Consequences of the Pauli exclusion principle for the Bose-Einstein condensation of atoms and excitons

S. M. A. Rombouts, L. Pollet, K. Van Houcke,[1]

[1]*Universiteit Gent - UGent, Vakgroep Subatomaire en Stralingsfysica*
Proeftuinstraat 86, B-9000 Gent, Belgium
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The bosonic atoms used in present day experiments on Bose-Einstein condensation are made up of fermionic electrons and nucleons. In this Letter we demonstrate how the Pauli exclusion principle for these constituents puts an upper limit on the Bose-Einstein-condensed fraction. Detailed numerical results are presented for hydrogen atoms in a cubic volume and for excitons in semiconductors and semiconductor bilayer systems. The resulting condensate depletion scales differently from what one expects for bosons with a repulsive hard-core interaction. At high densities, Pauli exclusion results in significantly more condensate depletion. These results also shed a new light on the low condensed fraction in liquid helium II.

Recent experiments with ultracold fermionic gases have demonstrated the gradual crossover between a Bose-Einstein condensate of two-fermion molecules and a BCS-like condensate of fermion pairs [1–3]. Turning this picture around, one might ask to what extent subatomic degrees of freedom play a role in Bose-Einstein condensates of bosonic atoms, because these atoms are made up of fermions: electrons and nucleons. From the energetic point of view there is no effect: subatomic excitation energies greatly exceed the thermal energy scale of Bose-Einstein condensation. Therefore one can safely assume that the subatomic degrees of freedom are completely frozen [4]. However, even for a frozen internal structure one has to take into account the correct symmetries: at the level of the many-electron wave function, quantum mechanics dictates antisymmetry, which makes that electrons can not overlap. As a consequence, the Pauli principle for the electrons limits the available phase space for the bosonic atoms, which can have an influence on the properties of the condensate [5–11]. It has been demonstrated before that a condensate of bosons made up of fermions has a maximum occupation number [12]. For hydrogen atoms, that number corresponds to a condensate density of the order of $1/(4\pi a_0^3)$, with a_0 the Bohr radius. Such high densities are not reached in present day experiments on Bose-Einstein condensates [13]. Still, the Pauli principle can have an effect also at lower densities, where it leads to condensate depletion. It is generally believed that it is sufficient to model this effect through an effective interaction for the bosons which is strongly repulsive at short distances, like a hard-sphere potential or e.g. the (unphysical) r^{-12} term in the Lennard-Jones potential. The condensate will be depleted, simply because of the excluded volume. However, the only physical parameter which determines the low-density properties of the condensate is the scattering length. It is demonstrated below that any bosonic interaction with the right scattering length fails to reproduce the Pauli exclusion effect at high densities. We show how Pauli exclusion puts an upper bound on the Bose-Einstein condensed fraction of ultracold atomic gases. The bound is made quantitative for hydrogen atoms, through the use of an exactly solvable pairing model.

The consequences for ultracold alkali gases, exciton condensates in semiconductors and liquid helium II are discussed.

Following Penrose and Onsager [17], one can define a Bose-Einstein condensate by looking at the one-boson density matrix $\rho_B(\mathbf{r},\mathbf{r}')$ of a many-boson system,

$$\rho_B(\mathbf{r},\mathbf{r}') = \langle\Phi_B|b_{\mathbf{r}'}^\dagger b_{\mathbf{r}}|\Phi_B\rangle, \qquad (1)$$

with $b_{\mathbf{r}}^\dagger$ the operator that creates a boson at position $\mathbf{r}$ and Φ_B the many-boson wave function. The system is said to exhibit Bose-Einstein condensation if the one-body density matrix has an eigenvector $\psi_B(\mathbf{r})$ with an eigenvalue λ_B of the same order as the total number of bosons, N. The ratio $f_B = \lambda_B/N$ gives the condensed fraction, and the eigenvector $\psi_B(\mathbf{r})$ corresponds to the order parameter of the condensate.

Taking into account that atomic bosons are actually made up of fermions, one realizes that the many-boson state $|\Phi_B\rangle$ corresponds at a more microscopic level to a many-fermion wave function $|\Phi_F\rangle$. For bosons made up of two fermions, the bosonic one-body density matrix of Eq.(1) can be related to the fermionic two-body density matrix. Grouping the fermion pair states in a single coordinate $\mathbf{R} = (\mathbf{r}_1,\mathbf{r}_2)$, one can write the fermionic two-body density matrix as a square matrix $\rho_F(\mathbf{R},\mathbf{R}')$. A Bose-Einstein condensate would show up as an eigenvector $\psi_F(\mathbf{R})$ of the fermionic two-body density matrix with a macroscopic eigenvalue λ_F [18]. If the bosons correspond to strongly bound pairs of fermions, one can expect the bosonic and the fermionic picture of the condensate to be equivalent [5], with

$$\int \psi_B(\mathbf{r})b_{\mathbf{r}}^\dagger d\mathbf{r} \equiv \int \psi_F(\mathbf{r}_1,\mathbf{r}_2)a_{\mathbf{r}_1}^\dagger a_{\mathbf{r}_2}^\dagger d\mathbf{r}_1 d\mathbf{r}_2, \qquad (2)$$

$$\lambda_B = \lambda_F. \qquad (3)$$

The fermionic model has the Pauli correlations between the fermions taken into account, while the bosonic model does not. Unfortunately, for any realistic model the fermionic many-body problem is too complicated in order to determine $\rho_F(\mathbf{R},\mathbf{R}')$ accurately.

A variational approach is feasible: λ_F is an eigenvalue of the many-body operator $B^\dagger B$, with the operator $B^\dagger$ defined as

$$B^\dagger = \int \psi_F(\mathbf{r}_1, \mathbf{r}_2) a_{\mathbf{r}_2}^\dagger a_{\mathbf{r}_1}^\dagger d\mathbf{r}_1 d\mathbf{r}_2. \qquad (4)$$

If one knows the structure of the order parameter ψ_F of the fermionic pair condensate, one can determine an upper limit for the eigenvalue λ_F:

$$\lambda_F = \langle \Phi_F | B^\dagger B | \Phi_F \rangle < |E_P|, \qquad (5)$$

where E_P is the ground state energy of a fermionic pairing Hamiltonian, $H_P = -B^\dagger B$. This Hamiltonian is not meant to be phenomenological, but it is useful here because it is integrable [19] and exactly solvable [20]. Therefore, given the pair function $\psi_F(\mathbf{r})$, one can determine the ground state energy $E_P(2N)$ of H_P for $2N$ fermions, and make a rigorous variational statement about the boson condensed fraction:

$$f_B \leq \frac{|E_P(2N)|}{N}. \qquad (6)$$

This upper bound is valid at all temperatures, but will be most stringent at zero temperature, because there one expects the largest condensed fraction. The resulting eigenstate is not meant as an approximation to the true state $|\Phi_f\rangle$, because Eq.(6) is a variational statement on the condensed fraction, not on the energy.

To determine the upper bound of Eq.(6), one needs to know the bosonic order parameter in terms of the fermionic degrees of freedom. Although the following treatment is based on a hydrogen 1s wave function, the procedure is more general. Alkali atoms such as ^{7}Li, ^{23}Na or ^{87}Rb can be seen as a paired state of a valence electron and a singly ionized core atom. Because the tail of the wave function for the valence electron is similar to the hydrogen 1s wave function, we expect qualitatively the same Pauli effects as for hydrogen atoms. The Bloch-Messiah theorem says that it is always possible to write a fermion pair wave function in the form $B^\dagger = \sum_i \psi_F(i) a_i^\dagger a_{-i}^\dagger$, where the index i does not necessarily refer to momentum states. The ground state energy of the corresponding pairing Hamiltonian $H_P = -B^\dagger B$ gives the upper bound for the condensed fraction. In the low-density limit this results in $f_B \leq 1 - N\sum_i |\psi_F(i)|^4$, while the upper bound in the high-density limit follows from the absolute maximum occupation number for the pair condensate [12]: $f_B \leq \frac{(\sum_i |\psi_F(i)|)^2}{4N}$. At intermediate densities one can evaluate the exact solution for the ground state energy of H_P (see below).

Here we consider hydrogen atoms in a cubic volume V with periodic boundaries, because for this case the wave function is known analytically, and therefore we can determine the upper bound of Eq.(6) easily. At low temperatures and densities, one can expect that the protons and electrons form hydrogen atoms and that all atoms are in a 1s state. Because of translational invariance, the bosonic condensate order parameter will be a uniform function in the center-of-mass coordinate of the atoms. The resulting pair operator can be written as

$$B^\dagger = \sum_{\mathbf{k}} \psi_F(\mathbf{k}) a_{\mathbf{k},p}^\dagger a_{-\mathbf{k},e}^\dagger, \qquad (7)$$

where the sum runs over all momentum states allowed by the periodic boundary conditions of the cubic volume V, with the subscripts p and e distinguishing between protons and electrons and $\psi_F(\mathbf{k})$ the pair wave function in momentum space,

$$\psi_F(\mathbf{k}) = \frac{\sqrt{Z}}{(1 + a_0^2 k^2)^2}, \qquad (8)$$

with $Z = 64\pi a_0^3/V$. As discussed above, one can assume the internal structure of the hydrogen atoms to be completely frozen at temperatures of the order of $1\mu K$, relevant for Bose-Einstein condensation [4]. Therefore we can rely on the pair operator of Eq.(7) in order to evaluate the upper bound of Eq.(6).

To find the ground-state energy of H_P for N pairs, one has to solve the following set of equations [20]:

$$\frac{1}{y_i} + \sum_{j \neq i} \frac{1}{y_i - y_j} = F(2Z y_i), \quad \forall i = 1, \ldots, N-1, \qquad (9)$$

with the function $F(x)$ given by

$$F(x) = \frac{32}{\pi} \int_0^{+\infty} \frac{q^2 dq}{x - (1 + q^2)^4}. \qquad (10)$$

The energy E_P is given by $E_P = \sum_{i=1}^{N-1} \frac{1}{y_i} - 1$. The low density limit is obtained by taking the limit $Z \to 0$ for a fixed number of pairs N. One obtains, for $n_B a_0^3 \ll 1$, that

$$f_B \leq 1 - \frac{33\pi}{2}(n_B a_0^3). \qquad (11)$$

The result of Eq.(11) was obtained without reference to interactions. At the bosonic level the Pauli blocking between the constituing fermions results in a repulsive interaction between atoms at short distances. This interaction can be modeled using a pseudopotential

$$V(r) = \frac{4\pi\hbar^2 a_s}{m} \delta(r) \frac{\partial}{\partial r} r, \qquad (12)$$

with m the atomic mass. For non-interacting hard spheres, one can identify the scattering length a_s with the radius [21, 22]. The Bogoliubov approximation, which applies in the low-density limit, results at zero temperature in a condensed fraction that scales as [22, 23]

$$f_B = 1 - \frac{8}{3\sqrt{\pi}} \left(n_B a_s^3\right)^{\frac{1}{2}}. \qquad (13)$$

The scaling with a power $1/2$ assures that the variational bound Eq.(11) is fulfilled. However, one observes that the Bogoliubov result is fundamentally different from the expression of Eq.(11) because it scales differently. This can be explained by the fact that the ground state wave function is not uniform. In fact, one can expect a higher amplitude for configurations where the bosons are well separated than for configurations where bosons nearly overlap [24]. Consequently, it turns out

that Pauli blocking is only a second order effect in the low-density limit. The leading order is determined by bosonic many-body physics, which not only tries to avoid overlapping atoms, but also tries to minimize the energy.

The high density limit can be derived from the maximal occupation number of the pair state [12], which for hydrogen results in $f_B \leq \frac{1}{4\pi n_B a_0^3}$. At high densities one has to assess the effects of the interactions with the other atoms. Calculations with a quantum Monte Carlo method [25] and a constrained variational method [26] have shown that for an interaction with scattering length a_s the condensed fraction is well described by the Bogoliubov approximation of Eq.(13), up to densities of the order of $n_B \sim 10^{-3} a_s^{-3}$. For hydrogen atoms the s-wave scattering length is given by $a_s = 0.41 a_0$ [27]. One can see in Fig. 1 that at a density of $n_B = 10^{-3} a_s^{-3} = 1.5 \times 10^{-2} a_0^{-3}$, the Pauli effect results in a condensate depletion of the order of 36%, while hard spheres with a radius a_s, and hence any interaction with the same scattering length, yield a depletion of only 5%. One can conclude that at densities of the order of $10^{-3} a_0^{-3}$ and higher the Pauli exclusion effect results in a significantly stronger condensate depletion than the hard-sphere potential, and that an effective two-body interaction for the bosons is not able to reproduce this effect.

However, at high densities one also has to consider another effect: due to interactions with electrons and protons in neighbouring atoms, the intrinsic wave function of the atoms might get deformed from the standard 1s wave function of Eq.(8). A way to take these interactions into account is to use a screened Coulomb potential, such as the Hulthén potential [28, 29]: $V_H(r) = -\frac{4\gamma V_0}{e^{2\gamma r/a_0}-1}$, with V_0 the hydrogen 1s binding energy and γ a dimensionless parameter proportional to the screening constant as defined in the Debeye-Hückel or Thomas-Fermi models. This is a phenomenological way to take screening into account, which could deviate from the true microsopic behaviour at high densities. The intrinsic wave function in momentum space becomes

$$\psi_F(\mathbf{k}) = \frac{\sqrt{Z(1-\gamma^2)}}{\left[(1-\gamma)^2 + a_0^2 k^2\right]\left[(1+\gamma)^2 + a_0^2 k^2\right]}. \tag{14}$$

In theory, at very high densities the system could undergo a Mott transition, where electrons and protons are no longer bound to each other in hydrogen atoms, but rather form a plasma. For cold hydrogen atoms this is not a realistic scenario, but it does apply to excitons in semiconductors, which have a similar intrinsic structure. The Mott transition occurs at an exciton density of $n_c \simeq 0.02 a_x^{-3}$, with a_x the exciton Bohr radius [30]. This behavior should be reflected in the density dependence of the screening parameter γ: at very low densities γ should tend to zero, in order to recover the hydrogen wave function, Eq.(8), from the Hulthén wave function, Eq.(14). The Mott transition, on the other hand, would require the bound states of the Hulthén potential to disappear around the transition density. As this happens at $\gamma = 1$, we follow Ref. [31] to take $\gamma = \sqrt{n_B/n_c}$. The variational bound for the condensed fraction can be obtained for any density by solving

the eigenvalue equations of the pairing Hamiltonian H_P with the screened pair structure of Eq.(14). Fig. 1 shows the results
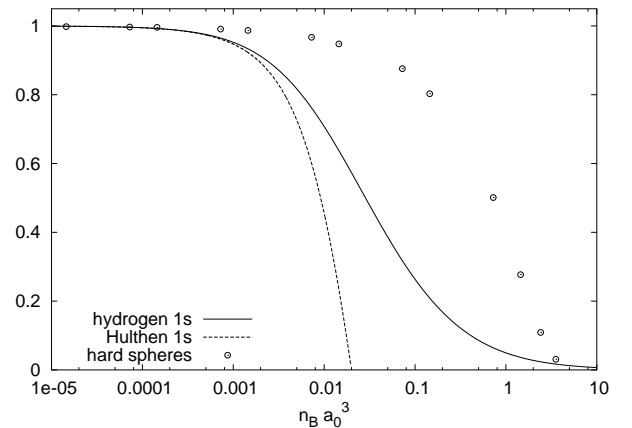

FIG. 1: Maximal condensed fraction for hydrogen 1s bosons (full line), Hulthén 1s bosons (dashed line), and bosonic hard spheres of radius a_s (circles), as a function of the density parameter $n_B a_0^3$. The hydrogen and Hulthén curves were calculated for 1000 bosons, the hard-sphere results were taken from Ref.[25].

for the hydrogen 1s and Hulthén 1s intrinsic wave functions. At the resolution of the figure the results for 1000 and 2000 particles are undistinguishable, meaning that convergence has been reached and that finite-size effects are negligible at these particle numbers. The screening effects enhance the condensate depletion even more. They become important only when the maximal condensed fraction is lower than 80%, so they are of secondary importance in the high density regime.

The densities where the Pauli effect becomes sizeable are probably out of reach for an ultracold atomic hydrogen gas, because three-body recombination processes would convert the atoms into molecules. However, the same pair structure also applies to Wannier excitons in semiconductors. There the Pauli effect might explain, together with biexciton recombination, why a clear signal of exciton condensation has not yet been observed in a three-dimensional structure. Bose-Einstein condensation of excitons has been observed in semiconducting bilayers [14]. The physics there is basically two-dimensional [15, 16]. There too the Pauli effect applies. Although an analytical expression for the intrinsic structure of these excitons is not readily available, one can estimate the Pauli effect by looking at the results of Fig. 2 for a two-dimensional hydrogen wave function, $\phi_h(k) \propto (1 + a_x^2 k^2/4)^{-3/2}$ and for a Gaussian wave function with the same low-density properties, $\phi_g(k) \propto \exp(-a_x^2 k^2/5)$, with a_x the two-dimensional excitonic Bohr radius.

If one has to treat more valence electrons independently, then the boson operator corresponds to a three- or higher-body fermion operator instead of a pair operator, and the resulting Hamiltonian $-B^\dagger B$ is no longer exactly solvable. Still, one can expect the Pauli principle to have qualitatively the same effect on e.g. ^{4}He condensates: the electrons will
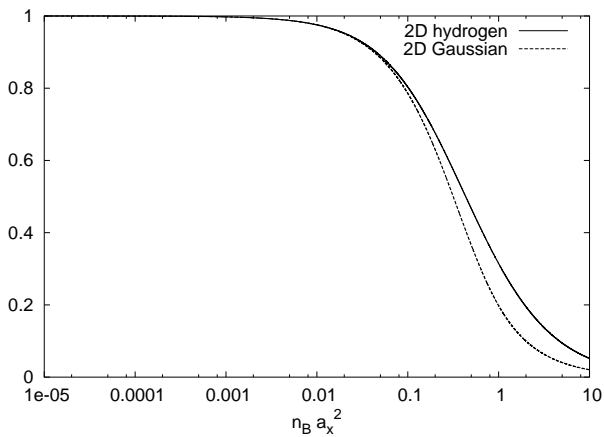
FIG. 2: Maximal condensed fraction for two-dimensional excitons with a hydrogen-like (solid line) or Gaussian (dashed line) intrinsic wave function, as a function of the exciton density parameter $n_b a_x^2$. The curves were calculated for 1000 excitons.

avoid overlap and hence limit the available phase space for the ^{4}He atoms. A uniform distribution of hard-core bosons can qualitatively explain the reduced condensed fraction in liquid helium II compared to an ultracold low-density Bose gas [17, 24]. The Pauli blocking of the underlying fermions offers a more microscopic view of this process. Pauli effects will result in a significant depletion of the condensate at densities of the order of 10^{-3} times the close-packing density or higher, and definitely at the density of liquid helium.

We have demonstrated here that Pauli blocking of the underlying electrons leads to condensate depletion in ultracold atomic gases and in exciton condensates. This effect might be measurable in systems where densities of the order of $10^{-3}a_0^{-3}$ can be reached, such as Wannier excitons in semiconductors or ^{4}He films adsorbed on porous Vycor glass [32]. This effect depends solely on the symmetry and internal structure of the wave-functions. In the high-density regime, this effect can not be modeled through an effective two-body interaction at the bosonic level. Interactions might change the internal wave function of the fermionic pairs. Our calculations based on a screened potential show that the Pauli effect dominates over the interaction effects at densities one or two orders of magnitude below the Mott transition density.

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