Molding Photon Emission with Hybrid Plasmon-Emitter Coupled Metasurfaces

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Directional emission of photons with designed polarizations and orbital angular momenta is crucial for exploiting the full potential of quantum emitters (QEs) within quantum information technologies. Capitalizing on the concept of hybrid plasmon-QE coupled metasurfaces, a holography-based design approach is developed allowing one to construct surface nanostructures for outcoupling QE-excitation regularly diverging surface plasmon polaritons (SPPs) into well-collimated beams of photons with desirable polarization characteristics propagating along given directions. Using the well-established simulation framework, the efficiency and versatility of the developed approach are demonstrated by analyzing different hybrid SPP-QE coupled metasurfaces designed for the generation of linearly, radially, and circularly polarized photons propagating in various off-axis directions. This work enables the design of single-photon sources with radiation channels that have distinct directional and polarization characteristics, extending thereby possibilities for designing complex photonic systems for quantum information processing. Moreover, it is believed that the developed scattering holography approach is generally useful when employing surface electromagnetic excitations, including SPP modes, as reference waves for signal wave reconstruction.

1. Introduction

Many efforts have been dedicated over recent years to improving the performance of quantum emitters (QEs) that constitute one of the key enabling technologies for quantum communication and information systems.[1–5] Various micro/nanostructures, including nanoantennas and nanocavities, have been designed in recent years to enhance the photon emission from QEs by making use of the Purcell effect[6–11] via engineering their immediate dielectric environment.[12–22] Although very large (even thousands fold) enhancement of brightness has been reported with different nanostructures, the polarization, direction, and wave front of the photon emission are still rarely addressed.[16–22] Very recently, the design route for optical metasurfaces has been introduced to efficiently mold the single-photon emission from QEs with hybrid plasmon-QE coupled configurations, consisting of dielectric nanoridges surrounding QEs on metal substrates, that appear most attractive from the viewpoint of combining high quantum yield and collection efficiency.[14–20] A common feature of these configurations (meta-atoms) is the “efficient” and “nonradiative” coupling of a QE to surface plasmon-polariton (SPP) modes that are subsequently outcoupled by a metasurfaces structure into free propagating waves.[23] Different designs of the outcoupling metasurface resulted in the generation of circularly (with different orbital angular momenta)[23,24] or radially[27] polarized single photons. Using displaced circular ridges, directional off-normal photon streaming for different, up to 20°, angles has also been experimentally realized.[28] At the same time, serious deficiencies in generated wave fronts were observed both in simulations and experiments, especially when producing off-normal photon streaming,[28] calling for the development of the general design approach that would ensure the realization of well-collimated beams of photons with desirable polarizations (e.g., linear, circular, and radial) propagating in given (off-axis) directions.

Holography is an approach enabling the recording and reconstruction of arbitrary wave fronts with the help of various types (amplitude, phase, transmission, reflection, etc.) of holograms.[29–34] The problem of generating complicated wave fronts with arbitrarily designed polarizations distributions is of great importance in photonics, in general, and in the field of optical metasurfaces, in particular.[35–40] Very recently, efficient approaches using local control of the phase and polarization of transmitted/reflected light have been developed for designing optical metasurfaces that perform rather complicated transformations of wave fronts and polarizations in transmission[41] and reflection[42] configurations. Although the main...
principles of holography and its many modalities are well known and established for free propagating (object and reference) optical beams, the problem of reconstructing beams with arbitrary wave fronts and polarizations when using (QE-excited) circularly diverging SPP fields have not been considered. In this work, we introduce a novel modification, vectorial scattering (computer-generated) holography, for the purpose of designing hybrid SPP-QE coupled metasurfaces suitable for the generation of well-collimated beams of single photons with desirable polarization characteristics propagating along given directions. In our modified method, spatial variations of QE-excited SPP fields used as a reference wave due to their divergence, absorption, and scattering are, for the first time to our knowledge, explicitly taken into account. With this holography-based approach at hand, we design metasurfaces for the generation of linearly, radially, or circularly polarized photons propagating in various off-axis directions and demonstrate the efficiency and versatility of our approach using simulations conducted within the well-established numerical framework.[23,24,27,28]

2. Results and Discussion

The basic configuration under consideration, representing the hybrid SPP-QE coupled metasurface, consists of dielectric nanoridges (forming a metasurface) formed atop a metal substrate covered with a dielectric spacer (Figure 1). We have previously demonstrated that such a meta-atoms can be designed to very efficiently (up to 90%) convert QE-excitations into free propagating and objective-collected photons.[27] In the simulations presented here, a QE considered is a nitrogen-vacancy (NV) center in nano-diamond (ND), whose emission peak wavelength is near 670 nm (when illuminated by a 532 nm laser beam) and whose radiative transition dipole is oriented perpendicular to the sample surface.[23,24,27,43] We would like to note that the proposed design route can be applied to and remains efficient for other QEs with different wavelengths as long as the QE dipole is perpendicular to the sample surface. The QE-excited SPPs propagate along silica (15 nm thickness) covered silver film, circularly diverging from the QE in a cylindrically symmetric fashion. Nanoridges made of hydrogen silsesquioxane (HSQ, 150 nm thickness) with a refractive index of 1.42[28,44] is positioned atop the silica spacer layer (protecting a silver film from oxidation). The pattern of nanoridges encircling the QE is designed in accordance with the holography-based approach developed in this work that enables the SPP outcoupling into a single-photon stream carrying a specific polarization, such as linear (LP), radial (RP), or right circular (RCP) polarization, and propagating along a given direction.

To design the nanoridge pattern of a hybrid SPP-QE coupled metasurface, we develop a modified vectorial scattering holography approach (Supporting Information Sections 1 and 2) with a cylindrically diverging SPP wave, $E_{\text{sp}}$, used to reconstruct a signal wave, $E_s$. In brief, a hologram in our approach is formed by “isotropic non-interacting dipolar nano-scatterers” (such as spherical nanoparticles), whose volumes and thereby scalar polarizabilities are proportional to the intensity of an interference pattern generated by a reference and signal waves. Discretizing the intensity pattern and fusing neighbor nanoparticles results in a pattern of constant-height nanoridges (of varying width) that can conveniently be fabricated with electron-beam lithography (EBL). Importantly and “contrary” to the conventional holography approaches,[29–34] we employ an “artificial” reference wave for calculating the interference pattern and designing thereby the nanoridge pattern. This artificial reference wave is taken in the form of a “radially increasing” SPP wave that can be expressed in the hologram plane as follows: $E_{\text{sp}} = E_{\text{sp}}^0 \sqrt{r_N} \exp(i k_{\text{sp}} r_N) \exp(-i k_{\text{sp}} r_N)$, where $E_{\text{sp}}^0$ is the SPP vectorial amplitude, $r_N$ is the length of radius vector, $k_{\text{sp}} = (2\pi/\lambda) N_{\text{sp}}$, with $N_{\text{sp}}$ being the SPP effective index at the operation wavelength $\lambda$, and $\alpha = 1/2L_{\text{sp}}$ with $L_{\text{sp}}$ being the SPP propagation length. The nanoridge pattern is then calculated by considering interference between the above artificial SPP wave and the signal wave (taken also in the hologram plane): $E_{s} = E_s^0 \exp(-i k_s r_N)$, where $E_s^0$ and $k_s$ are the amplitude and wavevector projection (on the hologram plane) of the signal wave, respectively (both quantities are assumed to be varying in the hologram plane). When the resulting nanoridge pattern is illuminated by the actual (QE-excited) SPP wave: $E_{\text{sp}} = E_{\text{sp}}^0 (1/\sqrt{r_N}) \exp(-i k_{\text{sp}} r_N) \exp(-i k_{\text{sp}} r_N)$, the scattered field in the $(x, y)$-plane near the hologram contains the corresponding (to the reconstructed signal wave) term in the following form (Supporting Information Section 2):

$$E_s \sim \ldots + \left(E_{\text{sp}}^0, E_s^0\right) E_{\text{sp}}^0 \exp(-i k_s^0 r_N).$$

Although the problem of reconstructing the signal wave polarization remains when one uses “isotropic” dipolar nano-scatterers (Supporting Information Section I), the spatial phase and amplitude distribution of the signal wave (encoded in the signal amplitude and wave vector projection spatial distributions) is expected to be reconstructed, ensuring the propagation of a specified beam in a specified direction. Note that the SPP depletion during the reconstruction, occurring due to the SPP scattering out of the surface plane by surface nanoparticles, can, in principle, also be accounted for in a similar manner by simply adding the SPP attenuation by scattering to the SPP attenuation by absorption: $\alpha^0 = \alpha + \alpha_{\text{scat}}$. In the
following we illustrate this approach for designing nanoridge patterns of hybrid SPP-QE coupled metasurfaces that generate differently polarized and propagating single-photon beams. In view of reconstruction problems associated with the polarization cross-talk (Supporting Information Section 1), we limit calculating the interference (and thereby designing the nanoridge pattern) to considering only in-plane \((x, y)\)-components of reference and signal waves. For reconstruction, we employ 3D finite difference time domain (FDTD) simulations using the well-established (theoretically and experimentally) numerical framework.[23,24,27,28]

First, we design metasurfaces for generating linearly polarized, along the \(x\)-axis (LPX), Gaussian beams of different radii \(w_0\) propagating normal to the metasurface plane \((k_{\gamma\gamma} = 0)\), so that the only non-zero signal field component is the \(x\)-component: \(E_{x} \sim \exp(-r_x^2/w_0^2)\). The considered beam radii are conveniently related to the SPP propagation length \((8 \, \mu\text{m})\), for example \(w_0 = L_{\text{SPP}}/\sqrt{2}\) (Figure 2), that was calculated using the average filling ratio \((0.39)\) of HSQ found from the designed nanoridge pattern. As expected, the nanoridges vanish near the \(y\)-axis (Figure 2a), where the polarization of the reference and SPP wave are orthogonal to each other, resulting in the reconstructed LPX beams becoming elliptical (less confined along the \(y\)-axis) in the cross-section (Figure 2b,c). Importantly, the power carried by the LPX beams is significantly larger than that of LPY (four-lobed) beams, with the total field along the normal to the hologram center being purely \(x\)-polarized. Finally, the beam divergence angle \(\vartheta\) is found decreasing when the Gaussian signal beam radius increases, closely following the theoretically expected dependence: \(\vartheta = \lambda/\pi w_0\) (Figure 2d), because wider signal beams result in larger intensity interference patterns and thereby hologram areas (Supporting Information Figure S5).

To realize the off-axis LPX photon emission, we consider the \(x\)-polarized signal field in the following form: 
\[
E_x = \exp(-r_x^2/w_0^2)\exp(ik_0 \sin \vartheta x) \exp(-r_y^2/w_0^2)\exp(ik_0 \sin \vartheta y) \exp(\rho^2/\sqrt{2}) \exp(ik_0 \sin \vartheta z),
\]

where \(k_0 = 2\pi/\lambda\), \(\vartheta\) is the emission angle in the \((x, z)\)-plane with respect to the surface normal and \(r^2 = (x \cos \vartheta)^2 + y^2\). Calculating the corresponding interference patterns, one notices that these and, consequently, nanoridge patterns become progressively more asymmetric for larger emission angles (Figure S6, Supporting Information).

At the same time, it is seen that the reconstruction remains faithful, reproducing well-confined LPX beams propagating in the designed direction even for rather large, up to \(\theta = 60^\circ\), angles (Figure 3), which is very important from the viewpoint of potential applications in quantum technologies.

Generation of radially polarized (RP) photon beams propagating normal to the surface is relatively straightforward with a simple bullseye nanoridge pattern[27,45,46] because of the same symmetry for in-plane \((x, y)\)-components of both QE-excited radially diverging SPP waves and RP beams. The situation becomes more complicated for off-axis RP emission: the Doppler-like design of displaced circular ridges does not work well, failing to reconstruct the doughnut shape even for small \((\theta < 10^\circ)\) angles.\[28\] Our unified design strategy suggests using the in-plane RP field distribution (signal wave) for calculating the interference pattern in the following form: 
\[
E_x = (E_{x+}E_{x-}) - (r_x/a_0)\exp(-r_x^2/w_0^2)\exp(ik_0 \sin \vartheta x)\cdot(\cos \theta \cos \varphi \sin \varphi - \sin \theta \sin \varphi),
\]

where \(\varphi\) is the polar angle: \(\tan \varphi = y/x\). It is seen that for relatively large angles, such as \(\theta = 15^\circ\), the doughnut intensity distribution is well reconstructed when using the designed patterns of hybrid SPP-QE coupled metasurfaces that generate...
nanoridge pattern (Figure 4a), although some distortion is visible (Figure 4b). The latter is believed to be due to relatively stronger forward SPP scattering (with respect to the backward one) by nanoridge arrays, a circumstance that results in changing the field balance along the RP beam circumference. Importantly, the RP field decomposition into circularly polarized fields reveals opposite orbital angular momenta or topological charges ($\pm 1$) in their phases (Figure 4c,d) as expected. For larger angles, the doughnut shape is progressively more distorted, but the emission angles remain very close to the designed values (Figure S7a, Supporting Information).

Generation of purely circularly right or left polarized (RCP or LCP) photon beams is impossible with simple spiral nanoridge patterns that would always generate RCP and LCP beams carrying orbital angular momenta (topological charges) different by 2. We can still use our approach targeting the generation of, for example, an RCP beam and thus using the following signal wave for calculating the interference pattern:

$$E_s = E_{\text{LP}} \sim \exp(ik_0 \sin \theta x) \cdot (\cos \theta : i)$$

It is seen that for relatively large angles, such as $\theta = 15^\circ$, the well-defined RCP beam and the doughnut-shaped (vortex) LCP beam, carrying the orbital angular momentum with the topological charge of 2, are both well reconstructed (Figure 5), i.e., showing the expected intensity distribution. Small distortion in the LCP intensity distribution is seen (Figure 5d) similarly to that observed for the RP case above and is also related to the asymmetry in the off-axis SPP scattering by nanoridge arrays. Note, that the resulting emission pattern (Figure 5b) consists of spatially separated RCP (Figure 5c) and LCP (Figure 5d) emission channels, representing thereby spatially separated entangled emission channels, a feature that is very important from the viewpoint of potential applications in quantum technologies. For larger angles, the doughnut LCP beam shape is progressively more distorted, but the RCP beam remains well-shaped and confined, with the emission angles being very close to the designed values (Figure S7b, Supporting Information).

### 3. Conclusion

In summary, we have developed a holography-based design approach for constructing nanoridge patterns of hybrid SPP-QE coupled metasurfaces enabling the generation of well-collimated single-photon beams with desirable wave...
front and polarization characteristics propagating along given directions. Conducting 3D-FDTD simulations using the well-established (theoretically and experimentally) numerical framework,[23,24,27,28] we have demonstrated the efficiency and versatility of the developed approach by analyzing different hybrid SPP-QE coupled metasurfaces designed for the generation of LP, RP, and CP polarized photon beams propagating in various off-axis directions. We would like to emphasize that the QE and metasurface material compositions considered here are not particularly important, representing only one possible realization, and other QE types and metasurface constituents can be analyzed within the same approach. We would also like to note that the presented approach can further be developed and generalized by using properly designed “anisotropic” (instead of isotropic) nano-scatterers (Supporting Information, Section 1). This generalization should enable increasing the generation efficiency up to the level of ≈90%[27] that is expected for the RP generation (when there is a perfect match between in-plane field components of the reference SPP and signal RP waves).

Overall, our work opens fascinating perspectives for designing single-photon sources with radiation channels that exhibit diverse (including vectorial with spin and orbital angular momenta) wave fronts and polarization characteristics, extending thereby possibilities for designing complex photonic systems for quantum information processing. Furthermore, the developed scattering holography approach allows one to achieve the faithful reconstruction of complex signal wave fronts in configurations that involve surface electromagnetic excitations, including SPP modes, as reference waves by accounting for “inevitable” (due to divergence, absorption, and scattering losses) spatial variations of reference wave magnitude (Supporting Information, Section 2).

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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