Quasicondensate Droplet Formation in a Gas of Trapped Atomic Hydrogen

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The formation of quasicondensate droplets in ultracold atomic gases is investigated as a first-order phase transition. Physical parameters and size distribution of metastable condensate droplets are derived from conditions of local equilibrium between quasicondensate and normal gas phases. Droplet characteristics are used for the quantitative description of recent measurements of enormous frequency shifts in the $1S$-$2S$ two-photon absorption in spin-polarized hydrogen in the normal gas phase. Theoretical evaluation of the line shape is in good agreement with experimental data.

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The experimental realization of Bose-Einstein condensation (BEC) in dilute alkali-metal atoms [1] and more recently in atomic hydrogen [2] has stimulated theoretical studies of the growth and time evolution of condensates [3]. The kinetics of the condensate formation from a strongly nonequilibrium initial state to a final macroscopic coherent state has been explored in many contexts [4], and particularly in dilute atomic gases [5,6]. Recently, effects of quasicondensates on the three-body recombination rate in a two-dimensional gas of atomic hydrogen have been observed [7]. The $1S$-$2S$ two-photon absorption line shapes measured at MIT [2] show significant distortions of the Doppler-free spectrum of the normal gas, which cannot be explained by the influence of the small fraction of atoms in the condensate. We argue that the origin of these distortions is the formation of metastable quasicondensate droplets.

In this paper, we develop a theoretical model based on a first-order phase transition treatment for the formation of metastable condensate droplets, and evaluate the effect on the $1S$-$2S$ line shape. We consider a weakly interacting trapped bosonic gas near the critical conditions for BEC. In the MIT experiment [2,8], a pronounced asymmetry in the Doppler-free spectrum of the normal gas was observed. This asymmetry appears at laser detunings corresponding to frequency shifts much larger than expected from the maximum density n_0 in the normal gas, and contains 20%–30% of the trapped atoms [9]. It implies that some fraction, much larger than the 5% of the atoms in the main condensate [2], is effectively in regions of higher density than n_0, and points to nonequilibrium density fluctuations.

The formation of metastable condensates as a first step in atomic BEC has been discussed in several theoretical models [3,5,6]. Direct experimental observations of the quasicondensate formation have not yet been reported. We show that the pronounced asymmetry in the normal gas Doppler-free spectrum [2] can be considered as a strong experimental evidence of quasicondensate droplet formation. We propose a model for the nucleation of metastable quasicondensate droplets where a droplet coherent many-body wave function covers only a small fraction of the whole volume.

The model takes into account nonequilibrium conditions that exist in the trapped gas during the measurements [10]. In the last stage of evaporative cooling, the gas is in the degenerate regime and up to 100 collisions are needed to thermalize [11]: about 10 s in [2]. During that time, the metastable gas can be described by a Boltzmann-like distribution [11], with a positive chemical potential [12] whose fluctuations provide the nucleation conditions for the quasicondensate droplets [13]. The lower states of the Bose-Einstein distribution are difficult to fill, and the distribution shows a Maxwell-Boltzmann character before equilibrium is reached [11].

We consider a spherical quasicondensate droplet locally formed by density fluctuations within the metastable gas. The droplet radius R_d is determined by the local mechanical equilibrium conditions: the pressure induced by the quasicondensate droplet equals the external pressure of the surrounding normal gas [14]. For the metastable bath gas with effective temperature T, the pressure is

$$P_g = n_g k_B T + \frac{2\pi a \hbar^2}{m} n_g^2 , \qquad (1)$$

where $n_g = n_g(\vec{r})$ is the local normal gas density, a is the scattering length, and m is the atom mass. We estimate the quantal pressure in the droplets using a solvable model for the dependence of the droplet energy E_d on the droplet radius R_d. Assuming, for simplification, that the coherent wave function of the quasicondensate is localized inside a spherical region [15] and that the quasicondensate containing $\tilde{N}$ atoms is in its ground state with entropy $S_d = 0$, we get

$$E_d = \tilde{N} \frac{\hbar^2 \pi^2}{2mR_d^2} + \frac{4\pi a \hbar^2}{mV_d} \frac{\tilde{N}(\tilde{N} - 1)}{2} \eta , \qquad (2)$$

where $V_d = 4\pi R_d^3/3$ and $\eta = 2.815$ is the correction for the finite size of the droplet. For $\tilde{N} \gg 1$, we get the droplet quantal pressure from $P = -(\partial E/\partial V)_{\tilde{N},S}$,

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$$P_d = \frac{\hbar^2\pi}{4m}\frac{\tilde{N}}{R_d^5} + \frac{9\eta}{8}\frac{\hbar^2 a}{\pi m}\frac{\tilde{N}^2}{R_d^6}. \quad (3)$$

Equating P_g and P_d, we obtain R_d as a function of T, $\tilde{N}$, a, and n_g. For hydrogen gas, $a = 1.2a_0$ and the second term in P_g is much smaller than $n_g k_B T$, and for $\tilde{N}$ not too large, we can consider the interparticle interactions as a small perturbation. Set $R_d = R_0 + \delta$, where R_0 is the droplet radius with $a = 0$, and δ is the correction due to interactions. Then

$$R_0 = \left(\frac{\hbar^2\pi\tilde{N}}{4mn_g k_B T}\right)^{1/5} = \lambda\left(\frac{\tilde{N}}{8\xi_g}\right)^{1/5}, \quad (4)$$

and $\delta \simeq (9a\eta/10\pi^2)\tilde{N}$, with the thermal de Broglie wavelength defined as $\lambda = \sqrt{2\pi\hbar^2/mk_B T}$ and $\xi_g = n_g\lambda^3$ is a dimensionless parameter. In [2], $T \sim 50\ \mu$K, and $n_g \sim 10^{14}\ \mathrm{cm}^{-3}$, so the condition $\delta/R_0 \ll 1$ is satisfied for $\tilde{N} \ll 9 \times 10^4$. From R_d, we obtain the density of the coherent phase inside a droplet

$$n_d = n_d^0\left(1 - \frac{3\delta}{R_0}\right) \quad \text{with } n_d^0 = \frac{3n_g}{\pi}\left(\frac{\tilde{N}}{\sqrt{2}\,\xi_g}\right)^{2/5}, \quad (5)$$

where n_d^0 is the droplet density for an ideal gas [16]. For large $\tilde{N}$, the droplet density is much larger than the surrounding normal gas density.

To determine the most probable number of atoms in a droplet, $\tilde{N}_d$, we follow a first-order phase transition treatment based on an activation energy similar to the phase nucleation of small droplets or bubbles in a metastable vapor-liquid system [17]. Small droplets ($\tilde{N} < \tilde{N}_d$) have large quantal energy and are dissolved back into the normal gas phase. We define an activation energy A as the difference between the free energies of the final and initial systems. For the initial system made of N atoms in the normal gas phase, the free energy is simply $F_{\mathrm{initial}} = F_g(N)$. In the final system, the appearance of one droplet containing $\tilde{N}$ atoms gives a free energy $F_{\mathrm{final}} = F_g(N - \tilde{N}) + F_d(\tilde{N})$. Assuming that the number of atoms $\tilde{N}$ converted from the gas state to the coherent state is small, $\tilde{N} \ll N$, we expand $F_g(N - \tilde{N})$ and use the definition of the chemical potential for the normal gas to obtain

$$A = F_d(\tilde{N}) - \tilde{N}\mu_g + \mathcal{O}(\tilde{N}/N). \quad (6)$$

For a weakly interacting bosonic gas, the chemical potential of the initial metastable gas phase is [18]

$$\mu_g = k_B T \ln\xi_g + \frac{4\pi a\hbar^2}{m} n_g, \quad (7)$$

and the quasicondensate droplet free energy $F_d = E_d - TS_d$ is simply given by E_d (since $S_d = 0$). Thus

$$F_d = \tilde{N}\frac{\hbar^2\pi^2}{2mR_d^2} + \frac{2\pi a\hbar^2\eta}{m}\frac{3\tilde{N}^2}{4\pi R_d^3}. \quad (8)$$

The activation energy A becomes (with $\delta/R_0 \ll 1$)

$$A = A^0 + \frac{2\pi a\hbar^2}{m}\left[\frac{2}{5}n_d^0\eta - 2n_g\right]\tilde{N}, \quad (9)$$

with the noninteracting contribution A^0 given by

$$A^0 = \tilde{N}\frac{\hbar^2\pi^2}{2mR_0^2} - \tilde{N}k_B T\ln\xi_g, \quad (10)$$

where R_0 and n_d^0 are both functions of $\tilde{N}$. In Fig. 1, A and A^0 are shown as a function of $\tilde{N}$ for various conditions. The effect of a is to reduce the probability of the formation of large size nascent droplets. The position $\tilde{N}_d$ of the maximum value of A is also illustrated in Fig. 1. For $\tilde{N} < \tilde{N}_d$, the droplet will minimize its free energy by lowering $\tilde{N}$ until it disappears; such droplets are unstable and will vaporize back into the normal gas phase. Droplets with $\tilde{N} > \tilde{N}_d$ will decrease their free energy by growing: they are stable [17]. The maximum of A gives a threshold value A_{th} which determines if a droplet is stable or not. This model is well behaved for $\xi_g > 1$: near the critical conditions for BEC for an ideal infinite homogeneous bosonic gas, $\xi_g = 2.612$.

Setting $dA/d\tilde{N} = 0$, we obtain $\tilde{N}_d$. In what follows, we consider the case of an ideal gas [19]. From $dA^0/d\tilde{N} = 0$, we get $\tilde{N}_d^0$ and the corresponding A_{th}^0

$$\tilde{N}_d^0 = \left(\frac{3\pi}{5}\right)^{5/2}\frac{\xi_g}{4}(\ln\xi_g)^{-5/2}, \quad (11)$$

$$\frac{A_{\mathrm{th}}^0}{k_B T} = \left(\frac{3\pi}{5}\right)^{5/2}\frac{\xi_g}{6}(\ln\xi_g)^{-3/2}. \quad (12)$$

The formation time τ_f of a droplet containing $\tilde{N}$ atoms can be estimated via $\tau_f \sim \tau_g\tilde{N}^{1/2}$ [5,20], where $\tau_g^{-1} = 4\pi\hbar a n_g/m$. In [2], $n_g \sim 10^{14}\ \mathrm{cm}^{-3}$, so $\tau_g = 200\ \mu$s. Although density fluctuations in the normal gas may create droplets of any size, the smallest stable droplets containing $\tilde{N}_d^0$ atoms are formed faster than the larger ones. The probability of having a nascent droplet with $\tilde{N}_d^0$ is given by $P(\tilde{N}_d^0) \propto \exp(-A_{\mathrm{th}}^0/k_B T)$, and its density n_d^0 by $n_d^0 = 9n_g/10\ln\xi_g$. For atomic hydrogen gas at

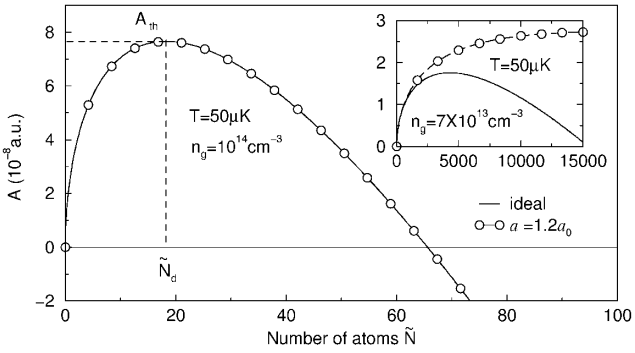

FIG. 1. Plot of A as a function of $\tilde{N}$ with and without interactions. The quantities A_{th} and $\tilde{N}_d$ regulate the stability of the droplets (see text). As shown in the inset, the activation energy is increased and the probability of droplet formation is reduced when the effect of repulsive interactions is considered.

$T = 50\ \mu$K, there is no droplet phase for $n_g < 6.7 \times 10^{13}\ \mathrm{cm}^{-3}$, which corresponds to $\xi_g < 1$.

Let us examine the effect of the droplet formation on the $1S$-$2S$ two-photon absorption line shape. The line profile is assumed to be due essentially to the local density shift $\Delta = \alpha n(\vec{r})$, where $\alpha = -3.8 \pm 0.8 \times 10^{-10}\ \mathrm{Hz\,cm}^3$ [8]. The line intensity at a given value of the laser detuning ω is proportional to the number of atoms absorbing the two photons with a frequency shift $\Delta = 2\omega = \alpha n_d$. The total number of atoms contained within droplets of density n_d is $\tilde{N}_d$ times the total number of droplets, assumed to be proportional to the probability $\exp(-A_{\mathrm{th}}/k_BT)$ of being formed. The droplet contribution to the line profile is therefore

$$I_d(n_g) = I_d^0 \tilde{N}_d(n_g) \exp\left(-\frac{A_{\mathrm{th}}(n_g)}{k_BT}\right), \qquad (13)$$

where I_d^0 is a normalization constant. Since the gas density n_g varies within the trap, the line profile as a function of $\omega = \Delta/2$ is

$$I_d(\omega) = \int_{n_{\min}}^{n_0} dn_g\, p(n_g) I_d(n_g) \delta[2\omega - \alpha n_d(n_g)], \qquad (14)$$

where $n_{\min} = \lambda^{-3}$ corresponds to $\xi_g = 1$. Here, $p(n_g)$ is the probability of having a local density of n_g in the line of sight of the laser beams, and it is given by

$$p(n_g) \equiv \frac{1}{N_g} \int d^3r\, n(\vec{r}) P_L(\vec{r}) \delta[n_g - n(\vec{r})], \qquad (15)$$

where N_g is the total number of atoms in the normal gas phase, and $P_L(\vec{r})$ is the laser intensity profile which depends on the geometry. For two-photon absorption of counterpropagating laser beams, $P_L(\vec{r}) = I_L^2(\vec{r})/I_0^2$, with axial symmetry [2,8] such that $I_L(\vec{r}) = I_L(\rho) = I_0 \exp(-\rho^2/2\sigma_L^2)$, where σ_L is the laser intensity half-width and I_0 its intensity. For the normal gas density, we consider $n(\vec{r})$ to be Gaussian with axial symmetry [21]

$$n(\vec{r}) = n(\rho, z) = n_0 \exp\left[-\frac{m}{2k_BT}(\omega_\rho^2 \rho^2 + \omega_z^2 z^2)\right], \qquad (16)$$

with the maximum density n_0 given by $n_0 = N_g (m/2\pi k_BT)^{3/2} \omega_\rho^2 \omega_z$. The probability $p(n_g)$ becomes

$$p(n_g) = \frac{2}{\sqrt{\pi}} \frac{1}{n_0} \left(\frac{n_g}{n_0}\right)^\nu \sqrt{\ln\left(\frac{n_0}{n_g}\right)}, \qquad (17)$$

where $n_g < n_0$, and $\nu = 1/3$ for the MIT experiment [22]. In Fig. 2, we illustrate $I_d(n_g)/I_d^0$ as well as $p(n_g)$ and $p(n_g) I_d(n_g)/I_d^0$ for an ideal gas.

Similarly, the normal gas line profile is given by [23]

$$I_g(\omega) = I_g^0 \int_0^{n_0} dn_g\, p(n_g) \delta(2\omega - \alpha n_g), \qquad (18)$$

$$= \frac{I_g^0}{|\alpha|} \left(\frac{2\omega}{\alpha}\right)^\nu \frac{2}{\sqrt{\pi}} \frac{1}{n_0} \sqrt{\ln\left(\frac{\alpha n_0}{2\omega}\right)}. \qquad (19)$$

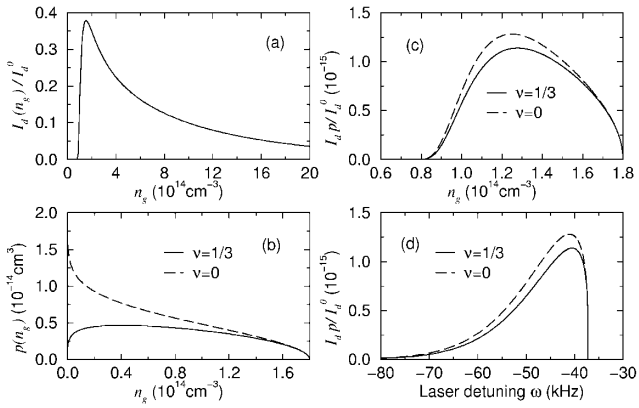

FIG. 2. Plot of (a) $I_d(n_g)/I_d^0$, (b) $p(n_g)$, and (c) $p(n_g) I_d(n_g)/I_d^0$ as a function of n_g for an ideal gas ($a = 0$) with $n_0 = 1.8 \times 10^{14}\ \mathrm{cm}^{-3}$ and $T = 50\ \mu$K: $p(n_g)$ cuts the long tail of $I_d(n_g)/I_d^0$. (d) shows the same as (c) but as a function of the laser detuning ω. In (b)–(d) we give the curves for two values of ν corresponding to the actual experiment geometry ($\nu = 1/3$) and the case of a uniform laser intensity ($\nu = 0$).

The frequency ω is limited to values between 0 and $\alpha n_0/2$. In Fig. 3, we show the contribution of the normal gas and quasicondensate droplet phases to the line profile of an ideal gas corresponding to the experimental conditions [2]. The normal line alone cannot reproduce

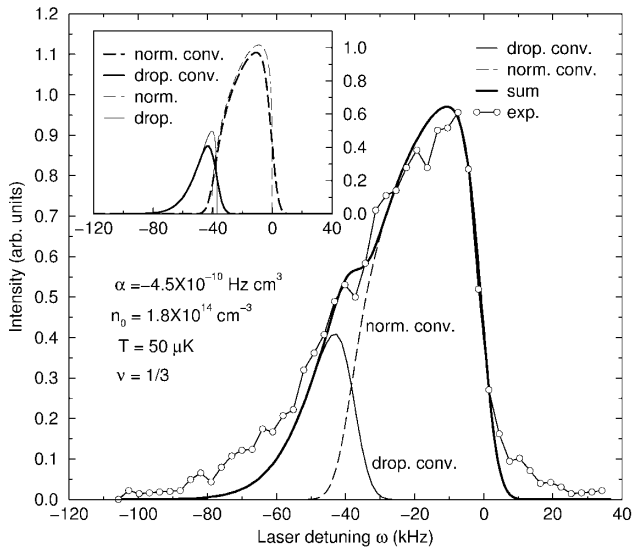

FIG. 3. Plot of $I_d(\omega)$ and $I_g(\omega)$ as a function of the laser detuning ω for an ideal gas with the experimental geometry. The normal and droplet lines have been convoluted with a Gaussian of 3 kHz width (corresponding to the experimental frequency step). The sum of those two lines is compared to the experimental signal obtained in [2]. The inset shows the normal line with (thick dashed line) and without (thin dashed line) convolution, and the droplet line with (thick solid line) and without (thin solid line) convolution. From the intensity of the droplet line, we determine that the droplets contain 20% of all atoms and that $\alpha = -4.5 \times 10^{-10}\ \mathrm{Hz\,cm}^3$.

the experimental curve: the droplet line contribution gives rise to a long tail at large detunings as well as a shoulder. The large asymmetry measured in the Doppler-free normal gas line shape is well reproduced by the model, and the experiment [2] provides a strong evidence of quasicondensate formation.

Because the data were taken over a period of a second [9], growth and decay of the metastable droplets may play an important role in explaining the large spread of the measured line. However, since the time evolution of the droplet line is slow (it can be observed for many seconds [9]), the static model presented here should be broadly applicable. A more thorough study, including the role of interactions, and the time evolution of the droplets, should help in the design of experiments to determine the detailed properties of the droplets.

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[12] Other studies [6] also use a system with depleted lower states and a positive chemical potential.

[13] Spatial density fluctuations of the metastable gas create fluctuations of the value of the positive chemical potential, which can be seen as an internal random potential for the single particle atomic wave function in the nonlinear equation.

[14] The nascent coherent phase, while attempting to spread throughout the trap, will interact with the surrounding thermal atoms, and its wave function final size will be attained as local equilibrium is reached.

[15] The surrounding normal gas restricts the coherent wave function spatial extension. If the infinite wall constraint is relaxed, the quasicondensate wave function penetrates the walls, and pressure and droplet density decrease.

[16] When $R_d \sim \lambda$, the extension of the quasicondensate wave function leads to a numerical factor β in the first term in Eq. (2), together with a different value of η. For a realistic box where the vertical wall is replaced with a Gaussian of width λ, we obtain $\beta \sim 0.8$ and $\eta \sim 1.5$, which do not greatly affect our results.

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[19] The $a \neq 0$ case needs to be solved numerically.

[20] The formation time can be estimated from local density fluctuations. See also [5].

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[22] This functional dependence is valid for σ_L larger than the equivalent width of the normal gas distribution. For $T = 50\ \mu$K, $n_0 = 1.8 \times 10^{14}\ \mathrm{cm}^{-3}$, and $\sigma_L = 50\ \mu$m, $\nu = 1/3$: this value varies with T, n_0, and σ_L. If the laser intensity is spatially constant (or $\sigma_L \to \infty$), $\nu = 0$.

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