Perspective

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Spaser or plasmonic nanolaser? – Reminiscences of discussions and arguments with Mark Stockman

https://doi.org/10.1515/nanoph-2021-0291
Received June 9, 2021; accepted June 28, 2021;
published online July 13, 2021

Abstract: This essay is my reminiscences of many interesting discussions I had with Mark Stockman over the years, mostly around the spaser, its meaning, and its relationship with plasmonic nanolasers.

Keywords: nanolaser; plasmonics; semiconductor laser; spaser.

The sad news of Mark Stockman's untimely death shocked me greatly. Barely a few months earlier, we were still communicating regularly on the paper [1] we were writing together in celebration of the 10th anniversary of the first experimental realizations [2–4] of the spaser, a landmark invention first proposed by Bergman and Stockman in 2003 [5]!

I first met Mark shortly after the publication of the now well-known Bergman–Stockman paper [5]. As a laser person, I was very intrigued by the idea of the stimulated generation of plasmons. To me, the idea was (and remains) a truly profound new physics concept. My group was working on semiconductor nanowire lasers as part of overall efforts in making ever-smaller lasers. Mark convinced me that their spaser had nothing to do with photons, rather, only confined plasmons were involved. I commented to him that the spaser would probably not be useful for my lasers, but that it would be good as a fully coherent electron source for a new generation of electron microscopes! I subsequently forgot about the concept until a few years later.

In the meantime, our efforts of making smaller lasers using semiconductor nanowires ran into serious difficulties. My colleague, Alan Chin, and I were able to demonstrate the first "nanowire" lasers in the infrared (λ = 1600 nm) [6] using, instead of nanowires, sub-wavelength-size wires of diameters on the order of 500 nm or larger. We had been struggling to shrink the diameter of nanowire lasers because the modes become less confined at smaller diameters [7]. My then postdoc, Alex Maslov, and I considered the concept of coating our nanowires with silver to use the surface plasmon polariton (SPP) modes instead of pure dielectric modes, which are always limited by the diffraction limit. Despite the initial skepticism from my laser colleagues, Alex's simulation showed promising results [8]: our semiconductor-metal core–shell structure was shown to support SPP modes for structures with core diameter as small as 10s of nm. Most important of all, the optical gain in the semiconductor core may overcome the expected large plasmonic loss of the shell. Our next task was to conformally coat a layer of silver onto semiconductor nanowires to form a shell; part of this work was in collaboration with Peidong Yang from Berkeley within our then DARPA NACHOS (Nanoscale Architectures for Coherent Hyper-Optic Sources) project. The task of applying a conformal silver coating onto a nanowire turned out to be quite a challenge!

Despite this challenge, I was optimistic enough about the core–shell SPP idea and the promising simulation results [8]. Based on our nanowire laser results and the core–shell SPP simulation, I started giving talks around 2007 with titles such as "Nanowire with and without a plasmonic shell: how small can a nanolaser be?". One of these talks was my IEEE Distinguished Lecture at the IEEE Benelux Chapter in Eindhoven, the Netherlands. Martin Hill, one of my local hosts, after seeing the announcement of my talk, informed me that he was working on similar concepts. Our meeting changed the trajectory of our approach to plasmonic nanolasers. Martin was also working on a core–shell structure based on a superior approach in terms of fabrication [9]. Instead of using free-standing nanowires as starting structures, he used the conventional lithographical approach to etch small pillars out of a conventional wafer, making his fabrication approach more compatible with mainstream III–V semiconductor processing techniques. Martin's original paper [9] showed lasing in dielectric modes in an otherwise similar core–shell

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structure. Thus, following one of the Silicon Valley mottos: “If you can’t beat them, buy them”, I was lucky enough to get Martin to join my then-new group at Arizona State to work on the core–shell laser in 2008. The result was the demonstration of lasing in a similar structure, but now in an SPP mode rather than an optical mode; the results were eventually published in Optics Express [2] after being rejected by a few other journals because of the apparent similarity with Martin’s earlier laser [9]. During the three to four years of our working on the core–shell SPP laser, I had completely forgotten about the Bergman-Stockman spaser concept [5]. I did not realize the close resemblance of SPP modes with the spaser concept initially. Nevertheless, Mark was very excited when we met again after the Optics Express paper was published [2]. He thought the results in the paper were proof of the spaser concept. I almost immediately expressed my disagreement with his thought. A few times at various conferences, Mark stood up to ask me or one of my students: “why don’t you call it spaser?” My answer was always: “because I don’t believe our device is a spaser.” The continuation of similar exchanges prompted me to consider the similarities between the spaser and what we called a plasmonic nanolaser. I thought a simple dispersion relation would be able to convince Mark of the difference between the two devices. At one of the subsequent conferences, I showed Mark the SPP dispersion curve shown in Figure 1 and asked him: “can we agree on this distinction between the SPP-nanolaser and the spaser?” Mark reluctantly agreed. Although similar arguments never occurred again, I knew Mark was not fully in agreement with me. During our last meeting, when he visited me at Tsinghua in 2019, we had some extended discussions. I showed him again the dispersion curve (Figure 1), but with a more blurred boundary between the SPP-laser regime and the SPASER regime. We finally agreed that there is no difference in the essential physics between the “SPP-laser” regime and the spaser regime (Figure 1). It is all a matter of degree in “plasmonicity”. Looking back, our use of the term "plasmonic nanolasers" was mostly a natural evolution of our previous nanowire-based nanolasers combined with the new ingredient of SPP modes. My reluctance of adopting SPASERs was also related to my incorrect perception of the spaser being at (or extremely close to) the SPP resonance (Figure 1). Mark’s original comment about the absence of photons in the spaser process in our very first meeting probably played a role also. I am pleased that we were able to reach an agreement at that Tsinghua meeting, as it turned out to be our last meeting ever.

Another related discussion I had with Mark involved the acronym spaser itself; the purpose of this discussion was not to challenge their creation of the acronym, but primarily to confirm my understanding of the physics processes involved and to understand the differences and similarities between the spaser and the laser. My understanding is presented in Figure 2. I showed him an earlier version of the lower row of Figure 2 around 2010 (see Fig. 10 in ref. [11]). I emphasized to him that there was no “Stimulated Emission of Radiation” because there are no propagating photons (radiation) being emitted, as he told me in our first meeting; rather, the process is “Stimulated Energy Transfer” from excited emitters to plasmons in metal

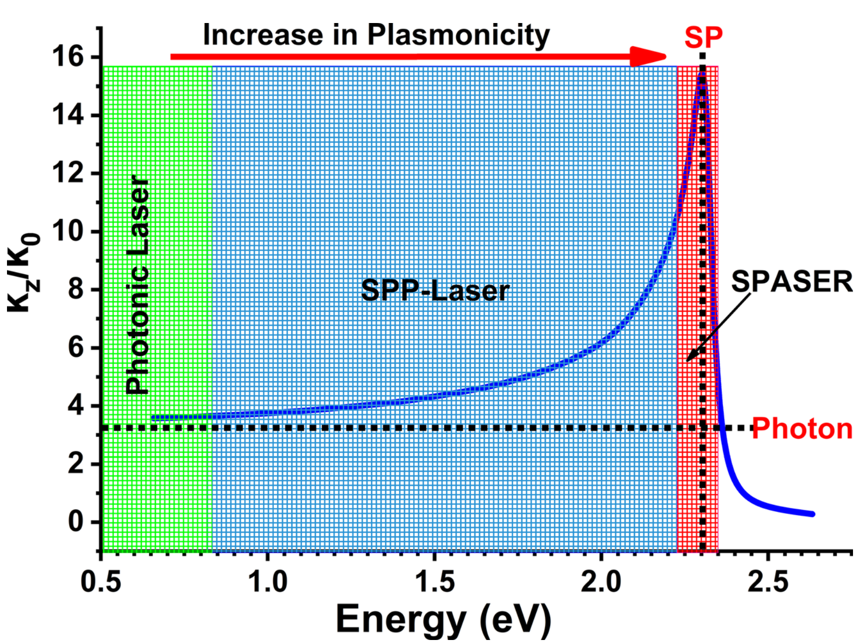

Figure 1: The normalized propagation wavevector versus energy plot for a semiconductor-silver interface; the range of energy is divided roughly into three regions depending on the “plasmonicity”, or the degree of proximity to the surface plasmon resonance (SP). We call the three regions: photonic laser (conventional laser), SPP-Laser, and SPASER. However, as Mark insisted and I initially opposed, there is no essential difference between the SPP-laser and the SPASER (figure adapted from Ref. [10]).

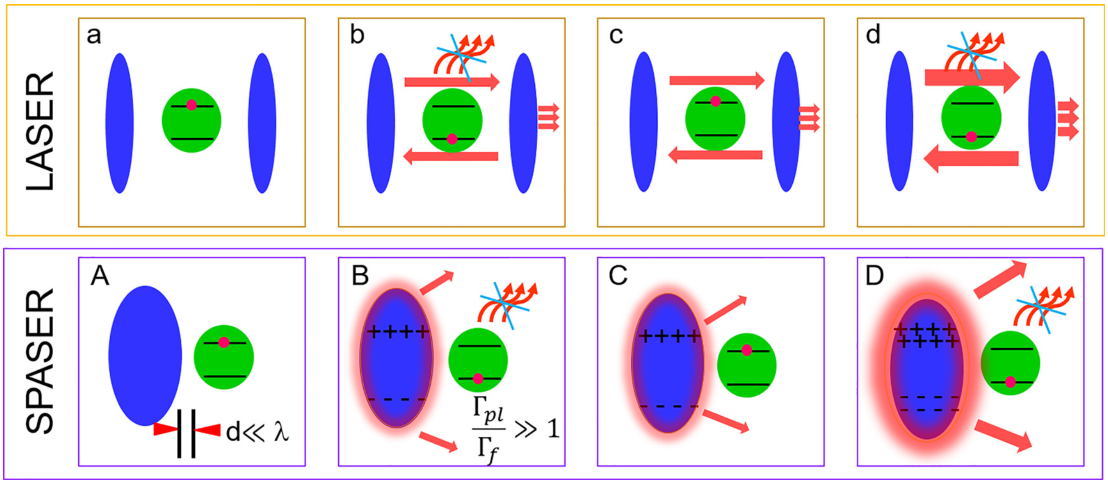

Figure 2: A step-by-step comparison between the conventional laser (top row) and the SPASER (bottom row) highlighting the key differences and similarities. The green circle represents an emitter and the blue elliptical mirrors (top row) or a plasmonic metal object (bottom row). a, A: the excited emitter (electron, the red dot, is in the upper state) in the presence of the cavity (a) or the plasmonic metallic structure (A) with distance, d, much smaller than the wavelength, λ. b, B: Due to the presence of the cavity (b) or plasmonic metal (B), spontaneous emission into cavity mode represented by the horizontal red arrows (b) or spontaneous energy transfer to the metallic structure to excite surface plasmon mode (the red cloud around the blue metallic structure) (B) dominates over spontaneous emission into free space as indicated by the crossed emission wiggles. Γ_{pl} and Γ_f stand for the rate of energy transfer to plasmons and that of emission into free space, respectively. c, C: The emitter is re-excited in the presence of pre-existing cavity mode (c) and plasmonic mode (C). d, D: The pre-existing cavity mode (d) or plasmonic oscillation (D) forces (stimulates) the excited emitter to emit the same photon (d) or plasmon (D) instead of emitting spontaneously into the free space. The continuation of step d (stimulated emission of radiation) or D (stimulated energy transfer) eventually leads to lasing (d) or spasing (D). For the Spaser in the lower row, we also indicate the possible far-field emission from the plasmons (red straight arrows in B, C, and D). It is important to note that such far-field originates from the plasmon oscillation from the metallic structure, rather than from the emitter directly. The bottom row is based on an earlier version in [11].

structures (Figure 2B, C, and D) and occurs exclusively in the nearfield, with no photon emission. Although I was joking that we should use the acronym SPASET for Surface Plasmon Amplification through Stimulated Energy Transfer, instead of spaser, I could tell that Mark was not pleased with my comment! I also expressed my opinion in my 2010 tutorial [11] by using a version of a figure similar to the lower row of Figure 2, mostly to highlight the new aspects of the spaser. However, my intention was never to change the acronym as I agree that the spaser is a better acronym

Figure 3: Mark Stockman and I were having a conversation during our last meeting at Tsinghua University on July 12, 2019.

than SPASET in many senses, especially in conveying the similarity to the laser. Although I did not mention the SPASET term after our first discussion, I did show him the comparison between the laser and the spaser as presented in Figure 2 at our Tsinghua mini-symposium. I was happy that we were in complete agreement about the differences and similarities between the laser and the spaser.

Mark was not only a brilliantly creative scholar but also an extremely warm and generous person. Many of us will forever miss his big hugs. Over the years, we had many discussions and arguments, sometimes quite heated, especially at the beginning of our plasmonic nanolaser research. However, we remained very friendly and enjoyed many meals together. I was very impressed with his love of good wines and lobster tails (I remember he ordered them at least twice over the years!). I was especially happy when he told me that he was coming to China in the summer of 2019 because that allowed me a chance to meet and discuss spaser physics with him again. We, together with Renmin Ma, held a mini-symposium to celebrate the first experimental observations [2–4] of spasers and plasmonic nanolasers at Tsinghua (see the picture in Figure 3). Unfortunately, that meeting became our last one.

On a lighter and more humorous note, one of our last interactions involved reimbursement of his flight ticket for that last trip to China via wiring the money to his US account. After a few rounds of emails with an accountant, who insisted that he must use his real name, Mark was frustrated and forwarded me their emails, attaching the image of the instructions in Chinese. The instructions in Chinese state that the recipient must be a real person and the name cannot be a "bank", "stock", or "security". I immediately realized the problem, resulting in the following email exchange:

> Me: Mark, I found out what was wrong. It is hilarious! You have the wrong family name that contains "Stock".
> Mark: Gosh, you are right! It is hilarious! Please explain to them that I am not traded on stock markets yet as of now.

Let us hope that, with our collective efforts, one day a company making spasers WILL be traded on stock markets!

Acknowledgment: The author thanks Alan Chin for his careful reading of the paper and for making corrections.
Author contributions: All the authors have accepted responsibility for the entire content of this submitted manuscript and approved submission.
Research funding: None declared.
Conflict of interest statement: The authors declare no conflicts of interest regarding this article.

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