Spectral engineering of cavity-protected polaritons in an atomic ensemble

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Multiple quantum emitters coupled to a single cavity mode appear in many situations, including quantum technologies and polaritonic chemistry. The ideal case of identical emitters is modelled in terms of symmetric states, and understood in terms of polaritons. In the practically relevant case of an inhomogeneous frequency distribution, this simple picture breaks down and new features emerge. Here we observe the transition from a disordered regime to a polaritonic one with only two resonances, using the high degree of control in a strongly coupled cold-atom system where the ratio between coupling strength and frequency inhomogeneities can be tuned. The polaritons are much narrower than the frequency distribution, as predicted in the context of cavity protection. We find that the concentration of photonic weight of the coupled light–matter states is a key parameter for this transition and demonstrate that a simple parameter based on the statistics of transmission count spectra provides an experimental proxy for this theoretical quantity. Moreover, we realize a dynamically modulated Tavis–Cummings model to produce a comb of narrow polariton resonances protected from disorder, with potential applications to quantum networks.

Although the foundations of cavity quantum electrodynamics (CQED) have been laid with single-emitter systems1,2, the extension to many emitters coupled to the same cavity mode holds interest for many different communities3–6. In solid-state systems, with emitters such as rare-earth ions, colour centres in diamond or semiconductor quantum wells, the N-emitter situation naturally arises and is exploited to enhance the light–matter coupling beneficial for quantum network implementations7–9. In atomic systems, the coupling between emitters via the cavity mode is a powerful approach for generating different forms of many-particle entanglement10–12 including spin-squeezed states for quantum metrology13–17. When combined with a high degree of control over the emitter qubits, this is the basis for an emerging form of quantum simulations of long-range coupled spin systems18–21. All these experiments operate in the regime of strong collective coupling, or even strong single-emitter coupling.

For many applications operating in the low-excitation limit, no more than one photon is present in the cavity. Central to such systems is the notion of a polariton22, the hybrid light–matter state composed of an intracavity photon and the ensemble of emitters sharing a single excitation. In homogeneous systems where all the emitters have the same resonance frequency \(\omega_0\), the energy spectrum of the coupled system is the well-known avoided crossing with only two resonances. Although many more, less symmetric atomic states exist, they do not participate in the system’s transmission or reflection spectra because they are ‘dark’: their collective coupling to the photonic mode vanishes. However, as soon as there is frequency inhomogeneity in the emitter ensemble, this destructive interference is no longer complete, and the formerly dark states now couple to the light field, becoming ‘grey’. Although frequency inhomogeneity is easily included in the foundational Tavis–Cummings Hamiltonian23, it leads to rich and often surprising physics, as shown by many recent advances driven by the interplay of theory and experiment. One spectacular example in the field of materials science started with the discovery that coupling an organic semiconductor to a cavity can profoundly change its material properties.

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trapping light with the 5 the main cavity mode (blue). The latter is in resonance with the | 4 the calculated photonic weight (PW) of the grey states. In addition, as 'unprotected' one where the amplitude of the polaritons decreases and 'unprotected' one where the amplitude of the polaritons decreases and many additional, randomly distributed resonances appear. Our analysis shows that the number of photon counts outside the two narrow polariton peaks acts as a robust experimental proxy that closely traces the calculated photonic weight (PW) of the grey states. In addition, as our system operates in the strong-coupling regime at the single-atom level, we are able to study this transition for a few tens of atoms, several orders of magnitude lower than previous experiments, highlighting the contribution of a finite number of dark states. Finally, we produce polaritons featuring a comb-like frequency spectrum by modulating the light-shifted frequency of the atoms on a very fast timescale in the protected regime. This allows us to shape the frequency spectrum of the system, with possible applications to quantum memories and quantum communications.

In our experiment, a cold ensemble of rubidium atoms is trapped inside a high-finesse Fabry–Perot microcavity with a length of 140 μm. The number of atoms loaded into the cavity can be varied between a few tens and a few thousands (Methods). Unless otherwise noted, atoms are initially prepared in |g⟩ = |S| 1/2, F = 2, mF = 2⟩ and |g⟩ = |P| 3/2, F = 2, mF = 2⟩. The probe beam exciting the cavity mode on the D2 line of 87Rb at 780 nm is circularly polarized, coupling |g⟩ to the excited state |e⟩ = |S| 1/2, F = 3, mF = 3⟩. The choice of circular polarization is possible owing to the negligible birefringence of our cavity. To realize a homogeneous and maximal coupling of all the atoms, they are trapped at the antinodes of the cavity mode. This is achieved by an intracavity standing wave at ωr = 2πcn/1,559 nm, where c0 is the speed of light, which is commensurate with the atomic line taking into account the Gouy phase shift and phase shift at reflection on the mirrors. The dipole light is linearly polarized and the trap depth of the intracavity lattice can be adjusted between 300 and 1,400 μK with atomic temperatures ranging from 50 to 190 μK. The maximum single-atom coupling is g/2π ≈ 76 megahertz (MHz); thermal motion leads to a temperature-dependent average single-atom coupling g/2π between 35 and 68 MHz (Supplementary Table I). This is larger than the cavity decay rate (half-width at half-maximum (HWHM)) of κ/2π ≈ 15 MHz and spontaneous emission rate of γ/2π ≈ 3 MHz, placing the system in the strong-coupling regime of CQED.
Due to a nearby resonance (the $^{19}F_2 \rightarrow 4D$ transition at 1,529 nm), the trapping light induces a light shift of the excited state of the D2 line that is ~50 times larger than the ground-state light shift\(^1\). Thus, it creates an adjustable shift and broadening of the frequency distribution of the $|g\rangle \leftrightarrow |e\rangle$ transition, the latter also depending on the finite temperature of the trapped atoms (Methods). We experimentally characterize the frequency distribution by illuminating the trapped atoms with a transverse beam and measuring the relative atomic losses as a function of beam frequency for various trap depths (Fig. 1c). It shows the broadening (up to 1 GHz) and shift due to the the 1,559 nm light power of $S_{1/2} F = 2 \rightarrow S_{1/2} F = 1, 2, 3$ transitions as well as level mixing at a high trap depth. A Monte Carlo simulation assuming an initial atomic population uniformly distributed between all the Zeeman sublevels, but no subsequent redistribution or depumping (Methods), is in qualitative agreement with the experimental loss curves.

To investigate the coherence properties of this inhomogeneous system, we consider the transmission spectrum of the cavity in the low-excitation limit, which is relevant for our experimental configuration. We start by describing the homogeneous case. For a cavity mode resonant with the atomic line in the strong-coupling regime, the spectrum exhibits the well-known vacuum Rabi splitting featuring two resonant with the atomic line in the strong-coupling regime, the spectrum. We start by describing the homogeneous case. For a cavity mode resonant with the atomic line in the strong-coupling regime, the spectrum exhibits the well-known vacuum Rabi splitting featuring two resonances, which broadens the averaged peaks by inducing a jittering of the collective coupling. The widths of the peaks should then be equal to the one of a homogeneous system given by $(\kappa + \gamma)/2 = 9$ MHz.

The fitted width of the peak $\Delta \omega/2\pi = 26 \pm 2$ MHz is much lower than the width $\Delta \omega/2\pi = 150 \pm 10$ MHz of the frequency distribution (Methods). The ratio $\Delta \omega/2\pi$ can be used to define a figure of merit for cavity protection. It allows a comparison between the measured width $\Delta \omega$ of the polaritonic peaks and width $\Delta \omega/2$ that would be obtained for a Lorentzian probability distribution inhibiting the protection effect\(^1\). This ratio is about 3 for our experiment, showing that despite strong inhomogeneities, the coherence of the polaritons can be preserved as long as $D = \Delta \omega$ for our frequency distribution.

We note that the measured widths are larger than the homogeneous limit, mainly due to shot-to-shot fluctuations of the atom number, which broadens the averaged peaks by inducing a jittering of the collective coupling. The widths are indeed the same for the different trap depths used in the experiment within the error margins. As shown in Fig. 2, the simulated spectra including atom number fluctuations are in excellent agreement with the experimental data.

Exploiting the high degree of control of our system, we explore the transition from the unprotected to protected regime. To exhibit this transition, we acquire the cavity transmission spectra as a function of number $N$ of trapped atoms in the cavity mode, for $10 \leq N \leq 550$. As we are dealing with a mesoscopic number of atoms, we can compute
the PW of the different eigenstates of the coupled system for the trap depth and atom temperature of the experiment. The cavity protection effect can be directly assessed by measuring the distribution of PWs over the different eigenstates. For a system with inhomogeneity of the emitter frequencies, the PW is distributed over more than two eigenstates. This leads us to introduce the sum $S_\text{PW}$ of PWs of all the eigenstates, except the two largest ones, to quantify the spreading of the PW and thus the coherence of the coupled system: $S_\text{PW}$ is zero for the homogeneous case and tends to 1 when the PW is distributed over an infinite number of dark states.

Experimentally, as we stay in the low-excitation limit, the small number of collected photons per spectrum prevents the direct measurement of PW of the eigenstates. Instead, from each spectrum, we extract the fraction $F_\text{out}$ of photon counts that are outside the two narrow frequency windows corresponding to the polariton peaks (Methods and Extended Data Fig. 2). The width $\Delta f$ of these windows is chosen such that they contain about 90% of the counts in the protected regime.

The results are shown in Fig. 3. Starting from zero atoms, $F_\text{out}$ increases with $N$, reaches a maximum for a measured coupling $\Omega/(2\pi) \approx 300$ MHz corresponding to $N \approx 25$ atoms and then decreases to an asymptotic value. As expected, we find that the shape of the $F_\text{out}$ data is robust against the choice of $\Delta f$ (Extended Data Fig. 3) and closely traces the calculated $S_\text{PW}$ (Methods). This allows us to interpret the common behaviour of $F_\text{out}$ and $S_\text{PW}$ in terms of two competing effects: the spread of the photonic excitation into the inhomogeneous atomic one, which is favoured by a large coupling, and the cavity protection effect where the synchronization between atoms benefits from a larger coupling. As $N$ increases, the dimension ($N + 1$) of the Hilbert space of the system rises and so does the number of states available to carry part of the photonic excitation outside $\Delta f$. The collective coupling $\Omega$ also increases (scaling as $\sqrt{N}$), and spreads the PWs to eigenstates close in frequency of $\Omega$ as long as the system stays in the unprotected regime ($\Omega < \Delta \omega$). Then, above a certain value of the collective coupling $\Omega_\text{in}$ roughly equal to the width of the atomic frequency distribution, the cavity protection effect starts to reduce $S_\text{PW}$ and $F_\text{out}$. In the large-collective-coupling limit, $S_\text{PW}$ and $F_\text{out}$ are low, because the PW—and thus the photon counts—concentrate in the two polaritonic states. To underscore the role of cavity protection, we simulate $S_\text{PW}$ for a Lorentzian distribution with the same average frequency and width as the experimental one. In this case (Fig. 3), after the initial increase, $S_\text{PW}$ stays on a plateau as it has been shown that no cavity protection occurs for this distribution featuring a slow decay with frequency [306, 313]. For our system, we note that the protection effect starts to play a role for a mesoscopic number of a few tens of atoms, which is several orders of magnitude lower than the typical number of emitters involved in solid-state-based systems [313]. This allows us to explore this effect for a finite-sized system, as well as directly observe the PW concentration in the two polaritons.

Because the coherence of polaritons is preserved by cavity protection, we can then harness the large sensitivity of the light-shifted atomic frequency to the trapping power to efficiently modulate the polaritonic frequencies. The intracavity dipole power is modulated by coupling two different frequencies of the dipole light into the cavity. The resulting beating at a frequency $\omega_\text{m}/2\pi$ leads to a modulation of the dipole potential and thus of the average light-shifted frequency of the atoms as $\omega_\text{at}(t) = \omega_\text{A}^0 + \beta_\text{s}\omega_\text{m} \cos(\omega_\text{m} t)$, where $\beta_\text{s}$ is the modulation index and $\omega_\text{A}^0$ is the average atomic frequency without modulation. The modulation frequency $\omega_\text{m}/2\pi = 125$ MHz is set to be larger than the width of the polaritons but smaller than the collective coupling $\Omega$. We record the transmission spectrum following the same procedure as before and average about 200 spectra. The results are presented in Fig. 4. Instead of the usual polariton doublet, the spectrum features two combs, each consisting of several peaks. The frequency splitting between the centres of the two combs is given by the collective coupling $\Omega/(2\pi)$, whereas the comb teeth are separated by modulation frequency $\omega_\text{m}/2\pi$. In contrast to the experiment in another work [306] where $\omega_\text{m} > \Omega$, the transmission spectrum does not result from the coupling between the cavity field and single-frequency atomic excitation given by one sideband of the modulated atomic transition. In our case, the photonic excitation couples to a multifrequency atomic excitation, yielding polaritons featuring multiple frequencies in their spectrum.

For $\omega_\text{m} < \Omega$, the theoretical cavity spectrum is well approximated by the following equation (Supplementary Section III):
where $J$ is the Bessel function of the first kind. $\omega_0 = \omega_c - \omega_A$ and $\gamma_e = \frac{\gamma_j}{2}$. Based on this expression, we define a modulation index for the polaritons given by $\beta = \beta_p / 2$. We have measured $\beta$ for different values of $\beta_p$ (Fig. 4, inset) and find very good agreement with the expected slope. The spectrum of the polaritons can, thus, be directly controlled by tuning the modulation index and/or modulation frequency. This is possible owing to the large sensitivity of the excited state to the trapping power. An important remark is that the width of each peak in multifrequency polaritons is similar to the ones obtained in the non-modulated case and is much narrower than the atomic frequency distribution as we operate in the cavity-protected regime. A slight asymmetry between the right and left part of each comb is clearly visible (Fig. 4). This asymmetry, stemming from the coupling between the two polaritons induced by modulation, is well reproduced in the simulated spectrum obtained by numerically integrating the master equation (Methods and Supplementary Information).

Polaritons with multifrequency components and yet small linewidth created using this new technique can lead to applications in quantum networks for the spectral shaping of single-photon emission. More generally, the highly controlled experimental platform described here is ideally suited to study the role of inhomogeneities and disorder in strong light–matter interfaces. It has already allowed us to observe and quantitatively describe the transition between protected and unprotected regimes, yielding a better understanding of the role of dark states. It can be extended to generate a new kind of entangled dark state through collective dissipation useful for quantum sensing and quantum simulation, as recently proposed, as well as to investigate the regime of higher excitation that has been little explored so far. Moreover, when combined with addressing capabilities, controllable disorder as demonstrated here will allow the study of transport in disordered ensembles, with the perspective to quantum simulate the properties of engineered polaritonic materials.

Note added in proof: While preparing this manuscript, we became aware of related results, and both groups decided to coordinate their submissions.

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Methods
Experimental setup
At the core of our experimental setup lies a high-finesse fibre Fabry–Perot optical microcavity with a length of 140 μm. The maximum single-atom coupling strength is $g_0/2\pi = 76$ MHz, larger than the cavity decay rate of $\kappa/2\pi = 15$ MHz and the spontaneous emission rate of $\gamma/2\pi = 3$ MHz ($x$ and $y$ are HWHM values); therefore, we operate in the strong-coupling regime of CQED at the single-atom level. To realize a homogeneous and maximal coupling of all the atoms with the cavity mode, the atoms are trapped at the antinodes of the cavity mode that is resonant with the D2 line of $^{87}$Rb at 780 nm (Fig. 1). We achieve this condition by using an intracavity standing wave at a wavelength of 1,559 nm, commensurate with the atomic line taking into account the Gouy phase shift and by optimizing the phase shift at reflection on the mirrors$^{41,42}$ such that the antinodes of both 1,559 and 780 nm standing waves overlap optimally.

To load the atoms into the cavity, we start with a three-dimensional magneto-optical trap of rubidium atoms positioned 12 mm below the cavity and loaded by a two-dimensional magneto-optical trap. The atoms are then trapped inside a horizontal dipole beam that we can vertically displace with an acousto-optical deflector$^{43}$. In 100 ms, the atoms are moved into the cavity. To load the atoms into the one-dimensional intracavity lattice, we increase the 1,559 nm intracavity power and decrease the intensity of the transport beams. We can adjust the number of atoms from a few tens up to 2,000 by varying the loading parameters of the magneto-optical trap. The atoms are first trapped in a 300 μK deep intracavity lattice at a temperature of 30 μK. We can then increase the trap depth by performing an adiabatic compression of the cloud.

Atomic frequency distributions probed by loss spectroscopy
The strong frequency broadening in our experiment results from the combination of finite atomic temperature, light shifts and level-mixing effects (Supplementary Information). To characterize the frequency distribution of the atoms, we measure the losses of the atom ensemble (prepared in the $F = 2$ hyperfine ground state) induced by a probe beam perpendicular to the cavity axis. This beam is switched on for 0.5 ms at a given frequency before performing the loss measurement. We repeat this process for frequency values across the distribution. To obtain reliable atom number measurements down to low atom numbers, the loss measurement is performed by measuring the vacuum Rabi splitting of the cavity transmission signal (see the ‘Cavity transmission spectrum’ section). The results are shown in Fig. 1. The power of the transverse beam is chosen as a compromise between getting a good signal-to-noise ratio for the trapped atoms’ broad distributions and limiting the losses for the much narrower free atoms’ distribution. As a result, for the latter, almost all the atoms are blasted close to the centre of the $F = 1$ and $F = 2$ peaks (at $-424$ and $267$ MHz, respectively), which is why there is no data point at the top of the corresponding Lorentzian fits. Because the atoms are excited with uncontrolled phases and amplitudes by this transverse beam, we expect (and observe) negligible coupling between the latter and the bright modes of the coupled atom–cavity system even though the resonance condition is fulfilled.

To generate the simulated frequency distribution curves (Fig. 1c), we assume that the atoms are in thermal equilibrium at each site of the optical lattice, described by a harmonic trap with radial frequencies $\omega_{x}$, $\omega_{y}$, and longitudinal frequency $\omega_{z}$. For a trap depth of 1.40 mK, $\omega_{x}/2\pi = 14.5$ kHz and $\omega_{y}/2\pi = 330.0$ kHz. For each atom of a given sample, we first draw $x$, $y$, and $z$ from a normal distribution with variance $\sigma_{x,y,z} = 8.0/\sqrt{F_{16}(|m_{F}|,2)}$. Then, we deduce the values of coupling $g(x,y,z)$ (based on the cavity parameters) and dipole trap intensity $I_{dip}(x,y,z)$ (based on the maximum value at the bottom of the trap, which is estimated from light-shift measurements and corroborated by a direct transmission measurement). The intensity $I_{dip}(x,y,z)$ seen by each atom is used to construct a $16 \times 16$ matrix representing the Stark operator in the $\{F, m_{F}\}$ basis of the $^{87}$Rb manifold (Supplementary Information). The output of the procedure for $N$ atoms is a collection of $16 \times N$ eigenvalues $\hbar \omega_{x,j}$ and eigenvectors $|\psi_{x,j}\rangle$, where $1 \leq j \leq 16$ and $1 \leq N$.

In addition, we assume that the initial atomic population is equally distributed between all the Zeeman sublevels of the $F = 2$ hyperfine ground state. For each atom (labelled by index $k$), we use the previous procedure to compute the $5 \times 16$ transition frequencies $\omega_{k,m_{F}}$ between the 5 ground states $|F = 2, m_{F}\rangle$ and 16 excited states $|\psi_{x,j}\rangle$ (with $-2 \leq m_{F} \leq 2$ and $1 \leq j \leq 16$). Each frequency $\omega_{k,m_{F}}$ is associated to a coupling strength $c_{x,j}(m_{F})$, defined as:

$$c_{x,j}(m_{F}) = \left| \langle \psi_{x,j} | \sum_{n=1}^{16} d_{n} \mathbf{E}_{n}^{x} |F = 2, m_{F}\rangle \right|^{2},$$

where $\mathbf{E}_{n}^{x}$ are the components of the electric field of probe beam $\mathbf{E}_{n}$ expressed in the spherical basis and $d_{n}$ are the dipole matrix elements (Supplementary Information). In the experiment, we have $\mathbf{E}_{n}^{x}/|\mathbf{E}| = 1/\sqrt{2}$ and $\mathbf{E}_{n}^{y}/|\mathbf{E}| = \pm 1/2$. The frequency distributions (Fig. 1c) are then obtained by sorting the transition frequencies $\omega_{k,m_{F}}$ in equal-width bins, weighted by their coupling strength $c_{x,j}(m_{F})$. For a given trap depth, we use an effective temperature $T$ (two to three times the initial experimental temperature) to match the positions of the simulated and experimental curves. The discrepancy stems from heating and depumping effects induced by the transverse beam. The amplitude of each curve is adjusted such that all of them have the same area.

Cavity transmission spectrum
Data acquisition. To measure the cavity transmission spectrum, we first prepare the atoms in the $\{F = 2, m_{F} = 2\}$ Zeeman sublevel by optical pumping. The trap depth is 1.400 ± 30 μK and the temperature is 190 ± 20 μK. To ensure that no atom remains in the other sublevels, we furthermore apply the following sequence: microwave transfer to $\{F = 1, m_{F} = 1\}$, blast of the remaining atoms in the $\{F = 2\}$ hyperfine level and microwave transfer back to $\{F = 2, m_{F} = 2\}$. We estimate that 800 atoms are in the $\{F = 2, m_{F} = 2\}$ state at the end of the preparation sequence. We then probe the cavity-atoms-coupled system by sweeping a tunable laser over a range of 8 GHz in 8 ms. Its intensity is chosen such that the average photon number in the cavity is smaller than unity ($\langle n_{w}\rangle = 0.14$), to be in the low-excitation limit. We measure the transmitted light with a single-photon-counting module. To calibrate the frequency axis of the spectrum, we simultaneously record the transmission signal of a 10-cm-long cavity providing a frequency ruler allowing us to compensate the slight nonlinearity in the frequency sweep and a saturated absorption signal of the probe laser that provides an absolute frequency reference. We estimate the uncertainty of the frequency of the probe laser to be ±8 MHz (1σ), well below the Rabi splittings that are typically 100 to 4,000 MHz.

As the single-shot spectra are strongly discretized (Extended Data Fig. 2), we average 200 such spectra to obtain the experimental data (Fig. 2). As experimental fluctuations of $N$ result into fluctuations of the collective coupling, we group and average the spectra according to their collective coupling $Q$ to avoid excessive broadening due to averaging. We used a 40 MHz bin centred on $Q/2\pi = 1,670$ MHz, to maximize the number of spectra in the bin. We independently fit the high- and low-frequency peaks of the averaged spectrum with a Voigt profile to extract the HWHM of the peaks. The HWHM width ($\delta \omega_{\text{FWHM}} = 28 \pm 2$ MHz) of the low-frequency peaks is a bit larger than the one of the high-frequency peak ($\delta \omega_{\text{FWHM}} = 24 \pm 2$ MHz), probably due to the presence of atom losses during the measurement. The HWHM value quoted in the main text ($\delta \omega_{\text{FWHM}} = 26 \pm 2$ MHz) is the average of the experimental polaritons’ widths.

Simulation of transmission spectrum and frequency distribution.
For a given atom $k$, the excited subspace of the $^{87}$Rb manifold dressed by the dipole light features 16 eigenvalues $\hbar \omega_{x,j}$ and 16 corresponding
For $F_{\text{out}}$, we get a cloud of points $(\Omega, F_{\text{out}})$, which we average with 30 MHz bins of $\Omega$. The vertical error bars are equal to $\pm \frac{\sigma}{\sqrt{N}}$, where $\sigma$ is the standard deviation (s.d.) of the $N$ points gathered within a bin. The resulting curve is plotted in Fig. 3. We have also checked that $F_{\text{out}}$ is robust with respect to the exact value of $A/2\pi$ (Extended Data Fig. 3).

Calculation of $S_{\text{pw}}$. Operating in the strong-coupling regime at the single-atom level enables us to explore the transition from the unprotected to protected regime for a mesoscopic number of atoms ranging from 10 to 550. For such numbers of emitters, we are far from the continuous frequency distribution of emitters usually encountered in solid-state systems and are sensitive to the finite size of the Hilbert space and to the discreteness of the spectrum of the coupled emitter–cavity system. In particular, we can directly compute the eigenvalues and eigenvectors of the coupled system and extract their properties.

In the low-excitation limit, we restrict the dynamics to the one-excitation manifold. For $N$ two-level atoms, it is spanned by the $N+1$ basis states $|1, G\rangle = |\{1\}, G\rangle$ with $|G\rangle = g_{1}\ldots \rangle$ and $|E\rangle = |e_{1}\ldots \rangle$, where $|g_{1}\rangle$ is the ground state of atom $k$.

Due to the 1,559 nm trapping light, a given atom $k$ is excited to the subspace of the $S_{\Omega 2}$ manifold featuring 16 eigenvalues $\omega_{k}$ and eigenstates $|\psi_{k}\rangle$. The single-excitation manifold is then spanned by the $16 \times N + 1$ basis states $|1,\ldots ,G_{k}\rangle = |\{1\},G_{k}\rangle |0,\ldots ,E_{k}\rangle$ with $|G_{k}\rangle = g_{1}\ldots ,g_{k}\ldots g_{16}$ and $|E_{k}\rangle = |e_{1}\ldots ,e_{k}\ldots ,e_{1}\rangle$. In this basis, the Tavis–Cummings Hamiltonian for multilevel atoms is given by an arrowhead matrix, from which we can numerically extract the $D = 16 \times N + 1$ eigenstates $\psi_{i}$ and eigenvalues $\epsilon_{i}$ ($i = 1, \ldots , D$) of the atom–cavity-coupled system. We can then compute the PW of eigenstates $\psi_{i}$ defined as the weight of the $|1, G\rangle$ component:

$$PW_{i} = (1, |\psi_{i}\rangle |\psi_{i}\rangle)^{2}.$$  

To simulate the theoretical curve shown in Fig. 3, we draw $N_{\text{atom}}$ random atomic positions (where $N_{\text{atom}}$ is the atom number), deduce the associated values of frequencies and cavity couplings ($\omega_{k}$, $g_{k}$) and compute the PW of the different eigenstates. We then calculate $S_{\text{pw}}$ (defined as the sum of all the PWs except the two largest ones): $S_{\text{pw}} = 1 - PW_{\text{max1}} - PW_{\text{max2}}$. For each value of $N_{\text{atom}}$, we average $S_{\text{pw}}$ over 300 repetitions.

Polariton modulation

Data acquisition. To modulate the polaritons, we modulate the average light-shifted frequency of the atoms $\omega_{k}(t) = \omega_{k0} + \beta_{k} \cos(\omega_{0} t)$ where $\beta_{k}$ is the modulation index, by modulating the intracavity dipole power at frequency $\omega_{0}/2\pi$. The spectrum in Fig. 4 is obtained by averaging ~200 spectra. Before averaging, we select the spectra with similar atom number $N$, by measuring the dispersive shift $\Delta\omega_{0}$ of the cavity frequency when the atoms are in the $|1, 0\rangle$ state, which is given by:

$$\Delta\omega_{0} = \frac{N_{\text{at}} g^{2}_{k}}{2(2\omega_{0}^{2} - \omega_{k0}^{2})}.$$  

We select the spectra within dispersive shift bin $\Delta\omega_{0}/2\pi = -258 \pm 5$ MHz, corresponding to $N = 1,130 \pm 25$, or to collective coupling $\Omega/2\pi = 2,020 \pm 20$ MHz. The trap depth is 1,400 $\mu$K and the temperature is 190 $\mu$K, similar to Fig. 2. The modulation frequency is $\omega_{0}/2\pi = 120$ MHz.

As we are in the regime where $\Omega \gg \omega_{0}$, the cavity spectrum is given by the following expression (as derived in equation (25) in the Supplementary Information):

$$S(\omega) \propto \sum_{n} \frac{F_{n}(\Omega/2)}{(\omega - \omega_{0} - n\omega_{m} + \Omega)^{2} + \gamma_{c}^{2}} + \sum_{n} \frac{F_{n}(\Omega/2)}{(\omega - \omega_{0} - n\omega_{m} - \Omega)^{2} + \gamma_{c}^{2}}.$$  


The cavity spectrum features two combs of peaks centred at ±Ω. The separation between two consecutive peaks is given by the modulation frequency ω_{m}. The amplitude of each peak is given by β_{p}(ω_{m}/2), and therefore, the modulation index for the polaritons is given by β_{s} = β_{p}/2. We are then able to directly control the modulation of the polaritons in frequency and amplitude with the modulation of the trapping light. The width of each peak in given by γ_{s} = γ_{s}/Δω, identical to the width of the non-modulated homogeneous case as we are in the cavity-protected regime.

As the number of atoms inside the cavity modes fluctuates, the transmission peak of the high-frequency polariton is best fitted by a Voigt function:

\[ \sum_{p} \beta_{p}(\omega_{m}) \text{Voigt}(\omega - \omega_{0} - n\omega_{m} \pm \Omega, \gamma_{s}, \omega). \tag{7} \]

where Voigt(\omega, γ_{s}, \omega) = \frac{1}{\pi}\frac{\gamma_{s}}{\omega_{s}^{2} + (\omega - \omega_{s})^{2}} and wofz is the Faddeeva function. This function provides a very good fit of the experimental data, except for the slight asymmetric shape (relative variation, ±7%) between the right and left part of each comb.

**Simulation of cavity transmission spectra.** To justify the function used to fit the experimental data and to confirm the analysis presented in the Supplementary Information, we simulate the cavity spectrum by numerically integrating the master equation using QuTiP^{32}, for an emitter–cavity system probed by a laser swept in frequency at the same rate as in the experiment (1 GHz ms⁻¹), and with the same power. We calculate the population of the state (1, G) after binning the simulated results to match the experimental resolution of 2.5 MHz. By using the parameters given by the experimental fit, we obtain a very good agreement between the experimental data and simulated spectra even for the asymmetrical shape of the comb.

**Modulation transfer measurement.** To measure the linear modulation transfer β_{s} = β_{p} (Fig. 4, inset), we measure ~100 spectra for each value of β_{p}. We reduce the frequency range of the probe laser scan by a factor of 4 to increase the frequency resolution. Before averaging the spectra, we frequency shift each spectrum so that the multiprime centres of all the spectra are aligned, to compensate for atom number fluctuations. Compared with raw averaging, this technique improves the accuracy of the relative amplitudes of the different peaks in the average spectrum, to which the value of β_{s} is very sensitive. We then fit the cavity spectrum with equation (5) to extract the value of β_{s}.

Since β_{p} is the result of a nonlinear fit, we resort to a non-parametric bootstrap method to determine its uncertainty. For each averaged spectrum (and thus each value of β_{p}), we generate 500 synthetic spectra A_{k}(1 ≤ k ≤ 500). Then, each synthetic spectrum A_{k} is fitted with equation (5), providing a fitted parameter β_{p,k}. The error bar for β_{p} is defined as ±s.d. of the set {β_{p,k}; 1 ≤ k ≤ 500}. To obtain the value of β_{s}, we measure the intensities of the different frequency components of the trapping light, using the transmission spectrum of the frequency-scanned cavity. Starting from \[ \delta \omega_{0}(t) = \delta \omega_{0}(t) - \omega_{s}^{0} = \beta_{0}\omega_{m}\cos(\omega_{m} t), \]

we get:

\[ \beta_{s} = \frac{\delta \omega_{0}^{\text{max}}}{\omega_{m}} = \frac{\delta \omega_{0}^{\text{max}}}{\omega_{s}^{0}} = \frac{\omega_{s}^{0}}{\omega_{m}} \delta \omega_{0}^{\text{max}} \frac{\delta \omega_{0}^{\text{max}}}{\omega_{s}^{0}} P_{D} \tag{8} \]

where \[ \delta \omega_{0}^{2n} = -1.320 \text{ MHz} \] is the average atomic light shift; \[ \delta \omega_{0}^{2n} = 130 \text{ MHz} \] is the modulation frequency; and \[ \delta \omega_{0}^{\text{max}} \] and \[ \omega_{s}^{0} \] are the constant and \[ \omega_{s}^{0} \] frequency-dependent components of the intensity of the trapping light, respectively. Uncertainties in \[ \beta_{p} \] are propagated from the experimental uncertainties of \[ \omega_{s}^{0}, \omega_{0}^{\text{max}} \] and \[ \delta \omega_{0}^{\text{max}} \].

**Code availability**

The data that support the findings of this study are available via Zenodo at https://doi.org/10.5281/zenodo.7308947.

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**Author contributions**

M.B., F.F. and R.L. built the experimental setup. M.B., P.-A.B., S.S., F.F. and R.L. performed the measurements and analysed the data. P.-A.B., S.S., J.R. and R.L. interpreted the results and wrote the manuscript with input from all the authors.

**Competing interests**

The authors declare no competing interests.

**Additional information**

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Extended Data Fig. 1 | Simulated atomic frequency distribution for different trap depths. For each trap depth $U_0$, the temperature $T$ used in the simulation corresponds to the typical experimental value based on time-of-flight measurements. When $U_0$ increases, the mean frequency of the distribution decreases linearly – as expected with red-detuned off-resonant light – and the width $\Delta \omega$ of the distribution increases. At low trap depth $U_0 = 310 \pm 10 \mu K$, the distribution has mainly one lobe, corresponding to the $|F = 2 \rangle \rightarrow |F' = 3 \rangle$ transition. For larger trap depths, two-photon couplings at 1559 nm mix the excited state hyperfine levels and two extra lobes appear in the distributions, at lower frequencies, corresponding roughly to transitions $|F = 2 \rangle \rightarrow |F' = 2 \rangle$ and $|F = 2 \rangle \rightarrow |F' = 1 \rangle$. This illustrates the tunability of the inhomogeneous distribution with the intensity of the trapping field.
Extended Data Fig. 2 | Single-shot experimental spectrum. As we probe the coupled system in the low excitation regime, we collect few photons in transmission and the spectrum is discretized (orange dots). For each spectrum, we compute the fraction $F_{\text{out}}$ of photons (identified with black triangles) that lies outside of a frequency window $\Delta f/2\pi$ (green colored area), centered on each peak distribution (red and blue dashed lines).
Extended Data Fig. 3 | Robustness of $F_{\text{out}}$ with respect to the size of the exclusion window. Here, we show $F_{\text{out}}$ for several values of $\Delta f/2\pi$ chosen to define the exclusion window, together with the simulated sum of the two highest PW, $S_{PW}$, as in Fig. 3 of the main text (red dots). The result is rather robust: when $\Delta f/2\pi$ decreases, the shape of $F_{\text{out}}$ remains the same and is shifted upwards as expected. Data are presented as mean values ± s.e.m. obtained with an average of 55 spectra per point.