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Minimizing the energy loss of perovskite solar cells with Cu^+ doped NiO_x processed at room temperature



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ABSTRACT

Room-temperature direct current magnetron sputtering was employed to prepare pristine and Cu^+ doped NiO_x films for perovskite solar cells (PSCs). Optimal power conversion efficiency (PCE) of about 15% was obtained with a 20 nm thick film with copper content of 5.7%. The solar cells based on as sputtered films demonstrated high reproducibility and stability. The origin of dramatically boosted photovoltaic performance of PSCs based on Cu doped NiO_x was analysed from energy loss considerations. The optical, carrier transport as well as recombination losses were discussed. According to our results, PSC with Cu doped NiO_x have lower recombination and transport loss which ensured the hysteresis free high efficiency.

1. Introduction

The pursuing of high photovoltaic performance is a struggling procedure to minimize the energy loss. This overall rule guided researchers in material choosing and device structure designing. In 1961, Shockley and Queisser discussed the band gap dependent power conversion efficiency (PCE) limit of single p-n junction solar cells [1]. Black body radiation of the device, recombination and spectrum loss were considered. As the solar energy not converted into electricity generally turned to heat, the inefficiency of the devices leads to the temperature increase. Blackbody radiation releases energy and hence equilibrium between heat generation and radiation eventually reaches. The practical temperature of a solar cell is often as high as 360 K, resulting in the degradation of operating PCE. Recombination in the solar device determines the maximum electron-hole generation rate [2]. The open circuit voltage (Voc) is determined by band gap (Eg) as well as recombination rate (relaxation of excited electrons to band edge) [3]. The relaxation increases with the decrease of band gap. Single junction devices generally could not cover the whole solar spectrum which limited the short circuit density (J_{SC}) . With the increase of band gap, the spectrum loss increases as the below band gap photon count dramatically rises [4]. A theoretical maximum of about 33.7% with an optimum light absorber band gap at 1.4 eV was proposed based on the balance of recombination, spectrum loss as well as other loss [5]. Their work provided an overall guide on active material choice for photovoltaic application. According to their results, semiconductors with band gaps between 0.9 and 1.65 eV are among the most efficient candidates for solar conversion with theoretical maximum efficiencies over 30% [6–8].

The energy losses of practical materials are quite different even though they may have similar band gaps. A simple and practical method to evaluate the basic energy loss of photovoltaic device with certain material is V_{OC}/E_g . Fig. 1 presented the V_{OC}/E_g evolution of different photovoltaic systems. Among various emerging photovoltaics, perovskite solar cell (PSC) has relatively high V_{OC}/E_g , a value of 74.8% was achieved with solution based process. In contrast, the basic energy losses in organic photovoltaic (OPV) [9] and dye-sensitised solar cell (DSSC) [10,11] are much higher, leading to the inefficiency of these devices. Although higher V_{OC}/E_g values are obtained by some other strategies, the demanding fabrication process or low efficiency remains as a problem resulting from the low defect tolerance of these materials.

Device structure is another factor that have great influence on

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Fig. 1. The V_{OC}/E_g values of solar cells based on different materials.

energy loss during solar-to-electricity conversion. For crystal silicon solar cells, the thickness of p and n doping layer realized the optimum carrier separation rate [10]. The antireflection coating and textured aluminium reflection back electrode enhanced the light capture efficiency. Scattering layer is adopted in dye-sensitised solar cells to maximize the absorption of photons in the available range [12]. The optimization of device structure would surely work in newly emerging perovskite photovoltaics.

Regarding to PSCs, the high light absorption coefficient of perovskite material ensured sufficient light harvesting with an active material of several hundred nanometres thick [13–15]. Extra scattering or reflection layers are unfavourable to enhance the light capture. Instead, developing a highly transparent film on transparent conductive oxide (TCO) layer would reduce the incident light loss and maximum the available photon flux for perovskite. Though many studies confirmed the long carrier diffusion lengths of perovskite material, the processing approaches set a limit to the practical film thickness in planar structured devices [16–18]. Most researchers fixed the perovskite absorber thickness to the range of 350–500 nm, which is a natural choice of balancing light absorption and carrier separation rate [19,20]. In this work, the light absorber thickness is also located in this range in order to lower energy losses.

Energy losses, in this context, includes incident light loss, basic energy loss, recombination loss and carrier transport loss. To reduce incident light loss, the carrier transport layer (CTL) on illumination side should be highly transparent, allow as much photons as possible to reach perovskite absorber for photon to electricity conversion. To decrease basic energy loss, the differences of conduction band edge (CBE) or lowest unoccupied molecular orbit (LUMO) of electron extraction layer (EEL) and valance band edge (VBE) or highest occupied molecular orbit (HOMO) of HEL should be maximised. Recombination loss could be minimized by optimise the perovskite carrier transport layer (CTL) interfaces to realize highest carrier separation rate. The carrier transport loss mainly takes place at CTL, and it could be decreased by improving the conductivity of CTL [21,22].

Therefore, CTL on the illumination side is of great significance not only because it extracts carriers from active layer but also it allows photons reach perovskite and transports carriers to FTO electrode. For its relevance on both photon losses as well as carrier losses, the preparation approach is important and could minimize the energy loss of PSCs. By fabricating a highly transparent, well-conductive CTL with suitable band structure, it is possible to dramatically decrease the energy losses in PSC and hence enhance the PCE.

In this work, we decreased the energy loss of NiO_x based PSC by Cu^+ doping at room temperature with a co-sputtering technique. The incident light loss remained almost the same after doping, however, the

basic energy loss, carrier transfer loss as well as carrier transport loss were dramatically decreased. As a result, the PCE of solar device was amazingly boosted by 53.4%. Our approach proposed here is of great potential for wide application. The direct current magnetron sputtering (DCMS) process is capable to economically deposit HELs with highly homogeneity without the limitation of size. Besides, the room temperature process would extend the perovskite photovoltaics to wider application fields based on flexible device.

2. Experimental section

2.1. Fabrication of solar cells

Fabrication of Cu doped $\rm NiO_x$ hole layers and perovskite solar cells are provided in the Supporting information.

2.2. Characterization

Scanning electron microscopy (SEM) was investigated on a field emission scanning electron microscope (FEI, Magellan 400). The surface morphology of the film was obtained by atomic force microscopy (AFM, SII Nano Technology Ltd., Nanonavi) in noncontact mode. The chemical composition and valence-band (VB) spectrum of the Cu:NiOx films surface were analysed by X-ray photoelectron spectroscopy (XPS) with monochromatic Al Ka radiation at a pass energy of 29.4 eV. The UV-vis transmittance spectra of Cu:NiOx film were recorded on UV-Vis spectrophotometer (HITACHI U-3010). Steady photoluminescence (PL) measurements were conducted at room temperature on a Horiba-Ltd. FluoroMax-4 device with an excitation wavelength of 470 nm. Time-resolved PL spectra were measured using a fluorescence lifetime spectrometer (Photo Technology International, Inc.). The PL lifetime of the CH₃NH₃PbI₃ films on Cu:NiO_x /FTO glass substrates were calculated by fitting the experimental decay transient data with bi-exponential decay model. Current - voltage characteristics of solar cells were measured under simulated AM1.5 G illumination of $100 \,\mathrm{mW \, cm^{-2}}$ with a Keithley-2420 source meter in combination with a Sol3A class AAA solar simulator IEC/JIS/ASTM equipped with an AM1.5 G filter and a 450 W xenon lamp. The light intensity was calibrated with a reference silicon solar cell (Oriel-91150). The J-V curves were respectively measured by applying an external voltage bias with a scan rate of 40 mV s-1. Incident photon-to-current conversion efficiency (IPCE) was measured on a SM-250 system (Bunkoh-keiki, Japan). The intensity of monochromatic light was measured with a Si photodiode (S1337-1010BQ).

3. Results and discussion

The deposition system of NiO_x was illustrated in Fig. 2a. The quality of NiO_x films was closely relate to the operation parameters. The sputtering power of nickel target was fixed at 300 W, and the chamber pressure was 1 Pa, with an oxygen argon ratio of 36:6. In order to realize optimal sputtering efficiency, the substrate surfaces were normal to the incident direction of ions (substrate target angle 37°, as shown in Fig. 2a) [23]. Based on DCMS NiO_x films, PSCs were fabricated. The thickness of NiO_x film was optimized by tuning the deposition time. The photovoltaic performance of PSCs based on NiO_x films with sputtering time of 1 min, 2 min, 3 min and 4 min, which was corresponded to the thickness of 10.2 nm, 21.5 nm, 33.1 nm and 45.6 nm, was listed in Table S1. Maximum PCE was achieved with a thickness of 21.5 nm, which was adopted as the 2 optimum value in this work.

Copper ion was doped in NiO_x film (Cu:NiO_x) by co-sputtering nickel and copper target. Ni and Cu targets were placed opposite to enhance the distribution homogeneity of Cu in the film. The crystal structure of Cu:NiO_x as well as NiO_x was presented in Fig. 2b and c. At room temperature, NiO_x has a space group of R-3 m (166). X-ray photoemission spectra (XPS) of O 1 s and Ni 2p of NiO_x and O 1 s, Ni 2p and Download English Version:

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