

Quantum Rotor Model for a Bose-Einstein Condensate of Dipolar Molecules

J. Armaitis,* R. A. Duine, and H. T. C. Stoof

Institute for Theoretical Physics, Utrecht University, Leuvenlaan 4, 3584 CE Utrecht, The Netherlands
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We show that a Bose-Einstein condensate of heteronuclear molecules in the regime of small and static electric fields is described by a quantum rotor model for the macroscopic electric dipole moment of the molecular gas cloud. We solve this model exactly and find the symmetric, i.e., rotationally invariant, and dipolar phases expected from the single-molecule problem, but also an axial and planar nematic phase due to many-body effects. Investigation of the wave function of the macroscopic dipole moment also reveals squeezing of the probability distribution for the angular momentum of the molecules.

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Introduction.—A promising new direction in the field of ultracold quantum gases is the study of dipolar gases with heteronuclear molecules [1–3]. Recent progress in this direction has already contributed to such diverse research areas as atomic and molecular physics, quantum computation, and chemistry [4–6]. Indeed, the unique combination of strongly anisotropic long-range interactions and the quantum nature in these systems has brought to light a number of striking phenomena, such as tunneling-driven [7] and direction-dependent [8] ultracold chemical reactions, as well as the shape-dependent stability of the gas cloud [9].

The novel ingredient of heteronuclear molecules as compared to neutral atoms is their large permanent electric dipole moment, which opens the possibility for a strong dipole-dipole interaction. Neutral atoms typically do have a permanent magnetic dipole moment, but this leads to a dipole-dipole interaction that is much weaker than in the case of heteronuclear molecules, although it nevertheless has observable effects in certain cases [10,11], in particular when the scattering length is made small using a Feshbach resonance [12–14]. In the absence of an external electric field, however, the average dipole moment in the laboratory frame is zero, since the rotational ground state of the molecule is spherically symmetric and the dipole moment is thus randomly oriented. For that reason, virtually all theoretical many-body studies are carried out in the limit of a large dc electric field. In that limit the molecules are completely polarized and the dipole moment in the laboratory frame is maximal [15]. One notable deviation from the large electric field limit is the discussion by Lin *et al.* [16], which considers the effects of an almost resonant ac electric field.

Going away from the large-field limit unmasks the subtle interplay between the quantum-mechanical rotation of the molecules, the long-range dipole-dipole interaction, and the directing static electric field, which is the main topic of this Letter. In particular, the molecular Bose-Einstein condensate turns out to be a ferroelectric material that is fully disordered by quantum fluctuations in the

absence of an electric field. This is illustrated by the phase diagram of a Bose-Einstein condensate of heteronuclear molecules in a harmonic uniaxial trap that is shown in Fig. 1. The system possesses four phases: two nematic phases (a planar nematic and an axial nematic phase), a dipolar phase, and a fully symmetric phase that are separated by smooth crossovers. Two order parameters are relevant for this system. First, a nonzero average dipole moment $\langle d_i \rangle$ defines the dipolar phase. Second, in the absence of an average dipole moment the nematic (or quadrupole) tensor $Q_{ij} = \langle d_i d_j - \delta_{ij} d^2/3 \rangle$ distinguishes the other three phases. In particular, the nematic tensor is equal to zero in the spherically symmetric phase. Two

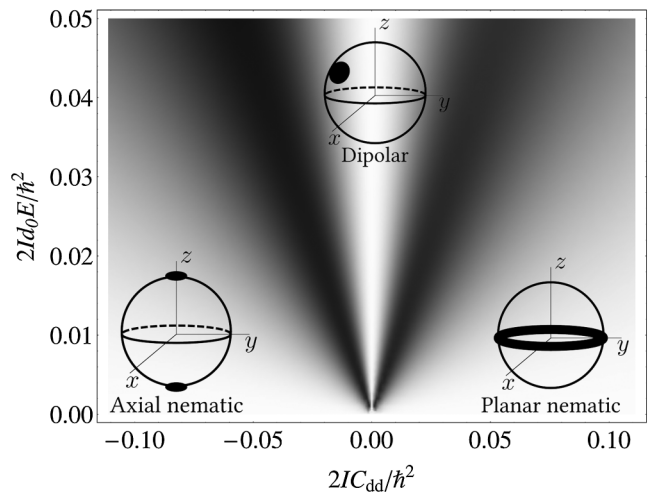


FIG. 1. Phase diagram of the axially symmetric Bose-Einstein condensate of heteronuclear molecules, where the probability distributions for the dipole moment on the unit sphere in the nontrivial phases are schematically indicated by the black areas on the spheres. The vertical axis is the external electric field, while the horizontal axis is the dipole-dipole interaction strength. In this diagram, the fully symmetric phase exists only in the origin. Shading corresponds to the “squeezing” parameter σ in Eq. (9), which runs from zero (white) to 0.09 (gray). The electric field is at a $\pi/4$ angle to the symmetry axis of the cloud.

eigenvalues are positive and one is negative in the planar nematic phase, whereas one eigenvalue is positive and two are negative in the axial nematic phase. It is worthwhile to notice that even in the absence of any electric field, many-body effects are crucial, giving rise to nematic ground states in strong contrast to the dipolar and fully symmetric ground states, expected from the single-molecule case. We finally remark that the predicted phase diagram is experimentally accessible by tuning three parameters in the laboratory, namely, the electric-field strength, the trap aspect ratio, and the number of particles.

Model.—We start from the single-molecule Hamiltonian

$$H_m = \frac{\mathbf{p}^2}{2m} + \frac{\mathbf{L}^2}{2I} - d_0 \hat{\mathbf{d}} \cdot \mathbf{E}, \quad (1)$$

where m is the mass of the molecule, $\mathbf{p} = -i\hbar\partial/\partial\mathbf{x}$ is the center-of-mass momentum operator with \mathbf{x} the center-of-mass position, I is the moment of inertia of the molecule, $d_0\hat{\mathbf{d}}$ is the electric dipole moment operator, $\mathbf{L} = -i\hbar\hat{\mathbf{d}} \times \partial/\partial\hat{\mathbf{d}}$ is the angular momentum operator, associated with the rotation of the molecules, and \mathbf{E} is the electric field. To describe the interactions between the molecules, we have to include both a contact (or s -wave) interaction term [17]

$$V_s = \frac{4\pi\hbar^2 a}{m} \delta(\mathbf{r}), \quad (2)$$

and a dipole-dipole interaction term

$$V_{\text{dd}} = -\frac{d_0^2}{4\pi\epsilon_0 r^3} (3\hat{\mathbf{d}}_1 \cdot \hat{\mathbf{r}} \hat{\mathbf{d}}_2 \cdot \hat{\mathbf{r}} - \hat{\mathbf{d}}_1 \cdot \hat{\mathbf{d}}_2), \quad (3)$$

where δ is the Dirac delta function, a is the s -wave scattering length, ϵ_0 is the electric permittivity of vacuum, $d_0\hat{\mathbf{d}}_1$ and $d_0\hat{\mathbf{d}}_2$ are the dipole moments of the two interacting particles, \mathbf{r} is the vector connecting them, and r is the distance between the particles. Finally, we consider the molecular gas to be trapped in a harmonic axially symmetric trapping potential

$$V_{\text{trap}} = m[\omega_{\perp}^2(x^2 + y^2) + \omega_z^2 z^2]/2, \quad (4)$$

where ω_{\perp} and ω_z are the radial and axial trapping frequencies, respectively.

For small electric fields, we are allowed to first solve for the spatial part of the condensate wave function by only including the effect of the s -wave interaction between the molecules. This leads to a Thomas-Fermi profile that depends on the s -wave scattering length [18]. The many-body ground state wave function is now $\Psi(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N; \mathbf{d}_1, \mathbf{d}_2, \dots, \mathbf{d}_N) = \prod_{i=1}^N \psi_{\text{TF}}(\mathbf{r}_i) \psi(\hat{\mathbf{d}})$, where N is the total number of molecules, and $\hat{\mathbf{d}}$ is the direction of $\mathbf{d} = (\sum_{i=1}^N \mathbf{d}_i)/N$. Hence, the dipole-dipole energy per particle is

$$V_{\text{dd}}^{\text{TF}} = -\frac{Nd_0^2}{4\pi\epsilon_0} \int d\mathbf{r} P(\mathbf{r}) \frac{1}{r^3} [3(\hat{\mathbf{d}} \cdot \mathbf{r})^2 - \hat{\mathbf{d}}^2 r^2], \quad (5)$$

where P is the probability to find two particles a certain distance apart. Subsequently, the many-body Hamiltonian per molecule in this so-called single-mode approximation [19] reduces to (cf. Sec. I of the Supplemental Material [20])

$$H = \frac{\mathbf{L}^2}{2I} - d_0 \hat{\mathbf{d}} \cdot \mathbf{E} + C_{\text{dd}}(3\hat{\mathbf{d}}_z^2 - \hat{\mathbf{d}}^2), \quad (6)$$

where C_{dd} is the effective dipolar interaction strength

$$C_{\text{dd}} = \frac{d_0^2 N}{4\epsilon_0} \int dz \rho d\rho P(R) \frac{1}{r^3} \left(\frac{3}{2} \frac{\rho^2}{r^2} - 1 \right), \quad (7)$$

and we have introduced the radius in cylindrical coordinates $r^2 = \rho^2 + z^2$, the dimensionless radius $R^2 = (\rho/x_{\text{TF}})^2 + (z/z_{\text{TF}})^2$, the radial size of the cloud x_{TF} , the axial size $z_{\text{TF}} = \lambda x_{\text{TF}}$, and the aspect ratio $\lambda = \omega_{\perp}/\omega_z$. We emphasize that even though Eq. (6) describes the dipole degree of freedom of the three-dimensional many-body system, it actually has a form of a single-particle (and thus effectively zero-dimensional) Hamiltonian, and the whole Bose-Einstein condensate acts as a single quantum rotor.

In the Thomas-Fermi approximation the probability P can be calculated analytically: $P(R) = 15(R-2)^4(32+64R+24R^2+3R^3)/7168\pi\lambda x_{\text{TF}}^3$ for $R < 2$ and zero otherwise. The analytic expression for $P(R)$ yields [21,22]

$$C_{\text{dd}} = -5Nd_0^2/[56\pi\epsilon_0 x_{\text{TF}}^3 \lambda(\lambda^2 - 1)^2] \\ \times \left(\lambda^4 + \lambda^2 - 2 + 3\lambda\sqrt{1 - \lambda^2} \text{arc cot} \left[\frac{\lambda}{\sqrt{1 - \lambda^2}} \right] \right), \quad (8)$$

which corresponds to one half of the mean-field dipolar energy per particle in the case of fully polarized electric dipoles [21,22]. Analogous results for magnetic dipoles were obtained by other authors for spinor Bose-Einstein condensates in the Gaussian approximation [23,24]. Note that C_{dd} depends on the number of particles N and the trap aspect ratio λ . Thus, the only effect of varying the number of particles is the change in C_{dd} .

The Hamiltonian in Eq. (6) represents a quantum rotor model for the macroscopic dipole moment of the molecular Bose-Einstein condensate, whose derivation is the main result of this Letter. Interestingly, a similar Hamiltonian applies to an atomic ferromagnetic spinor Bose-Einstein condensate (cf. Ref. [25] for a quantum rotor model of antiferromagnetic spinor condensates), but then without the quantum rotor term [23]. The reason for this difference is that the total (spin) angular momentum of the atoms is fixed, whereas in the case of interest here the wave function of the molecules is in general a superposition of states with an arbitrary (rotational) angular momentum, whose energy splitting is determined by the finite moment of inertia. Next we are going to investigate the ground-state properties of this quantum rotor model.

Results.—We have obtained the exact phase diagram pertaining to this Hamiltonian by expanding the dipolar

wave function in spherical harmonics (see Fig. 1). For zero electric field and no dipole-dipole interaction, the ground state of the system is a trivial spherically symmetric (non-dipolar) state. However, turning on \mathbf{E} or C_{dd} results in a very different state. For zero C_{dd} and nonzero \mathbf{E} , we obtain a dipolar state, where the probability distribution on the sphere is concentrated around the direction of the electric field. This state is classical in the sense that it is analogous to a classical dipole in the electric field. Another limiting case is $\mathbf{E} = 0$ and $C_{\text{dd}} < 0$, where we have an axial nematic phase, and the probability is concentrated around the north and south poles of the sphere. Finally, we have a planar nematic phase for $\mathbf{E} = 0$, $C_{\text{dd}} > 0$, where the high probability region is located around the equator of the sphere. The last two phases are quantum mechanical, as the ground state there is a coherent superposition of spherical harmonics with no average dipole moment. We observe smooth crossovers between the nontrivial phases, as expected due to the existence of quantum fluctuations in this effectively zero-dimensional situation.

In addition to the coordinate-space probability distribution $|\psi(\hat{\mathbf{d}})|^2$, we investigate the probability distribution with respect to angular momentum P_l . To that end, we expand our wave function in terms of spherical harmonics: $\psi(\hat{\mathbf{d}}) = \sum_{l,m} \alpha_{l,m} Y_{l,m}(\hat{\mathbf{d}})$. Hence, $P_l = \sum_{m=-l}^l P_{l,m}$, where $P_{l,m} = |\alpha_{l,m}|^2$ is the probability to occupy a state which has angular momentum quantum number l and azimuthal quantum number m . We find that this distribution has a peak at $l = 0$ for negative C_{dd} , and is peaked at $l \geq 0$ for positive C_{dd} or nonzero \mathbf{E} . For larger values of C_{dd} and $|\mathbf{E}|$, the peak shifts towards larger values of l . Moreover, due to the nature of the dipole-dipole interaction that conserves parity, at zero electric field P_l is zero for odd l . We have also investigated the distribution of probability between different $|l, m\rangle$ states (see Fig. 2). In general, this distribution is symmetric ($P_{l,m} = P_{l,-m}$) in every direction, implying that the average angular momentum $\langle \mathbf{L} \rangle$ is always zero, which is a consequence of time-reversal symmetry.

Noticing an anisotropic distribution of average dipole moment probability on the sphere in our system for certain parameters, it is natural to draw a parallel with the effect of spin squeezing [26]. To that end, we define a matrix $\langle L_i L_j \rangle$. This matrix describes the (Heisenberg) uncertainty in the angular momentum of the system. It has three eigenvalues that we order as follows: $|\lambda_0| \leq |\lambda_-| \leq |\lambda_+|$. Hence, we define a measure of angular momentum ‘‘squeezing’’ as

$$\sigma = \frac{|\lambda_+| - |\lambda_-|}{|\lambda_+| + |\lambda_-|}, \quad (9)$$

which tells us how anisotropic the uncertainty of angular momentum is (cf. Fig. 1). However, we must point out that, strictly speaking, this effect is not identical to squeezing in the usual sense, because $\langle L_i \rangle = 0$ and $P_{l,m}$ is not always a monotonically decreasing function of m (as can be seen from Fig. 2).

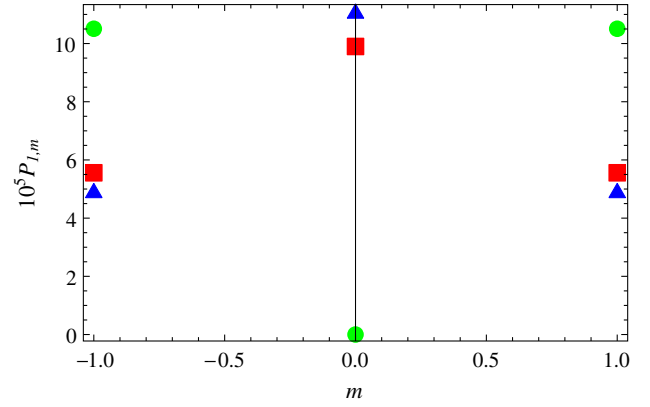


FIG. 2 (color online). Probability $P_{l,m}$ of occupying a state with total angular momentum 1 and its projection m . We have chosen $C_{\text{dd}} = 0.1\hbar^2/2I$ and $\mathbf{E} = 0.05\hbar^2/2Id_0$ (\mathbf{E} is at a $\pi/4$ angle to the z axis) in order to maximize the anisotropy of the state. The red squares correspond to the x' direction, the green circles correspond to the y' direction, and the blue triangles correspond to the z' direction, where the axes are defined such that the $\langle \hat{d}_i \hat{d}_j \rangle$ matrix is diagonal and has its smallest eigenvalue in the z' direction.

Discussion and conclusion.—It is interesting to compare the exact results described so far with mean-field theory techniques commonly employed for atomic Bose-Einstein condensates. Thus we turn to the Hartree approximation (which is equivalent to solving the Gross-Pitaevskii equation) for an analysis of the Hamiltonian in Eq. (6). To that end, we replace the operator \hat{d}_i^2 by $\hat{d}_i \langle \hat{d}_i \rangle$. The effect of the dipole-dipole interaction is then an additional static electric field of the form

$$\mathbf{E}_{\text{dd}} = \frac{C_{\text{dd}}}{d_0} (\langle \hat{d}_x \rangle, \langle \hat{d}_y \rangle, -2\langle \hat{d}_z \rangle)^T, \quad (10)$$

where the angle brackets indicate a quantum-mechanical average, and $N\langle \mathbf{d} \rangle \cdot \mathbf{E}_{\text{dd}}$ is the total average (Hartree) energy of all the classical dipoles with a density distribution given by the Thomas-Fermi profile. Therefore, we now have to solve the effective single-particle Hamiltonian

$$H_{\text{MF}} = \frac{L^2}{2I} - d_0 \hat{\mathbf{d}} \cdot \mathbf{E}_{\text{eff}}, \quad (11)$$

where $\mathbf{E}_{\text{eff}} = \mathbf{E} + \mathbf{E}_{\text{dd}}$ is the effective electric field, which now depends on the cloud geometry and the average dipole moment.

The average dipole moment in this approach is determined in two steps. First, we calculate the average dipole moment of the ground state $\langle \mathbf{d} \rangle(\mathbf{E})$ from Eq. (11) (see, e.g., Ref. [4]). Second, we write down a self-consistency condition, accounting for the effective electric field:

$$\langle \mathbf{d} \rangle = \langle \mathbf{d} \rangle(\mathbf{E}_{\text{eff}}(\langle \mathbf{d} \rangle)). \quad (12)$$

In the well-known case of a single molecule, C_{dd} is zero, Eq. (12) has a single solution, and $\langle \mathbf{d} \rangle$ always points in the

direction of \mathbf{E} . However, this is not the case for the whole (C_{dd} , \mathbf{E}) space and therefore requires a more thorough analysis. For small nonzero $|C_{dd}|$ and $\mathbf{E} = 0$, there still is only one solution, namely, $\langle \mathbf{d} \rangle = 0$. For $C_{dd} < 0$ and $E_z \neq 0$, the average dipole moment is always nonzero [Eq. (12) has a single solution], as then we are dealing with an Ising-like (easy-axis) model, and E_z couples directly to the order parameter $\langle \mathbf{d} \rangle$. In contrast to this, three solutions exist for $E_z = 0$ and $C_{dd}/|E_\perp|$ sufficiently large and negative. The two $\langle d_z \rangle \neq 0$ solutions are degenerate in energy, while the $\langle d_z \rangle = 0$ solution has a higher energy. On the other hand, for $C_{dd} > 0$ we are dealing with an XY -like (easy-plane) model and thus in that case one obtains a similar nontrivial situation for $E_\perp = 0$ and $C_{dd}/|E_z|$ large and positive.

When comparing the mean-field theory with the exact diagonalization of the Hamiltonian in Eq. (6), it is important to notice that the mean-field ansatz explicitly assumes that the average dipole moment is pointing in some direction. Therefore, the nematic phases are absent from the mean-field phase diagram. The fact that the dipole moment is zero under the asserted conditions can be intuitively understood, as the exact approach allows for a quantum superposition of states that have oppositely polarized dipole moments and are degenerate at the mean-field level. Finally, it is well known that the mean-field theory does not give reliable results in low dimensions because of the increased importance of quantum fluctuations. Since we are investigating an effectively zero-dimensional Hamiltonian, it is no surprise that the results of the mean-field theory differ significantly from the exact calculation.

In our analysis we have relied on the single-mode approximation, which is applicable to Bose-Einstein condensates with s -wave and dipole-dipole interactions [27]. However, we have not accounted for the dependence of the cloud aspect ratio z_{TF}/x_{TF} on dipole-dipole interactions. This limits the applicability of our analysis to the regime where the dipole-dipole interaction is much weaker than the mean-field s -wave interaction [23], i.e., $|\langle \mathbf{d} \rangle|^2 m / 4\pi\hbar^2 \epsilon_0 a \ll 1$. For a typical diatomic molecule with a mass of 80 atomic mass units, a scattering length of 5 Bohr radii, and an electric dipole moment of 1 D, this limits the external electric field strength to $|\mathbf{E}| \ll 1$ kV/cm, which translates to $2Id_0E/\hbar^2 \ll 0.05$ in the units of Fig. 1. We have also estimated that for a cloud of 10^7 particles with a linear extent of around $1 \mu\text{m}$, or radial trapping frequency of approximately $2\pi \times 80$ kHz, assuming a nearly two-dimensional trap with an aspect ratio of 1:10, $C_{dd} \approx 0.1\hbar^2/2I$, which corresponds to the energy of $2\pi\hbar \times 1$ GHz.

Besides the single-mode approximation, we have also made an assumption that the s -wave scattering length is independent of the dipole moment. Even though it has been shown that such a dependence is present [28–31], including it would merely add an extra self-consistency

equation to our approach. Its effect would be to change the Thomas-Fermi radii and thus map the system to a different point in the phase diagram. Therefore, all our results remain qualitatively unaffected.

In summary, we have considered an interacting Bose-Einstein condensate of dipolar molecules in a small static electric field. We have solved this problem exactly in the single-mode approximation and have also compared this with the mean-field (Gross-Pitaevskii) approach. We have found that the two approaches to the problem yield qualitatively very different results. Finally, we have put forward an experimentally accessible phase diagram and investigated the exact ground-state wave function both in coordinate and angular-momentum space.

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*j.armaitis@uu.nl

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