Rejection of stochastic background noise in low-level pulsed light scattering experiments

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We present an electronic scheme for reducing stochastic noise in the detection of low-level light signals in experiments where the signal coincides in time with a probe laser pulse. This scheme has been applied successfully in Raman experiments to reject the noise generated by afterpulsing in photomultiplier tubes as well as by unwanted laser-induced fluorescence.

I. INTRODUCTION

Among the methods for low-level light detection with photomultiplier tubes, digital photon counting and analog charge integration are the two most commonly used. Although photon counting can provide an excellent signal-to-noise ratio in continuous-wave experiments, charge integration must be used when the distribution of the arrival time of the photons is too narrow for digital photon counting. This is usually the case when short, intense laser pulses are used to achieve high temporal resolution. We have used charge integration with an Amperex XP2020Q high-gain photomultiplier to detect spontaneous Raman photons scattered from infrared multiphoton excited molecules.[1–3] Since the signal level is extremely low (0–3 photons per laser shot), good rejection of stochastic background noise is important.

There are two main sources of stochastic noise in our experiment: laser-induced fluorescence and afterpulses from the photomultiplier tube.[4] The laser-induced fluorescence, which is particular to our experiment, originates from dissociation fragments of the sample gas. Afterpulses originate from collisions between photoelectrons and residual atoms or molecules in the photomultiplier. The collisions generate positive ions which are accelerated toward the photocathode and induce the emission of secondary electrons. Afterpulses increase with time because of the diffusion of He atoms into the photomultiplier, especially in a He-rich environment. This is a general problem with any low-level light detection involving high-gain photomultiplier tubes. Both sources of noise are stochastic in nature and because of their high intensity, both dramatically reduce the signal-to-noise ratio, even if the noise appears in only a small percentage of the laser shots.

The desired spontaneous Raman signal coincides in timing with the 20-ns probe laser pulse. On the other hand, afterpulses in the photomultiplier appear *after* the real signal generated by the photoelectrons. The laser-induced fluorescence too appears after the probe laser pulse, because the lifetime of the fluorescence greatly exceeds the probe pulse. Therefore, to distinguish between the desired signal and the unwanted noise, one should monitor the coincidence of the photomultiplier signal with the probe pulse: photomultiplier output pulses not in coincidence with the probe pulse should be rejected as noise.

We developed a noise rejection system to monitor the coincidence between the photomultiplier output and the Raman probe pulse on a shot-by-shot basis. The design challenge is to both monitor the coincidence of the pulses and to measure their integrated intensity simultaneously. This requires processing the 0.01–0.05 V photomultiplier pulses in both a low-impedance nanosecond coincidence circuit *and* in a charge integrator requiring a (high impedance) current source. Specifically, to perform electronic logic operations with nanosecond precision the output of the photomultiplier must be preamplified using a timing amplifier. The system therefore needs to be designed in such a way that the timing amplifier does not affect the operation of the charge integrator.

II. ELECTRONIC SCHEME

Figure 1 shows a block diagram of the noise rejection system. The signal from the photomultiplier tube (PMT) is sent to a charge integrator consisting of a low pass filter and a low noise amplifier. For most of the frequency components of the photomultiplier pulses the charge integrator appears as a 50-Ω cable terminator, therefore it need not be placed very close to the photomultiplier. The photomultiplier signal is also amplified by a wideband 300-MHz timing amplifier, and the amplified output is sent to a constant-fraction discriminator (EG&G Ortec model 934) to record the arrival time of the photons. The time interval when the probe pulse is present is monitored by a photodiode and another constant-fraction discriminator. The outputs from both constant-fraction discriminators are compared in the coincidence circuit described below. If any of the photomultiplier output pulses appears outside a window determined by the probe pulse, the coincidence circuit sends a flag to the computer and the data are rejected.

Special care must be taken that the timing amplifier does not interfere with the charge integrator. If the input impedance of the timing amplifier is too high, it will not have the required fast temporal response. On the other hand, if the impedance is too low the charge-decay time of

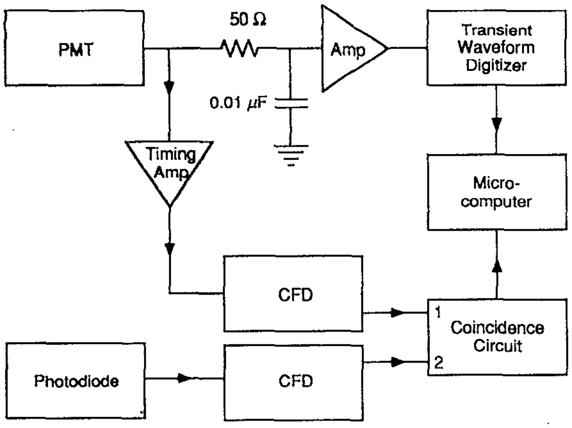

FIG. 1. Block diagram of the noise rejection system. PMT = Amperex XP2020Q photomultiplier, timing amplifier = modified ORTEC 574 amplifier. The constant-fraction discriminators are two of the four channels in an ORTEC 934 module. The transient wave form digitizer can be replaced by a sample-and-hold circuit. Details of the coincidence circuit are shown in Fig. 2.

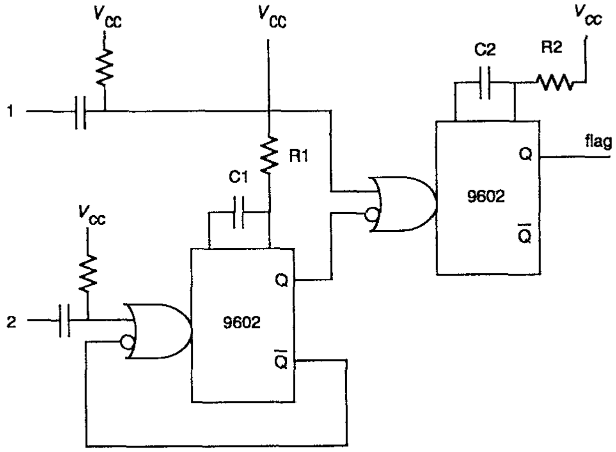

FIG. 2. Circuit diagram of the coincidence circuit. A pull-up resistor and a capacitor coupler are used in front of each one-shot (9602) circuit to shift the negative voltage input pulses into negative logic pulses. The first one-shot circuit triggers at the end of the input pulse from terminal 2 (the rising edge of a negative logic pulse). The output pulse of this one-shot lasts for 400 ns. If there are any pulses coming from terminal 1 during these 400 ns, the second one-shot circuit triggers and sends a flag signal to the computer.

the integrator becomes too short to record the integrated signal accurately. The timing amplifier used here is an ORTEC 574 amplifier, with its 50-Ω input terminator removed. The modified amplifier has a 2.5-kΩ input impedance, resulting in a 25-μs (2.5 kΩ × 0.01 μF) charge-decay time constant. This is long enough for a transient wave form digitizer or a sample-and-hold circuit to record the signal. In addition, because the 2.5-kΩ input impedance of the timing amplifier is much higher than that of the charge integrator (50 Ω), the amount of charge taken away from the charge integrator by the amplifier is negligible. When the amplifier is placed close to the integrator, its time response will not be affected by the cable configuration since the charge integrator terminates the cable correctly at 50 Ω.

Figure 2 shows the details of the coincidence circuit. It consists of two one-shot circuits. The first one triggers at the end of the probe pulse. Using R1 and C1, the output pulse is made long enough to cover the interval where afterpulses and fluorescence might appear (400 ns in our experiment). If any signal appears in this range, the second one-shot circuit triggers and the flag signal will be low. The length of the flag pulse can be adjusted with R2 and C2. Since the output pulse from the constant-fraction discriminator is of negative voltage, a pull-up resistor and a capacitor coupler are used in front of the one-shot circuits to shift the level. The output pulse width of the constant-fraction discriminator channel monitoring the photomultiplier tube should be made as short as possible to respond to the individual photons. The output pulse width of the second constant-fraction discriminator channel monitoring the photodiode, however, need not be identical to that of the probe pulse. The pulse width should be adjusted to compensate for the time delay due to the intrinsic path difference between the photomultiplier signal and photodiode signal.

The noise rejection system discussed above has been successfully applied in the Raman experiments described in Refs. 1–3. With this system, the signal-to-noise ratio of our experiment has been improved by at least a factor of 10. This has enabled us to extend our studies to higher levels of vibrational excitation, when laser-induced fluorescence from dissociation fragments tends to obscure the Raman signal. It also allows us to extend the useful life of expensive photomultiplier tubes by rejecting the afterpulse noise resulting from the aging of the tubes. The application of this noise rejection system is not limited to Raman experiments. It can be applied to any pump and probe experiment with very low-level light detection, as long as the desired signal is in coincidence with the probe.

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