Universal pair polaritons in a strongly interacting Fermi gas

Cavity quantum electrodynamics (QED) manipulates the coupling of light with matter, and allows several emitters to couple coherently with one light mode. However, even in a many-body system, the light–matter coupling mechanism has so far been restricted to one-body processes. Leveraging cavity QED for the quantum simulation of complex, many-body systems has thus far relied on multi-photon processes, scaling down the light–matter interaction to the low energy and slow time scales of the many-body problem. Here we report cavity QED experiments using molecular transitions in a strongly interacting Fermi gas, directly coupling cavity photons to pairs of atoms. The interplay of strong light–matter and strong interparticle interactions leads to well-resolved pair polaritons—hybrid excitations coherently mixing photons, atom pairs and molecules. The dependence of the pair-polariton spectrum on interatomic interactions is universal, independent of the transition used, demonstrating a direct mapping between pair correlations in the ground state and the optical spectrum. This represents a magnification of many-body effects by two orders of magnitude in energy. In the dispersive regime, it enables fast, minimally destructive measurements of pair correlations, and opens the way to their measurement at the quantum limit and their coherent manipulation using dynamical, quantized optical fields.

One of the most striking successes of quantum science is the ability to engineer the interaction between light and matter, culminating in the strong coupling regime of cavity quantum electrodynamics (QED). Cavity QED is already a corner stone of quantum networks and quantum information processing. However, it is now emerging as a new tool for quantum simulation with quantum gases, as it provides unique features such as long-range, collective interactions, controlled dissipation enabling novel non-equilibrium dynamics, and real-time measurements. However, the light–matter interaction has so far been limited to the dipole coupling between photons and individual atoms, which dominates the energy scales of quantum gases by several orders of magnitude. As a result, an interplay between many-body physics in a quantum gas and light–matter interactions could only be observed in the dispersive regime, effectively scaling down the light–matter interaction to the level of the motional degrees of freedom of atoms.

In a many-body system in free space, it is well known that photons can not only be absorbed and re-emitted by individual atoms but also exchanged between atoms, yielding a dipole–dipole interaction. Excited molecular states then form in the attractive branches of this interaction potential, yielding photoassociation (PA) resonances in the optical spectrum. When driving such a PA line, photons couple directly to pairs of free atoms separated by a distance $R_0$, the Condon length of the target molecular state, much shorter than typical interatomic distances in quantum gases. PA has been used for pair correlation measurements, however only through losses in the incoherent regime. Coherent Rabi oscillations on ultra-narrow PA transitions have been observed in weakly interacting gases of two-electron atoms, as shown in Fig. 1a. Our experiment brings PA into the strong coupling regime, where the interaction between photons and atom pairs overcomes all dissipative processes. In such a regime, pairs, molecules and cavity photons coherently hybridize into composite quasiparticles: pair polaritons. These coherent excitations inherit from their photonic component the fast dynamics of the resonant light–matter interaction, much faster than that of the many-body physics in the gas, and a weak dissipation channel into the environment enabling direct optical detection. From their matter, they inherit the universal properties of the short-distance pair correlations of the quantum gas, which are imprinted onto the optical spectrum.

This combination of light–matter and atom–atom interactions is embodied in the collective Rabi frequency $\Omega$ verifying

$$\Omega^2 = \Omega_0^2 \int dR \left| g(R) \right|^2 \int drr' f(r)f^*(r') \left\langle \psi_{\frac{r}{2}}(R + \frac{r}{2}) \psi_{\frac{r}{2}}(R - \frac{r}{2}) \right\rangle,$$

where $f(r)$ is an orbital describing the relative motion of the two atoms in the target molecular state, $\Omega_0$ is the single-photon–single-pair Rabi frequency and $g(R)$ the cavity mode function. $\psi_{\frac{r}{2}}(r)$ are field operators annihilating atoms in states 1 and 2 at point $r$ (see Methods).

In the linear response regime, the expectation value is taken in the unperturbed thermal state of the gas, and the integral counts the total ...
number of pairs of atoms overlapping with the excited molecular state. Thus, the pair correlations are directly scaled up by $\Gamma_{\text{pa}}$, itself proportional to the electric field, thereby amplifying the pair correlation signal to scales much larger than the Fermi energy. In the strong coupling regime, the cavity translates this signal in the optical spectrum, where $\Omega$ can be directly read out in the linear regime. In free space, equation (1) still holds but does not yield any spectral signatures in the low saturation regime, so that its use for measuring correlations is restricted to incoherent loss effects.

We use Fermi gases of $^6\text{Li}$ comprising $4.6 \times 10^5$ atoms equally populating the two lowest hyperfine states, denoted as $|1\rangle$ and $|2\rangle$, at a temperature $T = 0.08(2) T_F$ (where $T_F$ is the Fermi temperature), within the cavity mode of the high finesse cavity with the intensity decay rate $\kappa = 2\pi \times 77$ kHz. A homogeneous magnetic field $B$ set in the vicinity of the broad Feshbach resonance at 832 G brings the gas into the strongly interacting regime, where it explores the BEC–BCS crossover\textsuperscript{24,25}. In the PA process, the photons address the two-body wavefunction of atom pairs as shown in Fig. 1b. We probe the optical excitation spectrum of the system using transmission spectroscopy, with light linearly polarized along the magnetic field direction (see Methods). The frequency of the probe beam is swept through the resonance fast compared with the dynamical time scales of the gas, avoiding optomechanical effects\textsuperscript{26}.

Figure 2 shows a typical transmission spectrum for a gas at unitarity, where the detuning $\Delta_c/2\pi$ between the cavity and the atomic D2 $\pi$-transition spans 6 GHz. The spectrum is dominated by the strong coupling of photons with the atomic transition at $\Delta_c = 0$, yielding the smooth background variation of the transition resonance with respect to the empty cavity at $\Delta_c = 0$, with $\Delta_c$ the detuning of the probe with respect to the empty cavity resonance. For large negative values of $\Delta_c/2\pi$, we observe three successive avoided crossings corresponding to three PA transitions (Fig. 2c,d,e), denoted 1, 2 and 3. We observed similar patterns for several PA resonances corresponding to bound states in the $1^1\Sigma^+_g$ potential\textsuperscript{27}, including the vibrational level $v = 81$ located at $\Delta_c/2\pi = -25.998$ GHz, denoted PA4. The presence of clear avoided crossings confirms that the strong coupling is reached for each of these PA transitions.

We fit the spectrum around each PA transition using an analytical model (see Methods) that enables the extraction of the Rabi frequency and the atomic dispersive shift, providing the purely atomic contribution linking to the total number of atoms $N$ for each measurement. The Rabi frequencies extracted for PA1–PA4 are $2\pi \times 14.6(10), 11.0(4), 8.0(1)$ and 18.9(1) MHz, respectively, exceeding both the atomic and the cavity decay rates.

The correlation function in equation (1) depends on the shape and range of the molecular orbitals. We estimate that $R_c = 244 a_0, 210 a_0, 181 a_0, 164 a_0$, with the Bohr radius $a_0$, for PA1–PA4, respectively, such that $|f(r)|$ has a range that is much shorter than the interparticle distance, the de Broglie wavelength and the photon wavelength (see Methods).

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**Figure 1 | Concept of the experiment.**

**a.** A degenerate, strongly interacting two-component Fermi gas is placed inside a cavity. A beam linearly polarized along the bias magnetic field $B$ measures the transmission through the cavity. **b.** Sketch of the two-atom ground state wavefunction for the BEC, unitary and BCS situations (light blue, blue and dark blue solid lines, respectively) in the ground state, and wavefunction of a molecular state (solid black line) at the Condon point $R_c$. The collective Rabi frequency $\Omega$ of the process exceeds the rate of dissipation yielding a pair of resolved dressed states in the spectrum.

**Figure 2 | Strong coupling on PA transitions.**

**a.** Frequency diagram relevant to the experiment. We address three PA transitions at $\omega_{pa}$, $\omega_{pa}$, and $\omega_{pa}$ below the single-atom D2 $\pi$-transition at $\omega_{pa}$. The cavity–atom and probe–cavity detunings $\Delta_\text{c}$ and $\Delta_\text{a}$ are independently controlled by tuning the cavity resonance and the probe frequency. **b.** Cavity transmission spectrum of a unitary Fermi gas below the D2 $\pi$-transition. The solid lines indicate three PA transitions labeled as 1, 2 and 3 (as in a). A few narrower PA signals can also be seen, which we attribute to PA from other interaction potentials. c–e. Close-up view of the vicinity of the three PA lines showing avoided crossing patterns.
This suggests that the coupling to PA transitions depends only on the nature of the pairs in the ground state and not on the details of the molecular transition. A calculation using a simplified orbital model shows that the variations of Rabi frequency are equal to that of Tan’s contact up to a small correction accounting for the finite value of $R_c/a$ (see Methods), with $a$ the s-wave scattering length. Specifically, we expect $\Omega^2 = I_N k_c [1 - R_c/a]$, where $I$ is the dimensionless, trap-averaged contact and $k_c$ is the Fermi wave-vector. Figure 3b presents the evolution of $\Omega^2 = \frac{\Omega_\infty^2}{\Omega_\infty} [1 - \frac{R_c}{a}]^2$, normalized by its value at unitarity $\Omega_\infty$, for the different PA lines. We observe the data collapse on each other, a striking manifestation of the universal character of the interaction dependence. The solid and dashed lines in Figure 3b present the interaction dependence of the trap-averaged contact at $T = 0$ (refs. 29,39), without any fit parameter, in very good agreement with the data. We attribute the slight deviation in the BEC regime to the limited accuracy of the molecular orbital model. This demonstrates the direct mapping between the optical spectrum of the cavity and the many-body physics of the quantum gas. Remarkably, the Rabi splitting spans tens of MHz as the gas explores the BEC–BCS crossover, despite the fact that its Fermi energy is two orders of magnitude lower. Such an amplification is permitted by the scaling of $\Omega$ with the electric field, allowing the Rabi splitting to reach arbitrarily large values, limited only by the cavity mode volume and the dipole moment of the PA line.

The contact directly measures the number of pairs at short distance in a given volume of the gas. Equation (1) thus shows that the Rabi frequency scales with the square root of the number of pairs at a distance $R_c$, reminiscent of the scaling with the number of emitters in the Tavis–Cummings model. Having established the connection between the variations of $\Omega$ with interactions and the contact, we now consider the contact as known from theory and study the Rabi frequencies $\Omega$. Figure 3c presents the evolution of $\Omega$ as a function of the contact $\bar{c}$, for the four different PA transitions as a function of the contact (Fig. 3c). The square root dependence highlights the coherent coupling of Fermion pairs with photons. Using a simplified model of the molecular orbital to connect the Rabi frequency with the number of molecules at distance $R_c$ (see Methods), the fit provides an order-of-magnitude estimate for the single-photon–single-pair Rabi frequency $\Omega_\infty$. We obtain $\Omega/\Omega_\infty = 595(11)$, $492(5)$, $435(7)$ and $765(5)$ kHz for the four PA lines, respectively, comparable to the single-atom–single-photon Rabi frequency of $2 \pi \times 780$ kHz for the D2 line transition in the system. Although pairs in the unitary gas are inherently a many-body effect, and cannot be isolated individually, we nevertheless conclude that our cavity has a cooperativity for single pairs and single photons approaching one. This suggests that all of the quantum optics protocols designed in the context of single atom–photon interactions could be directly generalized to Fermion pairs in strongly interacting gases.

Figure 3 | Interaction-dependent photon–pair coupling. a, Transmission spectra around PA2 in the BEC, unitarity and BCS regimes. Horizontal axis is shifted by $\Delta/2\pi = -3.27, -3.20$ and $-3.14$ GHz, respectively, for visibility. The solid lines indicate the fitted locations of the PA line. The colour scale is as for Fig. 2. b, Magnetic field dependence of the collective Rabi frequencies $\Omega$ (inset) and the evolution of $\Omega^2$ (see the main text), normalized by its value at unitarity $\Omega^2_\infty$, for PA1 (green open circles), 2 (purple filled diamonds), 3 (orange open squares) and 4 (light blue crosses) as a function of $1/k_c a$. The black solid and red dashed lines represent the trap-averaged contact calculated by Gaussian pair fluctuations and quantum Monte Carlo, respectively, normalized by the value at unitarity. c, Scaling of $\Omega$ as a function of the trap-averaged contact $\bar{c}$, inferred from Gaussian pair fluctuations. The solid lines show the square root fit to the data. Error bars, standard deviation.
An example of raw photon counts obtained for a single interrogation is shown in Fig. 4b at \( \Delta /2\pi = -26.056 \) GHz. We fit the positions of the two resonances corresponding to the \( \pi \) and \( \sigma^\prime \) components using a double Lorentzian model. A typical spectrum corresponds to 16 intracavity photons on the transmission resonance, and of the order of one spontaneous emission event over the entire cloud. The positions of the resonances at a given \( \Delta /2\pi \) enables retrieval of both the atomic dispersive shift \( \delta \) and the collective Rabi frequency of the molecular transition \( \Omega \). The pair-polariton spectrum extrapolated from this single interrogation (Fig. 4a) shows excellent agreement with a full data set covering all detunings taken for clouds prepared in similar experimental conditions.

Such a single interrogation can be repeated 50 consecutive times on the same atomic ensemble, separated by 10 ms, allowing for equilibration of the cloud between each interrogation. Figure 4c shows \( \delta \) and \( \Omega \), extracted from each of these single consecutive interrogations. The retrieved values of \( \Omega \) reflect the time evolution of two-body correlations for a single atomic ensemble. We observe a decrease of \( \Omega /2\pi \) from 18 to 15 MHz after the 50 consecutive interrogations, and a decrease of \( \delta \) by 27%, reflecting atom losses. Approximately half of these losses can be attributed to the finite lifetime of the gas due to the strong interactions. The remaining measurement-induced atom losses are \( 1.9(1) \times 10^3 \) atoms per interrogation, 0.2% of the initial atom number per interrogation.

To confirm that the many-body physics is preserved in spite of the repeated measurements on a single atomic ensemble, we evaluate \( \Omega^2 = \Omega /Nk_b \), at each point in time. The result (Fig. 4d), normalized by its initial value, shows no discernible decay, where the noise originates predominantly from photon shot noise. An average of 20 traces of \( \Omega \) taken on different clouds and at different values of detuning in the dispersive regime of the PA4 resonance is shown in Fig. 4d. This quantity is directly proportional to the trap averaged contact \( \overline{T} \), and remains constant, demonstrating the weakness of the heating originating from the measurements. Independent temperature measurements (see Methods) indicate an initial temperature of \( 0.083(2)T_0 \) and a final temperature of \( 0.10(2)T_0 \) after the 50 interrogations, for which we would expect a decrease of the trap-averaged contact by about 4% (ref. 3).

The ability to observe pair correlations in time while preserving the many-body physics is an ideal starting point for future theoretical and experimental investigations of quantum noise and back-action mechanisms for correlation measurements in many-body systems. This will require a detailed understanding of technical and fundamental sources of noise, including noise originating from the background coupling to atomic density. Eventually, we expect the quantum noise spectra to carry fundamental information on higher order correlations, which will present the fascinating possibilities of combining quantum-limited sensing with strongly correlated Fermions. In our work, optomechanical effects originating from the motion of atoms triggered by the probe are absent, due to the short interrogation time. For slower probe sweeps, we will enter a regime in which optomechanical nonlinearities or even dynamical back-action will emerge.

Directly coupling the cavity field to the pair-correlation function in the dispersive regime suggests the possibility of engineering photon-mediated pair–pair long-range interactions, or pairing with a momentum space structure, which is uncharted territory in quantum simulation of strongly correlated materials. Beyond these fundamental questions, the weakly destructive and time-resolved character of the cavity-assisted measurement will be of immediate, practical interest in the study of correlations after quenches, such as spin diffusion, or during slow transport processes, complementing other high-efficiency methods. Finally, the addition of exquisite control over photons of a high finesse cavity to the existing cold molecules toolbox opens the way to dissipation engineering of cold chemistry.

**Online content**

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions...
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Methods

Experimental procedure
A quantum degenerate, strongly interacting Fermi gas of \(^{6}\text{Li}\) was produced following the method described in ref. 3. Atoms captured in a magneto-optical trap are first transferred into a standing wave optical dipole trap at 1,064 nm created by the TEM\(_{00}\) mode of the cavity. Before evaporative cooling, the magnetic field is ramped to 832 G, the location of the broad Feshbach resonance between the two lowest hyperfine states (I) and (2). Evaporative cooling is first performed in the cavity before transfer to a crossed optical dipole trap, formed by two running wave beams at 1,064 nm with a waist of 33 μm intersecting at an angle of 36° at the center of the cavity. This yields a spin-balanced unitary Fermi gas containing 4.6 × 10⁵ atoms with a temperature of \(T = 0.08(2)T_{\text{F}}\).

The cloud is recompressed in 50 ms followed by 50 ms hold time in a trap with frequencies of 2\(\pi \times 188(9)\), 579(28) and 608(30) Hz, with the weakest confinement along the cavity direction, before the cavity interrogation. This avoids unwanted trap losses and ensures maximal coupling of the cloud with the TEM\(_{00}\) mode of the cavity. The recompression is accompanied by a magnetic field ramp from 832 G, at which the whole evaporation is performed, to various values in order to change the interatomic interaction.

We send probe beam pulses at 671 nm mode-matched to the TEM\(_{00}\) mode of the cavity. Each probe pulse is 500 μs long and frequency-swept over 40 MHz, which controls \(\Delta /2\pi\). Transmission from the cavity is collected on a single-photon counter. At the end of an experimental mode of the cavity, each probe pulse is 500 μs long and frequency-swept before transfer to a crossed optical dipole trap, formed by two running wave beams at 1,064 nm with a waist of 33 μm intersecting at an angle of 36° at the center of the cavity. This yields a spin-balanced unitary Fermi gas containing 4.6 × 10⁵ atoms with a temperature of \(T = 0.08(2)T_{\text{F}}\).

The probe beam is linearly polarized and we set it either along or tilted from the magnetic field orientation depending on the measurements that we perform (see the main text). The polarization component collected on a single-photon counter. At the end of an experimental mode of the cavity. Each probe pulse is 500 μs long and frequency-swept before transfer to a crossed optical dipole trap, formed by two running wave beams at 1,064 nm with a waist of 33 μm intersecting at an angle of 36° at the center of the cavity. This yields a spin-balanced unitary Fermi gas containing 4.6 × 10⁵ atoms with a temperature of \(T = 0.08(2)T_{\text{F}}\).

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For a PA process, a minimal model of the light–matter coupling interaction can be written as

\[ \hat{F} = \int d\mathbf{R} g(\mathbf{R}) f(\mathbf{r}) \psi_m^\dagger(\mathbf{R}) \hat{\psi}(\mathbf{R} - \frac{\mathbf{r}}{2}) \hat{\psi}(\mathbf{R} + \frac{\mathbf{r}}{2}), \]

where \( \psi_m^\dagger(\mathbf{r}) \) is a field operator annihilating a Fermion in state 1 or 2 at position \( \mathbf{r} \), and \( \hat{\psi}(\mathbf{r}) \) annihilates a molecule with a center of mass at position \( \mathbf{r} \). \( g(\mathbf{R}) \) is the mode function of the cavity, and \( f(\mathbf{r}) \) describes the relative motion of atoms in the target molecular state. We evaluate the expectation value in equation (3) under the low saturation hypothesis \( \langle \hat{\psi}_m^\dagger(\mathbf{r}) \hat{\psi}(\mathbf{r}) \rangle \rightarrow 0 \), to obtain equation (1).

For the purposes of deriving a mean-field, low-saturation description, we consider the cloud as a collection of two-level systems, representing pairs and molecular states, described by a weakly fluctuating collective spin operator \( \hat{\sigma} \). We then explicitly introduce pair-polariton creation and annihilation operators \( \hat{\sigma}^\dagger \) and \( \hat{\sigma} \) through a Holstein–Primakov transformation

\[ \hat{\sigma} = \frac{\hat{F}}{\sqrt{\langle \hat{\sigma}, \hat{\sigma}^\dagger \rangle}}, \]

similar to ensembles of two-level systems described by the Tavis–Cummings model. The Hamiltonian is then expressed as

\[ \hat{H} = \delta a^\dagger a + \frac{\hbar \Omega_0}{2} \langle \hat{\sigma}, \hat{\sigma}^\dagger \rangle (\hat{a}^\dagger \hat{a}^\dagger - \hat{a} \hat{a}). \]

We thus recover a coupled harmonic oscillator model. The decay of the molecular state and cavity field can then be modeled by jump operators \( \hat{\sigma}^\dagger \hat{\sigma} \) and \( \hat{\sigma} \hat{\sigma}^\dagger \) in a master equation approach. At the mean-field level, at which we expect the model to be valid, this is strictly equivalent to the classical wave description, yielding equation (2) for the transmission coefficient.

**Molecular orbitals**

The molecular states that we target are bound in the \( 2S + 2P \) asymptotic potential. We work with \( \pi \)-polarized light, at strong magnetic fields and the binding energies of PA1, PA2 and PA3 are much smaller than the fine structure splitting of lithium and of the order of the Zeeman shift. Therefore, for these states we use a minimal model of the molecular potential originating from the D2 \( \pi \)-transition only, with transition dipole moments computed from the Breit–Rabi formula. This model suggests that they correspond to vibrational levels -12, -13 and -14 below the continuum, in the molecular potential resulting from the exchange of \( \pi \)-polarized photons between atoms.

The validity of this model can be tested using the Leroy–Bernstein formula connecting the van der Waals coefficient \( C_i \) to the location of the highly excited vibrational bound states. For the three PA lines PA1–PA3 used, a fit leaving the location of the continuum as a single free parameter describes the binding energies better than 5%. Note however that for most of the transitions that we observed, in particular closer to the D1 line or with other polarizations, we could not reproduce the spectrum with such a simple model. For PA4, the vibrational level \( \nu = 81 \) in the \( 1\Sigma_g^+ \) orbital, we use the documented \( C_i \) coefficient directly to calculate the Condon radius.

**Universality**

The connection between the pair correlations in equation (1) and Tan’s contact is standard. Following ref. 27, we write the pair correlation function as

\[ \langle \psi_m^\dagger(\mathbf{r}) \psi_m^\dagger(\mathbf{r}) \rangle = \sum_i n_i \phi^{(i)}(r_t, r_z) \phi^{(i)}(r_t, r_z), \]

where \( n_i \) are real positive coefficients and \( \phi^{(i)}(r_t, r_z) \) is a set of short-range, normalized two-body orbitals that obey the Bethe–Peierls boundary conditions

\[ \phi^{(i)}(r_t, r_z) \rightarrow A^{(i)} \left( \frac{1}{|r_t - r_z|} \right)^{\frac{1}{a}}, \]

where \( A^{(i)} \) is a normalization coefficient and \( a \) is the scattering length. Therefore, we get

\[ \langle \hat{\psi}_m^\dagger(\mathbf{r}) \hat{\psi}(\mathbf{r}) \rangle = \sum_i n_i \phi^{(i)}(r_t, r_z) \phi^{(i)}(r_t, r_z), \]

in the relevant range of relative distances. We introduce the contact as

\[ C(\mathbf{r}) = \frac{16\pi^2}{\hbar} \sum_i n_i A^{(i)} |\phi^{(i)}(r_t, r_z)|^2, \]

and thus express the commutator as

\[ \langle [\hat{F}, \hat{F}] \rangle = \int d\mathbf{R} |g(\mathbf{R})|^2 C(\mathbf{R}, a) \int_0^\infty \rho(r_t, r_z) \left( \frac{1 - r_t}{a} \right)^2. \]

This is the expected behaviour of a set of \( N_{2p} / 2 \) identical emitters coherently coupled to one mode of the field. Our experiment thus represents the PA counterpart of the celebrated Tavis–Cummings model.

To account for the finite value of \( R_C \) in the theory–experiment comparison, we follow ref. 28 and model the target molecular orbital as a square box of width \( L \) centered around \( R_C \). The overall coupling to the field is modulated by the mode function, such that on average only half the pairs contribute, and the Rabi splitting reads

\[ \Omega = \Omega_0 \left( \frac{N_p}{2} \right). \]

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The data points in Fig. 3 include the finite Condon radius correction, estimated from the \( C_i \) coefficient and the binding energies for the four different transitions. For data in the far BEC regime, the term accounting for the finite size of the molecule contributes approximately 15–20% to the scaling.

**Number of short-range molecules**

To connect the scaling with the contact observed in the experiment to the single-atom–single-pair coupling, we estimate the number of pairs in the gas addressed by the PA. To this end, we evaluate the length \( L \) introduced in the above theory. This can be done by supposing that...
the molecular potential close to \( R \) is approximately linear with the position \( r \) as 

\[ V(r) = -E_b \left( 1 - \frac{3r}{R_C} \right), \]

where we have expressed the \( C_3 \) coefficient in terms of the binding energy \( E_b \). Therefore, the molecular orbital is approximately an Airy function, with a width \( L = \left( \frac{\hbar^2}{3mE_b} \right)^{1/3} \), where \( m \) is the atomic mass and \( \hbar \) is the Planck constant divided by \( 2\pi \). We thus obtain \( L = 28.9a_0, 23.6a_0, 19.4a_0 \) and \( 12.6a_0 \) for PA1–PA4, respectively.

By virtue of Tan’s relations, for a distance \( s \) much smaller than other many-body length scales, the number of pairs \( dN_p \) with a volume \( d^3R \) centered around point \( R \) is

\[ dN_p(R, s) = \frac{\mathcal{C}(R)s}{4\pi} d^3R. \]

Integrating over the whole cloud we get the total number of pairs at distance \( s \) in terms of the integrated contact \( I_s \):

\[ N_p(s) = \frac{3s}{4\pi} = \frac{2N_k s}{4\pi}, \]

with \( I_s = \frac{3}{2} N_k \), and \( N \) the total atom number. Note that the factor of 2 due to the averaging over the \( \cos^2 \) mode function has already been incorporated in the scaling at the level of equation (4).

**Single-shot estimate of the Rabi frequency**

We model the position of the dressed states close to a PA transition by

\[ E_z = -\frac{\hbar(1-\alpha)(\Delta_a r - \Delta_{pa})}{2} \pm \hbar \sqrt{\Omega^2 + (\Delta_a r - \Delta_{pa})^2} + \hbar \delta_{at}, \]

where \( \Omega \) is the Rabi frequency, \( \alpha \) is the linear slope originating from the dressing of the cavity with the atomic transition, \( \Delta_{pa} \) is the position of the PA transition with respect to the atomic transition and \( \delta_{at} \) is the atom-induced dispersive shift. We found that both dressed states emerging from each polarization component fit this model with the same Rabi frequency.

Accounting for the presence of one resonance for each polarization, we determine \( \alpha, \delta_{at}, \) and \( \Omega \) from the measurement of the position of the two transmission resonances.

**Data availability**

All data files are available from the corresponding author upon request. Accompanying data, including those for figures, are available from the Zenodo repository (https://doi.org/10.5281/zenodo.4896757).
Extended Data Figure 1 | Fit to the spectrum. a, Spectrum of PA4 at 832 G averaged over three realizations. b, Spectrum reconstructed by equation (2) using the fit results. The solid and dashed lines indicate the fitted positions of the PA resonance and the dispersively shifted cavity resonance. The colour scale is identical to that in the main text.
**Extended Data Figure 2 | Magnetic field dependence of the binding energies.**

Positions of PA1–PA4 (green open circles, purple filled diamonds, orange open squares and light blue crosses, respectively) as a function of magnetic fields. The value at 730 G is subtracted for clarity. Linear fits presented by the solid lines yield 0.31, 0.67, 0.89 and $-0.83 \text{ MHz G}^{-1}$ for the four PA resonances, respectively.