Cavity Nonlinear Optics at Low Photon Numbers from Collective Atomic Motion

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(Received 7 June 2007; published 20 November 2007)

We report on Kerr nonlinearity and dispersive optical bistability of a Fabry-Perot optical resonator due to the displacement of ultracold atoms trapped within. In the driven resonator, such collective motion is induced by optical forces acting upon up to 10^5 ^{87}Rb atoms prepared in the lowest band of a one-dimensional intracavity optical lattice. The longevity of atomic motional coherence allows for strongly nonlinear optics at extremely low cavity photon numbers, as demonstrated by the observation of both branches of optical bistability at photon numbers below unity.

DOI: 10.1103/PhysRevLett.99.213601 PACS numbers: 42.50.Pq, 32.80.Pj, 42.50.Vk, 42.65.−k

Nonlinear optical phenomena occur typically at high optical intensities, or, equivalently, at high average photon numbers $\bar{n}$ in an optical resonator. Producing such phenomena at very low photon numbers, i.e., $\bar{n} \leq 1$, is desirable for applications ranging from optical communication to quantum computation [1,2]. In this low-intensity regime, nonlinear optics requires the use of materials with optical properties that are significantly altered even by single photons, and in which this alteration persists long enough to impact the behavior of subsequent photons interacting with the material.

Such requirements may be satisfied by atoms within a high-finesse, small-volume optical resonator. Under the condition of collective strong coupling, defined as $NC = N(g^2/2\kappa\Gamma) \gg 1$, atomic saturation on an optical transition induces nonlinear effects such as absorptive optical bistability [3–5] with $N \gg 1$, or cross phase modulation [6] and photon blockade [7] with $N \simeq 1$. Here, N is the number of atoms in the resonator, g the atom-cavity coupling frequency, and κ and Γ the cavity and the atomic coherence decay rates, respectively. Since strong nonlinearities (e.g., the upper branch of the optical bistability curve) occur beginning at $\bar{n}_{\rm nl} \simeq N(\Gamma/\kappa)$, the condition $\bar{n}_{\rm nl} \leq 1$ requires that the atomic coherence persist longer than the residence time of photons in the cavity. This condition is met with neutral atoms in state-of-the-art Fabry-Perot cavities [6,7] and with superconducting circuits within microwave resonators [8]. Strong nonlinearities are also achieved using the long coherence times for atomic Raman transitions [9–11]. The lowest reported resonator photon number for observing both branches of optical bistability is $\bar{n}_{\rm nl} \simeq 100$ [3,5].

The motional degrees of freedom of ultracold atomic gases represent a new source of long-lived coherence affecting light-atom interactions. This coherence leads, for example, to superradiant light scattering at low threshold intensities both in free space [12,13] and inside optical resonators [14]. The motion of single atoms in strong-coupling cavities has been observed and cooled [15,16], and the motion of atomic ensembles in weak-coupling cavities has been examined [14,17–20].

We report here on nonlinear optics arising from the long-lived coherent motion of ultracold atoms trapped within a high-finesse Fabry-Perot cavity. Optical forces exerted by light within the cavity displace the trapped atoms so as to significantly vary their position-dependent coupling to the cavity mode. The consequent refractive nonlinearity and bistability are observed in the transmission of probe light through the cavity. Because the coherence time (1 ms) for atomic motion is much longer than the residence time of photons in the cavity ($1/2\kappa = 120$ ns), strong optical nonlinearity is observed even at $\bar{n} = 0.05$ and is predicted to occur as low as $\bar{n} \simeq 10^{-4}$.

To explain this nonlinear optical behavior, we consider the one-dimensional motion of N atoms in a Fabry-Perot cavity in which the atom-cavity coupling frequency varies along the cavity axis as $g(z) = g_0 \sin k_p z$. A harmonic potential of the form $V(z) = m\omega_z^2(z - z_0)^2/2$ confines the atoms. For a trapping frequency $\omega_z \gg \hbar k_p^2/2m$, the position of atoms in the trap's ground state is determined to below the optical wavelength $2\pi/k_p$. In the dispersive regime, with the detuning $\Delta_{ca} = \omega_c - \omega_a$ between the bare-cavity and atomic resonances being large ($|\Delta_{ca}| \gg \sqrt{N}g_0$), the atomic ensemble presents a refractive medium that shifts the cavity resonance by $\Delta_N \simeq Ng(z_0)^2/\Delta_{ca}$.

Probe light in the cavity produces an additional, optical force on the atoms with average strength $f(z_0)\bar{n}$ where $f(z) = -\hbar\partial_z g^2(z)/\Delta_{ca}$. This force displaces the equilibrium position of the atoms by $(f(z_0)/m\omega_z^2)\bar{n}$. In turn, this displacement (assumed small) varies the atom-cavity coupling strength [Fig. 1(a)]. Taking $k_p z_0 = \pi/4$ for illustrative purposes, the cavity resonance shift changes to $\Delta_N(1 - \epsilon\bar{n})$ with $\epsilon = 2\hbar k_p^2 g_0^2/m\Delta_{ca}\omega_z^2$. Thus, this system is characterized by a Kerr nonlinearity, with the refractive index of the intracavity medium, $1 + (\Delta_N/\omega_p)(1 - \epsilon\bar{n}) = n_0 + n_2 I_p$, varying with the probe intensity $I_p \propto \bar{n}$.

This nonlinearity affects the frequency response of the driven cavity. Probing the cavity transmission with light detuned by Δ_{pc} from the bare-cavity resonance, and assuming that the atoms adiabatically follow the trap mini-

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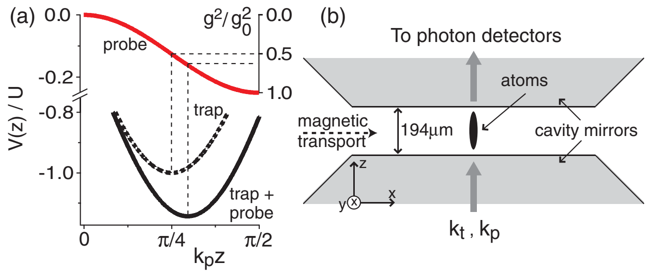

FIG. 1 (color). (a) The optical potentials due to the cavity probe light (red, here with maximum depth $U/4$ and $\Delta_{ca} < 0$) or the trapping light (black dashed line, with maximum depth U) are shown at one lattice site. Atoms in the combined optical potential (black, solid line) are displaced in the presence of the probe, changing the atom-cavity coupling strength [$\propto g(z)^2$], and, thereby, the cavity resonance frequency. (b) Experimental setup: ultracold ^{87}Rb atoms are trapped in an optical lattice formed by exciting a TEM$_{00}$ mode of a vertically oriented Fabry-Perot cavity with light at $k_t = 2\pi/(850\ \text{nm})$. The system is probed by detecting transmission through another TEM$_{00}$ mode at $k_p = 2\pi/(780\ \text{nm})$.

mum, the intracavity photon number $\bar{n}(\Delta_{pc})$ is given as

$$\bar{n}(\Delta_{pc}) = \frac{\bar{n}_{\max}}{1 + (\frac{\Delta_{pc} - \Delta_N(1-\epsilon\bar{n})}{\kappa})^2}, \tag{1}$$

where $\bar{n}_{\max}$ defines the probe intensity. This line shape is equivalent to that obtained for a damped nonlinear mechanical oscillator [21]. The deviation from the Lorentzian line shape of the bare cavity may be quantified by the parameter β that gives the maximum nonlinear shift of the cavity resonance in units of the cavity half-linewidth. Here $\beta = (\Delta_N \epsilon/\kappa)\bar{n}_{\max}$. In particular, for $\beta > 8\sqrt{3}/9 \simeq 1.54$, the cavity exhibits refractive bistability [22].

In our experiment [Fig. 1(b)], atoms were trapped not in a single harmonic trap, but rather in a one-dimensional optical lattice comprising many potential wells. This optical trap was formed by coupling frequency-stabilized laser light with a wave number of $k_t = 2\pi/(850\ \text{nm})$ to a TEM$_{00}$ mode of a Fabry-Perot cavity that was actively stabilized with this same light. The trap was loaded with an ultracold ^{87}Rb gas in the $|F = 1, m_F = -1\rangle$ hyperfine ground state that was transported magnetically to within the optical cavity prior to being transferred to the optical trap. Evaporative cooling in the optical trap yielded a gas of up to 10^5 atoms occupying $\simeq 300$ adjacent lattice sites and with a temperature of $T = 0.8\ \mu$K. We note that this temperature is just above the critical temperature $T_c \simeq 0.5\ \mu$K for Bose-Einstein condensation given our experimental parameters. The trap depth was set to $U \simeq k_B \times 6.6\ \mu$K, corresponding to axial and radial trapping frequencies of $\{\omega_z, \omega_{x,y}\} \simeq 2\pi \times \{42, 0.3\}$ kHz, about each minimum of the optical trap. Given $\hbar\omega_z/k_B = 2\ \mu\text{K} > T$, the gas resided predominantly in the ground state for axial vibration in each lattice site.

According to the specific value of k_t, another TEM$_{00}$ mode of the cavity was stabilized with $|\Delta_{ca}| = 2\pi \times (10\text{–}300)$ GHz detuning from the D2 resonance line at a wave number of $k_p = 2\pi/(780\ \text{nm})$. The cavity finesse for this mode was measured as $\mathcal{F} = 5.8 \times 10^5$, corresponding to a resonance half-linewidth of $\kappa = 2\pi \times 0.66$ MHz given the 194 μm spacing between the cavity mirrors. From the cavity mode waist of $w_0 = 23.4\ \mu$m and summing over all states excited by the σ^+ polarized probe light, we determine an atom-cavity coupling frequency of $g_0 = 2\pi \times 14.4$ MHz at the antinodes of the cavity field. Given the atomic half-linewidth of $\Gamma = 2\pi \times 3$ MHz, our cavity achieves single-atom strong-coupling conditions, with critical atom ($2\Gamma\kappa/g_0^2 = 0.02$) and photon ($\Gamma^2/2g_0^2 = 0.02$) numbers below unity.

After preparing the intracavity atomic medium, its nonlinear optical response was probed by detecting the transmission of light at wave number k_p through the cavity. For this measurement, the probe light was switched on and its frequency swept linearly across the cavity resonance at a rate of a few MHz/ms. The sweep rate was sufficiently slow so that trapped atoms adiabatically followed the varying intracavity optical potential, and sufficiently fast so that atom loss from probe-induced heating [23] was negligible. The transmitted probe light was separated from the trapping light using several filters, and then detected by single-photon counters. The overall efficiency of detecting an intracavity photon was $\simeq 0.05$.

As the power of the probe light was increased, the observed cavity transmission line shapes increasingly deviated from the symmetric line shapes observed for the bare cavity and at low powers [Fig. 2(a)]. This behavior can be described by adapting the model developed above to the experimental case with atoms trapped in a multitude of potential wells. Because of the difference in wavelengths of the trapping and the probing light, atoms in each well of the optical trap experience a different strength and gradient of the probe-induced ac Stark shift. Nevertheless, at all potential wells, this Stark shift displaces all atoms so as to either increase (for $\Delta_{ca} < 0$) or decrease ($\Delta_{ca} > 0$) their coupling to the cavity. That is, the regular crystalline arrangement of atoms in the confining optical lattice is distorted by probe light, somewhat analogously to the mechanism for Kerr effects in solids. Equation (1) remains valid for our experiment with ϵ halved to account for averaging over the many wells. The contribution of radial motion to this effect is negligible.

To compare these measurements with theoretical predictions, we also account for technical fluctuations in the probe detuning Δ_{pc}. The bare-cavity transmission line shape (Fig. 2, inset) is well approximated by the convolution of a Lorentzian with half-linewidth κ and a Gaussian with rms width of $\sigma = 2\pi \times 1.1$ MHz. Replacing the

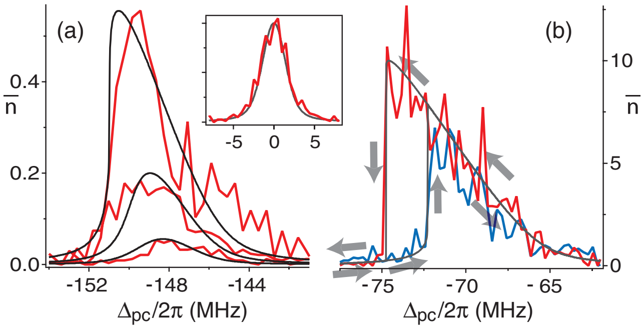

FIG. 2 (color). Nonlinear and bistable cavity line shapes shown as the intracavity photon number $\bar{n}$ vs detuning Δ_{pc} from the bare cavity at constant input power. (a) The cavity line shape [red, single (highest $\bar{n}_{\max}$) or average of 5 measurements] becomes increasingly asymmetric as the input intensity is increased. The triggering technique [see Fig. 3(a)] used to obtain these spectra resulted in variations of up to 1 MHz in $\Delta_N/2\pi$ between measurements. Model line shapes (black lines) using the Voigt profile indicated by the bare-cavity line shape (inset) are calculated with $\beta = \{0.37, 1.3, 3.7\}$ determined from the observed $\bar{n}_{\max}$, a common $\Delta_N = -2\pi \times 148$ MHz, and the known $\Delta_{ca} = -2\pi \times 30$ GHz, and $\omega_z = 2\pi \times 42$ kHz. (b) The lower (blue line) and upper (red line) branches of optical bistability are observed in a single experiment with probe light swept with opposite chirps (± 6 MHz/ms) across the cavity resonance, and match the expected behavior (black line) with $\beta = 9.5$, $\Delta_{ca} = -2\pi \times 101$ GHz, and $\omega_z = 2\pi \times 42$ kHz [29].

Lorentzian of Eq. (1) with this Voigt profile, and using values of β determined from measured experimental parameters, we obtain excellent agreement with the transmission measurements [Fig. 2(a)]. The contribution of internal-state dynamics alone to the observed nonlinear optical behavior is negligible [24].

At sufficiently strong refractive nonlinearity, the optical resonator becomes hysteretic and bistable. To observe clearly this bistability, we compared the cavity transmission for consecutive sweeps of the probe frequency with opposite chirp, i.e., sweeping linearly towards and then away from the bare-cavity resonance. For this measurement, the cavity was operated at $\Delta_{ca} = -2\pi \times 101$ GHz and contained $N \simeq 7 \times 10^4$ atoms. For $\bar{n}_{\max} = 10$, the parameter $\beta = 9.5$ is well within the bistable regime, which, for the Voigt profile relevant to our system, occurs for $\beta \gtrsim 3.7$. The observed line shapes [Fig. 2(b)] exhibit several hallmarks of bistability, such as the abrupt changes in $\bar{n}$ at the termini of the upper and lower branches of the bistability curve and the difference in the maximum $\bar{n}$ observed for the upward and downward frequency sweeps. These features were also observed in similar experiments with $\Delta_{ca} = -2\pi \times 10$ GHz and at the lowest intensities detectable in our system, with $\bar{n} = 0.05$.

In previous studies of bistability in strongly coupled atom-cavity systems [3–5], the operating range over which hysteretic behavior was observed was narrower than predicted theoretically. In contrast with those studies, here the cavity state is determined by atomic motion that varies only at a time scale of ω_z^{-1}. As this time scale is longer than both the $1/\kappa$ time scale for fluctuations of the intracavity field and also the typical ~ 10 μs time scale of variations in Δ_{pc}, such fluctuations do not destabilize the atoms-cavity system, and the entire range of bistability is observed.

As the detuning Δ_{ca} is reduced, the minimum photon number $\bar{n}_{\rm nl}$ for strong nonlinearities

$$\bar{n}_{\rm nl} = 4 \frac{\omega_z^2 \kappa}{\omega_{\rm rec} g_0^2} \frac{N g_0^2/2 + (\Delta_{ca}/2)^2}{N g_0^2}, \tag{2}$$

as defined by the condition $\beta = 1$, diminishes. Here, $\omega_{\rm rec} = \hbar k_p^2/2m$ is the probe recoil frequency and the effects of spatial averaging are included. For $\Delta_{ca} \to 0$, this number reaches a limiting value of $\bar{n}_{\rm nl} \simeq 10^{-4}$ for a minimum value for the trapping frequency of $\omega_z = 2\omega_{\rm rec}$ and other parameters of our experiment.

For $\bar{n}_{\rm nl} < \omega_z/2\kappa \simeq 0.03$, reached at $|\Delta_{ca}| \lesssim 2\pi \times 15$ GHz for our experimental conditions, the optical nonlinearity stems from the passage through the cavity of less than one photon per trap oscillation period. Photon number fluctuations may then cause a significant nonadiabatic atomic motional response. Taking the characteristic time for these fluctuations to be $1/2\kappa$, a single photon imparts an impulse $f(z)/2\kappa$ to an atom at location z. This impulse induces transient motional oscillations in the atomic medium that modulate the cavity resonance frequency. For a detuning Δ_{ca} at which $\bar{n}_{\rm nl} < \omega_z/2\kappa$, this frequency modulation becomes greater than the cavity half-linewidth and, thus, one may expect optical nonlinearities to manifest as strong temporal correlations imposed on photons transmitted through the cavity due to the light-induced atomic vibrations.

Transient oscillations may also be excited by diabatically varying the probe-induced optical force. We observed such oscillations as follows. We first loaded $N > 5 \times 10^4$ atoms into the cavity giving $|\Delta_N| > 2\pi \times 20$ MHz for the atom-cavity detuning of $\Delta_{ca} = -2\pi \times 260$ GHz, and then monitored the transmission of probe light at $\Delta_{pc} = -2\pi \times 17$ MHz. As atoms were lost from the trap, the atom number N diminished until the cavity resonance approached the probe frequency [Fig. 3(a)]. Once the cavity transmission exceeded a threshold value, establishing the value of $\Delta_N = -2\pi \times 19$ MHz, the probe was switched off for 10 ms and then turned back on within 5 μs at a high level of $\bar{n} = 6.5$. The potential minima within the combined intracavity optical potential were thus suddenly displaced, setting trapped atoms aquiver and causing the cavity resonance to be modulated over $2\beta = 0.75$ cavity half-linewidths at the atom trapping frequency of $2\pi \times 49$ kHz. The corresponding peak-to-peak oscillation amplitude for the atoms trapped where $k_p z_0 = \pi/4$ [Fig. 1(a)] is just 3 nm. For the Voigt profile relevant to our system, the modulation of the cavity reso-

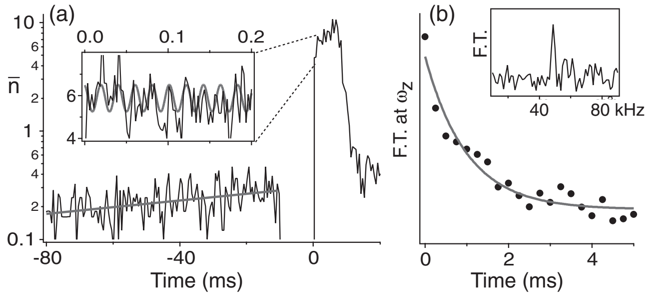

FIG. 3. Observation of collective atomic motion. (a) A record of the cavity transmission demonstrates the experimental sequence of a trigger, delay, and detection phase. Inset: Coherent atomic motion modulates the cavity transmission at $\omega_z = 2\pi \times$ 49 kHz (shown is the first 200 μs of the detection phase, 50-measurement average, 400 kHz bandwidth) at the expected amplitude and phase (gray curve, for $2\beta = 0.75$). (b) The Fourier spectral amplitude at ω_z, determined over 500 μs intervals of the transmission measurement (50-measurement average), decays within a $1/e$ time of 1.0(1) ms. Inset: Typical Fourier amplitude spectrum of the first 500 μs of the detection phase for a single measurement.

nance frequency varied the average cavity photon number by 1.3. This variation was visible in the temporal Fourier spectrum of the cavity transmission signal from single experimental runs [Fig. 3(b), inset]. By summing the transmission signals over 50 repeated measurements, a clear temporal variation of the cavity transmission, representing a record of the collective motion of atoms within the cavity, was obtained. The amplitude and phase of this motion agreed with the predictions discussed above. The observed motion decayed within $1/\Gamma_{\mathrm{osc}} = 1$ ms, a value consistent with the inhomogeneous broadening of ω_z due to the finite radial extent of the trapped gas. This decay rate for collective atomic motion imposes a technical limit of $\bar{n} = \Gamma_{\mathrm{osc}}/2\kappa \simeq 10^{-4}$ on the lowest light level at which motion-induced nonlinearities may be exploited in our system.

The present work highlights novel capabilities enabled by the long-lived coherence of atomic motion inside an optical cavity. Specifically, we observe strongly nonlinear optics, such as cavity bistability, occurring at extremely low light levels. The role of such nonlinearities in producing nonclassical and correlated quantum states of light, such as achieved by saturation in single-atom cavity quantum electrodynamics [6,7,25], warrants further investigation. Further, the influence of collective atomic motion over cavity properties may allow for quantum-limited measurement of that motion and for studies of quantum feedback [26–28].

We thank T. Purdy and S. Schmid for assistance in the early stages of this experiment and H. J. Kimble for helpful comments. This work was supported by AFOSR, DARPA, and the David and Lucile Packard Foundation.

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