

Optimized nanoplasmonics

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2015-2021

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We have developed a novel optimization methodology, which is based on the synthesis of finite element method (FEM, COMSOL) and an in-house developed algorithm (GLOBAL), and makes it possible to determine the optimal configurations of various nanophotonical systems [1, 2].

We have optimized complex nanophotonical structures, which make it possible to improve the single-photon sources' and detectors' efficiency and single-molecule sensors' sensitivity, to achieve superradiance, non-classical light generation and strong-coupling, to enhance few-cycle plasmonic fields and to balance the energy deposition. These results were delineated in framework of invited talks [3-6] and presented in several international publications on the specific topics [7-62].

1. Improvement of diamond color centers via monomer and dimer nanoresonators

A numerical method has been developed to optimize coupled emitter-nanoplasmonic nanoresonator (NR) systems. The emitters were different diamond color centers acting as single-photon sources. Our primary purpose was to determine the right objective function (OF) having a potential to maximize the fluorescence enhancement of SiV and NV diamond color centers, when they are coupled to individual silver or gold NRs. First, we examined two methods: Purcell factor (quantum efficiency (QE)) maximization by setting a criterion on the QE (Purcell factor) that have to be parallel met, respectively. Then optimization was performed to maximize excitation and emission separately with composite objective functions of radiative rate enhancements $\delta R_{\text{excitation}} = \text{Purcell}_{\text{excitation}} * \text{QE}_{\text{excitation}}$ and $\delta R_{\text{emission}} = \text{Purcell}_{\text{emission}} * \text{QE}_{\text{emission}}$, respectively. Finally, the optimization to maximize these phenomena simultaneously was realized with $\delta R_{\text{excitation}} * \delta R_{\text{emission}}$ nominated as P_x factor and parallel set conditions regarding the $\delta R_{\text{excitation}}$ and $c\text{QE}_{\text{emission}}$. In order to conclude about the right objective functions and criteria, as well as to obtain systems consistent with the material, structural and spectral limits, a comparative study was performed on the optimal systems determined by different OFs and criteria [7, 8, 9]. It was proven, that the achieved optimal configurations compose the same trajectory of optimal solutions, i.e. lie on the same curve in the high-dimensional parameter-space, but typically in significantly different regions for different optimization approaches.

In case of individual silver nanorods optimized to achieve the most efficient excitation, emission or simultaneous enhancement both (P_{crit} and QE_{crit}) approaches resulted in optimal configurations exhibiting peaks in radiative rate enhancement around the excitation, emission and both wavelengths, respectively. In case of individual gold nanorods, the resonance peaks are always detuned from the excitation wavelength, independent of the configuration type or method of optimization, which is a consequence of material limits. Because of the large gaps in the trajectory of optimal solutions obtained with QE_{crit} method, P_{crit} proved to be a more suitable optimization procedure [7].

It was demonstrated that the excitation and emission of diamond color centers can be enhanced either separately or simultaneously with individual metal nanorods, but with a characteristics and degree depending on the material [8]. A definite relation was evidenced between the studied systems: silver nanorod is more suitable to enhance fluorescence (either separately or simultaneously) and QE for both color centers. The emission and QE of SiV can be better enhanced by either of silver or gold individual nanorods, than the fluorescence of NV. At excitation SiV exhibited larger (smaller) enhancement in case of individual gold (silver) nanorods, which was explained by the perpendicularity of SiV dipoles at excitation and emission. It has been shown that the material limits arising from metal dielectric properties can be approached by simultaneous optimization, and the resulted configurations enhance the emission preferably [8].

The optimization methods using P_{crit} and Q_{crit} were used to determine the optimal configuration of diamond-metal core-shell NRs as well [9]. When the color center was centralized inside a core-shell monomer, a single Purcell factor and δR peak appeared. These peaks could be properly tuned to the excitation or emission wavelength, which proved the suitability of our optimization method in case of concave NRs as well. Decentralized dipole results in several higher order resonances in the Purcell spectrum, however caused by the low achievable QE, the δR radiative rate enhancement is small. Silver shell and SiV color center coupled systems proved to be better in terms of fluorescence and QE improvement among the core-shell NRs as well [9].

Further optimizations of fluorescence enhancement achievable with aluminum and copper concave and convex individual NRs revealed, that silver is the most promising in the inspected spectral interval. Therefore, in further studies silver and gold were used, the advantage of gold is that it is chemically inert [10, 11].

Although, optimization with P_{crit} and QE_{crit} criteria proved to be appropriate methods to determine efficient nanophotonical systems, maximization of the product of radiative rate enhancements at the excitation and emission (P_x factor) has an unambiguous physical meaning. In case of monomers, we have shown that there is a trade-off between the P_x factor and the corrected quantum efficiency (cQE), hence a minimal cQE criterion was set, that has to be met at emission.

The coupled systems of SiV color centers and concave diamond-silver core-shell monomer NRs have been optimized to maximize the fluorescence enhancement by maximizing the P_x factor with different cQE criteria (~20 – 30 – 40 and 50%) [12]. Spherical, ellipsoidal and rod-like core-shell resonators were investigated and the effect of emitter decentralization was also inspected. Optimization of spherical NRs showed that ~500-fold P_x factor increase can be achieved with ~50% cQE. A single δR peak (originating from a bonding resonance) appears around the emission wavelength, hence contribution of the excitation enhancement to the total fluorescence amplification is not significant. Decentralization is not beneficial despite the hexapolar resonance that can be tuned to the excitation due to symmetry breaking, caused by the nonradiative nature of the mode. By elongating the NRs, a bright transverse dipolar resonance can be tuned to the excitation wavelength, while a bright longitudinal dipolar resonance can enhance the emission. Due to degeneration lifting during appearance of transversal and longitudinal modes, three orders of magnitude larger P_x factor can be achieved with 50% cQE with a slightly decentralized emitter inside an ellipsoidal resonator. Due to the more pronounced antenna properties, significantly larger P_x factor can be achieved with a rod-like core-shell resonator. The conditional optimization revealed that even larger P_x factors can be achieved at the expense of cQE [12].

Two metal nanorods were placed close to each other to create a NR dimer. The P_x factor of a diamond color center deposited at the center of the resulting nanogap was conditionally maximized. The interacting nanorods result in a large **E**-field enhancement at the color center position in the nanogap, while properly tuned antenna modes facilitate to outcouple light into the far-field with larger efficiency. As a result, nanorod dimers are capable of promoting higher P_x factor and cQE than the corresponding monomers independent of the NR material and emitter type [13]. Two types of nanorod dimers were examined. First the identity of the individual nanorods forming the dimer was required (symmetrical nanorod dimer), second, they were allowed to be different (asymmetrical nanorod dimer). The predefined symmetry determines the charge, near-field and resistive heating distribution on the components. By allowing asymmetry, a higher P_x factor can be achieved in each case, according to the increased number of independently variable parameters [14].

Comparing highly asymmetric systems with their symmetric counterparts, up to an order of magnitude higher fluorescence enhancement is achievable. Co-existent antenna modes promote the excitation in all asymmetric silver nanorod dimers, while uniform modes accompanied by parallel dipolar surface modes enhance the emission. In case of gold nanorod dimer there is only a local surface charge separation at the excitation, while at the emission it depends on the type of color center, whether different modes co-exist (NV) or only uniform modes appear (SiV). With gold less significant enhancement can be achieved at the excitation wavelength, whereas in case of silver the specific configuration determines which phenomenon is more efficiently enhanced. In case of silver nanorod dimer, the moderated increase of P_x factor is accompanied by a decrease in cQE, while in the case of gold nanorod dimer, both the P_x factor and cQE are enhanced. The largest fluorescence and cQE is achieved with silver nanorod also in case of dimers. However, in contrast to monomers, for asymmetric silver dimers the NV color center outperforms SiV color center in terms of P_x factor [14]. Conditional optimization revealed that 90% cQE could be achieved with silver nanorod dimers with a penalty of significant P_x factor reduction. For gold nanorod dimers, the maximum available efficiency is around 60% [13, 14].

Optimization of spherical and ellipsoidal monomer and ellipsoidal dimer core-shell NRs was performed to maximize the P_x factor [15, 16, 17]. The hexapolar dark mode appearing on silver spherical monomer has no noticeable SiV excitation improving effect, accordingly enhancement at the emission is always larger than at the excitation [16]. One order of magnitude increase is achievable in the P_x factor, when silver monomer NR elongation is enabled [15, 16]. In contrast, significant increase (slight decrease) is noticeable in the SiV-gold monomer (NV-gold monomer) configuration's P_x factor, when elongation is enabled [17]. The larger SiV fluorescence enhancement originates dominantly from the excitation enhancement due to a quadrupolar resonance on silver ellipsoid monomer [16]. In contrast, the SiV fluorescence enhancement originates exclusively from the more strongly increased emission via gold ellipsoid monomer [17]. The SiV-ellipsoid monomer always outperforms the NV-ellipsoid monomer configuration in P_x and cQE as well [15-17]. In case of monomers more than one order of magnitude larger P_x factor is achievable with silver than with gold [17]. Dimers of silver ellipsoid NRs allow moderate SiV fluorescence but large QE enhancement at the excitation and emission as well. Enabling asymmetry in this configuration promotes to achieve larger P_x enhancement and allow smaller cQE, but the reached values still outperform the cQE achieved in case of SiV-silver monomers [16]. Compared to SiV coupled configuration the bright dimer mode enhances the fluorescence more efficiently in NV – silver dimer configuration, as a cost of moderated cQE [15]. Although, the positive effect of asymmetry is not significant, the largest (eight orders of magnitudes enhancement) P_x factor is achieved via the asymmetric NV – silver ellipsoidal dimer nanoresonator among all inspected coupled systems [15, 16, 17]. Although, gold core-shell dimer NRs provide significant P_x enhancement with respect to monomers, silver dimer NRs show orders of magnitude larger fluorescence enhancement and larger cQE as well [17].

A comparison of dimers composed of different metals shows that larger P_x factor can be achieved with silver in case of core-shells, except the SiV-symmetric silver dimer, whereas with gold in case of symmetric core-shell dimers and asymmetric nanorod dimers, independent of color center type [18]. In terms of quantum efficiency, the core-shell NR is better, except in NV-silver dimer configurations, which is due to the reduced amount of metal [18].

Correlations between cQE criterion, optical response and geometric properties of symmetric ellipsoid core-shell dimers were also examined [19]. The trade-off between the P_x factor and cQE remains valid for dimers as well. We have shown that 10^8 - 10^7 enhancement of P_x factor is reachable with a cQE of 38-92% via silver core-shell dimers.

Orders of magnitude smaller P_x factor but lower cQE of 11-55% are achievable via gold core-shell dimers. Although, the antenna outcoupling efficiency can be increased by increasing the NR size in both cases, larger antenna efficiency is achievable via more ellipsoidal and hollow NRs in case of silver, in contrast more spherical and solid NRs are preferred in case of gold [19].

Geometry of silver and gold core-shell dimers embedded into extended diamond medium have been optimized to maximize SiV and NV color center fluorescence [20]. The objective function was the P_x factor weighted by the directivity enhancement (nominated as D_x factor) to account for the symmetry breaking due to the single sided plane-wave illumination. During the optimization (i) symmetry of composing NRs was restricted, (ii) asymmetry was allowed to enlarge the parameter space and to maximize fluorescence. With D_x factor as the objective function and extended diamond medium the advantage of asymmetry was proven in case of silver and gold core-shell dimer coupled NV center [20].

Conclusion: We have developed a method, which makes it possible to extract the total decay rate and radiative decay rate of dipolar emitters in homogenous and plasmonic environments, as well as to compute the non-radiative losses by determining the Joule heating via frequency domain FEM in COMSOL. The numerical method allows conditional optimization with arbitrary composite objective functions and criteria. During optimization the geometry of NRs, as well as the location and orientation of the dipoles have been varied. The optimizations have been performed for the wavelengths of excitation and emission separately, as well as for both wavelengths simultaneously.

2. Superradiance of multiple diamond color centers in core-shell nanoresonators

Different (P_x and $P_x \cdot \text{cQE}$) objective functions were used to achieve superradiance (SR), via different (4 and 6) number of dipolar emitters modeling SiV diamond color centers, embedded into various (bare and coated) types of NRs with different (spherical and ellipsoidal) geometries, as well as in case of different (symmetrical, broken symmetry) properties of the emitter arrays were subjects of comparative studies.

2.1 Effect of objective function

Comparative study has been performed on different types of concave spherical NRs determined by using different objective functions [21]. We inspected diamond-silver core-shell (bare type) and diamond-silver-diamond core-shell-shell (coated type) spherical NRs seeded by 4 or 6 emitters. These systems were optimized either to maximize the total fluorescence enhancement (P_x factor), or the product of the P_x factor and the corrected quantum efficiency ($P_x \cdot \text{cQE}$) at the emission wavelength. NRs optimized by the $P_x \cdot \text{cQE}$ function have larger geometries and larger emitter distances from the metal. The quantum efficiency at the excitation and emission wavelength is larger compared to NRs optimized by P_x . The Purcell factor is smaller at both wavelengths, whereas the radiative rate enhancement is larger at excitation and smaller at emission. These NRs are more efficiently superradiant according to the larger FWHM, smaller Q factor and more well-defined bad-cavity characteristics achieved using the $P_x \cdot \text{cQE}$ function, which are advantageous in cooperative fluorescence enhancement. In contrast, using the P_x total fluorescence enhancement as the objective function results in high-Q resonators, which are more effective in non-cooperative fluorescence enhancement. All optimized systems are superradiant, namely the fluorescence enhancement becomes proportional to the N^2 of emitters. Moreover, the superradiance is accompanied by indistinguishability in case of NRs seeded with 4 emitters, whereas two branches appear in the Purcell factor in case of seeding by 6 emitters. Linewidth narrowing is achieved in all NRs (except when 4 emitters are embedded into coated spherical NRs).

2.2 Effect of array symmetry in spherical and ellipsoidal nanoresonators

Different types of concave spherical and ellipsoidal NRs were optimized by the $P_x \cdot cQE$ function to achieve superradiance: bare or coated type NRs of spherical and ellipsoidal geometry seeded by 4 or 6 emitters not only in symmetrical but also broken-symmetry arrays [22, 23]. NRs seeded by a broken-symmetry SiV diamond color center array were compared to their counterparts seeded by a symmetric array of emitters.

In case of bare type NRs the larger total volume allows SR in broken-symmetry arrays independently of the number of emitters and the shape of the NR, which results in larger scattering and extinction cross-section. In spherical bare NRs all of the quantum efficiency, the Purcell factor and the radiative rate enhancement are larger at the excitation wavelength (except the QE in case of 4 color centers). At the emission, the quantum efficiency is larger, whereas the Purcell factor and the radiative rate enhancement are smaller. In ellipsoidal bare NRs the quantum efficiency is larger, whereas the Purcell factor and the radiative rate enhancement is smaller/larger in case of 4/6 emitters at the excitation wavelength. In contrast, at the emission all quantities are smaller. In spherical bare NRs the P_x and $P_x \cdot cQE$ is smaller in broken-symmetry arrays. Smaller/equal P_x and $P_x \cdot cQE$ are achieved in broken-symmetry arrays of 4/6 emitters in ellipsoidal bare NRs.

The spherical coated NRs are considerably smaller for broken-symmetry arrays, which allows smaller scattering and extinction cross-section. In case of ellipsoidal coated NRs, the scattering and extinction cross-sections are larger for broken-symmetry arrays, similar to the bare type NRs. In spherical coated NRs both at the excitation and emission the quantum efficiency is smaller, whereas the Purcell factor and the radiative rate enhancement are larger (except the radiative rate enhancement at the excitation in case of 4 emitters). In ellipsoidal coated NRs the quantum efficiency is smaller, whereas the Purcell factor and the radiative rate enhancement is larger/smaller in case of 4/6 emitters at the excitation wavelength. At the emission, the quantum efficiency is larger/smaller, whereas the Purcell factor and the radiative rate enhancement are larger (except δR in case of 4 emitters). Larger P_x and $P_x \cdot cQE$ is achieved in broken-symmetry embedding coated NRs (except $P_x \cdot cQE$ in case of 6 emitters in spherical coated NRs, P_x and $P_x \cdot cQE$ in case of 4 emitters in ellipsoidal coated NRs). As a result, broken-symmetry array seeded coated NRs are dominantly better in superradiance.

Based on the superradiance performance both in symmetrical and broken-symmetry configurations bare type NRs are better in spherical geometry, whereas coated type NRs are preferred in ellipsoidal geometry (except when 6 emitters arranged into symmetric array, which turns out to be better in bare NR). Increasing the number of emitters is advantageous in both (bare) spherical (ellipsoidal) NRs.

2.3 Effect of nanoresonator geometry in case of symmetrical emitter arrays

Detailed comparative study has been performed on concave spherical and ellipsoidal NRs based on geometrical properties, optical responses and superradiance performance. We inspected diamond-silver core-shell (bare type) and diamond-silver-diamond core-shell-shell (coated type) type NRs seeded with 4 or 6 emitters in symmetrical arrays [24]. In ellipsoidal NRs the long axis is similar to the radius, whereas the silver shell is thicker compared to the spherical NRs. In spherical NRs a single peak appears on the scattering and extinction cross-section, as well as on the Purcell factor and radiative rate enhancement spectrum at the emission wavelength. In contrast the ellipsoidal NRs can enhance the excitation as well. Accordingly, in the (excitation and) emission configuration a global maximum appears near the (excitation and) emission wavelength in these quantities in (ellipsoidal) both NR geometry.

The spherical NRs exhibit a minimum in the quantum emission spectrum at the excitation, and a maximum at the emission wavelength. In contrast, the ellipsoidal NRs show a quantum efficiency maximum at both wavelengths in the corresponding configurations. The quantum efficiency is smaller (larger) in bare (coated) ellipsoidal NRs than in their spherical counterparts at the excitation wavelength. Larger (smaller) Purcell factor and radiative rate enhancement is achieved in ellipsoidal NRs than in the spherical NRs at the excitation (emission) wavelength. At the emission, the ellipsoidal NRs outperform the spherical ones in both types of NRs. The P_x and the $P_x \cdot cQE$ are larger in ellipsoidal NRs. All types of optimized NRs are superradiant, but the ellipsoidal NRs override the superradiance threshold better. More well-defined bad-cavity-characteristics is achieved in case of ellipsoidal NRs, which is advantageous in cooperative fluorescence enhancement. Indistinguishability is achieved in all NRs seeded with 4 emitters.

Based on SR performance bare type spherical and ellipsoidal NRs seeded by 6 emitters are better, since they exhibit better superradiance performance despite the smaller frequency pulling and larger fluorescence enhancement due to smaller detuning. These results on symmetric SiV diamond color center arrays embedded into concave NRs proved the advantage of ellipsoidal geometry in superradiance.

2.4 Effect of nanoresonator geometry in case of broken-symmetry emitter arrays

Comparison based on the geometrical and optical responses has been made on different types of concave spherical and ellipsoidal NRs: bare or coated type NRs seeded by 4 or 6 emitters were inspected [25, 26]. In both types of NRs the volume of the core is smaller in ellipsoidal geometry, whereas the metal layer thickness and the total volume is smaller (larger) in bare (coated) type NRs.

In case of bare type nanoresonators the quantum efficiency is smaller, whereas the Purcell factor and the radiative rate enhancement is larger at the excitation wavelength in ellipsoidal NRs. In contrast, at the emission wavelength the quantum efficiency is larger, whereas the other two quantities are smaller. Accordingly, the ellipsoidal bare NRs enable stronger excitation enhancement but allow weaker emission enhancement. As a result, the P_x and $P_x \cdot cQE$ are significantly larger in ellipsoidal bare NR. Linewidth narrowing occurs compared to the cold cavity in ellipsoidal (spherical) bare NRs in Purcell factor (radiative rate enhancement). Accordingly, the spherical bare NRs promote better collectivity, despite that the ellipsoidal bare NRs have low-Q-factor indicating more well-defined bad-cavity characteristics.

In case of coated type nanoresonators the quantum efficiency, Purcell factor and radiative rate enhancement is larger at the excitation wavelength in ellipsoidal NRs. At the emission wavelength the quantum efficiency is larger, whereas the other two quantities are smaller. Accordingly, the ellipsoidal coated NRs have stronger excitation enhancement, but weaker emission enhancement. As a result, the P_x and $P_x \cdot cQE$ are larger in ellipsoidal coated NRs, moreover exhibit a linewidth narrowing in Purcell factor and radiative rate enhancement, which is advantageous to achieve lasing. Accordingly, the ellipsoidal coated NRs promote better collectivity as well as they show more well-defined bad-cavity-characteristics.

In all inspected NRs 4 (6)-fold radiative rate enhancement is achieved with respect to the reference system in case of 4 (6) emitters. Bare type NRs enhance fluorescence in a broad spectral interval, except near 600 nm (500 nm), where there is a quadrupolar resonance in the reference system in case of spherical (ellipsoidal) geometry. Coated type NRs enhance fluorescence in a narrower spectral interval, and there is a quadrupolar resonance near 600 nm (525 nm) in the reference system of spherical (ellipsoidal) NRs. On every NRs hexagonal charge distribution appears at the excitation, and dipolar charge distribution develops at the emission wavelength.

The FWHM of all spectral responses is larger, accordingly the Q factor is more than two-times smaller, which indicates a more well defined bad-cavity characteristics for ellipsoidal NRs, that is advantageous to achieve superradiance. Ellipsoidal NRs exhibit larger frequency pulling in accordance with the smaller Q factor and better superradiance performance than their spherical counterparts (except the bare NRs seeded by 4 emitters). Based on these results ellipsoidal NRs are proposed to achieve superradiance via broken symmetry arrays as well.

In either NRs 6 emitters are not indistinguishable, there is a split in the Purcell factor distance dependency spectrum at the excitation wavelength in case of small emitter distance from the metal. This is caused by the cross-polarization of the **E**-field in the NRs. When the emitters are close to the metal shell, they behave non-collectively, and there are two sets of emitters: the two emitters on the x-axis and the four emitters rotated from the x-axis by the same angle. When the distance from the metal increases, the emitters start to behave collectively, and the split on the Purcell-factor distance dependency spectrum vanishes.

Conclusion: It was shown that both P_x and $P_x \cdot cQE$ optimization functions can be used, but the $P_x \cdot cQE$ is capable of promoting cooperative fluorescence due to the resulted more-well defined bad-cavity characteristics. Multiple emitter-plasmonic NR coupled systems were designed, which allow to reach emission on the level of superradiance. It was shown that the N^2 -fold P_x factor enhancement with respect to the reference system consisting of a single emitter is due to excitation and emission enhancement, which approximates and overrides N-fold increase, respectively. The distance dependent Purcell factor was inspected to prove the indistinguishable nature of 4 emitters, whereas the co-existence of two branches was proven either in symmetric and in broken symmetry arrays of 6 emitters.

3. Lasing and spasing via plasmonically enhanced stimulated emission

Various plasmonic NRs were designed to optimize the lasing phenomena, including a singlet convex nanoparticle, periodic and complex convex metal patterns covered by gain media, as well as a singlet concave core-shell nanoparticle, periodic and complex concave metallic patterns embedding gain media. The effect of the spectral overlap between the plasmonic resonances and the molecular excitation and emission lines on the lasing characteristics including the threshold behavior, slope efficiency, line-width narrowing, coherence properties, and transient oscillations was studied.

We have optimized the geometry of nanorods and core-shell nanoparticles coated by a gain medium to maximize stimulated emission in the near-field (NF-c-type NRs) and the out-coupling into the far-field (FF-c-type NRs) [27]. At moderate dye concentration the characteristic parameters, including the smaller lasing threshold, larger slope efficiency, larger achieved near-field intensity are better for both NR types based on nanorod. In addition, the FF-c-type nanorod possesses smaller threshold of negative gain absorption and amplified power outflow as well as smaller threshold of dip-to-peak transition in power outflow spectrum, moreover it exhibits a slightly larger external quantum efficiency. The advantage of NF-c-type and FF-c-type core-shell based NRs is that they show smaller bandwidth in the near-field and in the far-field, respectively. Post-optimization increase of the concentration up until an appropriate level (NF-c*-type NRs) ensure zero crossing in the extinction cross-section, which phenomenon is considered as the criterion of spasing transition, according to the literature. At high concentration the core-shell NR outperforms the nanorod by showing smaller lasing, power outflow, absorption and extinction cross-section thresholds, smaller near-field bandwidth and larger internal and external quantum efficiencies.

In comparison the nanorod remains better in larger slope efficiency and larger achieved near-field intensity. The novelty of our work is that the dipolar and quadrupolar modes' competition was uncovered and the achievable almost uniform far-field distribution was demonstrated.

Conclusion: The optimization method we have developed makes it possible to design NRs for user defined purposes, either to maximize the near-field enhancement or the far-field out-coupling via stimulated emission. The proofs of non-classical light generation can be extracted from numerical pump-and-probe computations. It can be demonstrated that by changing considerably one of the configuration parameters, e.g. the dye concentration, the optical response may undergo a significant modification, e.g. zero-crossing in the extinction cross-section can be achieved. As a result, transition between lasing and spasing operation regions can be controlled via configuration parameters' tuning. The uncovering of the mode competition opens novel avenues to realize Q-switching in plasmonic nanolasers. A robust method is provided to optimize plasmonic NRs for different application purposes.

4. Strong-coupling and lasing assisted by grating-coupled SPPs

Our studies on periodic patterns revealed that short-range and long-range SPPs (SR-SPP and LR-SPP) can be strongly coupled in the right azimuthal orientation of multilayers consisting of wavelength-scaled gratings, which results in two branches exhibiting a typical avoided crossing on the dispersion map. The upper branch corresponds to the LR-SPP mode, which shows a two-fold antisymmetric E_y component and E_{norm} accumulation on the right edge of the valley, accompanied by a weak-glass-side enhancement. The lower branch corresponds to the SR-SPP, the E_y component of which is horizontally antisymmetric but vertically slightly hybrid, and the E_{norm} shows a glass side enhancement along the edge of the valley (further details are provided in section #6 about bioplatforms).

We have inspected how the dipolar absorbers' and emitters' coupling to the SPPs is influenced by the oscillator frequency and strength, and by the symmetry of modes on the branch overlapping with it [28, 29]. The coupled modes dispersion characteristics was computed in reflectance and absorptance. The modes' symmetry was inspected at the extrema on the wavelength dependent optical responses computed at tilting corresponding to minima on the angle dependent reflectance interrogated at 532 nm.

Both the size of the split and the resulted modes characteristics depend on, whether the oscillator corresponds to an absorber or an emitter. The split increases by increasing the strength of an absorbing (emitting) molecular oscillator used to achieve strong-coupling (emission enhancement), however the degree of splitting depends on the oscillator frequency as well. In case of an oscillator with absorption band overlapping with the lower branch the split is larger at larger tilting and the mode arising on the secondary lower branch exhibits LR-SPP characteristics. In case of an oscillator with absorption band overlapping with the upper branch the split is larger at smaller tilting and the mode arising on the secondary lower branch exhibits LR-SPP characteristics. It is oscillator strength dependent, which tilting results in larger split.

An oscillator overlapping with the gap appearing on the dispersion map of the wavelength-scaled grating results in LR-SPP appearance on the secondary lower branch originating from the upper branch both at smaller and at larger tilting for both inspected oscillator strengths. In addition to this LR-SPP appears on the secondary upper branch originating from the upper branch for the case of smaller oscillator strength.

In case of split on the upper (lower) branch LR-SPPs arise on the lower (upper) secondary branch, accordingly the two-fold anti-symmetry is more often for the branches on the side of the gap. Typically, LR-SPPs arise on the lower secondary branch in case of strong-coupling of absorbing oscillators, whereas LR-SPPs promote lasing on the upper secondary branch of emitting oscillators.

Conclusion: Our studies have proven that both of the strong-coupling and lasing phenomena can be improved, when absorbing and emitting molecular oscillators interact with LR-SPPs, respectively.

5. Infrared single-photon detector (SNSPD) improvement

Superconducting nanowire single photon detectors (SNSPD), which are widely applied in quantum information processing, were integrated with one- and two-dimensional plasmonic structures to maximize absorption and to control the polarization contrast (PC) [1-3, 30-42]. Different types of integrated structures were optimized first to maximize the absorptance (A-SNSPD) [30, 31, 39], as well as the polarization contrast with (C-SNSPD) [3, 30, 35, 36] and without (P-SNSPD) a criterion regarding the absorptance that have to be parallel met [3, 31, 32, 33, 34, 35, 36]. The nanophotonical phenomena resulting in significant absorptance and polarization contrast have been compared for different types of plasmonic structure integrated SNSPDs. Devices optimized with a composite objective function of the product of absorptance and polarization contrast were also inspected (APC-SNSPD) [37, 38]. Finally, SNSPDs were designed to read out quantum information polarization independently (X-XX-DDX-O-SNSPD) [33, 35, 40, 41, 42]

5.1 SNSPDs to maximize p-polarized absorptance

The inspected A-SNSPD and C-SNSPD devices were nano-cavity (NCAI)-, nano-cavity-deflector (NCDAI)-, nano-cavity-double-deflector (NCDDAI)-, and nano-cavity-trench-(NCTAI)-array-integrated SNSPDs. The investigations have been performed in three different periodicity intervals, where Bragg scattering, Rayleigh phenomenon, and extraordinary transmission is expected. The highest absorptance was achieved via half-wavelength-scaled periodic structures, except in NCTAI-SNSPDs. The highest 95.05% absorption has been achieved via wavelength-scaled NCTAI-A-SNSPD, which ensures shorter reset time due to the larger periodicity. Different nanophotonical phenomena are at play at the absorptance maxima: plasmonic Brewster angle (PBA) / wide plasmonic pass band / crossing bands of localized cavity and grating coupled modes in NCAI-/NCDAI-/NCDDAI-SNSPD. In NCTAI-SNSPD the maximal absorptance is achieved inside an inverted plasmonic minigap, which appears due to the coexistence of coupled modes propagating forward and backward [30, 31]. Devices determined by optimizations performed to maximize absorptance resulted in A-SNSPD systems, which make it possible to achieve 10^2 - 10^3 - 10^4 - 10^2 polarization contrast.

5.2 SNSPDs to maximize polarization specificity by allowing different maximal tilting

Optimization performed to maximize the polarization contrast resulted in 10^2 - 10^{11} - 10^{13} - 10^3 & 10^5 values in integrated P-SNSPD systems optimized by allowing 85° and 80° maximal tilting [32, 33, 34]. Comparison of dispersion characteristics in p- and s-polarized absorptance shows that the achieved polarization contrast maximum is due to the s-polarized absorptance suppression at large tilting [32]. In all systems the polarization contrast exponentially increases with increasing tilting, except at local modulations originating from grating coupling. The PBA related light tunneling is more dominant in P-85-SNSPD and is just noticeable in P-80-SNSPDs [33, 34].

The PBA phenomena appear at the boundary of the second Brillouin zone. All optimized P-SNSPD devices show maximal polarization contrast at the maximal 85° and 80° tilting, i.e. close to the PBA. However, in NCTAI-P-85 the grating coupling promotes to reach maxima at tilting smaller than the PBA. One single exception is the wavelength-scaled NCTAI-P-80-SNSPD, where also the contrast maximum is reached at perpendicular incidence at the center of a minigap. The extremely large polarization contrast in NCDAI-P and NCDDAI-P-SNSPD indicates the polarization selection role of the inserted gold deflectors.

The polarization contrast exhibits correlation in all cases with the (extended) cavity length in quarter-wavelength units, additionally with the NbN/Au volume fraction except in NCAI-P-85 and with the maximal p-polarized absorptance, except in NCTAI-P-80, but correlates with the absorptance at the polarization contrast maximum only in NCDDAI-P-85.

5.3 SNSPDs to maximize polarization specificity with conditions on absorptance

We have performed conditional maximization as well, which have shown that a 3% penalty on absorptance allows to reach 10^2 - 10^7 - 10^{10} - 10^2 polarization contrast in C-SNSPDs [30]. In C-SNSPDs correlation between the polarization contrast and the NbN/Au volume fraction ratio as well as the normalized cavity length parameters was shown [3, 35].

The nanophotonical phenomena resulting in absorptance maxima in A-SNSPDs optimized to maximize absorptance were compared to the coupling of modes in devices optimized to maximize the polarization contrast with A_{\max} -1/2/3% (C1/C2/C3-SNSPD) and without (P-SNSPD) a criterion regarding the absorptance [36]. In NCAI-A-C-P-SNSPDs and NCDAI-C-P SNSPDs all of the maxima appearing at the PBA and in NCDAI-A-SNSPD at a transitional tilting are promoted by second order coupled modes having a wavelength slightly shorter than SPPs. In case of NCDDAI-A-SNSPD SPPs coupled at small tilting, in C-SNSPDs shorter wavelength modes coupled at large tilting, whereas in P-SNSPD Brewster-Zenneck waves excited at tilting corresponding to PBA promote the absorptance. In NCTAI-A and C1 / C2 and C3 / P-SNSPD coupling into SPPs and photonic modes in (+/-1) order at perpendicular incidence / into SPPs in (-2) order at PBA / into Brewster-Zenneck waves in (-2) order at a transitional tilting promotes absorptance. The increase of the NbN/Au volume fraction makes it possible to meet increasing absorptance criteria, with a few exceptions, where unique nanophotonical phenomena come to play. In P-SNSPDs compromised absorptance decrease of 5.14-16.84-22.43-25.97% makes it possible to reach 1.44 - $3.38 \cdot 10^8$ - $6.99 \cdot 10^8$ - $1.54 \cdot 10^1$ -fold contrast increase compared to A-SNSPDs.

5.4 SNSPDs to maximize absorptance and polarization specificity simultaneously

Optimization was performed by improving the mesh on A-SNSPDs and P-SNSPDs, as well as by using the product of absorptance and polarization contrast as the objective function ($FOM=A \cdot PC$), with absorptance criteria (APC1: A_{\max} -1%, APC3: A_{\max} -3%), and without a criterion regarding the accompanying absorptance (AP-SNSPD) [37, 38].

In NCAI- and NCDAI-SNSPDs optimized by applying a denser mesh the $A \cdot A \cdot PC \cdot PC$ maxima are promoted by second order coupled modes having a wavelength shorter than that of SPPs. The maximum in p-polarized absorptance appears at the PBA in all NCAI-SNSPD, whereas in NCDAI-SNSPDs the tilting increases and approximates gradually the PBA by modifying the absorptance criterion throughout A->P-SNSPDs transition. By decreasing the criterion regarding the absorptance, a gradually smaller NbN/Au fraction is allowed by the gradually widening and elongating deflectors.

In NCDDAI-A and APC1 (+/-1) order coupling of short-wavelength and SPP modes makes it possible to maximize the p-polarized absorptance close to perpendicular incidence, whereas the A*PC quantity is promoted by (-2) order coupling of short wavelength and SPP modes, respectively. In APC3-SNSPD the (-2) order coupling of SPP modes makes it possible to maximize the p-polarized absorptance and the A*PC at transitional tilting. In AP- and P-SNSPD (-2) order coupling of Brewster-Zenneck waves occurs at the maximum of p-polarized absorptance and of the A*PC (as well as of the PC) quantities. By gradually decreasing the absorptance criterion, first the deflector asymmetry increases and then the NbN/Au volume fraction decreases significantly in between the A and P devices.

In NCTAI-A (APC1-APC3-AP-P-SNSPD) (+/-1) order coupling occurs at perpendicular incidence into SPP (photonic modes). Exceptions are APC1 in A*PC and P-SNSPD in PC quantity, which maximum originates from +/-1 and -1 order coupling of short wavelength modes at small and zero tilting, respectively. In all NCTAI-SNSPDs the PC maxima appear close to or at perpendicular incidence, in contrast to NCAI-NCDAI-NCDDAI-SNSPDs, where the PC maxima appear uniformly at 85° and are promoted by (-2) order coupled modes. By decreasing the absorptance criterion the NbN/Au volume fraction non-monotonously decreases, however in APC1 and APC3 systems the asymmetry in deflector width is noticeably increased.

The largest 95.5% absorptance can be achieved in NCTAI-A-SNSPD at perpendicular incidence (which is improved due to the improved mesh-quality), whereas the highest $1.81 \cdot 10^{13}$ polarization contrast can be reached at the maximal 85° tilting in NCDDAI-SNSPD. The absorptance / polarization contrast / and the product of them was compared to the FOMs achieved in A-/ P-/ APC-SNSPDs. The comparative studies revealed that SNSPDs optimized to maximize the A*PC are the most suitable to read out the encoded quantum information.

5.5 SNSPDs to detect oxygen luminescence

Configuration of NCTAI-SNSPDs have been optimized to maximize absorptance at singlet oxygen luminescence [39]. Three integrated structures composed of a plasmonic wavelength-scaled grating were optimized for 1210 nm, 1270 nm and 1340 nm. Almost unity (~95%) absorptance was achieved at perpendicular incidence via plasmonic modes coupled in +/-1 order. The symmetry of the embedded vertical gold segments made it possible that the integrated devices are operable at perpendicular incidence, which is beneficial in medical applications. Implementation of these detectors into time correlated single-photon counting ensures the unambiguous detection of the singlet oxygen line with a specific decay characteristic.

5.6 SNSPDs to ensure polarization independent high absorptance

The wavelength-scaled NCTAI-SNSPD possesses the advantages of large period accompanied by small kinetic inductance resulting in short reset time, and the inserted trenches make possible to minimize the competitive gold absorptance. In this device both the metal-insulator-metal (MIM) modes squeezing and the forward/backward coupled SPPs contribute to absorptance maximization at perpendicular incidence. To achieve high absorptance independently of polarization two-dimensional crossed grating version of the NCTAI-SNDPD pattern was optimized. Based on primary studies the highest absorptance reached in NCTAI-X-SNSPD is 82-93% at perpendicular incidence and the corresponding polarization contrast is almost unity [3, 33, 35]. To minimize the polarization contrast in SNSPDs two mutually perpendicular and vertically shifted NbN patterns were integrated with two-dimensional gratings of gold segments terminated at the substrate plane and penetrating into it, by allowing asymmetry in vertical / all parameters.

The two-dimensional integrated patterns are nominated as nano-cavity-trench (NCTAI-X&XX) and nano-cavity-double-deflector (NCDDAI-X) / perfect nano-cavity-trench-deflector (NCTDAI-O) array integrated SNSPD, respectively [40, 41, 42]. The conditional optimization of the integrated SNSPD's dispersion characteristics made it possible to achieve near-unity absorptance maxima and polarization contrast close to perpendicular incidence in plasmonic pass-bands. The p and s-polarized absorptance in NbN stripes perpendicular to the **E**-field is mediated by neighboring gold antenna-segments through gaps parallel to **E**-field. This is promoted by MIM modes excitable via p-polarized illumination and the absorptance correlates with the ratio of the (extended) cavity-length - to - wavelength. The p-polarized absorptance is enhanced via (+/-1) order SPP coupling related pass-band, which crosses the flat band of resonant cavity modes. The overlap with a plasmonic pass-band of s-polarized illumination results in appearance of a depleted PC region. The achieved absorptance correlates, whereas the polarization contrast anticorrelates with the NbN/Au volume ratio.

The effects of vertical and tilted nano-cavity walls, as well as the substrate deepening gold antenna segments were compared. The X-SNSPDs exhibit the largest PC interval at perpendicular incidence, which is disadvantageous. Vertical-wall NCTAI-X-SNSPD exhibits the largest spectral bandwidth of high absorptance but the largest detuning as well. Tilted-wall NCTAI-XX-SNSPD ensures the smallest PC integrated in polar angle, but this is accompanied the largest absorptance interval at perpendicular incidence, the largest slope both of absorptance and PC and the highest PC integrated in azimuthal angle.

The NCDDAI-X-SNSPD makes it possible to achieve the largest (93%) absorptance with zero detuning and almost unity (1 ± 0.003) PC at perpendicular incidence, moreover the smallest PC interval and PC integrated in wavelength, widest spectral region of 1% PC deviation, as well as the smallest maximal PC value taken on throughout the inspected spectral band. Drawbacks are the tilting sensitivity and the small spectral bandwidth of high absorptance. These results indicate that this device has to be designed for a specific wavelength, and is tilting sensitive.

The enhanced degrees of freedom in NCTDAI-O-SNSPDs ensure zero detuning, the smallest absorptance interval at perpendicular incidence, the smallest slope both of absorptance and PC and the smallest PC integrated in azimuthal angle. Drawbacks are the smallest absorptance, largest PC integrated in polar angle, narrowest spectral region of high absorptance and 1% PC deviation, the largest PC interval and integrated value in wavelength. Interesting property is that both NbN patterns show PC sensitivity, but the added signals diminish it completely. Better performance is achievable by allowing difference in vertical parameters only, when the MIM cavity length is re-adjusted in order to achieve almost unity absorptance.

Conclusion: An optimization methodology was developed by using composite objective functions and appropriate constraints to achieve desired nanophotonical phenomena. It was shown that both the optimized systems and the nanophotonical phenomena resulting in maxima depend strongly on the objective function and on the criteria. Important result is uncovering that the dispersion relation is the two-dimensional function, which tailoring with appropriate criteria makes it possible to maximize the absorptance and to control the polarization selectivity in a specific wavelength interval. The same principle makes it possible to design structures capable of efficiently out-coupling light, which transfer encoded information. Integrated SNSPDs with extremely large absorptance and polarization contrast are designed for single-photon counting and to detect singlet oxygen lines with high efficiency.

6. Bioplatforms with enhanced sensitivity and FOM

6.1. *Bioplatforms based on periodic structures*

We have shown that the rotated grating coupling phenomenon, i.e. Bragg scattering, on a wavelength scaled grating in an optimal azimuthal orientation results in long-range surface plasmon polaritons' (LR-SPP) excitation [43]. The longitudinal **E**-field component of LR-SPP possesses two-fold anti-symmetry, i.e. it takes on zero value inside the metal, as a result it is possible to minimize losses, hence to maximize the propagation distance as well as the interaction cross-section with bio-molecules.

Considering that the biomolecules prefer to attach inside or along the side of the valleys, we have designed plasmonic biosensing platforms, which are based on the rotated grating coupling phenomenon [44]. To prepare the biochips, gratings with a periodicity commensurate with the SPP wavelength have been generated by two-beam interference on polymer film covered bimetal layers. Fluorescent lysozyme (LYZ) and gold-lysozyme bioconjugates (AuNP-LYZ) were seeded onto these chips. These bioplatforms were experimentally investigated in a modified Kretschmann arrangement. Based on angle interrogation attenuated total internal reflection (ATIR) measurements, the grating coupling results in double resonance minima on the reflectance curve.

By inspecting the optical response of such bioplatforms via FEM (COMSOL) the experimental chips' amplitude modulation parameters were determined by fitting the measured reflectance curves. Modeling of the fitted chips revealed that the measured polar angle shifts can be attributed to significantly larger amount of LYZ and smaller amount of AuNP-LYZ, respectively. This proves that the presence of Au-NP enhances the sensitivity due to the localized plasmon resonance, however the strength of this effect depends on the resonance extremum. Both the secondary and primary dips appearing at smaller and larger polar angles exhibit a larger shift in case of location at the side of the hills than at the bottom of valleys, except the primary dip for the Au-LYZ. This is governed by the extent of spatial overlap between the different amount of bio-moieties and the coupled modes of different symmetry. In case of secondary resonance minima originating from LR-SPP beside the smaller tilting and larger polar angle shift, the FWHM is also smaller, which allows better FOM. Monitoring of the secondary resonance is proposed to enhance sensitivity and to improve the FOM with respect to already existing SPP and LSPR based biodetection methods. According to these results all biochips based on structured plasmonic multilayers possess their own optimal configuration, which ensures the maximal resonance peak shift. For the optimal chip the azimuthal orientation is critical, since detuning causes disappearance one of coupled modes.

6.2 *Bioplatforms based on individual plasmonic nanoresonators and aggregates*

Our collaborators have prepared gold nanohybrid systems, which can be applied as bio-markers, and have shown that the fluorescence emission can be tuned by the chemical environment [45 - 47].

The fluorescence enhancement of dye molecules via plasmonic nanorods has been investigated theoretically via numerical computations and experimentally via STORM microscopy [48, 49]. A numerical optimization of coupled Cy5 dye molecule and gold nanorod systems was performed to determine configurations that are capable of resulting in an enhancement of excitation and emission, as well as both phenomena simultaneously. The coupled system geometry was determined via optimization by applying the fluorescence rate enhancement (i) at the excitation and (ii) emission, as well as (iii) at an intermediate wavelength as the objective function and setting criteria regarding the QE that have to be met.

The optimal location corresponds to a gradually increasing x coordinate and distance, whereas the optimal tilting gradually decreases. All geometrical parameters take on an intermediate value in the system optimized to enhance the excitation and emission simultaneously. It was concluded that the coupled system designed for the simultaneous improvement exhibited the highest total fluorescence enhancement proving that both the excitation and emission phenomena are maximized rather than compromised. However, the directional enhancement takes on an intermediate value due to that the out-coupling efficiency decreases, when the target wavelength increases.

Dependence of the fluorescence enhancement on the configuration parameters was inspected as well, by varying one and preserving two from the optimal location, distance and orientation parameter set. It was shown that the optimal location and orientation correspond to a compromised QE quantum efficiency and δR radiative rate enhancement, whereas the optimal distance corresponds to the threshold, above which the QE meets the criterion set at the emission. The system optimized for simultaneous improvement exhibited intermediate sensitivity to the dye molecule's parameters, except the location and orientation sensitivity at the excitation wavelength.

For proof-of concept experiments Au nanorods with a desired geometry were synthesized, Cy5 dye molecules were aligned at the optimal distance via hybridization of the Cy5-labelled oligonucleotide of controlled length, dSTORM was used to detect individual dye molecules that are stochastically on-state around the individual nanorods. The single on-state-molecule detection proved that an emission enhancement can be reached at the level, which is predicted theoretically at analogue distances, and larger fluorescence enhancement can be achieved, when a shorter DNA strand is used.

Sensing platforms were designed based on singlet NRs and on arrayed monometal and bimetal nanoparticles [48, 50]. The optimal parameters of the plasmonic NRs and of the primary artificial biolayers (DNA sequences and antigen compositions) were determined. These primary biolayers promote fluorescence enhancement as a result of capturing the complement targeted molecules (specific RNA and antibody) labelled by fluorescent dyes.

Aggregation based biosensors exhibit a spectral modification, when biomolecules' attachment triggers the aggregation process. Accordingly, aggregation of bare and thin biolayer (DNA strand) covered nanoparticles was inspected. Development of linear aggregates with different composition was primarily tested. By using monometal silver (gold) nanoparticles the absorption peak less (more) rapidly shifts to the red during the aggregation. We have inspected, how the spectral response is influenced by the thickness of the shell on a bimetal core-shell nanoparticle. Compared to the monometal Ag-NP, presence of (2) 4 nm silver shell results in (less) more rapid red shift. In contrast, 2 (4) nm gold shell results in significantly enhanced (reduced) shift compared to the monometal Au-NP. Due to the large modification of the resonance wavelengths, the aggregation process results in a visible color modification. These results proved that bimetal core-shell nanoparticles are more suitable to construct aggregation based bio-sensors compared to their monometal counterparts. The spectral response of nonlinear aggregates composed of triangular and hexagonal arrangement of the monometal and bimetal NPs was inspected as well. Nonlinear aggregates result in smaller extinction cross-sections with blue-shifted peaks. The "spectral position/number of particle" slope became smaller for both silver shell thicknesses. However, the 4 nm shell allowed a well detectable spectral shift. In case of 2 nm gold shell the slope was just slightly smaller in nonlinear aggregates. Accordingly, the 2 nm gold shell coated silver core can be proposed for aggregation sensor development, since the spectral modification is well detectable independently of the aggregate geometry.

Conclusion: It was shown that all biochips based on plasmonic multi-layers decorated by periodic structures possess their own optimal configuration, in which the plasmonic resonance related extremum shift is maximal. Long-range modes excitation promotes to achieve larger shift and enhanced sensitivity due to the large interaction cross-section as well as small resonance bandwidth and improved FOM. A complete kit was provided to design and prepare coupled nanosystems exhibiting plasmonically enhanced fluorescence for biodetection and STORM platform. Aggregation based biosensors are designed to detect the presence of specific virus RNS or the antibodies and applications were accomplished for proof-of-concept funds. The general method enables to design high sensitivity platforms to detect specific viruses and bacteria and to trace tumor markers.

7. Plasmonic spectral engineering via complementary complex structures

A novel integrated lithography methodology was patented in US, which makes it possible to fabricate complex structures, namely wavelength scaled arrays of nano-objects, on large scaled areas [51]. By illuminating colloid sphere monolayers using a single homogeneous perpendicularly and obliquely incident circularly polarized beam hexagonal patterns of nanorings and nanocrescents can be fabricated, whereas two interfering beams result in rectangular patterns of their miniarrays. We have demonstrated that six geometrical parameters (array period, nano-object distance, nanoring and nanocrescent diameter and thickness, nanocrescent opening angle and orientation) can be tuned independently. Concave and convex patterns can be directly fabricated via interferometric illumination of colloid sphere monolayers by applying circularly polarized light and by transferring these masks via a lift-off procedure, respectively. The possibility of plasmonic spectral engineering via complex structures consisting of spherical nano-objects was revealed by re-illuminating the complementary patterns by linearly and circularly polarized light in different configurations [52].

The spectral and near-field effects and the charge distribution of hexagonal patterns composed of uniform nanorings and horizontal nanocrescents were studied primarily to uncover the localized surface plasmon resonance (LRSP) related modes supported by the specific nano-objects. Comparative studies have been performed on complex concave and convex patterns composed of analogous nanorings and nanocrescents [53-57]. We have proven, when complementary patterns are illuminated by complementary beams the reflectance and transmittance are interchanged. Accordingly, the complementarity of the transmittance (reflectance) signal on convex patterns and the rectified reflectance (transmittance) signal on concave patterns has been shown. The extrema arising on the complementary signals were explained based on the complementary LSPR related charge distributions developing on the nano-objects and based on scattered photonic and coupled plasmonic modes on their periodic patterns in complementary configurations [53, 54].

Various rectangular patterns of miniarrays composed of a central nanoring and satellite nanocrescents have been designed. The azimuthal orientation dependent spectral effects of the pattern period, composing nano-object shape and size parameters have been uncovered. The results proved that precise plasmonic spectral engineering is realizable via these complex patterns. When the shape-parameter and radii of the nanocrescent are varied, the spectral effect is more pronounced in C-orientation, whereas the variation of the nanoring radii and period results in more well defined modification on the spectra in U-orientation [54]. Namely, by varying the period of the convex and concave patterns the FWHM varies in C-orientation, whereas in U-orientation local maxima appear due to the Rayleigh-Wood anomaly.

We have proven that it is more unambiguous to inspect the transmittance and reflectance on the convex and concave patterns, respectively [57]. Moreover, fingerprints of the plasmonic resonances are the most well defined in absorptance, accordingly we have investigated the absorptance and rectified absorptance spectra of various complementary convex and concave complex patterns [55-57]. Artificial rectangular patterns of different nano-objects were investigated to analyze the effect of nano-object interactions and the lattice type. The constituent singlet nanoring, singlet nanocrescent and a quadrumer of nanocrescents, as well as miniarrays of them arranged in 300 nm, 600 nm, 900 nm periodic rectangular patterns were the subjects of a comparative study [6, 54].

By comparing the spectra in $0^\circ - 90^\circ$ and $16^\circ - 106^\circ$ azimuthal orientations the LSPR, surface lattice resonance of photonic modes (SLR) and coupled surface plasmon polaritons (SPP) related origin of different extrema have been analyzed. By applying the Babinet principle regarding the complementary convex E_z and concave B_z modal profiles the complementary charge distributions accompanying the azimuthal orientation independent localized resonances on nanorings, and the orientation dependent U and C2 - C1 resonances on nanocrescents were determined. The Babinet principle predicts that the E_z distribution on the complementary convex pattern corresponds to the B_z distribution on the concave pattern and the accompanying E_z distribution reveals the charge distribution at the characteristic resonances on concave nano-objects. Surface lattice resonance and perfect grating coupling occurs, when the photonic mode is radiated along the \mathbf{k} vector ($\gamma=0^\circ$) and the plasmonic mode propagates along the \mathbf{k} vector ($\gamma=90^\circ$).

The hexagonal pattern of nanorings exhibits the “U-resonance” of the two coalesced crescent-shaped objects, which is independent of the \mathbf{E} -field oscillation direction due to the complex pattern symmetry properties. On the convex (concave) hexagonal array of horizontal nanocrescents the charge distribution and the corresponding near-field indicates U/C1 and C2 resonance in $\gamma=0^\circ/90^\circ$ ($\gamma=90^\circ/0^\circ$) azimuthal orientation. On the hexagonal pattern of singlet horizontal nanocrescents the convex absorptance and transmittance in $\gamma=0^\circ/90^\circ$ azimuthal orientation corresponds to the concave rectified absorptance and reflectance in $\gamma=90^\circ/0^\circ$ azimuthal orientation. In comparison, on the convex (concave) rectangular pattern of slightly rotated nanocrescents U/C1 and C2 resonances appear in $\gamma=16^\circ/106^\circ$ ($\gamma=106^\circ/16^\circ$) azimuthal orientation [55-57]. Caused by the orientation of nanocrescents inside the rectangular pattern of mini-arrays, the convex absorptance and transmittance in $16^\circ/106^\circ$ azimuthal orientation corresponds to the concave rectified absorptance and reflectance in $\gamma=106^\circ/16^\circ$ [55-57].

On convex patterns the nanophotonical phenomena are governed by the azimuthal orientation independent localized resonance on the nanoring and by the C2-C1 and U resonances on the nanocrescents excitable in case of \mathbf{E} -field direction perpendicular and parallel to their symmetry axes, respectively. Cross-coupled modes appear in case of non-perfect alignment of the \mathbf{E} -field with respect to the axis. As a result, cross-coupled quadrupolar U and dipolar C1 type modes related extrema appear in 106° and 16° azimuthal orientation of singlet horizontal nanocrescents, respectively. In contrast, on the quadrumer of slightly rotated nanocrescents enhanced preference to the quadrupolar r-U mode related distribution is noticeable in 90° azimuthal orientation, whereas along-tips-oriented dipolar C1 mode is cross-coupled in 0° azimuthal orientation. The interaction between LSPRs on individual nano-objects is weak, the miniarray absorptance equals to the sum of composing central nanoring and satellite nanocrescent quadrumer absorptance. The scattered photonic modes have a perturbative role at the Rayleigh anomaly on larger wavelength-scaled (600 nm and 900 nm) periodic rectangular patterns of miniarrays in 0° and 16° azimuthal orientation [6, 54].

The spectra are more complex on the concave patterns. On the rectangular array of the nanoring one single maximum appears in and close to C orientation (16° and 0°) independently of the period. The shoulder in 16° is due to the cross-coupling facilitated by the existence of E-field component along the \mathbf{k} vector. This results in appearance of a rotating reversed dipoles, instead of a horizontal steady-state r-U mode. In and close to U orientation (106° and 90°) a local-global maximum pair appears, which reveals interacting LSPR and a grating coupled SPP modes. This is due to that the E-field oscillation is almost and perfectly parallel to the \mathbf{k} vector. On the rectangular array of singlet nanocrescent a shoulder is followed by a global maximum originating from C1 mode in and close to C orientation. Enhanced preference to the quadrupolar mode reveals the cross-coupled c-U resonance at the shoulder close to C-orientation (16°). In and close to U orientation a local-global maximum pair appears due to interacting LSPR and SPP modes, which is followed by a shoulder originating from cross-coupled c-C1 resonance close to U-orientation (106°). Interestingly, the interacting C2 and C1 LSPR modes in the C orientation and the coupled U LSPR and SPP modes in the U orientation manifest themselves in similar split spectra. On the rectangular array of the satellite nanocrescents a shoulder and a global maximum appears in C-orientation, which is attributed to the interacting C2 and C1 modes and to the C1 mode. At the shoulder preference to the c-U mode related quadrupolar distribution is unambiguous close to C orientation. In U orientation of the quadrumer one single peak appears, in contrast to the singlet nanoring and horizontal nanocrescent. Disappearance of the grating coupling is caused by symmetry reasons on the quadrumer of slightly rotated nanocrescents. Close to U-orientation (90°) a shoulder is observable due to the cross-coupled C1 mode. In addition to this, further modulations appear on the wavelength scaled (600 nm) periodic pattern in the intervals of the Wood-Rayleigh anomalies. In case of miniarrays the local-global maxima are additive in C orientation in the sense that the former corresponds to the nanoring, whereas the latter originates from the quadrumer. In U orientation the distributions on the nanocrescents are perfectly undistinguishable. By comparing the sum of the composing nanoring and nanocrescent pattern's spectral responses with the spectra of their miniarrays it was concluded that the concave nano-objects' interaction is also weak.

The mapping of the dispersion characteristics of these complex patterns was also realized. On all convex patterns in 0° and 90° azimuthal orientation flat bands originating from the PPR, U, C2 and C1 LSPRs are identifiable. In addition to this on the dispersion map of the 600 nm periodic pattern the Rayleigh anomaly related band also appears due to the more/less efficient scattering of photonic modes in $(+/-1, 0)$ order along the \mathbf{k} vector in $0^\circ/16^\circ$ azimuthal orientation. On all concave systems the dispersion is similar in 0° azimuthal orientation to the characteristics of the convex counterpart patterns. In 90° azimuthal orientation the coupling (in $(-1,0)$, $(-2,0)$ and $(-3,0)$ order) via the \mathbf{k} grating vector of the (300 nm, 600 nm, 900 nm) pattern results in SPP band, which shows anticrossing with the LSPR band of the nano-objects. The differences between complementary convex and concave dispersion characteristics were attributed to the anticrossing of U resonances with SPP modes coupled on concave patterns. The relatively stronger (weaker) grating-coupling of the $(+/-1, 0)$ order SPP1 in 90° (106°) and 16° azimuthal orientation results in Wood-Rayleigh anomaly related bands on the 600 nm and 900 nm patterns. The different strength of Rayleigh and Wood anomaly related perturbations is due to different efficiencies of SLR and SPP coupling.

The fluorescence enhancing capability of these complex structures was demonstrated, by determining radiative rate enhancements of dipolar emitters based on the complementary transmission (reflection) and reflection (transmission) signals and on the complementary absorptance and rectified absorptance of the concave and convex patterns [53, 55, 56]. The effect of the location and orientation of dipolar emitters was inspected as well [53].

On complex patterns considerable fluorescence enhancement of dipolar emitters was achieved at spectral locations promoting the C and U resonances on the constituent nano-objects. Dipolar emitters can be significantly enhanced via these convex and concave structures at slightly shifted spectral locations with respect to the absorbance maxima arising in case of plane wave illumination.

The asymmetrical spectral lines were fitted by Fano models taking into account interacting localized and scattered and propagating modes. The potential application of the complex structures in metamaterial design and in enhancement and out-coupling of the stimulated emission was also demonstrated [57].

Conclusion: A novel integrated lithography methodology was developed and patented, that makes it possible to vary the geometrical properties of complex structures with high degrees of freedom, which is crucial in plasmonic spectral engineering. It was proven that the complementary concave and convex patterns exhibit complement optical responses as well as near-field and charge distributions at specific extrema according to the Babinet principle. A novel FEM and FDTD based method was developed, which can be used to optimize complex structures dispersion characteristics. Complex patterns can be designed to promote sensing of molecular emitters and lasing phenomena via embedded dye molecules.

8. Few-cycle plasmonic field generation

We have proven, that few-cycle localized plasmon oscillations can be induced in optical near-fields of various plasmonic NRs [58]. Our comparative study shows that the generation of few-cycle plasmonic transients is indeed possible, however this requires optimization of the NRs. During the optimization realized to maximize the near-field enhancement (NFE) while preserving the number of cycles (NOC) the scattering cross-section (SCS) was selected as the objective function, since this is accompanied by a large bandwidth in case of core-shell nanoparticles. The large SCS enables small absorption cross-section (ACS), as a consequence, a trade-off has to be found between the contradictory requirements of large near-field enhancement and short transients in case of optimization with the FOM = NFE/NOC .

A comparative study of gold and silver nanorod and core-shell monomers and dimers and bow-tie antennas illuminated by few-cycle pulses was performed. The number of cycles of the excited plasmonic field, the achieved near-field enhancement as well as their ratio (NFE/NOC) was analyzed, and the pulse length dependence of these quantities was also investigated.

Change from monomers to dimers is advantageous considering NOC in case of nanorods. The NFE achieved via nanorod and core-shell dimers is also considerably higher than that achieved via the corresponding monomers. The NFE/NOC ratios are significantly higher for dimers proving their advantage compared to the monomers. Comparing all dimers, the Ag bow-tie has the highest NFE. The NOC is the lowest in case of Ag core-shell dimers. Gold core-shell dimers outperform all other dimers in their NFE/NOC ratio due to the relatively low NOC and strong NFE.

Comparing all inspected NRs, all types have some advantageous characteristic property. Throughout the inspected pulse-length interval silica-gold and silica-silver core-shell monomers have the potential to preserve the NOC of the incoming pulse, silver bow-ties result in the highest NFE, whereas gold core-shell dimers have the highest NFE/NOC . Based on the analysis, silver bow-ties, gold core-shell dimers and silver nanorod dimers proved to be the most suitable for few-cycle near-field amplification.

Conclusion: The conditional optimization method that we applied in this study is an important forward step towards adaptive near-field control. These results have the potential to enhance applications in strong-field physics, such as the harmonic conversion of light in the vicinity of nanostructured surfaces.

9. Fusion targets doped by plasmonic nanoresonators

We have shown that the fusion targets absorptivity can be improved via plasmonic NRs [59-62]. The generalized aspect ratio of silica-gold core-shell nanoparticles was tuned to ensure absorption cross-section peaks at the ~ 800 nm central wavelength of the 1 ps laser pulse. When such particles are arranged in a sparse lattice, their monolayer exhibits an absorptance peak at the central wavelength, that can be further increased via double sided illumination. By implanting 10 layers of resonant core-shell nanoparticles into (1 ps) pulse length-scaled ($276 \mu\text{m}$) DT target in a sparse lattice, 33% and 66% absorptance corresponding to 114 cm^{-1} and 216 cm^{-1} absorption coefficients can be achieved via single and double sided illumination, respectively.

The uniform distribution of implemented plasmonic NRs is not proper to achieve uniform heating of the targets, since the ohmic loss (absorption) exhibits exponential decay according to the Lambert-Beer law. Different distributions, which result in more densely packed layers at the center than at the border of the target, are more appropriate. In Gaussian distributions via phase tuning of the counter propagating pulses (adjusting the position of the layers) 77% and 76% (98% and 99%) absorptance is achieved, which corresponds to 217 cm^{-1} and 216 cm^{-1} (216 cm^{-1} and 291 cm^{-1}) absorption coefficients [59].

We have shown that significant charge separation accompanies the plasmonic resonance, which leads to 10^8 C/m^3 average charge density on the nanorod in linear approximation at the peak of a 26 fs pulse already at $1.4 \cdot 10^{12} \text{ W/cm}^2$ that was reported as a threshold resulting in permanent damage of similar gold antennae [60].

The distribution of plasmonic NRs of core-shell composition and nanorod shape along a pulse-length scaled target was optimized to maximize the absorptance with the criterion of minimal absorption difference in between neighbouring layers. Successive approximation of layer distributions made it possible to ensure almost uniform deposited energy distribution up until the maximal overlap of two counter-propagating pulses. Although, the standard deviation of absorptance is larger in the uniform and in different predefined Gaussian distributions for core-shell NRs, it becomes smaller in the primarily optimized distributions (except for energy and power loss densities). In case of the adjusted distributions the standard deviation of the absorptance and energy is smaller for core-shell NRs, but becomes larger for energy density as well as for power loss and power loss density. Based on the larger absorptance and smaller uncertainty in absorptance and energy distribution, the core-shell NRs override the nanorods. However, optimization of both NR distributions has potential applications, where efficient and uniform energy deposition is crucial, including phase transitions and even fusion [61]. Further studies are in progress to determine the charge separation by taking the nonlinear phenomena arising at high intensities into account [62].

Conclusion: The energy deposition can be uniformized in different types of substrates, e.g. in fusion targets by using plasmonic structures or complex metamaterials.

Outlook: Demonstration of similar phenomena on predesigned metamaterial architectures is in progress.

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