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# Antimonene quantum dot-based solid-state solar cells with enhanced performance and high stability

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ABSTRACT

Quantum-dot-sensitized solar cells (QDSSCs) usually suffer from a strong recombination effect or low stability, resulting in a low efficiency and limiting their practical application. A facile liquid-phase synthesis of antimonene quantum dots (AMQDs) with unique morphology and size distribution is demonstrated, and applied as an effective photoactive material in QDSSCs for the first time. The intrinsic advantages of AMQDs, in association with a strong light–matter interaction, moderate energy band-gap in the visible range and antioxidation properties, enable an impressive photoelectric conversion efficiency (PCE) to be obtained, where the hysteresis-free AMQD-based solar cells can reach up to 3.07%. Furthermore, the as-fabricated solar cell shows a long-term stability, where the PCE value maintains to more than 90% of the initial value after 1000 h. In view of highly cost-effective synthesis and outstanding photoelectrochemical performance, we anticipate that AMQDs will have a promising future in solar cells and related photonics applications. AMQDs may provide a new direction for the rapidly developing photovoltaic technology, which is necessary for further developments in this field.

## 1. Introduction

Photovoltaic devices that can convert solar energy into electrical energy are considered to be effective for solving the fundamental problems of the increasingly serious energy requirements and the depletion of fossil fuels [1]. One of these devices, quantum-dot-sensitized solar cells (QDSSCs), has attracted much interest owing to the photoelectric conversion efficiency (PCE) that can theoretically reach up to 66% [2–4]. To date, the most widely adopted inorganic semiconductor quantum dot light harvesters have mainly been the Pb-based (PbS [5–7], PbSe [8]), Cd-based (CdTe [9–11], CdSe [12,13], $CdSe_xTe_{1-x}$ [14]), chalcogenide ($Sb_2S_3$ [15], $CuInS_2$ [16]), and other (CuInGaSe [17], AgInSe [18], ZnCuInSe [19]) materials [20–22]. Similar to the dye sensitized solar cells (DSSCs), electrons generated in the quantum dot sensitizers will be injected into a wide bandgap semiconductor, such as $TiO_2$, and the holes created in the quantum dot sensitizer will simultaneously pass through the hole conductor to the metal electrodes [23].

Traditional liquid quantum dot solar cells have been found to experience a strong corrosion effect on the quantum dots and the electrolyte can easily come into contact with the electron conductor to cause the generation of dark current, which greatly limit their service life and efficiency [24,25]. Therefore, QDSSCs with a solid electrolyte are expected to avoid leakage and evaporation, as well as the back-charge transfer problems associated with liquids, which results in the improvement of the long-term stability [26,27]. However, as the core of QDSSCs, the QD sensitizers are involved in the light trapping, charge separation and transmission of the solar cells. However, QD sensitizers usually possess the characteristics of photodegradation and photo-instability owing to the easy recombination of holes and electrons from the surface of the quantum dots [28]. Consequently, expanding the light absorption range and reducing electron recombination will be an effective approach to improve the photovoltaic power generation [1,9,29,30].

A number of investigations have shown that two-dimensional (2D) materials are suitable for optoelectronic devices such as QDSSCs [31]. 2D materials can be easily tuned by changing the number of layers, sizes, stress level to finely control the band gap, and thereby, researchers have been able to achieve an absorption of the near-infrared and short-wave infrared light [32]. In addition, 2D materials have

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attracted attention owing to their superior physicochemical properties [33,34]. Among them, graphene, graphene-like materials such as black phosphorus (BP) [31,35,36], $MoS_2$ [37,38], antimony (Sb) [39,40], and perovskites have been applied in the field of optoelectronics during the past decades [35,41–44]. Perovskites have a wide absorption range and excellent absorption coefficient, but perovskite solar cells (PSCs) usually suffer from a hysteresis effect in the current–voltage measurements, which leads to an inaccurate estimation of the device efficiency [45–47]. Additionally, PSCs generally exhibit stability problems, where many devices undergo decay during testing, which has slowed down the commercialization of PSCs [48,49].

Graphene quantum dots show a sizeable energy band-gap but suffer from complex manufacturing processes, poor flexibility, high cost, and poor sustainability [41,50]. BP QDs have been successfully prepared by using liquid crystal stripping technology with probe-type ultrasonic and water baths, and have been applied to solar cells [51,52]. An organic solvent has been used to improve the interface and has shown excellent photoelectrochemical properties with a PCE value of 6.58% [52]. Although great advancements have been made for BP photodetectors, the stability of BP remains as a serious drawback [53,54].

Sb belongs to the same group of elements as BP and its layered structure resembles that of BP [55]. However, Sb exhibits shorter out-of-plane atom-to-atom distances, which indicates a greater stability [56,57]. In addition, Sb is unique among other 2D materials because of its strong spin-orbit coupling [33]. However, the Sb semiconductors reported thus far feature band gaps that are 2.28 eV, which has greatly restricted their applications, especially in optoelectronic devices, because this band gap corresponds to a photoresponse in the blue and UV range [58]. Many comprehensive articles have shown that the band gap of antimony can be regulated by the number of layers, so that it can absorb from the visible to the infrared wavelengths of radiation [59,60]. Tao et al. [61] reported the effective strategy of liquid phase exfoliation to obtain few-layer antimonene quantum dots (AMQDs) for photothermal cancer therapy, which has facilitated the implementation of AMQDs in practical applications. Zhang et al. [34] took advantage of epitaxial growth methods to synthesize high-quality few-layer antimonene monocrystalline polygons, which can be applied to a flexible substrate photoelectric. These features, combined with the first-principles calculations, predict that antimony has excellent photoelectric properties, and its internal electron structure is the most stable buckle-like structure [58], with high carrier mobility (1000 $cm^2/Vs$) [32], high electrical conductivity ($10^4$ S $m^{-1}$) [34], excellent thermal conductivity (200 W $Mk^{-1}$) [43], superior refractive index, and directional optical transparency [32]. Moreover, Sb shows good optical transparency in the visible light range, which is promising for transparent conductive electrode applications [32]. These properties indicate that Sb may be potentially useful for optoelectronic devices or solar cell applications and has potential in the fabrication of new flexible substrate photovoltaic devices [62,63]. However, research on the photoelectric conversion of antimonene-based quantum dots is still required.

Herein, we develop a novel liquid exfoliation technique for preparing a high yield of AMQDs, which are then applied as the photoactive layer in QDSSC. AMQDs, with the general features of a 2D material, have not been studied in QDSSC as a sensitizer. However, AMQDs show good optical absorption owing to the band gap ($E_g$) of approximately 1.5 eV which is near to the ideal optical $E_g$ for a solar absorber [64], a wide absorption range, and high light stability. Hence, AMQDs are very promising photoactive candidates for improving the photovoltaic performance, device stability, and fabrication cost of QDSSCs. The PCE of QDSSCs based on AMQDs as sensitizers achieve a very encouraging PCE of 3.07% in the forward and reverse scan modes (without employing any interfacial optimization) and an improved ambient stability. This study demonstrates the great potential for achieving a cost-effective and stable photoactive sensitizer.

## 2. Experimental

### 2.1. Materials

Titania ($TiO_2$) paste (Dyesol 18NRT, 20 nm average particle size), dimethylformamide (DMF, AR, Aladdin), butanol (AR, Aladdin), *N*-methyl-2-pyrrolidone (NMP, AR, Aladdin), polyvinylidene fluoride (PVDF), tert-butanol, acetone, and ethanol, were used as received. The half-area that was etched on the transparent conductive glass (F-doped tin oxide (FTO)) was 7 Ω/square and the transmittance was 80%. 2,2′,7,7′-tetrakis (N,N-di-p-methoxyphenylamine)-9,9′-spirobifluorene (spiroOMeTAD) (≥ 99.0%, Aldrich), titanium diisopropoxide bis (acetylacetonate) (75 wt% in isopropanol), chlorobenzene (99.8%, Aldrich), lithium bis(trifluoromethylsulphonyl) imide (Li-TFSI, 95%), and 4-tert-butylpyridine (TBP, 96%) were used as received without any further purification.

### 2.2. Preparation of AMQDs

AMQDs were exfoliated from bulk antimony crystals by using a simple probe ultrasonic procedure combined with an ultrasound bath treatment (liquid exfoliation technique) [65]. A probe ultrasonic procedure uses the ultrasonic cell grinder to produce the dispersion effect of ultrasonic waves in the liquid, causing the liquid to cavitation, thereby breaking the solid particles in the liquid. Briefly, bulk Sb (50 mg) was added to NMP (50 mL), sealed in a conical plastic tube (80 mL), and ultrasonicated under a power of 800 W for 12 h. The temperature of the probe ultrasonic processing was controlled by ice and kept below 25 °C to avoid possible oxidation or degradation during exfoliation. The ultrasound probe worked for 2 s and then was stopped for 2 s. Then, the dispersion was centrifuged at a controlled speed of 7000 rpm for 20 min to remove the residual bulk Sb. The obtained liquid was centrifuged again for 30 min at 12,000 rpm to collect the supernatant containing the AMQDs. Finally, the top half of the supernatant of the AMQDs solution was centrifuged at 15,000 rpm for another 30 min to achieve the re-suspension in NMP.

### 2.3. Characterizations

X-ray diffraction (XRD) measurements were performed using a D8 Bruker diffractometer with Cu Kα (λ = 0.154 nm) radiation. Scanning electron microscopy (SEM) images were obtained on a JEOL JSM-7800F microscope. Transmission electron microscopy (TEM) images were obtained using an FEI Tecnai G2 F30, operated at 300 kV. Atomic force microscopy (AFM) was applied to characterize the morphology of the AMQDs on mica slice substrates using the MultiMode 8 at. force microscope (Bruker, USA) in air. Raman spectra were obtained on an InVia Reflex (Renishaw, United Kingdom) instrument with a 514 nm laser as the excitation source at room temperature. Photoluminescence (PL) spectra were obtained with an excitation wavelength of 325 nm. X-ray photoelectron spectroscopy (XPS, Escalab 250Xi) and ultraviolet photoelectron spectroscopy (UPS, Escalab 250Xi) measurements were used to determine the chemical compositions of the samples. The UV–Vis–NIR absorption spectra of the AMQDs solutions were measured with a UV-2550 (Shimadzu, Japan) UV–Vis–NIR spectrophotometer using quartz cuvettes at room temperature. The time-resolved PL was determined using a high-resolution monochromator and a hybrid photomultiplier detector assembly (Ultrafast systems).

### 2.4. Fabrication of QDSSCs

The FTO glass (1 × 1.2 cm) substrates were cleaned through sequential 30 min ultrasonication in detergent, ethanol, acetone, deionized water, and isopropyl alcohol, followed by treatment in a UV-$O_3$ chamber for 15 min to remove the organic components. Then, a compact layer of $TiO_2$ (~ 60 nm-thick, C-$TiO_2$) film was deposited through

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