



## UvA-DARE (Digital Academic Repository)

### Understanding losses in halide perovskite thin films

Adhyaksa, G.W.P.

**Publication date**

2018

**Document Version**

Other version

**License**

Other

[Link to publication](#)

**Citation for published version (APA):**

Adhyaksa, G. W. P. (2018). *Understanding losses in halide perovskite thin films*.

**General rights**

It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content license (like Creative Commons).

**Disclaimer/Complaints regulations**

If you believe that digital publication of certain material infringes any of your rights or (privacy) interests, please let the Library know, stating your reasons. In case of a legitimate complaint, the Library will make the material inaccessible and/or remove it from the website. Please Ask the Library: <https://uba.uva.nl/en/contact>, or a letter to: Library of the University of Amsterdam, Secretariat, Singel 425, 1012 WP Amsterdam, The Netherlands. You will be contacted as soon as possible.

---

## Summary

Semiconductors have been an integral element in our 21st century society. We find semiconductors at the heart of every electronic device such as microprocessor chip, transistor, as well as light-emitting diode (LED). Although, the seminal theory of semi-conductors was proposed by A.H. Wilson in 1931 after the birth of quantum mechanics, the main impetus of the fast development of semiconductors for devices, perhaps only came after several ground breaking experiments in transistors by W.B. Shockley, J. Bardeen, and W.H. Brattain who received the Nobel prize in 1956. Rapid progress in techniques to deposit and study semiconductor interfaces in the 1980s helped close the gap between our knowledge in solid-state physics semiconductor and light science, most recently exemplified by the LED. An energy-efficient LED can reduce electricity consumption by about 75%. High-efficiency LEDs are enabled by high quality GaN semiconductor films, firstly introduced by I. Akasaki, H. Amano, and S. Nakamura who received the Nobel prize in 2014. They introduced a growth technique to make uniform nucleation, non-selective growth, and good coalescence that leads to reduce defect density in GaN.

On the other hand, advances in thin-film semiconductor technology will also find applications in highly efficient and low-cost photovoltaics. Hybrid-halide perovskites have emerged over the unprecedented timeframe over the last 6 years as a promising class of materials for such applications. Most notably, their solar cells have achieved power conversion efficiencies above 20 % in the laboratory, even though many fundamental questions still remain unanswered. Therefore for halide perovskite thin-films to have an impact beyond the laboratory requires a systematic understanding and eliminating sources of losses.

In Chapter 1 we outline the methodology used in this thesis to improve our understanding, while pointing the way forward to eliminate the losses and ultimately design even better devices. Technically the development cycle involves controlled and clean experimental systems along with careful measurements and full optoelectronic modelling/theory to verify, and quantify the losses in halide perovskite thin-films. By quantifying and considering all of the losses, the ultimate goal is directed at designing a novel architecture device that potentially exceeds the performance and relaxes the limitations of traditional solar cell configurations.

In Chapter 2 we report experimental values of carrier diffusion lengths in halide perovskite thin films and describe how processing, composition, aging and surface passivation affect the results. Considering the many possibilities for all three ions in the halide perovskite structure, hundreds of distinct compositions have already been reported. Such compositional changes can alter the carrier diffusion length - a key parameter for solar cell performance. Given the large compositional and processing parameter space, a rapid and simple technique for directly measuring diffusion length is needed. Here we use a laser grating technique to screen the diffusion length in perovskite materials. First, in pure  $\text{CH}_3\text{NH}_3\text{PbI}_3$  we observe the diffusion length is largely dependent on the controlled processing conditions. Next, we partially replace iodide (I) with bromide (Br) and show that surprisingly, the diffusion length increases after aging for 1 month in air. Finally, we use a 4-nm  $\text{Al}_2\text{O}_3$  layer (atomic layer deposition) to passivate the surface of  $\text{CH}_3\text{NH}_3\text{PbBr}_3$ , leading to a remarkable increase in diffusion length from 201 nm to 532 nm. The correlation that we have established between materials processing and diffusion length offers guidance in how to improve materials for devices.

In Chapter 3 we describe the use of electron backscattering diffraction (EBSD) to properly determine the grain boundary locations in halide perovskite thin-films. Grain boundaries play a key role in the performance of thin-film optoelectronic devices and yet their effect in halide perovskite materials is still not understood. The biggest factor limiting progress is the inability to identify grain boundaries; the gold standard - EBSD - destroys halide perovskite thin films. Non-crystallographic techniques commonly misidentify grain boundaries, leading to conflicting literature reports about their influence. Here we solve this problem using a solid-state EBSD detector with 6,000 times higher sensitivity than the traditional phosphor screen and camera. We used the crystal misorientation data set from the EBSD to model the characteristics of grain boundary interface energy in  $\text{CH}_3\text{NH}_3\text{PbBr}_3$  thin-films, and show that the halide perovskite grains do not exhibit twinning. In addition, we find a peculiar case, where the grain boundary very likely consists of amorphous halide perovskites.

In Chapter 4 we attempt to correlate the true grain size with photoluminescence lifetime, carrier diffusion length, and mobility in halide perovskites. We find that the grain boundaries are not benign as is often claimed, but have a recombination velocity of 1670 cm/s, comparable to that of crystalline silicon. However, as with silicon, amorphous perovskite can passivate crystalline boundaries, leading to brighter photoluminescence and longer carrier lifetime without reducing diffusion length. This variable grain boundary character explains the mysteriously long lifetime and record efficiency achieved in small grain halide perovskite thin films, while pointing the way forward to even better performance.

In Chapter 5 we use our understanding of the losses to design a novel device architecture - nanoscale back-contact perovskite solar cell to improve tandem efficiency. Using coupled optical-electrical modelling, we optimize this architecture for a planar perovskite-silicon tandem, highlighting the roles of nanoscale contacts to reduce the required perovskite electronic quality such as minority carrier diffusion length. We discuss the advances of our design over the traditionally used two- (2-T) and four-

## Summary

---

terminal tandem (4-T), and point the way towards further improvements enabled by our design such as surface texturing, surface passivation and photoluminescence outcoupling.

---

## Samenvatting

Halfgeleiders zijn onmisbaar in onze 21e-eeuwse maatschappij. We kunnen halfgeleiders vinden in de kern van elk elektronisch apparaat, zoals de microprocessor chip, transistors en licht-emitterende diodes (LED). Hoewel de rudimentaire theorie van de halfgeleiders al was voorgesteld in 1931 door A.H. Wilson na de geboorte van de kwantummechanica, kwam de voornaamste impuls voor de snelle ontwikkeling van halfgeleiders voor apparaten kwam pas na verscheidene baanbrekende experimenten met transistors door W.B. Shockley, J. Bardeen, en W.H. Battain, die daarvoor in 1956 de Nobelprijs hebben ontvangen. Razendsnelle technologische ontwikkelingen tussen 1980 en 1990 om halfgeleiders te deponeren en de grensvlakken te bestuderen hielpen om de kloof te dichten tussen onze kennis van vaste-stof halfgeleiderfysica en de wetenschap van het licht ("fotonica"). Een goed voorbeeld hiervan is de LED. Een energiezuinige LED kan het elektriciteitsverbruik met ongeveer 75 % verminderen. LEDs met een hoge efficiëntie zijn mogelijk door het gebruik van een dunne laag gallium nitride (GaN) van hoge kwaliteit. Deze zijn voor het eerst geïntroduceerd door I. Akasaki, H. Amano, en S. Nakamura, die daarvoor in 2014 de Nobelprijs in ontvangst namen. Deze wetenschappers introduceerden een kristalgroeitechniek waarmee uniforme nucleatie, non-selectieve groei en goede samensmelting mogelijk werden wat vervolgens leidde tot een vermindering in de defectendichtheid in GaN.

Anderzijds zal de vooruitgang in dunne-film halfgeleidertechnologie ook toepassingen vinden in fotonica met hoge efficiëntie en lage kosten. Hybride-halide perovskieten zijn een categorie materialen welke in een uitzonderlijk korte tijdsperiode van 6 jaar een enorme opkomst hebben gemaakt voor dergelijke toepassingen. Hybride-halide perovskiet zonnecellen hebben een elektriciteitsopwekkingefficiëntie van 20 % bereikt in het laboratorium, ondanks dat vele fundamentele vragen nog steeds onbeantwoord zijn. Om dunne-film halide perovskite ook buiten het laboratorium een impact te laten hebben is het nodig om systematisch ons begrip te vergroten en oorzaken van energieverlies te elimineren.

In hoofdstuk 1 wordt de onderzoeksmethodologie van deze thesis geschetst. Hierin verbeteren wij onze kennis en wijzen wij de weg naar het elimineren van verliezen om uiteindelijk betere apparaten te ontwerpen. Vanuit een technisch oogpunt heeft het

ontwikkelingsproces gecontroleerde en schone experimentele systemen nodig, gecombineerd met zorgvuldige metingen en volledige optisch-elektronische modellering en bijbehorende theorie om de verliezen in halide-perovskiet dunne films te kunnen verifiëren en kwantificeren. Door alle verliezen te kwantificeren en doorgroondig te onderzoeken wordt het uiteindelijke doel gestuurd richting het ontwerpen van een apparaat met een nieuwe architectuur, die mogelijk de prestaties en beperkingen overtreft van traditionele zonnecelconfiguraties.

In Hoofdstuk 2 rapporteren wij experimentele waarden van energiedragerdiffusielengtes in halide perovskiet dunne films en beschrijven we hoe de verwerking, samenstelling, veroudering en oppervlaktepassivering invloed heeft op de resultaten. Omdat er veel verschillende samenstellingsmogelijkheden zijn voor de drie ionen in de halogenide perovskietstructuur zijn er al honderden verschillende samenstellingen gerapporteerd. Dergelijke wijzigingen in de samenstelling kunnen de energiedragerdiffusielengte veranderen - dit is een kernparameter voor zonnecelprestaties. Aangezien er vele mogelijkheden zijn voor de samenstelling en de verwerking is een snelle en eenvoudige techniek voor het direct meten van de diffusielengte nodig. Wij hebben gebruik gemaakt van een laser-traliewerk techniek om de diffusielengtes van perovskieten door te lichten. Ten eerste, in zuiver  $\text{CH}_3\text{NH}_3\text{PbI}_3$  observeren we dat de diffusielengte grotendeels afhankelijk is van de vervaardigingsomstandigheden. Vervolgens vervangen we gedeeltelijk de jodide (I) ionen met bromide (Br). Verrassend genoeg observeren wij dat de diffusielengte toeneemt na blootstelling aan de lucht gedurende één maand. Ten slotte deponeren wij een 4-nm  $\text{Al}_2\text{O}_3$ -laag (door middel van atomaire laag depositie; ALD) op het oppervlak van  $\text{CH}_3\text{NH}_3\text{PbBr}_3$  om deze te passiveren. Dit zorgde voor een opmerkelijke toename in diffusielengte van 201 nm naar 532 nm. De correlatie die we hebben vastgesteld tussen materiaalvervaardiging en diffusielengte biedt richtlijnen voor toepassing in apparaten.

In hoofdstuk 3 beschrijven we het gebruik van elektron-terugverstrooiingdiffractie (EBSD) om de korrelgrenslocaties in halide perovskiet dunne films nauwkeurig te bepalen. Korrelgrenzen hebben een grote invloed op de prestaties van dunne film opto-elektronische apparaten en toch wordt hun effect in halide perovskietmaterialen nog steeds niet begrepen. De grootste voortuitgangsbeperkende factor is het onvermogen om korrelgrenzen te identificeren; de gouden standaard - EBSD - vernietigt halogeen perovskiet dunne films. Niet-kristallografische technieken lijden onder het vaak misidentificeren van korrelgrenzen, wat leidt tot tegenstrijdige experimentele data in de literatuur over de invloed van deze korrelgrenzen. Dit probleem lossen wij op met behulp van een vaste-stof EBSD-detector met een 6000 maal hogere gevoeligheid dan een traditioneel fosforschermbild met een camera. Wij maken gebruik van de kristalmisoriëntatiedataset van de EBSD om de kenmerken van de korrelgrensvlak-energie in  $\text{CH}_3\text{NH}_3\text{PbBr}_3$  dunne films te modelleren. Deze gegevens laten zien dat halogenide perovskiet korrels geen tweelingeffecten vertonen. Verder vonden wij een bijzondere situatie waarbij de korrelgrens van dit geval zeer waarschijnlijk bestaat uit amorfe halogenide perovskieten.

In Hoofdstuk 4 proberen we de ware korrelgrootte te correleren met fotoluminescentielevensduur, de energiedragerdiffusielengte en -mobiliteit in halogenide perovskieten.

Verder ontdekken we dat de korrelgrenzen niet onschuldig zijn zoals vaak wordt beweerd. Deze korrelgrenzen hebben namelijk een recombinatie snelheid van 1670 cm/s. Dit is vergelijkbaar met de recombinatiesnelheid van kristallijn silicium. Echter kan amorf perovskiet, net als silicium, kristallijne grenzen passiveren, wat leidt tot helderdere fotoluminescentie en een langere levensduur van de ladingsdragers zonder de diffusielengte te verminderen. Dit variabele karakter van de korrelgrenzen verklaart de verrassend lange levensduur en recordefficiëntie welke bereikt wordt in klein-korrelige halide perovskiet dunne films. Verder wijst het ook de weg voorwaarts naar nog betere prestaties.

In hoofdstuk 5 gebruiken we ons begrip van de verliezen om een apparaat met een nieuwe architectuur te ontwerpen: perovskietzonnecellen met een nanoschaal contact aan de achterkant om zo de tandemefficiëntie te verbeteren. Met behulp van gekoppelde optisch-elektrische modellering, optimaliseren we deze architectuur voor een vlakke perovskiet-silicium tandem. Hierbij leggen we de nadruk op de rol van de nanoschaal contacten om de vereiste elektronische kwaliteit van perovskiet te verminderen, zoals de diffusielengte van minderheidsenergiedragers. Ook wordt de vooruitgang van ons ontwerp ten opzichte van de traditioneel gebruikte twee- (2-T) en vier-contacten (4-T) tandem zonnecellen besproken en wijzen we de weg naar verdere verbeteringen die mogelijk zijn met ons ontwerp zoals oppervlaktetextureren, oppervlaktepassivering en fotoluminescentieuitkoppelingsefficiëntie.

---

## Ringkasan

Semikonduktor sudah menjadi kesatuan utuh dalam kehidupan manusia abad-21. Kita menemukan semikonduktor dalam setiap komponen alat elektronik seperti mikroprosessor, transistor, termasuk didalamnya lampu LED. Dasar teori lahirnya semikonduktor pertama kali diajukan oleh A.H. Wilson pada tahun 1931 setelah lahirnya mekanika kuantum, meski dari itu penerapan semikonduktor itu sendiri baru berkembang pesat setelah beberapa eksperimen terobosan pada transistor yang dilakukan oleh W.B. Shockley, J. Bardeen, dan W.H. Brattain (penerima hadiah Nobel 1956). Perkembangan pesat berikutnya terjadi ditahun 1980an dimana pengetahuan tentang cara memproses, dan pengkarakterisasian semiconductor telah membantu kita dalam menjembatani dua bidang penting yaitu dasar fisika struktur dan ilmu cahaya. Contoh paling penting yaitu lahirnya lampu LED yang mampu mengurangi konsumsi listrik sebesar 75 %. Faktor keberhasilan utamanya terletak pada penggunaan GaN semikonduktor material berkualitas tinggi, yang pertama kali di pelopori oleh I. Akasaki, H. Amano, dan S. Nakamura (penerima hadiah Nobel 2014). Dimana mereka berhasil memperkenalkan cara membuat lapisan tipis GaN dengan proses pembibitan atom secara merata, tidak terlokalisasi, dan membentuk kesatuan kristal yang mampu mengurangi jumlah cacat material.

Pada sisi lain, kemajuan dalam teknologi lapisan tipis semikonduktor sudah terbukti untuk menghasilkan sel surya yang sangat efisien namun murah. Hibrida-halogen perovskait adalah jenis mineral baru, yang dalam waktu singkat 6 tahun sudah membuktikan diri memenuhi kriteria diatas. Terbukti dengan efisiensi sel surya yang dihasilkan sudah diatas 20 % di skala laboratorium, meskipun banyak dasar-dasar sains yang belum diketahui mengapa material ini begitu efisien. Maka dari itu agar Hibrida-halogen perovskait ini bisa berdampak lebih luas dibutuhkan pemahaman sains mendasar yang sistematis, serta cara untuk mengurangi jumlah cacat material yang dimilikinya.

Pada Bab 1 kami menjelaskan metodologi yang digunakan di disertasi ini untuk memahami sains mendasar tersebut. Sekaligus memberikan saran penting bagaimana mengurangi jumlah cacat material, yang pada akhirnya kami mengajukan terobosan desain baru untuk merancang solar cell yang lebih efisien. Secara teknis ini mencakup pembangunan ide bagaimana membuat sistem penelitian yang bersih, terpercaya dan teliti, yang kemudian menggabungkannya dengan teknik permodelan serta teori yang



utuh dengan tujuan untuk memverifikasi, mengidentifikasi, dan menghitung sumber sumber cacat dari hibrida-halogen perovskait. Dengan semua cara ini, kita pada akhirnya akan bisa membuat teknologi sel surya baru yang melebihi, dan mengurai keterbatasan dari teknologi yang ada sekarang.

Pada Bab 2 kami melaporkan hasil pengukuran seberapa jauh muatan listrik bisa mengalir di hibrida-halogen perovskait ini, dan menjelaskan bagaimana teknik pemrosesan, komposisi, penebaran, dan pelapisan permukaan mempengaruhi persepsi kita akan hasil penelitian. Walaupun hanya memiliki tiga jenis ion dalam struktur materialnya, namun kombinasi dari ketiganya mampu menghasilkan ratusan jenis kombinasi struktur yang berbeda-beda. Semua kombinasi ini akan mempengaruhi hasil pengukuran tersebut, terutama panjang muatan listrik yang mampu mengalir karena ini merupakan komponen penting untuk aplikasi sel surya. Kita mengajukan teknik pengukuran yang mampu mengukur parameter panjang muatan listrik ini dengan cara cepat dan tepat. Teknik ini menggunakan cahaya laser untuk membuat pola interferensi permukaan. Pertama, kita menemukan bahwa pada jenis material perovskait murni pun,  $\text{CH}_3\text{NH}_3\text{PbI}_3$ , pengukuran panjang muatan listrik bisa berbeda beda tergantung bagaimana kita membuatnya. Kemudian kita secara bertahap mengganti iodida (I) dengan bromida (Br) dan hasilnya panjang muatan listrik bisa bertambah setelah didiamkan selama 1 bulan di udara. Akhirnya, kami menggunakan lapisan tipis  $\text{Al}_2\text{O}_3$  sebesar 4-nm (pembuatan dengan cara atom per atom) untuk mengisolasi permukaan, dan hasilnya panjang muatan listrik bertambah dari 201 nm menuju 532 nm. Korelasi penting ini akan menjadi panduan penting bagaimana meningkatkan kualitas material untuk aplikasi yang dibutuhkan.

Pada Bab 3 kami menjelaskan penggunaan teknik elektron backskatering difraksi (EBSD) untuk mengidentifikasi dengan benar keberadaan dan lokasi dari kristal batas dari lapisan tipis hibrida halida perovskite. Pengetahuan akan kristal batas ini menjadi begitu penting mengingat perannya di hibrida halida perovskite sangatlah ambigu. Faktor penghalang utamanya yaitu ketidakjelasan akan konsensus yang digunakan dalam menentukan lokasi kristal batas ini. Konsensus standard seperti EBSD tidak mampu membantu karena biasanya langsung menghancurkan material ini ketika diterapkan. Kami menyelesaikan masalah ini dengan menggunakan detektor padat jenis baru yang memiliki sensitifitas 6000 kali detektor EBSD yang ada sekarang seperti kamera, dan jendela klasik fosforus. Kami menggunakan informasi dari misorientasi kristal untuk memodelkan dan mengkarakterisasi jenis kristal batas tersebut, dan kemudian total rugi-rugi energi yang ada di kristal batas tersebut. Kami menemukan kristal batas di  $\text{CH}_3\text{NH}_3\text{PbBr}_3$  tidak memiliki kristal kembar seperti material pada umumnya, namun kristal batasnya memiliki karakteristik yang sangat tidak jelas sekaligus penting.

Pada Bab 4 kami mencoba untuk mencari hubungan antara ukuran kristal yang asli dari informasi EBSD dengan waktu hidup emisi cahaya, panjang, dan mobilitas dari muatan listrik yang dihasilkannya. Kami berkesimpulan bahwa kristal batas di hibrida-halida perovskait tidaklah bermanfaat, sebagaimana diklaim sebelumnya, namun memiliki kecepatan rugi rugi sebesar 1670 cm/s, yang hampir setara dengan rugi-rugi di tradisional material seperti silikon. Namun, sebagaimana silikon keberadaan

kristal batas yang sangat tidak jelas ini mampu mengisolasi material, dan menghasilkan emisi cahaya yang lebih lama, dan mobilitas muatan listrik yang lebih jauh. Hasil dari penelitian ini menjawab semua misteri yang pernah ada sejauh ini tentang emisi cahaya yang begitu lama yang kaitannya dengan sel surya berefisiensi tinggi dari material ini, walaupun ukuran kristalnya begitu kecil. Penelitian ini memberi panduan jelas bagaimana membuat material ini lebih baik lagi.

Pada Bab 5 kami menggunakan pengetahuan yang telah kami dapat sejauh ini untuk mendesain jenis arsitektur baru untuk sel surya. Arsitektur ini kami namakan sel surya dengan semua colokan dibelakan berskala nano. Kami menggabungkan teori optik dan elektronik untuk memodelkan, dan mengoptimalkan arsitektur ini pada jenis sel surya perovskait-silikon berpermukaan datar, dengan menekankan pada peran colokan belakang berskala nano. Peran colokan nano ini adalah untuk mengurangi kriteria penggunaan material murni. Terlebih desain kami mampu menghilangkan halangan yang ada pada desain yang ada sekarang (2-colokan atau 4-colokan). Faktor penting yang membuat desain ini menarik adalah permukaan atasnya yang kosong bisa dimanfaatkan untuk desain permukaan struktur penangkap cahaya, strategi pengisolasian permukaan, dan pelepasan emisi cahaya keluar permukaan.

---

## Acknowledgements

During my sojourn in Holland, AMOLF has been the place providing helpful scientific exercises to raise myself to another level (you call it a PhD program). This section is dedicated to those who have helped me along that way.

My first thanks go to my advisor Erik Garnett. At the beginning I had little idea of what the group was really about, and I faced a hard time understanding his scientific approach. Nevertheless, he was still patient enough, both scientifically and personally, to bring me into sync. I have enjoyed the way he challenges me with problems: although to me it sounds noisy at first, it becomes clear at the end. I now realize after thinking about them very carefully, that in some cases the challenges were very visionary. The most important skill I learned from him is how to identify goals that are clear, important, and worth trying, and also how to demonstrate them in a convincing way. This is an important aspect that I will always carry in my future career. Although I did not finish all the projects he gave me, he is still kind enough to allow me to graduate. Therefore, I am truly honored.

Albert Polman, he is not only my PhD (co-)promotor, but also an inspiring leader connecting me with networks around him. Perhaps I only spent 1 % of my time during my PhD with him, but I find that this 1 % time has turned out to be productive experiences. One example is his sudden notification asking me to give a talk on the "COMPASS" project meeting. I found it overwhelming but at the end very useful sharpening my awareness of more details regarding surface passivation in Si PVs that eventually brought me in touch with some leading figures on this field in the Netherlands. I also thank for his help rectifying my Dutch summary, as well as what he provides for my future career.

I thank Prof. Rene Janssen, Prof. Jan Anton Koster, Prof Mark Golden, Prof. Wim Sinke, and Dr. Rene Williams for their time reading through my thesis draft carefully, providing comments for improvements, and serving as my committee members.

Members of the Nanoscale solar cell group: Jenny, Harshal, Biplab, Marc, Sven, Hongyu, Niels, Julia, and Forrest, I enjoyed your daily companion, and I am certainly going to miss this routine. Also the past members of the group, Sarah, Eric, Jia, Lai-Hung, Parisa, Haralds, Teo, Tade, and Shanti, thanks for sharing with me your similar

## Acknowledgements

---

mutual research interests. In particular to Sarah Brittman, and Eric Johlin, my extensive discussions with you were always very stimulating and helpful.

All AMOLF nanophotonics group members, especially the group leaders (Femius, Ewold, Bruno, Esther, and recently Said) who provided very useful and relevant questions to me over the last 4 years during colloquia, and poster seasons. In particular to Bruno Ehrler who consistently served as internal reviewer for my published works. Also members who are kind enough to provide direct help for my research: Sophie Meuret, and Nick Schilder for CL experiments, Benjamin Daiber, and Stefan Tabernig for troubleshooting bugs during my wide-field experiment, Jumin Lee for all detailed tips and tricks for material evaporation, Mark Aarts for AFM testing, Kevin Cognee for translating RCWA codes, Juha Muhonen for quite a discussion on measuring spin-diffusion. I have been pleased to share my research experience with Moritz Futscher, Lucie McGovern, and Loreta Muscarella on controlling perovskite grain sizes, David van der Flier for preparing samples for polariton lasing, and Jose Ocana for the CVD system. I was also supported in a friendly manner by amazing people from the nanoLab: Hans Zeijlemaker, Andries Lof, Dimitry Lamers, and Bob Drent on many projects. I also enjoyed the collaboration work with almost everyone in Wim Noorduyn's group: Hans, Lukas, Iarik, and my superb paranymph Arno. In particular the work on converting their minerals into halide-perovskites using the CVD system - I am glad that it works, and has become a standard routine. I should thank many current (past) members in Huib Bakker's group who were being helpful during my first year adaptation, especially Simona, Liyuan, Konrad, Biplab, Niklas, Artem, and also Johannes, Reinout for helping me with the XPS measurement.

I would not be able to stay focused on my research without the help from our supporting staff. The HR team: Wouter, Roos, Valeska, and Reshma for securing all of my legal documents for working in Holland. The communication department: Grace, Petra, and Floortje for all of your encouragement to have fun during AMOLF events. The Inkoop department: Andre, Tatiana, and Angela for fast helps delivering/receiving my important samples. The financial team: Arnelli, and Sebastiaan for always being patient to guide me over and over again for through many mistakes I made filling out forms for my business trips. The Mechanical design team: Illiya, Henk-Jan, Marnix, and Ricardo, thanks for your help with all details designing in my setup and other drawings. The ICT team: Carl, Rutger, and my fav-friendly person Wiebe for all helps troubleshooting my PC/network/software licences. The electronic department: I thank Jan Zomerdijk, Luc Blom, and Erik Clay for helping me to develop the SSPG setup. From the mechanical workshop department: I thank Jan van der Linden, Ricardo, Niels, Mark, and the specially friendly Tom Brouwer. The facility team: Clyde, and Karim for ensuring my office is comfortable enough all the time. Special thank to Silvia de Jong who amazingly managed to find every text book I requested to be available.

I also thank people outside of AMOLF, within the Netherlands and abroad, with whom I collaborated. From TU Eindhoven: Pim Veldhuizen, Yinghuan Kuang, and Dibya Koushik for tremendous help during my early years of the PhD. Pim introduced me to the SSPG world for the first time, something that fascinated me immediately and ever since. Yinghuan introduced me to the ALD method and magic recipes on it.

Dibya helped me with fabricating the first solar cell device. From TU Delft: Ibadillah Digdaya, Bartek Trzesniewski, and Wilson Smith who consistently made my Friday time extremely busy fabricating MIS samples for their water-splitting projects. I noticed that Digdaya has truly grown as scientist, and thanks for asking me a bunch of critical questions that surely deepened my understanding of silicon interfaces. From ECN: Piero Spinelli, and Paula Bronsveld who provide help and brainstorming ideas on fabricating the nanoscale back-contact tandem devices. From Maastricht University: Joel Keelor, and Sean Ellis for their help with mass-spec analysis of my samples, and I am happy that our frustration on the unexpected bad results turned out to be a supporting point in our other findings. From UC San Diego: Xueying Li, and David Fenning who provided helps with the nano-XRD on my samples, and confirmed the amorphous nature of grain boundaries. David thanks for your constructive questions on my analysis that I think hardly anyone could catch. From Washington University: Sarthak Jariwala, and David Ginger for an exciting proposal project on measuring carrier diffusion across different grain orientations. From Georgia Tech (USA) together with CSIRO (Australia): Giovanni DeLuca, Qicheng, Anthony Chesman, and Udo Bach on the theory project of microscale/hexagonal back-contact perovskite solar cells. I thank you for all of these encouraging and positive environments allowing me to really enrich my (limited) horizons across many disciplines.

My presence at AMOLF would not be possible without personal encouragement, and recommendations from my previous scientific mentors. In that regard, I thank Brian Yulianto, Jeungku Kang, Jung-Yong Lee, and Omar Yaghi who have inspired me in their own ways.

I thank many Indonesian friends in Holland for their, somehow good, distraction which helped me to relax a little bit. I will spill out your names on other occasions, and thanks that I was having fun. Also my football teammates who really forced me to stay healthy even in snowy weather. I am going to miss you all!

I personally would dedicate this thesis to my late grandpa(s) who (to me) were truly great scholars and inspired me to be the one. Next to them, also my grandma(s) who keep reminding me of being genuine. Ayu, thank you for your patience and love. On top of everything, the spirit in this thesis always belongs to my Bapak, Ibu, and Indra.

---

## Scientific portfolio

### **This thesis is based on the following publications:**

1. *The expanding world of hybrid perovskites: materials properties and emerging applications.* S. Brittman, G.W.P. Adhyaksa, and E.C. Garnett. *MRS Communications* **5**, 7-26 (2015). (**Chapter 1**)
2. *Carrier diffusion lengths in hybrid perovskites: processing, composition, aging, and surface passivation.* G.W.P. Adhyaksa, L.W. Veldhuizen, Y. Kuang, S. Brittman, R.E.I. Schropp, and E.C. Garnett. *Chemistry of Materials* **28**, 5259-5263 (2016). (**Chapter 2**)
3. *Understanding detrimental and beneficial grain boundary effects in halide perovskites.* G.W.P. Adhyaksa, S. Brittman, H. Abolins, A. Lof, X. Li, J.D. Keelor, Y. Luo, T. Duevski, S.R. Ellis, D.P. Fenning, and E.C. Garnett. under review (2018). (**Chapter 3 and 4**)
4. *Nanoscale back contact perovskite solar cell design for improved tandem efficiency.* G.W.P. Adhyaksa, E. Johlin, and E.C. Garnett. *Nano letters* **17**, 5206-5212 (2017). (**Chapter 5**)

### **Patent applications:**

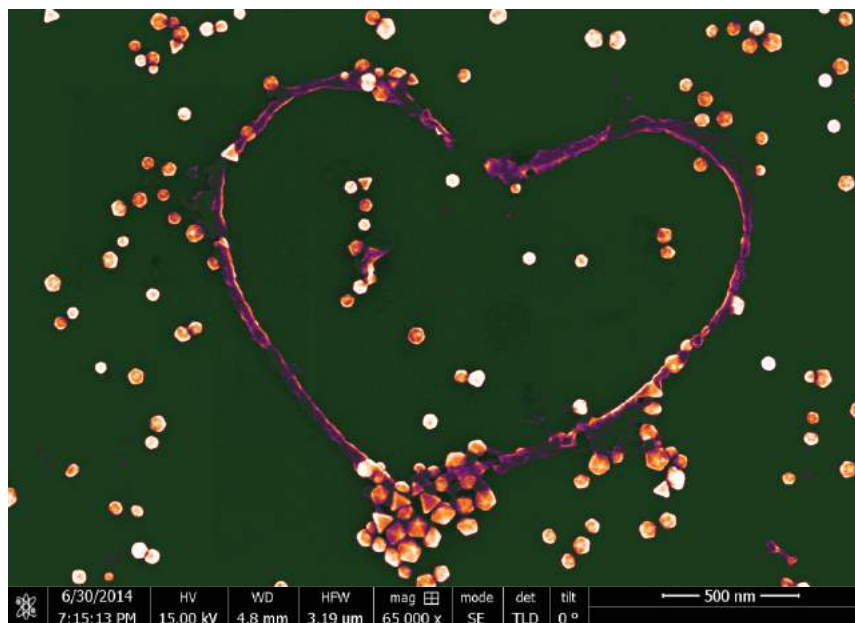
1. *Multi-junction back contacts solar cell.* E.C. Garnett, G.W.P. Adhyaksa, and L.J. Geerligs. EU Patent Application P6062079NL (2017).
2. *Method for fabricating an efficient bio-photovoltaic.* J.K. Kang, G.W.P. Adhyaksa, D.K. Lee, I.W. Ock. US Patent 9293265 B1 (2016).

**Other publications by the author:**

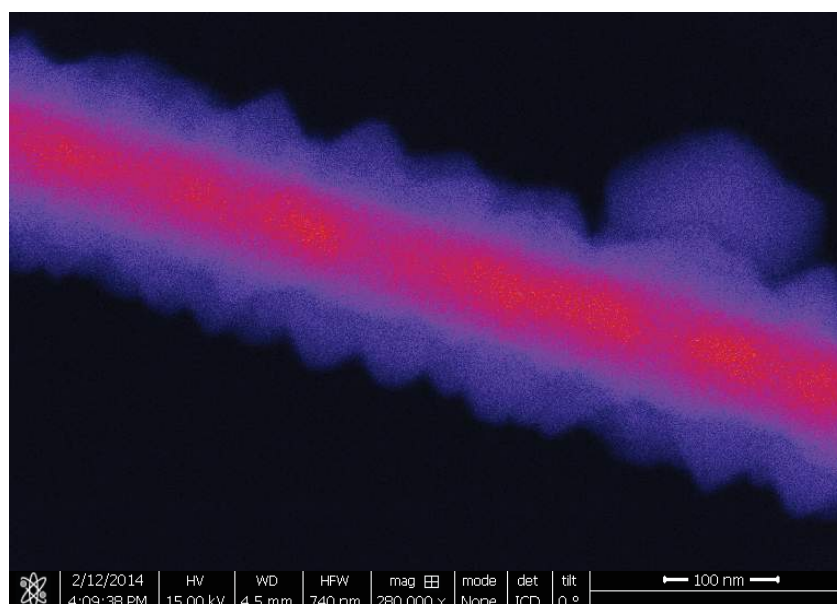
1. *Anisotropic carrier diffusion across different grain orientations in lead-iodide perovskites.* S. Jariwala, D.W. deQuilettes, G.W.P. Adhyaksa, J.T. Wang, H.J. Snaith, E.C. Garnett, and D.S. Ginger. in prep (2018).
2. *Transparent quasi-interdigitated electrodes for back-contact solar cells.* G. DeLuca, A.N. Jumabekov, G.W.P. Adhyaksa, A.N. Simonov, J. Lu, B. Tan, E.C. Garnett, U. Bach, E. Reichmanis, and A.S.R. Chesman. submitted (2018).
3. *Converting rationally designed mineral architectures into semiconductors.* T. Holtus, L. Helmbrecht, H.C. Hendrikse, I. Baglai, S. Meuret, G.W.P. Adhyaksa, E.C. Garnett, and W.L. Noorduin. Nature Chemistry, accepted (2018).
4. *General considerations for improving photovoltage in metal-insulator-semiconductor photoanodes.* I.A. Digdaya, B.J. Trzesniewski, G.W.P. Adhyaksa, E.C. Garnett, and W.A. Smith. J.Phys. Chem. C. **9**, 17-36 (2018).
5. *Interfacial engineering of metal-insulator-semiconductor junctions for efficient and stable photoelectrochemical water oxidation.* I.A. Digdaya, G.W.P. Adhyaksa, B.J. Trzesniewski, E.C. Garnett, and W.A. Smith. Nature Communication. **8**, 15968 (2017).
6. *Benchmarking photoactive thin-film materials using a laser-induced steady-state photocarrier grating.* L.W. Veldhuizen, G.W.P. Adhyaksa, M. Theelen, E.C. Garnett, R.E.I. Schropp. Progress in Photovoltaics: Research and Application. **25**, 605-613 (2017).
7. *Engineering the kinetics and interfacial energetics of Ni/Ni-Mo catalyzed amorphous silicon carbide photocathodes in alkaline media.* I.A. Digdaya, P.P. Rodriguez, M. Ma, G.W.P. Adhyaksa, E.C. Garnett, A.H.M. Smets, W.A. Smith. J. Mater. Chem. A **4**, 6842-6852 (2016).
8. *A light harvesting antenna using natural extract graminoids coupled with plasmonic metal nanoparticles for bio-photovoltaic cells.* G.W.P. Adhyaksa, E.C. Prima, D.K. Lee, I. Ock, S. Yatman, B. Yulianto, J.K. Kang. Advanced Energy Materials **4**, 18 (2014).
9. *Coupled near-and far-field scattering in silver nanoparticles for high-efficiency, stable, and thin plasmonic dye sensitized solar cells.* G.W.P. Adhyaksa, S.W. Baek, G.I. Lee, D.K. Lee, J-Y. Lee, J.K. Kang. ChemSusChem. **7**, 9 (2014).
10. *Broadband energy transfer to sensitizing dyes by mobile quantum dot mediators in solar cells.* G.W.P. Adhyaksa, G.I. Lee, S.W. Baek, J-Y. Lee, J.K. Kang. Scientific Reports **3**, 2711 (2013).

**Image galery:**

1. self-assembly "*Perovskite heart template*" with plasmonic nanoparticles (2014)\*.

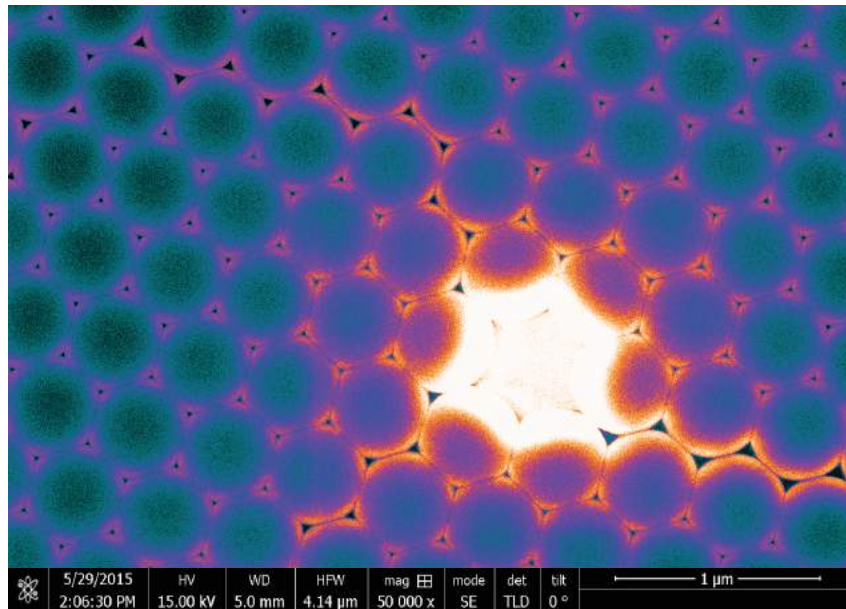


2. Plasmonic-perovskite core-shell nanowire (2014)\*.

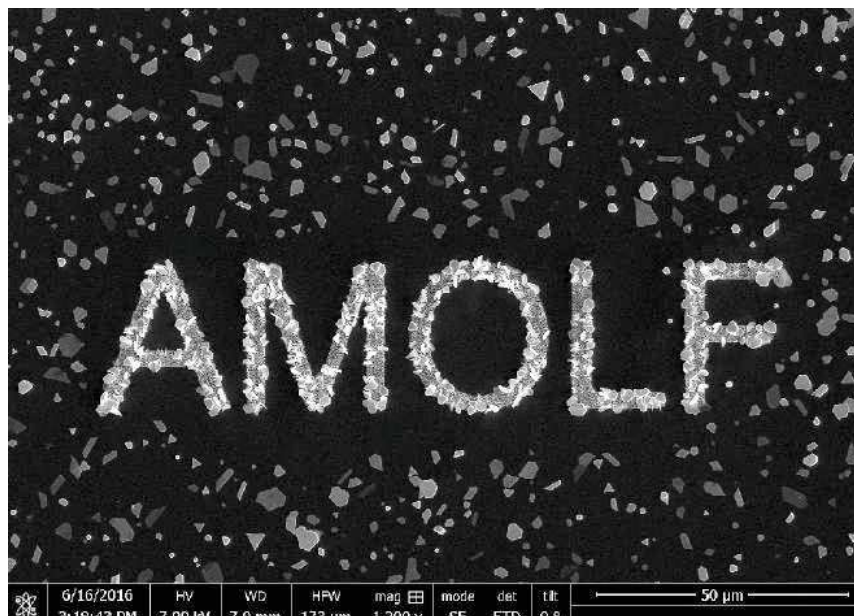




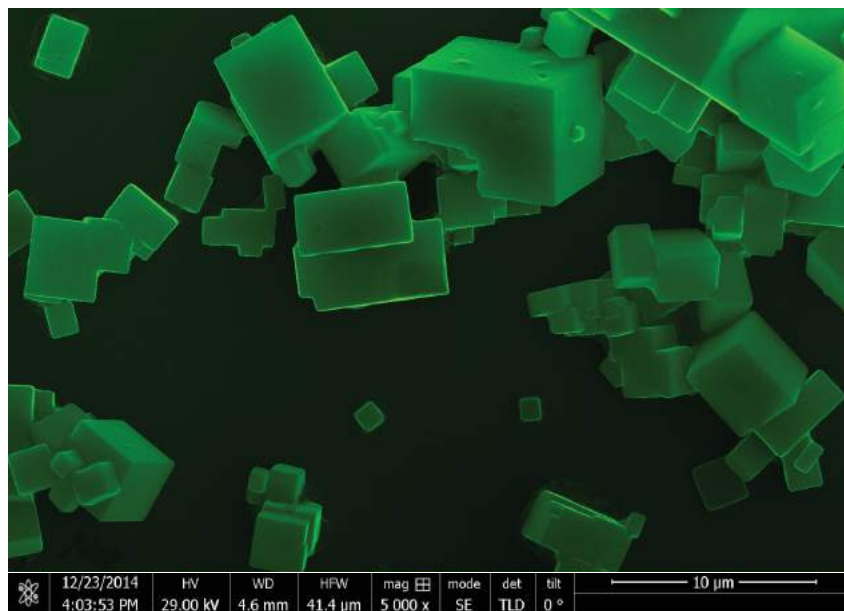
3. Vacancy defect inducing triple-plane boundaries in a hexagonal close packed structure (2015)\*.



4. Selective area growth of perovskite crystals on "AMOLF" gold marker (2016).



5. Cubic perovskites (2014)\*.



\*false color image

---

## About the author



Gede Adhyaksa was born in the island of Bali, Indonesia, on August 1, 1986. He studied Engineering Physics at Institut Teknologi Bandung in West Java, and graduated *Cum laude* (2008). Before starting his PhD at AMOLF (2014), during his master programs, he studied Engineering Physics at Technical University of Munich, Germany (2010), and Materials Science at Korea Advanced Institute of Science and Technology, Republic of Korea (2013). His research experience at AMOLF is presented in this book.

Besides research, he enjoys playing football, piano, traveling to unknown territories, and crafting aquatic plants at home.