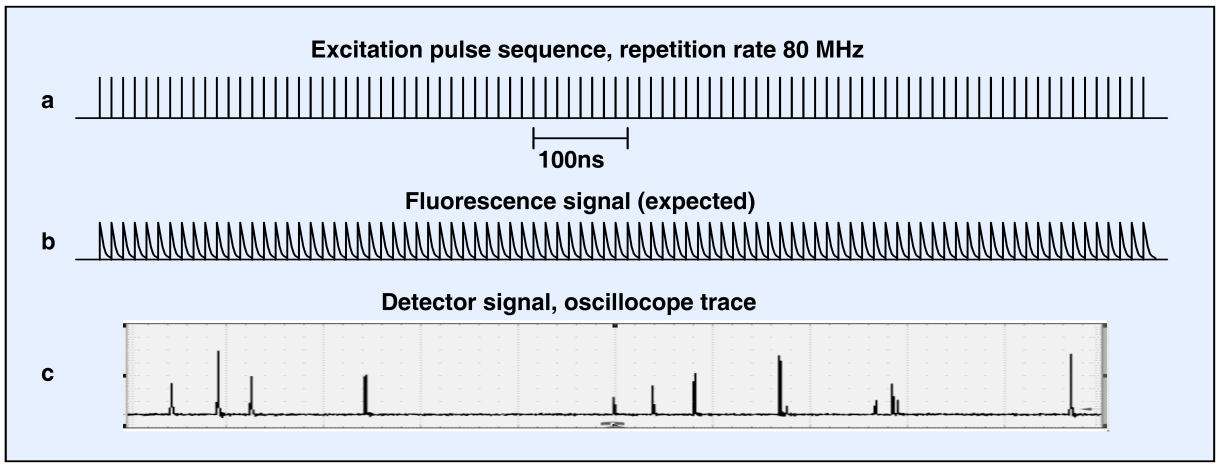
Photon counting can be taken as an opposite to the classical analog optical signal recording, consisting in direct digitizing of voltage or current signal from a detector. Several arguments for using photon counting are listed below:

- The light consists of photons. To measure the light means to absorb each photon in a detector. Why not to count them?
- Output of fast detectors (with time resolution in ns or ps range) is not a smooth signal even under constant illumination, but a composition of narrow peaks, each formed as a response to the photon arrival to the detector (see figure 1).
- Electric output of such a detector under a weak light illumination does not reproduce the original light signal at all, consisting of random sequence of pulses (see figure 2). The pulse shape is a characteristic of the detector. The analog digitization then produces totally erroneous result. The original signal can be reconstructed only by determination of the photon arrival time distribution.
- The time resolution achievable with the photon counting can be better than with analog electronic recorders. Whilst the time resolution of analog systems is determined by the width of a single electron response pulse, the time resolution of time-correlated single photon counting (TCSPC) can be substantially lower (typically 1/10 of the value). For processes occurring in ns- and ps- time scale, the resolution of analog recorders is insufficient.
- The S/N ratio can be better than in analog systems, when the noise is small compared to the amplitude of single photon pulses. This is due to the noise rejection in times, when no photons are registered.

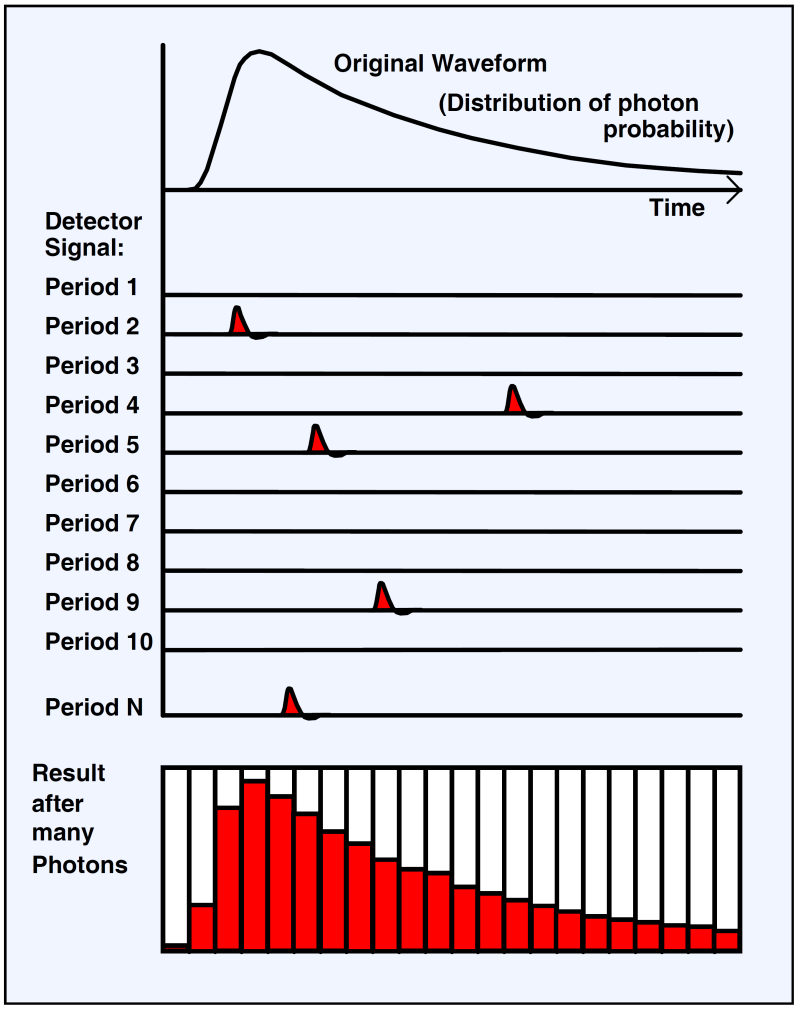


Obrázek 1: Single photon pulses from a fast PMT under varied illumination. Courtesy of W. Becker.

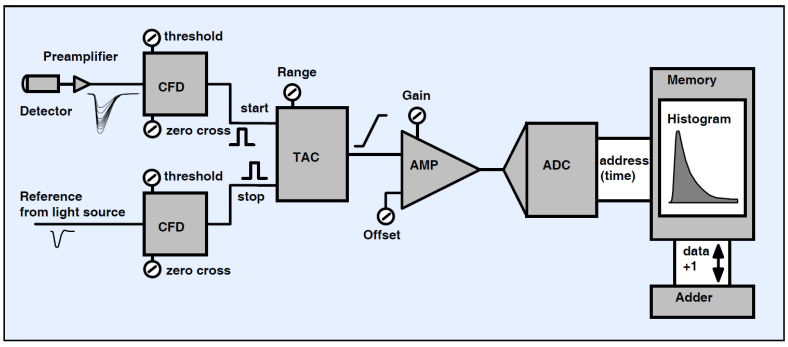
There are many ways how photon counting can be employed in the time-resolved measurement (using gates, multichannel scalers, event recorders etc). However, the highest time resolution is obtained with the time correlated single photon counting, which will be described in the next section.



Obrázek 2: Excitation laser signal (a), expected (b) and real (c) distribution of photons in a experiment of laser induced fluorescence. Reproduced from [1].



Obrázek 3: Reconstruction of a weak signal by counting the photons. Reproduced from [1].



Obrázek 4: So-called reversed start-stop TCSPC setup. Reproduced from [1].

Principle of TCSPC method

The basic principle of the TCSPC method can be understood from figure 3. Let's assume, that the we want to measure the light emitted during a periodical process, e.g. a photoluminescence initiated by a periodical laser pulse or during a discharge driven by high frequency AC voltage. For low level signals, the probability of a single photon detection during a single period is substantially less than one.[1] Then it is possible to count the photons, to measure their arrival times within the process period and to reconstruct the original optical signal from their temporal distribution (figure 3). This of course requires a repeatable and a highly reproducible experiment.

Time correlation An example of a technical solution of the TCSPC principle is displayed in figure 4, where a so-called reversed start-stop version of TCSPC setup is displayed. This means, that the registration of the photon in the detector starts the time measurement procedure. During this procedure, a voltage ramp is generated. The synchronization signal (SYNC), which is mostly taken from an experiment control (trigger from laser, AC voltage or a signal from another PMT) then stops the ramp generation. The final value of voltage is then translated into the time difference between the photon arrival and the SYNC pulse signal.

Times available in the selectable time windows of ramp generation (e.g. of 50 ns width) are mirrored in the memory of the TCSPC device as a series of memory bins. According to the measured time difference, the value of a specific memory bin is incremented by the unit to denote that the photon at this time was registered. The counting procedure is repeated until a photon distribution with sufficient number of collected photons is obtained.

During the routine of the photon counting and the time measurement, the single TCSPC device is unable to register another photon. This dead time then determines the maximal count rate achievable with the device. E.g., the dead time of our TCSPC instrument is 100 ns, which limits the counting rate to 10 MHz. The high counting rate is feasible in order to reduce the time of the experiment.

Detectors Several types of detectors can be used for single photon counting: photomultiplier tubes (PMT), avalanche photodiodes (AD), hybrid detectors combining PMT and AD. The PMTs with linearly focused dynodes or in form of a multichannel plate are often used. High voltage placed on the PMT ensures, that a single electron, which got off due to photoeffect from the photocathode of the PMT, is multiplied at the dynodes into a photocurrent with a sufficient magnitude. For example, our photomultiplier PMC-100, which is of multichannel plate type, has a single electron response (SER) in a form of voltage pulse with 50-200 mV amplitude and 1.5 ns width (for a gain of 0.9-1.0).

[1] Precisely, the photon count rates must be lower than $\approx 1\,\%$ of the signal frequency, otherwise the distortion of the signal by so-called pile-up effect happens.

Time resolution The time of the photon arrival, with respect to the synchronization signal, can be measured with an extremely high resolution (several ps). This is the advantage over multi-channel scalers, in which the time resolution is lower, limited by the slow memory bank switching. In order to achieve such high resolution, the TCSPC uses specific routines to determine precisely the photon pulse time. E.g., the time measurement must be insensitive to the statistically varying SER pulse amplitude. For this purpose, special constant fraction discriminators (CFD) are employed in the path processing the signals from the PMTs.

The time resolution of the counter of several ps is so high, that the resolution of the whole setup is finally limited by the used detector. As already mentioned above, the use of the time correlation avoids the limitation by the SER pulse width. The resolution is then dictated by the random effects occurring in the PMT during the electron creation and multiplication. These processes result in so-called transit time spread (TTS) in the PMT.

For example, the time resolution of our TCSPC counter (BH SPC-150) is 7 ps (FWHM). The real time resolution of the whole setup with the PMT (BH PMC-100) is given by the TTS value of 180 ps of the PMT. This seems but is not in contrast with the PMT SER pulse width of 1.5 ns. Hybrid detectors provide even better time resolution, down to 40 ps.

These resolutions are usually given by some mathematical construct, such as full width at the half maximum (FWHM) or rising slope from 10 to 90% of the signal peak and represent usually an artificial waveform test case. For stable repetitive light pulses however, changes in few tens of picoseconds can be recognised in the data (e.g. relative signal peak shift).

Applications

The main use of the TCSPC technique is currently probably in the biochemistry; photochemistry and photobiology [2]. A historical overviews of TCSPC early applications (and principles) can be found in [3, 4], the technique was at its origin called pulse fluorometry. TCSPC measurement of the fluorescence is one of the key method for identification of photoactive molecules in the samples [2]. Common technique for biochemical tissue investigation is the fluorescence lifetime imaging (FLIM) using time-resolved laser scanning confocal microscopy (with multiphoton absorption). This method can provide a spatial mapping of fluorescence decay times inside the cells. Lifetime changes induced by solvent interaction, conformational changes and different binding state to proteins, peptides or lipids are investigated. Simply stated, three pices of information can be found from the time-resolved fluorescence measurement: which photoactive molecules are inside (emitted wavelengths), what are their concentrations (fluorescence intensities) and in what type of environment they occur (fluorescence decay times).

Enhanced radiative decay rates and enhanced photostability of dyes conjugated to metallic nanoparticles were reported using the TCSPC method. Multi-spectral measurement of low-intensity fluorescence induced by ion beams were made as well [2].

For our scientific field, the low temperature plasma physics, several applications have been developed up to now. Probably the best known example is the application of the TCSPC onto the dielectric barrier discharge at atmospheric pressure air [5]. In that article unique results of sub-nanosecond and sub-millimetre resolved electric field distribution in the single filament of the barrier discharge were obtained. With this result, obtained by the optical emission spectroscopy enhanced by the TCSPC, the local ozone production in the barrier discharge was quantified. Other examples of TCSPC application in plasma physics take advance not only of method's temporal resolution, but also its high sensitivity given by the Poisson statistics driven process of the photon counting in the TCSPC [3]. The ultrafast ionisation process in Trichel pulse mode of negative corona was investigated and the electric field quantified in [6] with the resolution of few tens of picoseconds and 10 μm in space. The streamer head internal structure was revealed using the TCSPC method enhanced by the computer modelling in [7]. While the method of electric field determination in [5, 6] was dependent on kinetic model and required thus additional analysis, the determination of the electric field using direct method was done in [8]. There, the Stark polarisation

spectroscopy was applied onto the radio-frequency discharge in helium in γ-mode. Furthermore, the extremely high sensitivity TCSPC measurements of the pre-breakdown phase of the barrier discharge in helium enabled to detect very weak light emission of approx. 10^{-3} of active discharge signal with average count rate of 1 photon $s^{-1}nm^{-1}$ in [9]. These results revealed a presence of neutral bremsstrahlung in this atmospheric pressure discharge. All these cases demonstrate the power of the TCSPC technique for the applications in high-frequency plasma physics.

Experiments

In the following two experiments, the student will deal with two issues connected to the TCSPC technique. The first one is related to the test of the TCSPC setup itself, to check the resolution and the response delays of the TCSPC system from given defined light emitting source. The second one will be the application of the TCSPC technique onto the plasma physical sample - in this case highly repetitive light emitting discharge. The exact implementation of the below given points will be carried out under the supervision of the specialist.

The resulting report from these investigations should contain relevant short introduction based on the student individual literature research, data evaluation based on the recommendations and literature given by the supervisor and commented results and their discussion.

Tests of TCSPC setup

1. Using the highly repetitive light emitting source (typically a laser diode) for quantification of the TCSPC system resolution. The FWHM value will be determined from the TCSPC measurement for given wavelength of the detected light. This value should be compared with the resolution given as a technical parameter from the manufacturer. Possible deviations should be discussed in the text.
2. Determination of the main sources of the signal delay in the spectroscopic setup. The influence of the different media (air, optical fibres, photocathode of the photomultiplier etc.) under consideration will be quantified. Physical mechanisms responsible for these delays should be analysed in detail and discussed.
3. The limits of the applicability of the TCSPC detection system in relation to frequency ratio of the main and synchronisation signals will be probed. The pile-up effect will be investigated on which the basic idea of the TCSPC will be discussed. The statistical nature of the TCSPC method should be clarified.

Plasma diagnostics

1. High-frequency light emitting discharge setup will be prepared and under the supervision of the specialist started. Selected spectral lines for given discharge gas mixture will be recorded.
2. From the analysis of the two selected spectral signatures (spectral atomic lines or spectral bands of molecular spectra) the basic plasma parameters (electron density or electric field) will be estimated. The possible advantages and drawbacks of the used diagnostic methods will be discussed and their applicability for different plasma sources clarified.
3. Based on the excitation mechanisms of radiative species and estimated values of the basic plasma parameters the fundamental discharge mechanisms of the detected emitting system will be discussed. The Townsend theory and the theory of streamers and ionising waves should be taken into account. Basic discharge characteristics should be discussed in comparison to the know discharge types, such as glow, Townsend, capacitively coupled radio-frequency or streamer discharge.

Reference

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