Sodium Bose-Einstein Condensates in the $F = 2$ State in a Large-Volume Optical Trap

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We have investigated the properties of Bose-Einstein condensates of sodium atoms in the upper hyperfine ground state. Condensates in the high-field seeking $|F = 2, m_F = -2\rangle$ state were created in a large volume optical trap from initially prepared $|F = 1, m_F = -1\rangle$ condensates using a microwave transition at 1.77 GHz. We found condensates in the stretched state $|F = 2, m_F = -2\rangle$ to be stable for several seconds at densities in the range of 10^{14} atoms/cm^3. In addition, we studied the clock transition $|F = 1, m_F = 0\rangle \rightarrow |F = 2, m_F = 0\rangle$ in a sodium Bose-Einstein condensate and determined a density-dependent frequency shift of $(2.44 \pm 0.25 \pm 0.5) \times 10^{-12}$ Hz cm^3.

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So far, Bose-Einstein condensation in dilute atomic gases [1] has been achieved in all stable bosonic alkali isotopes except ^{39}K, as well as in atomic hydrogen [2] and metastable helium [3]. The physics that can be explored with Bose-Einstein condensates (BEC) is to a large extent governed by the details of interatomic interactions. At ultralow temperatures, these interactions not only vary significantly from one atomic species to another but can also change significantly for different internal states of a single species. While in ^{87}Rb, only minor differences of the collisional properties are observed within the ground state manifolds, in ^{7}Li, the magnitude of the scattering length differs by a factor of 5 between the upper and the lower hyperfine manifold and even the sign is inverted [4]. The behavior of ^{23}Na with a scattering length of 2.80 nm in the $|F = 1, m_F = \pm 1\rangle$ states and 3.31 nm in the $|F = 2, m_F = \pm 2\rangle$ states [5] is intermediate. Thus, ^{23}Na might provide a system in which the study of BEC mixtures of states with significantly differing scattering length is possible. Such a mixture would be a natural extension of earlier work on spinor condensates in ^{87}Rb [6] and in the $F = 1$ manifold of ^{23}Na [7].

In this Letter, we report the realization of Bose-Einstein condensates of ^{23}Na in the upper $F = 2$ hyperfine manifold in a large-volume optical trap [8]. In ^{87}Rb, condensates in both the $F = 1$ and $F = 2$ states had been achieved by loading atoms in either state into a magnetic trap and subsequent evaporative cooling. In contrast, sodium BECs have previously been produced only in the $F = 1$ state. Early attempts at MIT and NIST to evaporatively cool sodium in the $F = 2$ state were discontinued since the evaporative cooling scheme proved to be more robust for the $F = 1$ state. Instead of developing an optimized evaporation strategy for $F = 2$ atoms in a magnetic trap, we took advantage of an optical trap which traps atoms in arbitrary spin states [9]. After producing $F = 1$ condensates and loading them into an optical trap, we transferred the population into the $F = 2$ manifold using a single-photon microwave transition at 1.77 GHz. We found that a BEC in the stretched $|F = 2, m_F = -2\rangle$ state is stable on time scales of seconds at densities of a few 10^{14} atoms/cm^3. Simultaneous trapping of condensates in the $|2, -2\rangle$ and $|1, -1\rangle$ states for several seconds was also achieved. In contrast, at the same density, a condensate in the $|2, 0\rangle$ state decays within milliseconds. Nevertheless, we were able to observe the so-called clock transition $|1, 0\rangle \rightarrow |2, 0\rangle$ in a BEC, which is to lowest order insensitive to stray magnetic fields.

The basic setup of our experiment is described in [10,11] and is briefly summarized here. We have prepared condensates of more than 4×10^7 ^{23}Na atoms in a so-called "clover-leaf" magnetic trap with trapping frequencies of $\nu_x = 16$ Hz and $\nu_y = \nu_z = 160$ Hz by radio frequency evaporation for 20 s. After preparation of the condensate in the $|1, -1\rangle$ state, the radial trapping frequencies were adiabatically lowered by a factor of 5 to decompress the condensate. Subsequently, an optical trapping potential was superimposed on the condensate by slowly ramping up the light intensity. After turning off the remaining magnetic fields, nearly all atoms were loaded into the large-volume optical dipole trap. The resulting peak density reached 5×10^{14} atoms/cm^3, slightly higher than the density in the magnetic trap.

The large-volume optical trap was realized by shaping the output of a Nd:YAG laser (typically 500 mW at 1064 nm) with cylindrical lenses leading to an elliptical focus with an aspect ratio of approximately 25. At the location of the condensate, the focal size was ≈ 20 μm along the tight axis resulting in an optical trapping potential with typical trap frequencies of $\nu_x = 13$ Hz axially and $\nu_y = 36$ Hz and $\nu_z = 850$ Hz transversely. The trap axis with the largest trapping frequency was oriented vertically to counteract gravity. The pancake shape of the trap, which we had recently used to create (quasi-) 2D condensates [11], provided a much larger trapping volume than our previous cigar-shaped

optical traps [9,12] and thus significantly larger condensates could be stored.

Optically trapped condensates were observed by absorption imaging on the closed $|F = 2, m_F = -2\rangle \rightarrow |F' = 3, m'_F = -3\rangle$ cycling transition at 589 nm after sudden release from the trap, using light propagating parallel to the trap laser. The ballistic expansion time was typically 30 ms, after which the vertical size of the condensate had increased by more than a factor of 100 while the horizontal expansion was less than a factor of 2. To make sure that atoms in both the $F = 1$ and the $F = 2$ manifold could be detected simultaneously, a short laser pulse resonant with the $F = 1 \rightarrow F' = 2$ transition was applied to pump all atoms into the $F = 2$ manifold. State-selective detection could be achieved by applying a magnetic field gradient of several G/cm during the free expansion of the atomic cloud, leading to a spatial separation of spin states which differ in the orientation of the magnetic moment (Fig. 1).

As a first test of the intrinsic stability of the optical trap, we investigated condensates in the $|1, -1\rangle$ state [Fig. 2(a)]. Even after 70 s of dwell time, more than 10^6 atoms remained in the condensate. A fit to the data for $t \geq 40$ yields a one-body loss rate $k_1 = 0.029\ \mathrm{s}^{-1}$ and reveals a clear deviation from an exponential decay for shorter times. Generally, the decay of the number of atoms N in a condensate can be modeled by the rate equation

$$\frac{dN}{dt} = -k_1 N - k_2 N\langle n\rangle - k_3 N\langle n^2\rangle, \qquad (1)$$

where k_2 and k_3 are the two- and three-body loss coefficients and n is the condensate density. By setting either k_2 or k_3 to zero in Eq. (1), analytical fitting functions can be derived. Fits to the data in Fig. 2(a) yield as upper limits for the decay constants $k_2 = (5.26 \pm 0.45^{+0.53}_{-1.4}) \times 10^{-16}\ \mathrm{cm}^3\,\mathrm{s}^{-1}$, which is roughly an order of magnitude larger than theoretical predictions [13], and $k_3 = (2.12 \pm 0.16^{+0.42}_{-0.69}) \times 10^{-30}\ \mathrm{cm}^6\,\mathrm{s}^{-1}$, which is in fair agreement with a previously published experimental value of $(1.1 \pm 0.3) \times 10^{-30}\ \mathrm{cm}^6\,\mathrm{s}^{-1}$ [9]. The first error we give is statistical and the second one accounts for systematic uncertainties, e.g., in the determination of the cloud sizes. Since fits with k_1 as a free fit parameter yield $k_1 < 10^{-5}\ \mathrm{s}^{-1}$, which is unrealistic under our experimental conditions, we also fitted the data with a fixed $k_1 = 0.029\ \mathrm{s}^{-1}$ and incorporated the difference in the obtained values for k_2 and k_3 in the systematic error. Our analysis indicates that the observed loss is predominantly due to three-body collisions, though contributions of two-body processes cannot be ruled out.

Condensates in the $|2, -2\rangle$ state were produced by applying a microwave pulse at 1.77 GHz to an optically trapped $|1, -1\rangle$ condensate. The power and duration of the pulse determined the fraction of atoms transferred into the $|2, -2\rangle$ state. Figure 2(b) shows a measurement of the lifetime after complete transfer into the $|2, -2\rangle$ state. The lifetime in the $|2, -2\rangle$ state is still on the order of seconds but it is significantly shorter than the lifetime of a $|1, -1\rangle$ condensate [Fig. 2(a)]. The reduced lifetime can be attributed to much larger three- and/or two-body loss rates. Using the solutions of Eq. (1) we deduce as upper

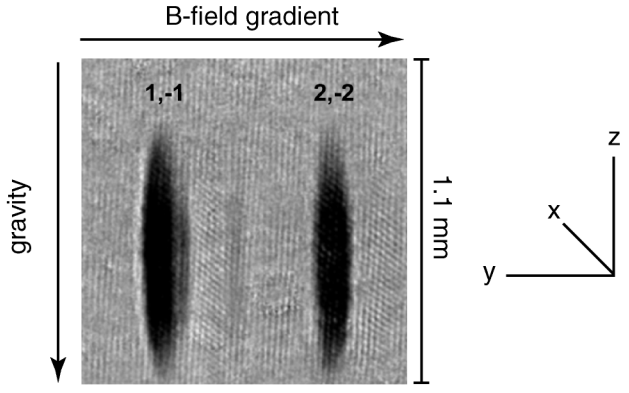

FIG. 1. Sodium condensates in the $|1, -1\rangle$ and $|2, -2\rangle$ state, 30 ms after release from the trap. After preparation of the mixture the atoms were held in the optical trap for 1 s. The horizontal separation of the spin states is due to application of a magnetic-field gradient during expansion.

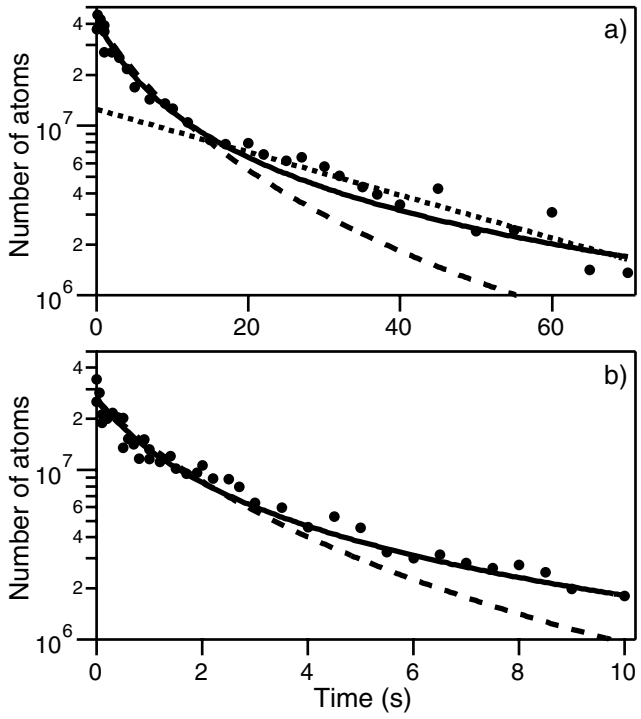

FIG. 2. Lifetime measurement of sodium BECs in the $|1, -1\rangle$ (a) and $|2, -2\rangle$ (b) states in a large-volume optical trap with an initial peak density of $\approx 5 \times 10^{14}\ \mathrm{atoms/cm}^3$. The number of atoms is determined from the measured size of the clouds after ballistic expansion. The dotted line is an exponential fit to data points with $t \geq 40$ s. The solid (dashed) lines are fits to all data points including also three-body (two-body) loss.

bounds for the loss coefficients $k_2 = (3.11 \pm 0.32^{+0.31}_{-0.36}) \times 10^{-15}\ \mathrm{cm^3\,s^{-1}}$ and $k_3 = (1.63 \pm 0.14^{+0.33}_{-0.34}) \times 10^{-29}\ \mathrm{cm^6\,s^{-1}}$. Both values are reasonably close to theoretical predictions [14,15] but the data are better fit by assuming a three-body decay. Though, at typical densities, the decay rate in the $F = 2$ state is roughly an order of magnitude larger than in the $F = 1$ state, it is still compatible with direct condensation in the $F = 2$ manifold, provided that the loss coefficients for the magnetically trapable $|2, +2\rangle$ state are similar to those for the $|2, -2\rangle$ state.

By transferring only part of the atoms into the upper hyperfine manifold, we could also observe mixtures of condensates in the $|1, -1\rangle$ and $|2, -2\rangle$ states (see Fig. 1). In the presence of small magnetic field gradients, we observed a rapid spatial separation of the two components in a time shorter than 100 ms due to the fact that the $|1, -1\rangle$ state is low-field seeking while the $|2, -2\rangle$ state is high-field seeking. During the separation, strong density modulations in both components were observed, which could be attributed to tunneling processes playing a role in the separation process [16]. Afterwards, the two components lived almost independently side by side in the trap and the individual lifetimes were not significantly affected. When we tried to compensate all stray magnetic field gradients, we still found that in steady state the two components tend to separate, i.e., we observed domains with only one component [17]. This indicates that the two states are intrinsically not miscible. While we found ^{23}Na BECs in the $|2, -2\rangle$ state as well as mixtures of $|1, -1\rangle$ and $|2, -2\rangle$ condensates to be stable for several seconds, nonstretched states in the $F = 2$ manifold as well as $F = 1$, $F = 2$ mixtures with $|m_1 + m_2| \neq 3$ decayed within several ms for typical condensate densities on the order of 10^{14} atoms/cm^3. This fast decay is probably due to (two-body) spin relaxation which is strongly suppressed in ^{87}Rb but occurs with rate constants on the order of $10^{-11}\ \mathrm{cm^3\,s^{-1}}$ in ^{23}Na [14].

A particularly interesting transition within the electronic ground state of alkali atoms is the magnetic-field insensitive transition $|F, 0\rangle \rightarrow |F + 1, 0\rangle$, often referred to as clock transition since its equivalent in cesium is used as the primary time standard. Shortly after the realization of laser cooling, the clock transition has been studied in laser-cooled [18] and optically trapped [19] atoms and today the most accurate atomic clocks are operated with laser-cooled atoms [20]. Therefore, it seems natural to investigate the use of a BEC with its significantly reduced kinetic energy for the study of the clock transition.

To observe the clock transition, we first completely transferred an optically trapped $|1, -1\rangle$ condensate into the $|1, 0\rangle$ state with a radio frequency Landau-Zener sweep. Selective driving of the $|1, -1\rangle \rightarrow |1, 0\rangle$ transition was achieved by applying a 3 G offset field which provided a large enough quadratic Zeeman shift to lift the degeneracy with the $|1, 0\rangle \rightarrow |1, +1\rangle$ transition. Subsequently, the magnetic field was reduced to a value of typically 100 mG which keeps the spins aligned and gives rise to a quadratic Zeeman shift of the clock transition of ≈ 20 Hz. The $|1, 0\rangle \rightarrow |2, 0\rangle$ transition was then excited by using a microwave pulse at 1.77 GHz with a duration between 2 and 5 ms. The fraction of atoms transferred into the $|2, 0\rangle$ state was kept below 20% in order to ensure a practically constant density in the $|1, 0\rangle$ state during the pulse. Immediately afterwards, the optical trap was turned off suddenly and the number of atoms which made the transition was detected by state-selective absorption imaging after 15–30 ms of ballistic expansion. A typical spectrum showing the number of transferred atoms as a function of microwave frequency (corrected for the calculated quadratic Zeeman shift) for a BEC with an average density of 1.6×10^{14} atoms/cm^3 is shown in Fig. 3(a). The density was determined by measuring the release energy [11] of $|1, -1\rangle$ condensates without applying a microwave pulse. The release energy E_{rel} is related to the chemical potential μ by $E_{\mathrm{rel}} = (2/7)\mu = (2/7) \times (h^2 a_{|1,-1\rangle|1,-1\rangle}/\pi m) n_o$ [21]. Here, $a_{|a\rangle|b\rangle}$ is the scattering length between two ^{23}Na atoms in states $|a\rangle$ and $|b\rangle$

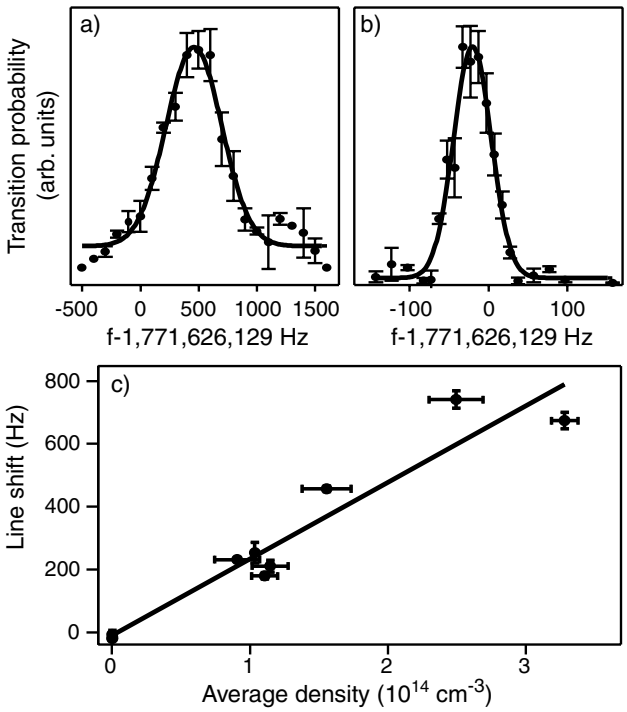

FIG. 3. Magnetic-field insensitive transition $|1, 0\rangle \rightarrow |2, 0\rangle$ in a BEC. (a) Spectrum in the trap at a mean density of 1.6×10^{14} atoms/cm^3. (b) Spectrum after 12.5 ms time of flight at a mean density of 4.3×10^{11} atoms/cm^3. The discrepancy between the center of the line and $\nu = 0$ is attributed to the uncertainty in the determination of the residual magnetic field. The solid lines are Gaussian fits. (c) Transition frequency as a function of density yielding a clock shift of $(2.44 \pm 0.25 \pm 0.5) \times 10^{-12}\ \mathrm{Hz\,cm^3}$.

($a_{|1,-1\rangle|1,-1\rangle} = 2.80$ nm), m is the ^{23}Na mass, h is Planck's constant, and n_0 is the peak density in the condensate related to the average density by $\bar{n} = (4/7)\, n_0$. The spectrum in Fig. 3(a) is significantly broadened compared to the one in Fig. 3(b), which is taken after ballistic expansion, and the transition frequency is shifted with respect to the unperturbed frequency $\nu_0 = 1,771,626,129$ Hz [18].

In the limit of weak excitation, the density-dependent shift of the clock-transition frequency is due to the difference in mean-field potential that atoms in the $|1, 0\rangle$ and $|2, 0\rangle$ state experience within a $|1, 0\rangle$ condensate. Taking into account the inhomogeneous density distribution of a trapped BEC, this leads to a line shape given by [22]

$$I(\nu) = \frac{15h(\nu - \nu_0)}{4n_0\Delta U}\sqrt{1 - \frac{h(\nu - \nu_0)}{n_0\Delta U}}, \qquad (2)$$

with

$$\Delta U = \frac{h^2}{\pi m}(a_{|2,0\rangle|1,0\rangle} - a_{|1,0\rangle|1,0\rangle}), \qquad (3)$$

where the center of the line is at $\nu_0 + 2n_0\Delta U/3h$ and the average frequency is $\nu_0 + 4n_0\Delta U/7h$. In our experiment, the line is additionally broadened and the asymmetry of Eq. (2) smeared out due to the finite width of the microwave pulse which was limited by rapid inelastic losses in the $|2, 0\rangle$ state. Therefore, we have used a (symmetric) Gaussian to fit the resonances where we have identified the fitted center frequency as the average frequency of the line. By taking spectra of the clock transition at different densities we have determined a density shift of $(2.44 \pm 0.25 \pm 0.5) \times 10^{-12}$ Hz cm^3 [Fig. 3(c)]. The first error is again statistical and the second one an estimate of systematic uncertainties originating from fitting the line with a Gaussian and from the determination of the density. Using Eq. (3) and $a_{|1,0\rangle|1,0\rangle} = 2.71$ nm [5], we determine the scattering length $a_{|2,0\rangle|1,0\rangle} = 3.15 \pm 0.05 \pm 0.1$ nm for collisions between two atoms in states $|1, 0\rangle$ and $|2, 0\rangle$.

In conclusion, we have prepared condensates in the upper $F = 2$ hyperfine manifold of the sodium ground state in a large-volume optical trap and observed a stable condensate in the high-field seeking stretched state $|2, -2\rangle$. Since only the stretched state exhibits reasonable stability, experiments with more complex spinor condensates do not seem to be possible. Furthermore, we have for the first time observed the alkali clock transition in a Bose-Einstein condensate and determined the value for the density-dependent mean-field shift. In present BEC experiments, the magnitude of the shift precludes the use of trapped condensates for precise atomic clocks. However, under circumstances where the condensate density can be drastically reduced as may be feasible in space-based experiments, the extremely low velocity spread of BECs might help improve the accuracy of atomic clocks.

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Note added.—After submission of this manuscript, a ^{23}Na BEC in the $|2, 2\rangle$ state was realized at MIT by direct evaporation in a magnetic trap [23].

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