

Magnetic phase transition in a mixture of two interacting Bose gases at finite temperature

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The miscibility condition for a binary mixture of two interacting Bose-Einstein condensates at zero temperature is shown to be deeply affected by interaction driven thermal fluctuations, which give rise to a first order phase transition to a demixed phase with full spatial separation of the two condensates. Explicit predictions for the spinodal temperature T_M , where the spin susceptibility diverges are provided. For a mixture of two sodium condensates occupying the hyperfine states $|F=1, m_F=1\rangle$ and $|F=1, m_F=-1\rangle$ respectively, T_M turns out to occur at about one half the usual BEC critical temperature. Analytic and numerical results for the transition temperature are obtained in the framework of Hartree-Fock theory and compared with predictions of universal relations, holding in the critical region of the superfluid transition. We further extend our analysis to two-dimensional Bose mixtures where a similar scenario occurs.

Introduction.—The miscibility of liquids and gases, and in particular its temperature dependence, is a topic of high relevance in the study of classical fluids [1]. For quantum mixtures, this question was addressed long time ago in the context of ^3He - ^4He liquids [2], and more recently for the mixtures of quantum gases [3, 4]. In particular, binary Bose gases occupying two different hyperfine states are the simplest, yet interesting example of quantum mixtures, offering an ideal playground for the investigation of diverse phenomena, including collective modes [5–7], superfluidity [8, 9], non-equilibrium dynamics [10–12], coherent coupling [13, 14] and quantum droplets [15, 16]. The miscibility of binary Bose condensates has been intensively investigated, both experimentally [17–21] and theoretically [22–31]. These theoretical studies have revealed that, at zero temperature, the mixture is stable against phase separation if the inequality $g_{12}^2 < g_{11}g_{22}$ holds, where g_{ij} is the coupling constant for the intra-species (g_{11} and g_{22}) and inter-species (g_{12}) interactions [3, 22–26]. At finite temperature, analysis have been mainly carried out for trapped systems, in the mean-field framework [27–29]. Although these studies clarified the effects of thermal atoms on the density profiles of the gas, it was found that for a Bose mixture confined in a harmonic potential, the zero temperature criterion is not significantly modified by the inclusion of the thermal component, because of its reduced spatial overlap with the condensate caused by the harmonic trapping. Instead for a homogeneous system, the authors of Refs. [29] and [30] have pointed out that the presence of thermal atoms may soften the criterion, so as to predict phase separation even for $g_{12}^2 < g_{11}g_{22}$. On the other hand, few works have been devoted on two-dimensional mixtures, and to our knowledge only the zero temperature stability has been discussed, within a renormalization group approach [32, 33].

In this Letter, we study the miscibility condition for uniform mixtures of bosonic atoms occupying two different hyperfine states, both in three and two dimensions,

by investigating the spin susceptibility. Throughout this work, we restrict ourselves to the simplest symmetric configuration where the number of atoms in each species as well as the intra-species interactions are the same, i.e. $N_1 = N_2 = N$ and $g_{11} = g_{22} = g$. For the three-dimensional (3D) system, we find that when the gas is in the Bose-Einstein condensed (BEC) phase and the inter-species interaction is close to, but still smaller than the intra-species one ($0 < \delta g = g - g_{12} \ll g$), the spin susceptibility exhibits a divergent behavior at a temperature T_M which is significantly smaller than the critical temperature for BEC in a single component atomic species. This singularity signals a transition to a phase-separated state even if the stability condition $g_{12} < g$ is ensured at zero temperature. Extension of the analysis for two-dimensional (2D) mixtures is discussed in the second part of the paper. We find that although BEC does not exist at finite temperature, the same magnetic instability occurs.

Magnetic susceptibility.—The magnetic susceptibility for a homogeneous binary mixture of Bose gases at finite temperature is defined through the Helmholtz free energy per unit volume V according to:

$$\kappa_M = \left(\frac{\partial^2 F/V}{\partial m^2} \right)_{m=0}^{-1}, \quad (1)$$

with $m = n_1 - n_2$ the magnetization density. For weak inter-species coupling, the free energy can be expressed as a sum of contributions from each species of the mixture and a temperature independent inter-species interaction term, $F = F_1(n_1) + F_2(n_2) + g_{12}n_1n_2$. By further introducing the isothermal compressibility for a single component $\kappa_{Ti} = [\partial^2(F_i/V)/\partial n_i^2]^{-1}$, with $i = \{1, 2\}$, Eq. (1) takes the convenient form

$$\kappa_M = \frac{2\kappa_T}{1 - g_{12}\kappa_T}, \quad (2)$$

with $\kappa_{T1} = \kappa_{T2} \equiv \kappa_T$ and we have assumed that the densities of the two components are equal: $n_1 = n_2 \equiv n$.

The onset of the magnetic instability is fixed by the simple condition $\kappa_T^{-1} = g_{12}$ and is favoured at finite temperature due to the increase of the isothermal compressibility of each species. It is worth mentioning that expression (2) only requires weak interaction between the two components, and the spin susceptibility is easily determined once the temperature dependence of the single-component isothermal compressibility is known.

For the single-species dilute Bose gas, an accurate description in the fluctuation region is provided by the universal relations (UR), which state that the equation of state in the vicinity of the critical density n_c , characterizing the onset of the superfluid phase transition, depends on a single variable X related to the interaction g and to the reduced chemical potential $\beta\mu$ ($=\mu/(k_B T)$), according to $n - n_c = f(X)$. Explicit results for the universal functions f in 3D and 2D were calculated from classical Monte-Carlo simulations in Refs. [34, 35]. However, the UR approach provides the proper description of the thermodynamic functions only in the critical region near the phase transition. We therefore also call on the mean-field Hartree-Fock (HF) and on Popov theories to provide a complementary description of thermodynamics.

The Hartree-Fock chemical potential for a single-component Bose gas below the superfluid phase transition, in three or two dimension, is given by [3]

$$\mu = g(n_0 + 2n'), \quad (3)$$

with $n = n_0 + n'$ the total atoms density. The non-condensed component is defined through the Bose distribution function $n' = V^{-1} \sum_{\mathbf{p}} [e^{\beta p^2/(2m)} z^{-1} - 1]^{-1}$, which takes into account the mean-field effect in the single particle energy through the expression $z = \exp[\beta(\mu - 2gn)]$ for the fugacity. It takes the explicit form

$$n' = \begin{cases} \lambda_T^{-3} g_{3/2}(z) & \text{in 3D} \\ -\lambda_T^{-2} \ln(1 - z) & \text{in 2D} \end{cases}, \quad (4a)$$

$$(4b)$$

where $g_p(z)$ is the usual Bose function and λ_T the thermal de Broglie wavelength. While in 3D, n_0 corresponds to the Bose-Einstein condensate density, this identification no longer holds in 2D, where BEC is ruled out at finite temperature. Instead n_0 in 2D corresponds to the quasi-condensate, which for sufficiently low temperature plays the same role as the genuine condensate [36]. In the following, we evaluate the single-component compressibility from the thermodynamic relation $\kappa_T = \partial n / \partial \mu$, for both the 3D and 2D systems, and discuss the resulting emergence of a magnetic instability at finite temperature.

Three dimensions.—In three dimensions, Hartree-Fock theory predicts BEC to occur at $k_B T_{\text{BEC}} = 2\pi\hbar^2/m(n/\zeta(3/2))^{2/3}$ with $\zeta(s)$ the Riemann zeta function. Below this temperature, the isothermal compressibility for a single component gas evaluated from Eqs.

(3) and (4a) takes the form

$$\kappa_T = \frac{1}{g} \frac{1 - g\beta g_{1/2}(z)/\lambda_T^3}{1 - 2g\beta g_{1/2}(z)/\lambda_T^3}. \quad (5)$$

At $T = 0$, Eq. (5) yields the well known result $\kappa_T = 1/g$, while the spin susceptibility (2) of the mixture takes the value $\kappa_M = 2/(g - g_{12})$, revealing its large increase near the miscible-immiscible phase transition occurring for $g_{12} = g$. For example, in the case of a mixture of ^{23}Na atoms occupying the hyperfine states $|F = 1, m_F = \pm 1\rangle$, one has $\delta g/g = 0.07$ yielding an increase of a factor ~ 14 of the $T = 0$ value of the spin polarizability with respect to the value obtained in the absence of interatomic interactions. The huge increase of the spin polarizability has been recently demonstrated experimentally in the case of a harmonically trapped mixture of sodium atoms [7, 8]. The presence of the factor 2 in the denominator of Eq. (5) is crucial for the increase of compressibility at finite temperature and thus for the description of the magnetic instability. This is the direct consequence of exchange effects which are responsible for the increase of the interaction energy with respect to the value predicted at $T = 0$ when the whole system is fully Bose condensed. This effect explicitly shows up in the temperature dependence of the chemical potential Eq. (3) and in the density dependence of the non-condensed component through the fugacity (see Eq. (4a)). If one employs the lowest order HF theory by taking $\mu \simeq g(n + \zeta(3/2)/\lambda_T^3)$, one misses the additional density dependence responsible for the enhancement of κ_T . This was the approximation made in the earlier work of Ref. [31], so that their prediction for the solubility condition turned out to be the same as at zero temperature. In Fig. 1(a) we report the temperature dependence of the isothermal compressibility κ_T of a single component Bose gas calculated from the HF expression (5), which explicitly reveals a significant increase in a useful region of temperature below T_{BEC} . The predictions of HF theory agree with the ones obtained from the universal relations of [34], as well as the ones of Popov theory [37, 38], plotted, respectively, as black circles and red dashed line in Fig. 1(a). This agreement motivates us to use the simplest HF theory for the discussion of magnetic instability in what follows.

Figure 1(b) shows the temperature dependence of κ_M in the case of the sodium mixture discussed above. The instability condition $\kappa_T^{-1} = g_{12}$ takes place at temperatures well below the critical temperature T_{BEC} , thereby revealing the occurrence of an experimentally visible effect. An analytic expression for the spinodal temperature T_M is obtained by expanding the Bose function in Eq. (4a) for small values of the coupling constant g . One finds for the single-component chemical potential

$$\mu \simeq g \left(n + \frac{\zeta(3/2)}{\lambda_T^3} \right) - g^{3/2} \frac{2\sqrt{\pi}}{\lambda_T^3} \sqrt{\beta(n - \zeta(3/2)/\lambda_T^3)}. \quad (6)$$

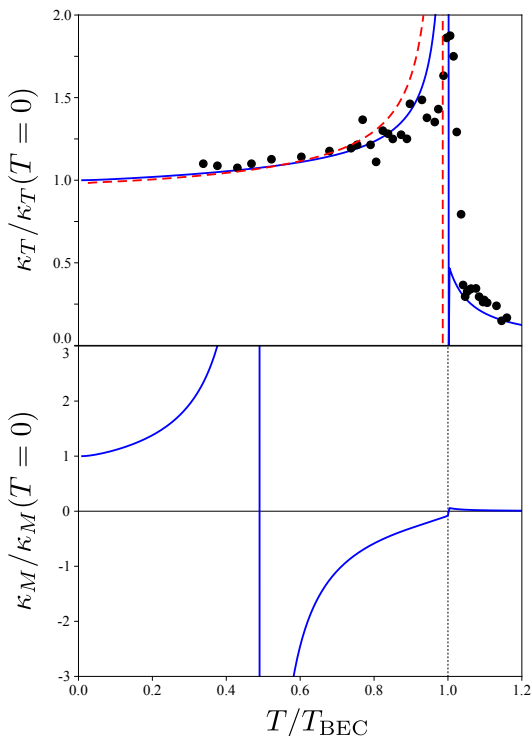


FIG. 1: (a) Single-component isothermal compressibility for a 3D Bose gas, with $gn/(k_B T_{\text{BEC}}) = 0.05$. The blue solid and the red dashed lines are the predictions of HF theory Eq. (5) and Popov theory [37], respectively. The black circles are results from the universal relations, calculated by numerically differentiating the data points for the equation of state given in Ref. [34]. (b) Spin susceptibility Eq. (2) in a 3D Bose mixture with interaction parameters $gn/(k_B T_{\text{BEC}}) = 0.05$ and $\delta g/g = 0.07$.

As already stressed, the emergence of a magnetic instability in the mixture is due to the last $g^{3/2}$ -term. The situation is analogous to that of quantum droplets, where the Lee-Huang-Yang (LHY) correction to the zero-temperature chemical potential is responsible for the stabilization of collapsing Bose mixtures [43]. While the LHY term originates from quantum fluctuations, the last term in Eq. (6) arises from interaction driven thermal fluctuations. From the HF expression (6) (or, equivalently, expanding the Bose functions in Eq. (5)) one can finally derive the following expression for T_M , holding for $gn \ll k_B T_{\text{BEC}}$ and $\delta g \ll g$:

$$\frac{T_M}{T_{\text{BEC}}} \simeq \zeta(3/2) \frac{\delta g}{g} \sqrt{\frac{1}{\pi} \frac{k_B T_{\text{BEC}}}{gn}}. \quad (7)$$

The calculation of T_M from the pole of Eq. (2), as well as from the analytical expression Eq. (7) are shown in Fig. 2 [39]. We briefly note that above T_{BEC} , when both species are in the normal phase, the chemical potential follows the ideal Bose gas behavior $\mu = \mu^{\text{BG}} + 2gn$, and the inverse isothermal compressibility takes the expres-

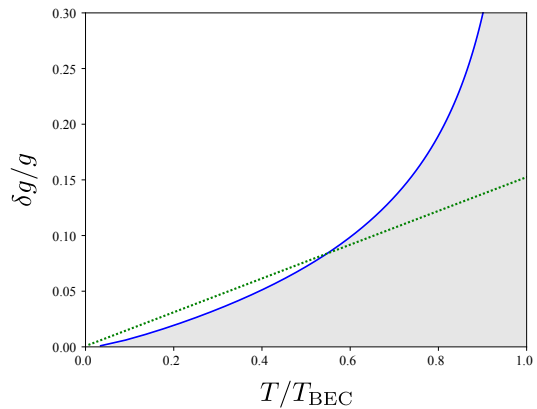


FIG. 2: Phase diagram for 3D binary condensates with $gn/(k_B T_{\text{BEC}}) = 0.05$. The blue solid and the green dotted lines are the magnetic instability temperature T_M , evaluated from the pole of Eq. (2) using the HF compressibility Eq. (5) and the small g expansion Eq. (7), respectively. The gray area corresponds to the region where the mixture is dynamically unstable against phase separation.

sion $\kappa_T^{-1} = 2g + (\beta g_{1/2}(z)/\lambda_T^3)^{-1}$. Consequently, the unpolarized state is stable as far as $g_{12} \leq 2g$.

The above results for the magnetic instability of the binary mixture suggest the occurrence of a first order phase transition, the value of T_M corresponding to the spinodal temperature above which the unpolarized uniform configuration of the mixture is dynamically unstable. The actual transition to a demixed configuration is then expected to take place at smaller values of the temperature and can be identified by comparing the free energy of the uniform unpolarized configuration with the one of the phase separated configuration. Consistent with the findings of Ref. [30], we find that the transition temperature lies very close to T_M for all values of $\delta g/g$, and the equilibrium configuration in the new phase is characterized by a full space separation of the Bose-Einstein condensed components of the two atomic species, their thermal components remaining instead mixed, but with a finite magnetization. A more complete discussion of the phase separation, including the propagation of spin sound, will be reported in a subsequent paper.

Before closing this section, let us briefly discuss the effects of inhomogeneity on the phase separation. So far, all the experiments on 3D binary Bose gases at finite temperature have been performed in presence of harmonic potentials (see for example Ref. [8]), and the phase transition discussed in this section has never been observed. This can be understood from the suppression of feedback between thermal and condensate atoms in a trapped gas. Indeed, in a trap the condensate atoms occupy the center of the gas, while the thermal ones are spread out, and consequently the overlap between the two components is greatly reduced. From the aforementioned free

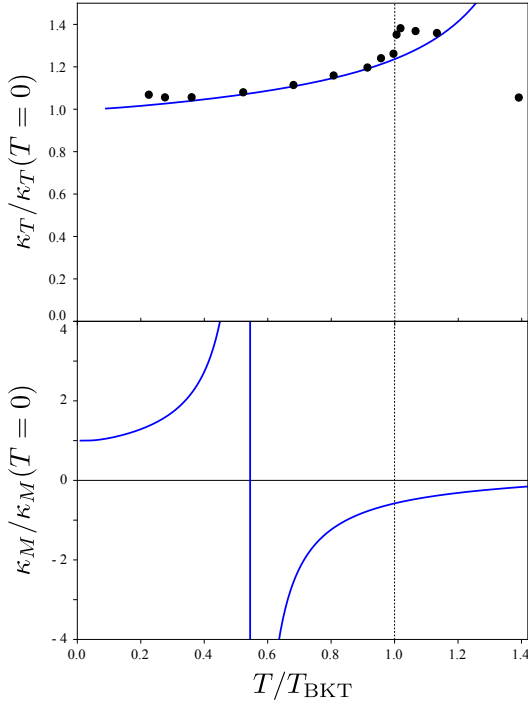


FIG. 3: (a) Single-component isothermal compressibility of a 2D Bose gas with $mg/\hbar^2 = 0.16$. The blue solid line is the prediction of HF theory Eq. (8), while the black circles are results from universal relations of Ref. [35], calculated in the same way as the 3D case. (b) Spin susceptibility Eq. (2) in a 2D Bose mixture with interaction parameters $mg/\hbar^2 = 0.16$ and $\delta g/g = 0.07$.

energy analysis, we have verified that by neglecting the coupling between the condensate and the thermal atoms, the phase separation does not occur for any values of T .

Two dimensions.—We now turn to the study of two dimensional Bose mixtures. Equation (4b) yields a divergent behavior for $\mu = 2gn$, in accordance with the fact that BEC does not exist in 2D at finite temperature. However, below the Berezinskii-Kosterlitz-Thouless (BKT) superfluid transition $k_B T_{\text{BKT}} = \frac{2\pi\hbar^2 n}{m} \ln^{-1}(380\hbar^2/(mg))$, one can safely use expression (3) for the chemical potential with the quasi-condensate $n_0 = n_{\text{qc}}$. Then, the isothermal compressibility of a single component 2D Bose gas is

$$\kappa_T = \frac{1}{g} \frac{1 - g\beta (e^{\beta g n_{\text{qc}}} - 1)^{-1} / \lambda_T^2}{1 - 2g\beta (e^{\beta g n_{\text{qc}}} - 1)^{-1} / \lambda_T^2}. \quad (8)$$

Comparing the above equation with the 3D expression (5), we find the same factor 2 in the denominator, arising from exchange effects. In Fig. 3(a) we compare κ_T calculated from Eq. (8) with the results of universal relations of Ref. [35], using a typical interaction parameter $mg/\hbar^2 = 0.16$ [44]. The Hartree-Fock theory is able to catch the qualitative behavior of the isother-

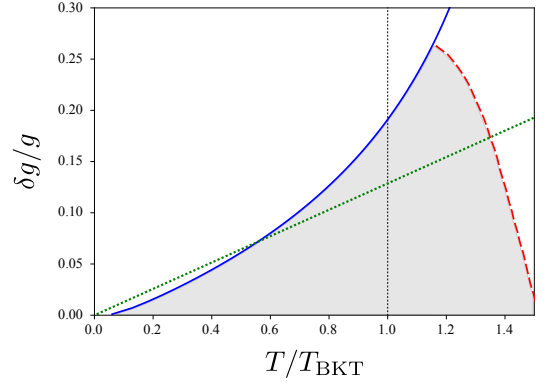


FIG. 4: Phase diagram for 2D binary condensates with $mg/\hbar^2 = 0.16$. The blue solid and the green dotted lines are the magnetic instability temperature T_M evaluated from the pole of Eq. (2) using the HF compressibility Eq. (8) and the small g expansion Eq. (9), respectively. The red dashed line is the UR prediction for the second magnetic instability temperature, obtained from an interpolation of the data points for the equation of state given in Ref. [35]. The gray area corresponds to the region where the mixture is dynamically unstable against phase separation.

mal compressibility, with its characteristic enhancement when T approaches the BKT transition, although, differently from UR, it does not predict the typical peak just above the critical point. Our result for the spin susceptibility Eq. (2) is reported in Fig. 3(b), for a mixture of ^{23}Na atoms with $\delta g/g = 0.07$. The decrease of κ_T above T_{BKT} predicted by the UR suggests that the condition $g_{12}\kappa_T = 1$ characterizing the magnetic phase transition should be satisfied also above the BKT transition.

In the same way as in 3D, one can derive an analytical expression for the spinodal temperature T_M below T_{BKT} , by using the small- g expression for the single component compressibility: $\kappa_T^{-1} \simeq g(1 - 2\mathcal{D}_{\text{qc}}^{-1})/(1 - \mathcal{D}_{\text{qc}}^{-1})$ with $\mathcal{D}_{\text{qc}} = \lambda_T^2 n_{\text{qc}}$. We briefly note that written in this form, the isothermal compressibility reflects the universal behavior of a 2D Bose gas. Indeed, $\kappa_T/\kappa_T(0)$ does not explicitly depend on the value of the coupling constant g and we have further verified that the same expression can be obtained within the modified Popov theory [40–42]. In the region $T \ll T_{\text{BKT}}$ for which $\mathcal{D}_{\text{qc}} \simeq \lambda_T^2 n \gg 1$, one obtains the simple estimate

$$\frac{mT_M}{2\pi\hbar^2 n} \simeq \frac{\delta g}{g}. \quad (9)$$

holding for $mg/\hbar^2 \ll k_B T_{\text{BKT}}$ and $\delta g \ll g$. The results for T_M from Eq. (9) as well as the numerical calculation from the pole of the spin susceptibility is reported in Fig. 4.

Conclusion.—In conclusion, we have investigated the magnetic phase transition in a uniform binary mixture of weakly interacting Bose gases. Our analysis revealed that phase separation of the homogeneous mixture can

occur, even if the zero temperature inequality $g_{12} < g$ is met, as a direct consequence of exchange effects and the coupling between condensate and thermal atoms beyond the lowest order. This phase transition is characterized by the divergence of spin susceptibility, which for binary mixtures of ^{23}Na atoms occupying the hyperfine states $|F=1, m_F=\pm 1\rangle$, occurs around $T \sim 0.5T_{\text{BEC}}$. The instability condition $\kappa_T^{-1} \leq g_{12}$ found in this paper indicates the crucial role played by the isothermal compressibility, a quantity for which experimental measurements are still lacking. Recently, box-like potentials yielding uniform trapping became available for both Bose [45] and Fermi [46] gases. Thus, the experimental possibility of observing the predicted magnetic phase transition is a realistic option.

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