Two-dimensional Bose gas of tilted dipoles: anisotropic roton-maxon spectrum and instabilities

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We predict effect of the roton instability for two-dimensional weakly interacting gas of tilted dipoles in a single homogeneous quantum layer. Being typical for strongly correlated systems, the roton phenomena occur to appear in a weakly interacting gas. In contrast to a system of normal to a wide layer dipoles, breaking of the rotational symmetry for a system of tilted dipoles leads to the convergence of the condensate depletion even up to the threshold of the roton instability, with mean-field approach being valid. Predicted effects can be observed in a wide class of dipolar systems. We suggest to observe predicted phenomena for systems of ultracold atoms and polar molecules in optical lattices, and estimate optimal experimental parameters.

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I. INTRODUCTION

Bosonic systems with the dipole-dipole interaction are highly promising for observation of novel quantum phases and many-body phenomena [1–3]. Due to significant experimental progress, several realizations of these systems have been considered: ultracold atoms [4] with large magnetic dipole moment (e.g., chromium, dysprosium and erbium), for which Bose-Einstein condensation (BEC) has been recently demonstrated [5, 6]; ultracold clouds of ground state diatomic polar molecules [7] in electric fields (e.g., KRb and RbOH); Rydberg atoms in electric fields [14]; and excitons with spatially separated electrons and holes in semiconductor layers [15, 16]. These systems are well controllable via external fields. In particular, s-wave scattering length can be controlled via Feshbach resonances [17, 18].

The anisotropy and the region of attraction of the dipole-dipole interaction provide a set of interesting collective phenomena. In the limit of strongly correlated (classical) system of in-plane dipoles, the ground state of the system has the chain structure [1]; the 3D system of parallel dipoles has the chain structure as well [19]. In Ref. [20], the roton-maxon spectrum has been predicted for normal to a wide layer (pancake) dipoles. The key feature of dipolar BEC is the character of the excitation spectrum [21] similar to that in superfluid 4He [22].

Interesting structural properties emerge close to the threshold of the instability. The stability criteria is the non-negativity of square of the Bogoliubov spectrum

\[ \varepsilon_p^2 = \frac{\varepsilon_p^2}{2m} \left( \frac{p^2}{2m} + 2n_0 U(p) \right) \geq 0, \]  

(1)

where \( p \) is the momentum, \( m \) is the mass of dipoles, \( n_0 \) is the condensate density, \( U(p) \) is the Fourier transform of an interaction pseudopotential. On the one hand, the stability problem have inspired great progress in investigation of superfluidity [23], density waves [24], phonon collapse [2, 25], vortices [26], behavior of the system in optical lattices [27], traps [28] and presence of disorder [29]. Monte-Carlo simulations have predicted that a 2D gas of dipoles exhibits a quantum phase transition to a triangle crystal phase at zero temperature [30]. Special attention [31–33] has been paid to a supersolid phase [34].

On the other hand, the condensate depletion in the system diverges at the threshold of the roton instability [35]. In other words, condensate disappears before the spectrum reaches zero even at zero temperature. Consequently, both the threshold of the roton instability and supersolid phase are unattainable [36].

The considered system of normal to the layer dipoles is rotationally invariant. Therefore, actual question is how this invariance affects the system behavior and stability properties. Example of a system with broken rotational symmetry is BEC of tilted dipolar particles [37–44], where it is broken by an external field.

In the present work, we consider dilute one-component BEC gas of 2D tilted at angle \( \theta \) dipolar bosons in a qua-
imply the tight-confinement quantization, i.e., $\hbar^2/mz_0^2$ is sufficiently larger than other energy scales of the problem, e.g., the interaction energy (for details, see [45, 46]). At last, we assume the independence of layers formed by the lattice potential (see, Fig. 1a), i.e., we suppose that the interlayer tunneling is sufficiently small and the interlayer interaction of dipoles is screened. Under these assumptions, the 3D Hamiltonian of the system reads

$$\hat{H}_{3d} = \hat{H}_0 + \hat{H}_{\text{int}},$$

where

$$\hat{H}_0 = \int d\vec{r} \hat{\Psi}^\dagger(\vec{r}) \left( -\frac{\hbar^2}{2m} \Delta_3 + V(\vec{r}) - \mu_{3d} \right) \hat{\Psi}(\vec{r}),$$

$$\hat{H}_{\text{int}} = \frac{1}{2} \int d\vec{r} d\vec{r}' U(\vec{r}, \vec{r}', \theta) \hat{\Psi}^\dagger(\vec{r}) \hat{\Psi}^\dagger(\vec{r}') \hat{\Psi}(\vec{r}') \hat{\Psi}(\vec{r}).$$

Here, $\hat{\Psi}(\vec{r})$ is the 3D field operator, $\Delta_3$ is the 3D Laplace operator, $\mu_{3d}$ is the 3D chemical potential, $V(\vec{r}) = V(\vec{r}) + V_{\text{ext}}(z)$ is the external potential, $V(\vec{r})$ is the 2D confinement potential in thin layer plane, $V_{\text{ext}}(z) = (m\omega^2z^2)/2$ is the 1D confinement potential in the tight direction ($\omega$ is the oscillator frequency), and

$$U(\vec{r}, \vec{r}', \theta) = V_{dd}(\vec{r}, \vec{r}', \theta) + g_{3d} \delta(\vec{r} - \vec{r}')$$

is the interaction potential; $\vec{r} = \{r, z\}$ is the 3D vector and $r = |\vec{r}|$, $|\vec{r}'| = \sqrt{r^2 + z^2}$.

In a sufficiently thin layer, the motion in the tight direction is frozen at the lowest energy state of the confining trap. Thus, in the representation of the 3D field operator in the basis $\{\varphi_k^{(i)}(z)\}$ in the tight direction

$$\hat{\Psi}(\vec{r}) \approx \varphi^{(0)}_k(z) \hat{\Psi}(\vec{r}), \quad \hat{\Psi}(\vec{r}) = \int dz \varphi^{(0)}_k(z) \hat{\Psi}(\vec{r}),$$

we take into account only $k = 0$ term. Here, $\hat{\Psi}(\vec{r})$ is the effective 2D field operator, which satisfies the standard bosonic commutation relations. The eigenfunctions
\[ \varphi_k(z) \text{ and eigenenergies } \mathcal{E}_k \text{ are determined from the 1D Schrödinger equation:} \]
\[ \left( -\hbar^2 \frac{d^2}{2m \, dz^2} + V_{ii}(z) \right) \varphi_k(z) = \mathcal{E}_k \varphi_k(z). \]

By substituting (4) in (3), we find the effective 2D Hamiltonian for the thin-layer motion
\[ \hat{H}_{2d} = \int d\mathbf{r} \hat{\Psi}^*(\mathbf{r}) \left( -\frac{\hbar^2}{2m} \Delta_2 - \mu + V(\mathbf{r}) \right) \hat{\Psi}(\mathbf{r}) + \int d\mathbf{r} d\mathbf{r}' U_{2d}(\mathbf{r} - \mathbf{r}', \theta) \hat{\Psi}^*(\mathbf{r}) \hat{\Psi}(\mathbf{r}) \hat{\Psi}^*(\mathbf{r}') \hat{\Psi}(\mathbf{r}') \] (5)

with the effective 2D interaction potential
\[ U_{2d}(\mathbf{r} - \mathbf{r}', \theta) = \int d\mathbf{z} d\mathbf{z}' U(\mathbf{r} - \mathbf{r}', \theta) |\varphi_0(\mathbf{z}) \varphi_0(\mathbf{z}')|^2. \] (6)

Here, \( \Delta_2 \) is the 2D Laplace operator, \( \mu = \mu_{2d} - E_0^i \) is the chemical potential, and
\[ \mathcal{E}_0^i = \hbar \omega / 2, \quad \varphi_i(z) = \exp(-z^2 / 2a_i^2) / \sqrt{\sqrt{\pi} a_i}. \]

Thus, we obtain the Fourier transform of the effective interaction potential of 2D dipoles (6) in the Born approximation
\[ U_{2d}(\mathbf{p}, \theta) = g_s - g_d + U_h(\mathbf{p}) \sin^2 \theta + U_v(\mathbf{p}) \cos^2 \theta, \] (7)
where
\[ U_h(\mathbf{p}) = \frac{2a^2}{\hbar} \int_{-\infty}^{\infty} \frac{p_x dp_x}{p_x^2 + p_y^2 + p_z^2} \exp \left( -\frac{p_x^2 a_0^2}{2\hbar^2} \right), \]
\[ U_v(\mathbf{p}) = \frac{2a^2}{\hbar} \int_{-\infty}^{\infty} \frac{p_y dp_y}{p_x^2 + p_y^2 + p_z^2} \exp \left( -\frac{p_y^2 a_0^2}{2\hbar^2} \right), \]
with coupling constants
\[ g_s = \frac{2\sqrt{2\pi \hbar^2 a_s}}{m a_0}, \quad g_d = \frac{2\sqrt{2\pi \hbar^2 a_d}}{m a_0} = \frac{2\sqrt{2\pi \hbar^2}}{3a_0}. \]

For Bogoliubov spectrum square (1) with effective interaction potential (7), we find the following regimes on the phase diagrams (see, Fig. 2):
(i) Effective potential \( U_{2d}(\mathbf{p}) > 0 \) is positive for all momenta \( \mathbf{p} \). Hence, the square of the Bogoliubov spectrum \( \varepsilon^2 \) is positive for all momenta as well. In this case, the homogeneous phase is stable at an arbitrary density.
(ii) At low momenta, the effective potential \( U_{2d}(\mathbf{p}) < 0 \) is negative. Therefore, the square of the Bogoliubov spectrum \( \varepsilon^2 \) drops below the zero point at momenta below some critical one. This regime is known as phonon instability in respect to a long-wavelength collapse [2, 25], which can appear in system at an arbitrary density.
(iii) At low momenta, the effective potential \( U_{2d}(\mathbf{p}) > 0 \) is positive, but \( U_{2d}(\mathbf{p}) < 0 \) is negative for a certain momentum range. In this case, at the certain density, square of the Bogoliubov spectrum \( \varepsilon^2 \) touches zero point of energy at non-zero momentum. This regime corresponds to the threshold of the roton instability.

III. CONDENSATE DEPLETION

For normal to the layer (\( \theta = 0 \)), close to the threshold of the roton instability (i.e., at \( p \approx p_t \)), we have the following relation for the spectrum
\[ \varepsilon^2 \approx 1 \frac{d^2 p}{2 dp} (p - p_t)^2. \] (8)

Substituting (8) to the condensate depletion
\[ \frac{n - n_0}{n_0} = \int \frac{dp}{8\pi^2 \hbar^2 n_0} \frac{p^2/2m + U(\mathbf{p}) - \varepsilon_p}{\varepsilon_p}, \] (9)
we find that the condensate depletion diverges at the threshold of the roton instability even at \( T = 0 \) [35].

In contrast, due to the anisotropy of the spectrum of tilted dipoles (Fig. 1), close the threshold of the instability, we obtain (see, Fig. 1c)
\[ \varepsilon^2 |_{p = \pm p_t} \approx 1 \frac{d^2 p}{2 dp} p^2 + \frac{1}{2} \frac{d^2 p}{dp_y^2} (p_y \mp p_t)^2 = A(\theta) p^2 + B(\theta) (p_y \mp p_t)^2, \] (10)
where at \( \mathbf{p} = \pm p_t \):
\[ A(\theta) = \frac{1}{2} \frac{d^2 \varepsilon^2}{dp^2} = \frac{p^2}{2m^2} + \frac{n_0 p^2}{2m} \frac{\partial^2 U_{2d}(\mathbf{p}, \theta)}{dp^2}, \]
\[ B(\theta) = \frac{1}{2} \frac{d^2 \varepsilon^2}{dp_y^2} = \frac{p^2}{2m^2} + \frac{n_0 p^2}{2m} \frac{\partial^2 U_{2d}(\mathbf{p}, \theta)}{dp_y^2}, \]
with \( \partial^2 \varepsilon^2 / dp_x dp_y = 0 \). By substituting (10) in (9), we obtain that at the threshold of roton instability the condensate depletion (9) converges (see, Fig. 3).
IV. EXPERIMENTAL REALIZATIONS

We suggest experimental realization of the roton minimum and the roton instability for dysprosium atoms and RbOH polar molecules. Details of our estimations for the threshold of the roton instability are:

(i) Dysprosium atoms [6]: \( m = 164 \text{ u}, \ z_0 = 150 \text{ nm} \) (\( h\omega = 130 \text{ nK}, \ \omega / 2\pi = 2.72 \text{ kHz} \), \( \theta = 72^\circ, \ a_d = 7 \text{ nm}, \ a_s = 5.5 \text{ nm}, \ a = 0.5 \text{ nm}, \ n_0 = 2.15 \times 10^{10} \text{ cm}^{-2} \) (\( \alpha = 11/14, \ \gamma = 17/5 \)), \( \mu = 10.6 \text{ nK}, \ n_0/n = 197/200 \).

(ii) Polar molecules RbOH [8]: \( m = 104 \text{ u}, \ z_0 = 200 \text{ nm} \) (\( h\omega = 116 \text{ nK}, \ \omega / 2\pi = 2.42 \text{ kHz} \), \( \theta = 57.7^\circ, \ a_d = 14 \text{ nm}, \ a_s = 5 \text{ nm}, \ a = 3 \text{ nm}, \ n_0 = 2.65 \times 10^9 \text{ cm}^{-2} \) (\( \alpha = 5/14, \ \gamma = 10/9 \)), \( \mu = 9.3 \text{ nK}, \ n_0/n = 74/75 \).

In experiments with ultracold molecules, difficulties are related to ultracold chemical reactions (e.g., KRb+KRB=K₂+Rb₂). In this way, we expect that RbOH polar molecules are preferred for long-lived Bose gases. In contrast to KRb, ultracold RbOH molecules does not react as RbOH+RbOH=Rb₂+H₂O₂ and RbOH+RbOH=2Rb₂O+H₂O, with merging of two RbOH molecules into a dimer being suppressed [47].

V. DISCUSSIONS AND CONCLUSION

Main emphasis of our work is accessibility of the threshold of the roton instability for tilted dipoles based on the challenging problem of validity of the Bogoliubov approximation. In turn, this problem is related to two conditions: absence of the divergence of the condensate depletion at the threshold of the roton instability and the negligibility of the loops diagrams.

For the situation of normal to the wide layer dipoles, it was shown [36] that the Bogoliubov approximation becomes inapplicable before the roton minimum reaches zero (i.e., at non-zero roton gap).

In contrast, for tilted dipoles the condensate depletion as we have shown above converges at the threshold of the roton instability. Consequently, the depletion \( n - n_0 \) can be small enough if the interactions in the system are sufficiently weak. In this case, we obtain \( n_0 \approx n \). Hence, for tilted dipoles, even at the threshold of the instability the Bogolubnov approximation is at least self-consistent.

The problem of negligibility of the loops diagrams in weak interaction regime at \( T = 0 \), when the roton minimum touches zero [48], is more complicated and it will be considered in another place. At the same time, in the wide sense, the Bogoliubov approximation as well as the mean-field approach can be valid in quasi-2D system on the mesoscopic scales, i.e., in the quasicondensate formalism (see, [49–51]). Therefore, when the coupling constants are sufficiently small, the roton minimum in the macroscopic system should be closer to zero than the width of the crossover of the roton instability at finite-size scales. Furthermore, the characteristic size \( \delta p \) for a domain of roton minimum should be smaller than the length \( \hbar/L \) of the touching point in case of scales of order of \( L \) [51].

Moreover, we admit there are a macroscopic population in \( |p - p_r| \lesssim \delta p \) region even though the true roton minimum is higher than zero. In this case, there are local density waves in a macroscopic tilted dipole system [52], i.e., both diagonal and off-diagonal short-range orders. These orders are totally controlled by external fields: the wave period \( \lambda = 2\pi \hbar/p_r \) is given by \( \alpha \) and \( \theta \), the wave direction is determined by an orientation of the polarizing field, and the number of waves is control of interaction weakness (i.e., the quantity \( n - n_0/n_0 \)). At the same conditions, global density waves can be absent in the system.

Experimentally, the local density waves can be observed (i) in weakly interacting system of size \( L < \hbar/\delta p \) or (ii) in measurements of short-range order in one-body density matrix or pair-correlation function.

In contrast to our case, for wide to the layer dipoles, the Bogoliubov approximation is not self-consistent. This results from the divergence of the condensate depletion at the threshold of the roton instability [35]. Besides, the above mesoscopic arguments do not justify the Bogoliubov approach even in the weak interaction regime. It is in agreement with recent results [36].

To summarize our results, we have considered the stability problem for BEC gases of 2D tilted dipoles in a thin layer. The excitation spectrum of the system has the roton-maxon character. We have obtained stability diagrams in which we find a stable homogeneous, phonon-collapsed and roton unstable phases. For tilted dipoles, we predict achievability of the threshold of the roton instability at the finite-size scales as well as a possibility of local density waves with controlled short-range order. According to our numerical estimation, the effects are achievable in experiments with ultracold atoms and polar molecules in optical lattices.

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Here we based on the following strict considerations. Let ∆θ be the parameters α, γ, and ρ of the problem be as follow that the roton gap ∆r is small but differs from zero. Then, in both the macroscopic and weak interaction limits at T = 0 all loop diagrams are vanishing [48]. In this case, the Bogoliubov approximation (if it is self-consistent) is valid even in the macroscopic system. Therefore, it is valid on the finite-size scales [50], e.g., in Lx × Ly box. Let us choose Lx and Ly so that the condition ∆r = A(θ/Lx)2 + B(θ/Ly)2 is fulfilled. Then, for Lx × Ly box at these θ, α, and γ we obtain (i) the roton minimum is touch zero (see Eq. (10)) and (ii) the Bogoliubov approximation is valid. Thus, in the mesoscopic formalism the Bogoliubov approximation is valid at the threshold of roton instability of tilted dipoles. Moreover, it is valid even though there is a macroscopic population in the region of the minimum.

In the mesoscopic formalism, the Bogoliubov approximation in regime of macroscopic population of the minimum region is valid [51]. This case evidently corresponds to occurrence of local density waves in the system (if, of course, there are only two roton minima and they are symmetrical relative to the line p = 0). Seemingly, this can take place in the reconstructed roton unstable phase.

[47] It is unlikely that the system merges into excited dimer RbOH+RbOH=Rb2H2O2 because of huge energy gaps in electronic degree (∼10 K) and rotational (∼0.3 K) degrees of freedom [10]. Hence, the chemical reaction of the merging of two molecules RbOH into the dimer should be accompanied by a photon emission. Therefore, reaction RbOH + RbOH = Rb2H2O2 + hν should be suppressed by the reaction barrier. Thus, the lifetime of the system significantly increases. Moreover, since all bonds of RbOH are saturated, at the merging of two molecules RbOH emitted photon has energy, which is much lower energy than at merging of two radicals. This fact suppress of merging into a dimer on value of order (hν) in compare with common in literature OH [12] and NH [13] radicals.

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