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Controlling light emission by a thermalized ensemble of colloidal quantum dots with a metasurface

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Abstract: We report an experimental and theoretical study of light emission by a patterned ensemble of colloidal quantum dots (cQDs). This system modifies drastically the emission spectrum and polarization as compared to a planar layer of cQDs. It exhibits bright, directional and polarized emission including a degree of circular polarization in some directions. We introduce a model of light emission based on a local Kirchhoff law which reproduces accurately all the features of the experiment. The model provides a figure of merit to assess quantitatively the emitted power. This work paves the way to the systematic design of efficient ultrathin light emitting metasurfaces with controlled polarization, spectrum and directivity.

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1. Introduction

The control of light emission from single emitters via resonators is a widespread topic in the scientific literature of the past 70 years. Theoretical concepts and tools have been developed as early as the 1940's, with Purcell's seminal paper on the modification of spontaneous emission probability at radio frequencies [1]. The process of cavity mediated emission has since then been extended to the visible domain of the electromagnetic spectrum [2]. Within the field of cavity quantum electrodynamics, milestones are the enhancement of spontaneous emission [3], its inhibition [4–6], the observation of the strong coupling regime through the signature of mode splitting [7] and the Rabi oscillation regime [8]. The concepts explored by cavity QED have been generalized in the community of nanophotonics with emitters interacting with other types of "cavities" such as antennas [9–11] or photonic crystals [12, 13], that can similarly tailor the electromagnetic environment to affect the properties of light emission. Among the properties that can be engineered through emitter-resonator interaction, one notes the spontaneous rate of emission [14–17], the directivity of emission [18–21], or the emission spectrum [22–26].

Demonstration of higher power and control of directivity at the single emitter level as well as the technical feasibility and reproducibility of fabrication processes naturally paves the way towards the design of light-emitting metasurfaces consisting of a large number of emitters embedded in a metasurface. Metasurfaces are based on planar arrays of antennas, whose modes are engineered to provide control over the spatial, spectral and polarization response. A survey of important contributions can be found in ref. [27] for light emission by fluorophores in metasurfaces and in ref. [28] for light extraction from semiconductors.

The purpose of this paper is to consider a particular case of metasurface consisting of an ensemble of colloidal quantum dots structured into a resonant dielectric grating. The system of

45 quantum dots is thus both an ensemble of incoherent quantum dots emitting light by spontaneous
46 emission and a resonant dielectric grating. We analyse how this system shapes the spectrum, the
47 directivity and the polarization of the emitted light.

48 When designing or analysing the interplay between single emitters and metasurfaces, the
49 standard approach outlined in ref. [27] is to consider that each emitter can be treated as a dipolar
50 emitter. Its environment is thus taken into account to analyse its emission, via, for instance, the
51 Purcell effect.

52 In our study, we are dealing with a metasurface, behaving as an extended resonator, and
53 fabricated using an ensemble of closely packed QDs. If the emitters have a broad emission
54 spectrum, an environment with narrower resonances may modify significantly the emission
55 spectrum [29]. This modification is position-dependent in a resonant metasurface. One could
56 thus first imagine to model the light emitted from the ensemble of emitters as an average of the
57 emission pattern of single emitters over position and dipole moment orientation. Nevertheless,
58 this approach leads to particularly lengthy calculations and misses a crucial point: in such a
59 system, the emitters are both sources but are also the very material that the resonator consists of,
60 shaping the environment that the emitters radiate into. The optical properties of the quantum dot
61 layer must be accurately accounted for. Additionally, the absolute amplitude of light emitted by
62 cQDs remains unknown when treated as single dipolar emitters: the "single emitter" approach
63 does not provide an upper bound of the emitted power.

64 The generalized Kirchhoff's law approach enables to overcome the previous limitations [30].
65 It is based on the description of the emission by first calculating the absorption features of the
66 structure. Under this formalism, ensemble of emitters are described not as randomly positioned
67 and randomly oriented dipoles, but as forming a layer with optical properties affecting the mode
68 structure of the system. Since the model is based on the local absorptivity within the structure,
69 an upper bound for absorption (and therefore for emission) can be assessed by maximizing the
70 absorptivity.

71 All in all, the generalized Kirchhoff's law approach is able to adequately model the interplay
72 between a metasurface and an *ensemble* of emitters, and explains all the resonance effects and
73 features observed experimentally, as will be shown in the main part of this paper. In this article,
74 we use this model to discuss our observations and analyze the data. We present the experimental
75 data in the next section. The third section is devoted to the introduction of the model, and we
76 compare data with theory in the fourth section.

77 2. Experiments

78 *System description*

79 We study in this paper the emission of light by the system depicted in Fig. 1. The major feature
80 of the structure is its top 1D quantum-dot grating (QDG). This grating was fabricated using a
81 template-stripping technique. In a first step, a dispersion of CdSe based cQDs was drop-casted
82 onto a silicon template in which the negative shape of the grating was etched away [31, 32]. After
83 evaporation of the solvent, a SiO₂ glass slide was attached with epoxy adhesive to the back of the
84 QDG. In a final step, the whole stack was removed from the silicon template using mechanical
85 cleavage.

86 The cQDs used in the experiment were synthesized in the laboratory, following procedures
87 and methods reported in previous works of the authors [31]. They consist of a CdSe core, inside
88 a CdS/ZnS shell (2 monolayers + 2 monolayers), for a total diameter of around 10 nm. This sets
89 the absorption peak maximum around 620 nm.

90 The whole structure is therefore made of a dense ensemble of cQDs whose solvent has
91 evaporated. It consists of the grating itself, with alternating lines and grooves with a groove
92 depth of 50 nm and a duty cycle of 50%. The period of the structure is denoted as a , and its
93 value taken as 400 nm or 450 nm depending on the sample under study. The grating lies atop a

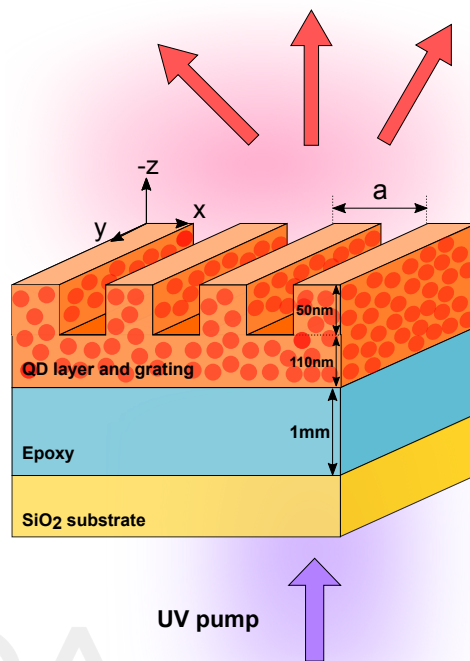


Fig. 1. **Schematic view of the QDG system.** The whole QDG is patterned on a SiO₂ substrate. The top layer, comprising both the grating and the supporting slab, is made of the very same closely packed quantum dots, left as a dense ensemble of emitters after evaporation of the solvent. The system is pumped by a UV LED illuminating the structure from the backside, via the substrate.

110 nm thick uniform layer of colloidal quantum dots (cQDs) with an absorption peak around 620 nm. Details regarding the cQDs and the fabrication process of the QDG are also available in the Supplemental Document.

The photoluminescence (PL) properties, and in particular, the spatial and spectral features of the light emission by such a system have been characterized experimentally using the setup depicted in Fig. 2. The cQD layer was optically excited by a UV LED illuminating the back of the sample. A microscope objective focuses the beam through the substrate and epoxy onto the cQD layer. The PL is then collected and collimated by another microscope objective facing the grating.

In this study, we are interested in getting information on three characteristics of light emission: spectrum, directivity, and polarization. All the information we are looking for is accessible by performing Fourier imaging and using a spectrometer.

2.1. PL intensity map

In a first set of experiments, we focus on spectral and angular features. We use appropriate optics to image the Fourier plane of the collection objective onto the input slit of a spectrometer. This slit selects a narrow area of the Fourier plane that corresponds to a wide range of emission directions in the grating direction, that is the x axis. Dispersion by the spectrometer grating forms an image on the CCD camera sensor that corresponds to a PL map of the system with respect to both wavelength and direction of emission.

Results are depicted on Fig. 3a, showing the total PL intensity as a function of wavelength

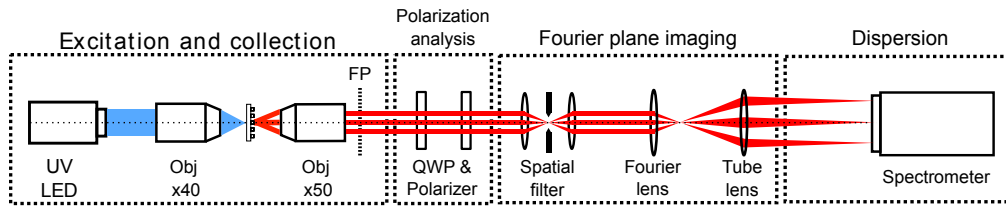


Fig. 2. **Schematic depiction of the experimental setup.** The setup uses a microscope to illuminate the back of the sample with UV light. The light is collected by a microscope objective with large numerical aperture (NA=0.8). The whole apparatus allows switching between direct imaging and back focal plane (hereafter called Fourier plane) imaging by the insertion of a Fourier lens. The Fourier plane can also be imaged on the input slit of a spectrometer. In this configuration, the image formed on the spectrometer detector is a wavevector versus wavelength plot of the PL collected from the excited sample.

114 and emission angle $\sin(\theta) = ck_x/\omega$, for a sample of period $a=400$ nm. Different features can be
 115 observed on this map. The most prominent feature is arguably a bright cross-shaped pattern,
 116 formed by the intersection of two branches. A second cross, fainter and blue-shifted, is also
 117 visible. The brightest spot of the main cross is located at the close to the crossing point, indicating
 118 that the maximum of emitted intensity occurs at a direction normal to the surface of the QDG, for
 119 a wavelength around 648 nm. More generally, the bright cross feature can be interpreted in terms
 120 of narrowband and directional emission by plotting vertical cross sections of the map. When
 121 one considers emission away from the crossing point, emission takes place mainly in two lobes,
 122 symmetric with respect to the normal direction.

123 The whole pattern is superimposed on a faint isotropic emission background ranging from
 124 610 nm to 670 nm. This emission has its maximum at 640 nm, and is consistent with the PL
 125 emission spectrum from a bare layer of cQDs. This is shifted from the position of the absorption
 126 maximum of a bare layer of cQD, located at around 620 nm (see Supplemental Document).
 127 Directional emission along the cross branches is experimentally observed up to a wavelength of
 128 690 nm where the background emission is below the noise level.

129 At 645nm, around 8% of the total emission within the objective's NA comes from the dispersion
 130 branches and represent funneled in the directional mode. This value goes up to 15% when
 131 looking at 660 nm, away from the center of the pattern, where the background emission has
 132 decreased more significantly than the emission in the cross branches. Thus, the contrast between
 133 the directional emission and the isotropic background tends to increase, thanks to this slight
 134 difference in spectral evolution of the two patterns. The luminance of the cross is 7 to 8 times
 135 larger than the luminance of the background at 645 nm, while this factor reaches 10 to 11 at
 136 660 nm. Additionally, one can note that the percentage of emission coming from the directional
 137 mode would be much higher if a lower NA objective is chosen, as it would result in a significant
 138 filtering of the background emission.

139 *Fourier imaging*

140 Fourier imaging allows for a representation of directivity and polarization features of the spectrally
 141 integrated emission. Fig. 3b depicts intensity in the Fourier plane. The picture shows that
 142 most of the intensity is emitted along two arcs with a peak at normal incidence. An analysis of
 143 the polarization features in the Fourier plane is performed using waveplates and polarizers to
 144 measure the Stokes parameters (S_0, S_1, S_2, S_3) of the emitted light. S_0 is the total intensity. The
 145 measurement of S_0 normalized by its maximum value is displayed in Fig. 3b. S_1/S_0 is displayed
 146 on Fig 3c, and is obtained by subtracting the two intensity maps measured when placing a

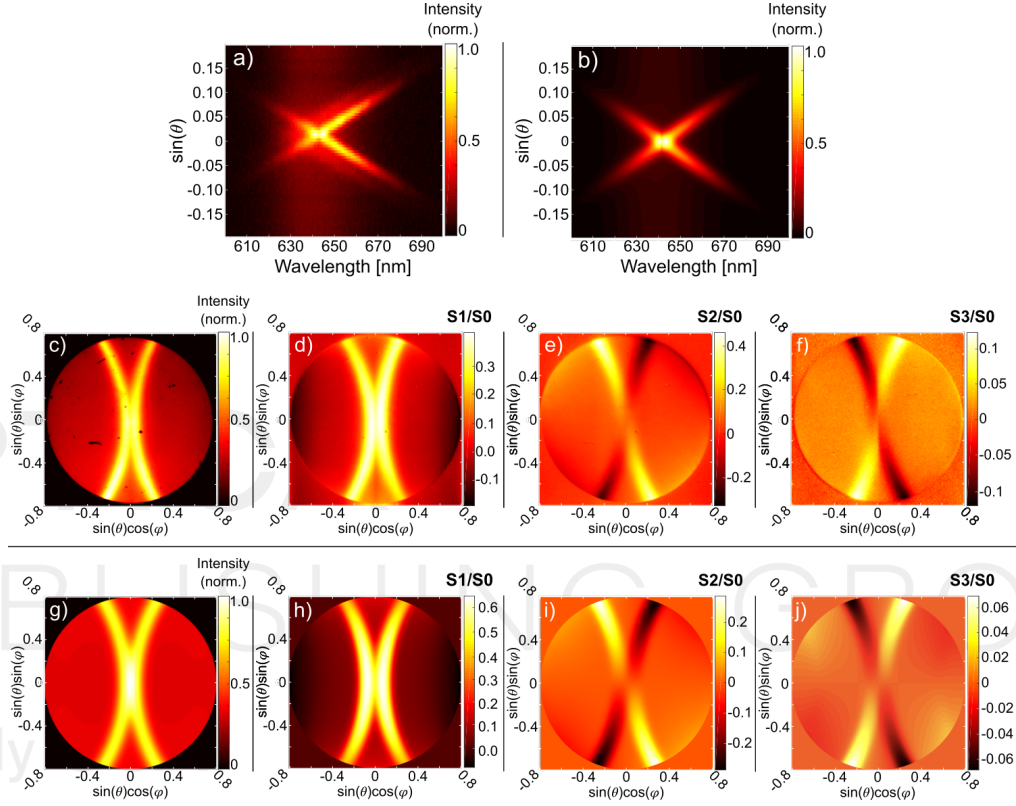


Fig. 3. Intensity, directivity and polarization characteristics of the QDG system PL for a period $a=400$ nm. **a)** PL intensity plotted as a function of wavelength (horizontal axis) and emission direction (vertical axis), as experimentally measured. **b)** Numerical simulations of the PL intensity following the generalized Kirchhoff's modelling procedure. **c)-f)** Stokes vector components S_0 , S_1/S_0 , S_2/S_0 , and S_3/S_0 of the emitted light, plotted as colormaps in the Fourier plane. These components fully represent the polarization state of the light emitted in all directions by the QDG system. The different maps are obtained by measuring the intensity distribution in the Fourier plane for different orientations of the quarter wave plate and polarizers, used for polarization analysis (see the experimental setup of Fig. 2). **g)-j)** Numerical simulations of the Stokes vector components, following the generalized Kirchhoff's modelling procedure.

147 polarizer with its axis either parallel or perpendicular to the grating grooves. Similarly, S_2/S_0
148 is obtained when placing the polarizer at $+45^\circ$ and -45° with respect to the grooves orientation (see
149 Fig. 3d). Finally, The degree of circular polarization S_3/S_0 is shown in Fig. 3e. The parameter
150 S_3 is obtained by subtracting the two intensity maps measured with a right and a left circular
151 polarizers formed by combining a polarizer and a quarter wave plate.

152 The observation of a degree of circular polarization may be surprising as its generation is
153 usually due to chiral samples. However, optical activity has been reported with non-chiral
154 metasurfaces illuminated in oblique incidence [33]. The behaviour observed in our work has the
155 same properties that in ref. [33]: dichroism is not seen at normal incidence; it changes sign in
156 opposite directions; it takes place close to a mode resonance.

157 The experimental observations show unambiguously that the structure under study is able to
158 drastically modify light emission, causing enhanced and directional outcoupling of the emission
159 and changes in the emission spectrum. Finally, light emission is strongly polarized and may even
160 be circularly polarized in some emission directions. In the next section, we discuss qualitatively
161 the physical process responsible for these effects.

162 *Qualitative interpretation of the PL features*

163 In this section, we show that the features of the PL can be interpreted using the mode structure of
164 the system. In our experiment, the cQDs form a slab with an effective refractive index much
165 larger than 1 so that guided modes are supported by the slab. The cQDs are pumped by the
166 UV light coming from the bottom of the structure so that they also behave as light emitters.
167 The emitters can relax by spontaneous emission: they can excite either the slab modes or the
168 radiative modes. In summary, the cQDs have two roles: on one hand, they form a corrugated
169 planar waveguide which supports leaky modes; on the other hand, they behave as fluorophores
170 embedded in the waveguide.

171 Without corrugation, light emission is only due to radiative modes. It is spatially incoherent,
172 and provides an isotropic, unpolarized background. In the presence of a periodic corrugation,
173 the slab modes are leaky modes so that light coupled to the slab mode is then emitted in a
174 well-defined direction. Thus, despite the fact that all emitters are incoherent, the system generates
175 spatially coherent (directional) emission. The origin of the spatial coherence of the field along
176 the surface is the propagation of the leaky mode. The spatial coherence length is thus dictated by
177 the decay length of the leaky mode [34, 35].

178 According to this mechanism, the emission should have the polarization of the leaky mode. This
179 can be easily checked by analysing the polarization of the light emitted in a plane perpendicular
180 to the grooves edges. We select light propagating in this plane by using the spectrometer slit
181 to collect components in the $k_y = 0$ direction of the Fourier plane (defining the Oxz plane of
182 incidence) and disperse them with the grating. The Fig. 4a provides PL maps measured as
183 function of $\sin(\theta)$, corresponding to momentum k_x in the plane $k_y = 0$, and as a function of the
184 wavelength. When placing a polarizer along the y direction, along the grooves, we select the TE
185 polarization. When placing the polarizer along the x direction, we select the TM polarization.
186 This was also confirmed by adding a half-wave plate after the linear polarizer. This rotates
187 together with the linear polarizer to ensure a constant polarization orientation of the light reaching
188 the spectrograph to remove any polarization dependence of the spectrometer's components. It
189 can be observed non-ambiguously that the bright cross shape is associated to emission in the TE
190 polarization, while the isotropic background remains unpolarized. A faint secondary cross can
191 be observed in the orthogonal polarization.

192 We now discuss the emission pattern observed in Fig. 3b. Given the rotational symmetry of a
193 plane, the dispersion relation of a guided mode at a given frequency has the form $k_{\text{gm}} = n_{\text{eff}}\omega/c$
194 where n_{eff} is the effective refractive index. The corresponding dispersion relation in the plane
195 (k_x, k_y) is a circle whose radius is larger than ω/c so that it is not coupled to the far-field. This

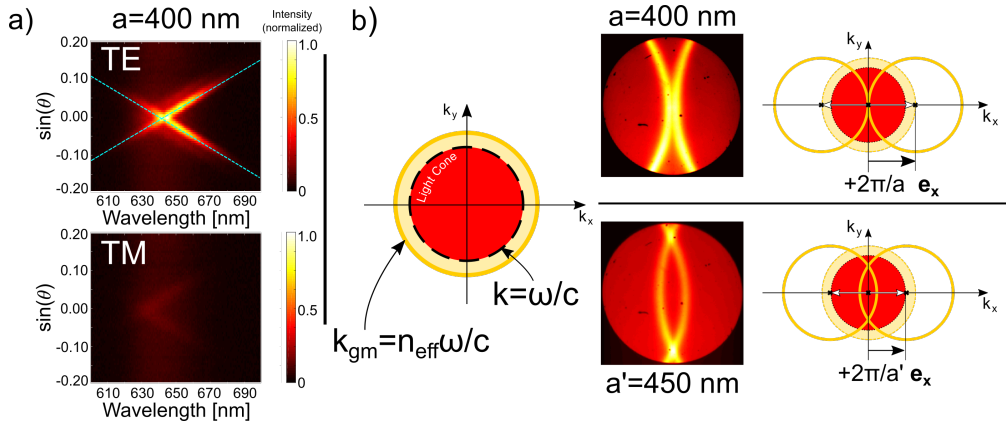


Fig. 4. **Qualitative interpretation of the experimental features in relation to the modal structure of the sample.** **a)** PL maps of the QDG structure for TE (top) and TM (bottom) polarization for $a=400$ nm. Dashed lines are plots of the dispersion relation of the guided mode for TE polarization, displaced by the grating into the light cone. **b)** Schematic description of the Fourier plane light distribution. When considered individually, the guided-mode propagation constant lies beyond the limit of the light line. The top lamellar grating couples the guided mode to modes $\mathbf{k} = \mathbf{k}_{\text{gm}} \pm \frac{2\pi}{a} \mathbf{e}_x$ corresponding to circles translated by $\pm \frac{2\pi}{a} \mathbf{e}_x$. The result is displayed for two different grating periods, 400 nm and 450 nm.

196 is where the grating comes into play: the guided mode dispersion relation is folded back into the
 197 light cone according to $\mathbf{k} = \mathbf{k}_{\text{gm}} \pm \frac{2\pi}{a} \mathbf{e}_x$ so that the dispersion relation is now given by two circles
 198 centered at $\pm \frac{2\pi}{a} \mathbf{e}_x$ as sketched in Fig. 4b. The branches of the circle that are inside the light
 199 cone become leaky and can be measured as shown in Fig. 4b for two different grating periods.

200 3. Light emission by a thermalized ensemble of quantum dots

201 The modelling of light emission by the system must include two key ingredients:

- 202 1. the emission spectrum by a homogeneous thermalized slab of emitters and
- 203 2. the emission through a resonator whose geometry and optical properties are influenced by
 204 the emitters themselves.

205 As discussed in ref. [27], the first step can be accounted for by using the emission spectrum of
 206 the fluorophore and the second step by computing light emission by a dipole placed in a given
 207 environment. We note that the refractive index of the environment may change under pumping
 208 so that both effects cannot be included independently under strong pumping.

209 A self-consistent model for light emission under pumping has been widely used for semicon-
 210 ductors. It is based on Kirchoff's law [36, 37]. The emitted power is given as the product of
 211 the absorptivity by the semiconductor and the blackbody radiance. The pumping intensity is
 212 accounted for by including what is known as the photon chemical potential μ in the Bose-Einstein
 213 distribution [37] and by using the refractive index in the presence of pumping to compute the
 214 absorptivity. In the case of an LED, μ is given by the difference of the quasi-Fermi levels in the
 215 conduction band and valence band which is equal to eV where e is the electron charge and V is
 216 the applied voltage. The absorptivity depends on the occupation of the electronic states given by
 217 the Fermi-Dirac distribution of the two bands. The Kirchoff model has enabled the design of
 218 very efficient silicon light-emitting diodes [38].

219 In the case of an LED, all emitting points have an equivalent environment. This is not the case
 220 for a metasurface which may have several localized resonances at different points and different
 221 frequencies. Hence, a local form of the Kirchhoff law is needed. It has been introduced in
 222 ref. [30] where a local absorption rate $\alpha(\mathbf{r})$ was introduced. The method has been applied to
 223 analyse the photoluminescence and electroluminescence of a layer of PbS cQDs on a periodic
 224 array of gold patches [39]. The Kirchhoff method assumes that the emitting system is thermalized.
 225 A discussion of the thermalisation of an ensemble of PbS cQDs as a function of the ligand size
 226 has been reported recently [40]. In the rest of the paper, we use this approach to model light
 227 emission by the QDG system. According to ref. [30], the power radiated in a specific direction θ ,
 228 at a wavelength λ and in a given polarization $i = \text{TE}$ or $i = \text{TM}$ can be cast in the form:

$$P_e(\lambda, \theta, i, \mu) = \iiint_V \alpha_{abs}(\lambda, \theta, i, \vec{r}, \mu) dV \frac{2hc^2}{\lambda^5} \frac{1}{\exp(\frac{hc}{k_B T \lambda} - \frac{\mu}{k_B T}) - 1}. \quad (1)$$

229 where k_B is Boltzmann constant, T is the temperature and $\alpha_{abs}(\lambda, \theta, i, \vec{r}, \mu)$ is a local rate of
 230 absorption. Let us emphasize that the absorption involved here is the absorption of the material
 231 under UV excitation so that the populations are out of equilibrium. This term contains the entire
 232 information on the mode geometry of the system. The second term of the expression describes
 233 the emission spectrum of the cQD layer considered as an ensemble of thermalized emitters. For
 234 $\frac{hc}{\lambda} \gg \mu$, we can replace the Bose-Einstein distribution by its Wien approximation:

$$P_e(\lambda, \theta, i, \mu) = \iiint_V \alpha_{abs}(\lambda, \theta, i, \vec{r}, \mu) dV \frac{2hc^2}{\lambda^5} \exp(-\frac{hc}{k_B T \lambda}) \exp(\frac{\mu}{k_B T}). \quad (2)$$

235 Thus, the calculation of the emitted power can be performed by numerically computing *absorption*
 236 of incoming light within the cQD layer, instead of calculating the power *radiated* by ensembles of
 237 radiating dipoles placed in the cQD slab. To proceed, we need to know the refractive index of the
 238 material under pumping. We emphasize that computing light emission from the knowledge of the
 239 absorption spectrum requires a great accuracy of the imaginary part of the refractive index due to
 240 the exponentially varying term $\exp(-\frac{hc}{k_B T \lambda})$. A method to measure the permittivity of TDBC
 241 J-aggregated molecules (5,5',6,6'-tetrachloro-1,1'-diethyl-3,3'-di(4-sulfobutyl)-benzimidazolo-
 242 carbo cyanine) in order to reproduce photoluminescence measurements has been reported
 243 recently [41]. In this work, we used the emission data to identify the permittivity of the effective
 244 medium made of the assembly of cQDs.

245 4. Numerical modelling and results

246 The field distribution in the cQD slab, and the subsequent local absorption, depends on the
 247 geometry of the whole structure. We study the structure in the spectral range [600 nm, 700 nm].
 248 We detail in the Supplemental document the procedure used to model the refractive indices of
 249 the different layers. We consider a duty cycle $f = 50\%$ for the top QDG in contact with air. The
 250 absorptivity is computed using a rigorous coupled-wave approximation (RCWA) method [42].
 251 The method gives the reflected and transmitted field amplitudes for TE and TM incident plane
 252 waves of arbitrary angle and frequency. The absorption is then derived from energy conservation.

253 The results of the numerical simulations of the PL, normalized by the maximum intensity,
 254 are plotted in Fig. 3g. The numerical simulations of the Stokes vector components normalized
 255 by S_0 are shown in panels Fig. 3h to Fig. 3j. To allow a direct comparison between the
 256 simulations (that represent light emitted by the system) and experimental measurements (that
 257 represent light collected by the detector at the end of the experimental setup), some experimental
 258 parameters were taken into account: the instrumental response and point spread function was
 259 included in our model, as well as an unbalance between responses in TE and TM polarizations.

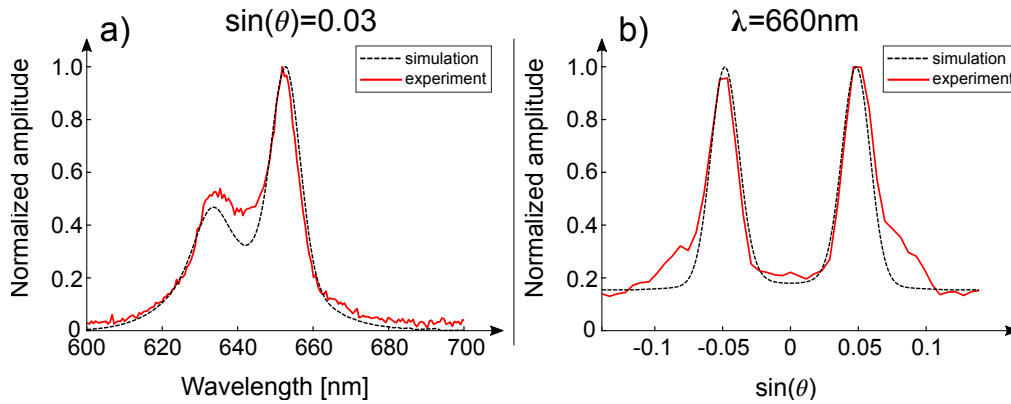


Fig. 5. **PL map cross-sections and comparison between experimental data and results obtained with the thermalized emitters model.** **a)** Spectral profile of light emitted at $\sin \theta = 0.03$. **b)** Emission diagram at $\lambda = 660$ nm. All plots are normalized to their respective maximum intensity.

260 Furthermore, to show quantitatively the differences between experimental measurements and
 261 numerical simulations, we plot different cross sections (experimentally measured from Fig. 3
 262 and numerically simulated from Fig. 3f) selecting a specific emission direction (Fig. 5a, with
 263 $\sin \theta = 0.03$) or a specific wavelength (Fig. 5b, with $\lambda = 660$ nm).

264 The numerical results reproduce all major features observed on the plot: directional and
 265 spectrally narrow emission over a faint isotropic background, as well as all the polarization
 266 characteristics. Small deviations are noticeable in the relative heights of emission peaks from
 267 Fig. 5a. Those deviations are likely to be connected to experimental artifacts such as uncorrected
 268 spectral response and impulse response of the whole instrumentation chain, as well as residual
 269 deviations in the refractive index model. One can also note broader emission lobes on Fig. 5b. The
 270 lobes are formed by the contribution of the faint remaining cross pattern in the TM polarization
 271 (see Fig 4a), which is slightly offset with the bright cross pattern of the TE configuration.

272 5. Conclusion

273 In this paper, we studied the light emitted by an ensemble of quantum dots arranged in a
 274 periodically corrugated slab which can support leaky modes. When the system was pumped in
 275 the UV range, strongly polarized, spectrally narrow and directional light emission was observed.
 276 A local form of Kirchhoff's law has been used to model the system photoluminescence. This
 277 approach involves the calculation of the absorptivity of the structure. It is well suited for the design
 278 of light sources with specified properties involving dense assemblies of thermalized emitters.
 279 We note that such a model is able to account for both the electromagnetic properties of the
 280 whole resonator architecture surrounding the emitter, and for modification of the electromagnetic
 281 properties of the emitter itself, through modifications of its imaginary permittivity induced
 282 by pumping. Therefore, this model can also accurately predict light emission by systems
 283 experiencing progressively induced transparency of the emitter layer that are observed when
 284 increasing the pump power.

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289 **Author contributions.** JJG and EDL conceived the project. HM, ALP and EB performed modelling
 290 simulations. EDL performed samples design, fabrication, and characterization with input from FP and DN.

291 AR assisted in samples preparation. JJG and BV supervised the project. HM and BV wrote the original draft
292 of the manuscript. All authors contributed to the discussion of the results and to the revision of the paper.

293 **Disclosures.** The authors declare no conflicts of interest.

294 **Data availability.** Data underlying the results presented in this paper are not publicly available at this time
295 but may be obtained from the authors upon reasonable request.

296 **Supplemental document.** See Supplement 1 for supporting content regarding materials and methods.

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