

Mark Stockman: Evangelist for Plasmonics

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ABSTRACT: Mark Stockman was a founding member and evangelist for the plasmonics field for most of his creative life. He never sought recognition, but fame came to him in a different way. He will be dearly remembered by colleagues and friends as one of the most influential and creative contributors to the science of light from our generation.

■ A SHORT BIOGRAPHY OF PROFESSOR MARK STOCKMAN

Mark Stockman was born on the 21st of July 1947 in Kharkov, a major cultural, scientific, educational, and industrial center in the mainly Russian-speaking northern Ukraine, a part of the Soviet Union at that point in time. His father, Ilya Stockman, was a mining engineer by training, who fought in World War II and became a highly decorated enlisted officer. After the war, Ilya Stockman embraced an academic career and eventually became a Professor at the Dnepropetrovsk Higher Mining School. He was from a Cantonist family descended from Jewish conscripts to the Russian Imperial army, who were educated in special “canton schools” for future military service.

Mark was an avid reader at school, and it was the undergraduate textbook on applied mathematics by Yakov Zeldovich, the famous theoretical physicist, who played a crucial role in the development of the Soviet Union’s nuclear bomb project that turned his attention to physics in a serious way.

Following successful participation in the national school physics competition, Mark was accepted into a highly selective institution for gifted children in Kiev known as the Republican Specialized Physics and Mathematics Boarding School. The school was established by the father of Soviet cybernetics Victor Glushkov. Mark left his family in Dnepropetrovsk and moved to Kiev as a boarding student. Upon graduating from the school, he successfully applied to the Physics Department of Kiev State University, aided by his reputation as a top student and links between the school and university academics: for a Jewish boy with no family connections, to enter this prestigious university in the Ukrainian capital was a formidable challenge in the Soviet Union. However, after his second year, feeling uncomfortable at the University in Kiev, Mark decided to leave the blessed city for Novosibirsk State University far away in Siberia where a more cosmopolitan atmosphere prevailed at that time. He studied for a diploma in the Institute of Nuclear Physics where after graduation he became a researcher registered as a Ph.D. student. He defended a dissertation on collective phenomena in nuclei under the supervision of Russian theoretical physicists Spartak Belyaev and Vladimir Zelelevinsky. While a Ph.D. candidate, Mark met and married Branislava Mezger, a junior research scientist in biomedicine, and in 1978 they had a son, Dmitry.

After a few years in the Institute of Nuclear Physics, Mark became disillusioned with nuclear physics, where research

projects involved large groups and implied very long experimental cycles. He moved to the neighboring Institute of Automation and Electrometry in Novosibirsk to work on the fundamentals of nonlinear optics with Sergey Rautian. He habilitated in 1989 with a D.Sc. dissertation on nonlinear optical phenomena in macromolecules.

Around this time, the political regime in the Soviet Union softened and the Iron Curtain was lifted. In 1990, on the invitation of Professor Thomas F. George, Mark was permitted to leave Russia with his family to take a research post at the State University of New York at Buffalo. He later followed Professor George to Washington State University and eventually settled with his family in Atlanta, Georgia, as Professor of Physics at Georgia State University. In 2012, he became the founding director of the Center for Nano Optics at Georgia State. Mark traveled widely, but never returned to Russia.

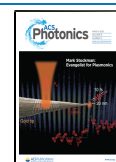
Professor Mark Stockman passed away in his beloved Atlanta, Georgia, U.S.A. on Wednesday, November 11, 2020. To honor Mark, a Virtual Issue organized by *ACS Photonics* features articles from *ACS Photonics*, *ACS Nano*, and *Nano Letters* authored by friends and colleagues of Mark.



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Mark Stockman posing a question to a speaker during a NANOP 2019 conference on Nanophotonics. Photo courtesy of PremC.

■ PLASMONIC LENS

Javier Aizpurua. Mark Stockman contributed intensely, sharply, and brilliantly to many subfields within nanophotonics. In the era when calculating the plasmonic response of a complex nanostructure was still a challenge, greatly mitigated nowadays by the capabilities of software tools available to solve Maxwell's equation in almost any architecture, Mark proposed several canonical plasmonic structures that had a strong impact in the further development of plasmonic nanoantennas later on. Among them, Mark and colleagues proposed a canonical plasmonic nanolens made of a self-similar chain of particles with decreasing size,¹ as a particularly efficient field-enhancing nanoantenna. The principle of this lensing effect was based on the concept of cooperative, or cascade, plasmonic enhancement. Within this simple concept, Mark et al. designed a set of plasmonic nanoparticles with gradually reduced size, where each smaller nanoparticle progressively localized the electromagnetic field induced by the immediately larger particle into a more confined space, and thus producing a much larger enhancement than the nanoparticles by themselves. This concept of a cascade plasmonic lens was successfully implemented experimentally by several groups in the community, which proved that the concept could be practically used in surface-enhanced spectroscopy and microscopy.^{2,3}

The near-field cascade effect introduced by Mark et al. can be understood as the discrete version of his adiabatic compression of a plasmon traveling along a tapered metallic guide toward its apex.⁴ When the plasmon oscillation is constrained into smaller transverse dimensions, the associated local field unavoidably gets enhanced. One can also understand the localization and enhancement of optical fields in atomic protrusions⁵ as an extreme case of the cascade effect also occurring at the atomic scale. Metal atoms protruding from a metallic nanostructure can be considered as smaller "polarizable particles" subjected to the local field induced by the larger hosting nanoantenna (particle, tip, gap, etc.), and thus, the original local field at the hosting nanoantenna can be further enhanced and localized around single protruding atoms due to this atomic-scale cascade effect.⁶ This type of architecture, recently termed a "picocavity",⁷ has allowed for confining light at atomic dimensions, pushing the limits of nanophotonics into the realm of picophotonics and emphasizing the relevance of the inspirational work by Mark on the plasmonic lens concept.

■ ACTIVE PLASMONICS

Harry A. Atwater. Mark Stockman, like the field of plasmonics, was extremely active. No speaker at a photonics conference would go unquestioned or unchallenged whenever Mark was sitting in the front row. But challenging the community was Mark's way of conveying his infectious love and enthusiasm for photonic science. His penetrating and deep scientific questions were also the ultimate compliments he could pay his colleagues and almost always gave a gift of insight or taught a physics lesson to the recipient. Those who also saw him or joined him on the ski slopes were surprised at how active Mark could be while navigating a black diamond run.

For some, the field of plasmonics has always had a dark side, arising from the inherent Ohmic losses of our beloved plasmonic

materials, limiting the achievable quality factor or degree of coherence of nanoconfined optical modes. But Mark's signature achievement of the plasmonic spaser embodies the old adage that one person's loss is another person's gain; in the case of the spaser, it is the lossy metal juxtaposed with a gain medium that is the key to making a subwavelength-scale nonradiative stimulated emission source. It was Mark's genius to see where others did not that this concept was not only interesting, but possible.

Mark's work on active plasmonics and the nanoconfinement of light was tremendously stimulating to me and my research group, who from the beginnings in 2000 shared Mark's conviction that combining subwavelength confinement with active media opens a myriad of possibilities for nanoscience. In our lab, this led to the first prototype for a subwavelength-scale nanoparticle waveguide,⁸ an all-optical plasmonic modulator,⁹ a transistor-like modulator for light,¹⁰ plasmonic light trapping in solar cells,¹¹ and achievement of unity-order modulation of the refractive index under electrical control,¹² which later gave rise to electrically reconfigurable reflectarray metasurfaces,¹³ both using lower loss plasmonic materials¹⁴ such as conducting oxides and tunable perfect absorbers,¹⁵ as well as phased arrays for beam steering¹⁶ of infrared radiation using actively tunable graphene elements. Mark's insistence to me over a breakfast in Japan once about the possibility of an infrared spaser composed of graphene active elements inspired me to begin exploring excited state radiative decay processes in graphene under ultrafast optical excitation. This effort, while not yet yielding a spaser, has recently demonstrated ultrabright mid-infrared spontaneous emission by plasmon generation from excited state relaxation in graphene.¹⁷ His work will continue to inspire students and researchers long after his passing.

■ NANOCAVITIES

Jeremy J. Baumberg. The energy and stimulation of Mark's influential explorations underpinned my move into plasmonics and, specifically, the approach of stacking nanoparticles to amplify optical field confinement¹⁸ and exploring spaser concepts.¹⁹ Our development of nanoparticle-on-mirror (NPoM) geometries and the elaborations of these to trap optical fields efficiently into nanogaps that can be mass produced uses many of these ideas of antenna and gap plasmon mode.^{20,21} His early publications on trapping light into random gaps on rough surfaces²² and tapered plasmon waveguides²³ helped clarify the cascades of confinement possible to couple free space photons into tiny gaps and motivated our search for the reliable field enhancements of 1000 now obtained in these sub1 nm nanogaps. We continue to explore placing of light emitters into such gaps, which has many surprising consequences with forerunners in Mark's papers. For instance, the selection rules of emitters are completely changed in such nanogaps.^{24,25}

Taking his ideas further opened up the understanding that another stage of confinement is possible in confined plasmonics, where the light can be trapped around single metal atoms⁷ or small metal defects.^{26,27} This opens up the interplay of surface science to plasmonics and allows the dynamics of single atom movements to be trapped with light,²⁸ far beyond what was thought possible a decade ago. Currently we are developing this into single-atom optical switches and studying catalysis a molecule at a time. Many intriguing futures are opened up by this focus since such atomic motions are fast ($<100 \mu\text{s}$), and the interactions and optical forces are so far not resolved. The extreme stability of these NPoM constructs allows detailed

investigations of these properties and dynamics, opening up the realm of picophotonics.

■ NANOFOCUSING OF ELECTROMAGNETIC RADIATION

S.I. Bozhevolnyi. Nanofocusing of electromagnetic radiation, that is, progressive reduction of the cross section of optical modes (far beyond the diffraction limit) propagating along tapered waveguides, requires the existence of waveguides supporting the corresponding (progressively confined) propagating optical modes, that is, requires nanoguiding. Although the possibility of guiding strongly confined radiation by surface plasmon-polariton (SPP) modes was made apparent over 50 years ago,²⁹ the concept of nanoguiding has been widely recognized much later, following the theoretical demonstration in 1997 of the possibility of subdiffraction SPP guiding in cylindrical metal nanowire or nanohole configurations.³⁰ Practically simultaneously with the latter, the idea of superfocusing with SPP modes was introduced by considering SPP propagation in wedge-like metallic structures.³¹ It was, however, not until the publication of the seminal paper by Mark Stockman on nanofocusing in 2004²³ that the idea of nanofocusing was truly accepted and taken to practice.³²

The paper by Mark has produced an enormous impact on the still young and explosively growing plasmonics community due to several profound insights opening fascinating perspectives for further explorations. First, Mark has clearly differentiated nanofocusing as a transport phenomenon in which the radiation energy is progressively concentrated during the SPP propagation along a tapered plasmonic waveguide from the concentration of radiation energy in nanoscale volumes by illumination of nanostructures (nanoantennas). Importantly, the requirement of adiabaticity in nanofocusing has been emphasized with the consequence of SPP propagation slowing down and asymptotic stopping, an important feature of adiabatic nanofocusing resulting in giant concentration of energy. Finally, a practical example of a silver cone was analyzed in detail, convincingly demonstrating the possibility of efficient coupling of far-field radiation to the near-field zone and boosting up the field intensity at the cone tip by 3 orders of magnitude. The “nanofocusing” paper by Mark can serve as an ideal (albeit very rare) case when practically all potential applications promised in the conclusion (“adiabatic nanofocusing promises to find various applications ... in particular, for probing, spectroscopy, detection, and modification on the nanoscale in physics, chemistry, biology, electrical engineering”) ²³ have indeed been realized.³²

Concluding, it should be mentioned that Mark has also introduced a hybrid plasmonic configuration that simultaneously displays the properties of a resonant nano-optical antenna and nanofocusing: a tapered chain of metal nanoparticles whose diameters decrease along the chain.¹⁸ This “snowman” configuration features an ingenious combination of a large nanoparticle, which both strongly interacts with an incident far-field radiation and is coupled to progressively smaller nanoparticles, delivering thereby the radiation energy from the first large nanoparticle to a nanoscale gap between the last two nanoparticles. As time goes by, the relative significances of different areas of nano-optics, plasmonics, and ultrafast science influenced by Mark’s insights will probably change, but his inception of nanofocusing will definitely remain one of the most significant contributions.

■ FROM PLASMON HYBRIDIZATION TO MIE-TRONICS

Mark L. Brongersma. Mark Stockman has played a critical role in developing our fundamental understanding of the way in which metallic nanostructures can be used to concentrate and manipulate light at the nanoscale through the excitation of surface plasmons.³³ Early work in the Brongersma group on plasmonic waveguides,³⁴ sources,³⁵ and plasmonic resonator antennas³⁶ was certainly inspired by Stockman’s unique ability to formulate basic plasmonic design principles that can capture the essence of their operation. His research on plasmon hybridization in metallic nanoparticle dimers³⁷ continues to stimulate new nanophotonics developments. In these systems, hybridization can lead to a desirable redistribution of optical fields and an intense light concentration. The extreme light confinement renders the dimer’s optical response also very sensitive to minute changes in gap size. Very recently, we harnessed this physics to create a nanoelectromechanical system to achieve low-power (~ 1 fJ/bit), high-speed (~ 10 MHz) manipulation of optical signals by dynamically modulating the gap of a dimer at the ultimate, atomic scale (~ 1 nm).³⁸ The availability of low-power, electrically tunable optical elements with a deep-subwavelength footprint can have a transformative impact on the development of dynamic flat optics and quantum information processing.

The large body of plasmonics research has shown that individual metallic nanostructures and assemblies thereof display a tremendous design flexibility. This flexibility underlies many scientific and technological triumphs in the nanophotonics field. In the hopes of reproducing the success of the plasmonics field, the Brongersma group was very eager to see whether the highly tunable optical resonances in high-index semiconductor nanostructures^{39–41} could bring similar advantages. Following Stockman’s early work in plasmon hybridization and with his great encouragement, the group started studying the optical coupling of semiconductor nanostructures and exploring the similarities and differences between the resonances in metallic and semiconductor nanostructures. This research has now grown into its own field of Mie-tronics, with many opportunities and applications complementary to plasmonics.⁴²

■ SERS IDENTIFICATION OF BACTERIAL PATHOGENS

Jennifer A. Dionne. As a first-year graduate student and newcomer to the field of plasmonics, I had the honor of hearing Mark Stockman present his research on plasmonic focusing, a single author paper published in *Phys. Rev. Lett.* in 2004.²³ Mark described a new concept for the “delivery and concentration of optical radiation energy on the nanoscale”, which had previously been “formidable because the wavelength of light is on the microscale, many orders of magnitude too large.” In many ways, this paper fueled my passion in nanophotonics for health and sustainability.

Inspired by Mark’s pioneering work to concentrate light at the nanoscale, we have been developing a low-cost platform to rapidly identify bacterial pathogens, their drug susceptibility, and their minimum inhibitory concentration. Bacterial infections do not often make headlines, yet they are responsible for more deaths than AIDS and many cancers and rank among the most expensive medical conditions to treat. Traditional bacterial identification and antibiotic susceptibility testing can span hours

to days, even in state-of-the-art laboratories using the most advanced technologies, delaying the use of targeted antibiotics, leading to overuse of broad-spectrum drugs, and accelerating the spread of infectious disease. According to the CDC, approximately 50% of patients are unnecessarily treated with antibiotics or are treated with the wrong type or dose, a prime contributor to the evolution of antibiotic-resistant pathogens and one of the most significant global threats to community health, sustainable food production, and the economy.

Our technique is based on surface-enhanced Raman scattering (SERS), inelastic photon scattering resulting from molecular vibrations, with single-cell sensitivity enabled by plasmonics and nanophotonics. Because of the unique molecular structure of a pathogen, each bacterial species has a specific SERS signature that reflects the identity of the bacteria and its real-time response to various environmental conditions. Using single-cell SERS, we have trained a convolutional neural network (CNN) to classify over 30 pathogenic species and strains from their single-cell Raman spectra, representing over 95% of patient infections at Stanford Hospital and the majority of infections in intensive care units worldwide. Even on low signal-to-noise spectra, we achieve average species and strain identification accuracies exceeding 82% and antibiotic treatment identification accuracies of $97.0 \pm 0.3\%$. We also show that this approach distinguishes between methicillin-resistant and -susceptible isolates of *Staphylococcus aureus* (MRSA and MSSA) with $89 \pm 0.1\%$ accuracy.

These results have been validated on clinical isolates from 50 patients; with just 10 bacterial cells from each patient, we achieve treatment identification accuracies of 99.7%. We have also extended the approach to liquid SERS of pathogens in order to understand pathogen–environment interactions, such as bacteria–antibiotic interactions. By optimizing the plasmonic nanoparticle geometry and nanoparticle-to-bacteria concentration ratios in liquid, we obtained large, uniformly enhanced SERS signatures from bacterial samples of *Escherichia coli*, *Serratia marcescens*, *Staphylococcus aureus*, and *Staphylococcus epidermidis* (compared to no signal with bacteria in water). Interestingly, this approach can provide information about the pathogens' antibiotic susceptibility and minimum inhibitory concentration, facilitating tailored and personalized antibiotic treatment.

Owing in large part to Mark's pioneering work, Raman spectroscopy now ranks among the most promising biomedical diagnostic tools. The technique provides a specific and sensitive fingerprint of biomarkers, with the potential for rapid, multiplexed analysis in a portable, low-cost platform. In the past, Raman spectroscopy has suffered from relatively low signal-to-noise ratios, complexity in spectral interpretation, and a lack of an efficient workflow from sample collection to spectral acquisition, challenging clinical translation. Today, Mark's legacy in nanophotonics, coupled with advances in machine learning and bioprinting, are enabling breakthroughs that can overcome each of these challenges.

■ NONLINEAR ULTRAFAST PLASMONICS

Harald Giessen. Mark Stockman tremendously influenced our efforts in the field of ultrafast plasmonics. During a guest professorship at the University of Stuttgart in the fall of 2008, we worked on ideas of how to coherently control plasmon oscillations using an ultrafast dual-pump and probe scheme. The idea was to generate a plasmonic charge oscillation in nanoantennas, which could be coherently enhanced or sup-

pressed by a second coherent pump pulse. A probe pulse after a few tens of femtosecond probe beam impinged at an angle and created third harmonic light, whose spatial direction could be selected in a four-wave mixing fashion. This allowed monitoring of the constructive as well as destructive effects of the second pump pulse on the coherent plasmon polarization. The paper was published with grad student Tobias Utikal as first author, who closely worked with Mark Stockman as second author who worked out the theory.⁴³

This work inspired many follow-up efforts in our group, including research that was geared toward understanding the influence of metallic nonlinearities and the third harmonic system that came from neighboring dielectric layers.⁴⁴ Mark Stockman's original idea of concentrating light in the nanofocus of a nanotip and measuring the nonlinear optical response was realized by using bowtie nanoantennas and placing different dielectrics into the gap.⁴⁵ It turned out that the metal itself already had an extremely high nonlinearity and was contributing nearly as much to the nonlinear signal as the dielectrics. The ultrafast nonlinearity could also be utilized in nonlinear sensing schemes.⁴⁶ Detailed investigations of material and polarization dependence, more complex hybrid geometries, band-structure dependencies of the used metals, utilizing double resonant conditions, nonlinear Fano schemes, and chiral nonlinearities, as well as creating ultranarrow resonances using dielectric fiber cavities, were carried out in our group,^{47–52} all resulting from the initial stimulus and inspiration of Mark Stockman.

■ HYBRIDIZATION PERSPECTIVE ON PLASMONIC HOMODIMERS

Naomi Halas and Peter Nordlander. We got to know Mark in 2002, during the infancy of plasmonics, and met frequently at workshops and conferences several times every year for many years afterward. During conference sessions, we often began lively discussions that sometimes would extend through dinner and well beyond. At the time, Naomi was investigating the photophysical properties of nanoshells, with their tunable plasmon resonances,⁵³ and their applications in surface-enhanced Raman scattering⁵⁴ and biomedicine.⁵⁵ Mark was very excited about the promise and potential of nanoshells for photothermal cancer therapy.⁵⁶ Peter was working on first-principles TDLDA modeling of plasmonic systems and, in particular, on the tunability of plasmonic nanoshells.^{57,58} This was also a topic of great interest for Mark who was particularly interested in how quantum mechanical effects may limit the field enhancements and plasmonic couplings in thin nanoshells and in ultranarrow plasmonic junctions. A particularly inspirational paper by Mark was his study of self-similar chains,¹⁸ a topic we discussed very frequently. This led to our sole collaborative work, where we applied plasmon hybridization theory to analyze the properties of two adjacent nanoparticles: a plasmonic homodimer.³⁷ At the time, our primary concern was actually how the plasmonic response of a sphere would scale with decreasing size and if quantum mechanical effects would introduce a minimum nanoparticle size beyond which the self-similarity argument would be invalid. Our TDLDA studies clearly showed that nonlocal and quantum size effects introduced such a lower limit, but our model was based on the jellium approximation and left open the question of how an atomistic description would influence, and most likely increase, this minimum size. That issue is still unresolved. However, the issue of the possible role of quantum mechanical effects on the coupling between nanoparticles, although difficult, was even-

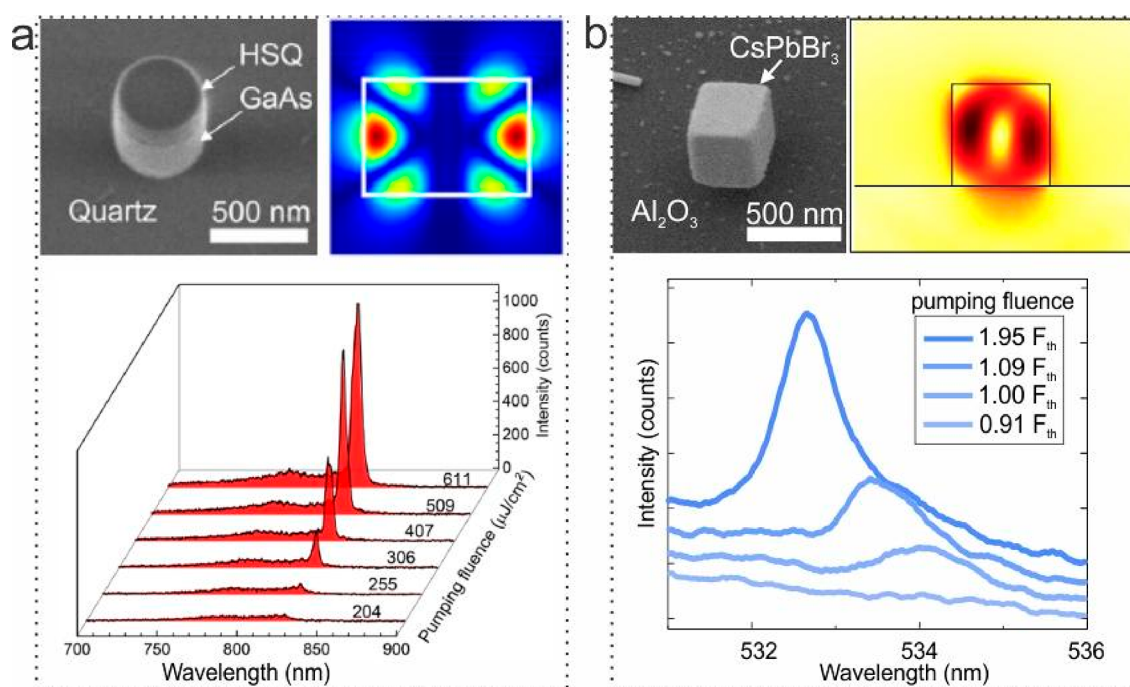


Figure 1. Two recent demonstrations of Mie-resonant nonplasmonic nanolasers. (a) Scanning electron image (SEM) of GaAs nanoscale cylinder covered by hydrogen silsesquioxane resist (HSQ); calculated electric near-field distribution of the cylinder at the lasing wavelength; and evolution of the emission spectrum of the resonator with diameter $D = 500$ and height $H = 330$ nm at different pumping fluence. Reprinted with permission from ref 63. Copyright 2020 The American Chemical Society. (b) SEM image of CsPbBr₃ perovskite nanocube and calculated electric near-field distribution at the lasing wavelength, and evolution of the emission spectrum of the perovskite nanocube with the side length of 310 nm at different pumping fluence. Reprinted with permission from ref 64. Copyright 2020 The American Chemical Society.

tually resolved with a first-principles TDLDA study of the plasmonic properties of a homodimer.⁵⁹ This work inspired a joint experimental–theoretical investigation of coupled pairs of nanoparticles: since no two nanoparticles are identical, this study addressed the broader case of heterodimers: mismatched nanoparticle pairs.⁶⁰

■ MIE-RESONANT SUBWAVELENGTH LASERS

Sergey V. Makarov and Yuri Kivshar. One of the great achievements of Mark Stockman was the suggestion of a spaser or plasmonic laser that confines light at a subwavelength scale.¹⁹ Spaser is a nanolaser with a very small footprint that can be modulated quickly and, combined with their small footprint, becomes a candidate for on-chip optical computing. Such on-chip light sources are critical for realizing integrated photonic circuits.

The suggestion of a spaser inspired many efforts to make its dielectric analogue. So far, the size of the smallest semiconductor lasers has been limited by a few micrometers. Further reduction of sizes to the nanoscale is really challenging due to radiative losses. Despite spasers help to reduce radiative losses and sizes,^{19,61} they also introduce parasitic Ohmic losses originating from their metallic parts.

Recently, active all-dielectric metaphotonics has emerged as a new and rapidly expanding field of subwavelength optics that is based on electric and magnetic optical Mie resonances excited in high-index dielectric nanoparticles.⁶² Similar to the spaser suggested by Mark Stockman,¹⁹ a dielectric nanolaser provides a tight confinement of local electromagnetic fields combined with low losses. Its physics is driven by multipolar interferences available in single dielectric nanoparticles supporting Mie resonances that provide novel tools to tailor the properties of

light at the subwavelength scale, often not available with plasmonics.

On the way to miniaturization of nanoscale laser sources, two major problems have to be solved: (i) optical gain of many materials is not high enough to compensate for losses in the nanosystem at room temperature; (ii) radiative losses in subwavelength nanocavities are usually very high. In two recent papers,^{63,64} a novel approach to overcome these challenges was demonstrated and it is based on the recent achievements in metaphotonics (Figure 1).

Namely, the far-field engineering allowing to design the so-called supercavity mode⁶⁵ was successfully employed for GaAs cylinders to suppress radiative losses as much as possible for subwavelength lasers without any metallic components.⁶³ The other strategy employed to go to the visible frequency range and room temperatures was the use of single-crystalline perovskite nanocuboids possessing low-order Mie modes as well as extremely high gain, even without cooling.⁶⁵ Both of these approaches are based on the excitation of Mie resonances in nonplasmonic resonators that correspond to a deeply subwavelength regime when the characteristic dimensions of the nanocavity become a fraction of the emitted wavelength of light. Further assembly of such particles into one- and two-dimensional structures opens additional possibilities for creating highly directional nanoscale lasers with tailorable directivity.

These recent developments in nanoscale optical control suggest many opportunities for subwavelength signal processing, optical computing, and biosensing.

■ ATTOSECOND NANOPHYSICS AND PETAHERTZ OPTOELECTRONICS

Matthias F. Kling and Ferenc Krausz. In the year 2007, Mark Stockman paid his first visit to the Max Planck Institute of Quantum Optics (MPQ). We immediately started discussions about the potential of attosecond spectroscopy for tracing electron dynamics in nanoscale materials. Within the one month that he spent at MPQ at the time, he initiated and worked out a proposal for attosecond nanoplasmonic microscopy,⁶⁶ which might have inspired the birth of attosecond nanoscience. The technique, also referred to as ATTO-PEEM, combines attosecond streaking spectroscopy and photoemission electron microscopy (PEEM). It involves a pump pulse inducing near-fields on a (metal) nanostructure. The near-fields are probed via photoemission using a time-delayed attosecond extreme-ultraviolet (XUV) pulse. Momenta of XUV-released electrons can be measured with a time-of-flight PEEM, providing nanometer spatial resolution. The space and time-dependent momenta sample the surface near-fields at the time of the electrons' release. His initial work, where Mark also pointed out important differences to conventional attosecond streaking, has led to a wealth of further studies on probing plasmons and other electronic excitations in nanostructures with attosecond spectroscopies (see, e.g., ref 67 and references therein). Tremendous efforts were directed toward implementing ATTO-PEEM experimentally. Despite significant progress,^{68,69} the technique is still awaiting its realization. This story exemplifies the way Mark has been making contributions to the emergence of new fields of science. In the case of attosecond nanophysics, a visionary concept has set the stage for interactions with many interested researchers, giving rise to the formation of a new community.

During the coming years, upon his repeated visits and via all possible communication channels, we discussed with Mark our first experimental observations of unusual behavior of dielectrics in strong laser fields. These observations included field-induced currents in silica-based Schottky junctions,⁷⁰ reversible changes in extreme-ultraviolet absorptivity and near-infrared reflectivity of dielectrics in strong fields,⁷¹ and characteristic cutoff energy modification for electron emission from silica nanoparticles above a threshold intensity.⁷² Searching for a possible interpretation of the results, Mark developed his revolutionary concept and basic theory for the semimetallization of solids in strong fields.^{73,74} This ground-breaking idea is a cornerstone of a field now known as petahertz optoelectronics.⁷⁵ In recent years, he has extended his work to strong-field interactions with other types of solids, including 2D materials such as graphene⁷⁶ and 2D transition metal dichalcogenides (TMDs).⁷⁷ As an example, he introduced strong-field valleytronics in TMDs and the use of such phenomena for information storage and processing in petahertz optoelectronics.⁷⁷ His pioneering work is stimulating a wealth of strong-field studies on solids, including probing their response via high-harmonic spectroscopy.⁷⁸ While petahertz optoelectronics is in its infancy, it holds the promise to revolutionize electronics by increasing information processing frequencies to the ultimate limit, given only by the speed of light. Mark Stockman's stipulation of new research directions in ultrafast and strong-field science has provided and continues to provide a strong inspiration for us and a steadily growing community in these areas.



Mark Stockman skiing with Matthias King. Photos courtesy of Matthias Kling.

■ ADIABATIC COMPRESSION OF SURFACE PLASMONS

Stefan Maier. The description of adiabatic compression of surface plasmons,²³ a unique way of channeling electromagnetic energy to nanoscopic dimensions, constitutes another inspiring work by Mark Stockman, which has had a significant impact beyond the nanophotonics community. It put forward a potentially highly efficient way for channeling radiation from the far- to the near-field, with a concomitant enhancement of the local electric field by up to 3 orders of magnitude. Intriguingly the original work predicted a wealth of applications in an interdisciplinary context, "in particular, for probing, spectroscopy, detection, and modification on the nanoscale in physics, chemistry, biology, electrical engineering" (quote from Mark's paper).

While various techniques for nanofocusing with surface plasmons were concisely reviewed by Gramotnev and Bozhevolnyi³² some time ago, it is the broadband nature of the adiabatic compression described by Mark Stockman that has enabled intriguing applications in spectroscopy, for example, for the exploitation of hot electron effects⁷⁹ and nanoscale chemical mapping.⁸⁰ The work has further inspired extension toward metamaterials and metasurfaces, such as the "trapped rainbow" light storage concept⁸¹ and extensions of spoof plasmon polariton waveguides to fiber waveguide geometries.⁸² The emerging area of topological plasmonics⁸³ should be a fertile ground for the further development of this pioneering work.

■ PLASMONIC NANOGAPS

Maiken Mikkelsen. In pursuit of the vision to realize ultrafast nanoplasmonics, nanogap structures have been studied extensively.²⁰ To experimentally realize nanogap structures with ultrasmall mode volumes in a reproducible manner, often a nanoparticle-on-mirror geometry has been utilized. For example, silver nanocubes have been placed 1–10 nm from a metal film, where a dielectric spacer layer occupies the gap region and defines the gap distance. Exciting such structures with light on resonance has shown to result in ~100-fold enhancement in the local electromagnetic field intensity in the gap region.⁸⁴ When emitters are embedded in this gap region, their local density of states is strongly modified, resulting in enhanced spontaneous emission rates up to 1000-fold for ensembles of fluorescent dye molecules.⁸⁵ Additionally, when colloidal quantum dots were embedded, this nanoplasmonic structure was shown to enable ultrafast spontaneous emission rates with lifetimes of less than 11 ps, limited by the detector resolution.⁸⁶ For single quantum dots or nitrogen-vacancy centers in diamond, the structure even enables an ultrafast⁸⁷ or ultrabright single photon source.⁸⁸ By tuning the plasmon resonance to overlap with the excitation wavelength, instead of

the emission wavelength, as done above, the absorption rate of the embedded emitters can be strongly enhanced, resulting in up to a 30000-fold increase in the fluorescence of dye molecules for the same excitation power,⁸⁹ which could play an important role for, for example, point-of-care biosensors.⁹⁰ Additionally, depending on the concentration of embedded emitters and their coupling to the cavity mode, strong coupling between the emitters and cavity mode can be observed both from large-area metasurfaces and individual nanogap cavities.⁹¹

■ SURFACE ENHANCED RAMAN SPECTROSCOPY

Martin Moskovits. A decade later, largely through the work of Mark Stockman, it became clear that for fractal nanoparticle aggregates, the self-similarity of the colloidal aggregates produced a kind of symmetry-breaking that, in general, led to a highly heterogeneous distribution of hot spots so that almost all of the SERS intensity originated from a rather small volume fraction of the aggregate. The predicted SERS localization was proved by direct near-field imaging. Our teacher, we will miss you greatly.

■ SPASERS

Teri Odom. Mark Stockman's seminal paper on spasers,¹⁹ surface plasmon amplified stimulation emission of radiation, introduced a concept elegant in simplicity but complex in experimental interpretation. This amplification of light should be localized to deep-subwavelength length scales, but would there be enough gain surrounding the metal nanoparticle core to overcome the losses and, then, could this coherent radiation be used in the far-field? Several years later, the prospects of directionality from arrays of resonators provided further guidance.⁹² We achieved lasing action from two-dimensional lattices of metal nanoparticles surrounded by molecular gain materials nearly a decade after the spaser was first proposed.⁹³ We found that nanocavity modes defined by surface lattice resonances, hybrid modes from the coupling of localized surface plasmons of individual nanoparticles to diffraction modes of a periodic array, could provide optical feedback for directional lasing at room temperature; critically, the stimulated emission mechanism was confined to the near-field (tens of nm) of the nanoparticle surface. The spatial coherence of these nanoparticle lattice lasers approached 1 mm.⁹⁴

With the spasing mechanism established, we achieved real-time tuning of the lasing wavelength with this open, distributed nanocavity architecture,⁹⁵ either by changing the refractive index environment around the nanoparticle lattice within a microfluidic device⁹⁶ or by mechanically stretching the devices to increase the separation between particles.⁹⁷ Low-symmetry lattice geometries with rhombohedral unit cells⁹⁸ also exploited the spasing mechanism, where changing the pump polarization resulted in different populations of molecules within the same electromagnetic hotspots contributing to lasing action and at very different wavelengths. Closing the loop on the original spaser design, we found that colloidal semiconductor quantum dots combined with plasmonic nanoparticle lattices could produce lasing action with both radially and azimuthally polarized light and with an emission beam at any desired angle.^{99,100}

■ PLASMONIC TAPERS

Albert Polman and Ewold Verhagen. One of Mark Stockman's key theoretical predictions was that extreme

focusing of optical near-fields would be possible in plasmonic tapers composed of tips with a nanoscale apex.²³ The idea that light could be funneled with high efficiency into arbitrarily small volumes served as a great inspiration to the developing field of nano-optics. This concept was experimentally tested soon after in various geometries.^{101–103} Phase- and polarization-resolved near-field microscopy of the focusing plasmon modes revealed their special antisymmetric nature that allowed these modes to focus adiabatically, unhindered by the diffraction limit.¹⁰⁴ Stockman also proposed very strong optical focusing in a plasmon lens composed of an arrangement of nanoscale plasmonic nanospheres with decreasing size.¹ Such structures with nanoscale plasmonic gaps were made using DNA-templating of Au nanobeads.¹⁰⁵

The nanofocusing that Stockman predicted found direct application in high-resolution nanoscopy with chemical sensitivity.⁸⁰ His insights into the relevance of plasmonic mode conversion and symmetries have had broad impact on the field; inspiring for example the development of transformation optics,¹⁰⁶ waveguide-based negative-index metamaterials,^{107,108} and ultrasensitive nanomechanical sensors based on the coupling of subwavelength plasmonic fields to the mechanical vibrations of nanostructures.^{109,110} At AMOLF, these insights also triggered a wider program to use nanophotonic concepts to create solar cells with improved efficiency and lower manufacturing costs.¹¹¹

■ FAREWELL TO A FRIEND

Cheng Wei Qiu. Professor Mark Stockman is an iconic figure in nanoplasmonics, spasers, and ultrafast physics. He is deeply recognized not only for his fundamental contribution in science, but also for his attitude toward the rigor in science. His comments and viewpoints were always resourceful, informative and inspirational. I always recall that he used to sit in the front row of every conference he attended, and a favorite moment of a session with Mark is when he started to question and comment, regardless of whether the speaker was a junior researcher or a quite senior one.

Mark was a very special scientist to me. I had just completed my Ph.D. when I first talked to him. I asked a few quite simple (obviously naïve to him) questions on spasers and plasmonic hot spots as well as their value to applications and limitations. I received very detailed and sophisticated answers from him while his fellow friends were waiting for him in the conference hall to go outside for lunch. Even though I did not hop onto research on spasers, I have benefited a lot from candid interactions with Mark since then via conference contacts and working emails. Many of his works identify and study the fundamental limits of plasmonics and nanophotonics,^{112,113} which inspired me to explore limits in plasmonics and metasurfaces and transcend the barriers and search for new possibilities thereafter.^{114–116} He shows us a role model for how to identify the intrinsic boundaries and limits of the knowledge and how to go all in to push that territory outward. His scholarly impact goes across generations, continents, and cultures. He has planted all sorts of seeds into the soil of our hearts. Let me end by quoting a poem "Farewell to a Friend" by Bai Li, a Chinese poet (701–762 AD) in Tang Dynasty.¹¹⁷ I translated it into English and particularly used some typical words to dedicate to our great friend Mark. We miss him sorely and dearly.

Farewell to a Friend – Bai Li

Evergreen mountains mark the northern landscape

Glistening creeks gird the eastern town
Here is the place to say goodbye
You'll drift like lonely dandelions,
taking a journey of ten thousands of miles
Over floating clouds, you ski away
With the twilight, you take our thoughts away
We wave as you start your way
The steeds still neigh, "Adieu, adieu"
As if they say, "Don't go too fast, don't go that fast."

■ INSPIRED BY THE SPASER OF A CANTONESE JEW

Mordechai Segev. Mark Stockman was an unusual scientist and person: A person I liked to argue with, a stubborn person that would argue even after he changed his mind, just for the sake of the argument. At the same time, he was a graceful person that truly cared about others. But if you asked Mark how he defines himself, he would tell you: I am first of all a Cantonese Jew. Who were the Cantonese Jews? They were not from the city of Canton (Guangzhou), China. They were not even from any of the 26 cantons of Switzerland. Cantonese Jews lived in the 19th century Imperial Russia and were Jewish boys who were drafted to military service at the age of 12 and placed for their six-year military education in cantonist schools.¹¹⁸ They were required to serve in the Imperial Russian Army for 25 years after the completion of their studies. During their long army service, discriminatory regulations ensured that those Cantonese Jews were held back in their army promotions. In fact, Jews who refused to convert were barred from becoming officers; only eight exceptions were recorded during the 19th century. They were required to distinguish themselves in combat in order to get promoted.

Mark took pride of being a descendent of a 19th century Jewish officer in the Tsar's army who remained Jewish despite the strong pressure to convert. This family history affected Mark's views on life and also specifically on science. Like his ancestors, Mark felt he had to distinguish himself when doing research. He was always searching for new conceptual ideas and hated secondary work. He has generated many original ideas, and we used to argue about them quite often, sometimes quite vocally. Of course, Mark was never wrong in anything ...

To me, Mark's single most important and creative work was the spaser, surface plasmon amplification by stimulated emission of radiation, a paper he wrote with Prof. David Bergman from Tel Aviv University.¹⁹ This paper described a crazy idea where the surface plasmon serve as the amplification mechanism that can support the emission of coherent light. The idea was considered controversial for quite a few years, but it inspired a series of papers on nanolasers, some published at the top journals. Since I never worked on plasmonics or on metamaterials, what caught my interest is the ability to go against the mainstream. Conventional lasers operate by virtue of population inversion, which provides the coherent amplification through stimulated emission, despite the fact that the atoms remain "thermal", lacking any kind of coherence among them. Mark's spaser does not rely on population inversion whatsoever, and it does not rely on stimulated emission in its usual sense, but the spaser does have similarities with laser action. What's more important, in Mark's language, "Spaser generates temporally coherent high-intensity fields of selected surface plasmon modes that can be strongly localized on the nanoscale". This sentence naturally inspired subsequent work on nanolasers.^{61,119,120}

In my own eyes, the most remarkable aspect was that Mark and David were able to match between seemingly unrelated phenomena: laser science and plasmonics. Naturally, plasmonics are anything but related to lasers. Plasmons mean metals, and metals mean loss to any electromagnetic wave propagating on a plasmonic surface. Lasers are the opposite of loss. In fact, the higher the loss, the higher the laser threshold and the less efficient the laser is. So why the heck match between lasers and plasmons? The scientific reasoning was size: plasmons can be confined to the nanoscale, so this meant that perhaps one can construct lasers smaller than the wavelength they emit. The nonscientific reasoning was Mark: he liked to be provocative, and this seemed a proper provocation. At the time, I had many arguments with Mark, also about the spaser. But a little more than a decade later, I took a similar path, matched between lasers and photonic topological insulators, and created the topological insulator laser.^{121–123} Last year, just before Mark passed away, he published a paper on topological spasers.¹²⁴

Rest in peace, Mark, my friend. We will always remember the high-speed skier, the highly creative super provocative scientist, the outspoken, often blunt, person with a good heart. The Cantonese Jew. From the land of your great forefathers.

■ COMPOSITE, ENGINEERED OPTICAL MATERIALS

Vladimir Shalaev. Mark Stockman performed the foundational work on the optics of random media that helped to define and propel the entire area of composite, engineered optical materials. Specifically, Stockman, together with Shalaev, conducted pioneering studies of linear and nonlinear optics of fractals, marked by an important discovery of how the fluctuation nature of fractals defines their optical properties as well as the important role of the enhanced local electromagnetic fields.^{125,126} Later Tsai, Moskovits, and Shalaev demonstrated that the localized plasmon modes in fractals¹²⁷ enable highly confined strong electromagnetic fields, the so-called "hot spots", which are responsible for the unique optical properties of fractals, including their greatly enhanced nonlinear responses.¹²⁸ Inspired by these early papers, Shalaev and Sarychev showed that the hot spots also occur in thin metallic/plasmonic films that are grown close to the percolation threshold, which consist of fractal clusters of all sizes, and that such hot spots result from the Anderson localization of plasmons.¹²⁹ The localized plasmons in random metal–dielectric films were later experimentally demonstrated in ref 130. Mark Stockman with coauthors also showed that the localized and delocalized modes in random metal–dielectric films can coexist,²² as was later verified experimentally.¹³¹ It should be noted that these early findings in the field of optics of random composites had later helped to initiate and mold a new field of optical metamaterials that had since become one of the most active and dynamic fields of all optics.

The theoretical prediction by Stockman and Bergman of the Surface Plasmon Amplification by Stimulated Emission Radiation (SPASER),¹⁹ which was later experimentally verified,^{61,119} was indeed one of the key breakthroughs in the field of plasmonics.¹³² This discovery inspired myriads of new studies on ultrafast plasmonic and nanophotonics and it will stay in the history of science.

■ SURFACE LATTICE RESONANCE SPASERS

Päivi Törmä. The idea of a spaser, originally proposed by Mark I. Stockman,¹⁹ has inspired work on nanoscale lasing in a

multitude of different systems. One of them is plasmonic nanoparticle arrays hosting collective modes of nanoparticle oscillations and diffracted orders of the periodic structure, called surface lattice resonance (SLR) modes. The arrays can be combined with organic gain media, for instance, dye molecules: in such systems, lasing has by now been observed by several research groups; for a review, see ref 133. As suggested by Mark, the features specific to plasmonic systems have led to the unique behavior of the observed lasing, such as ultrafast dynamics. The strong near-fields of the plasmonic particles are advantageous for strong light–matter coupling, which has been observed in these systems.¹³⁴ Moreover, since the nanoparticles can host both dipolar and quadrupolar modes, lasing in a dark quadrupolar mode (so-called bound state in continuum) can be realized. The nanoparticles can be arranged into a hexagonal geometry, and consequently, K-point lasing with specific polarization properties emerges.¹³⁵ Mark realized that, in this type of setting, one can observe chiral, valley-selective lasing;¹²⁴ this proposal will definitely inspire future work. Finally, utilizing the platforms developed for nanoparticle array lasing, Bose–Einstein condensation of the SLR excitations has been recently achieved;^{136,137} this shows how Mark’s visionary contributions to nanoscale lasing have led to developments that probably only a few could anticipate at the time his first spaser paper was published.

■ LOOKING UP CLOSE AT HOT SPOTS

Din Ping Tsai. Professor Mark Stockman was a pioneering researcher in plasmonics. I knew him before the word “plasmonics” was known to most people. In 1992, Professor Martin Moskovits advised me how to chemically produce silver nanoparticles. I still vividly remember that he taught me the specific recipe and to put the beaker into the ice-cold water sink to cool the chemical process, in the Lash Miller Chemical Laboratories around 28 years ago. As a postdoctoral research fellow, I laid the silver nanoparticle droplet on the clean coverslip and took the SEM images for various parameters in order to produce the best fractal silver clusters for near-field optical imaging experiments. We had a brilliant and eminent theoretical physicist, Dr. Vladimir Shalaev as our team member to calculate and simulate the collective electrons oscillation at the hot spots of the fractal silver nanoparticle clusters at that time. I used our homemade photon scanning tunneling microscope to measure the optical near-field images on the samples of the fractal silver nanoparticle clusters. The enhanced optical near-field was found at localized hot spots with clear dependence of the polarization. Professor Martin Moskovits submitted the manuscript to *Physics Review Letters* in 1993 and it immediately received attention from many, including Mark Stockman.

Mark published a research highlight article, “Photon Tunneling Microscope Reveals Local Hot Spots,” in 1994¹³⁸ to discuss the important physics of the localized surface plasmon discovered and reported in our *Physical Review Letters* paper.¹²⁷ I met him in his talk at the University of Toronto in 1993. Since then, we exchanged ideas and discussed some of the research and development of plasmonics. He visited us in Taiwan, and gave short course talks a few times. He always kindly provided his physics opinions and comments on our experimental studies^{139–141} in plasmonics. He founded the Plasmonics Conference of the SPIE Optics and Photonics Symposium in 2002. I was invited to help as the cochair of this Plasmonics conference since 2014. People respected very much

Mark’s great effort and perseverance to sustain this important annual plasmonics conference in San Diego over the last 20 years.

We are lucky to have met and interacted with Professor Mark Stockman over the last 28 years. We sincerely appreciate his great efforts and contributions to physics research and scholarship, especially in plasmonics. His smile, voice, and laugh are always kept in our memory.

■ HOT SPOTS

Anatoly Zayats. The field enhancement and localization are ubiquitous in plasmonics. At the dawn of modern plasmonics, when the attention shifted from the studies of idealized smooth films and single nanoparticles to more complex structures, it was Mark’s theoretical work that introduced the giant enhancement of electromagnetic fields in clusters of interacting nanoparticles, fractals and on rough plasmonic surfaces.¹⁴² These predictions were promptly confirmed experimentally with a newly developed technique of scanning near-field optical microscopy^{127,143} and become the foundation for the advancement of new types of the enhanced spectroscopies, such as surface-enhanced Raman spectroscopy with single molecule sensitivity and second-harmonic generation.¹⁴⁴ The studies of these so-called plasmonic hot-spots have revealed numerous fundamental properties of the electromagnetic near-fields such as Anderson localization and bright and dark plasmonic eigenmodes as well as was a push to development of nonlinear nano-optics.^{144,145} Moving from two-dimensional surfaces to three-dimensional nanoscale composites (aka metamaterials), Mark demonstrated how the localized optical fields can be used to achieve nonlinear optical response.¹⁴⁶ Nonlinear metamaterials and, in particular, those based on plasmonic nonlinearities, have been proven to provide some of the strongest and fastest third-order optical nonlinearities for controlling refractive index, propagation, and absorption of light.¹⁴⁷ Despite the absence of the bulk second-order nonlinear response in the majority of plasmonic metals, sculpturing of electromagnetic fields in metamaterials results in the effective second-order susceptibility comparable to conventional nonlinear crystals.¹⁴⁸ These few of many examples illustrate a universality of the concept of engineered hot spots in plasmonics and metamaterials, which were then extended to achieving strong nanoscale field localization in self-similar waveguides and high-harmonic generation and development of a concept of a spaser.

■ PLASMON NANOLASERS: FROM CONCEPT TO IMPLEMENTATION

Xiang Zhang. The invention of lasers has enabled a new capability in controlling light. During the past decades, lasers have had an enormous impact on our life and society, driving science and applications ranging from information technologies, defense, energy, imaging, and biomedicine. The current information explosion requires the miniaturization of optical devices and computing chips imposing a new challenge in laser research: that is, the physical and mode volumes of lasers must be miniaturized. However, the spatial concentration of the laser-radiation energy is fundamentally limited by the diffraction of light, with the smallest scale being at the half-wavelength scale (about a few hundred nanometers for visible light).

In 2003, David Bergman and Mark Stockman proposed the concept of spasers (Surface Plasmon Amplification by Stimulated Emission of Radiation).¹⁹ They theoretically

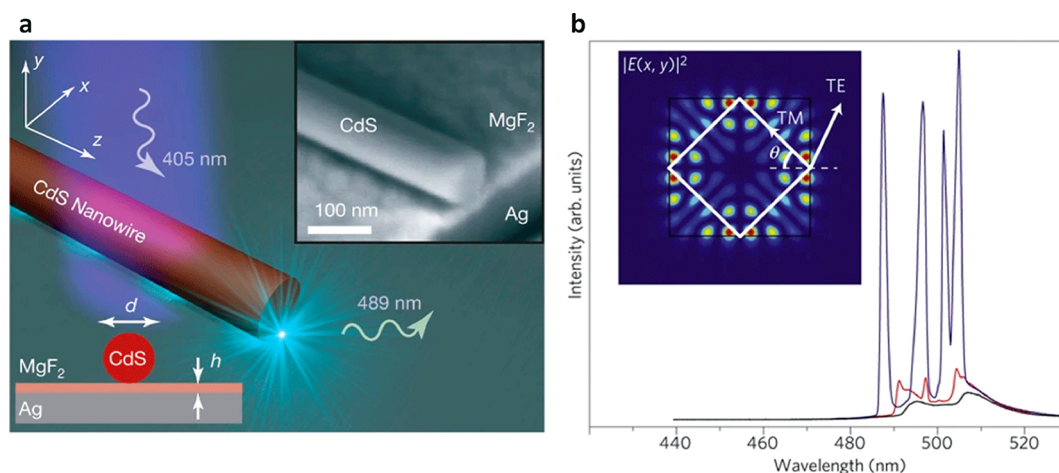


Figure 2. (a) The demonstration of hybrid plasmon laser designed with CdS-MgF₂-Ag configuration, maintaining ultrasmall optical confinement in dielectric gap region while mitigating the metal loss. Reprinted with permission from reference⁶¹ (b). Further suppressed cavity loss enabled by total internal reflections leads to a high temperature plasmon nanolaser, as evidenced by the observation of a transition from spontaneous emission (black) through amplified spontaneous emission (red) to full laser oscillation (blue) at room temperature. Reprinted with permission from ref 150. Copyright 2011 Nature.

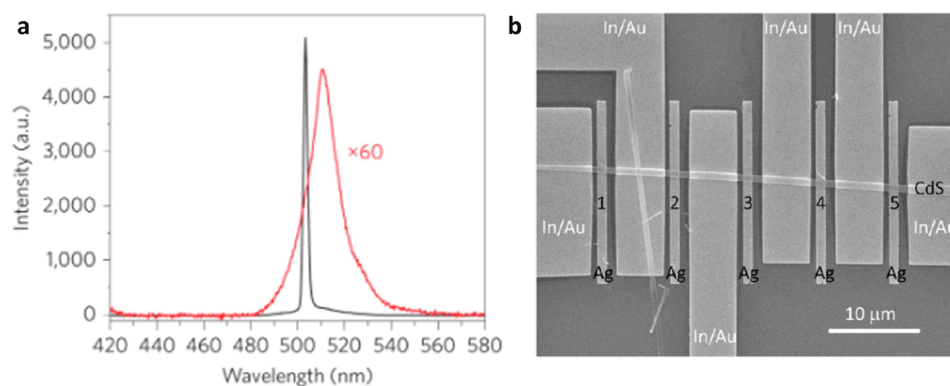


Figure 3. (a) Due to the loss compensation by active media, the sensitivity of lasing emission is approximately 300× higher than that of spontaneous emission. Reprinted with permission from ref 151. Copyright 2014 Nature. (b). Integrated plasmon nanolaser interconnect that enables wavelength-division-multiplexing and effective waveguiding is a promising step toward on-chip photonics and optoelectronics. Reprinted with permission from ref 152. Copyright 2012 American Chemical Society.

introduced surface plasmon quanta for stimulated emission of radiation that was amplified by surface plasmon polaritons (SPPs), confining the optical energy at the metal–dielectric interface with the spatial scale much smaller than a half-wavelength (at the order of ten nanometer and even smaller), well below the diffraction limit. Later on, this concept has been generalized to plasmon nanolasers, or plasmonic nanolasers. Inspiring from that, subwavelength spasers or plasmon lasers have been successfully demonstrated and implemented experimentally in 2009, opening a new door to ultracompact and ultrafast on-chip communications and information processing.^{61,119,149}

Yet, the surface plasmon polaritons are light coupled delocalized electron oscillations, which inevitably lead to the intrinsic metal/ohmic loss. To mitigate such metal losses, a hybrid plasmon approach that integrates dielectric waveguiding with plasmonics has been proposed and experimentally demonstrated.⁶¹ This hybrid plasmon laser consists of a dielectric nanowire separated from a metal surface by an extremely thin dielectric gap. The coupling between the plasmonic and waveguide modes across the gap enables “capacitor-like” energy storage that allows effective subwave-

length optical energy confined in the thin nonmetallic regions, which could significantly reduce metal loss while maintaining ultrasmall modes (Figure 3a). At the same time, the strong confinement of optical modes reveals a broadband enhancement of the exciton spontaneous emission rate by up to six times. In another aspect, the current applications of optoelectronic devices and photonics circuits critically relies on the small coherent light source working at the room temperature. However, the nanoscale plasmon lasers suffers from drastically increasing losses and deteriorating gains upon elevating the temperatures. Room-temperature operation of a plasmon laser poses another challenge in the field. By further mitigating cavity losses, a total internal reflection of SPPs in a whispering-gallery plasmonic cavity was realized, where sustained plasmonic lasing action at room temperature has been successfully demonstrated¹⁵⁰ (Figure 3b). In addition, high cavity quality factors together with strong $\lambda/20$ mode confinement lead to spontaneous emission rate enhancement by up to 18-fold. Thus, plasmon nanolasers offer the new opportunities in exploring extreme interactions between light and matter, opening up new avenues of active photonic circuits, sensing and quantum information technologies.

Based on the amplification of the surface plasmons, an active plasmon laser sensor has been achieved with a subpart-per-billion level of sensitivity, the lowest reported to date for plasmonic sensors.¹⁵¹ Due to the loss compensation by the active media, it leads to ultrasensitive detection of 300 times greater than traditional semiconductor sensor based on the spontaneous emission (Figure 2a). Such exceptional detection ability demonstrates the potential of actively excited surface plasmons for important applications in chemical/biodiagnostics, security, defense. With an unprecedented ability to localize the electromagnetic field, plasmon nanolasers can also play a key role in the scaling down of current photonic chips and circuitry. However, ever-increasing radiation divergence and challenges in the integration of current electronic functionality brings a fundamental hurdle for the ultracompact circuitry. A promising solution is to design a metal–insulator–metal waveguiding or multiplexed plasmon nanolaser interconnects (Figure 2b), in which an integrated waveguide embedded (WEB) plasmon laser that efficiently converts surface plasmons into directional laser emission and the metal strips simultaneously serve as the electrical contacts.^{152,153} Such a waveguide-integrated plasmon laser circuits can reach more than 70% directional emission coupling with dramatically enhanced radiation efficiency on-chip, illustrating the potential future applications for large scale, ultradense photonic computing and communications.

In the past, we have seen tremendous progress in bringing the plasmon laser to the deep subwavelength scale.¹³² Yet, several future challenges are still down the road. For example, the temporal characteristics of plasmon lasers has to be extensively explored for ultrafast computing and communications (e.g., Purcell enhancement factor, modulation bandwidth). On-chip electrical data input and optoelectronic integration begins the quest for an electrically driven plasmon nanolaser. The significant optical losses at electrical contacts and severe nonradiative losses as the current densities increase will need to be overcome. Moreover, single-crystalline processing and self-assembly offer opportunities to explore both new design and scalable applications of plasmon nanolasers.

■ FINAL THOUGHTS

Nikolay Zheludev. Mark Stockman became a household name in the photonics community in 2003, after publishing his seminal paper on the spaser.¹⁹ From that point on, his career was elevated to a new level: according to Web of Science, 95% of papers referencing Mark's works were written since 2003. We met and became friends at around that time and worked together on a number of occasions. His opinion was always extremely valuable to me, and it came as no surprise to me that he achieved such a colossal influence in the nanophotonics community: his regular journal papers and letters published since 2003 were each cited nearly 100 times on average (98). Every conference speaker would keep an eye on Mark sitting in the front row of the audience in his signature white shirt, anticipating his sharp, deeply physical question. Regardless of personalities, Mark was happy to challenge scientific concepts he did not agree with^{154,155} and keenly defended and promoted his own ideas, such as the spaser,¹⁹ plasmonic taper,²³ or the ultrafast topological all-optical gate¹⁵⁶ (one of his last papers), which he passionately believed can change technology. He earned the reputation of a theorist with a background, opinion, and ideas spanning a very wide range of subjects, from solid state physics to biomedicine.

I had the privilege of collaborating with Mark from 2004, when we started working on “active plasmonics”,¹⁵⁷ the ability to dynamically control the propagation of surface plasmon polaritons. In 2006, we together wrote the first experimental paper on the generation of traveling surface plasmon waves by free electron impact,¹⁵⁸ exactly as was originally predicted by the father of surface plasmonics, Rufus Ritchie, in 1957. And, in 2009, we wrote with Mark a paper demonstrating femtosecond all-optical switching of plasmon polaritons.¹⁵⁹ Excited and stimulated by Mark's work on the spaser, we developed it further and introduced the lasing spaser,⁹² a laser based on a metamaterial array of spasers, which was published in a paper that Mark saw and endorsed prior to publication and promoted on many subsequent occasions. Plasmonics was everything to Mark, and I am grateful to him for supporting our work that drew an analogy between plasmonics and superoscillatory fields, which he handled as editor at NPG Light Science and Applications.¹⁶⁰ In 2018, Mark organized a roadmap paper on plasmonics¹⁶¹ collectively authored by leading authorities in the field. In his own part of the roadmap, he passionately wrote: “Modern nanoplasmonics is a flourishing science, rich in ideas, fundamental achievements, and applications. Among them are biomedical and environmental sensing, detection of minute amounts of vapours from explosives, cancer diagnostics and treatment, etc. In my opinion, the future of nanoplasmonics is in the fundamental progress with further extensions into areas of strong, ultrafast, and extremely nanolocalized fields, where theory will need to become fully quantum mechanical to accurately predict and describe new phenomena. At the same time, the existing applications will be further improved, and commercialized, and new applications will be invented, and among those ultrafast optical computing may be one of the most important.” He has been the founding member and evangelist of plasmonics for most of his creative life. Mark Stockman never sought recognition, and he was never awarded big medals or prizes for his scientific work. Fame came to him in a different way. He will be remembered by colleagues and friends as one of the most influential and creative contributors to the science of light from our generation.



Mark Stockman with friends at Ein Gedi Kibbutz, Israel, 10 February 2019. Photo courtesy of Nikolay Zheludev.

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Notes

Views expressed in this editorial are those of the authors and not necessarily the views of the ACS.

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