Gold nanoparticle surface deposition induced quantum efficiency enhancement for Si-multipixel photon counters

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Abstract: With recent development of nanotechnology, novel devices with nanostructures arise to improve the performance of photodetectors. Here, we demonstrated that by surface decoration with gold nanoparticles on the active area, the quantum detection efficiency of a multi-pixel photon counter was increased due to surface plasmon resonance enhancement. The deposited gold nano-particles actually brought about almost the same enhancement factor for any photon-number fields. As a result, the photon-number-resolving capability of the multi-pixel photon counter was well reserved with the gold nano-particle deposition induced efficiency augment. This result provides guidance to the development of the high-efficiency photon-number-resolving detectors.

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1. Introduction

Recent research shows that the application of metallic nanoparticles covers an increasing variety of fields due to the striking features of the materials in nanometer scale [1–7]. Among all kinds of features, the surface plasmon resonance enhancement effects are well studied and implemented in different fields. In particular, Au or Ag nanoparticles have been used on the photovoltaic devices for plasmonic light harvesting [8–13]. And these techniques can also be employed to improve the performance of single-photon detection. For instance, single plasmons on chip were used to increase the single-photon detection speed [14]. The photon detection efficiency is one of the most important parameters of a single-photon detector, which is especially crucial in the quantum key distribution and photon-counting classical optical communication. The increase of the quantum detection efficiency can largely extend the communication distance. And with the advances in quantum information processing technique, single-photon detectors with photon-number-resolving capability are greatly in need. Multi-pixel photon counter (MPPC) is a recently developed photon-counting device, which is not only able to respond to a single photon but also capable of distinguishing the number of photons in each pulse. A typical MPPC consists of multiple Silicon avalanche photodiode (APD) pixels operating in Geiger mode. Each APD pixel of the MPPC produces an avalanche current pulse when it detects a photon. The output signal from the MPPC is the sum of the avalanche current from all APD pixels. The amplitude of the peak output voltage is proportional to the number of detected photons [15,16]. At present, the quantum detection efficiency of MPPC is not as high as a single Si-APD mainly due to the relatively low filling factor of the active area, which limits its applications in quantum optics.

In this letter, we report on surface plasmon induced augment of the detection efficiency of a photon-number-resolving MPPC. The surface plasmon was generated by the Au nanoparticles deposited on the active area of the MPPC. The detection efficiency has been increased by about 2.76 times with appropriate deposition of Au nanoparticles on the surface of the active area of the silicon MPPC. The quantum detection efficiency augment resulted in no deleterious effects on the photon-number-resolving capability, manifesting that the deposited Au nano-particles brought about a linear response for the surface plasmon enhancement of the photon-number resolving detection. This result may find promising applications in the design of high-efficiency photon-number-resolving detectors.

2. Experiment section



Fig. 1. (a) Topographical images of a single Au nano-particle. (b) Extinction spectrum of Au nanoparticles with diameter of 200 nm in water solution.

In order to increase the detection efficiency of the MPPC at longer wavelengths since the photon-number resolving detection at near infrared wavelengths is quite interesting in quantum optics not only for open-air quantum cryptography but also for quantum storage using alkali metal atoms, we chose the Au nanoparticles with diameter of 200 nm (NanoSeedzTM, HongKong) due to its absorption peak in the near infrared. The Au nanoparticles deposited on the MPPC were about 200 nm in diameter as shown in Fig. 1(a). In general, the resonant peak of the metallic nanoparticle is dependent on the size and shape of the nano-particle and the surrounding environment. Figure 1(b) displays the extinction spectrum of the Au nanoparticles in deionized water solution. There is a resonance peak at 579 nm with a long tail up to 850 nm. The resonance peak could be red-shifted with the increase of the Au nanoparticle size [17,18]. A drop of 1 µL Au nanoparticle solution was directly deposited on the surface of the active area of the MPPC. When the water was dried away, the Au nanoparticles rested on the surface. As observed in an atomic force microscope (JPK NanoWizard II), each drop of 1 µL solution was dried to produce about 18 Au nanoparticles per $10 \times 10 \ \mu\text{m}^2$ as shown in Fig. 2 (a). The density of the Au nanoparticles was changed by adding the solution each time after the device was totally dried. Figures 2 (a)-2(c) show the distribution of Au nanoparticles on the surface with different densities. At low densities, the nanoparticles were spread on the surface evenly, while the nanoparticles tended to bunch up with the increase of the nano-particle density as shown in Fig. 2(c).



Fig. 2. Topographical images the MPPC surface with different Au nano-particle densities of approximately 18 (a), 36 (b), and 72 (c) particles per $10 \times 10 \ \mu\text{m}^2$.

The MPPC used in the experiment consisted of 10×10 Si-APD pixels with pixel size of $100 \times 100 \ \mu\text{m}^2$ on the whole active area of $1 \times 1 \ \text{mm}^2$ (S10362-11-100U, Hamamatsu). And the effective filling factor was 78.5% for the silicon APDs occupying the whole active area.

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Fig. 3. (a) Experimental setup of quantum efficiency measurement for the MPPC with Au nano-particle deposition. AMP, amplifier; OSC, oscilloscope. (b) Counting rate of the MPPC with no Au nanoparticles (line A) and about 18 (line B), 36 (line C), and 72 (line D) Au nanoparticles per $10 \times 10 \ \mu\text{m}^2$, respectively. (c) Photon detection efficiency as a function of the Au nanoparticle density. (d) Enhancement factor of the photon detection efficiency as a function of the incident laser wavelength.

The laser emitting at 780 nm was focused on the active area of the MPPC by a convex lens after being attenuated to few-photon level. The laser power was measured by a power meter with precision of pW and calibrated density filters with fixed attenuation was inserted to decrease the intensity of the photon stream. In this way, the avarage photon number incident on the MPPC could be calculated. The beam spot on the MPPC was about 0.4 mm in diameter, within the active area of $1 \times 1 \text{ mm}^2$. The Si-APDs were operated in Geiger mode with the bias voltage of 69.80 V at 21 °C. The avalanche current was passively quenched as shown in Fig. 3(a). In order to lower the darkcount noise, we operated the MPPC at a bias voltage 0.4 V lower than the recommended voltage and the operation temperature was also lower than that in the datasheet. With these settings, the total dark count of the MPPC was 1.23×10^5 cps (counts per second) without any light illumination. The output signal of the MPPC was captured and recorded by an oscilloscope, where the photon counting rate and peak output voltage could be read out. Figure 3(b) shows the counting rate (including the afterpulses) of the MPPC tested as a function of incident laser powers at the wavelength 780 nm. The quantum detection efficiency was deduced by the slope of the curve. Line C shows the largest quantum detection efficiency, which was the experiment result with the density of the Au nanoparticles was about 36 particles per $10 \times 10 \ \mu\text{m}^2$. With different density of Au nanoparticles on the surface of the active area of the MPPC, the detection efficiency changed as shown in Fig. 3(c). The detection efficiency of the MPPC in the experiment was about 2.48% at 780 nm without surface decoration. Since the biase voltage was lower than the recommended value, the detection efficiency was lower thant the typical value in the datasheet. As the density of the Au nanoparticles on the surface of the active area increased, the detection efficiency augmented. The maximum of the detection efficiency was achieved to be 3.23% when the density of the Au nanoparticles was 36 particles per $10 \times 10 \ \mu\text{m}^2$. The detection efficiency was increased by about 1.30 times at 780 nm. With Au nanoparticle deposition, there was no change on the dark count rate. The forward incident light scattering from the Au nanoparticles increased the effective active region of the detector. The interference between the light scattered from the Au nanoparticles into the device and the light transmitted directly into the device affected the photocurrent generation of the MPPC

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device. Constructive node in the active region of the device would lead to the increase of the detection efficiency [19]. The MPPC used in the experiment was with revered structure where P^+ layer which was the absorption layer was on the incident surface. Therefore, it is a near field effect. However, when the density of the Au nanoparticles increased further, the detection efficiency dropped. The drop of detection efficiency at high nanoparticle densities could be ascribed to the fact that additional Au nanoparticles covered up the active area of the silicon APD instead of scattering light.

In order to verify the surface plasmon resonance enhancement on the quantum detection efficiency, the detection efficiency of the MPPC was measured with incident light of different wavelengths with and without the Au nanoparticles on the surface. The enhancement factor gwas calculated by

$$g = \eta_{Au} / \eta_b, \tag{1.1}$$

where η_{Au} and η_b represent the detection efficiency of the MPPC with and without the Au nanoparticles on the surface, respectively. Figure 3(d) plots the enhancement factor as a function of the incident laser wavelength. The maximum enhancement factor was 2.76 at 670 nm. It should be noticed that the enhancement factor matched the extinction spectrum in Fig. 1(b) except a red-shift of the peak from 579 nm in Fig. 1(b) to 670 nm in Fig. 3(d), which agrees with the fact that the resonance plasmon peak is red-shifted owing to the significant inter-aggregate coupling [20–23]. As the Au nanoparticles were separated far away from each other, interference effects played an important role. With the increase of the nanoparticle density, the Au nanoparticles became approached each other. As a result, two or more Au nanoparticles might be aggregated to form dimers, trimers, and even tetramers, as shown by the atomic force microscopic image given in Fig. 2(c). The approaching Au nanoparticles were featured by dominant inter-particle couplings along with observable red-shifts of the absorption peaks, large near-field enhancement within the inter-particle gap, as well as the increase of the far-field scattering.



Fig. 4. Histogram of the MPPC peak output voltages (a) with Au nanoparticle density of ~36 particles per $10 \times 10 \ \mu m^2$ (a), without Au nano-particles (b). Red circle represents the data measured in the experiment and the blue line represents the fitting data.

We next compared the photon-number-resolving capability of the MPPC with and without Au nano-particles. With Au nanoparticle density of \sim 36 particles per 10 \times 10 μ m² on the MPPC surface, the histogram of the peak output voltage from the MPPC was shown in Fig. 4(a) under the illumination of an attenuated laser to provide few-photons pulses. Those individual peaks indicating different photon numbers which separated obviously. The gap between two peaks was basically equal. The photon number obeyed the Poissonian distribution

$$P(V) = \sum_{n=0}^{\infty} p(\mu, n) \cdot \rho(n, V), \qquad (1.2)$$

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where $p(\mu, n)$ represents the probability containing n photons in a single pulse with the average photon number μ and $\rho(n, V)$ is avalanche voltage signal distribution. By fitting the experimental data in accordance with Eq. (1.2), we got the average detected photon number as 4.52. The photon-number-resolving capability of the MPPC was well reserved in the presence of Au nanoparticle deposition. As shown in Fig. 4(b), without the Au nanoparticles, the average detected photon number was 3.61 under the same light incidence. The enhancement of the detection efficiency was calculated to be 1.29, in agreement with the above-mentioned test.

3. Conclusion

In conclusion, the detection efficiency of MPPC-based photon-number-resolving detector was enhanced by surface decoration with Au nanoparticles. The enhancement of the quantum detection efficiency originated from the surface plasmon resonance enhancement from the Au nanoparticles on the surface of the active area of the MPPC. Meanwhile the photon-numberresolving capability was well reserved. It is a proven technology by using metallic nanoparticles integrated with photodiodes to enhance photocurrent. Our results demonstrated that this technology could be further applied to single-photon detection. It provides a promising method for the design and fabrication of high-efficiency photon-numberresolving detectors with nanotechnique.

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