BNL-113919-2017-JA

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Submitted to Proc. SPIE 10212

May 2017

Physics Department

Brookhaven National Laboratory

**U.S. Department of Energy
USDOE Office of Science (SC),
High Energy Physics (HEP) (SC-25)**

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Time stamping of single optical photons with 10 ns resolution

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ABSTRACT

High spatial and temporal resolution are key features for many modern applications, e.g. mass spectrometry, probing the structure of materials via neutron scattering, studying molecular structure, etc.^{1–5} Fast imaging also provides the capability of coincidence detection, and the further addition of sensitivity to single optical photons with the capability of timestamping them further broadens the field of potential applications. Photon counting is already widely used in X-ray imaging,⁶ where the high energy of the photons makes their detection easier.

TimepixCam is a novel optical imager,⁷ which achieves high spatial resolution using an array of 256×256 $55 \mu\text{m} \times 55 \mu\text{m}$ pixels which have individually controlled functionality. It is based on a thin-entrance-window silicon sensor, bump-bonded to a Timepix ASIC.⁸ TimepixCam provides high quantum efficiency in the optical wavelength range (400-1000 nm).

We perform the timestamping of single photons with a time resolution of 20 ns, by coupling TimepixCam to a fast image-intensifier with a P47 phosphor screen. The fast emission time of the P47⁹ allows us to preserve good time resolution while maintaining the capability to focus the optical output of the intensifier onto the 256×256 pixel Timepix sensor area. We demonstrate the capability of the (TimepixCam + image intensifier) setup to provide high-resolution single-photon timestamping, with an effective frame rate of 50 MHz.

Keywords: Timepix, TimepixCam, fast imaging, single photon imaging, photon counting

1. INTRODUCTION

The emergence of fast imaging devices with nanosecond-scale time resolution has enabled a multitude of novel research techniques, ranging from optical tomography in medicine, to quantum cryptography, thereby advancing information technologies. Many of these fields would benefit immensely from fast imaging devices which are sensitive to single optical photons.

Single photon counting has been successfully used, for example, in X-ray imaging, where incident photons have sufficiently high energy for the detectors to register photons without intrinsic amplification. While a single X-ray photon creates thousands of charge carriers in the sensor, it would require thousands of optical photons, i.e. a flash bright enough, to use the same approach for optical photons. This therefore poses a technological challenge for obtaining photon counting capabilities for individual optical photons.

This manuscript presents the timestamping of single optical photons with high time resolution. To achieve this goal, a novel imaging device, TimepixCam, that has demonstrated a time resolution of 20 ns, was paired with an image intensifier with a fast P47 phosphor screen.

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2. TIMEPIXCAM DESCRIPTION

TimepixCam is an optical imaging device which allows timestamping of incident photons with 10 ns-scale time resolution.⁷ It consists of a specialized thin-entrance-window sensor¹⁰ bump-bonded to a Timepix ASIC⁸ and read out by an X-Ray Imatek UNO camera. The camera is outfitted with a lens to project the image of the intensifier screen onto the sensor.

The readout chip is a 256×256 pixel grid, each pixel of which can be individually controlled. The size of each pixel is $55 \mu\text{m} \times 55 \mu\text{m}$, providing high spatial resolution. The Timepix chip can operate with a maximum readout clock rate of 100 MHz, corresponding to a time resolution of 10 ns.

2.1 Sensor

The TimepixCam sensor is a $300 \mu\text{m}$ thick p-on-n type silicon sensor with high resistivity. Figure 1 shows the sensor bump-bonded to the Timepix chip, and the assembly mounted on and wire bonded to a daughter board in the camera. The sensor can be easily depleted by reversely biasing the p-n junction.

Figure 1: Photograph of the TimepixCam chip wire bonded to the daughter board of the X-Ray Imatek UNO camera.

A key feature of the TimepixCam sensor is its thin entrance window, which allows for transmission of the incident photons to the sensitive volume of depleted silicon. Photodiode capacitance measurements have shown the typical voltage required for the depletion of the silicon to be about 25 V. A radiation-hard design developed for particle physics applications has been employed for the fabrication of the TimepixCam sensors, which makes them capable of withstanding bias voltages of up to 500 V. In our experiments, a bias of 80 V was used to ensure full depletion of the silicon, and therefore a better sensitivity to the shorter wavelength photons.

In order to improve the sensitivity, i.e. the conversion rate of the incident photons into photoelectrons in the sensor, a layer of anti-reflective coating (ARC) was applied to every sensor. The ARC properties were optimized for the 430 nm wavelength emitted by the P47 phosphor, employed in the image intensifier used in this study.

2.2 TimepixCam

The TimepixCam readout is based on the XRI UNO X-ray camera manufactured by X-Ray Imatek.¹¹ The bottleneck of the readout chain is the USB interface and associated camera software, which limits the maximum continuous readout rate to about 10 frames per second.

Figure 2 shows the image intensifier in a custom 3D-printed enclosure and TimepixCam, which was focused at the output window of the intensifier. The enclosure was needed to reduce the ambient light, and thereby the spurious single photon counts in the dark.

The camera is equipped with an external trigger, which is used to synchronize the shutter, i.e. the time period when the camera is sensitive to light, with the photon sources such as lasers or light emitting diodes (LED).

Figure 2: TimepixCam and the image intensifier in the custom enclosure to keep it from ambient light during the experiment.

2.3 Image Intensifier

In order to gain sensitivity to single optical photons, and at the same time preserve the high time resolution of TimepixCam, we used a P47 image intensifier, Hamamatsu V9569U71N270, to convert the single photons to light flashes for the camera. The intensifier has a GaAs photocathode with an effective area of $16 \times 16 \text{ mm}^2$ followed by a two-stage micro-channel plate (MCP) and a P47 phosphor. Light incident on the photocathode is converted to photoelectrons with approximately 25% efficiency. The electrons are accelerated by the 200 V potential between photocathode and MCP, with the voltage across the MCP controlling the gain of the image intensifier. Electrons exiting the MCP are accelerated by a 6 kV potential applied between the MCP and the P47 screen, and hit the screen, causing a phosphorescent flash that is then detected by the camera.

The advantage of the P47 phosphor is its fast response time, with a signal rise time of 7 ns,¹² which minimizes the image intensifier's contribution to the final time resolution. Its emission has maximum at a wavelength of 430 nm, with a decay time of about 110 ns.⁹

Figure 3 shows the quantum efficiency of the GaAs photocathode in the image intensifier.¹³ It is well suited for multiple applications, which need sensitivity for photons in the range 450 - 850 nm.

3. QUANTUM EFFICIENCY

Quantum efficiency (QE) is a key characteristic of photon detecting sensors. It is defined as the ratio of the number of photoelectrons produced by the photons striking the sensor to the number of those photons. The quantum efficiency can be estimated by direct measurement of the produced photocurrent and precise measurement of the incident flux with an absolutely calibrated source. Another estimator of the quantum efficiency is the reflectance of the sensor's surface in the wavelength range of 500 - 800 nm, where the absorption on the surface, and silicon's transparency are sufficiently small. In this region, we can express the QE as $1 - R$, where R is the reflectance.

Figure 4a shows the measured quantum efficiency for four photodiodes with different treatments of the surface, produced in the same process as the camera sensors.¹⁰ As expected, the samples with the ARC have a much higher QE; over 90% for most of the wavelength range. The effect of the thickness of the passivation layer on the quantum efficiency is also very pronounced. In both pairs of samples (with and without the ARC), the QE of the sensor with the thicker passivation is consistently lower. The roll-off in the QE curve at the longer wavelengths is due to the increased transparency of silicon for those wavelengths, and is also expected.

Figure 3: Quantum efficiency of the GaAs photocathode in the image intensifier as a function of the incident light wavelength.

Figure 4: Quantum efficiency of the TimepixCam sensors as a function of wavelength with and without ARC.

Figure 4b shows the reflectance measurement results. These measurements were made using a spectroscopic ellipsometer, J.A.Woollam M2000D. This device can measure the reflectance of the polarized incident light from 45 to 72 degrees from normal to the surface. The results of the $1 - R$ measurement, at 45 degree incidence angle, for the samples with and without the ARC, agree with the direct QE measurements. The calculated $1 - R$ result

for the zero degree incident light is also shown in Fig. 4b, which can be directly compared to the QE.

4. SINGLE PHOTON IMAGING

4.1 Single Photon Demonstrator

For the single photon counting experiment we used an LED synchronized with the TimepixCam shutter. The duration of the LED pulses was 50 ns and the pulse voltage was varied from 0 V to 10 V. The light pulse was reflected from a white surface to further reduce the number of incident photons, and was directed to the image intensifier through a 2 mm hole.

This experiment was intended to demonstrate the sensitivity of the intensifier/TimepixCam combination to single photons. A single photon incident on the image intensifier typically results in a cluster of hit pixels with a signal large enough to cross the threshold and thus record a timestamp. The number of clusters at a particular LED voltage should be distributed according to Poisson statistics around a corresponding mean value. If the photons follow Poisson statistics, the mean number of photons should be equal to their variance from frame to frame. This property should be independent of the photon flux, and can be expressed in a so-called photon transfer curve (PTC). Linearity of the PTC would demonstrate that we have signal from single photons which follow Poisson statistics.

Figure 5: Photon transfer curve. Each point represents the measurement with a particular LED voltage.

Figure 5 shows the PTC measurements, and for LED voltages up to 5 V it is linear with a slope of 0.95 ± 0.05 . In this region we can therefore claim good agreement with the predicted single photon counting result. At higher voltages, the number of clusters in the sensor increases and, due to the finite size of the clusters, they start merging. As a result, the distribution of the number of clusters diverges from being Poissonian. Thus, the PTC non-linearity at high LED voltages is an expected consequence of the finite size of the single photon clusters on the pixel array.

4.2 Perovskite Sample

With the demonstrator experiment showing a PTC in agreement with a single photon signal, we used our setup to perform a perovskite lifetime measurement. Perovskite crystals are promising photovoltaic materials for use as active layers in solar cells.¹⁴

The sample we used was a hybrid inorganic-organic perovskite halide photovoltaic. Lead methylammonium tri-iodide $CH_3NH_3PbI_3$ perovskites were prepared following published recipes.^{15,16} First we synthesized CH_3NH_3I by reacting methylamine and hydriodic acid in a round bottom flask for 2 hours while stirring. The resulting precipitate was recovered by placing in a rotary evaporator followed by filtration. The resulting solid was dried in a vacuum oven. The $CH_3NH_3PbI_3$ perovskite final solution was achieved by dissolving PbI_2 and CH_3NH_3I in N,N-dimethylacetamide for 12 hours while stirring. The perovskite solution was spin-coated onto solvent-cleaned soda lime glass (SLG) substrates using different deposition techniques, such as solvent-solvent engineering, hot casting, or thermal evaporation.

The sample was excited by a 10 kHz sapphire laser pulse at 532 nm. The fluorescent light was focused by an Olympus IX81 microscope with 40×0.6 NA lens and recorded by TimepixCam using the image intensifier.

Figure 6: Histogram of single photon timestamps for the perovskite sample. Fit lines : red - noise background, blue - other systematic effects, magenta - exponential decay function.

Figure 6 shows the preliminary results of the time distribution of measured single photons emitted by the sample. A simple fit of the signal lifetime represented by an exponential, a constant background and a Gaussian background, results in an estimate for the sample lifetime of 560 ± 15 ns, which is within expectations.

5. SUMMARY

In this work, TimepixCam was paired with a P47 image intensifier and the ability to image and timestamp single photons was demonstrated. The perovskite lifetime-measurement test-application, though still in need of more rigorous analysis, showed agreement with the expected lifetime of these photovoltaic materials.

ACKNOWLEDGMENTS

We would like to thank P.Takacs and T.Tsang for helping with the QE measurements. This work was supported by the BNL LDRD grant 13-006 and used resources of the Center for Functional Nanomaterials, which is a U.S. DOE Office of Science Facility, at Brookhaven National Laboratory under Contract No. DE-SC0012704.

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