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Weitao Lian

University of Science and Technology of China

Chen-Hui Jiang

University of Science and Technology of China

Yiwei Yin

University of Science and Technology of China

Rongfeng Tang

University of Science and Technology of China

Gang Li

University of Science and Technology of China

Lijian Zhang

University of Science and Technology of China

Bo Che

University of Science and Technology of China

Tao Chen (✉ tchenmse@ustc.edu.cn)

University of Science and Technology of China <https://orcid.org/0000-0002-3483-8341>

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**Experimental disclosing the composition- and
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antimony trisulfide materials**

**Weitao Lian¹, Chenhui Jiang¹, Yiwei Yin¹, Rongfeng Tang¹, Gang Li¹, Lijian
Zhang¹, Bo Che¹ and Tao Chen^{1*}**

¹Hefei National Laboratory for Physical Sciences at Microscale, CAS Key
Laboratory of Materials for Energy Conversion, Department of Materials Science
and Engineering, School of Chemistry and Materials Science, University of Science
and Technology of China, Hefei, Anhui 230026, P. R. China

*E-mail: tchenmse@ustc.edu.cn

23 Antimony trisulfide (Sb_2S_3) represents a kind of emerging light-harvesting material
24 with excellent stability and abundant elemental storage. Due to the low-symmetry,
25 theoretical investigation has pointed out that there exists complicated defect properties.
26 However, there is no experimental verification on the defect property. Here, we conduct
27 optical deep-level transient spectroscopy to investigate defect properties in Sb_2S_3 and
28 show that there are maximum three kinds of deep level defects observed, depending on
29 the composition of Sb_2S_3 . We also find that the Sb-interstitial (Sb_i) defect does not show
30 critical influence on the carrier lifetime, indicating the high tolerance of the one-
31 dimensional crystal structure where the space of $(\text{Sb}_4\text{S}_6)_n$ ribbons is able to
32 accommodate impurities to certain extent. This work provides basic understanding on
33 the defect properties of quasi-one-dimensional materials and a guidance for the
34 efficiency improvement of Sb_2S_3 solar cells.

Point defect engineering of semiconducting materials is essential for photovoltaic devices, comprehensive understanding of the defects formation mechanism and function enables achieving high-efficiency solar energy conversion. In specific, the point defect (intrinsic or extrinsic defect) determines the Fermi level (E_F), free carrier concentration and conductivity type (n or p) of semiconductors. Shallow-level defects with thermal energy of about $k_B T$ (where k_B is Boltzmann constant, T is temperature) from conduction band minimum (CBM) or valence band maximum (VBM) plays the major role to tune the carrier concentrations and conductivity type. In contrast, deep-level defects whose activation energy is much higher than $k_B T$ from CBM or VBM is detrimental to photogenerated carrier lifetime and transport (carrier mobility and diffusion length).¹ This defect results in the trap-assisted Shockley-Read-Hall (SRH) recombination^{2, 3} (dominant non-radiation recombination) in solar cells, which is the primary cause of open-circuit voltage (V_{OC}) loss.

Recently, binary antimony chalcogenides $Sb_2S(e)_3$ (including Sb_2S_3 , Sb_2Se_3 , $Sb_2(S,Se)_3$) have drawn increasing attention in solar cell applications, for their large visible light absorption coefficient ($> 10^5 \text{ cm}^{-1}$), earth-abundant compositional elements and tunable band gap in 1.1-1.7 eV. The efficiency breakthrough towards 10% in alloy-type antimony selenosulfide stimulating new interests in the development of this class of materials^{4, 5}. In particular, Sb_2S_3 with band gap of ~ 1.7 eV can be perfectly applied as top cell material for the construction of tandem solar cells. Different from previous photovoltaic materials such as silicon, $Cu(In, Ga)Se_2$, $CdTe$ and organic-inorganic hybrid perovskite, the Sb_2S_3 displays quasi-one-dimensional structure composed of

[Sb₄S₆]_n ribbons and could generate benign grain boundaries without dangling bonds along *c* axis (Fig. 1)^{6,7}. In practical, to transfer the distinctive structural advantage into efficient carrier transport and final power conversion efficiency (PCE), one must understand and control the defect properties, in particular the deep-level defects. This requirement has spurred intense interests in exploring the defect properties of such materials, especially from theoretical perspective, which revealed complicated defect characteristics although it has only two kinds of elements⁸⁻¹¹. However, to date there is a lack of experimental verification of defect properties associated with the structure and compositions, which induces ambiguity in the fabrication of high quality Sb₂S₃ films for efficient solar cells.

Herein, we experimentally uncover the defect characteristics by using deep level transient spectroscopy (DLTS) and identify the details of the defect. We examine both Sb-rich and S-rich Sb₂S₃ films for a comparative study which is prepared by thermal evaporation approach. To make clear conclusions, we carefully analyze the crystallinity, electronic structure and chemical composition (impurity) of the as-obtained films. In contrast to the theoretical study where complicated defect proposed in the Sb₂S₃⁸⁻¹¹, our experimental investigation shows only a few types of defect, and the defect type and concentration is sensitively dependent on anion/cation ratio.

Results

Structure and composition characterization of Sb₂S₃ films. Due to the high saturated vapor pressure of sulfur, the sulfur loss is inevitable and results in Sb-rich films during thermal evaporation deposition. To obtain S-rich Sb₂S₃ film, we apply a co-evaporation

equipment for the Sb_2S_3 film fabrication, in which S powders are co-evaporated with Sb_2S_3 for the generation S-rich Sb_2S_3 films. The synthesis details are provided in the Methods section. The as-obtained films at Sb-rich and S-rich conditions display similar morphology (Fig. 2a, 2b and Supplementary Fig. 1). X-ray diffraction (XRD) is then applied to characterize the crystallinity (Fig. 2c). Both of the films display diffractions at 15.6, 17.5, 24.9, 33.6 and 35.6 °, which are well assigned as (020), (120), (130), (330) and (240) crystal planes of orthorhombic Sb_2S_3 (JCPDS No. 42-1393, *Pbnm*)⁶. According to the energy dispersive X-ray spectroscopy (EDS), the atomic ratio of S/Sb in Sb-rich and S-rich Sb_2S_3 films are calculated to be 1.28 to 1.55 (Supplementary Table 1), respectively.

Impurity crucially influences the defect properties in a semiconductor. In this case, we carry out Raman scattering and X-ray photoelectron spectroscopy (XPS) characterizations to carefully examine the bulk and surface chemical compositions. Raman spectra show typical antisymmetric and symmetric bending vibration of S-Sb-S at 189 and 239 cm^{-1} (Fig. 2d), respectively. The antisymmetric and symmetric stretching vibration of Sb-S appear at 282 and 309 cm^{-1} . Furthermore, Raman shift at 126 and 156 cm^{-1} correspond to crystalline Sb_2S_3 phase¹². In addition, Raman scattering at 302 cm^{-1} is ascribed to the phonon scattering from CdS substrate¹³.

XPS spectra (Fig. 2e and f) display typical binding energy of 539.2 eV and 529.8 eV which are assigned to Sb 3d_{3/2} and 3d_{5/2} of Sb_2S_3 , respectively. Notably, there is no existence of SbO_x in both films, which is commonly generated as impurity in the synthesis of Sb_2S_3 and displays deep defects characteristics¹⁴. In addition, the binding

energy of 163.1 eV and 161.9 eV are ascribed to the S 2p_{1/2} and 2p_{3/2} of Sb₂S₃^{6, 14, 15}.

From both Raman scattering and XPS analysis, both Sb-rich and S-rich Sb₂S₃ films prepared via thermal evaporation are free of impurities, which thus provides excellent platform for the investigations of the defect properties.

Photovoltaic performance of devices. To test the optoelectronic quality of the as-synthesized film at device level, we examine the solar cell efficiency by using CdS and Spiro-OMeTAD as electron and hole transport layer (ETL & HTL), respectively (Fig. 3a and b). The current density-voltage (*J-V*) curves are obtained under standard AM 1.5G illumination (Fig. 3c). The optimal Sb-rich Sb₂S₃ based solar cell delivers PCE of 5.0%, with open-circuit voltage (*V*_{OC}), short-circuit current density (*J*_{SC}) and fill factor (*FF*) of 0.68 V, 15.4 mA cm⁻² and 47.6%, respectively. While the optimal S-rich Sb₂S₃ device generate a PCE of 6.2% (*V*_{OC} of 0.72 V, *J*_{SC} of 15.9 mA cm⁻² and *FF* 54.3%) with a net increase by 1.2% than the Sb-rich Sb₂S₃ based device. This PCE represents the highest value in thermal evaporation derived Sb₂S₃ solar cells^{16, 17}. The statistic parameters (*V*_{OC}, *J*_{SC}, *FF*, and PCE) of each type of devices show narrow distributions (Supplementary Fig. 2a-e), confirming the reliability of the film and device fabrication. Moreover, the external quantum efficiency (EQE) spectra (Fig. 3d and Supplementary Fig. 2f) display excellent photocurrent generation efficiency in the 500-700 nm, a decrease in the short wavelength is attributed to the light absorption loss induced by CdS since its band gap is 2.4 eV. On the ground of both *J-V* and EQE analysis, the as-prepared S-rich Sb₂S₃ films display high photovoltaic quality.

Bulk deep-level defects analysis. In this characterization, there is a typical concern on

traditional electric DLTS regarding the distortion of minority-carrier trap detection¹⁸. Whereas attaching optical pulse to DLTS is recognized as an effective means to improve the sensitivity and authenticity of minority-carrier traps detection^{19,20}. Herein, we adopt electric and optical double pulse mode, and both pulses are exerted simultaneously and share the same pulse width. In order to avoid the interference of electrons transport layer (ETL) and holes transport layer (HTL), a 635 nm-wavelength red laser is selected as the excited source of optical pulse. Moreover, the reverse bias is set at -0.3 V, while various pulse voltage (0.1~0.5 V) is applied to avoid the fault peaks caused by the capacitance bridge recovery delay at inappropriate pulse voltage sometimes. Meanwhile, the information of defects at various depths can also be probed by varying the pulse voltage.

Fig. 4a and b show the DLTS spectra of Sb-rich and S-rich Sb₂S₃ devices at different pulse voltages. The positive peaks in the DLTS spectra represent majority-carrier traps, while the negative peaks indicate minority-carrier traps¹⁸. Regarding n-type Sb₂S₃ (Supplementary note 1), the majority and minority-carrier traps are corresponding to electron and hole traps, respectively. Eventually, the statistical defects information of the two kinds of devices can be calculated and the results are summarized in Table 1, where E_T , σ , N_T , τ and N_S are trap energy level, capture cross section, trap density, carrier lifetime and shallow donor concentration (Supplementary note 2), respectively. It turns out that the Sb-rich film displays three electron traps, E1, E2 and E3 (donor defects), with the energy level of 0.31, 0.60 and 0.69 eV below the CBM (Fig. 4a and Fig. 5a). In contrast, the S-rich film exhibits two hole traps which are denoted as H1

and H2 (acceptor defects) with energy levels of 0.64 and 0.71 eV above the VBM (Fig. 4b and Fig. 5b). In comparison to the calculations on the defect energy levels^{8, 9, 21}, the traps E1, E2 and E3 are well assigned to Sb-interstitial (Sb_i), S-vacancy (V_S), and antisite of Sb for S (Sb_S) defects, respectively. The traps H1 and H2 in S-rich Sb_2S_3 are ascribed to Sb-vacancy (V_Sb) and antisite of S for Sb (S_Sb), respectively. This characterization suggests clearly the composition-dependent defect properties of Sb_2S_3 films.

Notably, according the energy level alignment (Fig. 5a and b), these are all deep-level defects whose energy level are more than 0.3 eV far from CBM or VBM. Therefore, they are supposed to act as carrier traps or recombination sites due to their high ionization energy. Specially, the trap E2, E3, H1 and H2 possess deeper energy level (close to intrinsic E_F), large capture cross section and high trap density (Table 1) which accord with basic characteristics of SRH recombination centers. Thus, they behave seriously as recombination centers to hinder carrier transport and shorten the carrier lifetime.

Carrier dynamic analysis. To study the charge transport kinetics, we perform transient absorption spectroscopy (TAS) measurement of the Sb-rich and S-rich Sb_2S_3 films deposited on soda-lime glass. We apply glass/ Sb_2S_3 films without any electron or hole extraction layer as sample for this study, which can reflect unambiguously that the exciton relaxes to ground state through charge recombination in films. The TAS is tracked at a time window of 5 -5000 ps using 400 nm pulse laser excitation for Sb-rich and S-rich Sb_2S_3 films (Fig. 6a and b, Supplementary Fig. 5). Strikingly, there appear

two photo-induced absorption peaks near 545 and 690 nm, both of which are assigned to a single broad feature of trapped carrier in Sb₂S₃. Especially, the peak at 690 nm is associated with the formation of sulfide radical^{22, 23}. Thus, the enhanced absorption at 690 nm for S-rich Sb₂S₃ could be attributed to abundant sulfide radical. Accordingly, the decay characteristics at 545 nm are fitted by biexponential model to study carrier decay kinetics (Fig. 6c and Supplementary Table 2). It is observed that the S-rich Sb₂S₃ exhibits much longer carrier lifetime (18.7 ns) than Sb-rich Sb₂S₃ (3.8 ns), the reduced defect type and concentration contribute the prolonged lifetime.

To gain further insight into the specific influence of each defect on the carrier dynamics, we extract the limited carrier lifetime associated with specific defect by trap-assisted SRH recombination according to equation 1.

$$\tau = (\nu_{th}\sigma N_T)^{-1}. \quad (1)$$

where the ν_{th} is thermal velocity for electron (hole) that is about 10^7 cm/s in bulk semiconductors at room temperature^{2, 3, 24}. Both σ and N_T can be obtained from DLTS. Consequently, the defect with large capture cross section, high trap density and deep trap energy level are much more detrimental to carrier lifetime.

In Sb-rich Sb₂S₃ film, the carrier lifetime is limited by trap E2 (V_S) and E3 (Sb_S) in Sb-rich Sb₂S₃ film which are estimated to be 64.6 and 28.4 ns (Table 1), respectively. The other defect E1 (Sb_i) present carrier lifetime of 2180 ns, much longer than those of E2 and E3. This result should be associated with the crystal structure where the Sb atoms enters into the space between (Sb₄S₆)_n ribbons is less detrimental to the carrier transport dynamics. In S-rich Sb₂S₃ film, the trap H1 (V_{Sb}) sets the limited lifetime of

48.3 ns, while the lifetime limited by trap H2 (S_{sb}) is 997 ns. Defect S_{sb} shows less detriment to carrier lifetime since the low concentration due to high formation energy⁹, even though it manifests deeper energy level than V_{sb} .

In the first sense, we compare the shortest carrier lifetime limited by E3 and H1, we suppose that this prolonged carrier lifetime is attributed to reduced capture cross section and trap density of H1. In fact, the recombination form in real semiconductors is mostly multi-level recombination²⁴. That is to say, the carriers leap among multiple trap level and get recombination ultimately. Therefore, the decreased defect numbers in S-rich Sb_2S_3 also contribute the longer carrier lifetime. It is worthy of noting that the calculated carrier lifetime estimated based on bulk trap-assisted SRH recombination model should be slightly exaggerated since it does not consider other non-radiation recombination such as Auger recombination and interface recombination. In any case, the carrier lifetime calculated by bulk trap-assisted SRH recombination model shows the similar trend at the same scale when compared with that obtained by TAS, suggesting that it is the intrinsic deep-level defects that restrict the carrier lifetime in Sb_2S_3 .

Discussion

For the Sb_2S_3 solar cell development, one of the major concern is the efficiency improvement. The rational engineering of the defect properties benefits the carrier transport and suppress the negative recombination, which further improve three device parameters, V_{OC} , J_{SC} and FF. In particular, it has been acknowledged that there is a serious V_{OC} loss in the Sb_2S_3 solar cell. Fundamentally, the V_{OC} of solar cell originates from the split of quasi-Fermi level for electron and hole (Fig. 5c)^{24, 25}. The trap E2 and

E3 in Sb-rich Sb_2S_3 films with large capture cross section and high trap density, especially trap energy level closer to Fermi level compared with H1 and H2 in S-rich Sb_2S_3 . There is high possibility that the electron quasi-Fermi level is pinned near trap E2 and E3 in Sb-rich Sb_2S_3 owing to inefficient extraction of trapped photo-excited carriers. In contrast, S-rich Sb_2S_3 displays decrescent capture cross section, defects density and defects numbers which jointly give rise to suppressive recombination and prolonged carrier lifetime. This characteristic is able to alleviate the Fermi level pinning effects and improving V_{OC} ultimately.

An interesting finding in this study is that the existence of Sb_i in the Sb-rich Sb_2S_3 film generates less detrimental effect on the carrier lifetime, which should be related to Q1D crystal structure where the space between $(\text{Sb}_4\text{S}_6)_n$ ribbons can afford impurities to certain degree. However, the S_i defect does not appear in the S-rich Sb_2S_3 film, it is most likely that the sulfur is easy to be evaporate out during the film deposition at high temperature. Finally, we find that the Sb-rich film displays two types of crucial defect, i.e. V_{S} and Sb_{S} , while S-rich Sb_2S_3 film shows only one kind of critical defect, V_{Sb} . Therefore, the S-rich Sb_2S_3 film seems more promising for achieving next efficiency breakthrough, provided that the V_{Sb} is well suppressed without introducing other deep-level defects.

Methods

Preparation of Sb_2S_3 thin films. The Sb_2S_3 films were deposited on FTO/CdS substrate preheated at 300 °C via thermal evaporation under pressure of 0.2~0.3 Pa. About 0.3 g Sb_2S_3 powder (99.9%, aladdin) was put into a tungsten boat equipped on

the DC evaporator source, and the evaporating temperature was tuned through the DC current. As for co-evaporation, another 0.1 g S powder (99.999%, Sinopharm group) was needed to evaporate simultaneously with Sb_2S_3 powder. The films were deposited at a rate of $3\text{--}5\text{ nm s}^{-1}$. The final thickness of films was controlled around 200 nm via a film thickness gauge. Finally, the as-deposited films were post-annealed at $350\text{ }^\circ\text{C}$ for 2 min on a preheated hot plate in a N_2 -filled glove box.

Fabrication of Sb_2S_3 solar cells. The FTO glass (TEC-A7) was cleaned by DI water, isopropanol, acetone, and ethanol sequentially. Then FTO substrate was cleaned for 15 min by UV ozone prior to use as well. Next, the FTO substrate coated with 60 nm CdS film as ETL by using chemical bath deposition (CBD) method⁴. After treatment with CdCl_2 (20 mg mL^{-1} methanol), then FTO/CdS substrate was heated at $400\text{ }^\circ\text{C}$ for 10 min in open air. Subsequently, the Sb_2S_3 film was deposited on FTO/CdS substrate by thermal evaporation mentioned above. Spiro-OMeTAD was utilized as HTL according to our previous report⁵. Finally, the Au back electrode was evaporated on the HTL under a pressure of $5 \times 10^{-4}\text{ Pa}$. The active area was defined as 0.12 cm^2 .

Films characterizations. The morphologies of Sb_2S_3 thin films were characterized by Zeiss G450 SEM equipped with an EDS (Bruker) module. The crystal structure was measured by XRD (Bruker Advance D8 diffractometer) with $\text{Cu K}\alpha$ radiation ($\lambda = 1.5406\text{ \AA}$). Raman spectroscopy (Horiba JobinYvon, LabRAM HR800) was applied to analyze chemical bonds in Sb_2S_3 with 532 nm laser excitation. XPS (Thermo Fisher) with a Monochrome Al $\text{K}\alpha$ (1486.6 eV , 15 kV) was used to characterize the surface composition. The work function and valence band binding energy were measured by

UPS (Thermo Escalab 250Xi, He I excitation 21.22 eV) which were recorded at 0 V samples bias in an ultrahigh vacuum chamber. TAS of Sb₂S₃ films were measured by a Helios setup, where a nondegenerate pump–probe configuration was applied to probe the transient dynamics (50 fs to 7 ns). Additionally, the pump and probe laser pulses were generated by frequency doubling the fundamental output (Coherent Vitesse, 80 MHz, Ti-sapphire laser) and white light generated with a sapphire plate, respectively. The decay characteristics were fitted by biexponential model $y = \sum A_i \exp(-x/t_i)$, while carrier lifetime (τ) was obtained by $\tau = \sum A_i t_i^2 / \sum A_i t_i$ ($i=2$).

Devices characterizations. The J - V characteristics were performed by Keithley 2400 apparatus under solar-simulated AM 1.5 sunlight (100 mW cm⁻²) with a standard xenon-lamp-based solar simulator (Oriel Sol 3A, Japan). Prior to test, the solar simulator illumination intensity was calibrated by a monocrystalline silicon reference cell (Oriel P/N 91150 V, with KG-5 visible color filter) which is also calibrated by the National Renewable Energy Laboratory (NREL). The EQE (model SPIEQ200) was measured using a single-source illumination system (halogen lamp) combined with a monochromator. The DLTS measurement was performed via a Phystech FT-1230 HERA-DLTS system equipped with a 10 mW laser of 635 nm wavelength. And the optical pulse was generated from the laser. The modified Boonton 7200 capacitance meter (high frequency, 1 MHz) was used to observe capacitance dynamically. The samples were placed in a liquid-helium cryostat (Lakeshore 335,336). The DLTS temperature scan range varies from 120 to 420 K at 2 K intervals. The pulse mode was set as electrical (fill voltage) and optical (laser excitation) pulse yielding simultaneously.

In detail, the reverse bias, pulse voltage, pulse width (electric & optical), and period width were −0.3 V, 0.1~0.5 V, 10 ms and 100 ms, respectively.

Data available

All data that support the findings of this study are available within the paper and the supplementary information.

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340 Author contributions

341 T. C. supervised the project, W. L. and T. C. conceived the original concept and
342 designed the experiments. W. L. fabricated the devices and conducted the photovoltaic
343 characterizations and DLTS measurement analysis, B. C. assisted with the DLTS

344 measurement and analysis. C. J. aid in TAS measurement. Y. Y. and R. T. helped the
345 fabrication of films and deives. G. L., L. Z. and C. Z. assisted with the electrochemistry
346 characterizations. W.L. and T.C. co-wrote the manuscript, all authors revised the
347 manuscript.

348 **Competing interests**

349 The authors declare no competing interests.

Fig. 1 Schematic diagram of quasi-1-dimensional structural Sb_2S_3 . Sideview (a) and aeroview (b) of $[\text{Sb}_4\text{S}_6]$ ribbons along c axis.

Fig. 2 Characterizations of Sb_2S_3 film. **a, b**, Surface SEM images of Sb-rich and S-rich Sb_2S_3 films. **c, d**, XRD patterns and Raman spectroscopy of Sb-rich and S-rich Sb_2S_3 films. **e, f**, Sb 3d and S 2p XPS spectra of Sb-rich and S-rich Sb_2S_3 .

Fig. 3 Device structure and photovoltaic performance. **a, b**, Cross sectional SEM image and corresponding device configuration of Sb₂S₃ solar cells. **c** $J-V$ characteristics of the optimal Sb-rich and S-rich Sb₂S₃ devices under standard one Sun illumination. **d** EQE spectra of the optimal Sb-rich and S-rich Sb₂S₃-based devices.

Fig. 4 Deep-level defects characterization. DLTS signals of Sb-rich (a) and S-rich Sb_2S_3 films (b) at pulse voltage ranging from 0.1 to 0.5 V, synergized with an identical pulse-width optical pulse.

Fig. 5 Schematic of band structure and heterojunction. a, b, Conduction band (E_C), valence band (E_V), Fermi level (E_F) and trap energy level (E_T) for Sb-rich and S-rich Sb_2S_3 films. c Schematic diagram of V_{OC} derived from heterojunction.

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378 Fig. 6 TAS study of Sb_2S_3 films. **a, b**, 2D color images of TAS of Sb-rich and S-rich
379 Sb_2S_3 films. **c** Transient kinetic decay and fittings (solid lines) monitored at 545 nm of
380 Sb-rich and S-rich films.

381

382 Table 1 Deep-level defect parameters (trap type, trap energy level (E_T), capture cross
383 section (σ), trap density (N_T), carrier lifetime (τ), shallow donor concentration (N_S)) of
384 Sb-rich and S-rich Sb_2S_3 films.

Sample	Trap	E_T (eV)	σ (cm^2)	N_T (cm^{-3})	τ (ns)	N_S (cm^{-3})
Sb-rich	E1	$E_c-0.31\pm0.02$	$(0.54\sim8.13)\times10^{-17}$	$(3.75\sim5.63)\times10^{14}$	2.18×10^3	7.71×10^{16}
	E2	$E_c-0.60\pm0.02$	$(0.26\sim4.68)\times10^{-16}$	$(1.57\sim3.31)\times10^{15}$	6.46×10^1	
	E3	$E_c-0.69\pm0.02$	$(0.11\sim1.75)\times10^{-15}$	$(1.38\sim2.01)\times10^{15}$	2.84×10^1	
S-rich	H1	$E_v+0.64\pm0.01$	$(0.46\sim1.31)\times10^{-15}$	$(0.45\sim1.58)\times10^{15}$	4.83×10^1	5.13×10^{16}
	H2	$E_v+0.71\pm0.02$	$(0.49\sim1.17)\times10^{-16}$	$(6.71\sim8.57)\times10^{14}$	9.97×10^2	

385

386

Figures

Figure 1

Schematic diagram of quasi-1-dimensional structural Sb₂S₃. Sideview (a) and aeroview (b) of [Sb₄S₆] ribbons along c axis.

Figure 2

Characterizations of Sb₂S₃ film. a, b, Surface SEM images of Sb-rich and S-rich Sb₂S₃ films. c, d, XRD patterns and Raman spectroscopy of Sb-rich and S-rich Sb₂S₃ films. e, f, Sb 3d and S 2p XPS spectra of Sb-rich and S-rich Sb₂S₃.

Figure 3

Device structure and photovoltaic performance. a, b, Cross sectional SEM image and corresponding device configuration of Sb₂S₃ solar cells. c J–V characteristics of the optimal Sb-rich and S-rich Sb₂S₃ devices under standard one Sun illumination. d EQE spectra of the optimal Sb-rich and S-rich Sb₂S₃-based devices.

Figure 4

Deep-level defects characterization. DLTS signals of Sb-rich (a) and S-rich Sb₂S₃ films (b) at pulse voltage ranging from 0.1 to 0.5 V, synergized with an identical pulse width optical pulse.

Figure 5

Schematic of band structure and heterojunction. a, b, Conduction band (EC), valence band (EV), Fermi level (EF) and trap energy level (ET) for Sb-rich and S-rich Sb₂S₃ films. c Schematic diagram of V_{OC} derived from heterojunction.

Figure 6

TAS study of Sb₂S₃ films. a, b, 2D color images of TAS of Sb-rich and S-rich Sb₂S₃ films. c Transient kinetic decay and fittings (solid lines) monitored at 545 nm of Sb-rich and S-rich films.

Supplementary Files

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