Cavity Nonlinear Optics at Low Photon Numbers from Collective Atomic Motion

Subhadeep Gupta[1], Kevin L. Moore[1], Kater W. Murch[1], and Dan M. Stamper-Kurn[1,2]
[1]*Department of Physics, University of California, Berkeley CA 94720*
[2]*Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720*
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We report on nonlinear optical phenomena caused by the collective motion of ultracold atoms within a Fabry-Perot cavity in a regime of single-atom strong coupling. Up to 10^5 rubidium atoms are trapped in the lowest band of a one-dimensional intracavity optical lattice. We observe large Kerr nonlinearity and dispersive optical bistability arising from probe-induced displacement of the trapped atoms. 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.

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Nonlinear optical phenomena occur typically at high optical intensities, or, equivalently, at high average photon numbers $\bar{n}$ in an optical resonator, because conventional materials mediate weak coupling between photons. 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. 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 [1, 2, 3] with $N \gg 1$, or cross phase modulation [4] and photon blockade [5] with $N \simeq 1$. Here, N is the number of atoms in the resonator, g the vacuum Rabi 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 [4, 5] and in recent experiments with superconducting circuits within microwave resonators [6]. Strong nonlinearities are also achieved using the long coherence times for atomic Raman transitions [7, 8, 9].

The motional degrees of freedom of ultracold atomic gases represent a new source of long-lived atomic coherence affecting light-atom interactions. This coherence leads, for example, to superradiant light scattering at low threshold intensities both in free space [10, 11] and inside optical resonators [12]. The motion of single atoms in strong-coupling cavities has been observed and cooled, [13, 14], and effects of motion in atomic ensembles in weak-coupling cavities have been examined [12, 15, 16, 17, 18].

We report here on nonlinear optics arising from 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 (1.3 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 $\bar{n} \simeq 0.05$, 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 vacuum Rabi 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 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 in the cavity that shifts the cavity resonance by $\Delta_N \simeq N g(z_0)^2/\Delta_{ca}$.

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 cavity as evident, for example, in the intracavity photon number $\bar{n}(\Delta_{pc})$ for probe light detuned by Δ_{pc} from the bare cavity resonance. Assuming the atoms adiabatically follow the trap minimum, one obtains

$$\bar{n}(\Delta_{pc}) = \frac{\bar{n}_{\rm max}}{1 + \left(\frac{\Delta_{pc} - \Delta_N(1-\epsilon\bar{n})}{\kappa}\right)^2} \qquad (1)$$

where $\bar{n}_{\rm max}$ defines the probe intensity. This lineshape is equivalent to that obtained for a damped nonlinear oscillator [19]. The deviation of $\bar{n}(\Delta_{pc})$ from the Lorentzian lineshape of the bare cavity may be quantified by the parameter $\beta = (\Delta_N \epsilon/\kappa)\bar{n}_{\max}$ that gives the maximum nonlinear shift of the cavity resonance in units of the cavity half-linewidth. In particular, for $\beta > 8\sqrt{3}/9 \simeq 1.54$, the cavity exhibits refractive bistability [20].

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 wavenumber of $k_t = 2\pi/850$ 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 at a temperature of $T = 0.8\,\mu$K. 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 predominantly occupied 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 $|\Delta_{ca}| = 2\pi \times (10-300)$ GHz detuning from the D2 resonance line at a wavenumber of $k_p = 2\pi/780$ nm. The cavity finesse for this mode was measured as $\mathcal{F}_{780} = 5.8 \times 10^5$, 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 light used in our experiment, we determine a vacuum Rabi 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, this cavity setup achieves the 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 wavenumber k_p through the cavity. For this measurement, the probe light was switched on and its frequency $\omega_p/2\pi$ was 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 variation in the intracavity optical potential, and sufficiently fast so that atom loss from probe-induced heating [22] was negligible. The transmitted probe light was separated from the trapping light using several filters, and was 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 lineshapes increasingly deviated from the symmetric lineshapes 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

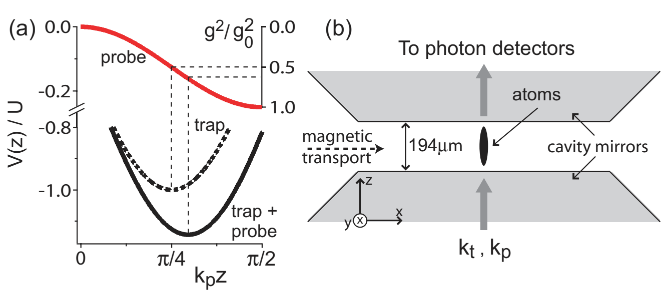

FIG. 1: (a) The optical potential due to either the cavity probe light (red, here with maximum depth $U/4$ and $\Delta_{ca} < 0$), or the trapping light (black dashed, with maximum depth U) is shown near the axial minimum of one lattice site. The trap minimum of the combined optical potential (black, solid) is 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 a standing wave of 850 nm light formed by exciting a TEM_{00} mode of a vertically oriented Fabry-Perot cavity. The system is probed by detecting transmission through another TEM_{00} mode at $k_p = 2\pi/780$ nm.

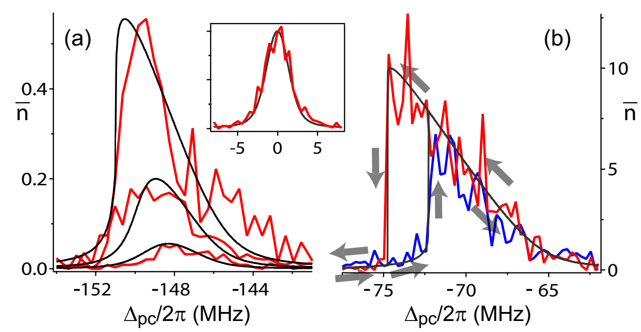

FIG. 2: Nonlinear and bistable cavity lineshapes shown as the intracavity photon number $\bar{n}$ vs. detuning Δ_{pc} from the bare-cavity at constant input power. (a) The cavity lineshape (red, single (highest $\bar{n}_{\max}$) or average of 5 (others) measurements) becomes increasingly asymmetric as 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 $\Delta_N/2\pi$ across measurements. Model lineshapes (black) using the Voigt profile indicated by the bare-cavity lineshape (inset) are calculated with $\beta = \{0.37, 1.33, 3.72\}$ determined from the observed $\bar{n}_{\max} = \{0.06, 0.20, 0.56\}$, 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) and upper (red) 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) with $\beta = 9.5$, $\Delta_{ca} = -2\pi \times 101$ GHz, and $\omega_z = 2\pi \times 42$ kHz [21].

multitude of potential wells. Due to the difference between the 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. Eq. 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 lineshape (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 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)).

At sufficiently strong refractive nonlinearity, the optical resonator becomes hysteretic and bistable. To observe clearly this bistability, we measured the transmission lineshape of the cavity resonance with 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 lineshapes (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 stability curves and the difference in the maximum intracavity photon numbers attained 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 cavity QED, 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 timescale of ω_z^{-1}. As this timescale is much longer than both the $1/\kappa$ timescale for fluctuations of the intracavity field and also the typical $\sim 10\,\mu$s timescale of variations in Δ_{pc}, such fluctuations do not destabilize the atoms-cavity system, and the full bistable region predicted by our theoretical treatment is observed.

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

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

as defined by the condition $\beta = 1$, diminishes. Here, $\omega_{\mathrm{rec}} = \hbar k_p^2/2m$ 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 $\bar{n}_{\mathrm{nl}} \simeq 10^{-4}$ for a minimum value for the trapping frequency of $\omega_z = 2\omega_{\mathrm{rec}}$ and other parameters of our experiment.

However, already for $\bar{n}_{\mathrm{nl}} < \omega_z/2\kappa \simeq 0.03$, the optical nonlinearity becomes strongly modified by the granularity of the optical forces upon the trapped atoms. During its typical residence time of $1/2\kappa$ in the cavity, a single photon imparts an impulse of $f(z)/2\kappa$ to an atom at location z. This impulse induces transient motional oscillations in the atomic medium that modulate the cavity resonance frequency. For our experimental parameters, with $N = 5\times10^4$ atoms, this modulation becomes greater than the cavity half-linewidth for $|\Delta_{ca}| \leq 2\pi \times 15$ GHz. Optical nonlinearities in this regime would manifest as temporal correlations imposed on photons transmitted through the cavity due to the light-induced atomic vibrations. Such photon correlations will be the subject of future work.

Transient oscillations may also be excited coherently, outside the aforementioned granularity regime, by diabatically varying the probe-induced optical force. We observed such oscillations as follows. With $\omega_z = 2\pi\times 49$ kHz, 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 came into coincidence with the probe frequency (Fig. 3(a)). Once the transmission signal

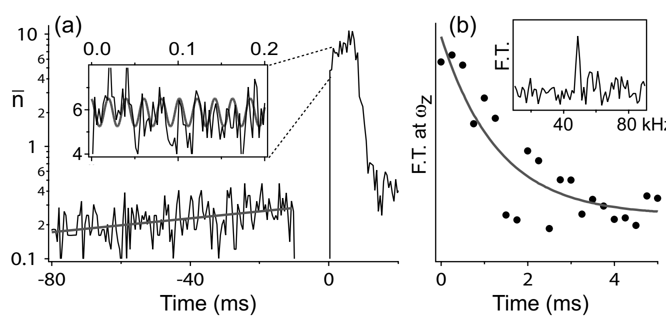

FIG. 3: Measurement of collective atomic motion. (a) A record of the cavity transmission demonstrates the experimental sequence of a trigger, delay and detection phase. Inset: coherent atomic motion modulates the cavity transmission at $\omega_z = 2\pi\times 49$ kHz (shown is the first 200 μs of the detection phase, 50-measurement average, 400 kHz bandwidth), in agreement with the gray theory curve showing the expected amplitude ($2\beta = 0.75$) and phase of the transmitted signal. (b) The transient oscillation is seen also in the transmission signal for a single measurement. The inset shows the power spectrum of the first 500 μs of the detection phase for a typical measurement. The spectral power at ω_z, determined over 500 μs intervals of the transmission measurement, decays within a $1/e$ time of 1.3(3) ms.

reached a threshold level, establishing conditionally 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. For the Voigt profile relevant to our system, the modulation of the cavity resonance 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 $1/\Gamma_{\rm osc} = 1.3$ ms, a value consistent with the inhomogeneous broadening of ω_z due to the finite radial extent of the trapped gas.

For the present system, the observed damping of the collective atomic motion imposes a technical limit of $\bar{n} = \Gamma_{\rm osc}/2\kappa \simeq 10^{-4}$ on the lowest light level at which these motion-induced nonlinearities may be exploited. However, this damping can be mitigated by radially confining the atoms more tightly or cooling them to lower temperatures. Longer-lived coherence may also be attained upon Bose-Einstein condensation, which would occur for our system below an atomic temperature of $T_c \leq 0.5\,\mu$K, close to that achieved experimentally.

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 QED [4, 5, 23], 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 [24, 25, 26].

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