Multi-spectral SWIR PbS Quantum Dot Pixels Realized Using Transfer Printing

Nayyera Mahmoud[†‡¶*], Willem Walravens[‡¶], Robin Petit[§], Michiel Van Daele[§], Christophe Detavernier[§], Zeger Hens[‡¶], and Gunther Roelkens[†‡*]

† Photonics Research Group, Ghent University-imec, Technologiepark-Zwijnaarde 126, B-9052 Zwijnaarde, Belgium
‡ Center for Nano and Biophotonics, Ghent University-imec, Technologiepark-Zwijnaarde 126, B-9052 Zwijnaarde, Belgium
¶ Physics and Chemistry of Nanostructures (PCN), Ghent University, Krijgslaan 281-S3, Ghent 9000, Belgium
§ Conformal Coating of Nanomaterials (CoCooN), Ghent University, Krijgslaan 281-S1, Ghent 9000, Belgium
nayyera.mahmoud@ugent.be, gunther.roelkens@ugent.be**

Abstract: We report on the transfer-printing-based integration of PbS QD pixels for multi-spectral imaging in the short-wave infrared. As a proof-of-concept, 8 multi-spectral pixels each consisting of 4 QD photodetectors operating in the SWIR are demonstrated. © 2019 The Author(s)
OCIS codes: 040.0040, 040.5160, 110.4234.

1. Introduction

There is an increased interest in advanced imaging systems in the short-wave infrared (SWIR), both for civilian and military applications such as non-invasive biomedical imaging, passive night vision, defense, quality control and product inspection in food and pharmaceutical industries. Multi-spectral and hyper-spectral imaging is an enabling technology for extracting "fingerprint" spectral data of the inspected objects by utilizing multiple IR spectral bands. Light is collected in multi-band detector arrays where each pixel of an array element has a different spectral response. In order to see a proliferation of this technology, the realization of cost-effective and scalable SWIR multi-spectral imaging systems is crucial. Existing infrared focal plane arrays (FPAs) are based on either II-VI compounds, namely mercury-cadmium-telluride (HgCdTe or MCT) or III-V compounds, such as indium gallium arsenide (InGaAs), indium arsenide (InAs), and indium antimonide (InSb). MCT is instrumental for the development of IR imagers by covering a very wide wavelength range (SWIR, MWIR and LWIR). Despite the superior detectivity D* and photoresponse, the imaging arrays are fairly small, expensive and have stability issues. III-V based imagers on the other hand offer a good alternative, especially in the short-wave infrared, but the cost remains an issue [1].

A fundamentally different approach is to use colloidal quantum dots (CQDs), which allows for substantially reduced cost. CQDs have enabled significant progress in optoelectronic applications because of their size-dependent band structure, spectrally narrow emission, broad absorption and high photoluminescence quantum efficiency [2]. The classical integration techniques of CQDs comprise either drop casting, spin coating or spray coating the colloidal solution on predefined metal contacts/electronic driver circuits. Post-processing steps are then required to pattern the integrated QD films in order to realize operational devices. Despite the facile fabrication of CQDs, and ease of integration into any substrate, established integration techniques are not designed to integrate patterns of multiple different QD films into the same device, as is required for a multi-spectral imager. A dual-color photodetector has been reported based on a tandem configuration of two PbS CQDs films of different nanocrystal size on two separate predefined metal contacts. However, scaling this approach to multiple bands is problematic.

In 2004 Menard et al. proposed micro-transfer-printing as a novel technique to transfer thin-film components from a source substrate to a target substrate, providing a scalable approach for device integration[3]. More recently, we reported the transfer printing of micro-scale patterns of Al_2O_3-capped PbS QD films to realize large-scale integrated photodetector arrays with a first excitonic absorption peak at 2.1 μm wavelength. The presence of the Al_2O_3 cap layer enhances the transfer printing process by forming an "exoskeleton" for the QD film and providing a fast and high-quality pickup of the QD patches. This led to the conclusion that the Al_2O_3 layer not only protects the QDs against oxidation, but also facilitates QD film patterning and improves the pick-up yield and printing throughput. Our approach provides a facile and selective pick-and-print of QD assemblies with high precision [4]. In this paper, we demonstrate the first SWIR multi-spectral imager pixels based on the transfer printing of arrays of Al_2O_3-capped PbS QD-based films. Each multi-spectral pixel consists of 4 QD photodetectors of different absorption cut-off wavelengths, offering a broadband operation in the SWIR range spanning from 1.2 to 2.2 μm.

2. Fabrication strategy and device characterization

The transfer printing is realized by a commercial micro-transfer-printer (X-Celeprint, model µTP-100). The process involves using a patterned elastomeric PDMS stamp to rapidly pick-up the material stack (Al_2O_3/PbS QDs) from a source substrate and print the stack on pre-fabricated interdigitated contact electrodes on the target substrate. Almost 100% printing yield can be obtained this way. One of the key advantages of transfer printing is the ability to transfer-print QD patches in a massively parallel way, by picking up and printing arrays of defined patterns at the same time. As a proof-of-principle we used a patterned PDMS stamp with 8 posts (60 × 60 µm^2 and pitch 250 µm). In this way, 8 multi-spectral pixels were fabricated by means of 4 successive transfers (one for each QD detector material) of "8 at a time" high-quality Al_2O_3-capped PbS QD patches to the desired positions on the target substrate (Figure 1). The scalability of this technique has been demonstrated in other work where several thousands of optoelectronic devices have been simultaneously transferred with high yield.

Four solutions of PbS CQDs with first excitonic peaks at 1.2, 1.55, 1.8 and 2.2 µm were fabricated (Fig. 1(a)). As previously described [4], for each type of CQD solution, a source substrate was prepared through a layer-by-layer deposition and surface ligand removal on a surface-modified ODTS-silicon substrate. The dried QD films were then capped through a thermal ALD deposition of Al_2O_3 (60 nm thick layer). Lastly, a dense array of Al_2O_3 patterns were realized on the source substrates through standard photolithography and subsequent wet etching. On the target substrate, planarized arrays of interdigitated Ti/Au contacts were defined on a thermally oxidized silicon wafer. The realized 8-pixel structure is shown in Fig. 1(b). The photoresponse of the multi-spectral pixels was investigated in the SWIR (Fig. 1(c)) enabling multi-band image sensors in the SWIR. Our recent work shows that the pixel dimensions can be substantially reduced from the 50x50 µm^2 pixel dimensions used in this work down to 2x2 µm^2 enabling high resolution image sensors.

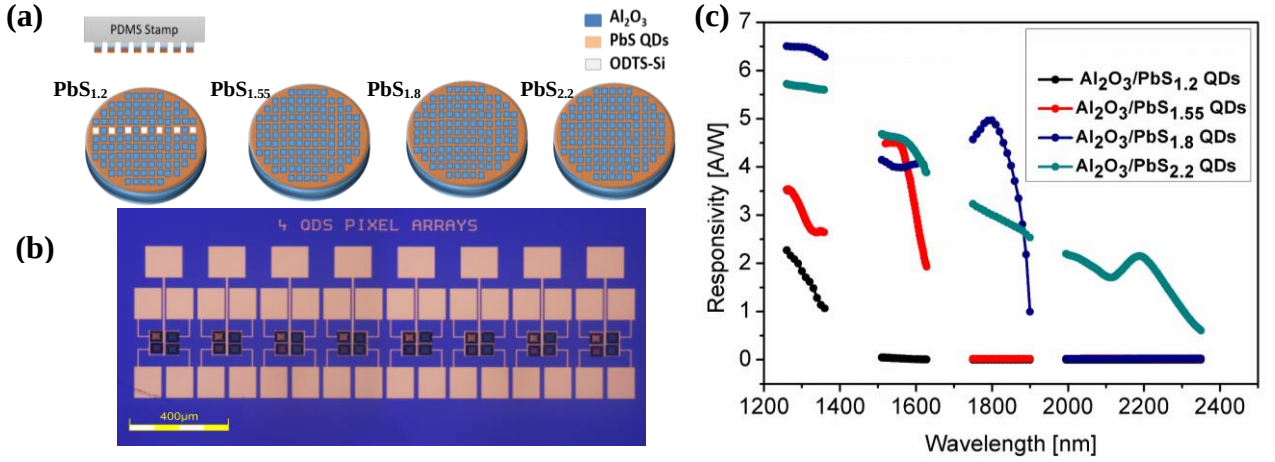

Fig. 1 . (a) A schematic illustration of the transfer-printing process flow from the four Al_2O_3/QD source substrates using arrayed PDMS stamp, (b) Optical image of a printed array of 8 multi-spectral Al_2O_3/PbS QD pixels, and (c) Spectral response measured at 1µW power level and 1 V bias

3. Conclusion

An array of multi-spectral pixels based on 4 different Al_2O_3-capped PbS QD photodetectors is realized using micro-transfer-printing. The arrays show a broadband and distinct multi-spectral response in the SWIR at low operation voltage (1V) and room temperature. Our developed integration strategy represents an excellent approach for broadband spectral sensors/imagers that could be potentially cheaper than current technologies.

4. References

[1] A. Rogalski, P. Martyniuk, and M. Kopytko, "Challenges of small-pixel infrared detectors: A review" *Rep. Prog. Phys.* **79** , 046501 (2016).
[2] G. Konstantatos, "Colloidal quantum dot photodetectors," in *Handbook of Colloidal Quantum Dot Optoelectronics and Photovoltaics,* G. Konstantatos and E. H. Sargent (Eds.), Cambridge: Cambridge University Press, 173–198 (2013).
[3] E. Menard, K. J. Lee, D.-Y. Khang, R. G. Nuzzo, and J. A. Rogers, "A printable form of silicon for high performance thin film transistors on plastic substrates," *Appl. Phys. Lett.* **84**, 5398–5400, (2004).
[4] N. Mahmoud, W. Walravens, J. Kuhs, C. Detavernier, Z. Hens, and G. Roelkens, "Micro-Transfer-Printing of Al_2O_3 -Capped Short-Wave-Infrared PbS Quantum Dot Photoconductors" *ACS Appl. Nano Mater.* **2**, 299-306 (2019).