

Tsuchime-like cost-efficient aluminum film to enhance absorption in thin and ultra-thin photovoltaic cells

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Abstract: Silicon solar cells account for more than ninety percent of the photovoltaic (PV) market, primarily due to convenience, cost-effectiveness, and long service life. However, the light absorption efficiency of PV cells remains a limiting factor, especially for thin and ultra-thin solar panels. Most methods for increasing light absorption in thin and ultra-thin solar cells are either cost-ineffective or technically challenging, given the thinness of the functional layers employed. In this article, we propose employing a lithography-free and cost-efficient approach to increase the light absorption in thin and ultra-thin solar cells using a Tsuchime-like self-forming nanocrater (T-NC) aluminum (Al) film. T-NC Al film can be obtained by the well-known process of electrochemical anodizing of Al followed by etching the nanoporous alumina (Al_2O_3). According to the current study, T-NC film boosts the average absorbance in the 400–1100 nm wavelength range by 85.9, 64.3, 46.3, and 41.8% for PV cells with silicon active layer thickness of 500 nm, 1 μm , 3 μm and 10 μm , respectively. Moreover, the wavelength range of increased absorption depends on silicon thickness, where peak of absolute increase of absorbance moves from 620 nm to 950 nm as the thickness of the silicon layer increases from 500 nm to 10 μm . Therefore, T-NC Al film allows for a significant increase in the efficiency of thin and ultra-thin solar cells without involving costly top-down techniques such as lithography.

1. Introduction

One of the fundamental challenges concerning global society is to reduce the emission of combustion products such as carbon dioxide (CO_2) into the atmosphere [1–5]. On the one hand, over the past decade, significant achievements have been made in reducing the energy consumption of household and industrial electrical devices [6–8]. However, demand for electricity continues to grow along with the current drift towards electric vehicles, as well as the ongoing surge in the field of artificial intelligence [9]. Additionally, it is worth noting the ongoing growth of visual information devices, when the area covered by displays keeps expanding every year, while the lifespan of handheld devices is constantly decreasing, and the augmented and virtual reality devices are becoming more accessible [10]. Undoubtedly, the source of clean and inexhaustible electrical energy becomes the cornerstone, one of the solutions to which is employing solar energy.

Silicon (Si) solar cells are ubiquitous in the market due to the well-established manufacturing process, cost-efficiency, and reliability [11]. Since the very first generation of Si photovoltaic (PV) cells, one of the most powerful ways to increase efficiency has been to maximize absorption within the cell since the light absorbed in the Si active layer directly

contributes to photocurrent generation [12–15]. However, weak light absorption due to low absorption coefficient and reduced active layer thickness cut down the performance of thin and ultra-thin PV cells [16–18]. Weak absorption results from optical losses such as reflections from front solar cell surface and very short optical path length due to thinness of the active layer [19,20]. To enhance the absorption in the structure, it is necessary to reduce the reflection on the surface of the solar panel and lengthen the optical path as much as possible. With the purpose of the latter, micro- and nanoscale photonic structures can be utilized, however fabricating such structures generally requires high-cost top-down processes such as lithography [16,21].

Here, we propose to use a self-forming nanopatterned aluminium (Al) film to significantly increase absorption in thin and ultra-thin Si solar panels. The nanostructure, resembling a nanoscale Tsuchime (or hammertone) patterned surface, results from the electrochemical nanoporous (NP) anodizing of Al followed by etching away the NP alumina (Al_2O_3) layer [22]. This method of Al NP anodizing followed by removing the oxide completely omits the use of costly equipment, such as lithography; the method easily scales, and allows tuning the diameter and depth of resulting nanocraters (NCs) in a wide range [22–24]. Moreover, it allows producing ordered micro/submicro- and nanopatterned surfaces by iterating the procedure in several steps of anodizing followed by etching – factually making prior NC pattern a mask for the following anodization [25,26]. Using our experience in fabricating and modelling nanostructured Al surfaces for transparent electrodes, we conveyed a theoretical study of Tsuchime like NC (T-NC) film as a functional layer of thin Si solar cells. We found the optimum diameter between the NCs in the nanopatterned film for different thickness of Si active layer for maximizing the optical path in the solar cell. Among the tested configurations of T-NC films, the best result was obtained when diameter of NCs (d_{NC}) was set to 800 nm in combination with 3000 nm thick Si layer (H_{Si}). However, the effect of using a T-NC film is the higher, the thinner the Si active layer of the solar panel. Applied in PV cells with Si active layer thickness of 0.5, 1, 3 and 10 μm , T-NC film boosts the average absorbance ($\Delta\lambda = 400\text{--}1100\text{ nm}$) by 85.9, 64.3, 46.3 and 41.8%, respectively. Moreover, the wavelength range of increased absorption depends on Si thickness. Peak of absolute increase of absorbance moves from 620 nm to 950 nm as the thickness of Si increases from 500 nm to 10 μm ; hence, thinner PV cells benefit in the visible spectrum, while thicker cells benefit in the near infrared range. Therefore, T-NC Al film allows to significantly increase the efficiency of thin and ultra-thin solar cells without the use of expensive lithography.

2. Methodology

Figure 1 schematically explains the concept behind increasing absorption in thin and ultra-thin Si solar cells using a T-NC Al film. When light interacts with planar Al surface [Figs. 1(a) and 1(b)], there is a single process of reflection of light. The effective optical path of the reflected light is minimal. In the case when light collides with the T-NC film [Figs. 1(c) and 1(d)], light is reflected in directions other than in case of a planar surface. This includes multiple reflections from the inner side of the NC. As a result of multi-reflecting, the effective optical path inside the Si active layer increases, which, in turn, leads to an increase in absorption in the cell. Fig. 1(e) shows the scanning electron microscope (SEM) image of the experimentally obtained T-NC Al surface with diameter of NC $d_{\text{NC}} = 300\text{ nm}$. As can be seen from the figure, the nanopatterned structure resembles a crater left by a hammer on metallic surface – so-called hammertone or Tsuchime technique [27]. Experimental details of electrochemical NP anodizing and etching of Al film are fully described in our previous work (Ref. 17). Fig. 1(f) shows the lateral SEM image of the same T-NC Al film.

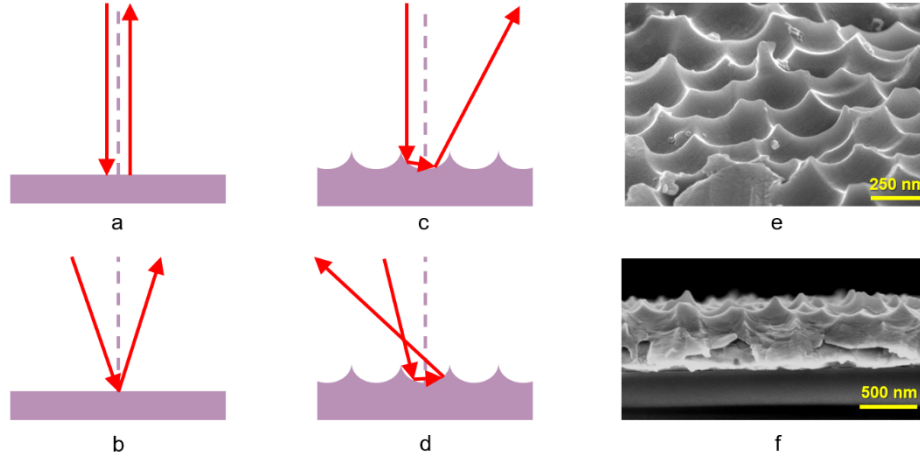


Fig. 1. (a-d) Schematic representation of the concept of increasing absorption in thin and ultra-thin Si solar cells using a T-NC Al film. The effect is achieved by elongating the optical path inside the active silicon layer. Here, (a) and (b) represent cases when light is reflected from plain Al surface; (c) and (d) represent cases when light faces multiple reflections from T-NC Al film surface, resulting in longer optical path. (e) SEM image of the T-NC Al film fabricated by our group [22]. (f) Lateral SEM image of same T-NC Al film. The manufacturing process is fully described in reference 17.

The unit cell of whole solar cell structures including Si active layer and Al planar electrode and electrode with Tsuchime-like nanostructured surface was transferred into the commercial COMSOL simulator [Figure 2(a) and 2(b)] [28]. The incident light source from 400 nm to 1.1 μm was illuminated along the Z-axis and was placed above the Si layer, while transmittance and reflectance monitors are located below Al layer and above the light source, accordingly. The periodic boundary conditions and perfectly matched layers (PML) were applied perpendicular and parallel to the Z-axis, respectively. Figure 2(c) shows 3D view of a single NC unit of T-NC Al film, where D is the diameter of NC. Optical constants of Si and Al were taken from Refs. [29,30].

Numerical optical simulations using finite element method or finite difference time domain includes coherent propagation of light, which may result in wave interference fringes when simulating multilayer structure with an effective thickness of one or more layer equal to or greater than the incoming wavelength. Such wave interference fringes are not observed experimentally in most cases because structure layers are not so perfectly planar. To avoid this issue some scientists applied phase matching and phase elimination approaches [31], but, unfortunately, they do not work for all cases. Here we proposed signal processing filter (Zero Phase Filter and ARIMA) to artificially smooth the undesired fringes of the absorbance spectra (see supplementary information for more details). Fig. S1 shows an example of applying such a filter for planar Si solar cell.

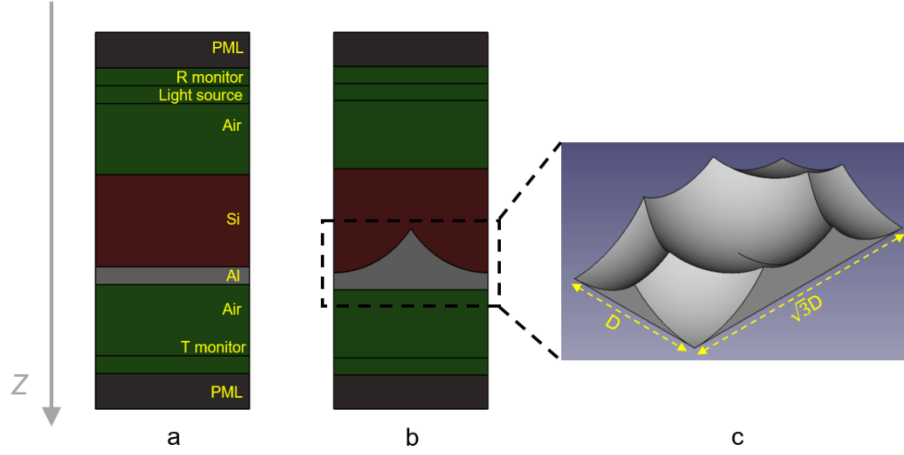


Fig. 2. (a) The unit cell of the simulated PV element including Si active layer and planar Al film, surrounded by air. Light source is placed above the structures layer, while transmittance and reflectance monitors are located below Al layer and above the light source, accordingly. The periodic boundary conditions and PML were applied perpendicular and parallel to the Z-axis. (b) The unit cell of the simulated PV element including Si active layer and T-NC treated Al film, surrounded by air. (c) 3D view of a single NC unit of T-NC Al film. Here, D is the diameter of NC.

3. Results and discussion

3.1 Diameter of nanocraters (d_{NC})

At the first phase of the study, we estimated the influence of the diameter of NCs (d_{NC}) on the level of light absorption at a fixed thickness of the Si active layer. Figure 3 shows the optical absorbance of simulated Si PV cells with planar Al and T-NC modified substrates, where the thickness of the Si active layer was set to 3 μm . The diameter of NCs (d_{NC}) was varied from 0 nm (planar Al) to 800 nm as such diameters can be easily obtained during Al anodizing [32]. Starting from $d_{NC} = 200$ nm, T-NC gains significant advantage over planar Al substrate under Si and reaches a maximum increase of 62.8% at $d_{NC} = 800$ nm as shown in Table S1. When d_{NC} is compatible to incoming wavelength then the light gets reflected repeatedly from the inner side of the NCs, and thus increases the reflective optical path and absorbance in the Si layer. However, the effect from employing T-NC film increases when the thickness of Si active layer decreases. For example, the absorbance of PV cell having 1000 nm thick Si active layer increases by 20% at $\lambda = 590$ nm (from 51.1 to 61.5%), while for PV cells having 2000 and 3000 nm thick Si active layer it increases by only 10.4 and 4.4%, respectively. Remarkable that the use of T-NC Al film effectively eliminates the need for a thicker Si active layer. While the difference between the absorbance in PV cells with 1000, 2000 and 3000 nm thick Si active layer is significant when the Al substrate is planar, it is virtually eliminated with the T-NC Al film where d_{NC} exceeds 200 nm. Indeed, the average absorbance of PV cells with 1000, 2000 and 3000 nm thick Si active layer jumps from 37.2, 41.5 and 44.3%, respectively, to over 60% when $d_{NC} = 800$ nm (at wavelength range $\Delta\lambda = 400\text{--}1100$ nm). In case of PV cell with 1000 nm thick Si active layer this translates into tangible 64.3% boost over the cell with planar Al substrate. T-NC-enhanced PV cells with 2000 and 3000 nm Si active layer gain 46.3 and 41.8% of increase in the average absorbance over solar cells with planar Al. However, the PV cell with 3000 nm thick Si active layer and $d_{NC} = 800$ nm reached the highest average optical absorption

in the given wavelength range ($>62.8\%$ at $\Delta\lambda = 400\text{-}1100\text{ nm}$). Table S1 summarizes the optical absorbances of thin and ultra-thin solar cells with planar Al and T-NC films.

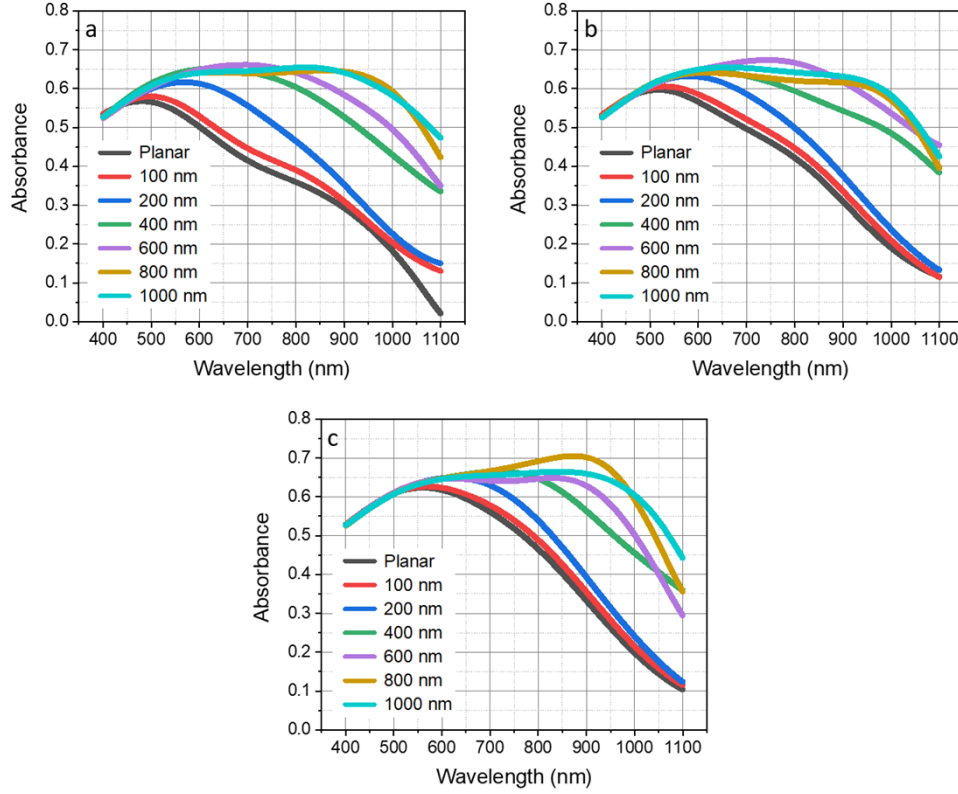


Fig. 3. Optical absorbance of thin and ultra-thin Si PV cells with planar Al and T-NC modified substrates with varied diameter of the NCs ($d_{NC} = 0\text{ nm to }1000\text{ nm}$). The thickness of Si active layer (H_{Si}) equals to (a) 1000 nm, (b) 2000 nm, and (c) 3000 nm, correspondingly. The insets show the diameter of the NCs (d_{NC}): ranging from 0 nm (planar Al) to 1000 nm.

3.2 Thickness of the silicon active layer (H_{Si})

During the second phase of the study, we determined the optimal thickness of the silicon active layer to provide synergy with the T-NC Al film. Fig. 4 shows the optical absorbance of simulated thin and ultra-thin Si PV cells with planar Al and T-NC modified substrates with varied thickness of Si active layer ($h_{Si} = 500\text{ nm to }10\text{ }\mu\text{m}$) and fixed $d_{NC} = 800\text{ nm}$. As can be seen in the Fig. 4, solar cells with planar Al surface beneath Si active layer fail to any of the PV cells with 800 nm or 1 μm NCs. Even the PV cell with planar Al surface having 5 μm Si active layer absorbs less than the thinnest cell with either 800 nm or 1 μm NCs, which has 1/10 of Si active layer thickness [as shown in Figs. 5(c) and 5(d)]. Nevertheless, it is obvious that 100-nm NCs are not enough to be categorically preferred over a solar cell with a sufficiently thick Si active layer [as shown in Fig. 5(b)]. Even though any configuration of Si active layer thickness (from 500 nm to 5 μm) benefits from residing over T-NC film compared to planar Al. Again, the less is the thickness of Si active layer – the more pronounced is the effect of T-NC film. PV cell with 500 nm thick Si active layer gains substantial 30.4% by using T-NC even with d_{NC} of 100 nm. However, the most dramatic difference is observed for a structure with an active layer thickness of 500 nm, when the d_{NC} becomes 1 micron. This configuration of PV

cell nearly doubles the average absorbance in 400-1100 nm wavelength range: absorbance of PV cell with 500 nm thick Si active layer without and with 1 μm NCs is 33.5 and 62.3%, respectively. Hence, forming T-NC Al structure below Si active boosts the absorbance by 85.9% compared to pristine cell. Worth mentioning that T-NC solar cells with the diameter of NCs $d_{\text{NC}} = 800$ nm perform nearly identical to cells with $d_{\text{NC}} = 1$ μm . For the thinnest Si active layer, 500 nm, T-NC film with $d_{\text{NC}} = 800$ nm increases the absorbance by 80.4% compared to pristine PV cell. In case of PV cell with $h_{\text{Si}} = 3$ μm the T-NC film enhances the absorbance by 41.8% compared to the cell without T-NC structure.

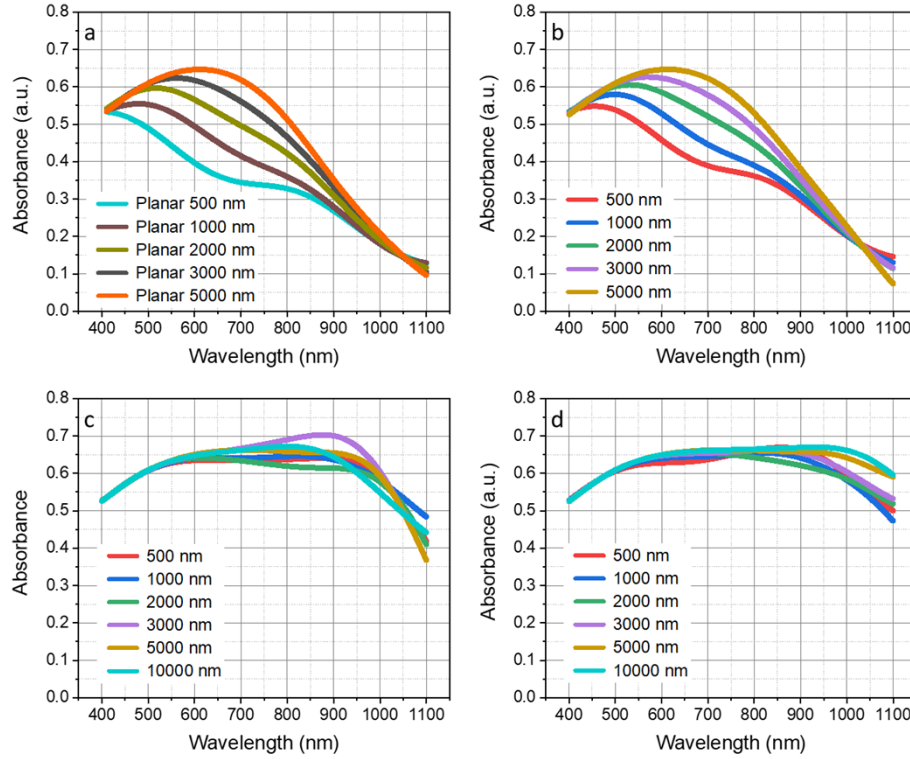


Fig. 4. Optical absorbance of thin and ultra-thin Si PV cells with planar Al and T-NC modified substrates with varied thickness of Si active layer ($H_{\text{Si}} = 500$ nm to 1 μm). The diameter of NCs (d_{NC}) equals to (a) 0 nm, planar, (b) 100 nm, (c) 800 nm, and (d) 1 μm , correspondingly. The insets show thickness of Si active layer (H_{Si}): ranging from 500 nm (planar Al) to 1 μm .

Moreover, the absorption range is directly dependent on the thickness of the silicon, which we will discuss in more detail. Fig. 5 shows the absolute difference in the optical absorbance of simulated thin and ultra-thin Si PV cells with planar Al and T-NC modified substrates with varied thickness of Si active layer ($H_{\text{Si}} = 500$ nm to 10 μm) and $d_{\text{NC}} = 800$ nm. As can be seen in Fig. 5a, peak absolute difference in absorbance is observed at $\lambda = 950$ nm: about 42% gain over the structure with a flat aluminum layer, regardless of the thickness of the silicon active layer. It is interesting to note that the peak absorption difference in Si active element [shown in Fig. 5(b)] migrates from the visible to the near infrared range as the Si thickness increases. The maximum absorbance difference inside the Si active element with a thickness of 500 nm is at $\lambda = 620$ nm, while for structures where $H_{\text{Si}} = 1, 3$, and 10 microns, it is at 680, 850, and 950 nm, respectively. Absolute increase in average absorbance of T-NC with 800 nm NCs over

planar Al is 27.3, 23.2, 17.8 and 2.6% for $H_{Si} = 500$ nm, 1 μm , 3 μm and 10 μm , respectively. These results are summarized in Table S2.

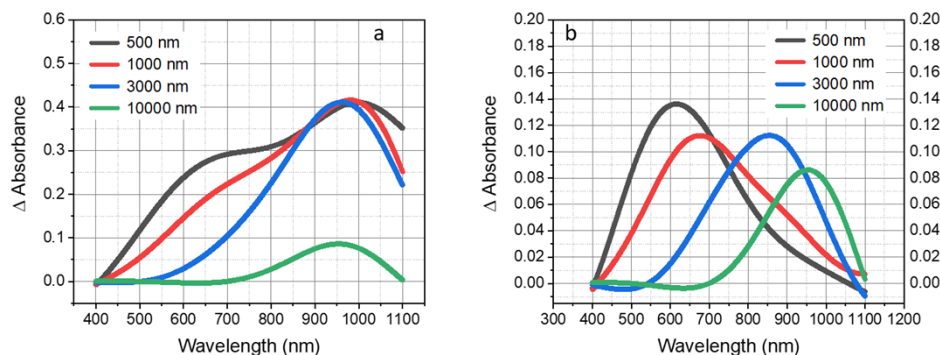


Fig. 5. Optical absorbance difference (absolute) of simulated thin and ultra-thin Si PV cells with planar Al and T-NC modified substrates with varied thickness of Si active layer ($H_{Si} = 500$ nm to 10 μm) and $d_{NC} = 800$ nm: (a) total; (b) in Si active layer.

4. Conclusion

Our study shows that a simple process of electrochemical anodizing of Al with followed by etching-off nanoporous Al_2O_3 allows to significantly improve the absorbance in thin and ultra-thin Si solar cells. T-NC film obtained after such treatment of Al works as reflection multiplier increasing the effective optical light path within the Si active layer. The effect is more pronounced for ultra-thin solar cells with the thickness of Si active layer of 1 μm and below. For instance, employing T-NC Al structure with the diameter of NCs from 800 nm to 1 μm below 500 nm thick Si active layer allows to gain 80.4 to 85.9% more absorbed light compared to the PV cell with T-NC layer, respectively. Absolute increase in average absorbance (ΔA_{ave}) of PV cells with T-NC structure with 800 nm NCs over PV cell with planar Al is 27.3, 23.2, 17.8 and 2.6% when height of Si active layer is 500 nm, 1 μm , 3 μm and 10 μm , respectively. Hence, T-NC film allows to significantly increase the efficiency of thin and ultra-thin Si solar cells – and without the need in utilizing costly top-down processes such as lithography.

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Data availability. Data underlying the results presented in this paper are available from the corresponding authors upon reasonable request.

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