Cavity Nonlinear Optics at Low Photon Numbers from Collective Atomic Motion

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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 $10^5$ $^{87}\text{Rb}$ 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.

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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. 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 $N \approx 1$. Here, $N$ is the number of atoms in the resonator, $g$ the atom-cavity coupling frequency, and $\kappa$ and $\Gamma$ 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 $n_{\text{sat}} \approx N(\Gamma/\kappa)$, the condition $n_{\text{sat}} \leq 1$ 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 $n_{\text{sat}} \approx 100$ [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 $\bar{n} \approx 10^{-4}$.

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 $\omega_z \gg h k_p^2/2m$, the position of atoms in the trap’s ground state is determined 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 $\Delta_N \approx N g(z_0)^2/\Delta_{ca}$.

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 $\Delta_N (1 - e^{i \bar{n}})$ with $e = 2h k_p^2 g_0^2/m \Delta_{ca} \omega_z^2$. Thus, this system is characterized by a Kerr nonlinearity, with the refractive index of the intracavity medium, $1 + (\Delta_N/\omega_z)(1 - e^{i \bar{n}}) = n_0 + n_2 f_p$, varying with the probe intensity $f_p \propto \bar{n}$.

This nonlinearity affects the frequency response of the driven cavity. Probing the cavity transmission with light detuned by $\Delta_{pc}$ from the bare-cavity resonance, and assuming that the atoms adiabatically follow the trap mini-
minimum of the optical trap. Given the atomic half-linewidth of $\Gamma = 2\pi \times 3$ MHz, our cavity achieves single-atom strong-coupling conditions, with critical atom ($2\Gamma / g_0^2 = 0.02$) 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/sms. 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 $\approx 0.05$.

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 (for $\Delta_{ca} > 0$) 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 $\epsilon$ 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 $\Delta_{pc}$. The bare-cavity transmission line shape [Fig. 2, inset] is well approximated by the convolution of a Lorentzian with half-linewidth $\kappa$ 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)]. 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 $N \approx 7 \times 10^4$ atoms. For $\tilde{n}_{\text{max}} = 10$, the parameter $\beta = 9.5$ is well within the bistable regime, which, for the Voigt profile relevant to our system, occurs for $\beta \approx 3.7$. The observed line shapes [Fig. 2(b)] exhibit several hallmarks of bistability, such as the abrupt changes in $\tilde{n}$ at the termini of the upper and lower branches of the bistability curve and the difference in the maximum $\tilde{n}$ 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 $\tilde{n} = 0.05$.

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 $\omega_c^{-1}$. As this time scale is longer than both the $1/\kappa$ time scale for fluctuations of the intracavity field and also the typical $\sim 10 \mu$s time scale of variations in $\Delta_{pc}$, such fluctuations do not destabilize the atoms-cavity system, and the entire range of bistability is observed.

As the detuning $\Delta_{ca}$ is reduced, the minimum photon number $\tilde{n}_{\text{nl}}$ for strong nonlinearities

\[
\tilde{n}_{\text{nl}} = \frac{4 \omega_c^2 \kappa}{\omega_N^2 g_0^2 / 2 \pm (\Delta_{ca}/2)^2} e^{\Delta_{ca}^2/4},
\]

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

For $\tilde{n}_{\text{nl}} < \omega_z / 2\kappa = 0.03$, 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 $\Delta_{ca}$ at which $\tilde{n}_{\text{nl}} < \omega_z / 2\kappa$, 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 $5 \mu$s at a high level of $\tilde{n} = 6.5$. 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 $3 \mathrm{nm}$. For the Voigt profile relevant to our system, the modulation of the cavity reso-
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 [5,25], 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].

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[24] For the large $\Delta_{\text{ca}}$ explored in this work, the equivalent nonlinearity parameter due to atomic saturation is $\beta = (\Delta_{\text{ca}}/\kappa)(\delta_0^{\text{eq}}/\Delta_{\text{ca}})^{1/2}/\delta_0$. For the parameters of Fig. 2(a), this would imply $\hat{n} > 10^6$ photons for significant nonlinearity.
[29] Background atom loss in our system occurs at 0.9 s$^{-1}$ as measured previously (K. W. Murch et al., arXiv:0706.1005). To account for this loss in between the two 25 ms long frequency sweeps, the second line shape (red) in Fig. 2(b) has been shifted by 1.7 MHz.