# Reflections on Mark Stockman and his contributions to nano-optics: guest editorial

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**Abstract:** A very brief tribute to Mark I. Stockman and his contributions to optical science.

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#### 1. Introduction

A giant tree fell in the forest–Professor Mark I. Stockman of Georgia State University, the driving force behind many recent groundbreaking developments in nanophotonics, and more specifically plasmonics, passed away in November 2020. This left a deafening silence, which we try to fill with our words of remembrance of Mark, his love of science, and his critical contributions to it. Mark had an unusually wide range of interests and his impact can be found in many diverse areas of photonics and condensed matter science. It is impossible to properly acknowledge all the contributions made in the course of five decades. In this editorial, we pay tribute to just three key areas where Mark's influence is most visible.

#### 2. Mark Stockman and plasmonic lasers (SPASERs)

Lasers have been around for over sixty years. When the laser was first demonstrated (by Theodore H. Maiman, in 1960 [1]), it was called the "solution looking for its problem" – a unique fundamental phenomenon without clear applications in sight. Sixty years later, lasers are everywhere, starting from the world's largest (three football fields can fit inside) and highest-energy laser at the National Ignition Facility (Lawrence Livermore National Laboratory, USA) to a laser pointer, which one can use to play with a cat. Many industries, including aviation, manufacturing, and telecommunication use lasers as standard tools or are simply impossible without lasers. Information processing can also become revolutionized by the advent of lasers, as their size shrinks and they both provide dense on-chip interconnects and potentially perform some switching functions currently reserved for transistors and this is where lays Mark's critical contribution.

LASER is an acronym for light amplification by stimulated emission of radiation. Two principal components of every laser are an active gain medium, providing for light amplification, and (commonly) two mirrors providing for a stimulated emission feedback. The smallest optical resonator supporting a fundamental standing wave is half-wavelength long. For visible light, this approximates a quarter of a micrometer – more than an order of magnitude larger than the gate width of a modern transistor currently as small as a few nanometers [2].

In 2003, Mark I. Stockman and his collaborator, David J. Bergman [3], had theoretically shown that the size of a resonator can be reduced down to a nanoscopic scale if a metallic nanoparticle supporting localized surface plasmon resonance(s) is used instead of a quarter-micrometer photonic cavity formed by mirrors. This device, called SPASER (the acronym in which SP standing for surface plasmon replaces L standing for light), was designed to generate not photons

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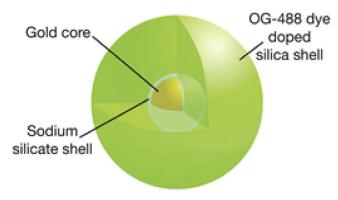
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but rather coherent surface plasmons (SPs), which are, arguably, more suitable in photonic circuits. Outcoupling of SP oscillations to far-field optical modes made the proposed device a nanolaser. Mark further developed the concept and the theory of a SPASER in several seminal publications [4,5] portending applications of SPASERs as an ultra-fast nanoscale switch and amplifier. These are Mark's key contributions to future photonic nano-circuitry and information technology.

Strong confinement of a SPASER mode to a volume of metallic particle, which is nanoscopic in all three dimensions, is the source of a Joule heat loss that requires large optical gain to overcome it. Two realizations of a SPASER, comprised of metallic nanoparticles surrounded by large numbers of dye molecules (e.g.  $3 \times 10^3$  molecules per nanoparticle in [6]) have been reported in the literature [6,7], as shown in Fig. 1. The demonstrated SPASERs (some of them researched by Mark) were suspended in water, preventing over-heating, and pumped optically.



**Fig. 1.** Diagram of the SPASER nanoparticle, showing the gold core (a resonator) and dye-doped silica shell (a gain medium). Adapted from [6].

Millions of such SPASERs can be injected into a bloodstream and used for medical diagnostics and treatment [7]. However, the prospective grand-scale application of SPASERs in future nano-circuitry operating at optical frequencies requires all-solid-state designs and electrical pumping. As it was shown in [8–10], electrical pumping of semiconductor-based SPASERs will require such high threshold current (~20  $\mu A$ ) that the dissipated heat will damage the device. When the size of a SPASER reaches a nanoscopic scale (~50 nm), its threshold no longer goes down with further size reduction, hindering scores of nano-circuitry applications.

Confinement of a mode to a plasmonic waveguide, whose size, at least in one dimension, exceeds a wavelength, is getting weaker and, correspondingly, the Joule loss becomes much smaller [11]. This enables a large variety of relatively low-threshold 2D (plane waveguides) and 1D (nanowires) miniature plasmonic lasers, with and without feedback, reported in the literature [12–15]. Note that while the concept of Surface Plasmon Polariton excitation was first introduced as early as 1989 [16] and a partial offset of the SPP loss by optical gain was experimentally demonstrated in 2005 [17], it was Mark's work that gave real impetus to the development of nanolasers.

As we have already mentioned, the ongoing research largely tries to resolve the inherent contradiction between the requirements for (i) a small physical size and tight mode confinement and (ii) a small loss in the metal. The authors of [18] have fabricated ~170 miniature photonic and plasmonic lasers and studied their performance at optical pumping. It has been shown that although photonic lasers are superior to plasmonic lasers at large mode volumes ( $\geq 5 \lambda^3$ ), plasmonic lasers have a smaller threshold and smaller power consumption as the mode volume is getting reduced to  $\leq \lambda^3$ . Plasmonic lasers also have a critical advantage in speed, which hopefully

will be exploited in future nanophotonics circuits, fulfilling Mark Stockman's dream that set so many researchers on the path to SPASERs.

#### 3. Mark Stockman and plasmonic nanofocusing

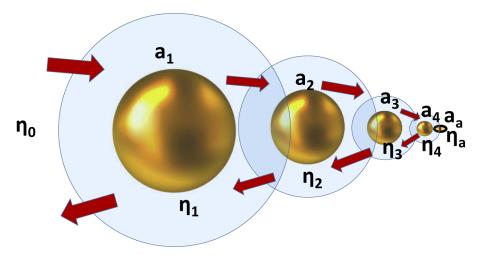
While Mark's work on coherent source of surface plasmons (SPASERs) is probably his most recognized, his contribution to nanophotonics goes well beyond it. In particular, his pioneering ideas in the field of nanoscopy and nano focusing deserve special attention. While the idea of nano focusing (i.e. concentrating the light into sub-wavelength volumes) using photonic and plasmonic nanoscale structures has been discussed for many years [19], Mark made seminal contributions to it by suggesting concepts of adiabatic focusing [20] and focusing using self-similar chains of nanoparticles [21]. These ideas have been further developed by the community and have established the means of both effective focusing of light into miniscule volumes [22] and, most remarkably, accelerating by orders of magnitudes the spontaneous emission rates via the Purcell effect [23,24]. These achievements are already finding applications in nano-scale sensing and in the development of efficient single photon sources. Let us now briefly review the issue of nano focusing and emission enhancement in order to both ascertain the obstacles facing anyone attempting these tasks, and the ingenuity of the solutions that Mark generated.

First of all, consider the concept of impedance matching that is omnipresent in every aspect of photonics. Impedance  $\eta = E/H$  relates the magnitudes of electric E and magnetic H fields and in purely dielectric media can be found as  $\eta = \eta_0/n$ , where  $\eta_0 = 377\Omega$  is a vacuum impedance and n is the refractive index. It is the mismatch of impedance that causes reflection at the boundary between two dielectrics thus complicating coupling into all-dielectric photonic structures. The same impedance mismatch plays a key role in electronics, where it is defined as voltage-to-current ratio and prevents effective coupling of typically high impedance electronic circuits to free space or transmission lines whose impedance is commensurable with  $\eta_0$ . This fact necessitates various impedance matching and antenna techniques. When it comes to coupling the light in and out of optically active atoms and molecules the same paradigm applies. If one considers a Lorentz dipole model of an emitter/absorber, its impedance can be found as roughly  $\eta_{em} \sim 1/\omega C$ , where capacitance  $C \sim \varepsilon_0 a_a$  and  $a_a \sim 1$  is characteristic size of the atom. Then,  $\eta_{em} \sim (\lambda/a_a)\eta_0$ , i.e. orders of magnitude higher than that of free space.

Therefore, to achieve efficient coupling of light into and out of optical emitters and absorbers, one must resort to the well-tried method of matching impedances, i.e. employing one or a series of photonic entities with intermediate impedances  $\eta_{em} > \eta > \eta_0$ . Alas, for any photonic structure with n > 1, it becomes an impossible feat, and one has to consider media with free carriers where n < 1 and in most cases has large imaginary part. Indeed, if one considers the structure with characteristic dimension  $a < \lambda/2$  (where  $\lambda$  is wavelength in dielectric) it follows from the Maxwell's equation  $\nabla \times \mathbf{H} = i\omega \varepsilon_0 n^2 \mathbf{E}$  that  $H \sim (2a/\lambda)\eta_0 E$ . If the structure is made of pure dielectric, no stable mode can exist because the magnetic energy, oscillating as  $\sin^2 \omega t$  is much smaller than the electric energy oscillating as  $\cos^2 \omega t$ , which violates energy conservation, and, this fact, of course, constitutes the diffraction limit [25]. If, however, free carriers are present, their kinetic energy oscillating in phase with magnetic energy restores the energy balance and permits self-sustaining oscillations which are of course, surface plasmon polaritons (SPPs) of two varieties—localized surface plasmons (LSP) on the sub-wavelength nanoparticles and propagating SPP's with effective wavelength  $\lambda_{SPP} < \lambda$ .

Clearly, the impedance of LSP or SPP is proportional to  $(\lambda/a)\eta_0$ , where a is the characteristic size of the mode and it can serve as an impedance transformer for achieving effective absorption or emission of light as well as enhancing various nonlinear phenomena, such as Raman scattering. This can be witnessed by the large trove of experimental results that accumulated in recent decades. However, the impedance mismatch between the free space (or a waveguide) and the atom or molecule is way too large  $(10^4)$  to achieve efficient coupling with just a single "transformer".

For instance, the concentrated field in a tiny SPP mode with effective size of a few nanometers will be coupled very effectively into the atom or molecule and that atom or molecule will also radiate into the SPP mode very efficiently due to the Purcell effect. However, the light will not couple in and out of this small volume SPP mode successfully due to large impedance mismatch, which in the presence of large nonradiative losses inherent to metal can be ruinous for the overall efficiency. To raise the efficiency, one has to somehow match the impedance between the small volume SPP and free space and that can be attained by using another impedance transformer, i.e. LSP with characteristic size between that of the first one and the wavelength, as shown in Fig. 2. Note that the LSP mode can reside either on a metal nanoparticle or in the gap between two of them.



**Fig. 2.** Impedance matching allows efficient coupling between the light in free space (or waveguide) and an excitation of an atom or molecule. The excitation travels thorough a chain of N LSP modes with characteristic dimensions  $a_n$  progressively reduced from the fraction of wavelength ( $\sim 100 nm$ ) to the few times atomic dimensions ( $\sim 1 nm$ ). As the dimension  $a_n$  gets smaller, the impedance increases as  $\eta_n \sim (\lambda/a_n)\eta_0$  from low impedance in the free space to the high impedance of the atomic dipole.

In 2003, Mark Stockman, working with colleagues Kuiru Li and David Bergman, was the first to come up with this groundbreaking idea (albeit formulated in a somewhat different way) by noticing that using a chain of self-similar (i.e. having the same shape) nanoparticles with progressively decreasing sizes, one can focus the light into the gap between the smallest nanospheres where the local fields are enhanced by orders of magnitude [21]. In this work, Mark and his colleagues skillfully utilized the fact that the resonant frequencies of sub-wavelength LPSs depend only on the shape and not on the size of the modes and that allows the efficient resonant transfer of power along the chain in both directions, i.e. can enhance both absorption and emission of light.

Shortly thereafter, in 2004, Mark expanded the concept of nanofocusing to propagating SPPs in the tapered plasmonic waveguide [20]. As the width of the waveguide and the effective wavelength of SPP get reduced, the impedance gradually increases and reaches values almost comparable with those of atomic and molecular dipoles. The key to efficient transfer of energy is the fact that the width (and hence impedance) are changing adiabatically, hence the reflection is almost nonexistent and the only limitation to efficiency is metal absorption. In his work, Mark has taken previous work on polariton nanofocusing [26] further to practical implementations achieved since his pioneering contribution [22].

Another contribution Mark made to the fertile field of nanofocusing, in collaboration with Peter Nordlander's group at Rice University, is his work on plasmon hybridization in nanoparticles [27], where he has pointed out that significant enhancement can be attained in plasmonic dimers. This was shown to come from the fact that the dimer acts as an optical antenna with low impedance while the gap mode in it acts as a high impedance cavity [28] and was exploited in achieving record high emission rate for quantum emitters [23].

Last but not least, one must acknowledge the great pedagogical role played by Mark by presenting a treasure chest of inspiring presentations and comprehensive elucidating reviews and tutorials [29,30] over the decades of his work in nanoplasmonics. These contributions have inspired new generations of scientists who will continue along the path blazed by Mark Stockman.

#### Mark Stockman and optics of random composites: hot spots and enhanced nonlinearities

Mark Stockman made his earliest contributions to nanophotonics in the 1980s and 1990s, long before plasmonics became fashionable. Mark performed the foundational work on the optics of random media that helped to define and propel the entire area of composite, engineered optical materials. In the late 80s, pioneering studies on linear and nonlinear optics of fractals were conducted [31,32]. Fractals are scale-invariant: they reproduce the structure of the whole on progressively smaller scales-either deterministically, as in the case of geometrically ordered structures, or statistically, on average, for random fractals, such as for example, for colloidal aggregates or finite clusters in random metal-dielectric films. Similar to the physics of phase transitions, such scale-invariance strongly affects the basic physical properties, which are often characterized by scaling, power-law dependencies. Fractals have asymptotically zero density (the larger the size of the fractal the smaller its density) so that fractal systems are 'fluctuational' by their nature. In that regard, one could expect that fluctuations should play a particularly important role in the physics of fractals, including their optical properties. The early papers on optics of fractals [31,32] by Mark and coworkers were marked by the 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.

Later, it was demonstrated that the localized plasmon modes in fractals enable highly confined strong electromagnetic fields, which they dubbed "hot spots" [33] (see also later related works [34,35]) (Fig. 3). Such hot spots are responsible for the unique optical properties of fractals, including their greatly enhanced nonlinear response [36–38]. The areas where plasmon modes are localized strongly vary in their local geometries and thus resonate at different frequencies [6,8]. The local responses are also polarization sensitive, resulting in strong spatial frequency and polarization selectivity of the light-fractal interaction, so that light at different frequencies and polarizations excites different sets of hot spots in a fractal. Because of the large variety in local geometries of the resonating structures, the plasmon modes in fractals cover all together a very broad spectral range [36,38], enabling dramatic enhancement of surface-enhanced Raman scattering (SERS) [39] and optical nonlinearities in the hot spots [6–8]. The higher the order of nonlinearity the stronger enhancement can be obtained from the localized plasmon modes so that the hot-spot enabled enhancement for third-order nonlinear processes such as four-wave mixing can be gigantic [38].

Inspired by these early papers, researchers have shown that hot spots associated with the localized plasmon modes also occur in thin metal-dielectric films in the vicinity of the percolation threshold and that such hot spots result from the Anderson localization of plasmons [40,41]. At percolation, such semi-continuous metal films consist of fractal clusters of all sizes, from structures with few particles only to the "infinite cluster," which connects the opposite ends of the film enabling the DC conductivity ("percolation") through the film. The localized plasmons in random metal-dielectric film were eventually experimentally demonstrated in [42]. Mark

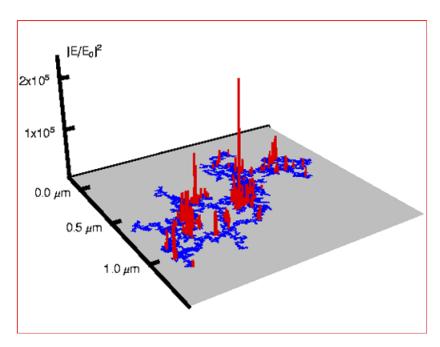


Fig. 3. Localized plasmon modes in fractals enabling high-local field areas – "hot spots" – introduced in [33]. Numerical simulations of the enhanced local-field intensity distribution in a fractal aggregate of silver nanoparticles deposited on a plane (adapted from Ref. [37]).

Stockman, with co-authors, also showed that the localized and delocalized modes in random metal-dielectric films can co-exist [43], which has been verified experimentally [44]. Similar to fractals, optical nonlinear responses are strongly enhanced in random metal-dielectric films close to percolation because of the hot spots resulting from the localized plasmon modes [38,40,45,46].

These early findings in the field of optics of random composites, where Mark Stockman was one of protagonists, later helped to initiate and mold a new field of optical metamaterials that has since become one of the most active and dynamic fields in optics.

#### 5. Coda

It will take some time for the full impact of Mark Stockman's pioneering work to be fully realized by the community, and even longer for the giant void left after Mark's untimely death to be filled. But filled it will be, as new generations inspired by Mark continue the work he started. We hope that this brief tribute to Mark's life and achievements will help serve this purpose.

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