

# Superabsorption of light via quantum engineering

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Almost 60 years ago Dicke introduced the term superradiance to describe a signature quantum effect:  $N$  atoms can collectively emit light at a rate proportional to  $N^2$ . Structures that superradiate must also have enhanced absorption, but the former always dominates in natural systems. Here we show that modern quantum control techniques can overcome this restriction. Our theory establishes that superabsorption can be achieved and sustained in certain simple nanostructures, by trapping the system in a highly excited state through transition rate engineering. This opens the prospect of a new class of quantum nanotechnology with potential applications including light harvesting and photon detection. An array of quantum dots or a molecular ring structure could provide a suitable platform for an experimental demonstration.

Superradiance can occur when  $N$  individual atoms interact with the surrounding electromagnetic field [1]. The atom would typically have a discrete dipole allowed transition, and so may also be a semiconductor quantum dot [2], a crystal defect, or a molecule [3]. Light-matter coupling depends on the square of the dipole moment, so that when  $N$  dipoles add coherently light can be emitted at the enhanced rate  $N^2$  [1, 4, 5]. Even for moderate  $N$  this represents a significant increase over the prediction of classical physics, and the effect has found applications ranging from probing exciton delocalisation in biological systems [6], to developing a new class of laser [7], and even to astrophysics [8].

Time-reversal symmetry of quantum mechanics implies that systems with enhanced emission rates will also have enhanced absorption rates. Emission dominates if an excited state of the collective emits into a vacuum, since there are no photons to absorb. Even in an intense light field where absorption and emission are closely balanced, a given transition remains more likely to emit than to absorb. Thus it might seem that the inverse of superradiance is physically forbidden. However, here we show that an often-overlooked interaction between the atoms allows us to control the quantum system such that a sustained superabsorbing state can exist. An appropriate realisation involves a ring structure (see Fig. 1) that is strikingly reminiscent of the photosynthetic light harvesting complex LH1 [9, 10], but the approach described here would outperform any known photosynthetic process.

The Hamiltonian of an ensemble of  $N$  identical atoms is ( $\hbar = 1$ ):

$$\hat{H}_S = \frac{\omega_A}{2} \sum_{i=1}^N (\mathbf{1}^i - \hat{\sigma}_z^i) = \omega_A \sum_{i=1}^N \hat{\sigma}_+^i \hat{\sigma}_-^i, \quad (1)$$

where  $\omega_A$  is the bare atomic transition frequency;  $\hat{\sigma}_z^i = |g\rangle_i \langle g| - |e\rangle_i \langle e|$ ,  $\hat{\sigma}_+^i = |e\rangle_i \langle g|$ , and  $\hat{\sigma}_-^i = (\hat{\sigma}_+^i)^\dagger$  are the usual Pauli operators defined with respect to the  $i$ th atom's ground,  $|g\rangle_i$ , and optically excited state,  $|e\rangle_i$ . When the wavelength  $\lambda$  of light is much larger than all interatomic distances  $r_{ij}$ , ( $\lambda \gg r_{ij}$ ), the atoms become indistinguishable and light interacts with the system collectively. The dynamics are then best described by collective operators:

$$\hat{J}_\pm = \sum_{i=1}^N \hat{\sigma}_\pm^i, \quad \hat{J}_z = \sum_{i=1}^N \hat{\sigma}_z^i, \quad (2)$$

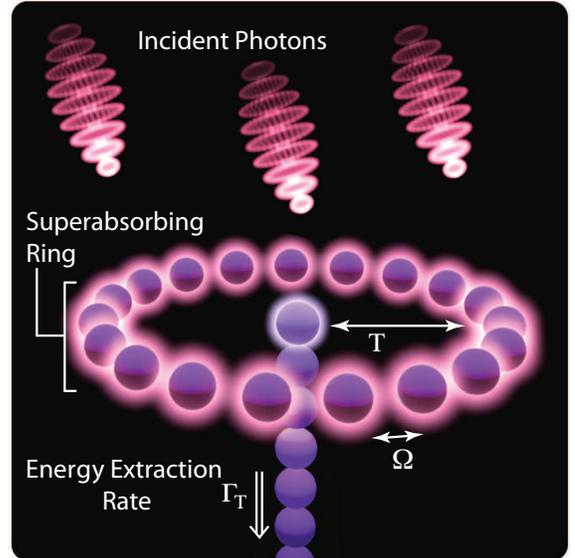


FIG. 1: One potential realisation of superabsorption. Photons absorbed by the ring give rise to delocalised excitons; ideally the ring maintains a specific exciton population. As new excitons are created they transfer from the ring to a central core absorber and then exit via (e.g.) a strongly coupled chain.

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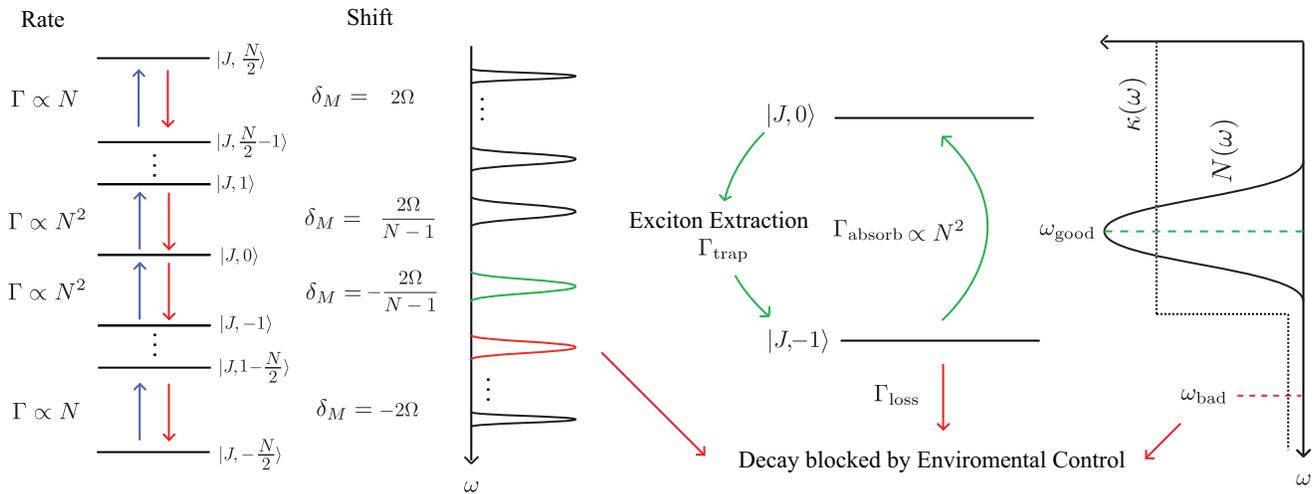


FIG. 2: Left: The ladder of Dicke states of an  $N$  atom system, with emission (red) and absorption (blue) processes. In the presence of interactions  $\Omega \neq 0$ , the frequency shift of each transition is given by  $\omega_A + \delta_M$ . Right: A scheme for using the environment to confine the ladder of states into an effective two level system either by tailoring the spectral density  $\kappa(\omega)$  or the mode occupation  $n(\omega)$ .

which generate transitions between the eigenstates of Hamiltonian (1) and obey  $SU(2)$  commutation relations. We can succinctly express the light matter interaction Hamiltonian as

$$\hat{H}_L = -\hat{E} d \left( \hat{J}_+ + \hat{J}_- \right), \quad (3)$$

where  $\hat{E}$  is the light field operator and  $d$  is the atomic dipole matrix element. Hamiltonian (3) causes the system to move along a ladder of states called the ‘Dicke’ or ‘bright’ states which are characterised by the eigenvalues  $J$  and  $M$  of  $\hat{J}^2$  and  $\hat{J}_z$ , respectively. In the absence of interactions between the atoms,  $\hat{J}^2$  commutes with  $\hat{H}_S + \hat{H}_L$  and thus its eigenvalue  $\frac{N}{2} \left( \frac{N}{2} + 1 \right)$  is a conserved quantity. The Dicke states form a ladder from  $|J, -\frac{N}{2}\rangle$  to  $|J, \frac{N}{2}\rangle$  shown on the left in Fig. 2, each rung of which corresponds to the fully symmetric superposition of  $N/2 + M$  excited atoms. The collective excitation operators explore this ladder of states,

$$\hat{J}_\pm |J, M\rangle = \sqrt{(J \pm M + 1)(J \mp M)} |J, M \pm 1\rangle, \quad (4)$$

and the transition rates between adjacent Dicke ladder states are then readily calculated:

$$\Gamma_{M \rightarrow M \pm 1} = \gamma \left( \frac{N}{2} \pm M + 1 \right) \left( \frac{N}{2} \mp M \right), \quad (5)$$

where  $\gamma = 8\pi^2 d^2 / (3\epsilon_0 \hbar \lambda^3)$  is the free atom decay rate.

If the system is initialized in the fully excited state  $|J, \frac{N}{2}\rangle$  with no environmental photons, then the system cascades down the ladder, as shown by the red arrows on the left side of Fig. 2. Upon reaching the midway point ( $M = 0$ ) its emission rate is given by

$$\Gamma_{M \rightarrow M-1} \approx \gamma \left( \frac{N}{2} \right)^2, \quad (6)$$

which is larger than the  $\gamma N$  rate expected of  $N$  uncorrelated atoms, for any  $N > 4$ . This is the essence of superradiance: constructive interference between the different possible decay paths greatly enhances the emission rate, producing a high intensity pulse. The enhancement is the result of simple combinatorics: near the middle of the ladder,  $|J, 0\rangle$ , there are a large number of possible configurations of excited atoms corresponding to each respective Dicke state.

Superradiance is not an intrinsically transient effect: steady state superradiance can occur through repumping [11], or in cavities [12, 13], and recently a superradiant laser with potential for extraordinary stability and narrow linewidth was demonstrated [7].

## I. SUPERABSORPTION

We introduce the term ‘superabsorption’ to describe the reciprocal phenomenon to superradiance, in which a collective system absorbs radiation at a rate that scales superlinearly with the number of individual absorbers. In natural systems this is an improbable process that can only last for a vanishingly short time. However, by employing *environmental quantum control* it can be extended into a physical phenomenon that can be exploited and used in technology. The crucial ingredient for achieving superabsorption is to engineer the transition rates in a way that primarily confines the dynamics to an *effective two-level system* (E2LS) around the  $M = 0$  transition, which exhibits the required quadratic absorption rate. Achieving net superabsorption for harvesting light adds a second requirement: a non-radiative channel to extract excitons from the upper of these two levels, turning them into useful work. Periodic reinitialisation, for

example through feedback control [14], enables devices capable of functioning as sustained photon detectors or harvesters.

Let us consider the first requirement. In order to ensure that most transitions take place within the E2LS we must either suppress the total loss rate from the E2LS or enhance the probability of transitions within the subspace. This becomes possible if the frequency of the E2LS transition becomes distinct from the other transitions; this would not be true in the case of a non-interacting set of atoms, but does occur once interactions are included. Dicke physics requires that the atoms remain identical but interactions are still permissible in certain symmetric geometries such as rings [4, 15], and these structures will still exhibit superradiance. We therefore consider the candidate superabsorber depicted in Fig. 1, in which we assume the interactions act between adjacent atoms only and are due to Foerster type coupling. This leads to a Dicke ladder of non-degenerate transitions whose dynamics in the presence of frequency selective (filtered) thermal light are found from the master equation:

$$\begin{aligned} \dot{\rho} = & -i[\hat{H}_S + \hat{H}_I, \rho] \\ & -\gamma \sum_{\beta \in \omega} \kappa(\omega_\beta) \left( (n(\omega_\beta) + 1) D[\hat{L}_\beta] \rho + n(\omega_\beta) D[\hat{L}_\beta^\dagger] \rho \right). \end{aligned} \quad (7)$$

$\kappa(\omega) = \sum_k |g_k|^2 \delta(\omega - \omega_k) = \chi(\omega) |g(\omega)|^2$  is the spectral density at frequency  $\omega$ ;  $n(\omega_\beta)$  is the occupation number of the  $\omega_\beta$  mode, and  $D[\hat{L}_\beta] \rho$  is the Lindbladian dissipator  $\hat{L}_\beta \rho \hat{L}_\beta^\dagger - \frac{1}{2} \{ \hat{L}_\beta^\dagger \hat{L}_\beta, \rho \}$ .  $L_\beta^\dagger$  moves the system up a Dicke ladder transition with frequency  $\omega_\beta$ .

Eq. (7) also features unitary dynamics due to the field interaction that comprises two components: the Lamb shift, accounted for by renormalising  $\omega_A$  in the system Hamiltonian  $\hat{H}_S$ , and the field induced dipole-dipole interaction

$$\hat{H}_I = \Omega_{i,j} \sum_{i \neq j}^N \left( \hat{\sigma}_+^i \hat{\sigma}_-^j + \hat{\sigma}_-^i \hat{\sigma}_+^j \right), \quad (8)$$

which describes energy conserving ‘hopping’ of excitons between sites mediated by (virtual) photon exchange. The hopping interaction strength  $\Omega_{i,j}$  is given by [4]

$$\Omega(i, j) = \frac{d^2}{4\pi\epsilon_0 r_{ij}^3} \left[ 1 - \frac{3(\hat{\epsilon}_a \cdot \vec{r}_{ij})^2}{r_{ij}^2} \right] \approx \frac{d^2}{4\pi\epsilon_0 r_{ij}^3}. \quad (9)$$

with  $\hat{\epsilon}_a$  a unit vector in the dipoles’ direction. For a circular geometry with dipoles perpendicular to  $\vec{r}_{ij}$  and retaining only nearest neighbour interactions [a good approximation for larger rings since  $\Omega(i, j) \propto r_{ij}^{-3}$ ],  $\Omega := \Omega(i, i+1)$  is a constant (see the Supplementary Information for a discussion of the non-nearest neighbour case). Owing to the high degree of symmetry of the ring geometry,  $\hat{H}_I$  does not mix the  $|JM\rangle$  eigenstates but only shifts their energies [4] according to

$$\delta E_M = \langle J, M | \hat{H}_I | J, M \rangle = \Omega \frac{J^2 - M^2}{J - \frac{1}{2}}. \quad (10)$$

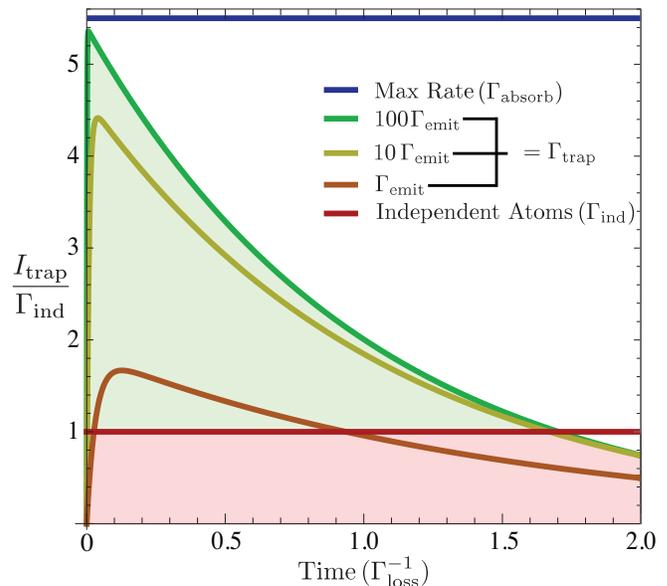


FIG. 3: The transient superabsorption of the effective two level system indicated in Fig. 2. The green shading indicates the superabsorption region, the red when the extraction rate is below what could be extracted from uncorrelated atoms; both are for a system of twenty atoms and mode occupancy  $n(\omega_{\text{good}}) = 10$ . The maximum extraction possible from independent atoms ( $\Gamma_{\text{ind}} = n(\omega_{\text{good}})N\gamma$ ) is used for comparison.

The shift of the transition frequencies is given by the difference of two adjacent levels  $E_M - E_{M-1}$ :

$$\omega_{M \rightarrow M-1} = \omega_A - 4\Omega \frac{M - \frac{1}{2}}{N - 1}. \quad (11)$$

These altered frequencies break the degeneracy in the Dicke ladder and each transition has a unique frequency. For example the transition frequency from the ground state to the first Dicke state is  $\omega_{-N/2+1 \rightarrow -N/2} = \omega_A - 2\Omega$ . Crucially, the Dicke states still form an eigenbasis for the system, yet each transition in the ladder now samples both  $\kappa(\omega)$  and  $n(\omega)$  at its own unique frequency.

## II. QUANTUM CONTROL

Our aim is to enhance transition rates at the frequency of the E2LS, which we shall call the ‘good’ frequency ( $\omega_{0 \rightarrow -1} =: \omega_{\text{good}}$ ) and suppress those for transitions directly out of the E2LS at the ‘bad’ frequency ( $\omega_{-1 \rightarrow -2} =: \omega_{\text{bad}}$ ). The required type of (incoherent) control of the environment is known as reservoir engineering [16]; in this problem we have a choice between tailoring  $\kappa(\omega)$ ,  $n(\omega)$ , or both. Tailoring the spectral density has the advantage that it can, in theory, completely eliminate the rate of loss from our E2LS when there is no mode of the right frequency present to allow decay. In practice this would require the device to be inside a photonic bandgap (PBG) crystal with a stop band at  $\omega_{\text{bad}}$

(see Fig. 2), where the required dimensionality of the PBG depends on the orientation of the optical dipoles. Suppression of emission rates by several orders of magnitude has already been demonstrated [17, 18]. Alternatively a narrow line width cavity could enhance  $\kappa(\omega_{\text{good}})$ .

Control of  $n(\omega)$  is easier to achieve, e.g. by using filtered thermal or pseudothermal [19, 20] light. However, this approach has the limitation that even in the optimal control regime, where  $n(\omega) = 0$  everywhere except at in a narrow region around  $\omega_{\text{good}}$ , spontaneous emission will still cause some loss from the E2LS.

Since both environmental control approaches rely on frequency selectivity, a sufficiently large detuning between adjacent Dicke transitions will be critical for achieving effective containment within the E2LS. In practice the environmental control will never be quite perfect and our system will over long times inevitably evolve away from the E2LS, thus requiring reinitialisation at regular intervals as a trade off for the enhanced absorption rate.

### III. TRAPPING

We have detailed how to create a system exhibiting sustained superabsorption. To achieve net superabsorption or for it to function as a light harvester we need to be able to extract the resultant excitons via an irreversible trapping process. We require a process that extracts only the excitons that are absorbed by the E2LS, as depicted in Fig. 2, and does not extract excitons from levels below the E2LS. The excitons are delocalised across the ring and need to be extracted collectively to preserve the symmetry of the Dicke states. In designing this process we take inspiration from natural light harvesting systems: A ‘trap’ atom is located at the centre of the ring and symmetrically coupled via a resonant hopping interaction to all the other atoms (see Fig. 1). The corresponding trapping Hamiltonian is

$$\hat{H}_T = g(\hat{J}_+ \hat{\sigma}_-^T + \hat{J}_- \hat{\sigma}_+^T) + \omega_{\text{trap}} \hat{\sigma}_+^T \hat{\sigma}_-^T, \quad (12)$$

where the superscript  $T$  denotes the trap site,  $g$  is the strength of the coupling between the ring and the trap, and the trap’s transition frequency  $\omega_{\text{trap}}$  ideally matches  $\omega_{\text{good}}$ . In this case the interaction is mediated by the electromagnetic field as described in the previous section, but it could have other physical origins depending on the system of interest. Once an exciton has moved to the trap site we assume that it is then removed into the wider environment by a process which irreversibly absorbs its energy. We note that more exotic and potentially far more efficient trapping implementations can be envisioned, such as e.g. a reservoir of excitons with an effective ‘Fermi level’ capable of accepting an excitons only above the energy level  $E_{-1}$ . However, at present our aim is to focus on the simplest system capable of exhibiting superabsorption.

The above trapping process is adequately described phenomenologically (see Supplementary Information) as collective exciton extraction from the mid point ( $M = 0$ ) by adding the dissipator  $D[\hat{L}_{\text{trap}}]\rho$  to the righthand side of Eq. (7) with  $\hat{L}_{\text{trap}} = \sqrt{\Gamma_{\text{trap}}} |J, -1\rangle \langle J, 0|$ , and where  $\Gamma_{\text{trap}}$  is the rate of the trapping process. The rate of exciton extraction  $I_{\text{trap}}$  is then simply given by the population of the trapping level multiplied by the trapping rate:

$$I_{\text{trap}}(t) = \Gamma_{\text{trap}} \text{Tr} [|J, 0\rangle \langle J, 0| \rho(t)]. \quad (13)$$

### IV. RESULTS

Consider an ideal E2LS realised by a PBG completely blocking  $\omega_{\text{bad}}$ , i.e. a vanishing  $\Gamma_{\text{loss}} := \kappa(\omega_{\text{bad}})(n(\omega_{\text{bad}}) + 1)\Gamma_{-1 \rightarrow -2}$ . Assuming a faster trapping than emission rate,  $\Gamma_{\text{trap}} \gg \Gamma_{\text{emit}} := \kappa(\omega_{\text{good}})(n(\omega_{\text{good}}) + 1)\Gamma_{0 \rightarrow -1}$ , our figure of merit  $I_{\text{trap}}$  matches the absorption rate  $\Gamma_{\text{absorb}} := \kappa(\omega_{\text{good}})n(\omega_{\text{good}})\Gamma_{-1 \rightarrow 0}$  for all  $t$ :

$$I_{\text{trap}}(t) = I_{\text{max}} \approx \Gamma_{\text{absorb}} \approx \mu \left( \frac{N}{2} + \frac{N^2}{4} \right), \quad (14)$$

where  $\mu = \gamma\kappa(\omega_{\text{good}})n(\omega_{\text{good}})$ . It is clear from this equation that under these conditions we achieve a superlinear scaling of the exciton current flowing out of the superabsorber.

When considering filtered thermal light to confine the total system to an approximate E2LS, or for an imperfect PBG, there will be some loss. This will lead to an exponential decay of  $I_{\text{trap}}(t)$  with the lifetime  $\Gamma_{\text{loss}}^{-1}$ , as shown in Fig. 3. The initial superabsorption rate still far exceeds that possible from uncorrelated atoms, however it is only a transient effect and the system needs to be reinitialised periodically to maintain its advantage. Reinitialisation could be achieved by exploiting a chirped pulse of laser light to re-excite the system, or through a temporary reversal of the trapping process. In practice there will be an energy cost associated with reinitialisation but, as we show below, in all but the most severe cases this cost is more than offset by the faster photon to exciton conversion rate during the transient superabsorption periods.

Perhaps the most elegant way of implementing the reinitialisation step would make use of quantum feedback control [14]: The superabsorption enhancement is derived from coherence between states that all possess the same number of excitons. Therefore, the number of excitons could be continually monitored (e.g. by monitoring fluorescence of a probe field tuned to a level or two below the desired manifold) without destroying the desired effect. A suitably designed feedback system could then feed in an excitation only when a loss event had

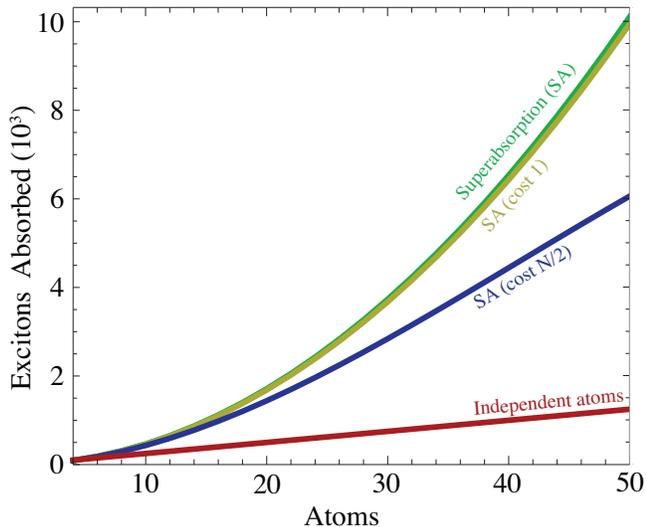


FIG. 4: How the total number of excitons absorbed in a given time (set to the lifetime of the smallest system  $\Gamma_{\text{loss}}^{-1}$  for  $N = 4$ ), scales with the number of atoms  $N$ , for various costs of reinitialisation (excitons). The red line shows the maximum extracted from independent atoms for comparison.

occurred, providing optimal efficiency

$$I_{\text{trap}} = \Gamma_{\text{absorb}} - \Gamma_{\text{loss}} = (\mu - \sigma) \left( \frac{N}{2} + \frac{N^2}{4} \right) + 2\sigma, \quad (15)$$

where  $\sigma = \gamma\kappa(\omega_{\text{bad}})(1 + n(\omega_{\text{bad}}))$ . Provided  $\mu > \sigma$  superabsorption will occur, and for  $\sigma = 0$  we recover the theoretical maximum of the idealised case in Eq. (14).

We now discuss a far simpler and much less demanding reinitialisation scheme where no such feedback knowledge exists. In this case, in order to account for the cost of reinitialisation, we need to quantify the total number of excitons absorbed in a given time. Let us fix the time at which reinitialisation is performed to the natural lifetime of the E2LS,  $\Gamma_{\text{loss}}^{-1}$ . Integrating the trapping rate  $I_{\text{trap}}(t)$  over one lifetime and subtracting the reinitialisation cost gives a fair measure of the number of excitons the system has absorbed within the given time. We consider the extreme limits of the reinitialization cost, from simply replacing a single lost exciton, to having to replace all of the  $N/2$  excitons that make up the superabsorbing state. A larger system requires more frequent reinitialisations, since its loss rate is also enhanced by the system size. However, the bias in favour of absorption created by the environmental control is sufficient to ensure this does not negate the superabsorption process. Fig. 4 shows how the number of excitons absorbed in a given time scales with the number of atoms, and for all cost models we have a superlinear scaling.

## V. DISCUSSION

We have shown that the absorptive analogue of quantum superradiance can be engineered in structures with suitably symmetric interactions. Interestingly, an ideal implementation would look remarkably similar to the ring structures found in Nature’s photosynthetic antenna complexes [9, 10]. As we discuss in the following, absorbing light beyond the limits of classical physics raises prospects for new types of technology, and the superabsorption effect could be realised in a broad range of candidate systems.

Light harvesting technologies immediately spring to mind as a potential application, and indeed our Fig. 4 indicates that one can obtain a net increase over conventional systems even allowing for the energy cost of sustaining the superabsorbing state. A caveat is that a given superabsorber achieves optimal performance only for a specific frequency range; however, one could engineer a range of different systems to jointly cover the solar spectrum. The technique may also be particularly suited to wireless power transfer using narrowband light, e.g. for remote sensor or biologically implanted devices, where wired electrical power is impractical.

A quite different application of the phenomenon may be in the context of optical or microwave sensors, either in future cameras or for scientific instruments. In addition to the obvious merits of being sensitive to weak light levels, the frequency specificity of the superabsorber may in fact be a desirable attribute. The small size of the ring structure and collective ‘antenna array’ could lead to high spatial and angular resolution, and the fact that the superabsorber is (re)initialised into its fully receptive state by an excitation pulse would allow excellent time resolution. For sensing applications the cost of reinitialisation is unimportant. Further, a trapping mechanism is not required if the number of excitons in the system can be monitored in a different way, e.g. with a quantum point contact.

There are multiple candidate systems for engineering the above applications. Molecular rings have the advantage of featuring intrinsically low levels of disorder and natural symmetry. Taking  $\Omega = 350 \text{ cm}^{-1}$  as appropriate for a B850 ring [21] with eight atoms produces transition wavelength shifts exceeding 6 nm, and a wavelength selectivity on the scale of nanometers is readily available with current laser and cavity linewidths. Of course, the dipole alignment of the B850 ring is not optimised for this purpose. However, complex ring structures can be designed and synthesised artificially (for example, porphyrin rings [22]) and this route should enable far superior properties. Alternatively, superradiance, long-range interactions, and optical control have been demonstrated in quantum dots [2, 23], and there has been recent progress in synthesising ring like clusters with high spectral and spatial order [24].

To demonstrate the effect of superabsorption as a novel physical phenomenon, initially without a trapping mech-

anism, several additional possibilities present themselves. For example, circuit QED experiments possess long coherence times and have already demonstrated superradiant effects [25, 26], and recent 3D structures [27] provide further flexibility. Bose Einstein Condensates offer similar properties but with much larger numbers of atoms [28, 29].

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# Supplementary Information: Superabsorption of light via quantum engineering

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## I. MASTER EQUATION DERIVATION

In the following we sketch the derivation of the master equation (7) from the main Article. Following the general procedure of Ref. [1], we generalise from a vacuum environment to one with a population distribution and structured spectral density. This allows for the presence of the superabsorption term and introduces some additional complexities.

We consider the interaction picture with respect to  $\hat{H}_S$  [Eq. (1) of the Article] and the free Hamiltonian of the electromagnetic field. After performing the standard Born-Markov approximation and tracing over the environment, the starting point for our derivation is [2] ( $\hbar = 1$ ):

$$\frac{d}{dt}\tilde{\rho}_S(t) = -i \int_0^\infty dt' \text{Tr}_E[\tilde{H}_L(t), [\tilde{H}_L(t-t'), \tilde{\rho}_S(t) \otimes \tilde{\rho}_E]], \quad (\text{S1})$$

where  $\tilde{\rho}_S(t)$  is the reduced interaction picture density matrix and  $\tilde{H}_L(t)$  denotes the interaction picture representation of the system-light-interaction Hamiltonian  $\hat{H}_L = -\sum_{i=1}^N \hat{\sigma}_-^i \vec{d} \cdot \hat{E}(\vec{r}_i) + \hat{\sigma}_+^i \vec{d}^* \cdot \hat{E}(\vec{r}_i)$ , [c.f. Eq. (3) of the main text]. Here,  $\vec{d}$  is the atomic dipole vector and the electric field operator is given by

$$\hat{E}(\vec{r}_i) = i \sum_{i=1}^N \sum_{\vec{k}, \lambda} \sqrt{\frac{2\omega_k}{V}} \vec{e}_\lambda(\vec{k}) \left( \hat{b}_\lambda(\vec{k}) e^{i\vec{k} \cdot \vec{r}_i} - \hat{b}_\lambda^\dagger(\vec{k}) e^{-i\vec{k} \cdot \vec{r}_i} \right), \quad (\text{S2})$$

where  $\vec{e}_\lambda(\vec{k})$  and  $\hat{b}_\lambda^\dagger(\vec{k})$  are the polarisation vector of the field and its annihilation (creation) operator, respectively. The system dynamics is then generically determined by the following master equation [2]

$$\frac{d}{dt}\rho_S(t) = -i[\hat{H}_S + \hat{H}_I, \rho_S(t)] + \sum_\omega \sum_{i,j} \left[ \Gamma_{i,j}(\omega) (\hat{A}_j \rho_S(t) \hat{A}_i^\dagger - \hat{A}_i^\dagger \hat{A}_j \rho_S(t)) + \text{H.c.} \right], \quad (\text{S3})$$

where H.c. denotes the Hermitian conjugate. The  $\hat{A}_i$  are the Lindblad operators given by  $\hat{\sigma}_-^i$  and  $\hat{\sigma}_+^i$ , and  $\Gamma_{i,j} = \int_0^\infty ds e^{i\omega s} \langle \vec{d}^* \cdot \hat{E}(\vec{r}_i, s) \vec{d} \cdot \hat{E}(\vec{r}_j, 0) \rangle$  is the spectral correlation tensor, which will be calculated in the following. We start by considering the expression

$$\langle \vec{d}^* \cdot \hat{E}(\vec{r}_i, s) \vec{d} \cdot \hat{E}(\vec{r}_j, 0) \rangle = \text{Tr}_E[\vec{d}^* \cdot \hat{E}(\vec{r}_i, s) \vec{d} \cdot \hat{E}(\vec{r}_j, 0) \rho_E], \quad (\text{S4})$$

where  $\rho_E$  denotes the thermal state of the environment, though allowing filtered thermal light later will not change the form of the result [3, 4]. Generally, for a thermalised environment it is well known that [2]

$$\langle \hat{b}_\lambda(\vec{k}) \hat{b}_{\lambda'}(\vec{k}') \rangle = \langle (\hat{b}_\lambda^\dagger(\vec{k}) \hat{b}_{\lambda'}^\dagger(\vec{k}')) \rangle = 0, \quad (\text{S5})$$

$$\langle \hat{b}_\lambda(\vec{k}) \hat{b}_{\lambda'}^\dagger(\vec{k}') \rangle = \delta_{\vec{k}\vec{k}'} \delta_{\lambda\lambda'} = (1 + n(\omega_k)), \quad (\text{S6})$$

$$\langle \hat{b}_\lambda^\dagger(\vec{k}) \hat{b}_{\lambda'}(\vec{k}') \rangle = \delta_{\vec{k}\vec{k}'} \delta_{\lambda\lambda'} = n(\omega_k). \quad (\text{S7})$$

Using these, the spectral correlation tensor can be written as:

$$\Gamma_{i,j} = \frac{2\pi}{V} \sum_{\vec{k}, \lambda} (\vec{d} \cdot \vec{e}_\lambda(\vec{k}))^2 \omega_k \left( (1 + n(\omega_k)) e^{i\vec{k} \cdot \vec{r}_{ij}} \int_0^\infty ds e^{-i(\omega_k - \omega)s} + e^{-i\vec{k} \cdot \vec{r}_{ij}} n(\omega_k) \int_0^\infty ds e^{i(\omega_k + \omega)s} \right), \quad (\text{S8})$$

where  $\vec{r}_{i,j}$  is the vector connecting atoms  $i$  and  $j$ . Converting the sum over  $\vec{k}$  to an integral ( $\omega_k = c|\vec{k}|$ ) yields

$$\frac{1}{V} \sum_{\vec{k}} \rightarrow \frac{1}{(2\pi)^3 c^3} \int_0^\infty d\omega_k \kappa(\omega_k) \omega_k^2 \int d\Omega, \quad (\text{S9})$$

where  $\kappa(\omega)$  is the spectral density given by the density of states weighted by the coupling strength,  $\kappa(\omega) = \sum_k |g_k|^2 \delta(\omega - \omega_k) = \chi(\omega) |g(\omega)|^2$ . The angular part of the integration gives a diffraction type function:

$$F(\omega \vec{r}_{ij}) = \frac{8\pi}{3} \left( j_0(\omega |\vec{r}_{ij}|) + \frac{1}{2} \left( 3 \cos^2(\theta_{\vec{d}\vec{r}_{ij}}) - 1 \right) j_2(\omega |\vec{r}_{ij}|) \right), \quad (\text{S10})$$

where  $j_n(x)$  is the  $n^{\text{th}}$  spherical Bessel function and the angle  $\theta_{\vec{d}\vec{r}_{ij}}$  between the atomic dipoles and pairwise connection vectors is

$$\cos^2 \theta_{\vec{d}\vec{r}_{ij}} = \frac{|\vec{d} \cdot \vec{r}_{ij}^2|}{|\vec{d}^2| |\vec{r}_{ij}^2|}. \quad (\text{S11})$$

Considering the geometry in Fig. 1 of the Article, we assume that all dipoles are parallel, and perpendicular to the plane defined by the ring. In this case, and for only nearest neighbour interactions,  $\theta_{\vec{d}\vec{r}_{ij}}$  is independent of  $i$  and  $j$ . Thus Eq. (S8) becomes

$$\Gamma_{i,j} = \frac{|d|^2}{(2\pi)^2 c^3} \int_0^\infty d\omega_k \kappa(\omega_k) \omega_k^3 F(\omega_k r_{ij}) \left( (1 + n(\omega_k)) \int_0^\infty ds e^{-i(\omega_k - \omega)s} + n(\omega_k) \int_0^\infty ds e^{i(\omega_k + \omega)s} \right), \quad (\text{S12})$$

which we separate into its real and imaginary parts  $\Gamma_{i,j} = \frac{1}{2} \gamma_{i,j}(\omega) + iS(\omega)$  with the help of the identity

$$\int_0^\infty ds e^{\pm i\epsilon s} = \pi \delta(\epsilon) \pm iP \frac{1}{\epsilon}. \quad (\text{S13})$$

The real terms  $\gamma_{i,j}(\omega)$  derive from the delta functions and give rise to the dissipative dynamics, i.e. optical transitions in this case. In the remaining term,  $F(\omega_k r_{ij})$  is evaluated at  $\omega_k = \pm\omega$ . We are working in the small sample limit, where the wavelength of light is far longer than the size of our nanostructure ( $\omega r_{ij} \approx 0$ ), and so  $F(\omega r_{ij}) \approx 8\pi/3$ . Hence  $\gamma_{i,j}$  is independent of the atomic indices to a good approximation:

$$\gamma_{i,j}(\omega) \approx \gamma(\omega) = \frac{4\omega^3 |d|^2}{3c^3} \kappa(\omega) (1 + n(\omega)) \quad (\text{S14})$$

The Planck distribution has the property that  $n(-\omega) = -(1 + n(\omega))$ . Thus we can combine the terms arising from  $\delta(\omega_k \pm \omega)$  and only run the sum over positive values. The second term on the righthand side of Eq. (S3) thus becomes

$$\sum_{\omega > 0} \sum_{i,j} \frac{4\omega^3 |d|^2}{3c^3} \kappa(\omega) \left( (1 + n(\omega)) (\sigma_j^- \rho \sigma_i^+ - \frac{1}{2} \{ \sigma_i^+ \sigma_j^-, \rho \}) + n(\omega) (\sigma_j^+ \rho \sigma_i^- - \frac{1}{2} \{ \sigma_i^- \sigma_j^+, \rho \}) \right). \quad (\text{S15})$$

By simply assuming all transitions have the same frequency splitting ( $\omega = \omega_A$ ), a vacuum environment state  $n(\omega) = 0$  and switching to the collective operators to express the sums  $\hat{J}_- = \sum_i \hat{\sigma}_i^-$ , we reproduce the ordinary superradiance master equation.

We now turn to the imaginary part  $S(\omega)$  of the spectral correlation tensor, which will be responsible for providing the detuning between different transitions; this is given by:

$$S(\omega) = \frac{|d|^2}{(2\pi)^2 c^3} P \int_0^\infty d\omega_k \kappa(\omega_k) \omega_k^3 F(\omega_k r_{ij}) \left( \frac{1 + n(\omega_k)}{\omega - \omega_k} + \frac{n(\omega_k)}{\omega_k + \omega} \right). \quad (\text{S16})$$

The  $i = j$  terms, for which  $F(0) = 8\pi/3$ , correspond to the ordinary Lamb shift of individual atom transitions; these can be accounted for by a renormalisation of the bare atomic frequency  $\omega_A$ . By contrast, the  $i \neq j$  terms correspond to the dipole-dipole interaction induced by the EM field. Evaluating this integral requires us to choose a specific form for the spectral density  $\kappa(\omega)$ . Here we consider two cases: first a flat spectral density, and second one that features a ‘stop band’ in the spectrum, blocking the ‘bad’ transition at frequency  $\omega_{\text{bad}}$ . We begin with the former case. We first separate out the term that is independent of  $n(\omega)$ , the Lamb shift  $S_L(\omega)$ , and evaluate it. In the small sample limit taken ( $\omega r_{ij} \ll 1$ ) we find

$$S_L(\omega) = \frac{d^2}{4\pi\epsilon_0 r_{ij}^3} [1 - 3 \cos^2(\theta_{dr})]. \quad (\text{S17})$$

After separating out the Lamb shift, we are left with the divergent integral corresponding to the Stark shift:

$$S_s(\omega) = \frac{|d|^2}{(2\pi)^2 c^3} P \int_0^\infty d\omega_k \kappa(\omega_k) \omega_k^3 F(\omega_k r_{ij}) \left( \frac{n(\omega_k)}{\omega - \omega_k} + \frac{n(\omega_k)}{\omega + \omega_k} \right). \quad (\text{S18})$$

This very seldom evaluated in the literature and is usually assumed negligible. In this work we are primarily concerned with controlling  $n(\omega)$  so that it is only significant for one mode, which has frequency  $\omega_g$ . In this case we set  $n(\omega_k) = \delta(\omega_k - \omega_g)$  and take the small sample limit:

$$S_s(\omega) = \lim_{r\omega_g \rightarrow 0} \frac{4\pi\omega (\sin(r\omega_g) (\cos(2\theta_{dr}) (3 - r^2\omega_g^2) + r^2\omega_g^2 + 1) - r\omega_g(3 \cos(2\theta_{dr}) + 1) \cos(r\omega_g))}{r^3(\omega - \omega_g)(\omega + \omega_L)} = 0. \quad (\text{S19})$$

Hence we can neglect the Stark shift and only retain the Lamb shift. Returning to the other spectral density we consider in the paper, that with the stop band, we can express the stop band with the following, simplistic spectral destiny:

$$\kappa(\omega) = 1 - T(\omega_b, \sigma), \quad (\text{S20})$$

where  $T(\omega_b, \sigma)$  is the ‘top hat’ function centred on  $\omega_b$  with a width  $\sigma$ . The factor of one produces the same result as for the flat spectral density  $S_s(\omega)$ . The top hat handles the effect of the gap  $S_{gap}(\omega)$ :

$$S(\omega) = S_s(\omega) - S_{gap}(\omega). \quad (\text{S21})$$

The top hat has the effect of confining the integral to a window around  $\omega_b$ :

$$S_{gap}(\omega) = \frac{|d|^2}{(2\pi)^2 c^3} P \int_{\omega_b - \sigma}^{\omega_b + \sigma} d\omega_k \omega_k^3 F(\omega_k r_{ij}) \left( \frac{1 + n(\omega_k)}{\omega - \omega_k} + \frac{n(\omega_k)}{\omega_k + \omega} \right). \quad (\text{S22})$$

This can be evaluated to yield a lengthy, but straightforward expression. For an ideal gap ( $\sigma \rightarrow 0$ ),  $S_{gap}(\omega) = 0$  and hence can be neglected. Collating these results, we are left with a familiar expression for the strength of the interaction between two dipoles (S17) multiplied by a hopping term introduced via the EM field:

$$\hat{H}_I = S_L(\omega) \sum_{i \neq j}^N \left( \hat{\sigma}_+^i \hat{\sigma}_-^j + \hat{\sigma}_-^i \hat{\sigma}_+^j \right). \quad (\text{S23})$$

In the paper we consider the nearest neighbour limit of this expression, although this is not a necessity (see below). We know that the EM field operators interact with the system collectively, causing them to explore the ladder of Dicke states defined by:

$$|J, M\rangle = \sqrt{\frac{(J+M)!}{N!(J-M)!}} \hat{j}_-^{(J-M)} |ee\dots e\rangle. \quad (\text{S24})$$

The highly symmetric geometry of our system means that the hopping interaction (S23) does not cause mixing of Dicke levels, but only shifts their energies (see Article). Thus an effective Hamiltonian for the subspace consisting of only the fully symmetric states of the Dicke ladder can be written as :

$$\hat{H}_S + \hat{H}_I = \frac{E_M}{2} \sum_{M=-J}^J |J, M\rangle \langle J, M|, \quad (\text{S25})$$

where  $E_M$  is the energy of the  $|J, M\rangle$  state now including the shift defined in Eq. (10) of the Article. Instead of having a generic ladder operator  $\hat{J}_\pm$  that moves any state  $|J, M\rangle \rightarrow |J, M \pm 1\rangle$ , with emission or absorption at  $\omega_A$ , we now have to break up this operator, because the Dicke transitions are no longer degenerate in energy. The generic ladder operators are thus replaced by a sum over operators, which take us between specific Dicke states, sampling the spectral density at the requisite frequency  $\omega_\beta$ :

$$\hat{L}_M = |J, M-1\rangle \langle J, M|, \quad (\text{S26})$$

which yields the result in the main paper Eq. (7):

$$\dot{\rho} = -i[\hat{H}_S + \hat{H}_I, \rho] - \gamma \sum_{\beta} \kappa(\omega_\beta) \left( (n(\omega_\beta) + 1) D[\hat{L}_\beta] \rho + n(\omega_\beta) D[\hat{L}_\beta^\dagger] \rho \right). \quad (\text{S27})$$

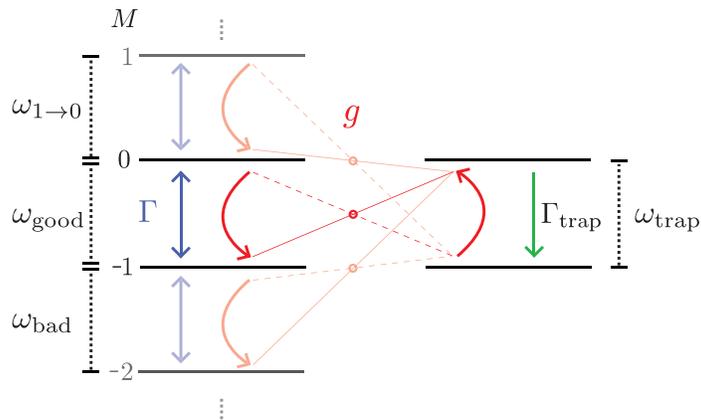


FIG. S1: A trap site is connected to the Dicke ladder. The trap’s transition frequency ideally matches that of the ‘good’ transition  $\omega_{\text{trap}} \approx \omega_{\text{good}}$ , and it is coupled to the Dicke transitions via a flip-flop interaction of strength  $g$ . This gives rise to ‘see-saw’ like oscillations between Dicke and trap transitions, but only the desired transition is resonant, all others are detuned and thus suppressed. To ensure that excitons hopping to the trap site are irreversibly removed instead of ‘see-sawing’ back and forth indefinitely, the trap is incoherently emptied at a rate  $\Gamma_{\text{trap}}$ . As long as  $\Gamma_{\text{trap}}$  is not so large that the trap transition experiences significant lifetime broadening approaching  $|\omega_{\text{good}} - \omega_{\text{bad}}|$ , and also assuming  $|\omega_{\text{good}} - \omega_{\text{bad}}| > g$ , the exciton extraction from the ‘bad’ transition in particular can be suppressed. The blue double-headed arrows indicate the (enhanced) optical emission and absorption processes from the main paper.

### Trapping

In the main text we describe the trapping process using a Lindblad operator, which takes the system from the state  $|J, 0\rangle \rightarrow |J, -1\rangle$  by irreversibly removing one exciton from the system. Numerous microscopic mechanisms can be conceived of that would produce this effect; all will have to involve a collective coupling of the atoms of the ring, followed by a process (through coupling to a wider external environment) which prevents the return of exciton to the primary system, or at least renders it very unlikely.

Here, we give an account of the simplest scenario one can envision: all atoms of the ring are coupled to a ‘trap’ atom at the centre, whose role is to first localise the energy and then irreversibly remove it. This simple trapping model is schematically depicted in Fig. S1: The trap atom possesses a transition frequency  $\omega_{\text{trap}} \approx \omega_{\text{good}}$  and is coupled to the ring by a field mediated hopping interaction of strength  $g$  (i.e. the same type of interaction which couples the ring’s atoms to each other). An exciton ‘hopping’ onto the trap site is subjected to an irreversible decay with rate  $\Gamma_{\text{trap}}$ , e.g. by being linked to a chain of exciton sites acting as a wire or lead. In natural light harvesting systems the trap would be the reaction centre and the decay a photochemical process.

When the trapping rate  $\Gamma_{\text{trap}}$  is sufficiently fast only negligible population exists in the 2LS forming the trap, hence its effect can, to a good approximation, be considered that of a Lindblad operator acting on the main system. For a slower rate  $\Gamma_T$  a decaying Rabi oscillation may take place, moving the exciton back and forth between trap and ring. However, the presence or absence of these oscillations does not significantly affect the superabsorption process.

This most simplistic trapping model introduces an extra contribution to the rate of loss from the E2LS: the finite lifetime  $1/\Gamma_{\text{trap}}$  means the energy of the trap is not perfectly sharply defined, reducing the trap’s frequency selectivity. Thus, it will occasionally also accept energy from the ‘bad’ transition  $\omega_{\text{bad}}$ , which increases the effective loss rate  $\Gamma_{\text{loss}}$ . However, one can optimise the parameters ( $\Gamma_{\text{trap}}, g, \omega_{\text{trap}}$ ) to minimise this undesirable side effect, whilst still meeting the condition that  $\Gamma_{\text{trap}} > \Gamma_{\text{emit}}$ .

### Resolution of Frequency Shifts

The enhanced absorption and emission rate in the middle of the Dicke ladder implies an increased lifetime broadening. One might thus worry whether the detunings obtained courtesy of the hopping interaction are then still sufficient to completely resolve adjacent transitions. A simple analysis shows that this is indeed the case [1], and the natural width ( $N^2\gamma$  around  $M = 0$ ) remains smaller than the shift  $\delta\omega$  provided the wavelength of the light is much greater than the size of the system. To see this, we shall assume that the transition is indeed well resolved,  $N^2\gamma < \delta\omega$ , and show that is essentially equivalent to the ‘small sample’ condition,  $r \ll \lambda$ , which underlies the phenomenon of superradiance in the first place.

Using the definition of  $\gamma$  from as the free atom decay rate (see Article), the greatest broadening and smallest energy shift at  $M = 0$  are, respectively,

$$N^2\gamma = \frac{8N^2\pi^2d^2}{3\epsilon_0\hbar\lambda^3}, \quad (\text{S28})$$

$$\delta_\omega = 4\frac{\Omega}{N-1} \approx \frac{d^2}{4\pi\hbar\epsilon_0r^3}. \quad (\text{S29})$$

Substituting Eqs. (S28) and (S29), the inequality  $N^2\gamma < \delta_\omega$  becomes

$$\frac{2N^3\pi^2d^2}{3\epsilon_0\hbar\lambda^3} < \frac{d^2}{4\pi\hbar\epsilon_0r^3} \iff 2N\pi r < \lambda, \quad (\text{S30})$$

where the righthand side follows after cancellation of several variables followed by taking the cubic root. This is equivalent to  $r \ll \lambda$ , up to moderate numerical factor (when  $N$  is not too large), accounted for by relaxing ‘ $\ll$ ’ to ‘ $<$ ’. For the present discussion,  $r$  is understood to be the nearest neighbour distance, having assumed energy shifts appropriate for only nearest neighbour interactions (for other interaction models, the detunings would be larger). We note that distinct shifted lines have also already been observed — and resolved — experimentally [5].

## II. INTERACTIONS BEYOND THE NEAREST NEIGHBOUR LIMIT

In the main text we assumed only nearest neighbour interactions are significant. For a symmetric ring geometry relaxing this condition leads to the same qualitative behaviour, but results in slightly larger detunings between adjacent transitions in the Dicke ladder. First, let us consider the opposite limit to the nearest neighbour case and allow all pairwise interactions with equal strength:

$$\langle J, M | \hat{H}_H | J, M \rangle = \langle J, M | \Omega \sum_{i \neq j} (\sigma_i^+ \sigma_j^- + \sigma_i^- \sigma_j^+) | J, M \rangle, \quad (\text{S31})$$

$$\delta E_m = \Omega \langle J, M | \sum_{i \neq j} (\sigma_i^+ \sigma_j^- + \sigma_i^- \sigma_j^+) | J, M \rangle, \quad (\text{S32})$$

which can be rewritten using the collective operators as follows

$$\delta E_m = \Omega \langle J, M | J_+ J_- + J_- J_+ - \sum_i (\sigma_i^+ \sigma_i^- + \sigma_i^- \sigma_i^+) | J, M \rangle. \quad (\text{S33})$$

The final two terms are added to remove the  $i = j$  terms implicit in the  $J_+ J_-$  terms, which count the number of excited and unexcited atoms, respectively. Hence,

$$\delta E_m = \Omega (\langle J, M | J_+ J_- + J_- J_+ | J, M \rangle - 2J). \quad (\text{S34})$$

The remaining two terms are easily calculated from Eq. (4) of the Article, yielding:

$$\delta E_m = 2\Omega(J^2 - M^2). \quad (\text{S35})$$

Thus the energy shifts are the same as in the nearest neighbour case Eq. (10) but lack the factor  $(J-1/2)^{-1}$ . Therefore, unlike in the nearest neighbour limit, increasing the number of atoms does not reduce the size of the frequency shift, which could help in blocking the transition at  $\omega_{bad}$  and ensuring frequency selectivity of a trapping mechanism.

The actual ring geometry with all pairwise dipole interactions included will fall somewhere in between these two limits, depending on ring size. The operators involved in the interaction remain the same, but their weights are altered as the size of the ring changes. The symmetry of the ring dictates that each atom will be subject to the same set of interactions with the rest of the ring. The condition of interchangeability of atoms is thus met regardless of the specific interaction model (i.e. nearest neighbour, next nearest neighbour etc.). For all cases, the hopping interaction only causes shifts of a variable size between the two limits we have discussed above; the size of the shifts given a particular ring size and interaction model is readily obtained numerically.

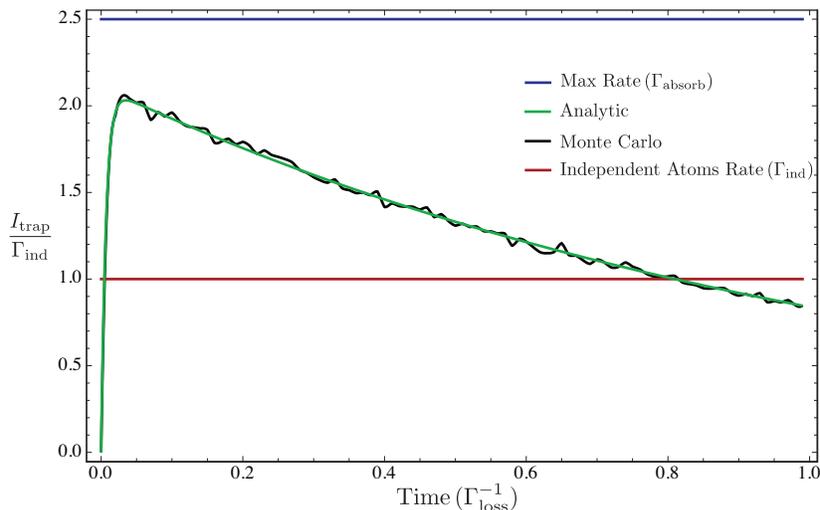


FIG. S2: Comparison of the Effective Two Level System (E2LS) model with numerical results from Monte Carlo simulations done using the Quantum Optics Toolbox in Python (QuTip). Parameters:  $N = 8$ ,  $N(\omega_{\text{good}}) = 10$ ,  $\Gamma_T = 10\Gamma_E$ ,  $\gamma = 1$ , trajectories 100,000.

### The Implicit interaction Approach

In the main paper, the hopping interaction emerged from the derivation of the master equation, as a direct consequence of embedding the absorbers into the photon field environment. Alternatively, we could add an interaction to the Hamiltonian, which may either be mediated by virtual photon exchange or have some other physical origin. The initial Hamiltonian then reads:

$$H_S = \omega_A \sum_{m=1}^N \hat{\sigma}_+^m \hat{\sigma}_-^m + \Omega \sum_{i,j}^N (\hat{\sigma}_+^i \hat{\sigma}_-^j + \hat{\sigma}_-^j \hat{\sigma}_+^i). \quad (\text{S36})$$

Such a Hamiltonian is diagonalised using the Jordan-Wigner transformation [6]. For example, a four atom system has eigenvalues:

$$\left\{ 0, \omega_A - 2\Omega, 2\omega_A - 2\sqrt{2}\Omega, 3\omega_A - 2\Omega, 4\omega_A \right\}, \quad (\text{S37})$$

resulting in the following transition frequencies:

$$\omega = \left\{ \omega_A - 2\Omega, \omega_A - 2\Omega \left( 1 + \sqrt{2} \right), \omega_A + 2\Omega \left( -1 + \sqrt{2} \right), \omega_A + 2\Omega \right\}. \quad (\text{S38})$$

These differ slightly from those derived using the approach in the main paper. Crucially, however, the degeneracy of the transition frequencies is broken in a similar way as before, so that the discussion of environmental control for confining the dynamics to a specific transition remains valid. For small length scale linear systems it has been noted that superradiance dynamics are not significantly altered [6], when compared to the traditional field mediated interaction approach [1], used in the main text.

### Numerical Results

The Effective Two Level System (E2LS) model in the main paper allowed us to reduce the complexity of the problem dramatically and made it analytically tractable. In order to verify this approach we compare it to an independent numerical model. Fig. S2 shows excellent agreement between the E2LS model and Monte Carlo simulations of the master equation (S27) [Eq. (7) of the Article] using QuTip [7]. The numerical model uses a phenomenological trapping. The agreement with the E2LS can be made arbitrarily close with increasing numbers of trajectories. The same agreement was also seen for systems with larger numbers of atoms.

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