

Self-Bound Superfluid Membranes and Monolayer Crystals of Ultracold Polar Molecules

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We investigate the physics of ultracold dipolar molecules using path-integral quantum Monte Carlo simulations, and construct the complete phase diagram extending from weak to strong interactions and from small to mesoscopic particle numbers. Our calculations predict the formation of self-bound quantum droplets at interaction strengths lower than previous estimates for molecular condensates. Strikingly, we observe that, for stronger interactions, the oblate quantum droplet transitions to a two-dimensional sheet or superfluid membrane with a thickness of a single molecule. As interactions are increased further the system continually loses superfluidity while correlations develop, and is eventually found to undergo a transition to a crystalline monolayer that remains self-bound without external confinement. The spontaneous formation of such two-dimensional phases from a three-dimensional quantum gas is traced back to the peculiar anisotropic form of the dipole-dipole interaction generated by microwave dressing of rotational molecular states. For sufficiently large particle numbers, crystallization takes place for comparably low interaction strengths that do not promote two-body bound states and should thus be observable in ongoing experiments without limitations from three-body recombination.

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Explorations of dipolar quantum matter have revealed a rich phenomenology of exotic quantum states that emerge from the long-range nature and anisotropy of dipole-dipole interactions [1,2]. Recent advances have been spurred by experiments on ultracold gases of atoms with large magnetic dipole moments [3,4], which demonstrated the formation of new states such as free-space quantum droplets [5,6] and supersolid phases [7–9], and probed their physical properties [10–15]. Experiments also highlighted the importance of interaction effects beyond simple mean field theory [16,17] that led to an improved understanding of the role of quantum and thermal fluctuations in weakly interacting quantum gases [15,18–21] and their successful modeling in terms of the so-called extended Gross-Pitaevskii equation [22–25].

Since electric dipole-dipole interactions generally exceed interactions of atomic magnetic dipoles significantly, ultracold polar molecules have long been anticipated to open new parameter regimes for dipolar quantum

matter [26–29] and for studying quantum magnetism [30,31]. Here, the application of static and microwave fields offers broad opportunities to engineer and control molecular interactions [32–36] by properly coupling their rotational states. Such techniques have been designed and used experimentally to stabilize molecular gases against short-range collisional losses [37–42] and ultimately enabled the realization of Bose-Einstein condensates of heteronuclear polar molecules [43]. Recent theory predicts [28,44,45] the formation of molecular droplet states that are similar to those of weakly interacting atomic gases but with different geometries.

In this Letter, we report path integral Monte Carlo (PIMC) simulations of microwave-dressed bosonic molecules and explore the transition from weak to strong dipole-dipole interactions. Our results reveal a characteristic change in the morphology of dipolar quantum droplets, which self-confine in the xy plane and form a two-dimensional monolayer superfluid, i.e., a free-space superfluid sheet that reflects the transition into the strong-interaction regime. At even stronger interactions, we eventually find a crystallization transition into a self-bound two-dimensional lattice of molecules. Here, particles crystallize into a single monolayer of ordered dipoles, which shares similarities with two-dimensional van der Waals materials [46] and forms in free space without additional confinement due to an in-plane minimum of the interaction potential [Fig. 1(b)] and strong out-of plane repulsion [Fig. 1(c)]. Our calculations suggest that this new phases of dipolar matter should be observable under typical

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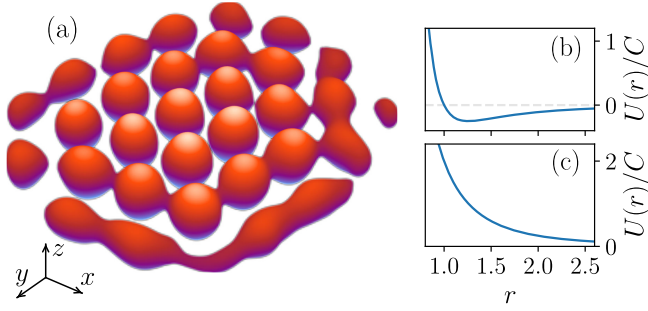


FIG. 1. (a) Illustration of a self-bound monolayer crystal, with a single molecule in each lattice site. Shown is the isosurface at a fixed particle density obtained from quantum Monte Carlo simulations for $N = 32$ molecules with a dimensionless interaction strength $C = 25$ [cf. Eq. (3)]. Panels (b), (c): antidipolar potential as a function of the intermolecular distance r , in (b) the xy plane and (c) along the z axis.

conditions of ongoing experiments with ultracold molecules. Importantly, we find that crystallization occurs for interaction strengths below the threshold for two-body field-linked bound states [32,47,54], avoiding detrimental three-body losses in experiments [55]. Interestingly, this scenario is akin to the unique behavior of low-temperature helium, where a solid phase emerges directly from an atomic quantum fluid [56], without the prior formation of a gas or fluid of bound pairs (molecules). Our results thus indicate the great potential of ultracold microwave-dressed molecules for exploring exotic many-body phenomena in quantum fluids with highly controllable interactions.

Among the different approaches to control molecular interactions with external fields, we focus here on the simplest configuration of a single microwave used to couple rotational molecular states and thereby induce a controllable dipole-dipole interaction. Importantly, the dressed molecules also feature a repulsive core that shields pairs of molecules from short-range collisions [35]. For weak dressing, the resulting interaction potential can be approximated by the simple expression [57]

$$V(\mathbf{R}) = \frac{C_6}{R^6} \sin^2\theta(1 + \cos^2\theta) + \frac{C_3}{R^3}(3\cos^2\theta - 1), \quad (1)$$

where $R = |\mathbf{R}|$ denotes the distance between two molecules and θ is the angle between the distance vector $\mathbf{R}$ and the orientation of the dipole, set by the applied microwave field. The interaction coefficients C_3 and C_6 are determined by the molecular dipole moment and can be controlled by the parameters of the applied microwave field [47]. The microwave dressing results in a peculiar antidipolar character of the interaction.

Choosing the dipole orientation along the z axis, the interaction potential features a minimum in the x - y plane [Fig. 1(b)] while it is purely repulsive along the orthogonal direction [Fig. 1(c)]. The position of the minimum is set by

the competition of the short-range repulsion and the long-range, attractive dipole-dipole interaction. The associated distance $R_0 = (C_6/C_3)^{1/3}$, at which the potential $V(R_0) = 0$ vanishes, provides a natural unit of length. Upon scaling energies by $E_0 = \hbar^2/(mR_0^2)$, and introducing $\mathbf{r} = \mathbf{R}/R_0$, the Hamiltonian for N interacting molecules with mass m can be written as

$$\hat{H} = -\frac{1}{2} \sum_{i=1}^N \nabla_{\mathbf{r}_i}^2 + \sum_{i<j} U(\mathbf{r}_i - \mathbf{r}_j), \quad (2)$$

with the dimensionless interaction potential

$$U(\mathbf{r}) = \frac{V(\mathbf{R})}{E_0} = C \left(\frac{\sin^2\theta(1 + \cos^2\theta)}{r^6} + \frac{3\cos^2\theta - 1}{r^3} \right), \quad (3)$$

that depends on a single interaction parameter $C = mC_3^{4/3}/(\hbar^2 C_6^{1/3})$. Hence, the equilibrium properties of the system are determined by only two parameters, the particle number N and the dimensionless interaction strength C .

We study the low-temperature quantum phases of the Hamiltonian (2) by using path integral Monte Carlo simulations (PIMC) [58]. Quantum Monte Carlo methods have been successfully applied to the description of dipolar bosonic systems in different geometries [25,45,59–70]. In PIMC, the partition function is sampled as a Monte-Carlo integral over a set of discrete imaginary-time trajectories, i.e., world lines associated to each particle. An efficient representation of the symmetrized many-body quantum state is possible through the so-called worm algorithm [71] that samples bosonic permutations by opening, closing and permuting world lines.

Just like other quantum Monte Carlo methods, PIMC simulations should, in principle, yield exact results, apart from statistical uncertainties. In practice, however, a judicious choice of initial configurations and a proper balance of different Monte Carlo moves is often required to achieve efficient convergence to equilibrium. As we discuss below, this is particularly important in the present case for a reliable prediction of phase boundaries and to avoid unphysical states associated with metastable world line configurations. Our simulations are performed at a low temperature $T = 0.01E_0/k_B$, and we have confirmed that further decreasing the temperature does not change the results reported here. Our results across a range of values of C and N are summarized in Fig. 2.

We simulate a finite number of particles in free space with open boundary conditions. Hence, the equilibrium phase for weak interactions $C \ll 1$ corresponds to an entirely delocalized gas with vanishing density and a finite kinetic energy proportional to the temperature T . As the value of C increases, the attractive part of the dipole-dipole interaction potential counteracts the kinetic energy of the

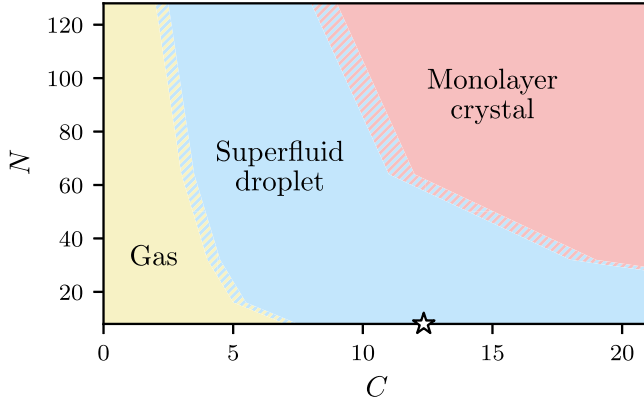


FIG. 2. Diagram showing the different phases of molecules in free space for different particle numbers N and interaction strengths C . The yellow region indicates the gas phase, the blue region indicates the self-bound, superfluid droplet phase, while the red region indicates the self-assembled monolayer crystal. Transition regions are represented by striped areas. The star marks the lowest value of C at which the potential admits a bound state (see Ref. [47]).

particles and eventually promotes the formation of self-bound quantum droplets, as previously found in Bose-Einstein condensates of magnetic atoms [5,6] and predicted for molecular interactions such as Eq. (1).

While the Monte Carlo simulations straightforwardly yield stable droplet states for sufficiently large interactions, an accurate determination of the boundary between the gas and droplet phases requires further consideration. To this end, we start from a localized initial configuration and track the changing state of the system during Monte Carlo moves by monitoring the spatial spread of the molecular gas, defined as $\sigma_x^2 = N^{-1} \int dx x^2 n_{3D}(\mathbf{r})$, and analogously for σ_y and σ_z , where n_{3D} is the density of molecules. In the droplet phase, the spatial spread converges to a finite value following an initial relaxation phase. In the gas phase, the initial configuration expands continuously, and σ_x asymptotically grows linearly with the number of Monte Carlo steps. The slope, D , of this linear growth can be used to discriminate between the gas phase ($D > 0$) and the droplet phase ($D = 0$), as illustrated in Fig. 3(a); the transition point is confirmed by the energy turning negative in the droplet phase. Plots of the size as a function of Monte Carlo steps are shown in Fig. 3(b).

In order to facilitate convergence of the simulations toward the droplet phase, we sample the starting position of the particles from a uniform distribution inside a cylinder of a given radius and height. We have thoroughly checked that the results do not depend on the choice of the initial cylinder [47]. However, we observe that if the initial configuration is too dilute, the molecules tend to fragment into smaller separate droplets. While such multidroplet states may appear to form an intriguing metastable phase [45], we find that they disappear upon including collective

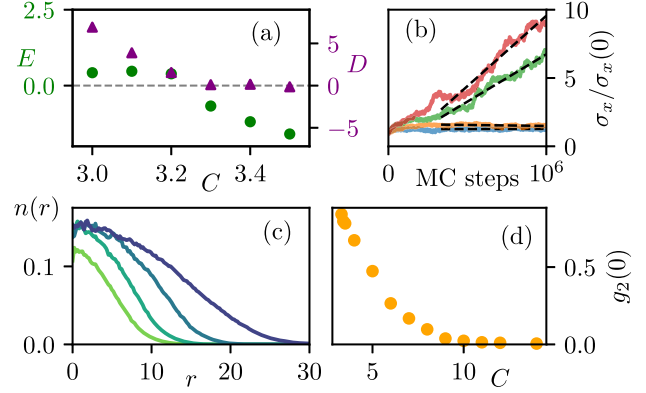


FIG. 3. (a) Total energy, E , (green circles) and slope, D , of σ_x (purple triangles) for $N = 64$ molecules at different values of C around the gas-to-droplet transition. The latter is obtained by fitting the size σ_x as a function of Monte Carlo steps, as illustrated in panel (b) for $C = 2.5$ (red), 3.0 (green), 3.5 (orange), 4.0 (blue). (c) Radial two-dimensional density $n(r) = \int dz n_{3D}(\mathbf{r})$ of the droplet for $N = 16$ (green), $N = 32$ (light blue), $N = 64$ (dark blue), $N = 128$ (purple) at C close to the gas transition. (d) Density-density correlation function in the xy plane at $\rho = 0$.

Monte Carlo moves of multiple particles [47]. Similar behavior was also found in the presence of harmonic confinement, indicating that long-lived multidroplet states do not represent the equilibrium state under the interaction Eq. (1) [47], although multidroplet states may form in experiments in out-of-equilibrium conditions.

In order to ensure the accuracy of our phase boundary for the gas-to-droplet transition, we also run simulations using as starting configuration the most weakly bound droplets observed at each N , while reducing C . In each case, we observe the disgregation of the droplet, confirming the result from the cylinder starting conditions. The critical interaction strengths are notably smaller than recent predictions, indicating that the described approach succeeds in finding lower energy states than previous variational estimates [44] and Monte Carlo simulations [45].

Close to the transition, the droplets feature a superfluid fraction close to unity and a quasi-2D geometry, extended in the x - y plane. The pancakelike shape arises due to the anisotropic form of the interaction potential (3), where the long-range attraction perpendicular to the dipole-orientation [Fig. 1(b)] facilitates self-trapping in the x - y plane, while the strong longitudinal repulsion [Fig. 1(c)] energetically favors an oblate droplet shape. The central density remains constant along the transition line [Fig. 3(c)], but increases away from it upon increasing the interaction strength C [Figs. 4(a)–4(f)]. Near the transition, the droplet size along the z axis, σ_z , exceeds the characteristic scales of the interaction potential [47] and the droplet represents an oblate superfluid with an approximately Gaussian density profile along all three directions. [Fig. 3(c)]. With increasing interaction strength, the droplet undergoes a structural transition and forms a two-dimensional superfluid composed

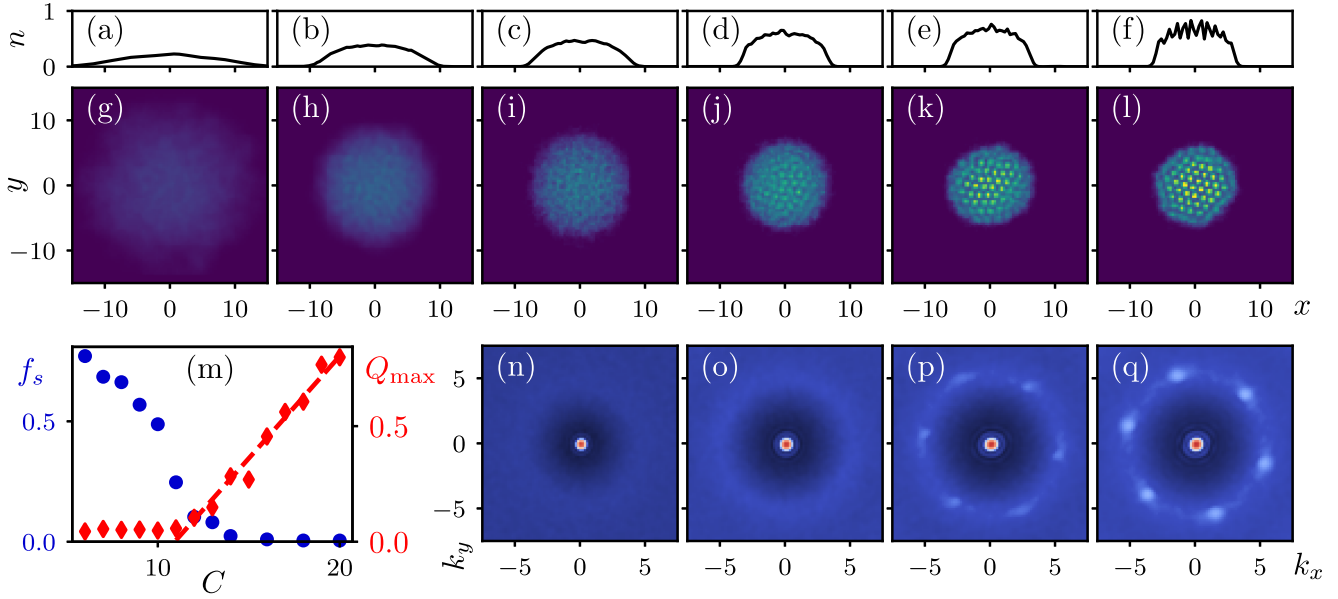


FIG. 4. Structural properties of the droplet in the strongly interacting regime. (a)–(f) Profiles of the 2D density n for $N = 64$ molecules at successive values of $C = 4, 6, 8, 12, 14$, and 18 . (g)–(l) n as a function of both x and y , for the same values of C . (m) Contrast $Q_{\max}$ (red diamonds) and superfluid fraction (blue circles) as a function of C . (n)–(q) Structure factor S for $C = 8, 12, 14$, and 18 , respectively.

of a single molecular layer—a superfluid sheet or membrane that remains stable in free space. One can trace this transition by inspecting the density-density correlation function in the xy plane, $g_2(r_\perp)$, with $r_\perp$ the distance between two molecules in the x – y plane [47]. In a 3D or quasi-2D geometry, particles can stack in the z direction, such that $g_2(r_\perp \rightarrow 0)$ is small but finite. Conversely, in a truly 2D geometry, where particle motion is confined to the x – y plane, the strong short-range repulsion leads to $g_2(r_\perp \rightarrow 0) = 0$. As shown in Fig. 3(d), $g_2(0)$ indeed decreases with growing interaction and vanishes prior to the transition to the crystalline phase, indicating an intermediate 2D superfluid phase, with a near-constant bulk density and a decreasing superfluid fraction with rising C [Fig. 4(m)]. This distinctively different geometry and lower superfluidity indicate a new self-bound phase of strongly correlated molecules.

Further increasing the interaction strength continually increases the droplet density, and the superfluidity eventually vanishes at a 2D droplet density of $n \approx 0.7$ [Fig. 4(m)], for which the interparticle spacing corresponds to the minimum of the in-plane potential well at $r_{\min} = 2^{1/3}$ seen in Fig. 1(b). At this point, the system undergoes a transition to an insulating crystalline monolayer of molecules, as illustrated by the density profiles shown in Figs. 4(g)–4(l). The finite superfluid fraction in the region from $C \approx 11$ to $C \approx 14$ can be ascribed to the finite size of the droplet: crystalline order emerges in the center first, while the edges initially remain superfluid. In order to quantify the crystallization transition, we define the structure factor

$$S(\mathbf{k}) = \left| \int d\mathbf{r} \rho(\mathbf{r}) e^{-i\mathbf{k} \cdot \mathbf{r}} \right|^2, \quad (4)$$

where we only consider in-plane momenta with $k_z = 0$. Upon entering the crystalline monolayer phase, $S(\mathbf{k})$ develops a peak structure at the lattice momentum $|\mathbf{k}| \sim 2\pi/r_{\min} \approx 5$, as shown in Figs. 4(n)–4(q). Its sixfold rotational symmetry reflects the triangular lattice of single molecules that forms in the bulk of the 2D droplet. We calculate the average $\bar{S}(k) = (1/2\pi) \int d\varphi S(\mathbf{k})$ to determine the angular contrast $Q(k) = (1/2\pi) \int d\varphi (S(\mathbf{k}) - \bar{S}(k))^2$, by integrating over the angle φ in the k_x – k_y plane. One can then use the maximum contrast $Q_{\max} = \max_k Q(k)$ to characterize the crystalline order of the droplet. As shown in Fig. 4(m), the order parameter $Q_{\max}$ starts to increase linearly with C coincidentally with the vanishing of the superfluidity. We can thus use a linear fit to determine the transition point, as illustrated in the figure.

The transition line between the superfluid droplet and the monolayer-crystal phase is shown in Fig. 2. The critical value of C decreases with the particle number and is expected to approach a constant value as N increases toward the thermodynamic limit. For sufficiently large particle numbers $N \gtrsim 60$, the crystallization occurs at interaction strengths $C < C_b \sim 12.3$ for which the potential does not promote two-body bound states. Remarkably, the considered system of microwave-dressed molecules thus resembles the rare phenomenology of low-temperature helium, where a solid phase occurs via crystallization of an atomic superfluid without the formation of a molecular

gas or fluid of two-body bound states. Interestingly, the two-body bound state is far more extended along the z direction [47] than the spatial spread σ_z of the droplet. The presence of the bound state is thus not expected to impact the physics of the droplet phase for the parameters considered in Fig. 2.

The absence of bound states is also important for potential experiments, as it should enable the observation of the predicted monolayer-crystal phase without detrimental losses from three-body recombination [55,72] into field-linked tetramer states of two bound molecules. Microwave dressing and shielding have been demonstrated in recent experiments with ultracold CaF [38], NaK [73], NaRb [42], and NaCs [40,43] molecules, using different field configurations. The simplest scheme based on a single field yields an interaction potential that is approximately given by Eq. (3), where the dimensionless interaction strength, C , is determined by the molecular dipole moment and mass, as well as the frequency detuning and Rabi frequency of the applied microwave [47]. Typical microwave Rabi frequencies of 10 MHz [38,40,42,73] yield interaction strengths of up to $C \sim 11$ for NaK molecules and even stronger interactions with up to $C \sim 55$ for NaCs molecules, such that the predicted phases appear to be within experimental reach for both molecular species.

In summary, we have explored the phases of bosonic ultracold dipolar molecules in free space. The peculiar form of the interaction potential that is induced by microwave dressing with a single applied field promotes the formation of self-bound quantum droplets with a characteristic pancakelike geometry. Our PIMC simulations predict this droplet formation to occur at weaker interactions than previously calculated [44,45]. As the droplet solution features a negative total energy, this result is conceptually consistent with the variational calculations of [44]. We point out that these droplets are strongly correlated, unlike dilute gases of magnetic atoms, and bear more resemblance to ^4He droplets [74]. Unlike helium, the high degree of quantum control in ultracold molecules allows for investigation of new phases by tuning the shape and strength of particle interactions. The simulations reveal a change in the droplet morphology upon approaching the strongly interacting regime. Hereby, the droplet forms a two-dimensional superfluid sheet before crystallizing into a triangular crystalline monolayer of individual molecules. Their physical origin and properties thus differ fundamentally from Wigner crystals of repulsive dipoles under strong three-dimensional confinement [33,61,75].

A careful variation and analysis of the starting conditions, the parameters of the PIMC algorithm, and the introduction of collective Monte-Carlo moves leads us to exclude the formation of multidroplet states in free space, and we have also found no evidence for such ground-state solutions under external confinement. Such multidroplet states or even cluster solids and supersolids may, however,

form under different forms of interactions that can be engineered with multiple dressing fields [43,76] and motivate future studies.

Our analysis suggests that the predicted monolayer crystals should be observable under typical conditions of ongoing ultracold molecule experiments. In particular, we show that they form for interaction strengths that do not promote two-body bound states and, thus, avoid detrimental effects of collisional losses in potential experiments. This appears reminiscent of the behavior of helium to crystallize directly from a superfluid state without an intermediate molecular gas or liquid phase of two-body bound states. This demonstrated analogy, together with the high tunability of interactions, indeed presents an exciting outlook for future explorations of exotic and long-elusive phases, such as defect-induced supersolids [77,78], in new interaction regimes that are now becoming accessible with ultracold dipolar molecules.

Note added—Self-bound droplets, very similar to the superfluid droplet phase predicted in this work, have been observed recently in experiments with microwave-shielded polar molecules [79,80].

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Data availability—The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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