Ultradilute quantum liquid drops

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Using quantum Monte Carlo methods we have studied dilute Bose-Bose mixtures with attractive interspecies interaction in the limit of zero temperature. The calculations are exact within some statistical noise and thus go beyond previous perturbative estimations. By tuning the intensity of the attraction, we observe the evolution of an N-particle system from a gas to a self-bound liquid drop. This observation agrees with recent experimental findings and allows for the study of an ultradilute liquid never observed before in Nature.

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The high tunability of interactions in ultracold Bose and Fermi gases is allowing for exploration of regimes and phases difficult to find in other condensed-matter systems [1]. By adjusting properly the applied magnetic field, Bose and Fermi gases are driven to Feshbach resonances with an increase of interaction practically at will, and with the possibility of turning the system from repulsive to attractive and vice versa. This is obviously not possible in conventional condensed matter where interactions are generally not tunable at this level. A significant example of this versatility has been the clean experimental realization of the unitary limit for fermions [2, 3] and the precise characterization of the BCS-BEC crossover [4, 5], which up to that moment, was only a theoretical scenario.

Recently, it has been possible to explore the formation of liquid/solid patterns in dilute gases by modifying the strength of the short-range interatomic interactions. Probably, the most dramatic example of this progress has been the observation of the Rosensweig instability in a confined system of $^{164}$Dy atoms with a significant magnetic dipolar moment [6]. By tuning the short-range interaction, Kadau et al. [6] have observed the spontaneous formation of an array of self-bound droplets remembering the characteristics of a classical ferrofluid. The observation of solid-like arrangements in dilute gases has also been possible working with highly-excited Rydberg atoms [7]. By direct imaging, Schauss et al. [7] have obtained ordered excitation patterns with a geometry close to the well known arrangements observed in few-body confined Coulomb particles.

In the line of obtaining other dense systems starting from extremely dilute Bose and Fermi gases, it is noticeable the mechanism suggested by Petrov relying on Bose-Bose mixtures [3]. According to this proposal, it is possible to stabilize a mixture with attractive interspecies interaction in such a way that the resulting system is self-bound, i.e., a liquid. Whereas a mean-field treatment of the mixture predicts a collapsed state, the first beyond mean field correction, the Lee-Huang-Yang (LHY) term, is able to stabilize the system by properly selecting the interspecies s-wave scattering length. Further work has shown that reducing the dimensionality of the setup to two or quasi-two dimensions may help to stabilize the liquid phase [9]. The LHY correction has also been used to account for the formation of dipolar drops [9] and then confirmed by full first-principles quantum Monte Carlo (QMC) simulations [11, 12].

The exciting idea of producing self-bound liquid drops by using interspecies attractive interaction acting as glue of the entire Bose-Bose mixture has been put forward by Tarruell and collaborators [13]. Results obtained with a mixture of $^{40}$K atoms in different hyperfine states have shown the formation of these drops, that do not release for a significant time when the confining trap is removed. Therefore, the theoretical prediction seems confirmed and thus a new window for exploring matter in unprecedented situations is open. On one side, it proves the way of forming liquid drops with large density in the world of cold gases and, on the other, makes possible the study of a liquid state of matter with an extremely low density, lower than any other existing liquid.

In the present work, we study the formation of liquid drops in a Bose-Bose mixture using the diffusion Monte Carlo (DMC) method, which solves stochastically the N-body Schrödinger equation in an exact way within some statistical uncertainties. The DMC method was extensively used in the past for determining the structure and energy properties of liquid drops of $^4$He [14, 15], $^3$He [16, 17], H$_2$ [18], and spin-polarized tritium [19]. At difference with previous perturbative estimates, DMC allows for an exact study of the quantum properties of the system relying only on its Hamiltonian. Our results confirm the LHY prediction on the stability of self-bound mixtures and determine quantitatively the conditions under which liquid drops are stable and how they evolve when the attractive interaction is increased. Within the regime here explored, we do not observe a full collapse of the drop but an increase of the density and reduction of the size, which is rather progressive.
The Bose-Bose mixture under study is composed of $N_1$ bosons of mass $m_1$ and $N_2$ bosons of mass $m_2$ with Hamiltonian

$$H = -\frac{\hbar^2}{2m_1} \sum_{i=1}^{N_1} \nabla_i^2 - \frac{\hbar^2}{2m_2} \sum_{j=1}^{N_2} \nabla_j^2 + \frac{1}{2} \sum_{\alpha,\beta=1}^{2} \sum_{i,j=1}^{N_1 N_2} V^{(\alpha,\beta)}(r_{i,j}),$$

(1)

with $V^{(\alpha,\beta)}(r)$ the interatomic interaction between species $\alpha$ and $\beta$. Our interest is focused on a mixture of intraspecies repulsive interaction, i.e., positive $s$-wave scattering lengths $a_{11} > 0$ and $a_{22} > 0$, and interspecies attractive potential, $a_{12} < 0$. To set up this regime, we use a hard-sphere potential of radius $a_{\alpha\alpha}$ for potentials $V^{(\alpha,\alpha)}(r)$ and an attractive square well of depth $-V_0$ and range $R$ for $V^{(\alpha,\beta)}(r)$. In the latter case, we fix $R$ and change $V_0$ to reproduce the desired negative scattering length; notice that we work with negative $a_{\alpha\beta}$ values and thus the attractive potential does not support a pair bound state.

The DMC method uses a guiding wave function as importance sampling to reduce the variance to a manageable level. We adopt a Jastrow wave function in the form

$$\Psi(R) = \prod_{1 \leq i < j}^{N_1} f^{(1,1)}(r_{ij}) \prod_{1 \leq i < j}^{N_2} f^{(2,2)}(r_{ij}) \prod_{i,j=1}^{N_1 N_2} f^{(1,2)}(r_{ij}),$$

(2)

with $R = \{r_1, \ldots, r_N\}$, $N = N_1 + N_2$. In the case of equal particles the Jastrow factor is taken as the scattering solution, $f^{(\alpha,\alpha)}(r) = 1 - a_{\alpha\alpha}/r$ for $r \geq a_{\alpha\alpha}$ and zero otherwise. If the pair is composed of different particles then we take $f^{(1,2)}(r) = \exp(-r/r_0)$, with $r_0$ a variational parameter.

In order to reduce the number of variables of the problem, keeping the essentials, we consider $m_1 = m_2$, $N_1 = N_2$, and $a_{11} = a_{22}$. In this way, our study explores the stability and formation of liquid drops as a function of $a_{12}$ and the number of particles $N$ ($N_1 = N_2 = N/2$).

The $s$-wave scattering length $a_{12}$ of an attractive well is analytically known, $a_{12} = R [1 - \tan(KR)/(KR)]$, with $K^2 = m_1 V_0/\hbar^2$. We take $a_{12} < 0$, which correspond to $KR < \pi/2$. In practice, we fix the range of the well $R$ and vary the depth $V_0$. As it is obvious from the equation for $a_{12}$, its value depends on the product $RV_{0}^{1/2}$ and thus increasing $R$ means to increase $V_0$. If for a fixed $a_{12}$ value we want to approach the limit $R \to 0$ then $V_0 \to \infty$, situation that makes our calculations extremely demanding in terms of accuracy and number of particles required to observe saturation. After some preliminary studies, we determined that $R = 4a_{11}$ is a good compromise between accuracy and reliability and thus the major part of our results are obtained with that. In the following, unless stated otherwise, all energies and lengths are given in $\hbar^2/(2m_1 a_{11}^2)$ and $a_{11}$ units, respectively.

The trial wave function $\Psi(R)$ depends on a single parameter $r_0$. This parameter is previously optimized using the variational Monte Carlo method. Its value increases with the total number of particles $N$; for instance, when $R = 4$ and $V_0 = 0.166$, $r_0$ increases monotonously from 106 up to 622 when $N$ grows from 100 to 2000. Our DMC algorithm is accurate up to second order in the imaginary-time step and uses forward walking to remove any bias of the trial wave function in the estimation of diagonal operators which do not commute with the Hamiltonian. Any systematic bias derived from the use of a finite time step and a finite number of walkers in the diffusion process is kept smaller than the statistical noise.

In Fig. 1 we report results for the energy per particle of the Bose-Bose mixture for different number of particles and as a function of the scattering length $a_{12}$. For each $N$, we observe a similar behavior when we tune $a_{12}$. There is a critical value which separates systems with positive and negative energies. When the energy is positive the $N$ system is in a gas phase and, by increasing $|a_{12}|$, it condenses into a self-bound system, that is, a liquid drop. Around the critical value the energy decreases linearly, the absolute value of the binding energy becomes...
scattering lengths \(a\) for binding on the number of particles: smaller drops show a clear dependence of the critical scattering length \(a\) on the number of particles \(N\). In Fig. 3, we report DMC results on the density profiles of the obtained drops. Notice that there is no self binding, \(a_{12}\) for unsaturated drops and then it stabilizes when saturation is reached. Our results show also this trend: for \(a_{12} = -3.09\), \(W = 15\) for the smallest drop and stabilizes then to \(W \approx 20\); for \(a_{12} = -3.81\), these values are \(W = 11\) and 18.

DMC allows for the study of the drops around the gas-liquid transition but can also show how the evolution towards a collapsed state happens. By increasing the depth of the attractive well \(V_0\) we can see the change in shape and size of a given drop. In Fig. 4 we report this evolution as a contour plot of the density profiles for a particular liquid drop with \(N = 200\) particles. The range of \(a_{12}\) values starts close to \(a_{12}^{\text{crit}}\), for this \(N\) value, and ends quite deep into the Feshbach resonance at a scattering length \(a_{12} \approx 40 a_{12}^{\text{crit}}\). Following this ramp, we observe an increase of an order of magnitude in the inner density and a shrinking of the size, with a reduction of the radius in a factor of three. Therefore, the drop becomes more dense but it is still a fully stable object which is not at all collapsed.

The microscopic characterization of the Bose-Bose liquid drops is not complete without the knowledge of the energy. As we commented before, it is the result of the energy which determines if an \(N\)-particle system is in a gas or liquid state. Once in the liquid phase, it is important to calculate the dependence of the energy on the number of particles. In Fig. 5 we report the DMC energies as a function of \(N\) and for three different \(a_{12}\) values. From intensive calculations carried out in the past on liquid \(^4\)He drops [14, 15], we know that the energy of the drops is well accounted for by a liquid-drop model. According to this, the energy per particle is

\[
E(N)/N = E_v + E_s x + E_c x^2 ,
\]

with \(x = N^{-1/3}\). The coefficients in Eq. (3), \(E_v\), \(E_s\), and \(E_c\) are termed volume, surface, and curvature energies, respectively. The term \(E_v\) corresponds to the energy of an infinitely large drop or, in other words, to the energy per particle of the bulk. The second term \(E_s\) is important because, from it, we can estimate the surface tension of the liquid \(t\) as

\[
t = \frac{E_s}{4\pi r_0^2}.
\]

The parameter \(r_0\) is the unit radius of the liquid, and can be estimated from the relation \(4\pi r_0^3 \rho_0 / 3 = 1\), with \(\rho_0\) the equilibrium density of the liquid.
It increases faster. From relation (4) and the critical value for self binding but, for larger potential values of the energy grows linearly with $V$.

In the three cases studied we obtain a high-fidelity fit. In the Figure we plot as lines the results of the liquid-drop model obtained as least-squares fit to the DMC energies. The errorbars are smaller than the size of the open symbols. Different sets correspond to different values of the interparticle scattering length $a_{12}$. The cross points at zero $x$-axis correspond to bulk calculations.

In Fig. 5, we plot as lines the results of the liquid-drop model obtained as least-squares fit to the DMC energies. In the three cases studied we obtain a high-fidelity fit. The open symbols are the DMC results and the lines are fits according to the liquid drop model. The errorbars are smaller than the size of the symbols. Different sets correspond to different values of the interparticle scattering length $a_{12}$. The cross points at zero $x$-axis correspond to bulk calculations.

We think that a comparison between the Bose-Bose drops here studied and the well-known properties of stable superfluid $^4$He drops can help to better visualize their extraordinary properties. We can consider a typical value for $a_{11}$ used in the experiments with ultracold mixtures of $^{39}$K, say $a_{11} = 50 a_0$, with $a_0$ the Bohr radius. Then, the saturation densities of the drops shown in Fig. 3 are $\sim 1.0 \cdot 10^{-6}$ and $1.4 \cdot 10^{-6}$ Å$^{-3}$. The saturation density of liquid $^4$He is $2.2 \cdot 10^{-2}$ Å$^{-3}$ implying that the Bose-Bose drops can be as dilute as $\sim 10^4$ times the $^4$He ones (a similar ratio happens when compared with water, with density $3.3 \cdot 10^{-2}$ Å$^{-3}$). For the same number of atoms, the Bose-Bose drop is much larger than the $^3$He one: $9.8 \cdot 10^{-2}$ μm for $V_0 = 0.150$ and $3 \cdot 10^{-3}$ μm for $^4$He with $N = 2000$. The surface of the dilute drop for this $N$ is $\sim 50\%$ of the total size, much larger than the $20\%$ value in $^4$He.

Summarizing, we have carried out a DMC calculation of Bose-Bose mixtures with attractive interspecies interaction. Relying only on the Hamiltonian, we describe the system without further approximations. As announced by Petrov using LHY approximate theory, it is possible to get self-bound systems by a proper selection of the interactions between equal and different species. The versatility of ultracold gases to change its interaction in magnitude and sign opens the possibility to explore new and very exciting physics. Our results clearly show the transition from a gas, with positive energy, to a self-bound system (liquid) and determine accurately the critical scattering lengths for the transition as a function of the number of particles. In the range of parameters here studied, we do not observe universality in the sense that the results depend only on the $s$-wave scattering lengths. For a same $a_{12}$ value we observe dependence on the range of the potential $R$. Preliminary calculations of the bulk phase seem to confirm this result, even carrying $R \rightarrow 0$.

Future studies can try to see if the inclusion of the effective range to better fit the model potential can help to devise a more precise description.

The experimental realization of Bose-Bose liquid drops opens the possibility of accessing denser systems than the usual trapped ultracold gases where quantum correlations can be much more relevant. The point of view from liquid state is however different: the liquid
that emerges from these mixtures is ultradilute, much less
dense than any other stable liquid in Nature. Therefore,
the liquid phase realm extends to unexpected regimes
never achieved before.

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