Supplemental Material to
"A superatom picture of collective nonclassical light emission and dipole blockade in atom arrays"

L. A. Williamson,[1] M. O. Borgh,[2] and J. Ruostekoski[1]

[1]Department of Physics, Lancaster University, Lancaster LA1 4YB, United Kingdom
[2]Faculty of Science, University of East Anglia, Norwich NR4 7TJ, United Kingdom
(Dated: June 18, 2020)

S.A.   Formalism

The dynamics in the interaction picture for an array of N two-level atoms driven by a coherent laser field is described by the many-body quantum master equation (QME) for the reduced density matrix ρ [S1, S2],

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}\sum_j [H_j, \rho] + i\sum_{j\ell(\ell\neq j)} \Delta_{j\ell}[\hat{\sigma}_j^+\hat{\sigma}_\ell^-, \rho]$$
$$+ \sum_{j\ell} \gamma_{j\ell}\left(2\sigma_j^-\rho\sigma_\ell^+ - \sigma_\ell^+\sigma_j^-\rho - \rho\sigma_\ell^+\sigma_j^-\right). \quad (S1)$$

Here $\hat{\sigma}_j^+ = (\hat{\sigma}_j^-)^\dagger = |e\rangle_{jj}\langle g|$, $\hat{\sigma}_j^{ee} = \hat{\sigma}_j^+\hat{\sigma}_j^-$ are the atomic raising (lowering) and excited state population operators, with ground $|g\rangle_j$ and excited $|e\rangle_j$ states of atom j. The Hamiltonian operator

$$H_j \equiv -\hbar\delta\hat{\sigma}_j^{ee} - \mathbf{d}\cdot\mathcal{E}^+(\mathbf{r}_j)\hat{\sigma}_j^+ - \mathbf{d}^*\cdot\mathcal{E}^-(\mathbf{r}_j)\hat{\sigma}_j^-. \quad (S2)$$

describes the dynamics of a single atom at position $\mathbf{r}_j$ with the dipole moment $\mathbf{d} \equiv \mathcal{D}\hat{\mathbf{d}}$. Here $\mathcal{D}$ is the reduced dipole matrix element that we assume is real without loss of generality. The atoms are driven by a plane-wave drive with positive frequency component $\mathcal{E}^+(\mathbf{r}) = \frac{1}{2}\mathcal{E}_0 e^{i\mathbf{k}\cdot\mathbf{r}}\hat{\mathbf{e}} = [\mathcal{E}^-(\mathbf{r})]^*$. The drive field frequency ω is detuned from the single-atom transition frequency ω_0 by $\delta \equiv \omega - \omega_0$. Here the atomic and light fields are slowly varying, such that the rapidly rotating phase factors $e^{\pm i\omega t}$ are removed by moving into an interaction picture and making the rotating wave approximation (by omitting the fast co-rotating terms $\hat{\sigma}_m^- e^{2i\omega t}$, $\hat{\sigma}_m^+ e^{-2i\omega t}$). The single-atom dynamics is thus described by H_j together with the decay terms $\gamma(2\sigma_j^-\rho\sigma_j^+ - \sigma_j^+\sigma_j^-\rho - \rho\sigma_j^+\sigma_j^-)$, where $\gamma \equiv \mathcal{D}^2 k^3/(6\pi\hbar\epsilon_0)$ is the single atom Wigner-Weisskopf linewidth.

The scattered light is given as a sum of the scattered light from all the atoms

$$\epsilon_0\hat{\mathbf{E}}_{sc}^+(\mathbf{r}, t) = \sum_j \mathsf{G}(\mathbf{r} - \mathbf{r}_j)\mathbf{d}\hat{\sigma}_j^-(t) \quad (S3)$$

where the dipole radiation kernel [S3],

$$\mathsf{G}(\mathbf{r})\mathbf{d} = -\frac{\mathbf{d}\delta(\mathbf{r})}{3} + \frac{k^3}{4\pi}\left\{(\hat{\mathbf{r}}\times\mathbf{d})\times\hat{\mathbf{r}}\frac{e^{ikr}}{kr}\right.$$
$$\left. - [3\hat{\mathbf{r}}(\hat{\mathbf{r}}\cdot\mathbf{d}) - \mathbf{d}]\left[\frac{i}{(kr)^2} - \frac{1}{(kr)^3}\right]e^{ikr}\right\}, \quad (S4)$$

represents the monochromatic positive frequency component of the scattered light at $\mathbf{r}$ from the dipole $\mathbf{d}$ located at the origin. The interaction terms in Eq. (S1) arise from each atom j

being driven by the light scattered from all other atoms $\ell \neq j$. These radiative dipole-dipole couplings have coherent $\Delta_{j\ell}$ and dissipative $\gamma_{j\ell}$ contributions given by the real and imaginary parts of

$$\Delta_{j\ell} + i\gamma_{j\ell} = \frac{1}{\hbar\epsilon_0}\mathbf{d}^*\cdot\mathsf{G}(\mathbf{r}_j - \mathbf{r}_\ell)\mathbf{d}. \quad (S5)$$

Note that Eq. (S5) gives $\gamma_{jj} = \gamma$. A proper calculation of Δ_{jj} would involve evaluation of the Lamb shift, and we assume this is incorporated to the single-atom detuning δ.

The total field at position $\mathbf{r}$ is given as a sum of the incident field $\mathcal{E}^+(\mathbf{r})$ and the scattered light $\hat{\mathbf{E}}_{sc}^+(\mathbf{r}, t)$. We assume that the incident field has been blocked before detection, for example by a thin wire as in the dark-ground imaging technique of [S4]. Hence only the scattered field is detected, with intensity

$$I_{sc}(\mathbf{r}, t) = 2\epsilon_0 c\langle\hat{\mathbf{E}}_{sc}^-(\mathbf{r}, t)\cdot\hat{\mathbf{E}}_{sc}^+(\mathbf{r}, t)\rangle. \quad (S6)$$

Integrating the scattered intensity over the detector surface S gives the total count rate, which is the expectation value of the operator

$$\hat{n}(t) = \frac{2\epsilon_0 c}{\hbar\omega_0}\int_S dS\,\hat{\mathbf{E}}_{sc}^-(\mathbf{r}, t)\cdot\hat{\mathbf{E}}_{sc}^+(\mathbf{r}, t) = \sum_{j,\ell} I_{mn}\hat{\sigma}_j^+(t)\hat{\sigma}_\ell^-(t). \quad (S7)$$

with interference integrals

$$I_{j\ell} \equiv \frac{2c}{\hbar\epsilon_0\omega_0}\int_S dS\,\left[\mathsf{G}(\mathbf{r} - \mathbf{r}_j)\mathbf{d}\right]^*\mathsf{G}(\mathbf{r} - \mathbf{r}_\ell)\mathbf{d} \quad (S8)$$

We assume the detector lies in the radiation zone $kr \gg 1$, hence we can expand the dipole radiation kernels to obtain [S5]

$$I_{j\ell} = \frac{3\gamma}{4\pi}\int_S d\theta d\phi\,\sin\theta\left(1 - |\hat{\mathbf{r}}\cdot\hat{\mathbf{d}}|^2\right)e^{ik\hat{\mathbf{r}}\cdot(\mathbf{r}_j - \mathbf{r}_\ell)}$$
$$= 2\gamma_{j\ell}. \quad (S9)$$

Hence we arrive at

$$\hat{n}(t) = 2\sum_{j\ell}\gamma_{j\ell}\hat{\sigma}_j^+(t)\hat{\sigma}_\ell^-(t) \quad (S10)$$

for the photon-number operator.

S.B.   Single-atom physics

For a single isolated atom, both the photon detection rate $\langle\hat{n}(t)\rangle$ and the second-order correlation function $g_2(\tau)$ can be

evaluated analytically to yield [S6–S9]

$$g_2(\tau) = 1 - e^{-3\gamma\tau/2}\left(\cosh\kappa\gamma\tau + \frac{3}{2}\frac{\sinh\kappa\gamma\tau}{\kappa}\right)$$

$$\langle\hat{n}(t)\rangle = \frac{I_{\mathrm{in}}}{I_{\mathrm{in}} + I_s}g_2(\tau) \tag{S11}$$

The parameter $\kappa = \frac{1}{2}\sqrt{1 - 8I_{\mathrm{in}}/I_s}$ depends on the ratio of the incident intensity to the single atom saturation intensity, and determines the spectral properties of the atom [S10]. For low incident light intensity $I_{\mathrm{in}} \ll I_s$, Eqs. (S11) are dominated by a term proportional to the single decay $e^{-\gamma t}$. Here the single atom linewidth exceeds the single atom Rabi frequency $\gamma\sqrt{2I_{\mathrm{in}}/I_s}$ and Rabi oscillations are suppressed. Conversely, when $I_{\mathrm{in}} \gtrsim I_s$, the parameter κ is imaginary and hence both the photon scattering rate and $g_2(\tau)$ display decaying Rabi oscillations.

S.C.   Limit of low light intensity

A consistent low light intensity (LLI) theory of Eq. (S1), can be obtained [S11] from the equations of motions by retaining terms containing at most one of either $\sigma_j^{\pm}$ or the incident field amplitude. The only remaining equations of motion for the expectation values of atomic operators from Eq. (S1) are those for $\langle\sigma_j^{\pm}\rangle$, which in the LLI are,

$$\frac{d\langle\sigma_j^-\rangle}{dt} = i\delta\langle\sigma_j^-\rangle + i\sum_\ell \mathcal{H}_{j\ell}\langle\sigma_\ell^-\rangle + i\frac{\mathbf{d}\cdot\mathcal{E}^+(\mathbf{r}_j)}{\hbar} \tag{S12}$$

with $\mathcal{H}_{j\ell} \equiv \Delta_{j\ell} + i\gamma_{j\ell}$ (with $\Delta_{jj} \equiv 0$; recall that $\gamma_{jj} = \gamma$). Hence the atom dynamics evolves linearly in terms of the drive. Here we expand the complex symmetric matrix $\mathcal{H}_{j\ell}$ in a complete basis of eigenstates u_m, $m = 1,...,N$, which are the LLI collective eigenmodes,

$$\sum_\ell \mathcal{H}_{j\ell}u_m(\mathbf{r}_\ell) = (\zeta_m + i\upsilon_m)u_m(\mathbf{r}_j), \tag{S13}$$

where the imaginary part, υ_m, of the eigenvalue gives the collective linewidth of the eigenmode u_m and the real part the line shift ζ_m from the single-atom resonance. Note that the eigenstates u_m are not necessarily orthogonal, however, they do satisfy the biorthogonality condition $\sum_j u_m(\mathbf{r}_j)u_n(\mathbf{r}_j) = \delta_{mn}$ (after appropriate normalization of the u_m) apart from possible rare cases when $\sum_j u_m(\mathbf{r}_j)u_m(\mathbf{r}_j) = 0$.

Given some steady-state values for the $\langle\sigma_j^-\rangle$, a measure of the occupation of the LLI collective mode u_m is given by [S12]

$$L_m \equiv \frac{\sum_j |u_m(\mathbf{r}_j)\langle\sigma_j^-\rangle|^2}{\sum_{j\ell} |u_\ell(\mathbf{r}_j)\langle\sigma_j^-\rangle|^2}. \tag{S14}$$

S.D.   Quantum trajectories

A direct way to solve the QME (S1) is via matrix exponentiation of the density-matrix evolution operator. This is convenient for small atom numbers. For larger systems, however, the size of the density matrix becomes prohibitively large ($\sim 2^{2N}$). A more profitable scaling is to employ the Monte Carlo wavefunction method of quantum trajectories [S13–S16]. The evolution of the density matrix is then represented as the ensemble average of many individual realizations of the evolution of a many-body wavefunction $\psi(t)$, whose size scales as $\sim 2^N$, under a non-Hermitian Hamiltonian operator

$$H_S - \frac{i\hbar}{2}\sum_j \hat{J}_j^\dagger \hat{J}_j, \tag{S15}$$

where $\hat{J}_j$ are jump operators derived from the dissipative terms of QME and H_S represents Hermitian Hamiltonian evolution. Incoherent evolution is incorporated via stochastic quantum jumps that happen with a probability proportional to the loss of norm of the wavefunction as it evolves under (S15). One can show that this formalism is exactly equivalent to QME for the operator expectation values [S16].

A many-body system supports multiple decay channels and unraveling of the QME into an explicit mixture of pure states subject to stochastic evolution can be done in several different ways, corresponding to different constructions of the jump operators, as long as the full incoherent evolution in Eq. (S1) is accounted for. For the driven array of two-level atoms of the QME (S1) we follow here the "source-mode" quantum trajectory formalism [S5, S17]. In the single-excitation limit, these jumps correspond to the emission of photons, while their physical interpretation is more convoluted at sufficiently high light intensities to cause multiple excitations when the jump operators become formal constructions that do not necessarily correspond to any specific measurement record. They, however, provide a straightforward mapping of Eq. (S1) to the evolution of quantum trajectories of state vectors.

To formulate the source-mode jump operators, the matrix $\gamma_{j\ell}$ is diagonalized to find its eigenvalues λ_j and the corresponding eigenvectors $\mathbf{b}_j = (b_{1j},...,b_{Nj})^T$. The jump operators are then defined as

$$\hat{J}_j = \sqrt{\lambda_j}\mathbf{b}_j^T\hat{\Sigma}, \quad \hat{J}_j^\dagger = \sqrt{\lambda_j}\hat{\Sigma}^\dagger\mathbf{b}_j, \tag{S16}$$

where

$$\hat{\Sigma} = \begin{pmatrix}\hat{\sigma}_1^- \\ \vdots \\ \hat{\sigma}_N^-\end{pmatrix}, \quad \hat{\Sigma}^\dagger = \begin{pmatrix}\hat{\sigma}_1^+, & ..., & \hat{\sigma}_N^+\end{pmatrix}. \tag{S17}$$

Then defining

$$H_S = \sum_j H_j + \hbar\sum_{j\ell(\ell\neq j)} \Delta_{j\ell}\hat{\sigma}_j^+\hat{\sigma}_\ell^-, \tag{S18}$$

the problem has been cast in the form of quantum trajectories and the corresponding non-Hermitian Hamiltonian for the wavefunction evolution follows from Eq. (S15). The quantum trajectory evolution can then be evaluated as described in, e.g., Ref. [S18]. Thanks to the source-mode unraveling, the dissipative component of Eq. (S1) is now diagonal in the jump operators $\hat{J}_j$, which is computationally expedient. Further, the
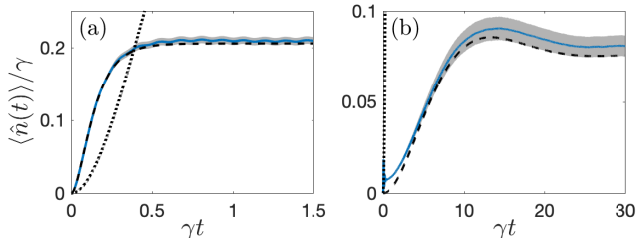
stochastic wavefunction evolution requires exponentiating a matrix of size 2^{2N}, as opposed to the 2^{4N} matrix governing the density matrix evolution, providing a significant numerical advantage as the system size increases beyond a few atoms.

FIG. S1. Transient dynamics of the photon detection rate for a 3×4 atom array with a drive field resonant with (a) the uniform superradiant ($\upsilon \approx 9.3\gamma$, $\mathcal{I} \approx 0.98N$) and (b) a subradiant ($\upsilon \approx 0.11\gamma$, $\mathcal{I} \approx 0.015N$) LLI collective eigenmode, with $NI_{\mathrm{in}} = 2I_s$, $a = 0.1\lambda$. The full quantum solution (blue solid line) agrees very well with the superatom (black dashed line); black dotted line shows the single isolated atom solution. The gray shading gives the standard error from $\sim 10^4$ quantum trajectories. Interestingly, examining just the incoherent contribution to the scattering rates in (b) gives even better agreement between the SAM and full quantum solution.
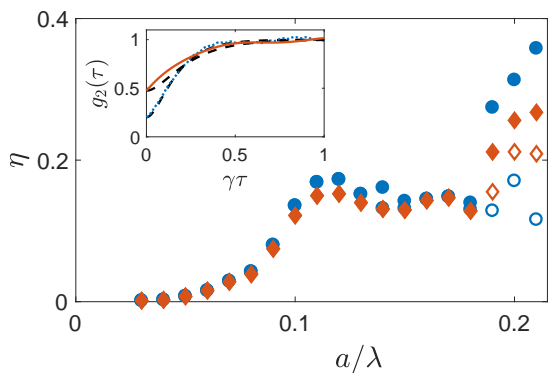

FIG. S2. Relative error η of the superatom picture as a function of lattice spacing for a field resonant with the uniform superradiant LLI mode for a 3×3 atom at $NI_{\mathrm{in}} = 0.08I_s$ (blue circles) and $NI_{\mathrm{in}} = 2I_s$ (red diamonds). Unfilled markers show results for $\eta \equiv \max |g_2(\tau)/[1 - g_2(0)] - g_2^{(\upsilon,\kappa')}(\tau)|_{\tau < \tau_0}$, where the deviation is calculated until τ_0, such that for all $\tau \lesssim \tau_0$, $g_2(\tau) < 0$, while filled markers for $\eta \equiv \max |g_2(\tau)/[1-g_2(0)]-g_2^{(\upsilon,\kappa')}(\tau)|_{\mathrm{all}\ \tau}$. The two deviate at $a \sim 0.2\lambda$ due to a persistent oscillation arising from a second mode at larger τ. Inset: Example $g_2(\tau)$ for $a = 0.08\lambda$ (blue dotted curve) and $a = 0.17\lambda$ (red solid curve) compared to the SAP results (black dashed curves), with the larger lattice spacing showing an oscillation.

[S1] R. H. Lehmberg, "Radiation from an N-Atom System. I. General Formalism," Phys. Rev. A **2**, 883–888 (1970).

[S2] G. S. Agarwal, "Master-equation approach to spontaneous emission," Phys. Rev. A **2**, 2038–2046 (1970).

[S3] John David Jackson, *Classical Electrodynamics*, 3rd ed. (Wiley, New York, 1999).

[S4] MR Andrews, M-O Mewes, NJ Van Druten, DS Durfee, DM Kurn, and W Ketterle, "Direct, nondestructive observation of a Bose condensate," Science **273**, 84–87 (1996).

[S5] HJ Carmichael and Kisik Kim, "A quantum trajectory unraveling of the superradiance master equation," Opt. Commun. **179**, 417–427 (2000).

[S6] B. R. Mollow and M. M. Miller, "The damped driven two-level atom," Ann. Phys. **52**, 464–478 (1969).

[S7] H J Carmichael and D F Walls, "Proposal for the measurement of the resonant Stark effect by photon correlation techniques," Journal of Physics B: Atomic and Molecular Physics **9**, L43–L46 (1976).

[S8] HJ Carmichael and DF Walls, "A quantum-mechanical master equation treatment of the dynamical Stark effect," J. Phys. B: At. Mol. Phys. **9**, 1199 (1976).

[S9] H. J. Kimble and L. Mandel, "Theory of resonance fluorescence," Phys. Rev. A **13**, 2123–2144 (1976).

[S10] B. R. Mollow, "Power spectrum of light scattered by two-level

systems," Phys. Rev. **188**, 1969–1975 (1969).

[S11] Janne Ruostekoski and Juha Javanainen, "Quantum field theory of cooperative atom response: Low light intensity," Phys. Rev. A **55**, 513–526 (1997).

[S12] G. Facchinetti, S. D. Jenkins, and J. Ruostekoski, "Storing light with subradiant correlations in arrays of atoms," Phys. Rev. Lett. **117**, 243601 (2016).

[S13] Jean Dalibard, Yvan Castin, and Klaus Mølmer, "Wave-function approach to dissipative processes in quantum optics," Phys. Rev. Lett. **68**, 580–583 (1992).

[S14] L. Tian and H. J. Carmichael, "Quantum trajectory simulations of two-state behavior in an optical cavity containing one atom," Phys. Rev. A **46**, R6801–R6804 (1992).

[S15] R. Dum, P. Zoller, and H. Ritsch, "Monte carlo simulation of the atomic master equation for spontaneous emission," Phys. Rev. A **45**, 4879–4887 (1992).

[S16] Klaus Mølmer, Yvan Castin, and Jean Dalibard, "Monte Carlo wave-function method in quantum optics," J. Opt. Soc. Am. B **10**, 524–538 (1993).

[S17] J. P. Clemens, L. Horvath, B. C. Sanders, and H. J. Carmichael, "Collective spontaneous emission from a line of atoms," Phys. Rev. A **68**, 023809 (2003).

[S18] Janne Ruostekoski, M. J. Collett, Robert Graham, and Dan F. Walls, "Macroscopic superpositions of bose-einstein condensates," Phys. Rev. A **57**, 511–517 (1998).