Supplemental Material: Collectively enhanced chiral photon emission from an atomic array near a nanofiber

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MASTER EQUATION TERMS CALCULATION

The interaction coefficients can be calculated as

$$V_{ij} = -\mathcal{P}\sum_{\nu} \left[\frac{G_{\nu i} G_{\nu j}^*}{\omega - \omega_{\rm a}} + (-1)^{\delta_{ij}} \frac{\tilde{G}_{\nu i}^* \tilde{G}_{\nu j}}{\omega + \omega_{\rm a}} \right], \quad (1)$$

and

$$\Gamma_{ij} = 2\pi \sum_{\nu} G_{\nu i} G_{\nu j}^* \delta(\omega - \omega_a).$$
⁽²⁾

Here, \mathcal{P} denotes the Cauchy principal value, δ_{ij} denotes the Kronecker delta function, and $\sum_{\nu} = \sum_{g} + \sum_{u}$ is a generalised sum over guided and unguided modes. The sum over the guided modes reads $\sum_{g} = \int_{0}^{\infty} d\omega \sum_{fl}$, where ω is the mode frequency, and $l = \pm 1$ denotes counterclockwise or clockwise polarization. Finally, $f = \pm 1$ denotes whether the mode propagates in the +z or -z direction, such that the sum over the guided modes can be broken down into the two directions along the fiber. For the unguided modes, we have $\sum_{u} = \int_{0}^{\infty} d\omega \int_{-k}^{k} d\beta \sum_{ml}$, where β is a continuous variable in the range $-k < \beta < k$ with $k = \omega_{a}/c$, $m = 0, \pm 1, \pm 2, \ldots$ denotes the mode order, and $l = \pm 1$ again denotes the mode polarization. $G_{\nu i}$ are the coupling strengths between the atom i and the mode ν which, for the guided and unguided modes, respectively, are given by

$$G_{gi} = \sqrt{\frac{\omega\beta_{f}'}{4\pi\hbar\varepsilon_{0}}} \left[\mathbf{d} \cdot \mathbf{e}^{(g)}(r_{i},\phi_{i}) \right] e^{i(f\beta_{f}z_{i}+l\phi_{i})},$$

$$G_{ui} = \sqrt{\frac{\omega}{4\pi\hbar\varepsilon_{0}}} \left[\mathbf{d} \cdot \mathbf{e}^{(u)}(r_{i},\phi_{i}) \right] e^{i(\beta z_{i}+m\phi_{i})}.$$
 (3)

Here, $\mathbf{e}^{(\nu)}$ denotes the profile function of the electric field part of ν , the explicit forms of which can be found, e.g., in [1]. Since we consider a single fundamental guided mode, HE₁₁, the value of the longitudinal propagation constant of the mode $\beta_{\rm f}$ and its derivative $\beta'_{\rm f} = \mathrm{d}\beta_{\rm f}/\mathrm{d}\omega$, which must be determined numerically as the solution of an eigenvalue equation [2], are the only ones in the range $k < \beta_{\rm f} \leq k n_{\rm f}$. The tilde in this notation (e.g. $\tilde{G}_{\nu j}$) serves to indicate that the dipole moment **d** is to be replaced with its complex conjugate. Note that the modes $\mathbf{e}^{(g)}$ and $\mathbf{e}^{(u)}$ are normalized in different manners, leading to dimensions of inverse distance $([m^{-1}])$ and root-time inverse distance $([t^{1/2}m^{-1}])$, respectively [3]. As required, the generalized sums have the same dimensions for both.

MODIFIED SINGLE-ATOM EMISSION

We characterize the emission from a single atom near a nanofiber via three quantities: the total decay rate (Γ), the fraction of the total photon emission that enters into the guided modes of the nanofiber (beta factor), and the degree of chirality of the emission (C).

The total decay rate Γ is obtained from (2) as $\Gamma_{ii} \equiv \Gamma$ (note that in the chain all atoms sit at the same height h above the nanofiber). This rate is the sum of the rates into the guided and unguided modes

$$\Gamma^{g/u} = 2\pi \sum_{g/u} G_{(g/u)} G^*_{(g/u)} \delta(\omega - \omega_a).$$

We will thus define the beta factor as Γ^{g}/Γ . To quantify the degree of directionality or *chirality* of the decay into the nanofiber, we define

$$C = \frac{\Gamma_{\rm R}^{\rm g} - \Gamma_{\rm L}^{\rm g}}{\Gamma^{\rm g}},\tag{4}$$

with R and L representing the rightward and leftwardpropagation direction of the mode, respectively. Here, the rate into the guide modes has been broken down as $\Gamma^{\rm g} = \Gamma^{\rm g}_{\rm R} + \Gamma^{\rm g}_{\rm L}$ with

$$\Gamma_{\rm R/L}^{\rm g} = \sum_{l} \frac{\omega_{\rm a} \beta' |_{\omega_{\rm a}}}{2\hbar\varepsilon_0} |\mathbf{d} \cdot \mathbf{e}^{(\omega_{\rm a}, \pm 1, l)}|^2.$$
(5)

Note that if the transition dipole moment is purely real (for example in the case of a linearly polarized dipole), then the coupling into each of the two guided modes (that possess circular polarization) propagating with opposite directions in the nanofiber is the same and there is no chirality (C = 0). For this reason, we will consider only circularly polarized dipole moments.

The overall modification of the decay rate, Γ/γ , is shown in Fig. 1a for a single atom with circularly polarized transition dipole moment $\mathbf{d} = (d/\sqrt{2})(i, 0, -1)$ placed at a distance h = 100nm above a silica nanofiber.

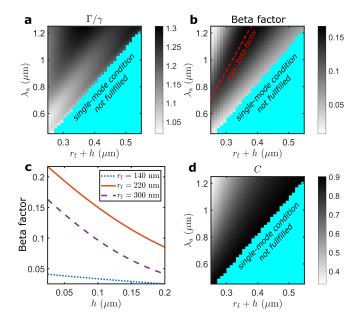


FIG. 1. Modified single atom decay properties. a: Single-atom decay rate Γ for a circularly polarised emitter placed h = 100nm from a silica nanofiber. The decay rate is scaled by the one in vacuum, γ , and plotted varying the transition wavelength $\lambda_{\rm a}$ and the fiber radius $r_{\rm f}$ (in the blue shaded area the single-mode condition is not fulfilled). b: Beta factor. The red line indicates the value of the nanofiber radius where the strongest coupling to the guided modes is found for each wavelength. c: Beta factor for various fixed values of $r_{\rm f}$ as a function of h at $\lambda_{\rm a} = 1\mu$ m. d: Chirality of the decay into the guided modes.

The largest value of the varying nanofiber radius $r_{\rm f}$ for each wavelength is determined by the single-mode condition. The exact dependence of Γ/γ with the nanofiber radius and wavelength is not straightforward. However, we do observe that in all cases decay rate is increased with respect to the free space value by the presence of the nanofiber, and that in general this effect is strongest for long transition wavelengths (i.e., when the atom-fiber separation h is much smaller than the transition wavelength).

The beta factor (Fig. 1b), displays a clearer trend: it has a constant value along the lines $(r_{\rm f} + h)/\lambda_{\rm a} =$ constant, achieving a maximum approximately at $(r_{\rm f} + h)/\lambda_{\rm a} \approx 1/4$ (red dashed-dotted line). Overall, we also note that the coupling to the guided modes is relatively weak, not increasing above 20% over the plotted parameter range. Even reducing the distance to the fiber, h, to values as small as a few tens of nanometers has only a limited effect on the maximum value achievable for the beta factor, as is reflected in Fig. 1c.

We investigate the directionality of the emission into the guided modes, C, in Fig. 1d. One can observe that the chirality of the emission is approximately proportional the ratio between the nanofiber radius and the wavelength $r_{\rm f}/\lambda_{\rm a}$, achieving values up to 0.9 close to the

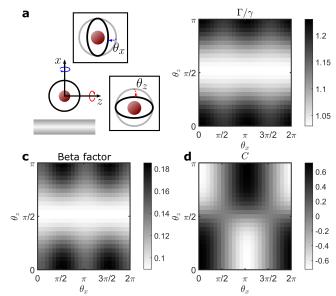


FIG. 2. Role of the polarization direction. a: Circularly polarised emitter with transition wavelength $\lambda_{\rm a} = 1 \mu {\rm m}$ placed $h = 100 {\rm nm}$ above a silica nanofiber with radius $r_{\rm f} = 0.22 \lambda_{\rm a}$. The transition dipole moment is rotated about the axis parallel to the fiber by an angle θ_z , or about the axis parallel to the atom-fiber separation by an angle θ_x . b: Modified decay rate. c: Beta factor. d: Chirality of the decay into the guided modes.

limit of the validity of the single-mode condition. Note, moreover, that in the example shown the chirality is positive, meaning that the guided emission goes prominently into the rightward-propagating guided mode.

Up to now, we have considered a fixed direction of the circularly polarized transition dipole moment **d**, contained in the *xz*-plane. Different directions of this dipole moment modify dramatically the coupling of the atom both to the free radiation and the guided modes. This effect is explored in Fig. 2 for an atom with transition wavelength $\lambda_a = 1\mu m$, where the nanofiber radius $r_f = 0.22\lambda_a$ is chosen such that the coupling to the guided modes is strongest for h = 100nm (red line in Fig. 1b). The dipole moment can be rotated about the axis parallel to the nanofiber by an angle θ_z , and about the axis parallel to the atom-fiber separation by an angle θ_x .

We see that rotating away from $\theta_z = 0$ leads to a reduction in both the modified decay rate Γ , which gets closer to γ , and the beta factor. The beta factor increases to a maximum at $\theta_x = \pi/2$, when the orientation of the dipole moment matches that of the guided mode. The overall decay rate is reduced under this same rotation, as the dipole couples less to the r component of the electric field and more to the weaker ϕ component. The chirality is also reduced, falling to zero at $\theta_x = \pi/2$ (see Fig. 2d) as the dipole moment is then aligned perpendicular to the mode propagation direction, leading to a left/right symmetry in the decay.

STATIONARY STATE IN THE SINGLE-EXCITATION SUBSPACE

The wave function that describes the state of the system can be written in the low excitation limit as $|\psi(t)\rangle = c_G(t) |G\rangle + \sum c_e^i(t) |e\rangle_i$, where $|G\rangle \equiv |g\rangle_1 \otimes |g\rangle_2 \cdots \otimes |g\rangle_N$ and $|e\rangle_i \equiv |g\rangle_1 \otimes |g\rangle_2 \dots |e\rangle_i \cdots \otimes |g\rangle_N$ are the many-body ground and single excitation states, respectively. In this subspace, the photon emission rates depend only on the stationary state value of the coefficients c_e^i , which can be found as a solution of the equation

$$\left(\Delta + i\frac{\Gamma}{2}\right)\mathbf{c}_e^{\rm ss} = H^{\rm eff}\mathbf{c}_e^{\rm ss} + \Omega_{\rm L}\mathbf{v}.$$
 (6)

Here, $\mathbf{c}_{e}^{\mathrm{ss}}$ is the vector that contains the stationary state coefficients c_{e}^{i} , $H_{ij}^{\mathrm{eff}} = V_{ij} - i\Gamma_{ij}/2$, and $v_{i} = e^{i\mathbf{k}_{\mathrm{L}}\cdot\mathbf{r}_{i}}$.

COLLECTIVE EMISSION CHARACTERIZATION

The characterization of the emission from the weakly driven atomic array is done through three quantities: The collective decay rate, the collective beta factor and the collective chirality. Here we introduce the definitions that are only indicated in the main text.

The collective decay rate $\Gamma_{\rm C}$ is defined as

$$\Gamma_{\rm C} = \frac{1}{\gamma} \int_{-\infty}^{\infty} d\Delta N_p(\Delta).$$
 (7)

In order to define a collective collective beta factor, we define first the collective guided rate as

$$\Gamma_{\rm C}^{\rm g} = \frac{1}{\gamma} \int_{-\infty}^{\infty} d\Delta N_p^{\rm g}(\Delta), \qquad (8)$$

such that the collective beta factor is defined as the fraction of the photons that are emitted into the nanofiber, i.e. the $\Gamma_{\rm C}^{\rm g}/\Gamma_{\rm C}$. Finally, the collective chirality is similarly obtained by breaking down the emission rates into the right and left directions of the nanofiber, i.e. $N_p^{\rm g}(\Delta) = N_p^{\rm gR}(\Delta) + N_p^{\rm gL}(\Delta)$. The collective emission rates into each direction are thus given by

$$\Gamma_{\rm C}^{\rm gR/L} = \frac{1}{\gamma} \int_{-\infty}^{\infty} d\Delta N_p^{\rm gR/L}(\Delta), \tag{9}$$

and the collective chirality is defined as

$$C_{\rm C} = \frac{\Gamma_{\rm C}^{\rm gR} - \Gamma_{\rm C}^{\rm gL}}{\Gamma_{\rm C}^{\rm gR} + \Gamma_{\rm C}^{\rm gL}}.$$
 (10)

Note that all three quantities defined here reduce to their single-atom counterparts in the limit of N = 1.

ELECTROMAGNETIC SIMULATION

A visual insight into the emission from the atoms can be gained by simulating the emission of radiation from an array of classical point dipoles placed near the nanofiber. We use the finite difference time domain simulation package MEEP [4], which propagates the electric and magnetic fields on a discretised grid in time steps. The simulation was run using nanoHUB [5]. A short video that shows the dynamics is part of the Supplemental Material.

In Fig.3a, we show the field generated from a single circular dipole $\mathbf{d} = (i, 0, -1)d/\sqrt{2}$ with transition wavelength $\lambda_a = 1\mu$ m at a distance h = 100nm from a nanofiber with $n_f = 1.45$ and $r_f = 220$ nm at an arbitrary time. Here one can observe that the circulating dipole emits preferentially in the forwards direction (chirality). This dipole radiation is equivalent to the photon emission from a single atom.

In Fig.3b we show the results of the same simulation for N = 15 circular dipoles. The spacing between dipoles is set to 800nm, and the relative phase between adjacent dipoles to $\Delta \phi = -1.5$. These parameters approximately reproduce the phase distribution of the superradiant mode depicted in Fig. 2e in the main manuscript (black crosses). Thus, the radiation pattern here approximates the one from this superradiant mode. It can be seen that the directionality of the emission is much improved from the single atom case and the improved β factor is visible in the reduced free space emission.

The laser field driving the atoms is not included in the simulation. The angle of this laser field is implicit in the relative phases between the dipoles.

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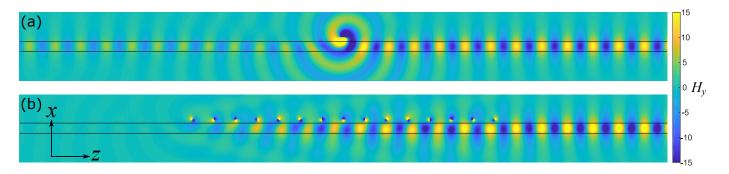


FIG. 3. Emission from classical circular dipoles. In the central plane of the fiber (y = 0) the nonzero field components of the light are E_x , E_z and H_y , the real part of the latter is shown (arbitrary units). The solid black lines represent the edges of fiber. **a:** Radiation from a single dipole. The emission is amplified ×8 to better share a colour scale with **b:** N = 15 dipoles.