Normal-Superfluid Phase Separation in Spin-Half Bosons at Finite Temperature

Li He, 1 Peipei Gao, 2 and Zeng-Qiang Yu 2, *

1 College of Physics and Electronic Engineering, Shansi University, Taiyuan 030006, China
2 Institute of Theoretical Physics, Shanxi University, Taiyuan 030006, China

For pseudospin-half bosons with inter-spin attraction and intra-spin repulsion, normal phase and Bose condensed phase can coexist at finite temperature. The homogeneous system is unstable against the spinodal decomposition within a medium density interval, and consequently, a normal-superfluid phase separation takes place. The isothermal equation-of-state shows a characteristic plateau in the $P-V$ (pressure-volume) diagram, which is reminiscent of a classical gas-liquid transition, although, unlike the latter, the coexistence lines never terminate at a critical point as temperature increases. In a harmonic trap, the phase separation can be revealed by the density profile of the atomic cloud, which exhibits a sudden jump across the phase boundary.

Introduction.— The relation between Bose-Einstein condensation (BEC) and gas-liquid condensation has been discussed for a long time [1]. In the Einstein’s seminal work, BEC was thought as a spatial separation that the condensate neither occupy any volume nor contribute to pressure [2]. Although the equation-of-state (EoS) of free bosons resembles that of a van der Waals gas in the transition region [1, 3], such similarity is merely a coincidence due to the absence of interparticle interaction. As pointed out by London, the condensation stemming from the Bose statistics is more appropriately understood in momentum space rather than in coordinate space [4]. Nowadays, it is well known that a BEC transition associates with the spontaneous breaking of U(1) symmetry and the emergence of off-diagonal long range order [5]; by contrast, the classical gas-liquid condensation gives rise to the change only in density but not in symmetry [6]. Despite their distinctions in nature, the two kinds of condensation are not incompatible. In 1960, Huang proposed that, for bosons with hard-core repulsion and long range attraction, BEC transition and gas-liquid transition can take place simultaneously [7]. While this theory was based on an unrealistic model, it qualitatively reproduced some features of the phase diagram of $^4$He, in which the liquid phases, either superfluid or not, can coexist with the gas phase at finite temperature [8, 9].

The recent realization of self-bound liquids with ultracold atoms opens up new perspective to explore the gas-liquid transition in the quantum degenerate regime [10–17]. Such liquids, which would collapse from the mean-field viewpoint, are stabilized by the many-body effects of quantum fluctuations [18]. At a balanced density, energy per particle reaches the minimum, and the liquid exhibits the unique self-bound character. For a finite size system, a liquid drop is stable against evaporation when its atom number exceeds a critical value. The bound-unbound transition has been experimentally observed in the dipolar condensates of magnetic atoms [12], as well as the binary Bose mixtures with short range $s$-wave interactions [14, 16, 17].

Previously, the liquid-like properties of two-component bosons have been investigated by many theoretical works. Most of them focus on the influence of quantum fluctuations at zero temperature [18–26]; few pay attention to thermal effects [24]. In this Letter, we study the thermodynamics of pseudospin-half bosons with inter-spin attraction and intra-spin repulsion at finite temperature. Our main results are summarized in Fig. 1. Reminiscent of classical gas-liquid condensation, the isothermal EoS shows a characteristic plateau in the $P-V$ diagram, and the normal and the BEC phase coexist in the transition region. This unusual normal-superfluid phase separation (PS) is mainly driven by thermal fluctuations, although, quantum fluctuations still play an important role when the attractive and the repulsive mean-field energy almost cancel out. Contrary to the case of spinless bosons, here the BEC transition is of first order and is accompanied by an abrupt change in density; in this sense, the condensation occurs not only in momentum space but also in coordinate space.

EoS of a Homogeneous System.— We consider weakly interacting bosonic atoms, which occupy two hyperfine sublevels labelled by the pseudospin $\sigma = \uparrow, \downarrow$. In the grand canonical ensemble, the Hamiltonian reads

$$\hat{H} = \sum_{\sigma} \int d^3 r \hat{\psi}_\sigma^\dagger (r) \left( -\frac{\hbar^2 \nabla^2}{2m} - \mu_\sigma \right) \hat{\psi}_\sigma (r) + \frac{g_{\sigma\sigma'}}{2} \int d^3 r \hat{\psi}_\sigma^\dagger (r) \hat{\psi}_{\sigma'}^\dagger (r) \hat{\psi}_{\sigma'} (r) \hat{\psi}_\sigma (r), \quad (1)$$

where $\hat{\psi}_\sigma$ and $\mu_\sigma$ are the field operator and the chemical potential, respectively, of $\sigma$-component, $m$ is the atomic mass, and $g_{\sigma\sigma'}$ are the interaction parameters in different spin channels. Within the Born approximation, $g_{\sigma\sigma'} = 4\pi \hbar^2 a_{\sigma\sigma'}/m$ with $a_{\sigma\sigma'}$ the corresponding scattering length. In the present work, we assume $\mu_\uparrow = \mu_\downarrow = \mu$ and $g_{\uparrow\uparrow} = g_{\downarrow\downarrow} = g$ (accordingly $a_{\uparrow\uparrow} = a_{\downarrow\downarrow} = a$). Under these considerations, the Hamiltonian possesses $U(1) \times Z_2$ symmetry in spin space, and the spontaneous magnetization would not emerge unless the symmetry is broken. Hereafter, we will focus on the regime of $a \gg -a_{\uparrow\downarrow} > 0$, where the normal-superfluid PS could take place. First, let us assume the system is homogeneous. At a
finite temperature \( T \), the importance of quantum degeneracy is governed by the phase space density \( n\lambda_T^2 \), with \( n \) the atomic density and \( \lambda_T = \sqrt{2\pi \hbar^2 / mk_B T} \) the thermal wavelength. For \( n\lambda_T^2 \ll 1 \), the effects of quantum degeneracy are negligible, and atoms behave like classical particles with the EoS given by

\[
P = nk_B T + \frac{1}{2} (2g + g_{\uparrow\downarrow}) n^2, \tag{2}
\]

where the first term coincides with the pressure of a classical ideal gas, and the second term corresponds to the leading order contribution of interactions. On the other hand, for \( n\lambda_T^2 \gg 1 \), the system is in the highly degenerate regime, where almost all the bosons condense in the zero-momentum state [27]. The spinor condensate breaks the rotation symmetry in the \( \sigma_x-\sigma_y \) plane with the wavefunction given by \( \varphi = \sqrt{n/2} (e^{-i\theta_1}, e^{-i\theta_2}) \). Within the Bogoliubov approximation, we obtain the EoS

\[
P = \frac{1}{2} g + \frac{3}{5\sqrt{\pi}} \left( g + \sqrt{n\lambda_T^2} + g - \sqrt{n\lambda_T^2} \right), \tag{3}
\]

with \( g_{\pm} = \frac{1}{2} (g \pm g_{\uparrow\downarrow}) \) and \( a_{\pm} = \frac{1}{2} (a \pm a_{\uparrow\downarrow}) \). The first term of (3) is the mean-field energy of the condensate, and the second term is the Lee-Huang-Yang (LHY) correction caused by the quantum fluctuations.

For a constant \( T \), as density varies from small to large, the EoS will change its form from (2) to (3). During this evolution, two phase transitions take place successively: one is the transverse ferromagnetic transition at critical density \( n_{MT} \), the other is the BEC transition at critical density \( n_C \). The transverse spin polarization is energetically favorable owing to the inter-spin attraction, and the corresponding phase transition occurs for arbitrary small negative \( a_{\uparrow\downarrow} \) [28, 29]. At the mean-field level, the onset of ferromagnetism is fixed by the condition [30]

\[
2a_{\uparrow\downarrow} \frac{L_{1/2}}{L} (e^{\beta'\mu}) + \lambda_T = 0, \tag{4}
\]

where \( L_{q} (z) = \sum_{n=1}^{\infty} z^\mu / \ell_1 \) is the polylogarithm function of \( q \) order, \( \beta = 1/k_B T \), and \( \mu' = \mu - (g + g_{\uparrow\downarrow}) n \). By combining (4) with the density equation \( n\lambda_T^2 = 2\frac{L_{1/2}}{L} (e^{\beta'\mu}) \), the critical point \( n_{MT} \) thus can be determined. Up to the first order of \( a_{\uparrow\downarrow} \), we find

\[
n_{MT} = n_C^{(0)} + 8\pi a_{\uparrow\downarrow}/\lambda_T^4, \tag{5}
\]

where \( n_C^{(0)} = 2\xi (\frac{3}{2}) \lambda_T^2 \approx 5.22\lambda_T^2 \) is the critical density of the BEC transition in the noninteracting case, with \( \xi (q) \) the Riemann zeta function.

Bose condensation emerges when density exceeds the higher threshold value \( n_C \). For weak inter-spin attraction, \( n_C \) are very close to \( n_{MT} \), and the leading difference between them is of order \( a_{\uparrow\downarrow}^2 / \lambda_T^4 \) [31]. In the BEC phase, both thermal atoms and condensate contribute to the transverse magnetization, and their spin polarization prefer to align in the same direction [28]. Without any loss of generality, we set the polarization along the \( \sigma_x \) axis. The magnetization \( M_x \) and the condensate fraction \( n_{0}/n \), which act as the order parameters associated with the respective phase transitions, can be obtained from the Popov theory or the Hartree-Fock (HF) theory [30]. Their rises with \( n\lambda_T^2 \) are displayed in Fig. 2(b).

From the EoS (2) and (3), it is easy to show that the isothermal compressibility \( \kappa_T = (\partial n / \partial P)/n \) is positive in both cases of \( n\lambda_T^2 \ll 1 \) and \( n\lambda_T^2 \gg 1 \), hence the system is mechanically stable under the corresponding densities at a given \( T \). However, such stability no longer survives when density is within a medium range. The onset of the spinodal decomposition occurs at \( n_{MT} \), where \( \kappa_T \) exhibits a discontinuity. For \( a_{\uparrow\downarrow} \ll \lambda_T \), we find [30]

\[
\kappa_T^{-1} \big|_{n=n_{MT}} = g_{\uparrow\downarrow}^2 M, \tag{6}
\]

\[
\kappa_T^{-1} \big|_{n=n_{MT}} = \left( g + \frac{1}{2} g_{\uparrow\downarrow} - C\lambda_T^2 k_B T \right) n_{MT}^2, \tag{7}
\]
with the constant $C = -1/2\zeta(\frac{1}{2}) \approx 0.34$. In the weakly interacting regime, the last term in the parentheses of Eq. (7) is dominant, therefore the sign of $nT$ changes across the ferromagnetic transition [32]. For the case of $|a_{\uparrow\downarrow}|$ comparable to $a$, compressibility can retain a negative value within a wide range of density extending from the normal phase to the BEC phase [see Fig. 2(a)].

**Normal-Superfluid Phase Separation.**— The mechanical instability discussed above implies a PS at finite $T$. The equilibrium pressure in an inhomogeneous state with the coexistence of the normal and the BEC phase can be fixed by the Maxwell construction [3, 6]. An example is illustrated in the inset of Fig. 2(a). Alternatively, one can determine the coexistence lines according to the balanced conditions for chemical potential and pressure

$$\mu(n_1, T) = \mu(n_2, T), \quad P(n_1, T) = P(n_2, T),$$

where $n_1$ and $n_2$ are densities of the normal and the BEC phase, respectively, in the mixed state $(n_1 < n_M, n_2 > n_C)$. For average density within the interval $n_1 < n < n_2$, the mixed state has a lower free energy than that of the homogeneous ones, and as a result, the normal-superfluid PS takes place. Remarkably, the isotherm exhibits a horizontal segment in the coexistence region, which is usually recognized as a characteristic of gas-liquid condensation [see Fig. 1(a)]. Using the thermodynamic relation $dP = s\,dT + n\,d\mu$ ($s$ is the entropy density), one can further show that the coexistence pressure satisfies the Clapeyron equation $dP/dT = L/T (n_1^{-1} - n_2^{-1})$, with $L$ the latent heat per particle of the phase transition [3, 6].

When the inter-spin attraction is tuned from weak to strong, the PS region extends to an increasingly large area in the phase diagram. For $|a_{\uparrow\downarrow}| < 0.9a$, the phase diagram predicted by the Popov theory and the HF theory are essentially the same, which suggests the PS in this regime is mainly driven by thermal fluctuations. For stronger inter-spin attraction, however, quantum fluctuations play an important role to affect the phase boundaries [see Fig. 1(b)]. In particular, at $a_{\uparrow\downarrow} = -a$, where the mean-field energy of the condensate totally vanishes, the HF theory wrongly predicts the BEC phase would always collapse at finite temperature [33]. This deficiency can be remedied by the Popov theory, in which the LHY correction due to the quantum fluctuations is properly taken into account [30].

For fixed $a$ and $a_{\uparrow\downarrow}$, the PS occurs at any non-zero $T$, and the density difference between the coexisting normal and BEC phase becomes more pronounced as $T$ increases (see Fig. 3). This feature is in bold contrast to the classical gas-liquid condensation. In the latter case, the coexistence lines terminate at a critical temperature, beyond which gas and liquid are indistinguishable. Actually, the two separated phases considered here differ not only in density but also in symmetry. Across the phase interface, the order parameters $n_0/n$ and $M_\sigma$ exhibit abrupt changes as well. In this respect, the normal-superfluid PS is more like the gas-liquid coexistence in $^4$He below the temperature of the $\lambda$ point, where the gas behaves al-
most classically, while the liquid (He-II) shows a unique quantum nature [8, 9].

It is well known that mean-field approaches, such as the Popov theory and the HF theory, usually lead to a BEC transition of first order. The normal-superfluid PS was also predicted for spinless bosons with purely repulsive interactions [34–36], in which case the false spinodal decomposition near the BEC transition is due to the unphysical multi-valued behavior of the mean-field EoS. Such an artifact differs from the results presented here, where the mechanical instability is caused by the inter-spin attraction. As the attraction strength increases, the density interval of spinodal decomposition enlarges, while the artificial multi-valued region tends to vanish [30]. For $|a_{↑↓}|$ and $a$ being comparable, the phase-separated densities in the mixed states (namely $n_1$ and $n_2$) turn out to be considerably away from $n_c$. In that case, the mean-field description is supposed to be qualitatively reliable. On the other hand, if the inter-spin interaction turns into repulsion, the BEC transition should be of second order as usual. Thus, a tricritical point is expected to appear at $a_{↑↓} = 0$ [37]. This interesting tricriticality can be revealed by more sophisticated methods beyond the mean-field level [38, 39], and we leave the issue for future study.

**Density Profiles in a Trap.**— Now, we discuss the experimental relevance of our theory. For ultracold bosons confined in a harmonic trap, the normal-superfluid PS can be readily observed through the density profile of the atomic cloud. Below the condensation temperature, the BEC phase, being of a relatively higher density, would occupy the central region of the trap and be surrounded by an outer rim of the normal phase. Such shell structure is illustrated in Fig. 4, based on the mean-field calculation combined with the local density approximation (LDA). In contrast to the usual bimodal distribution, both the total and the condensate density profile exhibit a sudden jump at the phase boundary, which is a clear signature of the PS [40]. According to the phase diagram given above, the density jump would be more evident for larger value of $|a_{↑↓}|/a$ and higher temperature. In experiments, the 3D density distribution can be achieved by means of in situ absorption imaging followed by an Abel transform. Previously, this method has been successfully employed to detect the PS in spin imbalanced Fermi gases [41–44].

Within the LDA, the featured EoS can also be extracted from the measured density profiles [45–49]. Along certain axial direction, say, the $x$ direction, the local pressure can be obtained via the formula [45, 46]

$$P(x,0,0) = m\omega_y\omega_z n_{ax}(x)/2\pi,$$  \hspace{1cm} (9)

where $\omega_i$ are the frequencies of the harmonic trap ($i = x, y, z$), and $n_{ax}(x) = \int dydz n(r)$ is the integrated axial density. The first-order nature of the normal-superfluid transition implies the discontinuity of the derivative of pressure, which results in a kink in the axial density profile at the phase boundary (see the inset of Fig. 4). This singular behavior can also be inferred from the relation $dn_{ax}(x)/dx = -2\pi x n(x, 0, 0)\omega_0^2/\omega_y\omega_z$ [45, 46], according to which, the abrupt change of the slope of $n_{ax}(x)$ is proportional to the jump amplitude of the 3D density.

**Discussion and Conclusion.**— It should be noted that in the transverse ferromagnetic BEC phase atoms occupy superpositions of the hyperfine sublevels labelled by $\uparrow$ and $\downarrow$. From the viewpoint of symmetry breaking, the emergence of transverse magnetization requires a weak perturbation of spin-flip. Experimentally, the ferromagnetic phase can be realized at sufficiently low temperature when an external radio-frequency field driving the inter-spin transition is adiabatically switched off. On the other hand, if the system is initially prepared without the inter-spin coupling, the transverse magnetization would not occur spontaneously, since the number of atoms in each hyperfine level is conserved separately. The incoherent mixture achieved in the latter way also undergoes a normal-superfluid PS, although, the non-magnetic BEC phase has a higher free energy than that of the ferromagnetic one described above [30]. For binary heteronuclear mixture, the situation is even more complicated, since the mass ratio of the constituent atoms may have additional effects on the mechanical and the diffusive stabilities. The possible occurrence of a similar PS in dual-species Bose mixtures, such as $^{41}$K–$^{87}$Rb [17], $^{39}$K–$^{87}$Rb [50], and $^{23}$Na–$^{87}$Rb [51], will be considered elsewhere.

In summary, we have shown that for spin-half bosons with both attractive and repulsive interactions, the normal phase can coexist with the superfluid BEC phase at finite temperature, and the isotherms exhibit characteristic plateaus in the transition region. Our predictions for the EoS and the phase diagram can be examined in current experiments with ultracold atoms. Further interesting issue concerns the extension of this study to the regime of $a_{↑↓} < -a < 0$, where the spinodal decomposi-
tion occurs even at zero temperature. Presumably, as the density of the normal phase tends to vanish, the quantum liquid will eventually become self-bound in free space.

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* Electronic address: zqyu.physics@outlook.com

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I. Transverse Ferromagnetic Phase Transition

For the spin-half Bose system with inter-spin attractive interactions, as its phase space density increases, the occurrence of transverse ferromagnetism precedes the BEC transition [1, 2]. In the Hartree-Fock approximation, the transverse magnetization in the non-condensed phase satisfies the self-consistency equation

\[ n\lambda_T^3 M_x = \text{Li}_{3/2}(e^{\beta\mu} - \beta g_{1\downarrow} n M_x/2) - \text{Li}_{3/2}(e^{\beta\mu} + \beta g_{1\uparrow} n M_x/2). \]  

(S1)

Here, without any loss of generality, we assume the spin polarization along the \( \sigma_x \)-axis. The magnetization \( M_x \) and the shifted chemical potential \( \mu' = \mu - (g + \frac{1}{2} g_{1\uparrow}) n \) can be evaluated from Eq. (S1) combined with the density equation

\[ n\lambda_T^3 = \text{Li}_{3/2}(e^{\beta\mu'} - \beta g_{1\downarrow} n M_x/2) + \text{Li}_{3/2}(e^{\beta\mu'} + \beta g_{1\uparrow} n M_x/2). \]  

(S2)

Apparently, \( M_x = 0 \) is always a possible solution of these equations, which corresponds to the non-magnetic state. Actually, this is the only solution for sufficiently small phase space density. When \( n\lambda_T^3 \) exceeds a critical value, an additional solution with finite magnetization and lower free energy appears, which gives rise to the emergence of the ferromagnetic order.

For \( |a_{1\downarrow}| \ll \lambda_T \), the non-condensed ferromagnetic phase exists only in a narrow window of \( n\lambda_T^3 \) with small magnetization. In that case, Eqs. (S1) and (S2) can be solved through the series expansion in powers of \( M_x \). Retaining the terms up to the third order, we find

\[ (g_{1\downarrow} n M_x)^2 = -24 \left[ g_{1\downarrow} \partial_x \text{Li}_{3/2}(e^{\beta\mu'}) + \lambda_T^3 \right] / g_{1\uparrow} \partial_x \text{Li}_{3/2}(e^{\beta\mu'}) \]  

(S3)

with \( \mu' \) determined by

\[ n\lambda_T^3 = 2 \text{Li}_{3/2}(e^{\beta\mu'}) - 6 \partial_x^2 \text{Li}_{3/2}(e^{\beta\mu'}) \left[ g_{1\downarrow} \partial_x \text{Li}_{3/2}(e^{\beta\mu'}) + \lambda_T^3 \right] / g_{1\uparrow} \partial_x \text{Li}_{3/2}(e^{\beta\mu'}) \]  

(S4)

The critical condition for the ferromagnetic transition is then obtained by setting \( M_x \) equal to zero in (S3), i.e.,

\[ g_{1\downarrow} \partial_x \text{Li}_{3/2}(e^{\beta\mu'}) + \lambda_T^3 = 0. \]  

(S5)

Using the relation \( \partial_x \text{Li}_q(e^x) = \text{Li}_{q-1}(e^x) \), we can rewrite the condition (S5) in the form of Eq. (4) of the main text.

With the function \( n(\mu') \) derived in (S4), the isothermal compressibility \( \kappa_T = (\partial n / \partial \mu)|_T / n^2 \) can be evaluated straightforward. At a given \( T \), when density approaches the critical point \( n_M \) from above, we find

\[ \left. \frac{\partial n}{\partial \mu'} \right|_{n \to n_M^+} = \kappa_T^{-1} \left\{ 2 \partial_x \text{Li}_{3/2}(e^{\beta\mu'}) - 6 \left[ \partial_x^2 \text{Li}_{3/2}(e^{\beta\mu'}) \right]^2 / \partial_x^3 \text{Li}_{3/2}(e^{\beta\mu'}) \right\} = \beta \lambda_T^{-3} \left[ 2 \zeta \left( \frac{3}{2} \right) + \mathcal{O} \left( \beta \mu' \right) \right], \]  

(S6)

where, in the second equality, we have used the fact \( \beta \mu' \) is negative small at the ferromagnetic transition, and accordingly, the polylogarithm function can be simplified as [3]

\[ \text{Li}_{3/2}(e^x) = \zeta \left( \frac{3}{2} \right) - 2 \sqrt{\pi} |x| + \zeta \left( \frac{3}{2} \right) x + \mathcal{O} (x^2), \quad (x \to 0^-). \]  

(S7)

By applying the expansion (S7) to the critical condition (S5), it is readily to show the omitted terms in (S6) are at least at order of \( a_{1\downarrow}^2 / \beta \lambda_T^2 \). Therefore, up to first order of the interaction parameters, the inverse compressibility can be written as Eq. (7) of the main text, namely

\[ \kappa_T^{-1} \big|_{n \to n_M^+} = (g + \frac{1}{2} g_{1\downarrow}) n_M^2 + n_M^2 \lambda_T^3 / 2 \beta \zeta \left( \frac{3}{2} \right) \]  

(S8)

For \( |a_{1\downarrow}| \ll \lambda_T \), the last negative term on the r.h.s of (S8) is dominant, which results in the mechanical instability of the ferromagnetic phase.
On the other hand, in the non-magnetic phase, \( \mu' \) satisfies the density equation \( n \lambda_T^3 = 2L_i3/2(e^{\beta \mu'}) \), which yields \( \partial n/\partial \mu' = 2\beta \lambda_T^3 L_i1/2(e^{\beta \mu'}) > 0 \). Hence, the compressibility \( \kappa_T \) exhibits a discontinuity with a sign change at the ferromagnetic phase transition.

### II. Popov Theory and Hartree-Fock Theory

We employ the Popov theory and the Hartree-Fock theory to study the thermodynamics of spin-half bosons at finite temperature. The formalism has been presented in a previous work by one of us [4]. In the transverse ferromagnetic BEC phase, we introduce the mean-field parameters \( \delta n \) and \( \delta M_x \) to denote the density and magnetization of thermal atoms, respectively. These quantities can be determined self-consistently from the equations

\[
\delta n = \sum_{\alpha=\pm} \int \frac{d^3k}{(2\pi)^3} \left[ (u_{k,\alpha}^2 + v_{k,\alpha}^2) f(E_{k,\alpha}) + v_{k,\alpha}^2 \right],
\]

\[
\delta n \delta M_x = \sum_{\alpha=\pm} \alpha \int \frac{d^3k}{(2\pi)^3} \left[ (u_{k,\alpha}^2 + v_{k,\alpha}^2) f(E_{k,\alpha}) + v_{k,\alpha}^2 \right],
\]

where \( f(E_{k,\alpha}) = 1/(e^{\beta E_{k,\alpha}} - 1) \) is the Bose distribution function with \( E_{k,\alpha} \) being the excitation energy of quasi-particles. In the Popov theory, \( E_{k,\pm} = \sqrt{\varepsilon_k (\varepsilon_k + 2g_a n_0)} \), \( E_{k,\pm} = \sqrt{(\varepsilon_k + g_a n_0 - g_{a \pm} \delta n \delta M_x)^2 - g_a^2 n_0^2} \), \( u_{k,\pm} \) and \( v_{k,\pm} \) are coefficients of the Bogoliubov transformation satisfying the relations \( u_{k,\pm}^2 - v_{k,\pm}^2 = 1 \) and \( 2u_{k,\pm} v_{k,\pm} = -g_a n_0 / E_{k,\pm} \). In the Hartree-Fock theory, \( E_{k,\pm} = \varepsilon_k + g_a n_0 \), \( E_{k,\pm} = \varepsilon_k + g_a n_0 - g_{a \pm} \delta n \delta M_x \), \( u_{k,\pm} = 1 \) and \( v_{k,\pm} = 0 \). In both cases, the condensate density \( n_0 = n - \delta n \), and \( \varepsilon_k = \hbar^2 k^2 / 2m \). When \( n_0 \) approaches zero, Eqs. (S9) and (S10) recover Eqs. (S2) and (S1), respectively, at the BEC transition.

In the representation of quasi-particles, we obtain the free energy density

\[
F = F_0 + F_Q - k_B T \sum_{\alpha=\pm} \int \frac{d^3k}{(2\pi)^3} \ln (1 + f_{k,\alpha}),
\]

with \( F_0 = \frac{1}{2} g_a n^2 + \frac{1}{4} g \delta n^2 + \frac{1}{2} g_{a \pm} \delta n^2 \delta M_x (2 - \delta M_x) \). \( F_Q \) vanishes in the Hartree-Fock theory, but takes a nontrivial form in the Popov theory

\[
F_Q = \frac{1}{2} \int \frac{d^3k}{(2\pi)^3} \left[ E_{k,\pm} + E_{k,\pm} - 2\varepsilon_k - g_a n_0 + g_{a \pm} \delta n \delta M_x + \frac{g_a^2 n_0^2}{4\varepsilon_k} + \frac{g_{a \pm}^2 n_0^2}{4\varepsilon_k} \right].
\]

Physically, it originates from the quantum fluctuations associated with the zero-point motion of quasi-particles. In the highly degenerate limit \( n \lambda_T^3 \gg 1 \), thermal atoms are negligible, accordingly, \( F_0 \) reduces to the mean-field energy density of the condensate \( \mathcal{E}_0 = \frac{1}{2} g_a n^2 \), and \( F_Q \) recovers the Lee-Huang-Yang correction to the ground state energy \( \mathcal{E}_{LHY} = 64 \left( g_a \sqrt{n a_\pm^2} + g_a \sqrt{n a_\pm^2} \right) n^2 / 15 \sqrt{\pi} \). For the case of \( |a_\pm| \) comparable to \( a \), the attractive and the repulsive mean-field energy mostly cancel out, and \( \mathcal{E}_{LHY} \) gives an important contribution to the equation-of-state.

Once free energy is obtained, chemical potential and pressure are readily evaluated according to the thermodynamic relations \( \mu = \partial F / \partial n \) and \( P = \mu n - F \). These quantities are used in the determination of the balanced conditions for the normal-superfluid phase separation [Eq. (8) of the main text].

### III. Multi-Valued Behavior Near BEC Transition

It is well known that the self-consistency equations in the Popov theory and the Hartree-Fock theory usually have multiple solutions around the BEC transition, which results in a non-monotonic change in condensate fraction [5, 6]. Such artifact also exists in our spin-half model. However, as shown in Fig. S1, the unphysical multi-valued region shrinks with increasing relative strength of inter-spin attraction. In particular, at \( a_\pm = -a \), the multi-valued behavior completely disappears in both the Popov theory and the Hartree-Fock theory \(^1\). On the other hand, the density interval

\(^1\) This result can be simply understood by noticing that, in the specific case of \( a_\pm = -a \), the lower branch of excitation spectrum \( E_{k,\pm} \) is identical to that of the noninteracting system.
of spinodal decomposition enlarges as the ratio $|a_{T\downarrow}|/a$ increases [see Fig. 1(a) in the main text]. These two opposite trends clearly show that the mechanical instability is a consequence of the inter-spin attractive interactions, rather than an artifact of the mean-field approximations.

IV. Thermodynamics of Incoherent Bose Mixture

As mentioned in the main text, if the inter-spin conversion is prohibited, the transverse ferromagnetic order would not emerge spontaneously. Experimentally, the system prepared under this condition is an incoherent binary mixture, in which the atom number of each component is conserved separately.

Now, let us briefly discuss the thermodynamic properties of such incoherent mixture. For simplicity, we consider atoms have the balanced population in each component and the interaction parameter $a + a_{T\downarrow}$ is not very close to zero. In that case, the contribution of quantum fluctuations can be safely ignored, and the equation-of-state predicted by the Hartree-Fock theory is qualitatively reliable.

We start by assuming that the mixture is homogeneous. Within the Hartree-Fock approximation, each component behaves like a single gas with the chemical potential having a mean-field shift due to the inter-component interactions. In the BEC phase, we have

$$
\mu_{T\uparrow} = \mu_{T\downarrow} = \frac{1}{2} g(n + \delta n) + \frac{1}{2} g_{T\downarrow} \lambda \delta n, \quad \text{(mixture)} \quad (S13)
$$

where $\delta n$ is the total density of thermal atoms given by

$$
\delta n = \sum_{\sigma} \int \frac{d^3 k}{(2\pi)^3} f(E_{k,\sigma}) = 2\lambda_T^{-3} \text{Li}_{3/2}(e^{-gn_0/2k_B T}), \quad \text{(mixture)} \quad (S14)
$$

with $E_{k,\uparrow} = E_{k,\downarrow} = \epsilon_k + gn_0/2$ being the excitation energy. For comparison, in the BEC phase of spin-half bosons, the chemical potential reads,

$$
\mu = \frac{1}{2} g(n + \delta n) + \frac{1}{2} g_{T\downarrow} (n + \delta n M_x), \quad \text{(spinor)} \quad (S15)
$$

where $\delta n$ and $\delta M_x$ are determined by Eqs. (S9) and (S10). One can see that, in the spinor system, the transverse magnetization of thermal atoms gives rise to an additional shift in chemical potential.

In Fig. S2, the isothermal $\mu-n$ curves of an incoherent Bose mixture and spin-half bosons are compared for the same interaction parameters. The two curves coincide in the non-degenerate regime but differ in the BEC phase. Combine this numerical result with the thermodynamic relation $F(n) = \int_0^n dn' \mu(n')$, we conclude that (at given $n$

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$^2$ Here, we still use $\uparrow$ and $\downarrow$ to label the two components of the mixture and assume $a_{T\uparrow} = a_{T\downarrow} = a$.

$^3$ In the limit $n\lambda_T^3 \gg 1$, both curves approach to the same asymptotic result $\mu = g_{T\uparrow} n$. 

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FIG. S2: Comparison of isothermal $\mu$-$n$ curves of an incoherent Bose mixture and spin-half bosons. Both systems are presumed to be homogeneous, and the predictions are based on the Hartree-Fock theory. Parameters: $a_{\uparrow\downarrow} = -0.8a$, and $T = 400\text{nK}$.

FIG. S3: Phase diagram of a balanced incoherent Bose mixture predicted by the Hartree-Fock theory. For comparison, the boundaries of phase separation region for spin-half bosons are also shown (dashed-lines). Parameters: $a_{\uparrow\downarrow} = -0.8a$.

and $T$) the free energy of the incoherent BEC mixture is higher than that of the spin-half bosons. Such difference in equation-of-state can be understood by noting that, in the spinor system, the inter-spin interactions produce an extra mean-field energy proportional to $\delta M^2$.

The non-monotonic behavior of the $\mu$-$n$ curve implies that the homogeneous state is unstable against the spinodal decomposition within a medium range of density. Therefore, the normal-superfluid phase separation also occurs in the incoherent mixture. The resulting phase diagram is displayed in Fig. S3. Compared with the spinor system, the area of coexistence region shrinks apparently at higher temperature. In the presence of a harmonic trap, such phase separation can be experimentally detected in the similar way as discussed in the main text.

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