Single photon timing system for picosecond fluorescence lifetime measurements[a)]

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A single-photon timing system is described which is capable of extracting fluorescence lifetimes as short as 25 ps. The system is an improved version of an earlier apparatus. The new system uses a synchronously pumped, mode-locked dye laser with 10-ps pulses operating at 82-MHz repetition rate. A fast photodetector and a leading-edge discriminator were developed to use with this light source. Also, a special rate reduction circuit was built to eliminate large oscillations in fluorescence decay spectra due to the excessive stop rates that overload commercial time-to-amplitude converters.

PACS numbers: 78.55. – m

The single-photon system described in this article was set up to study the very fast fluorescence decay kinetics present in photosynthetic systems.[1,2] The new system is a modification of our previous single-photon timing system[3,4] to include a synchronously pumped mode-locked dye laser. The output pulses of this laser have a full-width half-maximum (FWHM) of about 10 ps. The new system is capable of extracting fluorescence lifetimes as short as 25 ps.[1]

Major modifications to our earlier single-photon detection apparatus[3,4] were the design of new photodetector circuitry, a fast discriminator, and a new rate reduction circuit. The rate reduction circuit was required to eliminate oscillations in the measured fluorescence decay.

A block diagram of the system is shown in Fig. 1. The light source consists of a Spectra Physics SP 171 argon ion laser mode locked with a Spectra Physics SP 362 Ultrastable Mode Locker driving an acousto-optical mode-locking crystal. A modified Spectra Physics SP 375 dye laser, pumped by the mode-locked argon laser, produces light pulses of 10 ps FWHM[1] at an 82-MHz repetition rate.

The laser pulses illuminate samples in the single-photon detection apparatus.[3,4] An RCA C31034A photomultiplier detects the fluorescence photons. The constant fraction discriminator[4] shapes the single photon pulses generated by the photomultiplier. The original constant fraction discriminator needed no modification. A fast

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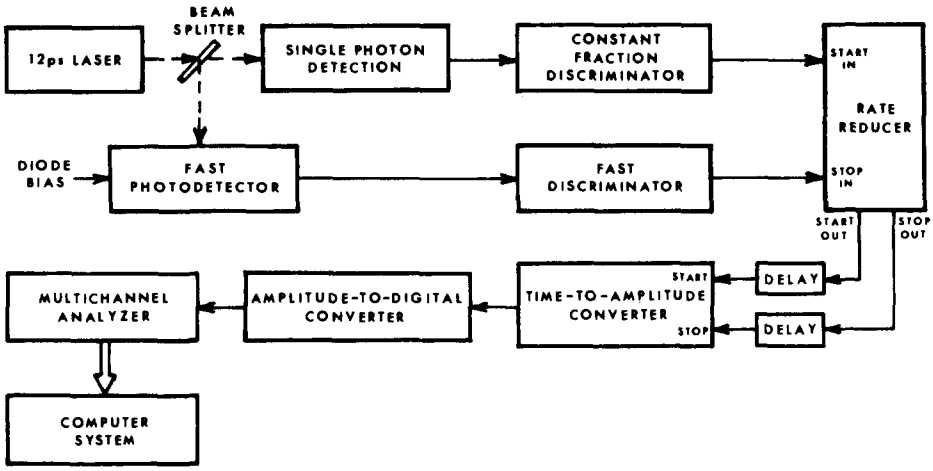

FIG. 1. Picosecond fluorescence lifetime-system block diagram.

diode photodetector picks up the excitation light from a beam splitter, producing subnanosecond pulses at a repetition rate of 82 MHz. The detector is a Texas Instruments TIED photodiode that has been mounted in a fashion similar to that described by Steinmetz.[5]

A fast tunnel diode discriminator was built and directly attached as an extension of the detector output connector. The losses and problems associated with transmission of such short pulses by a cable between the photodiode and a remote discriminator are thus eliminated. The discriminator circuit is shown in Fig. 2.

Conventional single-photon timing systems start a voltage ramp in the time-to-amplitude converter (TAC) upon each excitation pulse and stop the voltage ramp when a fluorescence photon is detected.[6] Because the 82-MHz repetition rate of the laser pulse is too high for the TAC to trigger a ramp on each pulse, we have adopted a reverse single-photon timing scheme. We start the TAC with the output of the constant fraction discriminator (fluorescence photon) and stop the TAC with the next pulse from the fast discriminator, i.e., with the next laser pulse. However, the very high stop rates present at the TAC input resulted in poor performance of two different commercial time-to-amplitude converters. A rate reduction circuit was built to eliminate this problem. This rate reducer gates out all of the stop pulses except the one immediately following the fluorescence photon start pulse. The converter thus never gets a stop pulse before the start pulse is first accepted. Normally, this will result in oscillation free performance of the TAC.

The discriminator outputs are as follows: a single-photon start rate usually less than 20 kHz and an extremely high stop rate of 82 MHz. Two different TACs were tried (ORTEC, Inc. Model 457 Biased Time-To-Pulse-Height Converter and Canberra Industries, Inc. Model 2043 Time Analyzer). Both converters show large oscillations in the measured fluorescence decay curve. The period of the oscillation was about 2 ns. Apparently the 82 MHz stop pulse train continually present at the stop input of the TAC interfered with the start input.

This problem could be minimized in some cases by shifting the start and stop pulses from one another by a fixed delay and away from the cross talk region. However, this was not possible in this case because the separation between the stop pulse is only 12 ns, limiting the useful time range to less than 10 ns.

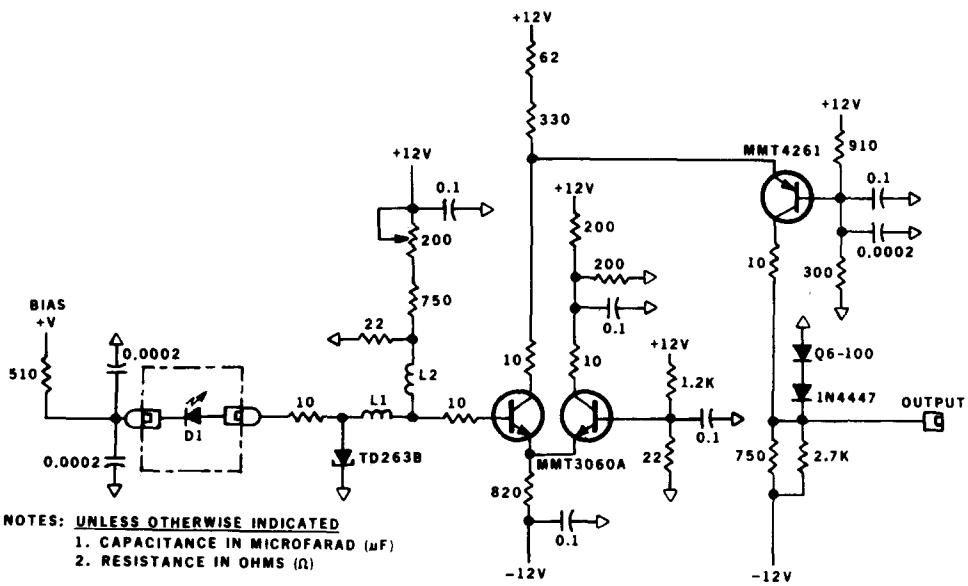

FIG. 2. Fast photodetector and tunnel diode discriminator circuit diagram.

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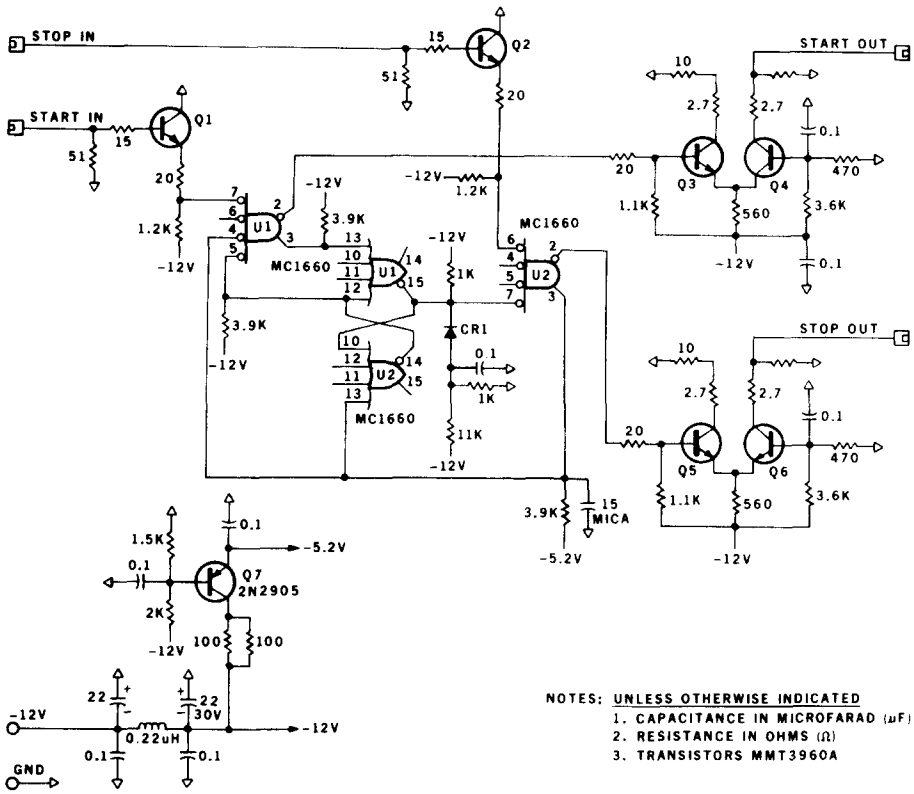

FIG. 3. Rate reducer circuit diagram.

A stop rate reduction circuit was built (Fig. 3), which gates out all of the stop pulses prior to the arrival of the single-photon pulse from the constant-fraction discriminator. The reducer is placed between the discriminators and the TAC (Fig. 1). Only one stop is generated after each single-photon count, eliminating cross talk in the TAC. The oscillations in the fluorescence decay spectrum are thus virtually eliminated.

The Canberra Model 2043 Time Analyzer was used because of the shorter deadtime (permitting higher processing rates) and superior immunity to cross talk oscillations. The analyzer had to be modified slightly.

Our picosecond fluorimeter has been used extensively to study fluorescence decay kinetics in photosynthetic systems.[1,2] Oscillation-free performance is only possible by using some form of rate reduction. We have reduced the rate of discriminator pulses reaching the TAC and thus were able to avoid the necessity of reducing the laser pulse rate with a cavity dumper. We have been able to resolve fluorescence decay kinetics of chloroplasts consisting of three exponential components in the range of lifetimes from 50 to 3000 ps. The published results[1,2] show excellent kinetic fits with little evidence of instrument distortion.

We wish to thank B. Leskovar for advice in the initial stages and Dr. Kerry K. Karukstis for helpful discussions and for her careful reading of the manuscript. This work was done in the Laboratory of Chemical Biodynamics and the Electronics Research and Development Group of the Lawrence Berkeley Laboratory, and was supported by the U.S. Department of Energy under Contract No. DE-AC03-76SF00098. Reference to a company or product name does not imply approval or recommendation of the product by the University or the U.S. Department of Energy to the exclusion of others that may be available.

[a] A more detailed version of this report is available as Lawrence Berkeley Laboratory Report LBL-14646.

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