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### Ion exchange, self-assembly, and light emission

*A dance of ions and light*

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**Publication date**

2020

**Document Version**

Other version

**License**

Other

[Link to publication](#)

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

Helmbrecht, L. (2020). *Ion exchange, self-assembly, and light emission: A dance of ions and light*. [Thesis, externally prepared, Universiteit van Amsterdam].

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# Acknowledgment

This endeavor began with the sight of beautiful colored SEM images of crystal flowers on the website of AMOLF. I was caught by the aesthetics and intrigued by the science behind it. At first sight, it did not seem to match my studies. Now, many adventures into this journey later, I know that this was a perfect fit. This thesis, the essence of those years in the lab, bears only my name on the cover. Yet we know that this book is the product of the many different relations between people. In this thesis, you saw the names of those I worked with at the beginning of each chapter. Beyond these scientific bonds, there were many helping hands and guiding minds involved. This is for my mentors who guided me, the scientists from whom I learned, the colleagues that created a warm atmosphere, the old friends who are still around, the new ones that made Amsterdam a home, and the family that always supported me.

First of all, my gratitude goes to Wim for giving me the opportunity to do my PhD with him. Wim, I still remember vividly how enthusiastic we both were during the interview. From that moment, I knew I wanted to do a PhD with you. You had to learn how to start a group, while I had to learn how to become an independent scientist. It has been an incredible journey in which you taught me how to spot exciting topics, make new discoveries, and finally, how to write about them in a way that creates impact. I am happy to have had the opportunity to visit many conferences and meet renowned researchers in our field. There are many highlights of our time together—the afternoon coffees, the trip to the GRC and your former lab, the discussions in our meetings, the moments in various labs together—just to name a few. Sharing the belief that science should feel exciting, we created beautiful figures and surprising experiments to convey this feeling. This is what made the years in the lab a lot of fun and turned this thesis almost into a picture book. Even when your group grew, you always found time to meet every single one of us, supporting us with your cheerful enthusiasm. You know how to choose the right balance between guiding and granting freedom. I cannot imagine having a better mentor for my PhD.

With Wim just having started his group, we had to figure out a lot of things on our own. Well, not entirely on our own. Martin was always there to help. Thank you, Martin, for your support throughout these years, your pragmatic advice helped to readjust the efforts and gave me confidence that with careful planning, everything will work out. Once, you suggested creating a "Lukas\_Thesis.tex" and adding the first paper—as you promised—this made things feel concrete, and here we have the final print of it.

Another implication of being the first PhD student is that writing a thesis is faster than getting a professor's title. Luckily, Huib became my official promotor. Thank you, Huib, for finding the time for me, even if that meant looking at my thesis during your vacations. I am impressed that although you manage a group and run the institute, you never lose your cheerful attitude.

I want to thank those that have opened this thesis not only for the pretty pictures and the acknowledgment. The Doctorate Committee took the time and effort to read this thesis down to the most minute detail. I want to thank prof. dr. E.C. Garnett, prof. dr. A.M. Brouwer, prof. dr. N.F. Shahidzadeh, dr. E.M. Hutter, prof. dr. M.L. van Hecke, and prof. dr. P. Levkin for accepting the invitation to be part of the committee and providing me with valuable feedback to eliminate the little typos before printing the thesis. I am looking forward to our scientific discussions.

Of course, I was not going to be Wim's only student. Foremost I want to thank you, Hans. Like scientific twins, we joined almost at the same time, became friends and are finishing practically together. Starting from what Tim and Daniel had set up in the first weeks of their internships, we began to build a lab together, we figured out how to be PhD students, traveled to conferences and conducted social experiments on how to create bonds between scientists. Through these years we were almost in sync with the challenges we faced, and chatting about them made everything easier. Not many PhD students get to work together with someone on the same topic towards the same goal, together with you making perovskites was a lot of fun!

Observing Wim as a mentor, I was keen on guiding students myself and got the chance to work with three excellent students. Thank you, Alicia (with your magical approach to experiments), Bruno (with your resolute effort in getting the perfect helix), and Ariane. Ariane, I am impressed by the sophistication you demonstrated already in your bachelor's research, and I am looking forward to seeing your beautiful crystal stars in a paper. Thanks for the liveliness while you were here and the friendship that followed.

From the outside—I have been told—our group sometimes looked like quite a wild mix of characters. But actually, this is what created a very enjoyable balance of personalities where each has its own place in the group. With you, Tim and Daniel, the early months when we had the freedom to decide which signature this lab will have in the future, were a lot of fun. Iarik, your pragmatic sarcasm made our lunch breaks fun and saved our meetings when we drifted off into pointless discussions. Arno, you are an infinite source of technical information—whatever the question was, you would help with something you read in a paper or you would have written some code to answer it. Marloes, your dedication to making the impossible work is fascinating, and with your caring attitude, you always created a warm atmosphere in the group. Christiaan, your weightless positivity always cheers me up, and your enthusiasm while explaining your experiments

when we met in the lab almost made me want to change topics. Sjoerd, your input to the challenges I face in the lab made me realize how important it is to routinely question everything you do in the lab. And of course, I want to thank Nesrine, Imane, Stivell, Pietro, and Miriam, who brought fresh perspectives into the group. During the many conferences and symposia, Liesbeth seemed almost part of the group. I want to thank you for the many words of advice you had on being a confident scientist. Esmee, it is exciting to see that as a secret member of the group, you bring out our science's artistic side. I find it remarkable that in our group, we always support each other and everyone will pause their own project for a moment if someone needs help. Without you all, this PhD would have been much harder and certainly less fun. I will always remember our coffees, ice-cream breaks, lunches, outings (in rain and wind), and conferences together. Our group wouldn't be complete with the indispensable help of Teresa, Karelia, and Sabine, who kept Wim's agenda together, as well as Marko, Marc, and Hincó, who kept the lab running and translated our thoughts into actual functional equipment. Marko, thank you for bringing in all the expertise on optics that I didn't have, and beyond that for the many lunch breaks together. Marc, you help with the many chemistry questions along the way was indispensable. Hincó, ik ben blij dat je altijd Nederlands tegen mij hebt gepraat en daarbij ook tegen iedereen hebt gezegd dat ze met mij Nederlands moesten praten. Dit heeft erg geholpen mijn Nederlands te verbeteren. Met veel plezier heb ik met je over het oplossen van de vele kleine technische uitdagingen van mijn PhD nagedacht. In een ander leven had de jouwe mijn droombaan kunnen zijn.

My path to becoming a scientist started long ago with many influential teachers along the way, who encouraged me in my curiosity for the world and the functioning of things. I want to thank my high school teachers, Herr Paas and Herr Landers, that got me interested in chemistry by teaching how to create color and explosions. Now, after a few years of PhD research, I am amazed by how many of the things I learned in my studies came in handy. I want to compliment dr. Mertin, prof. Bacher, and prof. Winterer for devising a university course that prepared me well enough to pursue a PhD in chemistry although studying nano-optoelectronics. Especially, I want to thank prof. Winterer and Notti for supervising both my bachelor and master theses. Listing everyone with whom I studied and from whom I learned during that time would fill a book by itself, but those who remember studying together hiding in the far corners of the library, in the Faschaftsraum with takeaway fries, or at home till deep in the night, you know who you are, and I want to thank you.

Like an apprenticeship as a craftsman is not complete without the *Wanderjahre*, an academic shall aim to learn from different mentors in different labs and cultures. I want to thank prof. Donghan Lee at Chungnam National University and his PhD student Jongmin, prof. Craig B Arnold at Princeton University and his PhD student Frederik, as well as prof. John Canning at the University of Sydney for

welcoming me to their labs and showing me their way to approach science. I learned how the friendship between people, the ambition, and the right questions are what develops a great scientist and enables one to make discoveries.

Having seen companies and universities, I was used to every group being a little kingdom of its own. AMOLF, however, felt more like a community with less hierarchy. It is this collaborative atmosphere that I am especially grateful for. So far, Wim's microarchitectures had been beautiful, the new mission was to make them functional. Wim and I set out to explore how they would interact with light. In our lab, we can create the self-assembled microarchitectures, but to explore their functionality, I needed help. It was clear that I needed Femius. Thank you, Femius, not only for the many funny conversations but specifically for bringing me back to the darkness of laser labs. Ruslan, thank you for introducing me to the puzzling world of Fourier optics and explaining what the light does whenever I got too puzzled. Bart, I want to thank you for introducing me to the world of crystals that twist light. It was a lot of fun to be in your lab at NYU together with Melissa, discovering how light can twist while it travels through our self-assembled helices. Melissa, your professionalism in the experiments impressed me. You were so good at finding helices and measuring them within minutes, I didn't even need to come back to New York (which was almost a little disappointing). Many thanks to the four of you; without you, there would be no chapter two.

The 3D perovskite architectures of chapter three were only possible because Erik had introduced Hans and me to his lab and the art of making perovskites. I want to thank you, Erik for the many discussions about this surprising material. Of course, we didn't do this alone and I owe a big thanks to Sarah, Gede, and Sophie. This work led to chapter four, in which I wanted to build an LED. I am thankful that Bruno welcomed me in his lab and that Moritz and Loreta did everything possible to help with knowledge and hands-on lab work to eventually make this happen. I also want to thank Louis for our many discussions with Wim and Alicia on manipulating the growth of the helices. I am looking forward to the day where you grow a field enantiomerically pure helices. The best thing that can happen to research is when it sparks someone's curiosity, and they decide to continue it. Therefore, I am honored that you, Igor, chose to introduce ion exchange reactions to your group and thank you Lucas for starting your postdoc by coming here to learn how to convert chalk into perovskite.

I also want to thank everyone who makes AMOLF a place where I only need to worry about getting my experiments working. My thanks go to Ad, Marjo, and Juliette whose warm welcome in the morning brightens my day already at its start, to Tineke and her team who keep things neat and tidy (especially to Kaya, who saves the plants and Jusuf), and to Floortje's team of the canteen. A PhD is full of technical challenges and this is a big thanks to those that helped to solve them all: Thank you, Hans, Bob, Andries, Dimitry, and Igor from the nanocenter. Without you, none of the beautiful SEM images would be here. Hincó, thank you for

always inventing something on the spot for the many technical challenges I faced. Dion, thanks for getting enthusiastic about my experiments, especially when you made the stamps. Niels, the many Christmas Lunch projects with you were a lot of fun. Ilya, Marnix, and Henk-Jan form the design department; your ability to transform a vague pencil sketch into a technical drawing is impressive. Thank you, Jan, Tom, Wessel, and the other fantastic craftsmen/craftswomen of the workshop, for bringing my ideas to reality and for the many interesting conversations we had. Jan, I enjoyed our many chats about the scientific and hobby projects you are mastering and am thankful for the advice you had for mine. Tom, the cheerfulness with which you tell about the things you are currently building makes me want to do an internship in the workshop. Wessel, it is always nice to pass by the workshop and hear about the things you are making and chat about photography equipment. Erny and Petra, thank you for promoting me to the official unofficial AMOLF photographer. I am honored that you gave me the possibility to grow into this role. I always enjoy passing by to hear about the events you are planning and to brainstorm about the next photo session for AMOLF news. Oana, thank you for your indispensable help in dealing with publishers, open access, and copyrights. Valeska, Reshma, Sharlene, Linda, and Wouter, thank you for having arranged such a smooth start into my Amsterdam life by sorting out my housing and since then taking care that PhD students don't need to do much paperwork. Thank you, Tatiana and Angela, for helping countless times with the many orders I wanted to place at obscure suppliers and even more for being the Christmas angels. Also, thanks to Tarik, Andre, and Clyde, I am happy that you always keep the building running so smoothly. Clyde, you are certainly one of the most iconic figures of AMOLF. Your authenticity brightens my day every time we meet. We met at all the fun PhD defense parties, and I am looking forward to seeing you at mine. Thank you, Floortje, Karelia, Teresa, Henriëtte, and Sabine; if it weren't for your effort to structure the group leaders' agendas and tell us where we can find them, no one would graduate on time.

AMOLF is not complete without the PV. Floortje, Niels, Hans, Konrad, Anemarie, Niels, Giorgio, Giulia, Sukanth, Christian, Lennard, Tom, Loreta, Jeffrey, and Isabelle. I want to thank you all for being such a great team and all the fun we had together with organizing the best borrels and most fantastical Christmas Lunches AMOLF has seen. We couldn't have organized events this big without all the helping hands that have helped setting up and tidying up. Thanks to you and thanks to those that showed up to the events. Of course, I also want to thank those that established the PV before us and those that continued once I left. Beyond the PV, I want to thank everyone who was part of the many other activities. Nasim, thank you for initiating a language café that reflects the international spirit of AMOLF. Thanks to those who met in the Climate Lunches to discuss what we scientists can do about the climate (thank you, Giada for kicking off the first

meeting) and those who would just start a spontaneous vrijdagmiddagborrel with a message in our life of pi whatsapp group.

The office I was in felt sometimes more like a botanical garden than an office. This little green oasis has hosted many friendly faces throughout these years. Katharina and Erçağ, the plant we chose together, has been my steady companion, who reminds me of the beginning at AMOLF and our time in the office. Erçağ, I like to recall the many productive hours in silence, interwoven with deep conversations, and occasionally compensated with a concert at Paradiso. Ariane, with your arrival in the office, the calm was replaced by positive energy with which you motivated everyone to leave their comfort zone. When Ariane left, it took till you, Hanne, arrived and with you, liveliness returned. Anne, you, together with Hanne, have brought the mood in our office to another cheerful peak. When I had to move into the corona-home-office, I realized how vital this atmosphere was for me. I missed the steady calm of you, Zhou, recalled the chats about life with Rob, and the days with Daniel, Nesrine, Alicia, and Imane, who once were part of the office. Said, Falco, Marnix, and Bob, you joined us so often, you feel like temporary office mates that I now miss. I am thankful to have had such lovely company throughout the years with whom we could work in silence and chat about life.

You will have noticed that the SEM images on the cover are the most colorful ones you have ever seen. These images were taken by Stefan Diller, purely motivated by the beauty of the microarchitectures. I want to thank you, Stefan for reaching out to us, taking such fantastic images, and even making 3D movies. This was the most surprising and most aesthetic collaboration of my PhD. And to the scientists with beautiful SEM specimen: make sure to tell Stefan about it. I am grateful to those that helped to make the inside of this thesis artistic. Henk-Jan, I like recalling the photoshoot for the sand dollars, spending a day in a dark room throwing sand dollars into an aquarium. Det, I want to thank you for teaching me how to make a coral from clay during the "Molding Nano" workshop with Giada. Rogier, thank you for passing by AMOLF to take these impressive photos of scientists, including me.

To my paranymphs Annemarie and Leonie—I chose you because you are smart enough to defend this thesis with words and fearless enough to defend it with a sword if that was still allowed. I am honored that you accepted my invitation, also because you both match my spirit of the PhD—always dedicated to perfection in the lab, and rarely frustrated about the frustrations of science and devoted to actively enjoying every moment outside of the lab. Annemarie, thank you for all the traveling we have done together. Independent of where we are, you always have the right advice and your sober realism has brought my mind back to solid ground many times. Leonie, thank you for the many conversations we had on the many aspects of life. From theater plays and unknown indie bands to shady

take out restaurants and beer at Joost, you are the loveliest companion to venture through the nights of Amsterdam while never losing our calm.

Most of the friendships I made at AMOLF started with an encounter at the coffee machine. If we had given the coffee machine a name, I would thank her for being such a good companion throughout the years. She started on the same day as me and made about 20.000 coffees a year. In front of the coffee machine, everyone is equal—master students can show senior professors how to make a good espresso, and group leaders can casually check in on their students. I recall many chats and want to thank Albert for his caring career advice, Bas for showing me how to froth milk properly, Pieter Rein for enthusiastic stories about his research, Femius for commenting on conversations with humorous facial expressions, Paula for stories on her travels, Kristina for discussions on gender equality, and Bruno and Erik for spontaneous advice on misbehaving perovskites.

Walking from the coffee machine to our lab always involves a brief but pleasant journey through the lab of Martin. On my way through, I often got stuck—distracted by this physics playground. Scott playing with bouncy balls, Anne crumpling plastic sheets, Corentin programming smiley faces in rubber, and Hadrien putting architectures in the hydraulic press, you always brightened my day with brainstorming about what else one could do with all those squishy or wrinkly rubber things. Thank you for showing me how fun mechanics can be!

I made many friends at AMOLF during my PhD. Some of you I saw only inside of AMOLF, others mostly outside. Without being able to mention all the names, I want to thank everyone who socializes at the coffee machine. Somehow there is always someone having a coffee to have a chat with, no matter if day or night. Moritz, when we met during my interview, little did I know back then what lasting friendship would develop. I like to look back at the places we explored together, from morning coffee with a panoramic view in Berlin to Zürich with Laura and Leo. Giada, thank you for bringing art and politics into AMOLF. With you, I enjoyed exploring the hidden sides of Amsterdam and learning how to make the best pizza in Italy with your parents. Now that our friendship has brought us safely through the last few months of intense thesis writing, I am looking forward to meeting again somewhere and starting another artistic project. Giorgio, thank you for the many coffee breaks we spent trying to get a decent coffee out of mediocre beans and the many evenings in which we thought we would write code but ended up cooking dinner. Your culinary dedication is amazing, and I am looking forward to many more dinners with Andrea and you. Nasim, I am always amazed by the endless source of cheerful energy inside of you. I couldn't imagine anyone else starting a language café and getting so many people excited about it. When the green oasis of my office faced the lockdown, you adopted the plants and gave them as much care and attention as you give your friends. Thank you for balancing my soul with delicious Persian dinners when there seemed to be only work, and the writing bound all my time. Dario, the dinner evenings



with you were great. I especially like to remember all the food adventures we had together—walking half an hour through town just to try another bar that someone told us would have the best jamón ibérico. Agustin, conversations with you always bring back my optimism. When you listen, you are always interested, yet nothing can surprise you. Jenny, where ever you are, things are cheerful. I always enjoyed how, wherever you go, your laugh signals your arrival. Carolyn, whenever you cook and bake, I feel reminded of home, and whenever I was gone, and you took care of my plants, they seemed happy as if they had been on vacation themselves. Giulia, many of my friends admire food, but your passion for original ingredients impresses me. Thank you for showing me these flavors and hosting dinners with Nicola. Cristina, thank you for the concerts together and the Zen you radiate. Felicio, thank you for sharing many beautiful techno experiences, no matter how far we had to travel.

In such a rainy country, sunny days need to be worshipped. On a sunny day, no one would risk wasting time late at AMOLF or even at home. The many summer barbecues were a great place to make and develop our friendships. Marco, Ale, Masha, Nicola, Mario & Olga, Lorenzo, Giorgio & Andrea, Agustin & Bram, Cristina, Jenny, Agata, Parisa, Carolyn, Laura, Dario, Eliane, Moritz & Laura, Giulia & Judith, Ruslan, Kevin, you were indispensable for a party mood, no matter whether in the park or at a party, you would make sure some dancing would happen. Federica, thank you for bringing the physics and the biophysics parts of AMOLF closer together when you first suggested merging two different BBQs.

I want to thank everybody that made my life at AMOLF richer—Kevin (for your sarcasm that lifted the mood of any coffee or lunch break), Christiaan M (for your support and the nice postcards), Loreta (for making me feel at home in Rome and never getting tired of my perovskite questions), Chris (for the grounding atmosphere that surrounds you), Marloes (for our shared fascination for the mind), Henk-Jan (for your advice on photography and the sand dollar photo shoot), Rob (for radiating kindness wherever you go), Sander and Sebastian (yours were the first defenses I addended and was looking forward to having that same confident seniority by the time I would finish my PhD), Roberto (for our many lunch time conversations about food and for briefing me for my Mexico travels), Mario (for the energy you brought into every party), Isabelle (for your dance moves and trying to teach them as well as sharing the fascination for photography), Ilya (for the concerts and parties we went to together and for adding liveliness to Eindhoven), Ruslan (for your endless enthusiasm, and for hosting fun house parties), Lucie (for your caring personality and the posters on influential female scientists in your office window), Susan (for bringing a sense of fashion into AMOLF), Steffen (for welcoming me at AMOLF in odd hours), Eline (for your support in perovskite questions and encouraging women to stay in science), Benjamin and Christian D. (for bringing up politics from a German and a Dutch perspective and sharing

the obsession with coffee), Beniamino and Charly (for your inspiring lively spirit that made me question the productivity obsession at AMOLF), Mareike (for the kind atmosphere you bring wherever you go), Bob (for inspiring me to bring more lab equipment to the kitchen) and Bob together with Dimitry (for encouraging everyone at the borrels to go wild), Tom (for the conversations on coffee and rebuilding a house), Alexander (for more conversations on coffee), Hugo (for the many conversations over lunch and the enthusiasm with which you explain your research here and in Zürich), Anne (for your positivity each day and our snack-sharing friendship), Verena (for your determination to make this world a better place), Eitan (for challenging me and everyone with questions and sarcasm), Said (for the lunches together, the interest in the people around you and the different perspective you bring to conversations), Luuk (for sharing your experiences about your life as an artist and advice how to bring that out in myself). Also, the many other little encounters in the daily AMOLF life really make a difference. I was always happy to run into you—Cesare, Laura, Marco, Yuval, Melle, Bart, Rene, Hongyu, Matthijs, Jian-Yao, Magda, Fotios, Aaron, Alexandra, Luca, Harmen, Matthias, Sven, and Nick. Some of you I especially remember for the shared passion for food—Nicola (for having the best food a defense has seen), Andrea (for constantly talking about coffee or food), Agatha (for importing polish sweets and sausages), Parisa (for bringing Safran from Iran to assure that I cook with quality ingredients), Anders (for making Haggis and the most elaborate cakes), Eliane (for shining at every cake baking contest), Harshal (for teaching me some Indian cooking skills), Jumin (for making a proper Korean bbq), and finally the bread-baking friends: Kelly, Moritz, Cristina, Giorgio and Verena.

The first days in Amsterdam, before even starting my PhD, I dedicated to learning Dutch. Not only would this be the basis for integrating into Amsterdam, I also made good friends. Nina and Johanna, I am happy we are still in touch this long after the course. Zita and Vainius, you were my first friends in Amsterdam and you are such a cosmopolitan couple with amazing friends. I am looking forward to seeing you again, somewhere on this globe. Alex J., for you, it took a while till you eventually moved here, but since then, we had many coffees and beers talking about the Dutch things we like and the German things we miss.

Amsterdam is a place of art and liveliness. Marlies, you showed me how colorful and fun that world is, and with you, I discovered many of those colorful aspects in myself. The energy with which you move around the globe to follow your passion for fashion inspires me to live as intensely as you and never get stuck in a place. Rosa, I always enjoyed creating and discovering with you. Steffi and Davide, whenever I met you two somewhere, I knew I had picked the right event for the evening. Ashley, thank you for showing me how different and yet similar a PhD student's life can be that has to do fieldwork instead of lab work. Ruth, we both still are surprised how randomly our friendship started and how it grew into a close bond over the course of a summer. You inspire me with the

ease with which you go through life and the many references to literature you use to comment on what happens around you. I enjoy recalling the trips we went on and am grateful for the support we give each other—you are the best. Ranes, it is beautiful how we met through music and how, ever since, we've been cooking well and made people dance. Paloma, you see the world with critical eyes; the Columbian anthropologist in you has made me aware of the often Eurocentric routine with which I look at the world. Imran, you are one of the most loving and kind persons I know. Thank you for our conversations without judgment. You truly know how to make everyone feel appreciated. Kat, our shared fascination for sound, light, and the mind has started a wonderful friendship. Egle, it is always fun to explore museums with you. Alex K., the BBQ's with you in Duisburg were always a culinary highlight. I am glad that now, you and Hanneke are just a few minutes away. There are many more that have made my time in Amsterdam unforgettable, Istvan, Joy, Jonathan, Doet, Miko, Willem, Szymon, Mohammad, and all the others that I may have forgotten to mention; thanks for your friendship!

Part of my time in Amsterdam was not actually in Amsterdam. Tobi, you challenged me to move into a home office before it was cool. And then take this home office with me around the world to meet you in Peru and Mexico. Thank you for showing me a different way of living and helping me to learn so much about myself.

This thesis would not have been possible without my amazing friends that supported me through school and university. I am grateful that our friendships continued, and so many of you came to visit me here in Amsterdam. I want to thank all of you for the many contributions to my life. Kati, I enjoy looking at the many years we spent together and how we grew with one another. Christian and Hendrik, by now, you are kind of family, making me feel at home whenever we meet. Together with Kati, Anna, Lina, Marcel, Eva, Katha, and Anka, you two always make me feel welcome in Witten. Jacke, I enjoy your reflective view on the world. Dominique, I keep being impressed by our friendship that lasted although we changed so much during these years. Chrisi, it has always been great fun to come back to Witten to your parties. Even more importantly, you also visited so regularly that I never felt like I really lived abroad, especially when Lena and Lauritz joined. Lünd, your critical perspective on current affairs is always a great start for conversations. Cornelius, I like creating art with you. I wish there was a way to add NanoTropics as a video at the end of this text. Marcel, thank you for never losing contact through these many years. Maria, I enjoy your directness and liveliness; thanks for still being there after these many years. Johannes, I am impressed that no bus ride can be long enough to discourage you from passing by. Jean, thank you for bearing even longer bus rides and sharing your fantastic taste in music. Eddie, I am inspired by the many lives you have lived and still remember when you wrote your first book full of illustrations. I am sure you will approve of this scientific equivalent. Henning, your drive and entrepreneurial

spirit keep me inspired to be open to the possibilities that make life exciting. Julien, thank you for balancing out the hectic ones with your peaceful atmosphere. Alex, I am amazed at the energy you go through life with. Especially with you, Zoey, you two become unstoppable. Around you two, life is not allowed to become a boring routine, thanks for the times together.

Studying NanoEngineering was quite an experiment, but going through this together with friends made it a memorable time. Seeing you again in Amsterdam, on my birthdays, at fusion, and in your new homes means a lot to me. Caner, your dedication and entrepreneurship keep impressing me—Kaiserberg for life! Mohamed, you are the best flatmate in the world. Andreas, imagining wild scientific ideas with you, always revives my fascination for science. Stevan, thank you for our many conversations about photography and food. Alex, your attitude makes conversations relevant, and when they pause, your taste in music pleasantly fills the room. Stephan, I enjoy listening to the stories of your adventures. Erik, thank you for first introducing me to Arduino projects and sharing a fascination with electronics. Eric, the many cooking evenings at yours are some of my favorite memories of the last Duisburg years. Domme, thank you for reuniting us every time around your birthdays and for the many music recommendations you (and your dad) gave. Samer, your fascination for fire and photography makes the conversations always exciting. Rene, thank you for our nighttime walks with beer and discussions in whichever city we met. Lana, you are always a great addition to those walks. Ina, thank you for always having a loving word of support for me and for all your friends. Freddy, your cheerful chaos always brightens my mood. Kevin, planning our fusion mascots together is the best part of fusion preparations. Tim, thank you for making sure that at some point in the night, everyone dances to psytrance. Lisa, thank you for the many weekends spent with music and good food. Patti, the friendship with you is the most valuable element our studies created. With you, I feel comfortable telling anything that is on my mind, and many times your challenging questions have brought clarity to my thoughts. From calm to wild, from fine teas on the couch to Kellertechno in concrete bunkers, time with you is extraordinary. Danke, dass du meine beste Freundin bist.

I am really fortunate to have a very supportive family, this includes everyone from siblings, parents, grandparents to aunts and uncles, cousins, and those that feel like family. Especially the infinite loving support of my parents made it possible to make it this far. Thank you, Mia and Detlef, for supporting me in every possible way—teaching me to be independent and think in a critical analytical, and creative way. By its very nature, there is more I am grateful for than these few lines can express, yet here are some thoughts that came to my mind. Detlef, you taught me how to overcome any problem that involves technology, code, or mathematics and inspired me with your fascination for sound and music. Mia, you are the origin of my passion for food and art. From you, I learned to dream and create. Oma, wenn ich sehe wie munter und aktiv du noch mit über neunzig

bist, habe ich keine Angst alt zu werden. Mit deinem Hühnersuppen Rezept rettetest du mich jedes Mal durch die verregneten Amsterdamer Winter. Opa, von dir habe ich gelernt gründlich zu arbeiten und Nickerchen zu machen. Granny and Jeddo, you inspired me to aim high, and I am proud to carry on your passion for food and festivities. Nelly, thank you for your moral support during the years of my PhD. We understand each other and the challenges the life as a scientist brings. Kilian, thank you for the many conversations in which we sorted our thoughts on the situations we are in. Through your analyses that that span from the details to the big picture you have opened my perspectives on the world. With both of you, I share the curiosity for the world and am amazed to see your ways of uncovering the unknown. Time with you two always grounds me, and your reflective view on the world sparks my enthusiasm for life. Mit euch allen teile ich nicht nur die Neugierde für die Welt, sondern auch die Leidenschaft für gutes Essen, eine Gemeinsamkeit die zusammen mit euch am meisten Freude bereitet.

Together, all of you played a role in making this thesis possible. And even beyond that, you shaped who I am now. Thank you!

