Atomic–Molecular Condensates with Large Positive Scattering Length

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We show that in the limit of large and positive atom–atom scattering length the properties of an atomic–molecular Bose–Einstein Condensate (amBEC) are determined by an universal energy density functional (EDF). We find that the optimal conditions for the formation of a stable amBEC are in the regime where there are no shallow trimers and the atom–dimer scattering length is negative and comparable in magnitude with the atom–atom scattering length. At temperatures lower than $T_e$, the chemical potentials for the atoms and molecules can be specified independently. Besides three–body recombinations processes into dimers of large size, inelastic processes involving the formation of deep bound small size molecular states are possible. These inelastic processes do not lead to an efficient heating of the amBEC and can be used for its mostly non–destructive monitoring.

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The theoretical prediction \cite{1}, along with the experimental confirmation \cite{2} of the fact that one can manipulate relatively easily the interaction of slow atoms by means of external fields, opened the doors to a long series of possible experiments. Perhaps among the most spectacular experimental results obtained lately are the creation of the so–called Bosenova \cite{3} and the evidence that related to a mixed atomic–molecular Bose–Einstein Condensate (BEC) was created \cite{4}, an object envisioned by theory several years before \cite{5–7}, see also Refs. \cite{8,9} for a couple of theoretical analyses of this experiment. In the present work we identify a new universal regime, which appears in the case when the two–body scattering length $a$ is positive ($a > 0$) and large, $a \gg r_0$, where $r_0$ is the effective range, appearing the the low energy parameterization of the s–wave phase shift $k \cot \delta(k) = -1/(a + r_0 k^2/2 + \ldots$ Typically $r_0$ is of the order of the so called van der Waals length $(C_6 m/\hbar^2)^{1/4}$ \cite{10}. The universality of this new regime is encoded in only two parameters, which define the effective Hamiltonian (except for the mass), one of them the two–body scattering length $a$ and the other a three–body characteristic, which can be chosen in various ways. As we will show below, in this regime, when $na^3 \ll 1$, the properties of a cold mixture of atoms and shallow dimers are described by a very simple theoretical model. In the other limit, when $a < 0$ and $|a| \gg r_0$, there is another universal regime and a new class of dilute quantum liquids exists \cite{11}. The basic ingredients of the model are derived from the well known Efimov effect \cite{12,13}, which pertains to the universal properties of the three–body systems with large two–body scattering lengths.

In order to derive the energy density of an atomic–molecular gas let us first consider the dilute regime where $nr_0^3 \ll 1$, with $n$ a typical atomic or molecular density and $a = O(r_0)$. The details of the potential are not important and the atom–atom interactions can be described by a contact term

$$H_a = -\psi_a^\dagger \frac{\hbar^2 \nabla^2}{2m} \psi_a + \frac{1}{2} \lambda_2 \psi_a^\dagger \psi_a^\dagger \psi_a^\dagger \psi_a + \frac{1}{3} \lambda_3 \psi_a^\dagger \psi_a^\dagger \psi_a^\dagger \psi_a,$$  \hspace{1cm} (1)

where $\psi_a^\dagger, \psi_a$ are creation and annihilation operators for atoms. The value of $\lambda_2$ is determined (after fixing a short distance regulator) by the scattering length $a$. $\lambda_3$ depends also on a genuinely three–body length scale, denoted below by $a_3$, that can be determined only through the value of a three–body observable. Terms with more derivatives or higher body forces can be included, but their effect on a dilute gas is small as long as the relevant momenta satisfy the condition $kr_0 \ll 1$ [14–17]. The Hamiltonian in Eq. (1) contains, in principle, all the information necessary to describe the system in the dilute regime, including the molecular states/dimers with binding energy $\hbar^2/ma^2$ and radius $\approx a$ when $a > 0$. In the case $a \gg r_0$ we are interested in, the presence of bound states within the regime of validity of Eq. (1) evidences the fact that perturbation theory breaks down. Since particle–particle diagrams should be summed up to all orders, a simple mean–field approximation is not legitimate as well and two–particle correlations have to be accounted for explicitly. Thus, even though $H_a$ contains all the information needed to describe molecular states, it is inconvenient to use it directly \cite{16}.

Let us consider now a regime even more dilute, where $na^3 \ll 1$. The relevant typical momenta are of the order $p = \hbar k \approx \sqrt{na^3 \hbar/a} \ll \hbar/a \ll \hbar/r_0$ \cite{14}. We will argue later that transitions altering the number of molecular states are “very slow” in this regime and, consequently, for time scales shorter than the transition rate, the number of atoms and molecules are separately conserved. An appropriate effective Hamiltonian describing the system in this regime is

$$H_{am} = -\psi_m^\dagger \frac{\hbar^2 \nabla^2}{2m} \psi_m - \psi_m^\dagger (\frac{\hbar^2 \nabla^2}{4m} - \varepsilon_2) \psi_m$$ \hspace{1cm} (2)

$$+ \frac{1}{2} \lambda_{aa} \psi_a^\dagger \psi_a^\dagger \psi_a^\dagger \psi_a + \lambda_{am} \psi_a^\dagger \psi_m \psi_a^\dagger \psi_m + \lambda_{mm} \psi_m^\dagger \psi_m^\dagger \psi_m.$$  

Both $H_a$ and $H_{am}$ are applicable in the regime $ka \ll kr_0 \ll 1$. $H_{am}$ has a great advantage over $H_a$ how-
ever. Perturbation or mean–field theory is not valid for $H_a$ when $p \sim \hbar/a$, but it is valid for $H_{am}$ since all non–perturbative physics occurring at the scale $\sim a$ and leading to the formation of the bound state (dimers of size $\sim a$) and to the Efimov effect (trimers of size $\sim a$) is already neatly encapsulated in the constants $\lambda_{aa}, \lambda_{am}$ and $\lambda_{mm}$. To make that more explicit, let us determine $\lambda_{aa}, \lambda_{am}$ and $\lambda_{mm}$ in terms of $\lambda_2$ and $\lambda_3$ by considering zero momentum scattering of two atoms, atom–molecule and two molecules. Using $H_a$ these processes are described by an infinite number of diagrams shown in Fig. 1, corresponding to the solution of the Lippmann–Schwinger, Fadeev and Yakubovski equations respectively [12,13,16,18]. The explicit solutions of the Lippmann–Schwinger and Fadeev equations are known. If we use dimensional regularization [19], where divergences proportional to powers of the momentum cutoff vanish, the same processes are described using $H_{am}$ simply by the Born amplitude, see the Fig. 1. This leads to the identification

$$\lambda_{aa} = \frac{4\pi^2 a}{m}, \quad \varepsilon_2 = -\frac{\hbar^2}{ma^2},$$

$$\lambda_{am} = \frac{3\pi^2 a}{m} - \frac{3\pi^2 a^3}{m} \left[ c_1 + c_2 \cot \left( s_0 \ln \frac{a}{a'_0} \right) \right],$$

$$\lambda_{mm} = \frac{2\pi^2 a^3}{m} = 4\pi^2 a^3 c_3,$$

where the values of the universal constants $s_0 \approx 1.00624$, $c_1 \approx 1.46$ and $c_2 \approx 2.15$ are known [20] and $|\varepsilon_2|$ is the true dimer binding energy. $a'_0 > 0$ is the value of the two–body scattering length for which a trimer is formed. Notice that the matching described above is that in most cases the dimer–dimer scattering length is $0$ regime only the atom–dimer scattering processes are described by an infinite number of diagrams built using Eq. (1) this expression corresponds to the exact summation of the linear and quadratic terms in densities of all the two–particle, three–particle and four–particle diagrams, computed with the initial Hamiltonian $H_a$. Using well–known arguments [10,15], it is straightforward to show that higher order corrections, due to mean–field fluctuations, are controlled by the diluteness parameter $\sqrt{na^3} \ll 1$. This EDF expression is remarkable, since, in the Efimov regime when $a \gg r_0$, it is universal. Only the exact value of the parameter $c_3$ for bosons is not explicitly known. Notice, however, that the condition $na^3 \ll 1$ excludes its use arbitrarily close to a Feshbach resonance. By changing the units of energy to $\hbar^2/ma^2$ and length to $a$ this EDF acquires the following dimensionless and thus universal form:

$$\mathcal{E} = 2\pi n_a^2 + \frac{3\pi}{2} n_a n_m + 2\pi c_3 n_m^2 - n_m,$$

with $\xi = c_1 + c_2 \cot \left( s_0 \ln (a/a'_0) \right)$ tunable.

In the $a > 0$ regime only the atom–dimer scattering length can change dramatically from negative to positive values and even become infinite. If $\lambda_{aa} > 0$, $\lambda_{mm} > 0$ and $\lambda_{aa} \lambda_{mm} - \lambda_{am}^2 > 0$, then a mixed dilute atom–molecule system is a metastable gas. This can be seen as well by computing the pressure

$$P = \frac{1}{2} \lambda_{aa} n_a^2 + \lambda_{am} n_a n_m + \frac{1}{2} \lambda_{mm} n_m^2.$$
Phase separation occurs when $\lambda > \sqrt{\lambda_{aa}\lambda_{mm}}$ and domain wall structures are expected to appear [23]. A (meta)stable trimer phase might also form if the (yet unknown) trimer–trimer interaction is repulsive. Another type of domain walls can appear in the opposite limit [24]. It is easy to see that there is no region where $P = 0$ and thus no stable liquid phase exists, unlike Refs. [6].

An amBEC system would be in chemical equilibrium if

$$
(2\lambda_{aa} - \lambda_{am})n_a + (2\lambda_{am} - \lambda_{mm})n_m = \varepsilon_2. \quad (9)
$$

This type of chemical equilibrium can be established through the reaction $A + A + A \rightarrow A + A + A$, which has a low rate for dilute systems $\propto h\alpha^2 n_a^2/m$, see Refs. [25] and below. At temperatures $T < T_c \propto (n\alpha^3)^{2/3} h^2/ma^2 \ll |\varepsilon_2| = h^2/ma^2$, the rhs of the above chemical equilibrium condition is parametrically much larger than the lhs when $n\alpha^3 \ll 1$. The reverse reaction $A_2 + A \rightarrow A + A + A$ is therefore exponentially suppressed, as it requires an activation energy larger then the thermal energy. Consequently, these type of collisions thus lead to a very small heating (and very slow chemical equilibration of the system) and its ultimate depletion [25]. Since these two reactions (and $A_2 + A_2 \leftrightarrow A_2 + A + A$, etc. as well) are very slow in an amBEC, the chemical potentials for atoms and dimers could be specified independently

$$
\mu_a = \lambda_{aa}n_a + \lambda_{am}n_m, \quad \mu_m = \lambda_{am}n_a + \lambda_{mm}n_m + \varepsilon_2.
$$

In a trap one has to add the trap potentials for atoms and dimers in the rhs of these two relations [10].

A number of other processes can occur in such a system. It is desirable to consider three-body inelastic collisions involving one or more dimers in the initial state. Particularly interesting are however the inelastic atom–dimer $A + A_2 \rightarrow A^* + A_2^*$ (see Ref. [26] for a rate calculation) and dimer–dimer $A_2 + A_2 \rightarrow A^* + A^* + A_2^*$ collisions, where $A^*$ and $A_2^*$ stand for a fast atom (with its kinetic energy significantly exceeding the thermal energy $3T/2$) and $A_2^*$ for a fast dimer respectively (with a binding energy of the order $\hbar^2/mr_0^2 \gg \hbar^2/ma^2$). If the temperature of the system is significantly lower then the dimer binding energy, i.e. $T < T_c \ll \hbar^2/ma^2$, these inelastic collisions are not leading to a significant heating of the system, but mostly to its depletion, in a manner similar to the depletion in the BEC of metastable helium [27]. If an atom and a dimer collide and this leads to the formation of a dimer with a binding energy of order $\hbar^2/mr_0^2 \gg \hbar^2/ma^2$ a relative large amount of energy is released (NB $T < T_c \ll \hbar^2/ma^2 \ll \hbar^2/mr_0^2$). It is conceivable that transitions to various final bound states could be resolved and thus the spectrum of fast ejectiles would be discrete. The momenta of the outgoing atom and of the deeply bound dimer are large as well, of the order $\hbar/r_0 \gg \hbar/a$, and such fast atoms and dimers will interact relatively weakly with the rest of the atoms and dimers. While the slow/thermal atoms and shallow dimers interact with each other with cross sections of the order of $a^2$, a fast atom and a deeply bound fast dimer will interact with the amBEC with a cross section of order $r_0^2 \ll a^2$. One can expect a similar behavior in a dimer–dimer inelastic reaction. The fact that the products of these inelastic processes are fast and weakly interacting objects can be used for a non-destructive monitoring of the amBEC. The rates of these inelastic processes are obviously controlled by the parameter $r_0^3/a^3 \ll 1$, since in order to form a deeply bound dimer two atoms in the shallow dimer have to change their separation from distances of order $a$ to distances of order $r_0$. One cannot help here but to make a parallel with the similar role played by neutrinos (which are also fast and weakly interacting products of inelastic processes) in studying the interior of the Sun.

A possible final channel of a dimer–dimer collision could be a trimer–atom final state if $a_{am} > 0$. If the trimer energy is close to the atom–dimer threshold, then the atom–dimer cross section could be significantly larger than the atom–atom and dimer–dimer cross sections ($|a_{am}| \gg a, a_{mm}$). If this is the case, one can expect that the atom–dimer elastic collisions would be chiefly responsible for the thermalization of the system. However, besides the elastic channel there is also an inelastic channel present if $a_{am} > 0$: a dimer–dimer collision leading to a shallow trimer and an atom in the final state. This process will lead to a relatively small release of energy since the trimer energy is very close to the threshold, see Fig. 2. With good accuracy the ground state energy of a trimer is given in this limit by $\varepsilon_3 \approx \varepsilon_2 - 3\hbar^2/4ma_{am}^2$. [12] Moreover, the trimer under these conditions is basically a very large two-body object of size $a_{am} \gg a$, made of a dimer and a relatively loosely bound atom to
it. In this limit the atoms and dimers will phase separate, since $\lambda_{am} \gg \sqrt{\lambda_{aa} \lambda_{mm}}$. A pure trimer phase could appear instead, iff the trimer–trimer interaction is repulsive. When $a_{am} > 0$ and $\lambda_{am} \leq \sqrt{\lambda_{aa} \lambda_{mm}}$ dimer–dimer collisions with a trimer in the final state will lead to an energy release of the order of $\hbar^2/ma^2$, and thus to the heating of the system, since the momenta of the emerging products are of the order $\hbar/a$ in this case. Consequently, the best conditions to create a stable amBEc are expected when $a_{am} < 0$ (no shallow bound trimers and thus no $A_2 + A_2 \rightarrow A_3 + A$ reactions) and $|a_{am}| < a$. As a last note, there is essentially no overlap between present results and those of Refs. [28].

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\[ E \propto a n(a^2)^{3/4}/\hbar^2ma^2 \]