Communication

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Active tuning of spontaneous emission by
Mie-resonant dielectric metasurfaces

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Abstract

Mie-resonant dielectric metasurfaces offer comprehensive opportunities for manipulating light fields with high efficiency. Additionally, various strategies to dynamically tune the optical response of such metasurfaces were demonstrated, making them important candidates for reconfigurable optical devices. However, dynamic control of the light emission properties of active Mie-resonant dielectric metasurfaces by an external control parameter has not been demonstrated so far. Here we experimentally demonstrate dynamic tuning of spontaneous emission from a Mie-resonant dielectric metasurface, which is situated on a fluorescent substrate and embedded into a liquid crystal cell. By switching the liquid crystal from the nematic state to the isotropic
state via control of the cell temperature, we induce a shift of the spectral position of the metasurface resonances. This results in a change of the local photonic density of states, which in turn governs the enhancement of spontaneous emission from the substrate. Specifically, we observe spectral tuning of both the electric and magnetic dipole resonances, resulting in a twofold increase of the emission intensity at $\lambda \approx 900$ nm.

Our results demonstrate a viable strategy to realize flat tunable light sources based on dielectric metasurfaces.

**Keywords**

all-dielectric nanophotonics, dielectric nanoantennas, Mie-resonances, emission enhancement, tunable metasurfaces, active metasurfaces, liquid crystals

Photonic metasurfaces, consisting of designed plasmonic or dielectric nanoresonators that are arranged in a plane, offer a wide range of opportunities for controlling the properties of light fields transmitted through them or reflected from them, such as the spectrum, polarization and wavefront\(^1\),\(^2\) Dielectric metasurfaces in particular have received a growing amount of attention recently owing to their low absorption losses, which allow for high-efficiency metasurface devices\(^3\)–\(^15\). For example, dielectric metasurfaces allow for the realization of flat and efficient beam-shapers\(^3\)–\(^4\),\(^6\)–\(^8\) beam deflectors\(^5\),\(^10\) focussing devices\(^6\),\(^11\) and holograms\(^12\),\(^13\) to name just a few. Most metasurfaces realized so far are passive and thus rely on external light sources in order to synthesize light fields with desired properties. However, the potential of resonant dielectric metasurfaces to enhance and tailor spontaneous emission was also investigated\(^16\),\(^17\) pointing towards their possible deployment as flat light sources with emission properties that can be tailored by the metasurface design. The key for the manipulation of spontaneous emission are the electric and magnetic dipolar Mie-type resonances supported by high refractive-index-contrast dielectric metasurfaces\(^18\), which can concentrate the electromagnetic near-fields inside or near the nanoresonators. Thereby, they can enhance the interaction of light with the composite material of the nanoresonator or its immediate
surroundings. An important special case of light-matter interaction enhancement is the Purcell effect, which was recently theoretically\textsuperscript{19} and experimentally\textsuperscript{20,21} studied for isolated dielectric Mie resonators. As such, the metasurface can be considered as a subwavelength array of dielectric nanoantennas,\textsuperscript{22} where the low absorption loss of the dielectric material directly translates into a high radiation efficiency and low heat conversion of the individual nanoantennas.\textsuperscript{23,24} As a consequence, a high level of control is obtained over the properties of the emitted light field without adding significant non-radiative channels to the system. This mechanism can also be harnessed to enhance and tailor other light-matter interaction processes, such as frequency conversion and other nonlinear optical processes.\textsuperscript{25,26}

Another important prospect of metasurfaces is their potential for the realization of tunable optical components that can be compact as well as cost- and energy-efficient. Various mechanisms were suggested for tuning of metasurfaces (see e.g.\textsuperscript{27} for a recent review). In particular, numerous tuning methods have been demonstrated for dielectric metasurfaces, such as mechanical stretching of the substrate,\textsuperscript{28,29} temperature tuning,\textsuperscript{30,31} integration of the metasurface into a liquid crystal cell,\textsuperscript{32,33} generation of free carriers,\textsuperscript{34,35} \( \chi^{(3)} \) nonlinearities\textsuperscript{36} and the use of phase change materials.\textsuperscript{37} Thus, by combining active tunability of the resonance positions with the resonant enhancement of spontaneous emission, resonant metasurfaces hold the potential to act as on-demand sources of complex tailored light fields. Using a plasmonic metasurface, dynamic spectral control of spontaneous emission was previously demonstrated, which achieved a spectral shift of 10 nm with an emission linewidth of 6 nm.\textsuperscript{38} However, the dynamic manipulation of spontaneous emission by dielectric metasurfaces has not been realized so far. The use of dielectric instead of plasmonic implementations is essential wherever one targets high efficiency solutions, such as in display applications or light sources.

Here, we demonstrate for the first time tuning and switching of spontaneous emission using a silicon metasurface exhibiting electric and magnetic dipolar Mie-type resonances around 900 nm wavelength. A sketch of our system and the deployed switching mechanism is shown
in Fig. 1. The intrinsic fluorescence from the glass substrate of the silicon metasurface is used as emitters. Autofluorescence of glass is a common phenomenon which can originate from point defects and impurities. The glass fluorescence is resonantly enhanced by the presence of the metasurface. In order to induce a spectral shift of the metasurface resonances, and thereby a change in the local photonic density of states governing the emission enhancement, we use the refractive index difference of a liquid crystal (LC) for the nematic and the isotropic state.

![Figure 1: Sketch of a silicon nanocylinder metasurface integrated into a liquid crystal cell. By heating the liquid crystal, it changes its state from nematic to isotropic, resulting in a spectral shift of the metasurface resonances, and tuning of the emission enhancement.](image)

For fabrication of the silicon metasurface samples, we first deposit a 182 nm thick layer of hydrogenated amorphous silicon (a-Si:H) on a standard microscope cover slip using plasma-enhanced chemical vapour deposition (PECVD). Square arrays (lattice constant of 560 nm) of nanocylinders with various diameters and a constant height, fixed by the a-Si:H layer thickness, are then defined by electron-beam lithography (EBL) in combination with inductively coupled plasma (ICP) etching of the silicon thin-film. To render the sample surface conductive for imaging in an electron microscope, we cover it with a thin (15 nm) transparent layer of indium tin oxide (ITO). A scanning-electron micrograph (SEM) of a typical sample is shown in the inset of Fig. 2 (a).

The linear-optical transmittance spectrum of the fabricated sample is shown in Fig. 2 (a).
The measurement was performed with a custom white-light spectroscopy setup attached to an optical spectrum analyzer (a sketch and more details available in the Supporting Information). The spectrum is referenced to the unstructured substrate next to the metasurface. It clearly shows two pronounced resonance minima at 689 nm and 797 nm, as well as a less pronounced resonance feature below 600 nm. In the following, we will concentrate on the discussion of the two more pronounced resonance minima. To compare our experimental results with theoretical expectations, we performed numerical calculations using the commercial software package CST Microwave Studio. We used experimental values for the dispersive a-Si:H refractive index as well as for the geometrical parameters of the metasurface taken from SEMs. The substrate refractive index was taken as \( n = 1.51 \). Furthermore, our model accounts for the formation of a low refractive index layer at the nanocylinder surface due to oxidation of the silicon during fabrication. The best agreement is observed for a layer thickness of \( t_{\text{SiO}_2} = 26.7 \) nm. This assumption is consistent with the observation of a thin low-contrast layer at the outer nanocylinder surfaces in focused ion beam (FIB) cross section images. The simulation results are shown in Fig. 2 (a, dashed-line) and show a good agreement with experimental data. The corresponding near-field distributions at the resonance positions are shown in Fig. 2 (b), allowing us to identify the two resonances as the fundamental electric and magnetic dipolar Mie-type resonances of the silicon nanodisk metasurface.
Figure 2: (a) Measured (solid line) and numerically calculated (dashed line) linear-optical transmittance spectra of the a-Si:H metasurface on a substrate in air, with a disk height of $h = 182\, \text{nm}$, a lattice constant of $a = 560\, \text{nm}$ and a diameter of $d = 237\, \text{nm}$. The inset shows an SEM of a similar sample with slightly smaller nanocylinder diameter. The two pronounced minima correspond to the electric (red circle) and magnetic (blue circle) dipole resonances of the metasurface. The corresponding calculated field distributions are displayed in (b). The white dotted lines mark the interface to the substrate, the outer borders of the nanocylinder and the optimized thickness of the low-index layer used in numerical calculations.

Next, we integrate the metasurface sample into an LC cell following the procedure described in our previous works.\textsuperscript{32,33,43} To this end, we sandwich a layer of $5\, \text{µm}$ of the nematic LC Licristal E7 from Merck between the metasurface sample and a glass substrate, which was coated with a brushed polymer layer used for planar alignment of the LC molecules. The layer consists of Nylon-6 dissolved with a mass concentration of 1% in 2,2,2-trichlorethanol (Sigma Aldrich). The cell is then attached to a transparent sapphire base substrate for thermal conductivity. A resistor (WELWYN, WH10 15R JI) and a PT-100 sensor are glued to the latter with a thermally conductive paste. These are connected to a temperature controller (ILX Lightwave, LDT-5980), together forming the heating unit of our cell. Note that our cell is fully transparent at visible and near-infrared frequencies, allowing us to probe the metasurface in transmittance at operation wavelengths around 900 nm, where we observe significant intrinsic emission from the sample, while the sapphire base still ensures an
efficient thermal contact between the heater and the sample.

The tuning properties of the metasurface sample are summarized in Fig. 3. Note that the presence of the LC leads to a redshift of both spectral resonance positions with respect to the spectra shown in Fig. 2 as well as a swap in the order of the two resonances (see Supporting Information). Fig. 3 (a) shows the colour-coded transmittance spectra for a systematic increase of the temperature from room temperature to 67°C. The incident light is polarized along the brushing direction, which coincides with the preferential direction of LC alignment and approximately the anisotropy axis of the LC at room temperature. For temperatures below the critical value $T_c$, the resonance positions depend weakly on the temperature, with the electric resonance showing a slight gradual blue shift and the magnetic resonance showing a slight gradual red shift. At the critical temperature of the LC of 58°C, where the LC transitions from nematic to isotropic phase, we observe a dramatic change of the transmittance spectrum. The transmittance now exhibits only a single minimum, resulting from a spectral shift of the two resonances towards each other in combination with an interplay of both resonances with the diffractive modes of the structure. The temperature dependence of the transmittance spectral position is weak for temperatures beyond $T_c$. All these observations are in accord with previous results on metasurfaces with operation frequencies in the telecom spectral range. To allow for a better quantitative assessment of the spectral changes associated with the phase transition of the LC, Fig. 3 (b) shows a direct comparison of the spectra for 32°C and 67°C.
Figure 3: (a) Transmittance spectra of the metasurface over a range of temperatures below and above $T_c$. The yellow dashed line indicates the LC phase transition temperature. (b) Metasurface transmittance for 32°C (nematic, blue) and 67°C (isotropic, red), where the insets illustrate the respective LC orientations.

Next, we study the emission properties of our metasurface sample in air using a commercial confocal microscope setup (Picoquant, MicroTime-200) with a NA=0.25 objective (Olympus, Plan N) coupled to a spectrometer (Horiba, iHR320).

A 532 nm laser with an average excitation power of 1.4 mW (Pulse:
\( \sim 100 \text{ps}, \ 80\text{MHz repetition rate} \) was used as the excitation source. The results are presented in Fig. 4. For reference, we also measured the emission from the bare glass substrate next to the metasurfaces (grey areas). Clearly, the emission spectrum is strongly reshaped by the presence of the metasurface. In particular, the two spectral maxima positions of the emission match well with the metasurface resonance positions observed in the linear-optical transmittance spectrum, which is included in the image as a dashed line for a direct comparison. These observations can be explained by an increase of the radiative rate through the Purcell effect\(^{45}\) caused by increased photonic density of states at resonance.

Note that the emission of the substrate with and without the metasurface coincide on the long-wavelength side of our measurement range, i.e. beyond the lowest-order Mie-type mode of our system, while the non-negligible background to the emission enhancement on the short-wavelength side of the electric dipole mode is likely due to the collective effect of various higher-order photonic modes.

Finally and as the central step of this work, we study the emission properties of the LC integrated metasurfaces as a function of temperature. The emission spectra for room temper-
ature and 67°C are shown in Fig. 5 (a). Heating the sample above the critical temperature leads to a pronounced spectral reshaping effect. Again, the spectral density of the emission correlates very well with the position of the resonances in the linear-optical spectra (dashed lines) for the respective temperature as seen in Fig. 5 (a). It becomes even more obvious in Fig. 5 (b), which shows the emission contrast, defined as the spectral change of the emission upon heating \( \Delta E_m \) referenced to the room temperature emission spectrum \( E_m(25°C) \). As the LC transitions into the isotropic phase, the emission intensity at the spectral position of the resulting single transmittance feature increases by up to 90%. The reproducibility of this effect was tested by recording the emission spectra for several subsequent cycles where the sample was first heated and then allowed to cool down again. Note that for the emission tuning measurements we observe a decrease of the LC phase transition temperature by about 10°C as compared to the transition temperature observed in transmission measurements. A detailed temperature scan can be found in the Supporting Information. To further support our experimental observations, we also performed exemplary numerical simulations based on the reciprocity principle. The results are also included in the Supporting Information.
Figure 5: (a) Emission spectrum and (b) emission contrast (solid lines) of the silicon nanocylinder metasurface after integration into the LC cell for room temperature (nematic case, Em. nem.) and 67°C (isotropic case, Em. iso.). The data was smoothed by averaging over neighbouring data points. The dashed lines show the corresponding transmittance spectra of the metasurface in the LC cell.

In conclusion, we have demonstrated active tuning of spontaneous emission using a Mie-resonant dielectric metasurface situated on a fluorescent glass substrate and embedded into a nematic LC cell. Heating of the sample above the critical temperature, where the LC transitions from nematic to isotropic phase, leads to a pronounced change in the metasurface transmittance spectrum. Since the fluorescence from the glass substrate is significantly
enhanced by the metasurface resonances owing to the Purcell effect, the phase transition induces a clear reshaping of the emission spectrum. In particular, the emission intensity almost doubles at 900 nm for the transition of the LC from nematic to isotropic. Our results demonstrate that tunable dielectric metasurfaces provide a viable and efficient route to dynamic control of spontaneous emission, which could find applications in next generation displays and smart light sources. While we concentrate on dynamic shaping of emission spectra in this work, the concept could be extended towards active control of the directional and polarization properties of emission.

Supporting Information Available

Sketch of the experimental temperature-dependent transmission measurement setup; numerical simulations of transmittance and emission for silicon nanocylinder arrays situated on a glass substrate and embedded into LC.

Author information

The authors declare no competing financial interest.

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