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Published in:
APL Photonics

DOI:
10.1063/1.5096270

Published: 01/07/2019

Document Version
Publisher's PDF, also known as Version of record

Please cite the original version:
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Large-area enhancement of far-field fluorescence intensity using planar nanostructures

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ABSTRACT
While nanoscale local enhancement of fluorescence is a well-studied phenomenon, enhancement of far-field radiation of large-area samples using nanostructures has been explored much less. In this work, we demonstrate planar nanostructures with exceptionally high far-field intensity enhancement factors and in addition show that the enhancement can be made controllable via the polarization state of the pump light. We also find that, counterintuitively, the Purcell effect plays only a small part in the observed far-field enhancement. The results of these studies can be applied, for example, in thin-film optical emitters, fluorescence sensors, and wide-field fluorescence microscopy.

I. INTRODUCTION
Fluorescence is spontaneous emission of light by optically excited quantum emitters, such as atoms, molecules, and quantum dots. It is widely used, for example, in the detection and analysis of chemical species, fluorescence microscopy, and light sources. Averaged over many emitters in a homogeneous medium, spontaneous emission results in an isotropic emission pattern. Thin-film and nanopatterning technologies make it possible to control the electromagnetic environment and thus the spontaneous emission characteristics in versatile ways. For example, nanoantennas and optical metasurfaces can increase the emission rate of a molecule via the Purcell effect, concentrate the emission into a certain direction, and enhance the rate of optical pumping. These effects are typically designed to affect emitters in the strong near-field “hot-spots” of plasmonic nanoparticles or microwaves. However, when emitters are deposited over large areas, most of them are outside the limited hot-spot locations, where the enhancement takes place, and thus are not affected. This can be a disadvantage, when high fluorescence intensities of extended samples are desired. Previous work on enhanced fluorescence emission of large-area sources has included the use of Fabry-Perot-type resonant multilayer structures and gratings, where changes in radiation pattern, spectrum, and fluorescence lifetime were observed. However, the investigations have often been limited to the control of the Purcell effect and the emission spectrum, with only occasional foray into the enhancement of the far-field intensity, which is an important parameter for light sources and detectors. Unexplored possibilities in this area include the combination of optical pump enhancement with the enhancement of directivity of emitted light.

In this work, we first develop a set of theoretical tools for designing nanostructures for far-field fluorescence enhancement. We then design and experimentally demonstrate planar metal nanostructures that enhance remarkably the far-field fluorescence intensity, which is the quantity of interest in the majority of applications. The structures enhance spontaneous emission to the far field by changing the spatial distribution of the electromagnetic modes,
while having only a small effect on the local density of states. The structure that we use as a reference in our experiments is a gold mirror exploiting intensity enhancement by reflection. Then, we demonstrate a layered metal-dielectric structure that combines the functions of a Fabry-Perot resonator and a metal waveguide at the cutoff frequency. The structure is able to enhance the far-field fluorescence intensity by a factor of 350. We directly measure an enhancement factor of 120. We also study a subwavelength grating metasurface that enhances the fluorescence of quantum emitters placed on top of it, with the enhancement factor significantly depending on the pump polarization. The polarization dependence enables, for instance, fast modulation of the fluorescence power.

II. THEORY AND DESIGN

Our design method is based on the extension of the existing theory of single-emitter fluorescence enhancement (see Refs. 26 and 14) to the far-field radiation of large ensembles of quantum emitters. The fluorescence enhancement factor \(F(r)\) is defined as the ratio of detected fluorescence power (or power spectral density) to the power detected in a reference case of molecules placed in an infinite homogeneous dielectric host medium; the emitter under consideration is located at coordinate \(r\). In our analysis, the factor consists of two parts, the pump rate enhancement factor \(F_p(r)\) and the emission enhancement factor \(F_e(r)\), such that \(F(r) = F_p(r)F_e(r)\). The emission enhancement factor can be expressed as (see the Appendix for details)

\[
F_e(r) = \frac{\gamma_\text{ff}(r)}{\gamma_{\text{ref}}} \frac{1 + \Gamma}{P(r) + \Gamma}, \tag{1}
\]

where \(\gamma_\text{ff}(r)\) is the rate of spontaneous emission into the detected far-field optical modes, \(P(r)\) is the Purcell factor, and \(\Gamma = 1/\eta - 1\) is the intrinsic loss coefficient of the emitter determined by the intrinsic quantum yield \(\eta\). The quantity \(\gamma_{\text{ref}}\) is the detected emission rate for the reference case of an emitter in a homogeneous dielectric environment. As we are interested in the enhancement of normally directed radiation intensity, the factor \(\gamma_\text{ff}(r)/\gamma_{\text{ref}}\) can be approximately calculated as \(\gamma_\text{ff}(r)/\gamma_{\text{ref}} = |E_e(r)|^2/|E_{\text{ref}}(r)|^2\), representing the enhancement of the coupling strength to the mode directed normally. Here, \(|E_e(r)|^2\) is the squared electric-field amplitude of a normally incident plane wave at \(r\), and \(|E_{\text{ref}}(r)|^2\) is the same in a reference case. This is a direct consequence of the Lorentz reciprocity principle. The Purcell factor \(P(r)\) may be found by calculating the overall power emitted by a time-harmonic electric dipole located at \(r\) and comparing it to the power radiated by the same dipole in the reference case. Note that in Eq. (1), counterintuitively, the Purcell factor appears in the denominator, implying that large \(P(r)\) leads to a lower far-field enhancement factor. This is indeed the case because \(P(r)\) includes the rates of emission into all electromagnetic modes including those that are not seen in the far field. Hence, a large \(P(r)\) without a correspondingly large \(\gamma_\text{ff}(r)\) reduces the far-field fluorescence enhancement. As an example, quantum emitters on metal surfaces show a high Purcell factor, but their far-field emission is quenched.

The enhancement of the pump absorption may be predicted from the intensity of an incident plane wave as well, but this time at the pump wavelength. The pump enhancement factor is \(F_p(r) = |E_p(r)|^2/|E_{\text{ref}}(r)|^2\). For continuously distributed emitters in a film, the measured fluorescence enhancement factor is the spatial average of the combined pump and emission enhancements,

\[
F = \frac{1}{V_{\text{ff}}} \iint_V \langle F_p(r)F_e(r) \rangle \, dr, \tag{2}
\]

where the integration is performed over the fluorescent medium with volume \(V_{\text{ff}}\). It is important to average over all possible orientations of the dipole moments of the molecules. The angle brackets denote this averaging. For the designed thin-film samples, we performed these calculations analytically by using a transfer-matrix method combined with an electric-current decomposition method (see also the Appendix), and for the grating sample, we used the finite element method-based computer software Comsol Multiphysics.

In the following, we consider three simple structures with which we aim to enhance both the optical pumping rate and the directivity of the fluorescence. The fluorescent medium in our experiments is a dye-doped polymer [poly(methyl methacrylate) (PMMA)] doped with the IR-780 dye. The structures are shown in the left column of Fig. 1. They are all pumped from the top with a plane wave incident at an angle.

The reference sample, shown in Fig. 1(a), is a 200 nm thick fluorescent film on a glass substrate. The structure of Fig. 1(c) is the same fluorescent film, but on top of a 50 nm gold film, which acts as a reflector. The gold film enhances the optical pump wave intensity because the incident and reflected waves form a standing wave (sinusoidally varying in the normal direction). For an emitter, the emission in the normal direction can be enhanced due to a similar interference effect. In the structure of Fig. 1(e), the structure of Fig. 1(c) has been coated with another gold film to form a resonator. The top film is semitransparent, and its thickness is chosen to obtain a desired enhancement factor and spectral width. Both the pump intensity and fluorescence directionality enhancement can be substantially increased, if both of the associated plane waves meet the Fabry-Perot resonance condition. Because the pump has a shorter wavelength than the fluorescence, this condition can be achieved if the pump wave is incident on the sample at a specific incidence angle. Furthermore, because the structure is only half a wavelength thick, it acts like a metal waveguide at its cutoff frequency. This prevents emission into plane waves that are not at normal incidence, further improving radiation directivity. Finally, the structure of Fig. 1(g) consists of a fluorescent film on top of a glass-covered silver grating. The grating aids in the excitation of surface modes (at the surface lattice resonance) by light polarized perpendicular to the metal strips, which increases the pump wave intensity. This is a polarization-dependent effect, enabling the modulation of fluorescence output by modulating the polarization of the pumping light.

The peak wavelength of fluorescence, the grating period is smaller than the wavelength in either air or glass, and diffracted or bound surface modes are not excited.

The quantum yield of IR-780 is assumed to be 0.076, corresponding to the value for IR-780 in methanol. The fluorescence and absorption spectra of the dye in PMMA host medium are presented later in the article. The dimensions of each of the samples (also shown in Fig. 1) were optimized for the fluorescence wavelength \(\lambda_0 = 780\) nm which is close to the peak of the dye’s emission spectrum. The pump wavelength was chosen as \(\lambda_0 = 650\) nm, except...
The purpose of the LiF film is to prevent dye molecules from adhering to the gold surface during the spin coating of the film. In the samples with mirror surfaces at the bottom, there is also a thin layer of lithium fluoride (LiF) between the Au and the fluorescent films.

The right column of Fig. 1 shows the calculated spectra of the fluorescence enhancement factors for the sample of Fig. 1(g) that was designed for 633 nm pumping. For the reference sample of Fig. 1(a), the enhancement factor is, by definition, $F = 1$ for all wavelengths [see Fig. 1(b)]. For the mirror-polymer structure of Fig. 1(c), the enhancement factor ranges from $F = 3.5$ to $F = 4$ in the wavelength range of interest [see Fig. 1(d)]. In the structure of Fig. 1(e), we predict a maximum enhancement factor of $F = 350$ at $\lambda_0 = 770$ nm for the optimal pump wave incidence angle of $57^\circ$. Consequently, we also pump the samples of Figs. 1(a) and 1(c) at this angle, which is accounted for in the calculations. The spectral shape of $F$ resembles the transmission spectrum of the resonator. In the grating structure of Fig. 1(g), using a TM-polarized pump wave (with the polarization perpendicular to the metal stripes of the grating), we obtain a total fluorescence enhancement of $F = 17$. For the TE-polarization, this factor is $F = 5$. This sample is pumped at normal incidence in contrast to the other samples. The thickness of the fluorescent film is only 50 nm because we want to maximize the polarization sensitivity of the sample and not the overall enhancement factor. For this sample’s reference, we also used a 50 nm thick fluorescent film. Finally, the spectra of the Purcell factor $P$ are shown for the first three samples. It is averaged over all dipole orientations and positions in the film. Its value is close to 1 for the first two samples. For the resonator sample, it ranges from 0.2 to 2.4, and at the enhancement peak, it is 1.5. The structure, therefore, only modestly changes the local density of states, whereas the spatial distribution of the states is very different from that in vacuum, which is reflected in the high value of $F$. Also, because IR-780 has a low quantum yield [and thus a large $F$ in Eq. (1)], the Purcell effect does not remarkably affect the value of $F$.

To analyze the radiation also at other exit angles besides the normal direction, we calculated the radiation directivity patterns of the resonator and grating samples at the 770 nm wavelength. These are seen in Fig. 2. The radiation pattern of the resonator in Fig. 2(a) is concentrated around the normal direction and has a full width at half maximum of about 40°. The directivity pattern of the grating in Fig. 2(b) is considerably wider, but still concentrated in the normal direction, except for two side lobes on the substrate side. These lobes arise from a prominent excitation of radiative surface plasmons on the grating stripes. They are fairly bright but carry less power than the main lobe around the surface normal.

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Figure 3 further clarifies the functionality of the resonator structure of Fig. 1(e). Figure 3(a) shows the numerically calculated intensity distribution resulting from a horizontally oriented electric dipole located at the center of the polymer layer and radiating at $\lambda_0 = 780$ nm. Because only the plane-wave components emitted along the normal direction undergo constructive interference, the emission is highly directive. This is seen in a wide, plane-wave-like intensity profile. Figure 3(b) shows the pump intensity distribution. The pump beam is TE polarized. A standing wave is visible above the structure because of interference of the incident and reflected waves. With optimal pumping, the pump intensity is increased by a factor of 10 at the center of the resonator. The emission enhancement factor, including the enhancement of directivity, is about 35 at maximum. Together they make up the predicted enhancement factor of 350. Figure 3(c) shows schematically the setup, in which the sample is pumped at an angle.

Figures 3(d) and 3(e) show the pump intensity distributions in the grating sample for the TM and TE polarizations, respectively. From these distributions, one can observe that for the TM polarization, the pump intensity is significantly enhanced inside the fluorescent film. For the TE polarization, the effect is much weaker, showcasing the designed polarization sensitivity.

III. EXPERIMENTS

The thin-film samples of Figs. 1(a), 1(c), and 1(e) were prepared on microscope slides. The slides were first cleaned by ultrasonication in four solvents, for 2 min in each: (1) a 5% solution of detergent (Micro-90, Sigma-Aldrich) in DI water, (2) DI water, (3) acetone, and (4) isopropanol. Gold films were deposited by e-beam evaporation together with a 5 nm Ti adhesion layer. Films of LiF were deposited by thermal evaporation. For each material, the deposition rate was about 0.3 nm/s. To fabricate the fluorescent film, 4% by weight PMMA (molecular weight 25 000, Polysciences, Inc.) was dissolved in toluene, and 10 mg/ml of IR-780 chloride (Sigma-Aldrich) was dissolved in methanol. The IR-780 solution was then added to the toluene-PMMA solution such that once the solvents are removed, the concentration of IR-780 in PMMA is 6.7 mmol/l.

The dye-doped PMMA films were deposited by spin coating and baked on a hotplate at 140 °C for 90 s. We measured the absorption and fluorescence spectra of a thick film (thickness 1.3 μm, dye concentration 7 mmol/l) of dye-doped PMMA (measurement setup described later). The spectra are presented in Fig. 3(f) by the green and red curves, respectively. The fluorescence spectrum was obtained by pumping at 650 nm (indicated by the green dashed line), and as stated previously, fluorescence enhancement is expected around the 780 nm wavelength (red dashed line). The pump wavelength is outside the fluorescence spectrum so that the spectrum can be measured without scattered pump light contributing to it.

For the grating structure [Fig. 1(g)], the pattern of the grating was exposed on a 200 nm thick double layer PMMA resist on a glass substrate using an electron beam writer. The bottom layer (molecular weight 250 000) is more sensitive than the top layer (molecular weight 950 000), resulting in an undercut that facilitates lift-off processing. A 30 nm thick layer of conductive polymer (eSpacer 300Z) was spin coated on top of the resist to prevent charging of the sample. The sample was developed after exposure, and 60 nm of silver was deposited by thermal vaporization. After a lift-off process, the metal remains on the substrate in the exposed regions. Immediately after the lift-off, the sample was coated with 120 nm of the spin-on-glass IC1-200 (Futurrex, Inc.) planarizing the sample. The immediate coating with the spin-on-glass minimized the oxidation of silver. Finally, a 50 nm layer of PMMA doped with IR-780 was spin coated from a solution with toluene. The dye concentration of the film was 20 mmol/l.

All spectral measurements were performed with a custom-built microscope. A 1-mW diode laser at a wavelength of 650 nm was used to pump the thin-film samples at an incidence angle of 57°. The fluorescence is collected at normal incidence by a microscope objective with a numerical aperture of 0.1 (corresponding to a collection angle of 6°). The objective collects the power of plane-wave components propagating close to normal incidence, and therefore, the collected power is proportional to the fluorescence enhancement factor $F$ as long as the collection angle of the objective is smaller than the width of the radiation pattern. Figure 2(a) demonstrates this to be true. Furthermore, the pump
respectively, we do not observe significant broadening. On the other hand, in the grating sample, we observe slight waviness of the surface (under atomic force microscope, images are not shown). The impact of this fabrication error is difficult to quantify as numerical simulations indicate that the waviness may increase or decrease the fluorescence enhancement depending on the exact shape of the PMMA film that is spincoated on top of a wavy spin-on-glass spacer.

We have found that the use of the LiF film to prevent metal-dye contact is very essential to preserve the fluorescence. We have fabricated similar samples but without the LiF film. The fluorescence intensity obtained with these samples was considerably smaller. For example, for the structure of Fig. 1(e), the measured enhancement was less than 10. We believe that, during the spincoating
of the polymer-dye film while the solution is still in fluid state, dye molecules diffuse to the metal surface and adhere to it as is commonly observed, e.g., in surface-enhanced Raman scattering studies.\textsuperscript{44,45} Hence, the LiF layer prevents fluorescence quenching of the molecules at the surface of the metal film.

We have verified the fabrication quality of our resonator sample [Fig. 4(c)]. Figure 5(a) presents an atomic force microscope image of the resonator sample. The image shows a 1 μm × 1 μm area. The surface is seen to be smooth save for a sparse distribution of small (approximately 30 nm in diameter) particles that presumably have small effect on the optical properties. Figure 5(b) shows the surface profiles along the two vertical red lines in Fig. 5(a). It can be seen that the surface roughness stays within a few nanometers. This verifies that depositing Au directly on PMMA without an adhesion layer yields good film quality. As for the grating sample of Fig. 4(d), the scanning electron microscope image in Fig. 5(c) confirms that the grating dimensions correspond to those of the design.

IV. CONCLUSIONS

We studied the intensity enhancement of far-field radiation of continuous large-area fluorescent samples by planar nanostructures. We have developed a theoretical framework for this effect and designed nanostructures that affect the entire sample instead of localized electric-field hot spots, utilizing the enhancements of emission directivity and optical pumping simultaneously. We fabricated and tested the structures experimentally. With a simple planar resonator structure, we demonstrated an exceptionally high far-field fluorescence enhancement factor of 120. Using a subwavelength grating, we obtained an enhancement factor of 6 for TM and 2 for TE polarization of the pumping light, allowing fluorescence intensity to be controlled by polarization.

As far-field radiation enhancement translates directly to an improved signal strength when collecting fluorescence light, it can be advantageous in many applications, such as frequency-shifting photodetection, detection of molecules, and chemical compounds via their fluorescence, as well as creation of bright spatially incoherent light sources for full-field optical coherence tomography,\textsuperscript{44} optical ghost-imaging microscopy,\textsuperscript{45} and other imaging techniques with such illumination requirements. We note that our selection of materials is only an example. The principles of large-area fluorescence enhancement that we put forward in this work, as well as the presented theoretical methods, can be applied also to other types of emitters, including quantum dots and other semiconductor light sources.

ACKNOWLEDGMENTS

M.N. acknowledges financial support from the Finnish Cultural Foundation (Grant No. 00170774). A.S. and M.K. acknowledge financial support from the Academy of Finland (Grant No. 308394). Part of the research was performed at the Micronova Nanofabrication Centre of Aalto University. This work has been partially supported by the University of Cologne through the Institutional Strategy of the University of Cologne within the German Excellence Initiative (QM$^3$).

APPENDIX: CALCULATION OF FLUORESCENCE ENHANCEMENT FACTOR

The fluorescence enhancement factor $F = F_p F_e$ is composed of the pump enhancement factor $F_p$ and the emission enhancement factor $F_e$. In the most general case, the emission enhancement factor can be defined (for a dipole emitter with a fixed orientation) as

$$F_e(r) = \frac{y_{\text{ff}}(r)}{y_{\text{opt}}(r) + y_{\text{nr}}},$$  \hspace{1cm} (A1)

where $y_{\text{ff}}(r)$ and $y_{\text{opt}}(r)$ are the rates of spontaneous emission into detected far-field optical modes and all available optical modes, respectively. The quantity $y_{\text{nr}}$ is the rate of de-excitation through nonradiative pathways such as internal conversion to nonfluorescent states. The quantities subscripted with a zero are the corresponding rates for the reference case of the emitter in a homogeneous environment. We identify $y_{\text{opt}}(r)/y_{\text{opt},0} = P(r)$ as the Purcell factor and $y_{\text{nr}}/y_{\text{nr},0} = \Gamma$ as the intrinsic loss coefficient of the emitter. The latter is related to the intrinsic quantum yield $\eta$ of the emitter as $\Gamma = 1/\eta - 1$. Therefore, $\Gamma$ is constant in our calculations; nonradiative decay modes through, e.g., fluorescence quenching at metal interfaces are included in the Purcell factor. Thus, the expression for $F_e(r)$ is simplified to

$$F_e(r) = \frac{y_{\text{ff}}(r)}{y_{\text{opt}}(r) + y_{\text{nr}}} \cdot \frac{1 + \Gamma}{P(r) + \Gamma},$$  \hspace{1cm} (A2)

For the samples that were composed of continuous thin films, we calculated the Purcell factor $P(r)$ by an analytical method. An electric dipole (that models the quantum emitter) is represented by the electric-current density distribution $J(r) = j_0(r)$, where $j$ is the electric-current amplitude directed along the dipole moment. To calculate $P(r)$, one must find the electric field at the position of the dipole, including the dipole’s self-field and the field back-scattered...
from the environment. We did this using a method of decomposition of electric currents and their fields into elementary waves. We calculate the back-scattered plane-wave electric-field amplitudes at the position of the dipole. In general, the emitter is located in a host film that is sandwiched between two stacks of thin films. The distances from the dipole to the boundaries of the host film are $d_1$ toward the first stack and $d_2$ toward the second stack. If the reflection coefficients of the two stacks are $\rho_1$ and $\rho_2$, then the plane-wave field amplitude at the emitter’s position will be

$$E = \tilde{E}_{em} e^{i k d_1} + \rho_1 e^{i k d_1} + 2 \rho_1 \tilde{E}_{bb} e^{i k d_1},$$

where $\tilde{E}_{em}$ is the emitted plane-wave amplitude obtained from the electric-current decomposition and $k$ is the component of the wave vector along the normal direction. The reflection coefficients were calculated using a transfer-matrix method. Finally, the plane-wave field amplitudes are summed together to obtain the back-scattered electric field $E_{bb}(r)$. The Purcell factor is then $P(r) = 1 - \text{Re}[\tilde{E}(r)/2\rho]$, where $\rho$ is the power radiated by the dipole in a homogeneous dielectric environment. We note that compared to alternative ways of calculating Purcell factors in layered media, this approach is advantageous if the host medium of the emitters is an anisotropic material, such as a liquid crystal, or even a spatially dispersive nanomaterial.

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