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Quantum dots/graphene nanohybrids photodetectors: progress and perspective

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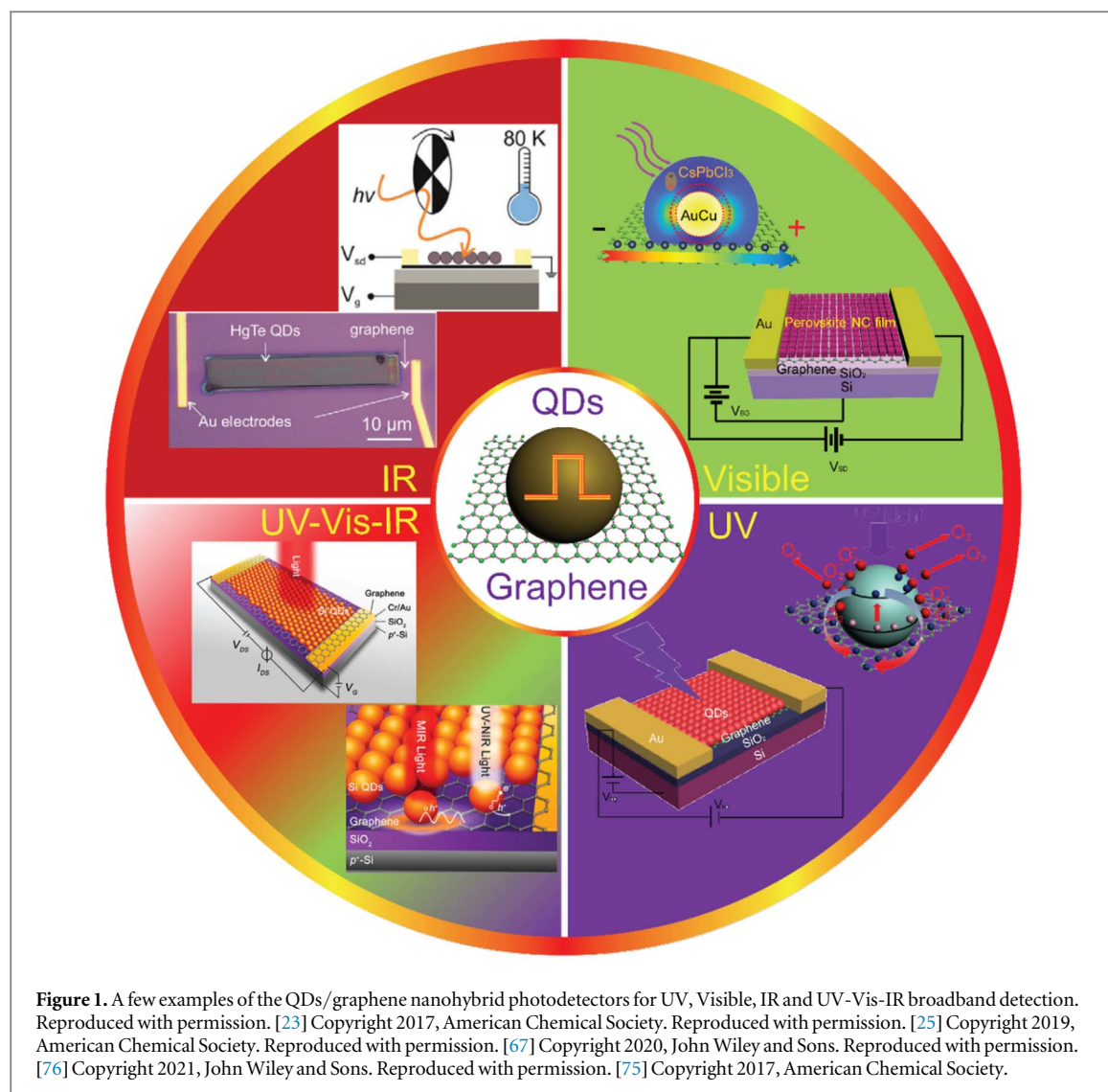
E-mail: jwu@ku.edu and gmg@ku.edu**Keywords:** quantum dots, graphene, heterostructure, nanohybrids, photodetectors**Abstract**

Semiconductor quantum dots/graphene heterostructure nanohybrids combine the advantages of the enhanced light–matter interaction and spectral tunability of quantum dots (QDs) and high charge mobility in graphene as a charge transport pathway, providing a unique platform for exploration of photodetectors with high performance. In particular, the QDs/graphene nanohybrids allow resolution to the critical issue of charge transport in QDs-only photodetectors stemming from the low charge mobility associated with both QD surface defect states and inter-QD junctions. Furthermore, the achieved capability in industrial-scale fabrication of graphene and colloidal QDs has motivated efforts in research of QDs/graphene nanohybrids focal plane arrays that are expected to be not only high performance and low cost, but also light-weight, flexible and wearable. This paper aims to highlight recent progress made in the research and development of QDs/graphene nanohybrid photodetectors and discuss the challenges remained towards their commercial applications.

1. Introduction

Graphene regards to a monolayer of carbon atoms arranged in a two-dimensional honeycomb lattice [1, 2]. Its discovery in 2004 has not only led the 2010 Nobel Prize in Physics to Geim and Novoselov, but also an intensive interest in research and development for a large spectrum applications taking advantage of graphene's unique physical properties of high charge carrier mobility predicted to be up to $200,000 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ at room temperature, broadband optical transparency of 97.7% from near ultraviolet to mid-infrared, high mechanical strength, flexibility and chemical stability [1–4]. Graphene can be viewed as an atomically thin (thickness $\sim 0.34 \text{ nm}$) film and can be grown using chemical vapor deposition in large area [5, 6] with compatibility to the established microfabrication processes [7] for graphene-based devices and circuits for applications in electronic, photonic, optoelectronic, sensors, etc [8–16].

Graphene-based photodetectors [1, 17–19] present a particular interesting topic considering the broadband absorption of $\sim 2.3\%$ per graphene sheet. However, graphene is a semimetal with a zero bandgap E_g [4], which has limited its use in a similar way to conventional semiconductors of well-defined E_g . This has motivated a recent exploration of quantum dots (QDs) on graphene heterostructure nanohybrids (QDs/graphene, which is a specific form of more general graphene/semiconductor nanohybrids) for photodetection with a high photoconductive gain and hence high responsivity [20–35]. These QDs/graphene nanohybrids build on the exciting progress made recently in colloidal QDs of a larger number semiconductors [36–52] and graphene [1–4, 53–56]. The QDs/graphene nanohybrids take advantage of the strong quantum confinement of QDs with enhanced light–matter interaction, spectral tunability by controlling their composition, shape, dimension, functionality, and carrier doping [57, 58], and of the extraordinary charge mobility of graphene at room temperature [1, 3, 53, 56]. In these devices, there are two main steps to realize the optical-to-electrical signal conversion. First, the QDs play the role of photosensitizers and absorb the incident photons. The produced excitons (or electron-hole pairs) can then be dissociated at the QDs/graphene interface by the built-in electric



field associated to the QD/graphene band-edge alignment. Secondly, the free charge carriers are transferred from QDs to graphene channel under the same built-in field and collected by the source and drain electrodes on the channel. The high mobility in exceeding $1.0 \times 10^4 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ experimentally achieved in graphene at room temperature [18, 59], which could be further enhanced through sample optimization, can lead to high photoconductive gain (G) $> 10^{10}$ in various QDs/graphene nanohybrids photodetectors [23, 25, 27, 60–67] and hence high photoresponsivity (R) and figure-of-merit specific detectivity (D^*) [24, 26, 27].

After the first report of Pubs QDs/graphene photodetectors by Konstantinos *et al* in 2012 [27], many QDs/graphene nanohybrid photodetectors have been explored in ultraviolet (UV), visible (vis), and infrared (IR) spectra [23, 26, 27, 68–72]. High D^* values in the range of $10^{11} - 10^{16}$ Jones have been reported in a broad spectrum ranging from IR (0.8 – 3 μm) [27], visible (400 nm – 700 nm) [73] to UV detections (300 nm – 400 nm) [25–27, 73], figure 1 illustrates several examples of the QDs/graphene nanohybrid photodetectors [23, 25, 66, 74, 75]. On an all-printable ZnO QDs/graphene UV photodetector, the presence of an atomically thin insulating surface layer on ZnO QDs was found to block the charge transfer from QD to graphene, and removing such a layer has demonstrated critical to the achievement of high $G \sim 3.6 \times 10^9$, $R \sim 9.9 \times 10^8 \text{ A W}^{-1}$, and D^* of 1×10^{14} Jones in this device [23]. Gong *et al* reported a broadband photosensitizer based on FeS₂ nanocubes (NCs) that exhibits a strong localized surface plasmonic resonance (LSPR) effect with enhanced light absorption and expanded spectral range beyond the cutoff of FeS₂ covering the UV–visible–SWIR (SWIR regards to short-wave IR) broadband with responsivities reaching $1.08 \times 10^6 \text{ A W}^{-1}$ [26]. Ni *et al* reported broadband UV to mid-infrared (MIR) photodetection using plasmonic Si QDs doped with boron (B) for LSPR enhancement. These B-doped Si QDs/graphene photodetectors exhibited high $G \sim 10^{12}$, $R \sim 10^9 \text{ A W}^{-1}$, and $D^* \sim 10^{13}$ Jones [74]. Besides doping semiconductor QDs to produce LSPR for broadband and enhanced absorption, metal/semiconductor core/shell QDs use plasmonic metal cores for light trapping, which can lead to improved light absorption on semiconductor shell. Using a template modulated colloidal approach for

synthesizing AuCu core (~ 7.1 nm in diameter) [76–78], followed with growth of metal-halide perovskite shell (CsPbCl_3) shell [66], enhanced light absorption was observed in the AuCu/ CsPbCl_3 core/shell QDs as compared to CsPbCl_3 QDs of comparable total diameter ~ 10 nm. A remarkable 30 times-enhanced photoresponse in AuCu/ CsPbCl_3 core/shell QDs/graphene nanohybrid photodetectors, as compared to the counterparts of CsPbCl_3 QD/graphene, was observed and attributed to the light trapping by AuCu core and enhanced light absorption in CsPbCl_3 shell, as confirmed in a Finite-Difference Time-Domain (FDTD) simulation [79]. A recent work by Alamri *et al* [80] combined the plasmonic effects from WS_2 nanodiscs grown on graphene and from Ag nanoparticles (AgNPs), leading to a seven-fold enhancement in photoresponse. Recently, Grotevent *et al* employed the HgTe QDs combined with graphene forming HgTe QDs/graphene nanohybrids photodetectors. The spectral sensitivity of this device has been extended to MIR range up to $3\text{ }\mu\text{m}$. At 80 K, the $D^* \sim 6 \times 10^8$ Jones at a wavelength of $2.5\text{ }\mu\text{m}$ and a frequency of 67 Hz was obtained [75].

QDs/graphene nanohybrids can be fabricated by depositing QDs on graphene using various low-cost, scalable methods including spin-coating [52, 74, 81–84], inkjet printing [16, 23, 85–89] and direct growth [80, 90–92], which illustrates additional advantages of QD/graphene nanohybrids including low cost, monolithic, flexible, wearable, etc In addition, different QDs targeting at different spectral ranges may be mixed or pixelated for multi-color, broadband photodetection [24, 85]. Moreover, a 288×388 channel focal plane array of PbS QDs/graphene nanohybrid photodetectors have been integrated with Si-based readout circuit, illustrating the potential of QD/graphene nanohybrid photodetectors for practical applications [21].

Despite progress made in research and development of QDs/graphene nanohybrid photodetectors, challenges remain and must be addressed before commercial applications of these photodetectors can be realized. A major challenge is in atomic-scale control of the QDs/graphene heterojunction interface that directly affects the exciton dissociation and charge transfer, which in turn impact the device performance including both photoresponse and response speed. For example, an atomically thin Zn acetate or Zn(Ac) layer on the ZnO QDs can completely block the charge transfer from ZnO QDs to graphene and lead to negligible photoresponse to UV light. When this surface layer is removed, improved photoresponse by orders of magnitude can be obtained [23]. On the other hand, many QDs such as PbS and CsPbX_3 ($X = \text{Cl}, \text{Br}, \text{I}$), have surface states due to the presence of defects and dangling bonds on the QDs surface, which not only lead to QDs decomposition in ambient but also behave as charge traps at the QDs/graphene interface that degrade both photoresponse and response speed by orders of magnitude [67, 93]. Ligand-exchange has been adopted for passivation of such surface states and hence improvement of photodetector performance [23, 24, 27, 94, 95]. Vafaie *et al* developed an efficient ligand-exchange route that tailors the halide passivants and removes unwanted organic species, and improves charge transport. In PbS QDs based photodetectors, this ligand-exchange protocol gave rise to a high external quantum efficiency of 80% at 1550 nm, $D^* \sim 8 \times 10^{11}$ Jones, and a 10 ns response time [96]. Gong *et al* replaced insulating oleic acid and oleylamine with conducting 3-mercaptopropionic acid ligands in the CsPbCl_3 QDs/graphene photodetectors with significantly improved device stability and performance [25]. It should be noted that the approaches developed for such interface engineering are QDs specific and hence cannot be applied universally. This means a continuous effort in research and development of various new interface engineering approaches for high-efficiency charge transfer across the QDs/graphene interface and for longevity of the QD/graphene nanohybrids devices will be a focus of future research with discovery of new functional QDs. Another major challenge is the limited light absorption in QDs/graphene nanohybrids that have a thin layer of QDs. It should be realized that multilayer QDs may have enhanced light absorption while the benefit can be significantly reduced by the inter-QD junctions that are hurdles to charge transport [24, 29, 97]. Development of schemes for light management will be important in future research to enhance light absorption of the very thin QD layer. Finally, an additional challenge is associated with device nonuniformity due to lack of a precise control in QDs (dimension, surface states, distribution on graphene, stacking in multilayer cases, etc) and QDs/graphene interface. All of the above present a series of roadblocks that must be removed to achieve high performance, uniformity and yield in commercial application of QDs/graphene based focal plane arrays.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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