Molecular Signatures in Hybrid Atomic/Molecular Bose-Einstein Condensates

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As it was proposed and recently verified experimentally, the mechanism of Feshbach resonance in a condensate can create a second condensate component of molecules that coexists with the atomic condensate. In this work we investigate signatures of the presence of the molecular condensate through the equilibrium properties and collective excitations of the hybrid system of atoms and molecules, subjected to a trap, employing a time-dependent variational ansatz. We show that the shape of the condensate changes significantly by the presence of the molecules and that modes unique to this hybrid system appear at observable frequencies.

The recent realization of Bose-Einstein condensation (BEC) in a dilute gas of alkali atoms has open a new opportunity for the theoretical and experimental investigation in quantum degenerate many-body systems. In contrast to superfluid helium, these weakly interacting gases are much more amenable to theoretical prediction and quantitative analysis. After the observation of such condensates, a new generation of more complex experiments has been done involving the production of multiple species BEC. Such mixture may consist of different hyperfine states of the same atom or different spin state of the same atomic species. In the former case the composition of mixture remains fixed and in the latter case the internal conversion leads to important phenomena. It has been proposed that the interaction through the Feshbach resonance that bring a binary atom system to an intermediate state molecule recently observed in BEC, create in the dilute atomic Bose-Einstein condensate (BEC) a second molecular condensate. We should mention also the case of a dilute atomic BEC coupled to a molecular Bose gas by coherent Raman transitions, which may lead to what is coined superchemistry.

Recently, several groups have been working in order to detect a signature of molecule formation in BEC with Feshbach resonance. Since these systems allow experimentalists to change the magnetic filed (and consequently the detuning) one can probe these systems in two different ways: 1 - adiabatic, where the detuning can be changed slowly (waiting for the system to equilibrate) and 2 - non-adiabatic, where a sudden change of the magnetic field is performed (usually with a pulse). The non-adiabatic change leaving the system out of equilibrium will produce oscillations, among other effects, in the number of atoms, and that was indeed observed.

In the adiabatic change the system is always at the equilibrium configuration. In this paper we show that in this regime the shape of the equilibrium condensate cloud of atoms and the behavior of the excitation energies of the collective modes can be a signature of the presence of molecules. It will be shown that the effects of the presence of molecules are more important and could be detected experimentally for weak resonances.

We start by reviewing the many body theory for a condensate on a Feshbach resonance. In a two component picture the mean field Hamiltonian density describing the trapped system is

$$\mathcal{H} = \phi_a^* \left[-\frac{\hbar^2 \nabla^2}{2m} + V_a + \frac{\lambda_a}{2} \phi_a^* \phi_a \right] \phi_a$$

$$+ \phi_m^* \left[-\frac{\hbar^2 \nabla^2}{4m} + V_m + \varepsilon + \frac{\lambda_m}{2} \phi_m^* \phi_m \right] \phi_m$$

$$+ \lambda_{am} \phi_a^* \phi_a \phi_m^* \phi_m + \frac{\alpha}{\sqrt{2}} \left[ \phi_m^* \phi_a \phi_m + \phi_m^* \phi_a \phi_m \right],$$

where $\phi_{am}$ denote the macroscopic wave function of the atom (molecule) condensate, $\varepsilon$ is the detuning parameter, $\alpha$ is the Feshbach resonance parameter introduced in Ref., and we assume that the confining potential for the atoms and molecules are harmonic with axial symmetry, $V_a = \frac{m \omega^2}{2} (\beta_{a} x^2 + \beta_{a} y^2 + \beta_{a} z^2)$, $V_m = m \omega^2 (\beta_{m} x^2 + \beta_{m} y^2 + \beta_{m} z^2)$. As usual, the Gross-Pitaevskii equations of motion for this model can be derived by imposing that the action

$$\Gamma = \int dt \left\{ \frac{d^3 \hat{r}}{2} \left[ \phi_a^* \phi_a + \phi_m^* \phi_m \right] - E[\phi_a, \phi_m] \right\},$$

where $E[\phi_a, \phi_m] = \int d^3 \hat{r} \mathcal{H}$, is stationary with respect to the variation of $\phi_{am}$, subject to the normalization condition constraint $\int d^3 \hat{r} |\phi_a|^2 + 2 |\phi_m|^2 = N$. By looking for solutions of the form $\phi_a(r,t) = e^{-i \frac{\mu a}{\hbar} t} \phi_a(r)$ and $\phi_m(r,t) = e^{-i \frac{\mu m}{\hbar} t} \phi_m(r)$ we find the coupled time-independent GP equations,

$$\mu \phi_a = \left( -\frac{\hbar^2}{2m} \nabla^2 + V_a + \lambda_a |\phi_a|^2 + \lambda_{am} |\phi_m|^2 \right) \phi_a$$
\[ 2\mu \phi_m = \left( -\frac{\hbar^2}{4m} \nabla^2 + \varepsilon + V_m + \lambda_{am}|\phi_a|^2 + \lambda_m|\phi_m|^2 \right) \phi_m + \frac{\alpha}{\sqrt{2}} \phi_a. \]  

(4)

Now we turn our attention to the limit where the two-component theory reduces to an effective one-component theory. This limit is reached when the detuning is large enough that \( \varepsilon \) greatly exceeds the kinetic energy as well as any interaction energies, i.e., \( \varepsilon \gg \lambda_{an}n_m, \lambda_{am}n_a, \lambda_an_a \), where \( n_a \) and \( n_m \) are the atomic and molecular densities respectively, plus the additional hypothesis that the number of molecules are small compared to the number of atoms \( n_m/n_a << 1 \). These assumptions allow us to neglect the interaction terms and the chemical potential \( (\mu \approx \lambda_a n_a) \) in the Gross-Pitaevskii equation \( \phi \), leading to a simple relation between the molecular and atomic condensate

\[ \phi_m \approx -\frac{\alpha}{\sqrt{2\varepsilon}} \phi_a^2. \]  

(5)

The insertion of this expression into equation \( \phi \) yields an effective single condensate Gross-Pitaevskii equation for \( \phi_a \)

\[ \mu \phi_a = \left[ -\frac{\hbar^2}{2m} \nabla^2 + V_a + \lambda_{eff} \phi_a^2 \right] \phi_a \]  

(6)

with

\[ \lambda_{eff} = \lambda_a - \frac{\alpha^2}{\varepsilon}. \]  

(7)

Note that, from \( \phi \), the limit \( |\phi_m|/|\phi_a| << 1 \) implies \( \varepsilon >> \alpha \phi_a \). As \( \lambda_a \) is positive and we want to vary \( \varepsilon \) in a way that \( \lambda_{eff} \) is always positive, it follows from \( \lambda_{eff} \) that we must set \( \varepsilon > \alpha^2/\lambda_a \). Assuming that all the interaction strengths are of the same order of magnitude, the conditions of validity of the one-component theory can be summarized by the inequalities: \( \varepsilon >> \lambda_a n_a, \varepsilon >> \alpha \sqrt{n_a} \) with \( \varepsilon > \alpha^2/\lambda_a \). In what follows we will consider two regimes characterized by the parameter \( r \),

\[ r = \frac{\alpha}{\lambda_a \sqrt{n_a}}. \]  

(8)

which relates the characteristic energies of our system,

\[ \alpha^2/\lambda_a = r \alpha \sqrt{n_a}, \alpha \sqrt{n_a} = r \lambda_a n_a \]  

(9)

i) Strong resonance limit-When \( r > 1 \) the characteristics energies satisfy the set of inequalities \( \alpha^2/\lambda_a >> \alpha \sqrt{n_a} >> \lambda_a n_a \) and for the entire range of variation of the detuning, even up to \( \lambda_{eff} \approx 0 \), the conditions of validity of the effective one-component theory is satisfied. Therefore, when the parameter \( r \) satisfies the above inequality one can picture the system as being one condensate of atoms with an effective scattering length. We will show, in our calculations that with values of \( r \) in this range, the two component and the effective one component theory agree even when the effective scattering length is almost zero, the molecules are practically inexistent in this regime.

ii) Weak resonance limit- On the other hand when \( r \approx 1 \) it follows that all the characteristics energies are of the same order of magnitude \( \alpha^2/\lambda_a \approx \alpha \sqrt{n_a} \approx \lambda_a n_a \) and for \( \varepsilon \approx \alpha^2/\lambda_a \), the conditions of validity of the effective one-component theory is not satisfied. This is corroborated by our calculations where we show that, for detunings of the order of \( \alpha^2/\lambda_a \) that correspond in the single condensate effective theory, to almost zero effective scattering length, we obtain significantly different equilibrium configuration and also have unique modes to the hybrid system that are relatively low in energy \( (\omega_{exc} \sim \omega_{trap}) \).

The weak resonance regime proves to be an ideal place to experimentally detect a clear signature of molecule formation. The expression for the parameter \( r \) illustrates that there are three ways of approaching this regime: by increasing the condensate density, having systems with a higher asymptotic atom-atom scattering length and/or reducing the value of \( \alpha \).

Next we will show, as pointed out above, that the equilibrium spatial distribution of the atoms and the excitation energies of specific collective modes have different behaviors as a function of the detuning \( \varepsilon \) depending on the regimes discussed above and can be a signature of the presence of molecules in the system.

The standard approach to investigate the properties of a hybrid system would be to solve the equations \( \phi \), \( \phi_a \) and \( \phi_m \) to find the equilibrium shapes and to solve the linear approximation of its time dependent extension to find the collective excitations. However in this paper we use a simpler approach, the variational method. In this method we employ a Gaussian ansatz \( \phi_a(m) \), \( \phi_m(m) \) to parameterize the time dependence of the condensate wave function as

\[ \phi_a(m) = A_a(m) e^{iF_a(m)} \]  

(10)

where the amplitudes of the condensate wave functions are given by

\[ A_a = \sqrt{\frac{N_a}{\pi^{3/2}} \prod_{j=1,2,3} 1 \sqrt{q_j} e^{-\frac{(x_j-r_j)^2}{2q_j}}} \]  

(11)

\[ A_m = \sqrt{\frac{N_m}{\pi^{3/2}} \prod_{j=1,2,3} 1 \sqrt{Q_j} e^{-\frac{(x_j-R_j)^2}{2Q_j}}} \]  

(12)

and the phase as

\[ F_a = \theta_a + \sum_{j=1,2,3} \pi_j (x_j - r_j) + \frac{p_j}{2q_j} (x_j - r_j)^2 \]  

(13)

\[ F_m = \theta_m + \sum_{j=1,2,3} \Pi_j (x_j - R_j) + \frac{P_j}{2Q_j} (x_j - R_j)^2 \]  

(14)

The variational parameters \( q_j, (Q_j) \), \( r_j^c, (R_j^c) \) correspond respectively to the widths and translation of the atomic...
and molecular condensate clouds, which have an ellipsoidal shape \( p_j(P_j) \), \( \pi_j^f(\Pi_j^f) \) are the corresponding momenta. The \( N_{a(m)} \) are the number of particles of each component \( \theta_{a(m)} \) the corresponding phase.

When we replace the condensate wave function parametrized as in eqs. (10-14) into equation (2), \( \Gamma \) reduces to a classical action

\[
\Gamma = \int dt \left\{ \sum_{j=1,2,3} N_a \pi_j^f \dot{p}_j^f + \frac{N_a}{4} \left(p_j q_j - \dot{p}_j q_j\right) + \sum_{j=1,2,3} N_m \Pi_j^f \dot{P}_j^c + \frac{N_m}{4} \left(P_j \dot{Q}_j - \dot{P}_j Q_j\right) + \theta_a \dot{N}_a + \theta_m \dot{N}_m - E \right\}
\]

(15)

where the energy \( E \) has the form \( E = E_a + E_m + E_i + E_{FR} \), with \( E_{a(m)} \) being the usual Gaussian Gross-Pitaevskii energy for a single atomic(molecular) condensate \( E_i \) are the interaction energies between these two species and \( E_{FR} \) is the Feshbach component.

The variation of the action with respect to the twenty-eight parameters leads to Hamiltonian type equations of motion for the time evolution of these parameters. Denoting collectively the parameters by \( X, i = 1, \ldots, 28 \), the equations of motion can be written as

\[
\sum_i \Gamma_{kl}(X) \dot{X}_i = \frac{\partial E}{\partial X_k}(X),
\]

(16)

where \( \Gamma_{kl}(X) \) is the anti-symmetric matrix coming from the variation of the “kinematic” piece of the action (14) \( E(X) \) depends only on the phase difference, \( \theta = \theta_a - \theta_m / 2 \) and as a consequence the quantity \( N = N_a + 2 N_m \) is a constant of the motion. This reduces the number of degrees of freedom to thirteen which we take as the relative phase \( \theta = \theta_a - \theta_m / 2 \) and the relative population \( n = (N_a - 2 N_m)/2 \), besides the translational and shape degrees of freedom.

i) Equilibrium configuration

The equilibrium configuration is determined by the condition \( X_k = 0 \), where \( k = 1, \ldots, 26 \) which leads to

\[
\frac{\partial E}{\partial X_k}(X_0) = 0,
\]

(17)

where \( X_0 \) represents the equilibrium values of the parameters. Thirteen of the equations (17) lead immediately to \( p_j = 0(P_j = 0), \pi_j^c = 0(\Pi_j^c) = 0 \) and \( \theta = \pi/2 \) which is the phase of the minimum configuration. Six of the remaining thirteen equations refers to the equilibrium displacement of the condensate clouds. These equations have the trivial solution \( r_j^c = 0(\Pi_j^c = 0), j = 1, 2, 3 \), which is the one considered in this paper. Therefore we are left with seven equations to find the equilibrium widths of the condensate clouds and of the relative population \( n \), \( n = (N_a - 2 N_m)/2 \).

We proceed by presenting the results of the static calculations obtained by solving (17) in the weak and strong resonance limits respectively. In the strong resonance limit we used \( r \approx 10 \). We computed the equilibrium radial and axial widths \( (q_x, q_y, q_z) \) for the atomic condensate for different values of the detuning. We varied the detuning from \( 2\pi \times 105 \text{ MHz} \) to \( 2\pi \times 22 \text{ MHz} \) corresponding, in the effective theory, to a \( \lambda_e \) varying from 80% to 5% of the asymptotic interaction strength, respectively. The results are shown on Table I.

One can see no difference between the two component molecular theory and the single condensate effective theory. This shows that all attempts, through an adiabatic change of the detuning, to detect molecular signatures in this regime will fail, because for all practical purposes the number of molecules formed is quite insignificant. To achieve the weak resonant limit we used \( r \approx 1 \). The detuning values were changed from \( 2\pi \times 41.7 \text{ KHz} \) to \( 2\pi \times 8.7 \text{ KHz} \) corresponding to the same effective interaction strength as in the strong resonance case (from 80% to 5% of the asymptotic interaction strength). The results, shown on Table II, indicate a very different behavior between the molecular and the single condensate effective theories, when the detuning approaches \( \alpha^2 / \lambda_a \).

In the two component theory, the size of the atomic condensate will not change significantly when we vary the detuning, due to the presence of molecules in contrast with a drastic change in the single condensate effective picture. We conclude that this regime proofs to be ideal for probing molecular signatures provided the detuning is adiabatically changed. The weak resonance limit results are displayed in Fig.1.
ii) Excitations

The excitations are associated with the normal modes of the linear approximation of the equations of motion \( \hat{H} \). In this limit \( \hat{H} \) reduces to

\[
\sum_l \Gamma_{kl}(X_0) \dot{\eta}_l = \sum_l \mathcal{H}_{kl}(X_0) \eta_l .
\]

(18)

where \( \mathcal{H}_{kl}(X_0) \) is the Hessian matrix at equilibrium \( \mathcal{H}_{kl} = \frac{\partial^2}{\partial X_k \partial X_l} \mathcal{H}(X^0) \) and \( \eta_l \) are the displacement from the equilibrium value \( X_l = X_{0I} + \eta_l \). The normal modes correspond to oscillatory solutions of (18), and the eigenfrequencies are given by the characteristic equation \( \det(i\Gamma \Omega - \mathcal{H}) = 0 \). The roots of the characteristic equation are real and come out in pairs \( \Omega, -\Omega \), each pair associated to one normal mode.

In the single-condensate case we have six normal modes which are related to the translations of the condensate clouds and to shape oscillations \( \mathcal{H}_{kl} \). For the later, two of these correspond to axially symmetric oscillations where the axial and radial oscillation are in phase (mode c) and the other where they are out of phase (mode b), the third one being non-axially symmetric with the oscillations in \( x \) and \( y \) directions out of phase and the axial direction at its equilibrium value (mode a).

In the two component case the normal modes will be linear combinations of the modes described above for each component plus one mode related to the dynamics of population exchange between atoms and molecules, giving a total of thirteen normal modes. In what follows we will investigate how the eigenfrequencies depend on the Feshbach resonance parameter \( \alpha \) and the detuning \( \varepsilon \).

In our calculation we obtain the following general results, independent of the value of \( r \) : a) The six translational degrees of freedom are decoupled from the shape and number degrees of freedom, originating three atomic and molecular translations in phase (center of mass modes) and three out of phase (dipole modes). b) The remaining seven modes can be divided into two groups. The first group, where the shape and number oscillations are decoupled, consists of two modes which correspond to an in phase and an out of phase atomic and molecular oscillations where both components are in mode a. The second group consists of coupled shape and number oscillations with each component in modes b and/or c.

(2) In the single component theory we have six modes, three translational and three shape oscillations. The three translational modes correspond to the three center of mass modes of the hybrid system and the three shape oscillations correspond to the three lowest energy modes not counting the center of mass modes.

For the strong resonance limit, \( r \approx 10 \), the three lowest energy modes, disrecred the translational ones, for the hybrid system correspond to in phase atomic and molecular shape oscillations \( (b+b) \), \( (a+a) \) and \( (c+c) \) respectively, and have the same behavior in both theories as exemplified on Table I for the mode \( (a+a) \).

In the weak resonance limit \( r \approx 1 \) of the three modes only the mode \( (a+a) \) behaves differently depending on which theory is used. In the single component theory the a mode approaches abruptly to the c mode when \( \lambda_{eff} \rightarrow 0 \), because of the degenerescence present in the ideal gas limit. On the other hand for the hybrid theory this abrupt change is not present. The absence of a near degenerescence is a signature of the presence of the molecules. The values of this frequency are shown on Table II and two of the three lowest energy modes, for both single and hybrid theories are displayed in Fig.2.

Another possible signature of a molecular condensate is the presence of the dipole frequency in the two component theory. The frequency of the center of mass oscillation motion of the hybrid BEC is entirely determined by the frequencies of the trap and does not depend on the interatomic interaction \( \varepsilon \). On the other hand the frequencies of the dipole oscillation of the hybrid system depend on the Feshbach resonance parameter \( \alpha \), and on the detuning parameter \( \varepsilon \). Since this mode is unique to the hybrid system its presence is a clear signature of the presence of molecules. This being an interesting phenomenon to be explored experimentally. Of the three dipole modes the radial ones are degenerate and have a higher frequency than the axial one. Using the same parameters for the weak limit we plot in Fig.3 the frequency of this mode as a function of the detuning. From this results we see that the frequency of this mode, unique to the hybrid system, decreases when the detuning approaches \( \alpha^2/\lambda_{eff} \) reaching observable values.

In conclusion, we have shown, that provided we work in the weak resonance limit, the hybrid atom molecule BEC system behaves qualitatively differently from a single condensate with an effective scattering length, exhibiting different behaviours for the equilibrium shapes.
FIG. 3: Plot showing the dipole frequency, in units of the radial trap frequency, as a function of the detuning for the hybrid condensate. The lower curve corresponds to an axial oscillation and the upper curve radial oscillations. The parameters are the same as in figure 1. See text for details.

TABLE I: Comparison between the single condensate theory and the hybrid theory in the strong resonant limit. The Feshbach resonance parameter in this case is \( \alpha = 5 \times 10^4 \mu m^3/2Hz \) and the other parameters are the same as in figure 1.

| \( \varepsilon (2\pi \times MHz) \) | \( \lambda/\lambda_{eff} \) | \( q_x(\mu m) \) | \( q_z(\mu m) \) | \( \omega_a/\omega_r \) | \( q_x(\mu m) \) | \( q_z(\mu m) \) | \( \omega_a/\omega_r \) |
|---|---|---|---|---|---|---|---|
| 105 | 0.8 | 1.7 | 23.9 | 1.5 | 1.7 | 23.9 | 1.5 |
| 30.0 | 0.3 | 1.4 | 19.3 | 1.5 | 1.4 | 19.3 | 1.5 |
| 26.0 | 0.2 | 1.3 | 17.5 | 1.6 | 1.3 | 17.5 | 1.6 |
| 22.0 | 0.1 | 1.1 | 12.6 | 1.7 | 1.2 | 12.8 | 1.7 |

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TABLE II: Comparison between the single condensate effective theory and the hybrid molecular theory in the weak resonant limit.

| $\varepsilon$ (2πKHz) | $\lambda/\lambda_{eff}$ | $q_z (\mu m)$ | $q_z (\mu m)$ | $\omega_2/\omega_r$ | $q_r (\mu m)$ | $q_r (\mu m)$ | $\omega_a/\omega_r$ |
|------------------------|-----------------------|---------------|---------------|-------------------|---------------|---------------|-------------------|
| 41.7                   | 0.8                   | 1.7           | 23.9          | 1.5               | 1.7           | 23.8          | 1.5               |
| 11.9                   | 0.3                   | 1.4           | 19.3          | 1.5               | 1.6           | 22.0          | 1.5               |
| 10.4                   | 0.2                   | 1.3           | 17.5          | 1.6               | 1.6           | 21.8          | 1.5               |
| 8.7                    | 0.1                   | 1.1           | 12.6          | 1.7               | 1.5           | 21.6          | 1.5               |

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