Supplementary information

Many-body cavity quantum electrodynamics with driven inhomogeneous emitters

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Supplementary Information

1 System parameters

The ion-cavity coupling rate $g$ has a particular distribution, arising from inhomogeneity of the cavity field across the device. Simulating the cavity structure in COMSOL, the total YVO$_4$ volume is 3.058 um$^3$, with total coupling rate $\Omega = 2\pi \times 9$ GHz, calculated assuming uniformly distributed Yb ions across the YVO$_4$. In order to obtain the distribution of $g$, we generate the histogram of varying $g$ across the entire nanobeam volume of 3.058 um$^3$. Since there are many ions with very small $g$, we ignore ions with $g < 1.39$ MHz which retains 98.4% of $\Omega$, resulting in a total coupling rate of $\Omega = 2\pi \times 8.86$ GHz and the distribution shown in Fig. S1. The considered ions occupy a physical volume of 0.636 um$^3$ and their total number is $\approx 7 \times 10^5$. From this, the root mean square of $g$ is estimated to be $\sqrt{\langle g^2 \rangle} = \frac{\Omega}{\sqrt{N}} \approx 2\pi \times 10.6$ MHz, used later in the estimation of the number of ions participating in superradiance. Additionally, we calculate the optical mode volume to be $V_{\text{mode}} = 0.0918$ um$^3$, based on the maximum field strength.

![Figure S1: The simulated probability distribution of cavity-ion coupling strength $g$ in the device.](image)

| Parameter Description                           | Symbol | Value       |
|-----------------------------------------------|--------|-------------|
| Optical Frequency                             | $\omega$ | $2\pi \times 304500$ GHz |
| Spontaneous decay rate                        | $\gamma_s$ | $2\pi \times 0.6$ kHz |
| Excess dephasing rate                         | $\gamma_d$ | $2\pi \times 6$ kHz |
| Optical inhomogeneous linewidth (FWHM)        | $\Delta_{\text{inh}}$ | $2\pi \times 150$ MHz |
| Cavity energy decay rate                      | $\kappa$ | $2\pi \times 44$ GHz |
| Cavity external coupling rate                 | $\kappa_c$ | $2\pi \times 8.8$ GHz |
| Maximum Yb ion-cavity coupling rate           | $g$ | $2\pi \times 35$ MHz |
| Number of Yb ions in cavity                   | $N$ | 700000 |
| Cavity mode volume                            | $V_{\text{mode}}$ | $0.0918\mu$m$^3$ |
| Ensemble cooperativity                        | $C_A(C_I)$ | 12(24) |

Table S1: Relevant system parameters.

2 Theoretical Analysis of CIT

To understand the physical origins of the Collectively Induced Transparency (CIT) dip observed within the broad DIR peak, we start by analyzing simple cases with only a few ions, followed by the large ensemble case with an inhomogeneous distribution.
2.1 General formalism

We start with the Tavis-Cummings Hamiltonian for $N$ two-level systems coupled to a single cavity field under the rotating-wave approximation (setting $\hbar = 1$):

$$ H = \Delta_c a^\dagger a + \frac{1}{2} \sum_{j=1}^{N} \Delta_j \sigma_j^+ + \sum_{j=1}^{N} g_j (a^\dagger \sigma_j^- + \sigma_j^+ a) - i\sqrt{\kappa_c} A_{\text{in}} (a^\dagger - a). \quad (S1) $$

In our case, the two-level system consists of the ground ($|g\rangle$) and excited state ($|e\rangle$) of the A transition (see main text). Here $g_j$ is the $j^{th}$ ion-cavity coupling strength, $a$ is the bosonic cavity field operator, $\sigma_j^+$, $\sigma_j^-$ are the $j^{th}$ spin ladder operators and the Pauli-Z operators, respectively. Note that here we consider the general case where each ion has a different coupling strength $g_j$. $\Delta_c$ is the cavity-laser detuning and $\Delta_j$ is the $j^{th}$ ion-laser detuning, accounting for inhomogeneous broadening. $\sqrt{\kappa_c} A_{\text{in}}$ is the coupling from the input power $P_{\text{in}}$, which is associated with the cavity mean photon number in the absence of ions $\mu$, as:

$$ \sqrt{\kappa_c} A_{\text{in}} = \frac{\kappa_c}{2}\sqrt{\mu} \quad (S2) $$

where $\mu$ is defined as

$$ \mu = \frac{\kappa_c}{(\kappa/2)^2 + \Delta_c^2 \hbar^2} \approx \frac{\kappa_c}{(\kappa/2)^2 \hbar^2}. \quad (S3) $$

Here, three variables $A_{\text{in}}$, $P_{\text{in}}$ and $\mu$ all represent the excitation power, and they are related to each other by system parameters. We will use $\mu$ to represent power in the following theoretical analysis for brevity.

Starting with Eq. S1, we obtain the equations of motion for $a$, $\sigma_j^-$ and $\sigma_j^+$ in the Heisenberg picture, with dissipation terms, given as:

$$ \dot{a} = -i(\Delta_c + \frac{\kappa_c}{2})a - i\sum_{j=1}^{N} g_j \sigma_j^- - \frac{\kappa_c}{2}\sqrt{\mu} \quad (S4) $$

$$ \dot{\sigma}_j^- = -(i\Delta_j + \gamma)\sigma_j^- + ig_j \sigma_j^+ a \quad (S5) $$

$$ \dot{\sigma}_j^+ = 2ig_j (a^\dagger \sigma_j^- - \sigma_j^+ a) - \gamma_s (1 + \sigma_j^-) \quad (S6) $$

where $\gamma = \frac{\gamma_p}{2} + \gamma_d$. Using Eq. S2 and input-output formalism, $A_{\text{out}} = \sqrt{\kappa_c} \theta + A_{\text{in}}$, we obtain the cavity reflection:

$$ R = \left| \frac{\langle A_{\text{out}} \rangle}{\langle A_{\text{in}} \rangle} \right|^2 = \left| \frac{2\kappa_c}{\kappa}\sqrt{\mu} (\theta + 1) \right|^2 \quad (S7) $$

where cavity field $\theta$ is determined by solving Eqs. S4-S6 and thus depends on the state of ions.

2.2 Weak-excitation case (DIR)

Under the weak-excitation condition ($\langle \sigma_z \rangle \approx -1$), an analytical expression for the cavity reflection $R(\omega)$ as a function of laser frequency $\omega$ is derived in [1] as:

$$ R(\omega) = \left| 1 - \frac{i\kappa_c}{\omega - \omega_c + i\kappa/2 - W_A(\omega) - W_E(\omega) - W_I(\omega)} \right|^2 \quad (S8) $$

where $W_X$ accounts for the coupling of an $X$ ($X=A,E,I$) transition to each of the inhomogeneously broadened ensembles:

$$ W_X(\omega) = \frac{\Omega_X^2}{\omega - \omega_X + i\gamma_X + i\Delta_X/2}. \quad (S9) $$

Here, $\Delta_X$ is the full-width half-maximum (FWHM) of a Lorentzian distribution, $\Omega_X = \sqrt{\sum g_j^2}$ is the total ion-cavity coupling rate, $\gamma_X$ is the dephasing rate, and $\omega_X$ is the center frequency of an $X$ transition. From COMSOL simulations, we estimate that $\Omega_A = \Omega_E = \Omega_I/\sqrt{2} = 2\pi \times 4.4$ GHz, where the $1/\sqrt{2}$ factor in $\Omega_I$ comes from the double degeneracy in that ground state of the I transition. We plot the cavity reflection using Eq. S8 in Fig. S2, which qualitatively matches the experimental data in Extended Data Fig. 2c. From this, the cooperativity $C_X = \frac{W_X(\omega)/\kappa^2}{\kappa^2}$ [2] of the three transitions is computed as $C_{A,E} \approx 12, C_I \approx 24$. 

Figure S2: Theoretical cavity reflection as a function of laser detuning in the low excitation regime (DIR), showing ion-cavity coupling. Three DIR peaks (orange) corresponding to the A, E, and I transitions of a Yb ion are identified using Eq. S8, showing high cavity reflectivity.

2.3 Driven single-ion case

We would now like to see if CIT appears for just a few ions, starting by considering a single ion coupled to the cavity. Since there is only one ion, the subscript $j$ is dropped. In the fast-cavity limit, we adiabatically eliminate the cavity mode by setting $\dot{a} = 0$, and using the fact that $\Delta_c \ll \kappa$ we get

$$
a = \frac{-ig\sigma^+ - \frac{\kappa}{2} \sqrt{\mu}}{i\Delta_c + \frac{\kappa}{2}} \approx -\frac{ig\sigma^+}{\frac{\kappa}{2}} - \sqrt{\mu}.
$$

(S10)

Substituting Eq. S10 into Eq. S5 and Eq. S6, we obtain linear differential equations for $\sigma^z$ and $\sigma^-$:

$$
\dot{\sigma}^- = -(i\Delta + \gamma + \frac{2g^2}{\kappa})\sigma^- - ig\sigma^z \sqrt{\mu} - \frac{1}{4g^2\mu} \left( \gamma + \frac{2g^2}{\kappa} \right) \left( \frac{\gamma + \frac{2g^2}{\kappa}}{\left( \Delta + \gamma + \frac{2g^2}{\kappa} \right)^2} \right)
$$

(S11)

$$
\dot{\sigma}^z = -(\gamma_s + \frac{4g^2}{\kappa})(1 + \sigma^z) + 2ig\sqrt{\mu}(\sigma^+ - \sigma^-).
$$

(S12)

Then we solve the expectation value of the operators in the steady-state $\langle \dot{\sigma}^z \rangle = \langle \dot{\sigma}^- \rangle = 0$ as:

$$
\langle \sigma^z \rangle = \frac{ig\sqrt{\mu}}{\left( i\Delta + \gamma + \frac{2g^2}{\kappa} \right)} \left( 1 + \frac{4g^2\mu}{\left( \gamma_s + \frac{4g^2}{\kappa} \right) \left( \Delta + \gamma + \frac{2g^2}{\kappa} \right)^2} \right)
$$

(S13)

$$
\langle \sigma^- \rangle = \frac{ig\sqrt{\mu}}{\left( i\Delta + \gamma + \frac{2g^2}{\kappa} \right)} \left( 1 + \frac{4g^2\mu}{\left( \gamma_s + \frac{4g^2}{\kappa} \right) \left( \Delta + \gamma + \frac{2g^2}{\kappa} \right)^2} \right).
$$

(S14)

The cavity reflection is

$$
R = \left| \frac{4i\kappa_c}{\kappa^2 \sqrt{\mu}} g(\sigma^-) - \frac{\kappa_c}{i\Delta_c + \frac{\kappa}{2}} + 1 \right|^2
$$

(S15)

where $\frac{4i\kappa_c}{\kappa^2 \sqrt{\mu}} g(\sigma^-)$ is the contribution from the ion, which is related to the power, and $-\frac{\kappa_c}{i\Delta_c + \frac{\kappa}{2}} + 1$ is the contribution from the bare cavity. We plot the cavity reflection for varying $\sqrt{\mu}$ using Eq. S15 in Fig. S3a. We see that in the weak-excitation regime ($\sqrt{\mu} \to 0$, black), there is a DIR centered at the ion frequency, reaching unit cavity reflection. Upon increasing the power, the entire DIR profile decreases, and finally disappears, as the cavity reflection reaches the bare cavity value of $R = \left| \frac{\kappa_c}{i\Delta_c + \frac{\kappa}{2}} + 1 \right|^2$ ($R \approx 0.36$ for our case). This occurs when we fully saturate the ion.

2.4 Driven two and three ion cases

Since there is no dip for the single ion case, we add another ion, where the ions are now symmetrically detuned around 0 MHz, with detunings $\pm 2\pi \times 0.048$ MHz. The cavity reflection for $\sqrt{\mu} \to 0$ is plotted using an analytical expression under the
We define $\langle \sigma_j^z \rangle$ using the master equation (Section 3.1). First, we note that a weak-excitation condition, similar to Eq. S8. Additionally, we sweep the power from $\sqrt{\mu} = 10^{-4}$ to $\sqrt{\mu} = 10^{-2}$ and simulate using the master equation (Section 3.1). First, we note that $\sqrt{\mu} = 10^{-4}$ is very close to the weak-excitation limit. In this case, we observe two DIRs centered around each ion frequency. The two DIRs overlap at zero detuning where the ions destructively interfere, producing a narrow window even with low excitation, indicating that this is not exactly CIT (Fig. S3b). When $\sqrt{\mu}$ is increased, both DIRs decrease, similar to the single ion case.

We now add a third ion between the two ions at zero detuning, where a third DIR peak appears (Fig. S3c). At the same time, two sharp dips appear between the first/second ion and second/third ion. When $\sqrt{\mu}$ is increased, all three DIR peaks decrease where the center peak decreases the fastest. This eventually leads to a single, wide dip at $\sqrt{\mu} = 10^{-2.5}$. From the results of the two and three ion cases, we see hints of the origins of CIT from the destructive interference between different ions.

### 2.5 Driven multi-ion case

#### 2.5.1 Analytical derivation

From the simple cases analyzed above, we get the intuition that an inhomogeneously broadened ensemble of ions is required for a narrow dip to form. To see this more rigorously, we derive the analytical expression for CIT under certain conditions. We first set the steady state of Eqs. S4 - S6 ($\dot{\sigma} = \dot{\sigma}^z = \dot{\sigma}^+ = 0$), which gives $2N + 1$ equations for $2N + 1$ variables:

$$-(i\Delta_c + \frac{K}{2}) \langle a \rangle - i \sum_{j=1}^{N} g_j \langle \sigma_j^+ \rangle - \frac{K}{2} \sqrt{\mu} = 0 \quad (S16)$$

$$-(i\Delta_j + \gamma) \langle \sigma_j^- \rangle + ig_j \langle \sigma_j^z \rangle \langle a \rangle = 0 \quad (S17)$$

$$2ig_j (\langle a^1 \rangle \langle \sigma_j^- \rangle - \langle \sigma_j^+ \rangle \langle a \rangle) - \gamma_s (1 + \langle \sigma_j^z \rangle) = 0. \quad (S18)$$

Note that here we adopt the hypothesis that quantum correlation between atomic and cavity field operators can be neglected ($\langle \sigma_j^z a \rangle = \langle \sigma_j^z \rangle \langle a \rangle$ and $\langle a^1 \sigma_j^- \rangle = \langle a^1 \rangle \langle \sigma_j^- \rangle$) [3]. Eliminating $\langle \sigma_j^- \rangle$, we get the equation relating $\langle \sigma_j^z \rangle$ and $\langle a \rangle$ as:

$$\langle a \rangle = \frac{-\sqrt{\mu}}{1 - \sum_{j=1}^{N} \frac{2g_j^2 \langle \sigma_j^z \rangle}{\kappa(\gamma + i\Delta_j)}} \quad (S19)$$

$$\langle \sigma_j^z \rangle = -\frac{1}{1 + \frac{4g_j^2 \langle a^1 \rangle \langle a \rangle}{\gamma_s} \left(\frac{\gamma^2 + \Delta_j^2}{\gamma^2 + \Delta_j^2 + \gamma^2}\right)} \quad (S20)$$

We define $\langle a \rangle = -\frac{\sqrt{\mu}}{1 + \frac{\gamma^2}{\Delta_j^2}}$, where

$$x = -\sum_{j=1}^{N} \frac{2g_j^2 \langle \sigma_j^z \rangle}{\kappa(\gamma + i\Delta_j)} = \sum_{j=1}^{N} \frac{2ig_j \langle \sigma_j^- \rangle}{\kappa \langle a \rangle} \quad (S21)$$

Figure S3: a, Cavity reflection spectrum of a single ion coupled to the cavity with rate $g = 2\pi \times 35$ MHz, using Eq. S15. DIR is observed for small powers, and decreases with increasing powers. b, Simulation of the cavity reflection spectrum of two symmetrically detuned ions at $\pm 2\pi \times 0.048$ MHz. There are two DIR peaks at each of the ion frequencies, and destructive interference between two ions form a dip at 0 MHz. c, Three ion simulation, adding an ion at 0 MHz. The dip from the two ion case disappears at low powers due to DIR from the added third ion.
which represents the ions’ response to the cavity field. Here, we also ignore \( \Delta_c \) for simplicity based on the fact that \( \Delta_c \ll \kappa \).

We can get an implicit equation for \( x \) as:

\[
x = \sum_{j=1}^{N} \frac{2g_j^2}{\kappa (\gamma + i \Delta_j)} \left( \frac{1}{1 + \frac{4g_j^2 \mu}{\gamma (\gamma + \Delta_j)}} \right)
\]  
(S22)

where the distribution of coupling strength \( g_j \) is not correlated with the distribution of ion frequency detuning \( \Delta_j \), as the physical position of the ion in the cavity is unrelated to its resonance frequency. We first consider \( g_j = g \) and define \( y(x) = \frac{4g^2 \mu \gamma}{1 + x^2 \gamma} \) for simplicity, and convert the summation to integral in the limit of large \( N \):

\[
x = \frac{2Ng^2}{\kappa} \int \frac{\rho(\omega) d\omega}{(\gamma + i(\omega - \omega_L)) \left( 1 + \frac{y(x)}{\gamma + (\omega - \omega_L)^2} \right)}
\]  
(S23)

where \( \omega_L \) is laser frequency and \( \rho(\omega) \) is the probability distribution of a given ion frequency \( \omega \). Given \( x \), the cavity reflection is now simply

\[
R = \left| - \frac{2\kappa_c}{\kappa (1 + x)} + 1 \right|^2.
\]  
(S24)

We consider a Lorentzian distribution of ions with \( \rho(\omega) = \frac{\Delta_{\text{inh}}/2}{\pi (\omega - \omega_0)^2 + \frac{1}{4}} \) where \( \Delta_{\text{inh}} \) is the FWHM and \( \omega_0 \) is the center frequency of the ion distribution, which gives us:

\[
x = \frac{N \Delta_{\text{inh}} g^2 \pi \kappa}{\mu \gamma} \int_{-\infty}^{+\infty} \frac{(\gamma - i(\omega - \omega_L)) d\omega}{(\gamma^2 + (\omega - \omega_L)^2 + y(x)) \left( (\Delta_{\text{inh}}/2)^2 + (\omega - \omega_0)^2 \right)}.
\]  
(S25)

Making the approximations \( \Delta_{\text{inh}} \gg |\omega_L - \omega_0|, \gamma, \sqrt{y(x)} \) and \( y(x) \gg \gamma^2 \) as well as \( \sqrt{\mu} > \frac{2Ng \gamma}{\kappa \Delta_{\text{inh}}} \) (summarized and justified below), allows us to solve for \( x \) explicitly:

\[
x = \frac{1}{\Delta_{\text{inh}} \sqrt{\frac{\mu \gamma}{\pi \kappa}}} \left( 1 + \frac{8i(\omega_L - \omega_0)}{\Delta_{\text{inh}} \sqrt{\gamma + \Delta_{\text{inh}}}} \right).
\]  
(S26)

Plugging Eq. S26 into Eq. S24, we get the explicit expression of the reflectivity relative to the laser frequency \( \omega_L \) as:

\[
R = 1 + \frac{\kappa_c (\Delta_{\text{inh}}/C)^2}{\frac{\Delta_{\text{inh}}}{4g^2} \left( 1 - \frac{C \Delta_{\text{inh}}}{\Delta_{\text{inh}}} \right)}
\]  
(S27)

where \( C = \frac{|W(\omega=\omega_0)|}{\kappa/2} \) (\( = \frac{4Ng^2}{\kappa \Delta_{\text{inh}}} \) when \( \Delta_{\text{inh}} \gg \gamma \)) and

\[
\Delta_{\text{CIT}} = C \frac{\Delta_{\text{inh}}}{1 - C \sqrt{\frac{\gamma \gamma}{4g^2 \mu}}}.
\]  
(S28)

According to Eq. S27, it is apparent that the profile of the CIT dip is a Lorentzian function with width \( \Delta_{\text{CIT}} \) and depth \( \eta_{\text{CIT}} \), which is determined by the reflectivity at \( \omega_L = \omega_0 \) as:

\[
\eta_{\text{CIT}} = \frac{A}{(\Delta_{\text{inh}}/2)^2 \eta_{\text{bare}}}
\]  
(S29)

\[
= \frac{1}{(1 - \frac{\kappa_c}{\kappa})} \left( 1 - C \sqrt{\frac{\gamma \gamma}{4g^2 \mu}} \right) \left( 1 - C \sqrt{\frac{\gamma \gamma}{4g^2 \mu}} \right)
\]

where the depth has been normalized against the bare cavity depth \( \eta_{\text{bare}} = 1 - (1 - \frac{\kappa_c}{\kappa})^2 \). The lower bound of \( \Delta_{\text{CIT}} \) is \( \Delta_{\text{CIT, min}} = \frac{\Delta_{\text{inh}}}{4g^2} \), as the laser power \( \mu \) becomes large. Additionally in the same limit, the depth reaches 1. Here we summarize the assumptions made to derive the analytical expressions of the CIT width and depth.

- \( C \gg 1 \) High cooperativity
- \( \frac{\Delta_{\text{inh}}}{\gamma} \gg \mu \) Intermediate power
- \( \frac{\Delta_{\text{inh}}}{\gamma} \gg C \) Appreciable inhomogeneity and good coherence
We note that for a uniform (rectangular) ensemble distribution, the CIT depth and width have the same power and cooperativity dependence up to a factor of $\frac{\pi}{2}$. From this we anticipate CIT to be a ubiquitous phenomenon for any distribution satisfying Eq. S30.

### 2.5.2 Inhomogeneity of $g$ distribution

For simplicity, we have so far assumed that all ions are coupled equally to the cavity ($g_j = g$). Now we consider inhomogeneously distributed $g$, with a probability distribution given by $p(g)$. Using the fact that $p(g)$ is uncorrelated with $\rho(\omega)$, Eq. S23 can be written as:

$$x = \frac{2}{\kappa} \int N g^2 p(g) dg \int \rho(\omega) d\omega \frac{\rho(\omega)}{\gamma + i(\omega - \omega_L)} \left(1 + \frac{y(x, g)}{\gamma^2 + (\omega - \omega_L)^2}\right).$$

(S31)

Note that $y(x, g)$ is now also a function of $g$. We can first calculate the integral over frequency $\omega$, then over $g$ to get the solution of $x$ similar to Eq. S26:

$$x = \frac{1}{\frac{\Delta_{\text{inh}}^2}{2 N g_{\text{inh}}} \sqrt{\frac{\mu}{\gamma^2}} - 1} + \frac{8 i \omega_L}{\Delta_{\text{inh}}^2}$$

(S32)

where $\omega^2 = \int N g^2 p(g) dg$, and we define $g_{\text{avg}} = \int gp(g) dg$. Now, we obtain the width

$$\Delta_{\text{CIT}} = \Delta_{\text{inh}} \frac{\Delta_{\text{inh}}}{1 - 2 N g_{\text{inh}} \sqrt{\frac{\gamma^2}{\mu}}}.$$ (S33)

From this, we can see that the CIT profile and the minimum width do not depend on the specific distribution of cavity-ion coupling strength, but only on the total coupling strength. Ensemble cooperativity $C \propto \frac{\Delta_{\text{inh}}}{\kappa_{\text{inh}}}$ also only depends on the total coupling strength $\Omega^2$, so the expression of the minimum FWHM relative to the cooperativity will still be $\Delta_{\text{CIT, min}} \sim \frac{\Delta_{\text{inh}}}{C}$, regardless of the specific distribution of $g$.

### 2.5.3 Intuitive understanding

From the above derivations, we see that for high cooperativity ($C > 1$), we will get a CIT dip with width approximately equal to the inhomogeneous linewidth divided by $C$. Looking back at Eq. S21, the contribution of each ion to $x$ can be extracted using Eq. S17 (still assuming $g_j = g$ since $g_j$ and $\Delta_j$ are uncorrelated):

$$\langle \sigma_{\Delta_j} \rangle = \langle a \rangle = \frac{i g \Delta_j (\gamma - i \Delta_j)}{\gamma^2 + \Delta_j^2}.$$ (S34)

Here, we use notation $\langle \sigma_{\Delta_j} \rangle = \langle \sigma_{\Delta_j} \rangle_a$ and $\langle \sigma_{\Delta_j}^* \rangle = \langle \sigma_{\Delta_j}^* \rangle_a$, where the subscript now denotes the ion-laser detuning. For ions with detuning much larger than a homogeneous linewidth $|\Delta_j| \gg \gamma$, if a ion is blue-detuned ($\Delta_j > 0$), its atomic coherence phase $\arg \left(\frac{\sigma_{\Delta_j}}{\langle a \rangle}\right) \rightarrow -\pi$ while its red-detuned pair has $\arg \left(\frac{\sigma_{-\Delta_j}}{\langle a \rangle}\right) \rightarrow 0$. Their amplitudes are $\left|\langle \sigma_{\Delta_j} \rangle_a\right| \sim \left|\langle \sigma_{-\Delta_j} \rangle_a\right| \sim 1$.

We can see that for the above pair of ions with $\pm \Delta_j$, the phase difference will be $\pi$, such that their phases will cancel and their contribution to the cavity field will be

$$\langle \sigma_{\Delta_j} \rangle \sim \frac{2 g \gamma}{\gamma^2 + \Delta_j^2} \sim \frac{2 g \gamma}{\Delta_j^2} |\langle \sigma_{\Delta_j} \rangle_a^*|.$$ (S35)

Note that the phase we talk about here is the phase of $\langle \sigma_{\Delta_j} \rangle$ relative to $\langle a \rangle$. For the Fig. 2a in the main text, we add a global phase of $\frac{\pi}{2}$ to the ion phases for visual clarity.

To summarize,

1. The contribution from any ion to cavity reflection will decrease as the ion gets excited ($\langle \sigma_{\Delta_j} \rangle > -1$), and finally disappear when the ion is saturated to the completely mixed state where $\langle \sigma_{\Delta_j} \rangle = \langle \sigma_{-\Delta_j} \rangle = 0$.

2. The phase of each ion relative to the cavity field is determined by the detuning of an ion relative to the laser frequency. For blue-detuned ions ($\Delta_j > 0$), the phases are between $-\frac{\pi}{2}$ to $-\pi$, while for the red-detuned ions ($\Delta_j < 0$), the phases are between $-\frac{\pi}{2}$ to $0$.

3. A pair of symmetrically detuned ions relative to the laser frequency will have a reduced contribution to the cavity field compared to a single ion due to the phase cancellation, by a factor of $\sim \frac{1}{\Delta_j^2}$. This is obtained by comparing Eq. S35 to Eq. S34 in the case of $|\Delta_j| \gg \gamma$. 
2.5.4 Mean-field approximation

In the above derivation of CIT, we have used the mean-field approximation [4, 5] where the mean values of the products of the operators $a$ and $\sigma^z_j$ are factorized: $\langle \sigma^z_j a \rangle = \langle \sigma^z_j \rangle \langle a \rangle$. In the following, we will justify why such an approximation can be made for this system when considering CIT. To start, we introduce the term $\langle \sigma^z_j a \rangle - \langle \sigma^z_j \rangle \langle a \rangle$ which considers the correlations between $a$ and $\sigma^z_j$, and we would like to show that this term is negligible. Using adiabatic elimination $a \approx -\frac{2i}{\kappa} \sum_{k=1}^N g_k \sigma^z_k$ we expand this as:

$$\langle \sigma^z_j a \rangle - \langle \sigma^z_j \rangle \langle a \rangle = -\frac{2i}{\kappa} \sum_{k=1}^N g_k \left( \langle \sigma^z_j \sigma^z_k \rangle - \langle \sigma^z_j \rangle \langle \sigma^z_k \rangle \right)$$

$$= -\frac{2i}{\kappa} \left( g_j \left( \langle \sigma^z_j \sigma^z_j \rangle - \langle \sigma^z_j \rangle \langle \sigma^z_j \rangle \right) + \sum_{k \neq j} g_k \left( \langle \sigma^z_j \sigma^z_k \rangle - \langle \sigma^z_j \rangle \langle \sigma^z_k \rangle \right) \right)$$

(S36)

The first term is the self-correlation between $\sigma^z_j$ and $\sigma^z_j$ of ion $j$, and the second term is the cross-correlation between different ions. Setting the second term $\langle \sigma^z_j \sigma^z_k \rangle - \langle \sigma^z_j \rangle \langle \sigma^z_k \rangle = 0 (k \neq j)$ indicates that the final states of different ions are not entangled. For the first term (self-correlation), we use the commutation rule between $\sigma^z_j$ and $\sigma^z_j$ ($\sigma^z_j \sigma^z_j = -\sigma^z_j \sigma^z_j$) to rewrite it as

$$-\frac{2ig_j}{\kappa} \left( \langle \sigma^z_j \sigma^z_j \rangle - \langle \sigma^z_j \rangle \langle \sigma^z_j \rangle \right) = \frac{2ig_j}{\kappa} \left( 1 + \langle \sigma^z_j \rangle \right) \langle \sigma^z_j \rangle$$

(S37)

It is true that this term can be nonzero based on the solution in CIT. However, we now want to show that the influence of this term of the final cavity field $\langle a \rangle$ is very small. To do this, we introduce $\delta(\langle \sigma^z_j \rangle)$, which is the modification to $\langle \sigma^z_j \rangle$ if we were to consider this self-correlation term. Using Eq. S17,

$$\delta(\langle \sigma^z_j \rangle) = -\frac{2g_j^2}{(i \Delta_j + \gamma)\kappa} (1 + \langle \sigma^z_j \rangle) \langle \sigma^z_j \rangle$$

(S38)

We can then plug this back into our expression for the cavity field $a$ using Eq. S16, and get the modification to the cavity field introduced by this self-correlation:

$$\delta(\langle a \rangle) = -\frac{2i}{\kappa} \sum_{k=1}^N g_k \delta(\langle \sigma^z_k \rangle)$$

$$= \frac{4i}{\kappa^2} \sum_{k=1}^N g_k^2 (1 + \langle \sigma^z_k \rangle) \langle \sigma^z_k \rangle$$

$$= -\frac{4}{\kappa^2} \sum_{k=1}^N g_k^4 \left( \frac{1}{(i \Delta_k + \gamma)^2} (1 + \langle \sigma^z_k \rangle) \langle \sigma^z_k \rangle \langle \sigma^z_k \rangle \right)$$

(S39)

If we plug solution Eq. S20 into Eq. S39, we get

$$\delta(\langle a \rangle) = \frac{4}{\kappa^2} \sum_{k=1}^N g_k^4 \left( \frac{1}{(i \Delta_k + \gamma)^2} \frac{4g_j^2 \langle \sigma^z_j \rangle^2 \gamma}{\gamma (\gamma^2 + \Delta_k^2)} \right)$$

(S40)

Assuming $g_k = g$ for all the ions, we can obtain the relative change of $\langle a \rangle$ is

$$\left| \frac{\delta(\langle a \rangle)}{\langle a \rangle} \right| < \frac{16g^6 \gamma}{\kappa^2 \gamma_s} \sum_{k=1}^N \frac{|\langle a \rangle|^2}{(i \Delta_k + \gamma)^2 (\gamma^2 + \Delta_k^2)} \sim \frac{16Ng^6 \gamma}{\kappa^2 \gamma_s \Delta_{in}^4} |\langle a \rangle|^2 \sim \frac{16Ng^6 \gamma}{\kappa^2 \gamma_s \Delta_{in}^4} \mu$$

(S41)

Using Eq. S30b where $\mu \ll \left( \frac{\Delta_{in}}{g} \right)^2 \frac{2N}{\gamma}$ we can then get $\left| \frac{\delta(\langle a \rangle)}{\langle a \rangle} \right| \ll \frac{Cg^2}{4\kappa \Delta_{in}} \ll 1$, which is validated both in experiment and simulation for our system. The conclusion is that $\left| \frac{\delta(\langle a \rangle)}{\langle a \rangle} \right| \ll 1$, or that the fractional change in the cavity field due to the self-correlation term will be very small.
2.5.5 Description of CIT in the good-cavity regime

We have so far focused on the resonant bad-cavity regime, such that \( \Delta_c = \omega_c - \omega_L \), the cavity detuning from the laser, was ignored. Here we generalize the CIT derivation to the case where \( \Delta_c \) is considered, and S23 becomes:

\[
x = \frac{2Ng^2}{\kappa + 2i\Delta_c} \int \frac{\rho(\omega) d\omega}{(\gamma + i(\omega - \omega_L))(1 + y(x)/(\gamma + i(\omega - \omega_L))^2)}
\]

(S42)

where now \( y(x) = \frac{4g^2\gamma}{(1+x^2\gamma^2)(1+4\Delta_c^2/\kappa^2)} \approx \frac{4g^2\gamma}{(1+x^2\gamma^2)(1+4\Delta_c^2/\kappa^2)} \), keeping the first order of \( \Delta_c/\kappa \). Solving for \( x \):

\[
x = \frac{1}{2g/C} \left( 1 - \frac{2i(\omega_c - \omega_L)}{\kappa} \right) + \frac{2i(\omega_L - \omega_0)}{\Delta_{\text{min}}/C}
\]

(S43)

Using this, we can write our cavity reflection coefficient as

\[r = 1 - \frac{2\kappa_c}{(\kappa + 2i(\omega_c - \omega_L))(1 + x)} = 1 - \frac{\Delta_{\text{CIT, min}} \kappa_c/\kappa}{\Delta_{\text{CIT}}/2 + i(\omega_L - \omega_{\text{center}})}\]

(S44)

From this the center of CIT and width of CIT are:

\[
\omega_{\text{center}} = \left( \frac{\omega_0 C}{\Delta_{\text{inh}}} - \frac{\omega_c}{\kappa} \right) \left( \frac{\Delta_{\text{inh}}}{\kappa} \right) \frac{1}{1 - \left( \frac{\Delta_{\text{inh}}}{\kappa} \right)} = \frac{1}{1 - \left( \frac{\Delta_{\text{inh}}}{\kappa} \right)} \omega_0 - \frac{\Delta_{\text{inh}}}{\kappa} \omega_c
\]

(S45)

\[
\Delta_{\text{CIT}} = \frac{\Delta_{\text{inh}}}{\kappa} \left( \frac{1}{1 - \left( \frac{\Delta_{\text{inh}}}{\kappa} \right)} \right) \Delta_{\text{min}}/C \rightarrow \Delta_{\text{CIT, min}} = \frac{\Delta_{\text{inh}}}{\kappa} \left( \frac{1}{1 - \left( \frac{\Delta_{\text{inh}}}{\kappa} \right)} \right)
\]

(S46)

The above derivations show the following:

1. The CIT spectrum is similar to a cavity resonance, with \( \Delta_{\text{CIT}} \) linewidth and input relative coupling ratio \( \Delta_{\text{CIT, min}} \kappa_c/\kappa \) . This means that CIT is a mechanism in which a new resonance can be produced from a cavity, where this new resonance will be much narrower than the original cavity, whilst maintaining the input relative coupling ratio.

2. This narrow feature can be used for precision sensing. The effective cavity (CIT) fluctuation is reduced by a factor of \( \frac{\Delta_{\text{inh}}}{\kappa_c} \rightarrow \Delta_{\text{inh}}/C/\kappa \) compared to the original cavity fluctuation.

3. The above derivation requires \( \Delta_{\text{inh}}/\kappa_c \ll 1 \) (besides the conditions in Eq. S30), which gives \( \Delta_{\text{inh}} \ll 2\sqrt{Ng} \). However, we can see that there is no individual requirement for \( \kappa_c \), so the CIT phenomenon is not necessarily restricted to the bad cavity regime, and can work for cavities with higher quality factors. We note that if the cavity becomes extremely narrow, the mean-field approximation may not hold and a more exact model may be required. It is unclear, however, if we must be in the mean-field regime for CIT to occur.

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Figure S4: **a**, CIT spectrum for different cavity detunings, fit to a Fano resonance. **b**, Relation between CIT detunings and cavity detunings, showing the expected linear dependence. The error bar is from the fit.
2.5.6 Response time of CIT and application for optical switch

In the previous sections, CIT has been investigated in the steady state regime. However, CIT has a finite response time, which is important to explore for certain applications such as an optical switch. To this end, we park the laser at the center of the CIT, and measure the reflect as a function of time (Extended Data FIG. 10).

We find a fast response time of 600 ns, and posit that a two-port optical switch with an integrated filter can be realized using CIT. This current device has not been optimized for this particular application, hence we are unable to measure the other port (transmission) nor apply a transverse pump. Additionally, the contrast suffers from reflection due to an imperfect beam overlap with our optical coupler and our cavity being under-coupled. However, this problem can be solved by critically coupling and coupling the laser better into the cavity. This will result in increased contrast, important for improving the extinction ratio of the switch. For the best extinction ratio, the cavity should be designed as critical-coupled where \( \kappa_1 / \kappa = \kappa_2 / \kappa = 0.5 \) where \( \kappa_1 \) and \( \kappa_2 \) are the coupling rate from port 1 and 2. We note that the pump light is to create the CIT, which is an absorptive process where the ions absorb light and get excited. In contrast, as long as the signal is weak enough, the optical switch can be non-absorptive. Furthermore, we believe the current switching time is limited by how fast the ions can reach saturation, which can be faster by engineering a larger \( g \).

2.5.7 Validity of the Tavis-Cummings model

It has been shown in [6] that large collective coupling rates can lead to a breakdown of the standard Tavis-Cummings model. The relevant condition for the validity of the Tavis-Cummings model is \( \text{FSR} \gg W \), where FSR is the free-spectral range of the cavity, and \( W \) is the ensemble absorption derived in Eq. S9. Plugging in the parameters of our system, we find \( \text{FSR} = 2\pi \times 25 \) THz, and \( W = 2\pi \times 0.5 \) THz, thus satisfying the above inequality, and validating our use of the Tavis-Cummings model.

3 Theoretical modelling for dynamics

3.1 Master equation formalism

For simulating the dynamics of a driven inhomogeneous ensemble, we use the same Hamiltonian in Eq. S1, and introduce various dissipative mechanisms through the Lindblad operators:

\[
\mathcal{L}_{\text{cav}} = \kappa (a \rho a^\dagger - \frac{1}{2} a^\dagger a \rho - \frac{1}{2} \rho a a^\dagger) \quad \text{(S47)}
\]

\[
\mathcal{L}_{\text{em}} = \gamma_s \sum_{j=1}^{N} (\sigma_j^- \rho \sigma_j^+ - \frac{1}{2} \sigma_j^+ \sigma_j^- \rho - \frac{1}{2} \rho \sigma_j^+ \sigma_j^- ) \quad \text{(S48)}
\]

\[
\mathcal{L}_{\text{deph}} = \gamma_d \sum_{j=1}^{N} (\sigma_j^z \rho \sigma_j^z - \frac{1}{2} \sigma_j^z \sigma_j^z \rho - \frac{1}{2} \rho \sigma_j^z \sigma_j^z ) = \gamma_d \sum_{j=1}^{N} (\sigma_j^z \rho \sigma_j^- - \rho) \quad \text{(S49)}
\]

where \( \mathcal{L}_{\text{cav}} \) is the cavity dissipation, \( \mathcal{L}_{\text{em}} \) is the local spontaneous emission, \( \mathcal{L}_{\text{deph}} \) is the local dephasing, \( \gamma_s \) is the single atom spontaneous emission rate, and \( \gamma_d \) is the excess dephasing rate. We operate in the fast cavity limit, where \( \kappa \gg g, \gamma_s, \gamma_d \), and adiabatically eliminate the cavity mode by setting \( a = 0 \). This allows us to replace the cavity field operator \( a \) with atomic operators as:

\[
a = \frac{-i \sum_{j=1}^{N} g_j \sigma_j^- - \frac{\kappa}{2} \sqrt{R}}{i \Delta_c + \frac{\kappa}{2}}. \quad \text{(S50)}
\]

We then rewrite \( H \) and \( \mathcal{L}_{\text{cav}} \) to \( H_{\text{at}} \) and \( \mathcal{L}_{\text{col}} \) (collective dissipation) in terms of atomic operators:

\[
H_{\text{at}} = \frac{\Delta_c}{(\kappa/2)^2 + \Delta_c^2} \sum_{j=1}^{N} g_j \sigma_j^+ \sum_{k=1}^{N} g_k \sigma_k^- + \Delta_c \mu + \frac{1}{2} \sum_{j=1}^{N} \Delta_j \sigma_j^z - \sum_{j=1}^{N} g_j \mu (\sigma_j^+ + \sigma_j^-) \quad \text{(S51)}
\]

\[
\mathcal{L}_{\text{col}} = \frac{\kappa}{(\kappa/2)^2 + \Delta_c^2} \sum_{j,k} g_j g_k (\sigma_j^- \rho \sigma_k^+ - \frac{1}{2} \sigma_j^+ \sigma_k^- \rho - \frac{1}{2} \rho \sigma_j^+ \sigma_k^- ). \quad \text{(S52)}
\]

We can further simplify \( H_{\text{at}} \) and \( \mathcal{L}_{\text{col}} \) by noting that \( \kappa \) is large and the cavity is tuned to be on resonance with our atomic transition \( (\Delta_c \rightarrow 0) \) as:
\[ H_{\text{at}} \approx \frac{1}{2} \sum_{j=1}^{N} \Delta j \sigma_j^z - \sum_{j=1}^{N} g_j \mu (\sigma_j^+ + \sigma_j^-) \]  

(S53)

\[ \mathcal{L}_{\text{col}} \approx \frac{4}{\kappa} \sum_{j,k} g_j g_k (\sigma_j^- \rho \sigma_k^+ - \frac{1}{2} \sigma_j^+ \sigma_k^- \rho - \frac{1}{2} \rho \sigma_j^+ \sigma_k^-). \]  

(S54)

The system master equation now reads:

\[ \dot{\rho} = -i [H_{\text{at}}, \rho] + \mathcal{L}_{\text{col}} + \mathcal{L}_{\text{em}} + \mathcal{L}_{\text{deph}}. \]  

(S55)

We solve this system using QuTIP [7]. For simulations of system sizes greater than 8, we make use of Permutational Invariance (PIQS, [8]) enabling larger simulations. However, since this requires atoms to be identical, we stick to the full simulation in cases where we add inhomogeneities or want to look at correlations between ions.

### 3.2 Excitation pulse length dependence

Here we study the non-steady state regime where the driving pulse length is short. We first theoretically investigate the atomic states during the driving period. In the case of 6 identical atoms, we plot the normalized cavity mean photon number \( \langle a^\dagger a \rangle \propto \langle J^+ J^- \rangle \) during the drive for three different powers (Fig. S5a).

For a low drive (Fig S5a, red), we see that \( \langle a^\dagger a \rangle \) quickly and monotonically increases to reach steady state. The steady-state population at this power consists primarily of lower excitation states, as the system is unable to be excited into the higher Dicke state manifolds. As we increase our drive, we see the appearance of Rabi-like oscillations among the Dicke states (Fig. S5a, yellow) and then a decay into steady state. The origin of this decay is two-fold: one is the dampening of the oscillations. This is due to collective decays (which go as \( g^2/\kappa \)) that drive the system towards equal population within a single vertical Dicke ladder. In other words, even in the absence of individual decay and dephasing, collective emission will spread the population out evenly among the superradiant states, leading to damped oscillations. Another decay type is the overall decay of the magnitude of \( \langle a^\dagger a \rangle \). This is due to the coupling to the subradiant subspace through individual decay and dephasing, similar to the experimentally observed decrease in peak counts with power in regime II.

![Figure S5](image_url)

**Figure S5:** a, Simulation of the cavity field population \( \langle a^\dagger a \rangle \) during a 50 \( \mu s \) pulse applied to a system of \( N = 6 \) identical ions, for different powers. The power increases from red (0.6 a.u.) to blue (1.0 a.u.) to yellow (1.5 a.u.) curves. Regardless of the power, here 50 \( \mu s \) is long enough for the cavity to enter steady state. b, Experimental data for pulse length dependence of peak counts, with 10 nW of power, showing steady-state behavior at 50 \( \mu s \).

We explore this experimentally by measuring the excitation pulse length dependence of peak counts, shown in Fig. S5b. We find a qualitative match to the simulation of the initial build up of photons as we populate the superradiant states, and then a decay towards a steady state as we pump into the subradiant subspace. However, despite increasing the pump power, we do not observe the oscillations seen in Fig. S5a for high power (yellow). We attribute this to an averaging effect over our ensemble, as the oscillation frequency depends on \( g \) and ion detuning, both inhomogeneous in our system.

### 3.3 \( N \)-dependence of S-curve

Using PIQS, we simulate the dependence of the S-curve with varying system size \( N \) (Fig. S6b). We find that as expected, the turning point between regimes I and II shifts towards larger power, as our Dicke space expands and it requires more power to populate the subradiant subspace. As a result, at smaller powers, larger number of ions will have smaller peak emission,
Figure S6: a, Extended data of Fig. 4b, showing more burning powers. For higher burning power, $N_{\text{res}}$ is larger and S-curve shifts towards the higher probe power. b, Simulation of varying ion number for the S-curve, showing a shift of the S-curve to higher powers. We find that, as expected, the turning point between regimes I and II shifts towards larger power, as our Dicke space expands and it requires more power to populate the subradiant subspace. As a result, at smaller powers, larger number of ions will have smaller peak emission, while the global maximum of peak emission increases for more ions.

while the global maximum of peak emission increases for more ions. This qualitatively matches what we see in the experiment (Fig. S6a). We note that regime III is not reproduced in this simulation as we do not consider the detuned ions.

3.4 Comparison to the saturation effect

An increase and subsequent decrease in emission can also occur due to the saturation of the atomic coherence (below, saturation effect), which occurs even for a single atom and is unrelated to the Dicke model. Here, we will show that we do not see the saturation effect, and explain the difference and the connection to the saturation effect. First, let us consider the single ion case, where we have already derived the analytical solution in Eq. S11 and Eq. S12.

We can see that the intensity of the coherence ($|\sigma^-|^2$) first increases and then decreases with power where power $P \propto g^2 \mu$. However, what is measured from cavity emission is

$$
\langle a^+a^- \rangle = \frac{4g^2}{\kappa^2} \left( \sum_{i=1}^{N} \langle \sigma_i^+ \sigma_i^- \rangle + \sum_{i \neq j}^{N} \langle \sigma_i^+ \sigma_j^- \rangle \right) = \frac{4g^2}{\kappa^2} \left( \sum_{i=1}^{N} \left( \langle \sigma_i^z \rangle + 1 \right) \frac{1}{2} + \sum_{i \neq j}^{N} \langle \sigma_i^+ \sigma_j^- \rangle \right)
$$

(S56)

Here we have applied Eq. S10 and the commutation rule. Then, by plotting both $|\sigma^-|^2$ and $\frac{|\sigma_{z+1}|}{2}$, we can see that while $|\sigma^-|^2$ has the non-monotonic trend, our measure $\frac{|\sigma_{z+1}|}{2}$ doesn't (Fig. S7). In other words, since we are not directly measuring the coherence by detecting the cavity emission, our S-curve cannot be explained by saturation of coherence.

Once we move to the multiple ions case, the measure becomes

$$
\langle a^+a^- \rangle = \frac{4g^2}{\kappa^2} \left( \sum_{i=1}^{N} \langle \sigma_i^+ \sigma_i^- \rangle + \sum_{i \neq j}^{N} \langle \sigma_i^+ \sigma_j^- \rangle \right) = \frac{4g^2}{\kappa^2} \left( \sum_{i=1}^{N} \left( \langle \sigma_i^z \rangle + 1 \right) \frac{1}{2} + \sum_{i \neq j}^{N} \langle \sigma_i^+ \sigma_j^- \rangle \right)
$$

(S57)

as we analyzed in Eq. 17 in the main text. The first term is monotonically increasing relative to driving power, so the existence of the second term is necessary to observe S-curve shape. This second term is correlation between distinct ions, and will be absent for the single ion case, hence distinct from the saturation effects.

4 Justification for photon emission fit function

Here we provide justification for the function used in Fig. 3 to fit the decay curves. To find the best fit, one could start with a naive guess of a bi-exponential, to capture the superradiant and subradiant decays. A representative curve with a bi-exponential fit is shown in Fig. S8a. We see that the fit fails due to the strong multi-exponential nature of the subradiant decay. We then try
Figure S7: Comparison between excited state population, $|\langle \sigma_z \rangle + 1|$, and the intensity of coherence, $|\langle \sigma^- \rangle|^2$, of a single atom as a function of excitation power.

a single stretched exponential fit in Fig. S8b. However, we still see that the fit fails at the crux between the slow and fast decay, shown in the inset.

To capture both the multi-exponential fast and slow decay, we now attempt a fast stretched exponential + slow stretched exponential (Fig. S8c). In particular, the fit function is $A_1 \exp[-(t/\tau_1)^{x_1}] + A_2 \exp[-(t/\tau_2)^{x_2}] + b$, where there is a fast decay with subscript 1 and slower decay with subscript 2. We find that this fits the data well in all power regimes, and most importantly does so with fit parameters that match our qualitative expectations from the picture provided in Fig. 3f in the main text. Additionally, while there are 7 fit parameters, we take several steps to minimize any overfitting of the data. First, the background $b$ is set to be the count level at a time much longer than the longest observed lifetime, essentially the dark counts and any leakage through our setup. Second, the existence of two decays is only relevant in regime II where there is a clear distinction between a fast and slow decay. In both regime I (III), the fast (slow) decay component is dominant over the other, effectively resulting in just a single stretched exponential.

Figure S8: Fit of an emission curve to a a, Bi-exponential, b, Stretched exponential, and c, Addition of two stretched exponentials. The blue and orange lines are from experiments and fits, respectively. Insets show early-time comparisons between the data and the fits.

5 Estimation of the number of ions participating in superradiance

From Fig. 3d, we see that $x$ approaches 1 as the decay becomes single exponential for very low powers. In this case, we expect to be primarily exciting the single-excitation manifold, leading to a decay rate of $N\Gamma_c$, where $N$ is the number of atoms in our Dicke space, and $\Gamma_c$ is the Purcell enhanced decay rate of a single atom. While $N$ (and thus $\tau$) changes continuously in this regime due to power broadening, we still can estimate an effective Dicke space.

In order to estimate $N$, we first show that the superradiant decay rate is $N$ times the average Purcell decay rate, given that our system has inhomogeneous $g$. To this end, we simulate the time dynamics of 6 ions with varying $g$, excited with low power (Fig. S9). We find that as expected, the superradiant decay time from the lowest manifold is given by $\frac{4N(g^2)}{\kappa}$, indicated by the overlap of the simulation and analytical time decay curves. From Section 1, we estimate the average Purcell enhancement by using $\sqrt{\langle g^2 \rangle} = 2\pi \times 10.6$ MHz, giving an average Purcell decay time of 15.6 µs. Meanwhile, we measured a decay time of
Figure S9: Master equation simulation of the decay of $N = 6$ ions with inhomogeneous $g$ (red) with low excitation. The three dashed lines (pink, black, green) are single exponential decays with decay rate $\frac{4N\langle g^2 \rangle}{\kappa}, \frac{4N\langle g^2 \rangle}{\kappa}, \frac{4N\langle g^3 \rangle^{2/3}}{\kappa}$, respectively. The results show that, as expected, the superradiant decay from the single excitation manifold is governed by the average of $g^2$ for the case of inhomogeneous $g$.

270 ns in the low power regime with $x \approx 1$ (Fig. 3d), from which we can estimate the effective number of ions participating in superradiance at this particular power to be $\sim 50$.

6 Cooperativity required to observe superradiance

For a general symmetric distribution of ions, $iW(\omega=\omega_0)/\rho(\omega=\omega_0) = \pi g^2$ [1] where $\rho(\omega = \omega_0)$ is the ion probability distribution at $\omega = \omega_0$, from which we obtain $C = |W(\omega=\omega_0)| = 2\pi g^2 \rho(\omega=\omega_0)/\kappa$. This ensemble cooperativity represents the ratio of the absorption rate to the cavity decay rate, which indicates the number of ions a photon can interact with before it leaks out of the cavity. The condition to observe superradiance in an inhomogeneous ensemble is roughly given by $\tau_R < T^*_2$ [9], where $\tau_R$ is the slowest superradiant decay time in the absence of inhomogeneity, and $T^*_2$ is the inhomogeneous dephasing time. Assuming there are $N_{\text{eff}}$ ions participating in superradiance, spanning a frequency $\Delta_{\text{eff}}$, this leads to the requirement that the superradiance decay rate $N_{\text{eff}} \frac{4g^2}{\kappa}$ must be larger than the effective bandwidth $\Delta_{\text{eff}}$ of the participating ions. For the ions around $\omega_0$, we know $N_{\text{eff}} = \Delta_{\text{eff}} N \rho(\omega = \omega_0)$. Using $N_{\text{eff}} \frac{4g^2}{\kappa} > \Delta_{\text{eff}}$, we obtain $4\pi g^2 N \rho(\omega = \omega_0) > 1$, giving an estimate of the cooperativity required to observe superradiance as $C > \frac{\pi}{2}$.

7 Cavity emission and reflection of the entire spectrum

Here we show pulsed excitation measurements at different laser frequencies, covering all of the transitions (Fig. S10). The laser frequency is swept from the lower frequency side of the A transition to the higher frequency side of I. In Fig. S10a we plot the reflected pulse amplitude and in Fig. S10b we show the integrated total emission counts. There are two aspects of the plot in Fig. S10b. Firstly, we find three tall peaks corresponding to A, E, and I. Zooming into one of these transitions, we can see that the spectral width of the emission is narrower than the inhomogeneous linewidth of the ensemble, and resembles more a flat top than a Gaussian/Lorentzian (see zoom in of Fig. S10b). This is because CIT creates a transparency window in the center of the inhomogeneous line, allowing more light to enter through that window. Thus, the CIT shapes the incoming light with width approximately equal to the CIT width (zoom in for Fig. S10a). Secondly, between the A and E, and E and I transitions, at detuning around 1.5 and 5.5 GHz, there are elevated counts where there are no resonant ions. This is related to the phase cancellation effect observed in CIT. Specifically, when there are balanced ions on either side of the laser frequency, more light can enter the cavity, resulting in enhanced off-resonant excitation for the same input power.

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Figure S10: Pulsed excitation with 47 nW as a function of laser detuning, where the a, reflected pulse amplitude and b, integrated emission counts after the pulse are extracted. The x-axis denotes the laser detuning relative to A transition. The other sharp peaks at ~ 4 and ~ 7 GHz correspond to E and I transitions, respectively. The green shaded area indicated the inhomogeneous ion distribution, clearly showing that the emission is nonlinear and narrower than the ion distribution due to phase cancellation.

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