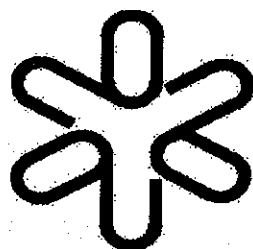


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**FESHBACH RESONANCES IN ATOMIC  
BOSE-EINSTEIN CONDENSATES**

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# Feshbach Resonances in atomic Bose-Einstein condensates

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## Abstract

The low energy Feshbach resonances recently observed in the inter-particle interactions of trapped ultra-cold atoms involve an intermediate quasi-bound molecule with a spin arrangement that differs from the trapped atom spins. Variations of the strength of an external magnetic field then alter the difference of the initial and intermediate state energies (i.e. the 'detuning'). The effective scattering length that describes the low energy binary collisions, similarly varies with the near-resonant magnetic field. Since the properties of the dilute atomic Bose-Einstein condensates (BEC's) are extremely sensitive

to the value of the scattering length, a 'tunable' scattering length suggests highly interesting many-body studies.

In this paper, we review the theory of the binary collision Feshbach resonances, and we discuss their effects on the many-body physics of the condensate. We point out that the Feshbach resonance physics in a condensate can be considerably richer than that of an altered scattering length: the Feshbach resonant atom-molecule coupling can create a second condensate component of molecules that coexists with the atomic condensate. Far off-resonance, a stationary condensate does behave as a single condensate with effective binary collision scattering length. However, even in the off-resonant limit, the dynamical response of the condensate mixture to a sudden change in the external magnetic field carries the signature of the molecular condensate's presence: experimentally observable oscillations of the number of atoms and molecules. We also discuss the stationary states of the near-resonant condensate system. We point out that the physics of a condensate that is adiabatically tuned through resonance depends on its history, i.e. whether the condensate starts out above or below resonance. Furthermore, we show that the density dependence of the many-body ground state energy suggests the possibility of creating a dilute condensate system with the liquid-like property of a self-determined density.

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## I. INTRODUCTION

The recent observation of Feshbach resonances in the inter-particle interactions of a dilute Bose-Einstein condensate of  $Na$ -atoms by Ketterle's group at MIT [1] was an eagerly anticipated event. The significance of this experimental breakthrough appears of singular importance as its consequences are far-reaching in two subfields of physics: (i). Atomic and Molecular Physics. Although predicted theoretically, technical difficulties had previously prevented the observation of the low-energy Feshbach resonances [2]. This situation abruptly changed when the experimental efforts recently culminated in the observation of resonances in ultra-cold  $^{23}Na$  at MIT, in  $^{85}Rb$  by Heinzen's group at U.T. Austin [3] and by the Wieman-Cornell collaboration at JILA [4], as well as in Cesium by Chu's group at Stanford [5]. This cascade of results indicates that the field of atomic trapping and cooling has achieved the necessary amount of control and precision to carry out systematic studies of the resonances in a variety of atomic systems. (ii). BEC-physics An important distinguishing feature of the MIT-experiment [1], is that the Feshbach resonances were observed in an atomic BEC-system [6]- [8]. The resonances were observed by varying an external magnetic field, thereby altering the 'detuning' (defined as the difference between the initial and intermediate state energies). Similarly, the effective scattering length that describes the low energy atom-atom interaction, varies with magnetic field and is consequently 'tunable'. As all quantities of interest in the atomic BEC's crucially depend on the scattering length, a tunable interaction suggests very interesting studies of the many-body behavior of condensate systems [9].

Previous searches for the low energy Feshbach resonances had been unsuccessful [2]. The difficulties that had to be overcome were multiple: the magnetic traps can only trap high field seeking states, the resonant magnetic field strengths are rather high and the margin of error on the predicted values for the resonant magnetic fields were considerable due to the uncertainties in the interatomic potential curves. Conversely, the measured values of the resonant fields and the 'widths' will be of great help in refining the potential curves

that characterize the inter-atomic interaction. This, in turn, has important applications in spectroscopy, high-frequency resolution measurements and atomic clocks.

The Feshbach resonance has been listed as one of the prospective schemes to alter the effective inter-particle interactions of the cold-atom systems [10]. A variable interaction strength is a highly unusual degree of freedom in experimental studies of many-body systems. Especially for the atomic condensate system, the 'tunability' of this parameter suggests highly interesting applications as virtually all observable quantities, as well as the stability of the system, sensitively depend on its value.

Among the many applications suggested by this novel degree of freedom, we name but a few: (i). Study of negative scattering length condensates. A dilute gas condensate of bosons that experience an effective mutual attraction is unstable in the absence of an external potential. A trapping potential can 'stabilize' a negative scattering length condensate of limited number of bosons [11]- [15]. Once the population of the condensate exceeds this critical number, the condensate collapses. Although most theories agree on the collapse and the critical number of particles, the details and the mechanism of the collapse are still being debated in the current literature (for a discussion see, for example, Ref. [15]). Measurements taken by Hulet's group at Rice university with  $^7\text{Li}$  provide valuable data on this interesting system, but a study of the collapse at variable values of the interaction strength will give definitive tests of the theoretical predictions and yield much needed insight in the dynamics of the collapse. (ii). Study of the condensate phase separation. Overlapping condensates are unstable if the strength of the unlike boson interactions exceeds the geometric mean of the like-boson interaction strengths. Mutually repelling condensates then separate spatially and act as immiscible fluids. As the phase separation criterion depends solely on the interaction strengths, one could by varying one of the strengths, render the condensates miscible or immiscible. Whether the BEC-systems will have practical applications remains to be seen, but a two-fluid system that can be made miscible or immiscible at will, does suggest applications in areas such as data-storage, or perhaps even quantum

computation. (iii). Study of Josephson oscillations. A mixture of same species condensates in different internal states can exchange bosons coherently, for example by interacting with coherent near-resonant laser light [19]-citepr21. Such coherent inter-condensate particle exchange processes are often referred to as inter-condensate 'tunneling' processes because of the strong analogy with Josephson tunneling [22] - [23]. Interestingly, unlike the condensed matter Josephson junctions, the dilute condensate mixtures can actually probe the non-linear regime of the Josephson oscillations [19]- [22]. Like the DC Josephson junction, the number of bosons in each condensate oscillates when the values of the chemical potentials of the respective condensates differ. For a single dilute condensate, the chemical potential is equal to the product of the interaction strength and the density. Thus, a sudden change of the interaction strength effects precisely such a chemical potential difference. (iv).

Condensate Dynamics. While the near-equilibrium dynamics of dilute single condensate systems are well-understood, at least in the low-temperature limit, the far-from equilibrium condensate dynamics poses a problem that has not been satisfactorily resolved. A detailed understanding of the condensate formation, in particular of the formation time [24] - [27], will have important implications in a variety of fields, such as field theory and early universe theories. Experimental studies with a variable interaction strength will give a firm understanding of the role of the inter-particle interactions.

Clearly, each of the above applications represents an exciting prospect. It is in this context, the creation of a tunable interaction strength, that the motivation for much of the previous research on Feshbach resonances has been situated [9], [28]- [33]. However, we would urge caution in interpreting the effects of the Feshbach resonance on the condensate solely as altering the inter-boson interaction. We believe that the effects of the Feshbach resonance are considerably more profound.

In particular, in this paper, we review some of our previous research results ( [34], [35], see also ref. [36] for a brief review of the tunneling aspect) that show that in a near-resonant BEC, the Feshbach resonant atom-molecule coupling creates a second condensate component of quasi-bound molecules. The many-body dynamics predicts that the expectation value of



the molecular field does *not* vanish in a near-resonant magnetic field,  $\langle \hat{\psi}_m \rangle \neq 0$ : the hallmark of Bose-condensation. Whether the presence of the molecular condensate predicts a behavior that differs from that of a single condensate with altered scattering length depends on the values of several relevant parameters: the detuning, the rate of change of the detuning (in a dynamical experiment with changing magnetic field), the lifetime of the quasi-bound molecules, limited—most importantly—by collisions with other atoms or molecules that change the molecular state.

Off-resonance, the stationary condensate system does behave as a single condensate with a scattering length that has the value predicted by the binary collision description. However, even in the off-resonant limit, the dynamical response of the condensate system to a sudden change of the external magnetic field, can differ significantly from that of a single condensate with effective scattering length. We find that, subsequent to a sudden change of the external magnetic field, the number of atoms and molecules that occupy the respective condensates oscillate. These oscillations are damped out on the time scale of the single molecule lifetime. The off-resonant condensate lives much longer than the individual molecules, since the atomic condensate acts as a reservoir of atoms, continually replenishing the molecular condensate. The oscillations can then be observed by illuminating the condensate with light that is near resonant with a transition of the quasi-bound molecule. The oscillations in the molecular condensate population then reveal themselves as an oscillating intensity of the image. The oscillations, like the oscillating current observed in Josephson junctions, are caused by the coherent inter-condensate exchange of particles. Unlike the Josephson tunneling, the exchange involves boson pairs. This, as we shall show, has a profound effect on the stationary state properties of the BEC system. For instance, we find that, for a near-resonant detuning, the homogeneous ground state system is *always* unstable in the limit of ultra low atomic particle density. Unlike the negative scattering length condensate, this instability does not necessarily lead to collapse. At higher densities the near-resonant condensate system can be stabilized by the inter-particle interactions of the atoms and molecules. In that

case, the many-body energy, as a function of the atomic particle density, goes through a minimum. The many-body system can find this minimum by decreasing its volume and, if given enough time, can relax to the state of minimal energy and self-determined density, a typical liquid-like property. This self-determined density would still be of the order of  $10^{15} \text{cm}^{-3}$ , so that these considerations suggest the possibility of creating the world's first rarified liquid!

The paper is organized as follows. In section II, we review the binary collision theory of low energy Feshbach resonances. In section III, we specialize to the magnetically controlled Feshbach resonances recently observed in atomic traps. In that section, we set up the Hamiltonian for the many-body problem and we argue that a second molecular condensate component is formed. A compelling argument follows from the equations that describe the many-body dynamics. We also discuss the effects of the most important destructive influence that the molecular condensate undergoes: collisions of the molecules with other atoms or molecules that change the vibrational state of the quasi-bound molecule. In section IV, we investigate the stationary states of the near-resonant condensate system, neglecting, for the time being, the effects of particle-loss. We point out that the state that the near-resonant condensate system finds itself in, depends on its history. If the system was brought near-resonance by adiabatically increasing the detuning, its state differs from that of the system created by lowering the detuning. Furthermore, we show that, in contrast to the effective scattering length description, the condensate system does not have to collapse, as it is tuned adiabatically through resonance. Finally, we conclude in section V.

## II. LOW ENERGY FESHBACH RESONANCES

### A. Introduction

In this paper, we focus on the implications of a Feshbach resonance on the many-body behavior of Bose-Einstein condensed systems. To this purpose, we develop the necessary

theoretical framework to describe the relevant binary collision physics. The following discussion does not aim at being comprehensive with regards to Feshbach resonance physics. For such treatment, we refer the reader to more specific articles [37]- [39].

By definition, Feshbach resonances involve intermediate states that are quasi-bound, so that they are sometimes referred to as closed-channel collisions. These intermediate states are not bound in the true sense of the word, as they acquire a finite lifetime due to the interaction with continuum states of other channels (such as the channel of the incident projectile/target system). For example, in electron-atom and electron-ion scattering, the intermediate states generally decay by ejecting the electron captured in the intermediate state. These states are known as auto-ionization states. In the atom-atom scattering Feshbach resonances of interest here, the intermediate states are molecules with electronic and nuclear spins that have been rearranged from the spins of the colliding atoms by virtue of the hyperfine interaction. The intermediate molecular states interact with the continuum states of the incident channel that are the scattering states of the single channel atom-atom scattering problem. In the next paragraph, we discuss these single channel scattering states.

## B. Low-energy potential scattering

As the interactions of interest involve bosonic atoms at ultra-low translational energies, the collision physics reduces to the description of  $s$ -wave scattering. Specifically, the angular momentum potential barrier of 'height'  $\sim (\hbar^2/ML^2)$ , where  $L$  is the range of the interatomic interaction and  $M$  the mass of a single atom, prevents colliding atoms with relative motion of lower kinetic energy and non-vanishing angular momentum from entering the inter-atomic interaction region. The magnitude of the angular momentum potential barrier height is of order  $(\hbar^2/ML^2) = (\hbar^2/m_e a_0^2) \times (a_0/L)^2 \times (m_e/M) \sim 10 - 100mK$ , where  $m_e$  represents the electron mass,  $(m_e/M) \sim 10^{-4} - 10^{-5}$ , and where  $a_0$  denotes the usual Bohr radius,  $(L/a_0) \sim 10$ . Thus, in cold atom samples of temperature below  $1mK$ , bosonic atoms undergo pure  $s$ -wave scattering. The atomic Bose-Einstein condensates have temperatures

of the order of  $1\mu K$ .

Before we proceed with the treatment of low-energy Feshbach resonances, we describe the solution to the single channel scattering problem for the collision of two of such indistinguishable atoms. In the center-of-mass frame the problem reduces to an integration of the radial s-wave Schrodinger equation with the corresponding molecular potential. The resulting regular solution,  $u(r)$ , where  $r$  denotes the internuclear distance and 'regular' means that  $\lim_{r \rightarrow 0} u(r)$  is finite, has to be normalized. We normalize  $u(r)$  to  $u_N(r)$  by requiring its asymptotic behavior to be similar to that of the zeroth-order spherical Bessel-function, which is the regular solution to the free atom Schrodinger equation:

$$u_N(r) \sim \sin(kr + \delta_0)/(kr), \quad r \rightarrow \infty, \quad (1)$$

where  $\delta_0$  is the s-wave phase shift.

For future reference, we relate  $u_N(r)$  to the s-wave components of scattering states that have been normalized using alternative schemes. One useful normalization consists of separating the scattered wave into an incident plane wave of wave vector  $\mathbf{k}$  and an outgoing spherical wave:

$$\varphi_{\mathbf{k}}(\mathbf{r}) \sim \exp(i\mathbf{k} \cdot \mathbf{r}) + f \frac{\exp(ikr)}{r}, \quad r \rightarrow \infty, \quad (2)$$

where  $f$  is the scattering amplitude. By comparing its asymptotic behavior to that of  $u_N$ , Eq.(1), we find the usual expression for the s-wave scattering amplitude:

$$f = \frac{[\exp(2i\delta_0) - 1]}{2ik}, \quad (3)$$

where  $k$  denotes the wavenumber, which is the magnitude of the  $\mathbf{k}$ -vector. Furthermore, the s-wave component of  $\varphi_{\mathbf{k}}$  is equal to

$$[\varphi_{\mathbf{k}}(\mathbf{r})]_s = \exp(i\delta_0)u_N(r). \quad (4)$$

An alternative normalization that we shall consider, requires the regular solution to the Schrodinger equation to be a superposition of incident and outgoing spherical waves:

$$\varphi^{(+)}(r) \sim \exp(-ikr)/r - S \exp(ikr)/r, \quad r \rightarrow \infty, \quad (5)$$

where the coefficient of the outgoing wave,  $S$ , is the scattering (or  $S$ )-matrix for the single channel s-wave scattering problem,  $S = \exp(2i\delta_0)$ . By comparing the asymptotic behaviors of  $u_N$  and  $\varphi^{(+)}$ , we find that

$$\varphi^{(+)}(r) = -2ik \exp(i\delta_0) u_N(r). \quad (6)$$

The alkali atoms in the atomic-trap condensates interact through molecular potentials that support bound states. In accordance with Levinson's theorem,  $u_N(r)$  has then nodes in the inter-atomic interaction region. The number of nodes is equal to the number of bound states of the corresponding potential. Furthermore,  $u_N(r)$  for the ultra-cold collision energies is essentially independent of the energy. To see that, we start by noting that for the relevant collision energies the de Broglie wavelength,  $(2\pi/k)$ , vastly exceeds the range  $L$  of the inter-atomic potential. Outside the range of the inter-atomic interaction,  $r > L$ , but well within the de Broglie wavelength,  $r < k^{-1}$ ,  $ru(r)$  is approximately linear:  $ru(r) \propto r - a$ , where  $a$  is the scattering length. By scaling  $u(r)$  to the normalized function  $u_N$ , which in this region of the internuclear distance,  $L < r < k^{-1}$ , takes on the form  $u_N(r) \approx 1 + (\delta_0/kr)$ , we see that  $\delta_0 = -ka$  and

$$u_N(r) \approx 1 - (a/r) \quad \text{where } L < r < k^{-1}, \quad (7)$$

independent of the collision energy.

### C. Low energy Feshbach resonances

In describing the Feshbach resonant collision, we distinguish the channels of the continuum incident projectile/target state and the closed (molecular) channels to which it is coupled. To this purpose, we introduce the projection operators  $P$  and  $M$  that denote, respectively, the projections onto the Hilbert subspace of the incident channel and the subspace of the closed (molecular) channels. These projection operators satisfy the usual projection operator properties

$$0 = MP = PM$$

$$P^2 = P, \text{ and } M^2 = M . \quad (8)$$

The time-independent Schrödinger equation, satisfied by the total scattering state  $|\Psi\rangle$  of the binary atom system,

$$(E - H) |\Psi\rangle = 0 , \quad (9)$$

takes on the form of coupled equations:

$$(E - H_{PP}) P|\Psi\rangle = H_{PM} M|\Psi\rangle \quad (10)$$

$$(E - H_{MM}) M|\Psi\rangle = H_{MP} P|\Psi\rangle , \quad (11)$$

where we use the notation  $PHP = H_{PP}$  etc. ...

We may obtain the projection of the scattering state onto the Hilbert space of the quasi-bound molecules,  $M|\Psi\rangle$ , by formally inverting Equation (11):

$$M|\Psi\rangle = (E - H_{MM})^{-1} H_{MP} P|\Psi\rangle . \quad (12)$$

The substitution of Eq.(12) into the projection of the Schrodinger equation onto the continuum channel (10) then yields an effective Schrodinger equation for the continuum scattering state,  $(E - H_{\text{eff}})P|\Psi\rangle = 0$ , with an effective Hamiltonian,

$$H_{\text{eff}} = H_{PP} + H_{PM} \frac{1}{E - H_{MM}} H_{MP} , \quad (13)$$

that exhibits a strong dependence on the energy,  $E$ , of the colliding particles.

For the purpose of treating the scattering problem, it is, in fact, more instructive to start by inverting Equation (10) for  $P|\Psi\rangle$ , by means of the propagator for outgoing waves,

$$g_P^{(+)}(E) = (E - H_{PP} + i\eta)^{-1} , \quad (14)$$

where  $\eta$  is an infinitesimally small positive number. This inversion leads to

$$P|\Psi\rangle = |\varphi_P^{(+)}\rangle + g_P^{(+)}(E) H_{PM} M|\Psi\rangle . \quad (15)$$

The  $\varphi_p^{(+)}$ -state in the above expression is a scattering state of the single channel ( $P$ ) scattering problem,  $(E - H_{PP})|\varphi_p\rangle = 0$ . In addition, we choose the asymptotic boundary condition of the scattering state so that  $\varphi_p^{(+)}$  is the superposition of incident and outgoing spherical waves shown in Eq.(5) for the  $P$ -channel,  $\lim_{r \rightarrow \infty} \varphi_p^{(+)}(r) = \exp(-ikr)/r - S \exp(ikr)/r$ . Upon insertion of the expression for  $P|\Psi\rangle$  from Eq.(15) into Eq.(11), we find the effective Schrodinger equation satisfied by  $M|\Psi\rangle$ :

$$(E - H_{MM}) M|\Psi\rangle = H_{MP}|\varphi_p^{(+)}\rangle + H_{MP} g_p^{(+)}(E) H_{PM} M|\Psi\rangle . \quad (16)$$

Finally, substitution of the formal solution for  $M|\Psi\rangle$  from Eq.(16),

$$M|\Psi\rangle = \frac{1}{E - H_{MM} - H_{MP} g_p^{(+)}(E) H_{PM}} H_{MP} |\varphi_p^{(+)}\rangle , \quad (17)$$

into the initial expression for  $P|\Psi\rangle$  of Equation (15) yields the following result:

$$P|\Psi\rangle = |\varphi_p^{(+)}\rangle + \frac{1}{E - H_{MM} - H_{MP} g_p^{(+)}(E) H_{PM}} H_{MP} |\varphi_p^{(+)}\rangle . \quad (18)$$

The asymptotic dependence of  $P|\Psi\rangle$  on the radial inter-nuclear distance  $r$  supplies the  $S$ -matrix that characterizes the low energy collision with Feshbach resonance. At the end of this section, we determine  $S$  in this manner.

#### D. Width

The experimentally observed low energy Feshbach resonances are narrow - each individual resonance is well-separated in frequency space from the other resonances. Near a particular resonance  $m$ , with a single intermediate molecular state  $|\varphi_m\rangle$  of specific ro-vibrational quantum number, we may further simplify the expression (18) by keeping only a single diagonal matrix element in evaluating the energy denominator of Eq.(18). Specifically, we replace

$$\frac{1}{E - H_{MM} - H_{MP} g_p^{(+)}(E) H_{PM}} \rightarrow \frac{1}{|\varphi_m\rangle \frac{1}{E - E_m + i\Gamma_m/2} \langle \varphi_m|} , \quad (19)$$

where the energy  $E_m$  and width  $\Gamma_m$  of the resonance are equal to

$$\begin{aligned} E_m &= \text{Re}\langle \varphi_m | H_{MM} + H_{MP} g_p^{(+)}(E) H_{PM} | \varphi_m \rangle \\ \frac{\Gamma_m}{2} &= -\text{Im}\langle \varphi_m | H_{MP} g_p^{(+)}(E) H_{PM} | \varphi_m \rangle . \end{aligned} \quad (20)$$

Furthermore, the coupling between the continuum and molecular states for these resonances is weak enough that we may evaluate  $|\varphi_m\rangle$  in the spirit of perturbation theory as the eigenstate of the molecular potential, and approximate  $E_m$  by its eigenvalue.

In particular, the low energy dependence of the width is of importance to the observed resonances and we will evaluate  $\Gamma_m(E)$  in detail. The expansion of  $g_p^{(+)}(E)$  in continuum states  $|\mathbf{k}\rangle$  gives with Eq.(14) the following expression:

$$\begin{aligned} \frac{\Gamma_m}{2} &= -\text{Im} \left\{ \sum_{\mathbf{k}} \frac{|\langle \varphi_m | H_{MP} | \mathbf{k} \rangle|^2}{E - E_{\mathbf{k}} + i\eta} \right\} \\ &= \pi \sum_{\mathbf{k}} |\langle \varphi_m | H_{MP} | \mathbf{k} \rangle|^2 \delta(E - E_{\mathbf{k}}) . \end{aligned} \quad (21)$$

In this equation, the continuum states,  $|\mathbf{k}\rangle$ , are not plane waves, but properly normalized scattering states  $\varphi_{\mathbf{k}}$  (introduced in equation (2)). We shall work in box normalization so that in coordinate space

$$\begin{aligned} \langle \mathbf{r} | \mathbf{k} \rangle &= \varphi_{\mathbf{k}}(\mathbf{r}) / \sqrt{\Omega} \\ &\sim \frac{[\exp(i\mathbf{k} \cdot \mathbf{r}) + f \frac{\exp(i\mathbf{k}r)}{r}]}{\sqrt{\Omega}} , \quad r \rightarrow \infty , \end{aligned} \quad (22)$$

where  $\Omega$  represents the macroscopic volume to which the binary atom system is confined. At the ultra-low collision energies of interest, the matrix elements  $\langle \varphi_m | H_{PM} | \mathbf{k} \rangle$ , are dominated by the s-wave component of the  $|\mathbf{k}\rangle$ -wave. Within the molecular interaction range, the amplitudes of the higher partial waves have all but vanished at these energies as the colliding particles lack the energy to overcome the angular momentum potential barrier. Furthermore,



the low-energy  $s$ -wave,  $u_N(r)$  is essentially energy independent, so that  $\langle \varphi_m | H_{PM} | \mathbf{k} \rangle$  is not only independent of the direction of the  $\mathbf{k}$ -vector, but also of its magnitude:

$$\begin{aligned} \langle \varphi_m | H_{MP} | \mathbf{k} \rangle &\approx \frac{\exp(i\delta_0)}{\sqrt{\Omega}} \int d^3r \varphi_m(\mathbf{r}) \hat{H}_{MP} u_N(r) \\ &= \frac{\exp(i\delta_0)}{\sqrt{\Omega}} \alpha, \end{aligned} \quad (23)$$

where the  $\alpha$ -parameter denotes the integral over the relative internuclear position,  $\alpha = \int d^3r \varphi_m(\mathbf{r}) \hat{H}_{MP} u_N(r)$ . Consequently, the width is proportional to the square of  $\alpha$  and the remaining 'phase space factor'  $\sum_{\mathbf{k}} \delta(E - E_{\mathbf{k}})/\Omega$ :

$$\begin{aligned} \frac{\Gamma_m(E)}{2} &= \pi \alpha^2 \frac{1}{\Omega} \sum_{\mathbf{k}} \delta(E - E_{\mathbf{k}}), \text{ or} \\ \Gamma_m(E) &= \alpha^2 \left( \frac{M}{2\pi\hbar^2} \right) k_E, \end{aligned} \quad (24)$$

where  $M$  is the mass of a single atom and  $k_E$  the wave number corresponding to the relative velocity of a pair of atoms with total kinetic energy  $E$  in the center-of-mass frame. Note that the width depends on the energy of the colliding atoms through the phase space factor, which is a measure of the phase space volume available to the binary atom system after the collision. Evidently, this remark is of importance to the Feshbach resonances in the ultra-cold atoms systems, where the relative velocity of the interacting atoms rigorously vanishes.

It is customary to introduce a 'reduced width',  $\gamma$ , which makes the  $k$ -dependence of the width explicit:

$$\Gamma_m(E) = 2\gamma k, \text{ where } \gamma = \alpha^2 \left( \frac{M}{4\pi\hbar^2} \right), \quad (25)$$

where it is understood that  $k$  is the wave number corresponding to  $E$ . Under condensate-conditions,  $k \rightarrow 0$ , and  $\Gamma_m(E) \rightarrow 0$ , although the value of the coupling constant,  $\alpha$ , remains constant. The corresponding phase shift also vanishes linearly with  $k$ , but the effective scattering length tends to a well-defined finite value, as we shall see below.

## E. Scattering matrix

We evaluate the continuum scattering wave  $P|\Psi\rangle$ ,

$$P|\Psi\rangle = |\varphi_p^{(+)}\rangle + g_p^{(+)}(E) H_{PM}|\varphi_m\rangle \times \frac{1}{E - E_m + i\Gamma_m/2} \langle\varphi_m|H_{MP}|\varphi_p^{(+)}\rangle . \quad (26)$$

As discussed above,  $|\varphi_p^{(+)}\rangle$  is normalized by requiring its asymptotic  $r$ -dependence to take on the form  $\varphi_p^{(+)}(r) = \exp(-ikr)/r - \exp(2i\delta_0) \exp(ikr)/r$ , where  $\delta_0$  is the  $s$ -wave phase shift. In the low energy regime,  $\delta_0 = -ka$ , where  $a$  is the scattering length. Furthermore, we remarked in Eq.(6) that  $\varphi_p^{(+)}(r) = -2ik \exp(i\delta_0) u_N(r)$ , where  $u_N$  is the regular solution to the Schrodinger equation with  $u_N(r) \approx 1 - r/a$  outside the range of the molecular potential.

In evaluating the state (26) in the asymptotic region of coordinate space, the following asymptotic expansion of the  $s$ -wave component of the  $g_p^{(+)}$ -propagator is useful:

$$\begin{aligned} [g_p^{(+)}(E; \mathbf{r}, \mathbf{r}') ]_s \rightarrow & \\ & - \left( \frac{M}{4\pi\hbar^2} \right) \exp(i\delta_0) \frac{\exp(ikr)}{r} u_N(r') , \\ \text{where } r \rightarrow \infty , & \end{aligned} \quad (27)$$

and where we replaced the mass in the usual expression for the propagator by the reduced mass,  $M/2$ . The asymptotic behavior of  $g_p^{(+)}(E)H_{PM}|\varphi_m\rangle$  of Eq.(26) is then given by

$$\begin{aligned} \lim_{r \rightarrow \infty} \langle \mathbf{r} | g_p^{(+)}(E) H_{PM} | \varphi_m \rangle & \\ = - \left( \frac{M}{4\pi\hbar^2} \right) \frac{\exp(ikr)}{r} \exp(i\delta_0) \times & \\ \int d^3r u_N(r) \hat{H}_{PM} \varphi_m(\mathbf{r}) & \\ = - \left( \frac{M}{4\pi\hbar^2} \right) \frac{\exp(ikr)}{r} \exp(i\delta_0) \alpha . & \end{aligned} \quad (28)$$

Finally, with  $\langle\varphi_m|H_{MP}|\varphi_p^{(+)}\rangle = -2ik \exp(i\delta_0)\alpha$ , we obtain the desired asymptotic behavior:

$$\begin{aligned} \langle \mathbf{r} | g_p^{(+)}(E) H_{PM} | \varphi_m \rangle \langle \varphi_m | H_{MP} | \varphi_p^{(+)} \rangle & \\ = i \exp(2i\delta_0) \left( \frac{M}{2\pi\hbar^2} \right) \alpha^2 k \frac{\exp(ikr)}{r} & \\ = i \exp(2i\delta_0) \Gamma_m(E) \frac{\exp(ikr)}{r} . & \end{aligned} \quad (29)$$

Consequently, we obtain the following expression for the asymptotic  $r$ -dependence of the scattering state of Eq.(26):

$$\langle \mathbf{r} | P | \Psi \rangle \sim \frac{\exp(-ikr)}{r} - \left[ 1 - \frac{i\Gamma_m(\bar{E})}{E - E_m + i\Gamma_m(E)/2} \right] \times \exp(2i\delta_0) \frac{\exp(ikr)}{r} . \quad (30)$$

By identifying the factor in square brackets in Eq.(29), with the scattering matrix  $S$ , we obtain

$$S = \exp(-2ika) \left[ 1 - i \frac{\Gamma_m(E)}{E - E_m + i\frac{\Gamma_m(E)}{2}} \right] , \quad (31)$$

where we used that  $\delta_0 = -ka$ . Note that the  $S$ -matrix is unitary,  $|S| = 1$ , as befits scattering without loss-channels. As a consequence, we can describe the scattering by means of an effective scattering length,  $S = \exp(-2ia_{eff}k)$ , where  $a_{eff} = a + a'$ , with

$$\begin{aligned} \exp(-2ika') &= 1 - i \frac{\Gamma_m}{E - E_m + i\Gamma_m/2} \\ &= \frac{E - E_m - i\Gamma_m/2}{E - E_m + i\Gamma_m/2} . \end{aligned} \quad (32)$$

Thus, we find that the effective scattering length for arbitrary collision energy  $E$  is equal to

$$a_{eff}(E) = a + \frac{1}{2k} \tan^{-1} \left[ \frac{\Gamma_m \times [E - E_m]}{(E - E_m)^2 + \Gamma_m^2/4} \right] . \quad (33)$$

In the ultra-low energy limit appropriate for condensate systems,  $E$  and  $\Gamma_m(E)$  vanish and  $E - E_m \rightarrow -\epsilon$ , where we will refer to  $\epsilon$ , the energy of the molecular state relative to the continuum level of the  $P$ -channel, as the detuning of the Feshbach resonance. In this limit, we need to expand Eq.(33) to lowest order in  $k$ . With  $\Gamma_m = 2\gamma k$ ,  $E = \hbar^2 k^2/M$  and  $\tan^{-1}(x) \approx x$  if  $x \ll 1$ , we find

$$\lim_{E \rightarrow 0} a_{eff}(E) = a - \frac{\gamma}{\epsilon} . \quad (34)$$

Furthermore, in describing the weakly interacting many-body system, it is notationally more convenient to work with the interaction strength  $\lambda$  than with the scattering length  $a$ . If the

inter-particle scattering can be described in the Born approximation, then the  $\lambda$ -parameter represents the zero-momentum Fourier component of the inter-particle interaction potential. The zero momentum Fourier component is related to the scattering length  $a$ , calculated in the same approximation as  $\lambda = (4\pi\hbar^2/M)a$ . However, the Born-approximation cannot be used in describing the low energy binary atom collisions. In that case, the interaction strength is still proportional to the scattering length, although the latter has to be determined more accurately from the full potential scattering problem. In the same spirit, we may introduce an 'effective' strength,  $\lambda_{eff} = (4\pi\hbar^2/M)a_{eff}$ , to describe the binary atom interaction, related to  $\lambda$  as

$$\lambda_{eff} = \lambda - \frac{\alpha^2}{\epsilon}. \quad (35)$$

In Eq.(35) we made use of the expression for the reduced width,  $\gamma = \alpha^2(M/4\pi\hbar^2)$ .

### III. FESHBACH RESONANCES IN ATOMIC CONDENSATE SYSTEMS

#### A. Magnetically controlled, hyperfine induced Feshbach resonance

In each of the experiments, the low-energy Feshbach resonances were observed by studying the behavior of the ultra-cold atom systems under variations of an external magnetic field. The resonance in the binary-atom interactions is caused by the hyperfine interaction which flips the electronic and nuclear spins of one of the colliding atoms, bringing the collision system from the continuum ( $P$ )-channel into a closed channel,  $M$ , of different spin arrangement. The  $M$ -channel is closed by the external magnetic field which has raised the continuum level of the binary spin flipped atom system. While in the  $M$ -channel, the colliding atoms reside in a quasi-bound molecular state  $m$ . A second hyperfine-induced spin flip breaks up the molecule, returning the system to the initial  $P$ -channel. If the energy of the intermediate quasi-bound molecule is equal to the continuum level of the  $P$ -channel, the above described collision process is 'on resonance'. Variations of the external magnetic field shift the relative energies of the  $M$  molecule and the  $P$ -continuum level and tune, or

detune, the interacting atoms close to, or far from resonance. We now describe the physics of this collision in more detail.

We find it instructive to discuss the collision first in the limit of high external magnetic field, although this is not the regime in which the experiments were conducted. A high magnetic field,  $\mathbf{B} = B\hat{z}$ , aligns the electronic ( $s$ ) and nuclear ( $i$ ) spins of the atoms. To be definite, we consider a system of ultra-cold  $^{23}\text{Na}$  atoms ( $i = 3/2$ ) with spin projections  $m_i = -1/2$ , and  $m_s = -1/2$ . The interaction of two such atoms is described by the triplet potential, where 'triplet' refers to the total electronic spin of the interacting atoms. In the triplet state, the valence electrons of the atoms behave as indistinguishable fermions and 'avoid each other', thereby reducing the Coulomb repulsion of the electrons. In contrast, if the spins of the colliding atoms are arranged in a singlet state, the valence electrons do not avoid each other and the Coulomb repulsion generally reduces the depth of the inter-atomic potential, as compared to the triplet potential. Thus, the inter-atomic interaction depends on the magnitude of the total electronic spin,  $S$ , where  $\mathbf{S} = \mathbf{s}_1 + \mathbf{s}_2$ . In this case, the spins of the initial binary atom system are in a state  $|S_{in}\rangle = |m_s = 1/2, m_i = -1/2; m_s = 1/2, m_i = -1/2\rangle$ , with electronic spins arranged in a pure triplet state,  $S = 1$ . Consequently, the atoms interact through the molecular triplet potential. However, the binary atom hyperfine interaction,  $V_{hf} = (a_{hf}/\hbar^2) \times [\mathbf{s}_1 \cdot \mathbf{i}_1 + \mathbf{s}_2 \cdot \mathbf{i}_2]$ , does *not* commute with  $\mathbf{S}^2$  and can flip the electronic spins of a triplet state to a singlet configuration. At large internuclear separation, this singlet channel corresponds to the binary atom system with a single spin flipped atom. Consequently, the continuum of the singlet channel lies an energy  $\Delta$  above the continuum of the incident triplet channel, where  $\Delta = B[2\mu_e + \mu_N]$ , and where  $\mu_e$  and  $\mu_N$  denote the electronic and nuclear magnetic moments. Under near-resonant conditions, the singlet potential supports a quasi-bound molecular state  $\varphi_m(\mathbf{r})|S_{j'}\rangle$ , of energy  $E_m$  near the continuum of the  $P$ -channel and total (electronic and nuclear) spin state  $|S_{j'}\rangle$ .

In this context, the atom-molecule coupling,  $H_{PM} + H_{MP}$ , is provided by the binary atom hyperfine interaction,  $V_{hf}$ . The corresponding  $\alpha$ -parameter that indicates the strength

of this inter-channel coupling is the product of the spin matrix element and the overlap of the regular triplet wave function with the molecular singlet wave function,

$$\alpha = \langle S_{j'} | V_{hf} | S_{in} \rangle \times \int d^3r \varphi_m(\mathbf{r}) u_N(r) , \quad (36)$$

which characterizes the resonance.

The observed resonances were created at intermediate values of the magnetic field for which the actual spin state of the individual atoms are *not* states of good  $m_i$  and  $m_s$ -quantum number. Instead the single atom spin degrees of freedom occupy a state that diagonalize the spin dependent part of the single atom Hamiltonian:

$$H_{Spin(1)} = \left( \frac{a_{hf}}{\hbar^2} \right) \mathbf{s}_1 \cdot \mathbf{i}_1 + \mathbf{B} \cdot \frac{[2\mu_e \mathbf{s}_1 - \mu_N \mathbf{i}_1]}{\hbar} . \quad (37)$$

The first term on the right-hand side of Eq.(37) is the single atom hyperfine interaction, characterized by  $a_{hf}$ , an energy that depends on the isotope (e.g.  $a_{hf} = 42.5 \text{ mK}$  for  $^{23}\text{Na}$ , see, e.g. ref. [40]). At zero magnetic field,  $\mathbf{B} = 0$ , the diagonalization yields the hyperfine states of good 'f' quantum number, where f represents the 'total' single atom spin,  $\mathbf{f} = \mathbf{s} + \mathbf{i}$ . At high magnetic field strengths  $B \gg a_{hf}/[\mu_e \hbar]$ , the hyperfine interaction can be treated as a perturbation and, to lowest order, the electronic and nuclear spins are in states of good  $m_i$  and  $m_s$  quantum number. At arbitrary values for the magnetic field, the atoms find themselves in a linear combination,  $c_{-1/2} |m_i = m + 1/2; m_s = -1/2\rangle + c_{+1/2} |m_i = m - 1/2; m_s = +1/2\rangle$  with coefficients  $c_{\pm 1/2}$  that diagonalize  $H_{Spin(1)}$  (37). The system of two alkali atoms, each atom in this particular spin state, is then generally in a linear combination of a singlet and triplet state. We represent the interatomic potential interaction  $\hat{V}$  to the binary-atom Hamiltonian by means of projection operators  $\hat{\Pi}_S$ , projecting onto states of good total electron spin S:

$$\hat{V} = \sum_S V_S(\mathbf{r}) \hat{\Pi}_S . \quad (38)$$

We also write the scalar spin product of the binary atom hyperfine interaction in terms of the total electronic and nuclear spins,  $\mathbf{I} = \mathbf{i}_1 + \mathbf{i}_2$ :

$$\begin{aligned}
\mathbf{s}_1 \cdot \mathbf{i}_1 + \mathbf{s}_2 \cdot \mathbf{i}_2 &= \\
&= [(\mathbf{s}_1 + \mathbf{s}_2) \cdot (\mathbf{i}_1 + \mathbf{i}_2) + (\mathbf{s}_1 - \mathbf{s}_2) \cdot (\mathbf{i}_1 - \mathbf{i}_2)] / 2 = \\
&= [\mathbf{S} \cdot \mathbf{I} + \mathbf{S}_d \cdot \mathbf{I}_d] / 2 ,
\end{aligned} \tag{39}$$

where  $\mathbf{S}_d$  and  $\mathbf{I}_d$  denote the corresponding 'spin difference' operators:  $\mathbf{S}_d = \mathbf{s}_1 - \mathbf{s}_2$ , and  $\mathbf{I}_d = \mathbf{i}_1 - \mathbf{i}_2$ . The  $\mathbf{S}_d$  and  $\mathbf{I}_d$  operators, antisymmetric under the interchange of the subscripts, couple states with  $I$  and  $S$  quantum numbers differing by 1. The spin dependent Hamiltonian of the colliding binary atom system takes on the form:

$$\begin{aligned}
H_{Spin(1,2)} &= \left( \frac{a_{hf}}{2\hbar^2} \right) \mathbf{S} \cdot \mathbf{I} + \mathbf{B} \cdot \frac{[2\mu_e \cdot \mathbf{S} - \mu_N \mathbf{I}]}{\hbar} + \\
&\quad \sum_S V_S \hat{\Pi}_S + \left( \frac{a_{hf}}{2\hbar^2} \right) \mathbf{S}_d \cdot \mathbf{I}_d \\
&= H_{Spin(1,2)}^{(0)} + \sum_S V_S \hat{\Pi}_S + \\
&\quad \left( \frac{a_{hf}}{2\hbar^2} \right) \mathbf{S}_d \cdot \mathbf{I}_d .
\end{aligned} \tag{40}$$

The expression of Eq.(40), suggests that a convenient partitioning into collision channels divides the Hilbert space of the binary atom/spin system into subspaces  $j$  of states of good  $I$  and  $S$ -quantum numbers with the proper superposition of  $|I, S; M_I, M_S\rangle$  states that diagonalizes the  $H_{Spin(1,2)}^{(0)}$ -operator. The corresponding eigenvalues determine the continuum levels of the  $j$ -channels. By construction, the atom-atom potential interaction,  $\hat{V}$ , is diagonal in these channels, but not the  $\mathbf{S}_d \cdot \mathbf{I}_d$ -contribution. In describing the collision, we project the spin of the incident binary atom system,  $|S_{in}\rangle$  onto the collision channels  $j$ :  $|S_{in}\rangle = \sum_j |S_{in}; j\rangle$ . The Feshbach-resonance occurs if the interaction potential of another, 'spin flipped', binary atom channel of spin state  $|S_{j'}\rangle$ , accessible from the  $j$ -channel by virtue of the  $\mathbf{I}_d \cdot \mathbf{S}_d$ -interaction, supports a bound state  $\varphi_m(\mathbf{r})|S_{j'}\rangle$  of energy  $E_m$  near the continuum level of the incident  $j$ -channels. The corresponding  $\alpha$ -parameter (defined as in section II) is equal to

$$\begin{aligned}
\alpha_{j,j';m} &= \left( \frac{a_{hf}}{2\hbar^2} \right) \langle S_{j'} | \mathbf{S}_d \cdot \mathbf{I}_d | S_{in}; j \rangle \times \\
&\quad \int d^3r \varphi_m(\mathbf{r}) u_{N,j}(r) .
\end{aligned} \tag{41}$$

Since the strength of the magnetic field is the experimental knob that controls the resonance detuning, it is helpful to make the magnetic field dependence explicit. The detuning,  $\epsilon$ , is the energy difference of the quasi-bound molecular state,  $E_m$ , and the continuum of the incident atoms, pictured in Fig.(1). When  $B$  takes on its resonant value,  $B_m$ , the energy of the quasi-bound molecule lines up with the continuum of the incident atoms so that  $\Delta$  equals the binding energy of the bound state,  $\Delta = E_m$ . Near resonance,  $\Delta \approx E_m + (\partial\Delta/\partial B) \times [B - B_m]$ , and thus  $\epsilon = \Delta - E_m \approx (\partial\Delta/\partial B) \times [B - B_m]$ .

Consequently, the near-resonant effective scattering length of Eq.(33) depends on the external magnetic field strength as

$$a_{eff} = a \left[ 1 - \frac{\Delta B}{B - B_m} \right],$$

$$\text{where } \Delta B = \frac{\gamma}{a \times \partial\Delta/\partial B} \quad (42)$$

Thus, the dependence on the magnetic field strength is 'dispersive'. Likewise the effective interaction strength of Eq.(34) takes on the form

$$\lambda_{eff} = \lambda \left[ 1 - \frac{\Delta B}{B - B_m} \right] \quad (43)$$

## B. Feshbach resonant interactions in many-body systems

We treat the many-body physics of the atoms in second quantization. In describing the 'homogeneous' many-body system, it is convenient to work with the creation and annihilation operators  $\hat{c}_{\mathbf{k}}$  and  $\hat{c}_{\mathbf{k}}^\dagger$  of the single particle plane wave states. In a more general description, we work with the field operators  $\hat{\psi}(\mathbf{r}, t)$ , which, for the homogeneous system, take on the form  $\hat{\psi}(\mathbf{r}, t) = \sum_{\mathbf{k}} \exp(i\mathbf{k} \cdot \mathbf{r}) \hat{c}_{\mathbf{k}}(t) / \sqrt{\Omega}$ , where we use Box-normalization, and where  $\Omega$  represents the macroscopic 'box' volume. The dynamics of the single species low-temperature dilute gas can be taken to be governed by a Hamiltonian operator  $\hat{H} = \int d^3r \hat{\mathcal{H}}(\mathbf{r})$ , where we denote the Hamiltonian density operator by  $\hat{\mathcal{H}}$ ,

$$\hat{\mathcal{H}}(\mathbf{r}) = \hat{\psi}^\dagger(\mathbf{r}) \left[ -\frac{\hbar^2 \nabla^2}{2m} + V_{ext}(\mathbf{r}) \right] \hat{\psi}(\mathbf{r}) +$$



$$\frac{\lambda}{2} \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}(\mathbf{r}) \hat{\psi}(\mathbf{r}) , \quad (44)$$

and where  $V_{ext}$  represents the external potential experienced by the atoms.

In the ultra-dilute atomic-trap systems of density,  $n \sim 10^{13} - 10^{15} \text{ cm}^{-3}$ , giving  $na^3 \sim 10^{-8} - 10^{-4}$ , particles interact essentially as binary atom systems. Specifically, the atom-atom interactions can be described as in binary collisions, partly because the collision complex is so short lived that its interaction with other particles may be neglected. This assumption is a crucial ingredient of the formal many-body treatment which reintroduces the binary atom scattering length as a result of the 'ladder approximation' in treating the atom-atom interactions. The final result of this treatment is the result quoted above Eq.(35):  $\lambda = (4\pi\hbar^2/M)a$ . The low density of the gas might suggest the prescription  $\lambda \rightarrow \lambda_{eff}$ , where  $\lambda_{eff}$  is the effective interaction strength of Eq.(43) that describes binary atom collisions,

$$\lambda \rightarrow \lambda_{eff} = \lambda - \frac{\alpha^2}{\partial\Delta/\partial B} \frac{1}{B - B_m} , \quad (45)$$

to describe the effects of the Feshbach resonance. However, this recipe, which we shall refer to as the  $\lambda_{eff}$ -description, becomes problematic near resonance,  $\epsilon \rightarrow 0$ . One reason, of course, is that the system does not remain dilute in this description,  $na_{eff}^3 \rightarrow \infty$ . Alternatively, we may consider the time that two atoms, approaching each other at relative velocity  $v$ , spend in each others presence during the collision. If we estimate this time by the delay time,  $\tau_D = \hbar(\partial\delta_0/\partial E)$ , we can naturally break up  $\tau_D$  into a contribution that describes the time that the atoms spend in each others potential well,  $|\tau_{D,p}| = a/v$ , and the time that the atoms spend in the intermediate molecular state,  $|\tau_{D,m}| = |\gamma/(\epsilon v)|(k \rightarrow 0)$ . Note that  $\lim_{\epsilon \rightarrow 0} |\tau_{D,m}/\tau_{D,p}| = \infty$ , which means that the atoms spend an infinitely longer time in the intermediate molecular state near resonance. The validity of approximating the interactions as 'one-on-one' collisions, is then not self-evident. Furthermore, as we discuss in section IV, the  $\lambda_{eff}$ -description leads to unphysical predictions for the many-body behavior of a stationary on-resonant condensate.

In an approach that avoids making any 'a priori' assumptions, we reformulate the many-body problem while treating the particles as compound particles. We account for the Fesh-

bach resonances by including the intermediate state molecules explicitly, as well as the spin flip interactions that access the molecular states. We include the compound character of the particles by adding a subscript to the field operators or annihilation/creation operators,  $\hat{\psi}_a$  and  $\hat{c}_{a,k}$  for the atoms and  $\hat{\psi}_m$  and  $\hat{c}_{m,k}$  for the  $m$ -state molecules. The hyperfine induced spin flips that create the molecules formed in the intermediate state of the Feshbach resonance, are then described by the following term in the many-body Hamiltonian of a homogeneous system:

$$\hat{H}_{MP} = \sum_{\mathbf{K}, k', k} \left\{ \frac{1}{\sqrt{2}} \langle \mathbf{K}, m | V_{hf} | \mathbf{k}, \mathbf{k}' \rangle \times \hat{c}_{m, \mathbf{K}}^\dagger \hat{c}_{a, k} \hat{c}_{a, k'} \right\}, \quad (46)$$

where  $V_{hf}$  denotes the two-particle hyperfine interaction, and where the interaction matrix element is an integral over the unsymmetrized two particle wave functions of the  $m$ -molecule with center-of mass momentum  $\mathbf{K}$ ,  $|\mathbf{K}, m\rangle$ , and the binary atom continuum state of  $a$ -atoms colliding with momenta  $\mathbf{k}$  and  $\mathbf{k}'$ . The factor of  $1/\sqrt{2}$  has to be included to insure that the matrix elements of  $H_{MP}$  calculated in first and second quantization are equal, as follows from the example of an  $\hat{H}_{MP}$ -matrix element with single molecule bra and a binary atom ket-state. Likewise, the hyperfine induced break up of the molecules is described in the Hamiltonian by  $\hat{H}_{PM}$ , the hermitian conjugate of  $\hat{H}_{MP}$ .

We now evaluate the matrix element explicitly in coordinate space. The wave functions then depend on the positions  $\mathbf{r}_1$  and  $\mathbf{r}_2$  of both nuclei. The dependence of the molecular state on the relative position,  $\mathbf{r}_1 - \mathbf{r}_2$ , is given by the molecular wave function,  $\varphi_m(\mathbf{r}_1 - \mathbf{r}_2)$ , and the dependence on the center-of-mass position by the box normalized plane wave state  $\exp(i\mathbf{K} \cdot [\mathbf{r}_2 + \mathbf{r}_1]/2)/\sqrt{\Omega}$ . The binary atom state depends on the relative coordinates through a scattering state with  $s$ -wave component  $u_N(|\mathbf{r}_1 - \mathbf{r}_2|)$ . Its dependence on the center-of-mass coordinate is expressed by the plane wave  $\exp(i[\mathbf{k} + \mathbf{k}'] \cdot [\mathbf{r}_1 + \mathbf{r}_2]/2)/\Omega$ . Using the notation introduced in the previous section, the resulting expression for the matrix element of  $\hat{H}_{MP}$  is equal to

$$\begin{aligned}
\langle \mathbf{K}, m | V_{hf} | \mathbf{k}, \mathbf{k}' \rangle &= \frac{1}{\Omega^{3/2}} \left( \frac{a_{hf}}{2\hbar^2} \right) \langle S_{j'} | \mathbf{S}_d \cdot \mathbf{I}_d | S_{in}; j \rangle \times \\
&\exp(i\delta_0) \int d^3 r_1 \int d^3 r_2 \varphi_m^*(\mathbf{r}_1 - \mathbf{r}_2) \exp(-i\frac{\mathbf{K}}{2} \cdot [\mathbf{r}_1 + \mathbf{r}_2]) \\
&u_N(|\mathbf{r}_1 - \mathbf{r}_2|) \exp(i[\mathbf{k} + \mathbf{k}'] \cdot [\mathbf{r}_1 + \mathbf{r}_2]/2) .
\end{aligned} \tag{47}$$

The substitution to 'sum'  $[\mathbf{r}_1 + \mathbf{r}_2]/2 = \mathbf{R}$  and 'difference' coordinates,  $\mathbf{r}_1 - \mathbf{r}_2 = \mathbf{r}$ , then factorizes the integral into the product of an  $\mathbf{R}$ -integral,  $\int d^3 R \exp(-i[\mathbf{K} - \mathbf{k} - \mathbf{k}'] \cdot \mathbf{R}) = \Omega \delta_{\mathbf{K}, \mathbf{k} + \mathbf{k}'}$ , and an  $\mathbf{r}$ -integral, proportional to the Feshbach resonance  $\alpha$ -parameter:

$$\langle \mathbf{K}, m | V_{hf} | \mathbf{k}, \mathbf{k}' \rangle = \frac{1}{\sqrt{\Omega}} \delta_{\mathbf{K}, \mathbf{k} + \mathbf{k}'} \alpha , \tag{48}$$

where we have used that  $\exp(i\delta_0) \approx 1$ . Consequently, the Feshbach-resonance interactions, in second quantization, are described by

$$\begin{aligned}
\hat{H}_{MP} &= \frac{1}{\sqrt{\Omega}} \sum_{\mathbf{k}, \mathbf{k}'} \frac{\alpha}{\sqrt{2}} [\hat{c}_{m, \mathbf{k} + \mathbf{k}'}^\dagger \hat{c}_{a, \mathbf{k}} \hat{c}_{a, \mathbf{k}'}] \\
&= \frac{\alpha}{\sqrt{2}} \int d^3 r \hat{\psi}_m^\dagger(\mathbf{r}) \hat{\psi}_a(\mathbf{r}) \hat{\psi}_a(\mathbf{r}) .
\end{aligned} \tag{49}$$

Similar to the 'elastic' inter-atomic interactions, the low energy conditions imply a coupling strength to the molecular channel that is independent of the momentum transfer. Consequently, the Feshbach resonant interactions are also characterized by a single parameter: the atom-molecule coupling strength  $\alpha$ .

In accordance with these results, we generalize the Hamiltonian density for the many-body system, Eq.(44), to describe the many atom/molecule system:

$$\begin{aligned}
\hat{\mathcal{H}} &= \hat{\psi}_a^\dagger \left[ -\frac{\hbar^2 \nabla^2}{2M} + \frac{\lambda_a}{2} \hat{\psi}_a^\dagger \hat{\psi}_a \right] \hat{\psi}_a + \\
&\hat{\psi}_m^\dagger \left[ -\frac{\hbar^2 \nabla^2}{4M} + \varepsilon + \frac{\lambda_m}{2} \hat{\psi}_m^\dagger \hat{\psi}_m \right] \hat{\psi}_m(\mathbf{r}) + \\
&\lambda \hat{\psi}_a^\dagger \hat{\psi}_a \hat{\psi}_m^\dagger \hat{\psi}_m + \frac{\alpha}{\sqrt{2}} [\hat{\psi}_m^\dagger \hat{\psi}_a \hat{\psi}_a + \hat{\psi}_m \hat{\psi}_a^\dagger \hat{\psi}_a] ,
\end{aligned} \tag{50}$$

where the position dependence of the operators is understood, and where  $\lambda_a$ ,  $\lambda_m$  and  $\lambda$  represent the strengths of the atom-atom, molecule-molecule and atom-molecule interactions respectively. In Eq.(50), we have assumed that the atoms and molecules do not experience

external potentials. We note that the same Hamiltonian correctly reproduces the Feshbach resonant behavior of a binary atom system.

### C. Dynamics of the Condensate Mixture

A new phenomenon occurs in the many-body physics: the appearance of a molecular condensate component, which follows from the many-body dynamics. A convenient starting point to describe this dynamics are the Heisenberg equations of motion for the atomic and molecular field operators:

$$\begin{aligned} i\hbar\dot{\hat{\psi}}_a(\mathbf{x}, t) &= [\hat{H}, \hat{\psi}_a(\mathbf{x}, t)] \quad , \\ i\hbar\dot{\hat{\psi}}_m(\mathbf{x}, t) &= [\hat{H}, \hat{\psi}_m(\mathbf{x}, t)] \quad . \end{aligned} \quad (51)$$

With the following expressions for the relevant commutators,  $[\hat{\psi}_i^\dagger(\mathbf{r}, t), \hat{\psi}_j(\mathbf{x}, t)] = \delta_{i,j} \delta(\mathbf{r} - \mathbf{x})$ , and  $[\hat{\psi}_i(\mathbf{r}, t), \hat{\psi}_j(\mathbf{r}, t)] = 0$ ,  $i, j = a$  or  $m$ , we obtain the following coupled operator equations:

$$\begin{aligned} i\hbar\dot{\hat{\psi}}_a &= -\frac{\hbar^2\nabla^2}{2M}\hat{\psi}_a + \lambda_a\hat{\psi}_a^\dagger\hat{\psi}_a\hat{\psi}_a \\ &\quad + \lambda\hat{\psi}_m^\dagger\hat{\psi}_m\hat{\psi}_a + \sqrt{2}\alpha\hat{\psi}_m\hat{\psi}_a^\dagger \\ i\hbar\dot{\hat{\psi}}_m &= -\frac{\hbar^2\nabla^2}{4M}\hat{\psi}_m + \epsilon\hat{\psi}_m + \lambda_m\hat{\psi}_m^\dagger\hat{\psi}_m\hat{\psi}_m \\ &\quad + \lambda\hat{\psi}_a^\dagger\hat{\psi}_a\hat{\psi}_m + \frac{\alpha}{\sqrt{2}}\hat{\psi}_a^\dagger\hat{\psi}_a^\dagger \quad , \end{aligned} \quad (52)$$

where it is understood that all field operators depend on the same position. The operator equations (52), which provide an 'exact' description of the many-problem, are generally very difficult to solve. However, for dilute condensates, we obtain a closed set of equations for the condensate fields,  $\phi_a(\mathbf{r}) = \langle \hat{\psi}_a(\mathbf{r}) \rangle$  and  $\phi_m(\mathbf{r}) = \langle \hat{\psi}_m(\mathbf{r}) \rangle$ , by taking the expectation value of Eqs. (52). Furthermore, we assume that for the dilute gas systems considered here, we may take the condensed fields to be totally coherent in the sense that the expectation value of the products is equal to the product of the expectation values, e.g.  $\langle \hat{\psi}_a\hat{\psi}_a \rangle \approx \phi_a^2$ . This corresponds to a particular Gaussian trial wave functional in the Dirac time dependent variational scheme [41]. We find

$$\begin{aligned}
i\hbar\dot{\phi}_a &= \left[ -\frac{\hbar^2\nabla^2}{2M} + \lambda_a|\phi_a|^2 + \lambda|\phi_m|^2 \right] \phi_a + \\
&\quad \sqrt{2}\alpha\phi_m\phi_a^* \\
i\hbar\dot{\phi}_m &= \left[ -\frac{\hbar^2\nabla^2}{4M} + \epsilon + \lambda_m|\phi_m|^2 + \lambda|\phi_a|^2 \right] \phi_m + \\
&\quad \frac{\alpha}{\sqrt{2}}\phi_a^2.
\end{aligned} \tag{53}$$

These coupled non-linear equations replace the usual Gross-Pitaevskii equation that describes the time evolution of the dilute single condensate system [42] - [43]. Note that the  $\phi_m$ -field has a source term  $\propto \phi_a^2$  so that the expectation value of the molecular field operator is forced to take on a finite value when  $\phi_a \neq 0$ : the atom-molecule coupling creates a molecular condensate component in the presence of an atomic condensate.

The Gaussian trial wave function, which leads to an effective classical Hamiltonian density,

$$\begin{aligned}
\mathcal{H}_{eff} &= \phi_a^* \left[ -\frac{\hbar^2\nabla^2}{2M} + \frac{\lambda_a}{2}|\phi_a|^2 \right] \phi_a + \\
&\quad \phi_m^* \left[ -\frac{\hbar^2\nabla^2}{4M} + \epsilon + \frac{\lambda_m}{2}|\phi_m|^2 \right] \phi_m + \\
&\quad \lambda|\phi_a|^2|\phi_m|^2 + \frac{\alpha}{\sqrt{2}} \left[ \phi_m^*\phi_a^2 + \phi_m\phi_a^{*2} \right],
\end{aligned} \tag{54}$$

gives equations of motion identical to Eq.(53).

#### D. Effects of particle-loss

The experimental lifetime of the  $m$ -molecules is not only determined by the hyperfine induced spin flips, but also by collisions with other atoms, or even molecules. The importance of such three-body collisions is particularly pronounced as the recent experiments 'resonate' on a molecular state  $m$  of high vibrational quantum number (e.g.  $\nu = 14$  for the MIT experiment). The created quasi-bound molecules are consequently fragile and a collision with a third particle, atom or molecule, likely causes the molecule to decay into a state of lower vibrational quantum number. Such collisions that 'quench' the internal molecular

state are a potential problem for molecular condensation. They are also the most likely culprit for the particle loss that served as a signal to detect the Feshbach resonances in the MIT experiment.

Particle loss in atomic traps is usually described to sufficient accuracy, by very simple rate equations:

$$\begin{aligned}\dot{n}_a &= n_a [-c_{aa}n_a - c_{am}n_m] \\ \dot{n}_m &= n_m [-c_{ma}n_m - c_{mm}n_m] .\end{aligned}\tag{55}$$

In the above equation,  $n_a$  and  $n_m$  represent the particle densities of atoms and molecules,  $n_i = |\phi_i|^2$ , where  $i = a$  or  $m$ , if all particles are Bose condensed. The  $c_{i,j}$ -coefficients represent the rate coefficients for collisions between particles  $i$  and  $j$  that change the internal state of the  $i$ -particle. Typical values for the alkali-atoms are  $c_{aa} \sim 10^{-13} - 10^{-14} \text{cm}^3 \text{sec}^{-1}$ . The fragility of the loosely bound alkali dimers is expressed by atom-molecule and molecule-molecule state changing collision rates that exceed the atom-atom rates by several orders of magnitude:  $10^{-9} - 10^{-11} \text{cm}^3 \text{sec}^{-1}$ , where these numbers are estimates based on calculations with Hydrogen molecules [44]. A 'pure' molecular condensate of density  $10^{14} \text{cm}^{-3}$  of such dimers is then not expected to survive longer than  $10^{-3}$  seconds. Nevertheless, this time scale might actually suffice to study interesting molecular condensate physics. Furthermore, off-resonance, the molecular condensate is significantly smaller than the atomic condensate, which keeps 'replenishing' the small molecular condensate with bosons. The lifetime of the condensate mixture is then equal to the molecular lifetime divided by the fraction of molecules, e.g. a condensate system with a 1 percent molecular condensate can survive 100 times longer than a single molecule embedded in an atomic condensate of the same density.

Of equal importance is the question whether, and how, the atomic/molecular condensate system reaches its equilibrium as the detuning  $\epsilon$  is altered by varying the magnetic field. From the two-field coupling in the equations of motion, Eq.(53), we expect, as we discuss below, that a sudden change of the detuning is followed by oscillations in the atomic and molecular populations. From Eqs.(53), it would appear as if these oscillations persist indefi-

nately, or, at least until the condensates have disappeared due to state changing collisions or other three-body recombination processes. However, we point out that a correct treatment of the effects of state-changing collisions on the condensate predicts that the number oscillations damp out on a time scale that is the lifetime of a single molecule. As we noted above, the molecular condensate can survive for a much longer time and the condensate system can reach a 'quasi-equilibrium' state on a time scale small compared to its lifetime.

To build in the effects of state-changing collisions, we treat the loss-processes by considering the channels of all chemical reactions that remove particles from the atomic and molecular condensates. The elimination of the two-body collision channels in perturbation theory modifies the equations of motion (53) in a predictable way: the interaction strengths become absorptive with an imaginary part that determines the loss-rates. In the  $\phi_m$ -equation, for instance, the interaction strengths are replaced by  $\lambda_m \rightarrow \lambda - i\hbar c_{mm}/2$  and  $\lambda \rightarrow \lambda - i\hbar c_{ma}/2$ . We define the off-resonant regime to correspond to values of the detuning that exceed, in absolute value, the molecular kinetic energy, as well as any of the single particle interaction energies  $\epsilon \gg \lambda_m n_m, \lambda n_m, \lambda_a n_a, \lambda n_a$  and  $n_m \ll n_a$ . In this regime, we can neglect the variations of an initially constant atomic density and the main effect of the atomic condensate is that of a coherent (i.e. one that preserves the phase information) reservoir of atoms for the molecular condensate. In that limit, the molecular field equation of motion is linear:

$$i\hbar\dot{\phi}_m = \left[ \epsilon(t) + \lambda n_a - i\frac{\gamma_m}{2} \right] \phi_m + \frac{\alpha}{\sqrt{2}} \phi_a^2(t) , \quad (56)$$

where  $\gamma_m$  corresponds to the molecule loss-rate:  $\gamma_m/\hbar = c_{ma}n_a$ . In the same off-resonant limit, the atomic condensate field, to lowest order, propagates without feeling the effect of the molecular condensate:  $\phi_a(t) \approx \sqrt{n_a} \exp(-i\lambda_a n_a t/\hbar)$ . The solution to Eq.(56) for a detuning that is suddenly shifted from its initial value to  $\epsilon_f$  then gives the time dependent field  $\phi_m(t)$ :

$$\phi_m(t) = \phi_\infty \exp \left[ -\frac{it}{\hbar} (2\lambda_a n_a) \right] +$$

$$[\phi_0 - \phi_\infty] \times \exp \left[ -\frac{it}{\hbar} (\epsilon_f + \lambda n_a) \right] \exp \left( -\frac{\gamma_m t}{2\hbar} \right) , \quad (57)$$

where  $\phi_0$  represents the initial value of the molecular field,  $\phi_0 = \phi_m(t = 0)$ , and  $\phi_\infty$  is the value that the molecular field tends to at large times,  $t \gg (\hbar/\gamma_m)$ ,  $\phi_\infty = -\alpha n_a / [\sqrt{2}(\epsilon_f + \lambda n_a - 2\lambda_a n_a - i\gamma_m/2)] \approx -\alpha n_a / (\sqrt{2}\epsilon_f)$ , which is the quasi-equilibrium value of the molecular condensate field. Note that the molecular condensate density has an oscillating contribution  $\sim 2|(\phi_0 - \phi_\infty)\phi_\infty^*| \cos[(\epsilon_f + \lambda n_a - 2\lambda_a n_a)t/\hbar] \exp(-\gamma_m t/2\hbar)$ . This oscillation in the molecular population is caused by the interference of the propagation of the initial molecular field amplitude with the amplitude stemming from the coherent inter-condensate exchange of atom pairs. Thus, the oscillations are pure quantum effects and, if observed, provide strong evidence for the presence of a molecular condensate. Similarly, the atomic condensate density will oscillate with twice the amplitude of the molecular density oscillations, since each molecule that appears takes out two atoms from the condensate. These oscillations of the population imbalance are similar to the oscillating current observed at Josephson junctions. Here, due to the additional concern of boson-decay, the oscillations damp out on a time scale of  $(\hbar/\gamma_m)$ . Thus, in addition to leading to particle loss, the state changing collisions serve as relaxation processes, allowing the molecular condensate to approach its 'static' value  $\phi_\infty \approx -\alpha n_a / (\sqrt{2}\epsilon)$ . For a more general variation of the detuning with time, the oscillations appear if the rate of change of the detuning  $|\dot{\epsilon}|$  exceeds  $|\epsilon\gamma_m/\hbar|$ , or alternatively, if the rate of change of the magnetic field exceeds  $|\dot{B}/[B - B_m]| \gg (\gamma_m/\hbar)$ . In the opposite limit,  $|\dot{\epsilon}/\epsilon| < \gamma_m/\hbar$ , the system adiabatically follows its quasi-equilibrium state,  $\phi_m(t) \approx -\alpha n_a(t) / [\sqrt{2}\epsilon(t)]$ .

With the expression for the static value of the molecular field, we can also obtain the loss-rate of the condensate system in the off-resonant regime. We define the loss of the condensate system as the loss of atomic particles that occupy the atomic/molecular condensate. The total density of atomic condensate particles,  $n$ , is then equal to  $n = n_a + 2n_m$ , where we count each molecule as two atomic particles. The corresponding loss-rate of the atomic condensate particles is given by



$$\dot{n} = \dot{n}_a + 2\dot{n}_m = -n_a[c_{aa}n_a - 2c_{ma}n_m] , \quad (58)$$

where, in the off-resonant regime, we only account for atom-atom and molecule-atom collisions,  $\dot{n}_a \approx -c_{aa}n_a^2$  and  $\dot{n}_m \approx -c_{ma}n_m n_a$ . In the case of a slowly varying magnetic field,  $n_m \approx [\alpha^2 n_a^2 / 2\epsilon^2]$ , so that, with  $n_a \approx n$ , we find

$$\dot{n} = -c_{aa}n^2 - \frac{c_{ma}\alpha^2}{\epsilon^2}n^3 . \quad (59)$$

As a three-body collision process, the molecular-state changing collisions contribute a term  $b n^3$ , proportional to  $n^3$ . From Eq.(59) we find that the three-body collision rate is equal to  $b = c_{ma}\alpha^2/\epsilon^2$ .

We note that the above expressions are only valid in the far off-resonant limit. Closer to resonance, the nonlinearities of the equations become important and have to be included. However, in a qualitative sense, the response of a condensate to a sudden variation of the magnetic field agrees with the off-resonant behavior: out-of-phase oscillations of the atomic and molecular densities are damped out, after which the total condensate density decays exponentially, as shown in Fig.(2).

### E. Conclusions and remarks regarding molecular condensate formation

The population imbalance oscillations, discussed above in the far-off resonance regime, illustrate that the presence of a molecular condensate leads to observable effects. Thus, the appearance of a molecular condensate component is not a semantic issue but one that implies fundamental and observable differences in the many-body physics of the condensate system.

Before proceeding, we point out that the formation of a molecular condensate component does not defy common sense. For instance, in the limit that the molecules are destroyed extremely rapidly, the oscillations that occur in response to a sudden change of the magnetic field are 'overdamped', and we won't find a signal of the molecular condensate. It is

furthermore interesting and important to note that the lifetime of the molecular condensate can greatly exceed the lifetime of a single molecule.

We also point out that the appearance of a condensate of molecules does *not* violate conservation of energy. It is true that in the binary collision the energy and momentum the molecule receives from the incident atoms are generally 'off-shell', so that the creation of the molecule can only be understood in the sense of a 'virtual' particle. However, this consideration does not preclude the appearance of a molecular condensate. In a condensate system, it is not possible to assign a definite energy to a single particle component of the full wave function. Nevertheless, we may assign a chemical potential to each condensate:  $\mu_a$  represents the chemical potential of the atomic and  $\mu_m$  the chemical potential of the molecular condensate. These chemical potentials include the non-linear self-interaction terms (e.g.  $\lambda_a |\phi_a|^2$  is one contribution to  $\mu_a$ ) and  $\mu_m$  includes the single molecule energy  $\epsilon$ . The energy required to make a single molecule is then equal to  $\mu_m - 2\mu_a$ . Interestingly, in the equilibrium condensate mixture,  $\mu_m = 2\mu_a$ , as we discuss in the next section. Thus, it costs no energy to make a molecule in the equilibrium condensate mixture, even though  $\epsilon$  may be different from zero. In that sense the energy and momentum of the molecules are 'on shell' and the molecules may be treated as long-lived 'real' particles without violating any conservation laws.

#### IV. STATICS

Under the present experimental conditions, particle loss is an important effect to be reckoned with. Nevertheless, in the near-future, it might be possible to 'resonate' on less fragile molecular states (i.e. with lower values for the  $c_{ma}$ -collision rates). Even with the present systems, it might be possible to let a near-resonant condensate relax to its quasi-equilibrium state. In this section, we discuss relevant aspects of this equilibrium. We find, for example, that some of the unphysical predictions of the  $\lambda_{eff}$  description on resonance are avoided in the condensate mixture description. Furthermore, we show that the stationary

state of the system depends on its history. For instance, the state of the system at a near-resonant value for the detuning may be different if it was obtained by an adiabatic lowering or by an adiabatic increase of the detuning. Near resonance, the homogeneous BEC ground state is unstable in the limit of very low atomic particle density. At higher densities, on the other hand, the condensate can be stable with respect to density fluctuations, in contrast to the predictions of the  $\lambda_{eff}$ -description. At higher atomic particle densities,  $n$ , the inter-particle interaction dominates over the atomic-molecular condensate coupling and can stabilize the system. As a function of the density  $n$ , the energy goes through a minimum at a density where the inter-condensate coupling interaction is of the order of the inter-particle interaction energy per particle. The appearance of a minimum suggests the possibility of creating condensate systems with the liquid-like property of a self-determined density. A low density condensate would spontaneously adjust its volume to settle in a state with density equal to the value at which the energy is minimized. From previously calculated numbers, it appears that the corresponding density is only of the order of  $10^{14} - 10^{15} \text{ cm}^{-3}$ , and the resulting system would be a rarified liquid!

In the  $\lambda_{eff}$ -description of the on-resonant BEC, the dispersive dependence of the effective scattering length on the magnetic field gives a diverging inter-boson interaction. For the purpose of visualizing the inter-particle interactions, one can picture the bosons as hard spheres with radius equal to the scattering length (assuming  $a > 0$ ). The fraction of space occupied by the spheres diverges as  $\epsilon$  approaches zero,  $\epsilon \rightarrow 0$ , and the system is not a weakly interacting single condensate. Not surprisingly, a naive extrapolation of the dilute condensate results leads to unphysical predictions for macroscopic measurable quantities in this limit. For instance, the chemical potential,  $\mu$ , and the pressure,  $P$ , of dilute condensates of interaction strength  $\lambda$  and density  $n$  are given by

$$\begin{aligned} \mu &= \lambda n \quad , \\ P &= \frac{\lambda}{2} n^2 \quad . \end{aligned} \tag{60}$$

Hence, a simple replacement  $\lambda \rightarrow \lambda_{eff}$  gives diverging values that change discontinuously

from  $+\infty$  to  $-\infty$  at the pole,  $\epsilon = 0$ . Of course, one can argue that near resonance, the scattering length description of the collision breaks down, or, if nothing else, that the expressions (60), valid only for weakly interacting condensates, do not apply. However, in the hybrid atomic/molecular condensate picture, we find that the unphysical divergencies do not occur. Near and on-resonance, the equilibrium system simply arranges the atomic and molecular densities so as to insure that  $\mu_m = 2\mu_a$ , where  $\mu_m$  and  $\mu_a$  respectively denote the chemical potentials of the atoms and the molecules.

### A. Static Equations

In this section, we will assume that the condensate system has relaxed to a stationary state and that the effects of particle loss are negligible on the time scale that the system is observed. We determine the stationary state variationally. A first remark of relevance to the determination of the ground state of the system is that the inter-condensate coupling interaction ( $\hat{H}_{PM} + \hat{H}_{MP}$ ) necessarily lowers the ground state many-body energy. Indeed, the coupling contribution,  $\sim \alpha\phi_m^*\phi_a^2 + h.c.$ , sensitive to the relative phase of the atomic and molecular condensate fields, can and will be negative in the lowest energy state. As usual in describing the static system, we may choose the fields to be real, but the relative sign of the ground state fields is determined by minimizing  $\alpha\phi_a^2\phi_m$ . Without loss of generality, we will assume that  $\alpha > 0$ , and we will choose the phase of  $\phi_a$  to be zero,  $\phi_a > 0$ , so that the ground state value of  $\phi_m$  is negative,  $\phi_m < 0$ .

If we neglect particle loss, the total number of atomic particles,  $N = \int d^3r n(\mathbf{r}) = \int d^3r [|\phi_a(\mathbf{r})|^2 + 2|\phi_m(\mathbf{r})|^2]$ , is a conserved quantity. To account for this, we may vary the Hamiltonian with a Lagrange multiplier  $\mu$ ,  $\delta(H_{eff} - \mu N)/\delta\phi_a^*(\mathbf{r}) = 0$ , and  $\delta(H_{eff} - \mu N)/\delta\phi_m^*(\mathbf{r}) = 0$ . These equations take on the form of coupled time-independent Gross-Pitaevskii equations,

$$\mu\phi_a = \left[ -\frac{\hbar^2\nabla^2}{2M} + \lambda_a\phi_a^2 + \lambda\phi_m^2 \right] \phi_a + \sqrt{2}\alpha\phi_m\phi_a ,$$

$$2\mu\phi_m = \left[ -\frac{\hbar^2\nabla^2}{4M} + \epsilon + \lambda_m\phi_m^2 + \lambda\phi_a^2 \right] \phi_m + \frac{\alpha}{\sqrt{2}}\phi_a^2, \quad (61)$$

of the form of Eq.(53) with the time-derivatives replaced by the corresponding chemical potential,  $i\hbar\dot{\phi}_i \rightarrow \mu_i\phi_i$ . Note that  $\mu$  in the  $\phi_a$  equation plays the role of the atomic chemical potential,  $\mu_a = \mu$ , and  $2\mu$  in the  $\phi_m$  equation plays the role of the molecular chemical potential,  $\mu_m = 2\mu$ . Thus, the condition of chemical equilibrium is satisfied:  $\mu_m = 2\mu_a$ .

In the limit of large detuning, by which we mean here that  $|\epsilon|$  greatly exceeds the kinetic energy as well as any of the molecule interaction energies,  $|\epsilon| \gg \lambda_m n_m, \lambda n_a$ , we find that the corresponding approximations in the Gross-Pitaevskii equation of the molecular condensate lead to:

$$\phi_m \approx -\frac{\alpha}{\sqrt{2\epsilon}}\phi_a^2, \quad |\epsilon| \gg 0, \quad (62)$$

in agreement with the value of the quasi-equilibrium molecular field found in Eq.(57). The insertion of this expression into the  $\phi_a$ -equation of (61) yields an effective single condensate Gross-Pitaevskii equation for  $\phi_a$ :

$$\mu\phi_a = \left[ -\frac{\hbar^2\nabla^2}{2M} + \left(\lambda_a - \frac{\alpha^2}{\epsilon}\right)n_a \right] \phi_a. \quad (63)$$

Thus, in the off-resonant limit, the stationary atomic condensate behaves effectively as a single atomic condensate with an inter-atomic interaction described by the effective interaction strength Eq.(35) of the binary atom system. However, the effective single condensate Gross-Pitaevskii equation (63) does not describe the appearance of the small molecular condensate, and consequently, cannot describe the interesting dynamical effects discussed in the previous section.

Note that the molecular condensate in the off-resonant limit of negative detuning takes on the same sign as the atomic condensate. This does not contradict the remark that the  $H_{PM}$  interaction is minimized by fields of opposite sign. The variational procedure with Lagrange multiplier  $\mu$  finds an extremum, but does not guarantee that the extremum is a minimum. In fact, the actual ground state in the off-resonant regime of negative detuning is the 'trivial'

all-molecule solution to Eqs.(61),  $\phi_a = 0$ . Interestingly, the state  $\phi_m \approx -\alpha n_a / \sqrt{2\epsilon}$ ,  $\epsilon \ll 0$ , represents a local *maximum* of the energy.

To see that, we reformulate the variational problem. Instead of introducing the chemical potential, we insure particle conservation by substituting  $n_a = n - 2n_m$ . If the system is large enough to neglect surface effects, we may omit the kinetic energy and work with an energy density,

$$u = \frac{\lambda_a}{2} n_a^2 + \frac{\lambda_m}{2} n_m^2 + \lambda n_m n_a + \epsilon n_m + \sqrt{2} \alpha n_a \phi_m . \quad (64)$$

We parametrize the 'population imbalance' or the fraction of atoms that are converted to molecules by a scaled molecular field  $x$ ,  $\phi_m = x\sqrt{n/2}$ , which can take on values between  $-1$  and  $+1$ . In terms of the molecular field parameter, the atomic condensate density is  $n_a = n(1 - x^2)$ , and the energy per particle,  $e = u/n$  is a simple quadratic polynomial in  $x$ :

$$e = \frac{\lambda_a}{2} n + \alpha \sqrt{n} x + \{[-\lambda_a + \lambda/2]n + \epsilon/2\} x^2 - \alpha \sqrt{n} x^3 + [\lambda_a/2 - \lambda_m/4 - \lambda/2] n x^4 . \quad (65)$$

Minimization of the energy  $e$ , yields the value of the ground state expectation value of  $x$ .

In the interest of simplicity, we start by considering Feshbach resonances in ideal gas condensates, i. e. we consider the limit  $\lambda = \lambda_a = \lambda_m = 0$ . In that case the energy per atomic particle is equal to

$$e_{id} = \alpha \sqrt{n} \left[ \epsilon' \frac{x^2}{2} + x(1 - x^2) \right] , \quad (66)$$

where we introduced the scaled detuning,  $\epsilon' = \epsilon / [\alpha \sqrt{n}]$ . We determine the expectation value of the molecular condensate field,  $\phi_m = x\sqrt{n/2}$ , for a system of atomic particle density  $n$ ,  $n = n_a + 2n_m$ , variationally by finding the extrema of  $e_{id}(x, n)$  for fixed value of  $n$ ,  $\partial e_{id} / \partial x = 0$ . Within the limitation that  $|x| \leq 1$ , we find two solution for any given value of  $\epsilon' = \epsilon / [\alpha \sqrt{n}]$ :

$$x_+(\epsilon') = \left[ \epsilon' + \sqrt{\epsilon'^2 + 12} \right] / 6 , \text{ if } \epsilon' \leq 2 ,$$

$$\begin{aligned}
&\text{and } x_+(\epsilon') = +1, \text{ if } \epsilon' > 2, \\
&x_-(\epsilon') = [\epsilon' - \sqrt{\epsilon'^2 + 12}] / 6, \text{ if } \epsilon' \geq -2, \\
&\text{and } x_-(\epsilon') = -1, \text{ if } \epsilon' < -2.
\end{aligned} \tag{67}$$

The  $x_+ = +1$  and  $x_- = -1$  correspond to the above mentioned trivial all-molecule solution,  $\phi_a = 0$ . Although the all-molecule solution represents a third solution in the detuning interval  $-2 < \epsilon' < +2$ , the analysis of the next paragraph shows that this solution is unstable in this detuning region. The previously obtained off-resonant limit (62),  $\phi_m \approx -\alpha n / \sqrt{2}\epsilon$  or  $x \approx -1/\epsilon'$ , corresponds to  $x_-$  if  $\epsilon \gg 0$  and  $x_+$  if  $\epsilon \ll 0$ . The molecular field solution of positive sign,  $x_+$ , corresponds to a maximum of the energy, as we can see from Fig.(3). Nevertheless, as we discuss below, the molecular condensate field value  $x$  cannot simply 'roll down' on the energy curve. Such changes of the relative population of the atomic and molecular condensates also involve a change of the relative phase. From the dynamical treatment of the population changes, it follows that both solutions,  $x_+$  and  $x_-$ , are stable with respect to small perturbations of the condensate populations. This is a significant conclusion, since it suggests that the near-resonant BEC-system can reside in two distinct stationary states.

## B. Near-Equilibrium Population Dynamics

How can a maximum in the energy correspond to a dynamically stable state? To gain insight into this issue, we construct the classical Hamiltonian that describes the dynamics of position independent atomic and molecular fields. Again, we limit the discussion to the 'ideal gas' condensate mixture,  $\lambda = \lambda_a = \lambda_m = 0$ , although the treatment can easily be generalized to include the non-linear interactions. In terms of the density and phase variables,  $\phi_a = \sqrt{n_a} \exp(i\theta_a)$ ,  $\phi_m = \sqrt{n_m} \exp(i\theta_m)$ , we obtain the following expression for the Lagrangian density of the Dirac variational principle:

$$\mathcal{L} = \frac{i\hbar}{2} [\phi_a^* \dot{\phi}_a - \dot{\phi}_a^* \phi_a + \phi_m^* \dot{\phi}_m - \dot{\phi}_m^* \phi_m] - \mathcal{H},$$

$$\begin{aligned}
&= -\hbar\dot{\theta}_a n_a - \hbar\dot{\theta}_m n_m - \epsilon n_m \\
&\quad - \alpha \sqrt{2n_a} \sqrt{n_m} \cos(\theta_m - 2\theta_a) .
\end{aligned} \tag{68}$$

We now simplify the expressions by making use of particle conservation,  $n_a = n - 2n_m$ . We also introduce the relative phase variable,  $\theta = 2\theta_a - \theta_m$ . In terms of the relative phase and the total number of molecules,  $N_m$ , the Lagrangian-per-particle,  $L = \int d^3r \mathcal{L}/N$ , reads

$$\begin{aligned}
L &= -\hbar\dot{\theta} + \hbar\dot{\theta}(N_m/N) - \epsilon(N_m/N) \\
&\quad - \alpha \sqrt{n}(N_m/N)[1 - 2(N_m/N)] \cos(\theta) ,
\end{aligned} \tag{69}$$

where  $N$  represents the total number of atoms. The fraction of molecules ( $N_m/N$ ), which can take on values from 0 to 1/2, is the (dimensionless) conjugate momentum of the relative phase variable,

$$\frac{\partial L}{\partial \dot{\theta}} = \hbar(N_m/N) = \hbar p . \tag{70}$$

The  $\dot{\theta}_a$ -dependence of the Lagrangian does not contribute to the equations of motion, expressing the fact that one of the condensate phases can be chosen arbitrarily and that  $\theta$  is the only dynamical phase variable. From Eqs.(69) and (70) we find the classical Hamiltonian  $H$  that governs the time evolution of the two-field system by identifying  $H$  from  $L = \dot{\theta}p - H$ :

$$H = \epsilon p + \alpha \sqrt{n} \sqrt{2p} (1 - 2p) \cos(\theta) . \tag{71}$$

The corresponding equation of motion,

$$\begin{aligned}
\hbar\dot{\theta} &= \frac{\partial H}{\partial p} = \epsilon + [\alpha \sqrt{n}] \left( \frac{1 - 6p}{\sqrt{2p}} \right) \cos(\theta) ; \\
\hbar\dot{p} &= -\frac{\partial H}{\partial \theta} = -[\alpha \sqrt{n}] \sqrt{2p}(1 - 2p) \sin(\theta) ,
\end{aligned} \tag{72}$$

lead to the stationary states  $x_+, x_- < 1$ , by requiring  $\partial H/\partial p = \partial H/\partial \theta = 0$  at the equilibrium solution  $(\theta_0, p_0)$ . The  $\dot{p} = 0$ -equation gives  $\sin(\theta_0) = 0$ , corresponding to atomic and molecular fields of the same,  $\theta_0 = 0$ , or opposite,  $\theta_0 = \pi$ , signs. The second requirement,  $\dot{\theta} = 0$ , results in a quadratic equation for  $\sqrt{2p} = |x|$  (where  $x$  denotes the scaled molecular field of the previous paragraph). The solution to this equation gives the stationary states:



$$\sqrt{2p_0} = [\sqrt{\epsilon'^2 + 12} + \cos(\theta_0)\epsilon'] / 6, \quad (73)$$

identical to the solutions of Eq.(67) in the regime  $|x_{\pm}| < 1$ . Since at  $(\theta_0, p_0)$ ,  $\partial^2 H / \partial p \partial \theta = 0$ , the small amplitude fluctuations,  $\tilde{\theta} = \theta - \theta_0$  and  $\tilde{p} = p - p_0$ , evolve in time according to the following linearized equations of motion:

$$\begin{aligned} \hbar \ddot{\tilde{\theta}} &\approx \frac{\partial^2 H}{\partial p^2} \tilde{p} = -\cos(\theta_0) \left[ \frac{1+6p}{(\sqrt{2p})^3} \right] \alpha \sqrt{n} \tilde{p} \\ \hbar \dot{\tilde{p}} &\approx -\frac{\partial^2 H}{\partial \theta^2} \tilde{\theta} = -\cos(\theta_0) \sqrt{2p} [1-2p] \alpha \sqrt{n} \tilde{\theta}. \end{aligned} \quad (74)$$

Consequently,  $\ddot{\tilde{\theta}} = -\omega^2 \tilde{\theta}$ , where  $\omega$  is a real-valued frequency, regardless of the sign of  $\cos(\theta_0) = \pm 1$ :

$$\begin{aligned} \hbar^2 \omega^2 &= \frac{\partial^2 H}{\partial p^2} \frac{\partial^2 H}{\partial \theta^2}, \\ &= [\alpha \sqrt{n}]^2 \frac{(1+6p_0)(1-2p_0)}{2p_0}. \end{aligned} \quad (75)$$

Thus, the fluctuation variables,  $\tilde{\theta}$  and  $\tilde{p}$  describe a harmonic motion in phase space, and the position-independent field fluctuations experience a restoring force, both at  $\theta_0 = 0$  (corresponding to  $x_+$ ) and at  $\theta_0 = \pi$  (corresponding to  $x_-$ ). The reason for the dynamical stability of the fluctuations around the  $\theta_0 = 0$ , in spite of the local maximum in the energy, follows from the analysis. Although the 'inverse mass' is negative,  $m^{-1} = \partial^2 H / \partial p^2 < 0$ , so is the 'spring constant',  $K = \partial^2 H / \partial \theta^2 < 0$ . The resulting frequency,  $\hbar \omega = \sqrt{K/m}$  is consequently real-valued.

Likewise, we may study the stability of the the 'all-molecule' solution,  $\phi_a = 0$ . In that case, the relative phase  $\theta$  is not a meaningful quantity, and it is more convenient to start from the field equations of motion:

$$\begin{aligned} i\hbar \dot{\phi}_a &= \sqrt{2} \alpha \phi_m \phi_a^* \\ i\hbar \dot{\phi}_m &= \epsilon \phi_m + \frac{\alpha}{\sqrt{2}} \phi_a^2. \end{aligned} \quad (76)$$

These equations have, in fact, been solved analytically in the field of non-linear optics [45]. Near the all-molecule solution,  $\phi_a = 0$ , the molecular field is approximately equal to

$\phi_m \approx \sqrt{n/2} \exp(-i\epsilon t/\hbar)$ . In the same limit, the fluctuations of the  $\phi_a$ -field are approximately decoupled from the  $\phi_m$ -fluctuations, and evolve according to

$$i\hbar\dot{\phi}_a = \alpha\sqrt{n} \exp(-i\epsilon t/\hbar)\phi_a^* . \quad (77)$$

Substitution of the following linear combination of positive and negative frequency fields of small amplitude,

$$\phi_a = \exp(-i\epsilon t/2\hbar)[a_+ \exp(i\omega t) + a_- \exp(-i\omega t)] , \quad (78)$$

into Eq.(77) yields a set of linear equations in the  $a_+$  and  $a_-$ -coefficients. The frequency at which the system near the all-molecule state oscillates after it has been perturbed by breaking up a few molecules into atoms, follows from the requirement of a non-trivial solution to the linear set of equations in  $a_+$  and  $a_-$ :

$$\hbar^2\omega^2 = (\epsilon/2)^2 - (\alpha\sqrt{n})^2 . \quad (79)$$

Thus,  $\omega$  is imaginary if  $|\epsilon| < 2\alpha\sqrt{n}$ , and the all-molecule system is unstable in the regime where both  $|x_+| < 1$  and  $|x_-| < 1$ .

Thus, at any value of the detuning, we find two (and not three) different stationary states the system can occupy. Which state the system will find itself in, depends on its history. For instance, if the condensate was brought near-resonance by adiabatically increasing the detuning from 'below resonance', the system is expected to reside in the  $x_+$ -state. In Fig.(4), we plot the molecule fraction of both stationary states as a function of the detuning. Interestingly, the analysis of the next paragraph shows that the all-molecule branch of the  $x_+$ -solution is unstable with respect to density fluctuations. In the figure, we indicate the instability by plotting the unstable branch in a dotted line.

### C. Considerations regarding the mechanical stability

The previous analysis does not give insight into the stability of the homogeneous system with respect to position dependent fluctuations of the densities. That this is a major concern in describing the dynamics of these systems will become clear from the discussion below.

A first example that illustrates the importance of position dependent field fluctuations, is the instability of the homogeneous all-molecule solution if  $\epsilon > 2\alpha\sqrt{n}$ . To see the instability, we allow for position dependent fluctuations and solve for plane wave  $\phi_a$ -fluctuations around the all-molecule solution,

$$\phi_a = \exp(-i\epsilon t/2\hbar)[a_+ \exp(-i[\mathbf{k} \cdot \mathbf{r} - \omega t]) + a_- \exp(i[\mathbf{k} \cdot \mathbf{r} - \omega t])] . \quad (80)$$

Upon insertion of this expression in the equations of motion (74), after restoring the kinetic energy term, we find that the resulting frequency,

$$\hbar^2 \omega_k^2 = \left( \frac{\hbar^2 k^2}{2M} - \frac{\epsilon}{2} \right)^2 - (\alpha\sqrt{n})^2 , \quad (81)$$

does become imaginary for a finite range of wavenumbers if  $\epsilon > 2\alpha\sqrt{n}$ . Specifically, the modes with  $\mathbf{k}$  vectors of magnitude  $\sqrt{M(\epsilon - 2\alpha\sqrt{n})} < \hbar k < \sqrt{M(\epsilon + 2\alpha\sqrt{n})}$  grow exponentially: the homogeneous  $x_+ = +1$ -branch is unstable and the system spontaneously generates a finite atomic condensate. Consequently, we show the corresponding branch of the  $x_+$ -state in Fig.(4) in dotted line, to indicate that the system will not remain in that state. Whether the other branches are stable, is a question that can be answered from a more general RPA-study, and will be the subject of further research.

However, even without the knowledge of the excitation modes, we can make statements about the mechanical stability of the system by investigating the pressure, for example. Whereas we could omit the inter-particle interactions to get an accurate qualitative picture of the population dynamics, their effects in the mechanical stability of the homogeneous system are all-important and they have to be considered. Indeed, the ground state of the homogeneous near-resonant ideal gas-condensate is always unstable. This follows from the pressure,  $P$ , which we determine from the energy per atomic particle,  $e$ ,  $P = n^2(de/dn)$ :

$$\begin{aligned} P &= n^2 \frac{\partial e}{\partial x} \frac{dx_0}{dn} + n^2 \frac{\partial e}{\partial n} \\ &= n^2 \frac{\partial e}{\partial n} , \end{aligned} \quad (82)$$

where  $x_0$  represents the actual value of the scaled molecular field, which follows from  $\partial e/\partial x = 0$ . In the ideal gas condensate,  $e = e_{id} = (\epsilon/2)x^2 + \alpha\sqrt{n}(1-x^2)x$ , we find

$$P_{id} = n \frac{\alpha\sqrt{n}}{2} x_0(1-x_0^2), \quad (83)$$

so that its pressure has the same sign as the molecular field. In the ground state system, for which  $x_0 = x_- < 0$ ,  $P$  is negative and the homogeneous system is unstable. Furthermore, we expect the stationary state of positive molecular field,  $x_+$ , to be equally unstable, as its energy can be lowered by spatially separating the atomic and molecular condensates.

The inter-particle interactions can stabilize the homogeneous condensate system, but only at higher values for the density or in the off-resonant regime. The pressure that follows from Eq.(82) can be written as

$$P = \frac{\lambda_a}{2} n_a^2 + \frac{\lambda_m}{2} n_m^2 + \lambda n_m n_a + \frac{\alpha\sqrt{n}}{2} n x (1-x^2). \quad (84)$$

In this expression, we recognize the 'elastic' inter-particle interaction contributions in addition to the atom-molecule Feshbach resonant coupling term that was the sole contribution to  $P_{id}$ . The inter-particle interaction contributions are proportional to  $n^2$ ,  $\lambda_r n^2/2$ , where  $\lambda_r$  is an interaction strength of a magnitude that is representative of  $\lambda_a$ ,  $\lambda_m$  and  $\lambda$ . The  $\alpha$ -contribution to the ground state pressure, on the other hand, is of order  $-\alpha n^{3/2}$  if the system is near-resonance,  $\epsilon \approx 0$ , and of order  $-\alpha n^{3/2} \times (\alpha\sqrt{n}/[2\epsilon])$  in the off-resonant regime. Near-resonance, the inter-particle interaction and atom-molecule coupling contributions are of the same order of magnitude if  $n \approx n_r = (\alpha/\lambda_r)^2$ . In the limit of vanishing density,  $n \ll n_r = (\alpha/\lambda_r)^2$ , the density-dependent contribution to  $e$  is dominated by the Feshbach resonant interaction and the system behaves as the near-resonant ideal gas system. In contrast, at 'high' densities,  $n \gg n_r$ , the inter-particle interactions dominate, so that the molecular population is determined by the fraction that minimizes the inter-particle interaction. At these higher densities, the Feshbach resonant interactions can be treated as a perturbation and generally do not determine the stability of the system. Thus, at higher densities the condensate system will be stable if, for example,  $\lambda_a, \lambda_m > 0$  and  $\lambda^2 < \sqrt{\lambda_a \lambda_m}$

( [18]). Note that this behavior is very different from the predictions that follow from the  $\lambda_{eff}$ -description,  $\lambda \rightarrow \lambda_{eff} = \lambda_a - \alpha^2/\epsilon$ , which predicts that the homogeneous system has a negative pressure,  $P = \lambda_{eff}n^2/2 < 0$ , if  $0 < \epsilon < (\alpha^2/\lambda_a)$ , regardless of the density.

If the homogeneous near-resonant condensate system is stable, the slope of the many-body ground state energy (proportional to the pressure) changes sign as the atomic particle density  $n$  is increased. The corresponding change in sign of the pressure, occurs in the density regime of  $n \sim n_r = (\alpha/\lambda_r)^2$  for near-resonant detuning.

Remarkably, the existence of a minimum in the energy-versus-density curve implies that the negative value of the pressure at low densities does not necessarily lead to collapse of the condensate system. Indeed, the unstable system that was initially at very low density  $n < n_r$  can decrease its volume until its density reaches the value at which the energy is minimized. In the assumption that the system can relax to its stationary ground state, the system does not collapse, but settles at the volume that corresponds to the self-determined density. Interestingly, with the parameters calculated for the observed  $^{23}\text{Na}$ -resonances the value of this density  $\sim n_r$ , is only of the order of typical atomic-trap condensate densities,  $n_r \sim 10^{14} - 10^{15} \text{cm}^{-3}$ . Moreover, the atomic-trap set-up lends itself very well to the observation of this unusual property. After switching the trap-potential off, the condensate of self-determined density does not fly apart, but stays together as a 'blob' of approximately uniform density.

The above considerations suggest that the equilibrium Feshbach-resonant BEC can be an ultra-dilute gas with the liquid-like property of a self-determined density. Whether the liquid-like system is really stable, will have to follow from an RPA-study of its excitations. Nevertheless, it is certainly a fascinating thought that the dilute atomic condensates with Feshbach resonances might yield the first example of a rarified liquid!

## V. CONCLUSIONS

In this paper, we discussed the theoretical description of the hyperfine-induced low-energy Feshbach resonances in the neutral atom interactions of condensates. Such resonances were recently observed in atomic-traps, of the type used to achieve Bose-Einstein condensation. In fact, one of the observations of the resonance was carried out in a BEC [1].

We pointed out that the Feshbach resonances affect the condensate physics in a more profound manner than the alteration of the effective inter-particle interaction. The atom-molecule coupling that gives rise to the resonance in binary-atom collisions, creates a second molecular condensate component in the many-body BEC-system. Even if the molecular condensate density remains small in the off-resonant detuning regime ( $\alpha^2 n / \epsilon^2 \sim$  a few percent or so), the presence of the molecular condensate can still be detected by suddenly varying the magnetic field and observing oscillations in the atomic and molecular populations. If the molecular condensate survives long enough to reach its equilibrium, the resulting double condensate displays fascinating properties: (i). The state of the near-resonant condensate depends on its history. For instance, the behavior of the BEC that is taken through the resonance by adiabatically varying the detuning, will be different if the system starts out above or below the resonance. (ii). Although, in the limit of vanishing density, the ground state of the near-resonant BEC can experience negative pressure, in agreement with the  $\lambda_{eff}$ -description, the homogeneous higher density system can be stabilized by the inter-particle interactions of the atoms and molecules. In that case, the ground state energy goes through a minimum as the atomic particle density is increased. This suggests the remarkable, liquid-like property of self-determined density. Thus, by tuning an external magnetic field near resonance, an atomic condensate could become the first rarified liquid to be observed in nature.

## VI. ACKNOWLEDGEMENTS

This paper is dedicated to the memory of R. Slansky, who, we think, would have been amused by this application of field theory to the many-body condensate problem.

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## Figure Captions

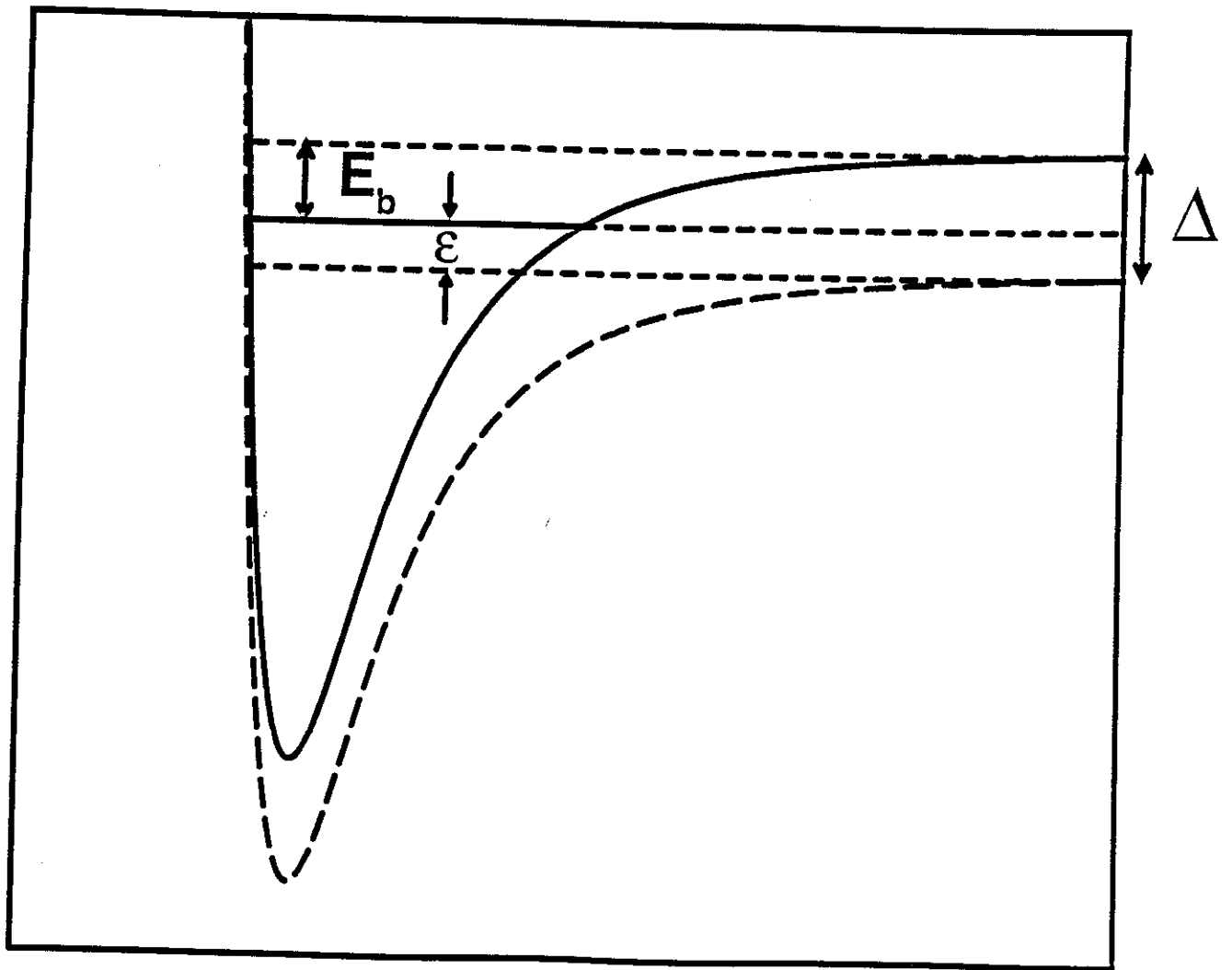
Fig.1 : Schematic representation of the molecular potentials of the incident and intermediate state channels. The energy difference of the continuum levels,  $\Delta$ , is the sum of the binding energy, denoted here by  $E_b$ , of the quasi-bound state and the 'detuning'  $\epsilon$ .

Fig.2 : 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  $n = 10^{14} \text{cm}^{-3}$  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,  $\lambda n = \lambda_m n = \lambda_a n = \alpha\sqrt{2n} = 10^5 \text{ Hz}$ , and of the decay parameters,  $c_{ma} = c_{mm} = 5 \times 10^{-10} \text{cm}^3 \text{sec}^{-1}$  (while neglecting the atomic decay) are realistic.

Fig.3 : The energy per atom particle of a homogeneous mixture of Feshbach resonant coupled atomic and molecular condensates, as a function of the scaled molecular field  $x$ , ( $\phi_m = x\sqrt{n/2}$ ), at a fixed detuning, chosen so that  $\epsilon' = \epsilon/[\alpha\sqrt{n}] = 0.5$ . The energy is shown in units of the tunneling energy,  $\alpha\sqrt{n}$ , and the curve was calculated in the assumption of non-interacting, or 'ideal', atomic and molecular condensates ( $\lambda = \lambda_a = \lambda_m = 0$ ).

Fig. 4 : Plot of the molecule fraction,  $(N_m/N)$ , as a function of the detuning, scaled by the 'tunneling energy',  $\alpha\sqrt{n}$ , for both stationary state solutions, called  $x_+$  and  $x_-$  in the text to denote a molecular field of, respectively, positive and negative sign. The atomic and molecular condensates were assumed to be 'ideal' in this calculation, i.e.  $\lambda = \lambda_a = \lambda_m = 0$ . The all-molecule branch,  $(N_m/N) = 1/2$ , of the  $x_+$ -state is shown in dotted line to indicate that the homogeneous  $x_+$ -branch is unstable in this regime, as explained in the text.

Energy



Internuclear distance

