Phase Separation of Bose-Einstein Condensates

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We distinguish two types of spatial separation exhibited by atomic trap Bose-Einstein condensates: potential separation, in which case the condensates diffuse into each other as the trap is opened adiabatically, and phase separation, in which case the separation persists in the absence of external potentials. We discuss relevant features of the dynamics and statics of the phase separation of dilute condensates. [S0031-9007(98)07928-9]

PACS numbers: 03.75.Fi, 05.30.Jp, 32.80.Pj, 67.90.+z

As dilute gases, the atomic trap Bose-Einstein condensates [1] occupy a unique position among the superfluid systems. In this context, the study of multiple atomic condensates is particularly interesting, as they represent the first laboratory mixtures of distinguishable boson superfluids [2–6]. At the present time, two experimental groups have observed trapped multiple condensates [7– 9]. In both experiments, condensates have been observed to partially separate in space.

In addition to the insights gained from other theoretical work [10-12], we point out that it is useful to distinguish two types of spatial separation: (1) potential separation, caused by the external trapping potentials in much the same way that gravity can separate fluids of different specific weight. If the trap is opened adiabatically, potential separated condensates diffuse into each other. (2) Phase separation, which persists in the absence of external potentials. In the fluid analogy, phase separated condensates can be compared to a system of two immiscible fluids, such as oil and water.

This analogy also suggests an important qualitative difference in the dynamics of both types of separated condensates: "stirring" phase separated systems, such as oil and water, can produce more droplets but does *not* mix the fluids homogeneously, unlike stirring potential separated fluids. In discussing the phase separation dynamics quantitatively, we consider a mixture of two condensates that is initially homogeneous [13]. The wave functions, ϕ_1 and ϕ_2 , of two interacting condensates evolve according to

$$i\hbar\dot{\phi}_{1} = \left[-\frac{\hbar^{2}\nabla^{2}}{2m_{1}} - \mu_{1} + \lambda_{1}|\phi_{1}|^{2}\right]\phi_{1} + \lambda|\phi_{2}|^{2}\phi_{1},$$

$$i\hbar\dot{\phi}_{2} = \left[-\frac{\hbar^{2}\nabla^{2}}{2m_{2}} - \mu_{2} + \lambda_{2}|\phi_{2}|^{2}\right]\phi_{2} + \lambda|\phi_{1}|^{2}\phi_{2},$$

(1)

where μ_j (j = 1, 2) represents the chemical potential of the *j* bosons. The interaction strengths, λ_j and λ , are determined by the scattering lengths for binary collisions of like and unlike bosons: $\lambda_j = 4\pi\hbar^2 a_j/m_j$ and $\lambda = 2\pi\hbar^2 a/m_{\rm red}$, where $m_{\rm red}^{-1} = m_1^{-1} + m_2^{-1}$. The excitations of the static homogeneous condensates, $\phi_j(\mathbf{r}, t) = \phi_j^{(0)}$, are described by fluctuations of the fields; $\phi_j(\mathbf{r}, t) = \phi_j^{(0)} + \delta \phi_j$, which evolve according to the Gross-Pitaevski equations (1), linearized in $\delta \phi$ and $\delta \phi^*$. Decomposing the field fluctuations into Fourier components, $\delta \phi_j = \sum_{\mathbf{k}} c_{j,\mathbf{k}} \exp(i\mathbf{k} \cdot \mathbf{r})$, we obtain the equations of motion for the *c* amplitudes [14],

$$i\hbar\dot{c}_{1,\mathbf{k}} = [\hbar^{2}k^{2}/2m_{1} + \lambda_{1}n_{1}]c_{1,\mathbf{k}} + \lambda_{1}[\phi_{1}^{(0)}]^{2}c_{1,-\mathbf{k}}^{*} + \lambda\phi_{1}^{(0)}[\phi_{2}^{(0)*}c_{2,\mathbf{k}} + \phi_{2}^{(0)}c_{2,-\mathbf{k}}^{*}], \qquad (2)$$

where $n_j = |\phi_j^{(0)}|^2$ and where we have used that $\dot{\phi}_j^0 = 0$. A similar equation for $i\hbar\dot{c}_2$ is obtained by exchanging the 1 and 2 subscripts. Alternatively, we can introduce the phase and density of the condensate field, $\phi = \sqrt{\rho} \exp(i\theta)$, the fluctuations of which, $\rho = n + \delta\rho$ and $\theta = \theta^{(0)} + \delta\theta$, account for the field fluctuations, $\delta\phi = \phi^{(0)}[\delta\rho/2n + i\delta\theta]$. The last term of Eq. (2) then represents a density fluctuation, $\delta\rho_{\mathbf{k}} = [\phi^{(0)*}c_{\mathbf{k}} + \phi^{(0)}c_{-\mathbf{k}}^*]$ whereas the difference, $\delta\Pi_{\mathbf{k}} = [\phi^{(0)*}c_{\mathbf{k}} - \phi^{(0)}c_{-\mathbf{k}}^*]/2i$, represents a phase fluctuation. With Eq. (2), we find

 $\hbar \delta \dot{\rho}_{1,\mathbf{k}} = 2[\hbar^2 k^2/2m_1] \delta \Pi_{1,\mathbf{k}},$

$$\hbar \delta \dot{\Pi}_{1,\mathbf{k}} = -\frac{1}{2} [\hbar^2 k^2 / 2m_1 + 2\lambda_1 n_1] \delta \rho_{1,\mathbf{k}} \qquad (3)$$
$$= \lambda n_1 \delta \rho_{2,\mathbf{k}}$$

The substitution of $\delta \rho_{j,\mathbf{k}}(t) = \delta \rho_{j,\mathbf{k}} \cos(\Omega_{\mathbf{k}} t)$ into the time derivative of the $\delta \dot{\rho}$ equations of Eq. (3) leads to the normal mode equations for the coupled density fluctuations,

$$-\Omega_{\mathbf{k}}^{2}\delta\rho_{1,\mathbf{k}} = -\omega_{1,\mathbf{k}}^{2}\delta\rho_{1,\mathbf{k}} - \lambda n_{1}\frac{k^{2}}{m_{1}}\delta\rho_{2,\mathbf{k}},$$

$$-\Omega_{\mathbf{k}}^{2}\delta\rho_{2,\mathbf{k}} = -\omega_{2,\mathbf{k}}^{2}\delta\rho_{2,\mathbf{k}} - \lambda n_{2}\frac{k^{2}}{m_{2}}\delta\rho_{1,\mathbf{k}},$$

(4)

where $\hbar \omega_{j,\mathbf{k}} = \sqrt{(\hbar^2 k^2/2m_j)^2 + (\hbar^2 k^2/m_j)n_j\lambda_j}$ (we assume $\lambda_j > 0$) denotes the usual single condensate Bogoliubov dispersion. The requirement that Eq. (4) has nontrivial solutions gives the dispersions of the double condensate excitations,

$$\Omega_{\pm,\mathbf{k}}^{2} = \frac{\left[\omega_{1,\mathbf{k}}^{2} + \omega_{2,\mathbf{k}}^{2}\right]}{2} \\ \pm \frac{\sqrt{\left[\omega_{1,\mathbf{k}}^{2} - \omega_{2,\mathbf{k}}^{2}\right]^{2} + 4(\lambda^{2}/\lambda_{1}\lambda_{2})c_{1}^{2}c_{2}^{2}k^{4}}}{2}, \quad (5)$$

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where c_j is the sound velocity of the *j* condensate, $c_j = \sqrt{n_j \lambda_j / m_j}$.

In the long wavelength limit, we find with $\omega_{j,\mathbf{q}} \approx c_j q$ that the double condensate dispersions of Eq. (5) are also phononlike, $\Omega_{\pm,\mathbf{k}} \approx c_{\pm}k$ $(k \rightarrow 0)$, with "sound velocities" c_+ and c_- equal to

$$c_{\pm}^{2} = \frac{[c_{1}^{2} + c_{2}^{2}] \pm \sqrt{[c_{1}^{2} - c_{2}^{2}]^{2} + 4(\lambda^{2}/\lambda_{1}\lambda_{2})c_{1}^{2}c_{2}^{2}}}{2}.$$
(6)

If $\lambda^2 > \lambda_1 \lambda_2$, the intercondensate interaction repels the energy levels so strongly that c_-^2 becomes negative, giving long wavelength modes that grow exponentially: the homogeneous system is unstable [15]. Interestingly, if the masses of the distinguishable bosons are equal, the double condensate dispersions are Bogoliubov-like,

$$\Omega_{\pm,\mathbf{k}}^2 = c_{\pm}^2 k^2 + (\hbar k^2 / 2m)^2.$$
(7)

In the case of instability, $c_{-}^{2} < 0$, the *k* modes of $\Omega_{-,\mathbf{k}}^{2} < 0$ grow initially at a rate $|\Omega_{-,\mathbf{k}}|$. The fastest growing modes have wave number $k_{f} = \sqrt{2} m |c_{-}|/\hbar$ and grow initially at a rate $m |c_{-}|^{2}/\hbar$. We expect that these modes set the length scale of the pattern that is formed, so that repelling condensates ($\lambda > 0$) would separate into single condensate regions a distance $2\pi/k_{f} = h/(\sqrt{2} m |c_{-}|)$ apart (on average). By the same token, we can roughly estimate the relevant time scale τ_{D} for the formation process of droplets of single condensate regions: $\tau_{D} \sim |\Omega_{-,k_{f}}|^{-1} = \hbar/(m |c_{-}|^{2})$.

Eventually, the droplets "gather," forming an equilibrium system with two single condensate regions [16]. We describe the resulting static situation by minimizing the free energy. If the spatial variations of the condensates are slow and the kinetic energy contributions may be neglected, the (zero-temperature) free energy is the integral over the free energy density $F(\mathbf{r})$,

$$F(\mathbf{r}) = \frac{\lambda_1}{2} n_1^2(\mathbf{r}) + \frac{\lambda_2}{2} n_2^2(\mathbf{r}) + \lambda n_1(\mathbf{r}) n_2(\mathbf{r})$$
$$- \mu_1(\mathbf{r}) n_1(\mathbf{r}) - \mu_2(\mathbf{r}) n_2(\mathbf{r}), \qquad (8)$$

where the effective chemical potentials $\mu_j(\mathbf{r})$ can include the external trap potentials $v_j(\mathbf{r})$ experienced by the *j* bosons, $\mu_j(\mathbf{r}) = \mu_j - v_j(\mathbf{r})$ [17]. Minimizing *F* with respect to the densities, $\delta F / \delta n_j(\mathbf{r}) = 0$, gives the Thomas-Fermi equations

$$\mu_1(\mathbf{r}) = \lambda_1 n_1(\mathbf{r}) + \lambda n_2(\mathbf{r}), \qquad (9)$$

$$\mu_2(\mathbf{r}) = \lambda_2 n_2(\mathbf{r}) + \lambda n_1(\mathbf{r}).$$

When $v_j(\mathbf{r}) = 0$, the Thomas-Fermi condensate densities of Eq. (9) are homogeneous. However, equating firstorder derivatives to zero gives only a minimum provided the second-order derivatives satisfy $(\partial^2 F/\partial n_j^2) > 0$ and $(\partial^2 F/\partial n_1^2) (\partial^2 F/\partial n_2^2) - (\partial^2 F/\partial n_1 \partial n_2)^2 > 0$. The latter condition implies that the Thomas-Fermi equations (9) give only a minimum provided the stability criteria $\lambda_1 >$ $0, \lambda_2 > 0$, and $\lambda^2 < \lambda_1 \lambda_2$ are satisfied. To see that "strongly" repulsive condensates ($\lambda > 0$, $\lambda^2 > \lambda_1 \lambda_2$) lower their free energy by distributing the condensates inhomogeneously, we write the free energy density of Eq. (8) in the absence of external potentials $v_i(\mathbf{r}) = 0$ as

$$F(\mathbf{r}) = \frac{\lambda_1}{2} [n_1(\mathbf{r}) + n_2(\mathbf{r})\sqrt{\lambda_2/\lambda_1}]^2 + [\lambda - \sqrt{\lambda_1\lambda_2}]n_1(\mathbf{r})n_2(\mathbf{r}) - \mu_1 n_1(\mathbf{r}) - \mu_2 n_2(\mathbf{r}).$$
(10)

Starting from the homogeneous overlapping condensate system, redistributing bosons 1 and 2 spatially while keeping $[n_1(\mathbf{r}) + n_2(\mathbf{r})\sqrt{\lambda_2/\lambda_1}]$ constant over space can lower the energy by decreasing the overlap integral $\int d^3r n_1(\mathbf{r})n_2(\mathbf{r})$. The lowest value is reached by spatially separating the two condensates. Although the Hamiltonian is translationally invariant $[v_j(\mathbf{r}) = 0]$, the resulting double condensate system is not: the system spontaneously breaks translational symmetry. Strongly attracting condensates ($\lambda < 0, \lambda^2 > \lambda_1 \lambda_2$) can decrease the free energy by increasing their mutual overlap: collapse. The similarity to the behavior of a single condensate of negative scattering length suggests that a trapping potential can still create small systems of strongly attracting condensates that are stable or metastable.

The free energy of the phase separated condensate system in a macroscopic volume *V* is the sum of the single condensate free energies, condensate 1 confined to a volume V_1 , and condensate 2 confined to $V - V_1$. Minimizing the total free energy with respect to V_1 gives the equilibrium condition of equal pressures exerted by both condensates. With the pressure $P_j = \lambda_j n_{j,s}^2/2$, where $n_{j,s}$ denotes the density of the single condensate regions, we are led to the equivalent condition for the condensate densities $n_{1,s} = n_{2,s}\sqrt{\lambda_2/\lambda_1}$.

We note that the assumption of slowly varying condensate wave functions, necessary to justify neglecting the kinetic energy in the free energy of Eq. (8), is violated at the boundary of the two condensates. The kinetic energy contribution gives a boundary region of finite size b, in which the wave functions smoothly tend to zero as the condensates cross the boundary. We assume that the condensates are so large that the boundary region which separates them can be approximated locally as a planar region with densities that vary spatially as functions of the coordinate zwith the z axis perpendicular to the boundary surface of area A. The kinetic energy contribution, $E_{kin}(b)$, is then approximately equal to $E_{kin}(b) \approx (A\hbar^2/2b)[n_1/m_1 + n_2/m_2] = 4AP[l_1^2 + l_2^2]/b$, where l_j represents the single condensate coherence lengths $l_j = \hbar / \sqrt{4m_j n_{j,s} \lambda_j}$ and where we have used that $P_1 \approx P_2 = P$. The overlap of the condensates in the boundary region increases the interaction energy by an amount $E_{int}(b)$, which we estimate by modeling the condensate densities in the boundary region, $z \in (0, b)$, crudely as $n_1(z) \approx n_{1,s}(b - z)/b$

and $n_2(z) \approx n_{2,s}z/b$. We find that $E_{int}(b) \approx A(\lambda - \sqrt{\lambda_1 \lambda_2})n_{1,s}n_{2,s}b/6 = AP(\lambda - \sqrt{\lambda_1 \lambda_2})b/3$. To estimate the actual boundary size \overline{b} , we minimize the boundary energy $E_b(b) = E_{kin}(b) + E_{int}(b)$ with respect to b and find

$$\overline{b} = 2\sqrt{3}\sqrt{\frac{[l_1^2 + l_2^2]}{[\lambda/\sqrt{\lambda_1\lambda_2} - 1]}}.$$
(11)

The boundary contribution to the energy is a surface energy $E_b(\overline{b}) = \sigma A$, where the surface tension σ is equal to $\sigma = (2/3)P\overline{b}[\lambda/\sqrt{\lambda_1\lambda_2} - 1] = (4/\sqrt{3}) \times \sqrt{[l_1^2 + l_2^2][\lambda/\sqrt{\lambda_1\lambda_2} - 1]P}$. In the absence of external potentials, a "droplet" of

In the absence of external potentials, a "droplet" of condensate 1 immersed in a much larger condensate 2 of density n_2 minimizes E_b by taking on the shape of a sphere of radius R, $V_1 = (4\pi/3)R^3$. We can now imagine creating the double condensate system starting from a single condensate of type 2 and replacing condensate 2 bosons in the droplet volume V_1 by condensate 1 bosons. The energy ΔE required in the replacement is equal to $\Delta E = [\lambda_1 n_{1,s}^2/2 - \lambda_2 n_{2,s}^2/2]V_1 + E_b$. Minimizing the "replacement energy" ΔE with respect to V_1 and realizing that $E_b \propto V_1^{2/3}$, we find

$$\frac{\lambda_1 n_{1,s}^2}{2} = \frac{\lambda_2 n_{2,s}^2}{2} + \frac{2}{3} \frac{E_b}{V_1}.$$
 (12)

The previous result, $\lambda_1 n_{1,s}^2/2 = \lambda_2 n_{2,s}^2/2$, obtained by ignoring the boundary energy, is therefore accurate provided the size of the droplet exceeds $R_s = 2\sigma/P = (8/\sqrt{3})\sqrt{[l_1^2 + l_2^2][\lambda/\sqrt{\lambda_1\lambda_2} - 1]}$. The energy per droplet particle, $\Delta E/N_1$, with Eq. (12) is equal to $\Delta E/N_1 = 5E_b/[3n_{1,s}V_1] = 5\sigma/[Rn_{1,s}]$, a function that decreases monotonically as N_1 increases. Consequently, it is energetically favorable for condensate 1 to gather in a single region of space, in accordance with the picture of single condensate droplets gathering in the second stage of the phase separation dynamics described above.

To describe separated double condensates in traps, we subtract the overlap term $\lambda n_1 n_2$ and include the boundary surface energy in the expression of the free energy [Eq. (8)]. The validity of this description rests on two conditions: (i) The local coherence length within each condensate is much less than the length scale on which the condensates vary spatially. (ii) The change of the potential energy across the intercondensate boundary $|\mathbf{f}_i|\overline{b}$, where $\mathbf{f}_i = -\nabla v_i$ represents the external force experienced by bosons j near the boundary, is much less than the local chemical potential, $|\mathbf{f}_i| \overline{b} \ll \lambda_i n_{i,s}$ (j = 1, 2). If these conditions are satisfied, the physics of the phase separation is similar to the above $v_i(\mathbf{r}) =$ 0 case, and we can immediately address interesting issues regarding trapped phase separated condensates. For instance, if we add a droplet of condensate 1 to a trapped condensate 2, does it "sink" to the middle of the trap, or does it remain "floating" on the surface of condensate 2?

To find the answer, we apply a reasoning that is very similar to that underlying Archimedes law for ordinary fluids. We assume that the size of the droplet is large enough to neglect the boundary surface energy and small enough to neglect the spatial variation of the density inside the droplet. The previously defined replacement energy ΔE depends on the center of mass position **R** of the droplet through the external potentials $v_1(\mathbf{r}) = v(\mathbf{r})$ and $v_2(\mathbf{r}) = \alpha v(\mathbf{r})$. Since the pressures inside and outside the droplet are equal, $\lambda_1 n_{1,2}^2/2 - \lambda_2 n_{2,s}^2/2 \approx 0$, we find that

$$\Delta E(\mathbf{R}) = \int_{V_1} [n_1(\mathbf{r})v_1(\mathbf{r}) - n_2(\mathbf{r})v_2(\mathbf{r})] d^3r$$
$$\approx N_1 v(\mathbf{R}) [1 - \alpha \sqrt{\lambda_1 / \lambda_2}].$$
(13)

Thus, even though both bosons experience a trapping potential, if $\alpha \sqrt{\lambda_1/\lambda_2} > 1$, the force on the droplet, $-N_1[1 - \alpha \sqrt{\lambda_1/\lambda_2}]\nabla v$, is directed outwards; the droplet floats.

The thinner the layer of the floating condensate, the more important are the effects of the boundary surface energy. Indeed, with only a few condensate 1 particles, the energy will be minimized by covering a fraction of the surface of condensate 2 (thereby reducing the surface boundary energy). Here we focus on the case where enough bosons have been added for condensate 1 to "wrap" around condensate 2, and we can ignore boundary surface energy effects. Within the single condensate regions, the condensates are described in the Thomas-Fermi approximation of Eq. (9) (putting $\lambda = 0$), $n_1(\mathbf{r}) =$ $[\mu_1 - v(\mathbf{r})]/\lambda_1$ and $n_2(\mathbf{r}) = \alpha [\mu_2 - v(\mathbf{r})]/\lambda_2$, where μ_1 and $\alpha \mu_2$ are the chemical potentials. In that case, the boundary surface is the equipotential surface $v(\mathbf{R}) = \mu_b$ where the pressures of both condensates, $\lambda_i n_i^2(\mathbf{r})/2$, are equal. This leads to

$$\mu_b = \mu_2 - \frac{[\mu_1 - \mu_2]}{[\alpha \sqrt{\lambda_1 / \lambda_2} - 1]}.$$
 (14)

In Fig. 1, we show a typical density profile for two separated condensates in a spherically symmetric trap. With $\lambda_j n_j(R_b) = \mu_j - v_j(R_b)$, we find for the case shown in Fig. 1 that $\mu_1 - \mu_2 = \lambda_1 n_1(R_b) - \lambda_2 n_2(R_b)/\alpha > 0$. Using $[n_1(R_b)/n_2(R_b)] = \sqrt{\lambda_2/\lambda_1}$ we then find that $\alpha \sqrt{\lambda_1/\lambda_2} > 1$, so that condensate 1 should indeed float on top of condensate 2. Of course, the experimentally relevant quantities are the number of boson particles, N_1 and N_2 , rather than μ_1 and μ_2 . The chemical potentials can be determined by inverting $N_1 = \int_{V_1} d^3r n_1(\mathbf{r})$ and $N_2 = \int_{V_2} d^3r n_2(\mathbf{r})$, where the boundary between V_1 and V_2 is defined by Eq. (14), to give $N_j(\mu_1, \mu_2)$.

In summary, we distinguish two types of spatial separation that can be exhibited by multiple condensates. Of these, we have discussed phase separation, the spatial separation that occurs even in the absence of external potentials. The small amplitude analysis of the homogeneous systems shows that an initially homogeneous multiple condensate, which satisfies the phase separation criterion,

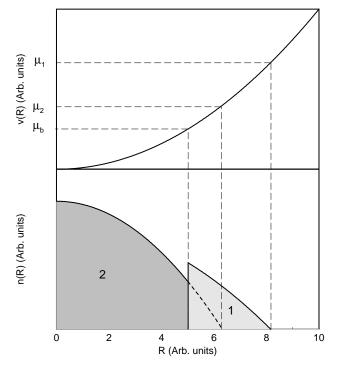


FIG. 1. Plot of a typical phase separated double condensate in a spherically symmetric trap. In reality, the boundary of condensates 1 and 2 [at $R = R_b$, where $v(R_b) = \mu_b$] is not infinitely sharp and the condensates overlap over a region of size *b* that is estimated in the text.

separates out by forming single condensate droplets which gather to form a stable separated condensate system with two single condensate regions of equal pressure. The same analysis gives the relevant time and length scales for the initial droplet formation process. In the resulting equilibrium system, the condensates are separated by a region of partial overlap, the size and surface tension of which we estimated analytically. Finally, we described phase separation of large condensates in atomic traps and discussed relevant features, such as a version of Archimedes law for condensates.

The author acknowledges fruitful interactions with Dr. P. Milonni, Dr. P. Tommasini, Professor E. Heller, Professor A. Dalgarno, and Professor K. Huang. The work of the author is supported by the NSF through a grant for the Institute for Atomic and Molecular Physics at Harvard University and Smithsonian Astrophysical Observatory. *Present address: T-4, MS B-268, Los Alamos National Laboratory, Los Alamos, NM 87545.

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