

Emerging Quantum Dots Spotlight on Next-Generation Photovoltaics

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Jingxuan Chen, Xinyi Mei, and Xiaoliang Zhang*

School of Materials Science and Engineering, Beihang University, Beijing 100191, China

* Corresponding author, E-mail: xiaoliang.zhang@buaa.edu.cn

Abstract

Semiconducting quantum dots (QDs) received considerable attention for application in optoelectronic devices, such as solar cells, photodetectors and light-emitting diodes, due to their unique fundamental properties, including solution processability, size-dependent bandgap energies, high stability and low cost. Specifically, the suitable bandgap energy of QDs with strong light absorption in the visible and near-infrared regions makes them a kind of competitive photovoltaic materials toward next-generation photovoltaics. Herein, the advantages of emerging QDs, including infrared lead sulfide QDs and perovskite QDs, are highlighted for new generation photovoltaics, and the possible challenges and opportunities approaching high-performance solar cells are also proposed.

Key words: Quantum dot; Solar cell; Energy conversion; Photovoltaic

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Photovoltaic devices can directly convert solar energy into electricity at a low cost, which is believed as an important strategy to efficiently explore solar energy in a feasible way. The optoelectronic materials, working as photoactive materials in the photovoltaic devices, to a large extent affect the power conversion efficiency (PCE) of solar energy-to-electricity. Quantum dots (QDs), a kind of nano-sized semiconductors, received considerable contention for application in solar cells, light-emitting diodes (LEDs) and photodetectors due to their unique properties, such as solution processability, tunable bandgap energies (E_g), high stability and low cost.^[1,2] Specifically, for photovoltaic applications, by controlling the size of QDs, their light absorption spectra can be finely tuned in the visible and near-infrared wavelength regions, offering large freedom for constructing tandem or multijunction solar cells.^[3,4] Moreover, the multiple-exciton generation in QDs that more than one electron-hole pair can be produced after absorbing one high-energy photon provides a potential avenue to overcome the Shockley-Queisser theoretical limitation on the PCE of single-junction solar cells, approaching further usage of the solar energy.^[5] Notably, the theoretical PCE of a single-junction solar cell is generally determined by the E_g of photoactive materials (Figure 1a), and the ideal E_g for efficient solar cells is in a range of 1.1-1.4 eV to maximize its photovoltaic performance, which can be essentially obtained from QDs.

In a typical QD solar cell (QDSC), QD solid film is sandwiched between the electron transport layer (ETL) and hole transport layer (HTL), as shown in Figure 1b. Under the light illumination, the photon absorbed by the QD solid film is converted to be an electron-hole pair, which is subsequently sep-

arated as free charge carriers under the built-in-electric field and extracted by charge transport layers, finally transporting to external circuits through the electrodes.^[6] In the past few years, significant efforts were made to boost the photovoltaic performance of QDSCs by controlling the surface chemistry of QDs through surface ligand engineering of QDs and exploring novel charge transport materials or interface engineering (Figure 1c).^[7-10] Consequently, remarkable advances were obtained that the lead sulfide-QDSCs (PbS-QDSCs) with a PCE of 13.8% and high stability was achieved,^[11] showing high potential for next-generation photovoltaics.

Recently, perovskite quantum dot (Pe-QD) was also studied as a novel member of the QD family. For the typical ABX_3 (A= methylammonium (MA), formamidinium (FA) or Cs, B=Pb, X=Cl, Br or I) perovskite structure, the corner-sharing $[BX_6]$ octahedra form the frameworks of the perovskite lattice and larger A-site cations fill in the void.^[12] In 2014, all-inorganic Pe-QDs were synthesized for the first time using the hot-injection method.^[13] Due to the easy-controlling preparation process and outstanding optoelectronic properties of Pe-QDs, they have been expected to be an excellent candidate as light absorbers for solar cells, and they also show high potential for electroluminescence, laser, and optical detection devices.^[14] Compared with the bulk perovskite materials, Pe-QDs possess tunable E_g by controlling the experimental conditions for the synthesis of Pe-QDs, such as the injection temperature of precursors and the composition of halide in the Pe-QDs.^[15] Meanwhile, the bulk inorganic perovskites, such as $CsPbI_3$ perovskites, generally suffer from phase stability under ambient conditions due to their low structural tolerance factor.^[16] In contrast, Pe-QDs exhibit higher phase stability than bulk

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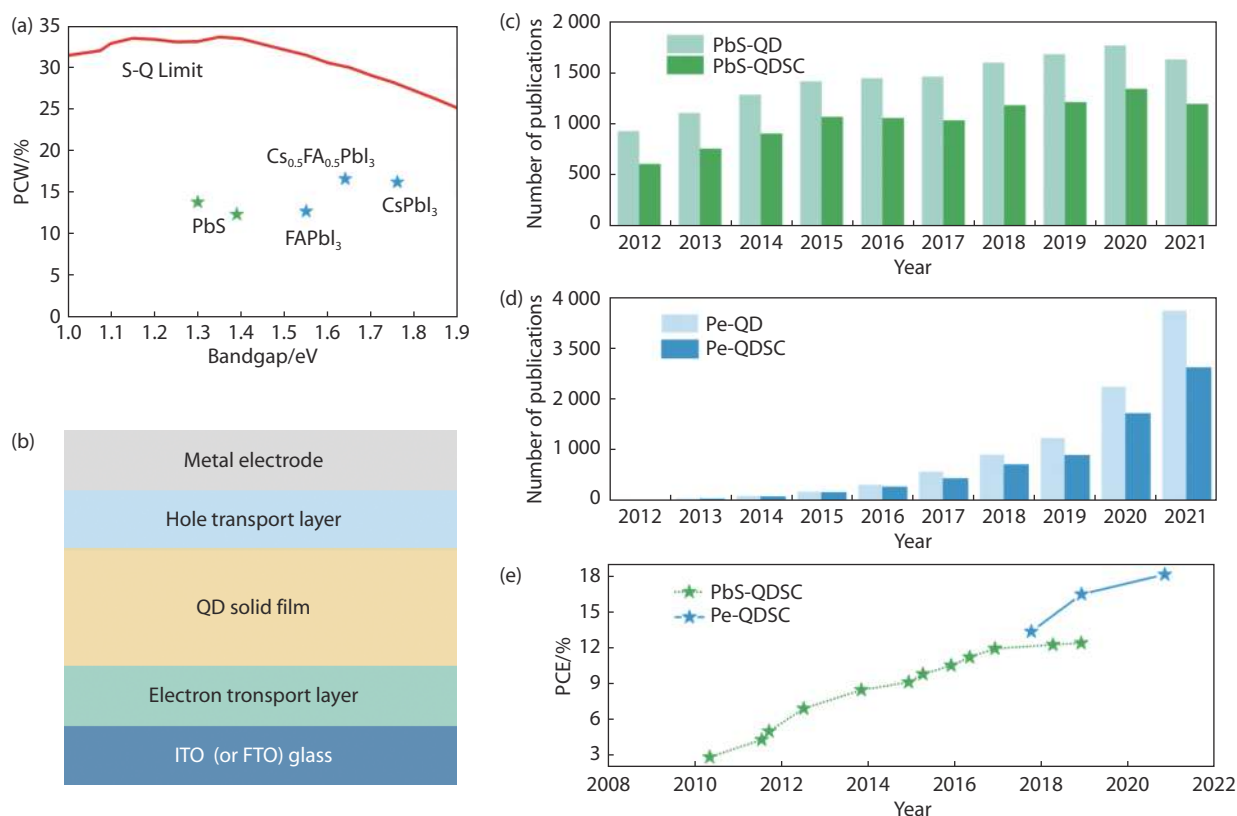


Fig. 1 (a) The Shockley-Queisser theoretical efficiency limitation of single-junction solar cells and the highest PCEs of QDSCs.^[11,18-21] (b) The device structure of QDSCs. (c) The number of annual publications (from 2012 to 2021) of PbS-QD (searched using the keyword: PbS quantum dot) and PbS-QDSC (searched using the keyword: PbS quantum dot solar cell). The data were retrieved from Scopus. (d) The number of annual publications (from 2012 to 2021) of Pe-QD (searched using the keyword: perovskite quantum dot) and Pe-QDSC (searched using the keyword: perovskite quantum dot solar cell). The data were retrieved from Scopus. (e) Evolution of the certified PCEs of QDSCs (from National Renewable Energy Laboratory (NREL)).^[22]

perovskites due to the size-induced lattice strain and controlled surface chemistry of Pe-QDs.^[17] As such, the phase stability of CsPbI₃ Pe-QDs was significantly improved, and CsPbI₃ Pe-QDSC (Pe-QDSC) with a high PCE was obtained.^[15] The Pe-QDs and Pe-QDSCs are now receiving significant attention and the number of publications is also increasing (Figure 1d).

Compared with the PbS-QDSCs, the CsPbI₃ Pe-QDSCs exhibit a higher open-circuit voltage (V_{oc}) with a lower short-circuit photocurrent density (J_{sc}) due to a high E_g of Pe-QDs, and thus the Pe-QDSCs could harvest fewer infrared photons in the solar spectrum than PbS-QDSCs.^[35,36] Recently, studies revealed that the light absorption spectra of Pe-QDs can be expanded to the near-infrared region through adjusting the composition of Pe-QDs, providing a chance for further improving the light absorption of Pe-QDSCs in the infrared region.^[37] Very recently, the certified efficiency of Pe-QDSCs was reported (PCE=16.6%),^[19] which is very promising for new generation QDSCs (Figure 1e). The detailed photovoltaic parameters of these high-performance QDSCs were summarized in Table 1.

The PbS- and Pe-QDs capped with long-chain ligands, such as oleic acid (OA) and oleylamine (OAm), are generally synthesized in a liquid condition forming a stable colloidal system. When constructing QDSCs, the QD solid films were deposited from the QD colloidal solution and the long-chain ligands were removed or replaced by short ones to facilitate the

charge transfer within the QD solid films.^[38,39] Meanwhile, the large surface area of QDs results in numerous defects forming on the QD surface, which need to be well passivated during the ligand exchange. Thus, various ligand exchange approaches and post-treatment of QD solid films were studied to increase the carrier mobility and lower the defect density of Pe-QD solid films.^[12,40] However, compared with the bulk films, the defect density of QD solid films is still very high, which could capture the photoinduced charge carriers, pre-

Table 1. The device photovoltaic performances of the certified PCEs of QDSCs by the NREL.

QD type	PCE (%)	V_{oc} (V)	J_{sc} (mA/cm ²)	FF	Ref
PbS-QDs	2.94	0.59	8.92	0.55	[23]
PbS-QDs	4.4	0.52	18.14	0.46	[24]
PbS-QDs	5.1	0.54	14.6	0.62	[25]
PbS-QDs	7	0.60	20.1	0.58	[26]
PbS-QDs	8.55	0.55	24.2	0.64	[27]
PbS-QDs	9.61	0.63	22.95	0.66	[28]
PbS-QDs	9.88	0.63	21.6	0.71	[29]
PbS-QDs	10.6	0.61	24.3	0.71	[30]
PbS-QDs	11.28	0.61	27.23	0.68	[31]
PbS-QDs	12.01	0.65	29.04	0.64	[32]
PbS QDs	12.33	0.66	27.5	0.67	[20]
PbS-QDs	12.47	0.65	28.63	0.66	[33]
Pe-QDs	13.43	1.16	15.25	0.76	[34]
Pe-QDs	16.6	1.17	18.30	0.78	[19]

dominantly affecting the charge carrier extraction of QDSC devices. Thus, further studies to diminish the defect density of QD solid films are highly required in the future.

Along with the fundamental studies on the surface chemistry of QDs and insight into the device physics of QDSCs, the photovoltaic performance of PbS- and Pe-QDSCs was largely improved. However, both PbS- and Pe-QDs hold different advantages for next-generation solar cells. For instance, PbS-QDSCs show high stability with relatively low efficiencies due to the high surface defect density and covalent bond of PbS-QDs, whereas Pe-QDSCs demonstrate higher efficiencies with relatively low stability owing to the electrovalent bond of Pe-QDs. Meanwhile, the PCEs of resulting QDSCs are still largely lagging behind their theoretical efficiencies, suggesting that there is a large room to improve the photovoltaic performance of QDSCs (Figure 1a). The long-term operational stability of QDSCs is also highly required for commercial applications, which is due to the humidity and oxygen corrod- ing during the device operation under continuous illumination. With the encapsulation of QDSCs and/or the improve- ment of surface passivation of QDs, we believe that the stabil- ity of QDSCs could be improved.

The QD shows a tunable light absorption spectrum by con- trolling QD size, suggesting that the QDSCs could be coupled with other photovoltaic materials to complementarily har- vest photons from the solar spectrum. For instance, the in- frared PbS-QDSC could be integrated with the visible solar cells, such as perovskite solar cells or organic solar cells, to construct tandem solar cells, extending the light absorption spectrum of these solar cells and thus enhancing photovolta- ic performance.^[3] The Pe-QDSC with a high V_{oc} could also be stacked with the infrared solar cells, such as silicon solar cells, to further improve their photovoltaic performance. The QDs with different E_g could also be applied for the fabrication of tandem or multijunction solar cells, opening a new avenue for device structure engineering.

Notably, the highly efficient QDSCs with small sizes are mostly studied under lab conditions using the feasible spin- coating method for the deposition of QD solid films. Theoret- ically, the pre-synthesized QDs can be applied for the scal- able fabrication of QDSCs using the solution-processed pro- duction approaches, such as roll-to-roll spraying or blade- coating methods. Since the deposition of the large area QD solid film with high quality and uniformity remains a chal- lenge, the studies on the large-sized QDSCs are still rarely conducted, which could be overcome through the solvent engineering of QD solutions or advanced deposition tech- niques of QD solid films.

Overall, the substantial advances and insight into the device operation of QDSCs have forwarded the QDSCs to a new arena and made QDSCs more competitive among emer- ging solar cells, paving a strong cornerstone and big step to- ward commercialization. We believe that with more funda- mental studies made on the surface chemistry of QDs and feasible scalable fabrication of QDSCs, as well as device archi- tecture engineering, QDSCs will become more spotlighting on next-generation solar cells.

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Conflict of interest

The authors declare no conflict of interest.

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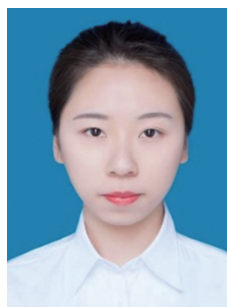
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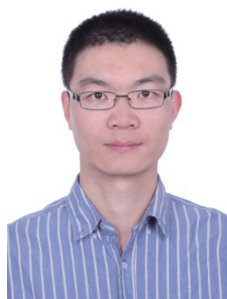
Biographies



Jingxuan Chen is a PhD student at Beihang University, China. She received her Bachelor's degree in materials science and engineering from Nanjing University of Aeronautics and Astronautics, China, in 2018. Currently, she is conducting her PhD studies under the guidance of Prof. Xiaoliang Zhang. Her current research interest mainly focuses on utilizing surface management to improve the photovoltaic performance of quantum dot solar cells.



XinYi Mei earned her Bachelor's degree from Central South University, China, in 2019. She is now pursuing her PhD degree in Materials Physics and Chemistry at Beihang University, under the supervision of Prof. Xiaoliang Zhang. Her current research interest mainly focuses on the novel physical properties of low-dimensional optoelectronic materials, such as quantum dots, and their application in light-emitting devices.



Xiaoliang Zhang is a professor at Beihang University, China, and directs a research group working on optoelectronic materials and devices. He received his PhD degree in Materials Physics and Chemistry from Beihang University in 2013. Then, he joined Uppsala University, Sweden, as a postdoctoral researcher and subsequently was promoted as a senior researcher at Uppsala University. He joined Beihang University as a full professor in 2018. He has a particular interest in semiconducting quantum dots for application in new generation optoelectronic devices.