

TIME RESPONSE ANALYSIS OF PEROVSKITE/SI TANDEM SOLAR CELLS

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ABSTRACT: Perovskite and Silicon solar cells have recently been shown to be perfect partners for tandem devices with potentially very high efficiency at low additional costs over standard Si cells. Several aspects must be considered regarding the characterization of these novel devices. Spectral response or external quantum efficiency measurements of perovskite/Si tandem devices present additional challenges compared to single junction ones due to the series connection of the junctions. Light bias intensity is expected to affect the quantum efficiency of each junction in the tandem since the appropriate current limitation must be applied each time. In this paper, external quantum efficiency measurements have been implemented at different light bias conditions in an attempt to establish the minimum recommended light bias conditions during quantum efficiency measurements of Silicon bottom junction. Furthermore, due to the slow photogenerated characteristics of the perovskite cells, the chopping frequency during quantum efficiency measurements of those devices is expected to be different compared to conventional Silicon cells. In an attempt to investigate this, external quantum efficiency measurements have been conducted at different chopping frequencies.

Keywords: Perovskite, Tandem, Spectral Response, Characterization

1 INTRODUCTION

Multi-junction solar cells have been the most promising approach to achieve efficiencies higher than those of their single-junction counterparts. Detailed balance limit calculations revealed that tandem solar cells can reach energy conversion efficiencies exceeding 70% if an ideal material combination is selected. Recently, perovskite/Si tandem solar cells have attracted significant attention due to their very high energy conversion efficiencies up to 28% [1]. The rapid improvement of perovskite/Si tandem performances and the related growing interest in this emerging technology clearly indicate the need for methods and procedures to reliably measure and characterize perovskite/Si tandem solar cells. Electrical parameter measurements of these devices under standard testing conditions (STC) can be accurately made only if sufficient time is allowed to complete the photocurrent generation. This time is usually much longer for perovskites than for typical crystalline Silicon (Si) devices [2]. The external quantum efficiency (EQE) measurements of perovskite/Si tandem devices are challenging due to the photocurrent decay of perovskite which can lead to systematic distortion of the EQE spectral shape during those measurements. Considering the slow response of perovskite solar cells, suitable chopping frequencies of the monochromatic beams need in spectral responsivity (SR) and EQE measurements should be determined carefully to avoid underestimation of the short-circuit current when the standard AC method is applied according to IEC 60904-8. In this paper we investigate the response for photocurrent generation of a $\text{FA}_{0.85}\text{CS}_{0.15}\text{PbI}_{2.5}\text{Br}_{0.5}$ perovskite/Si tandem solar cell at different chopping frequencies.

Moreover, this paper aims to contribute to the improvement of perovskite/Si characterization by reporting the effect of light bias at different testing conditions. In tandem cells, due to the series connection of junctions, the subcells are not accessible separately and the EQE of a certain subcell has to be measured by using the effect of current limitation [3]. Therefore, the

intensity of light bias plays crucial role for the appropriate current limitation of the junction of interest and the minimization of measurement artifacts during the EQE process. Moreover, light bias EQE measurements present another main problem: Since the tandem performance is very sensitive to light bias, those measurements are strongly dependent on the degradation level of the cell. In this case the optimum light bias conditions are different for different degradation levels of the cells. The EQE results shown here correspond to a perovskite/Si tandem cell that presents a short-circuit current of 26 mA under STC conditions (around 52% degradation from its initial short circuit current). In the paper, the optimum light bias conditions for EQE measurements of Si junction in the perovskite/Si tandem solar cell are reported.

2 MEASUREMENT APPARATUS

For the time response analysis measurements described here we followed the same procedure presented in [4]. Two different monochromatic light sources (465 nm, 808 nm) and a chopper have been used to apply chopped monochromatic light to a Si reference device and to a perovskite/Si tandem device. One of the light sources lies in the wavelength region of the perovskite top junction while the other light source lies in the wavelength region of the bottom Si junction. A beam splitter allowed the simultaneous measurement of the Si reference device and the tandem perovskite/Si cell. The voltage signals of both devices were collected through transimpedance amplifiers with oscilloscopes. The frequency of the chopper was varied from 5-80 Hz. 5 Hz is the minimum operating frequency of the chopper used while 70 Hz -80 Hz is the frequency usually used during the SR measurements of a typical solar cell.

For the light bias EQE measurements, the typical AC method for EQE measurements has been utilized. The EQE measurement set-up at the Photovoltaic Laboratory, University of Cyprus, consists of a steady-state Quartz-

Tungsten-Halogen light source in series with a monochromator to produce the monochromatic light input, which is then chopped by a chopper, superimposed over continuous bias light and measured by digital lock-in-amplifiers. The monochromatic light is separated by a beam splitter and allows the simultaneous measurement of the test device and a reference cell of known absolute EQE. Plano-convex lenses are located between the beam splitter and the sample in order to focus the light onto the cells. The test device temperature is kept stable at 25°C by using a temperature-controlled base and is stabilized using a PID control algorithm running on ARDUINO hardware. The reference device used was a NIST traceable calibrated Si photodiode which is sensitive across the visible and into the near infrared spectrum. A set of colored light sources have been used to saturate the non-measured junction and subsequently achieve current limitation by the junction of interest. In particular, a blue and a purple LED have been used for the measurement of the bottom Si junction.

Two identical perovskite/Si tandem devices of active area of 4 cm² were involved in the measurements presented. The one of the devices was used in the time response analysis while the other one was used for the light bias dependent EQE measurements. The two devices under test are monolithic FA_{0.85}CS_{0.15}PbI_{2.5}Br_{0.5} perovskite/double-planar homo-junction Si tandem cells. The 4 cm² devices achieve a power conversion efficiencies of 15.6% with an open-circuit voltage of 1.58V, a short-circuit current density of 13.6mA/cm² and a Fill Factor of 72%. More details about the cells can be found elsewhere [5].

3 RESULTS AND DISCUSSION

In order to investigate and find the optimal chopping frequency for these devices, we have performed the time response analysis method. A quantitative time response analysis method was applied according to [4]. The voltage signals of both devices were collected through transimpedance amplifiers with oscilloscopes. The frequency of the chopper was varied from 5-80 Hz. 5 Hz is the minimum operating frequency of the chopper used while 80 Hz is the typical frequency used during EQE measurements of conventional solar cells. The signals were acquired for at least three periods at each frequency and the peak-to-peak amplitude variation was measured over the whole frequency range. An example of the signals of the device under test (DUT) perovskite/Si tandem at two representative chopping frequencies (10 Hz and 80 Hz) and at different excitation wavelengths (465 nm and 808 nm) are shown in Fig. 1.

As shown in Fig. 1 both devices demonstrate constant response at chopping frequencies of 10 Hz and 80 Hz and in both monochromatic light sources. The measurements reveal that the perovskite/Si tandem cells are not presenting slow photo-generation characteristics since no reduction of the peak-to-peak amplitude was obtained with chopping frequency. Therefore, despite the slow photogenerated characteristics of the perovskite/Si tandems the frequency does not determine the EQE measurements. The choice of the chopping frequency is irrelevant during EQE measurements of perovskite/Si cells.

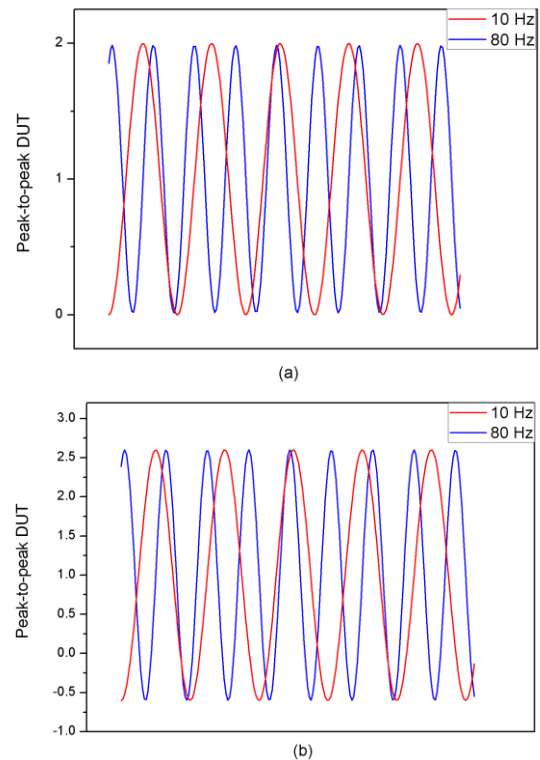


Figure 1: Photocurrent signals of the perovskite/Si tandem (DUT) at 10 Hz and 80 Hz illuminated by a monochromatic chopped light of (a) 465 nm and (b) 808 nm.

Light bias EQE measurements of the Si subcell were conducted afterwards. Light bias measurements are useful to define the low and ideal light bias conditions that should exist during the EQE studies of these junctions. It is important to note that the EQE measurements presented here correspond to EQE values of a degraded tandem cell. Therefore, the ideal and minimum light bias conditions determined correspond to the EQE values of the cell at the moment of measurement. These condition measurements will change according to the performance of the cell at the time of the measurement. More measurements have to be conducted in perovskite tandems with different degradation levels in the future in order to extract the dependence of the light bias conditions with the given EQE of the perovskite/Si devices.

During the measurement of the bottom Si junction a combination of a purple and a blue LED at 405 nm and at 465 nm has been applied to the device. The bias light applied to the devices was measured using a calibrated photodiode located next to the tandem device during the light bias studies. Fig. 2 demonstrates the EQE of the Si bottom junction at different light bias intensities. Light bias was varied from around 20 W/m² to 413 W/m² and EQE of the Si junction was collected each time. For comparison purposes, the dark EQE of the device is also plotted in Fig. 2. Dark EQE is the EQE of the tandem measured in the absence of light and indicates the relative shunt resistances of the junctions in the tandem [6]. The measurements show that the EQE signal of the junction is marginally increased with increasing light bias and no considerable change is obtained over a wide range of light biases. Particularly, only 3% increase is obtained on the EQE in the presence of light bias (compared to the

dark EQE case) indicating that the light bias is not a critical issue during the measurement of the bottom Si junction. One important observation arising from Fig. 2 is the reduction of the EQE Si signal in the response region of perovskite (600-750 nm) in the presence of light bias. However, the EQE in that region does not tend to zero at higher light bias intensities since Si presents some response in that wavelength region as well. The photocurrent produced by the tandem device during these measurements is also demonstrated in Fig. 2. This current corresponds to the photocurrent produced by the Si bottom junction since that junction is the current limiting junction in those conditions. No voltage bias was applied on the device in this case. During the EQE measurements of Si bottom junction at high light bias conditions (413 W/m^2), no luminescence coupling effects have been observed. The luminescence coupling causes underestimation of the signal in the response region of the bottom junction and overestimation in the response region of the top junction owing to the transfer of carriers from the top to the bottom junctions. Such an effect was observed during the EQE measurements of the InGaP/InGaAs/Ge triple junction devices [7]. Absence of that phenomenon in our measurements indicates that it is negligible at light bias conditions up to 413 W/m^2 .

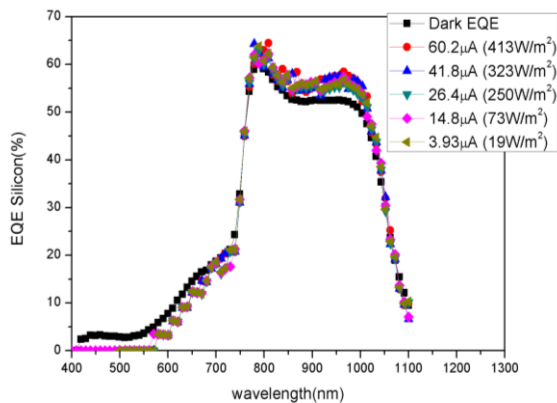


Figure 2: Measured EQE of the Si bottom junction under variable light bias intensity.

4 CONCLUSIONS

The paper demonstrates the impact of chopping frequency on the time response of the cells as well as the effect of light bias during EQE measurements of the Si bottom junction in the perovskite/Si configuration. Chopping frequencies in the range from 5-80 Hz have been applied on perovskite/Si multi-junction solar cells. Quantitative time response analysis was undertaken using oscilloscopes at various chopping frequencies ranging from 5-80 Hz. Time response of the perovskite/Si tandems was found not to be affected by chopping frequency indicating that the frequency might not be an issue during the EQE measurements of perovskite/tandem solar cells.

Furthermore, light bias measurements have demonstrated that the minimum light bias conditions for measuring the Si bottom junction in the perovskite/Si configuration is 20 W/m^2 . Above this value no improvement in the EQE signal was obtained.

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