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Abstract: Wide-bandgap all-inorganic CsPbIBr$_2$ solar cells with MoO$_x$/Ag/TeO$_2$ composite transparent electrode have been firstly used to construct a 4T perovskite/Si-heterojunction tandem cells. The calculation results showed that the long wavelength transmission (600 to 1100 nm) of MoO$_x$/Ag film can be enhanced remarkably by adding a TeO$_2$ optical capping layer. This resulted in an obviously increase of the PCE ($J_{SC}$) from 10.70% (17.56 mA/cm$^2$) to 14.76% (24.67 mA/cm$^2$) for corresponding bottom Si cell in CsPbIBr$_2$/Si devices, although the PCE of top CsPbIBr$_2$ cell decreased from 17.52% to 16.58%, the overall PCE of tandem device has increased from 28.22% to 31.34%. This $J_{SC}$-dependent PCE improvement mainly came from the more balanced light absorption, which can be understood by the optical interference induced light field redistributions in CsPbIBr$_2$/Si devices. Specially, comparing with the original Si cell, the $J_{SC}$ and PCE losses in bottom Si cell have been suppressed to about 33% and 32%, which are superior to most the reported pervoskite/Si tandem cells. Therefore, the tandem of Si cell and CsPbIBr$_2$ cell with optimized MoO$_x$(20 nm)/Ag(8 nm)/TeO$_2$(50 nm) transparent electrode could be the promising cost-effective photovoltaic in the future. This work is instructive to the fabrication of perovskite/Si tandem solar cells.

Index Terms: Wide-bandgap, CsPbIBr$_2$, MoO$_x$/Ag/TeO$_2$, four-terminal, perovskite/Si tandem solar cells.
1. Introduction

Metal halide perovskite solar cells have attracted great interest due to the advantages of low cost, simple process, adjustable absorption range, flexibility, and high conversion efficiency (PCE) of 25.5% (certified) [1]–[5]. Particularly, as the PCE of crystal silicon (Si) solar cells is closing to the commercial limits [6], [7], the perovskite/Si tandem devices have been verified as a promising cost-effective strategy to break the Shockley-Queisser (SQ) efficiency limit of Si and realize the 30%-efficiency photovoltaic applications [8]–[12]. Recently, the record PCE of two-terminal (2T) perovskite/Si tandem cells has reached 29.15% [13] and the four terminal devices also achieved an efficiency of 27.7% [14]. However, due to the limits of current match in 2T devices and spectrum match in 4T devices, the short circuit current density \( J_{SC} \) of bottom Si cell mostly was about 14 to 19 mA/cm\(^2\), which is much lower than that of the original Si cells (about 40 mA/cm\(^2\)), thus half of their output power would be lost (Table S2). In fact, it has been predicted in the 4T Si tandem devices that, this deficiency can be improved by employing top cells with larger bandgap width [15], which can be attributed to the more balanced light absorption in sub cells. Simultaneously, to obtain the similar overall PCEs, the PCE requirement of wide-bandgap top cell could be obviously reduced. These have been further confirmed by experimental perovskite/Si tandem cells from the reports [16], [17].

Nowadays, most researchers focused on the organic-inorganic mixed perovskite top cells, which possess higher PCE, narrower bandgap, and poorer intrinsic stability [18]. In fact, by replacing organic ions with inorganic metal cations, the all-inorganic perovskite cells (CsPbI\(_{x}\)Br\(_{3-x}\)) not only have wider bandgap (>2 eV) but also better stability, as well the record PCEs of 18% [19], [20], [21]. What is more, the theoretical efficiency of 4T wide-bandgap perovskite/Si tandem cells could exceed 40% [22], as the sub cells can be fabricated independently, the issues of current match and process compatibility in 2T structure could be addressed. Consequently, the 4T wide-bandgap perovskite/Si tandem cells have been another important options for efficient tandem photovoltaic. Among the typical CsPbI\(_{x}\)Br\(_{3-x}\) devices, the CsPbBr\(_2\) (~2.05 eV) cells owned the balance between PCE and stability [23], thus a candidate top cell in the 4T perovskite/Si tandem cells.

In this work, the semitransparent CsPbBr\(_2\) solar cells with ultrathin Ag film electrode has been firstly used to construct a 4T perovskite/Si heterojunction (SHJ) tandem cells, their optical and electrical performance had been systemically investigated by the Silvaco Atlas device simulator. The ultrathin Ag film electrodes with an optical capping layer were successfully utilized in our previous semitransparent or bifacial perovskite solar cells [24], [25], [26], [27], [28]. For the 4T tandem applications, the optical transmission of Ag electrode at long wavelength is strongly related to the light absorption of bottom Si cell and also the overall PCE of tandem devices. Here the oxide/metal/oxide [16], [29], [30], [31] composite electrodes were employed to carefully modulate the transmission of Ag film and the light field distributions in the CsPbBr\(_2\)/Si tandem devices. It has been found that, when the optimized MoO\(_x\)(20 nm)/Ag(8 nm)/TeO\(_2\)(50 nm) electrode was introduced, the PCE of tandem device reached 31.34% with a 16.58%-top CsPbBr\(_2\) cell and 14.76%-bottom Si cell. Specially, comparing with the original Si cell, the \( J_{SC} \) and PCE losses of bottom Si cell have been suppressed to about 33% and 32%, which is superior to most the reported perovskite/Si tandem cells. Therefore, the 4T wide-bandgap CsPbBr\(_2\)/SHJ tandem devices could be the promising cost-effective tandem photovoltaic in the future.

2. Methods

The optical and electrical calculations were performed by the Silvaco Atlas device simulator. The 4T tandem solar cell consists of a wide bandgap, semitransparent, inorganic CsPbBr\(_2\) top cell and a commercial heterojunction bottom Si cell. In the CsPbBr\(_2\) top cell, the typical TiO\(_2\)(20 nm) was selected as electron transport layer, the MoO\(_x\)(20 nm) acted as hole transport layer, the ultrathin Ag film (8 nm) was chosen as the transparent electrode, and the outer optical capping layer employed the TeO\(_2\) or MoO\(_x\) with high refractive index to modulate the transmission of Ag electrode. The bottom Si cell includes a ITO transparent conductive layer (10 nm), p-type amorphous Si (a-Si,
20 nm), intrinsic a-Si (15 nm), crystal Si (c-Si, 250 μm), n-type a-Si (20 nm) layer, and a Ag rear electrode (180 nm). Particularly, a refractive match layer [14] with a refractive index about 1.4 was introduced to reduce the light loss at the interface between sub cells. The performance of semitransparent CsPbIBr\(_2\) top cell and overall PCEs of tandem devices have been carefully optimized. For the optical calculation, the inner optical field distribution and carrier generation rate have been calculated by the transfer matrix method (TMM) [32]. While the Poisson’s equation, carrier continuity equation, and drift-diffusion equations were adopted to simulate the electrical characteristics. And the Shockley–Read–Hall and Auger recombination mechanisms have been considered for the CsPbIBr\(_2\)/Si tandem devices. The current density voltage (JV) curves were estimated under the AM 1.5G solar spectrum. All the optical constants of the each layer were derived from the references [28], [33]–[35], the main electrical parameters had been listed in the supporting information, and the calculation method can be found in our previous works. [24], [28], [36]

3. Results and Discussion

Fig. 1(a) showed the 4T tandem structure calculated in this work. It is known that the PCE and long wavelength transmission of CsPbIBr\(_2\) top cell are critical to improve the PCE of bottom Si cell and overall performance of tandem devices. Thus in the typical TiO\(_2\)/CsPbIBr\(_2\)/MoO\(_x\) structure, the transmission and conductivity of both front and rear electrodes determine the performance of CsPbIBr\(_2\) top cell. Usually the front electrode employed the commercial ITO or FTO coated glass substrate, while the rear electrode should be deposited at the last step of device fabrication. It has been proved that the ultrathin Ag film electrode is superior to oxide electrodes in electrical conductivity, especially in the large-area cells, and the obstacle for metal electrode comes from how to enhance its optical transmission. Nowadays, in most reports and our previous works, the transmission of Ag film in the range from 300 to 800 nm has been optimized to increase the PCE of perovskite or organic solar cells. But in the Si tandem devices, the transmission from 800 to 1100 nm must be further improved. Here, the TeO\(_2\) was firstly used to constitute a MoO\(_x\)/Ag/TeO\(_2\) composite transparent anode for the CsPbIBr\(_2\) top cell, and the corresponding device performed
better than the reported MoO\textsubscript{x}/Ag/MoO\textsubscript{x} electrodes, which will be discussed later. As shown in Fig. 1(b), when a 20 nm thick TeO\textsubscript{2} film was introduced, although the transmission of MoO\textsubscript{x} (20 nm)/Ag(11 nm)/TeO\textsubscript{2} film below 650 nm was decreased, its long wavelength transmission from 650 to 1100 nm was remarkably improved. As the absorption cutoff edge of CsPbIBr\textsubscript{2} material locates at around 600 nm, the triple layer anode would be the better candidates for a 4T CsPbIBr\textsubscript{2}/Si tandem devices.

According to Fig. 1(c), the calculated J\textsubscript{SC} of CsPbIBr\textsubscript{2} top cell increased with the thickness of CsPbIBr\textsubscript{2} absorber and approached saturation when its thickness beyond 600 nm, thus the 600 nm-thick CsPbIBr\textsubscript{2} was chosen to optimize the device performance. To obtain better transmission for Ag electrode, a thinner 8 nm Ag has been selected as it was the lowest electrode thickness in experimental reports [37]. Then the transmission of CsPbIBr\textsubscript{2} top cell with varied thicknesses of TeO\textsubscript{2} from 10 to 90 nm were simulated, as shown in Fig. 1(d), the transmission increased obviously from 600 nm, which is in line with the optical bandgap (~2.05 eV) [23] of CsPbIBr\textsubscript{2}. The transmission curves climbed up and then declined with the increase of TeO\textsubscript{2} thickness, which can be attributed to the optical interference at material interfaces with different refractive index. Fortunately, the average transmission of Ag electrode approached 80% at long wavelength. While the photon absorption rates of bottom Si cell in Fig. 1(e) agreed well with the results in Fig. 1(d), and about 70% photons ranging from 600 to 1100 nm could be absorbed by the bottom Si cell, which is favor to increase the power output of Si cell and improve the total PCE of 4T tandem devices.

In order to achieve better overall performance for tandem devices, the J\textsubscript{SC} of bottom Si cell and total PCE of tandem device were monitored to optimize the thickness of TeO\textsubscript{2} capping layer. It can be seen in Fig. 2 that the total PCE in all cases exceeded 30%, beyond the SQ limit of Si cell, and it can be higher than 31% when the thickness of TeO\textsubscript{2} was controlled between 10 to 60 nm; the J\textsubscript{SC} values beyond 24 mA/cm\textsuperscript{2} have been obtained for bottom Si cell with the TeO\textsubscript{2} thickness increased from 20 to 70 nm; and the optimized thickness of TeO\textsubscript{2} was about 50 nm, at which the total PCE over 31% and J\textsubscript{SC} over 24.5 mA/cm\textsuperscript{2} could be obtained. Further, the simulated JV curves under AM 1.5G solar spectrum and corresponding photovoltaic parameters were summarized in Figs. 3(a), 3(b), and Table 1, respectively. For the tandem device without TeO\textsubscript{2} capping layer, the CsPbIBr\textsubscript{2} top cell showed a PCE of 17.52% with V\textsubscript{OC} = 1.59 V, J\textsubscript{SC} = 13.24 mA/cm\textsuperscript{2}, and FF = 83.32%; while the bottom Si cell obtained a PCE of 10.7% with V\textsubscript{OC} = 0.76 V, J\textsubscript{SC} = 17.56 mA/cm\textsuperscript{2}, and FF = 80.46%; thus the total PCE of tandem device was 28.22%. When a 50 nm TeO\textsubscript{2} capping layer was introduced, the J\textsubscript{SC} of bottom Si cell increased to 24.67 mA/cm\textsuperscript{2} (by 40%) and the PCE was enhanced to 14.76% (by 38%), although the PCE of CsPbIBr\textsubscript{2} top cell decreased to 16.58%, the total PCE of tandem device has been promoted to 31.34%. It is noted that the J\textsubscript{SC} and PCE losses in bottom Si cell were the lowest in the reported 4T perovskite/Si tandem devices (Table S2). The improvement of J\textsubscript{SC} for bottom Si cell can be explained by the remarkably enhanced photon absorption rate shown in Fig. 3(c), this is in line with the higher transmission
Fig. 3. Calculated JV curves of CsPbBr₂ top cell and bottom Si cell when the TeO₂ thicknesses were 0 (a) and 50 nm (b); (c) absorption of bottom Si cell with different TeO₂ thickness; (d) comparing of top cell transmission and bottom cell absorption.

| Transparent electrode | Sub cells          | Jsc (mA/cm²) | Voc (V) | FF (%) | PCE (%) | Total PCE (%) |
|-----------------------|--------------------|--------------|---------|--------|---------|---------------|
| aMoO₃/Ag              | CsPbI₂ Br₂        | 13.24        | 1.59    | 83.32  | 17.52   | 28.22         |
|                       | Si                 | 17.56        | 0.76    | 80.46  | 10.70   |               |
| bMoO₃/Ag/MoO₃        | CsPbI₂ Br₂        | 12.53        | 1.59    | 83.75  | 16.67   | 29.36         |
|                       | Si                 | 20.97        | 0.76    | 79.48  | 12.69   |               |
| cMoO₃/Ag/TeO₂        | CsPbI₂ Br₂        | 12.44        | 1.59    | 83.87  | 16.58   | 31.34         |
|                       | Si                 | 24.67        | 0.77    | 78.25  | 14.76   |               |
| dMoO₃/Ag/TeO₂        | CsPbI₂ Br₂        | 11.99        | 1.58    | 77.36  | 14.69   | 28.96         |
|                       | Si                 | 23.79        | 0.76    | 78.54  | 14.27   |               |
| Reference Si cells    |                    | 36.83        | 0.77    | 0.859  | 24.40   |               |

*a* Semitransparent top CsPbI₂ Br₂ cells were illuminated from ITO side. *b* Semitransparent top CsPbI₂ Br₂ cells were illuminated from Ag side.

of MoO₃/Ag/TeO₂ electrode and CsPbI₂ Br₂ top cell. Further, in Fig. 3(d), the bottom Si cell could absorb most of the incident photons ranging from 600 to 1050 nm, which induced the obviously improved Jsc and PCE of Si cell.

Furthermore, the 3D optical electric filed distributions of 4T tandem device with and without 50 nm TeO₂ capping layer have been displayed in Fig. 4. When the photons entered into the CsPbI₂ Br₂ top cell from ITO side, the very low intensity of optical electric filed at 300 to 600 nm suggested the efficient absorption of CsPbI₂ Br₂ and the introducing of TeO₂ cannot enhance the performance of CsPbI₂ Br₂ top cell in this case. Simultaneously, the significantly enhanced field intensity in the wavelength range from 600 to 1100 nm was out of the active absorption range of CsPbI₂ Br₂, but increased the absorption possibility of long wavelength photon for bottom Si cell. The relatively low filed intensity at about 920 nm and 620 nm can be understood by the optical interference of incident light from ITO cathode and reflective light from Ag anode. All of these are in line with
Fig. 4. Distributions of normalized optical electric field for CsPbI$_2$Br$_2$ top cell in the tandem devices with 0 and 50 nm TeO$_2$ optical capping layer.

Fig. 5. Calculated JV curves of CsPbI$_2$Br$_2$ top cell and bottom Si cell when adopting the MoO$_x$/Ag/MoO$_x$ electrode (a) and MoO$_x$/Ag/TeO$_2$ electrode (b). It is noted that the top cell was illuminated from ITO side (a) and Ag side (b).

the results in Fig. 3 and Table 1. According the above discussions, comparing with MoO$_x$/Ag, it is suggested that the transmission improvement of MoO$_x$/Ag/TeO$_2$ anode dominated the performance enhancement of bottom Si cell and 4T tandem devices, which provides a strategy to fabricate 30% efficiency 4T perovskite/Si tandem solar cells with more stable but inefficient perovskite top cell (about 16%); markedly decreased PCE loss of bottom Si cell (<40%); and high conductivity transparent Ag electrodes (<10 $\Omega/\square$). Thus the 4T CsPbI$_2$Br$_2$/Si tandem device may be the front runner in the commercialization process of perovskite/Si tandem solar cells.

In addition, we also simulated the typical MoO$_x$/Ag/MoO$_x$ (50 nm) electrode based CsPbI$_2$Br$_2$ top cell and bottom Si cell. As shown in Fig. 5(a) and Table 1, the top cell displayed the $J_{SC}$ of 12.53 mA/cm$^2$, $V_{OC}$ of 1.59 V, FF of 83.75%, and a PCE of 16.67%; the bottom Si cell obtained
a PCE of 12.69% with the $J_{SC} = 20.97$ mA/cm$^2$, $V_{OC} = 0.761$ V, and $FF = 79.48$%; thus a total PCE of 29.36% for tandem device. Compared to the device with MoO$_x$/Ag electrode, the overall performance has been improved but was slightly lower than the device with MoO$_x$/Ag/TeO$_2$ electrode. Therefore, the TeO$_2$ should be another efficient capping layer to modulate and enhance the transmission of Ag film electrode at long wavelength, which broadens the applications of Ag film electrode in tandem solar cells. Simultaneously, the designed semitransparent CsPbIBr$_2$ top cell can absorb light from both ITO and Ag electrode sides, thus a bifacial cell. In other words, we can reverse the stacking direction of CsPbIBr$_2$ top cell and construct different tandem devices. As shown in Fig 5(b) and Table 1, when the CsPbIBr$_2$ top cell was illuminated from Ag side, a PCE reached 14.69% with $J_{SC} = 11.99$ mA/cm$^2$, $V_{OC} = 1.58$ V, and $FF = 77.36$%, and this lower PCE of top cell can be explained by the relatively lower $J_{SC}$ and poorer transmission of MoO$_x$/Ag/TeO$_2$ than that of ITO electrode at low wavelength, shown in Fig. 1(b) and previous works [24]. While the bottom Si cell also achieved a $J_{SC}$ of 23.79 mA/cm$^2$ and PCE of 14.27%, which was very close to the performance of device illuminated ITO side; and the total PCE of tandem device also reached 28.96%. Thus the bifacial CsPbIBr$_2$ semitransparent cell with MoO$_x$/Ag/TeO$_2$ composite electrodes could be the candidate top cell to fabricate efficient 4T perovskite/Si tandem devices. To further improve the performance of tandem devices and promote their application, the antireflective nano-structure [38], plasmonic effect [39], maximum power point tracker [40] should be promising strategies in the future work.

4. Conclusion

Semitransparent wide-bandgap CsPbIBr$_2$ solar cells with ultrathin Ag electrode have been firstly used to construct a 4T perovskite/Si heterojunction tandem cells, in which the top CsPbIBr$_2$ cell absorbed the light from 300 to 600 nm and the bottom Si cell harvested the photons ranging from 600 to 1100 nm. Here the MoO$_x$(20 nm)/Ag(8 nm)/TeO$_2$(50 nm) composite transparent electrode was utilized to address the drawback of poor transmission of Ag film at long wavelength, and the resulted PCE of CsPbIBr$_2$/Si tandem device reached 31.34% with a 16.58%-top CsPbIBr$_2$ cell and 14.76%-bottom Si cell. The simulation results shown that this PCE improvement mainly depended on the increased $J_{SC}$ of bottom Si cell, which was derived from the optical interference induced light field redistributions in CsPbIBr$_2$/Si devices. What interest is, comparing with the original Si cell, the $J_{SC}$ and PCE losses of in bottom Si cell have been suppressed to about 33% and 32%, which are superior to most the reported pervoskite/Si tandem cells. Therefore, the 4T wide-bandgap CsPbIBr$_2$/Si tandem devices could be the promising cost-effective photovoltaic in the future. This work is instructive to the fabrication of pervoskite/Si tandem solar cells.

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