P H Y S I C A L   R E V I E W   L E T T E R S

Zero-Temperature Phases of Many-Atom Bose Systems

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We show that a many-atom Bose system at zero temperature has, in general, a liquid phase in addition to its well-known gaseous phase. A universal phase diagram is presented that is applicable to all Bose systems with a $-C_6/r^6$ type of interaction at large interparticle separations. We show that the predicted phase structure has implications on the stability of a gaseous Bose-Einstein condensate (BEC) even at dilute densities that are routinely achieved under existing experimental conditions. We also predict that ^{4}He should have a gaseous BEC phase below a critical density of 5.58×10^{15} $1/\text{cm}^3$.

For decades, our understanding of many-atom quantum systems has been limited to weak coupling and low density regimes characterized by $\rho a_0^3 \ll 1$ and $\rho \beta_6^3 \ll 1$, where $\rho = N/V$ is the atomic number density, a_0 is the s wave scattering length, and $\beta_6 = (mC_6/\hbar^2)^{1/4}$ is the length scale associated with the van der Waals interaction, $-C_6/r^6$, between two atoms. As a result, there have been no general microscopic quantum theories of liquids beyond those specific to liquid helium (see, e.g., [1,2]). Even for dilute Bose gases, the theory has been limited to small scattering lengths as required by $\rho a_0^3 \ll 1$ [3,4].

In addition to the intrinsic difficulties associated with a many-body quantum system, this lack of progress can also be attributed to the fact that until recently, there has been no new systematic understanding of atomic interaction beyond the aging effective-range theory [5]. With the development of the angular-momentum-insensitive quantum-defect theory (AQDT) for diatomic systems [6–8], a new understanding of atomic interaction has emerged, along with a new, nonperturbative framework for studying and uncovering universal properties at different length scales in quantum two-atom, few-atom, and many-atom systems [8–10]. For a many-atom quantum system, in particular, it extends our understanding from weak coupling to arbitrary scattering length and from densities restricted by $\rho \beta_6^3 \ll 1$ to $\rho \beta_6^3 \sim 10$, or a typical density of 10^{20} $1/\text{cm}^3$, for alkali-metal atoms [10].

In this work, we apply this methodology [8–10], which we call the method of effective potential, or the method of renormalization in coordinate space, to show that a many-atom Bose system at zero temperature, even one with positive scattering length, has a liquid phase in addition to its well-known gaseous phase. The two phases correspond to different branches of many-atom states, with the same long-range, but different short-range correlations. A zero-temperature universal phase diagram is presented that is applicable to all Bose systems with a $-C_6/r^6$ type of long-range interaction. The result shows, for example, that a gaseous Bose-Einstein condensate (BEC) becomes unstable and undergoes a first-order phase transition to the

liquid phase either beyond a critical density or beyond a critical scattering length. The implications of this result for both alkali-metal and ^{4}He Bose systems will be discussed as examples.

Consider an N-body Bose system with pairwise interaction $v(r)$ that has the property of $v(r) \rightarrow -C_n/r^n$ $(n > 3)$ at large r. We have shown in a recent publication [10] that such a system, at sufficiently small densities, follows a universal equation of state (energy per particle as a function of density), called the universal equation of state at length scale $\beta_n = (mC_n/\hbar^2)^{1/(n-2)}$, which, other than scaling, is uniquely determined by the exponent n of the long-range interaction and a scaled scattering length. Specifically

$$E_s/N = \Omega^{(n)}(\rho_s, a_{0s}), \tag{1}$$

where $E_s = E/s_E$ is a scaled energy with energy scale $s_E = (\hbar^2/m)(1/\beta_n)^2$, $\rho_s = \rho \beta_n^3$ is a scaled density, $a_{0s} = a_0/\beta_n$ is a scaled s wave scattering length, and $\Omega^{(n)}$ is a universal function that is uniquely determined by the exponent n of the van der Waals interaction. Here the "sufficiently small density" requires only that $v(r)$ at the average interparticle separation, $r_\rho = (4\pi\rho/3)^{-1/3}$, is well represented by its asymptotic form of $-C_n/r^n$. For a many-atom system with $n = 6$, this range of densities can be estimated, from, e.g., $\beta_8/\beta_6 < \sim 1/2$ for a typical atom [11,12], to correspond to a range of $0 < \rho < \sim 10/\beta_6^3$. [Here β_8 is the length scale that corresponds to the C_8 term in the expansion $v(r) = -C_6/r^6 - C_8/r^8 + \ldots$.] For higher densities, there may still be universal properties for some classes of atomic systems at shorter length scales such as β_8. Beyond that, at densities where the electronic wave functions for different atoms start to overlap significantly with each other, no universal properties are generally expected, as nature has revealed to us through different crystal structures (solid phases) for different atoms. The pairwise interaction approximation also fails for such systems.

We restrict ourselves here to densities where the universal equation of state at length scale β_n is expected to be

applicable. Our theory for $\Omega^{(n)}$ is based on a theory of many-body Bose systems as outlined in Ref. [10] and is called here the nearest-neighbor theory (NNT) of many-body Bose systems. In this formulation, the energy per particle and the pair distribution function $g(r)$ for an N-particle Bose system with pairwise interaction $v(r) \rightarrow -C_n/r^n$ ($n > 3$) at large r are given by

$$E/N = \lambda/2 - \frac{2\pi C_n \rho}{(n-3)d^{n-3}}, \qquad (2)$$

and $g(r) = |F(r)|^2$, respectively. Here $F(r) \equiv u(r)/r$ is a pair correlation function, λ is a pair correlation energy, and d is a healing distance beyond which $F = 1$. They are determined by the solutions of

$$\left[-\frac{\hbar^2}{m}\frac{d^2}{dr^2} + v(r) - \lambda \right] u_\lambda(r) = 0, \qquad (3)$$

with a density-dependent normalization

$$4\pi\rho \int_0^d u_\lambda^2 dr = 1, \qquad (4)$$

and boundary conditions $(u_\lambda'/u_\lambda)|_{r=d} = 1/d$, and $u_\lambda(r \geq d) = r$. This formulation for a many-body Bose system has a clear physical interpretation. The first term in Eq. (2), $\lambda/2$, represents the energy of a particle as altered by its interaction with its nearest neighbor. The second term represents the mean field due to the van der Waals interaction with all other particles. Equation (4) gives the healing distance d a physical interpretation in NNT as being the distance within which the nearest neighbor is to be found. This theory was first deduced [10] from a constrained variational method [13,14] based on the Jastrow wave function [15]. A different derivation, which does not rely on the Jastrow wave function and has a greater potential to be generalized to a nearest q-neighbor theory ($q > 1$), is also possible and will be presented elsewhere. We stress here only that NNT is a general theory of a many-body Bose system with little assumption other than that only the nearest neighbors are correlated, an excellent approximation for all densities of interest here, as has been tested previously [10,14].

Systematic solutions of Eqs. (3) and (4) for the densities of interest take advantage of the following two results. (a) For any u_λ that satisfies Eq. (3), one can show that

$$\int_0^d u_\lambda^2 dr = \frac{\hbar^2}{m}[u_\lambda(d)]^2 \left[-\frac{d}{d\lambda}(u_\lambda'/u_\lambda) \right]\bigg|_{r=d}, \qquad (5)$$

which means that the normalization integral can be determined solely from u_λ and the energy dependence of its logarithmic derivative at the boundary. (b) At any radius where $v(r)$ is well represented by $-C_n/r^n$, the solution of Eq. (3) can be written as [6,8,10]

$$u_{\lambda_s}(r_s) = A[f_{\lambda_s l=0}^{c(n)}(r_s) - K^c g_{\lambda_s l=0}^{c(n)}(r_s)]. \qquad (6)$$

Here A is a normalization constant. K^c is a short-range K matrix that is related, for $n > 3$, to a_{0s} by [8,9]

$$a_{0s} = \left[b^{2b}\frac{\Gamma(1-b)}{\Gamma(1+b)} \right]\frac{K^c + \tan(\pi b/2)}{K^c - \tan(\pi b/2)}, \qquad (7)$$

where $b = 1/(n-2)$. $f_{\lambda_s l}^{c(n)}$ and $g_{\lambda_s l}^{c(n)}$ are the AQDT reference functions for $-C_n/r^n$ potential [6,10,16]. They depend on r only through a scaled radius $r_s = r/\beta_n$ and on energy only through a scaled energy $\lambda_s = \lambda/s_E$.

From these results, it is easy to show that the equation of state follows a universal behavior, independent of the nature of short-range interactions except through a_{0s}, provided that the density is sufficiently small that the $v(r)$ at r_ρ and beyond (same as d and beyond, since d has the same order of magnitude as r_ρ from its very definition) is well represented by $-C_n/r^n$. The corresponding $\Omega^{(n)}$, namely, the $\Omega^{(n)}$ in NNT approximation, is given by

$$E_s/N = \Omega^{(n)}(\rho_s, a_{0s}) = \lambda_s/2 - \frac{2\pi\rho_s}{(n-3)d_s^{(n-3)}}, \qquad (8)$$

with the λ_s and $d_s = d/\beta_n$ pair determined from

$$\left(\frac{1}{u_{\lambda_s}}\frac{du_{\lambda_s}}{dr_s} \right)\bigg|_{d_s} = \frac{1}{d_s}, \qquad (9)$$

$$d_s = \left\{ 4\pi\rho_s \left[-\frac{d}{d\lambda_s}\left(\frac{1}{u_{\lambda_s}}\frac{du_{\lambda_s}}{dr_s} \right)\bigg|_{d_s} \right] \right\}^{-1/2}, \qquad (10)$$

and Eq. (6) for $u_{\lambda_s}(r_s)$.

Figure 1 illustrates a typical equation of state for a many-atom Bose system ($n = 6$) with a large positive scattering length. It shows that the universal equation of state has in general multiple branches, corresponding to multiple solutions of Eqs. (9) and (10) for the same scaled density, each representing a different short-range correlation function. In particular, a Bose system with a positive scattering length has a liquid branch in addition to its well-known gaseous branch. This liquid branch, characterized by a negative energy per particle (self-binding) and a
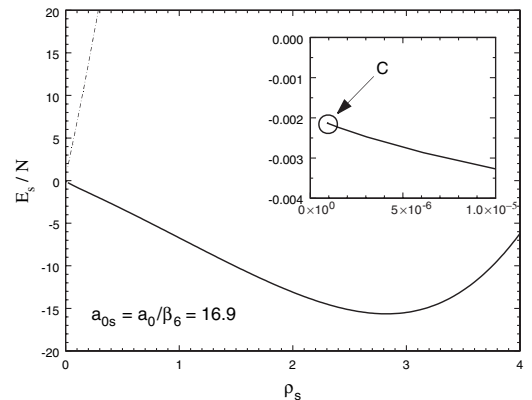

FIG. 1.   Two branches of universal equation of state at length scale β_6, computed in NNT, for a many-atom Bose system with $a_{0s} = a_0/\beta_6 = 16.9$, describing ^{4}He or any other system with the same scaled scattering length. Solid line: liquid branch. Dash-dotted line: gaseous branch. The liquid branch terminates at a critical point C, as shown in the inset.

negative pressure at small densities, differs from that for a negative scattering length [10] in one important aspect. Unlike the liquid branch for a negative scattering length that exists in the limit of zero density, the liquid branch for a positive scattering length terminates at a critical point C, corresponding to a scaled critical density ρ_{cs} that depends on the scaled scattering length a_{0s}. The physical consequence of this result is clear. Below ρ_{cs}, the gaseous BEC phase is the only phase that exists and is stable. (More precisely, it is metastable with metastability determined by the rate of molecular formation through three-body recombination [17,18].) Beyond the critical density, with the emergence of liquid states that have lower energy per particle, a gaseous BEC becomes unstable and can condense (or sometimes called "collapse") into a liquid state.

The universal equation of state as shown in Fig. 1 can be computed for any scaled scattering length. Putting together the critical densities for different scaled scattering lengths gives us the phase diagram as shown in Fig. 2. Since all the critical densities stay within the range ($\rho_s < 10$) in which the universal equation of state at length scale β_6 is expected to be followed, this phase diagram is also universal and applicable to all atomic Bose systems with $v(r) \to -C_6/r^6$ at large interatomic separations.

The phase diagram of Fig. 2 has a number of physical implications. It shows, for example, that (a) a gaseous BEC at a fixed positive scattering length becomes unstable and undergoes a first-order phase transition beyond a critical density. (b) A gaseous BEC at a fixed density becomes unstable beyond a critical scattering length. (c) No stable gaseous BEC phase exists beyond a density of $3.93\beta_6^3$, the maximum critical density corresponding to the zero scattering length. The most dramatic feature of this phase diagram is the sensitive dependence of the critical density on the scaled scattering length. It decreases by about 6 orders of magnitude as a_{0s} is varied from 0 to 10. As a result, this phase diagram has implications on the stability of a gaseous BEC even at dilute densities that are easily achieved under existing experimental conditions [19].
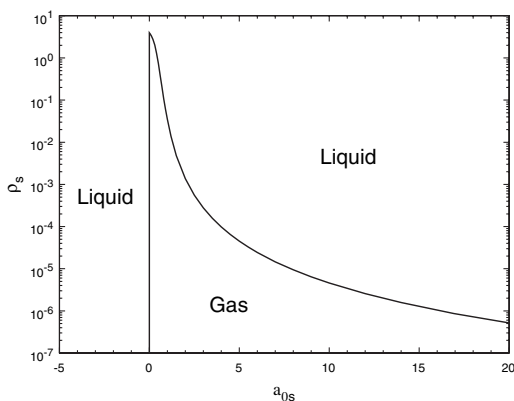

FIG. 2.   The universal phase diagram for a many-atom ($n = 6$) Bose system at zero temperature. Note the logarithmic scale for the density. Systems with negative scattering lengths have been discussed in Ref. [10].

Table I gives the numbers for some specific points on the phase diagram that can be of immediate experimental interest. For $a_{0s} = 16.9$, a critical scaled density of $\rho_{cs} = 8.92 \times 10^{-7}$ is predicted. This value of a_{0s} corresponds to that of ^{4}He. Here we have used $C_6 = 1.461$ a.u., with the corresponding scaling parameters listed in Table II, and $a_0 = 172$ a.u., which is computed from the HFD-B(HE) potential of Aziz *et al.* [11]. From the parameters in Table II, $\rho_{cs} = 8.68 \times 10^{-7}$ leads to the prediction that ^{4}He should have a metastable gaseous BEC phase below a density of $\rho_c = 8.68 \times 10^{-7}(1/\beta_6^3) = 5.58 \times 10^{15}$ 1/cm^3.

We emphasize that the results such as $\rho_{cs} = 8.68 \times 10^{-7}$ for $a_{0s} = 16.9$, and the corresponding equation of state shown in Fig. 1 (including its predictions of equilibrium density and equilibrium energy per particle for the liquid phase [10], which will be studied in more detail elsewhere) are all universal properties at length scale β_6, applicable not only to ^{4}He, but any system with the same type of long-range interaction and the same scaled scattering length. For example, they would apply to ^{85}Rb with its scattering length tuned, via a Feshbach resonance [23,24], to $a_0 = 16.9\beta_6 = 2780$ a.u. For such a system, the same $\rho_{cs} = 8.68 \times 10^{-7}$ would translate into a critical density of $\rho_c = 8.68 \times 10^{-7}(1/\beta_6^3) = 1.32 \times 10^{12}$ 1/cm^3. This result also implies that a gaseous ^{85}Rb BEC with a density of 1.32×10^{12} 1/cm^3 would become unstable beyond a critical scattering length of 2780 a.u. As another example, for $a_{0s} = 10$, the theory predicts a critical scaled density of $\rho_{cs} = 4.59 \times 10^{-6}$. From the scaling parameters in Table II, this translates into a critical density of 6.98×10^{12} 1/cm^3 for ^{85}Rb, assuming its scattering length is tuned to $a_0 = 10\beta_6 = 1643$ a.u. For ^{23}Na, it translates to a critical density of 4.27×10^{13} 1/cm^3 for a scattering length tuned to $a_0 = 10\beta_6 = 898.6$ a.u. Such densities and scattering lengths are easily achievable under existing experimental conditions [19].

These results are consistent with and offer an explanation for the experiment by Claussen *et al.* [19], in which excessive loss of atoms from a gaseous condensate (more than what can be attributed to three-body recombination [17,18]), has been observed for large scattering lengths. It is easily verified that all the experimental conditions under which this excessive loss has been observed in [19] correspond to having scattering lengths greater than the critical scattering length. The theory also explains the similarity of behaviors observed in experiments in which the scattering length is tuned to either a negative value [25] or a large positive value [19], both leading, in our picture here and in Ref. [10], to a quantum phase transition from a gaseous to a liquid phase. In fact, a more systematic experimental study, similar to the one by Claussen *et al.* [19], should allow for a direct experimental measurement of a considerable portion of the universal phase diagram corresponding to large scaled scattering lengths.

While the phase diagram that we have presented may be of more direct experimental interest, also important are the

TABLE I. Selected data of critical densities and other results. The first two columns give the critical density as a function of the scattering length. The inverse of these results gives the critical scattering length as a function of density. E_{cs}/N is the energy per particle at the critical density. λ_{cs} is the pair correlation energy at the critical density. ϵ_{ms} is the energy of a molecule in its least-bound s state, as determined from the scattering length using the quantum-defect theory of Ref. [6]. Note that λ_{cs} is always smaller than ϵ_{ms}. The numbers are all scaled quantities and applicable to all quantum systems with the same scaled scattering length.

a_{0s}	ρ_{cs}	E_{cs}/N	λ_{cs}	ϵ_{ms}
16.9	8.68×10^{-7}	-2.12×10^{-3}	-4.24×10^{-3}	-3.80×10^{-3}
14.0	1.58×10^{-6}	-3.15×10^{-3}	-6.31×10^{-3}	-5.64×10^{-3}
10.0	4.59×10^{-6}	-6.45×10^{-3}	-1.29×10^{-2}	-1.15×10^{-2}
5.00	4.51×10^{-5}	-2.99×10^{-2}	-5.98×10^{-2}	-5.31×10^{-2}
0.00	3.93	-147	-129	-72.8

conceptual insights the theory is starting to provide us on the very nature of the liquid states. (a) It resolves the apparent contradiction that ^{4}He has a liquid BEC phase but a positive scattering length, a characteristic usually associated with a gaseous BEC. The answer is that they both exist, depending on the density. (b) It shows explicitly that the gaseous and liquid states are different branches of many-atom states with the same long-range, $F = 1$, but different short-range correlations. (c) The liquid states can be correlated to the molecular states. In particular, the liquid states discussed here and illustrated in Fig. 1, which we call the first branch of liquid, evolve from the least-bound (the most highly excited) vibrational state of a diatomic molecule. A liquid state is first formed when the average interparticle separation becomes sufficiently small that the wave function of a would-be molecule in its least-bound state starts to overlap with other atoms. This is the physical origin of both the existence of the critical density and its sensitive dependence on the scattering length. Beyond the critical density, the liquid states emerge from the least-bound molecular state by allowing the atoms in a would-be molecule to move free from each other, further lowering their energy, as illustrated in Table I, which shows that the pair correlation energy at the critical density is always lower than the energy of a molecule in its least-bound vibrational state. (d) For any system other than helium, we expect that there would be other branches of liquid states, evolving from molecular vibrational states lower than the least-bound state, to emerge at higher critical densities.

In conclusion, we have presented a theoretical framework that gives a unified understanding of both the gaseous and the liquid states of a many-atom Bose system at zero temperature. A universal phase diagram, amenable to experimental verification, has been presented that is applicable to any Bose system with a $-C_6/r^6$ type of interaction at large interparticle separations. The theory provides us with considerable insights into the nature of liquid states that we are only beginning to understand.

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TABLE II. Scaling parameters for ^{4}He and selected alkali-metal atoms. β_6, $1/\beta_6^3$, and $s_E = (\hbar^2/m)(1/\beta_6)^2$ are the length, density, and energy scales, respectively. They are determined by the C_6 coefficient and the atomic mass.

Atom	C_6 (a.u.)	β_6 (a.u.)	$1/\beta_6^3$ (1/cm^3)	s_E (K)
^{4}He	1.461[a]	10.16	6.433×10^{21}	0.4192
^{23}Na	1556[b]	89.86	9.300×10^{18}	9.331×10^{-4}
^{85}Rb	4707[c]	164.6	1.522×10^{18}	7.558×10^{-5}

[a]From Ref. [11].
[b]From Ref. [20].
[c]From Refs. [21,22].