Ultrafast three-photon counting in a photomultiplier tube

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We demonstrate experimentally ultrafast three-photon counting by three-photon absorption in a GaAsP photomultiplier tube at the wavelength range of 1800–1900 nm, and analyze its sensitivity and time response. Pulse energy of ~500 fJ is shown to be detectable for ultrafast 170 fs pulses. The presented three-photon counter may serve as a unique tool for ultrafast quantum state characterization as well as for ultrasensitive third-order temporal measurements.   © 2011 Optical Society of America
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Photon counting is the key enabler of the revolution in experimental quantum optics, including the variety of quantum information applications achieved over the past decade [1]. It also plays an important role in classical applications involving extremely low light intensities in a wide range of fields, including biological imaging [2] and optical communications [3,4]. Single-photon states are widely employed in quantum information technologies such as quantum key distribution over optical fibers, where the ability to detect single photons is crucial, and many applications require photon number resolving (PNR) detectors [5]. Moreover, significant progress has been made in realization and manipulation of higher-photon-number states [6–9], which provide unique solutions for enhanced-sensitivity metrology [10–12] and quantum computing [13]. There have been several approaches to developing photon counters that can detect high-photon-number states. Some approaches are based on splitting a single optical mode into several temporal [14,15] or spatial modes [16], allowing number resolving by electronic coincidence measurements among several Geiger-mode avalanche photodiodes (APDs), by an APD array [17], or by a streak camera [18]. Other approaches employ characterization of the electrical response to different photon numbers in superconductor detectors [19], APDs [20], or visual light photon counters [21].

Although very high performance has been achieved in terms of efficiency and low dark counts, the time resolution of all the existing detectors is limited by the electronic response to be longer than several picoseconds, so that two single-photon pulses separated by less than the time resolution of the detector will be detected as a two-photon state by a PNR detector of the types described above. Furthermore, the best performance is obtained for short-wavelength light in the visible range, whereas IR photon counting remains extremely difficult, requiring sophisticated approaches, such as upconversion [22,23]. Nonlinear optical frequency conversion has the advantage of the ultrafast femtosecond-scale response; however, the weak nonlinearities of insulating materials limit the efficiency of such schemes. Semiconductor multiphoton absorption processes, where several low-energy photons are absorbed in a single-electron transition, have the advantage of being low-order resonant transitions and resulting in direct conversion of optical to electrical signals, nevertheless maintaining the femtosecond time resolution of nonlinear optics. Two-photon absorption

(2PA) in a photomultiplier tube (PMT) has made a breakthrough in high-sensitivity ultrafast optical measurements [24]. This method was recently exploited for ultrafast and wideband quantum characterization of different light sources [25–27], which could not have been achieved by conventional methods, and was shown to enable laser-assisted photon counting in the IR [28,29]. The process of three-photon absorption (3PA) in semiconductors can be used as a unique tool for ultrafast detection of photon triplets (Fig. 1 inset). Moreover, 3PA allows the extraction of the temporal shape of a femtosecond pulse without direction-of-time ambiguity and with no need for spectral measurements [30,31], much more efficiently and compactly than the traditional third-harmonic generation [32]. Therefore, enhancing the sensitivity of 3PA opens a wide range of applications in ultrafast and quantum optics.

Here we present a three-photon counter, in which input photons are absorbed as triplets by 3PA in a PMT with a semiconductor-based photocathode. We characterize the sensitivity and response time of the three-photon counter at the short-IR wavelength range of 1800–1900 nm with 170 fs pulses generated by an optical parametric oscillator (OPO) at 81 MHz. The detector
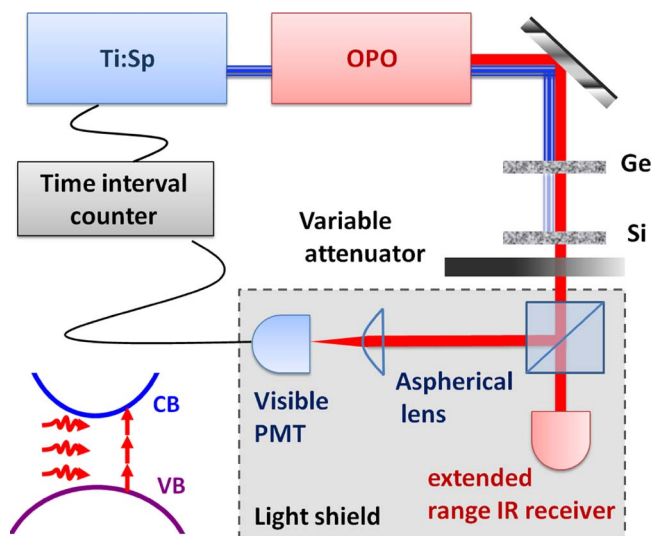

Fig. 1.   (Color online) Experimental setup for three-photon counting. The inset is a schematic diagram of the resonant third-order transition of 3PA between the valence band (VB) and the conduction band (CB).

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employed in our experiment is a commercial GaAsP PMT (Hamamatsu H7421-40) with an optimal dark-count level below 50 counts/s at 273 K, designed for efficient single-photon detection at the wavelength range of 300–720 nm. Its bandgap enables significant 3PA at the examined wavelength range. The photocurrent emitted from the GaAsP photocathode is in general given by

$$J(t) = \alpha P(t) + \beta \frac{P(t)^2}{S} + \gamma \frac{P(t)^3}{S^2}, \tag{1}$$

where P is the incident power; S is the spot size on the detector; and α, β, and γ are the one-, two-, and three-photon coefficients of the semiconductor. In our experiments, because photon energies are well below half the energy of the bandgap, considering the band tail, the first two terms in Eq. (1) are negligible. In order to assure that no residual visible or near-IR light from the OPO remains in the detected beam, both Si and Ge filters were used (Fig. 1). As can be seen from Eq. (1), the sensitivity of the three-photon detection is improved by the spot size; therefore, an aspherical lens with a relatively high NA, 0.66, was used to focus the beam on the photocathode.

Measurement of the number of counts due to 3PA at 1800 nm, as a function of the mean power monitored simultaneously through a beam splitter using InGaAs photoreceiver extended to 2.2 μm (Fig. 1), resulted in a cubic dependence (Fig. 2) and a clear signal above the dark-count level for pulse energies down to 500 fJ. The responsivity of the detector was measured for the spectral range of 1800–1900 nm and a dark-count level of 500 counts/s was subtracted (Fig. 2 inset), showing good agreement with theory [33], giving for the leading term:

$$\gamma \propto (3E_{ph} - E_g)^{\frac{1}{2}}, \tag{2}$$

where E_{ph} is the energy of the photon and E_g is bandgap energy, taken in our calculations to be 1.95 eV, as was estimated from the behavior of the one-photon absorption (1PA).

Because the presented detector is highly attractive for ultrafast measurements, its time response characteristics are of great importance. Although the absorption time is much faster than response times of any electronic circuitry, the time jitter of the PMT response is affected by two major factors: the time spread in the electron multiplication process, and the time spread in the photoelectron emission from the photocathode determined by the drift and diffusion times to the surface of the photocathode. These times are dependent on the spatial distribution of the charge generations within the semiconductor photocathode. 1PA occurs on a scale of 1 μm due to the large absorption coefficient. Because the 3PA coefficient is small, the three-photon effective absorption length is determined by the Rayleigh length, which is estimated in our experiment to be on a scale of tens of micrometers. Therefore, the three-photon generated carrier distribution should have a much wider spread than the one-photon distribution, causing a possible increase in the time spread of the photoelectron emission from the photocathode.

To characterize the 1PA and 3PA detection time jitter, we have performed time-resolved photon counting measurements. For this characterization we used a start–stop setup where time interval histograms between photon detections and generated femtosecond pulses were measured using a time interval and frequency counter (SR620 Stanford Research Systems) (Fig. 3). For 1PA, the measured time jitter has an exponential decay of ~210 ps (FWHM 350 ps), whereas the 3PA detection has a time jitter with an exponential decay of ~250 ps (FWHM 390 ps). Therefore, the photoelectron diffusion in the photocathode was shown to have only a minor effect on the overall 3PA detection time response relative to the 1PA detection. It should be noted that every point on the 3PA graph in Fig. 3 represents absorption of three photons that occurred on a time scale of femtoseconds, and the relatively large time scale of the jitter is just a consequence of the processes following the absorption; therefore, it does not prevent ultrafast measurements, similar to those conducted in [26].
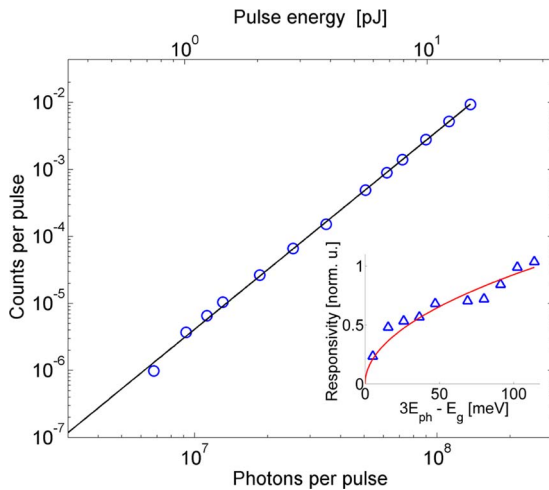

Fig. 2. (Color online) Three-photon counting efficiency on a logarithmic scale. The solid black line is a fit to a power of 2.95. The inset is the normalized detector responsivity for the wavelength range of 1800–1900 nm, equivalent to energies of 5–115 meV above the bandgap.
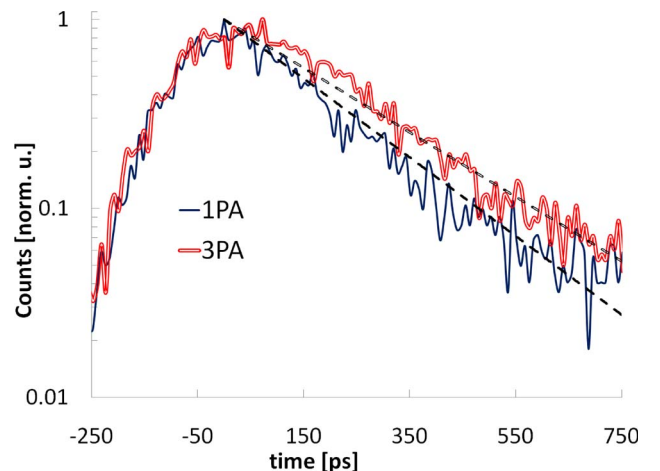

Fig. 3. (Color online) Time-resolved one-photon and three-photon counting. The dashed lines are exponential fits with 210 and 250 ps time constants for 1PA and 3PA, respectively.

The efficiency of the three-photon counter can be further enhanced by several simple technical improvements, namely, decreasing the dark count to its optimal value, adding suitable antireflection coating on the front glass of the detector, and redesigning the distance between the glass and the photocathode to allow better focusing. Another enhancement can be achieved by employing gated measurements synchronized with the source for pulsed light. The time-resolved measurement of Fig. 3, for example, results in 1 order of magnitude higher sensitivity. The method can also be easily implemented for the telecom wavelengths around $1.55\,\mu$m by bandgap engineering.

In conclusion, we have demonstrated experimentally three-photon counting by 3PA in a commercial GaAsP PMT and characterized its spectral and temporal response. This novel detector may pave the way for classical and quantum applications in ultrafast optics and for unique quantum characterization of light sources.

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