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**Bozhevolnyi, Sergey I.; Mortensen, N. Asger**

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Sergey I. Bozhevolnyi and N. Asger Mortensen*

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**Abstract:** Expanding the frontiers of information processing technologies and, in particular, computing with ever-increasing speed and capacity has long been recognized as an important societal challenge, calling for the development of the next generation of quantum technologies. With its potential to exponentially increase computing power, quantum computing opens up possibilities to carry out calculations that ordinary computers could not finish in the lifetime of the universe, whereas optical communications based on quantum cryptography become completely secure. At the same time, the emergence of Big Data and the ever-increasing demands of miniaturization and energy-saving technologies bring about additional fundamental problems and technological challenges to be addressed in scientific disciplines dealing with light-matter interactions. In this context, quantum plasmonics represents one of the most promising and fundamental research directions and, indeed, the only one that enables the ultimate miniaturization of photonic components for quantum optics when being taken to extreme limits in light-matter interactions.

**Keywords:** quantum plasmonics; extreme plasmonics; light-matter interactions; quantum technology.

Expanding the frontiers of information processing technologies and, in particular, computing with ever-increasing speed and capacity has long been recognized as an important societal challenge, calling for the development of the next generation of quantum technologies [1]. With its potential to exponentially increase computing power, quantum computing opens up possibilities to carry out calculations that ordinary computers could not finish in the lifetime of the universe, whereas optical communications based on quantum cryptography become completely secure. At the same time, the emergence of Big Data and the ever-increasing demands of miniaturization and energy-saving technologies bring about additional fundamental problems and technological challenges to be addressed in scientific disciplines dealing with light-matter interactions. In this context, quantum plasmonics represents one of the most promising and fundamental research directions [2, 3] and, indeed, the only one that enables the ultimate miniaturization of photonic components for quantum optics when being taken to extreme limits in light-matter interactions (Figure 1). Whereas plasmon phenomena are inherently of quantum nature, surface plasmons (SPs) [4] have, for decades, been successfully explored with a mindset of classical electrodynamics [5] (bottom left corner). However, fundamental problems found in all corners of this schematic chart (Figure 1) increase exponentially their complexity when coming to experience also fundamental size limitations posed by the very nature of both light and matter, both consisting of quanta and thereby being discrete. With this perspective, we anticipate an emerging new era of "quantum plasmonics" (top right corner), where both light and matter exhibit quantum mechanical effects. We expect numerous challenges emerging out of the "treasure trove" while also providing the fuel for emerging quantum technologies. Below, we discuss some intriguing aspects of the different regimes outlined in Figure 1.

Recently, there have been significant efforts in what we here term "extreme plasmonics" (bottom right corner), where extremely confined plasmonic modes [6] are promoting electrodynamics beyond the accurate description of the local-response approximation of light-matter interactions [7]. Intriguing and classically unexpected experimental observations include frequency blue-shifts of plasmon resonances in few-nanometer silver nanoparticles [8, 9] and gold plasmonic gap structures [10] as well as gap-dependent broadening and onset of charge-transfer plasmons in dimers and gap structures with sub-nanometer gaps [11–13]. Theoretical explanations range from hydrodynamic nonlocal response [14, 15] to ab initio quantum mechanics [16, 17], accounting for quantum effects such as quantum pressure waves, Landau damping, quantum spill-out, and tunneling. In the other extreme, "quantum optics and single-photon phenomena" (top left

***Corresponding author: N. Asger Mortensen,** Department of Photonics Engineering, Technical University of Denmark, DK-2800 Kongens Lyngby, Denmark, e-mail: asger@mailaps.org; namo@fotonik.dtu.dk
**Sergey I. Bozhevolnyi:** Centre for Nano Optics, University of Southern Denmark, Campusvej 55, DK-5230 Odense M, Denmark

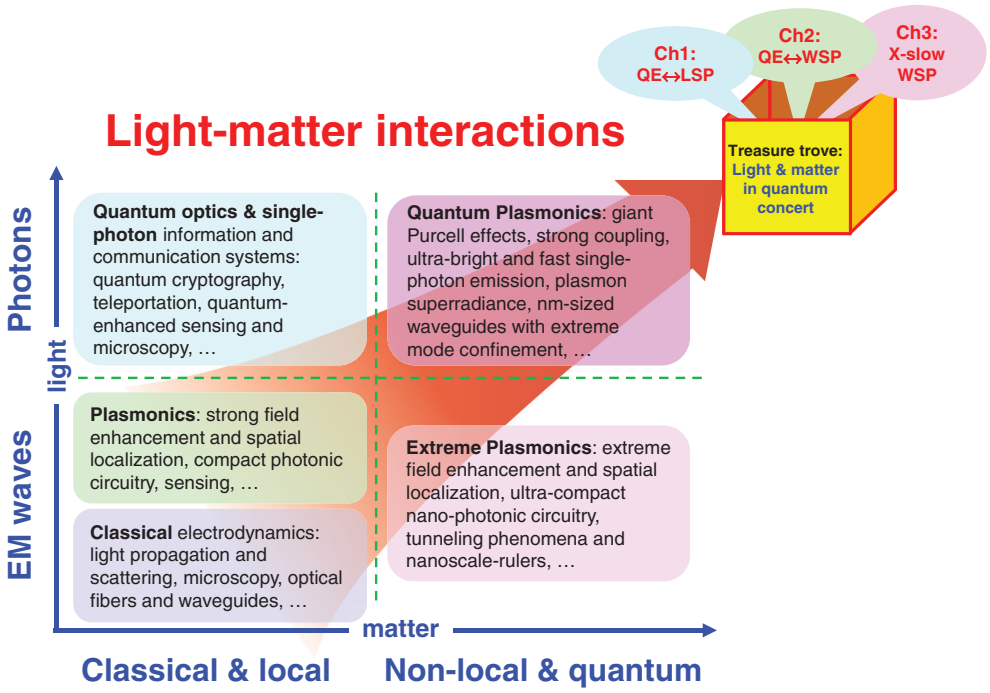


**Figure 1:** Quantum aspects of plasmonics exhibit both in the quantum nonlocal response of matter and in the quantized light fields. With the developments of quantum plasmonics, we anticipate a "treasure trove" of emerging quantum technologies that harvest from the quantum concert of both light and matter. In the main text, we outline three challenges associated with this.

corner) have been interfaced and enhanced by the classical electrodynamics of plasmonic nanostructures [18, 19]. Here, the quantumness is in the nonclassical states of the electromagnetic field or in the quantum properties of the light emitters [20, 21].

It is the merging of quantum optics with extreme plasmonics that now drives us to new exciting "quantum plasmonics" (top right corner), being quantum in a double sense: both light and matter exhibit quantum mechanical effects. As an appetizer from this almost virgin territory of quantum plasmonics, we here mention the recent exploration of single-molecule strong coupling dynamics in subnanometer plasmonic gap cavities [22].

Many crucial issues in modern plasmonics revolve around the fact that the most exciting and unique feature of plasmonic modes, viz., the feasibility of broadband and extreme (down to atomic scale) mode confinement as well as associated enormous field enhancement by the use of metal nanostructures, is directly associated with the energy loss (via radiation absorption) in metal that progressively increases with the field confinement [23]. The fundamental problem is then to understand to what extent this unique feature, enabling both ultimate miniaturization [24] and ultra-strong coupling to quantum emitters (QE) [25], should be exercised before the inevitable energy loss destroys the outcome of its exercise. Enormous challenges emerge already on the way to proper formulation of the associated problems, as the whole well-developed macroscopic treatment of light-matter interactions breaks down and no longer works on the nanoscale, requiring nonlocal and quantum effects to be taken into account.

The whole set of fundamental problems associated with extreme plasmonics can be factorized into several tradeoffs representing conflicting tendencies that are being enormously enhanced when reaching extreme limits. A careful analysis and assessment of these tradeoffs is indispensable for the accurate mapping of the field boundaries and potential developments. In this context, the exploration of fundamental limits in light-matter interactions can be categorized into several major research challenges that are all related to the aforementioned key issues.

**Challenges** It is known [25] that placing QEs in optical resonators with high-quality factors ($Q$) and/or very small volumes (V) results in the spontaneous emission (SE) enhancement described by the Purcell factor $F \sim Q/V$. Whereas extremely high $Q$ values ($>10^6$) can be attained with photonic crystal microcavities, the rate of single-photon QE emission out of such a resonator is inevitably limited due to its high-quality factor [25]. Alternatively,

localized surface-plasmon (LSP) excitations enable extreme light confinement (down to nanometer-scale), thereby providing unique possibilities for SE enhancement [2]. The quality factors of SP resonators are, contrary to the previous case, rather limited ($Q<100$), and ultrabright single-photon sources become feasible by squeezing the LSP volume ($V\to 0$). At the same time, extreme LSP confinement leads to increasing QE quenching and thus decreasing the QE quantum yield, so that the issue of ultimate SE enhancement remains open (Ch1). It should be noted that the most promising results (ultrafast SE emission) have been obtained with special SP resonators, viz., metal-insulator-metal nanocavities supporting gap SPs [26], whose unique features require very careful theoretical analysis with nonlocal and quantum effects being taken into account.

The problem of SE enhancement in the case of QE coupled to propagating waveguide SPs (WSP) is somewhat similar to and yet very much different from the above (Ch2). In this case, the Purcell factor is determined by the ratio between the waveguide mode group index and the mode area (i.e. $F\sim n_g/S_m$). Whereas very high group indexes ($>10^2$) can be attained with photonic crystal waveguides, the bandwidth available for QE emission into such a waveguide is inevitably limited due to its proximity to the (prohibited) band gap. Contrary to that, various WSP modes, including the gap and channel SP modes [27], can be confined (practically at any wavelength) within extremely small cross-sections, thereby increasing the QE-WSP coupling efficiency [28]. At the same time, however, the loss-related problems would also become significant. In fact, one should simultaneously maximize the Purcell factor, the coupling to propagating SP modes (rather than to lossy SPs), and the normalized SP propagation length [19]. The record of the corresponding product is currently ~6.6 [19], but its fundamental limit is yet to be established. A novel issue in this context is the problem of unidirectional and efficient SE into WSP modes [29], as one would ideally couple all emitted photons into WSP quanta propagating in the same direction.

Another unique feature of SP-based waveguides (Ch3) is related to the possibility of seamless interfacing of electronic and photonic circuits by employing the same metal circuitry for both guiding the optical radiation and transmitting the electrical signals that control the guidance [30]. The latter implies that metal electrodes used for radiation control are located at the maximum of the WSP mode intensity (reached at the metal-dielectric interface), maximizing the controlling efficiency and thus ensuring considerably lower power requirements. This feature opens unique perspectives for substantial reductions in sizes and energy of SP-based photonic components and circuits while extending the operation bandwidth [31]. It also allows one for the realization of conceptually new functionalities and exploitation of new materials with fascinating properties, such as single-crystalline gold films [32] or graphene [33], as demonstrated, for example, by realizing hybrid SP-graphene waveguide modulators [34]. The degree of WSP modulation can be enhanced by squeezing the WSP mode area and thereby increasing the slowdown factor. The associated increase in the WSP absorption has to be carefully investigated and analyzed to take advantage of the enormous potential of the aforementioned unique WSP characteristics. Proper analysis requires the further development of the theoretical models tailored suitably for dealing with nanostructured light-matter interactions.

**Spinoff directions** Investigations of the underlying physics and fundamental limitations associated with LSP excitations would be greatly beneficial for the whole field of flat optics based on phase and amplitude-gradient metasurfaces [35], allowing one to mould the radiation with an unprecedented control over its polarization and propagation characteristics as well as to produce surface coloration at nanoscale [36]. In the field of sustainable energy sources, LSP-induced resonance energy transfer becomes increasingly important for solar energy conversion [37]. Moreover, recent groundbreaking discoveries emphasize that even inevitable light absorption in plasmonics can be turned into gain within various topics: from plasmon-enhanced optical tweezing to thermal therapy and data storage [38]. Finally, mastering unique plasmonic features associated with the excitation of both LSP and WSP would most certainly provide additional possibilities for pairing photons with phonons in quantum optomechanics [39]. It is thus clear that studies related to the above objectives would have major consequences and implications to many subfields of modern nanoscience and emerging quantum technologies [40], including radiation-lifetime engineering, plasmon-enhanced chemistry, single-molecule sensing, quantum microscopy, quantum optics, and optomechanics in general and single-photon sources in particular.

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