## **Observation of Collective Friction Forces due to Spatial Self-Organization of Atoms:** From Rayleigh to Bragg Scattering

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We demonstrate that emission-induced self-organization of two-level atoms can effect strong damping of the sample's center-of-mass motion. When illuminated by far-detuned light, cold cesium atoms assemble into a density grating that efficiently diffracts the incident light into an optical resonator. We observe random phase jumps of  $\pi$  in the emitted light, confirming spontaneous symmetry breaking in the atomic self-organization. The Bragg diffraction results in a collective friction force with center-of-mass deceleration up to 1000 m/s<sup>2</sup> that is effective even for an open atomic transition.

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The dramatic success of laser cooling and trapping of neutral atoms has relied on light forces and scattering processes that act independently on the individual atoms in the sample [1]. Making use of Rayleigh or Raman scattering from a diffuse gas, conventional Doppler and polarization gradient cooling techniques deteriorate as the optical density of the atomic sample increases. The need for cycling transitions in these cooling processes has also limited the number of accessible atomic species. If the atomic sample is spatially ordered, however, Rayleigh scattering by individual atoms does not correctly describe the light-atom interaction. Rather, interference in the light emitted by the sample that is associated with the atomic position and momentum distribution can give rise to greatly enhanced scattering rates, as in Bragg scattering. This raises the intriguing possibility of light forces on an ordered atomic sample that cannot be modeled as the sum of single-atom forces, and whose performance may improve as the atom number increases.

From the point of view of optical gain, spatial ordering of two-level atoms has been invoked to explain apparently inversionless gain mechanisms, as in recoil-induced resonances and the collective atomic recoil laser [2-8]. Inside an optical resonator, "self-oscillation" in the absence of a probe beam has been observed for a driven sample [7,8]. There, the initial atomic density grating formation is due to field and atomic density fluctuations in an atom-field system characterized by a collective instability [7]. However, few experimental studies of the atomic motion itself [9] have been conducted.

The atomic self-organization and collectively enhanced scattering are of particular interest in light of efforts to use an optical resonator's modification of scattering rates to achieve frictional light forces [10–15]. Along these lines, Domokos and Ritsch have recently predicted self-organization of driven two-level atoms in the regime of strong atom-cavity coupling, accompanied by trapping and cooling of the atomic sample [15]. They further argue that, as the atoms organize, they make a collective, symmetry-breaking choice between two equivalent but spatially offset lattice configurations.

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We experimentally confirm the predicted stochastic lattice formation by observing random  $\pi$  jumps in the time phase of the Bragg-scattered intracavity light. Moreover, we observe collectively enhanced friction forces damping the center-of-mass (c.m.) motion of two-level Cs atoms at peak decelerations of  $10^3 \text{ m/s}^2$ and light-atom detunings up to -6 GHz. Because the collective damping force is present even on an open transition and in the weak-coupling regime, we anticipate the possibility of slowing samples of other polarizable particles.

The experimental apparatus [Fig. 1(a)] and procedure are similar to those described in Ref. [14]. A nearly confocal optical resonator with finesse F = 1000 and length L = 7.5 cm is oriented along the vertical z axis. The cavity has a free spectral range of c/2L = 2 GHz and a multimode transmission spectrum width of about 200 MHz [14]. A retroreflected laser beam along the horizontal x axis with waist size  $w_0 = 2$  mm intersects the cavity center. This y-polarized pump beam, tuned near the cesium D<sub>2</sub> transition wavelength  $\lambda = 2\pi/k = 852$  nm, is derived from a tunable diode laser and amplified to a power of 160 mW in a semiconductor tapered amplifier, yielding single-beam intensities up to  $I/I_s = 4.6 \times 10^3$ , where  $I_s = 1.1$  mW/cm<sup>2</sup> is the D line



FIG. 1. (a) Schematic experimental setup. Atoms falling along the cavity axis are illuminated by horizontal pump beams and self-organize into a density grating, emitting into the cavity. (b) Cs D<sub>2</sub> line. The atom approximates an open two-level system  $F_g = 4 \rightarrow F'_e$  since the light-atom detuning is much greater than the excited state hyperfine splitting.

saturation intensity. The laser bandwidth is narrowed to less than 10 kHz by feedback from an optical resonator. A weak auxiliary beam, detuned by -150 MHz from the atomic  $F_g = 3 \rightarrow F_e = 2$  hyperfine transition, is coupled into the cavity to electronically stabilize its length. Tuning the pump beam's frequency  $\omega_i$  changes both the laser-cavity detuning  $\Delta_c = \omega_i - \omega_c$  and the laser-atom detuning  $\delta_a = \omega_i - \omega_a$ , where  $\omega_c$  and  $\omega_a$  are the frequencies of the nearest TEM<sub>00</sub> cavity resonance and the atomic  $F_g = 4 \rightarrow F_e = 5$  transition, respectively.

The experiment begins with  $N \approx 10^7$  Cs atoms in a magneto-optical trap (MOT) on the cavity axis, prepared at velocities between 0 and 30 cm/s by dropping them from variable height 0 to 4 mm above the cavity center. We illuminate the atoms with the pump beam detuned in the range -5.7 GHz  $\leq \delta_a/2\pi \leq -1.6$  GHz for a duration between 50  $\mu$ s and 2 ms. The atoms are then allowed to fall freely, and after 40 ms a time-of-flight (TOF) measurement of the vertical velocity distribution is carried out by fluorescence from a horizontal light sheet. Except when we investigate an open transition, a repumper beam tuned to the  $F_g = 3 \rightarrow F_e = 4$  transition retains the atoms in the  $F_g = 4$  hyperfine state. A photodiode measures the power exiting the cavity.

Above a threshold pump intensity, we observe greatly enhanced emission from the atoms into the cavity, with a ratio of scattering rate into the cavity  $\Gamma_c$  to scattering rate into free space  $\Gamma_{\rm fs}$  as large as  $2\eta_c = \Gamma_c/\Gamma_{\rm fs} = 200$  near  $\Delta_c = 0$ . This scattering ratio contrasts with a maximum  $2\eta_s = 0.1$  for scattering into the cavity without collective enhancement, and  $2\eta = 1$  for intracavity Raman lasing driven by near-detuned light [14]. The collective emission occurs for any pump frequency within the cavity's multimode resonance structure. The threshold increases with temperature and decreases with atom number, as expected for a cooperative effect that relies on spatial ordering. For a typical MOT temperature  $T = 6 \ \mu K$  and atom number  $N = 10^7$ , the Rayleigh scattering rate per atom into free space at threshold is  $\Gamma_{\rm fs} \approx 10^4 \, {\rm s}^{-1}$ . The emitted light has the same linear y polarization as the pump field, and the emission lasts typically 500  $\mu$ s and maximally 2 ms.

The collective emission into the cavity can be explained by a gain process in which the pump beam undergoes Bragg diffraction from a self-organizing atomic density grating [2,15]. Below threshold, gain spectra obtained by coupling a weak probe beam into the cavity while the pump illuminates the atoms indicate that the gain is described by a recoil-induced resonance [2]. Above threshold, the emission becomes a runaway process: scattering strengthens the intracavity field, which in turn improves the atomic localization, leading to greater Bragg scattering into the resonator. A heterodyne measurement shows that, for a stationary atom cloud, the frequency of the light emitted above threshold is identical to that of the driving light (to within our 60 Hz resolution), as expected for Bragg diffraction.

organization is the spontaneous breaking of translational symmetry in a standing-wave cavity by the atoms' organization into either of two grating configurations [15]. Consider first atoms in a single yz plane. For  $\delta_a < 0$ , light forces associated with the intracavity standing wave cause localization in cavity antinodes with spatial period  $\lambda/2$ . The fields Rayleigh scattered by atoms at adjacent antinodes differ in phase  $\phi$  by  $\pi$  and interfere destructively. A macroscopic intracavity field that scales linearly with atom number N, rather than with the fluctuation in atom number  $\sqrt{N}$ , can build up only for a grating of period  $\lambda$ . The symmetry between preferred occupation of even- and odd-numbered antinodes is broken by fluctuations in the atomic position distribution [15]. Since the time phase of the Bragg diffracted light differs by  $\pi$  for the two gratings, it can be used to observe the symmetry breaking. When the atomic distribution along x is included, the atoms organize into either of two possible 2D rectangular lattices oriented at the angle bisecting the pump and cavity axes, again with a  $\pi$  time phase difference in the emission from the two configurations. The two possible lattices are then interlaced in a chessboard pattern of period  $\lambda$  (see inset of Fig. 3).

A significant prediction of a model for self-

To test the self-organization model of Ref. [15], we measure the relative time phase  $\phi$  between intracavity and pump light by heterodyning the cavity emission with frequency-shifted pump light. For a nearly stationary atomic ensemble, the phase of the intracavity light remains approximately constant, with drifts of less than  $\pi/9$  over 1 ms. If a randomizing force is exerted on the atoms by applying the MOT light during the emission—thus extending the emission duration—periods of nearly constant phase are interrupted by sharp phase jumps of approximately  $\pi$  [Fig. 2(b)]. Because these phase jumps must correspond to spontaneous rearrangement of the atomic spatial distribution, during which Bragg scattering is briefly suppressed, the amplitude of the emission also drops briefly when the phase jumps [Fig. 2(a)].

To further verify the atomic self-organization and determine the corresponding diffusion time, we measure the relative phase  $\Delta \phi$  between the emitted fields due to two pump pulses of 300  $\mu$ s duration, separated by a variable time  $T_{\text{off}}$ . We expect to find  $\Delta \phi = 0$  if the atoms remain in the same lattice between the two pulses, and equal probabilities of observing  $\Delta \phi = 0$  and  $\Delta \phi = \pi$  if the atoms diffuse randomly during  $T_{\text{off}}$ . The probability of observing intermediate values of relative phase,  $0 < \Delta \phi < \pi$ , should be correspondingly suppressed.

For each different  $T_{\text{off}}$ , we measure the relative phases  $\Delta\phi$  of 1000 pairs of pulses and histogram the phases with a  $\pi/5$  bin size. This data is taken with the MOT light on at all times to accelerate atomic diffusion. In Fig. 3, we plot the fractions  $f_0$ ,  $f_{\pi/2}$ , and  $f_{\pi}$  of the pulse pairs for which the relative phase is  $\Delta\phi = 0$ ,  $\Delta\phi = \pi/2$ , and  $\Delta\phi = \pi$ , respectively, as a function of pulse separation. Our observations confirm the symmetry breaking predicted in [15].

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FIG. 2. Simultaneous time traces of the intracavity intensity (a) (arbitrary units) and field phase (b). The parameters are  $N = 8.2 \times 10^6$ ,  $\delta_a/2\pi = -1.59$  GHz,  $\Delta_c/2\pi = -20$  MHz, and  $I/I_s = 440$ .

For short  $T_{\text{off}}$ , the relative phase is predominantly  $\Delta \phi = 0$ . As  $T_{\text{off}}$  increases, relative phases of  $\Delta \phi = \pi$  appear, and the difference  $f_0 - f_{\pi}$  decays with a time constant  $\tau = (11 \pm 3) \ \mu$ s. Optical-path-length drifts in the interferometer then cause a slow decay in  $f_0$  and  $f_{\pi}$ , and an increase in  $f_{\pi/2}$  to the 10% random-phase level.

Having confirmed a mechanism for the emission, we investigate the forces on falling atoms interacting with the pump light after the MOT is extinguished. The collective emission into the cavity leads to strong damping of the c.m. motion of the atoms, an effect qualitatively similar to that predicted by Gangl *et al.* for atoms in a multimode ring cavity [11]. The TOF measurements indicate that, following emission, a large fraction of the atoms is substantially decelerated (inset of Fig. 4). For atoms with an initial velocity  $v_0 = 15$  cm/s at the time of illumination, decelerations of about 300 m/s<sup>2</sup> are observed, with typically a third of the atoms being slowed. The TOF trace shown indicates a final velocity of 4 cm/s, while those with the largest delays indicate that the cloud is stopped before the emission ends.

For driven atoms falling at velocity v > 0, the measured intracavity power is modulated at a frequency given initially by twice the atomic Doppler effect  $2\omega_d = 2kv$ (Fig. 4). This modulation can be explained as the beat note between fields emitted upward and downward, which are Doppler shifted by  $\pm \omega_d$ . Alternatively, it can be explained by the spatial variation with period  $\lambda/2$  of the atom-cavity coupling, whereby falling atoms cannot emit at a node of the intracavity standing wave. The changing modulation frequency then indicates the atomic acceleration, as displayed in Fig. 4. These measured accelerations are consistent with the TOF measurements. The peak accelerations, up to  $-1000 \text{ m/s}^2$ , occur shortly after the onset of the collective emission. Heating of the delayed atoms is consistent with recoil heating at the photon scattering rate into the cavity. While most of our data are taken in the range  $-1.77 \le \delta_a/2\pi \le -1.57$  GHz, 203001-3



FIG. 3. The fraction of pulse pairs with relative phase shift  $\Delta\phi$  is plotted versus pulse separation time. The solid circles, open circles, and solid squares correspond to  $\Delta\phi = 0 \pm \pi/10$ ,  $\pi \pm \pi/10$ , and  $\pi/2 \pm \pi/10$ , respectively. The parameters are the same as for Fig. 2, except here  $N = 1.3 \times 10^7$ . The inset shows the two possible lattice configurations producing relative phase shift  $\Delta\phi = \pi$  in the emitted light.

we observe damping for pump-atom detunings up to  $\delta_a/2\pi \approx -5.7$  GHz, limited only by the pump intensity needed to reach threshold. For blue pump-atom detunings up to  $\delta_a/2\pi \approx 2.5$  GHz there is collective emission but no c.m. damping.

Since the pump-atom detuning  $\delta_a$  is much larger than the atoms' excited-state hyperfine splitting [Fig. 1(a)], the transition  $F_g = 4 \rightarrow F_e$  can be treated as an open, twolevel system with decay to the  $F_g = 3$  ground state. If the repumper counteracting the decay is turned off during the application of the pump beam, the ensuing TOF traces are similar to that displayed in the inset of Fig. 4, with a similar fraction of 40% of the sample being slowed. For an initial c.m. velocity of  $v_0 = 15$  cm/s the Bragg scattering lasts up to 200  $\mu$ s, slowing the sample to a final



FIG. 4. The emitted power (thin line) during the illumination of a falling atomic cloud. The beat frequency is used to calculate the deceleration (thick line). The inset shows the time-of-flight signal, in arbitrary units, of atomic clouds without (gray line) and with (black line) a 400  $\mu$ s exposure to the pump beam. Here the pump  $I/I_s = 420$ ,  $\delta_a/2\pi =$ -1.580 GHz, and  $\Delta_c/2\pi = -10$  MHz. The atom number is  $N = 2.6 \times 10^7$ .

velocity of 4 cm/s. This change in atomic momentum corresponds to 33 photon recoils per atom, which is much larger than the average number of five photons necessary to depump the atoms to the  $F_g = 3$  dark ground state. This implies that, due to the large cavity-to-free-space scattering ratio  $2\eta_c$ , the momentum change  $\Delta p$  per atom can far exceed the impulse that could be produced by the radiative force on an open transition. Therefore this technique may prove useful for slowing of a particle beam on an open transition.

We propose that the observed damping of the c.m. velocity can be understood in terms of different magnitudes of the blue and red Doppler sidebands produced by combined amplitude and phase modulation of the intracavity light [16]. If the blue Doppler sideband's magnitude exceeds that of the red one, energy conservation requires that kinetic energy be extracted from the atomic sample. The spatially modulated atom-light interaction in a standing-wave mode implies that for a falling atomic density grating the Bragg diffraction into the cavity is modulated, resulting in double-sideband, carrier-free amplitude modulation of the intracavity field with frequency  $\omega_d = kv$ . At the same time, the atomic index of refraction detunes the cavity when the atoms pass through an antinode, resulting in phase modulation of the intracavity field at a rate  $2\omega_d$ . In analogy to single-sideband modulation, the interference between these two modulation processes results in asymmetric Doppler sidebands of the intracavity light, with the sign of the asymmetry determined by the sign of the light-atom detuning  $\delta_a$ . For the present parameters, this model predicts slowing for  $\delta_a < 0$ at rates in agreement with our experimental results [16].

Finally, we speculate on applications to the creation of cold, slow molecular or atomic ensembles. By implementing a fast but cold, supersonic beam and slowing the beam using an open transition, one could extend the range of trapable species. For c.m. Doppler effects much larger than the cavity linewidth, appropriate red pump-cavity detuning should allow enhancement of only the blue Doppler sideband, leading to optimal sideband asymmetry and correspondingly large forces [12]. An application to the magnetic trapping of atomic chromium [17] illustrates this idea. If a cold beam of Cr is created by a method similar to that demonstrated for iron [18], the present damping technique could be used to stop the beam and load a magnetic trap. Ordinary laser cooling is possible on the  ${}^{7}S_{3} \rightarrow {}^{7}P_{4}$  transition at  $\lambda = 425.6$  nm, but decay to the  ${}^{5}D_{3}$  and  ${}^{5}D_{4}$  states allows the scattering of only  $2.5 \times 10^3$  photons per atom [17], limiting the c.m. velocity change to about 45 m/s. For a scattering ratio of  $2\eta_c = 200$  and optimum sideband asymmetry, a cold beam with c.m. velocities up to  $10^3$  m/s could be stopped. As our technique at large detuning is independent of shifts and broadening of the atomic lines, it could be performed inside a deep magnetic trap.

Our evidence indicates damping of the atomic centerof-mass motion, but not cooling of all motional degrees 203001-4 of freedom. There may, however, exist other modes of atomic motion which are damped as well, albeit with a smaller collective enhancement [11]. More promising is a proposal originally intended for stochastic laser cooling of atoms [19]. If the atomic sample is allowed to evolve in a harmonic potential, then the rotation of the position and momentum phase-space coordinates causes the c.m. mode to be mixed with all translational degrees of freedom, allowing all modes to be cooled.

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