A superfluid-droplet crystal and a free-space supersolid in a dipole-blockaded gas

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A novel supersolid phase is predicted for an ensemble of Rydberg atoms in the dipole-blockade regime, interacting via a repulsive dipolar potential "softened" at short distances. Using exact numerical techniques, we study the low temperature phase diagram of this system, and observe an intriguing phase consisting of a crystal of mesoscopic superfluid droplets. At low temperature, phase coherence throughout the whole system, and the ensuing bulk superfluidity, are established through tunnelling of identical particles between neighbouring droplets.

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The search for novel phases of matter drives much of the current research in condensed matter physics. Of particular interest are phases simultaneously displaying different types of order. A chief example, of great current interest, is the so-called *supersolid*, namely a phase featuring crystalline order, and also capable of sustaining dissipation-less flow. Attempts to observe experimentally a supersolid phase of matter, primarily in a crystal of solid helium, have spanned four decades since early theoretical predictions [1]. The most credible claim of such an observation to date [2, 3], has been subjected to in-depth scrutiny over the past few years, and it seems fair to state that agreement is lacking at the present time, as to whether experimental findings indeed signal a supersolid phenomenon [4].

A new, fascinating avenue to the observation of supersolid and other phases of matter not yet observed (or even thought of), is now opened by advances in cold atom physics, providing not only remarkably clean and controlled experimental systems, but also allowing one to "fashion" artificial inter-particle potentials, not arising in any known condensed matter system. This allows one to address a key theoretical question, namely which two-body interaction potential(s), if any, can lead to the occurrence of a supersolid phase. While this is well established for bosons in an optical lattice, it remains an open question how to realize this, and other novel, exotic phases, in free space.

In this Letter, we show that interaction potentials which combine a long-distance repulsion with a short-distance cutoff, will lead to the appearance of a novel self-assembled crystalline phase of mesoscopic superfluid droplets in a system of bosons. Furthermore, such a crystal can turn supersolid in the $T \to 0$ limit, as tunneling of particles across neighbouring droplets takes place, and superfluid phase coherence is established across the whole system, as individual separate Bose condensates (droplets) organize into a single, global condensate. Thus supersolidity, as arising in this system, is of a fundamentally different kind with respect to the one, defect-induced, originally envisioned by Andreev and Lifshitz [1]. Specifically, we consider the following two-body potential:

$$v(r) = \begin{cases} D/a^3 & \text{if } r \le a \\ \\ D/r^3 & \text{if } r > a \end{cases} , \qquad (1)$$

D being the characteristic strength of the interaction. This interaction potential can be realized with cold Rydberg atoms in the dipole-blockade regime [5], where D and a are parameters which can be controlled with external fields [6, 7], as shown below.

Our system of interest comprises N identical bosons of mass m, confined to two dimensions [17]. The many-body Hamiltonian is the following (in dimensionless form):

$$\mathcal{H} = -\frac{1}{2}\sum_{i=1}^{N}\nabla_i^2 + \sum_{i>j} v(r_{ij}) \qquad (2)$$

where $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$ is the distance between particles i and j, and v is given by Eq. (1). All lengths are expressed in terms of the characteristic length $r_\circ = mD/\hbar^2$, and we introduce a dimensionless cutoff $R_c = a/r_\circ$ for the potential (1). The system is enclosed in a square cell of area A, with periodic boundary conditions. The particle density is $n = N/A$, but we shall express our results in terms of the (dimensionless) inter-particle distance $r_s{=}1/\sqrt{nr_\circ^2}$. The energy scale is $\epsilon_\circ = D/r_\circ^3 = \hbar^2/mr_\circ^2$.

The low-temperature phase diagram of such a system has been explored by means of first principles numerical simulations, based on the Continuous-space Worm Algorithm [8, 9]. It is important to note at the outset that, while the numerical results presented here were obtained with the two-body potential (1), the main physical conclusions do not depend on its detailed form. Indeed, we have observed the same physical behaviour with potentials featuring a smoother merge of short- and long-range behaviours, as well as a different long-range tail than Eq. (1) – we come back to this point below.

Numerical results shown here pertain to simulations with a number of particles N varying between 50 and

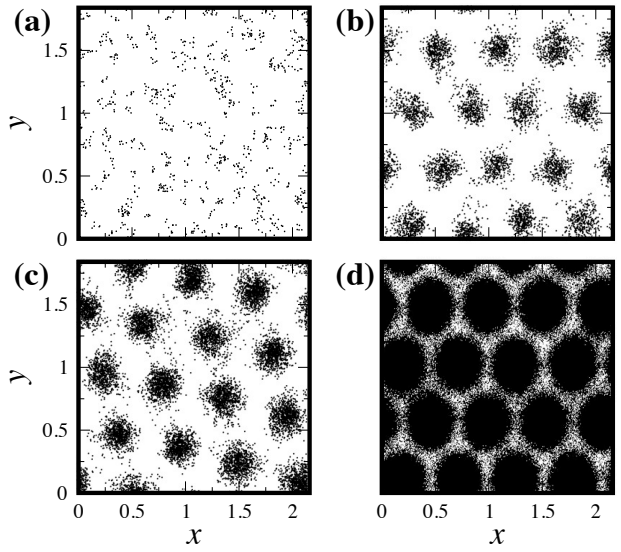

FIG. 1: Snapshots of a system of bosons interacting via potential (1), at the four different temperatures 200 (a), 20 (b), 1.0 (c) and 0.1 (d), expressed in units of $\epsilon_\circ$. Points shown are taken along individual particle world lines. The nominal value of r_s in this case is 0.14, whereas the cutoff of the potential (1) is R_c=0.3.

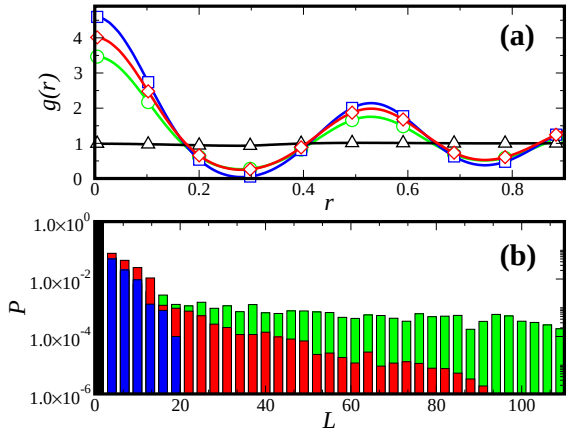

FIG. 2: (Color online) *Top*: pair correlation function $g(r)$ at a temperature T= 200 (triangles), 20 (squares), 1.0 (diamonds) and 0.1 (circles), expressed in units of $\epsilon_\circ$. The nominal value of r_s in this case is 0.14, whereas the cutoff of the potential (1) is R_c=0.3. The simulated system comprises N=200 particles. *Bottom*: Relative frequency of occurrence of permutation cycles of length L at the same four temperatures. Longer permutation cycles occur at lower temperature.

400, in order to carry out extrapolation of the results to the thermodynamic limit. Our ground state estimates are obtained as extrapolations of results at finite temperature. Details of the simulations are standard, as the use of the potential (1) entails no particular technical difficulty.

In the limit $R_c \ll r_s$, the truncation of the dipolar potential at short distances does not play an important role, and the low temperature phase diagram of (2) is that of purely dipolar bosons in two dimensions, investigated previously by several authors [10, 11]. It is known that for $r_s \lesssim r_s^C = 0.06$ the ground state of the system is a triangular crystal, whereas for $r_s \gtrsim r_s^L = 0.08$ it is a uniform superfluid (in the intermediate density range a more complex scenario is predicted [12]). As we show below, a very different physics sets in when $R_c \gtrsim r_s$, in the density ranges which correspond to either the crystalline or superfluid phase in the purely dipolar system.

Fig. 1 shows typical configurations (i.e., particle world lines) produced by Monte Carlo simulations of a system of bosons interacting via the potential (1), at a nominal density corresponding to $r_s = 0.14$, at different temperatures spanning three orders of magnitude. The value of the cutoff R_c in this case is 0.3. At the highest temperature, a simple classical gas phase is observed, as shown by the pair correlation function $g(r)$, shown in Fig. 2 (a), which is just a constant (note that $g(r)$ does not vanish at the origin, owing to the flattening off of the potential at short distance). As T is decreased, an intriguing effect takes place, namely particles bunch into mesoscopic droplets, in turn forming a regular (triangular) crystal. This is shown qualitatively in the snapshots in Fig. 1, but also confirmed quantitatively by the structure of the $g(r)$ as well (Fig. 2(a)), which displays pronounced, broad maxima, as well as well-defined dips, where the function approaches zero. We henceforth refer to this phase as the *droplet-crystal* phase.

The formation of such droplets is a purely classical effect, that depends on the flattening off of the repulsive inter-particle potential below the cutoff distance. In fact, a simple estimate of the number N_d of particles per droplet, can be obtained by considering a triangular lattice of point-like dipoles, each one of strength $\propto N_d$ (as it comprises N_d particles), and by minimizing with respect to N_d the potential energy per particle, for a fixed density. The result is

$$N_d = \gamma \left(\frac{R_c}{r_s} \right)^2 \qquad (3)$$

where $\gamma \approx 2.79$. Eq. (3) furnishes a fairly accurate estimate of N_d for the (wide) range of values of the parameters r_s and R_c explored here. For instance, using the parameters of Fig. 1, we find from (3) $N_d \approx 13$, which agrees quite well with our simulation result. It is worth noting that a similar sort of pattern formation, due to competing interactions, has been previously established for classical colloidal systems [13, 14].

In the $T \to 0$ limit, long exchanges of identical particles can take place, as a result of particles tunneling from one droplet to an adjacent one. Long exchanges of particles can result in a finite superfluid response throughout

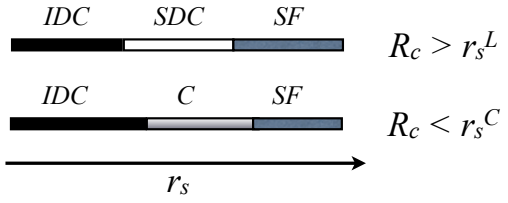

FIG. 3: Schematic ground state phase diagram of (2) as a function of r_s. The superfluid droplet crystal (SDC), namely supersolid phase, is sandwiched between an insulating droplet crystal (IDC) and a superfluid (SF). For $R_c \lesssim r_s^C$, an IDC, a single-particle crystal (C) and a superfluid phases are observed. The widths of the SDC and C regions depend on the value of R_c.

the whole system [18], and indeed for $R_c \gtrsim r_s^L$ we observe such a bulk superfluid signal, in a range of values of r_s in the vicinity of $R_c/2$. Because superfluidity arises in concomitance with the droplet-crystal structure, the denomination *supersolid* seems appropriate. At T=0, such a phase is sandwiched between an insulating droplet crystal at high density (i.e., lower r_s) and a homogeneous superfluid phase at lower density. For $R_c \lesssim r_s^C$, only two insulating phases are observed, namely the insulating droplet crystal at high density and the crystal of single particles, already detected in Refs. [10, 11], as well as a superfluid phase at lower density. All of this is summarized in the schematic phase diagram shown in Fig. 3.

It is important to stress that supersolid behaviour in this system does *not* originate from highly mobile point defects, such as vacancies or interstitials. Rather, tunnelling of particles between droplets which are themselves individually superfluid occurs, and the individual superfluid droplets connect to form a bulk superfluid. This is reminiscent of the phase-locking mechanism in a (self-assembled) array of Josephson junctions.

In order to establish that droplets are individually superfluid, one may consider the statistics of permutation cycles. Fig. 2 (b) shows the frequency of occurrence of exchange cycles involving a varying number L of particles ($1 \leq L \leq N$), at three different temperatures, at the physical conditions of Fig. 1. As one can see, as the temperature is lowered exchange cycles involving growing numbers of particles occur, involving almost all particles N at the lowest temperature; however, even at a higher temperature (e.g., T=20 in Fig. 2) one observes exchanges comprising a number of particles up to $\sim N_d$, i.e., particles inside an individual droplet. This is evidence that droplets are individually Bose condensed and superfluid, even though the system as a whole does not display superfluidity. This is observed to be the case at low T, for all values of R_c and r_s for which droplets form. That droplets should always be superfluid at low T is not surprising, given that particles in a droplet are essentially

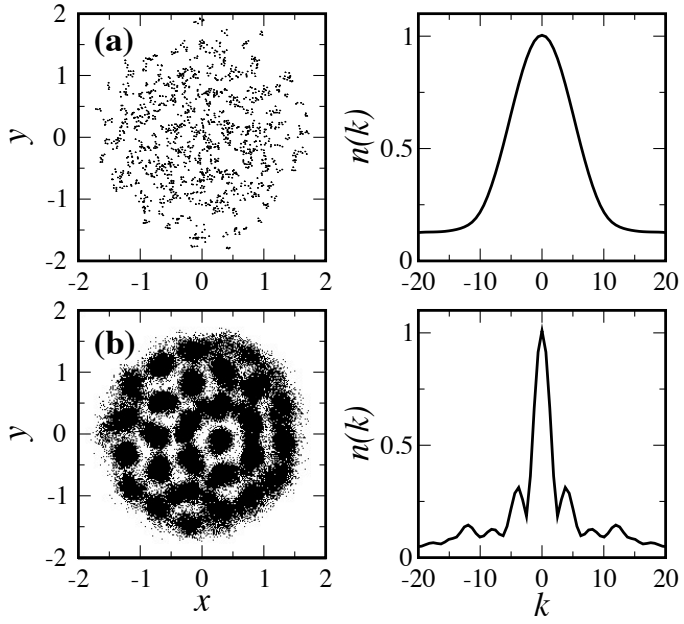

FIG. 4: (Color online) (a) Monte Carlo configurations of a system of $N = 400$ particles, interacting via the potential (1), confined in a harmonic trap of strength $\Gamma = 500\ \epsilon_\circ$, at the two temperatures T=100 $\epsilon_\circ$ (top) and T=0.5 $\epsilon_\circ$. Right panels show the corresponding momentum distributions, all normalized to unity for comparison purposes. The value of R_c in this case is 0.3. The development of secondary peaks at low temperature signals the occurrence of a supersolid phase.

non-interacting, due to the flatness of the potential at short distance. However, that droplets are themselves superfluid does not imply that a bulk supersolid phase will *always* occur in the $T \to 0$ limit, as discussed above.

The results discussed so far pertain to numerical simulation of the system described by Eq. (2) in its bulk phase. However, in any experiment aimed at probing the physics of such a system, the assembly of particles must necessarily be finite (a few thousand particles is a typical number for current experiments with cold dipolar atoms), held together by an external potential, due to the purely repulsive nature of the interaction. In order to enable a direct comparison with possible future experiments, we have performed simulations of the same system spatially confined *in-plane* by a harmonic trap, i.e., the term $\Gamma \sum_i \mathbf{r}_i^2$ is added to Eq. (2), $\Gamma \equiv m\omega^2/2$ being the strength of the trap.

Fig. 4 shows typical many-particle configurations of a trapped system comprising N=400 particles, at two different temperatures. Also shown are the associated momentum distributions $n(k)$, which are obtained by Fourier transforming of the *spherically and translationally averaged* one-body density matrix, computed by Monte Carlo. The momentum distributions are all nor-

malized to unity, for comparison purposes.

Clearly, one can readily identify the same droplet crystal structure predicted for the bulk, at low temperature. Droplets with a well-defined average number of particles form, and organize themselves on a triangular lattice. Correspondingly, the momentum distribution, which is directly observable experimentally by time-of-flight measurements [15], develops a sharp central peak, with additional structure on its sides. The secondary peaks correspond to oscillations in the one-body density matrix, in turn reflecting particle tunnelling to adjacent droplets. They are therefore connected to the appearance of the supersolid phase, as explained above.

Summarizing, accurate numerical simulations of a system of dipolar particles interacting via a potential softened at short distance, reveal the existence of a low temperature crystalline phase of superfluid droplets. This phase turns superfluid (supersolid) at $T \to 0$ through a mechanism of tunnelling of particles between adjacent droplets. The interaction that underlies such an intriguing, until now unobserved physical behaviour, can be realized with dipolar atoms in the dipole-blockade regime. We conclude by commenting on the dependence of the results on the particular form of potential utilized here, namely Eq. (1), which displays an abrupt, sharp cutoff at $r = a$. We have obtained qualitatively similar results with different potentials, featuring a smoother merge of long- and short-range behaviours, for example

$$V(r) = \frac{D}{a^3 + r^3} \tag{4}$$

This interaction potential is naturally realized in a cold gas of alkali atoms by weakly dressing the groundstate $|g\rangle$ of each atom with an excited Rydberg state $|r\rangle$ with a large dipole moment d, in the kDebye range [6]. Using a laser coupling with effective Rabi frequency Ω and red detuning $|\Delta| \gg \Omega$, the dressed groundstate reads $|\tilde{g}\rangle \sim |g\rangle - (\Omega/2\Delta)|r\rangle$, providing a small fraction $(\Omega/2\Delta)^2$ of excited Rydberg atoms. The cutoff $a \simeq (d^2/\hbar|\Delta|\epsilon_0)^{1/3}$ arises because of the well-known Rydberg-blockade mechanism, and has typical values in the hundreds of nm, while $D = \Omega^4 d^2/(8\pi|\Delta|^4\epsilon_0)$ [5].

Spontaneous emission rates γ_r from $|r\rangle$ are strongly reduced to values of at most $\gamma \simeq (\Omega/\Delta)^2\gamma_r$, which can be of the order of several tens of second for coupling to Rydberg states with large principal quantum number n, allowing for the observation of the phases above [6, 7]. For example, for a ^{87}Rb in an electric field $F = 25$kV/m, with $n = 45$ and experimentally reasonable $\Omega/2\pi \times 10$MHz and $\Delta/2\pi = 180$MHz, one obtains $a \simeq 500$nm and $\gamma \simeq (\Omega/\Delta)^2\gamma_r = 0.1$s. Collective many-body effects in the Rydberg-blockade regime [16] not described by Eq. (2) should be negligible provided $(R_c/r_s)^2 \ll (2\Delta/\Omega)^2$, which is readily satisfied for parameters as in Fig. 1.

It is worth noting that the droplet crystal phase does not crucially depend on the dipolar form of the interaction at long distances. Indeed, it is also observed in our simulations for an interaction of the type discussed in Ref. [7], namely $v(r) \sim 1/r^6$. All of this suggests that this phase should be observable experimentally, using cold atoms, under relatively broad conditions.

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[1] A. F. Andreev and I. M. Lifshitz, Sov. Phys. JETP. **29**, 1107 (1969).
[2] E. Kim and M. H. W. Chan, Nature **427**, 225 (2004).
[3] E. Kim and M. H. W. Chan, Science **305**, 1941 (2004).
[4] N. Prokof'ev, Advances in Physics **56**, 381 (2007).
[5] M. Lukin, M. Fleischhauer, R. Cote, L. Duan, D. Jaksch, J. I. Cirac, and P. Zoller, Phys. Rev. Lett. **87**, 037901 (2001).
[6] G. Pupillo, A. Micheli, M. Boninsegni, I. Lesanovsky, and P. Zoller, ArXiv:1001.0519, Phys. Rev. Lett., in press (2010).
[7] N. Henkel, R. Nath, and T. Pohl, ArXiv:1001.3250, Phys. Rev. Lett., in press (2010).
[8] M. Boninsegni, N. Prokof'ev, and B. Svistunov, Phys. Rev. Lett. **96**, 070601 (2006).
[9] M. Boninsegni, N. V. Prokof'ev, and B. V. Svistunov, Phys. Rev. E **74**, 036701 (2006).
[10] H. P. Büchler, E. Demler, M. Lukin, A. Micheli, N. Prokof'ev, G. Pupillo, and P. Zoller, Phys. Rev. Lett. **98**, 060404 (2007).
[11] G. E. Astrakharchik, J. Boronat, I. L. Kurbakov, and Y. E. Lozovik, Phys. Rev. Lett. **98**, 060405 (2007).
[12] B. Spivak and S. Kivelson, Phys. Rev. B **70**, 155114 (2004).
[13] Y. H. Liu, L. Y. Chew, and M. Y. Yu, Phys. Rev. E **78**, 066405 (2008).
[14] A. J. Archer, C. Ionescu, D. Pini, and L. Reatto, Journal of Physics: Condensed Matter **20**, 415106 (2008).
[15] M. Greiner, O. Mandel, T. Esslinger, T. W. Hänsch, and I. Bloch, Nature **415**, 39 (2002).
[16] J. Honer, H. Weimer, T. Pfau, and H. P. Büchler, ArXiv:1004.2499 (2010).
[17] A system of cold dipolar atoms or molecules can be confined to two dimensions using a tight (magnetic or optical) trap along z.
[18] We compute the superfluid fraction through the usual *winding number* estimator for the bulk system.