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Rarified liquid Properties of hybrid atomic-molecular BEC

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Abstract

The interactions that bring a binary atom system to an intermediate state molecule in the Feshbach resonance create in the dilute atomic Bose-Einstein condensate (BEC) a second molecular condensate component. The atomic and molecular condensates interact by coherently exchanging pairs of atoms. We discuss a signature of this coherent inter-condensate tunneling: Josephson-like oscillations of the atomic and molecular populations in response to a sudden change of the detuning. The tunneling energy depends explicitly on the volume and its dependence suggests that the ground state is a dilute BEC with the liquid-like property of a self-determined density.

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As dilute gases, the atomic Bose-Einstein condensates [1]– [3] are amenable to atomic manipulation and observation techniques. As superfluids [4], these dilute condensates exhibit an unusual degree of flexibility, promising novel studies of macroscopic quantum coherence. For instance, the notion that external fields can alter the inter-atomic interactions [5] suggests the study of condensates with inter-particle interactions that can be experimentally controlled [6]. Of the proposed schemes, the low energy Feshbach resonance [7] – [12] has attracted much attention and experimental efforts recently led to the observation of such resonances [13]– [16]. In this letter, we point out that the Feshbach resonance affects the condensate system more profoundly than altering an effective inter-particle interaction: the molecules, formed in the intermediate state of the binary atom resonance, occupy a second condensate component in the many-body BEC system. The atomic and molecular condensates interact in part by coherently exchanging pairs of atoms. This interaction implies interesting and unusual properties, of which we discuss two particularly relevant examples: (i) The molecular condensate, even if it is small in the off-resonant regime, can reveal its presence by means of Josephson-like oscillations of the atomic and molecular populations in response to a sudden change of the detuning. (ii) Close to resonance, the coherent inter-condensate tunneling binds the dilute many-body BEC to a system with the liquid-like property of a self-determined density. Thus, not only can macroscopic quantum coherence be observed differently, its manifestation can be truly novel.

In the low-energy Feshbach resonance, trapped alkali atoms interact in an external magnetic field $\mathbf{B}$. A rearrangement of the electronic spins brings the binary atom system to an intermediate bound molecular state. As states of different spin arrangement, the detuning ϵ, which is the energy difference of initial and intermediate states, depends on the magnetic field strength (ϵ proportional to the deviation of $|\mathbf{B}|$ from its resonant value). The magnetic field then provides the knob to alter and control the effective binary atom interactions.

The low-energy binary atom interactions are described by an effective scattering length that includes a resonant term. However, in spite of the BEC's diluteness, the particles in a near-resonant condensate do not interact as binary atoms. One indication that a binary

collision description becomes problematic, is the vanishing of the resonance width in the limit of zero relative velocity. As a consequence, the resonant contribution of the effective scattering length, $\sim \epsilon^{-1}$, diverges and the two interacting atoms can spend an infinite time in the molecular state as $\epsilon \to 0$.

Thus, rather than starting from the *a priori* assumption of an effective scattering length, we include the molecular states explicitly in the many-body description [17]. In second quantization, the coupling to the closed channel that creates the intermediate state molecule in the binary atom system gives the following contribution to the Hamiltonian:

$$\hat{H}_{at-mol} = \frac{\alpha}{\sqrt{2}} \int d^3r \, \left\{ \hat{\psi}_m^\dagger(\mathbf{r}) \hat{\psi}_a(\mathbf{r}) \hat{\psi}_a(\mathbf{r}) + h.c. \right\} \quad , \tag{1}$$

where $\hat{\psi}_a$ and $\hat{\psi}_m$ denote the field operators of the atoms and the quasi-bound molecules, respectively, and α represents the molecular coupling matrix element, which is energy independent in the relevant low-energy limit. In the binary atom system, Eq.(1) reproduces the correct resonant behavior. In the many-body system, we account for the elastic atom-atom, molecule-molecule and atom-molecule interactions by means of the corrresponding interaction strengths, λ_a, λ_m and λ [18]. Measuring energy relative to that of noninteracting atoms, the detuning ϵ makes its appearance as the single molecule energy, and the many-body Hamiltonian is

$$\begin{aligned} \hat{H} = & \int d^3r \, \hat{\psi}_a^\dagger \left[-\frac{\hbar^2 \nabla^2}{2m} + \frac{\lambda_a}{2} \hat{\psi}_a^\dagger \hat{\psi}_a \right] \hat{\psi}_a \\ & + \int d^3r \, \hat{\psi}_m^\dagger \left[-\frac{\hbar^2 \nabla^2}{4m} + \epsilon + \frac{\lambda_m}{2} \hat{\psi}_m^\dagger \hat{\psi}_m \right] \hat{\psi}_m + \lambda \, \hat{\psi}_m^\dagger \hat{\psi}_m \hat{\psi}_a^\dagger \hat{\psi}_a \\ & + \frac{\alpha}{\sqrt{2}} \int d^3r \left\{ \hat{\psi}_m^\dagger \hat{\psi}_a \hat{\psi}_a + \hat{\psi}_m \hat{\psi}_a^\dagger \hat{\psi}_a^\dagger \right\} \quad , \end{aligned} \tag{2}$$

where the position dependence of the field operators is understood. Bose-Einstein condensation is generally characterized by a non-vanishing order parameter, $\langle \hat{\psi} \rangle = \phi \neq 0$, which we refer to as the condensate field. We obtain the time evolution of the fields most easily from the Heisenberg equations, $i\hbar \dot{\hat{\psi}}_a = [\hat{\psi}_a, \hat{H}]$, $i\hbar \dot{\hat{\psi}}_m = [\hat{\psi}_m, \hat{H}]$. Under the assumption of complete coherence, e.g. $\langle \hat{\psi}_a \hat{\psi}_a \rangle \approx \phi_a^2$, appropriate for the dilute zero temperature conden-

sates, the expectation value of the Heisenberg equations yields coupled non-linear equations of motion that replace the single-condensate Gross-Pitaevskii equation [19]– [20]:

$$i\hbar\dot{\phi}_a = \left[-\frac{\hbar^2\nabla^2}{2m} + \lambda_a n_a + \lambda_m n_m\right]\phi_a + \sqrt{2}\alpha\phi_m\phi_a^* \quad ,$$
$$i\hbar\dot{\phi}_m = \left[-\frac{\hbar^2\nabla^2}{4m} + \epsilon + \lambda_m n_m + \lambda n_a\right]\phi_m + \frac{\alpha}{\sqrt{2}}\phi_a^2 \quad , \tag{3}$$

In the above equations, n_a and n_m denote the atomic and molecular condensate densities: $n_a = |\phi_a|^2$ and $n_m = |\phi_m|^2$. The α coupling terms describe coherent inter-condensate exchange of atom pairs. The source term $\alpha\phi_a^2/\sqrt{2}$ experienced by the molecular field, indicates that the molecular condensate forms dynamically [22] in the presence of an atomic condensate.

The current experiments resonate on molecular states of high vibrational quantum number ν. Consequently, interactions that change the molecular state (decreasing ν) are particularly relevant. The 'fragility' of these molecules is expressed by high rate constants $c_{m,a}$ and $c_{m,m}$ for state-changing collisions with atoms and other molecules (estimates [23] give $c_{m,a} \sim 10^{-9} - 10^{-11} cm^3 sec^{-1}$, as compared to $c_{a,a} \sim 10^{-14} cm^3 sec^{-1}$ for alkali atom state changing collisions). For non-condensed systems, the loss-processes can be accounted for by a rate equation, $\dot{n}_m = -[c_{m,a}n_a + c_{m,m}n_m]n_m$. For BEC systems, the same collisions affect the coherent dynamics. A lowest-order perturbation treatment modifies the $\dot{\phi}_m$-equation by rendering the interaction strengths absorptive: $\lambda \to \lambda - i\hbar c_{m,a}/2$ and $\lambda_m \to \lambda_m - i\hbar c_{m,m}/2$.

In the off-resonance limit, which we define here as $\epsilon >> \lambda n$ and $\epsilon >> \lambda_m n$, where n denotes the atomic condensate particle density, $n = n_a + 2n_m$, the molecule fraction is small and the state-changing molecular collisions destroy the condensate slowly. In spite of the smallness of the molecular condensate, the dynamical response to a sudden change of the magnetic field strength (i.e. detuning) carries a detectable signature of its presence: Josephson-like oscillations of the atomic and molecular populations. In this limit, propagation of the atomic condensate is hardly affected by the molecular condensate: $\phi_a \approx \sqrt{n}\exp(-i\lambda_a nt/\hbar)$. In the same approximation, the molecular condensate field evolves according to a linear equation:

$$i\hbar\dot{\phi}_m = \left[\epsilon(t) + \lambda n - i\frac{\gamma_m}{2}\right]\phi_m + \frac{\alpha}{\sqrt{2}} n \exp(-2i\lambda n t) \ , \tag{4}$$

where γ_m denotes the decay rate of a single molecule imbedded in the atomic condensate: $\gamma_m/\hbar = c_{ma}n$. Under a sudden change of the detuning, from $\epsilon = \epsilon_{in}$ to $\epsilon = \epsilon_f$ at $t = 0$, the molecular field, initially at $\phi_m(t = 0) = \phi_0$, evolves as

$$\phi_m(t) = \phi_\infty \exp\left[-\frac{it}{\hbar}(2\lambda_a n)\right] + \ [\phi_0 - \phi_\infty] \times \exp\left[-\frac{it}{\hbar}(\epsilon_f + \lambda n)\right] \exp\left(-\frac{\gamma_m t}{2\hbar}\right) \ , \tag{5}$$

where ϕ_∞ is the molecular field value at large times, $t >> (\hbar/\gamma_m)$, $\phi_\infty = -\alpha n/[\sqrt{2}(\epsilon_f + \lambda n - 2\lambda_a n - i\gamma_m/2)] \approx -\alpha n_a/(\sqrt{2}\epsilon_f)$. The molecular population oscillates with angular frequency $\epsilon_f + \lambda n - 2\lambda_a n \approx \epsilon_f$, and magnitude $2|\phi_\infty(\phi_0 - \phi_\infty)| \exp(-\gamma_m t/2\hbar)$, damped out on the scale of the lifetime of a single molecule. At a later time, the system has relaxed to its 'quasi-equilibrium' and the molecular field takes on its stationary value, ϕ_∞. For more general variations of the detuning in the off-resonance regime, populations oscillate if $|\dot{\epsilon}/\epsilon| << \gamma_m/\hbar$, or, since the detuning is proportional to the magnetic field variation from its resonant value B_0, if $|\dot{B}/(B - B_0)| << \gamma_m/\hbar$. In the opposite limit, $|\dot{\epsilon}/\epsilon| << \gamma_m/\hbar$, the system follows its equilibrium value adiabatically: $\phi_m(t) \approx -\alpha n(t)/\sqrt{2}\epsilon(t)$. For sudden near-resonant detuning changes [24], the qualitative picture remains: damped population oscillations appear, while molecular decay depletes the condensate, as illustrated in Fig.(1). The oscillations can be detected by near-resonant imaging of the molecules, as they give a modulation to the image intensity that oscillates at the same frequency. The cause of these oscillations, coherent inter-condensate boson exchange, is akin to Josephson inter-condensate tunneling but differs from it in a fundamental aspect: each tunneling event combines two atomic bosons to form a single molecular condensate boson. This difference spectacularly alters the near-resonant stationary state properties, as we show below.

We assume that boson decay is negligible on the time scale of observation, and that the system has reached its quasi-equilibrium [25]. The tunneling energy contribution, $\langle \hat{H}_{at-mol} \rangle = (\alpha/\sqrt{2}) \int d^3r \ \{\phi_m^* \phi_a^2 + c.c.\}$, depends on the relative phase of both condensates. This energy also exhibits an explicit dependence on the volume. To see that, we write the energy per atomic particle, E/N, of a homogeneous BEC, confined to a volume Ω:

$$\frac{E}{N} = \frac{\langle \hat{H} \rangle}{N} = \frac{N}{\Omega}\left\{\frac{\lambda_a}{2}(N_a/N)^2 + \frac{\lambda_m}{2}(N_m/N)^2 + \lambda(N_m/N)(N_a/N)\right\}$$
$$- \sqrt{\frac{N}{\Omega}}\left\{\alpha\sqrt{2(N_m/N)}\,(N_a/N)\right\} + \bar{\epsilon}\,(N_m/N) \quad , \tag{6}$$

where N_a (N_m) denotes the average number of atoms (molecules). Since $(N_a/N) = 1 - 2(N_m/N)$, the energy is a simple function of the molecule fraction. The ground state fraction follows from minimizing E/N. In Eq.(6), the interaction contribution is proportional to N/Ω, the tunneling contribution to $\sqrt{N/\Omega}$, and the detuning term does not depend explicitly on the volume.

In the ultra-dilute limit, $\Omega >> N(\lambda_r/\alpha)^2$, where λ_r is representative of the interaction strengths (λ_a, λ_m and λ), the interaction energy $\sim \lambda_r(N/\Omega)$ can be neglected. Close to resonance, $|\epsilon| << \alpha\sqrt{(N/\Omega)}$, the detuning is also negligible and the molecule fraction that minimizes the energy is $(N_m/N) \approx 1/6$ – a third of the atoms is converted to molecules. However, the pressure of the on-resonant ultra-dilute system, $P = -\partial E/\partial\Omega = -\alpha\sqrt{n}n/(3\sqrt{3})$, is negative and the system is mechanically unstable. Interestingly, this instability does not imply collapse. As the system responds to the negative pressure by decreasing its volume, the interaction energy ($\sim \Omega^{-1}$), growing faster than the tunneling energy ($\sim \Omega^{-1/2}$), can stabilize the system. In Fig.(2), we show the energy as a function of the atomic particle density. For near-resonant detuning, the tunneling energy dominates in the ultra-dilute limit, causing the energy to decrease with increasing density. In the high density limit, $\Omega << N(\lambda_r/\alpha)^2$, the interaction energy dominates and, for the interaction strengths of Fig.(2), the energy increases linearly with density. In between, the energy reaches a minimum in the density region $\sim (\alpha/\lambda_r)^2$, where interaction and tunneling energies compete. The minimum suggests that the unconfined physical system relaxes to its lowest energy state by adjusting its volume to take on the density of minimum energy. Such self-determined density is a typical liquid-like property, but the calculated values for scattering lengths and α-parameter (e.g. for Na) give self-determined densities that are not greater than the current atomic trap condensate densities: $\sim 10^{14} - 10^{15} cm^{-3}$. The low value is interesting, as the Feshbach

resonance could give the first example of a rarified liquid. The importance of the low density lies in the fact that a liquefied condensate could survive spin flip and recombination long enough for the system to be studied. Finally, we note from Fig.(2) that the self-determined density depends on the detuning, so that a variation of the detuning causes the liquefied condensate to readjust its volume.

A direct observation of the liquid-like behavior suggests itself: after switching off the trapping potential, the liquefied condensate remains as a droplet. Eventually, the droplet would fall under gravity. For this and other reasons, it might be desirable to study the system in a trap. The distinct density profile of a trapped liquefied condensate provides an alternative signature. Here we consider a shallow trap with a liquefied condensate that is large enough to justify a Thomas-Fermi description. The local density then follows from equating a local chemical potential to the difference of the system's chemical potential and the potential energy. The chemical potential depends on density *and* detuning, $\mu(n,\epsilon)$. Furthermore, not all atomic particles experience the same external potential: we assume an optical trap in which the atoms experience a trapping potential $V(\mathbf{r})$, but the molecules do not. Alternatively, we may pretend that all atomic particles experience the $V(\mathbf{r})$-potential, as long as we subtract the molecule contribution by assigning a local detuning $\epsilon - 2V(\mathbf{r})$ to the molecules. The Thomas-Fermi equation then reads:

$$\mu(n;\epsilon - 2V) = \mu_T - V \quad , \tag{7}$$

which has to be solved for the density at each position within the droplet, while requiring the pressure to vanish at the boundary (hence the density abruptly decreases from its self-determined value to zero at the edge). If the liquefied system is not too compressed, the energy per particle of a homogeneous system can be approximated as

$$(E/N) = e_s + \frac{1}{2\kappa n_s}\left(\frac{n-n_s}{n_s}\right)^2 \quad , \tag{8}$$

where the self-determined density n_s, the minimum energy e_s and the compressibility κ ($\kappa^{-1} = -\Omega\partial P/\partial\Omega = n_s^3\partial^2(E/N)/\partial n^2$) all depend on the effective detuning. The chemical potential $\mu(n;\epsilon)$, corresponding to Eq.(8), $\mu = \partial E/\partial N$, is

$$\mu = e_s + \frac{1}{\kappa n_s}\left(\frac{n-n_s}{n_s}\right) + \frac{3}{2\kappa n_s}\left(\frac{n-n_s}{n_s}\right)^2 . \tag{9}$$

Finally, in a shallow trap of potential variation significantly less than $\alpha\sqrt{n_s}$, the spatial variation of the effective detuning may be neglected, $\epsilon - 2V \approx \epsilon$, and Eq.(7) gives:

$$n(\mathbf{r}) = \frac{n_s}{3}\left[2 + \sqrt{1 + 6\kappa n_s[V_m - V(\mathbf{r})]}\right] , \tag{10}$$

where V_m denotes the potential at the equipotential surface of the boundary. The trap compresses the middle of the droplet causing a density increase that is determined by the compressibility. Note that a measurement of the profile determines the compressibility.

In summary, we have pointed out that the ground state of a Feshbach resonant BEC is a hybrid atomic/molecular condensate. The atomic and molecular condensates interact by coherently exchanging pairs of atoms, and we have discussed two of the most important implications of this manifestation of quantum coherence: Josephson-like population oscillations in response to a sudden change of the detuning, and the near-resonant liquefaction of the condensate ground state.

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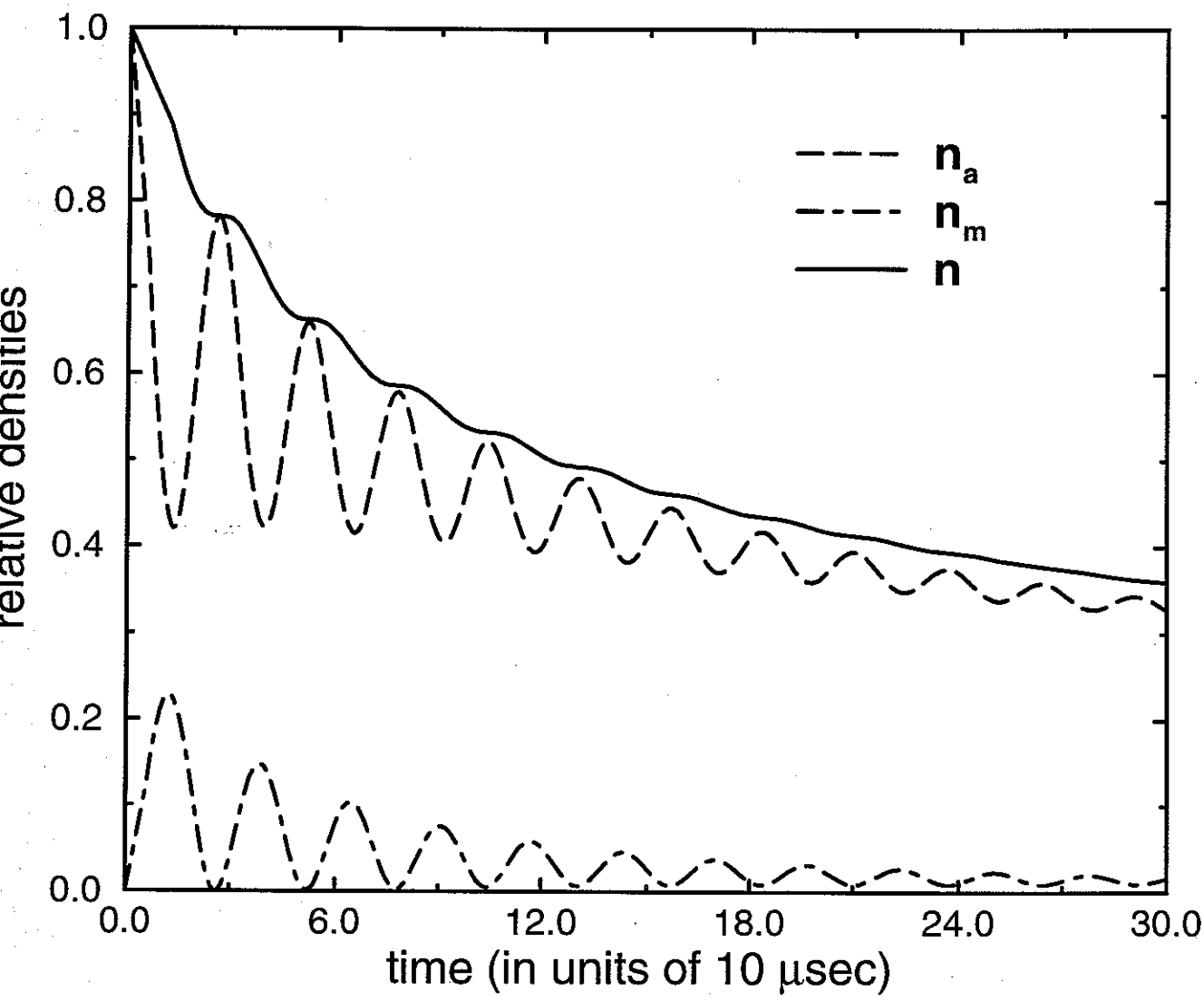
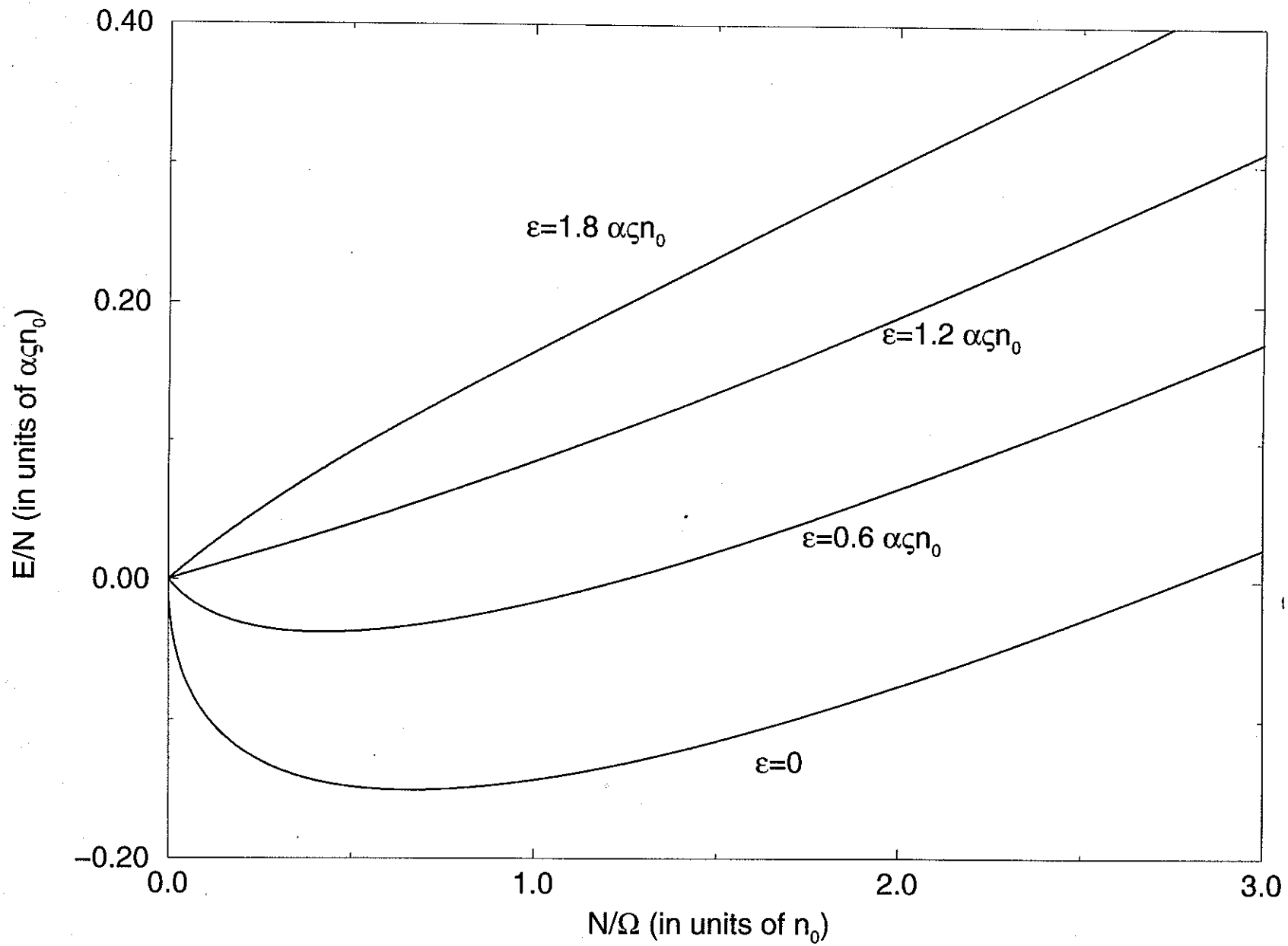
Figure Captions

Fig.1 : Plot of the particle densities: the total condensate density, $n = n_a + 2n_m$, in full line, the atomic density n_a in dashed line and the molecular density n_m in dash-dotted line. The calculation is for a homogeneous BEC that was initially in equilibrium at density $n = 10^{14} cm^{-3}$ when the detuning experienced a sudden shift from $\epsilon = 50\lambda n$ to $\epsilon = 2\lambda n$. The order of magnitude of the interaction parameters, $\lambda n = \lambda_m n = \lambda_a n = \alpha\sqrt{n} = 10^5$ Hz, and of the decay parameters, $c_{ma} = c_{mm} = 5 \times 10^{-10} cm^3 sec^{-1}$ (while neglecting atom decay) are realistic.

Fig.2 : Plot of the ground state energy per atomic particle as a function of the density at different values of the detuning, ϵ. The curves were calculated using realistic values for the parameters, with a reference density $n_0 \sim 10^{14} cm^{-3}$, and with interaction strengths $\lambda_a = \alpha/\sqrt{n_0}, \lambda_m = 2.\ \alpha/\sqrt{n_0}$, and $\lambda = 0.2\ \alpha/\sqrt{n_0}$. The densities at which the minima occur for the two curves of lowest detuning are the self-determined densities that a 'free' condensate would adopt in the ground state.


1.0
0.8
0.6
0.4
0.2
0.0
relative densities
0.0
6.0
12.0
18.0
24.0
30.0
time (in units of 10 μsec)
n_a
n_m
n


Energy per atomic particle
0.40
0.20
0.00
−0.20
0.0
1.0
2.0
3.0
E/N (in units of αςn0)
N/Ω (in units of n0)
ε=1.8 αςn0
ε=1.2 αςn0
ε=0.6 αςn0
ε=0