Supporting Information

Coherent Fluorescence Emission by Using Hybrid Photonic-Plasmonic Crystals

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The details of the toy model.

Consider *N* emitters located on a surface or inside a very thin layer, as shown in Figure 1a. For simplicity, the scalar approximation is assumed. U_n^0 corresponds to the field of the *n*-th emitter close to the surface (near field approximation). We assume that there is no correlation between any two emitters, which is $W(r_n, r_m) = \langle U_n^{0*} U_m^0 \rangle = 0$, if $n \neq m$.

When the surface or the thin layer supports propagating quasi-two-dimensional optical mode, such as surface plasmon or slab-guided mode, the cross spectral density between the fields around two different locations could be nonzero, because the field U_i at r_i , is contributed by all the emitters, as

$$U_i = \sum_N \frac{1}{\sqrt{2\pi x_{ni}}} U_n^0 e^{iK_S x_{ni}}$$

where $K_s = K_s^r + iK_s^i$ is the wavevector of the mode, and $x_{ni} = x_{in} = |r_n - r_i|$.

The cross spectral density of the fields between two locations, is

$$W(r_{i},r_{j}) = \left\langle U_{i}^{*}U_{j} \right\rangle = \left\langle \left\{ \sum_{n \in [1,N]} \frac{1}{\sqrt{2\pi x_{ni}}} U_{n}^{0*} e^{-iK_{S}^{*} x_{ni}} \right\} \left\{ \sum_{m \in [1,N]} \frac{1}{\sqrt{2\pi x_{mj}}} U_{m}^{0} e^{iK_{S}^{*} x_{mj}} \right\} \right\rangle.$$

,

Considering the fact of $\langle U_n^{0*}U_m^0 \rangle = 0$ and assuming $\langle U_n^{0*}U_n^0 \rangle = \langle U_m^{0*}U_m^0 \rangle = S^0$,

$$W(r_{i}, r_{j}) = \sum_{N} \frac{1}{2\pi \sqrt{x_{ni} x_{nj}}} \langle U_{n}^{*} U_{n}^{*} \rangle e^{-S nj} = S$$
$$= \frac{S^{0}}{2\pi} \sum_{N} \frac{1}{\sqrt{x_{ni} x_{nj}}} e^{-K_{S}^{i}(x_{ni} + x_{nj})} e^{iK_{S}^{r}(x_{nj} - x_{ni})}$$

To obtain the spatial correlation function, the self cross spectral density is also needed, $S = W(r_i, r_i) = \frac{S^0}{2\pi} \sum_N \frac{1}{x_{ni}} e^{-2K_S^i x_{ni}} .$

Therefore,

$$\Gamma(r_{i},r_{j}) = \frac{W(r_{i},r_{j})}{\sqrt{W(r_{i},r_{i})W(r_{j},r_{j})}} = \frac{\sum_{N} \frac{1}{\sqrt{x_{ni}x_{nj}}} e^{-K_{S}^{i}(x_{ni}+x_{nj})} e^{iK_{S}^{r}(x_{nj}-x_{ni})}}{\sum_{N} \frac{1}{x_{ni}} e^{-2K_{S}^{i}x_{ni}}}$$

Assuming our system to be symmetric,

$$\begin{split} &\Gamma(r_{i},r_{j}) = \frac{\sum_{N} \frac{1}{\sqrt{x_{ni}x_{nj}}} e^{-K_{3}^{L}(x_{ni}+x_{nj})} e^{iK_{3}^{r}(x_{nj}-x_{ni})}}{\sum_{N} \frac{1}{x_{ni}} e^{-2K_{3}^{L}x_{ni}}} \\ &= \frac{\frac{1}{2} \left\{ \sum_{N} \frac{1}{\sqrt{x_{ni}x_{nj}}} e^{-K_{3}^{L}(x_{ni}+x_{nj})} e^{iK_{3}^{r}(x_{nj}-x_{ni})} + \sum_{N'} \frac{1}{\sqrt{x_{n'}x_{n'j}}} e^{-K_{3}^{L}(x_{n'}+x_{n'j})} e^{iK_{3}^{r}(x_{n'j}-x_{n'j})} \right\}}{\sum_{N} \frac{1}{x_{ni}} e^{-2K_{3}^{L}x_{ni}}} \\ &= \frac{\frac{1}{2} \left\{ \sum_{N} \frac{1}{\sqrt{x_{ni}x_{nj}}} e^{-K_{3}^{L}(x_{ni}+x_{nj})} e^{iK_{3}^{r}(x_{nj}-x_{ni})} + \sum_{N} \frac{1}{\sqrt{x_{nj}x_{ni}}} e^{-K_{3}^{L}(x_{nj}+x_{nj})} e^{iK_{3}^{r}(x_{nj}-x_{nj})} \right\}}{\sum_{N} \frac{1}{x_{ni}} e^{-2K_{3}^{L}x_{ni}}} \\ &= \frac{\frac{1}{2} \left\{ \sum_{N} \frac{1}{\sqrt{x_{ni}x_{nj}}} e^{-K_{3}^{L}(x_{ni}+x_{nj})} (e^{iK_{3}^{r}(x_{nj}-x_{ni})} + e^{iK_{3}^{r}(x_{ni}-x_{nj})}) \right\}}{\sum_{N} \frac{1}{x_{ni}} e^{-2K_{3}^{L}x_{ni}}} \\ &= \frac{\frac{1}{2} \left\{ \sum_{N} \frac{1}{\sqrt{x_{ni}x_{nj}}} e^{-K_{3}^{L}(x_{ni}+x_{nj})} (e^{iK_{3}^{r}(x_{nj}-x_{ni})} + e^{iK_{3}^{r}(x_{ni}-x_{nj})}) \right\}}{\sum_{N} \frac{1}{x_{ni}} e^{-2K_{3}^{L}x_{ni}}} \\ &= \frac{\sum_{N} \frac{1}{\sqrt{x_{ni}x_{nj}}} e^{-K_{3}^{L}(x_{ni}+x_{nj})} \cos[K_{3}^{r}(x_{nj}-x_{ni})]} {\sum_{N} \frac{1}{x_{ni}}} e^{-2K_{3}^{L}x_{ni}}} \\ &= \frac{\sum_{N} \frac{1}{\sqrt{x_{ni}x_{nj}}} e^{-K_{3}^{L}(x_{ni}+x_{nj})} \cos[K_{3}^{r}(x_{nj}-x_{ni})]} {\sum_{N} \frac{1}{x_{ni}}} e^{-2K_{3}^{L}x_{ni}}} \\ &= \frac{\sum_{N} \frac{1}{\sqrt{x_{ni}x_{nj}}} e^{-K_{3}^{L}(x_{ni}+x_{nj})} (e^{iK_{3}^{r}(x_{nj}-x_{ni})} + e^{iK_{3}^{r}(x_{ni}-x_{nj})})} {\sum_{N} \frac{1}{x_{ni}}} e^{-2K_{3}^{L}x_{ni}}} \\ &= \frac{\sum_{N} \frac{1}{\sqrt{x_{ni}x_{nj}}} e^{-2K_{3}^{L}x_{ni}}} {\sum_{N} \frac{1}{x_{ni}}} e^{-2K_{3}^{L}x_{ni}}} \\ &= \frac{\sum_{N} \frac{1}{\sqrt{x_{ni}x_{nj}}} e^{-2K_{3}^{L}x_{ni}}} {\sum_{N} \frac{1}{x_{ni}}} e^{-2K_{3}^{L}x_{ni}}} \\ &= \frac{\sum_{N} \frac{1}{\sqrt{x_{N}x_{N}}} e^{-2K_{N}^{L}x_{Ni}}} {\sum_{N} \frac{1}{x_{N}}} e^{-2K_{N}^{L}x_{Ni}}} \\ &= \frac{\sum_{N} \frac{1}{\sqrt{x_{N}x_{N}}} e^{-2K_{N}^{L}x_{Ni}}} {\sum_{N} \frac{1}{x_{N}}} e^{-2K_{N}^{L}x_{Ni}}} \\ &= \frac{\sum_{N} \frac{1}{\sqrt{x_{N}x_{N}}} e^{-2K_{N}^{L}x_{Ni}}} {\sum_{N} \frac{1}{x_{N}}} e^{-2K_{N}^{L}x_{Ni}}} \\ &= \frac{\sum_{N} \frac{1}{\sqrt{x_{N}x_{N}}} e^{-2K_{N}^{L}x_{Ni}}} {\sum_{N} \frac{1}{x_{N}}} e^{$$

The final spectral degree of coherence

$$\Gamma(r_{i},r_{j}) = \frac{W(r_{i},r_{j})}{\sqrt{W(r_{i},r_{i})W(r_{j},r_{j})}} = \frac{\sum_{N}^{N} \frac{1}{\sqrt{x_{nj}x_{ni}}} e^{-K_{S}^{i}(x_{ni}+x_{nj})} \cos[K_{S}^{r}(x_{nj}-x_{ni})]}{\sum_{N} \frac{1}{x_{nj}} e^{-2K_{S}^{i}x_{ni}}}$$
(1)

in general could be non-zero. Note that in the derivation, the cross term $\langle U_n^{0*}U_m^0 \rangle$ is zero for different emitters and has no contribution to Equation 1. It corresponds to the premise that each spontaneous emission event is independent, in other words, correlations between emitters is not the reason for the obtained coherence. The results shown in Figure 1b are calculated based on Equation 1.



Fig. S1. Reflection spectra of the proposed structure as functions of wavelength and incident angle. Light is incident along the Γ -X direction. The left (right) panel is for the p (s) polarized incident light.



Fig. S2. (a) The emission enhancement for S101 fluorescent molecules on top of the structure as a function of wavelength and emission angle. (b) The Young's double-slit experimental results for the case of a 7 μ m double-slit distance on the sample. (c) The same as (b) but for the case of a 4 μ m double-slit distance on the sample. The emission light is detected along the Γ -X direction. The upper (lower) panels are for p (s) polarized emission light.



Fig. S3. The Young's double-slit experimental results of the emission without the structure for the case of a 7 μ m double-slit distance on the sample. For the fluorescence on top of the flat metallic film case, there are two parts of fluorescence emission, one part of the emission can couple to the surface plasmon modes via near field coupling, the other part of the emission does not couple to the surface plasmon modes and directly emits to the far field. For the directly emission part, there is certainly no spatial coherence properties in the far field. For the part coupled to the surface plasmon, as we explained in the toy model (shown in the Fig. 1 and supporting information), there is indeed spatial coherent properties in the near field mediated by the surface modes, however, one can not observe it in the far field. Furthermore, due to the imperfections on the metallic film, the coupled surface plasmon modes can re-radiate to the far field, but they do not contribute in this way to coherence. The reason is that the scattering by the randomly distributed imperfections is a non-coherent process. As we discussed in the MS, to maintain those near-field coherent properties, coherent scattering process by such as grating structures is highly required.



Fig. S4. (a) Reflection spectra of the PS spheres array on top of a glass substrate as functions of wavelength and incident angle. Here, the reflection directly from a bare glass substrate is used to be the reference. (b) The emission spectra for S101 fluorescent molecules on top of the PS spheres array on a glass substrate as a function of wavelength and emission angle. (c) The Young's double-slit experimental results for the case of a 7 μ m double-slit distance on the sample. The upper (lower) panels are for p (s) polarized light case. Light is incident along the Γ -J direction.



Fig. S5. The results of the single (a and b) and double (c) slit experiments for p-polarized emission light. In a and b, one slit (black part) is blocked. Here the double-slit distance and the width of the single slit are 7 μ m and 2 μ m on the sample. In d, the double-slit is removed from the experiment. The yellow dashed line shown in the figure corresponds to the gap position appearing around the Γ point.



Fig. S6. Polar plot of the emission around 597 nm (red) and 610 nm (black). The upper (lower) panel corresponds to the p (s) polarized emission light from the structure.