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# Theoretical insights into highly transparent multi-sized conducting films with high-haze and wide-angular scattering for thin film solar cells

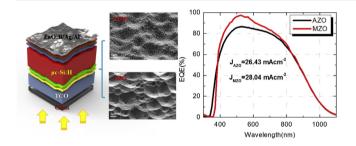


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#### HIGHLIGHTS

- MZO films consisting of micro-sized craters with nano-sized pyramids are fabricated.
- MZO films extend the scattering angular domain and enhance the scattering intensity.
- MZO films can significantly improve the current output of solar cells.

#### G R A P H I C A L A B S T R A C T



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# ABSTRACT

Recent advances in light trapping schemes open up new gateways for enhancing the absorption of solar energy that approaches and overcomes the Yablonovitch  $4n^2$  limit based on isotropic Lambertian scatterers. Achieving wide-angular scattering while maintaining a strong scattering intensity is the key to realize a Lambertian-like scatterer that may have a great potential to approach the absorption upper limit. However, few current light trapping strategies can experimentally extend the scattering angular domains in absorbers while maintaining a high scattering intensity. In this paper, we theoretically and experimentally investigate multi-sized transparent conducting oxide (TCO) films, which are comprised of micro-sized, magnetron-sputtered and chemically etched aluminum-doped zinc oxide (ZnO:Al), coated with metal organic chemical vapor deposition (MOCVD) deposited nano-sized, boron-doped zinc oxide (ZnO:B) pyramids. We demonstrate that the multi-sized TCOs in this study can efficiently increase the total transmittance in the visible spectral range, enhance the scattering intensity, successfully extend the scattering angular domains to  $90^\circ$ , and improve the short-circuit current density and power output of solar cells. The combination of these factors endows the TCOs with the significant potential for realizing a Lambertian-like scatterer. Accordingly, the multi-sized architecture may inspire fresh ideas for realizing more innovative light-trapping architectures.

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## 1. Introduction

Light harvesting in the field of photovoltaic (PV) has attracted extensive attention by allowing for either thickness reduction [1], material savings, and light-induced stability improvement [1–3] or stronger light absorption and thus higher current output. Transparent conducting oxide (TCO) films - which are widespreadly utilized as essential components in displays, cameras, touchscreens, and solar cells - play a key role in the light harvesting capacity of solar cells. Highly transparent and scattering TCOs while maintaining a high electrical conductivity, which decreases the ohmic heat loss [4] while reducing the electrical deterioration effect on the deposited cells [5], should be developed according to the requirements of both the optical and electrical points of view. The light scattering properties of TCOs are strongly dependent on the feature sizes of the surface morphologies that affect the light phase variation and scattering angles [6]. Such surface morphologies can be experimentally tuned to modify the light scattering properties of TCOs. With the aim of harvesting more photons in absorbers, various TCOs with specially designed surface morphologies are being developed, such as a wide range of periodic surface architectures, including triangular or pyramidal gratings [7,8], nanowires [9,10], and nanocones [11] et al., and randomly textured surface architectures [12,13]. More importantly, improved efficiencies of solar cells with scattering layers have also been observed [14–18]. Periodically textured TCOs allow for the incident light to be selectively diffracted into certain angles [19] and the absorption can be efficiently enhanced beyond the Yablonovitch  $4n^2$  (n is the refractive index of the material) limit [20] of the randomly textured surfaces at certain wavelengths  $(8\pi n^2/\sqrt{3})$  for the triangular geometry,  $4\pi n^2$  for the square geometry, and  $2\pi n$  for the one dimensional (1D) grating) [21,22]. Unlike the limited light harvesting capacity of the periodically textured TCOs, the randomly textured TCOs allow for the continuous scattering of the broadband incident light in a broad scattering angular domain [23] and a broadband absorption enhancement with an upper limit of  $4n^2$  can be acquired on the basis of an ideal Lambertian scatterer [20]. With the expectation of approaching the upper limit of the absorption enhancement, a Lambertain-like scatterer that isotropically scatters the incident light in the 0°-90° angular range should be experimentally acquired. However, the conventional randomly textured TCOs with single-scale textures – including tin oxide (SnO<sub>2</sub>), indium-tin oxide (ITO), chemically-etched aluminum-doped zinc oxide (ZnO:Al), and metal organic chemical vapor deposition (MOCVD) as-deposited boron-doped zinc oxide (ZnO:B) substrates [24-26] - are either far from being Lambertian-like scatterer, or have scattering intensities that are too weak to qualify as Lambertian-like behavior. How to experimentally fabricate a Lambertian-like scatterer while maintaining a high scattering intensity is therefore the focus of the research.

R.Dewan et al. theoretically studied the effects of the height and width of 2D periodic triangular profiles on the current output of microcrystalline silicon solar cells ( $\mu$ c-Si:H) and pointed out that the diffraction angles of the period of triangular units is reduced as the period increases [27]. This represents a theoretical observation that is not only suitable for the periodically textured surfaces, but also allows for the modulation of the scattering angular domains of randomly textured surfaces, the lateral and vertical feature sizes of which can be represented with the correlation length ( $l_c$ ) and roughness ( $\sigma_{rms}$ ) respectively [28]. Therefore, a combination of features with large and small lateral sizes may potentially produce a scattering distribution in a full angular domain — that is to say, Lambertian-like scattering behavior.

To date, the deliberate design of multiscale electrode architectures to guide light into the effective absorbers has emerged as an

active area of research with a wide variety of proposed multiscale architectures [29–31]. For instance, M. Boccard et al. combined nanoimprint lithography and self-textured ZnO to develop the multiscale electrodes [29]. Furthermore, S. S. Yang et al. proposed a double-period electrode comprised of a small-period self-textured MOCVD ZnO:B film deposited on a large-period, texture-etched ZnO:Al substrate [30]. However, many studies only account for the experimental fabrication of the multi-sized electrodes. To date, few theoretical studies on the optical properties of multi-sized electrodes — such as the angular scattering behavior — have been conducted.

In this study, we experimentally and theoretically investigate a type of multi-sized TCO film consisting of micro-sized magnetron-sputtered and chemically-etched ZnO:Al crater-like features decorated with nano-sized MOCVD deposited pyramids. Our analysis shows that the multi-sized TCOs can efficiently increase the transmittance in the visible spectral range, extend the scattering angular domains into  $90^\circ$ , enhance the scattering intensity, and eventually improve the current and power output of the  $\mu$ c-Si:H single-junction solar cells with enhanced light coupling and trapping. The broadened properties of scattering angular domains while maintaining a high scattering intensity make the multi-sized architecture have a great potential to achieve a Lambertian-like scatterer.

### 2. Experimental details

Magnetron-sputtered ZnO:Al substrates were chemically etched for 40 s in 0.5% diluted hydrochloric acid (HCl) solution to produce randomly-textured surfaces. ZnO:B films were deposited with an MOCVD process, diethylzinc (DEZ) and water ( $\rm H_2O$ ) vapours were employed as precursors with additional diborane ( $\rm B_2H_6$ ) as the doping gas.

Pin-type  $\mu$ c-Si:H single-junction solar cells with intrinsic thicknesses of about 2000 nm were simultaneously deposited on substrates with very high frequency plasma enhanced chemical vapor deposition (VHF-PECVD). The substrate temperature was 200 °C. A mix of hydrogen (H<sub>2</sub>) and silane (SiH<sub>4</sub>) source gases was used to prepare the  $\mu$ c-Si:H intrinsic layers. P- and n-type layers were deposited by adding additional diborane boroethane (B<sub>2</sub>H<sub>6</sub>) and phosphine (PH<sub>3</sub>) source gases, respectively. The substrate temperature and chamber pressure were carefully controlled to ensure that the experimental conditions were identical for all samples. ZnO:B/Ag/Al back contacts, which acted as mirror reflectors, were deposited on the  $\mu$ c-Si:H cells. The resulting cells had an active area of 0.25 cm<sup>2</sup>.

The illuminated J-V characteristics of the cells were measured with a Wacom WXs-156s-12 dual-beam solar simulator under 1 Sun illumination (AM 1.5, 25 °C, 100 mW/cm<sup>2</sup>). An excellent simulation of the solar spectrum with the standard AM1.5 spectrum is obtained from the superposition of the simulator's two filtered light sources after adequate calibration. External quantum efficiency (EQE) curves of all the cells were measured to characterize the effective response of the incident light at different wavelengths. Because of the deviation between simulated and real AM1.5 spectra, which might possibly lead to an imprecise determination of the short-circuit current density  $(J_{SC})$  values, the  $J_{SC}$  values of the cells were determined by integrating the EQE curves with a standard AM1.5 spectrum. The surface morphologies of the substrates in this study were characterized with atomic force microscope (AFM) images measured with SPA 400 AFM (SII Nanotechnology Inc., Tokyo, Japan) over a scanning area of 10  $\mu$ m  $\times$  10  $\mu$ m. For characterizing the optical transmittance and scattering properties, the total optical transmittance  $T_t(\lambda)$  and specular transmittance  $T_s(\lambda)$  of the films, which were deposited on Eagle XG glasses, were

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