High Sensitivity Crystalline Selenium-based CMOS Image Sensor Using Avalanche Multiplication

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Abstract We present our work on a complementary metal oxide semiconductor (CMOS) image sensor that uses crystalline selenium as the photoconversion layer and enables avalanche multiplication at low voltage, with the goal of realizing a high-definition, high-sensitivity camera. Gallium oxide, used as a hole blocking layer, and nickel oxide used as an electron blocking layer effectively prevents the increase of external dark current caused by carrier injection from an external electrode. In addition, a new crystallization method was developed to improve the crystallinity of selenium for the fabrication of crystalline selenium films. We were able to capture high-quality images in a crystalline selenium-based CMOS image sensor and confirm signal amplification by a factor of approximately 1.4 at a reverse bias voltage of 22.6 V by using these film structures and deposition conditions.

Keywords: image sensor, photoconversion layer, crystalline selenium, gallium oxide, nickel oxide, avalanche multiplication.

1. Introduction

4 K and 8 K broadcasting began in 2018, providing ultra-high image quality far superior to that of current high-definition television. The ultra-high-definition image will soon become a familiar part of people’s lives. We have been conducting research and development for the realization of the next-generation ultra-realistic broadcasting system “8 K Super Hi-Vision”. Even after the start of this broadcast, we are continuing our research into improving the performance of 8K cameras so that they can adapt to more diverse shooting environments. In particular, with regard to the miniaturization of pixels in imaging devices accompanying high-definition imaging, pixel sizes below 1 µm have already been reported1, 2. Such a trend results in a decrease in the amount of incident light per pixel, in other words, a decrease in the imaging device’s sensitivity, which is regarded a serious issue as a factor limiting the shooting conditions. We propose a high-sensitivity imaging device in which a photoconversion layer capable of avalanche multiplication of signal carrier is stacked on a complementary metal oxide semiconductor (CMOS) circuit (Fig. 1). Various methods for increasing sensitivity in conventional imaging devices, such as reducing readout noise and improving optical aperture ratio, have been tried, but these methods have yet to significantly solve the decrease in sensitivity caused by pixel miniaturization.

The use of signal amplification by avalanche

**Fig.1** Schematic cross section of the stacked CMOS image sensor overlaid with a photoconversion layer.
Multiplication to increase the sensitivity of imaging devices is expected to be a technology that has the potential to significantly improve such sensitivity degradation. Avalanche multiplication is a phenomenon in which the carrier is accelerated by a strong electric field, resulting in the formation of new electron-hole pairs one after another through impact ionization. The high-gain avalanche rushing amorphous photoconductor (HARP) imaging tube, which uses amorphous selenium (a-Se) as the photoconversion layer, has been put to practical use as an imaging device using avalanche multiplication. This technology has been applied not only in broadcasting, but also in various fields such as low-dose X-ray imaging detectors and biomedical scientific cameras. However, in order to meet the demand of higher resolutions such as 8 K, avalanche multiplication must be applied to solid-state imaging devices. For this purpose, there is an urgent need to develop a low voltage multiplication type photoconversion layer that can operate below the breakdown voltage of the solid-state imaging device.

In this study, we have focused on crystalline selenium (c-Se) as a multiplication type photoconversion layer material that can operate at low voltages, and by reducing the dark current, we were able to develop an 8 K CMOS image sensor laminated with a c-Se-based photoconversion layer using avalanche multiplication.

2. Low voltage Operation of Avalanche Photoconversion Layer

2.1 Crystalline Selenium

The c-Se is fabricated by annealing a-Se deposited on a substrate by vacuum evaporation. Fig. 2 shows the wavelength dependence of the absorption coefficient of a-Se, c-Se, and silicon (Si) in the visible light region. As is clear from the comparison of the photographs of the prepared a-Se and c-Se samples, the crystallization improves the absorption of long-wavelength light and shows optimal absorption properties as a material for visible light. In addition, the absorption coefficient is more than one order of magnitude higher than that of Si, a conventional photoconversion material, throughout the visible region, so that the film can absorb light sufficiently even if the thickness of the film is thinner than that of Si photodiode. Even when the same external voltage is applied, a thinner film can induce a stronger electric field inside, which is an important feature for realizing low voltage multiplication. We herein used Se composed of polycrystals, which has no restrictions on the choice of substrate, and its crystallization temperature is below 200°C, which is lower than that of other inorganic crystalline materials, allowing it to be fabricated directly on the CMOS readout circuit.

2.2 Fabrication Method for Crystallinity

Enhancement

A Se used in this study is hexagonal, which is the most stable Se crystal structure. In the preparation of c-Se, a small amount of tellurium (Te), which serves as a nucleation layer, can be inserted before the formation of a-Se to prevent film peeling during annealing. Furthermore, Te not only prevents film peeling, but also plays an important role in determining the crystal orientation of Se and contributing to improved crystallinity. Te is deposited through vacuum evaporation. It was discovered that depositing Te at a higher Te-source temperature than Te melting temperature (450°C) without substrate heating can significantly improve the crystal orientation of not only Te but also Se, which grows on Te as a nucleation layer. Fig. 3 shows X-ray diffraction (XRD) patterns of the 500-nm-thick c-Se films grown on 1-nm-thick Te nucleation layers deposited at different Te temperatures of 380°C and 490°C on glass substrates. As can be seen in
the figure, all reflection peaks in the XRD pattern of the c-Se film can be indexed to the (100)-oriented hexagonal Se (PDF#00-006-0362) and the intensity of the c-Se (100) peak increases drastically with increasing TTe. From the results, it is found that the crystal (100) preferred orientation of the c-Se films strongly depends on the crystallinity of the Te nucleation layer and the crystallinity of Se fabricated on a variety of substrates can be controlled by the fabrication method of Te. The inset shows the hexagonal selenium unit lattice, and it can be seen that the fabricated samples grow in the (100) plane direction perpendicular to the c-axis, even on an amorphous substrate.

3. Effects of Blocking Structures on Dark Current Reduction

In order to realize a high-sensitivity imaging device with avalanche multiplication, the suppression of the dark current in the photoconversion layer, which greatly affects the noise characteristics of the imaging device, is an important issue. The main cause of dark current under a strong electric field is the injection current from the external electrode into the photoconversion layer by overcoming the energy barrier between Se and the external electrode. As a countermeasure, the introduction of a blocking layer with large bandgap energy \( E_g \) between the c-Se and the external electrode is effective. In this chapter, we describe the development of hole and electron blocking layers to prevent carrier injection.

3.1 Hole Blocking Layer

The properties required for hole blocking materials are sufficient transmission characteristics of incident light and a large bandgap necessary to block the hole injection. We used gallium oxide (Ga2O3) as an effective hole blocking layer, which is an n-type oxide semiconductor with high transparency for visible light and very large \( E_g \). The Ga2O3 was formed by RF sputtering at room temperature. The crystallinity and \( E_g \) of the prepared films were evaluated by XRD and optical absorption spectra, respectively, and it was found that Ga2O3 is amorphous with an \( E_g \) of 4.9 eV\(^1\). The n-type Ga2O3 formed a pn junction with the p-type c-Se, and the measurement was performed by applying a reverse bias voltage to the photoconversion layer.

To verify the effect of Ga2O3 as a hole blocking layer, we fabricated two types of test devices: one with c-Se sandwiched between indium tin oxide (ITO) electrodes (Fig. 4a), and the other with Ga2O3 inserted as a hole blocking layer between ITO electrodes and c-Se (Fig. 4b) on glass substrates. Fig. 5 shows the current-voltage characteristics of each device in the dark condition. As can be seen in the figure, the dark current sharply decreased from \( 1.5 \times 10^{-7} \) A/cm\(^2\) to \( 5.5 \times 10^{-11} \) A/cm\(^2\) at a reverse bias voltage of 5 V because Ga2O3 blocked the carrier injection from the external electrode.

On the other hand, it was found that doping Ga2O3 with Sn could reduce the operating voltage of avalanche multiplication by increasing the carrier concentration in Ga2O3 and promoting the formation of depletion layers in p-type c-Se whose carrier concentration is \( 1 \times 10^{16} \) cm\(^{-3}\). Fig. 6\(^2\) shows the signal current-voltage characteristics of Ga2O3/c-Se heterojunction photodiodes with different Sn doping concentrations of 5 and 5.6 mol% on the ITO-patterned glass substrates (light intensity: 2.5 µW/cm\(^2\), light wavelength: 450 nm). Sn-doped Ga2O3 was deposited using a Ga2O3-SnO\(_2\) sputtering target. Owing to the effective blocking performance, we were able to confirm signal amplification by avalanche multiplication, in which the once-saturated signal current increases again as the voltage increases. Furthermore, the starting voltage of avalanche multiplication was reduced with the increase of Sn doping into Ga2O3.

By applying the above hole blocking structure, a photoconversion layer was then fabricated on a CMOS
readout circuit and the performance of the stacked image sensors was evaluated. Fig. 7a and 7b\(^{13}\) shows a schematic of a pixel of the fabricated image sensor and the photomicrograph of the chip. The pixel has the typical three-transistor configuration consisting of a reset transistor (MR), an amplifying transistor (MA), and a select transistor (MS). The developed image sensor has 8 K resolution (7472(H) \times 4320(V) pixels) in a Super 35 mm optical format with a pixel size of 3.2 µm \times 3.2 µm. All pixel electrodes are made of molybdenum (Mo) and embedded with silicon dioxide (SiO\(_2\)) and contact pads for ITO are prepared. The surface is planarized by chemical mechanical polishing so that the films can be stacked directly. Fig. 8a and 8b\(^{13}\) show the dark images (100 \times 100 pixels) extracted from the images captured by the fabricated image sensors with a photoconversion layer including Ga\(_2\)O\(_3\) and the dark current histograms obtained from the images, which correspond to the outputs of 12-bit (4096 greyscale level) images with different reverse bias voltages, respectively. The dark image is filled with white spots with a reverse bias voltage of 19.6 V as shown in Fig. 8a, and the histogram indicates the existence of white spots as pixels with saturated signals as shown in Fig. 8b. This is due to the local concentration of the electric field adjacent to the edges of the pixel electrodes in c-Se.

Furthermore, the peak of the histogram gradually becomes smaller and shifts to the higher signals as the reverse bias voltage increases. This is caused by the electron injection from an external electrode due to the lack of the electron blocking layer. In order to eliminate the white spots and suppress the increase of dark current under high electric field, we need to develop an electron blocking layer that also serves as a buffer layer.

### 3.2 Electron Blocking Layer

We mentioned in the previous chapter that the electron blocking layer needs to have the role of an electrical buffer layer while providing high blocking performance. On the other hand, because there are few p-type wide-gap materials in contrast to n-type wide-gap materials, which offer abundant options, the development of an electron blocking layers has been challenging. Nickel oxide (NiO) (\(E_g\): 4.0 eV\(^{14}\)) is one of the few p-type, wide-gap semiconductors used in various applications, such as visible-transparent UV photodetectors\(^{15, 16}\), visible-transparent solar cells\(^{17, 18}\), and UV-visible light-emitting diodes\(^{19, 20}\), and has great potential as an electron blocking layer. When applying NiO, defect control is important because the presence of Ni defects is a major factor in lowering the effective \(E_g\) and deteriorating the blocking properties. Because it has been reported that the carrier concentration in NiO can be reduced by changing the oxygen (O\(_2\)) to argon (Ar) flow rate ratio in the sputtering gas\(^{21}\), we have investigated the O\(_2\) concentration in NiO on the dark current of the photoconversion layers. Fig. 9 shows the comparison of the relative dark current-voltage characteristics for the test devices consisting of the photoconversion layers without and with NiO (O\(_2\) fraction in the sputtering gas of 10% and 3%,
respectively) fabricated on glass substrates\textsuperscript{13}. As shown in the figure, the introduction of NiO reduced the dark current, and following the reduction in the amount of O\textsubscript{2} introduced, the dark current was further reduced.

### 3.3 Avalanche multiplication in 8 K image sensor

Based on the above results, we developed an 8 K image sensor with hole blocking and electron blocking layers. In addition to a 20-nm-thick Ga\textsubscript{2}O\textsubscript{3} hole blocking layer, we applied a 20-nm-thick NiO electron blocking layer with the deposition condition of O\textsubscript{2} fraction of 3\% to the fabrication of stacked image sensors overlaid with a photoconversion layer. Fig. 10\textsuperscript{13} shows the dark current density-voltage characteristics of the photoconversion layer with and without\textsuperscript{22} NiO on the CMOS readout circuits. By applying the optimized NiO, we were able to reduce the dark current in the multiplication region at a reverse bias voltage of 22.6 V by a factor of 30 or less. Fig. 11\textsuperscript{a and b} show the dark images (100 × 100 pixels) extracted from images captured by the fabricated image sensors with photoconversion layers including NiO and the dark current histograms obtained from the images with different reverse bias voltages, respectively. Introduction of NiO prevented the appearance of white spots owing to the buffering effects and suppressed the increase of the dark current due to the effective electron blocking effects. Fig. 12\textsuperscript{13} shows the current density-voltage characteristics of a photoconversion layer with NiO (O\textsubscript{2} fraction of 3\%) stacked onto the CMOS readout circuit, in the dark and illuminated through a blue filter. The signal current increases steeply with increasing reverse bias voltage and saturates at around 5 V. Increasing the voltage further, we were able to confirm signal amplification by avalanche multiplication where the voltage exceeds approximately 15 V. The signal current multiplication factor is calculated as the ratio of the actual signal current (I\textsubscript{M}) to the extrapolated current (I\textsubscript{S}) in the saturation region at the same voltage, and this time, 1.4 times was achieved at a reverse bias voltage of 22.6 V, while the dark current remained 2.6 nA/cm\textsuperscript{2}. Fig. 13\textsuperscript{a and b} show partial images extracted from the original 8K images captured by the fabricated image sensors at reverse bias voltages of (a) 12.6 V (unmultiplied) and (b) 22.6 V (multiplied). We were able to obtain brighter...
images with avalanche multiplication without significant degradation due to white spots or dark current thanks to the effective blocking and buffering effect.

4. Conclusion

Using avalanche multiplication, we have developed a c-Se-based photoconversion layer stacked high-sensitivity CMOS image sensors. In order to achieve high-sensitivity, we improved the crystallinity of Se and introduced a wide-gap blocking layer to reduce the dark current. For the improvement of crystallinity, by depositing the Te nucleation layer at high temperature, high crystal orientation could be achieved. Furthermore, the introduction of Ga2O3 as a hole blocking layer and NiO as an electron blocking layer effectively prevents the increase of injection current from an external electrode. Signal amplification by avalanche multiplication was successfully achieved with a multiplication factor of 1.4 at a reverse bias voltage of 22.6 V by suppressing the increase of dark current and the generation of white spots, enabling bright images to be acquired without any degradation in image quality. The results of this research show the possibility of not only increasing the sensitivity of image sensors, but also increasing the performance and diversification of devices using hybrid structures made of Si and non-Si materials.

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