The Application of Perovskite Quantum Dots in Photodetectors

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Abstract: Perovskite quantum dots (PQDs) can adsorb visible light and give out fluorescence or excited electron to the surroundings. Therefore, some kinds of PQDs can be used for light detection. Due to the appropriate band gap, cesium bromide and iodide-based perovskite PQDs (i.e. CsPbX₃) are especially suitable for visible light detection. Some other kinds of PQDs with different photoluminescence spectrum can be used to detect ultra-violet light or infrared light. In this paper, PQD-based photodetectors are divided into three categories: photoresistors, photodiodes and field effect transistors. Herein, the device structures, characteristics and mechanism of these three types of PQD-based photodetectors are briefly reviewed. Other applications of PQDs are also briefly introduced, including selective ion detection and organic molecular detection associated with molecular imprinted polymers.

1. Introduction
Quantum dots (QDs), also known as nanocrystals[1], is a kind of zero-dimensional nanostructure materials. Usually their size is about 10 nm, and their shape is quite regular. The facets of these QDs are similar to their extended crystal counterparts[2]. Since the size is so small that the proportion of atoms on the surface is much greater than that on the large crystals, the surface energy of QDs is much bigger, leading to a series of unique properties compared with large crystals including band gap[3] and melting point[4]. Many of these characteristics can be greatly affected by the size of QDs[5]. Because the excess of surface energy gives QDs the tendency of aggregation, capping methods are essential for stabilizing QDs, and these methods can affect the properties of QDs to a certain extent[6].

Perovskites, as emerging semiconductor materials, recently attract much attention due to its great absorption coefficient, high emission efficiencies and fabulous charge transport properties[7-11]. The composition of perovskite can be expressed as a common formula AMX₃. Generally, A = CH₃NH₃⁺ (MA), CH(NH₂)₂ (FA) or Cs⁺; B = Pb²⁺ or Sn²⁺, and X refers to halide anions (Cl⁻, Br⁻ or I⁻), and different permutations and combinations result in various compositions[12]. When made into QDs, perovskite quantum dots (PQDs) show great potential[13]. The photoluminescence quantum yields (PLQYs) of some PQDs are even over 90%[14]. In view of the supreme performance and controllability of PQDs, they can be widely applied as active materials including solar cells[11, 15], light-emitting diodes[16-19], lasers[20] and photodetectors[21-23].

In this review, the application of PQDs in photodetectors is systematically summarized. PQD photodetectors are divided into three categories according to the device structures of photodetectors: photoresistors, photodiodes and field effect transistors (FETs). If divided by the functions of PQDs, here are two categories: direct detection and down-conversion. The device structures, mechanisms and performances of these devices are also discussed.
2. Photodetectors

2.1. A brief introduction to the mechanism

In photodetectors, PQDs can be used with two methods. The first one is making PQDs be excited by the photon, giving out electric signals, which is direct detection. It is the mechanics for most of PQDs photodetectors. And the other one is transforming short-wave-light into long-wave-light, which is called down-conversion. For the first method, PQDs can be often used as the only active material, while for the other method other active materials are normally required to turn light signals into electric signals.

The active materials in photodetectors that transform light signals into electric signals can function as the photoresistor or the intrinsic semiconductor layer in the positive-intrinsic-negative (P-I-N) photodiode. After excited by photons, the electrons in the valence band of active materials will be lifted up into the conduction band and leaving holes in the valence band. If an electric field is given, the electron-hole pairs can be separated. When the active material is used in photodiodes, the electron will go into the n-type semiconductor layer and the hole will go into the p-type layer, giving out an electric signal in the external electric circuit. In most circumstances, the photocurrent is quite weak, so sometimes PQD layer is embedded in FET in order to amplify the electric signal. When PQD layers are used as photoresistors, the separation of exciton leads to the increase in the concentration of carrier and the decrease of resistance. Therefore, given the same bias voltage, the current will be amplified by the light.

![Figure 1](image_url)

Figure 1 a) The absorption, photoluminescence excitation and emission spectra of CsPbX₃-based PQDs with various I/Br ratios[24]. b) The band gap of different kinds of PQDs with various halide contents[25]. c) The scheme showing the categories of PQD photodetectors.

The mostly used PQDs are cesium lead halide (CsPbX₃) QDs. As shown in Figure 1a)[24], the absorption and photoluminescence (PL) emission spectra vary with the molar ratios of halide anions, since the band gap varies with halide anions. Figure 1b)[25] shows the band gap of PQDs with different compositions. It’s obvious that the band gap shrinks with the increase in average radius of the anion. The range of visible light spectrum is about 380-750 nm, which means that the range of photon energy
is about 1.65-3.26 eV. As shown in Figure 1(b), the band gap of CsPb(Cl/Br) 3 QDs is too wide to cover the visible light spectrum. Therefore, that kind of PQDs can be used to detect ultra-violet (UV) light, while bromide and iodide-based or mixed-halide perovskites are more often used for visible light detection.

2.2. Direct detection

2.2.1. Photoresistors. The simplest structure of PQD photodetectors using PQDs as resistors is shown in Figure 2a), and the energy structure is shown in Figure 2b) 

The separation of electron and hole is owing to the external bias voltage. As the exciton is separated, holes go to the cathode and electrons go to the anode, leading to the increase in carrier concentration and photocurrent. The structure of this kind of photodetectors is quite simple, so it can be built into mini-devices. It can even be used to build flexible and printable photodetector arrays. Liu et al. fabricated printable inorganic PQD-based X-ray detector arrays using this kind of structure, and the device showed great tolerance of bending if it’s printed on flexible polyethylene terephthalate (PET) substrate. However, the responsivity is limited by the conductivity of QD layer itself. The commonly used method of fabricating QD layer is drop-coating or spin-coating. Comparatively speaking, spin-coating can construct a smoother and more uniform QD layer, however, the layer is still not dense enough. Since QDs are not good conductors, electrons can be easily trapped in this layer and recombine with holes, which causes the loss of electric signals. Possible solutions are as follows: using centrifugal-casting to build a dense QD layer [22], and using interdigital electrodes to decrease the distance between two electrodes [23]. It can be observed in Figure 2d) that the denser the electrodes are, the greater the responsivity is. Besides, when Au nanocrystals (NCs) are introduced into the device, the localized surface plasmon resonance (LSPR) of Au presents as a plasmonic absorption peak at 530 nm, which is perfectly consistent with CsPbBr3 QDs. These methods significantly enhance the performance of PQD photodetectors. The responsivity and external quantum efficiency (EQE) doubled after introducing the Au NCs.
Zhou et al. developed an all-inorganic PQD-based metal/semiconductor/metal (MSM) photodetector, whose structure is shown in Figure 3a)\textsuperscript{[26]}. The mesoporous TiO$_2$ (mp-TiO$_2$) layer is the resistor, and its resistance is controlled by the PQD layer. The mp-TiO$_2$ layer is an n-doped semiconductor, and the energy diagram is shown in Figure 3b). The energy of the conduction band in PQDs is higher than that in mp-TiO$_2$, and there is no barrier formed in the interface under bias, the excited photoelectrons go into the mp-TiO$_2$ layer. The electron holes are trapped on the surface of PQDs due to the large energy barrier. Therefore, the carrier concentration in mp-TiO$_2$ layer is enhanced under illumination until hole extraction or recombination occurs. However, the injection of electron from PQD layer to mp-TiO$_2$ layer takes time, resulting in long rise and decay time. According to that, they changed the ligand on the surface of PQDs\textsuperscript{[27]}. 3-mercaptopropionic acid (MPA) is used as the ligand to connect PQDs and mp-TiO$_2$ layer. The structure of the QDs–MPA–TiO$_2$ system is shown in Figure 3c). The average time required for electron injection is reduced from over 290ns to 41ns. Device performances are greatly improved, including responsivity, rise/decay time and on/off ratio. Besides, TiO$_2$ can also be built as nanotube arrays\textsuperscript{[28]}. The nanotube arrays, as resistors, can respond to UV light, because their band gap is larger than PQDs, as shown in Figure 3. Thus, when cooperating with PQDs, the detection range of the device is larger than normal.
Figure 3. a, b) Schematic and energy diagram of metal/semiconductor/metal (MSM)\textsuperscript{[26]}, c) The structure and kinetic scheme of QDs–MPA–TiO\textsubscript{2} system. The process of electron transfer is denoted by the black array with the time of transfer process. d, e) Optical photograph and SEM image of TiO\textsubscript{2} NTs/MAPbI\textsubscript{3} QDs heterostructure\textsuperscript{[28]}. f) Energy diagram of TiO\textsubscript{2} NTs and MAPbI\textsubscript{3} QDs.

The band gap of PQDs is so wide that the frequency of infrared light is less than the cut-off frequency of PQDs. So, if it’s used to detect infrared light, up-conversion material that convert infrared light into visible light is needed. Zhang et al. covered the α-CsPbI\textsubscript{3} QDs layer with a layer of 2.0 mol\% Er\textsuperscript{3+} and 20 mol\% Yb\textsuperscript{3+}-co-doped NaYF\textsubscript{4} QDs (NaYF\textsubscript{4}:Yb,Er QDs)\textsuperscript{[23]}. The structure of the device is shown in Figure 4a). The PL spectra of NaYF\textsubscript{4}:Yb,Er QDs is shown in Figure 4c). This kind of QDs absorb near-infrared light and emit visible light at with a wavelength of 550nm and 650nm. The emission peak of NaYF\textsubscript{4}:Yb,Er QDs correspond with the absorption spectra of α-CsPbI\textsubscript{3} QDs. It can be noticed in Figure 4d) that this device gains a weak response over near infrared (NIR) and the detection range of this device is broadened to 260nm-1100nm.
2.2.2. Photodiode. Bi et al. used CsPbI3 QDs to fabricate a photodiode, and the device configuration and energy band diagram is shown in Figure 5a) and 5b)\textsuperscript{[29]}. PQD layer is the intrinsic layer. Under illumination, excited electrons in PQD layer go into fluorine-doped tin oxide (FTO) electrode via TiO\textsubscript{2}, the electron transport layer, and holes go into the Ag electrode via Spiro-OMeTAD, the hole transport layer. Besides, since purification process of QDs will enhance the defect density at the surface of QDs, they used 2-Aminoethanethiol (AET) to passivate the surface defects and reduce the nonradioactive recombination (Figure 5c). The AET-CsPbI3 QDs show hydrophobicity and great stability under UV irradiation. Dark current and responsivity are also improved by this ligand exchange reaction.
2.2.3. FETs. Yu et al. fabricated an vertical field-effect phototransistors (VFEPTs) with different kinds of PQDs [30]. The structure, as shown in Figure 6a, is based on a FET. FET uses positive-negative-positive (PNP) or negative-positive-negative (NPN) type of semiconductors to connect three terminals: source, drain and gate. For normal FETs, source and drain terminals are set at different sides of transistors, and gate terminal is set in the middle layer of transistor. The electric potential of gate source can be used to control the depletion range of the transistor, and drastically influence the current between source and drain terminal. This device is different, because gate terminal doesn’t contact with the PQD layer, but still be able to control the carrier concentration. Au/Ag nanowires are used as the source electrode, and the gate terminal is a layer of silicon covered by a 300-nm-thick SiO2 layer (Figure 6b). A voltage is applied between the gate and drain terminals in order to build an electric field which can separate the photoinduced excitons. When PQDs are excited by the light, separated excitons lead to the increase of carriers and then form a current under the effect of electric field. Greater light intensity leads to greater current, until the PQD layer is saturated.
There are also another two kinds of FET PQD photodetectors, which make silicon and SiO$_2$ layers under the device as gate terminal, but drain and source terminals are set at different sides of PQD layer. The first device produced by Zheng et al.$^{[31]}$ uses a layer of single-walled carbon nanotube (SWNT) on the SiO$_2$/Si substrate, then a layer of CsPbI$_3$ PQDs was spin-coated. When photon create electron-hole pair in PQDs, holes are transferred to the SWNT but electrons remain in the PQDs layer, thus generating a photocurrent. The other device is produced by Pan et al.$^{[32]}$ The structure and mechanism of this photodetector is almost the same as the previous one, but when excited, holes are trapped in the PQDs and electrons are transferred out because the channel material here is graphene. Besides, in this structure, gate is not necessary, so Zheng et al. changed the substrate into polymethyl methacrylate (PMMA). Since PMMA and SWNT are flexible, this device shows great tolerance and stability of bending.

Figure 7 Schematic diagram of two kinds of FETs by a) Zheng et al.$^{[31]}$ and b) Pan et al.$^{[32]}$

This FET fabricated by Wu et al.$^{[33]}$ is a little different from the FETs mentioned above, because its gate is based on Schottky Barrier. Therefore, its structure is more similar to metal-semiconductor field effect transistor (MESFET). When PQDs are excited by light, the electrons are injected into MoS$_2$ layer, and form a photocurrent under external bias voltage. But apart from bias voltage, the photocurrent is also affected by the Schottky Barrier. Schottky Barrier is the energy barrier between semiconductor and metal, due to the effect of Fermi level pinning. This barrier obstructs the electron flow crossing the heterojunction, including both the photocurrent and the thermionic and tunneling current between source and drain. Since the barrier height can be controlled by the Fermi level, gate terminal is set to adjust the Fermi level of MoS$_2$ layer. When the Fermi level of MoS$_2$ layer is raised by the gate, Schottky Barrier will greatly suppress the thermionic and tunneling current, and let the photocurrent predominate the current. That is the OFF state of this device where the dark current can be strongly reduced. In contrast to OFF state, there is also an ON state where the Fermi level and Schottky barrier is lowered. This state will increase all kinds of current, leading to a great responsivity.
2.3. **Down conversion**

As mentioned above, besides the characteristic of absorbing light, we can also use the characteristic of PL of PQDs to detect light. In this kind of devices, the function of transforming light signal into electric signal is taken by other active materials. The function of PQDs here is just transforming shortwave light into longwave light, which is called down-conversion. This kind of device often use PQDs to enhance the detectivity of UV light, so wide-bandgap PQDs can be used here. Zou et al.\cite{34} fabricated perovskite photodetector arrays, and the structure of devices belongs to photodiode. However, different from ordinary photodiode, there is another layer of CsPbBr$_3$ QDs. The UV light detection performance of the device is greatly improved by this layer of PQDs. Lu et al.\cite{35} fabricated a hydrogenated amorphous silicon radial junction (a-Si:H RJ) over silicon nanowire (SiNW) structure. The structure of radical junctions (RJs) is shown in Figure 9c). The detector itself has a comparatively low respond to UV light. Then they covered the RJs by spin-coating a layer of CsPbX$_3$ (X = Cl/Br = 1:2) QDs, and the EQE of UV lights is lifted up. The electric signals of RJs with QDs and without QDs are both detected. The difference of these two signals is the signal produced by UV light.
3. Other applications

In addition, the fluorescence of PQDs can be used to detect other things such as various kinds of ions or even pesticides. These kinds of detection are accomplished through fluorescence quenching rather than by giving out electric signals. However, fluorescence quenching means the fluorescence of PQDs goes down under the infection of other materials.

Liu et al. [36] covered MAPbBr₃ PQDs with two kinds of ligand: n-octylamine (OA) and 6-amino-1-hexanol (AH). They call this kind of PQDs the dual-ligands-capped PQDs (DL-PQDs). OA makes the PQDs stably emit fluorescence, and AH selectively responds to fluoride due to the hydrogen bond of hydroxyl group. As shown in Figure 10 a), the strong hydrogen bond of fluoride leads to the aggregation of DL-PQDs, and can be detected by fluorescence quenching. Ding et al. [37] doped Cs₂Bi₂Br₉ QDs with Eu³⁺. This kind of PQDs can selectively adsorb Cu²⁺ on its surface. Normally after excitation, the excited electron should transfer from PQDs to Eu³⁺, but when Cu²⁺ is adsorbed on the surface, electron will transfer to Cu²⁺ instead of Eu³⁺. Therefore, the PQDs are selectively quenched by Cu²⁺. Zhang et al. [38] encapsulated MAPbBr₃ QDs inside metal-organic frameworks (MOFs). After encapsulation, the PQDs show great resistance to water and excellent thermal stability under high temperature and a wide range of pH. Besides, it’s highly sensitive to different heavy metal ions, making it can be used as aqueous heavy metal ion detector. All these quenching can be quantitatively described by Stern-Volmer Equation, so the concentration of ions can be calculated through the fluorescence intensity.
PQDs can also detect small organic molecules under the aid of molecular imprinted polymers (MIPs). According to previous reports, MIPs are a kind of materials that are highly affinitive and selective to the target molecule, which is also called template. The preparation process of MIP is shown in Figure 11a). Huang et al. designed and fabricated a novel MIPs: APTES-capped CsPbBr₃ QDs to detect omethoate (OMT). They use APTES to cover the PQDs. APTES is hydrolyzed by the moisture in the air, and form a transparent silica matrix layer protecting PQDs. The functional groups can also form hydrogen bond with OMT. The fluorescence of PQDs can be selectively quenched by OMT, and this MIP is suspected to provide a path to transfer excited electron from PQDs to OMT. Tan et al. designed a similar MIP to detect phoxim. The sol-gel process of encapsulation is also a hydrolysis reaction driven by moisture in the air. The template phoxim formed a complex with functional monomer BUPTEOS via hydrogen bond and π-π accumulation.
Figure 11a) The schematic illustration of preparation of MIP. b) Schematic representation of the MIPs@CsPbBr3 QD sensor designed by Huang et al.[40]. c) Schematic illustration of the preparation of MIP/CsPbBr3 QDs composites designed Tan et al.[41].
Wang et al.\cite{42} encapsulated CsPbBr$_3$ perovskite quantum dots (CPBQD) into PMMA substrate, forming a nanoscale fiber membrane (CPBQD/PMMA FM) by electro-spinning method. This membrane can provide great durability for water and UV light. What’s more, it can be used for fluorescence resonance energy transfer (FRET) based detection. If the fluorescence spectra of a donor group coincide with the absorption spectra of an acceptor group, the excited donor can transfer energy to acceptor through nonradiative dipole-dipole coupling. This energy transfer mechanism is called FRET. In their work, they find proper molecular probe to adsorb target ion or molecular on the surface of CPBQD/PMMA FM. Then the FRET process occurs between the CPBQD/PMMA FM and target, leading to the quench of donor.

![Figure 1.2 Schematic illustration of (a) trypsin, (b) Cu$^{2+}$ and (c) pH fluorescence detection, based on the CPBQD/PMMA FM\cite{42}.](image)

4. Summary and outlook
In this review, the device structure, characteristics and mechanism of PQD photodetectors are systematically summarized, and some representative works are concluded in Table 1. PQD photodetector has a long way to go before being put into large-scale manufacture. There is still large space for improvement in the responsivity and stability of PQD photodetectors, and the response time can also be enhanced if the structure of device can be optimized. Besides, these great FET photodetectors need to become tiny enough and printable in order to be fabricated into photodetector arrays and utilized in specific scenes such as cameras. If those photodetector units are small enough, it will also be easier for electrons to transfer to electrodes before recombining with holes, and the response time will also decline. If PQD photodetectors can be used to manufacture cameras, the performance of the camera should be quite competitive.

PQDs can be improved in another direction when safety is taken into consideration. Most perovskite materials, including PQDs, are lead-based or tin-based. They are toxic due to the presence of heavy metals, but the toxicity can be prevented by various kinds of encapsulation, according to recent researches\cite{43}. But for quantum dots, the surface ligand layer can be regarded as encapsulation layer, which can be replaced by better ligand via ligand-exchange process\cite{29, 36}. Another layer of encapsulation can also be added such as MOFs\cite{38}. These kinds of encapsulation can improve the stability and performance of devices, and may also help to reduce the toxicity.

| Active Materials type | detection range (nm) | Response time rise/decay | responsivity (A/W) | Detectivity (Jones) | references |
|----------------------|----------------------|--------------------------|--------------------|---------------------|------------|
| colloidal CsPbBr$_3$ PQDs | photoresistor | X-ray | 30ms/27ms | 1450 $\mu$C | [21] |
| CsPbBr$_3$ NCs/Au NCs  | photoresistor | <540 | 0.2ms/1.2ms | 0.01 | 4.56E+08 | [22] |
|------------------------|---------------|-------|-------------|-------|-----------|-------|
| CsPbBr$_3$            | photoresistor | 9000ms/7000ms | 3.5 |       |          |       |
| CsPbBr$_3$-MPA(mp-TiO$_2$) | photoresistor | <500 | 4700ms/2300ms | 24.5 | 8.90E+13 | [26] |
| MAPbI$_3$ QDs on TiO$_2$ NTs array | photoresistor | 300-800 | 2-7s/1-4s | 0.2-1.3 | ~1E+12 | [28] |
| α-CsPbI$_3$ QDs/NaYF$_4$:Yb,Er QDs | photoresistor | 260-1100 | <5ms/<5ms | 1.5 |       | [23] |
| AET-CsPbI$_3$ QDs     | diode         | <700  | 0.5         |       | 5.00E+13 | [29] |
| CsPbCl$_{1.5}$Br$_{1.5}$ | FET            | ~450 | 7.5 ms /8 ms | 2.50E+04 |       |       |
| CsPbBr$_3$            | FET            | <540 | 6.5 ms /7 ms | 4.00E+04 | 2.00E+17 | [30] |
| CsPbBr$_{1.5}$I$_{1.5}$ | FET            | ~632 | 6.5 ms /7 ms | 2.50E+04 |       |       |
| SWNT/CsPbI$_3$        | FET            | <680 | <500m/400ms  | <2.8  |       | [31] |
| FAPbBr$_3$/Graphene   | FET            | <650 | 58ms/60ms(635nm) | 1.15E+05 |       |       |
| PQDs/MoS$_2$/MvdWH    | MESFET         | <670 | 590ms/320ms  | 7.70E+04 | 5.60E+11 | [33] |
| CsPbBr$_3$ QDs/MAPbI$_1$ | diode (down conv.) | 200-280 | <70ms/<70ms | 1.40E-03 | 2.40E+11 | [34] |
| CsPbX$_3$ QDs (X = Cl/Br = 1/2) | diode (down conv.) | 200-400 | 0.48ms/1.03ms | 0.054 |       | [35] |

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