



High efficiency perovskite/crystalline silicon thin-film tandem solar cell from a highly textured MgO [111] buffered glass substrate: a proposal

来自于高度织构氧化镁[111]缓冲玻璃基板的高效率钙钛矿/结晶硅薄膜串列式太阳能电池：一个建议

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Abstract - In this paper a proposal for high efficiency perovskite/crystalline silicon thin-film tandem solar cells is presented. The proposal is based on the successful fabrication of a highly textured MgO [111] thin-film on glass substrate which is a promising indicator for successful growth of highly textured (c-axis aligned) silicon films on which textured perovskite films can then be deposited for high efficiency tandem solar cells.

Keywords – perovskite solar cells, thin-film crystalline silicon solar cells, MgO substrates.

I. INTRODUCTION

In today's solar cell market high power conversion efficiency (PCE) is the most important factor in any photovoltaic technology.ⁱ It was only recently that cost was considered to be the primary factor, and of course it still remains paramount, but PCE may now even be something manufacturers will pay extra for because efficiency lowers the balance of system costs (BOS) and therefore the overall price per Watt installed (currently at \$3.00 USD per Watt without subsidies with the goal of \$1.00).ⁱⁱ Ideally, both factors – PCE and low cost -- can be achieved, and here a proposal to accomplish this by fabricating a perovskite/crystalline silicon thin-film tandem solar cell is presented for the first time. To date, to the best of our knowledge, there have been no proposals or reports on a perovskite/silicon thin-film tandem solar cell device whether amorphous thin-film silicon or crystalline thin-film silicon.

II. BACKGROUND

Recently papers were published by Solar-Tectic LLCⁱⁱⁱ reporting the growth of a highly textured MgO [111] film on soda-lime glass,^{iv} crystalline Al₂O₃ on this same MgO substrate,^v and silicon film growth also on this MgO/soda-lime glass substrate.^{vi} Additionally, a paper was published reporting crystalline silicon film growth on glass without an MgO buffer layer, where the glass was a flexible glass made by a leading glass manufacturer.^{vii} While these papers prove that inexpensive deposition of crystalline silicon thin-films on ordinary glass is possible, their efficiency when converted to solar cells will be necessarily limited by the single absorber layer. This is true even if a single crystal thin-film of silicon without defects could be grown on glass, a long standing objective in the materials research community, which remains to be accomplished.^{viiiix}

Also recently, much attention within the solar cell community has been focused on a relatively new material called perovskites, specifically metal halide organic-inorganic hybrid perovskites. It has been suggested that a tandem solar cell consisting of a silicon sub-cell layer (of wafer thickness) combined with a perovskite layer would provide a solution to solving both the efficiency and cost problems which are keeping solar energy from truly competing with fossil fuels, without subsidies.^x Here we propose an alternative which is potentially less expensive, non-toxic, and equally efficient to a perovskite/silicon wafer tandem solar cell structure of the kind proposed thus far: a tandem solar cell consisting of a thin-film of crystalline silicon on soda-lime glass and perovskite film

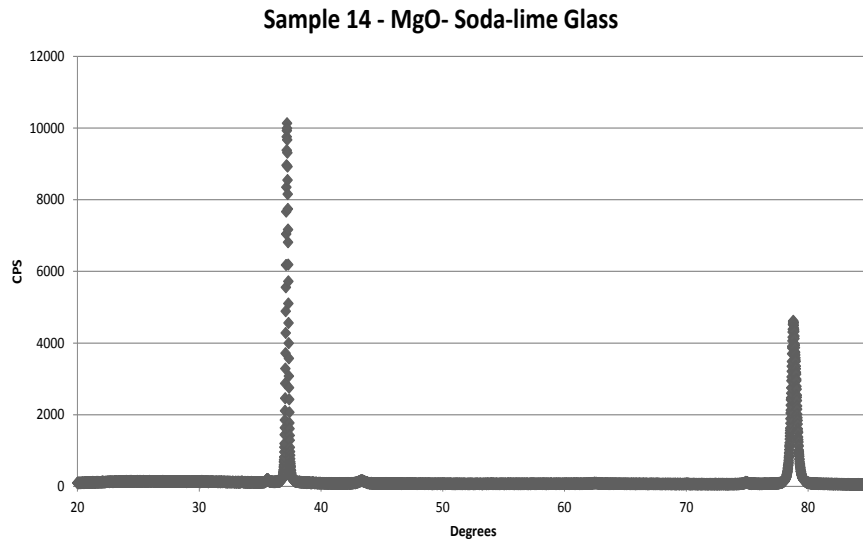


Fig.1, XRD of 7µm MgO [111] on soda-lime glass with associated- [222] peak at ~78 degrees.

grown on this film for a planar heterojunction high efficiency solar cell. Following the recipe (recently patented) of the late Dr. Praveen Chaudhari, a textured silicon film is grown on an MgO [111] buffered soda-lime glass substrate from a silicon-tin eutectic melt and a tandem solar cell is formed by the addition of a layer of another material with better light conversion efficiency, i.e. with a higher bandgap. In the new process disclosed here for the first time by Solar-Tectic LLC (patent pending) the Sn from the Si-Sn eutectic alloy melt which has segregated on the surface of the crystalline silicon film after deposition is then used as the metal in the perovskite structure thus replacing the toxic lead, and facilitating a planar heterojunction device formation for high efficiency, low cost.

In the following pages, data will be provided regarding the first and second steps in this architecture, namely the MgO [111] on glass substrate and the crystalline silicon film grown on it; and in the second half of the paper a proposal regarding the third and final step – the deposition of the perovskite film on the crystalline silicon film, will be described, though many details remain and this is only an introduction to a new process invented which will be developed.

III. STEP 1: TEXTURED MgO [111] FILM ON SODA-LIME GLASS

For the perovskite/crystalline silicon tandem solar cell a textured buffer layer that serves as a diffusion barrier for the soda-lime glass, preventing glass impurities from entering the silicon, as well as a template to induce texture in the silicon film, is preferable as it is known that textured materials have distinct electronic advantages, and this includes perovskites.^{xixii} Although the MgO [111] film on glass is highly transparent (see figure 2), transparency is not a

requirement for the device architecture since light must enter first through the top perovskite layer which has the higher bandgap. It is the combination of the higher bandgap perovskite with the lower bandgap silicon that allows for higher voltage and PCE.

The textured MgO [111] film on glass serves the purpose of the required buffer layer for the tandem device. As already stated a previous paper reported the successful deposition of the MgO [111] film on soda-lime glass, and here we present some additional data regarding this accomplishment which has not been made public until now. Figure 1 shows a second XRD that was performed on the same sample as reported on in the previous paper, but includes the associated parallel [222] peak thereby indicating and/or confirming that a good crystalline formation (better than if there had only been one [111] peak) was obtained. To further investigate the quality of the crystalline MgO [111] film, TEM was performed (see figure 3) clearly evidencing alignment or high texture. A rocking curve was also performed on a highly textured (almost perfectly aligned) sample of 3µm MgO [111] film deposited on a quartz substrate at 800°C using the same invented process^{xiii}. This particular sample was chosen because of the near perfect alignment and thinner film, and it showed a large FWHM (see figures 4 and 5).

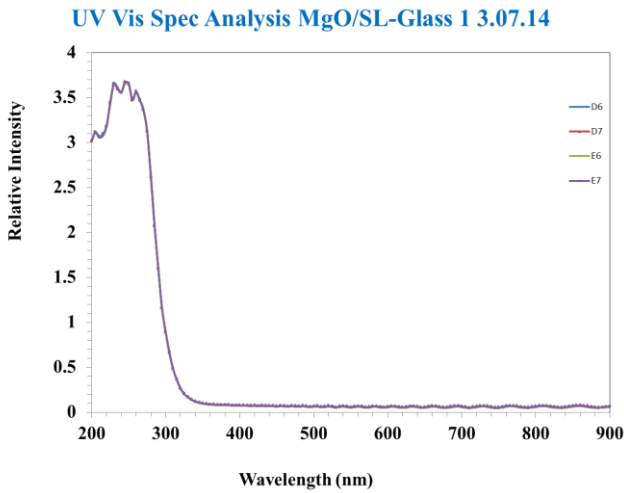


Figure 2: Transparency of MgO on glass. The layer here is 7µm thick and shows high transparency, or ~99%.

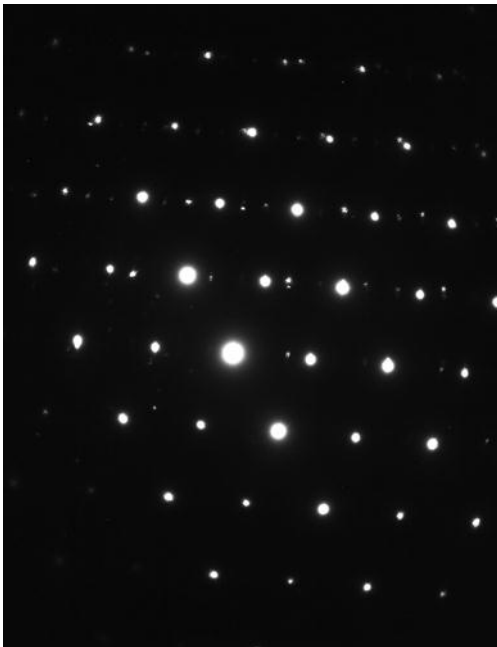


Figure 3: Selected area diffraction from TEM of 7µm MgO [111] film on soda-lime glass showing a c-axis aligned or highly oriented [111] film was grown directly on glass.

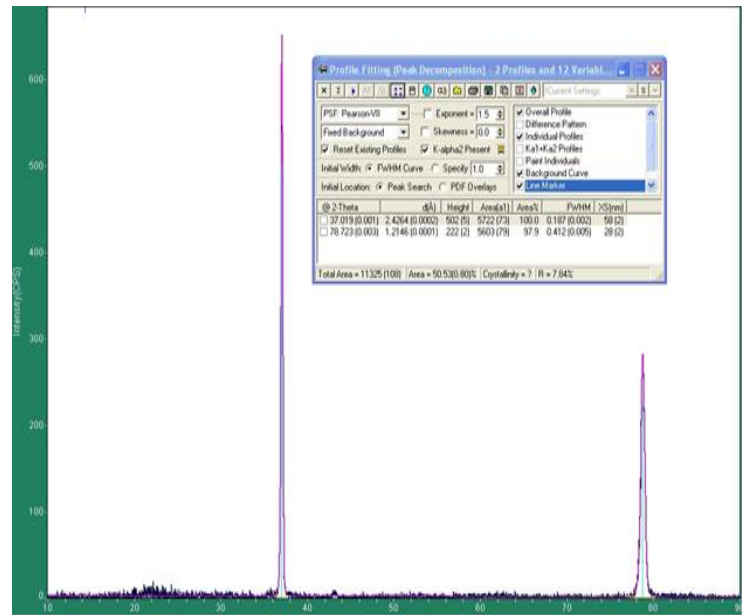


Fig.4: XRD of 3µm of MgO [111] on quartz. No peaks besides the MgO [111] and MgO [222] parallel peak (at ~78 degrees) are visible indicating near perfect out-of-plane or c-axis alignment.

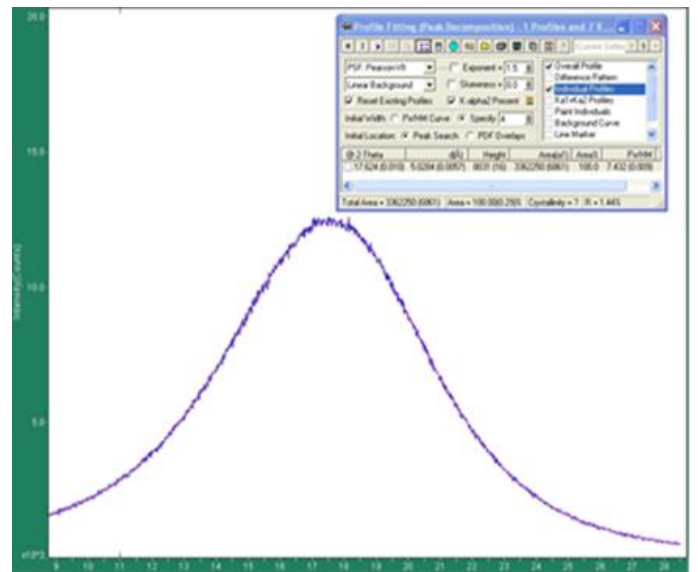


Fig.5, Rocking Curve of 3µm MgO [111] on quartz at 800°C 7.4 degrees.

It was hypothesized that higher temperature may improve the crystalline quality compared to the sample reported on in the previous paper since generally crystallinity of materials improves with higher temperature. It was therefore considered to be a “best case” scenario. The FWHM of the rocking curve indicates the range of tilt angles through which the MgO 111 planes are oriented, which was determined to be 7.4 degrees, a wide range. For comparison, single crystals usually give a FWHM of 0.1 deg. using the same configuration. Additional investigation and a rocking curve of the 7µm MgO [111] film on soda-lime glass samples reported on the previous paper is planned in the near future.

IV. STEP 2: CRYSTALLINE SILICON DEPOSITION ON THE MGO/SODA-LIME GLASS SUBSTRATE.

For deposition of thin-films on a textured substrate from a eutectic alloy melt, the energetics of the interface with the substrate and/or the vacuum or gas must be considered, and it is not only the chemical interaction of the metal or Si with the substrate that is important but also its crystallographic orientation. This is because the surface or interface energy depends upon orientation of the grains.^{xiv} Here the MgO has a strong texture which is [111] oriented, and should therefore be advantageous to silicon [111] film growth. However, another concern is the difference in lattice match between the nucleating film and the substrate which can lead to strain induced energy that is minimized by either inducing defects or not growing uniformly in thickness across the substrate surface. There is a lattice mismatch between MgO and silicon that may be adverse to achieving high quality films. MgO has a lattice constant of 4.212Å and silicon 5.431 Å. These factors determine if silicon is likely to deposit on the substrate (heterogeneous nucleation) or nucleate and form small crystals in the liquid (homogenous nucleation).^{xv}

When considering the texture of the MgO on glass, it is important to determine whether it is uniaxial or biaxial. In other words, whether it has in-plane or out-of-plane texture. In order to determine this, EBSD was performed on the samples, presented here for the first time. It was clear (see figure 6) that the sample was highly textured [111] out-of-plane. It was also evident that there was little or no in-plane texture, which is not surprising since the films were deposited on an amorphous substrate without ion beam assistance. It should be noted that it is well known that MgO films with both in-plane and out-of-plane texture can be grown using ion beam assisted deposition (IBAD). But this process is more expensive than the process we report on here. Moreover, it is not clear that biaxial orientation and epitaxy would be necessary in the process since epitaxial growth with both in-plane and out-of-plane texture is not always necessary and preferentially oriented polycrystalline films are often sufficient.^{xvi} Also, if one looks at this process as a kind of Vapor-liquid-solid thin-film process (VLS-TF) where the film is essentially a nanowire-like, and therefore -- because nanowires are single crystal -- single crystal-like thin-film, which depends more on the out-of-plane crystallographic

orientation of the film for growth (necessarily the case with nanowire growth), one can see how in-plane texture may not be so important.^{xvii} Also, with nanowire growth the [111] direction is crucial. (For details on this VLS-TF process by P. Chaudhari please see McMahon et al, “Textured [111] crystalline silicon thin film growth on flexible glass by e-beam evaporation”). Indeed, it may be for this reason that the film growth reported on in a recent paper where silicon was deposited on the MgO/glass substrate was not oriented as thought. It was polycrystalline but there was no texture. Initial XRD scans seemed to indicate the film was textured, as there was only one silicon peak in the XRD, but TEM later showed the film to be polycrystalline (see figure 7) without oriented silicon. We reason that the lack of orientation could be due to the amount of Al that was used. Too much Al would prohibit nanowire-like film growth and would not lead to oriented film growth, failing to replicate the high texture in the MgO substrate clearly evidenced in the XRD. On the other hand, the process disclosed by P. Chaudhari is probably closer to a modified liquid-phase epitaxy method. And therefore it may be necessary to grow biaxially textured substrates in order to obtain the quality of crystallinity that is the aim of his invention, and indeed is a part of the prescription. However, the plan view TEM of the same sample, presented here for the first time (see figure 8), showed a completely continuous film with crystals of various sized completely covering the MgO substrate and no pinholes. The film was etched to remove the Al from the Si film.

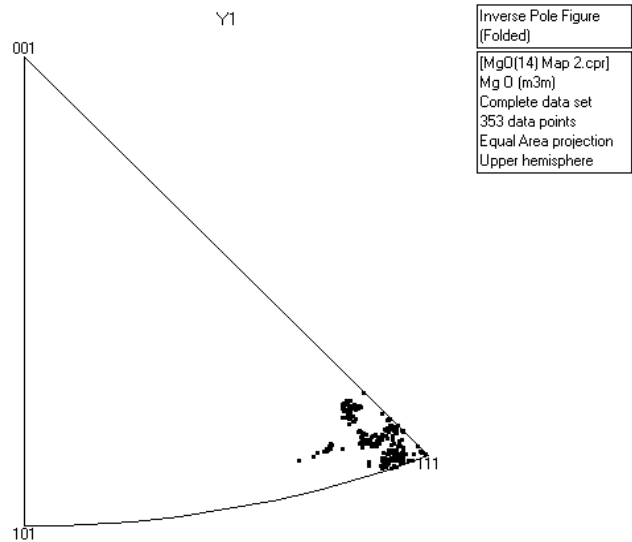


Figure 6: Inverse pole figure from EBSD confirming a highly oriented [111] MgO crystalline film has been grown.

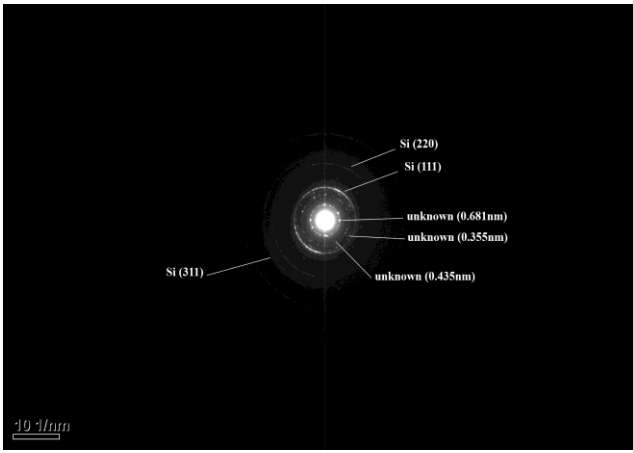


Figure 7: TEM SAED diffraction pattern showing a polycrystalline Si film with perhaps some in-plane orientation but otherwise no texture, despite the strong intensity of the [111]. (Cornell University, CCMR, 2016).^{xviii}

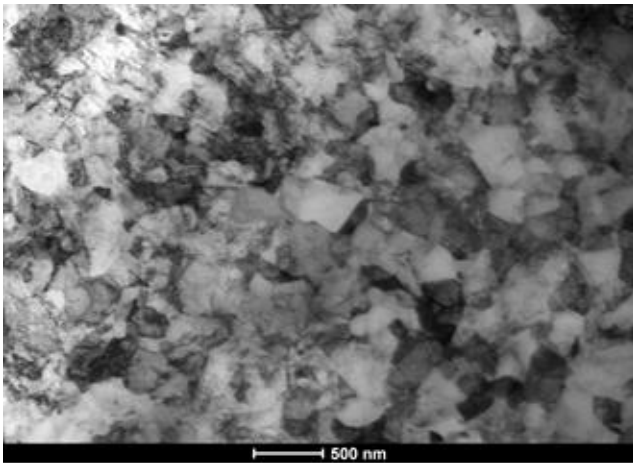


Figure 8: Plan view TEM of same sample as in figure 7, showing a completely continuous Al etched polycrystalline Si film covering the MgO/glass substrate. No pin holes are evident. (Cornell University, CCMR, 2016).^{xix}

V. STEP 3: PEROVSKITE FILM GROWTH ON THE SI FILM.

As already stated, solar cell efficiency is now of primary importance and in fact also lowers overall costs since the BOS costs are reduced with increased PCE. Many now believe that the best way to achieve a highly efficient and cost effective solar cell is by fabricating a tandem (or multi-junction) solar cell, consisting of two layers, where one layer has a bandgap higher than the other, and the layers work in tandem towards higher efficiency light conversion. The question is which materials should be used for this structure. Silicon has a near ideal bandgap for a sub-cell (1.1eV),^{xx} and the more difficult question is which material should be used

for the top layer (with higher bandgap). Interestingly, there is only one example found in the literature of a tandem solar cell consisting of crystalline thin-film silicon layer, and it is with germanium (on the bottom). A proposal and simulation for this was reported with maximum efficiencies of 28% predicted.^{xxi} However, very recently, perovskites have gained much attention due to the rapid increase in efficiency from just 3.8% in 2009 to over ~20% at present. It has been suggested that combining perovskites with silicon for a tandem cell is a viable way to gaining market entry in a silicon wafer dominated industry; and initial tandem device models predict that 36% to 37% efficiencies are achievable for perovskite/silicon tandems.^{xxii} However, these are not thin-film devices since the Si is wafer thickness or fabricated by the same process as Si wafers, namely at very high temperatures.

For a thin-film crystalline Si tandem solar cell, a perovskite top absorber layer is an attractive material because perovskites are easy and cheap to make.^{xxiii} But there are several significant problems that prevent perovskites entry into the market place at this point^{xxiv}. One such problem is the use of Pb (lead) as the main metal, which is toxic. Here we present a solution to this problem by replacing lead with Sn (tin) in a tandem structure. This has actually been demonstrated before in a perovskite solar cell,^{xxv} but not with a thin-film crystalline Si under layer. A crystalline Si under-layer can be deposited at low temperature using eutectics and requires much less material^{xxvi}. It is therefore is less expensive than c-Si wafers. So far the only tandem solar cell reported which used crystalline thin-film Si as an under-layer was with a P3HT (polymer).^{xxvii} Here for the first time we propose a process that uses Sn as the metal catalyst for low temperature crystalline silicon thin-film growth that not only makes use of the highly efficient perovskite material combined with silicon, but in using a thin-film of silicon rather than a wafer, additional cost savings are achieved since less silicon material is needed. Moreover, by using a Sn-Si eutectic alloy (rather than the more common Al-Si used in Aluminum Induced Crystallization) to deposit the silicon at low temperature on the MgO [111] buffered glass, the Sn can be used as the metal (the “B” in ABX₃) in the perovskite material, thus avoiding the otherwise necessary step of etching the Sn from the surface of the silicon layer. The process is simple, and can take place at low temperature – a crucial advantage often cited in connection with perovskite technology too -- using known deposition methods and non-toxic materials and processes throughout. Depositing the silicon film by e-beam evaporation avoids the costly and dirty manufacturing process associated with silicon wafers, both polycrystalline and monocrystalline. Since silicon has a global market share of about 90%, there is obviously a huge market potential for silicon thin-films with comparable quality to c-Si but with additional efficiency (beyond the 25.6% Si wafer current record) due to the tandem perovskite layer.^{xxviii} And this holds true for any material with a higher bandgap such as CZTS which is another contender for a top layer due to its high bandgap as well as closely matching lattice parameter with that of silicon.

As for details of the perovskite material, the family of perovskite materials adopt the chemical formula ABX_3 , where A and B are cations of dissimilar sizes, and X are anions. B is the metal cation and would in this case be Sn instead of Pb.^{xxx} Either a hybrid perovskite (organic-inorganic) or a non-organic perovskite material can be used, though it is known that the non-organic perovskites have bandgaps that are too large for high PCE.^{xxxii} Therefore, a likely material would look like $MASnI_3$. And the deposition process for combining the MA and I_3 with the Sn would be that known in the art and involve common processes, and while this requires much more development and detailed investigation, there is reason to be optimistic because perovskites are versatile and there is room for various configurations.

The versatility of perovskites makes them highly attractive as they can form multidimensional structures pertaining to the same chemical formula through use of different combinations of various components. There is therefore flexibility as to the exact perovskite compound.^{xxxiii} They are known to easily form solar cells layers. “One of the real beauties of perovskite is it almost doesn’t care how you make it. Put a few drops of perovskite precursor, in liquid form, on a piece of aluminum foil on your desk; leave it to evaporate for a day; then plug in a metal probe and you have an operating PV cell — cruddy output, but output nonetheless” as one industry expert put it, perhaps somewhat unscientifically!^{xxxiii}

Despite the seeming ease of fabricating perovskite films described above, there are potential hurdles. The B metal cation sites in hybrid perovskites are occupied by the group IVA metals in a divalent oxidation state (Pb^{2+} , Sn^{2+} , Ge^{2+}). Lead (Pb) has proven the superior constituent compared to tin (Sn), both in terms of performance and stability, and consequently has been the most widely employed of the group IVA metals.^{xxxiv} In the new process presented here, one concern is this issue of Sn stability. Another issue is how exactly the remaining components of the perovskite structure would be combined with the Sn layer on Si. Finally, if there are any impurities in the Sn from the Si (per Si-Sn eutectic melt), how will this affect the perovskite material?

As far as the solar cell structure is concerned, planar heterojunction perovskite/silicon solar cells have already been achieved. And they can be made with an electron transport layer between the silicon and the perovskite, or without. For example, a perovskite/silicon multi-junction solar cell may also be constructed via monolithic integration where a thin film perovskite sub-cell is deposited directly onto the c-Si sub-cell without a recombination layer.^{xxxv} In the case of the process we propose here the Sn on the surface of the silicon (the Sn segregates to the surface of the Si film in the process) film can be oxidized to form a SnO_2 electron transport layer onto which the perovskite film is then deposited. SnO_2 has already been proven to be a viable substitute for the common TiO_2 .^{xxxvi} This SnO_2 film is also likely to have texture if the underlying silicon film does.

While texture would be advantageous, it is not necessary for an efficient perovskite layer. Textured crystalline silicon and perovskite layers in a tandem cell would however be likely to have improved efficiencies as it is known that in order to achieve highly efficient perovskite layers good quality crystallinity is key. That is, crystallization of the perovskites, including the orientation, grain size, and morphology, determines the device performance of perovskite solar cells^{xxxvii}. There has been one report of textured perovskite growth.^{xxxviii} However, at the present time, there are no reports of any perovskite/crystalline or amorphous silicon thin-film results (or patent applications). And this includes both a-Si and crystalline Si from aluminum induced crystallization. Work has however been done on perovskite/thin-film CIGS tandem structures^{xxxix}. Yet a crystalline silicon thin-film would have many advantages over CIGS, not least because CIGS uses selenide (H_2Se) which is highly toxic. Also the manufacturing process is much more complicated than the crystalline silicon thin-film deposition process reported on here which is a straightforward e-beam or CVD (or HW-CVD) deposition method already used widely in the display industry, for example.

III. CONCLUSION

In conclusion, additional characterizations of the MgO [111] film growth on soda-lime glass have been provided, confirming the high texture growth, but also clarifying the texture is limited to out-of-plane or uniaxial as per EBSD results and analysis. Also, further investigation and analysis of the rocking curve (FWHM) should be undertaken and its significance reported. Secondly, further characterizations of the crystalline silicon film grown on this MgO [111] film have been reported and have clarified that in fact the Si film is likely not textured as originally thought, though it is polycrystalline and TEM showed that it is completely continuous even after etching of the Al. Such a film, if deposited using Sn instead of Al at the catalyst, could serve as the bottom layer in a tandem cell where perovskite film could be deposited. Moreover, the Sn segregated on the Si film could be used to form the $MASnI_3$ material thus saving the step of etching the Sn and also perhaps improving the perovskite material. Such a perovskite/crystalline silicon thin-film solar cell could be an even better solution than the current perovskite/c-Si design recommended by a number of groups, since lower temperature processing could be sufficient all around, not just for the perovskite film. Details of this proposal will be reported on as R&D commences in the near future.

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