Supersolid structure and excitation spectrum of soft-core bosons in 3D

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By means of a mean-field method, we have studied the zero temperature structure and excitation spectrum of a three-dimensional soft-core bosonic system for a value of the interaction strength that favors a crystal structure made of atomic nano-clusters arranged with FCC ordering. In addition to the longitudinal and transverse phonon branches expected for a normal crystal, the excitation spectrum shows a soft mode related to the breaking of gauge symmetry, which signals a partial superfluid character of the solid. Additional evidence of supersolidity is provided by the calculation of the superfluid fraction, which shows a first-order drop, from 1 to 0.4, at the liquid-supersolid transition and a monotonic decrease as the interaction strength parameter is increased. The conditions for the coexistence of the supersolid with the homogeneous superfluid are discussed, and the surface tension of a representative solid-liquid interface is calculated.

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I. INTRODUCTION

A supersolid is a phase of matter that shows both crystalline and superfluid properties, i.e. the simultaneous breaking of continuous translational and global gauge symmetries, as originally proposed in Ref.[1], resulting in the formation of an ordered crystal structure with a phase coherence that allows for partial superfluid flow through the solid [2, 3].

Theoretical and experimental efforts have been focused in recent years to investigate the most natural candidate for supersolidity, i.e. ⁴He at low temperature, especially after the apparent observation of Non-Classical Rotational Inertia (NCRI) effects by Kim and Chan [4]. Consensus is lacking, however, as to whether the experimental data are really a manifestation of supersolidity in ⁴He, and more recent measurements cast some doubts on this hypothesis [5] (for a recent review about possible supersolidity in ⁴He and other system, see Ref.[6, 7]).

The possibility of formation of a solid structure simultaneously possessing crystalline and superfluid properties has been associated long ago [8] with an excitation spectrum of the liquid phase characterized by a roton minimum at finite q-vector, the liquid to "supersolid" transition being triggered by softening of the roton minimum.

A necessary condition for developing a roton minimum is that the interaction pair-potential has a Fourier transform that becomes negative in some range of the q-vector. This can be understood by recalling the Bogoliubov dispersion for the uniform liquid of bosonic particles interacting via a pair potential V(r)

$$\hbar\omega(q) = \sqrt{\left(\frac{\hbar^2 q^2}{2M}\right)\left[\frac{\hbar^2 q^2}{2M} + 2\rho \tilde{V}(q)\right]}$$
 (1)

where $\tilde{V}(q)$ is the Fourier transform of the interaction potential V(r) and ρ is the liquid density. When $\tilde{V}(q)$ has a negative contribution in some range of q-vectors,

thus balancing the quadratic q-term in the above expression, a roton minimum may develop, and the roton gap decreases with increasing the density, ultimately vanishing at some critical value where ω becomes imaginary. This marks the onset of a dynamical instability at which density modulations may spontaneously develop with no energy cost. Such softening instability could be equivalently reached in a flowing superfluid with a non-zero roton gap, as predicted for ⁴He flow [9], and later seen in Density Functional simulations [10]. A similar effect arises in metastable superflow states of soft-core bosons in 2D [11].

If a roton minimum is present, spontaneous solidification into a crystal structure is actually possible even before the roton gap disappears [8, 12]. As numerically found, for instance, in Ref.[12], by increasing the density, the roton gap decreases until a critical value is reached where the system undergoes a first-order phase transition to an ordered structure, which may have supersolid nature. This happens for instance in the case of ⁴He where, by applying pressure, the roton gap decreases because of the associated increase in the density, and the formation of a crystal occurs well before the roton gap disappears [13].

Supersolid phases have been recently predicted for confined condensed spinless bosons in 2-dimensions interacting via a broad class of soft core repulsive potentials (i.e. short range interactions which do not grow arbitrarily large at short distance but remain instead at a finite value, say V_0), which might be experimentally realized via strongly enhanced Van der Waals interaction between Rydberg atoms [14, 15], making them ideal ingredients to realize exotic quantum many-body phases of matter [14–18]. At low temperature, such systems are predicted to make a transition from the condensed superfluid phase to a crystal structure made of atomic clusters arranged in an ordered lattice superstructure, resulting in a number of supersolid phases [15, 19], the phase co-

herence being established through quantum hopping of atoms across adjacent clusters.

Classically, cluster formation comes as a consequence of the soft-core interaction, i.e. the energy cost for forming close particle pairs is bound by V_0 , so that the energy cost associated with the particle overlap remains finite. This may enable the formation of "solid" structures made of clusters of atoms when the density is so high that multiply occupied states become energetically favored upon increasing the lattice constant. In the associated quantum bosonic system, two important effect are added, i.e. the possibility of particle "hopping" between adjacent clusters and the possibility of exchange-driven Bose-Einstein condensation, resulting in supersolidity.

zero-temperature phase diagram of twodimensional Bosons with finite range soft-core interactions has been theoretically described in Ref. [20, 21], where both mean-field and first principles Monte Carlo simulations have been used. This model is known to admit in 2-dimensions a ground state which is a "cluster crystal", and moreover it supports NCRI; three sound (gapless) modes are expected in the solid phase, identifiable as the Goldstone bosons associated with the breaking of continuous global symmetries. Besides the two phonon branches of a "normal" two-dimensional crystal associated with the breaking of translational invariance in 2-dimensions, a third mode may appear, associated with the breaking of global gauge invariance which leads to the phase coherence of a superfluid Quantized vortices are also expected in a supersolid, just like in the superfluid phase, as shown in the numerical simulations of Ref. [18]. A supersolid can thus be defined as any inhomogeneous structure with translational long-range order, which exhibits an excitation spectrum with the characteristic features described above [18].

The excitation spectrum of a soft-core bosonic supersolid has also been studied using first principles method in Ref.[22] for a 2-dimensional supersolid structure, showing both the phonon modes appropriate to a solid structure and a softer collective excitation related to broken translational and gauge symmetry, respectively.

In Ref.[23] the excitation spectrum of a 2-dimensional soft-core supersolid has been computed at a mean-field level by solving the Bogoliubov-deGennes equations, clearly showing the presence of the mode associated with the superfluid fraction of the solid. In particular, a rather good agreement was found between the mean-field predictions and the results of Quantum Monte Carlo simulations [22, 23].

In Ref.[24] a Bose-Einstein supersolid phase in 2-dimensions with a step-like pair interaction showing a soft-core below a given core radius and a dipolar repulsive term at larger distances has been studied and the phase diagram obtained. The nucleation of vortices in a dipole-blockade 2-dimensional supersolid condensate and its effect on the superfluid fraction has been investigated in Ref.[25]. Again, evidence of a supersolid phase of a

2-dimensional dipolar crystal has been found from zero-temperature Quantum Monte Carlo simulations [26].

All of the theoretical results mentioned above are in 2-dimensions except for the mean-field study of the ground-state structural properties of the 3D nonuniform phase of bosons interacting through a dipole-blockade type interaction $V(r) = C_6/(r^6 + R_c^6)$ [14], which constitutes a strong candidate to exhibit supersolid properties. In a recent work Saccani et al [22] show that, in 2D, the simple model of "soft spheres" features the same basic physics as produced by interaction with a $1/r^n$ interaction outside the soft sphere.

It seems thus timely to investigate in a quantitative way, even if only at the mean-field level, a threedimensional system of bosons interacting through a softcore potential, to establish the range of values of the interaction strength that favors a crystal structure made of atomic nano-clusters, and to study both the static properties of the supersolid structure and its excitation spectrum. Unlike the case of 2D soft-core boson supersolids where both mean field [18, 23] and Monte Carlo calculations [22, 23] have been performed, no ab-initio results exist for the three-dimensional system which may serve as a benchmark to assess the accuracy of the present calculations. For this reason we have performed some preliminary path integral ground state (PIGS) Monte Carlo simulations of the 3D system to substantiate our mean field findings. On the basis of the excellent agreement between mean-field and ab initio methods for the 2D system of soft-core bosons [23], both for the critical point of the superfluid-supersolid transition and for the dispersion relations of collective excitations, and on the preliminary results of our PIGS simulations, we expect that our calculations are accurate enough to provide a reliable picture which might be helpful to understand the properties of this prototype system in preparation of experiments in the near future.

In the following, we try to understand some basic questions such as: a) which is the ground state structure of the supersolid? b) how large a number of particles per unit cell is requested to realize the supersolid, given the interaction parameters? c) how does the superfluid fraction in the supersolid phase depend on the interaction parameter? d) is it possible to have coexistence of the supersolid and the superfluid? and what is the solid-liquid interface energy?

II. METHODS AND CALCULATIONS

We consider N bosonic atoms of mass M interacting through a pair-potential V(r) represented by a simple "soft-sphere" interaction:

$$V(r) = V_0 \Theta(R_c - r) \tag{2}$$

where V_0 and R_c are the height and width of the potential (Θ is the Heavyside step function and r is the length of the 3D vector $\mathbf{r} \equiv (\mathbf{x}, \mathbf{y}, \mathbf{z})$). While the above

interaction does not occur naturally the so-called Rydberg dressing [14, 17] of atomic BEC constitutes, as described in the previous Section, a promising approach for an experimental realization of this interaction. The actual potential in Rydberg-blockade interaction has an additional long-range Van der Waals repulsive tail which decays rapidly with distance, rather than being abruptly cut as in the simpler soft-core model. However, on the basis of numerical simulations [21, 23] it has been shown that the formation of supersolid is largely insensitive to the actual shape of the repulsive interaction, provided it produces a roton minimum in the dispersion relation.

We assume that all the N atoms of the system are in a Bose-Einstein condensate described by the wavefunction $\Phi(\mathbf{r})$. The energy of the system at the mean-field level is thus expressed by the functional

$$E[\Phi, \nabla \Phi] = \frac{\hbar^2}{2M} \int |\nabla \Phi(\mathbf{r})|^2 d\mathbf{r} + \frac{1}{2} \int \int V(|\mathbf{r} - \mathbf{r}'|) |\Phi(\mathbf{r})|^2 |\Phi(\mathbf{r}')|^2 d\mathbf{r} d\mathbf{r}'$$
(3)

Functional minimization of the above energy leads to the following Euler-Lagrange equation

$$\hat{H}\Phi \equiv \left[-\frac{\hbar^2}{2M} \nabla^2 + \int V(|\mathbf{r} - \mathbf{r}'|) |\Phi(\mathbf{r}')|^2 d\mathbf{r}' \right] \Phi(\mathbf{r}) = \mu \Phi(\mathbf{r})$$
(4)

where \hat{H} is defined by the terms in square brackets, and where μ is a Lagrange multiplier whose value is determined by the normalization condition $\int |\Phi(\mathbf{r})|^2 d\mathbf{r} = N$.

This equation will be solved numerically, as explained in the next Section, to yield the lowest energy state Φ describing the condensate in the ground-state.

Upon scaling lengths by R_c and energies by \hbar^2/MR_c^2 , and once the wave function is scaled as $\Phi/\sqrt{\rho}$, the above model has a single dimensionless parameter Λ that determine the solutions of Eq.(4). In 3D it reads:

$$\Lambda \equiv M \rho V_0 R_c^5 / \hbar^2. \tag{5}$$

 Λ can be varied, e.g., by changing the density of the system, although from the experimental point of view the optimal control parameter is V_0 , which can be easily varied since it depends strongly on the quantum number of Rydberg states [14, 17].

The mean-field equation (4) is valid when the quantum fluctuations in the region inside the range of the potential are relatively small, which occurs when the average particle number inside this range is large. In the present case this means that the number of particles within each cluster forming the solid structure should be large. However, as shown in Ref.[23], the mean-field description turns out to be rather accurate even when this number is relatively small (of the order of a few atoms).

In order to compute the excitation spectrum, we make the usual Bogoliubov transformation to a Hamiltonian describing a collection of non-interacting quasi-particles for which the condensate is the vacuum:

$$\Psi(\mathbf{r},t) = e^{-i\mu t/\hbar} [\Phi(\mathbf{r}) + u_{n,\mathbf{k}}(\mathbf{r})e^{-i\omega t} - v_{n,\mathbf{k}}^*(\mathbf{r})e^{i\omega t}]$$
(6)

where $u_{n,\mathbf{k}}(\mathbf{r})$ and $v_{n,\mathbf{k}}(\mathbf{r})$ are the wavefunctions of the excitation mode with band index n and wavevector \mathbf{k} and $\Phi(\mathbf{r})$ is the solution of Eq. (4). Substituting this form into the time-dependent Schrodinger equation:

$$i\hbar \frac{\partial}{\partial t} \Psi(\mathbf{r}) = \left[-\frac{\hbar^2}{2M} \nabla^2 + \int V(|\mathbf{r} - \mathbf{r}'|) |\Psi(\mathbf{r}')|^2 d\mathbf{r}' \right] \Psi(\mathbf{r})$$
(7)

associated with the Hamiltonian in Eq.(4) and keeping only terms linear in the functions u, v, one obtains the following coupled equations:

$$\hbar\omega u_{n,\mathbf{k}}(\mathbf{r}) = \left[-\frac{\hbar^2}{2M} \nabla^2 - \mu + \int V(|\mathbf{r} - \mathbf{r}'|) |\Phi(\mathbf{r}')|^2 d\mathbf{r}' \right] u_{n,\mathbf{k}}(\mathbf{r})$$

$$+\Phi(\mathbf{r}) \int V(|\mathbf{r} - \mathbf{r}'|) \Phi^*(\mathbf{r}') u_{n,\mathbf{k}}(\mathbf{r}') d\mathbf{r}'$$

$$-\Phi(\mathbf{r}) \int V(|\mathbf{r} - \mathbf{r}'|) \Phi(\mathbf{r}') v_{n,\mathbf{k}}(\mathbf{r}') d\mathbf{r}'$$

$$-\hbar\omega v_{n,\mathbf{k}}(\mathbf{r}) = \left[-\frac{\hbar^2}{2M} \nabla^2 - \mu + \int V(|\mathbf{r} - \mathbf{r}'|) |\Phi(\mathbf{r}')|^2 d\mathbf{r}' \right] v_{n,\mathbf{k}}(\mathbf{r})$$

$$-\Phi^*(\mathbf{r}) \int V(|\mathbf{r} - \mathbf{r}'|) \Phi^*(\mathbf{r}') u_{n,\mathbf{k}}(\mathbf{r}') d\mathbf{r}'$$

$$+\Phi^*(\mathbf{r}) \int V(|\mathbf{r} - \mathbf{r}'|) \Phi(\mathbf{r}') v_{n,\mathbf{k}}(\mathbf{r}') d\mathbf{r}'$$
(8)

We expand the (real) function Φ and the complex functions u,v in the Bloch form appropriate to a periodic system:

$$\Phi(\mathbf{r}) = \sum_{\mathbf{G}} \Phi_{\mathbf{G}} e^{i\mathbf{G} \cdot \mathbf{r}} \tag{9}$$

$$u_{n,\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} \sum_{\mathbf{G}} u_{\mathbf{k}+\mathbf{G}}^{(n)} e^{i\mathbf{G}\cdot\mathbf{r}}$$
 (10)

$$v_{n,\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} \sum_{\mathbf{C}} v_{\mathbf{k}+\mathbf{G}}^{(n)} e^{i\mathbf{G}\cdot\mathbf{r}}$$
 (11)

In the above expansions, the **G**-vectors are the reciprocal lattice vector appropriate to the space symmetry of the cluster-crystal structure. By making the above substitutions in Eq.(8) (and omitting the band index n for clarity) one gets:

$$\left[\frac{\hbar^{2}}{2M}(\mathbf{k}+\mathbf{G})^{2}-\mu-\hbar\omega\right]u_{\mathbf{k}+\mathbf{G}} + \sum_{\mathbf{G}',\mathbf{G}''}\Phi_{\mathbf{G}''-\mathbf{G}'}\Phi_{\mathbf{G}-\mathbf{G}''}\tilde{V}_{\mathbf{k}+\mathbf{G}''}u_{\mathbf{k}+\mathbf{G}'} + \sum_{\mathbf{G}',\mathbf{G}''}\Phi_{\mathbf{G}''-\mathbf{G}'}\Phi_{\mathbf{G}-\mathbf{G}''}\tilde{V}_{\mathbf{k}+\mathbf{G}''}u_{\mathbf{k}+\mathbf{G}'} - \sum_{\mathbf{G}',\mathbf{G}''}\Phi_{\mathbf{G}''-\mathbf{G}'}\Phi_{\mathbf{G}-\mathbf{G}''}\tilde{V}_{\mathbf{k}+\mathbf{G}''}v_{\mathbf{k}+\mathbf{G}'} = 0 - \left[\frac{\hbar^{2}}{2M}(\mathbf{k}+\mathbf{G})^{2}-\mu+\hbar\omega\right]v_{\mathbf{k}+\mathbf{G}} - \sum_{\mathbf{G}'}\tilde{U}_{\mathbf{G}-\mathbf{G}'}v_{\mathbf{k}+\mathbf{G}'} - \sum_{\mathbf{G}',\mathbf{G}''}\Phi_{\mathbf{G}''-\mathbf{G}'}\Phi_{\mathbf{G}-\mathbf{G}''}\tilde{V}_{\mathbf{k}+\mathbf{G}''}v_{\mathbf{k}+\mathbf{G}'} + \sum_{\mathbf{G}',\mathbf{G}''}\Phi_{\mathbf{G}''-\mathbf{G}'}\Phi_{\mathbf{G}-\mathbf{G}''}\tilde{V}_{\mathbf{k}+\mathbf{G}''}u_{\mathbf{k}+\mathbf{G}'} = 0 \right] (12)$$

 $\tilde{V}_{\mathbf{q}}$ is the Fourier transform of the soft-core interaction:

$$\tilde{V}_{\mathbf{q}} \equiv \int V(r)e^{i\mathbf{q}\cdot\mathbf{r}}d\mathbf{r} = 4\pi V_0 R_c^2 j_1(qR_c)/q$$
 (13)

where $j_1(x) = \sin(x)/x^2 - \cos(x)/x$ is the spherical Bessel function of the first kind.

The quantities $U_{\mathbf{G}}$ in Eqns.(12) are defined through

$$\int V(|\mathbf{r} - \mathbf{r}'|) |\Phi(\mathbf{r}')|^2 d\mathbf{r}' = \sum_{\mathbf{G}} \tilde{U}_{\mathbf{G}} e^{i\mathbf{G} \cdot \mathbf{r}}$$
(14)

It is useful to introduce the following matrices (with dimensions $(n_r^3 \times n_r^3)$, where n_r is the real space mesh used to integrate the stationary equation, see the following Section):

$$\mathbf{A}_{\mathbf{G},\mathbf{G}'} \equiv \delta_{\mathbf{G},\mathbf{G}'} \left[\frac{\hbar^2}{2M} (\mathbf{k} + \mathbf{G})^2 - \mu \right] + \tilde{U}_{\mathbf{G}-\mathbf{G}'} + \sum_{\mathbf{G}''} \Phi_{\mathbf{G}''-\mathbf{G}'} \Phi_{\mathbf{G}-\mathbf{G}''} \tilde{V}_{\mathbf{k}+\mathbf{G}''}$$
(15)

$$\mathbf{B}_{\mathbf{G},\mathbf{G}'} \equiv -\sum_{\mathbf{G}''} \Phi_{\mathbf{G}''-\mathbf{G}'} \Phi_{\mathbf{G}-\mathbf{G}''} \tilde{V}_{\mathbf{k}+\mathbf{G}''}$$
(16)

The system (12) can thus be written in the matrix form:

$$\begin{bmatrix} \mathbf{A} & \mathbf{B} \\ -\mathbf{B} & -\mathbf{A} \end{bmatrix} \begin{pmatrix} \mathbf{u} \\ \mathbf{v} \end{pmatrix} = \hbar \omega \begin{pmatrix} \mathbf{u} \\ \mathbf{v} \end{pmatrix}$$
 (17)

The excitation frequencies $\omega(\mathbf{k})$ can be determined from the solutions of the above non-Hermitian eigenvalue problem. This can be reduced to a non-Hermitian problem of *half* the dimension (thus largely reducing the computational cost of diagonalization) by means of a unitary transformation [27]:

$$(\mathbf{A} - \mathbf{B})(\mathbf{A} + \mathbf{B})|\mathbf{u} + \mathbf{v}\rangle = (\hbar\omega)^{2}|\mathbf{u} + \mathbf{v}\rangle$$
 (18)

If needed, one may calculate the separate \mathbf{u} , \mathbf{v} by properly combining the eigenvectors of Eq. (18) with those of the associated eigenvalue problem

$$(\mathbf{A} + \mathbf{B})(\mathbf{A} - \mathbf{B})|\mathbf{u} - \mathbf{v}\rangle = (\hbar\omega)^2 |\mathbf{u} - \mathbf{v}\rangle, \tag{19}$$

again of reduced dimensions.

In order to provide a quantitative benchmark for the mean field results presented here, we have performed also some preliminary path integral ground state (PIGS) Monte Carlo simulations [28]. PIGS is an $exact\ T=0$ K method that allows to obtain the ground state of a given microscopic Hamiltonian by projecting in imaginary time a trial wave function. The PIGS method is unbiased by the choice of such a trial wave function and the only inputs are the interparticle potential and the approximation for the imaginary time propagator [29]. Here we consider a system of N Bosons in a cubic box with periodic boundary conditions interacting via the soft-core

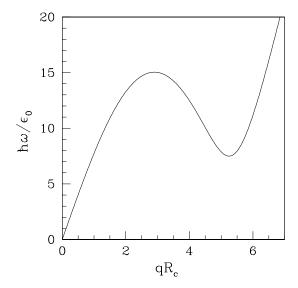


FIG. 1: Dispersion relation for the homogeneous system, for $\Lambda = 15.8$. Energies are expressed in units of $\epsilon_0 \equiv \hbar^2/(MR_c^2)$.

potential (2); we project a constant wave function and we consider the Suzuki pair approximation for the imaginary time propagator [29]. Despite the possibility of reducing the computational cost of PIGS simulation by choosing a trial wave function as close as possible to the ground state of the system, Quantum Monte Carlo simulations of the 3-dimensional system studied here are still much more computationally expensive than mean field methods. Encouraged by the consistency of our preliminary PIGS results with the mean field ones, we choose to perform only a limited number of calculations with the aim to give support to the present mean field study, leaving a systematic full Quantum Monte Carlo investigation of 3D soft Boson systems for a future work.

III. RESULTS AND DISCUSSION

In Fig.1 the dispersion relation (1) for the uniform liquid is shown for a value of the interaction strength $\Lambda=15.8$. Energies are expressed in units of $\epsilon_0=\hbar^2/(MR_c^2)$. For sufficiently large values of Λ , $\tilde{V}(q)$ has a negative contribution around $q_{rot}\sim 2\pi/R_c$ and a roton minimum appears. The roton gap decreases upon increasing Λ until a threshold value Λ_r is reached where Landau's critical velocity (defined as the tangent at the roton minimum) becomes zero, i.e. when

$$(\rho M/q)dV(q)/dq = -1/2 \tag{20}$$

This condition (closing of the roton gap) marks the onset of a roton instability at which density modulations may develop without energy cost, as discussed in the Introduction. The condition (20) can be recast in a form involving only the adimensional strength parameter Λ , giving

$$\Lambda_r = 21.71 \tag{21}$$

This value could be achieved, e.g., with the following choice of parameters: $^{87}{\rm Rb}$ condensate of density $2.2 \times 10^{-11}\,a_0^{-3}$, with $R_c=6000\,a_0$ and $V_0=183\,nK$.

As shown in the following, and similarly to what occurs in the 2-dimensional case [15, 21, 23], the homogeneous liquid phase destabilizes well before the roton instability condition (21) is reached, spontaneously converting the liquid phase into an ordered solid-like structure ("cluster crystal").

A. Ground-state properties

We have numerically solved Eq.(4) by propagating it in imaginary time, i.e. by solving the equation

$$\frac{\partial \Phi}{\partial t} + (\hat{H} - \mu)\Phi(\mathbf{r}) = 0 \tag{22}$$

The wave function $\Phi(\mathbf{r})$ is represented on a threedimensional uniform mesh in real space, with periodic boundary conditions imposed on the system. By studying the convergence in energy of the solution with increasing number of points in the mesh, we verified that a relatively coarse grid made of n_r^3 points with $n_r = 20$ is sufficient to accurately describe $\Phi(\mathbf{r})$. To compute the spatial derivatives appearing in the previous equation, we used a 11-point finite-difference formula [30]. The convolution integral in the potential energy term of Eq.(4) is efficiently evaluated in reciprocal space by using Fast Fourier transform techniques.

Depending on the value of Λ , the lowest energy structure described by $\Phi(\mathbf{r})$ is either a uniform liquid or a structured system with long-range order. For sufficiently large values of Λ , in fact, the system spontaneously breaks the translational invariance leading to the appearance of a crystalline phase made of individual clusters of atoms arranged in an ordered structure. This phase, as discussed in the following, can exhibit a supersolid behavior by decoupling a superfluid component from the crystalline structure.

We have studied the relative stability of the liquid vs. solid phase as a function of the interaction parameter Λ . We find that, upon increasing Λ , once a critical value

$$\Lambda_{ss} = 15.2 \tag{23}$$

is reached and well before the roton instability (i.e. the disappearance of the roton gap) occurs, the structure spontaneously converts into an ordered arrangement of

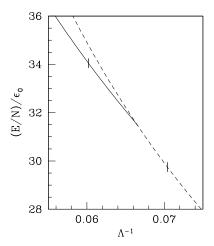


FIG. 2: Energy/atom for the liquid and solid phases, shown as a function of Λ^{-1} . Energies are in units of $\epsilon_0 \equiv \hbar^2/(MR_c^2)$. Dashed line: liquid phase; solid line: FCC cluster crystal. The two vertical ticks show the two values of Λ at coexistence as obtained by a double-tangent construction (see the text).

clusters, each containing a number of atoms that increases as Λ is further increased.

To assess the relative stability of different possible solid phases we have studied different crystal structures (SC. FCC and HCP) and optimized, for each structure, the associated lattice parameter for a given value of Λ . We find that face-centered-cubic (FCC) and hexagonal-closepacked (HCP) ordering are almost degenerate in energy, with the FCC structure only slightly favored. We have checked, by using a finer mesh than the one we eventually used to compute the excitation spectrum, that the FCC structure is indeed always (slightly) favored over the HCP one in a wide range of values of the interaction parameter Λ . The FCC arrangement is the one produced also by PIGS computations. This is not surprising, due to the short range nature of the interactions involved, since FCC and HCP structures differ only in the second nearestneighbor atoms arrangement. This is similar to what occurs in the three-dimensional boson system interacting through the Rydberg blockade type interaction [14] which contains also a long range repulsion, where again FCC ordering has the lowest possible energy per atom.

We compare the energy per atom (expressed in units of $\epsilon_0 \equiv \hbar^2/(MR_c^2)$) of the liquid and solid phases in Fig.2. The liquid-solid transition occurs at $\Lambda_{ss} = 15.2$. For higher values, the solid structure is always favored. We plot ϵ as a function of Λ^{-1} rather than of Λ for a reason that will be clarified in the next Subsection. Our preliminary PIGS simulations locate the liquid-solid transition between $\Lambda = 15.0$ and $\Lambda = 15.5$, in good agreement with

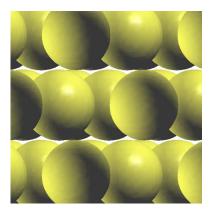


FIG. 3: (Color online) Equilibrium equidensity plot for the $\Lambda=15.8$ FCC cluster solid. Each cluster contains approximately 10 atoms. The surface of equal density is drawn at the value $\rho/2$.

our mean-field results.

Note that the energy per atom is an increasing function of Λ (i.e. of the density, for a given V_0 and R_c), which is an obvious consequence of the repulsive character of the interparticle interaction: therefore all the phases discussed here, with a finite density, are meaningful only in confined systems, a condition which is certainly realized in cold gases experiments.

As an example, in Fig.3 we show the resulting 3-dimensional density plot for the optimized FCC cluster solid obtained with $\Lambda=15.8$. For clarity, we also show in Fig.4 the density profile by means of contour lines in the (100) plane. The cell shown is the conventional cubic cell for FCC structures, containing four equivalent sites. Each cluster contains about 10 atoms. The equilibrium interatomic distance is $d \sim \rho^{-1/3} = 0.595\,R_c$, while the cluster-cluster distance is $d_c = 1.465\,R_c$. For comparison, the characteristic wavelength associated with the roton minimum in the dispersion relation for the homogeneous system is $2\pi/q_{min} = 1.208\,R_c$.

By increasing Λ (i.e. by increasing the atomic density at constant V_0 and R_c), the number of atoms inside each cluster grows, while the overlap between adjacent cluster decreases, suggesting that the superfluid fraction (which is associated to a global coherence maintained by hopping of atoms between adjacent clusters) is also decreasing with increasing the density. This will be confirmed by the calculation of the superfluid fraction, as shown in the following.

The liquid-solid transition which occurs at $\Lambda_{ss}=15.2$ is first-order in character, being accompanied by a discontinuity in the derivative of the energy per atom. The nature of the transition is better appreciated by looking at the occupation fraction of the lowest finite momentum, i.e. the lowest G-vector component $\Phi_{\mathbf{G}}$ of the wavefunction $\Phi(\mathbf{r})$ corresponding to the FCC reciprocal lattice vector $4\pi/a_{ss}$ ($a_{ss}=2.07\,R_c$ being the equilibrium lattice constant of the cubic structure shown in Fig.(4)). Fig.5 clearly shows the first-order jump in $\Phi_{\mathbf{G}}$ as the

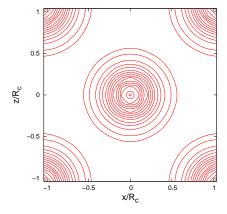


FIG. 4: Equilibrium density contour map for the $\Lambda=15.8$ FCC cluster solid in the (100) plane, shown in the conventional cubic cell containing four clusters. Each cluster contains approximately 10 atoms. The inner contour line is drawn at the value 16ρ ($\rho=N/\Omega$ being the density of the liquid phase), while the external contour line are drawn at the value $\rho/2$.

transition value Λ_{ss} is reached.

From the dependence of the energy per atom $\epsilon \equiv E/N$ on the atomic density one can compute, e.g., the inverse of thermodynamic compressibility, as

$$\kappa^{-1} = \rho \Lambda \frac{\partial \mu}{\partial \Lambda} \tag{24}$$

where the chemical potential μ is computed in turn from the data shown in Fig.(2) as:

$$\mu = \frac{\partial E}{\partial N} = \epsilon + \rho \frac{\partial \epsilon}{\partial \rho} = \epsilon(\Lambda) + \Lambda \frac{\partial \epsilon}{\partial \Lambda}$$
 (25)

We show the resulting inverse compressibility in Fig.(6), for $\rho=2.2\times 10^{-11}a_0^{-3}.$ A drop occurs at the liquid-solid transition, showing that the cluster solid is remarkably softer than the liquid phase at the same density. The inverse compressibility is related to the low-q average sound velocity c through the relation

$$\kappa^{-1} = \rho M^* c^2 \tag{26}$$

where M^* is some effective mass due to the periodic potential. The drop in Fig.(6) is thus associated to an overall softening of the sound velocity. As we will show in the following, this behavior is associated with the presence, in addition to the usual phonon modes, of a soft Bogoliubov mode which is the signature of the supersolid phase.

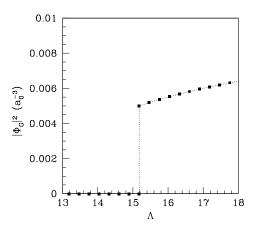


FIG. 5: Occupation fraction of the lowest finite momentum state (see the text).

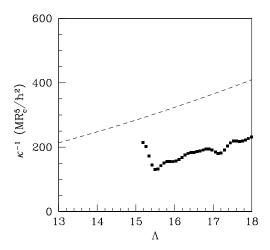


FIG. 6: Inverse compressibility for the case $\rho=2.2\times 10^{-11}a_0^{-3}$: homogeneous liquid (dashed line); cluster crystal (filled squares).

B. Superfluid fraction

Because of its condensate character, the cluster crystal described above can naturally exhibit superfluidity, resulting in a nonzero Non-Classical Rotational Inertia (NCRI) [3]. Numerical proof that this is indeed the case for soft-core bosons has been provided in Ref.[31]. Equivalently [32], one can instead consider the momentum of

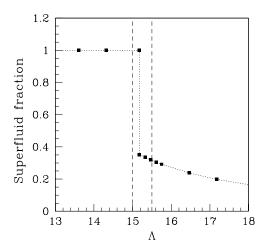


FIG. 7: Calculated superfluid fraction as a function of Λ . The vertical dashed lines indicate the Λ range for the liquid-solid transition as obtained by *exact* PIGS Monte Carlo method.

the crystal under a Galilean boost ${\bf v}$ and solve the associated time-dependent Schrodinger equation in the comoving frame of reference:

$$i\hbar \frac{\partial}{\partial t} \Psi = \left[\hat{H} + i\hbar \mathbf{v} \cdot \nabla \right] \Psi \tag{27}$$

By computing the linear momentum of the system one can define the superfluid fraction f^{ss} as the tensor [33]

$$f_{ik}^{ss} = \delta_{ik} - \lim_{|v| \to 0} \frac{1}{N} \frac{\partial P_i}{\partial v_k}$$
 (28)

where ${\bf P}=-(i\hbar/2)\int (\Psi^*\nabla\Psi-\Psi\nabla\Psi^*)d{\bf r}$ is the total momentum.

We show in Fig.(7) our calculated values for the diagonal part of this tensor along the direction of the velocity boost \mathbf{v} . A sudden, finite drop from 1 to ~ 0.4 occurs at the liquid-supersolid transition, while the superfluid fraction decreases in a monotonic way as Λ is further increased. We mention that for the supersolid triangular structure realized in 2-dimensions by a system of softcore bosons [23], the corresponding jump, computed using Path Integral Monte Carlo methods, is from 1 to ~ 0.6 . For comparison, we show with dashed lines the interval of Λ values where our preliminary PIGS Monte Carlo calculations allow to locate the liquid-solid transition.

Our calculated superfluid fraction are probably overestimating to some extent the actual superfluid fraction, since mean-field methods tend to overestimate the superfluid fraction compared to the predictions of *ab initio* Monte Carlo methods [34], mainly due to the neglect of

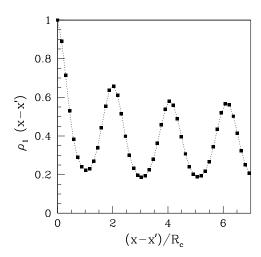


FIG. 8: One-body density matrix ρ_1 computed with PIGS at $\Lambda = 15.72$. x is along the (100) direction.

fluctuations that tend to suppress superfluidity. However, in 3D the effect of quantum fluctuations should be less than they are, e.g., in 2-dimensional system of bosons[23].

The PIGS method does not allow a direct evaluation of the superfluid fraction, so we cannot directly compare with our mean field results; however, PIGS provides an estimate of the condensate fraction n_0 [29] whose finite value, at T = 0 K, is a sufficient condition for NCRI [35]. The condensate fraction n_0 is obtained as the large distance limit of the one-body density matrix ρ_1 . We have computed ρ_1 for $\Lambda = 15.72$, i.e. very close to the liquidsolid transition, and we find that the large distances tail of the one-body density matrix is converging to a finite value, with large superimposed oscillations. Such oscillations are a typical signature of solid phase, thus confirming, on one hand, the crystalline structure of the system, but on the other hand they are so large to prevent us from a precise evaluation of n_0 in systems of tractable sizes. We show our computed values for ρ_1 in Fig.(8).

Similarly to what happens in 2D [33] the superfluid fraction decreases exponentially with the square root of Λ : $log f^{ss} \propto -\Lambda^{-1/2}$ very close to the transition; for higher values of Λ it follows instead (approximately) a power law: $f^{ss} \propto \Lambda^{-\gamma}$.

C. Supersolid-superfluid coexistence

We show here that when Λ is greater than Λ_{ss} , such that a supersolid phase is expected, but close to Λ_{ss} , the spontaneous formation of interfaces between solid and

liquid patches might be energetically favored over the realization of a single extended solid phase. A similar conclusion was drawn in Ref.[14] based on numerical calculations, where coexistence between small crystallites and liquid embedding them was observed in a system of bosons interacting through a soft-core Rydberg blockade interaction for values of the coupling strength very close to the transition point.

The possibility of liquid-solid coexistence in the system studied here is proved by a double-tangent construction on the data shown in Fig.2. We recall that a double-tangent construction on a plot where the energy per atom ϵ is drawn as a function of $(1/\rho)$ for the two phases, is equivalent to imposing the equality of pressure and chemical potential, i.e. the conditions for the thermodynamic coexistence of the two phases. The two vertical ticks in Fig.2 show the values of Λ characterizing the two coexisting phases as obtained by such construction, $\Lambda_s = 16.6$ and $\Lambda_l = 14.2$ for the solid and liquid phase, respectively.

To verify this prediction we have explicitly realized such two-phase system. We show in Fig.(9) the (100) planar interface, which we have computed by solving Eq.(4) with $\Lambda=15.3$ (i.e. just above Λ_{ss}) in a slab geometry with a fixed total number of atoms N in a cell of total volume Ω (such that $\rho=N/\Omega$), separating a cluster solid phase (upper part of the Figure) from a coexisting liquid phase. For the particular interface shown in Fig.(9) the average density in the solid phase (far from the interface) $\rho_{s,c}$, and the density in the liquid phase (far from the interface) $\rho_{l,c}$ correspond to the values of Λ_s and Λ_l as obtained by the double tangent construction described above

One can estimate the width of the liquid (L_l) and solid (L_s) region as $L_l = (L_s + L_l)(\rho - \rho_{s,c})/(\rho_{l,c} - \rho_{s,c})$, $L_s = L - L_l$.

The interface tension can thus be calculated (the factor 2 takes into account the presence, in a slab geometry, of two solid-liquid interfaces)

$$\sigma = [E - (E_s + E_l)]/(2A) \tag{29}$$

where A is the area of the transverse section of the supercell parallel to the interface, E is the total energy of the configuration shown in Fig.(9) and $E_s = \epsilon(\rho_{s,c})L_s$, $E_l = \epsilon(\rho_{l,c})L_l$ ($\epsilon = E/N$ being the energy per atom shown in Fig.(2)).

In this way we find the value $\sigma_{\{100\}} = 300 \, M R_c^4 / \hbar^2$ for the interfacial energy, i.e. for the energy cost to create a (100) solid-liquid interface of unit area.

D. Bogoliubov excitation spectrum

The excitation spectrum of the supersolid phase described above will be discussed next. As already stated in the Introduction, a consequence of the breaking of a global gauge symmetry is the emergence of a new Goldstone boson, i.e. a gapless mode in the excitation spectrum (which we will call Bogoliubov mode in the follow-

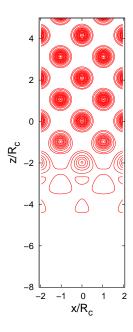


FIG. 9: (100) solid-liquid interface for $\Lambda=15.3$. Only a portion of the supercell used in the calculation is displayed for clarity.

ing), in addition to the usual phonon branches. To check for the appearance of such a mode, we have computed the frequency spectrum of the supersolid structure described in the previous Section by numerical diagonalization of the system (18).

Our results are shown in Fig.10 where $\omega(\mathbf{k})$ is plotted along symmetry lines in the 1^{st} Brillouin Zone (BZ), as shown in the inset of the figure. At low k-values we find four modes: three correspond to the usual phonon bands (one longitudinal and two transverse modes), while the fourth, softer mode is associated with the broken gauge symmetry. These modes can be seen, e.g., along the Γ -X direction where the first four small-q modes from lower to higher frequencies in Fig.(10) are the Bogoliubov mode, the (doubly degenerate) transverse phonon mode and the longitudinal phonon mode, respectively.

To unambiguously make the above assignments we computed [22] the effective potential corresponding to the cluster solid structure, $V_{eff}(\mathbf{r}) = \int V(|\mathbf{r} - \mathbf{r}'|) |\Phi(\mathbf{r}')|^2 d\mathbf{r}'$ (see Eq.(4)) and defined an effective force constant by fitting V_{eff} , close to its minima, with a quadratic curve. We then obtained the phonon frequencies by solving the dynamical matrix for a harmonic crystal with such force constants and an atomic mass equal to that of a cluster (i.e. $\sim 10\,M$). We find in this way two (degenerate) transverse and one longitudinal modes, whose dispersion agree within 10-15% at low q-values with the corresponding dispersion curves shown in Fig.(10).

The local density and phase fluctuations for a given \mathbf{k}

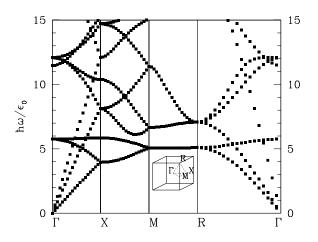


FIG. 10: Calculated excitation dispersion along symmetry lines of the cubic lattice 1^{st} Brillouin zone. Energies are expressed in units of $\epsilon_0 \equiv \hbar^2/(MR_c^2)$.

value and a given band n are provided by [23]

$$\Delta \rho_{n,\mathbf{k}}(\mathbf{r}) = |u_{n,\mathbf{k}}(\mathbf{r}) - v_{n,\mathbf{k}}(\mathbf{r})|^2$$

$$\Delta \phi_{\mathbf{k}+\mathbf{G}}(\mathbf{r}) = |u_{n,\mathbf{k}}(\mathbf{r}) + v_{n,\mathbf{k}}(\mathbf{r})|^2$$
(30)

By studying the spatial distribution of both density and phase fluctuations we find that, while the longitudinal phonon branch contributes mainly to density fluctuations $\Delta \rho_{\mathbf{k}+\mathbf{G}}$, the low-k Bogoliubov mode, which is also longitudinal in nature, contributes mainly to the phase fluctuations $\Delta \phi_{\mathbf{k}+\mathbf{G}}$, being associated with the superfluid response. These results parallel the similar findings in the 2-dimensional triangular supersolid phase studied in Ref.[23].

From the calculated excitation spectrum $\omega(\mathbf{k})$, one may obtain the density of states (DOS) as:

$$D(\omega) = \sum_{\mathbf{k}} \delta(\omega - \omega(\mathbf{k})) \tag{31}$$

where the summation is restricted to the 1^{st} BZ of the crystal. Fig.(11) shows our calculated DOS. To compute it we have used a uniform mesh of 82 points within the irreducible part of the 1^{st} BZ (shown with dotted lines in the inset of Fig.(10)), corresponding to a sampling of 4032 points in the whole 1^{st} BZ.

One can recognize two main peaks in the DOS: the one at low frequencies (below $\sim 10\,\epsilon_0$), comes mainly from the zone-edge Bogoliubov soft mode, while the other (above $\sim 20\,\epsilon_0))$ comes from the zone-edge longitudinal phonon modes.

The two curves (solid and dotted lines) shown in Fig.(11) correspond to two different values of Λ , i.e.

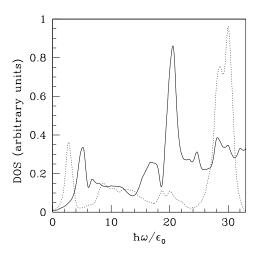


FIG. 11: Calculated Density of States for two different values of Λ . Solid line: $\Lambda=15.8$, dotted line: $\Lambda=21$.

 $\Lambda=15.8$ and $\Lambda=21$, respectively. We notice that by increasing Λ (e.g. by increasing density at constant V_0 and R_c) the phonon-like excitation peak moves to higher frequency because the particles get more localized around the FCC lattice sites, making the crystal stiffer, whereas the lower peak, associated with the Bogoliubov mode, shifts to lower frequencies, reflecting the loss of superfluid fraction.

IV. CONCLUSIONS

Quantum solid of clusters might be the prototypical system to realize and study the supersolid phase of matter. The system spontaneously breaks the translational invariance leading to the appearance of a crystalline phase of individual superfluid droplets governed by a global macroscopic wavefunction. This system can exhibit supersolid behavior by decoupling a superfluid component from the crystalline structure.

Using mean-field approach, we have carried out a numerical study of the structure and excitation spectrum of a soft-core model for a supersolid. We have computed the lowest energy structure: for values of the interaction

parameter Λ smaller than $\Lambda_{ss}=15.2$ the ground state is a uniform, superfluid phase, while for larger values it is a crystalline phase with FCC symmetry. Each unit cell contains clusters of atoms whose number increases with the system density, for a given value of the interaction parameter.

Our results for the critical value of Λ are in quite good agreement with exploratory PIGS Monte Carlo results. Due to the high computational cost of the *exact* Quantum Monte Carlo simulation, the present results are intended to be precursory of a full ab-initio study of the 3D softcore model that we leave for a future work.

We have also found a range of values of the interaction parameter which would allow coexistence of the two (superfluid and supersolid) phases, and calculated the corresponding interface tension.

We have computed the supersolid excitation spectrum within the Bogoliubov theory. Besides the usual low-k phonon-like excitations a new gapless, softer mode associated with the presence of a finite superfluid fraction appears, whose velocity decreases with increasing interaction parameter. The presence of this extra mode signals the breaking of gauge symmetry in the crystalline phase. We have calculated how the fraction of superfluid density varies as a function of the interaction parameter, finding a discontinuous drop from 1 to $\simeq 0.4$ at Λ_{ss}

In addition to the PIGS calculations mentioned above, the results of our calculations should provide a useful guide for experimental measurements. As proposed in Ref. [36] in fact, direct experimental measurements of the quasi-particle excitation of a condensate system are possible in principle by applying weak harmonic perturbation to the trapping potential at some probe frequency at the end of the cooling cycle and then probing the condensate shape by allowing the condensate to expand ballistically. By repeating the measurement with an incremented value of the probe frequency until the maximum distortion of the condensate is found relative to the case where no perturbation is applied, one determines the resonance lines. Another possible experimental way to probe superfluidity in cold gas systems is represented by Bragg scattering [37].

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E.P.Gross, Phys. Rev. 106, 161 (1957).

^[2] G.V.Chester, Phys. Rev. A 2, 256 (1970)

^[3] A.J.Leggett, Phys. Rev. Lett. **25**, 1543 (1970).

^[4] E.Kim and M.H.W. Chan, Nature 427, 225 (2004); Science 305, 1941 (2004).

^[5] D.Y.Kim and M.H.W. Chan, Phys. Rev. Lett. 109, 155301 (2012).

^[6] M.Boninsegni and N.Prokovev, Rev. Mod. Phys. 84, 759-776 (2012).

^[7] M.Boninsegni, J. Low Temp. Phys. 168, 137 (2012).

- [8] D.Kirzhnits and Y.Neopomnyashchii, JETP 59, 2203 (1970).
- [9] L.P.Pitaevskii, Pis'ma Zh. Eksp. Teor. Fiz. 39, 423 (1984) [JETP Lett. 39, 511 (1984)].
- [10] F.Ancilotto, F.Dalfovo, L.P.Pitaevskii and F.Toigo, Phys. Rev. B 71, 104530 (2005).
- [11] M.Kunimi and Y.Kato, Phys. Rev. 86, 060510 (2012).
- [12] Y.Pomeau and S.Rica, Phys. Rev. Lett. 72, 2426 (1994).
- [13] M.Rossi, E.Vitali, L.Reatto and D.E.Galli, Phys. Rev B 85, 014525 (2012).
- [14] N.Henkel, R.Nath and T.Pohl, Phys. Rev. Lett. 104, 195302 (2010).
- [15] F.Cinti et al., Phys. Rev. Lett. **105**, 135301 (2010).
- [16] P.Schauss et al., Nature 491, 87 (2012).
- [17] G.Pupillo et al., Phys. Rev. Lett. **104**, 223002 (2010).
- [18] C.Josserand, Y.Pomeau and S.Rica, Phys. Rev. Lett. 98, 195301 (2007).
- [19] B.Spivak and S.A.Kivelson, Phys. Rev. B 70, 155114 (2004).
- [20] F.Cinti, T.Macri, W.Lechner, G.Pupillo and T.Pohl, arXiv:1302.4576v1 [cond-mat.quant-gas] 19 Feb 2013.
- [21] S.Saccani, S.Moroni, M.Boninsegni, Phys. Rev. B 83, 092506 (2011).
- [22] S.Saccani, S.Moroni, M.Boninsegni, Phys. Rev. Lett. 108, 175301 (2012).
- [23] T.Macri, F.Maucher, F.Cinti and T.Pohl, Phys. Rev. A 87, 061602(R) (2013).

- [24] X.Li, W.Liu and C.Lin, Phys. Rev. A 83, 021602 (2011).
- [25] P.Mason, C.Josserand and S.Rica, Phys. Rev. Lett. 109, 045301 (2012).
- [26] I.L.Kurbakov, Y.E.Lozovik, G.E.Astrakharchik and J.Boronat, Phys. Rev. B 82, 014508 (2010).
- [27] M.Nooijen and R.J.Bartlett, J. Chem. Phys. 106, 6449 (1997).
- [28] A.Sarsa, K.E.Schmidt and W.R.Magro, J. Chem. Phys. 113, 1366 (2000).
- [29] M.Rossi, M.Nava, L.Reatto and D.E.Galli, J. Chem. Phys. 131, 154108 (2009).
- [30] M.Pi, F.Ancilotto, E.Lipparini and R. Mayol, Physica E, 24, 297 (2004).
- [31] A.Aftalion, X.Blanc and R.L.Jerrard, Phys. Rev. Lett. 99, 135301 (2007).
- [32] N.Sepulveda, C.Josserand, S.Rica, Phys. Rev. B 77, 054513 (2008).
- [33] N.Sepulveda, C.Josserand, S.Rica, Eur. Phys. J. B 78, 439 (2010).
- [34] A.van Otterlo and K.-H. Wagenblast, Phys. Rev. Lett. 72, 3598 (1994).
- [35] A.J.Leggett, Physica Fennica 8, 125 (1973).
- [36] P.A.Ruprecht, M.Edwards, K.Burnett and C.W.Clark, Phys. Rev. A 54, 4178 (1996).
- [37] R.Mottl et al., Science 336, 1570 (2012).