INTERVIEW

Solar cells combining hot carriers and multijunctions: synergies and insights from Maxime Giteau, Samy Almosni, and Daniel Suchet

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JPE editor-in-chief Sean Shaheen (Univ. of Colorado, Boulder, US) with JPE authors, (bottom, left) Maxime Giteau (CNRS, Ctr. de Nanosciences et de Nanotechnologies, Palaiseau, France; NextPV, Univ. of Tokyo, Japan), (top, right) Samy Almosni (Saule Tech., Wrocław, Poland), and (bottom, right) Daniel Suchet (CNRS, NextPV, Univ. of Tokyo, Japan; École Polytechnique, Institut Photovoltaïque d'Île-de-France, Paliseau, France). Readers are also invited to enjoy the interview in video format, https://bcove.video/3az5dNx.

Sean Shaheen: Hello everyone and welcome! I'm Sean Shaheen at the University of Colorado Boulder. As editor-in-chief of the *Journal of Photonics for Energy*, it's my pleasure to host this interview highlighting our recent paper, "Hot-carrier multijunction solar cells: sensitivity and resilience to nonidealities" (https://doi.org/10.1117/1.JPE.12.032208). In general, the *Journal of Photonics for Energy* publishes works that apply to photovoltaics (PV) and photonic concepts related to renewable energy. This work that we'll talk about today is part of a special section on novel photovoltaic materials and devices (https://www.spiedigitallibrary.org/jpe-novel-photovoltaic-device-architectures).

We have with us today three of the authors from the paper, Maxime Giteau, Samy Almosni, and Daniel Suchet. Our goal here is to highlight the work and learn a little more about it, how it came to be, a little bit of the inside story, and learn about the authors as well. So, with that, we'll dive right in and I'll ask the authors present to introduce themselves, tell a little bit about themselves, and then we'll start with the interview. So, Maxime, do you want to start?

Maxime Giteau: Yes, it's a pleasure to be here! My name is Maxime Giteau. I'm a postdoctoral researcher at ICFO in Barcelona currently, and I did this work when I was at the University of Tokyo in Japan, where did my PhD and also partly in France at C2N. I work in high efficiency

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photovoltaic concepts, in particular hot-carrier solar cells, as well as ultrathin solar cells and light trapping.

Sean Shaheen: Wonderful, welcome! Samy, do you want to go next?
Samy Almosni: Yes, thanks a lot for the invitation! I'm Samy Almosni. I did my PhD on multijunction solar cells, especially dilute nitride on silicon. Then I met Maxime and Daniel in Tokyo, in the framework of the NextPV collaboration. Since then, I've been working on perovskite solar cells—currently at Saule Technologies, on inkjet-printed perovskite solar cells.

Sean Shaheen: Great, welcome! This is a very international team we have. Daniel, please go next.
Daniel Suchet: Thank you very much and thank you again for the invitation! I'm Daniel Suchet. I'm a professor in physics at École Polytechnique in France and I work at the IPVF, L'Institut Photovoltaïque d'Île de France, where my research concerns mostly thermodynamics for solar energy conversion and optical characterization of material and devices. I had, also, the opportunity to work at the University of Tokyo through the international NextPV collaboration. And I'll also say a few words about our fourth author, who cannot join us today. Jean-François Guillemoles is a research director at CRNS also working here at IPVF, and he led this collaboration, where we all met few years ago.

Sean Shaheen: Wonderful, thank you for that! So, we'll begin. I'll ask actually Maxime to briefly introduce the paper and tell us a little bit about its implications.
Maxime Giteau: Sure, so there are two high efficiency technologies which are called multijunction solar cells and hot-carrier solar cells, which promise very high efficiencies much beyond the famous Shockley–Queisser limit. In this paper, we introduce a hybrid concept called the multijunction hot-carrier solar cell, the aim of which is to bring together the advantages and purposes of both technologies. The multijunction solar cell is well developed, as this is the technology that achieves the highest efficiencies, but it's quite constrained in terms of the materials you can stack together, in terms of the efficiencies—how to get beyond the current efficiencies. The hot-carrier solar cell hasn't led to high efficiency devices—as of yet it remains mainly a concept.

What we did in this paper is to show how bringing the two technologies together really benefits from the synergies between these two. And mainly, regarding the multijunction solar cells, considering hot carriers, strongly relaxes the constraint between the band gap of the different junctions that you need to have, so you have much more freedom in terms of which materials you can stack on top of one another. In particular, you don't even need to have a higher band gap material on top of a lower band gap second material. For the hot-carrier solar cell, we showed that it's much less difficult to make a functioning hot-carrier solar cell if it's on top of a second junction than if it's by itself.

And the second part of the paper was looking more at the resilience of the device to operational conditions, namely the solar radiation. The fact is that real solar cells don't operate under some standard solar spectrum 24/7, but the sun moves in the sky, and so the efficiency varies throughout the year, and so you have to calculate what the average of that is and if a cell is optimized for a certain condition, it might not be ideal all the time. We show, essentially, that the different types of technology, they're not that much impacted by non-ideal illumination conditions and the hot-carrier multijunction solar cell is actually quite robust to these variations.

Sean Shaheen: Yeah, perfect! So, actually a few questions came up during your highlight there. One is that if you can reverse the ordering of the high band gap and low band gap, what does that give you? Does that mean you have more flexibility in depositing materials, in terms of lattice mismatch if it's a crystalline material—or, what does that provide?
Samy Almosni: Maybe I would like to add one point on that. Especially using low band gap material, what is very interesting is we can reduce truly the thickness of the top junction, which is actually of double interest in terms of commercialization, because as the number of the junctions is increasing, the thickness of the stack is increasing, in classical multijunction, which is making

the growth here more time-consuming and more costly. And the second thing is that by reducing the thickness, it also allows us to reduce the constraint on the lattice mismatch. [When using low bandgap top junction, the thickness required to absorb enough light and achieve current matching with the bottom cell is very low (in the order of 30 to 200 nm). This allows the use of highly mismatched combinations of materials. Indeed, the highly mismatched top junction can be grown with thickness below the "critical thickness" where strain relaxation starts to occur and dislocations form.]
Daniel Suchet: And I would perhaps add a third point, which illustrates what Maxime mentioned as the synergies between the approaches. If we want to reach the efficiency of hot-carrier devices, we need to have a slow cooling rate. Ideally, the cooling rate would be zero. Then we would not care about the actual thickness of the material as soon as it absorbs enough light. But it appears that materials have a finite cooling rate and that making the material thinner tends to improve the hot-carrier properties of the material. But, of course, making the material too thin, if it's just a single hot-carrier absorber, making the absorber too thin would lead to reduced absorption and so reduced efficiency. So, there is a tradeoff—the usual tradeoff between being electrically thin and optically thick—and with hot-carriers we have this additional constraint that we need to be acoustically thin enough, as well, to favor hot-carriers. Now this trade off, which is hard to handle in a single hot-carrier absorber, we walk the issue around here by considering that the photons, which will not be absorbed in the hot-carriers absorber, are still going to be harvested in the underlying subjunction.

Sean Shaheen: Yes, that's part of the design flexibility. So, I'm having all sorts of interesting creative ideas. Could you use the same band gap for the different junctions? What if you had, you know triple junction—same band gap, but very thin absorbers. Could that still be a benefit?
Samy Almosni: Actually, that was the original idea. The very first time we discussed, the idea was to make (homo-)multijunction solar cells with hot carriers. So, this was the original idea.
Maxime Giteau: I would actually add to that: in practice, the first junction is going to get most of the hot-carrier effects, so it's difficult to have two junctions consecutively that would behave as hot-carrier solar cells. So, if you want to consider three junctions, I would say that the bottom and the middle are going to be the same as in a traditional multijunction device. But the top junction, you can really make it much thinner and much lower bandgap to harvest, as much as possible, the hot-carrier effect.

Sean Shaheen: So that may limit the total number of junctions that would be useful to include in the device, in the stack.
Maxime Giteau: Yes. With two you can already do much more than with this double multijunction classical solar cell, because you get the hot-carrier effect on top. So that's the benefit I would say.

Sean Shaheen: Now, you wrote the paper agnostic as to materials, but if you had to build one of these today, what material set would you try to start with? Or is that unknown?
Maxime Giteau: We have several ideas.
Samy Almosni: Yes, we do have several ideas. For me, I really believe in silicon–germanium alloy, for two reasons. 1. Because I think we could grow it—we could grow thick enough layers to have a good absorption. 2. I'm not a hot-carrier expert—I'm more in the multijunction solar cells, so maybe the others could correct me—but what I have read on those types of silicon–germanium alloy is they can have very high mobility and even sometimes ballistic transport. And so this, from my old knowledge of interaction between phonon and electron, means if you have ballistic transport and very high mobility, you have very low interaction between the phonon and electron and thus, I guess, a low thermalization rate. So, I would really bet on this type of high mobility materials.

Sean Shaheen: And you can tune the band gap as well, of course.
Samy Almosni: Yes!

Sean Shaheen: Maxime or Daniel, what would be your favorite material? Without giving away any trade secrets.
Maxime Giteau: I'll say the more conservative one, maybe, which is III-V multijunction because it's the established technology. So, using different III-V layers than the traditional three junction solar cell with germanium–gallium arsenide band gap, the advantage of going with our approach is we can switch, actually, to indium-phosphide-based materials because we don't need the high band gap material anymore. So, we could have, if you think about a triple junction, indium–gallium arsenide low band gap with two indium phosphide layers on top, for example.

Sean Shaheen: Yeah! You know, there was just recently a new record III-V multijunction at 1-sun illumination. I think it was 39%, something like that. So, it's really moving along! Daniel, did you have a favorite material?
Daniel Suchet: It's also chance here to emphasize the idea that our proposal builds on the fact that the systems are not ideal, because if you would have ideal multijunctions with an infinite number of junctions, it would work better than what we're suggesting. If you had an ideal hot-carrier solar cell, single solar cell but ideal, it would also work better than what we have. But our idea was really to take into account how materials are not ideal, how designs might be nonoptimal, and how we could still manage to get to higher efficiencies even moving away from the most ideal conditions—both from the design and the operating situation.

Sean Shaheen: Yeah, I think that's an important philosophical point. If we're always trying to achieve the absolute optimal, we have kept failing, for the last 40 to 50 years. So having a resilient design and shooting for a little bit less than optimal, which is what you've done, may be the best approach. I think that's a key point.

Okay, so I could ask lots more technical questions, but let me go back to more about how the work came about and ask what challenges or key obstacles did you have to overcome to bring it to fruition? And I'll ask Maxime to start again.
Maxime Giteau: One of the challenges is in modeling this kind of system because we're dealing with nonideal devices. So ideal devices we know how to model with the Shockley-Queisser limit, and you can add hot carriers, and if you assume there's no thermalization, it works quite well. But we had to add the minimum number of nonidealities that really made sense from our point of view. As Daniel mentioned previously, the fact that what will happen is we want some thermalization, so we don't have an ideal hot-carrier device, so we want to introduce some thermalization and we wanted to introduce the fact that to reduce this thermalization, you need to reduce the thickness of the cell, and that reduces the absorption. So, we needed an absorption model and a thermalization model. And so, we took from several of our previous works, and also from the literature, in order to build a consistent model that could take into account these different effects and, of course, adding to that the fact that the model had to consider several junctions, one on top of the other.

Sean Shaheen: So, there are other nonidealities that you could think about, perhaps in a follow-up paper.
Maxime Giteau: Exactly. Those were the nonidealities that were specific to the technologies that we were considering. We didn't consider nonradiative recombinations, for example, because we could say it's happening everywhere so we're going to assume all the cells are ideal from this point of view and saying if there is nonradiative recombination, it will impact all the technologies in the same way, relatively speaking.

Sean Shaheen: Absolutely.
Daniel Suchet: Maybe also, to build on what Maxime just explained, once this model was developed and had many parameters, to figure out what would make sense in terms of study and what to show in a paper that would make sense and that would make our point clear and test the actual resilience of the system. That was also something which we had to wrap our heads around. What actual system to consider, what to keep fixed as parameters, and what to vary so as to create the maps that have been shown in the highlight of the paper, for instance (Fig. 1).

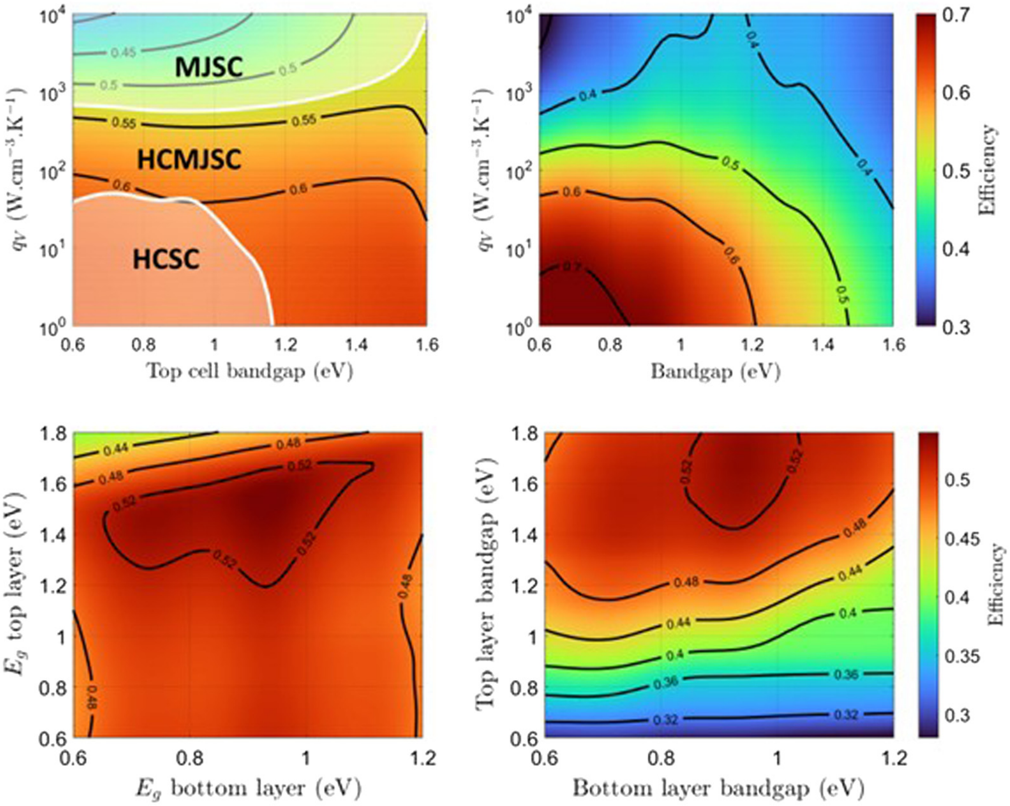

Fig. 1 Efficiencies of the MJSC, HCMJSC and HCSC panels as a function of the bandgap and thermalization coefficient. The novel HCMJSC design, investigated by the researchers, shows higher resilience to nonoptimal bandgaps and less constrained thermalization requirements, widening the scope of candidate materials for its design. Image credit: Giteau et al., doi 10.1117/1.JPE.12.032208.

Sean Shaheen: That's a really good point. So, it wasn't obvious in the beginning which figures, which plots you should produce, what the narrative of the work was. Is that correct?
Samy Almosni: Yes.
Daniel Suchet: Absolutely.

Sean Shaheen: I think that's an important point for any beginner researchers or new grad students: that aspect also requires a lot of thought.
Samy Almosni: Maybe on that point, we could say and acknowledge the work of some previous researchers, especially the work that was done at NREL on the modeling of gallium indium phosphide, the two junction solar cells on top of gallium arsenide because they were already discussing about this tradeoff between the band gap of the top junction and its thickness and its impact on the defects (generation) in the cell (during) growth. And to make a good device and a reliable device. So, I think we have been strongly influenced by this type of paper.

Sean Shaheen: And, of course, that work continues today. We already mentioned a new record in that area and the same questions hold that the researchers are working on.
Samy Almosni: I would like just to, on the difficulties, maybe just add one quick point. For example, especially to find those thermalization coefficients, from what I could see, people in the hot-carrier community were characterizing the hot-carrier relaxation in very different ways. Finally, it was difficult to find the data for the thermalization coefficient that we could use for our modeling. I think the approach was, for example, we are giving a value that was found in a previous work, actually from Maxime, and I think this approach was very interesting in terms of,

let's say, a more universal way of characterization, to do the characterization of hot-carrier solar cells, so that then we can compare the properties of different materials.

Sean Shaheen: I think that that's an important point. With any emerging technology being able to compare across papers across materials that's oftentimes a challenge. I'll tell you from our own work here in Boulder, if we use a little bit different assumptions in the fitting, we can get quite different thermalization constants. Where you do the fit and what the formalism is, what the equation is. So, I think that's still an unsolved problem. Although Maxime's earlier work definitely helps but there's more to be done, I think, in actually determining the thermalization rate constant.

Daniel Suchet: It also it also goes with the topic reaching a critical mass, in terms of experimental results that leads to some kind of standardization of experiments. That leads to practices which are shared among groups all over the world, and I guess, in this specific topic of hot careers, that's a call which we need to make and to find ways of sharing practices that helps comparing results obtained on different samples and different applications.

Sean Shaheen: So, we need a hot-carrier "round robin" where we have the one material that we send to everyone, and everyone measures it and fits it to a model. Okay! I'm coming up with all sorts of new work for us here but that's good. But I think you're right, the community of hot-carrier researchers and third generation researchers is still pretty small. It's nothing compared to the silicon industry, for instance, or maybe even just III-V industry, so I think that's an important point. If we could grow the community, then we'll get better reproducibility.

Next question: Did anything really unexpected come out? Were there any big surprises or "Aha!" moments that you weren't expecting?

Maxime Giteau: I would say that when we set out to do this work and combine the multijunction with the hot-carrier effect, we didn't really know if it was going to work because, okay, we thin down the top junction and we reduce the thermalization but is it going to be beneficial or is it going to be a loss, all the way? So, seeing that there was a benefit, and it was like an order of magnitude reduction in terms of thermalization rate that we needed. So, it was a huge benefit that was, I wouldn't say unexpected, but that was a very pleasant surprise. We didn't know what to expect from that point of view, so it was on the high end of what we could hope for, for sure.

Sean Shaheen: That's exciting! That's what you hope for.

Samy Almosni: And I would add maybe one point. Especially the first time, when Maxime was showing us the efficiency map as a function of the top junction and we were very surprised to see how resilient the concept was to the non- ideal band gap. We were really surprised because, you see, the sweet spot for multijunction solar cell is very narrow, but in the case of hot-carrier multijunction solar cells, it was very wide range of band gap we could use for the bottom and top junction. I really think it was a very surprising result for us.

Sean Shaheen: So, in several aspects it worked better than you might have anticipated when you started?

Samy Almosni and Maxime Giteau: Yeah.

Daniel Suchet: In a complementary approach, maybe to the band gap discussion which Samy just presented, I was amazed by the resilience on the cooling rate for hot carriers. And as Maxime said before, so far we have not reached material quality which would allow us to overcome the Shockley equation limit based on hot-carrier devices and yet, even with values which are optimistic but realistic as compared to what we know today, we can have a net gain as compared to a usual cold tandem cell. And so, that was quite striking to see that this is where we can have a net gain from having hot carriers, which is something we have trouble getting in other proposals. So that was unexpected there again in a surprising and positive way.

Sean Shaheen: So really in many aspects there's a lot of possibilities moving forward with this work, and hopefully, a lot of researchers in multijunctions and in new materials will see this and say "hey, there's some real possibilities here to bring this to fruition." So that's really exciting!

Along those lines, what are the next steps, from your point of view, to continue the work? Is there more modeling to be done? Well, that's a rhetorical question… I'm sure there's more modeling to be done. But at which point do you incorporate some experimentation? And what are your thoughts on that? Maybe Maxime can start it.
Maxime Giteau: Yeah, I can start. So, we need to figure out how to go from this very general material, agnostic proof of the benefit of the technology to an actual device that would benefit from this, so we need to go step by step. We have devices in which we get hot carriers and we have multijunctions that work really well, so we need to start by putting those two together to really get a multijunction device where we have hot carriers—probably starting with III-V materials. In general, experimental work would really highlight the value of this work, because so far it remains very general.

Sean Shaheen: Now I assume you're not going to get a 40% device on the first try, but what would you consider success with an experimental demonstration?
Maxime Giteau: I would say that, for the hot-carrier community in general, when you make a very good device is when you can show that you are getting extra efficiencies thanks to the hot carriers. So, comparing a device, basically by changing the intensity of lights, you can see that you start getting the hot-carrier effect and really a deviation from the behavior we'd expect from not having hot carriers. So that's really where you break the line between two worlds. Very good, very impressive results.

Sean Shaheen: And did you test some of the nonidealities that you have in the work?
Daniel Suchet: Maybe I can elaborate a little bit on that. I would say there are really different kinds of nonidealities that should be considered. Some nonidealities regarding the design, because it might be that for practical reasons you cannot get the absolute best gap with the absolute best absorption coefficient, and so on and so forth. There is a resilience or ability to deal with defects in the material, nonradiative recombination, and so on, and there's the issue of the actual real operating conditions and the fact that we will not be working under infinite concentration and so on. So, all these things have been taken into account, to some extent, in our work. Absorption is not perfect, concentration is not full sky.

You mentioned the neverending modeling work and my goal here would be to say this notion of resilience needs to be extended. And it needs to be extended on this work. It probably can be extended, also, to other concepts as well. We've been working before on investigating the resilience of intermediate band solar cells and trying to find out how this could be improved, and I really think that this notion—not only to aim at the best possible efficiency in ideal conditions where everything is perfect, but to try and see in more realistic cases how things go—that would be the way I'd like also to keep on pushing after this work.

Sean Shaheen: So, could this formalism apply to any photovoltaic material or technology? Would it make sense to think about, for instance, a silicon perovskite tandem, through the lens of nonidealities?
Samy Almosni: Sure, yeah it would make sense! It would make sense to take a look at those parameters. So, I just wanted to add one more point: we still have to work on the triple junction, which we believe, I think, will gain even more because the absorption in the blue part of especially low bad gap material is very strong, so we should gain even a lot on the thickness of the device. Also, we still didn't study the problem of the selective contact for this type of modeling. And maybe one more point on the interface thermalization that was not taken into account into this work yet: there is a lot of work and a lot of possibility to publish for the community. So, I think we tried to open some doors. Please feel free to go in and just have some easy papers, because it's a very new field and easy to do.

Sean Shaheen: Yeah, the point about the contacts, that's critical. So, you need an energy-selective contact with a very narrow energy band, it's got to be positioned correctly. And I assume, under your work, that the optimal value is different than done earlier, than you find in the textbooks, for instance.

But on your second point, I think that's really exciting and thank you for opening that door! You know, it sounds like there's a huge body of work that could be done here, and hopefully grad students and postdocs and faculty will get to get excited and dive into this space. So, thank you for opening that door for everybody—that's really exciting!

Daniel Suchet: And maybe actually we can just build upon the idea that you bring in. What you mentioned is the ideal, energy-selective contact, right, the perfect selectivity and the perfect location. Well, what about that? What if you can make it semiselective? It's much easier to make and certainly more resilient, more robust in case the system doesn't work exactly as expected.

Sean Shaheen: So that's another nonideality: the ESC layer all by itself. There's three papers to be written on that!

Maxime Giteau: Exactly.

Sean Shaheen: We've all got to get to work. You guys better get to work!

The last question to wrap it up here is more from a personal perspective: How has the work influenced your career and changed your thinking? And, you know, what impact has it had on your career trajectories? And I'll ask Maxime again to start.

Maxime Giteau: Sure. So, the work is very fresh so we don't have really the retrospect to see that—

Sean Shaheen: Ask you again in five years, I guess?

Maxime Giteau: Yeah, I would say, if you really want to see how it has impacted my career. But I can project that coming up with this new concept is something I can very, very easily valorize. It really started here, the idea didn't exist before so, we have a patent coming along with this work so, I think this is really something we can valorize and we can hope also to get funding for going further into this research, which will lead to more publication and overall just pushing forward the topic of high efficiency solar cells, which is very exciting.

Sean Shaheen: Wonderful. Samy, what do you think? How does this change your career?

Samy Almosni: Yeah, so as Maxime said, so far it didn't change my career yet, but at least it has helped me. At first, I had to learn about hot-carrier solar cells because I was really not involved in this type of topic. So, for me, it was very interesting to dig a little bit into the literature and remember about those photon–electron interactions, which is already a good thing for me. And then we finally applied for this patent, which made me learn this process. I think, for the three of us, it's the first time we've applied for a patent as young researchers, so it's taught us many things in that respect.

Daniel Suchet: And I would say, it also gives us a specific position regarding pushing fundamental topics, basic topics, in a field where applications and industry-oriented research tends to be dominant and showing that stemming from basic research, we can go to patent application, we can make proposals which include actual real-life considerations that improve robustness, and which push forward ideas that are quite generic when it comes to improving devices. I think this also strengthened our positions investigating these topics in the field and in other collaborations which we have, which often go a little bit too fast to the short-term research and to improve short-term efficiencies. I think we have shown the relevance of having, also, these longer-term research programs.

Sean Shaheen: So, you can go from fundamental thermodynamics to an application and through a fairly direct route. And collaboration between the academia and industry can be facilitated in that way.

Wonderful. And those are all really, really great points so thanks for those answers. I think with that we'll wrap it up. I want to thank you for your time! First of all, your time in doing the project and writing the paper and submitting to the *Journal of Photonics for Energy*. Then, obviously, today for taking the interview questions. I think this has been incredibly insightful. I've learned so much just in the last half hour, that was not in the paper! We're really getting insights from all of you.

Let me thank you all once again and we look forward to future work and future directions in this area and best success to all of you.
Samy Almosni: Thanks a lot.
Daniel Suchet: Thank you very much!
Daniel Suchet: Thank you very much!
Maxime Giteau: Thank you very much, Sean.

Sean Shaheen: Thank you.