Research article

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Improving the efficiency of silicon solar cells using *in situ* fabricated perovskite quantum dots as luminescence downshifting materials

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Abstract: Luminescence downshifting (LDS) layer integration has been proven to be an efficient way to ameliorate the poor UV-blue spectral response and improve the power conversion efficiency (PCE) for solar cells (SCs). By employing an *in situ* fabricated $\text{CH}_3\text{NH}_3\text{PbBr}_3$ (CH_3NH_3 = methylammonium, MAPbBr₃) quantum dot/polyacrylonitrile (PAN) composite film as the LDS layer, we observed a clear enhancement in the external quantum efficiency (EQE) for silicon SCs, predominantly in the UV-blue region. With a theoretically calculated intrinsic LDS efficiency (η_{LDS}) of up to 72%, silicon SCs with the LDS layer exhibited an absolute value of 1% for PCE improvement in comparison to those without the LDS layer. The combination of easy fabrication and low cost makes it a practical way to achieve photovoltaic enhancement of Si-based SCs.

Keywords: perovskite; quantum dots; composite films; luminescence downshifting; silicon solar cells.

1 Introduction

Photovoltaic (PV) technologies based on silicon semiconductor have made considerable strides in the past decades

and been most widely applied in improving the power conversion efficiency (PCE), which has successfully gone beyond 20% either in the laboratory or in large-scale production [1–5]. Limited by the indirect bandgap of 1.2 eV, silicon-based solar cells (SCs), no matter crystalline or amorphous, show almost transparent spectral response toward photons with energy below its bandgap. A number of works have therefore been devoted to extend the spectral response of silicon-based SCs to infrared region so as to enhance the photocurrent as well as the PCE [6–12]. However, because of the high reflection coefficient (reflective index) and shallow penetration depth of UV-blue irradiation (e.g. less than 100 nm for 400 nm wavelength) [13–15], another challenging approach to further boost the PCE of silicon is to strengthen the UV-blue responsivity in order to utilize more high-energy photons for photocurrent generation instead of substantial thermal losses [16–19].

Since silicon SCs have relatively strong absorption in the middle wavelength region in comparison to UV-blue shortwave length region of visible irradiation, luminescence downshifting (LDS), which converts the higher energy photons that cannot be sufficiently utilized into lower energy photons that can be well used for photocurrent generation, should be a straightforward route to improve the UV-blue response. By dispersing luminescent species, such as organic dyes, rare earth element luminophores, and quantum dots, in a transparent matrix and applying them in front of the SCs, LDS integration does not need any adjustment in device architecture or output electrical circuit for given SCs [20]. Generally, the ideal LDS integration for SC integration should satisfy the following essential requirements. First, the luminescent species should have high photoluminescence quantum yield (PLQY) but little absorption overlap in the response region of PV materials. Organic dyes have impressive photoluminescence (PL) properties, but photobleaching and the overlap between absorption and emission bands is a great drawback, which results in

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serious self-absorption within the LDS layer [21–24]. The narrow absorption band and extremely low absorption coefficient require rare earth element luminophores to have ligands or absorbers for transferring energy into the luminescent ion, but extending the absorption range is still an issue [25–29]. Because of the quantum confinement effect, quantum dots (QDs) not only possess excellent optical properties but also have easy tunability for both absorption and emission [30–35]. Nevertheless, QDs with high specific surface energies are prone to aggregation after incorporation in host materials, leading to PL quenching during film processing [36]. Second, matrix materials have to exhibit high transmittance and suitable refractive index ($n=1.4\text{--}2.4$) to prevent scattering and absorption, especially in the spectral region where the SCs have strong response [37, 38]. Third, the matrix also needs to provide the optimum dissolution capability for dispersing the luminescent species in order to ensure that the LDS layer uniform [30, 39, 40]. In addition, simple process, low cost, and fabrication scale-up of the LDS layer indeed are beneficial for large-scale integration with silicon SCs. Therefore, LDS integration by combining LDS composition with the appropriate matrix is a challenging but impactful means for improving the performance of SCs.

Recently, because of their outstanding optical properties, perovskite quantum dots (PQDs) have attracted much attention. They also offer an unparalleled level of tunability of their absorption and emission spectra due to the matched refractive index [41–43]. More importantly, different from other LDS layer processing schemes, thermal injection synthesis of QDs followed by solution dispersion in the matrix [44, 45] allows PQDs to be uniformly embedded in a polymer matrix by *in situ* fabrication, as we have previously shown [46, 47], which can efficiently overcome the issue of irreversible nanoparticle aggregation and undesirable luminescence quenching. In this work, we first demonstrate an easy but efficient route to integrate *in situ* fabricated PQD/polyacrylonitrile (PAN) composite films with commercial silicon SCs. With a theoretically calculated intrinsic LDS efficiency (η_{LDS}) of up to 72%, silicon SCs with the LDS layer exhibited an absolute value of 1% PCE improvement in comparison to those without the LDS layer due to the strengthened spectral response in the UV-blue region. Besides, *in situ* fabrication from a solution precursor ensures that such composite films are greatly beneficial for simplifying the LDS layer fabrication and scale-up in comparison to other LDS integration methods.

2 Experimental section

2.1 Fabrication

For fabricating the PQDs/polymer composite film (PQDCF), a precursor solution was prepared by dissolving a mixture of 0.8 mmol $\text{CH}_3\text{NH}_3\text{Br}$ (MABr), 0.4 mmol PbBr_2 , and 1 g PAN powder into 8 ml *N,N*-dimethylformamide (DMF). The precursor solution was then spin-coated onto the surface of quartz substrates and PV devices, forming a transparent layer. After that, the substrates precoated with the transparent film was moved into a vacuum oven and kept at 40°C for 30 min to remove the residual solvents. The thickness of the PQDCFs could be controlled from 1 to 4 μm by varying the spinning speed.

2.2 Material characterization

UV-vis transmittance spectra of the PQDCFs were measured on a UV-6100 UV-vis spectrophotometer (Shanghai Mapada Instruments Co., Ltd., China). The PL spectra were recorded using an F-380 fluorescence spectrometer (Tianjin Gangdong Sci. & Tech. Development Co., Ltd., China). The absolute PLQYs of PQDCFs were determined using a fluorescence spectrometer with an integrating sphere (C9920-02, Hamamatsu Photonics, Japan) and excited at the wavelength of 397 nm using a UV LED light source. Time-resolved PL (TRPL) measurements were made using a fluorescence lifetime measurement system (C11367-11, Hamamatsu Photonics). The thickness and refractive index of PQDCFs were measured by an Ambios XP-200 surface profiler and an ellipsometer (M-2000D), respectively. Ultrathin section samples were analyzed using a JEOL-JEM 2100F transmission electron microscope (TEM) operating at an acceleration voltage of 200 kV. X-ray diffraction (XRD) measurements were carried out on a Bruker D8 FOCUS X-ray diffractometer. The Fourier transform infrared (FTIR) measurements were made by using a Thermo Fisher Scientific Nicolet 8700 spectrometer in transmission mode. X-ray photoelectron spectroscopy (XPS) measurements were carried out on a PHI QUANTERA-II SXM X-ray photoelectron spectrometer.

2.3 Electrical measurements

The illuminated current density vs. voltage (*J*-*V*) characteristics were measured under AM 1.5G illumination (100 mW/cm², 25°C calibrated by solar simulator

XES-155S1Class AAA, San-Ei Electric). The EQE spectra were obtained with an incident photon-to-current conversion system (QE-R, Enlitech).

3 Results and discussion

PAN films were selected as matrix materials because of their following unique features to meet the requirements for the LDS layer: First, PAN exhibits high transmittance, particularly in the region where the SCs have strong response. Second, PAN provides an optimum environment for the dispersion of PQDs to suppress agglomeration. Third, high thermal conductivity and UV stability of PAN favor PV applications. In addition, PAN can form very uniform, high-quality films through spin-coating [48, 49]. As shown in Figure 1A, from the transmittance and PL emission spectral characterization of PQDCF, an impressively high transmittance >90% could be observed in visible-infrared region, which would hardly influence the light harvest of silicon SCs. The good transparency of the composite film indicates the *in situ* formation of small-sized MAPbBr₃ PQDs with uniform distribution in the polymeric matrix. Meanwhile, PQDCF exhibits a brilliant green emission peak at 531 nm with a full width at half-maximum (FWHM) of 21 nm under UV irradiation and the maximum PLQY of up to 95 ± 1% at 397 nm. The optical properties of PQDCFs with various thicknesses on the quartz substrates were also examined (see Figures S1 and S2). The results prove that PQDCF should be an ideal LDS layer for improving the UV-blue spectral response without disturbing any other light utilization for SCs.

To further understand the dynamics of excitons in PQDCFs, we used TRPL to measure the PL lifetime of PQDCFs. Figure S2B shows the dynamics of PQDCFs with different thicknesses monitored at 531, 529.2, 528.6, 526.8, and 525 nm. The decay curves at different thicknesses exhibit nearly a single-exponential behavior with the

corresponding decay times of 11.96, 10.99, 10.25, 10.01, and 8.79 ns (Figures S1–S5). All samples showed a much shorter average lifetime than that of bulk perovskite films (~100 ns) [41], which indicates that PL decay of PQDs mainly occurs by geminate electron-hole recombination due to the spatial confinement of electron-hole pairs inside the PQDs [50]. Excitation power-dependent integrated PL intensity of PQDCF is presented in Figure S2C. The PL is centered at around 531 nm with 405-nm laser excitation, and its intensity increases with the excitation power. A nearly linear relation between the integrated PL intensity and the excitation power was observed, which suggested high-quality PQDs embedded in the PAN matrix [51]. The refractive indices of pure PAN film and PQDCF were extracted via ellipsometry at each wavelength, as shown in Figure 1B. Compared to pure PAN film, PQDCF has a high value of refractive index in the wavelength range 370–900 nm, which could provide better coupling efficiency for Si₃N₄-coated silicon SCs. In addition, it is worth mentioning that, considering the application as LDS layers for PV devices, we determined the stability of the as-fabricated PQDCFs against UV radiation. PQDCFs show only a ~4% decrease of PLQY under UV 365 nm irradiation (5 W, SL8300-UV, Hong Kong Sunlonge INT'L CO., Limited) for 60 days, as shown in Figure 1C. Such photostability can be attributed to the barrier property of compact PAN matrix, which could also improve the life of LDS-integrated SCs. Therefore, the combination of high PLQY, high transparency, high photostability, and matched refractive index make the PQDCFs very promising for down-shifting applications.

The phase structure of PQDCFs was carefully examined to further understand the micro-interaction between PQDs and the PAN matrix. The TEM image of PQDCFs in Figure 2A shows 3–5 nm MAPbBr₃ QDs with good dispersion in the PAN matrix. According to the high-resolution TEM (HRTEM) image in Figure S3A and the corresponding fast Fourier transform (FFT) image in Figure S3B, interplanar distances of 2.07 and 3.09 Å were identified, corresponding to the

Figure 1: Optical properties and photostability of PQDCFs.

(A) Transmittance (black) and PL (red) spectra of PQDCF with the corresponding photographs under ambient daylight (left) and under a UV 365 nm lamp (right), shown in the inset. (B) Refractive index of PAN and PQDCF in wavelength region 370–900 nm. (C) Photostability of a typical PQDCF against UV illumination (UV 365 nm lamp, 5 W).

Figure 2: Morphological and structural characterization of PQDCFs.

(A) Typical TEM image of MAPbBr₃ QDs in PAN matrix. (B) XRD patterns of the pure PAN film and PQDCFs. (C) FTIR transmission spectra of pure PAN and PQDCFs.

(200) and (220) planes of MAPbBr₃, respectively. The formation of MAPbBr₃ QDs in the polymeric matrix can be identified from the XRD patterns shown in Figure 2B. Pure PAN has one sharp crystalline peak at $2\theta = 17^\circ$, corresponding to the orthorhombic PAN (110) reflection [52, 53]. In contrast, PQDCFs showed diffraction peaks at 14.9° , 21.3° , 30.2° , 33.7° , 43.2° , and 45.9° , corresponding to the lattice planes of (100), (110), (200), (210), (220), and (300) of MAPbBr₃ (space group: *Pm3m* no. 211). XPS was further used to characterize the formation process of MAPbBr₃ QDs in the PAN matrix. As shown in Figure S4, the detected Br 3d and Pb 4f peaks of PQDCF in comparison with pristine PAN film clearly revealed the existence of MAPbBr₃ QDs in the *in situ* fabricated PQDCF. Besides, on the basis of FTIR spectroscopy

measurements shown in Figure 2C, the vibration peak at 2245 cm^{-1} is assigned to the C≡N stretching of PAN and the peak at 1662 cm^{-1} together with that at 1732 cm^{-1} can be assigned to C=O stretching [52]. Moreover, an absorption band emerges at around 2340 cm^{-1} for the mixtures, which is due to the dipole-dipole interaction between PAN and DMF molecules [54]. In the case of PQDCFs, we also can clearly detect the N-H stretch ($3000\text{--}3300\text{ cm}^{-1}$), C-H bend ($1450\text{--}1550\text{ cm}^{-1}$), and C-H stretch ($2850\text{--}3000\text{ cm}^{-1}$), which also confirms the presence of MAPbBr₃ QDs [50]. All these results are consistent with previous observations of high-quality PQDs embedded in PAN matrix.

Figure 3A shows a schematic of the configuration of a silicon SC coated with PQDCF and the concept of solar

Figure 3: Photovoltaic performance of c-Si and mc-Si SCs with and without PQDCFs.

(A) Schematic diagram of PQDCF to enhance the PCE of SCs. (B) Photograph of PQDCF coated on the silicon SC. (C, D) J-V curves of c-Si and mc-Si SCs coated with and without PQDCFs at scan speed 10 mV s^{-1} . (E, F) EQE curves of c-Si and mc-Si SCs coated with and without PQDCFs.

spectrum downshifting in an LDS. In order to examine the effect of PQDCFs on SC application, we directly *in situ* coated the PQDCFs onto the surface of textured silicon SCs (area: $0.8 \times 2.5 \text{ cm}^2$) with Si_3N_4 antireflective layer by spin coating, as shown in Figure 3B. The current density vs. voltage (J - V) characteristics of single-crystalline silicon (c-Si) and multicrystalline silicon (mc-Si) with and without the PQDCFs integration are shown in Figure 3C and D, and all the data have been summarized in Table 1. Compared to the reference cells, a synergistic enhancement could be observed in LDS-integrated devices in the open-circuit voltage (V_{oc}), short-circuit current (J_{sc}), and fill factor (FF), which is an absolute value of almost 1% increment in PCE of SCs. Clearly, because of the LDS and antireflection effects, PQDCF could improve the photon utilization of the radiation by converting the high-energy photons in UV-blue region, which will be insufficiently used, into low-energy photons in the visible region where the SCs have strong spectral response. This is equivalent to extending the response range and improving the radiation intensity for silicon SCs. In that case, the increase in PCE should be mainly due to increased photocurrent generation. To check repeatability, the performance of 10 mc-Si SCs and 5 c-Si SCs coated with LDS layers was measured randomly (Tables S2 and S3). The PCEs of mc-Si SCs and c-Si SCs were in the range 10.3%–11.8% and 12.7%–15%, respectively, and the efficiency gain varied from 0.4% to 1%, indicating good reproducibility (Figure S5). To further verify the spectral modulation of PQDCF, we carried out EQE measurements to examine the wavelength-dependent response of silicon SCs coated with and without PQDCF (Figure 3E, F). Obviously, in comparison to the reference, the EQE values rose from 70% to 80% for c-Si and from 40% to 60% for mc-Si at 400 nm. Improved EQE values in the short wavelength region led to improvement in J_{sc} , which is in accordance with J - V measurements.

In order to promote the practical application of the PQDCF, we further integrated the PQDCFs onto the surface of standard large commercial c-Si (Code no. T1S2-GC061) and mc-Si (Code no. T1M2-V0761) SCs (area: $15.6 \times 15.6 \text{ cm}^2$, Tainergy Tech. Co., Ltd., Taiwan), as shown in Figure 4A and B, respectively. The J - V curves of the standard

commercial c-Si and mc-Si SCs with and without the LDS layer are shown in Figure 4C and D, from which we also find a clear improvement of PCE from 16.1%/16.7% to 16.4%/17.2% for mc-Si and c-Si SCs, respectively. These results indicate that the photoresponse of silicon SCs in the UV-blue region could be efficiently enhanced by PQDCF integration, which realized efficient downshifting of the high-energy photons beyond 2.5 eV and improved the photon utilization.

Further, a theoretical model to simulate the effect of the LDS layer on the performance of the PV cell was constructed for theoretical evaluation [31, 55]. A brief description of the theoretical model and the related equations are given in Supplementary Section S10. A theoretical simulation was performed according to the EQE of the reference PV cell, the measured PLQY, and the absorption and emission spectrum of the LDS layer. The efficiency of a complete SC with an LDS layer, $\eta(\lambda)$, can be modeled by using the absorption and emission spectra of the LDS [Eq. (1)]:

$$\eta(\lambda) = [1 - A(\lambda)] \cdot \eta_b(\lambda) + A(\lambda) \cdot \eta_{LDS} \cdot \max(\eta_b), \quad (1)$$

where $\eta(\lambda)$ is the EQE of the LDS layer-coated SC at a specific wavelength, $\eta_b(\lambda)$ is the EQE of the reference SC, $A(\lambda)$ is the fraction of solar spectrum absorbed in the LDS layer, η_{LDS} is the intrinsic LDS layer efficiency, and $\max(\eta_b)$ is the maximum external quantum efficiency of the reference SC.

$$\eta_{LDS} = \text{PLQY} \cdot (1 - R) \cdot (1 - \eta_{out}) \cdot (1 - \eta_{abs}) \quad (2)$$

The LDS layer efficiency, η_{LDS} [Eq. (2)], is governed by the PLQY of the LDS layer, the reflectivity (R), out-coupling loss (η_{out}), and re-absorption loss (η_{abs}). The out-coupling loss η_{out} due to reflection on the air-LDS layer interface is calculated for a refractive index of 1.56, which corresponds to ~12% loss according to Snell's law. The re-absorption loss η_{abs} is calculated to be ~8% based on the spectral overlap of the absorption and emission spectra (Supplementary Section S10). Based on theoretical calculation, η_{LDS} is estimated to be 72%.

The theoretical model was further used as a predictive tool to evaluate the effect of PLQY, as illustrated in Figure S6. It shows the simulated EQE curves for c-Si SC with various PLQYs, compared to the experimentally determined EQE of the PQDCF-integrated devices. The simulations show that the EQE in the UV-blue spectral region obviously increases with the increase of PLQY, and such theoretical simulation results closely match the measured EQE of c-Si SC with PQDCF. Based on the outcome of the theoretical prediction, it was concluded that developing

Table 1: Comparison of PV parameters of silicon SCs with LDS layer and reference SCs.

Device	V_{oc} (V)	J_{sc} (mA/cm ²)	PCE (%)	FF (%)
c-Si reference SC	0.59	38.67	14.05	61.6
c-Si SC with LDS layer	0.60	40.23	14.99	62.1
mc-Si reference SC	0.57	32.35	10.44	56.6
mc-Si SC with LDS layer	0.59	33.02	11.32	58.1

Figure 4: Photovoltaic performance of standard commercial c-Si and mc-Si SCs with and without PQDCFs. (A, B) Photographs of PQDCFs coated on standard commercial c-Si and mc-Si SCs under UV irradiation. (C, D) J-V curves of standard commercial c-Si and mc-Si SCs coated with and without PQDCFs with scan speed 10 mV s^{-1} .

PQDCF with high PLQY is essential for PV applications and that our model is suitable for predicting the effect of PQDCF as the LDS layer on the performance of SCs.

4 Conclusion

In summary, we demonstrated the *in situ* fabrication of PQDCFs as LDS layers with excellent optical properties of high transparency ($>80\%$ at $\lambda > 500 \text{ nm}$), high PLQY ($>95\%$), matched refractive index of 1.56, and excellent photostability. By integrating such PQDCFs with commercial silicon SCs, an absolute value of about 1% improvement in PCE could be observed either in c-Si or in mc-Si SCs as a result of the extended spectral response and increased photocurrent. Theoretical simulations revealed that a high PLQY leads to an increase in EQE in the UV-blue region, which was supported by experimentally obtained results of SCs integrated with PQDCFs and provide a

theoretically calculated η_{LDS} of intrinsic LDS layer of up to 72%. Overall, based on its low cost and excellent optical properties, we believe that such *in situ* fabricated PQDCF must be a strong candidate for further application in photoelectric device optimization.

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