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

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For pseudospin-half bosons with interspin attraction and intraspin repulsion, the 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.

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Introduction.- The relation between Bose-Einstein condensation (BEC) and gas-liquid condensation had been discussed for a long time [1]. In 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 interactions. As first pointed out by London, the condensation stemming from the Bose statistics is more appropriately understood in momentum space rather than in coordinate space [4]. Now, it is well known that a BEC transition associates the emergence of off-diagonal long-range order [5], while the classical gasliquid condensation gives rise to the change only in density [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]. Based on an unrealistic model, this theory qualitatively reproduced the phase diagram of ⁴He, 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 gasliquid 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. Owing to the finite-size effect, a liquid drop is stable against evaporation only when its atom number exceeds a critical value. The boundunbound transition has been experimentally observed in the dipolar condensates [12], as well as the binary Bose mixtures [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 spin-half bosons with interspin attraction and intraspin repulsion at finite temperature. Our main results are summarized in Fig. 1. Reminiscent of classical gasliquid condensation, the isotherms exhibit characteristic plateaus in the P - V diagram, and the normal and the BEC phase coexist in the transition region. This unusual normalsuperfluid 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 labeled by the pseudospin $\sigma = \uparrow, \downarrow$. The grand-canonical Hamiltonian reads

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

where $\hat{\psi}_{\sigma}$ and μ_{σ} are the field operator and the chemical potential, respectively, of σ component, *m* is the atomic



FIG. 1. (a) Isothermal EOS of spin-half bosons. The horizontal segments correspond to mixed states of the normal and the transverse ferromagnetic BEC phase. Coexistence lines (short-dashed) and spinodal lines (short-dotted) are plotted for $-1 \le a_{\uparrow\downarrow}/a \le -0.3$. They are supposed to meet at a tricritical point (denoted by \odot) when the interspin attraction vanishes. (b) Phase diagram in terms of phase space density $n\lambda_T^3$ and interaction parameter $a_{\uparrow\downarrow}/a$. Here, EOS and phase diagram are calculated based on the Popov theory; for comparison, phase boundaries predicted by the Hartree-Fock theory are also shown [dashed lines in (b)]. Parameters: T = 400 nK; for all the figures of this Letter, the mass of boson takes the value of ³⁹K atom, and the intraspin scattering length $a = 65a_0$ (with a_0 the Bohr radius).

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) × Z₂ symmetry in spin space, and the spontaneous magnetization would not emerge unless the symmetry is broken. Hereafter, we will focus on the regime of $a \ge -a_{\uparrow\downarrow} > 0$, where the normal-superfluid PS could take place.

At finite temperature *T*, the importance of quantum degeneracy of a homogeneous system is governed by the phase space density $n\lambda_T^3$, with *n* the atomic density and $\lambda_T = \sqrt{2\pi\hbar^2/mk_BT}$ the thermal wavelength. For $n\lambda_T^3 \ll 1$, the quantum effects are negligible, and atoms behave like classical particles with the EOS given by

$$P = nk_BT + \frac{1}{4}(2g + g_{\uparrow\downarrow})n^2, \qquad (2)$$

where the first term coincides with the pressure of an ideal classical gas, and the second term corresponds to the leading order contribution of interactions. On the other hand, for $n\lambda_T^3 \gg 1$, 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 about the σ_z axis with the wave function given by $\varphi = \sqrt{n/2} (e^{-i\theta_{\uparrow}}, e^{-i\theta_{\downarrow}})^{\dagger}$. Within the Bogoliubov approximation, we obtain the EOS

$$P = \frac{1}{2}g_{+}n^{2} + \frac{32n^{2}}{5\sqrt{\pi}}\left(g_{+}\sqrt{na_{+}^{3}} + g_{-}\sqrt{na_{-}^{3}}\right), \quad (3)$$

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 Eq. (3) is the mean-field energy of the condensate, and the second term is the Lee-Huang-Yang correction originating from the quantum fluctuations.

For a constant *T*, as density varies from small to large, the EOS will change its form from Eqs. (2) to (3). During this evolution, two phase transitions take place successively: one is the transverse ferromagnetic (TFM) transition at critical density n_M , the other is the BEC transition at critical density n_C . The transverse spin polarization is energetically favorable owing to the interspin attraction, and the resulting 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]

$$2a_{\uparrow\downarrow}\mathrm{Li}_{1/2}(e^{\beta\mu'}) + \lambda_T = 0, \qquad (4)$$

where $\operatorname{Li}_q(z) = \sum_{\ell=1}^{\infty} z^{\ell} / \ell^q$ is the polylogarithm function of q order, $\beta = 1/k_B T$, and $\mu' = \mu - (g + \frac{1}{2}g_{\uparrow\downarrow})n$. By combining Eq. (4) with the density equation $n\lambda_T^3 = 2\operatorname{Li}_{3/2}(e^{\beta\mu'})$, the critical point n_M thus can be determined. Up to the first order of $a_{\uparrow\downarrow}$, we find

$$n_M = n_C^{(0)} + 8\pi a_{\uparrow\downarrow} / \lambda_T^4, \tag{5}$$

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

Bose condensation emerges when density exceeds the higher threshold value n_C . For weak inter-spin attraction, n_C is very close to n_M , and the leading difference between them is of order $a_{\uparrow\downarrow}^2/\lambda_T^5$ [30]. 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 σ_x axis. The magnetization M_x and the condensate fraction n_0/n , which act as the order parameters associating with the respective phase transitions, can be obtained from the Popov theory or the Hartree-Fock (HF) theory [30]. Their rises with $n\lambda_T^3$ are displayed in Fig. 2(b).



FIG. 2. (a) Isothermal P - n curve of a hypothetically homogeneous system predicted by the Popov theory. Short-dashed lines labelled by ① and ② correspond to the asymptotic EOS Eqs. (2) and (3), respectively. Inset: the Maxwell construction in the coexistence region. (b) Transverse magnetization M_x and condensate fraction n_0/n as functions of $n\lambda_T^3$. Inset shows the critical points n_M and n_C are very close. Parameters: $a_{\uparrow\downarrow} = -a$, and T = 400 nK.

From EOS Eqs. (2) and (3), it is easy to show that the isothermal compressibility $\kappa_T = (\partial n / \partial P)_T / n$ is positive in both cases of $n\lambda_T^3 \ll 1$ and $n\lambda_T^3 \gg 1$, hence the system is mechanically stable under the corresponding densities. However, such stability no longer survives when density is within a medium range [33]. The onset of the spinodal decomposition occurs at n_M , where κ_T exhibits a discontinuity. For $|a_{\uparrow\downarrow}| \ll \lambda_T$, we find [30]

$$\kappa_T^{-1}|_{n \to n_M^-} = g n_M^2, \tag{6}$$

$$\kappa_T^{-1}|_{n \to n_M^+} = \left(g + \frac{1}{2}g_{\uparrow\downarrow} - C\lambda_T^3 k_B T\right) n_M^2, \tag{7}$$

with $C = -1/2\zeta(\frac{1}{2}) \simeq 0.34$. In the weakly interacting regime, the last term in the parentheses of Eq. (7) is dominant, therefore the sign of κ_T changes across the TFM transition. For $|a_{\uparrow\downarrow}|$ and *a* being comparable, compressibility retains a negative value within a wide range of density extending from the normal phase to the BEC phase [see Fig. 2(a)].

Normal-superfluid PS.—The mechanical instability discussed above implies a PS between the normal and the ferromagnetic BEC phase at finite temperature. The coexistence region can be fixed by the Maxwell construction [3,6], with an example illustrated in the inset of Fig. 2(a). Alternatively, one can determined the transition line according to the balanced conditions for chemical potential and pressure

$$\mu(n_1, T) = \mu(n_2, T), \qquad P(n_1, T) = P(n_2, T),$$
 (8)

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 shows 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 = $sdT + nd\mu$ (s is the entropy density), one can verify 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 interspin 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 results 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 [see Fig. 1(b)]. For stronger interspin attraction, however, quantum fluctuations play an important role to affect the phase boundary. In particular, at $a_{\uparrow\downarrow} = -a$, where the mean-field energy of condensate totally vanishes, the HF theory wrongly predicts the BEC phase would always collapse at finite temperature. This deficiency can be remedied by the Popov theory, in which the Lee-Huang-Yang correction due to the quantum fluctuations is properly taken into account [30].

For fixed *a* and $a_{\uparrow\downarrow}$, the PS occurs at any nonzero *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 case of classical gas-liquid condensation, where gas and liquid become indistinguishable above a critical temperature. 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_x exhibit abrupt changes as well. In this respect, the normal-superfluid PS is more like the gas-liquid coexistence in ⁴He below the lambda point, where the gas behaves almost 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



FIG. 3. Isotherms at different temperatures for $a_{\uparrow\downarrow} = -0.8a$. Coexistence lines (short-dashed) never terminate as *T* increases. Inset: phase diagram in the T - n plane. For parameters used here, the results predicted by the Popov theory and the Hartree-Fock theory are almost identical.

interactions [34–36], in which case the false spinodal decomposition near the BEC transition is due to the unphysical multivalued behavior of the mean-field EOS. Such an artifact differs from our situation, where the mechanical instability is attributed to the interspin attraction. As the attraction strength increases, the density interval of spinodal decomposition enlarges, while the artificial multivalued region tends to vanish [30]. For $|a_{\uparrow\downarrow}|$ and a being comparable, the phase-separated densities n_1 and n_2 turn out to be considerably away from $n_{\rm C}$. In that case, the mean-field description is supposed to be qualitatively reliable. On the other hand, the critical fluctuations beyond the mean-field level will become crucial when the PS region shrinks to small. Intuitively, the BEC transition should be of second order as usual if the interspin interaction turns into repulsion. Thus, a tricritical point is expected to appear at $a_{\uparrow\downarrow} = 0$ [37]. This interesting tricriticality can be revealed by more sophisticated methods [38,39], and we leave the issue for future study.

Density profiles in a trap.-Now, we discuss the experimental relevance of our theory. 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]. Experimentally, the 3D density distribution can be achieved by means of in situ absorption imaging followed by an inverse Abel transform. This method has been previously employed to detect the PS in spin imbalanced Fermi gases [41-44]. We note that, in actual experiments, the phase interface would become less sharp due to the surface tension effects. Nevertheless, a steep change in



FIG. 4. Density and condensate density profiles of $N = 5 \times 10^5$ atoms in an isotropic harmonic trap with trapping frequency $\omega = 2\pi \times 150$ Hz. The vertical short-dashed line indicates the phase boundary. Inset: integrated axial density profiles. Parameters: $a_{\uparrow\downarrow} = -0.8a$, and T = 400 nK.

density distribution would be still discernible for sufficiently large systems.

Within the LDA, the featured EOS can also be extracted from the density profiles [45-49]. Along certain axial direction, say, the *x* axis, the local pressure can be obtained via the formula [45,46]

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

where ω_i are the frequencies of the harmonic trap (i = x, y, z), and $n_{ax}(x) = \int dy dz n(\mathbf{r})$ 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 profiles at the phase boundary (see the inset of Fig. 4). This singular behavior can also be inferred from the relation $dn_{ax}(x)/dx = -2\pi xn(x, 0, 0)\omega_x^2/\omega_y\omega_z$ [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 labeled by \uparrow and \downarrow . From the viewpoint of symmetry breaking, the emergence of the TFM order requires a weak perturbation of spin flip. Experimentally, such circumstance can be realized when an applied radio-frequency field driving the interspin transition is adiabatically switched off, and the magnetization then can be measured by using a $\pi/2$ pulse, which rotates the transverse spin to the σ_z direction [50–53].

On the other hand, if the system is initially prepared without the interspin coupling, the transverse ferromagnetism would not occur spontaneously, as the atom number in each hyperfine sublevel is individually conserved. The incoherent mixture achieved in this way also undergoes a normal-superfluid PS, although, the coexistence region shrinks with respect to the spin-half system, and the superfluid phase has a higher free energy than that of the ferromagnetic BEC [30]. For heteronuclear mixtures, 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 ⁴¹K-⁸⁷Rb [17], ³⁹K-⁸⁷Rb [54], and ²³Na-⁸⁷Rb [55], will be considered elsewhere.

We finally mention that the PS studied here differs from the immiscible phenomenon, which is governed by the same Hamiltonian, but with repulsive interspecies interactions [56–69]. In that case, the two separated phases are both superfluid, and across their interface the longitudinal magnetization shows an abrupt change. The miscibleimmiscible transition occurs when the positive $a_{\uparrow\downarrow}$ reaches a critical value. Recent investigation reveals this critical condition is significantly affected by the interaction driven thermal fluctuations [69].

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 the characteristics of gas-liquid condensation. Our predictions for the EOS and the phase diagram can be examined in current experiments with ultracold atoms. A further interesting issue concerns the extension of this study to the regime of $a_{\uparrow\downarrow} < -a < 0$, where the spinodal decomposition 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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