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Room-temperature cavity QED with strongly-coupled Dicke states

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Collective light-matter interactions are fundamental to cavity quantum electrodynamics (cQED) [1]. A key feature is the *strong coupling regime*, where excitations are coherently transferred between different quantum systems over timescales significantly shorter than their lifetimes. A striking example is the Dicke state [2], where excitations are symmetrically distributed throughout an ensemble of spins or two-level atoms. Dicke states coupled to electromagnetic radiation have been realised experimentally for atomic gases in high-finesse optical cavities [3], emitters near plasmonic nanoparticles [4], electron spins coupled to superconducting qubits [5, 6] and sub-Hertz linewidth super-radiant lasers [7, 8]. Typically in these applications, cryogenic cooling is required to polarize the spin population and mitigate decoherence. Here we report strong coupling and collective Rabi oscillations at room-temperature between an optically excited spin-triplet ensemble within pentacene-doped *p*-terphenyl and a cavity mode supported by a strontium titanate dielectric resonator at 1.45 GHz. The spin ensemble becomes highly correlated through stimulated emission, resulting in cascading entangled Dicke states with suppressed decoherence due to a cavity

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29 **protection effect [9–11].**

30 The strong coupling regime can be achieved for single quantum emitters interacting with
 31 photons within electromagnetic fields through their electric dipole moment [12], however the
 32 magnetic coupling strength of single spins to photons is typically far too weak at microwave
 33 frequencies. By placing a spin in a resonant cavity, the coupling can be enhanced through
 34 the Purcell effect [13] where the local density of photonic states is modified by the geometry.
 35 The coupling strength of a single spin to a photon is then given by $g_s = \gamma\sqrt{\mu_0\hbar\omega_c/2V_m}$, where
 36 γ is the electron gyromagnetic ratio, μ_0 is the permeability of free-space, \hbar is the reduced
 37 Planck constant, ω_c is the angular frequency of the cavity field and V_m is the magnetic mode
 38 volume. Furthermore, the spin-photon coupling strength g_e for an ensemble of N spins is
 39 enhanced by a factor \sqrt{N} [6, 14]. Hence, for a sufficient number of spins N and small mode
 40 volume V_m , it should be possible for the ensemble spin-photon coupling strength to exceed
 41 both the decay rate of the cavity mode κ_c and the spin dephasing rate of the spins κ_s , taking
 42 the system into the strong coupling regime.

45 In order to satisfy this condition, we utilise a system comprising spin-triplets in pentacene
 46 molecules to generate a polarized (inverted) population of N spins and a cavity with small
 47 magnetic mode volume, $V_m \sim 10^{-5}\lambda^3$, where λ is the free-space wavelength. Following photo-
 48 excitation of pentacene in zero magnetic field, the non-degenerate X, Y, Z sub-levels of the
 49 T_1 spin-triplet are rapidly populated in the ratios 0.76 : 0.16 : 0.08, respectively [15] (see
 50 Fig. 1a). The initial inversion is $S^z \approx 0.8N$, where N is the number of pentacene molecules
 51 excited into either the $|e\rangle \equiv |X\rangle$ or $|g\rangle \equiv |Z\rangle$ states. The state $|Y\rangle$ does not play a significant
 52 role and shall not be considered further. At zero magnetic field the $|e\rangle$ and $|g\rangle$ states have
 53 a frequency splitting of $\omega_s/2\pi \approx 1.45$ GHz. The cavity is similar to the one reported for
 54 a miniaturised room-temperature maser which uses a hollow cylinder of strontium titanate
 55 (STO), but houses a pentacene-doped p -terphenyl crystal with a much higher concentration
 56 of 0.053% mol/mol than in previous studies [16, 17]. The $TE_{01\delta}$ mode has a frequency tuned
 57 to the $\langle e\rangle \leftrightarrow \langle g\rangle$ spin transition, $\omega_c \approx \omega_s \approx 2\pi \times 1.45$ GHz. This mode has a magnetic field
 58 dipole directed along the cylindrical axis (Fig. 1b) which, via the S^y spin-operator, induces
 59 transitions between the $|e\rangle \leftrightarrow |g\rangle$ states in suitably aligned pentacene molecules. STO has
 60 a high electric permittivity ($\epsilon_r \approx 320$) that allows a sub-wavelength ($\lambda_0/18 \sim 1$ cm) cavity to
 61 be constructed with a mode volume V_m of 0.25 cm³. An optical parametric oscillator (OPO)
 62 generated $\lambda \sim 592$ nm pulses of 5.5 ns duration and energy up to 15 mJ at a repetition rate

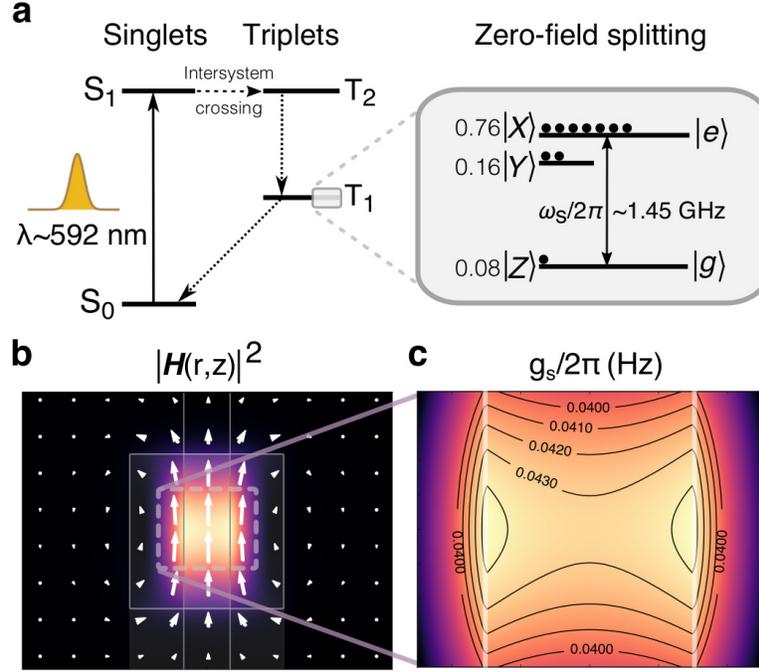


FIG. 1. **Pentacene:*p*-terphenyl/STO cQED system.** **a**, Simplified Jablonski diagram showing optical pumping scheme that prepares an inverted population. The $|X\rangle \equiv |e\rangle$ and $|Z\rangle \equiv |g\rangle$ triplet sub-levels constitute a two-level system of pseudo spin-1/2 particles with initial relative population 9:1. The difference in energy between the $|e\rangle$ and $|g\rangle$ states is $\hbar\omega_s$, where $\omega_s \approx 2\pi \times 1.45$ GHz. **b**, Distribution of magnetic energy density within a cross-section of the TE_{01δ} mode supported by the cavity. White arrows indicate the magnetic field vector. **c**, Region illuminated by optical pulse. The magnetic field directly maps to the single spin-photon coupling g_s , for a cylindrical region of diameter 3 mm and height 4 mm, where the pentacene:*p*-terphenyl crystal resides.

63 of 10 Hz. The 4 mm diameter (Gaussian profile) beam of the OPO was focused onto the 3
 64 mm diameter pentacene-doped *p*-terphenyl crystal by the high refractive index ($n \sim 2.6$) of
 65 the STO. Numerical modelling of the penetration of optical pulses into the pentacene-doped
 66 *p*-terphenyl allowed the number N of excited pentacene molecules to be estimated [18] and
 67 revealed that the spin-triplet yield is a linear function of the incident optical pulse energy
 68 (see Supplementary Information). For an optical pulse energy of 15 mJ, the number of
 69 pentacene molecules N excited into the $|e\rangle$ and $|g\rangle$ states was estimated to be $\sim 7 \times 10^{14}$,
 70 with an initial inversion $S^z \sim 6 \times 10^{14}$.

71 The microwave magnetic energy-density (Fig. 1b) can be mapped directly to the single
 72 spin-photon coupling in the central region of the cavity housing the pentacene molecules (Fig.

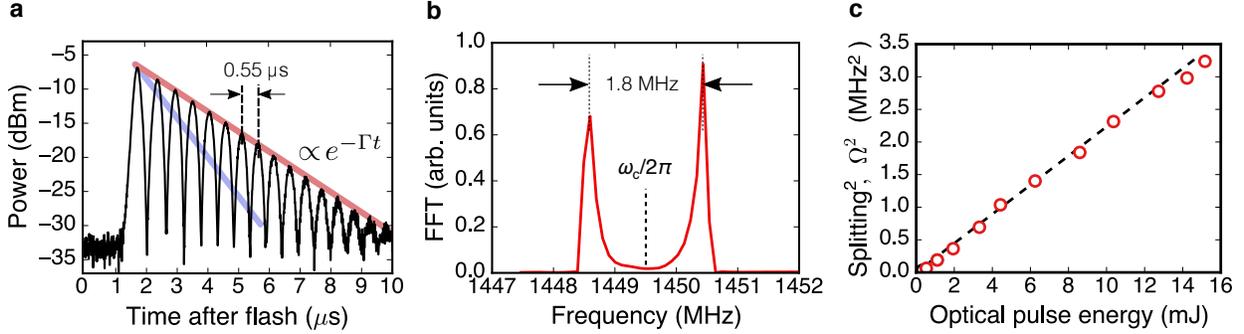


FIG. 2. Measured microwave emission. **a**, Rabi oscillations from photo-excited pentacene:*p*-terphenyl spin-ensemble coupled to the STO cavity. The initial burst builds up from thermal photons and follows classical laws since the spin-spin correlation and spin-photon coherence has yet to build up. The peak microwave output power is -6.8 dBm. Subsequent oscillations in the microwave power, with period $0.55 \mu\text{s}$ and decay rate Γ , are due to the collective exchange of energy between the correlated ensemble Dicke state and the cavity mode. The blue line shows the expected decay of an empty cavity ($\propto e^{-\kappa_c t}$). **b**, Fourier analysis of the signal for a cavity frequency of $\omega_c/2\pi = 1.4495 \text{ GHz}$ and 15 mJ optical pulse energy reveals that ensemble spin-coupling has split the normal modes, yielding a Rabi frequency of $\Omega/2\pi \sim 1.8 \text{ MHz}$. **c**, Increasing the laser pulse energy excites more pentacene molecules into spin-triplets and therefore increases the ensemble spin-photon coupling (and hence normal-mode Rabi splitting) with a \sqrt{N} dependence. This graph confirms that the square of the splitting is linearly dependent on the optical pump pulse energy and hence N .

73 1c). Within the region illuminated by the optical pulse, the single spin-photon coupling is
 74 $g_s/2\pi = 0.042 \pm 0.002 \text{ Hz}$. An estimate of the ensemble spin-photon coupling strength is
 75 therefore $g_e \approx g_s \sqrt{N} \approx 7 \mu\text{s}^{-1} \approx 2\pi \times 1.1 \text{ MHz}$. The cavity mode decay rate (linewidth)
 76 was measured to be $\kappa_c = 1.1 \mu\text{s}^{-1}$ and the spin decoherence rate of the transition was taken
 77 from reported room-temperature free induction decay measurements of the spin dephasing
 78 time, $T_2^* = 2.9 \mu\text{s}$ [19], yielding a rate of $\kappa_s = 2/T_2^* \approx 0.7 \mu\text{s}^{-1}$. The spin-lattice relaxation
 79 rate and decay rates of the triplet sub-levels back to the singlet ground state are at least
 80 an order of magnitude slower than the spin dephasing rate so can be neglected [20]. Thus,
 81 the system is expected to be within the strong coupling regime since the predicted ensemble
 82 spin-photon coupling is an order of magnitude greater than both the cavity decay and spin
 83 dephasing rates, $g_e \gg \kappa_c, \kappa_s$, with cooperativity $C = 4g_e^2/\kappa_c\kappa_s \approx 250$.

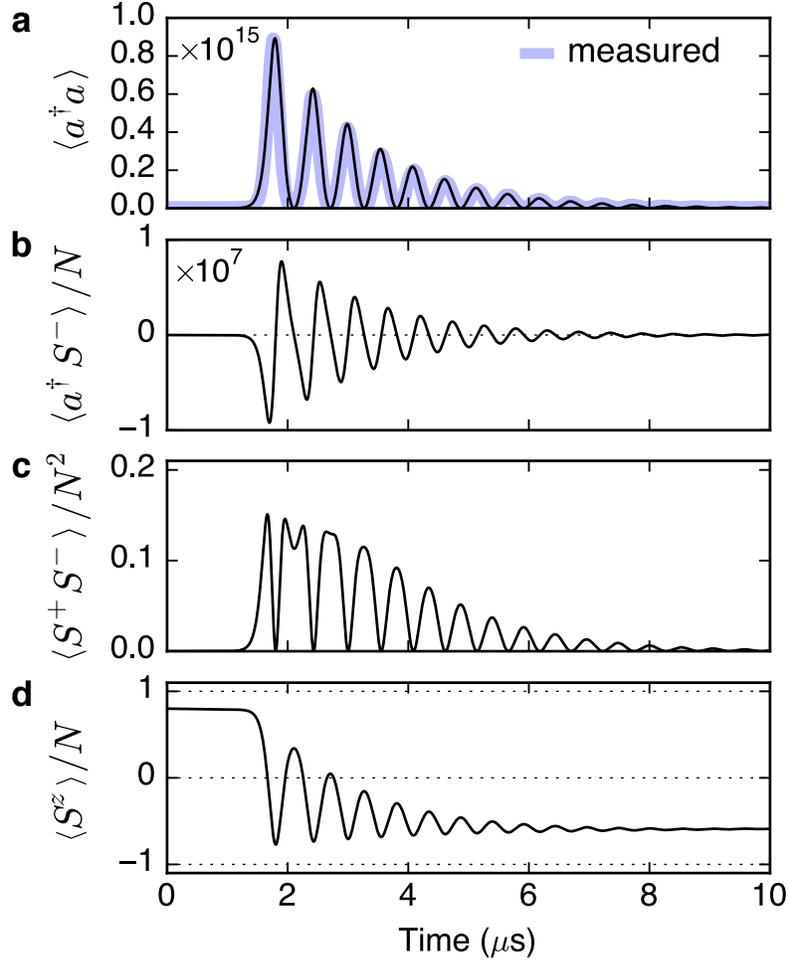


FIG. 3. **Modelled dynamics using master equations.** **a**, The expectation value of the cavity photon number $n = \langle a^\dagger a \rangle$, agrees well with the inferred population from measurement of the microwave power. **b**, Negative values of the spin-photon coherence $\langle a^\dagger S^- \rangle$ correspond to exchange of excitations from the collective spins to the cavity mode and positive values from the cavity mode to the collective spins. **c**, The spin-spin correlation $\langle S^+ S^- \rangle$, a measure of the degree of multipartite entanglement of the spins, is maximal when the inversion is zero. **d**, The inversion S^z , initially at $0.8N$, stays fairly constant until the emergence of the first microwave burst. It subsequently oscillates through the Rabi cycles, coming to rest in a negative yet still spin-polarized state, where the majority of electrons reside in the lower $|g\rangle$ state.

84 Stimulated emission, due to the population inversion established by the optical pulse,
 85 amplifies the thermal cavity photon population ($\bar{n} \sim 4 \times 10^3$), resulting in a buildup of
 86 electromagnetic energy in the cavity, *i.e.* masing [21]. The instantaneous microwave power
 87 coupled out of the cavity tuned to a frequency of $\omega_c = 2\pi \times 1.4495$ GHz and an optical pulse

88 of energy 15 mJ is shown in Fig. 2a. There is a short delay of the order of a microsecond
 89 until the microwave signal emerges, after which it oscillates with a period of around half a
 90 microsecond, whilst decaying at rate Γ over the course of 10 μs . Fourier analysis revealed
 91 a normal mode-splitting (Rabi frequency) of $\Omega = 2g_e \approx 11.3 \mu\text{s}^{-1} \approx 2\pi \times 1.8 \text{ MHz}$ (see
 92 Fig. 2b), indicating that the cavity mode and collective spin state have hybridised to form
 93 a collective spin-photon polariton and that the strong coupling condition is satisfied, with
 94 cooperativity $C \sim 190$. As the optical pump pulse energy was varied from 0 to 15 mJ to
 95 excite increasing numbers of pentacene molecules, the normal-mode splitting increased as
 96 \sqrt{N} as shown in Fig. 2c, confirming the ensemble spin-photon coupling scaling predicted
 97 by the Tavis-Cummings model for a spin-ensemble [22].

98 For a single excitation at resonance ($\omega_c = \omega_s$), the eigenstates are a coherent superpo-
 99 sition of two basis states: the spin-mode and the cavity mode. If $|0\rangle_c$ and $|1\rangle_c = a^\dagger |0\rangle_c$
 100 are the possible states of the cavity mode and $|0\rangle_s$ and $|1\rangle_s = a^\dagger |0\rangle_s$ are the possible
 101 states of the spin-mode, then the two eigenstates are $|+\rangle = \frac{1}{\sqrt{2}} (|1\rangle_s |0\rangle_c + |0\rangle_s |1\rangle_c)$ and
 102 $|-\rangle = \frac{1}{\sqrt{2}} (|1\rangle_s |0\rangle_c - |0\rangle_s |1\rangle_c)$, separated in energy by $\hbar\Omega = 2\hbar g_e$. In this system, there
 103 are many more excitations so to describe the dynamical behaviour of the Dicke system we
 104 derived Lindblad master equations for the reduced spin-photon density matrix within the
 105 Born-Markov approximation [7, 23]. Here we used the Tavis-Cummings Hamiltonian and
 106 a Liouvillian that accounted for decoherence due to cavity decay, spin dephasing and spin-
 107 lattice relaxation. A system of coupled differential equations was derived for the expectation
 108 values of relevant variables using a cumulant expansion truncated to third-order [24] (see
 109 Supplementary Information). The dynamical behaviour of the expectation values of the
 111 cavity photon population, spin-photon coherence, spin-spin correlation and inversion are
 112 shown in Fig. 3, where the photon population $\bar{n} = \langle a^\dagger a \rangle$ is in excellent agreement with that
 113 inferred from the measured microwave power. The delay between photo-excitation and the
 114 emergence of the first microwave burst can be explained in terms of the spin-spin correlation
 115 $\langle S^+ S^- \rangle$. Initially, the spins are not correlated but through stimulated emission of photons
 116 into the cavity become increasingly correlated, leading to the buildup of a macroscopic
 117 collective dipole. The maximum expectation value of the modelled spin-spin correlation,
 118 $\langle S^+ S^- \rangle / N^2 \approx 0.15$ is not much less than the theoretical maximum of 0.25 (see Supplemen-
 119 tary Information). The discrepancy being mostly due to the imperfect initialisation of the
 120 spin population polarization. The collective spin-spin correlation $\langle S^+ S^- \rangle$ reveals the degree

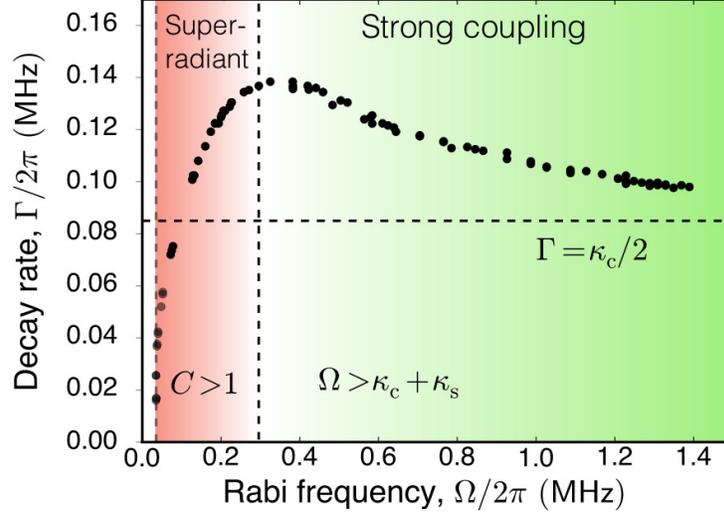


FIG. 4. **Cavity protection due to inhomogeneous broadening.** The vertical dashed lines marks the onset of super-radiant masing (where the cooperativity $C > 1$) and the strong coupling regime where $\Omega = 2g_e > \kappa_c + \kappa_s$, the Rabi frequency exceeds the sum of cavity decay and spin decoherence rates. As the Rabi frequency increases, Γ , the decay rate of the collective spin-photon polariton decreases, asymptotically approaching half the cavity decay rate $\kappa_c/2$ (horizontal dashed line).

121 of transient multipartite entanglement within the symmetric Dicke states, the entanglement
 122 resulting from the ambiguity in being able to assign emission or absorption of photons to
 123 specific molecules [1]. For the $N = 2$ case, the maximally entangled $(|e, g\rangle + |g, e\rangle)/\sqrt{2}$ state
 124 is a Bell state. Once the collective dipole emerges there is coherent oscillatory exchange of
 125 excitations with the cavity mode at the Rabi frequency Ω , persisting for up to $10 \mu\text{s}$.

126 The collective spin-photon polariton decay rate Γ as a function of Rabi frequency (for
 127 optical pump pulses in the 0 to 10 mJ range) is shown in Fig. 4. Above maser threshold
 128 where the cooperativity $C > 1$ and below the strong coupling regime threshold, the system
 129 is super-radiant and the rate of emission is proportional to N^2 . The decay rate Γ increases
 130 until the Rabi frequency is equal to the sum of the cavity decay and spin decoherence rates,
 131 $\Omega = 2g_e \sim \kappa_c + \kappa_s$ and the decay rate $\Gamma \sim (\kappa_c + \kappa_s)/2$. As the Rabi frequency (and ensemble
 132 spin-photon coupling) increases and the system goes further into the strong-coupling regime,
 133 the ensemble spin-photon polariton decay rate Γ begins decreasing asymptotically towards
 134 half the cavity decay rate, $\kappa_c/2$, implying that the spin dephasing rate is suppressed. This so-
 135 called *cavity protection* effect, associated with non-Markovian memory effects, is attributed

136 to inhomogeneous broadening of spin transitions with non-Lorentzian lineshapes [9–11].

137 To conclude, this system, which was recently used to demonstrate a solid-state room-
138 temperature maser [16, 17, 21], also shows promise as a platform for exploring cavity quan-
139 tum electrodynamics, spin memories for quantum information processing [14] and quantum-
140 enhanced technologies such as metrology, sensing and communications.

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147 AUTHOR CONTRIBUTIONS

148 Experiments were performed by J.D.B., E.S., J.S. and C.W.M.K. Data was processed
149 by J.D.B. and C.W.M.K. Theory was developed by J.D.B. who also performed simulations
150 of quantum master equations, optical pulse penetration and cavity design. The paper was
151 written by J.D.B. with assistance by C.W.M.K. and additional editing by E.S. and N.McN.A.
152 The study was conceived by J.D.B. E.S., N.McN.A. and C.W.M.K.

153 AUTHOR INFORMATION

154 The authors declare no competing financial interests. Readers are welcome to comment on
155 the online version of this article at www.nature.com/nature. Correspondence and requests
156 for materials should be addressed to J.D.B. (jonathan.breeze@imperial.ac.uk).

157 METHODS

158 **Experimental setup.** The cavity was constructed from a hollow cylindrical single-
159 crystal of strontium titanate (STO) containing a 0.053% pentacene-doped *p*-terphenyl crystal
160 (diameter 3 mm, height 8 mm). The STO cylinder was placed upon a cylindrical sapphire

161 disc and housed within a cylindrical copper enclosure. The cavity was directly coupled
 162 to a digital storage oscilloscope (Keysight MSOX6004A, 20 GSa/s sampling-rate, 6 GHz
 163 bandwidth) using a small loop antenna with coupling coefficient, $k = 0.2$. An additional
 164 weakly coupled (-35 dB) antenna, directional coupler (-20 dB) and amplifier (40 dB) allowed
 165 transmission measurements of the cavity to be made using a vector network analyzer (Agilent
 166 8520E), revealing the resonant frequency and loaded quality-factor (Q) of the $TE_{01\delta}$ mode
 167 to be $\omega_c \sim 2\pi \times 1.45$ GHz and 8,500 respectively. A Nd:YAG pumped optical parametric
 168 oscillator (OPO, Continuum Surelite Plus SL I-20) generated 592 nm wavelength optical
 169 pulses of 5.5 ns duration and energy up to 15 mJ at a repetition rate of 10 Hz. The optical
 170 pump pulse energies were measured using a beam splitter and an optical energy meter
 171 (Gentec-EO Maestro).

172 **Crystal growth.** The pentacene:*p*-terphenyl crystal was grown as per the previously
 173 reported method [16, 17] but with higher concentration of pentacene (see Supplementary
 174 Information for details).

175 **Cavity photon number from microwave power.** The expectation value of the pho-
 176 ton number as a function of time was extracted from the measured microwave power $P(t)$
 177 using the expression $\bar{n}(t) = \langle a^\dagger a \rangle = P(t)(1 + k)/\hbar\omega_c\kappa_c k$, where k is the coupling coefficient
 178 ($k = 0.2$), \hbar is the reduced Planck constant and ω_c and κ_c the cavity frequency and decay
 179 rate respectively.

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