Spin-Polarized Photon Emission by Resonant Multipolar Nanoantennas

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Supporting Information

**ABSTRACT:** We demonstrate nanoscale spin control of photons emitted by an atomic system coupled to a compact plasmonic nanoantenna supporting phase-locked interference of different multipolar moments within a single resonance. Experimentally we observe chiral light emission from quantum dots over split-ring resonant nanoantennas, where the spin of the emitted photons is locked to their transverse momentum. We demonstrate that the polarization can vary from linear to elliptical with ellipticity reaching ±0.5 for emission into opposite halves of the symmetry plane of the nanoantenna.

**KEYWORDS:** nanoplasmics, emission control, quantum dots, split-ring resonator, Stokes coefficients

Light emission from molecular or semiconductor fluorophores is at the foundation of molecular fluorescent spectroscopy and detection of biomolecules and proteins. 1 In recent years, the increased sensitivity of fluorescence spectroscopy techniques has allowed the deterministic detection of molecules with low abundance down to even single-molecule level. 2, 3 This has opened a number of important applications, including DNA sequencing, 4 fluorescence nanoscopy of living cells, 5 and molecular labeling. 6 At the heart of fluorescent single-molecule detection is the use of plasmic nanoantennas, which enable the efficient transfer of the fluorophore emission from the near- to the far-field and vice versa. 7, 8 Furthermore, plasmic nanoantennas enable enhancement of the spontaneous emission rates 9–13 and increased directionality of fluorescent emission, 14 thereby dramatically enhancing the detection efficiency.

Most optical nanoantennas to date draw their functionality from the excitation of electric dipole moments of its constituent elements, hence neglecting higher-order contributions. These contributions, however, become increasingly important for more complex plasmonic nanoparticles supporting strong higher-order multipolar moments, 15–17 thus presenting intriguing opportunities for conceptually new nanoantennas. So far, the research on multipolar plasmonic nanoantennas has focused only on studies of emission enhancement or directivity, including important examples of off-resonant multipolar nanoantennas for directional emission. 18, 19

However, multipolar nanoantennas may also manipulate the polarization state of emission of fluorophores and even control the spin of the emitted photons. Nevertheless, this unique functionality remains largely unexplored to date. Importantly, the emission of photons with different spin by a fluorophore through the use of optical antennas promises a broad range of applications in the control of atomic systems by the photonic environment. Such control can be of interest when selection rules in atomic and molecular transitions have to be obeyed. In particular, the employment of nanoantennas can possibly discriminate between emission of different chiral enantiomers or manipulate the spin of photons emitted by an atom.

Here, we propose and demonstrate the manipulation of the polarization state of light emission in a system of semiconductor quantum dots (QDs) coupled to single-element multipolar nanoantennas. In particular, we show that coupling of QDs to a plasmonic nanoantenna, which is characterized by a superposition of several resonant multipolar contributions (electric/magnetic dipole moments, quadrupole moments, etc.) with a fixed phase difference, leads to the emission of chiral light of opposite handedness from the coupled QD–nanoantenna system. This selective spin-polarized far-field emission from the antenna–emitter system makes our multipolar nanoantenna an ideal candidate for fluorophore detection or advanced light generation. As such, our concept opens a new route to practical nanoscale light sources of spin-polarized photons as well as for simple recognition of emission from chiral emitters of different handedness.

The conventional way for the generation of circularly polarized radiation of different handedness that is well known from radio physics is to employ helical antennas. Transferring this design rule to optics has led to a surge of activities in the field of passive chiral plasmonic structures 20–24 and so-called extrinsically chiral systems. 25–27 Several concepts for chiral light
generation have been explored. These include the circular emission from QDs coupled to surface plasmons on spiral grooves in metal films, as well as selective directional coupling of circularly polarized emitters to metal slot waveguides and hyperbolic media. However, all these examples represent micrometer-sized structures, which contrast with the concept of compact nanoantennas for localized coupling to emitters. Other approaches for generation of circularly polarized radiation are based on phase-retardation techniques; however, all they are highly sensitive to the exact positioning and orientation of the emitter. Altogether, none of the above concepts of chiral light emission allow for reliably controlling the spin of photons (ellipticity and handedness of polarization) emitted from a nanoscale light source.

We overcome these limitations by applying a new approach for circularly polarized light generation based on a multipolar nanoantenna driven in the near-field by an electric dipole emitter, e.g., a QD. Since, in this configuration, the electric and magnetic multipoles of the nanoantenna are normally significantly larger than the dipole moment of the QDs, the far-field emission pattern of the coupled system is dominated by the radiation pattern of the nanoantenna’s multipole contributions. Hence, the superposition of the radiative patterns from the dominating multipole contributions of a nanoantenna provides a unique way to generate circularly polarized light. For this purpose, an excitation of at least two multipolar components is required.

We expand the far-field emission of the nanoantenna into a multipolar series and take into account the first three terms of the expansion: the electric dipole moment $p$, the magnetic dipole moment $m$, and the electric quadrupole moment $Q$, which at resonance can be written in the form

$$E(r) = \frac{k_0^2 e^{ik_0r}}{4\pi \varepsilon_0 r} \left( [\mathbf{n} \times [\mathbf{p} \times \mathbf{n}]] + \sqrt{\varepsilon_0 \mu_0} [\mathbf{m} \times \mathbf{n}] + \frac{i k_0}{6} [\mathbf{n} \times [\mathbf{n} \times \mathbf{Q} \mathbf{n}]] + \ldots \right),$$

where $k_0$ is the photon wavevector, $\mathbf{n} = k_0/|k_0|$ is the unit vector in the direction of emission, $r$ is the coordinate vector, and $r = |r|$. As the polarization of the field emitted by each multipole is linear, the following requirements need to be fulfilled to obtain circular polarization of the emitted light. First, the electric fields from two multipoles need to be orthogonal to each other. Second, the fields need to have a $\pm \pi/2$ phase difference. Third, the fields need to have the same amplitude. To illustrate how the requirement of orthogonality of fields from the two multipole contributions can be fulfilled, we consider the case where the electric dipole moment $p$ is oriented along the $x$-axis, the magnetic dipole $m$ points in the $z$ direction, and the quadrupole tensor has two nonzero components, $Q_{yz} = Q_{zy}$ (see Figure 1). For this case we can see from the cross products in eq 1 that the electric fields in the $x$-$z$ plane from the electric dipole $E_y \propto [n \times [p \times n]]$ is perpendicular to the fields from the magnetic dipole $E_m \propto [m \times n]$ as well as to the electric quadrupole $E_Q \propto [n \times [n \times Q_n]]$. From eq 1 it also follows that a $\pi/2$ phase difference naturally occurs between the electric dipole term $E_p$ and the electric quadrupole term $E_Q$ as $E_Q \propto i = \exp(i\pi/2)$. In addition, we also expect a $\pi/2$ phase difference between $E_m$ and $E_Q$ for any plasmonic structure that can be described by an LC-circuit model, as for example a U-shaped split-ring resonator (SRR) since, in an LC-circuit picture, the magnetic moment is then given by $m_{LC} \propto -i p_{LC}$. Notably, for both cases, we expect the electric-field amplitudes of the superimposed contributing multipole moments to be different in each direction in $k$-space. This generally leads to the emission of elliptically polarized light with different values of ellipticity for each direction. Due to the orientation and symmetry of the multipole radiation patterns [as reflected in the cross products in eq 1], the polarizations of emission with $k_x < 0$ and $k_y > 0$ have ellipticity with opposite sense of rotation; hence, for emission with $k_x < 0$ and with $k_y > 0$ the photons with opposite spin are dominant. Importantly, the effect of circularly polarized light generation relies on the coherent superposition of multipolar components of a single nanoparticle resonance, which is an inherent property of the resonant antenna and does not require coherence of the excitation process itself.

To prove the concept of spin-polarized emission experimentally, we employ SRRs as multipolar nanoantennas coupled to semiconductor QDs emitting at around 800 nm wavelength (see Figure 2). Since SRRs have been a subject of extensive research in the last 10 years, their resonant properties and multipole moments are well understood. On the basis of this knowledge we chose SRRs as a convenient platform for multipolar nanoantenna studies; however, our results remain general and are valid for other types of antennas.
that support multipolar resonances. We fabricate gold SRRs in a periodic lattice with the lattice constant of \( a = 300 \text{ nm} \) on a glass substrate and cover them with a MgF\(_2\) spacer. A uniform layer of QDs is then spin-coated on top of the MgF\(_2\) layer, with a thickness of 12 nm has been chosen to achieve high coupling efficiency between the QDs and the SRRs (Figure 2b). Direct application of the QDs onto the metallic nanostructures could lead to PL quenching, whereas large separation distances would lead to a reduction of the coupling strength. The spin-coating leads to a randomized positioning of the QDs. We therefore investigate how the generation of spin-polarized light is affected by the positioning the emitter in a fixed plane above the SRR. We find that the dominant contribution for spin-polarized emission originates from the emitters positioned close to the center of a SRR (see Figure S5 of the Supporting Information).

To characterize the optical response of the SRRs, first, we collect their extinction spectra \( \ln(1 - T) \) using a white-light source for excitation. Figure 3a shows the spectra for two orthogonal linear polarizations of the incident plane wave (see inset in Figure 3a). The spectrum is dominated by three resonances (resonances 1–3). For vertical polarization [black line] resonance 1 is excited featuring only a strong electric dipole moment along the \( y \) direction. For horizontal polarization [red line] resonances 2 and 3 are excited. These two resonances are often referred to as the SRR higher-order resonances.\(^{13}\) The fundamental magnetic SRR resonance (not shown) appears at around 1400 nm wavelength and is dominated by an electric dipole moment along the \( x \) direction and a magnetic dipole moment along the \( z \) direction (see Figure S1 of the Supporting Information), corresponding to the schematic shown in Figure 1a. However, for a typical SRR the scattering cross section of the magnetic dipole is smaller than the scattering cross section of the electric dipole at the fundamental SRR resonance,\(^{16}\) leading to only a small fraction of the emitted light exhibiting elliptical polarization. Resonance 2, on the other hand, is characterized by a superposition of an electric dipole along the \( x \) direction, an electric quadrupole with two nonzero components \( Q_{ex} = Q_{ey} \), and a relatively weak magnetic-dipole moment along the \( z \) direction,\(^{16}\) which is neglected in the following. These resonance characteristics correspond to the case shown in Figure 1b. Importantly, for resonance 2 the scattering cross sections of the electric dipole and of the electric quadrupole reach comparable magnitudes,\(^{16}\) rendering resonance 2 well-suited for the observation of spin-polarized photon emission. Therefore, we focus on this resonance in the following.

To measure the emission of the QD−SRR nanoantenna system, we optically excite the QDs at \( \lambda = 532 \text{ nm} \), collect their emission from the substrate side of the sample, and measure the photoluminescence (PL) spectra. Since we use off-resonant pumping of the QDs well below the SRR resonances, the average QD excitation rate is not dependent on the SRR resonances and, hence, independent of the polarization of the pump beam. The QD PL spectrum is shown with gray shading in Figure 3a. Notably, the QD emission overlaps spectrally with the SRR resonances 1 and 2. Figure 3b shows the measured PL signal from the QD−SRR, normalized to the reference PL of the QDs on the plane glass substrate with the MgF\(_2\) layer next to the arrays. Importantly, we observe that even though the SRRs show a pronounced extinction maximum, i.e., a transmission minimum around 800 nm for both polarizations, the emission intensity of the QDs on top of SRRs is approximately a factor of 2 higher than the emission of QDs on the substrate (without SRRs). This enhancement at the resonance wavelengths is a clear signature of coupling between the QDs and the SRRs. Note also that for longer wavelengths at \( \lambda \approx 850 \text{ nm} \), the normalized PL spectrum shows a depletion dip, which is another signature of coupling.\(^{39}\)

Next, we investigate the polarization properties of the PL signal in Fourier space (\( k \)-space), i.e., with regard to different directions of emission. To this end, we perform the same excitation of the QDs and collect the PL signal from the substrate side within the available angle of about 44 degrees that is limited by total internal reflection inside the substrate. After filtering out the excitation signal, we introduce an achromatic quarter-wave plate and a linear polarizer into the optical path to detect a specific polarization state of light. We then record the image of the back-focal plane of the collecting
objective with a camera (see Figure S6 of the Supporting Information).

The obtained back-focal-plane images allow us to analyze the polarization properties of the emission pattern in $k$-space via the Stokes parameters formalism in terms of its total intensity $I_{tot}$ (fractional) degree of polarization $p$, the polarization inclination angle $\psi$, and the ellipticity $\tan(\psi)$ (see Tables S1 and S2 of the Supporting Information for full set of Stokes parameters and coefficients).

From the Stokes parameters we calculate the polarization state of light for every point of the experimentally accessible part of the $k$-space and plot the corresponding values for the ellipticity of the resulting polarization state in Figure 4c: an ellipticity of zero corresponds to linear polarization, while ellipticity values of $\pm 1$ correspond to right-handed/left-handed circular polarizations, respectively. Our results show that along the symmetry plane of the SRRs ($k_x = 0$) the emission of the system is vertically polarized and consequently has zero ellipticity, while for $k_x \neq 0$ the emission becomes elliptically polarized. The polarization pattern is mirror-symmetric to the $k_x = 0$ plane. Both the ellipticity and the polarization inclination increase with increasing $k_x$. The maximum ellipticity experimentally obtained is $\pm 0.5$. For $k_x < 0$ the emission is right-handed elliptically polarized, while for $k_x > 0$ it is left-handed elliptically polarized. Furthermore, we find that the polarization inclination qualitatively follows this behavior, exhibiting opposite inclinations in opposite ($\pm k_x$) semiplanes in $k$-space. We also observe a non-negligible amount of depolarization in the experiment.

In order to support our experimental results, we perform numerical calculations using CST Microwave Studio. We place a small dipole source over a single SRR on top of a MgF$_2$ spacer and subsequently extract the polarization state of the emission. We find that the main contribution for the spin-polarized emission of photons can be attributed to QDs that are situated in the center of a single SRR (for details see the Supporting Information). Since in our experiment we average over all orientations of QDs, we calculate the average values for the three cases of the dipole pointing in $x$, $y$, and $z$-direction. The results are shown in Figure 4b and well reproduce our experimental measurements of Figure 4c. Our calculations also show that for a single dipole emitter with an optimized orientation and position with respect to the SRR, the emission can be purely circularly polarized in specific directions (see Figure S4d in the Supporting Information). We therefore attribute the experimentally observed depolarization and reduction of the effective overall ellipticity to a maximum value of 0.5 to the incoherent superposition of resonances 1 and 2 of the SRRs, as well as to the random positioning and orientation of QDs.

In conclusion, we have demonstrated that, by using single-element multipolar plasmonic nanoantennas, we can create spin-polarized light emission from a subwavelength light source created by semiconductor QDs coupled to SRRs. We make use of the phase-locked superposition of the radiation fields of the electric dipole and quadrupole contributions of a single SRR resonance to control the direction-dependent polarization state of emission. The polarization state can be further controlled by directional filtering in $k$-space.

Importantly, the effect of spin-polarized emission is pronounced even for an array of SRRs with randomly distributed QDs. Our novel concept of generating spin-polarized light emission is generally applicable to other types of nanostructures that support multipolar resonances, and it enables the design of compact optical nanoantennas based on single nanoparticles.

Finally, the observed effect of spin-controlled light emission can be linked to the properties of thermal emission of spin-controlled metasurfaces where anisotropic optical antenna patterns result in spin–orbit interaction of light and emission of spin-polarized light with opposite spin in different directions. This emission is considered analogous to the optical spin Hall effect studied recently for passive plasmonic metasurfaces and plasmonic chains. As such, our results on spin-controlled light emission with achiral nanoantennas exhibiting two or more multipolar moments open a new efficient route toward spin-controlled photonics with a range of possible applications in molecular detection, quantum light sources, or display technologies.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information gives details on the resonances and multipole contributions of the split-ring resonators, as well as on the scenarios of localized dipole excitation. Further details are also provided on the fabrication procedures and the experimental setup for back-focal-plane measurements of the Stokes coefficients. This material is available free of charge via the Internet at http://pubs.acs.org.
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Notes
The authors declare no competing financial interest.

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REFERENCES