## Generation of Macroscopic Pair-Correlated Atomic Beams by Four-Wave Mixing in Bose-Einstein Condensates

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By colliding two Bose-Einstein condensates, we have observed strong bosonic stimulation of the elastic scattering process. When a weak input beam was applied as a seed, it was amplified by a factor of 20. This large gain atomic four-wave mixing resulted in the generation of two macroscopically occupied pair-correlated atomic beams.

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In a gaseous Bose-Einstein condensate (BEC), all the atoms occupy the ground state of the system [1]. Once BEC has been achieved, the initial well-defined quantum state can be transformed into other more complex states by manipulating it with magnetic and optical fields. This can result in a variety of time-dependent macroscopic wave functions [1], including oscillating condensates, multiple condensates moving relative to each other, an output coupler, and rotating condensates with vortex lattices. Such macroscopically occupied quantum states represent classical matter-wave fields in the same way an optical laser beam is a classical electromagnetic wave. The next major step involves engineering nonclassical states of atoms that feature quantum entanglement and correlations. These states are important for quantum information processing, subshot noise precision measurements [2], and tests of quantum nonlocality.

Quantum correlations in the BEC ground state have been observed in a BEC held in optical lattices [2-4]. The repulsive interactions between the atoms within each lattice site force the occupation numbers to equalize, resulting in a number squeezed state. Alternatively, correlations in a BEC can be created in a dynamic or transient way through interatomic collisions. At the low densities typical of current experiments, binary collisions dominate, creating correlated pairs of atoms. Because of momentum conservation, the pair-correlated atoms scatter into modes with opposite momenta in the center-of-mass frame, resulting in squeezing of the number difference between these modes [5-7]. Our work is an implementation of the suggestions in Refs. [5,6]. However, we use elastic scattering processes instead of spin flip collisions to create pair correlations because the elastic collision rate is much higher than the spin flip rate. This was essential to observe large amplification before further elastic collisions led to losses.

Elastic scattering between two BEC's produces a collisional halo [8], where the number of atoms moving into opposing solid angles is the same, corresponding to number squeezing. Once these modes are occupied, the scattering process is further enhanced by bosonic stimulation. The onset of such an enhancement was observed in Ref. [8].

In this paper, we report strong amplification, corresponding to a gain of at least 20. Based on a theoretical prediction [6], which drew an analogy to optical superradiance [9], we expected to obtain a highly anisotropic gain using our cigar-shaped condensate. However, this mechanism of mode selection proved to be irrelevant for our experiment, because atoms do not leave the condensate during amplification (see below). Instead, we preselected a single pair-correlated mode by seeding it with a weak third matter wave, and observed that up to 40% of the atoms scattered into it. Because the scattered atoms are perfectly pair correlated, the only fluctuations in the number difference between the two beams stem from number fluctuations in the initial seed. Therefore, an observed gain of 20 implies that we have improved upon the shot-noise limit by a factor of  $\sqrt{40}$ , although this was not directly observed. Such a four-wave mixing process with matter waves had only been observed previously with a gain of 1.5 [10,11].

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This experiment was performed with sodium condensates of ~30 million atoms in a cigar-shaped magnetic trap with radial and axial trap frequencies of 80 and 20 Hz, respectively. Such condensates had a mean field energy of 4.4 kHz, a speed of sound of 9 mm/s, and radial and axial Thomas-Fermi radii of 25 and 100  $\mu$ m, respectively. The second condensate and the seed wave were generated by optical Bragg transitions to other momentum states. Figure 1 shows the geometry of the Bragg beams. Four laser beams were derived from the same laser that was 100 GHz red detuned from the sodium  $D_2$  line. The large detuning prevented optical superradiance [9]. All beams propagated at approximately the same angle of  $\sim 0.35$  rad with respect to the long axis of the condensate, and could be individually switched on and off to form beam pairs to excite two-photon Bragg transitions [12] at different recoil

The seed wave was created by a weak 20  $\mu$ s Bragg pulse of beams with momenta  $\mathbf{p}_3$  and  $\mathbf{p}_2$ , which coupled 1%–2% of the atoms into the momentum state  $\mathbf{k}_s = \mathbf{p}_3 - \mathbf{p}_2$ , with a velocity ~15 mm/s. Subsequently, a 40  $\mu$ s  $\pi/2$  pulse of beams  $\mathbf{p}_1$  and  $\mathbf{p}_2$  splits the condensate into two strong source waves with momenta  $\mathbf{k}_1 = 0$ 

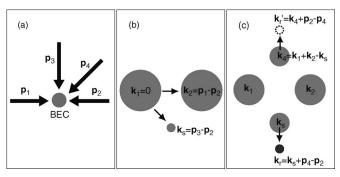


FIG. 1. Arrangement of laser beams to generate three atomic wave packets: (a) Four Bragg beams intersected at the condensate. (b) Two source waves and a small seed were created with Bragg beam pairs. (c) The four-wave mixing process amplified the seed and created a fourth wave. Both were subsequently read out with another Bragg pulse. The figures are projections on a plane perpendicular to the condensate axis. All wave packets move within this plane.

and  $\mathbf{k}_2 = \mathbf{p}_1 - \mathbf{p}_2$  (see Fig. 1b), corresponding to a relative velocity of  $\sim 20$  mm/s. The four-wave mixing process involving these three waves led to an exponential growth of the seed wave, while a fourth conjugate wave at momentum  $\mathbf{k}_4 = \mathbf{k}_1 + \mathbf{k}_2 - \mathbf{k}_s$  emerged and also grew exponentially (Fig. 1c). The Bragg beams were arranged in such a way that the phase matching condition was fulfilled [the sum of the kinetic energies of the source waves  $(\sim 11 \text{ kHz})$  matched the energy of the seed and the fourth wave]. The effect of any energy mismatch on the process will be discussed later. The four-wave mixing process was analyzed by absorption imaging [1]. Figure 2c shows the key result of this paper qualitatively: A small seed and its conjugate wave were amplified to a size where a significant fraction of the initial condensate atoms had been transferred into this pair-correlated mode.

To study this process, we applied "readout" beams  $\mathbf{p}_2$  and  $\mathbf{p}_4$  for 40  $\mu$ s, interrupting the amplification after a variable growth period between 0 and 600  $\mu$ s. {Turning off the trap after a variable amount of time is insuf-

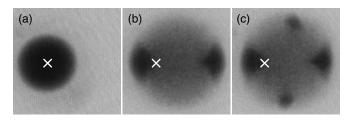


FIG. 2. High-gain four-wave mixing of matter waves. The wave packets separated during 43 ms of ballistic expansion. The absorption images [1] were taken along the axis of the condensate. (a) Only a 1% seed was present (barely visible), (b) only two source waves were created and no seed, and (c) two source waves and the seed underwent the four-wave mixing process where the seed wave and the fourth wave grew to a size comparable to the source waves. The gray circular background consists of spontaneously emitted atom pairs that were subsequently amplified to around 20 atoms per mode. The crosses mark the center position of the unperturbed condensate. The field of view is 1.8 mm wide.

ficient in this case because the density decreases on the time scale of the trapping period [1/(80 Hz)], while the amplification occurs more rapidly.} The frequency difference between the two readout beams was selected such that a fixed fraction of the seed (fourth) wave was coupled out to a different momentum state  $\mathbf{k}_r = \mathbf{k}_s + \mathbf{p}_4 - \mathbf{p}_2$  ( $\mathbf{k}_r' = \mathbf{k}_4 + \mathbf{p}_2 - \mathbf{p}_4$ ) (Fig. 1c).  $\mathbf{k}_r$  or  $\mathbf{k}_r'$  did not experience further amplification due to the constraint of energy conservation and therefore could be used to monitor the atom number in the seed (fourth) wave during the fourwave mixing process (Fig. 3).

The growth of the 2% seed and the fourth wave are shown in Fig. 4. As expected, the growth rates were found to increase with the mean field energy. Eventually, the amplification slowed down and stopped as the source waves were depleted. This is in contrast to Ref. [10,13], where the mixing process was slow (due to much lower mean field energy), and the growth time was limited by the overlap time of the wave packets. In our experiment, the overlap time was  $\geq 1.8$  ms, whereas the growth stopped already after  $\leq 500 \ \mu s$ .

A simple model describes the salient features of the process. The Hamiltonian of a weakly interacting Bose condensate is given by [11]

$$H = \sum_{\kappa} \frac{\hbar^2 \kappa^2}{2m} \, \hat{a}_{\kappa}^{\dagger} \hat{a}_{\kappa} + \frac{2\pi \hbar^2 a}{mV} \sum_{\kappa_1 + \kappa_2 \atop = \kappa_1 + \kappa_4} \hat{a}_{\kappa_1}^{\dagger} \hat{a}_{\kappa_2}^{\dagger} \hat{a}_{\kappa_3} \hat{a}_{\kappa_4} \,, \quad (1)$$

where  $\kappa$  denotes the wave vectors of the plane wave states, m is the mass, V is the quantization volume,  $\hat{a}_{\kappa_i}$  is the annihilation operator, and a=2.75 nm is the scattering length. If two momentum states  $\mathbf{k}_1$  and  $\mathbf{k}_2$  are highly occupied relative to all other states (with occupation numbers  $N_1$  and  $N_2$ ), the initial depletion of  $\mathbf{k}_1$  and  $\mathbf{k}_2$  can be neglected. Therefore, the only interactions are mean field interactions (self-interactions) and scattering involving  $\mathbf{k}_1$  and  $\mathbf{k}_2$ . In the Heisenberg picture, the difference between the

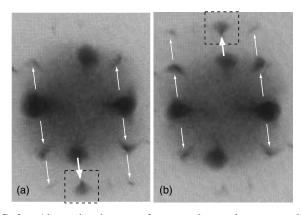
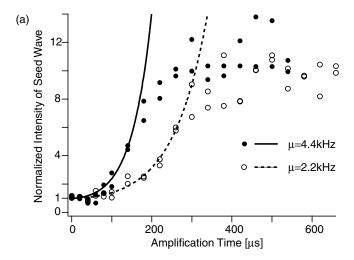


FIG. 3. Absorption images after a readout pulse was applied to (a) the seed wave and (b) the fourth wave. The thick arrows indicate the readout process. The readout pulse was kept short (40  $\mu$ s), resulting in a large Fourier bandwidth and off-resonant coupling to other wave packets indicated by the narrow arrows. However, this did not affect the readout signal (atoms in the dashed box).

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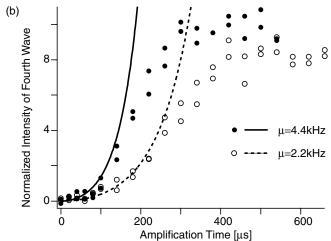


FIG. 4. Generation of pair-correlated atomic beams. The growth of (a) a 2% seed and (b) its conjugate fourth wave are shown for two different chemical potentials  $\mu$ . The intensities of the waves were determined by counting the number of atoms in the dashed boxes in Fig. 3 and were normalized to the intensity of the initial seed. The solid lines and dashed lines are fits to the initial growth according to Eq. (4) with growth rates of  $(170~\mu s)^{-1}$  and  $(100~\mu s)^{-1}$ , respectively.

occupations of the mode pairs  $\Delta \hat{n} = \hat{a}_{\kappa_1}^{\dagger} \hat{a}_{\kappa_1} - \hat{a}_{\kappa_2}^{\dagger} \hat{a}_{\kappa_2}$  is time independent for any  $\kappa_1 + \kappa_2 = \mathbf{k}_1 + \mathbf{k}_2$ . Therefore, the fluctuations in the number difference  $\langle \Delta \hat{n}^2 \rangle - \langle \Delta \hat{n} \rangle^2$  remain constant even though the occupations grow in time. The result is two-mode number squeezing. This is equivalent to a nondegenerate parametric amplifier; the Hamiltonians for both systems are identical [14].

When calculating the occupations  $\langle \hat{a}_{\kappa_1}^{\dagger} \hat{a}_{\kappa_1} \rangle$ ,  $\langle \hat{a}_{\kappa_2}^{\dagger} \hat{a}_{\kappa_2} \rangle$  and the correlation  $\langle \hat{a}_{\kappa_1} \hat{a}_{\kappa_2} \rangle$ , the relevant physical parameters are

$$\overline{\mu} = \sqrt{\mu_1 \mu_2}, \qquad \mu_i = \frac{4\pi \hbar^2 a}{mV} N_i \qquad (i = 1, 2), 
\Delta \omega = \frac{\hbar \kappa_1^2}{2m} + \frac{\hbar \kappa_2^2}{2m} - \frac{\hbar \mathbf{k}_1^2}{2m} - \frac{\hbar \mathbf{k}_2^2}{2m} + \frac{\mu_1}{\hbar} + \frac{\mu_2}{\hbar},$$
(2)

where  $\overline{\mu}$  is the (geometric) average mean field energy of the two source waves, and  $\hbar \Delta \omega$  is the energy mismatch for the scattering of atoms from states  $\mathbf{k}_1$  and  $\mathbf{k}_2$  to states

 $\kappa_1$  and  $\kappa_2$ . One obtains exponential growth for  $\kappa_1$  and  $\kappa_2$  if  $\overline{\mu} > \hbar \Delta \omega / 4$ , and the growth rate is given by

$$\eta = \sqrt{\left(\frac{2\overline{\mu}}{\hbar}\right)^2 - \left(\frac{\Delta\omega}{2}\right)^2}.$$
 (3)

For our initial conditions with s atoms in the seed wave  $\mathbf{k}_s$  and an empty fourth wave  $\mathbf{k}_4$ , the correlation  $\langle \hat{a}_{\mathbf{k}_s} \hat{a}_{\mathbf{k}_4} \rangle$  starts to grow as

$$|\langle \hat{a}_{\mathbf{k}_s} \hat{a}_{\mathbf{k}_4} \rangle| = rac{2\overline{\mu}\sqrt{4\overline{\mu}^2\cosh(\eta t)^2 - rac{\Delta\omega^2}{4}}}{\eta^2} imes \sinh(\eta t)(s+1).$$

This leads to exponential growth of the occupation numbers [10,13]:

$$\langle \hat{a}_{\mathbf{k}_{s}}^{\dagger} \hat{a}_{\mathbf{k}_{s}} \rangle = \left[ \frac{2\overline{\mu}}{\hbar \eta} \sinh(\eta t) \right]^{2} (s+1) + s,$$

$$\langle \hat{a}_{\mathbf{k}_{4}}^{\dagger} \hat{a}_{\mathbf{k}_{4}} \rangle = \left[ \frac{2\overline{\mu}}{\hbar \eta} \sinh(\eta t) \right]^{2} (s+1).$$
(4)

Equations (3) and (4) show that, for a large mean field energy  $\overline{\mu}$ ,  $\Delta\omega$  can be quite large without suppressing the four-wave mixing process. When  $\Delta\omega > 4\overline{\mu}/\hbar$ , one has to replace the hyperbolic sine functions in Eq. (4) with sine functions and  $\eta$  in Eq. (3) with  $\sqrt{(\Delta\omega/2)^2 - (2\overline{\mu}/\hbar)^2}$ . The occupations in states  $\mathbf{k}_s$  and  $\mathbf{k}_4$  still grow initially, but then they begin to oscillate. The above solution also applies to initially empty modes with s=0.

We can estimate the maximum growth rate  $(2\overline{\mu}/\hbar)$  for our experiment by using the average mean field energy across the condensate to obtain  $(53~\mu\text{s})^{-1}$  for high and  $(110~\mu\text{s})^{-1}$  for low mean field energy. The experimental data exhibit a somewhat slower growth rate of  $(100~\mu\text{s})^{-1}$  and  $(170~\mu\text{s})^{-1}$ , respectively. This discrepancy is not surprising since our theoretical model does not take into account depletion and possible decoherence processes due to the finite size and inhomogeneity of a magnetically trapped condensate. We also observed that the angles of the Bragg beams and therefore the energy mismatch  $\Delta\omega$  could be significantly varied without substantially affecting the four-wave mixing process, confirming the robustness [see Eq. (3)] of four-wave mixing.

In addition to the four distinct wave packets, Fig. 2 also shows a circular background of atoms that are scattered from the source waves  $\mathbf{k}_1$  and  $\mathbf{k}_2$  into other pairs of initially empty modes  $\kappa_1$  and  $\kappa_2$  ( $\kappa_1 + \kappa_2 = \mathbf{k}_1 + \mathbf{k}_2$ ). The scattered atoms lie on a spherical shell in momentum space centered at  $(\mathbf{k}_1 + \mathbf{k}_2)/2$  with a radius  $|\mathbf{k}|$  close to  $|\mathbf{k}_1 - \mathbf{k}_2|/2$  and a width  $|\Delta \mathbf{k}| \sim m\sqrt{2\pi\overline{\mu}/t\hbar^3}/|\mathbf{k}|$  [13]. As time progresses, the thickness  $|\Delta \mathbf{k}|$  narrows due to the exponential gain.

Eventually, the population of these background modes contributes to the depletion of the source waves. One can estimate the depletion time  $t_d$  of the source waves by comparing the total population in these modes to the original number of atoms. This sets a theoretical limit on the gain  $G = e^{4\overline{\mu}t_d/\hbar}/4$  given by  $4G/\sqrt{\ln(4G)} = \sqrt{2\pi}/|\mathbf{k}|a$ . For

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our geometry  $|\mathbf{k}|a = 0.01$  and G = 160. In our condensate of  $3 \times 10^7$  atoms, this maximum gain is achieved when all the atoms are scattered into the  $9 \times 10^4$  pair modes in the momentum shell. With a 1% seed, the source waves are depleted earlier, leading to a maximum gain of 37, where we measured a gain of 20. For a 2% seed the measured gain was 10 (see Fig. 4).

In our experiment, we deliberately reduced the velocity between the two source waves to twice the speed of sound in order to increase G and also the overlap time between the two source waves. Under these circumstances, the thickness of the shell  $|\Delta \mathbf{k}|$  becomes close to its radius, accounting for the uniform background of scattered atoms rather than the thin s-wave halo observed in Ref. [8]. For velocities around or below the speed of sound, the condensate will not separate from the other waves in ballistic expansion.

Once the amplified modes are populated, losses due to further collisions occur at a rate  $\Gamma \sim 8\pi a^2 n\hbar |\mathbf{k}|/m$  per atom (n is the number density of atoms). In order to have net gain, the growth rate  $\eta$  should be greater than  $\Gamma$ , which is the case since  $\eta/\Gamma = 1/|\mathbf{k}|a = 100 \gg 1$ . Furthermore, we begin to lose squeezing when s + 1 atoms are lost from the mode pair that occurs approximately at a gain of  $e^{4\overline{\mu}t/\hbar}/4 = 1/|\mathbf{k}|a$ . At this point, the condensate is already highly depleted. In our experiment, however, the shell of amplified modes is so thick that it includes many of the modes into which atoms are scattered and increases the scattering rate by bosonic stimulation. Ideally, the atomic beams should separate after maximum gain is achieved. However, for our condensate size, the waves overlap for a much longer time and suffer collisional losses. This is visible in Fig. 2, where 40% of the atoms were transferred to the seeded mode pair, but only  $\sim 10\%$  survived the ballistic expansion.

The collisional amplification process studied here bears similarities to the superradiant Rayleigh scattering of light from a Bose condensate [9], where correlated photon-atom pairs are generated in the end-fire mode for the photons and the corresponding recoil mode for the atoms. However, there are significant differences between the two processes. In optical superradiance, the scattered photons leave the condensate very quickly, causing only the recoiled atoms to maintain the coherence and undergo exponential growth. This physical situation is reflected in the Markov approximation adopted in Refs. [6,15]. In contrast, the atoms move slowly in collisional amplification, and the Markov approximation does not hold (although it was applied in Ref. [6]). The energy uncertainty  $\Delta E = \hbar/\Delta t$  for a process of duration  $\Delta t$  gives a longitudinal momentum width of  $\Delta E/v$ , where v is the speed of light for photons or the velocity of the scattered atoms. This shows that optical superradiance is much more momentum selective: The shell in momentum space is infinitesimally thin, and only the atomic modes with maximal overlap with this shell are selected. In contrast, the shell in collisional amplification is many modes thick and does not lead to strong mode

selection. Moreover, in optical superradiance the light is coherently emitted by the entire condensate, whereas collisional amplification reflects only local properties of the condensate, because the atoms do not move significantly compared to the size of the condensate. Therefore, features like growth rate, maximum amplification, and even whether mode pairs stay squeezed do not depend on global parameters such as size or shape.

In conclusion, we have observed high gain in atomic four-wave mixing and produced pair-correlated atomic beams. We have also identified some limitations for using collisions to create such twin beams, including loss by subsequent collisions, and competition between other modes with similar gain.

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