

Collective light shifts of many longitudinal cavity modes induced by coupling to a cold-atom ensemble

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We experimentally study the interaction between a cold-atom cloud and many longitudinal modes of a high-quality Fabry-Perot cavity, by measuring signatures of collective light shifts in the cavity transmission spectrum of an optical frequency comb probe. Using a resonator coupled to more than 10^5 intracavity atoms, we detect significant shifts of ~ 100 cavity modes simultaneously, which is a direct manifestation of physics beyond the hitherto explored regime of cavity-cold atom interaction with only single or few longitudinal modes at a time. For the cavity mode closest to the atomic resonance, we demonstrate a bistability in the transmission spectrum, arising due to a combined coupling of the cloud to an external pump laser and a cavity mode probed by the optical frequency comb. These results provide a first step toward deeper exploration of multifrequency cavity quantum electrodynamics, where ultrashort pulsed sources could be used for optical manipulation, cooling, and entanglement of cold atoms in a resonator.

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I. INTRODUCTION

Optical resonators have been a mainstay of atomic physics for the last four decades. The considerable enhancement of light-atom interaction, provided by coupling to a high-quality optical cavity, has led to unprecedented levels of quantum control, down to single atoms and photonic quanta [1–3]. For an ensemble of cold atoms, the dispersive regime of this interaction, occurring when resonator frequency is detuned sufficiently far from the optical transitions that resonant photon absorption is negligible, has also proven crucial for inducing spectacular collective effects such as cavity cooling [4–7], squeezing [8,9] and self-organization [10–13]. However, up to now collective effects in the dispersive regime of cavity-cold atom interaction were experimentally studied only using single and few longitudinal [14,15] and many nearly degenerate transverse [16–18] cavity modes with continuous-wave (cw) laser driving.

In contrast, recent theoretical studies highlight the exceptional promise of longitudinally multimode optical cavities with multifrequency drives, e.g., an optical frequency comb (OFC), for advancing cavity quantum electrodynamics (cQED). For instance, using an OFC excitation of many cavity modes can create a complex and highly controllable optical potential inside the cavity, which can be precisely shaped by tuning the OFC parameters, enabling the realization of effective atom-atom potentials beyond the all-to-all coupling

paradigm [19,20]. An OFC-based approach has been proposed in Ref. [21] as a possible multimode cavity platform that can realize supersolids and droplets with phononlike excitations. Multifrequency excitation with an OFC has also been suggested for implementing quantum annealing and Hopfield associative networks [22], where for a K -site lattice the full site-to-site interaction matrix can be dynamically controlled by shaping up to $K(K+1)/2$ lasers derived from an OFC. Furthermore, driving a cold-atom cloud with multifrequency laser light can lead to spontaneous formation of clusters in atomic density, the size of which is a direct signature of the drive and free spectral range of the cavity [23]. These numerical results also show that multifrequency driving can enhance cavity cooling compared to a single cw drive, which could enable cooling of atomic and molecular species with transitions at wavelengths below the optical region, where only ultrafast radiation sources are available [24]. Finally, a recent theoretical work has suggested that driving with weak optical pulses containing multiple spectral lines coinciding with cavity resonances can be used to project an atomic ensemble into an entangled state, which could allow heralded generation of complex nonclassical atomic states by detecting a single photon [25].

A basic ingredient of all these proposals is longitudinally multimode cavity-atom coupling and multifrequency driving of the cavity, which was never before demonstrated for many modes in a cold-atom setup. We have now realized a platform exhibiting such features, demonstrating appreciable dispersive coupling between atoms and approximately 100 longitudinal cavity modes by measuring the transmission of an OFC through the cavity. Moreover, for the cavity mode nearest to the optical transition, we observe that the atomic saturation due to the presence of an external cooling laser driving the atoms leads to an optical bistability in the transmitted comb

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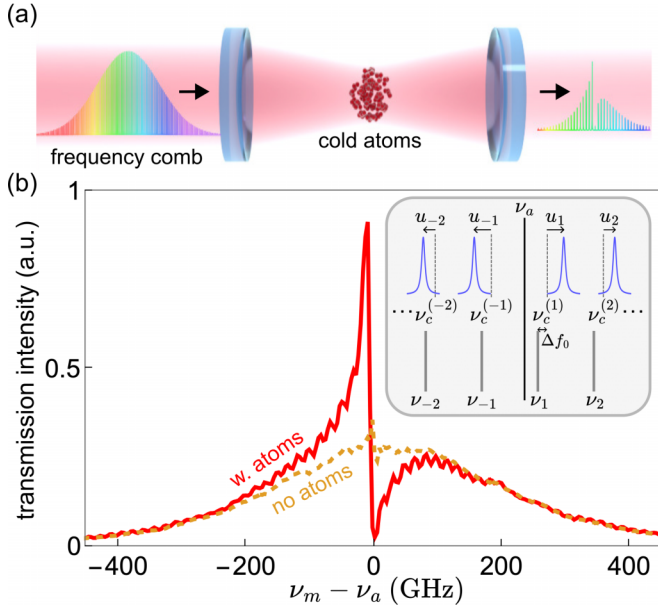


FIG. 1. Detecting collective light shifts of many longitudinal cavity modes induced by coupling to a cold-atom ensemble. (a) Illustration of the experimental setup. Cavity transmission of an optical frequency comb beam is measured using an optical spectrum analyzer. (b) Measurement results for an empty cavity (gold dashed line) and with atoms present (red solid line), under the condition of $\Delta f_0 = -200$ kHz. Inset: ν_m —comb line frequencies, $\nu_c^{(m)}$ —bare cavity mode frequencies, ν_a —atomic transition frequency, and u_m —collective cavity light shifts induced by coupling to a cold-atom ensemble, $\Delta f_0 = \nu_1 - \nu_c^{(1)}$.

line, which extends the OFC-driven cavity-atom interaction into the nonlinear regime.

II. CAVITY-TRANSMITTED COMB SPECTRA

In the first set of experiments, we detected the signatures of collective light shifts of many longitudinal cavity modes by measuring the transmission spectra of the OFC through the cavity-atom system. The basic principle of the experiment is schematically depicted in Fig. 1(a). An OFC, with each 24th comb line tuned near a longitudinal (i.e., TEM₀₀) resonance of an empty cavity, is coupled into a two-mirror resonator with cold atoms loaded at its center. We denote each 24th OFC line's frequency as ν_m [see inset of Fig. 1(b)], where, e.g., $m = 1$ ($m = -1$) is the index of the nearest blue (red) detuned line from the ^{87}Rb D2-line $F = 2 \rightarrow F' = 3$ atomic optical transition ν_a . Analogously, the empty cavity mode frequency near the corresponding OFC line is denoted as $\nu_c^{(m)}$, and its collective light shift (see below) is denoted as u_m . Another important parameter is the detuning $\Delta f_0 = \nu_1 - \nu_c^{(1)}$, which can be precisely tuned in our setup (see below).

The OFC radiation transmitted through the cavity is collected onto an optical spectrum analyzer (OSA). The transmission spectra of the OFC beam at fixed OFC-cavity detunings Δf_0 contain information about the resonances in the atom-cavity system. By comparing to the empty cavity transmission and the theoretical results, the influence of atom-induced collective light shifts on many longitudinal cavity

TABLE I. Experimental parameter values for the Fabry-Perot cavity (FSR—free spectral range, F —finesse, g_0 —cavity-atom coupling strength, and κ —decay rate) and the optical frequency comb (f_{rep} —repetition frequency, λ_{center} —central wavelength, and N_{modes} —number of modes).

	FSR (GHz)	F	$g_0/2\pi$ (kHz)	$\kappa/2\pi$ (kHz)
Cavity	1.93	1.2×10^4	140	150
	Bandwidth (THz)	f_{rep} (MHz)	λ_{center} (nm)	N_{modes}
OFC	~ 2.5	80.5	780	$\sim 3 \times 10^4$

modes can then be inferred using a single OFC transmission measurement. The number of cavity modes shifted due to the presence of the atomic ensemble can thus be used to estimate how many longitudinal modes significantly participate in the atom-cavity coupling, which is a quantity of interest with regard to the theoretical proposals of Refs. [21–25].

A. Experimental setup

We now describe our experimental setup, with the main parameters for the cavity and OFC given in Table I and more details given in Ref. [26]. The cavity with length $L = 7.76$ cm, mirror reflectivities $r = 99.98\%$, and dispersion $\epsilon = 18$ Hz (see the Supplemental Material for details [26]) is set up in the near-confocal configuration and stabilized via the Pound-Drever-Hall method. Every 24th OFC mode with frequency ν_m is matched to a cavity mode of frequency $\nu_c^{(m)}$, where the OFC is spatially matched to the TEM₀₀ cavity mode profile. The power per comb mode at the cavity input is ≈ 0.26 μW . A power enhancement factor of up to $G \sim 400$ is reachable in our setup, resulting in a cavity-enhanced peak intensity of 332 mW/cm² per comb, where the mode waist radius is $w_0 = 100$ μm . The OFC beam entering the cavity is circularly polarized. As the OFC frequency stabilization operates independently from the cavity stabilization, the OFC light can be freely switched on or off, and frequency detuned by a specified value Δf_0 relative to the cavity modes, using an acousto-optic modulator (AOM).

The light transmitted through the cavity was collected onto an OSA with frequency resolution of 7.5 GHz, such that individual transmitted OFC lines are not spectrally resolved. Note here that the bare cavity losses reduce the transmitted OFC power by a factor of ~ 8 with respect to the input. Also, the noise visible in the measured cavity transmission intensities in Figs. 1–3 was due to aliasing between the FSR and the resolution of the OSA.

The number of longitudinal cavity modes significantly shifted due to collective light shifts for $\Delta f_0 = -200$ kHz can be used to estimate the number of modes interacting appreciably strongly with the atomic ensemble, and was calculated from our measurements as follows. As shown in Fig. 1, the presence of atoms loaded into the center of the cavity induces frequency shifts of the cavity modes, resulting in a modification of the transmitted spectrum. Subtracting the transmission spectra measured with and without atoms reveals a differential signal that is nonzero in the frequency regions where mode shifts lead to changes in transmission [see Figs. 2(c) and 2(d)].

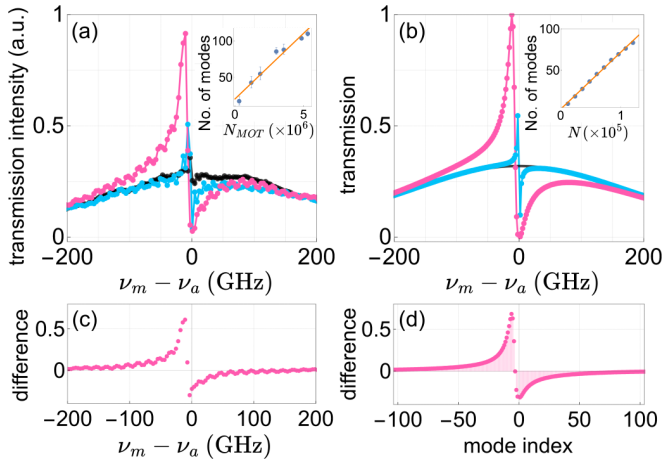


FIG. 2. Atom number dependence of the OFC-cavity transmission spectrum. (a) Experimental and (b) simulation data for empty cavity (black), partially loaded MOT (blue), and fully loaded MOT (magenta). Lines are guide to the eyes. Insets: Atom number scans of (a) experimental and (b) theoretical number of cavity modes significantly shifted due to the presence of atoms (see text). (c) Experimental and (d) simulated difference in the transmission value with respect to empty cavity for the highest number of atoms. Experimental parameters: MOT atom number $N_{\text{MOT}} = (0, 1.2, 5.3) \times 10^6$ for black, blue, and pink, $\Delta f_0 = -200$ kHz. Simulation parameters: FSR = 1.93 GHz, $g_0 = 2\pi \times 140$ kHz, $\Delta_a^{(1)} = 2\pi \times 495$ MHz, $\epsilon = 18$ Hz, mirror transmission coefficient $t = 0.0125$, $N = (0, 0.06, 1.2) \times 10^5$ for black, blue, and magenta, and $\Delta f_0 = -220$ kHz.

In our analysis, a threshold of 5% of the maximum transmitted signal in the presence of atoms is used to define a deviation of the differential signal from zero. The number of affected cavity modes is then obtained by dividing the corresponding frequency interval by the FSR.

The atoms are loaded into a magneto-optical trap (MOT) positioned to overlap with the center of the TEM₀₀ cavity mode. The MOT produces a cloud of $\approx 7 \times 10^6$ atoms at a temperature of around 70 μ K and a radius of ≈ 0.55 mm. As the repumper beam was turned on during these experiments, nearly all population was in the $F = 2$ ground state. Since the $F = 2 \rightarrow F' = 3$ transition is dominant in the ^{87}Rb D2 line, we here treat the cloud as a two-level medium.

B. Theoretical model

The atom-cavity behavior can be described using cQED [27]. The Hamiltonian modeling OFC-driven longitudinal cavity modes coupled to N static atoms placed at the cavity mode intensity maxima can be written as (see Ref. [26] for details of the derivations in this subsection):

$$\hat{H}(t) = - \sum_m \Delta_{c,m} \hat{a}_m^\dagger \hat{a}_m + g_0 \left[\sum_m \hat{a}_m e^{-i\delta_{p,m}t} \hat{\sigma}_{eg} + \text{H.c.} \right] + i \sum_m \eta_m (\hat{a}_m^\dagger - \hat{a}_m). \quad (1)$$

Here, m is the cavity mode index with the summation going over all relevant longitudinal modes, $\hat{a}_m$ is the cavity destruc-

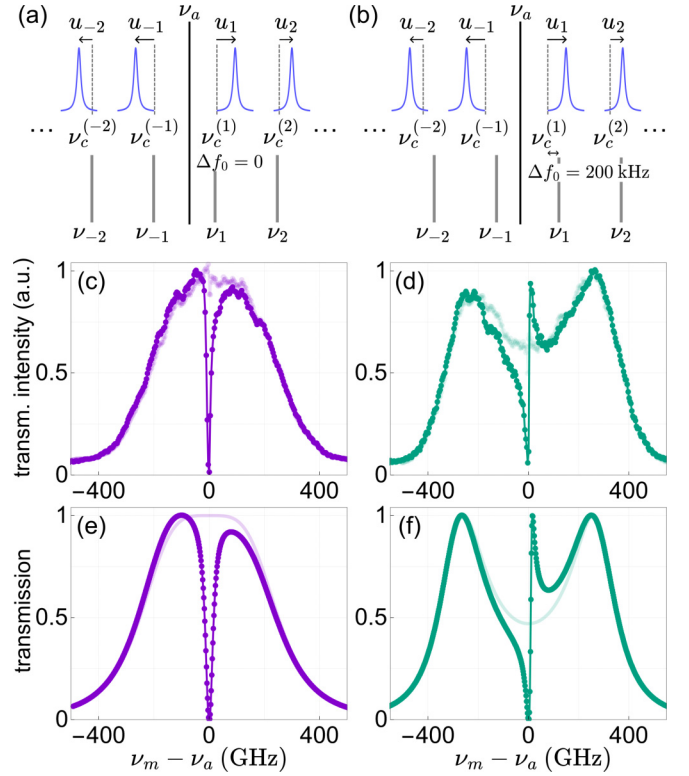


FIG. 3. Cavity transmission spectra for OFC tuned resonantly and to the blue of the empty cavity modes. Frequency schematics of the first several modes for (a) $\Delta f_0 = 0$ and (b) $\Delta f_0 = 200$ kHz. (c), (d) Experimental and (e), (f) simulation data for the resonant and blue-detuned cases, respectively. Thick lines—cavity with atoms loaded, semitransparent lines—no atoms in the cavity. Experimental parameters: See text. Simulation parameters: FSR = 1.93 GHz, $g_0 = 2\pi \times 140$ kHz, $\Delta_a^{(1)} = 2\pi \times 495$ MHz, $\epsilon = 18$ Hz, mirror transmission coefficient $t = 0.0125$, $N = 1.2 \times 10^5$, and $\Delta f_0 = 0, 160$ kHz for panels (e) and (f), respectively.

tion operator, $\eta_m \approx \eta$ is the driving rate (see below for the justification of the approximation), $\Delta_{c,m} = 2\pi \times (\nu_m - \nu_c^{(m)})$ is the cavity-comb line detuning, $\Delta_a^{(m)} = 2\pi \times (\nu_c^{(m)} - \nu_a)$ is the cavity-atom detuning, $\delta_{p,m} = 2\pi \times (\nu_m - \nu_a)$ is the comb line-atom detuning, $\hat{\sigma}_z = \hat{\sigma}_{ee} - \hat{\sigma}_{gg}$, $\hat{\sigma}_{ge}$ are the collective atomic optical inversion and coherence operators for the N atoms, and g_0 is the cavity-atom coupling strength.

The effective Hamiltonian of Eq. (1) in the dispersive limit can be calculated using the Schrieffer-Wolff transformation [28] while keeping the terms $\propto g_0^2/\Delta_a^{(m)}$, $g_0\eta/\Delta_a^{(m)}$ and neglecting the cavity-mediated dipole-dipole interaction [29] and cavity mode-mode scattering [30]. This Hamiltonian is given by

$$\begin{aligned} \hat{H}_{\text{eff}}(t) = & - \sum_m \Delta_{c,m} \hat{a}_m^\dagger \hat{a}_m + i \sum_m \eta (\hat{a}_m^\dagger - \hat{a}_m) \\ & - \sum_m \frac{g_0^2}{\Delta_a^{(m)}} \left(\hat{a}_m^\dagger \hat{a}_m + \frac{1}{2} \right) \hat{\sigma}_z \\ & + \sum_m \left(\frac{ig_0\eta}{\Delta_a^{(m)}} e^{i\delta_{p,m}t} \hat{\sigma}_{ge} + \text{H.c.} \right). \end{aligned} \quad (2)$$

The first two terms in $\hat{H}_{\text{eff}}(t)$ describe the cavity modes and OFC driving. The third term describes the collective light shifts of the cavity modes and the light shift of the atomic transition, arising due to atom-cavity coupling. The last term describes the effective optical driving of the atomic transition due to the cavity modes pumped by the OFC, similarly to free-space case [31–35].

Using $\hat{H}_{\text{eff}}(t)$ for calculating the steady-state mean-field cavity mode population $|\alpha_m|^2 = \langle \hat{a}_m^\dagger \hat{a}_m \rangle / N$ in the thermodynamic linear optics limit leads to

$$|\alpha_m|^2 = \frac{|\eta'|^2}{(\Delta_{c,m} - U_m)^2 + \frac{\kappa^2}{4}}, \quad (3)$$

where $\eta' = \eta/\sqrt{N}$ and κ is the cavity decay rate. The m th cavity mode thus experiences a collective light shift with respect to the empty cavity given by $U_m = 2\pi \times (v'_{c,m} - v_c^{(m)}) = 2\pi \times u_m = N g_0^2 / \Delta_a^{(m)}$. We note that although the shifts were here derived using cQED, their classical interpretation is the refractive index of the cold atomic medium changing the optical length of the resonator and altering the cavity resonance condition [26].

C. Experimental results

For an empty cavity [see gold dashed line in Fig. 1(b)], each of the narrow OFC lines with frequency near a cavity resonance follows the characteristic Lorentzian transmission curve of linewidth $\kappa/2$, while other OFC modes are reflected. An important quantity here is Δf_0 , defined by the difference of the first OFC mode from its nearest cavity mode; see inset of Fig. 1(b). At some fixed value of Δf_0 , the OFC transmission through the m th cavity resonance does not equal the transmission through the first cavity resonance, due to cavity dispersion ϵ caused by the mirror dispersion [26,36]. Instead, the detuning of the m th OFC line's frequency ν_m from its nearest empty cavity mode $\nu_c^{(m)}$ is given by $\nu_m - \nu_c^{(m)} = \Delta f_0 - |m|(|m| - 1)\epsilon/2$. This means that the transmitted signal will have a bandwidth significantly smaller than the input OFC. This justifies the approximation $\eta_m \approx \eta$ in Eq. (2), since the input OFC spectrum of bandwidth 2.5 THz varies negligibly on the cavity transmission spectrum of bandwidth ~ 400 GHz. It is worth noting that in our experiment the OFC spectrum can be frequency shifted by an AOM before being coupled into the cavity (see Ref. [26]). This enables precise matching of a selected comb mode to the corresponding cavity resonance, e.g., ν_1 to mode $\nu_c^{(1)}$, as well as controlled scanning of the comb mode around that resonance.

Deriving the Lorentzian profile (3) and shift u_m for each longitudinal mode with index m allows now to explain the spectral shape of the OFC transmission through the cavity, shown for a particularly notable case of $\Delta f_0 = -200$ kHz in Fig. 1(b). For cavity modes to the red of the atomic transition ($\Delta_a^{(m)} < 0$), the collective shift will reduce the cavity frequencies, and conversely increase them for the blue-detuned cavity modes; see inset of Fig. 1(b). For this Δf_0 , the small $|\Delta_a^{(m)}|$ modes with $\Delta_a^{(m)} < 0$ shift the cavity resonances closer to the OFC lines, resulting in a strong increase in transmission. In contrast, for the small $|\Delta_a^{(m)}|$ modes with $\Delta_a^{(m)} > 0$, the transmission is suppressed nearly to zero due to collective

light shifts moving the cavity modes away from OFC lines. As detailed below, this constitutes clear signatures of simultaneous dispersive cavity-atom interaction with many longitudinal cavity modes.

We now discuss the experimental results of cavity transmission spectrum measurements for $\Delta f_0 = -200, 0, 200$ kHz and give comparisons to the theoretical predictions for the total OFC transmission signal, calculated by summing over the transmission profiles of each cavity mode [see Eq. (S25) of Ref. [26]]. We note that all of the experimental data for different Δf_0 's have been normalized to the same maximum. For $N = 0$, the theoretical curves differ from the experimental ones near $\nu_m - \nu_a = 0$. This small central feature in the OFC transmission signal for a nominally empty cavity is a consequence of the residual background atomic vapor coupling to the cavity and shifting the cavity modes.

The $\Delta f_0 = -200$ kHz results for different N values are depicted in Fig. 2. When cold atoms are loaded into the cavity, a clear signature of the collective light shifts can be detected in the strong increase of cavity transmission for m values in the approximate range $-50 < m \leq -4$. For modes in the range $-4 < m < 45$, the collective light shift pushes the cavity modes away from the OFC modes, which results in a reduction of cavity transmission, reaching values near zero. For m 's outside of these two ranges, $|\Delta_a^{(m)}|$ increases and $|u_m|$ is reduced far below $\kappa/2\pi$, such that eventually the transmission spectrum resembles that of an empty cavity. The transmission curves are in good agreement with the theoretical calculations shown in Figs. 2(b) and 2(d). The number of cavity modes that are simultaneously shifted due to the cavity-atom interaction grow proportionally to the number of atoms loaded into the cavity waist; see insets of Figs. 2(a) and 2(b). From these data, we conclude that appreciable simultaneous collective light shifts of more than 100 cavity modes are achieved in the setup for the highest number of atoms in the MOT. Note that here the MOT number N_{MOT} and cavity atom number N differ by a factor of $\gtrsim 10$. This is a consequence of both the atomic motion inside the cavity modes reducing the effective atom-cavity coupling g_0 and the nonoptimal overlap of the cloud density distribution with radius 500 μm to the mode waist with radius $w_0 = 100$ μm .

The results for $\Delta f_0 = 0$ are given in Figs. 3(a), 3(c), and 3(e). For an empty cavity, the transmission peak here reaches a maximum value, theoretically equal to unity, near $\nu_m = \nu_a$. When atoms are loaded into the cavity, a transmission dip appears in the center of the spectrum, which is caused by the collective light shift pushing the cavity modes out of resonance with frequencies in the approximate range $-100 \text{ GHz} < \nu_m - \nu_a < 100 \text{ GHz}$. The slight asymmetry in the left and right transmission peaks is a consequence of the asymmetry in $\Delta_a^{(1)} = 2\pi \times 495 \text{ MHz} = 81.6\Gamma$ and $\Delta_a^{(-1)} = -2\pi \times 1437 \text{ MHz} = -236.6\Gamma$, which causes larger $|u_m|$ values to appear for modes with $m > 0$.

The transmission for $\Delta f_0 = 200$ kHz is shown in Figs. 3(b), 3(d), and 3(f). For an empty cavity, the transmission is small at frequencies near the center and there are two symmetric peaks at values $\nu_m - \nu_a \approx \pm 260 \text{ GHz}$, where the cavity dispersion results in resonances for the OFC. A slight asymmetry in the peak heights in the experimental results is

caused by a mismatch between the position of the center of the OFC emission spectrum and ν_a . When atoms are loaded into the cavity, the cavity modes near the center shift toward higher frequencies for positive m , and toward lower frequencies for negative m values. This causes a transmission increase for OFC modes with $m > 0$ and a decrease for $m < 0$, resulting in a curve with an inverted shape to the case of $\Delta f_0 = -200$ kHz for small $|m|$ values. The theory reproduces the experimental data reasonably well, with the main discrepancies stemming from uncertainties in estimating experimental parameters such as the detuning of the relative OFC-cavity detuning Δf_0 , cavity dispersion ϵ and reflectivity r , and cavity linewidth κ .

III. RESOLVING INDIVIDUAL CAVITY RESONANCES

A. Experimental setup

In the second set of experiments, we directly measured the transmission curves of cavity modes with $m = \pm 1, \pm 2$ in the single comb line coupling regime, where only one OFC mode is coupled to the cavity at a time; see Ref. [26] for details. The very narrow linewidths of individual comb lines (~ 10 kHz) allow for transmission measurements that can resolve the shape of individual cavity modes.

To achieve the regime of effectively single comb line cavity coupling, the empty cavity resonances were shifted by u_m via loading the MOT, after which $\nu_m - \nu_c^{(m)}$ was scanned to find the OFC-cavity resonance conditions for each $m = \pm 1, \pm 2$ mode. Since u_m for such small m values exceeds $\kappa/2\pi$, shifting the OFC-cavity detuning $\nu_m - \nu_c^{(m)}$ by u_m tunes all other comb modes away from their corresponding cavity resonances; i.e., only a single comb mode is resonant with a cavity mode at a time, excluding the far-detuned background modes with very large m . The single comb coupling condition was experimentally verified by measuring the cavity transmission spectrum using an OSA, which consisted of a narrow spectral feature limited by the resolution of the OSA, along with a wide background for large $|\nu_m - \nu_a|$ values. Due to this background signal, the OFC-cavity transmission cannot be detected using the standard methods, and it was necessary to implement a heterodyne detection technique, similar to that in Ref. [37], and described in detail in Ref. [26], which enabled measurements of the cavity-transmitted electric field of a single m th comb line at different values of $\nu_m - \nu_c^{(m)}$.

B. Experimental results

The measurement results for the single comb mode coupling regime are given in Fig. 4(a). Just as in the previous section, the presence of atoms shifts the central frequencies of the cavity modes by the collective light shifts given by $u_m = N g_0^2 / (2\pi \Delta_a^{(m)})$. For modes with $m = -1, \pm 2$, the cavity resonance positions follow this analytic prediction derived from the linear optics approximation in Sec. II B. The transmission curves of the $m = \pm 2$ modes (and higher $|m|$ modes) follow a Lorentzian shape (3), also measured for an empty cavity. Note that the empty cavity linewidth is measured to be $\kappa/2\pi = 240$ kHz, which differs from the value of $\kappa/2\pi = 150$ kHz measured by cavity ringdown (see Table I), where the broadening is here induced by the cavity length stabilization. In contrast, for the $m = 1$ mode, and to a lesser degree

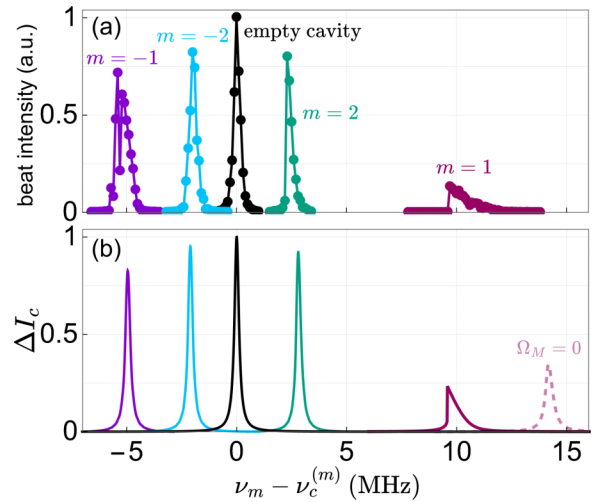


FIG. 4. Cavity transmission of individual comb lines in the single comb mode coupling regime. (a) Experimental measurements of transmission curves for modes with $m = 1$ (red), $m = -1$ (purple), $m = 2$ (green), $m = -2$ (blue), and for an empty cavity mode (black). (b) Simulated difference in mean cavity photon number when the comb beam is turned on and off (see text), normalized to the empty cavity value. For comparison, the curve for the $m = 1$ mode with $\Omega_M = 0$ is shown. Experimental parameters: See text. Simulation parameters: $N = 3.7 \times 10^5$, $\kappa = 2\pi \times 0.24$ MHz, $g_0 = 2\pi \times 140$ kHz, $\Omega_M = 2\pi \times 1.7$ MHz, $\eta/\sqrt{N} = 2\pi \times 0.06$ MHz, $\Gamma = 2\pi \times 6.066$ MHz, $(\Delta_a^{(1)}, \Delta_a^{(-1)}, \Delta_a^{(2)}, \Delta_a^{(-2)}) = 2\pi \times (495, -1437, 2427, -3369)$ MHz, and $\Delta_M^{(m)} = \Delta_a^{(m)} + 2\Gamma$.

the $m = -1$ mode, there is a deviation from the Lorentzian cavity resonance shape. Particularly, the shape of the $m = 1$ resonance is characteristic of bistable nonlinear cavities, for which multiple transmission values coexist at the same input frequency and intensity [38,39]. Note that this bistable transmission for $m = 1$ should be present also in the results of Figs. 1–3. However, a single mode influences the entire optical spectrum of the cavity-transmitted OFC beam sufficiently little that it can be neglected on the larger scale, such that the linear model reproduces the data well.

Describing the transmission shape of the $m = 1$ cavity mode requires thus a treatment beyond the linear optics regime. The peak cavity intensity per comb mode is ~ 330 mW/cm², which results in a detuned saturation parameter of $s \sim 0.005$ for the far-detuned ν_1 comb line with $I_s = 2.5$ mW/cm², such that the population in the excited state is $\sigma_{ee} \approx s/2 = 0.0025$. The bistable transmission curve in Fig. 4(a) should thus not be a consequence of saturating the two-level transition by only the ν_1 comb line. Magnetic sublevel pumping nonlinearities have been known to cause bistable transmission at low powers for a cavity coupled to a cold-atom cloud [40]. However, the ^{87}Rb D2-line $F = 2 \rightarrow F' = 3$ transition has a self-defocusing magnetic pumping nonlinearity for blue detuning [41], which would result in a cavity transmission curve with reversed shape compared to the one in Fig. 4(a) for the $m = 1$ mode. Additionally, reducing the repumper beam intensity did not change the shape of the $m = 1$ transmission curve in our experiments, provided that the MOT atom number did not decay. This indicates that

the optical nonlinearity is not a consequence of pumping the atoms into the $F = 1$ state.

We identify the main cause of the nonlinearity to be the presence of MOT cooling beams driving the $F = 2 \rightarrow F' = 3$ transition, kept on during the measurements. This interpretation is corroborated by additional transient transmission measurements for a single comb line at a fixed frequency corresponding to the peak of the $\nu_c^{(1)}$ cavity mode transmission [see Fig. 4(a)]. In this experiment, the MOT laser beams were switched on and off using an AOM, and the resulting time dependence of the cavity transmission was recorded. We observed that the transmitted signal at the selected frequency appears only when the MOT beams are on. When the MOT beams are turned off, the cavity transmission rapidly vanishes, caused by a shift of the cavity mode to a frequency different from the one being probed [see also dashed line in Fig. 4(b)]. The vanishing of cavity transmission occurs on timescales $\ll 1$ ms, such that the density of the cold-atom cloud does not change appreciably. These preliminary measurements thus indicate that the cooling beam helps drive the saturation of the two-level transition and cause the bistable transmission curve, as corroborated by simulation results below. The measured cavity bistability will be explored in more detail in a subsequent publication.

C. Numerical simulations

As the cooling beams were kept on during the measurements, we here model the system using a cavity-atom interaction Hamiltonian of the form $\hat{H}_{\text{int}}(t) = (g_0\hat{a} + \Omega_M e^{i\Delta_M t})\hat{\sigma}_{eg} + \text{H.c.}$, where $\hat{\sigma}_{eg}$ is the collective atomic “spin” raising operator and $\hat{a}$ is the cavity mode annihilation operator of the single cavity mode that is excited [26]. In the thermodynamic mean-field limit, where $\langle \hat{O}_1 \hat{O}_2 \rangle \rightarrow \langle \hat{O}_1 \rangle \langle \hat{O}_2 \rangle$ and $\langle \hat{a} \rangle \rightarrow \sqrt{N}\alpha$, $\langle \hat{\sigma}_{ij} \rangle \rightarrow N\sigma_{ij}$, the equations of motion are given by

$$\begin{aligned} \frac{\partial \alpha}{\partial t} &= \left(i\Delta_c - \frac{\kappa}{2}\right)\alpha - ig_N\sigma_{ge} + \frac{\eta}{\sqrt{N}}, \\ \frac{\partial \sigma_{ee}}{\partial t} &= -\Gamma\sigma_{ee} + [i(g_N\alpha^* + \Omega_M^* e^{-i\Delta_M t})\sigma_{ge} + \text{H.c.}], \\ \frac{\partial \sigma_{eg}}{\partial t} &= -\left(i\Delta_a + \frac{\Gamma}{2}\right)\sigma_{eg} + i(g_N\alpha^* + \Omega_M^* e^{-i\Delta_M t})(1 - 2\sigma_{ee}), \end{aligned} \quad (4)$$

where $g_N = \sqrt{N}g_0$. Here, a single comb line is driving the cavity mode while a second laser, detuned from the comb line by Δ_M , is driving the atomic two-level transition directly with Rabi frequency Ω_M . The above equations are solved numerically, and the quantity ΔI_c , defined as the temporally averaged steady-state difference between the cavity mode populations $|\alpha|^2 = \langle \hat{a}^\dagger \hat{a} \rangle / N$ when the comb beam is turned on and off, is plotted in Fig. 4(b). For $m = \pm 2$, the simulations reproduce the cavity mode shifts and shapes of the experiment, while for $m = -1$ the measured transmission line is broadened with respect to the simulated curve. The reason for this discrepancy is currently under investigation.

For the $m = 1$ mode, the cavity photon number has a notably different shape and resonance position when turning on the cooling beam. At $\Omega_M = 0$ (dashed curve), the cavity

resonance has a Lorentzian shape. Increasing the value of Ω_M (solid curve) to $\Omega_M = 2\pi \times 1.7$ MHz, the simulations reproduce the shape and position of the experimental transmission curve well. Indeed, in simulations with $\Omega_M = 0$, the maximum σ_{ee} reaches the experimentally estimated value of 0.0025 (see above), while for the finite Ω_M the value is ~ 0.2 , which indicates the presence of the nonlinear regime in the two-level atomic response, as also experimentally expected from the cooling beam intensities (see below). We note here that the only free parameter used in the $m = 1$ simulations is Ω_M , while κ , N , and η values were determined from the empty cavity transmission values, the $m = -1, \pm 2$ transmission values, and the experimental saturation parameter, respectively, with g_0 , $\Delta_a^{(m)}$, Γ taken directly from experiment.

Noting that the Rabi frequency of the MOT laser is given by $\Omega_M = \Gamma\sqrt{I_M/2I_s}$ [42], for saturation intensity $I_s = 2.5$ mW/cm² the intensity for $\Omega_M = 2\pi \times 1.7$ MHz is 0.4 mW/cm². This value is smaller than the total experimental cooling beam intensity of 20 mW/cm², which was taken from power measurements after an optical fiber. These measurements overestimate the total MOT beam intensity at cavity center as they did not include effects such as losses at optical elements and vacuum chamber windows, along with the absorption of the cooling beam within the cold cloud.

IV. CONCLUSION

We have experimentally demonstrated the interaction between cold atoms and ~ 100 longitudinal cavity modes by measuring the signatures of collective light shifts in the transmission of an incoming OFC beam. Additionally, we have measured a bistable transmission for the cavity mode nearest to the atomic transition, demonstrating nonlinear interaction due to combined coupling of the atoms to an external pump laser and the OFC-driven cavity.

By combining three experimental platforms hitherto never used together in the same setup, (1) a cold-atom ensemble coupled to (2) an optical resonator with (3) ultrafast laser driving, our experiment has the potential to establish a promising avenue for studying light-matter interaction. The advantage offered by each platform, narrow atomic transition linewidths, interaction enhancement via the cavity, and multifrequency interfacing via the OFC, could enable a host of developments in the field, as envisioned by recent theoretical proposals toward utilizing this system for studying multifrequency cavity QED [21–23,25].

The experiment can readily be extended into several intriguing directions. For instance, at mirror distances tuned close to the confocal regime, transverse cavity modes with frequency separations on ~ 10 MHz scales can be achieved [16]. Using an OFC pump with tailored f_{rep} [43] to drive the atoms in such a configuration could lead to additional flexibility in designing complex effective cavity-mediated interaction Hamiltonians. Moreover, effects such as mode-mode scattering for nearly degenerate transverse cavity modes could be extended to the regime of OFC driving, potentially enabling multiplexed information processing at many cavity frequencies [30]. Also, utilizing two-photon atomic transitions in the multifrequency cavity context could lead to other interesting effects in analogy with the free-space configuration [34].

Regarding intensity squeezing of light with a nonlinear medium in a cavity [44], using OFC driving with very high power per comb mode could here lead to frequency multimode squeezed light interfaced with atoms in the cavity—currently, a highly sought-after goal in quantum photonics [45,46].

Finally, we note that at present there is a great need for experimental techniques for manipulating atoms with transitions in the vacuum- and extreme-ultraviolet regions [47], where only pulsed radiation sources are available [48,49], along with molecules with complex transition level structures, where frequency combs could address multiple degrees of freedom with a single laser [24]. Exploration of cavity-cold atom interaction with OFC driving is a step toward developing experimental methods applicable in such situations.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [50].

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