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Development of semitransparent Perovskite Solar Cells with double electron transport layer and modified top electrode

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Abstract. In this work we report on semitransparent perovskite solar cell fabrication using double electron transport layer and double top electrode. Such devices may be used in tandem solar cells which are made to overcome Shockley-Queisser limit for one junction solar cell and improve efficiency of the device. One of the main issues of this task is to make transparent top electrode since it is usually made of metal. In this work the combination of ITO/Ag is used in which silver is deposited via special mask to improve conductivity. Top ITO electrode was deposited via magnetron sputtering – the process was optimized for room temperatures to avoid perovskite and organic degradation. ZnO nanoparticles are incorporated in device as second electron transport layer to protect all below layers from ITO sputtering damage. This layer was deposited via spin-coating process, which is much easier and faster than atomic layer deposition, which is usually used for ZnO deposition. The best semitransparent perovskite solar cell made with these modifications showed 11.5% efficiency with high V_{OC} value of 1.1 V.

Keywords: Perovskite solar cells, photovoltaics, tandem solar cells, semitransparent electrode, double electron transport layer

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Материалы конференции
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Разработка полупрозрачных перовскитных солнечных элементов с двойным электронным транспортным слоем и модифицированным верхним электродом

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Аннотация. В данной работе был получен полупрозрачный перовскитный солнечный элемент с двойным электронным транспортным слоем и двойным верхним электродом. Такое устройство может быть использовано в тандемном солнечном элементе. Основной сложностью в процессе синтеза полупрозрачных устройств является замена непрозрачного верхнего электрода на прозрачный. В данной работе для этой цели использовалась комбинация ITO/Ag, серебро напылялось через специальную маску, чтобы увеличить проводимость. Нанесение верхнего слоя ITO проводилось с помощью магнетронного напыления, процесс был оптимизирован для работы в условиях комнатной температуры для защиты перовскита и органики от термического разложения. Наночастицы ZnO использовались как дополнительный электронный транспортный слой для защиты всех нижних слоев от ионов в процессе напыления ITO. Этот слой был нанесен с помощью метода центрифугирования, который значительно проще и быстрее атомно слоевого осаждения, которым обычно получают пленки ZnO. Лучшее полупрозрачное устройство, полученное в таких условиях, обладало 11,5% эффективности и высоким значением $V_{х.х.}$, равным 1,1 В.

Ключевые слова: перовскитные солнечные элементы, фотовольтаика, тандемные солнечные элементы, полупрозрачный электрод, двойной электронный транспортный слой

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Introduction

Nowadays perovskite solar cells (PSCs) are the most promising photovoltaic devices. The first working cell was produced in 2009 [1], and in only fifteen years the power conversion efficiency has reached the value more that 26.1% [2]. Such fast development is possible due to excellent physical properties of perovskite material. Perovskite is originally $CaTiO_3$, but now the whole class of artificial materials with crystal structure ABX_3 is referred to perovskites. The ability of bandgap tuning with composition changing is the key advantage of these materials, the whole visible plus IR and UV parts of spectrum may be obtained using different combinations of A-cations or X-halides [3].

There is a theoretical limit of efficiency of one-junction solar cell named Shockley-Queisser limit [4], and in case of perovskite solar cells its value is around 33%. Therefore, tandem systems with two or more active materials should be produced to overcome this efficiency value. Materials with certain bandgap values should be combined to obtain the highest possible power conversion efficiency (PCE). Since the most common perovskite material $CH_3NH_3PbI_3$ ($MAPbI_3$) has bandgap of 1.55 eV, it is possible to obtain ~40% PCE when combining it with ~1.1 eV cell [5]. This value matches well with Si or CIGS cells; hence, highly efficient tandem systems Si/perovskite and CIGS/perovskite may be produced.

Since perovskite material has greater bandgap that Si or CIGS, this cell is used as top cell, and it should be therefore semitransparent to pass the light to the bottom cell. The main problem here is top electrode which is usually made of metal. It is possible to use thin metals [6] or another materials such as carbon nanotubes [7], PEDOT:PSS and indium tin oxide (ITO). The last one is the most popular transparent electrode, which is usually used as bottom electrode in PSCs. It is usually deposited via magnetron sputtering technique under high (> 200 °C) temperature, and in case of perovskite devices such conditions may damage all organic and active layers. Also, there are high energy ions while this process, and all below layers should be protected from them with some inorganic film. Therefore, there are two main problems which should be solved to have semitransparent perovskite solar cell with ITO as top electrode: the problem of perovskite degradation under high temperature, and the problem of sputtering damage to organic transport layers.

In this work, the process of ITO sputtering was optimized to room temperature. ZnO nanoparticles were incorporated in structure to protect organic electron transport layer from ion damage while sputtering.

Materials and Methods

Perovskite solar cells in this work had the following *p-i-n* structure: ITO/PTAA/perovskite/PCBM/ZnO/ITO/Ag. All layers except ITO and Ag were deposited via spin-coating process, classical $MAPbI_3$ was used as perovskite, solution was obtained combining MAI and PbI_2 salts in DMF/DMSO. Top ITO film was sputtered, and Ag was thermally evaporated. JV curves under dark and light conditions were measured to characterize the devices, in case of ITO films transparency spectrum, resistivity and thickness were measured while optimization work.

Results and Discussion

ZnO nanoparticles were spun onto the PCBM to protect it from sputtering damage, such combination works as double electron transport layer (ETL), enhancing transport properties compared to the single ETL. The first step of the work was to optimize the ZnO incorporation process. Device with PCBM/BCP double ETL was used as reference having the best efficiency of 16.9%. Different parameters of spin-coating process or annealing were changed during the experimental work. After the optimization, the champion device with ZnO showed high PCE value of 17.2% (Fig. 1), which can be used as base device for next ITO deposition.

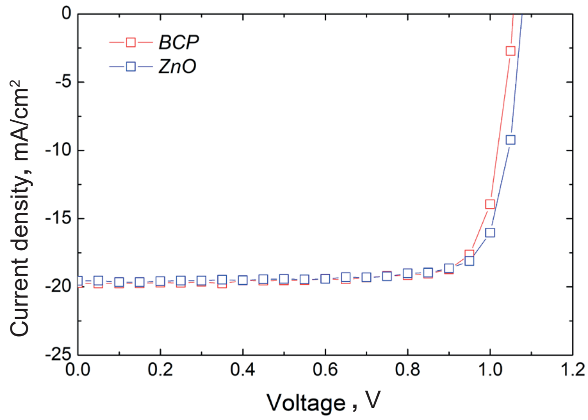

Fig. 1. JV curves of reference devices with BCP and ZnO layers

Since protective double ETL in perovskite solar cells was achieved, work on optimization of ITO sputtering under room temperature has begun. There are several parameters, which can be tuned in this process: flux of the gases (Ar and O_2), target power, process time etc. First main goal was to achieve ITO film under room temperature with low resistivity and high transparency. If O_2 flux was equal to 5 sccm, resistivity was found to be 109 Ohm/sq, almost transparent yellowish films were obtained. In such conditions the first semitransparent devices were made, and PCE of only 6.2% was obtained (Fig. 2). Spatial silver frame was used to improve the conductivity, it was deposited on the edges of the pixel, therefore there are still semitransparent ITO windows.

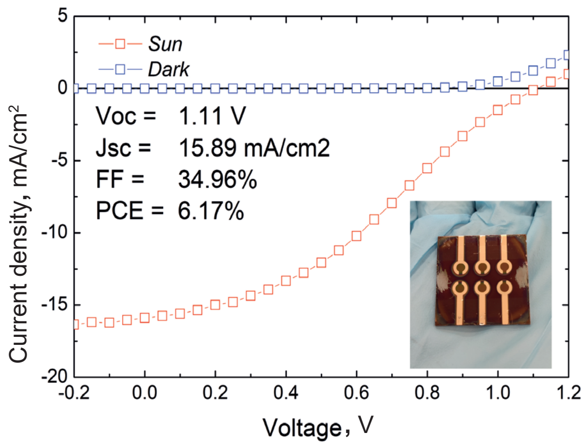

Fig. 2. JV curves of the first semitransparent device and photo of the device (inset)

However, fill factor (FF) of the device was too law – only 35%, while V_{OC} had normal value of 1.1 V. The reason of low FF is high series resistance in the device, which occurred due to additional ITO film. After this, the work on ITO film improvement was done. It was found that changing O_2 flux value may affect the resistivity of the film, 11 sccm led to only 38 Ohm/sq, *T* spectrum showed also better transparency (Fig. 3, *a*). In such conditions champion semitransparent device with PCE of 11.6% was achieved with high FF value of 62% (Fig 3, *b*).

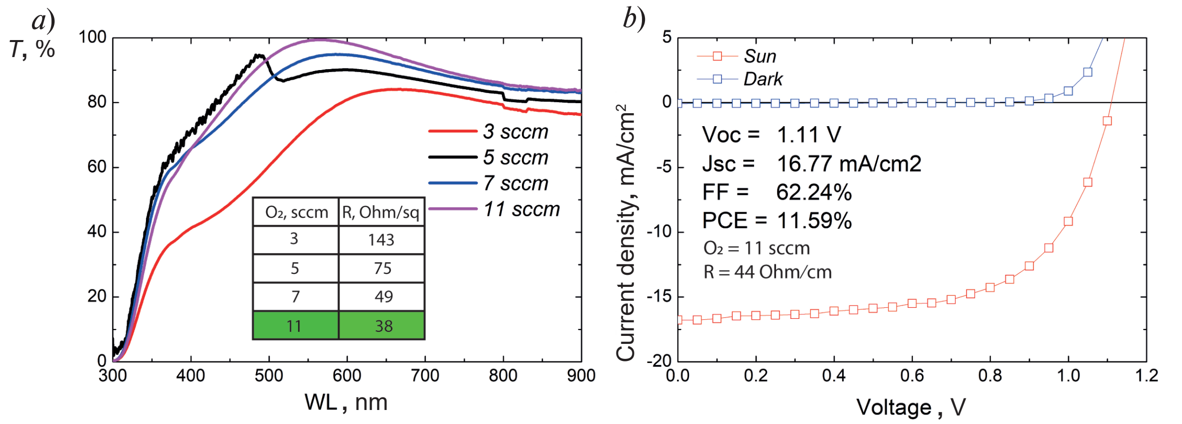

Fig. 3. *T* spectrum of different ITO films (*a*) and JV curves of the best semitransparent device (*b*)

In order to incorporate such solar cell in tandem with narrow bandgap material, the device should be transparent in IR part of spectrum. Transmittance was measured for a pixel from semitransparent window of the device (Fig. 4).

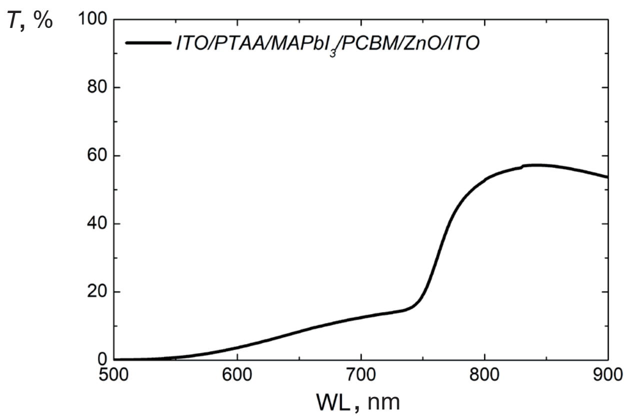

Fig. 4. T spectrum of the whole device measured through ITO window

It can be seen from the spectrum that obtained device passes radiation of IR part through itself. There are losses and *T* value is not high, but this device already may be used in experimental tandem systems with Si solar cells since it is semitransparent for long wavelengths.

Conclusion

In this work, semitransparent perovskite solar cells with double ETL and electrode were fabricated. High V_{OC} value of 1.1 V and FF of 62% led to PCE of 11.6%. Protective inorganic ZnO nanoparticles were spun onto PCBM to avoid high energy ion damage while sputtering process. Changing gas flux under room temperature conditions led to low resistance and high transparency of ITO films. Further optimization of ITO sputtering process should be done to improve J_{SC} of the semitransparent device, and after tandem solar cells of Si/perovskite or CIGS/perovskite may be produced.

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