Spinor Dynamics in an Antiferromagnetic Spin-1 Condensate

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We observe coherent spin oscillations in an antiferromagnetic spin-1 Bose-Einstein condensate of sodium. The variation of the spin oscillations with magnetic field shows a clear signature of nonlinearity, in agreement with theory, which also predicts anharmonic oscillations near a critical magnetic field. Measurements of the magnetic phase diagram agree with predictions made in the approximation of a single spatial mode. The oscillation period yields the best measurement to date of the sodium spin-dependent interaction coefficient, determining that the difference between the sodium spin-dependent s-wave scattering lengths \(a_{f=2} - a_{f=0}\) is 2.47 ± 0.27 Bohr radii.

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Atomic collisions are essential to the formation of Bose-Einstein condensates (BEC), redistributing energy during evaporative cooling. Collisions can be coherent and reversible, leading to diverse phenomena such as superfluidity [3] and reversible formation of molecules [2] in BECs with a single internal state. When internal degrees of freedom are included (as in spinor condensates), coherent collisions lead to rich dynamics [3, 4] in which the population oscillates between different Zeeman sublevels. We present the first observation of coherent spin oscillations in a spin-1 condensate with antiferromagnetic interactions (in which the interaction energy of colliding spin-aligned atoms is higher than that of spin-antialigned atoms.)

Spinor condensates have been a fertile area for theoretical studies of dynamics [3, 6, 7, 8], ground state structures [9, 10], and domain formation [11]. Extensive experiments on the ferromagnetic \(F=1\) hyperfine ground state of \(^{87}\text{Rb}\) have demonstrated spin oscillations and coherent control of spinor dynamics [3, 12]. Observation of domain formation in \(^{23}\text{Na}\) demonstrated the antiferromagnetic nature of the \(F=1\) ground state [13] and detected tunneling across spin domains [14]; no spin oscillations have been reported in sodium BEC until now. The \(F=2\) state of \(^{87}\text{Rb}\) is thought to be antiferromagnetic, but a cyclic phase is possible [15, 16]. Experiments on this state have demonstrated that the amplitude and period of spin oscillations can be controlled magnetically [4].

At low magnetic fields, spin interactions dominate the dynamics. The different sign of the spin dependent interaction causes the antiferromagnetic \(F=1\) case to differ from the ferromagnetic one both in the structure of the ground-state magnetic phase diagram and in the spinor dynamics. Both cases can exhibit a regime of slow, anharmonic spin oscillations; however, this behavior is predicted over a wide range of initial conditions only in the antiferromagnetic case [8]. The spin interaction energies in sodium are more than an order of magnitude larger than in \(^{87}\text{Rb} F=1\) for a given condensate density [3], facilitating studies of spinor dynamics.

The dynamics of the spin-1 system are much simpler than the spin-2 case [4, 15, 16], having a well-developed analytic solution [8]. This solution predicts a divergence in the oscillation period (not to be confused with the amplitude peak observed in \(^{87}\text{Rb} F=2\) oscillations).

This Letter reports the first measurement of the ground state magnetic phase diagram of a spinor condensate, and the first experimental study of coherent spinor dynamics in an antiferromagnetic spin-1 condensate. Both show good agreement with the single-spatial-mode theory [10]. To study the dynamics, we displace the spinor from its ground state, observing the resulting oscillations of the Zeeman populations as a function of applied magnetic field \(B\). At low field the oscillation period is constant, at high field it decreases rapidly, and at a critical field it displays a resonance-like feature, all as predicted by theory [8]. These measurements have allowed us to improve by a factor of three the determination of the sodium \(F=1\) spin-dependent interaction strength, which is proportional to the difference \(a_{f=2} - a_{f=0}\) in the spin-dependent scattering lengths.

The state of the condensate in the single-mode approximation (SMA) is written as the product \(\phi(r)\zeta\) of a spin-independent spatial wavefunction \(\phi(r)\) and a spinor \(\zeta = (\sqrt{\rho_+}e^{i\theta_+}, \sqrt{\rho_0}e^{i\theta_0}, \sqrt{\rho_-}e^{i\theta_-})\). We use \(\rho_-\), \(\rho_0\), and \(\rho_+\) (\(\theta_-\), \(\theta_0\), and \(\theta_+\)) to denote fractional populations (phases) of the Zeeman sublevels \(m_F = -1, 0, 1\), so that \(\sum \rho_i = 1\). The spinor’s ground state and its nonlinear dynamics may be derived from the spin-dependent part of the Hamiltonian in the single-mode and mean-field approximations, subject to the constraints that total atom number \(N\) and magnetization \(m = \rho_+ - \rho_-\) are conserved [8]. The “classical” spinor Hamiltonian \(E\) is a function of only two canonical variables: the fractional population \(\rho_0\) and the relative phase \(\theta = \theta_+ + \theta_- - 2\theta_0\). It is given by

\[
E = \delta(1-\rho_0) + \rho_0 \left(1-\rho_0 + \sqrt{(1-\rho_0)^2 - m^2} \cos \theta\right),
\]

where \(\delta = \hbar \times (2.77 \times 10^{10}\text{Hz/T}^2)B^2\) is the quadratic
Zeeman shift $\vec{B}$ with $\hbar$ the Planck constant. (The linear Zeeman shift has no effect on the dynamics.) The spin-dependent interaction energy is $c = c_2 \langle n \rangle$, where $\langle n \rangle$ is the mean particle density of the condensate and

$$c_2 = \frac{4\pi \hbar^2}{3M} (a_{f=2} - a_{f=0})$$

is the spin-dependent interaction coefficient $\left[8, 17\right]$. Here $M$ is the atomic mass. $a_{f=2}$ and $a_{f=0}$ are the s-wave scattering lengths for a colliding pair of atoms of total spin $f = 2$ and $f = 0$, respectively; Bose symmetry ensures there are no s-wave collisions with total spin of 1. If $c_2$ is positive (negative), the system is antiferromagnetic (ferromagnetic). The spinor ground state and spinor dynamics are determined by Eq. 1.

The apparatus is similar to that described previously [18]. We produce a BEC of $10^5$ $^{23}$Na atoms in the $F=1$ state, with an unobservably small thermal fraction, in a crossed-beam 1070 nm optical dipole trap. The trap beams lie in the horizontal $xy$ plane, so that the trap curvature is nearly twice as large along the vertical $z$ axis as in the $xy$ plane. By applying a small magnetic field gradient with the MOT coils (less than 10 mT/m) during the 9 s of forced evaporation, we fully polarize the BEC: all atoms are in $m_F = +1$. Conservation of spin angular momentum ensures that the magnetization remains constant once evaporation has ceased; a state with $\rho_+ = 1$ persists for the lifetime of the condensate, about 14 s.

We then turn off the gradient field and adiabatically apply a bias field $B$ of 4 to 51 $\mu$T along $\hat{x}$, leaving the BEC in the $\rho_+ = 1$ state. To prepare an initial state, we apply an rf field resonant with the linear Zeeman splitting; typically the frequency is tens to hundreds of kilohertz. Rabi flopping in the three-level system is observed [19], and controlling the amplitude and duration of the pulse can produce any desired magnetization $m$, which also determines the population $\rho_0$. The flopping time is less than 50 $\mu$s, much shorter than the characteristic times for spin evolution governed by Eq. 1. Using this Zeeman transition avoids populating the $F=2$ state, thus avoiding inelastic losses, which are much greater for $^{23}$Na than for $^{87}$Rb.

We measure the populations $\rho_i$ of atoms in the three Zeeman sublevels by Stern-Gerlach separation and absorption imaging [20]. The Stern-Gerlach gradient is parallel to the bias field $\vec{B}$, while the imaging beam propagates in the $\hat{z}$ direction. The phase $\theta$ is not measured.

To measure the ground state population distribution as a function of magnetization and magnetic field, we first set the magnetization using the rf pulse. We then ramp the field to a desired final value over 1 s, wait 3 s for equilibration, and measure the populations as above.

Figure 1(b) displays the measured ground-state magnetic phase diagram. The theoretical prediction in Fig. 1(a) is the population $\rho_0$ that minimizes the energy, Eq. 1. Such minima always occur at $\theta = \pi$ for antiferromagnetic interactions. The measurements agree well with the prediction, which is made for spin interaction energy $\epsilon = \hbar \times 20.5$ Hz (determined by spin dynamics as described below).

The first term of Eq. 1 depends on the external magnetic field and tends to maximize the equilibrium $\rho_0$ population. The second, spin dependent, term has the same sign as $c_2$ and in the antiferromagnetic case tends to minimize the equilibrium $\rho_0$ population. The phase transition indicated by the thick line in Fig. 1b arises at the point where these opposing tendencies cancel for $\rho_0 = 0$. Along the transition contour, $\rho_0$ rapidly falls to zero. By contrast, the ferromagnetic phase diagram has $\rho_0 = 0$ only at $m = 1$. In the region $B < 15$ $\mu$T and $m > 0.6$, there should be virtually no population in $m_F = 0$ for antiferromagnetic interactions, and popula-
tions up to \( \rho_0 = 0.34 \) for ferromagnetic interactions (assuming the same magnitude of \( c \)). For our equilibrium data, the reduced \( \chi^2 \) with respect to the antiferromagnetic (ferromagnetic) prediction in this region is 2 (20). This demonstrates that sodium \( F=1 \) spin interactions are antiferromagnetic, as previously shown by the miscibility of spin domains formed in a quasi-one-dimensional trap [13].

Across most of the phase diagram, the scatter in the population is consistent with measured shot-to-shot variation in atom number. This variation is 20%, implying a 8% variation in the mean condensate density according to Thomas-Fermi theory. The variance of results is not due to the magnetic field (calibrated to a precision of 0.2 \( \mu T \)), nor to residual field variations across the BEC (less than 250 \( \mu T \)). Uncertainties in setting the magnetization are obviated, as the magnetization is measured for each point as the difference in fractional populations \( m = \rho_+ - \rho_- \). Discrepancies between theory and experiment at low magnetic fields may be attributed to the inhomogeneity of the condensate. The initial relative phase is determined by prior determinations of \( c_2 \) and our knowledge of the condensate density. The initial relative phase is not the equilibrium value \( \theta = \pi \), due to our rf preparation. For a three-level system driven in resonance with both transitions, the relative phase is \( \theta = 0 \) at all times during the rf transition, as we derive from Ref. [19]. Small deviations from initial \( \theta = 0 \) could be caused by an unequal splitting between the levels, from e.g., the quadratic Zeeman shift.

The spinor dynamics are described by the Hamilton equations for Eq. (1) [8]:

\[
\dot{\rho}_0 = -\frac{2}{\hbar} \frac{\partial E}{\partial \theta} \quad \text{and} \quad \dot{\theta} = \frac{2}{\hbar} \frac{\partial E}{\partial \rho_0} \quad (3)
\]

The system is closely related to the double-well “bosonic Josephson junction” (BJJ) [21, 22] and exhibits a regime of small, harmonic oscillations and, near a critical field \( B_c \), predicted to display large, anharmonic oscillations. At \( B_c \) the period diverges (where \( \delta(B_c) = c(1 - \rho_0) + \sqrt{(1 - \rho_0)^2 - m^2 \cos \theta} \)), with \( \rho_0 \) and \( \theta \) taken at \( t = 0 \) [8]. The critical value corresponds to a transition from periodic-phase solutions of Eq. (3) to running-phase solutions. At the critical value it is predicted that the population is trapped in a spin state with \( \rho_0 = 0 \). This phenomenon is related to the macroscopic quantum self-trapping that has been observed in the BJJ [22]. However, very small fluctuations in field or density will drive \( \rho_0 \) away from 0. Observing a ten-fold increase in the period above its zero-field value would require a technically challenging magnetic field stability of better than 100 \( \mu T \).

Figure 2 plots the period and amplitude of oscillation as a function of magnetic field. An example of the oscillating populations is shown in the inset. The spinor condensate is prepared with initial \( \rho_0 = 0.50 \pm 0.01 \) and \( m = 0.00 \pm 0.02 \), and a plot of \( \rho_0 \) versus time is taken at each field value. Qualitatively, the period is nearly independent of magnetic field at low fields, with a small peak at a critical value \( B_c = 28 \mu T \), followed by a steep decline in period. The amplitude likewise shows a maximum at \( B_c \). Oscillations are visible over durations of 40 ms to 300 ms. Beyond these times, the amplitude of the shot-to-shot fluctuations in \( \rho_0 \) is roughly equal to the harmonic amplitude. This indicates dephasing due to shot-to-shot variation in oscillation frequency, probably associated with the variations in magnetic field and condensate density, rather than any fundamental damping process. At even longer times, we observe damping and equilibration to a new constant \( \rho_0 \); the damping time varies with magnetic field from 200 ms to 5 s.

For the theoretical prediction in Fig. 2 the initial value of \( \rho_0 \) and \( m \) are obtained experimentally. We treat only \( c \) and \( \theta(t=0) \) as free parameters; \( c \) is also predicted by prior determinations of \( c_2 \) and our knowledge of the condensate density. The initial relative phase is not the equilibrium value \( \theta = \pi \), due to our rf preparation. For a three-level system driven in resonance with both transitions, the relative phase is \( \theta = 0 \) at all times during the rf transition, as we derive from Ref. [19]. Small deviations from initial \( \theta = 0 \) could be caused by an unequal splitting between the levels, from e.g., the quadratic Zeeman shift.

The best fit to the data in Fig. 2a and b is obtained by using \( c = h \times (21 \pm 2) \) Hz and \( \theta(t=0) = 0.5 \pm 0.3 \) (with no other free parameters). Away from the critical field \( B_c \), agreement with theory is good. The fitted value of \( c \) implies that \( B_c = 27 \mu T \), in reasonable agreement with the apparent peak observed at 28 \( \mu T \). Our ability to observe strong variations in period near \( B_c \) is limited by density fluctuations (8%) and magnetic field fluctuations (0.2 \( \mu T \)). Near \( B_c \), typically only one cycle is visible before dephasing is complete. Such rapid dephasing can, itself, be taken as evidence of a strongly \( B \)-dependent period, as expected near the critical field.

To include the known fluctuations in density and magnetic field in our model, we perform a Monte Carlo simulation of the expected signal, based on measured, normally distributed shot-to-shot variations in values of \( c \), \( \delta \), \( m \) and \( \rho_0(t=0) \). At each value of \( B \) in Fig. 2 we generate 80 simulated time traces, with each point in the time trace determined from Eq. (3). We fit the simulated traces using sine waves and record the mean and standard deviation of the amplitude and period of the fits. The results (shaded regions in Fig. 2) show a less sharp peak in the period. The smoothing of the peak at \( B_c \) is consistent with our data.

\[1\] All uncertainties in this paper are one standard deviation combined statistical and systematic uncertainties.
of the inset to Fig. 2 is \( \langle n \rangle = 8.6 \pm 0.9 \times 10^{13} \) cm\(^{-3}\). From this we calculate \( a_{f=2} - a_{f=0} = (2.47 \pm 0.27)a_0 \), where \( a_0 = 52.9 \) pm is the Bohr radius. This is consistent with a previous measurement, from spin domain structure, of \( a_{f=2} - a_{f=0} = (3.5 \pm 1.5)a_0 \) and is smaller than the difference between scattering lengths determined from molecular levels, \( a_{f=2} = (55.1 \pm 1.6)a_0 \) and \( a_{f=0} = (50.0 \pm 1.6)a_0 \). A multichannel quantum defect theory calculation gives \( a_{f=2} - a_{f=0} = 5.7a_0 \).

Finally, we consider the validity of the spatial single-mode approximation. The SMA was clearly violated in previous work on \(^{23}\)Na and \(^{87}\)Rb. F = 1 spinor condensates where spatial domains formed. Spatial degrees of freedom decouple from spinor dynamics when the spin healing length \( \xi = 2\pi\hbar/\sqrt{2m(c^2)}n \) is larger than the condensate. From our density measurements we find typical Thomas-Fermi radii of \((9.4, 6.7, 5.7)\) \( \mu \)m. The spin healing length, based on our measurements of \( c \), is typically \( \xi = 17 \) \( \mu \)m. We therefore operate within the range of validity of the SMA. Furthermore, Stern-Gerlach absorption images show three components with identical spatial distributions after ballistic expansion, indicating that domain formation does not occur.

In conclusion, we have studied both the ground state and the spinor dynamics of a sodium \( F=1 \) spinor condensate. Both agree well with theoretical predictions in the SMA. By measuring the spin oscillation frequency at low magnetic field, we have determined the difference in spin-dependent scattering lengths. The observed peak in oscillation period as a function of magnetic field demonstrates that the spinor dynamics are fundamentally nonlinear. It also suggests the existence of the predicted regime of highly anharmonic spin oscillations at the center of this peak, which should be experimentally accessible with sufficient control of condensate density and magnetic field. Observation of anharmonic oscillations, as well as population trapping and spin squeezing effects, could be aided by a minimally destructive measurement of Zeeman populations to reduce the effects of magnetic field drifts and shot-to-shot density variations.

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