



Ultradilute Quantum Droplets

A new class of quantum mechanical liquids is stabilized by an elegant mechanism that allows them to exist despite being orders of magnitude thinner than air.

Igor Ferrier-Barbut

In his PhD thesis from 1873, Johannes van der Waals devised a theoretical framework to describe the gas and liquid phases of a molecular ensemble and the phase transition from one to the other. That work resulted in the celebrated equation of state bearing his name. To this day, the van der Waals theory is still the prevailing picture in most physicists' minds to explain the emergence of the liquid state. It asserts that the liquid state arises at high densities from an equilibrium between attractive interatomic forces and short-range repulsion. Now, a new type of liquid has emerged in ultracold, extremely dilute atomic systems for which the van der Waals model does not predict a liquid phase.

Using the tools of laser cooling and trapping, experimenters can reach the ultracold regime to create atomic quantum gases.¹ Quantum interference effects between atoms are an important part of the statistical descriptions of those systems. However, if a monatomic ensemble is simply cooled, any chemical species will form a liquid instead of a gas due to van der Waals forces and the system will never reach the quantum regime. So to see quantum effects, the classical liquid state must be avoided. That requires extremely low densities that keep the distances between atoms much larger than the range of attractive forces that would bind the liquid. But keeping the atoms far apart traps them in a dilute, metastable state. A whole

new mechanism is needed for atoms in such dilute conditions to form a liquid phase.

Mean-field quantum gases

Atoms in the quantum regime must be described as waves rather than classical point-like objects. They come in two flavors, bosons and fermions. That characterization dictates particles' collective behavior: Bosons interfere constructively, whereas fermions do so destructively. In the materials used to make ultradilute liquids, constructive bosonic interference leads to the accumulation at very low temperature of all the atoms into the same quantum state with zero momentum. That collective



For ultracold experiments, atoms are held in vacuum by a glass cell, shown here. The atoms are then cooled to ultralow temperatures to create a Bose–Einstein condensate. (Photo courtesy of Wolfram Scheible.)

state is known as a Bose–Einstein condensate (BEC) and is now routinely produced experimentally (see, for example, the article by Keith Burnett, Mark Edwards, and Charles Clark, *PHYSICS TODAY*, December 1999, page 37).

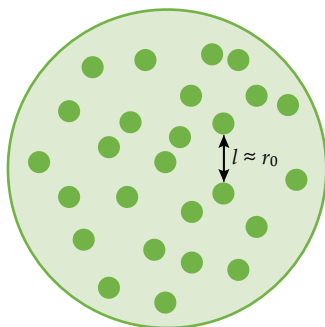
Bose–Einstein condensation is a pure quantum interference effect that requires no interaction between atoms. Interatomic forces do still affect that state, but rather than the familiar attractive potential with repulsive core, interactions take a much simpler form in ultracold and ultradilute conditions. Forming a BEC requires the interparticle distance l to be much larger than the typical interaction range r_0 to prevent the material from forming a classical liquid. Additionally, the thermal de Broglie wavelength λ_{dB} , which describes the typical atomic wavelength, needs to be larger than l . Together those conditions require that λ_{dB} be much larger than r_0 . In other words, the atomic waves cannot resolve the details of the interaction potential (see figure 1). As a result, the particles behave as if they were interacting through a zero-range contact potential that can be written as $V(r) = g\delta(r)$, where $\delta(r)$ is the Dirac delta function, g is a coupling constant, and r is the interatomic separation distance.

Despite the conceptual simplicity of the interaction, calculating the ground state of N bosons interacting through a contact potential is difficult and requires approximations. First is the mean-field approximation, which assumes that atoms all still occupy only one state, as they would in the absence of interactions; the interactions only modify the state with respect to the single-particle case. The ground-state energy E of an ensemble with uniform density n in a volume V is then simply $E/V = 1/2gn^2$. That equation says that the BEC can exist only in the gas phase: If g is positive, meaning that the particles are repulsive, then the energy of the system is lowest when n is minimized—in practice, that means the system always expands, which is why external trapping potentials are needed to confine BECs. On the other hand, if g is negative and the particles are attractive, the energy is minimized when n is maximized, so the ensemble collapses on itself. Both situations have been observed experimentally, but neither forms a liquid.

A game-changing correction

As usual, corrections to a quantum ensemble’s energy go beyond the mean-field approximation. The first of those corrections was calculated in the 1950s. At the time, the goal was to develop a theoretical description of superfluid helium. (For more information on superfluid helium droplets, see the article by Peter Toennies, Andrej Vilesov, and Birgitta Whaley, *PHYSICS TODAY*, February 2001, page 31.) The He–He interaction potential is far too complex to analytically solve in the many-body limit,

a Van der Waals liquid



b Ultracold atoms

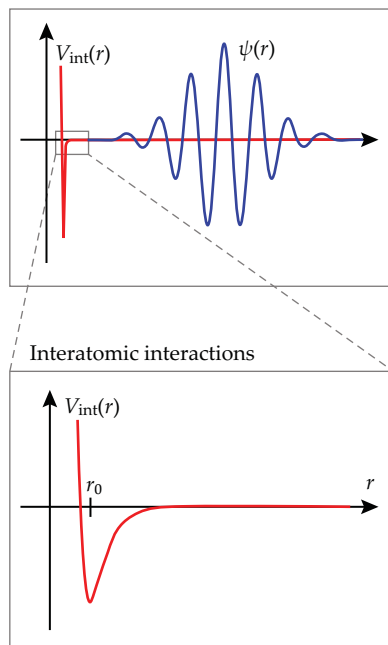


FIGURE 1. (a) IN A VAN DER WAALS

LIQUID, the nearest-neighbor interatomic distance l is on the order of the typical distance r_0 over which the interaction potential varies. **(b)** In the ultracold regime, two interacting atoms are described as matter waves $\psi(r)$. Their wavelengths are too large to resolve the details of the effective interaction potential $V_{int}(r)$ that only extends a radial distance r_0 from the atom. The effective interaction therefore has zero range, and van der Waals liquids cannot be found in ultracold conditions.

so a zero-range potential was used as an academic exercise. The first exact calculation of the next leading-order term in the energy came from Tsung-Dao Lee, Kerson Huang, and Cheng-Ning Yang in 1957 and is thus termed the LHY correction.² At that level of approximation, the ground state is composed of a large fraction of atoms still in the zero-momentum condensed state and also of a small non-condensed fraction, known as the quantum depletion, in higher momentum states. The interpretation of the LHY correction is that it accounts for the fact that the collective modes of the BEC are not fully at rest, even in the ground state, but undergo zero-point fluctuations as dictated by Heisenberg’s uncertainty principle.

Accounting for the zero-point energy in the ground state leads to a modified energy, $E/V = 1/2gn^2 + \alpha_{LHY}(gn)^{5/2}$, where α_{LHY} depends only on the atomic mass and Planck’s constant. The new expression recovers the mean-field term from before, along with an extra term from accounting for the quantum mechanical nature of the fluid. The correction becomes more important at higher densities,³ as shown in figure 2a. The beyond-mean-field theory at the LHY level is in excellent agreement with experiments that have observed the quantum depletion and measured the LHY energy correction.^{4,5} However, since the correction depends only on g and is repulsive, just like the mean-field term, the energy minimization works the same way and the atomic ensemble remains a gas.

The crucial ingredient for qualitatively altering the nature of the BEC was first laid out in 2015 in an imaginative theoretical proposal by Dmitry Petrov at CNRS in Orsay, France, and was incidentally experimentally observed shortly afterward by a group headed by Tilman Pfau at the University of Stuttgart in Germany.^{6,7} The two papers used different systems but with the same idealized situation: Imagine a bosonic system described by two separate interactions rather than one, with coupling constants g and g' . The energy is just the sum of the two contributions, so if the interactions both have the same sign, no qualitative change in behavior occurs.

An interesting situation arises when the two interactions are competing, meaning one is attractive (negative) and the other re-

pulsive (positive). The mean-field energy becomes $E/V = 1/2 \delta g n^2$ where $\delta g = g - g'$. Assuming g and g' are of the same order of magnitude, the mean-field energy is reduced but the qualitative behavior of the system does not change. Because of that reduction, however, the beyond-mean-field corrections are not necessarily negligible. The total energy is now given by $E_{\text{tot}}/V = 1/2 \delta g n^2 + \alpha'_{\text{LHY}} (gn)^{5/2}$, where α'_{LHY} depends on the ratio g'/g . As long as g and g' are individually not small, the LHY correction remains relatively large even as the mean-field term shrinks. The presence of two interactions can create collective high-energy excitations that have a large zero-point energy, which allows the LHY correction to be largely repulsive even as the mean-field term remains attractive.

Importantly, the first term in the expression for E_{tot}/V depends on δg , whereas the second depends on g and g'/g . When g and g' are of the same order and $g' > g$, the mean-field term is attractive ($\delta g < 0$) and the LHY correction is repulsive ($g, \alpha'_{\text{LHY}} > 0$). The resulting energy, shown in figure 2b, reaches a minimum at a finite density by balancing the weakened mean-field attraction at low n and the beyond-mean-field repulsion at high n . That competition enables the formation of a self-bound liquid.

A liquid and a gas differ essentially by their density.⁸ In this article, we define a gas by its expansion to fill the whole available volume; a liquid does not fill the whole volume, but instead forms a self-bound droplet with a fixed density. The peak density of a droplet in infinite volume can be thought of as the order parameter for the liquid–gas phase transition. It takes a nonzero value in the liquid phase but vanishes for a gas.

Making an ultradilute droplet

The question is, what experimental system is ruled by two different interactions? In general, only one type of contact inter-

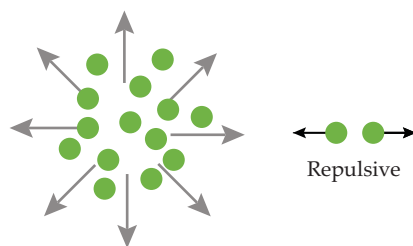
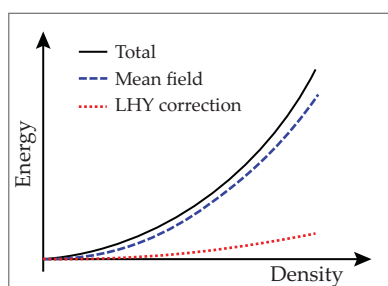
action results from the details of the short-range forces. Petrov's proposal to overcome that was to mix two types of bosons in which like atoms repel each other with one coupling constant, g_l , and unlike atoms attract each other with a coupling constant g_u . Using two different species allows the system to be effectively described by two different interactions, both coming from short-range forces.

Petrov showed that when the previously described conditions for g and g' (here g_l and g_u) were met, the mixture would form a liquid. Remarkably, the liquid would still be extremely dilute, so the interatomic distance l remains much larger than the interaction range r_0 . Another consequence of the low density is that the quantum depletion remains weak, so the LHY-level approximation remains valid. The existence of such a liquid is not explained by a van der Waals-like mechanism but instead stems from a many-body effect that is a consequence of the quantum mechanical nature of the bosonic ensemble. Petrov's proposal identified several concrete atomic mixtures in which such intraspecies repulsion and interspecies attraction could be found, which suggested that a liquid BEC could be realized in contemporary experiments.

Instead of two species, the Stuttgart experiments were performed on a single species of atoms with two different interactions. The experiments used dysprosium atoms, which have a large magnetic moment. As a result, they are subject not only to a repulsive contact interaction but also to an anisotropic dipole–dipole interaction. The dipole interaction is longer-ranged than the contact interaction⁹ and characterized by a coupling constant g_d that, in the experiments, was slightly larger than the contact coupling g . When the atoms are mostly distributed head-to-tail, the attractive dipole interaction leads to the same competition between attractive and repulsive interactions as in the two-species system.

The mean-field energy at the center of a droplet again reads $E/V \approx 1/2 \delta g n^2$, although the equality is no longer exact because the effective dipolar interaction is slightly altered when the dipoles are not exactly head-to-tail, and it predicts collapse because $\delta g < 0$. However, in experiments the bosonic system formed stable, long-lived droplets—as before, once beyond-mean-field effects are accounted for, the ensemble forms a liquid.^{10,11} Following the observation of liquid droplets with dysprosium,

a Gas



b Liquids

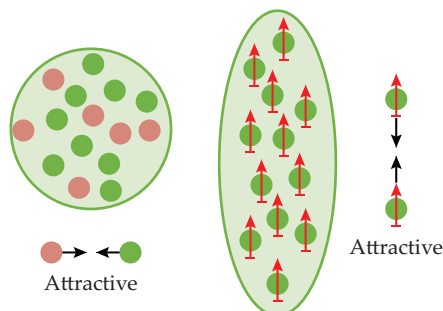
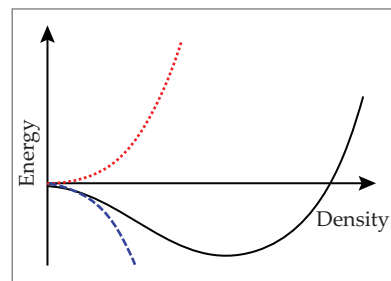
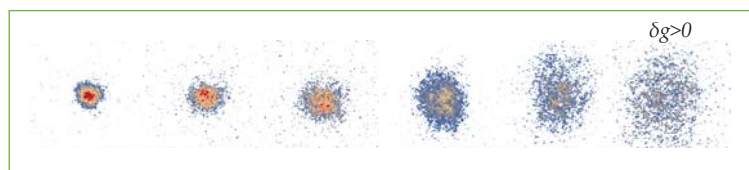


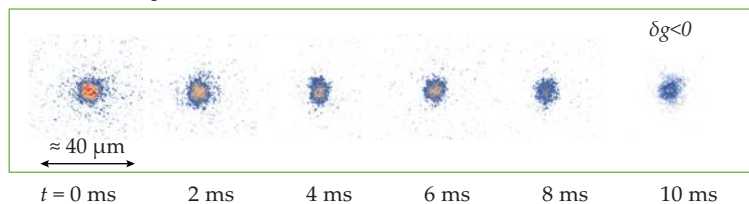
FIGURE 2. (a) A TYPICAL SINGLE-SPECIES BOSE-EINSTEIN CONDENSATE has repulsive short-range interactions. The sum of the mean-field energy and the Lee-Huang-Yang (LHY) correction is positive and therefore repulsive, so the atoms are not bound and form a gas. **(b)** Bose-Einstein condensates of atomic mixtures or magnetic atoms can have both attractive and repulsive interactions. When the mean-field energy and the LHY correction have opposite signs, the total energy can develop a minimum at finite density, which causes the atoms to form self-bound liquid droplets.

QUANTUM DROPLETS

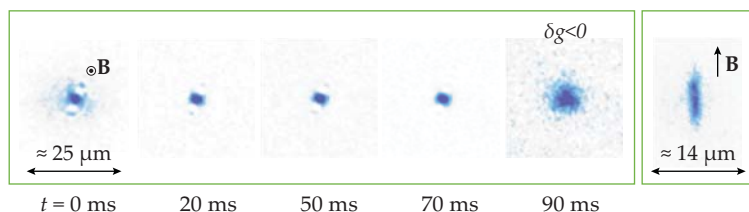
a Repulsive gas



Self-bound droplet



b Top view



Side view

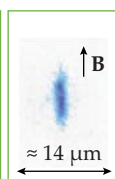


FIGURE 3. SELF-BOUND DROPLETS in the absence of an external trapping potential can be imaged experimentally. The densities of two such systems are shown here. **(a)** When the mean-field energy of a Bose–Bose mixture is repulsive (top row), it results in a gas phase, and the droplet expands over time. When the mean-field energy is attractive (bottom row), the size of the self-bound droplet remains constant over time, although its density, shown in false color, decays due to three-body losses. (Adapted from ref. 13.) **(b)** A droplet of dipolar magnetic atoms stays confined until the atom number decays below the critical atom number, at which point it starts expanding as a gas. The side view of the droplet clearly shows its elongation along the magnetic field **B**. (Adapted from ref. 14.)

tion captures the liquid and gas phases, but it completely ignores finite-size and surface effects by assuming a uniform density n . In any real liquid droplet, the density is not uniform; it increases from zero at the droplet's edge to a peak value at its center. For matter waves such as BECs, such density gradients cost kinetic energy, as dictated by the

Schrödinger equation. In droplets of dilute quantum liquids, kinetic energy acts as a surface tension, contributing an additional energy that depends on the density gradient at the surface. The consequence can be dramatic, because if the surface tension shifts the total energy from negative to positive, then the self-bound solution no longer exists and the ground state is a gas. Quantum liquids thus have another very peculiar feature: Kinetic energy, accounting for single-particle quantum fluctuations, can drive a liquid-to-gas transition.

Another way to think about the effective surface tension is that the density distribution created by all the atoms in a droplet acts as a trapping potential on each individual atom because of the effectively attractive interaction. If the trapping potential is strong enough to hold a bound state, then it supports a self-bound liquid solution. If not, then the ensemble forms a gas. A third possibility is that the liquid exists in a metastable state, but for low enough atom number, the trapping becomes too shallow and only the gas exists. The depth of the effective potential is determined by the number of atoms, N , and the difference between the two interaction strengths, δg . For larger values of N and more negative δg , the trapping volume is larger and the attraction is stronger, so the effective potential becomes more binding. A liquid–gas phase diagram can therefore be drawn as a function of system size and interaction strength,^{6,15} as shown in figure 4. The critical atom number N_c that marks the gas–liquid transition varies as $|\delta g/g|^{-2/5}$, so the minimal atom number to sustain a self-bound droplet grows dramatically as δg approaches zero.

The structure of the phase diagram has been confirmed experimentally^{13,14} using a mechanism that experimenters usually try to avoid: three-body losses. The number of atoms in a liquid drop decreases as a result of collisions that recombine two atoms into one molecule. To respect conservation of energy and momentum, such a recombination can only happen if three atoms are involved in the collision, so the loss rate grows strongly with density. When a liquid droplet is created, losses

experiments at the University of Innsbruck, Austria, under the direction of Francesca Ferlaino showed the same stabilization with erbium atoms,¹² which, like dysprosium, have a large magnetic moment. The experiments confirmed that the stabilization mechanism is general to atoms that possess competing short-range repulsion and longer-range dipole interactions.

Many experimental groups can produce bosonic atomic mixtures with a variety of elements and isotopes. The mixture that won the race for the first observation of a two-component liquid phase was a blend of two internal states of the same isotope of potassium. By creating the proper mixture of internal states in the right magnetic field, two teams, one led by Leticia Tarruell at the Institute of Photonic Sciences in Barcelona, Spain, and the other by Marco Fattori at the University of Florence, Italy, both observed the ultradilute quantum liquid phase.¹³ They confirmed their findings by removing all external trapping potentials, thus placing the BEC in an infinite volume. If the condensate were still a quantum gas, it would have expanded until the density was too thin to be measurable. Instead, the researchers saw self-bound droplets that did not expand in free space and could easily be observed for long times.¹³

The experimental results shown in figure 3 for Bose mixtures and magnetic atoms¹⁴ are visual proof of the gas–liquid phase transition. The theoretical prediction and later observation of quantum liquids marked a paradigm shift because they showed that the LHY correction, which was thought to be a small quantitative shift due to weak quantum fluctuations in a many-body system, can stabilize a liquid phase. That phase would be impossible under mean-field conditions. The diluteness of those liquids is remarkable, with typical densities being about four orders of magnitude lower than air and about eight orders of magnitude lower than liquid helium at room pressure.

Oscillating and disappearing droplets

Ultradilute liquids also exhibit another feature rooted in their quantum mechanical nature. Their simplified energy descrip-

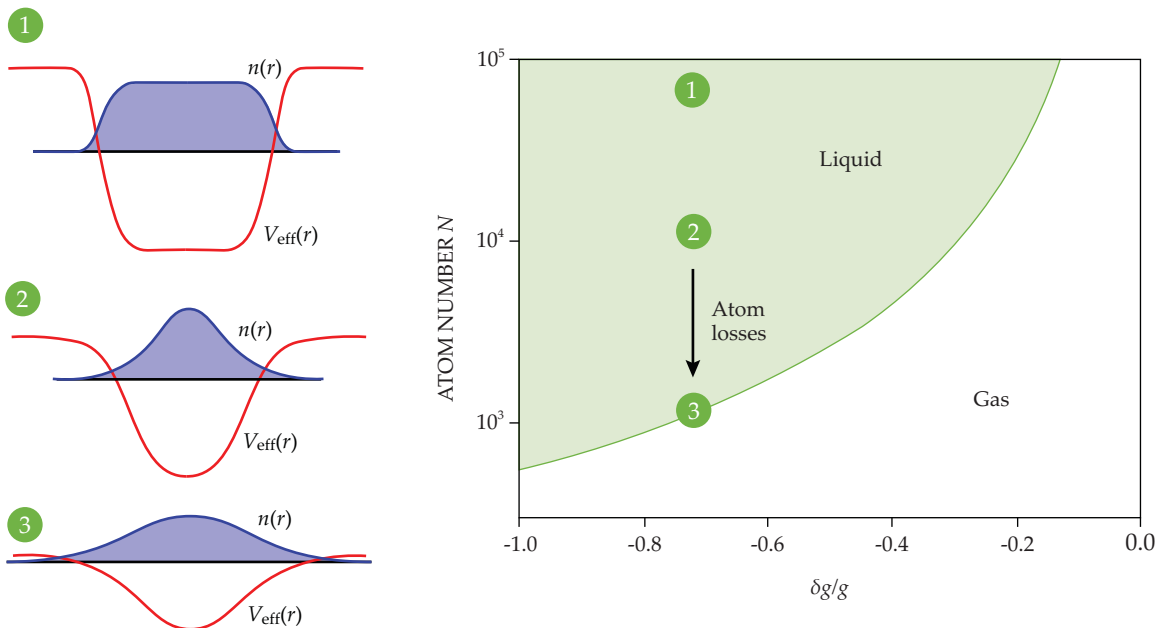


FIGURE 4. THE PHASE DIAGRAM FOR AN ULTRADILUTE DROPLET depends on the atom number N , the contact coupling g , and the difference between the mean-field and beyond-mean-field coupling constants δg . At high N (point 1 in the phase diagram), the density $n(r)$ of a droplet has a flat-top profile, which creates a deep self-binding potential $V_{\text{eff}}(r)$ and a liquid droplet. Experiments obtain lower N (point 2), and the density does not reach the flat top, but the self-trapping remains relatively deep. Atom losses deplete the droplet over time until the self-bound state barely exists (point 3). When N decreases further, the droplet crosses the phase boundary and becomes a gas.

typically limit its lifetime to between a few and tens of milliseconds. During that time, the atom number decays until it reaches the liquid–gas phase transition. At that point the self-bound liquid turns into a gas, the atoms expand in space, and the density immediately drops, as in the final frame of figure 3b (90 ms).

Once the liquid transitions to a gas, the three-body losses stop, and the number of atoms stays constant. Experimenters can therefore readily identify the critical atom number N_c for the liquid–gas transition at a given δg . They can also adjust the attraction strength using so-called Feshbach resonances, which vary the coupling constant g for contact interactions by means of a magnetic field. By varying the coupling constant and measuring the critical atom number, researchers mapped the phase diagram for the different experimental quantum liquids.

Although the two-component and dipolar liquids share the same stabilization mechanism, each also has its own characteristics. Quantum liquids of dipolar atoms are anisotropic: For the dipolar interaction to be attractive, the atoms need to be aligned. As a result, droplets are elongated along the dipole direction, as can be seen experimentally in figure 3. The shape of the dipolar droplets results from a competition between dipolar interactions trying to align the atoms and a surface tension that favors a round droplet. In atomic mixtures, the density ratio between the two species is locked to a value fixed by the precise short-range interactions. However, one species can end up in overabundance, which causes a gas halo of untrapped majority atoms to form around the droplet.

The elongation of dipolar droplets and the fixed density ratio for mixtures lead to specific, collective oscillation modes, illustrated in figure 5. Dipolar quantum droplets feature a scissor-like oscillation that corresponds to an angular oscillation of the droplet around the dipole's axis.¹⁶ Quantum mixture droplets, on the other hand, exhibit excitations in which the two components move either in or out of phase relative to each

other. Accurately mapping their spectrum of collective excitations should yield precious information about their precise equation of state beyond the current description.

The future of ultradilute liquids

The discovery of physically realizable ultradilute liquids highlights the strengths of ultracold atom experiments. Using exquisite control of the constituents of a many-body system and the interactions that characterize it, such experimental setups can expose the key mechanism that underpins the many-body state. The family of ultradilute quantum liquids will likely continue to grow because the same stabilization mechanism can be found in other systems.¹⁷ Theoretical proposals have already been laid out for mixtures of other constituents, such as bosons and fermions.

Exploring the possibility of making such liquids in lower dimensions is also of great interest because quantum fluctuations are enhanced, so any attractive potential allows for a self-bound liquid solution. Additionally, quantum droplets are localized matter waves in three dimensions, so they bear similarities to matter-wave solitons, which can be fully accounted for within mean-field theory and have been observed solely in lower dimensions. In strongly confined systems, competition between solitons and quantum droplets can lead to a crossover or abrupt transition between the two states.¹⁸ Localized matter waves could also be useful for performing interferometry, and the prospect of using quantum droplets experimentally to avoid trapping potentials remains to be investigated. (For an experimental example of matter-wave interferometry, see PHYSICS TODAY, April 2015, page 12.)

The exploration of the properties of ultradilute quantum liquids is in its infancy. The quantum depletion and LHY correction are, in theory, accompanied by quantum entanglement, and observing the presence of entanglement in the liquid phase

QUANTUM DROPLETS

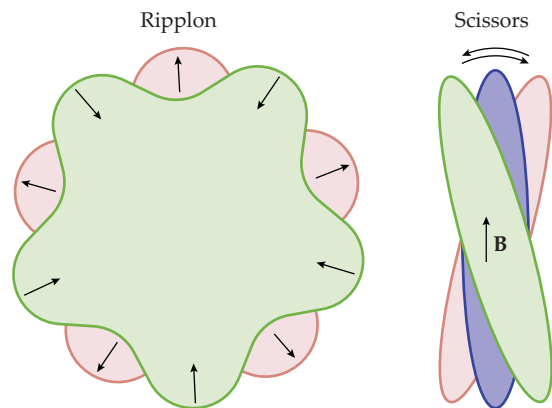


FIGURE 5. ISODENSITY CUTS OF QUANTUM DROPLETS at different times show their collective oscillation modes. Ripplons are typical for normal liquid droplets. They arise from surface tension, which creates a restoring force when the droplet is deformed, and are expected to exist in mixture droplets.⁶ The scissors mode is a signature of the breaking of rotational invariance in dipolar droplets. It consists of an angular oscillation of the elongated droplet around the direction of the magnetic field $\mathbf{B}$ and has been observed in dysprosium droplets.¹⁶

would be remarkable. The thermodynamics of such systems is also unknown—it is not yet clear whether or how thermal equilibrium is reached within a droplet.

While the theoretical descriptions of ultradilute liquids have progressed and approximations that include the LHY correction allow for an analytical expression for their energy,

many-body theories still lack a precise description of such liquids. However, in some existing experimental systems the usually dominant mean-field energy is masked, so beyond-mean-field effects that include interactions can be effectively magnified and measured. Corrections beyond the LHY description of the bosonic ensemble remain unobserved, but ultradilute quantum liquids finally provide a new testing ground for theories of quantum many-body interactions.

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