The case for quantum plasmonics

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The discrete quantum nature of plasmons may be exploited to make efficient single-photon sources. Despite the losses associated with metallic resonators, advantages over dielectric counterparts exist when it comes to producing efficient quantum emitters.

Plasmonics offers unique possibilities for the manipulation of light at the nanoscale resulting in extreme light concentration and giant local-field enhancements[1], while encountering enormous problems in taking advantage of these possibilities due to inevitable absorption losses[2]. Although some applications where this absorption can be put to use have already been identified[3], the relative merits of using plasmonic structures versus employing the more mature dielectric-based (photonic) structures have to be carefully examined, with the absorption effects being properly taken into account. (We use 'photonics' when referring to purely dielectric structures.) This approach is also required in the emerging field of quantum plasmonics[4].

Since plasmonics entails interactions between electromagnetic fields in dielectrics and free-electron oscillations in metals, the term 'quantum plasmonics' has been used whenever either of these two interacting parts exhibit quantum features. Thus, it has often been broadly applied to all situations where quantum-mechanical effects (quantum confinement, electron tunnelling and so on) may play a role, although not necessarily portend new phenomena. Here, we consider a more focused definition of 'quantum' effects as those in which the bosonic nature of plasmons (or plasmon–polaritons) and photons into which these polaritons decay becomes prominent. The discrete quantum nature of bosons reveals itself when they are generated discretely, hence the term quantum emitter (QE). In general, QEs are needed for the realization of single-photon sources that are exploited in a number of exotic yet potentially very practical phenomena such as quantum sensing, cryptography, entanglement and teleportation, all pertaining to explosively developing quantum information technologies. When making the case for quantum plasmonics, we thereby concentrate on unique perspectives for the single-photon generation that are opened by plasmonics as compared with photonics.

QE coupling to environment

For single-photon sources to become a truly enabling technology for quantum communication and information processing, sufficiently high emission rates have to be secured. Meanwhile, QE intrinsic radiative lifetimes being of the order of 10 ns are certainly too long to meet stringent and ever-increasing requirements of optical-communication and information-processing systems. The QE spontaneous emission (SE) rate can however be increased by placing a QE in a suitable photonic environment with an increased electromagnetic local density of states[5].

The plasmonic alternative to photonics in this context has generally been considered from the point of view of boosting the Purcell factor, which represents essentially a ratio between the resonator quality factor and its volume normalized by the diffraction-limited volume of $(\lambda/2)^3$ (ref. 5), by making use of extreme field confinement and, in doing so, trying to exceed the performance of (diffraction-limited) photonic resonators, whose quality factors can easily amount to many orders of magnitude. Meanwhile, it seems that in the quest for the highest Purcell factors and, consequently, for the strongest coupling between QEs and surface plasmon (SP) modes, a very important objective of this quest, namely, speeding up the SE rate of QEs, was moved to the background by the objective of reaching the 'strong coupling' regime[6].

The strong-coupling regime, which claims so much attention of the quantum plasmonics community, is, in its essence, a classical phenomenon of coupling between two oscillating modes associated, respectively, with a QE and a resonator (containing the QE). This regime is reached when the coupling strength, characterized by the Rabi frequency Ω_R, which is proportional to the QE dipole transition moment increasing for smaller resonator volumes, exceeds the damping rates of these two oscillators[6]. In the opposite limit of strong damping for at least one oscillator (as is usually the case in plasmonics), the coupling is weak, resulting only in the SE rate modification characterized by the Purcell factor. Comparing plasmonics and photonics from the perspective of strong coupling, one quickly realizes that plasmonics with its inherently high absorption losses can hardly compete with photonics. Indeed, very fast dissipation of localized SP (LSP) modes determined by the electron collision frequency γ_m requires enormous coupling strength for the Rabi frequency Ω_R to reach γ_m (refs 6,7). Dielectric cavities based on photonic crystal structures do not impose this stringent requirement and have successfully been exploited for reaching the strong-coupling regime for single QEs, quantum dots, back in 2004[8,9]. The coupling constant scales with the square root of the number of oscillators[5], and the strong coupling in plasmonics was first realized (also in 2004) with a QE ensemble, cyanide dye J-aggregates, deposited on a silver film supporting the SP propagation[10]. Note that in the case of plasmonics, the strongly coupled modes give rise to new quasiparticles usually termed plasmon–exciton–polaritons, whose properties can be adjusted through their light and matter content, thereby opening new routes for light manipulation[11].

At the same time, the prospects of strong coupling between single QEs and plasmonic nanoparticles have increasingly started to appear rather bleak due to the necessity of bringing a QE very close to a metal nanoparticle supporting a strong LSP[7,12]. The strong-coupling regime (at room temperature and in ambient conditions) for a single QE was finally claimed in plasmonics this year by utilizing gap plasmon resonators with high quality factors and scaling the cavity volume to less than 40 nm^3, with the key evidence of single-molecule strong coupling being deduced from statistical analysis of vibrational spectroscopy time series and dark-field scattering spectra[13]. It should be emphasized that the utilization of gap plasmon resonators is pivotal in this experiment because of their unique feature of combining strong field confinement and high-quality resonance due to the dominant nature of the magnetic dipole resonance[14]. Note that gap plasmon resonators were also exploited in a recent demonstration of ultrafast room-temperature single-photon emission[15].

Emission rate enhancement

The fact that the strong-coupling regime is much easier to reach with photonics than with plasmonics might mislead one into assuming that the situation with boosting the SE rates is the same, simply because in both cases one has to reach large Purcell factors. But it may come as a big surprise that the situation with boosting the SE rates is quite the opposite, in other words, plasmonics allows one to exceed the diffraction-limited SE rate enhancement of photonics by two orders of magnitude[16]. The key point is that for both photonic and plasmonic configurations, the SE rate can be enhanced only up to the rate with which photons are leaving the system (cavity or plasmonic nanostructure). Further increase of the Purcell factor leads inevitably to a decrease in the rate of emitted out-of-system photons, while resulting in establishing well-developed Rabi oscillations. In general, the maximum SE rate is simply equal to the vacuum Rabi frequency under the condition of the SE rate and the out-of-system emission rate being equal[16]. This interplay is illustrated in Fig. 1, where the out-of-cavity SE rate enhancement, $\Gamma = \gamma^{\rm out}_{\rm SE}/\gamma_0$, is mapped as a function of the normalized cavity or LSP volume, $V_{\rm norm} = V(2/\lambda)^3$, and the quality factor of a cavity, $Q = \omega/\gamma_{\rm cav}$, or an LSP, $Q = \omega/\gamma_{\rm m}$, taken in the electrostatic limit along with adopting the Drude model for describing the metal dielectric function. Here, γ_0 is the SE rate in vacuum, ω is the cavity resonant frequency and $\gamma_{\rm cav}$ is the cavity emission rate. The borders between the regimes of weak and strong coupling for plasmonic and photonic configurations show the maximum values of the out-of-system SE rate enhancement.

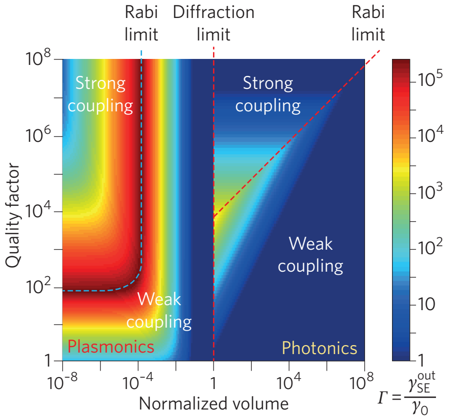

Figure 1 | Cavity-enhanced single-photon emission rate. The enhancement of the out-of-cavity SE rate as a function of the normalized volume and quality factor for both plasmonic (lossy) and photonic (diffraction-limited) configurations, calculated for the radiation wavelength of 1 μm and the vacuum SE rate $\gamma_0 = 10^8$ s^{-1} corresponding to the lifetime of 10 ns, and the gold plasma wavelength of 145 nm (ref. 4).

The map of SE rate enhancement highlights several important features, with the most prominent being the stark difference, by two orders of magnitude, between the SE rates achievable with plasmonic and photonic structures[16]. It is also seen that the maximum SE enhancement by photonic structures can be realized only with the diffraction-limited cavity, $V = (\lambda/2)^3$, while the maximum SE enhancement by plasmonic structures remains practically the same, $\Gamma^{\rm lim}_{\rm plas} \cong (\omega_{\rm P}/\gamma_0)^{2/3}$, for all parameters along the Rabi-limit line when $\gamma_{\rm m} + \gamma_{\rm rad} = (\omega_{\rm P}^2\gamma_0)^{1/3}$. Here, $\omega_{\rm P}$ is the plasma frequency in the considered metal and $\gamma_{\rm rad}$ is the rate of radiative LSP dissipation, which is proportional to the LSP volume. It should be clarified that this limit can only be achieved for sufficiently good metals, those that are characterized by the electrostatic resonance quality factor of close to or larger than 100 (Fig. 1). Perhaps the most nontrivial (surprising) and important feature of the considered map is that there is no need to strive for very small (lossy or lossless) plasmonic resonators. The optimum volume of an electrical dipole-like lossless plasmonic resonator found previously, ~10 nm^3 (ref. 16), can simply serve as the smallest plasmonic volume sufficient to ensure the highest SE rates. Smaller nanostructures are of course needed to reach the strong-coupling regime[13], but might be detrimental with respect to the SE rate enhancement.

Quenching and QE emission rates

Our physically transparent analytical model of single-photon emission in resonant structures[16] also helps to clarify and correct an existing misperception of the enhancement achievable in plasmonic cavities. Although many publications (for reviews see refs 5 and 17) have been devoted to evaluations of the SE enhancement in photonic[18,19] and plasmonic[15,20,21] structures, the direct comparison between these two classes is lacking, and, furthermore, it is not clear what is understood as 'enhancement' in many works. Quite often the enhancement of the SE rate is being confused with the enhancement the SE (luminescence) efficiency, which is strongly affected by the phenomenon of quenching. Quenching is caused by the fact that aside from the main ('bright') LSP dipole mode, SE also occurs into higher-order ('dark') LSP modes at a rate that can be at least as high, if not higher, than the emission into the bright mode, especially when a QE is placed very close to the metal surface[22]. Since all energy emitted into the dark modes eventually dissipates in the metal, the quantum efficiency of the out-of-system radiation is severely limited (quenched). Because of quenching, plasmonic nanostructures can only enhance the efficiency of originally very inefficient processes, such as Raman scattering. In contrast, if one assumes that the QE can be excited at a sufficiently high rate, quenching does not affect the out-of-system SE rate that can indeed be very high[16], although when it comes to the efficiency enhancement, plasmonic structures are far less appealing. Finally, quenching can still be harmful and limit the achievable SE rates as it can cause thermal damage when large excitation powers are used, thus one should avoid placing a QE in very close proximity to metal surfaces.

Parting thoughts

More work is required to refine the analytical description of single-photon emission in resonant structures[16] and improve its accuracy beyond the order-of-magnitude estimations. The treatment should be extended to the plasmonic resonators based on gap SP modes[13–15] with the dominant contribution coming from the magnetic dipole resonance[14]. Potential problems associated with the thermal damage mentioned above need to be considered. We are confident that that physical trends outlined here are faithfully described and provide pivotal guidelines for the development of new plasmonic configurations and materials to be exploited in quantum plasmonics. The key conclusion from the viewpoint of developing efficient and bright single-photon sources is that plasmonic resonant nanostructures, despite absorption loss in metal, hold inherent advantages over photonic (dielectric-based) resonators when it comes to enhancing the QE emission rate up to the sufficiently high levels required for quantum communication and computation systems. ❐

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References

1. Gramotnev, D. K. & Bozhevolnyi, S. I. *Nat. Photon.* **8,** 13–22 (2014).
2. Khurgin, B. *Nat. Nanotech.* **10,** 2–6 (2015).
3. Ndukaife, J. C., Shalaev, V. M. & Boltasseva, A. *Science* **351,** 334–335 (2016).
4. Tame, M. S. *et al. Nat. Phys.* **9,** 329–340 (2013).
5. Pelton, M. *Nat. Photon.* **9,** 427–435 (2015).
6. Torma, P. & Barnes, W. L. *Rep. Prog. Phys.* **78,** 013901 (2015).
7. Trügler, A. & Hohenester, U. *Phys. Rev. B* **77,** 115403 (2008).
8. Reithmaier, J. P. *et al. Nature* **432,** 197–200 (2004).
9. Yoshie, T. *et al. Nature* **432,** 200–203 (2004).
10. Bellessa, J., Bonnand, C., Plenet, J. C. & Mugnier, J. *Phys. Rev. Lett.* **93,** 036404 (2004).
11. Fernández-Dominguez, A. I., García-Vidal, F. J. & Martín-Moreno, L. *Nat. Photon.* **11,** 8–10 (2017).
12. Delga, A., Feist, J., Bravo-Abad, J. & García-Vidal, F. J. *Phys. Rev. Lett.* **112,** 253601 (2014).
13. Chikkaraddy, R. *et al. Nature* **535,** 127–130 (2016).
14. Pors, A. & Bozhevolnyi, S. I. *Opt. Express* **21,** 27438–27451 (2013).
15. Hoang, T. B., Akselrod, G. M. & Mikkelsen, M. H. *Nano Lett.* **16,** 270–275 (2016).
16. Bozhevolnyi, S. I. & Khurgin, J. B. *Optica* **3,** 1418–1421 (2016).
17. Aharonovich, I., Englund, D. & Toth, M. *Nat. Photon.* **10,** 631–641 (2016).
18. Noda, S., Fujia, M. & Asano, T. *Nat. Photon.* **1,** 449–458 (2007).
19. Lin, Z., Pick, A., Loncar, M. & Rodriguez, A. W. *Phys. Rev. Lett.* **117,** 107402 (2016).
20. Russell, K. J., Liu, T.-L., Cui, S. & Hu, E. L. *Nat. Photon.* **6,** 459–462 (2012).
21. Akselrod, G. M. *et al. Nat. Photon.* **8,** 835–840 (2014).
22. Khurgin, J. B. & Sun, G. *Appl. Phys. Lett.* **95,** 171103 (2009).