Rapid Communications

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.

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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 in 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 [6–8]. By tuning the short-range interaction, these works have observed the spontaneous formation of an array of self-bound droplets reminiscent of the characteristics of a classical ferrofluid. The observation of solidlike arrangements in dilute gases has also been possible working with highly excited Rydberg atoms [9]. By direct imaging, Schauss *et al.* [9] 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, the suggestion by Petrov relying on Bose-Bose mixtures [10] is very stimulating. 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 quasitwo dimensions may help to stabilize the liquid phase [11]. The LHY correction has also been used to account for the formation of dipolar drops [12] and then confirmed by full first-principles quantum Monte Carlo simulations [13,14].

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 two experimental teams [15,16]. Results obtained with a mixture of ^{39}K atoms in different internal states have shown the formation of these drops whose size remains constant for tens of milliseconds 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 high 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 Rapid Communication, 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 [17,18], ^{3}He [19,20], H_2 [21], and spin-polarized tritium [22]. By differing with 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 a gradual and simultaneous increase in density and reduction of size.

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,

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

with $V^{(\alpha,\beta)}(r)$ as the interatomic interaction between species α and β. We focus 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 $V^{(\alpha,\alpha)}(r)$ 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; note 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

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

with $\boldsymbol{R} = \{\boldsymbol{r}_1, \ldots, \boldsymbol{r}_N\}$, $N = N_1 + N_2$. 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 $r \geqslant a_{\alpha\alpha}$ 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 as 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 Rapid Communication 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 corresponds to $KR < \pi/2$. In practice, we fix the range of well R and vary depth V_0. As is obvious from the equation for a_{12}, its value depends on the product $RV_0^{1/2}$ and then decreasing R means to increase V_0. If for a fixed a_{12} value we want to approach the limit $R \to 0$ ($V_0 \to \infty$), our calculations become 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 is obtained with that. In the following, unless stated otherwise, all energies and lengths are given in $\hbar^2/(2m_1a_{11}^2)$ and a_{11} units, respectively.

The trial wave function $\Psi(\boldsymbol{R})$ (2) 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 [23] 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 [24]. 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.

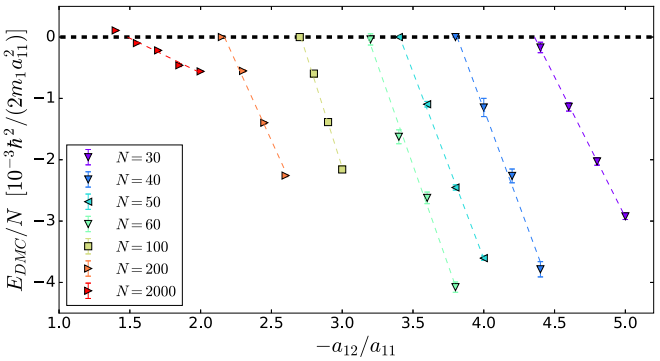

FIG. 1. Energy per particle of the Bose-Bose mixture as a function of the scattering length a_{12}/a_{11}. The different symbols and lines correspond to DMC calculations with different numbers of particles.

In Fig. 1, we report results for the energy per particle of the Bose-Bose mixture for a 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. Our results show a clear dependence of the critical scattering length for binding on the number of particles: Smaller drops require more attraction (larger V_0) than larger ones.

The dependence of the critical scattering length for self-binding $a_{12}^{\rm crit}$ on the number of particles is shown in Fig. 2. Plotted as a function of $1/N$ we observe a decrease in $a_{12}^{\rm crit}$, reaching in the thermodynamic limit ($N \to \infty$) a value slightly larger than one. In fact, LHY theory has been applied to the formation of Bose-Bose drops around this value of $|a_{12}| \sim a_{11}$ corresponding to drops with a very large number of particles [10]. In the same figure, we show results derived using a different range of $R = 10$ of the attractive well. As we can see, the dependence of $a_{12}^{\rm crit}$ with N is slightly different, but it seems that both curves tend to approach for the largest accessible $N = 2000$ value.

The calculation of the density profiles $\rho(r)$ allows for a better knowledge of the shape and size of the formed drops.

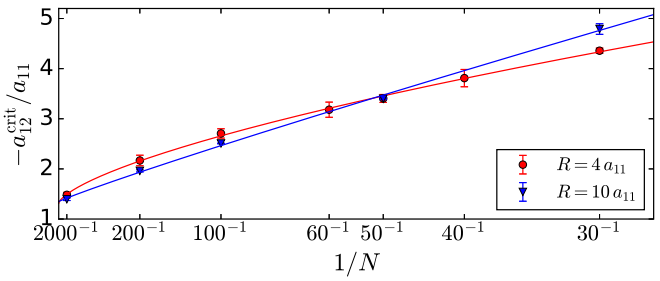

FIG. 2. Critical values $a_{12}^{\rm crit}$ for liquid-drop formation as a function of $1/N$. The circles and triangles stand for different sizes of the potential well. The lines correspond to fits to the DMC results.

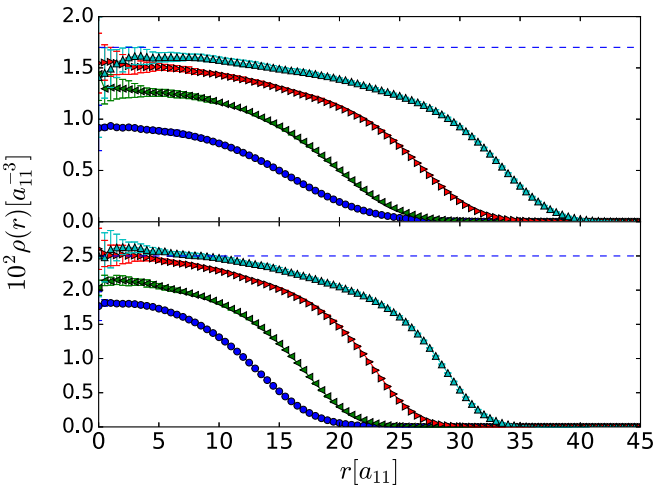

FIG. 3. Density profiles of the Bose-Bose liquid drops for a different number of particles. The top and bottom panels correspond to $V_0 = 0.150$, $a_{12} = -3.09$ and $V_0 = 0.166$, $a_{12} = -3.81$, respectively. From small to large drops, $N = 200$, 400, 1000, and 2000. The dashed lines correspond to the equilibrium density of the bulk phase.

In Fig. 3, we report DMC results on the density profiles of the obtained drops. Notice that there is no difference between the partial density profiles due to our election of interactions and masses $\rho^{(1)}(r) = \rho^{(2)}(r) = \rho(r)/2$. The two cases shown in Fig. 3 correspond to scattering lengths $a_{12} = -3.09$ (top) and $a_{12} = -3.81$ (bottom). When the number of particles increases one observes that both the central density and the radius of the drop grow. This is expected to happen until the central density reaches the equilibrium density of the bulk phase. In fact, the full shape of the profile evolves with N from a slow decay of the density to zero to a steeper one, trying to optimize volume and surface contributions to the total energy. Once the drop saturates, only the radius increases with the addition of more particles. The density profiles, shown here for two illustrative examples, correspond to very dilute liquids because we need $\sim$2000 particles to reach saturation. In the figures, we have also shown the equilibrium densities that we have obtained for the same potentials in preliminary calculations of the bulk phase. By increasing $|a_{12}|$, i.e., by making the system more attractive, we observe that the central density increases and the size of the drop squeezes. A direct comparison of our results with LHY theory [10] is not possible because our drops do not fulfill their validity condition $|a_{12}| \sim a_{11}$.

Apart from the central density one can also extract from the density profiles the surface width, usually measured as length W over which the density decreases from 90 to 10% of the inner density. It is expected that W increases with N 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 $W \simeq 20$; 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 the 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 $a_{12}^{\rm crit}$ for this N value and ends quite deep into the Feshbach resonance at a scattering length of $a_{12} \simeq 40a_{12}^{\rm crit}$. Following this ramp, we observe an increase in an order of magnitude in the inner density and a shrinking of the size with a reduction of the radius in a factor of 3. Therefore, the drop becomes denser, but it is still a fully stable object which is not at all collapsed. It is worth noting that, following this ramp, the interparticle distance approaches the range of the attractive interaction R making our results sensitive to the model potential (in the densest case of Fig. 4, $\langle r_{12}\rangle \simeq 2R$).

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 [17,18], 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

$$E(N)/N = E_v + E_s x + E_c x^2, \tag{3}$$

with $x \equiv N^{-1/3}$. 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 $t = E_s/(4\pi r_0^2)$. 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 ρ_0 as the equilibrium density of the liquid.

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

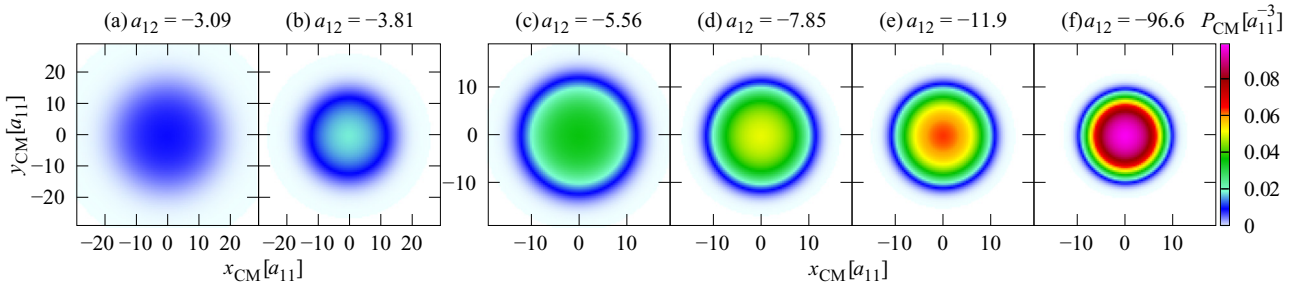

FIG. 4. Contour plots of the density profiles of a liquid drop with $N = 200$ as a function of a_{12}.

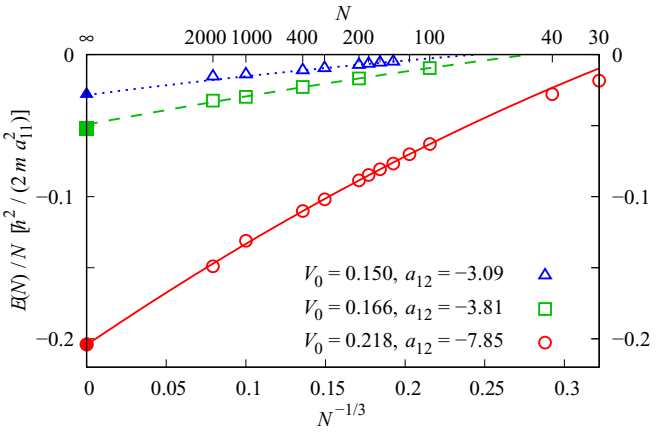

FIG. 5. Energy per particle of the Bose-Bose drops as a function of $N^{-1/3}$ and for different interparticle scattering lengths a_{12}. The open symbols are the DMC results, and the lines are fits according to the liquid-drop model (3). The error bars are smaller than the size of the symbols. The points on the zero x axis correspond to bulk calculations.

cases studied we obtain a high-fidelity fit. In the figure, we plot on the zero x axis the energies of the bulk liquid in the same conditions as the drops. These results are not included in the fit (3), but they are completely coincident with the energies E_v obtained solely from the drop energies. This is in fact a stringent test of accuracy on the calculations of the liquid drops. In the figure we see the effect of the potential on the energy of the drops for the three selected cases. The self-binding of a given N drop becomes stronger as depth V_0, and thus $|a_{12}|$, increase. We have verified that the absolute value of the energy grows linearly with V_0 close to the critical value for self-binding but, for larger potential depths, it increases faster. From the fits using the liquid-drop model, we estimate that the surface tension for the three cases shown in Fig. 5 are 0.18×10^{-2}, 0.37×10^{-2}, and 2.41×10^{-2} [in units $\hbar^2/(2m_1a_{11}^4)$] when $a_{12} = -3.09$, -3.81, and -7.85, respectively.

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 as the Bohr radius. Then, the saturation densities of the drops shown in Fig. 3 are $\sim1.0\times10^{-6}$ and 1.4×10^{-6} Å^{-3}. Near the critical scattering length for a given size, the drops are even more dilute, e.g., for $N = 2000$ and $a_{12} = -1.75$, the central density is about 3×10^{-8} Å^{-3}. The saturation density of liquid ^{4}He is 2.2×10^{-2} Å^{-3}, implying that the Bose-Bose drops can be as dilute as $\sim10^4$ times the ^{4}He ones (a similar ratio happens when compared with water with a density of 3.3×10^{-2} Å^{-3}) [25]. For the same number of atoms, the Bose-Bose drop is much larger than the ^{4}He one: 9.8×10^{-2} μm for $V_0 = 0.150$ and 3×10^{-3} μm for ^{4}He with $N = 2000$ [26]. The surface of the dilute drop for this N is $\sim50\%$ 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. 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. The experimental realization of Bose-Bose liquid drops [15,16] 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 the 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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[1] C. J. Pethick and H. Smith, *Bose-Einstein Condensation in Dilute Gases* (Cambridge University Press, Cambridge, UK, 2008).
[2] K. M. O'Hara, S. L. Hemmer, M. E. Gehm, S. R. Granade, and J. E. Thomas, Science **298**, 2179 (2002).
[3] *The BCS-BEC Crossover and the Unitary Fermi Gas*, edited by W. Zwerger (Springer, Heidelberg, 2012).
[4] J. Carlson, S. Y. Chang, V. R. Pandharipande, and K. E. Schmidt, Phys. Rev. Lett. **91**, 050401 (2003).
[5] G. E. Astrakharchik, J. Boronat, J. Casulleras, and S. Giorgini, Phys. Rev. Lett. **93**, 200404 (2004).
[6] H. Kadau, M. Schmitt, M. Wenzel, C. Wink, T. Maier, I. Ferrier-Barbut, and T. Pfau, Nature (London) **530**, 194 (2016).
[7] I. Ferrier-Barbut, H. Kadau, M. Schmitt, M. Wenzel, and T. Pfau, Phys. Rev. Lett. **116**, 215301 (2016).
[8] M. Schmitt, M. Wenzel, F. Böttcher, I. Ferrier-Barbut, and T. Pfau, Nature (London) **539**, 259 (2016).
[9] P. Schauss, M. Cheneau, M. Endres, T. Fukuhara, S. Hild, A. Omran, T. Pohl, C. Gross, S. Kuhr, and I. Bloch, Nature (London) **491**, 87 (2012).
[10] D. S. Petrov, Phys. Rev. Lett. **115**, 155302 (2015).
[11] D. S. Petrov and G. E. Astrakharchik, Phys. Rev. Lett. **117**, 100401 (2016).
[12] F. Wächtler and L. Santos, Phys. Rev. A **93**, 061603(R) (2016).
[13] H. Saito, J. Phys. Soc. Jpn. **85**, 053001 (2016).
[14] A. Macia, J. Sánchez-Baena, J. Boronat, and F. Mazzanti, Phys. Rev. Lett. **117**, 205301 (2016).
[15] C. R. Cabrera, L. Tanzi, J. Sanz, B. Naylor, P. Thomas, P. Cheiney, and L. Tarruell, Science **359**, 301 (2018).
[16] G. Semeghini, G. Ferioli, L. Masi, C. Mazzinghi, L. Wolswijk, F. Minardi, M. Modugno, G. Modugno, M. Inguscio, and M. Fattori, arXiv:1710.10890.

[17] V. R. Pandharipande, J. G. Zabolitzky, S. C. Pieper, R. B. Wiringa, and U. Helmbrecht, Phys. Rev. Lett. **50**, 1676 (1983).
[18] S. A. Chin and E. Krotscheck, Phys. Rev. B **45**, 852 (1992).
[19] R. Guardiola and J. Navarro, Phys. Rev. A **71**, 035201 (2005).
[20] E. Sola, J. Casulleras, and J. Boronat, Phys. Rev. B **73**, 092515 (2006).
[21] P. Sindzingre, D. M. Ceperley, and M. L. Klein, Phys. Rev. Lett. **67**, 1871 (1991).
[22] I. Bešlić, L. Vranješ Markić, and J. Boronat, J. Chem. Phys. **131**, 244506 (2009).
[23] J. Boronat and J. Casulleras, Phys. Rev. B **49**, 8920 (1994).
[24] J. Casulleras and J. Boronat, Phys. Rev. B **52**, 3654 (1995).
[25] Preliminary DMC calculations of the bulk liquid show that equilibrium densities as low as 4×10^{-11} Å^{-3} can be achieved. These would correspond to very large N drops, which are hardly accessible with direct DMC simulations. These densities are even below the ones observed experimentally in Ref. [15].
[26] M. Barranco, R. Guardiola, S. Hernández, R. Mayol, and M. Pi, J. Low Temp. Phys. **142**, 1 (2006).