Roton softening and supersolidity in Rb spinor condensates

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Superfluids with a tendency towards periodic crystalline order have both a phonon and roton like spectrum of collective modes. The softening of the roton spectrum provides one route to a supersolid. We show that roton softening occurs in \textsuperscript{87}Rb spinor condensates once dipolar interactions and spin dynamics are taken into account. By including the effects of a quasi-two-dimensional geometry and rapid Larmor precession, we show a dynamical instability develops in the collective mode spectrum at finite wavevectors. We construct phase diagrams showing a variety of instabilities as a function of the direction of the magnetic field and strength of the quadratic Zeeman shift. Our results provide a possible explanation of current experiments in the Berkeley group Phys. Rev. Lett. 100:170403 (2008).

The experimental elusive supersolid state has long been of great interest as an exotic quantum state of matter. Such a state offers the possibility of periodic crystalline order breaking translation invariance familiar from solids and long range phase coherence breaking global gauge invariance familiar from superfluids coexisting in the same material [1, 2, 3, 4, 5, 6, 7, 8]. Theoretical interest in the supersolid state has a long history dating back to early studies of superfluidity in \textsuperscript{4}He. Landau suggested that the excitation spectrum consists of two parts: a sound like long wavelength mode and a roton spectrum \(\varepsilon(p) = \Delta + (p - p_0)^2/2m\) for \(p \approx p_0\) indicative of a tendency towards crystalline order. In a system with both a phonon and roton spectrum, the softening of the roton gap where \(\Delta\) approaches zero provides one means of possibly realizing a supersolid [7, 8]. Recent experiments suggesting the existence of the supersolid phase in \textsuperscript{4}He are a subject of intense debate [9, 10, 11, 12].

In this paper, we demonstrate roton softening takes place in quasi-two-dimensional \(F = 1\) ferromagnetic condensates such as ultracold \textsuperscript{87}Rb once dipolar interactions and spin dynamics are taken into account. Pattern formation due to dipolar interactions is well known in condensed matter physics [13], but spinor condensates such as \textsuperscript{87}Rb present several novel effects without condensed matter analogs. In addition to dipolar interactions, the spin degrees of freedom also experience competing interactions including spin dependent contact interactions and the quadratic Zeeman shift [14]. Moreover, the spin dynamics of rapid Larmor precession and confinement to a two-dimensional geometry play an important role in modifying the effective dipolar interaction. Several works have previously analyzed the role of dipolar interactions in polar molecules [14, 15, 16, 17, 18, 19] and spinor condensates [20, 21, 22]. However, these studies do not take into account the combined effects of dynamical spin degrees of freedom, rapid Larmor precession, and reduced dimensionality.

We show that for an initially uniform ferromagnet, the excitation spectrum has roton like parts for both spin and density branches (generally each branch involves both spin and density degrees of freedom, so we define them by their behavior in the long wavelength limit) with the roton on the spin branch becoming imaginary at finite wavevectors. Macroscopic occupation of such roton excitations should lead to a state which breaks global gauge invariance, spin rotational and translational symmetries. Hence such instability suggests the likely formation of a supersolid phase. This instability has a simple physical origin in terms of lowering the classical dipolar interaction energy of a uniform ferromagnet through periodic modulation of the magnetization \(\mathbf{F}\) as shown in Fig. 1. However, the modulation direction is different for the longitudinal component along the direction of the magnetic field \(\mathbf{B}\) versus the perpendicular components because only the latter precesses around the external magnetic field (see discussion below). Different components of the spin are conjugate variables which couple to each other through the commutation relation \([\mathbf{F}^i, \mathbf{F}^j] = i\epsilon^{ijk}\mathbf{F}^k\), a rich variety of instabilities leading to possible striped and checkerboard supersolid phases can arise. This simple physical picture also gives a rough estimate for the length scale \(\lambda\) of the instability by equating the kinetic energy cost with the gain in dipolar interaction energy \(1/2m\lambda^2 + 1/2md^2 = 2\mu g_d n_0/3\) where \(m\) is the mass, \(g_d\) gives the strength of dipolar interactions, \(n_0\) is the density, and \(d\) is the thickness of the condensate. With typical experimental parameters \(d = 2\mu m\) and \(g_d n_0 = 10\) Hz this gives an estimate of \(\lambda \approx 10\mu m\). The length scale and structure of unstable modes agree quantitatively with current experiments on dipolar effects in spinor condensates [22].

RESULTS

We consider a quasi-two-dimensional spinor condensate as shown in Fig. 2. The unit vectors \(\hat{x}, \hat{y}\) are in the plane while \(\hat{z}\) is out of the plane. A uniform magnetic field points along \(\mathbf{B}\) in the \(\hat{n}, \hat{x}\) plane at an angle \(\alpha\) with respect to \(\hat{n}\). The initial condensate is either prepared with a uniform magnetization or with a non-uniform spiral spin texture with magnetization winding along \(\hat{\kappa}\) and
FIG. 1: We consider a quasi-two-dimensional spinor condensate with magnetic field \( \hat{B} \) along the \( \hat{z} \) direction in the plane, and uniform magnetization \( \hat{F} \). Larmor precession of \( \hat{F} \) about \( \hat{B} \) means \( \hat{F} \) spends half the time along the \( \hat{1} \) direction in the plane and half the time along the \( \hat{2} \) direction out of the plane. Magnetization fluctuations \( \delta \hat{F} \) with periodic modulation can lower the effective dipolar interaction energy and drive an instability towards a possible supersolid state. \( \delta \hat{F} \) along \( \hat{3} \) (parallel to \( \hat{B} \)) favor modulation along \( \hat{1} \) (perpendicular to \( \hat{B} \)). In contrast, \( \delta \hat{F} \) along \( \hat{1} \) or \( \hat{2} \) (perpendicular to \( \hat{B} \)) favor modulation along \( \hat{3} \) (parallel to \( \hat{B} \)). All components couple to each other through canonical commutation relations and give rise to a variety of instabilities towards striped phases modulated along \( \hat{1} \) or \( \hat{3} \) as well as checkerboard phases modulated along \( \hat{1} \) and \( \hat{3} \).

wavevector \( |\kappa| \). The Hamiltonian is given by

\[
\mathcal{H} = \int d^3x \Psi_\alpha^\dagger \left[ -\frac{\nabla^2}{2m} - \mu + B_0 \hat{B} \cdot \hat{F} + q \left( \hat{B} \cdot \hat{F} \right)^2 \right] \Psi_\alpha \\
+ \int d^3x \left[ \frac{g_0}{2} : \Psi^\dagger_\alpha \Psi^\dagger \Psi^\dagger_\beta \Psi + \frac{g_0}{2} : \Psi_\alpha \Psi^\dagger \Psi_\beta \Psi^\dagger \right] \\
+ \int d^3x d^3\tilde{x} \frac{g_d}{2} h_{3D}^{ij}(\tilde{x} - \tilde{x}') : \Psi^\dagger_i \hat{F}^j \Psi^\dagger_\alpha \Psi^\dagger_\beta \Psi^\dagger_\gamma \Psi^\dagger_\delta \Psi^\dagger_\epsilon \Psi^\dagger_\zeta \Psi^\dagger_\eta \Psi^\dagger_\xi \Psi^\dagger_\nu \Psi^\dagger_\sigma \Psi^\dagger_\tau \Psi^\dagger_\upsilon : \\
\tag{1}
\]

where \( : \) denotes normal ordering. We denote \( \Psi_\alpha \) with \( \alpha = 1, 2, 3 \) as annihilation operators for \( F = 1 \) bosons with mass \( m \) and \( \hat{F} \) hyperfine spin operators with \( \hat{F}_{ijk} = -i \epsilon_{ijk} \). Throughout, we use a matrix notation with suppressed indices where \( * \), \( T \), and \( \dagger \) denote the complex conjugate, transpose, and the conjugate transpose, respectively. For example, \( \Psi_\alpha (\Psi^\dagger_\alpha) \) is a column (row) vector while \( \hat{F}^T \) is a matrix.

The chemical potential \( \mu \) is a Lagrange multiplier controlling the density \( n_{3D} = \langle \Psi^\dagger \Psi \rangle \) and we work with fixed longitudinal magnetization \( n_{3D} f_B = \langle \Psi^\dagger \hat{B} \cdot \hat{F} \Psi \rangle \). The magnetic field induces Larmor precession about \( \hat{B} \) at a frequency \( B_0 \) and a quadratic Zeeman shift \( q \). With typical magnetic fields \( B \) of zero up to hundreds of mG, \( q = 70 \) Hz \( G^{-2} B \) ranges from zero to tens of Hz. AC Stark shifts can further tune \( q \), in particular to negative values. A harmonic trapping potential along \( \hat{n} \) confines the condensate to a thickness \( d_n \). We take typical values of \( B_0/2\pi = 115 \) kHz and \( d_n = 2 \) \( \mu \)m.

The spin independent and spin dependent contact interaction strengths are given by \( g_0 = 4\pi \hbar^2 a_0/m \), \( g_d = 4\pi \hbar^2 (a_0 - a_2)/3m \) in terms of the s-wave scattering lengths \( a_F \) for two atoms colliding with total angular momentum \( F \). For \( ^8\text{Rb} \), \( a_0 = 101.8a_B \) and \( a_2 = 100.4a_B \) where \( a_B \) is the Bohr radius giving positive \( g_0 \) and ferromagnetic interactions. The dipolar interaction strength is given by \( g_d = \mu_B g_F \mu_B^2 \) where \( \mu_0 \) is the vacuum permeability, \( g_F \) is the Landé g-factor, and \( \mu_B \) is the Bohr magneton. The dipolar interaction tensor is given by

\[
h^{ij}_{3D}(\tilde{x}) = |\tilde{x}|^{-3} \left[ \delta^{ij} - 3\tilde{x}^i \tilde{x}^j \right], \quad h^{ij}_{3D}(\tilde{k}) = -\frac{4\pi}{3} \left[ \delta^{ij} - 3\tilde{k}^i \tilde{k}^j \right] \\
\tag{2}
\]

in real and momentum space with the Fourier transform regularized as in Methods:Dipolar Interaction. For typical peak three-dimensional densities of \( n_{3D} = 2.2 \times 10^{14} \) cm\(^{-3} \) the interaction strengths are \( g_0 n_{3D} = 1.7 \) kHz, \( g_d n_{3D} = 8 \) Hz, and \( g_d n_{3D} = 10 \) Hz.

Notice the clear separation of energy scales for the above Hamiltonian. The quadratic Zeeman energy, spin dependent contact interaction, and dipolar interaction all compete at the lowest energies on the order of tens of Hz. The spin independent contact interaction and harmonic trapping along \( \hat{n} \) are both hundreds of Hz implying the density and out of plane dynamics are effectively frozen. At the highest energies, Larmor precession occurs at hundreds of kHz implying one should average over this rapid precession for the low energy properties.

**Dipolar Interaction**

As illustrated in Fig. 2, the bare dipolar interaction is modified by spatial and time averaging due to confinement along \( \hat{n} \) and Larmor precession, respectively. Here we briefly summarize the resulting effective dipolar interaction that emerges with more details in Methods:Dipolar Interaction.

We first consider the effect of confinement along \( \hat{n} \). Since the thickness of the condensate \( d_n \) and the spin

FIG. 2: Schematic of experiment with unit vectors \( \hat{x} \), \( \hat{y} \) (\( \hat{n} \)) in the plane (out of the plane). The magnetic field \( \hat{B} \) (spiral wavevector \( \hat{k} \) makes an angle \( \alpha \) (\( \beta \)) with respect to \( \hat{n} \) (\( \hat{x} \)). The effective dipolar interaction takes into account Larmor precession of the magnetization \( \hat{F} \) and the confinement of the condensate along \( b \) to thickness \( d_n \).
healing length $\xi$ are comparable $d_n, \xi \sim 2 \mu m$, we assume the condensate is frozen along $\hat{n}$ and take
\[ \Psi_x \rightarrow \sqrt{\rho(x_n)} \Psi_x \tag{3} \]
where $\Psi_x$ on the left-hand (right-hand) side is a three-dimensional (two-dimensional) field. Here $\rho(x_n)$ is the normalized to $\int dx_n \rho(x_n) = 1$ and for definiteness we take a gaussian form $\rho(x_n) = \exp(-x_n^2/2d_n^2)/\sqrt{2\pi d_n^2}$ with $x_n$ the coordinate along $\hat{n}$. Integrating over $x_n$ in Eq. 1 gives a two-dimensional Hamiltonian with
\[ \rho \]
with the constant $C = 1/\sqrt{2}$ is determined by normalization. However, the dipolar interaction tensor $\hat{h}_{2D}^2(\Delta x) \rightarrow \hat{h}_{2D}^2(\Delta x)$ is modified appreciably.

In addition to confinement along $\hat{n}$, rapid Larmor precession also modifies the dipolar interaction tensor. In experiments, the strong uniform component of the magnetic field causes precession of magnetization along $\vec{B}$ while a weak gradient along $\hat{k}$ induces a spiral order of the magnetization [23]. We take this into account by going to a co-moving frame via the unitary transformation
\[ \Psi_x \rightarrow R(t, \vec{x}) \Psi_x, \quad R(t, \vec{x}) = \exp \left[ i \theta(t, \vec{x}) \vec{B} \cdot \vec{F} \right] \tag{4} \]
with $\theta(t, \vec{x}) = -B_0 t + |\vec{k}| \vec{k} \cdot \vec{x}$ where $B_0$ is the uniform component of the magnetic field and $|\vec{k}|$ is the spiral wavevector. Applying this unitary transformation simply yields the substitutions $B_0 \rightarrow -i|\vec{k}|/m \vec{k} \cdot \nabla$, $q \rightarrow q + |\vec{k}|^2 / 2m$ which arise from the Berry’s phase and kinetic energy terms. Time averaging over the rapid precession then yields a modified dipolar interaction tensor $\hat{h}_{2D}^2(\Delta x) \rightarrow \hat{h}_{2D}^2(\Delta x).

We focus on the resulting two-dimensional time-averaged interaction in momentum space given by
\[ \hat{h}_{2D}^2(\vec{k}) = -\frac{4\pi}{3} \left[ \hat{h}_{2D}^2(\vec{k}) P_{ij}^B \hat{h}_{2D}^2(\vec{k}) P_{ij}^B \right] \tag{5} \]
with the momentum independent projection operators
\[ P_{ij}^B = \hat{B}^i \hat{B}^j, \quad P_{ij}^\perp = \delta_{ij} - \hat{B}^i \hat{B}^j \tag{6} \]
and recall $\vec{F}_{ij} = -ie\epsilon_{ijk}$ while the functions $\hat{h}_{2D}^2(\vec{k})$, $\hat{h}_{2D}^2(\vec{k})$, $\hat{h}_{2D}^2(\vec{k})$ carry the momentum dependence and are given explicitly in Eqs. 24, 25, 26 of Methods:Dipolar Interaction. Notice the spin dependent part carrying the $i, j$ indices only depends on $\hat{B}$ and not $\vec{k}$. Essentially, time-averaging over the fast Larmor precession selects a preferred direction in spin space along $\hat{B}$. Compare this to the bare dipolar interaction of Eq. 2 where the spin and momentum dependence are not separable. In real space, the bare dipolar interaction (without averaging over Larmor precession) favors spins aligned head-to-tail and anti-aligned side-by-side. In particular, small distortions of a mean-field condensate with uniform magnetization in the plane are energetically unfavorable since they destroy the favorable head-to-tail order already present.

These considerations change once rapid Larmor precession is included. Precession causes the energetically favorable head-to-tail order to rotate into the energetically unfavorable side-by-side order half of the time. Heuristically, this gives rise the instabilities of the effective dipolar interaction. Fig. 1 illustrates the fluctuations $\delta \vec{F}$ for the magnetization $\vec{F}$ that lower the effective dipolar energy with $\vec{B}$ in the plane. $\delta \vec{F}$ parallel to $\vec{B}$ favors modulation perpendicular to $\vec{B}$. This is as expected even for the bare dipolar interaction which favors head-to-tail alignment (uniform parallel to $\vec{B}$) and side-by-side anti-alignment (modulation perpendicular to $\vec{B}$). However, both components of $\delta \vec{F}$ perpendicular to $\vec{B}$ favor modulation parallel to $\vec{B}$. For $\delta \vec{F}$ in the plane and perpendicular to $\vec{B}$, this can be understood in terms of the bare dipolar interaction favoring head-to-tail alignment (uniform perpendicular to $\vec{B}$) and side-by-side anti-alignment (modulation parallel to $\vec{B}$). Larmor precession affects $\delta \vec{F}$ out of the plane and perpendicular to $\vec{B}$ the most as precession rotates this component into the plane half the time. This leads to the same type of behavior as $\delta \vec{F}$ in the plane and perpendicular to $\vec{B}$: uniform perpendicular to $\vec{B}$ and modulation parallel to $\vec{B}$. Compare this to arguments appealing to the bare dipolar interaction for this component which suggest modulation along both directions since $\delta \vec{F}$ out of the plane gives the energetically unfavorable aligned side-by-side arrangement.

Collective Modes

We now turn to quantitative analysis of the collective mode spectrum focusing on the two-dimensional time-averaged case. Starting from mean field solutions with uniform magnetization, we study the collective mode spectrum describing its small fluctuations. For details see Methods:Collective Modes. Recall we transformed to a frame co-moving with possible spiral order so that co-moving frame uniform states describe both lab frame uniform and spiral states.

We take $\hat{B}$ as the quantization axis and parametrize
\[ \Psi_x = \sqrt{n_x} \left[ e^{i\eta \phi} \sin(\phi + i \chi) e^{i\eta \nu} \sin(\phi + i \chi) \right] \tag{7} \]
with $n$ the two-dimensional density, $\eta$ the global phase, $\rho, \chi, \nu$ controlling the magnitude of the magnetization, and $\phi$ the orientation of the transverse magnetization. We take $\Psi_x = \Psi$ independent of $\vec{x}$ and show in Methods:Collective Modes these mean-field states only depend on
\[ Q = \frac{q}{2g_\perp n_{3D} C}, \quad g_\perp = g_s - g_d \hat{h}_{2D}^2(0) \tag{8} \]
where we use $n_{3D} = np(0)$.
To study collective modes, we take $\Psi_x = \Phi + \delta \Psi_x$ and find the linearized equations of motion for the fluctuations $\delta \Psi_x$ given by

$$i\hbar \begin{bmatrix} \delta \Phi_x \\ \delta \Phi^*_x \end{bmatrix} = \begin{bmatrix} M_x & N_x \\ -N_x^* & -M_x^* \end{bmatrix} \begin{bmatrix} \delta \Phi_x \\ \delta \Phi^*_x \end{bmatrix}$$

(9)

with $M_x$ and $N_x$ given in Methods: Collective Modes. The ansatz $\delta \Phi_x(t) \sim e^{i\omega t}$ gives an eigenvalue equation for the excitation energies $\omega_x$. Analysis of $\omega_x$ gives the spectrum for small fluctuations above the mean field solution. In particular, imaginary $\omega_x$ indicates a dynamical instability where such fluctuations grow exponentially.

The above collective mode analysis describes the general case for arbitrary $f_B, |\kappa| \neq 0$. Here we focus on the analytically tractable case of zero longitudinal magnetization and no spiral order $f_B, |\kappa| = 0$ and later discuss the effects of finite $f_B, |\kappa|$. Instead of the three component complex field $\Psi_x$, it will be convenient to use the six component real field

$$\Phi_x = \begin{bmatrix} n_x, \eta_x, \rho_x, \nu_x, \phi_x, \chi_x \end{bmatrix}^T$$

(10)

where Eq. [4] defines $\Psi_x$ as a function of $\Phi_x$. For the collective mode analysis, we take

$$\delta \Phi_x = \frac{\partial \Phi}{\partial \Phi} \delta \Phi_x$$

(11)

with the derivative evaluated at the mean-field parameters and derive equations of motion for $\delta \Phi_x$ from Eq. [9]. The $\phi$ and $\chi$ modes decouple from the $n, \eta, \rho, \nu$ modes. Since $\phi$ controls the orientation of the transverse magnetization and $\chi$ controls the magnitude of the longitudinal magnetization, they form a conjugate pair of variables which we denote as the spin mode. The other four degrees of freedom form two pairs of conjugate variables which we denote as the charge and magnetization modes.

At long wavelengths, we find for the spin, charge, and magnetization modes

$$\omega_k^c = \sqrt{[1 + Q][\alpha(Q - 1)g_s + (3 - Q)g_{\perp}(\alpha)]/\rho(0)}$$

(12)

$$\omega_k^s = \sqrt{2[g_0 + g_\perp(Q - 1)/\rho(0)]}$$

(13)

$$\omega_k^q = 2[1 - Q^2g_{\perp}(\alpha)]$$

(14)

where $\alpha(\theta)$ is the angle between $\hat{B}$ and $\hat{n}, (\hat{k}$ and $\hat{d})$. In the above, we write out the explicit $\alpha$ dependence of Eq. [8] as

$$g_{\perp}(\alpha) = g_s - \frac{2\pi}{3}g_d \left[1 - 3\cos^2(\alpha)\right]$$

(15)

The spin mode scales with $\sqrt{k}$ and is highly anisotropic due to the long-ranged and anisotropic nature of dipolar interactions. The spin mode can develop a roton minimum indicating a tendency towards crystalline order and has a strong dependence on $\alpha$. The charge mode scales with $k$ and describes phonon excitations of the superfluid. Notice the superfluid velocity depends primarily on the spin-independent contact interaction $g_0$. The magnetization mode is gapped and describes fluctuations in the magnitude of $F$. Notice the gap can vanish as a function of $Q$ and $\alpha$.

We plot a representative collective mode spectrum in Fig. 3 illustrating the spin and charge modes for $\alpha = 0.24\pi, \theta = \pi/2$ and $q = q_c + \delta q$ with $q_c = -0.87$ and $\delta q = 1.0, 0.5, 0.0, -0.5$ Hz from top to bottom.

![Fig. 3: Collective mode spectrum for $\alpha = 0.24\pi, \theta = \pi/2$ and $q = q_c + \delta q$ with $q_c = -0.87$ and $\delta q = 1.0, 0.5, 0.0, -0.5$ Hz from top to bottom.](image)

We then analyze the imaginary part of the collective mode spectrum in momentum space as in the left of Fig. 4. This indicates the structure of instabilities and we construct the phase diagrams for the spin and charge mode in Fig. 2. The uniform ferromagnet mean-field solution exists within the green lines and the polar state with no magnetization is outside. Dynamical instabilities above the red line imply the mean-field state is unstable to small fluctuations. In general, spin mode instabilities are stronger with larger $\text{Im} \omega_x^c$ than charge mode instabilities except near the boundaries to the polar state where they are comparable.

Notice the boundaries to the polar state intersect near $\alpha_c = 0.35\pi$ indicating the uniform ferromagnet is not stable for any $q$ even at the mean-field level. This is analogous to the magic angle effect familiar from NMR [26]. At this magic angle, $g_{\perp}(\alpha_c) = 0$ and rapid precession of the spins cancels out the combined effect of dipolar and spin dependent contact interactions which stabilize the
ferromagnet.

In the phase diagrams, $D_\alpha$ ($C_\alpha$) denotes regions of unstable modes disconnected from (connected to) the origin $k = 0$ and $R$ indicates a ring of unstable modes. There are several trends to notice in the phase diagrams. The uniform ferromagnet is stable only for $\alpha < \pi/4$ away from the negative $q$ boundary. For $\alpha < \pi/4$ near the negative $q$ boundary the spin mode is unstable along the direction perpendicular to $\hat{B}$. This suggests an instability towards a striped phase modulated along this direction. From Fig. 1 we see it is driven by transverse fluctuations $\delta \vec{F}$ of the magnetization $\vec{F}$ which modulation perpendicular to $\hat{B}$. There is also an area of ring instabilities near the lower left hand corner. Generally, modes perpendicular to $\hat{B}$ are more unstable than modes parallel to $\hat{B}$.

For $\pi/4 < \alpha < \alpha_c$, the primary instability is in the spin mode although the charge mode is also unstable in limited regions near the polar state. Again, transverse fluctuations $\delta \vec{F}$ drive the spin mode instability towards modulation perpendicular to $\hat{B}$ near the negative $q$ boundary to the polar state. However, longitudinal fluctuations $\delta \vec{F}$ drive the spin mode instability towards modulation parallel to $\hat{B}$ near the positive $q$ boundary. This suggests striped phases perpendicular and parallel to $\hat{B}$ near the negative and positive $q$ boundaries, respectively. In between these boundaries, both transverse and longitudinal fluctuations $\delta \vec{F}$ are important. This indicates a tendency towards a checkerboard phase modulated along both directions. Above the magic angle $\alpha_c < \alpha$, both types of instabilities are important throughout indicating a tendency towards checkerboard phases.

The above analysis for $f_B, |\kappa| = 0$ also holds qualitatively for general parameters $f_B, |\kappa| \neq 0$. As an illustration, we plot in Fig. 5 the imaginary part of $\omega_\kappa$ for the experimentally relevant parameters $\alpha = \pi/2$, $q = 0$, $\beta = 0$ in four cases: $f_B = 0$ and $|\kappa| = 0$ (top left), $f_B = 0.8$ and $|\kappa| = \kappa_{\text{max}}$ (bottom left), $f_B = 0$ and $|\kappa| = 0$ (top right), $f_B = 0.8$ and $|\kappa| = \kappa_{\text{max}}$ (bottom right). Here $\kappa_{\text{max}} = 2\pi/60 \mu m^{-1}$ is the largest spiral wavevector (tightest winding) obtainable in current experiments. In general, increasing $f_B$ suppresses the dynamical instability by decreasing of Im $\omega_\kappa$. Increasing $|\kappa|$ introduces additional instabilities with a length scale set by $2\pi/|\kappa|$ which is typically 60 $\mu m$ or greater. They are well-separated from instabilities due to dipolar interactions which are typically at a length scale of 10 $\mu m$. Experimentally, the instability also appears to occur on shorter or longer timescales depending on whether the length scale of the spiral winding is smaller or larger, respectively, than the largest length scale of the condensate. We do not find a strong dependence of the instability timescale on the wavelength of the spiral winding which was reported in [23]. One possibility for this discrepancy is that in experiments, spiral states are prepared dynamically by applying a strong gradient of the magnetic field. Winding the spins dynamically may introduce noise and fluctuations into the system and facilitate the development of fragmentation.

**FIG. 4:** Spin mode in momentum space (left) illustrating regions of unstable modes with shading indicating magnitude of Im $\omega_\kappa$ and $\hat{B}$ the magnetic field for current experimental parameters $\alpha = \pi/2$, $q = 0$ [23]. The red marker indicates these parameters in the spin and charge mode phase diagrams (middle and left) which have similar plots illustrating regions of unstable modes in various phases (see text for description). The blue arrow indicates parameters for Fig. 3.

**DISCUSSION**

Our analysis shows the intriguing complexity of excitation spectrum of spinor condensates when dipolar interactions are taken into account. In particular we demonstrated that softening of the roton gap is associated with
transverse fluctuations of the magnetization. This lowers the dipolar interaction energy and such fluctuations are distinct from phonon excitations of the superfluid. Earlier studies on dipolar effects in polar molecules treating the spin degrees of freedom as frozen obtained an excitation spectrum with one branch mixing the phonon and roton parts.

Dynamical spin degrees of freedom have been considered before in studies of ground state properties of spinor condensates but only with the bare dipolar interactions. Our analysis demonstrates rapid Larmor precession and reduced dimensionality significantly modify the effective dipolar interaction. Using this effective interaction, we have shown uniform mean-field condensates have instabilities towards a possible supersolid with stripe or checkerboard crystalline order.

Our work opens several new directions in the study of spinor condensates. We demonstrated that the uniform phase is unstable toward spin modulation for a whole range of wavevectors. However competition between unstable modes and the resulting stable state still need to be explored. Possible candidate phases include both states with small modulation and states with spiral spin winding. Another interesting question to be explored is the interplay of Bose condensation and spin modulation in the presence of thermal fluctuations. At the mean-field level spin modulation should appear simultaneously with the appearance of the condensate. However spin modulation breaks translational symmetry and is a distinct symmetry breaking from the Bose condensation. Hence a priori the two may have different transition temperatures.

The main significance of our work is developing a microscopic theory which successfully explains the checkerboard pattern observed in recent experiments of the Berkeley group. Previous theoretical studies have addressed these experiments without taking into account the precession averaged dipolar interaction and have not explained the observed pattern of spin fragmentation. Although we analyzed the simplest geometry of an infinite two dimensional layer, our results are in good agreement with the current experimental data. We expect however that the two dimensional model may not capture some effects which may be present in real systems such as pinning of the instabilities by the in plane trapping potential.

In conclusion, we have considered the combined effects of dynamical spin degrees of freedom, Larmor precession, and reduced dimensionality on the collective mode spectrum of spinor condensates with dipolar interactions. Starting from a mean-field state with uniform or spiral magnetization, we demonstrated the presence of a phonon and roton spectrum of collective modes. Softening of the roton gap suggest instabilities towards a possible supersolid state with stripe or checkerboard periodic crystalline order.

**METHODS**

**Dipolar Interaction**

Here we outline the derivation of the effective two-dimensional time-averaged dipolar interaction. The effect of the transformation in Eq. 4 is clearest in the bosonic coherent state path integral formalism for $\mathcal{U}$ the evolution operator

$$\mathcal{U} = \int \mathcal{D}\Psi^\dagger \mathcal{D}\Psi e^{i\int dt\mathcal{L}}, \quad \mathcal{L} = i \int d^3x \Psi^\dagger \partial_t \Psi - \mathcal{H} \quad (16)$$

where $\mathcal{H}$ is given in Eq. 3. Only the time derivative, spatial derivative and dipolar interaction tensor transform non-trivially. The transformed time and spatial derivatives

$$\partial_t \rightarrow \partial_t + B_0 \hat{B} \cdot \hat{F},$$

$$\frac{\nabla^2}{2m} \rightarrow \frac{\nabla^2}{2m} - i \frac{|\kappa|\nabla}{m} \hat{B} \cdot \hat{F} + \frac{|\kappa|^2}{2m} \left( \hat{B} \cdot \hat{F} \right)^2 \quad (17)$$

can be absorbed into Hamiltonian as modified linear and quadratic Zeeman shifts.
For the dipolar interaction tensor, the transformation needs to be expressed as one acting on \( h^{ij}_{3D}(\vec{x}) \) instead of \( \Psi_x \). Recall \( \tilde{F}_ij_{jk} \) are \( SO(3) \) Clebsch-Gordon coefficients projecting the tensor product of two spin-1 representations (lower indices) onto the spin-1 component (upper index). In particular,
\[
R^T_{ij} \tilde{F}^{ij}_{jk} R_{jk} = R_{i'i'} \tilde{F}^{i'i'}_{jk}
\]
for an arbitrary \( SO(3) \) rotation \( R \) implying \( R \) acting on the lower two indices is equivalent to \( R \) acting on the one upper index. This gives
\[
\tilde{h}^{ij}_{3D}(\vec{x}-\vec{x}') = \int_{-\pi/B_0}^{\pi/B_0} B_{0dt} \left[ R(t, \vec{x})^T_{ii'} h^{i'i'}_{3D}(\vec{x}-\vec{x}) R(t, \vec{x'})_{jj'} \right]
\]
where the bar denotes time-averaging and the explicit dependence on \( t \) and \( \vec{x} + \vec{x}' \) is removed as a result. Here \( \tilde{h}^{ij}_{3D}(\vec{x}) \) is given in Eq. 2. The short-distance singularity \( |\vec{x}|^{-3} \) in the Fourier transform
\[
\tilde{h}^{ij}_{3D}(\vec{k}) = \int d^3 x e^{-i\vec{k}\cdot\vec{x}} \tilde{h}^{ij}_{3D}(\vec{x})
\]
is regularized with the prescription
\[
|\vec{x}|^{-3}_{reg} = \begin{cases} 
0 & |\vec{x}| \leq b \\
|\vec{x}|^{-3} & |\vec{x}| > b
\end{cases}
\]
and taking \( b \to 0 \) at the end.

To take into account confinement along \( \hat{n} \) to thickness \( d_n \), we consider a general trial waveform of the form in Eq. 3 with
\[
\rho(x_n) = \frac{1}{d_n} f \left( \frac{x_n}{d_n} \right)
\]
where \( x_n \) is the coordinate along \( \hat{n} \) and \( f(x) \) is normalized to \( \int dx f(x) = 1 \). This results in
\[
\tilde{h}^{ij}_{2D}(\vec{k}) = \int \frac{d\vec{k}_n dxdx' \tilde{h}^{ij}_{3D}(\vec{k}, \vec{k}_n) \epsilon^{k_n a d_n (x-x')} f(x) f(x')}{\int dx f^2(x)}
\]
with \( \vec{k} (k_n) \) the two-dimensional (one-dimensional) coordinate perpendicular (parallel) to \( \hat{n} \). We obtain Eq. 6 with
\[
\tilde{h}^B_{2D}(\vec{k}) = 1 - 3g(\vec{k}, 0)
\]
\[
\tilde{h}^+_{2D}(\vec{k}) = -\frac{1}{2} + \frac{3}{4} q(\vec{k}, +|\vec{k}|) + \frac{3}{4} q(\vec{k}, -|\vec{k}|)
\]
\[
\tilde{h}^-_{2D}(\vec{k}) = -\frac{3}{4} q(\vec{k}, +|\vec{k}|) + \frac{3}{4} q(\vec{k}, -|\vec{k}|)
\]
and the function \( g(\vec{u}, \vec{v}) \) given by
\[
g(\vec{u}, \vec{v}) = (\vec{B} \cdot \vec{u} + \vec{v})^2 w(|\vec{u} + \vec{v}|) d_n - 2 \int_{|\vec{u}|d_n}^{|\vec{u} + \vec{v}|d_n} \frac{dq}{q} w(q) + (\vec{B} \cdot \hat{n})^2 [1 - w(|\vec{u} + \vec{v}|d_n)]
\]
and the following function
\[
w(q) = \frac{q \int dx dx' e^{-q|\vec{x}-\vec{x}'|} f(x) f(x')}{2 \int dx f^2(x)}
\]
the only quantity dependent on \( f(x) \). The asymptotic behavior of \( w(q) \) is
\[
w(q) = \begin{cases} 
\frac{1}{2D_0} q - \frac{D_{11}}{2D_0} q^2 & q \ll 1 \\
1 - \frac{D_{11}}{D_0} q^2 & q \gg 1
\end{cases}
\]
with \( D_0 = \int dx f^2(x) \), \( D_{11} = \int dx dx' f(x)f(x')|x-x'|^n \), \( D_{-n} = \int dx f(x)(-i\partial_x)^n f(x) \). For a gaussian trial waveform
\[
f(x) = \frac{e^{-x^2/2}}{\sqrt{2\pi}}, \quad w(q) = 2q \int_0^\infty dke^{-(k^2 + 2kq)}
\]
and we stress the qualitative behavior of \( w(q) \) is rather insensitive to the detailed form of \( f(x) \).

Collective Modes

Next we outline the mean-field and collective mode analysis. Taking \( \Psi_x = \Psi \) with all quantities independent of \( \vec{x} \) in Eq. 7 and extremize the free energy at fixed density \( n \) and longitudinal magnetization \( f_B \). The free energy is independent of \( \eta \) and \( \phi \) and we find two classes of solutions
\[
\nu = 0, \quad Q\tau^3 + (1 - Q)\tau = f_B
\]
\[
\nu = \frac{\pi}{2}, \quad Q\tau^3 - (1 - Q)\tau^{-1} = f_B
\]
with \( \sin(\rho)^2 = \frac{f_B}{2Q} (\tau + \tau^{-1}) \). The constraints \( 0 \leq \sin(\rho)^2 \leq 1 \) and \(-1 \leq \tau \leq 1 \) select the unique root for the cubic equations
\[
\tau = \begin{cases} 
\frac{2Q \sinh \left[ \frac{\pi}{4} \arcsinh \left( \frac{f_B}{2Q\tau} \right) \right]}{f_B^{1/3}}, & Q < 1 \\
\frac{2Q \text{sign}(f_B) \cosh \left[ \frac{\pi}{4} \arccosh \left( \frac{|f_B|}{2Q\tau} \right) \right]}{f_B^{1/3}}, & Q > 1
\end{cases}
\]
\[
\tau^{-1} = \frac{2Q \text{sign}(f_B) \cosh \left[ \frac{\pi}{4} \arccosh \left( \frac{|f_B|}{2Q\tau} \right) \right]}{f_B^{1/3}}
\]
with \( Q = \sqrt{1 - Q^3/3Q} \) and \( Q \) given by Eq. 5 for the \( \nu = 0, \pi/2 \) solutions, respectively. For \( g_\perp(\alpha) > 0 \) or \( \alpha > \alpha_c \) (\( g_\perp(\alpha) < 0 \) or \( \alpha < \alpha_c \)), the \( \nu = 0 \) (\( \nu = \pi/2 \)) solution has a lower mean-field free energy. However, we stress both solutions are stationary points for all \( \alpha \).

The above constraints also imply the \( \nu = 0 \) solution exists for \( Q > -\frac{1}{2} \left( 1 + \sqrt{1 - f_B^2} \right) \) while the \( \nu = \pi/2 \) solution exists for \( Q < \frac{1}{2} \left( 1 - \sqrt{1 - f_B^2} \right) \). In general, both types of solutions support transverse magnetization.
except the fully polarized state along \( f_B = \pm 1 \) and the polar state along \( Q > 1 \), \( f_B = 0 \) (\( Q < 0 \), \( f_B = 0 \)) for \( \nu = 0 (\nu = \pi/2) \). We focus on the \( \eta = 0 \) solution for \( -1 < Q < +1 \) as it supports a finite transverse magnetization at \( f_B = 0 \) as observed in experiment.

Turning to the collective mode analysis, the linearized equations of motion are given by Eq. [9] with

\[
M_{\xi} = \frac{\hbar_k^2}{2m} - \mu - \left( p - \frac{|\hbar_k|}{m} \right) \hat{B} \cdot \hat{F} + \left( q + \frac{|\hbar_k|^2}{2m} \right) (\hat{B} \cdot \hat{F})^2
\]

\[+ g_0 \rho(0) C \Psi \Psi^T + g_0 \rho(0) C \Psi \Psi^T + 2g_* \rho(0) C \Psi \Psi^T + C \Psi \Psi^T \Phi^j + \Phi^j \Psi \Psi^T \Phi^j (35)\]

\[
N_{\xi} = g_0 \rho(0) C \Psi \Psi^T + g_0 \rho(0) C \Psi \Psi^T + C \Phi^j \Phi^j + \Phi^j C \Psi \Psi^T \Phi^j (36)\]

with \( C = 1/\sqrt{2} \) and \( \hbar^2_{2D}(k) \) by Eq. [5].

We acknowledge useful discussions with D. Stamper-Kurn, M. Vengalatorre, A. Lamacraft, V. Gritsev, G. Shlyapnikov. This work was supported by NDSEG and NSF Graduate Research Fellowship, Harvard-MIT CUA, DARPA, MURI, and the NSF grant DMR-0705472.

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