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Making Quantum Emitters Emit Vector Vortex Beams

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Abstract – How far can we go in shaping the luminescence of quantum emitters? It is well known that by using cavities or nano-antennas, one can beam their luminescence into well-controlled directions of space. Here, we push this control to a level of complexity usually achieved with laser radiation: we make an assembly of colloidal quantum dots spontaneously emit vector vortex beams, which are swirls of light combining a non-zero angular momentum and an inhomogeneous field structure. The key ingredient to obtain this result is to increase the spatial coherence of the emitted photons. To fulfill this condition, we design and use plasmonic holograms that also impart a non-zero orbital angular momentum and a radial polarization state to the beam.

I. Introduction

Scalar vortex beams are characterized by a chiral helical wavefront associated with a non-zero orbital momentum [1]. The phase varies spatially with the azimuthal angle $e^{im\theta}$, where m is the topological charge of the vortex, resulting in a phase singularity at the center of the beam. This phase singularity implies a null point of intensity, giving to the vortex an annular intensity profile. Vector vortex beams are even more complex because they also possess a polarization singularity in addition to the phase singularity. Both scalar and vector versions of optical vortices are associated with a trove of discoveries in fluorescence imaging, optical trapping, high-speed telecommunications and quantum technologies [2]-[3].

A necessary condition to construct optical vortices is to use light that possesses spatial coherence, so as to create the specific set of interferences that defines them. It is therefore

perhaps not surprising, then, that the realizations found in the literature are focused on lasing light. For example, phase plates, diffractive holograms, SLM, or optical metasurfaces can produce a phase singularity, while birefringent crystals or polarizing prisms can impart a polarization singularity to the laser beam.

Contrarily to lasers, the spontaneous emission of quantum emitters has little spatial coherence. However, there is a well-known remedy to this problem that consists in coupling them with propagating surface plasmons. Surface plasmons have coherence lengths that are typically limited by their propagation distance, which can be tens of microns in the near infrared. These long coherence lengths explain why one can make surface plasmons emit light into very narrow beams using gratings carved at the metal surface [4]. Here, we revisit this approach to shape the spontaneous infrared emission of PbS colloidal quantum dots (cQDs) into vector vortex beams.

II. Results and Discussion

As illustrated on the inset of Fig. 1(a), our sample consists of a gold hologram on a gold substrate, with a thin but compact layer of cQDs coated on top. The samples are fabricated in a cleanroom with electron beam lithography followed by metal evaporation, the thickness of the gold hologram being equal to 25nm. First, it is crucial to notice that we do not observe a quenching of the photoluminescence, usually observed when a single quantum emitter is put in direct contact with a metallic surface. This unintuitive behavior has been recently explained by the realization that an ensemble of cQDs is very different from the single emitter case and that they respond to a new regime of light-matter interactions described by a local Kirchhoff law [5]. Then, as explained in the introduction, in order to reshape the spontaneous of cQDs, we need to maximize their spatial coherence. Here the quantum emitters are coupled to spatially coherent surface plasmon modes. More especially, after pumping the cQDs with a HeNe laser excitation at the center of the spiral, they populate surface plasmon modes that propagate radially at the surface of the sample and interact with the hologram. To make the cQDs outcouple as a vector vortex beam, the hologram must possess a specific design which

is obtained by computing the interference pattern between the radial plasmonic wave and the optical vortex:

$$H(r, \theta) = \exp[i \operatorname{Re}(2\pi r n_{\text{eff}} / \lambda)] + \exp(im\theta)^2, \quad (1)$$

where n_{eff} is the effective refractive index seen by the plasmons. The resulting pattern is a spiral with a number of arms equal to the topological charge of the vortex. A SEM picture of a symmetric 1-arm spiral is displayed on Fig. 1(a). To characterize experimentally the sample, we use a 4f optical setup to measure the angular distribution of the photoluminescence beam in the Fourier plane. An objective collects the photoluminescence signal, a dichroic mirror isolates it from the laser pump and then a pair of lenses images the Fourier plane of the objective on an IR InGaAs camera. A narrow spectral filter is inserted in front of the IR camera to select one single wavelength. As illustrated in the Fig. 1(b), the photoluminescent beam appears with an annular distribution. Due to the dispersive nature of our structure, the diameter of the ring changes with the wavelength filter, as discussed in Ref. [6] for vector vortex beams with higher topological charges (NB: the results presented in this paper are fully original). We have also established an analytical model that calculates the intensity and phase distribution of the far-field diffraction pattern in the scalar Fraunhofer approximation. As depicted in Fig. 1(c-d), we notice a very good agreement between the theoretical and experimental intensity distributions, while the theoretical phase varies from 0 to 2π and exhibits the expected characteristics of a 1st order vortex.

Fig. 1. a) Inset: Schematic cross-section of the sample. SEM image of a 1-branch spiral hologram, scale bar: $1\mu\text{m}$. The radial pitch of the spiral is equal to $\lambda/n_{\text{eff}} = 1095\text{nm}$. b) Experimental image (in false colors) of the photoluminescent beam intensity distribution in the Fourier plane. The spectral filter in front of the IR camera is centered on $\lambda=1050\text{nm}$. The dashed circle corresponds to the numerical aperture of the objective $\text{NA}=0.65$. c-d) Theoretical intensity and phase distributions of the photoluminescent beam in the Fourier plane.

The ring intensity profile of the photoluminescence beam is not a sufficient experimental proof to prove the generation of a vortex, an additional demonstration of the phase singularity is necessary. Thus we have considered interferometric structures, which consist of the superposition of two different spirals, generating two interfering vortices with different topological charges. In Fig. 2, two vortices with opposite topological charges equal to $m_1=+4$ and $m_2=-4$ interfere and give rise to a star-shaped interference pattern. This interference pattern can be generalized for any combination of vortices with a number of arms equal to $|m_1 - m_2|$, and are very well reproduced by our theoretical model. Additional interferometric patterns obtained with other values of m_1 and m_2 can be found in [6].

Fig. 2. a) SEM image of the interferometric spiral hologram, scale bar: $1\mu\text{m}$. The geometry consists of two 4-arm spirals with the same radial pitch $1.095\text{ }\mu\text{m}$ but with opposite winding. b) Interference pattern of the photoluminescence beam visualized experimentally in the Fourier plane and c) comparison with the theoretical predictions.

Finally, we have performed polarization measurements to probe the field structure of the photoluminescence beam. As depicted on Fig. 3, when a polarizer is added in front of the camera to select one specific polarization state, the intensity pattern is reduced to two lobes, and these two lobes rotate with the angle of the polarizer. Hence, the photoluminescent vortex exhibits a radial polarization distribution associated with a polarization singularity in its center. In other words, the beam displays all the features of a vector vortex beam. The polarization properties are explained by the field structure of the radial surface plasmons that are involved in the process [7]. This result can be generalized to arbitrary propagation directions, as shown for example on Fig. 3(b) for a sample that produces a vector vortex beam at an oblique incidence of 10 degrees with respect to the normal.

Fig. 3. a) Experimental Fourier plane images measured for the same structure and with the same spectral filter as the one used in Fig. 1 but with a linear polarizer inserted in front of the IR camera. The orientation of the polarizer is indicated with double arrows. b) Same observations for an asymmetric spiral generating a vortex with a 10° angle with respect to the normal.

III. Conclusion

In conclusion, we have experimentally demonstrated the generation of vector vortex beams from the incoherent photoluminescence of cQDs—a feat usually achieved with lasers. Following the same approach, other complex forms of light could be generated including vortex knots or links [8]. Moreover, the optical excitation of the cQDs could be replaced by electrical pumping, opening up the way to LED sources generating optical vortices. Such vortex emitting diodes would offer tremendous advantages for integrated systems.

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