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Observation of anomalous spin-state segregation in a trapped ultra-cold vapor

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We observe counter-intuitive spin segregation in an inhomogeneous sample of ultra-cold, non-condensed Rubidium atoms in a magnetic trap. We use spatially selective microwave spectroscopy to verify a model that accounts for the differential forces on two internal spin states. In any simple understanding of the cloud dynamics, the forces are far too small to account for the dramatic transient spin polarizations observed. The underlying mechanism remains to be elucidated.

Imagine a near-ideal gas consisting of a mixture of two distinct molecular species. Apply a differential force that tends to pull the two species apart. What determines whether the two species segregate, or remain mixed? From a kinetics point of view, the question becomes whether the segregating drift velocity, a function of the differential force and the mean free path, is larger or smaller than the effective velocity of remixing diffusion. The “diffusive velocity” depends on the characteristic size of the sample. The equilibrium point of view focusses instead on whether the energetic benefit derived from separating in the differential potential outweighs the corresponding entropic cost. Again, for a given magnitude force, the ultimate outcome hinges on the size of the sample. Both points of view lead to the same quantitative criterion – is the differential potential across the sample large or small compared to the thermal energy \( k_B T \)? As an example, a mixture of gaseous oxygen and nitrogen in a room-sized box does not undergo separation under the differential force of Earth’s gravity, because the gravitational potential of the differential mass (4 amu), evaluated over a few meters, is very small compared to \( k_B T \).

In this paper, we study the segregation behavior of a gaseous mixture. Our system consists of magnetically trapped ultra-cold rubidium gas in two distinct spin states. These states can be interconverted in the presence of an applied microwave magnetic field, but in the absence of the applied drive the two states do not interconvert and can be thought of as two distinct fluids \( \mathbb{1} \). Inter-atomic interactions and differential magnetic moments give rise to a small differential potential that would tend to separate the two species, except that it is a thousand times smaller than \( k_B T \). In contrast to the above arguments we observe that the initially homogenized sample nonetheless undergoes a transient segregation.

In the first part of this paper, we describe the use of spatially selective Ramsey spectroscopy to characterize a differential potential acting on the two spin states. In the second part, we describe our observations of the atomic motion induced by the differential potential over longer timescales. The atomic motions are much too large to be described simply as arising from an acceleration from the differential potential.

The experimental apparatus includes a single magneto-optical vapor-cell trap which precools a sample of \(^{87}\text{Rb}\) atoms for transfer into a quadrupole magnetic trap. The electro-magnetic quadrupole trap coils are mounted on a servo-controlled linear track that mechanically moves the atoms to a UHV region where they are transferred into a Ioffe-Pritchard magnetic trap for further cooling via rf evaporation. Our Ioffe-Pritchard trap uses permanent magnets to produce strong radial confinement. Electromagnetic coils generate a stable and adjustable bias field and a weak confining potential in the axial direction. The trapping frequencies are \( \{7, 230, 230\} \) Hz. Typical sample sizes are \( 10^6 \) atoms at a temperature of 600 nK (factor of 2 above the Bose-Einstein condensation temperature).

The two hyperfine states of interest are \( |1\rangle \equiv |F = 1, m_J = -1\rangle \) and \( |2\rangle \equiv |F = 2, m_J = 1\rangle \). We coherently couple the two hyperfine states via a two-photon drive \( \mathbb{2} \). The drive consists of a microwave photon (\( \sim 6.8 \) GHz) and a rf photon (1-3 MHz). The sum frequency is detuned 200 Hz from the transition, and the effective two-photon Rabi frequency is 2.5 kHz. The drive is turned on for \( \sim 100 \) \( \mu \)s to transfer half of the atoms (initially all in the \( |1\rangle \) state) to the \( |2\rangle \) state (a \( \frac{\pi}{2} \) pulse). A second \( \frac{\pi}{2} \) pulse is applied after a variable delay \( \mathbb{3} \). The final population and spatial distribution in either spin state can be separately determined using absorption imaging \( \mathbb{4} \).

We determine the transition frequency between states \( |1\rangle \) and \( |2\rangle \) by varying, in each realization of the experiment, the delay time between the two \( \frac{\pi}{2} \) pulses and measuring the relative population in the \( |1\rangle \) state. Typical delay times range from 1-60 ms. The frequency of the resulting Ramsey fringes is the difference between the atomic transition and the oscillator frequency of our two-photon drive \( \mathbb{5} \). In order to study the spatial variation of the transition frequency between the two states, we break up the images into 9 bins along the axial direction and integrate the number of atoms in each bin (Fig. \( \mathbb{6} \).
There are two different mechanisms that shift the transition frequency. The first, dominant in the limit of low atomic density, is the differential Zeeman effect. The atoms are confined in the parabolic minimum of the magnetic field and each spin state individually experiences a Zeeman shift, which varies by 30 kHz from the center of the cloud to the edge of the cloud [5]. States |1⟩ and |2⟩ were selected because the Zeeman shifts cancel to great accuracy. The Breit-Rabi equation [6] predicts that the frequency splitting is first-order independent of field at 3.23 G, increasing quadratically with magnetic field about that minimum. The magnetic field in turn has a quartic shift from trap center to sample edge [5]. States |1⟩ ≈ 3.23 G, increasing quadratically with magnetic field at 3.23 G. We are unable to account for this systematic offset.

For a particular peak density we measure the axial spatial variation in frequency by binning the cloud into several sections and determining a frequency for each bin. This measurement is done at $B_{bias} = 3.23$ G to minimize the magnetic contribution to the inhomogeneity. The transition frequency vs. axial position along the cloud is shown in Fig. 1c.

The magnetic contribution to the inhomogeneity. The axial oscillation period, on the other hand, is long compared to the collision time. To further reduce the effect of axial density inhomogeneity, we use the data only "frozen" in the axial direction. To further reduce the effect of axial density inhomogeneity, we use the data only from the center bin, a region of relative uniformity. We create clouds with various peak densities and measure the transition frequency of the center bin. The slope of the frequency vs. density fit ($-0.52 \times 10^{12}$ Hz/cm$^3$) is more negative than a prediction based on photo-associative data by 20% [7]. However, it is entirely possible that our analysis of our cloud images could have systematically underestimated the cloud density by as much as 20%. More troubling is that the zero-density extrapolation, at 683467818.0(7) Hz at 3.23 G, is too high by 4.4(7) Hz. We can change the frequency shift across the cloud and nearly cancel both of the frequency shifts by oper-
ating at different a value of $B_{bias}$. The cancellation of the mean-field and magnetic frequency shifts can not be perfect because the cloud has a gaussian density profile and the magnetic field shift is essentially parabolic across the cloud. However, for a cloud with $1 \times 10^{9}$ atoms at a temperature of 500 nK there will be less than 1 Hz shift across the cloud at $B_{bias} = 2.83$ G (Fig. 2).

![FIG. 2. The transition frequencies at different positions along the cloud are plotted as a function of $B_{bias}$. The cloud has a peak density of $4.0 \times 10^{13}$ cm$^{-3}$. Offset frequency is defined as the transition frequency - 6834682610.9 Hz. The frequency shift at 3.23 G (negligible magnetic contribution to inhomogeneity) has a gaussian shape. At $B_{bias} = 3.37$ G the magnetic shift and the mean-field shift are in the same direction and therefore the inhomogeneity is across the cloud. However, for a cloud with $1 \times 10^{9}$ atoms at a temperature of 500 nK there will be less than 1 Hz shift across the cloud at $B_{bias} = 2.83$ G (Fig. 2).

Conversely, if we operate at $B_{bias} > 3.23$ G, the curvature of the magnetic contribution to the frequency inhomogeneity adds with the same sign to that from the density contribution, and we see enhanced total frequency shift across the cloud (Fig. 3). Near the center of the cloud, the frequency shift is parabolic with displacement from the center, and the curvature of the differential potential can be conveniently characterized by

$$\nu_{diff} = 1/2\pi \sqrt{\hbar/m} \times (d^2 \nu_{12}/dz^2)$$

(2)

where $m$ is the mass of $^{87}$Rb and $\nu_{12}$ is the transition frequency. $\nu_{diff}$ may be thought of as the axial motional frequency of a hypothetical test atom, in state $|2\rangle$, under the influence of only the differential magnetic and mean-field forces. For the data shown in Fig. 3, measured $\nu_{diff}$ ranges from 0.15 Hz for $B_{bias} = 3.37$ G, down through 0.1 Hz for $B_{bias} = 2.83$ G.

We have used spectroscopic methods to characterize the spatial dependence of the differential potential experienced by states $|1\rangle$ and $|2\rangle$ in our trap. The data are quantitatively accounted for by a simple model involving magnetic and mean-field potentials. We now describe the observed mechanical effects that the gradients of these potentials have on the distribution of the relative atomic densities of states $|1\rangle$ and $|2\rangle$.

In the twin-pulse spectroscopy described so far, the first pulse sets up a coherent superposition of the two states; the delay time between the two pulses allows the relative internal phase, $\phi$, in the coherent superposition $(|1\rangle + e^{i\phi}|2\rangle)$ to evolve under the inhomogeneous potential, and the second pulse then converts the spatial pattern in $\phi$ into a pattern of relative density for ready imaging. In the experiments described below, the second $\pi/2$ pulse is omitted, and the delay between the first pulse and the density imaging is increased to allow time for the atoms to move under the influence of the developing spatial gradient in $\phi$. The patterns in the density distribution which we image must result from the physical displacement of atoms during the period after the $\pi/2$ pulse.

The results of this class of experiment are summarized in Fig. 3. Starting around 50 ms after the $\pi/2$ pulse, the atoms in the $|1\rangle$ state are observed to move outward along the trap axis, while the atoms in the $|2\rangle$ state move inward. Maximum separation is realized at about 100 ms, then the two clouds relax back to their original, well-overlapped distribution by around 200 ms. No further evolution is observed. The spin segregation is dependent on the curvature of the differential potential. For $\nu_{diff} < 0.05$ Hz (Fig. 3(ii)), no separation is observed above the imaging noise. For $\nu_{diff} > 0.18$ Hz (Fig. 3(iv)), we observe what we call “higher-order” effects, with the relative spin-density developing multiple stripes.
FIG. 3. (a) Spin segregation appears in the spatial distribution of atoms in the $|1\rangle$ state at various times after the $\pi$ pulse. $|1\rangle$ state atoms move outward, then relax back to their initial condition. Initial cloud conditions are peak total density $1.8 \times 10^{14} \text{cm}^{-3}$, $T=630 \text{ nK}$, $\nu_{diff} = 0.1 \text{ Hz}$. (b) Same as for (a), but showing the $|2\rangle$ state distribution. $|1\rangle$ and $|2\rangle$-state motions are complementary, and leave the total density profile unchanged. (c) $|1\rangle$-state axial density profile imaged at times after the $\pi$ pulse, listed on the left column. For all columns, $T = 600 \text{ nK}$. $\nu_{diff}$ values are listed at the bottom of each column. The first column of cross sections has an imaginary $\nu_{diff}$ corresponding to negative curvature of the frequency. Initial density (in $10^{15} \text{ cm}^{-3}$) and $B_{bias}$ (in Gauss) are for each column (i) 1.82, 2.99 (ii) 1.82, 3.12 (iii) 1.82, 3.52 (iv) 1.82, 3.79 (v) 0.254, 3.35 (vi) 1.16, 3.31 (vii) 2.05, 3.27 Columns (i)-(iv) are at fixed density and progressively higher $\nu_{diss}$, showing turn-on of effect with increasing $\nu_{diss}$. Columns (v)-(vii) are at fixed $\nu_{diss}$ and at progressively higher density, showing turn-on of segregation effect with density.

We find the segregation to be startling. While the mean-field and magnetic forces can combine to provide a small buoyant force that tends to push the $|1\rangle$ atoms out towards the lower density regions of the cloud, the magnitude of the observed effect is entirely disproportionate. For $\nu_{diff} = 0.1 \text{ Hz}$, for instance, the total frequency shift $\Delta \nu_2$ from center to edge of cloud is 6 Hz. This should be compared to $k_B T / h \approx 10 \text{ kHz}$. The criterion for separation discussed in the introduction is thus clearly not satisfied. This objection would be surmounted if the initially well-homogenized and non-condensed populations in each spin-state nonetheless moved collectively (the relevant inequality would then be $N_{atoms}(\Delta \nu_2 h) \gg kT$ rather than $\Delta \nu_2 h \ll kT$) [10]. One would still need to account, however, for the rate of segregation. If the atoms move ballistically in the differential potential, the minimum time for the peak segregation to develop should be at least $1/4\nu_{diff}$ [10]. But the observed segregation times are at least 10 times faster.

As seen in Fig. 3, to induce segregation one needs not only a sufficiently large value of $\nu_{diff}$ but independently one needs adequate density. It is possibly significant that the segregation seems to disappear when the density gets so low that the cloud is no longer collisionally thick in the axial direction. While the observed segregation rate is fast compared to $4 \nu_{diff}$, it is slow compared to $4 \nu_z$, where $\nu_z$ is the axial trap frequency. That is, the random thermal speed of the atoms, if it were selectively directed, is more than adequate to move the atoms the required distance. We speculate that as the internal phase $\phi$ develops a spatial gradient, it somehow acts as a “switch” to control velocity-changing collisions such that atoms in the $|2\rangle$ state are preferentially scattered inward and $|1\rangle$ atoms outward.

Finally, the observation of non-diffusive evolution of spin populations compels a comparison with the spin-wave phenomenon observed [11] and discussed extensively [12] in the 1980s. We understand the earlier work to be an effect of the transverse spin. The present work describes evolution of the relative population of energy eigenstates, a longitudinal spin effect. Additional study may lead to connections between the two spin effects.

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[3] We image the atoms in the $|1\rangle$ state by transferring them to the $|F = 2, m_F = -2\rangle$ state using a microwave frequency adiabatic rapid passage. The antitrapped $|F = 2, m_F = -2\rangle$ atoms expand rapidly in the radial direction for 3-5 ms. We then probe the atoms on the $|F = 2, m_F = -2\rangle \rightarrow |F' = 3, m_F' = -3\rangle$ transition. To image the atoms in the $|2\rangle$ state, we apply a $\pi$ pulse, converting the atoms in the $|2\rangle$ state to the $|1\rangle$ state and vice versa, and immediately proceed as with state $|1\rangle$.


[5] We arbitrarily define the “edges” of the cloud to correspond to $\pm 1.5 \times HWHM$.

[6] From the Breit-Rabi equation we expand the relative Zee- man shift about the minimum: $\nu = 6834678113.59(2) \text{ Hz} + 431.35957(9) \text{Hz}/G^2 \times (B - 3.228917(3) \text{ G})^2$. These values and associated uncertainties are derived from the zero-field hyperfine splitting $6834682610.90434(3)$ and the nuclear g-factor $0.0009951414(10)$ [13].


[8] We use a density weighted frequency curvature defined by $\langle \frac{d^2 \phi}{dz^2} \rangle = \int_{-\infty}^{\infty} n(z) \frac{d^2 \phi}{dz^2}(z)dz / \int_{-\infty}^{\infty} n(z)dz$ where $n(z)$ is the axial gaussian density profile.

[9] In the absence of applied microwave, spin population does not spontaneously interconvert. The energy released by a single atom converting from state $|2\rangle$ to state $|1\rangle$, if it were thermalized among the sample of $10^6$ atoms, would result in a 50% increase in sample temperature. This warming is not observed. The spin populations must thus each independently obey a mass continuity equation.

[10] Indeed exactly such behavior was observed in the collective motion of a two-level BEC [14]. The observed condensate segregation rate was in good agreement with the corresponding value of $\nu_{diff}$. That is, the condensate's segregation was at least 10 times slower than the present...
non-condensate effect.


