

# Homogeneous Bose Gas of Dipolar Molecules in the Mean Field Approximation

Robert E. Zillich

*Institute for Theoretical Physics, Johannes Kepler Universität Linz,  
Altenbergerstraße 69, A-4040 Linz, Austria*

K. Birgitta Whaley

*Department of Chemistry and Pitzer Center for Theoretical Chemistry,  
University of California, Berkeley, California 94720*

(Dated: November 2, 2016)

## Abstract

We present a mean field analysis of the effects of molecular rotation on the excitation spectrum and stability of ultracold dipolar gases. For an unpolarized homogeneous gas interacting with a pure dipole-dipole interaction, we find that for the rotational state  $L = 1$  the dipole-dipole interaction causes a splitting of the translation-rotation energy levels into a single  $M = 0$  and a doubly degenerate  $M = \pm 1$  excitation. For all other rotational states, the dipole-dipole interaction does not lead to coupling of translations and rotations and therefore has no effect on the rotational degeneracy of the excitations. The addition of arbitrarily small electric fields is found to introduce instabilities similar to those known to arise in the fully polarized dipolar gas. As in the case of a fully polarized gas, addition of a large enough short range repulsive potential is seen to stabilize the system, with the critical value of the repulsive interaction required for stabilization being larger when rotations are included.

## I. INTRODUCTION

Ultracold dipolar gases constitute a rapidly expanding area of study in cold quantum gases. These systems possess anisotropic long ranged interactions that can have significant effects on the properties of degenerate quantum gases. Initial interest focused on atomic ensembles of atoms possessing magnetic moments<sup>1-4</sup>. However with much recent experimental progress in both formation of and cooling of dipolar molecules<sup>5-13</sup>, attention has focused on investigating the properties of the more strongly interacting dipolar molecule ensembles. The molecular constituents bring additional interesting complexities to the behavior of cold gaseous ensembles, by virtue of their internal rotational and vibrational degrees of freedom that are absent in atomic systems. In particular, the presence of rotational degrees of freedom introduces fascinating questions of the interplay between rotation and translational motion in the quantum degenerate regime. Despite this strong motivation however, theoretical studies of trapped dipolar molecules have been to date mostly restricted to analysis of fully polarized systems in the presence of strong external fields, i.e., taking into account only translational degrees of freedom. For such polarized molecular systems, considerable theoretical effort has been devoted to developing an understanding both of the states of ultracold ensembles trapped in harmonic potentials<sup>14-22</sup> and of the phases of self-assembled aggregates<sup>23,24</sup>. The former studies of trapped dipolar gases have revealed the effects of the long range anisotropic dipolar interaction on the stability, spatial structure and excitations<sup>16,17,21,22,25,26</sup> of trapped Bose condensates, predicting interesting phenomena such as formation of bi-concave molecular ensembles<sup>21</sup> and excitation spectra possessing roton-like minima that signal the onset of instabilities at finite wave numbers<sup>16,17,26</sup>. In optical lattices, polar molecules are predicted to exhibit a rich variety of phases, including a supersolid phase<sup>27,28</sup>.

In this work we investigate the effect of molecular rotational degrees of freedom on a homogeneous Bose gas of dipolar molecules (dipolar molecular Bose gas, DMBG) within a mean field analysis in which translational and rotational degrees of freedom are treated on the same footing. This represents the first step in a systematic program of study of how the rotational kinetic energy influences the properties of ultracold dipolar molecules and constitutes a useful reference point for further work with more advanced theoretical approaches such as Quantum Monte Carlo or quantum many-body methods. After introducing

the mean field formulation we first briefly recall the mean field analysis of a fully polarized gas of aligned dipoles, for which compressional excitations perpendicular to the molecular orientation are known to lead to instability<sup>15</sup>. We then characterize the excitation spectrum and stability of an unpolarized gas of dipolar molecules as a function of the density, finding that in contrast to the polarized situation, the unpolarized gas is found to be stable for all excitations along all directions and furthermore displays a free rotor spectrum for all except the  $L = 1$  molecular rotational states which is split due to the dipolar interaction. We then develop the mean field solutions for excitation energies in the presence of a partially polarizing electric field. This analysis shows that, within a mean field analysis, while an unpolarized DMBG is stable with respect to excitations, the presence of an arbitrarily small electric field gives rise to instabilities similar to those of the fully polarized system, implying that the rotational kinetic energy does not stabilize the system for all wavevectors. This result raises interesting questions for further study beyond mean field.

## II. MEAN FIELD APPROACH TO DIPOLAR MOLECULAR BOSE GAS

In the mean field approximation, the many-body Schrödinger equation for the ground state of a Bose gas is approximated by the non-linear Gross-Pitaevskii (GP) equation in which the particle-particle interaction is replaced by an effective “averaged” interaction<sup>29–32</sup>. We consider here a homogeneous gas of dipolar molecules that are free to translate and rotate in three dimensions. The long range interactions of such a gas are given by pairwise dipole-dipole interactions of the form

$$V(\mathbf{r}_1 - \mathbf{r}_2, \Omega_1, \Omega_2) = \frac{d^2}{4\pi\epsilon_0} \frac{\Omega_1 \cdot \Omega_2 - 3(\Omega_1 \cdot \hat{\mathbf{r}})(\Omega_2 \cdot \hat{\mathbf{r}})}{r^3}, \quad (1)$$

where  $\mathbf{r}_i$  is the position of dipole  $i$  (corresponding to the center of mass of the  $i$ -th molecule),  $\Omega_i$  is the unit vector defining the orientation of the dipole, i.e., of the molecular orientation,  $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$ , and  $d$  is the bare dipole moment. In this work, we take the effects of short range intermolecular interactions into account by addition of a contact pseudo-potential  $g\delta(\mathbf{r})$ . For atomic Bose gases, there is a simple relation between  $g$  and the s-wave scattering length  $a$ ,  $g = 4\pi\hbar^2 a/m$ , where  $a$  is determined by both the short range part of the interaction and the dipole moment<sup>33,34</sup>. In the present case of molecule with rotational degrees of freedom, little is known about the appropriate pseudo-potentials. In the strong field limit

of fully polarized dipoles, Derevianko has derived velocity dependent anisotropic pseudo-potentials<sup>34</sup>. Close coupling calculations carried out in this regime have also shown that the low energy scattering is relatively insensitive to the short range interaction<sup>35,36</sup>. However, to our knowledge no attempt has been made to construct pseudo-potentials for interactions of molecules possessing only partial rotational polarization. We adopt the simplest, isotropic form here since the focus of this study is to characterize the *generic* effects of various, possibly competing potential energy features on the excitation energies rather than to make accurate calculations for specific systems. In particular, we seek to investigate the consequences of competition between long range dipolar interaction, short range repulsion, and the effect of rotational kinetic energy, as the electric field varies over a wide range of values. We shall therefore employ the simple delta function form  $g\delta(\mathbf{r})$ , treating  $g$  as a positive valued parameter that adjusts the strength of the short range repulsion. More realistic forms of short range interaction potentials could be produced for specific systems by performing close coupling calculations, which is beyond the scope of the present work.

The GP equation that we shall solve for a DMBG possessing both translational and rotational degrees of freedom and characterized by mass  $m$  ( $\lambda = \frac{\hbar^2}{2m}$ ), rotational constant  $B$  and bare dipole moment  $d$ , is then given by

$$\begin{aligned} \mu\Psi(\mathbf{r}_1, \Omega_1) = & -\lambda\nabla^2\Psi(\mathbf{r}_1, \Omega_1) + B\hat{L}^2\Psi(\mathbf{r}_1, \Omega_1) + V_{\text{ext}}(\mathbf{r}_1, \Omega_1)\Psi(\mathbf{r}_1, \Omega_1) \\ & + g \int d\Omega_2 |\Psi(\mathbf{r}_1, \Omega_2)|^2 \Psi(\mathbf{r}_1, \Omega_1) + U(\mathbf{r}_1, \Omega_1)\Psi(\mathbf{r}_1, \Omega_1). \end{aligned} \quad (2)$$

Here  $U(\mathbf{r}_1, \Omega_1)$  is the mean interaction that each molecule sees as a result of its dipole interactions with the other molecules and is given by

$$U(\mathbf{r}_1, \Omega_1) \equiv \int d^3r_2 d\Omega_2 V(\mathbf{r}_1 - \mathbf{r}_2, \Omega_1, \Omega_2) |\Psi(\mathbf{r}_2, \Omega_2)|^2. \quad (3)$$

As pointed out by Lahaye et al.<sup>37</sup>, it is not necessarily justified to set up a GP equation containing a real potential and a pseudopotential on equal footing. However, as noted above, we regard the particular short range term used here merely as a placemaker that provides a non-zero repulsive short range interaction, and do not attempt to justify its detailed form. The general issue of developing a consistent representation of both short and long range interactions for unpolarized dipolar gases is best addressed in the context of future studies for specific systems.

Solving the GP equation eq. (2) yields the effective single-molecule wavefunction  $\Psi = \Psi_0(\mathbf{r}, \Omega)$  (sometimes referred to as the “macroscopic wave function”) and the chemical potential  $\mu$ . Numerical solution of eq. (2) typically involves propagation of an initial trial form for  $\Psi$  in imaginary time, thus evolving to the ground state of this non-linear Schrödinger equation. The spatial integration of the convolution product in the last term may be avoided by Fourier transforming this and making use of the Fourier transforms of  $|\Psi(\mathbf{r}_2, \Omega_2)|^2$  and  $V$  (see eq. (A2)). However, in the homogeneous case that is of interest here, solving eq. (2) for the ground state becomes trivial, as we explain below. In the mean field approach, excitation energies are then estimated by linearizing the GP equation, leading to Bogoliubov-deGennes (BdG) equations. We shall see below that the incorporation of molecular rotation in eq. (2) increases the complexity of the resulting linearized equations. The major result of this work is thus the derivation of the linearized GP equations with rotation and solving the associated eigenvalue problems.

Under the influence of a finite external electric field (assumed to be parallel to the  $z$ -direction), dipolar molecules will possess a non-zero dipole moment with an average orientation parallel to the direction of the external field. The commonly used approximation for this situation is to neglect the rotational kinetic energy of dipolar molecules and to treat the system as a fully aligned dipolar Bose gas of particles *without* rotational degrees of freedom, with the dipoles held fixed in the field direction,  $\Omega = \mathbf{e}_z$ . In this approximation, the bare dipole moment  $d$  would overestimate the strength of the dipole-dipole interaction. Instead, one typically uses the value of the average dipole moment of the molecule in the external field,  $\langle \mathbf{d} \rangle$ , which is always smaller than  $d$  because of the non-zero angular distribution of a molecule in a finite field (the resulting states are often referred to as “pendular”<sup>38</sup>). The resulting BdG equations for a homogeneous, fixed orientation dipolar gas lead to the Bogoliubov spectrum<sup>15,39,40</sup>

$$\epsilon(\mathbf{k}) = \left[ (\lambda k^2)^2 + 2\lambda k^2 \rho \left( g + \frac{4\pi}{3} \langle d \rangle^2 (3 \cos^2 \theta_k - 1) \right) \right]^{1/2} \quad (4)$$

where the Fourier transformation of the dipole-dipole interaction in the fully aligned geometry,  $V_{\mathbf{k}} = \frac{4\pi}{3} \frac{d^2}{4\pi\epsilon_0} (3 \cos^2 \theta_k - 1)$ , eq. (A3), has been used. This leads to the well known result that, in the absence of a repulsive contact interaction, a homogeneous and fully aligned dipolar gas is unstable even for an arbitrarily small external field, since eq. (4) predicts that for  $g = 0$  the excitation energy will be imaginary for sufficiently small wave vector  $\mathbf{k}$

perpendicular to the external field<sup>15</sup>. Only for  $g > \frac{4\pi}{3}\langle d \rangle^2$ , will  $\epsilon(\mathbf{k})$  be real for all  $\mathbf{k}$  and the homogeneous system be stable. The situation is more complicated for inhomogeneous fully polarized dipolar gases in a trap, where the excitation energies can no longer be determined analytically, but need to be obtained numerically. It is known both from experiment<sup>3</sup> and from mean field calculations<sup>21</sup> that confinement in the direction of polarization has a stabilizing effect.

In the remainder of this paper we shall investigate how this instability of the polarized DMBG is affected by the rotational kinetic energy of the molecules in both unpolarized and partially polarized DMBGs. Following earlier practice in discussions of rotational motion of molecules in the presence of anisotropic interactions with other species or an environment, we shall refer to the rotational motion of the interacting molecules in the ground state as rotational zero-point motion of the molecules<sup>41</sup>.

### III. UNPOLARIZED BOSE GAS

In the homogeneous limit, we set the external potential  $V_{\text{ext}}$  to zero. For a homogeneous system, without loss of generality, we may assume a normalized, spatially homogeneous ground state solution  $\Psi_0(\mathbf{r}, \Omega; t) = \sqrt{\frac{\rho}{4\pi}} e^{-i\mu t/\hbar}$ . We immediately see that the chemical potential  $\mu$  is solely determined by the short range interaction, since the average dipole mean field potential vanishes in this homogeneous limit. When the short range interaction is approximated by the isotropic contact interaction as above, we obtain the chemical potential  $\mu = \rho g$ .

The excitation spectrum is obtained by linearizing the time-dependent GP equation (2), assuming small fluctuations around the ground state  $\Psi_0$ <sup>40</sup>,

$$\Psi(\mathbf{r}, \Omega; t) = \Psi_0(\mathbf{r}, \Omega) + \delta\Psi(\mathbf{r}, \Omega; t). \quad (5)$$

We now generalize the classic Bogoliubov analysis of a homogeneous Bose gas of point particles<sup>39</sup> to molecules possessing rotational degrees of freedom  $\Omega$ . From translational invariance, we know that the spatial dependence of  $\delta\Psi(\mathbf{r}, \Omega; t)$  can be written as plane wave fluctuations

$$\delta\Psi(\mathbf{r}, \Omega; t) = e^{-i\mu t/\hbar} [u_{\mathbf{k}}(\Omega) e^{i\mathbf{k}\mathbf{r}} e^{-i\omega t/\hbar} - v_{\mathbf{k}}(\Omega) e^{-i\mathbf{k}\mathbf{r}} e^{i\omega t/\hbar}] \quad (6)$$

Linearization of the dipole mean field (3) leads to

$$U(\mathbf{r}_1)\Psi(\mathbf{r}, \Omega; t) = \frac{\rho}{4\pi} \int d^3r_2 d\Omega_2 V(|\mathbf{r}_1 - \mathbf{r}_2|, \Omega_1, \Omega_2) (\delta\Psi(\mathbf{r}_2, \Omega_2) + \delta\Psi^*(\mathbf{r}_2, \Omega_2))$$

After also linearizing the mean field term of the contact interaction  $g \int d\Omega_2 |\Psi(\mathbf{r}_1, \Omega_2)|^2 \Psi(\mathbf{r}_1, \Omega_1)$ , we insert the plane wave ansatz, eq. (6). This allows the BdG equations to be derived with use of the Fourier transform of the orientation dependent dipole-dipole potential  $V_{\mathbf{k}}(\Omega_1, \Omega_2)$ , resulting in equations for the plane wave fluctuations  $u_{\mathbf{k}}(\Omega)$  and  $v_{\mathbf{k}}^*(\Omega)$ . We expand  $u_{\mathbf{k}}$  and  $v_{\mathbf{k}}^*$  in spherical harmonics

$$u_{\mathbf{k}}(\Omega) \equiv \sum_{L,M} u_{LM} Y_{LM}(\Omega) , \quad v_{\mathbf{k}}^*(\Omega) \equiv \sum_{L,M} v_{LM}^* Y_{LM}(\Omega). \quad (7)$$

omitting the index  $\mathbf{k}$  for brevity, and thereby obtain the BdG equations for  $u_{LM}$  and  $v_{LM}^*$ :

$$\omega u_{LM} = \lambda k^2 u_{LM} + BL(L+1)u_{LM} \quad (8a)$$

$$+ \frac{\rho}{4\pi} \int d\Omega_1 d\Omega_2 Y_{LM}(\Omega_1) V_{\mathbf{k}}(\Omega_1, \Omega_2) (u_{\mathbf{k}}(\Omega_2) - v_{\mathbf{k}}^*(\Omega_2)) + \rho g \delta_{L,0} \delta_{M,0} (u_{00} - v_{00}^*)$$

$$\omega v_{LM}^* = -\lambda k^2 v_{LM}^* - BL(L+1)v_{LM}^* \quad (8b)$$

$$- \frac{\rho}{4\pi} \int d\Omega_1 d\Omega_2 Y_{LM}(\Omega_1) V_{\mathbf{k}}^*(\Omega_1, \Omega_2) (v_{\mathbf{k}}^*(\Omega_2) - u_{\mathbf{k}}(\Omega_2)) + \rho g \delta_{L,0} \delta_{M,0} (v_{00}^* - u_{00}).$$

It is immediately apparent that the BdG equations do not couple states of different angular momentum  $L$ . Furthermore, the full orientation dependent dipole-dipole potential  $V_{\mathbf{k}}(\Omega_1, \Omega_2)$  contains only  $L = 1$  components of the molecular orientations, eq. (A2). This implies that the mean field dipolar interaction in eqns. (8a) and (8b) vanishes for all angular momentum quantum numbers except  $L = 1$ , i.e., within the mean field approximation, the dipole-dipole interaction has no effect on molecular states with  $L \neq 1$ . Eqns. (8a) and (8b) also show that the short-range repulsion  $g$  only affects the rotational ground state,  $L = 0$ , and that the energies of all other rotational states  $L > 0$  are independent of the short range repulsion when this is described by an isotropic pseudopotential. For the  $L = 0$  state we then recover the well-known Bogoliubov spectrum of a homogeneous Bose gas,  $\omega_0 = \sqrt{(\lambda k^2)^2 + 2\lambda k^2 \rho g}$ , while for  $L \geq 2$ , the spectrum of a freely rotating and translating molecule is obtained.

We now analyze the case of greatest interest,  $L = 1$ , where the dipole-dipole interaction induces a non-zero mean field coupling between molecular rotational states. For  $L = 1$ , the

BdG equations are

$$\begin{aligned}\omega u_{1M} &= \lambda k^2 u_{1M} + 2B u_{1M} + \rho \frac{d^2}{4\pi\epsilon_0} \frac{(4\pi)^2}{9} \sum_{m'm} Y_{2m'}(\Omega_k) \begin{bmatrix} 2 & 1 & 1 \\ m' & M & -m \end{bmatrix} (u_{1m} - v_{1m}^*) \\ \omega v_{1M}^* &= -\lambda k^2 v_{1M}^* - 2B v_{1M}^* - \rho \frac{d^2}{4\pi\epsilon_0} \frac{(4\pi)^2}{9} \sum_{m'm} Y_{2m'}(\Omega_k) \begin{bmatrix} 2 & 1 & 1 \\ m' & M & -m \end{bmatrix} (v_{1m}^* - u_{1m}),\end{aligned}$$

Solving this 6-dimensional eigenvalue problem becomes trivial if we note that without an external field the system is isotropic. Then we can choose  $\mathbf{k}$  to be parallel to the  $z$  axis and since  $Y_{2m'}(\hat{\mathbf{e}}_1) = \sqrt{\frac{5}{4\pi}} \delta_{m',0}$ , we obtain

$$\begin{aligned}\omega u_{1M} &= \lambda k^2 u_{1M} + 2B u_{1M} + \rho \frac{d^2}{4\pi\epsilon_0} \frac{(4\pi)^2}{9} \sqrt{\frac{5}{4\pi}} \begin{bmatrix} 2 & 1 & 1 \\ 0 & M & -M \end{bmatrix} (u_{1M} - v_{1M}^*) \\ \omega v_{1M}^* &= -\lambda k^2 v_{1M}^* - 2B v_{1M}^* - \rho \frac{d^2}{4\pi\epsilon_0} \frac{(4\pi)^2}{9} \sqrt{\frac{5}{4\pi}} \begin{bmatrix} 2 & 1 & 1 \\ 0 & M & -M \end{bmatrix} (v_{1M}^* - u_{1M}).\end{aligned}$$

These equations are both diagonal in  $M$ , indicating that the angular momentum projection on any selected axis  $z$  is a good quantum number and allowing the secular equation to be solved for each  $M$  separately. This results in two distinct eigenvalues, one for  $M = 0$ , and a doubly degenerate eigenvalue for  $M = \pm 1$ :

$$\omega_{10} = \sqrt{(\lambda k^2 + 2B)^2 + 2\pi\rho(\lambda k^2 + 2B) \frac{8}{9} \frac{d^2}{4\pi\epsilon_0}} \quad (9a)$$

$$\omega_{1,\pm 1} = \sqrt{(\lambda k^2 + 2B)^2 - \pi\rho(\lambda k^2 + 2B) \frac{8}{9} \frac{d^2}{4\pi\epsilon_0}}. \quad (9b)$$

This mean field analysis of our model eq.(2) thus predicts that for the unpolarized homogeneous DMBG, the dipole-dipole interaction couples the translational degrees of freedom and the rotational degree of freedom for the molecular rotational states  $L = 1$  but does not affect any other rotational states. We find that the 3-fold  $M$  degeneracy of the  $L = 1$  state of a linear rotor is lifted and the states split into a lower doubly degenerate level and an upper non-degenerate level. While  $\omega_{10}$  is strictly real, we see that, in principle, the excitation energy  $\omega_{1,\pm 1}$  could become imaginary, corresponding to an instability. However, it is easy to verify that this does not happen for realistic values of  $B$  and  $\rho d^2$  of naturally occurring dipolar molecules. For example for a density of  $\rho = 10^{14} \text{cm}^{-3}$  and a relatively large dipole moment of 5 Debye, the rotational constant  $B$  of the molecule would have to be smaller than  $35 \mu\text{cm}^{-1}$  which is much smaller than the  $B$  values of diatomic molecules. We stress that the



dispersion relation depends on the short range interaction only for  $L = 0$  (see above), and hence eqns. (9a) and (9b) are independent of  $g$ .

For purely rotational modes, i.e., for  $k = 0$ , expanding the eigenvalues to linear order in  $\frac{\rho d^2}{B}$  yields the rotational excitation energies

$$\begin{aligned}\omega_{10} &= 2B \left( 1 + \frac{2}{9} \frac{\rho d^2}{2B\epsilon_0} \right) \\ \omega_{1,\pm 1} &= 2B \left( 1 - \frac{1}{9} \frac{\rho d^2}{2B\epsilon_0} \right),\end{aligned}$$

which show that the rotational degeneracy of the  $L = 1$  state of an isolated molecule is split by the dipole-dipole interaction between the molecules of a DMBG. For  $k = 0$ , the energy splitting is  $\Delta = \frac{\rho d^2}{3\epsilon_0}$ , which is independent of the molecular rotation constant  $B$  but depends linearly on the density. In order to give an example of the possible magnitude of the pure rotational splitting, we assume a density of  $\rho = 10^{14} \text{cm}^{-3}$  and a dipole moment of 5 Debye. This yields an energy splitting  $\Delta = 5.3 \times 10^{-5} \text{cm}^{-1} = 1.6 \text{MHz}$ , i.e., in the radio frequency range. We note that if such a splitting could be measured, it would provide a local probe of the density  $\rho$ , when the molecular dipole moment  $d$  is known.

The linear dependence of the line splitting  $\Delta$  on density and the strength of the dipole-dipole interaction  $d^2$  is reminiscent of the broadening of microwave or magnetic resonance spectra due to electric or magnetic dipole interactions. The dipolar broadening of magnetic resonance spectra in crystals, i.e. for spins on a lattice, is known to result in a linear dependence of the line width on  $\rho$  and  $d^2$ <sup>44</sup> and a similar dependence is seen in rotational resonance broadened microwave spectra of linear dipolar molecules<sup>42,43</sup>. Our mean field analysis of the DMBG results in a line splitting for a given  $k$  value. At this time we cannot rule out the possibility that additional broadening of the  $L = 0 \rightarrow 1$  transition of a DMBG might be predicted by a more accurate theory.

#### IV. PARTIALLY POLARIZED BOSE GAS

We now derive the BdG equations for the excitations of a homogeneous DMBG with both translational and rotational degrees of freedom, in a finite valued external electric field that is too weak to fully align the dipoles. Numerical solutions of these equations will show that in the mean field treatment, an arbitrarily small external alignment field destabilizes a homogeneous DMBG when it interacts only via the long range dipole-dipole interaction

and there is no short range repulsion, i.e., when  $g = 0$ , but that it can be stabilized by the presence of a large enough finite value of  $g$ . Furthermore, we will see that neglecting the rotational degrees of freedom and using the average dipole moment  $\langle \mathbf{d} \rangle$  in the fully polarized approximation eq. (4) is a good approximation for typical dipole moments and densities: only for large values of  $\rho d^2/B$  do the deviations of this approximation from the full treatment become significant.

The one-body Hamiltonian for a single rotating and translating electric dipole in a constant external field  $\mathbf{E}$  pointed in the  $z$ -direction is given by

$$H_0 = -\lambda \nabla^2 + B \hat{L}^2 + \varepsilon Y_{10}(\Omega), \quad (10)$$

where  $\varepsilon = -|\mathbf{E}|d$ . The GP equation for a homogeneous system of such dipoles is obtained by adding the mean fields of the contact interaction and of the dipole-dipole interaction, leading to eq. (2) with  $V_{\text{ext}}(\mathbf{r}, \Omega) \equiv \varepsilon Y_{10}(\Omega)$ . In the homogeneous limit, the ground state wave function  $\Psi_0$  does not depend on  $\mathbf{r}$ . However, unlike the previous situation for unpolarized dipoles, under the influence of the external field  $V_{\text{ext}}$ ,  $\Psi_0$  now depends on the molecular orientation  $\Omega$ . Note that just as in the absence of an electric field, the average dipole-dipole interaction vanishes, despite the presence of the field:

$$U = \int d\Omega_2 |\Psi_0(\Omega_2)|^2 \int dr r^2 d\Omega_r V(\mathbf{r}, \Omega_1, \Omega_2) = 0 \quad (11)$$

This follows from the substitution of the expansion (A1) for the potential  $V$ . Thus the dipole-dipole interaction vanishes in the mean field approximation, even if the ground state  $\Psi_0(\Omega)$  is anisotropic because of the external field. We note that this is a consequence of the homogeneity of the DMBG considered here and will not be the case for a DMBG in a trap. The mean field of the short range contact interaction is just a constant,  $\rho g$ , which follows from eq. (2) and the normalization of  $\Psi_0(\Omega)$ ,  $\int d\Omega |\Psi_0(\Omega)|^2 = \rho$ . Consequently, the GP ground state  $\Psi_0(\Omega)$  that results from solution of the 1-body equation  $H_0 \Psi(\Omega) = (\mu - \rho g) \Psi(\Omega)$  is equivalent to the single-molecule wave function in the external field with ground state energy  $E_0 = \mu - \rho g$ . Since we define the  $z$ -direction to be the direction of the external electrical field,  $\Psi_0(\Omega)$  does not depend on the azimuthal angle  $\phi$ , but only on the polar angle  $\theta$ . The equation  $H_0 \Psi = (\mu - \rho g) \Psi$  can readily be solved by expanding  $\Psi$  in spherical harmonics

$$\Psi_0(\Omega) = \sqrt{\rho} \sum_{\ell} a_{\ell} Y_{\ell 0}(\cos \theta), \quad (12)$$

yielding a discrete eigenvalue problem with lowest eigenvalue  $E_0$  and corresponding eigenvector  $\{a_\ell\}$ . The chemical potential  $\mu$  is then simply the sum of  $E_0$  and the contribution  $\rho g$  deriving from the short range repulsion. Without loss of generality, the ground state wave function  $\Psi_0(\Omega)$  can be assumed to be real.

As before, the BdG equations for a partially polarized DMBG are obtained by linearizing the GP equation, eq. (2), and inserting  $\Psi_0(\Omega)$ , eq. (12), as the ground state in the ansatz eqns. (5) and (6). Using again the fact that the averaged dipole-dipole interaction vanishes, the linearized mean field interaction then becomes

$$U(\mathbf{r}_1)\Psi(\mathbf{r}_1, \Omega_1; t) = \int d^3r_2 d\Omega_2 V(|\mathbf{r}_1 - \mathbf{r}_2|, \Omega_1, \Omega_2) \Psi_0(\Omega_1)\Psi_0(\Omega_2) [\delta\Psi(\mathbf{r}_2, \Omega_2) + c.c.] + \mathcal{O}(\delta\Psi^2),$$

which results in the coupled BdG equations

$$\begin{aligned} \omega u_{\mathbf{k}}(\Omega_1) &= (\lambda k^2 - E_0)u_{\mathbf{k}}(\Omega_1) + B\hat{L}^2 u_{\mathbf{k}}(\Omega_1) + \varepsilon Y_{10}(\Omega_1)u_{\mathbf{k}}(\Omega_1) \\ &\quad + \Psi_0(\Omega_1) \int d\Omega_2 V_{\mathbf{k}}(\Omega_1, \Omega_2) \Psi_0(\Omega_2)(u_{\mathbf{k}}(\Omega_2) - v_{\mathbf{k}}^*(\Omega_2)) \\ &\quad + g\Psi_0(\Omega_1) \int d\Omega_2 \Psi_0(\Omega_2)(u_{\mathbf{k}}(\Omega_2) - v_{\mathbf{k}}^*(\Omega_2)) \\ \omega v_{\mathbf{k}}^*(\Omega_1) &= -(\lambda k^2 - E_0)v_{\mathbf{k}}^*(\Omega_1) - B\hat{L}^2 v_{\mathbf{k}}^*(\Omega_1) - \varepsilon Y_{10}(\Omega_1)v_{\mathbf{k}}^*(\Omega_1) \\ &\quad - \Psi_0(\Omega_1) \int d\Omega_2 V_{\mathbf{k}}^*(\Omega_1, \Omega_2) \Psi_0(\Omega_2)(v_{\mathbf{k}}^*(\Omega_2) - u_{\mathbf{k}}(\Omega_2)) \\ &\quad - g\Psi_0(\Omega_1) \int d\Omega_2 \Psi_0(\Omega_2)(v_{\mathbf{k}}^*(\Omega_2) - u_{\mathbf{k}}(\Omega_2)). \end{aligned}$$

Just as we did for the BdG equations of the unpolarized DMBG, we expand  $u$  and  $v^*$  in spherical harmonics, eq. (7). Using the expansion eq. (12) of  $\Psi_0(\Omega)$  and the expansion (A2) of the dipole-dipole potential allows us then to cast the BdG equations into a form convenient for calculations, namely

$$\begin{aligned} \omega u_{LM} &= (\lambda k^2 - E_0)u_{LM} + BL(L+1)u_{LM} + \varepsilon \sum_{L'} \begin{bmatrix} L & 1 & L' \\ -M & 0 & M \end{bmatrix} u_{L'M} \\ &\quad + \rho \sum_{L', M'} V_{LM}^{L'M'} (u_{L'M'} - v_{L'M'}^*) + \rho g \delta_{M,0} \sum_{L'} a_L a_{L'} (u_{L'0} - v_{L'0}^*) \end{aligned} \quad (13a)$$

$$\begin{aligned} \omega v_{LM}^* &= -(\lambda k^2 - E_0)v_{LM}^* - BL(L+1)v_{LM}^* - \varepsilon \sum_{L'} \begin{bmatrix} L & 1 & L' \\ -M & 0 & M \end{bmatrix} v_{L'M}^* \\ &\quad - \rho \sum_{L', M'} V_{LM}^{L'M'} (v_{L'M'}^* - u_{L'M'}) - \rho g \delta_{M,0} \sum_{L'} a_L a_{L'} (v_{L'0}^* - u_{L'0}), \end{aligned} \quad (13b)$$

with

$$V_{LM}^{L'M'} = \frac{d^2}{4\pi\epsilon_0} \frac{(4\pi)^3}{9} \sum_{\substack{\ell=L\pm 1 \\ \ell'=L'\pm 1}} a_\ell a_{\ell'} \begin{bmatrix} L & \ell & 1 \\ -M & 0 & M \end{bmatrix} \begin{bmatrix} 1 & \ell' & L' \\ -M' & 0 & M' \end{bmatrix} \begin{bmatrix} 2 & 1 & 1 \\ m' & M & -M' \end{bmatrix}_{m'=M'-M} Y_{2,M'-M}(\Omega_k). \quad (14)$$

Inspection of these equations shows that the dipole-dipole interaction, the short range repulsion, and the external field all act to couple the molecular rotation states, but the latter two couple only the rotational quantum numbers  $L$  and do not couple different  $M$  states. A more general anisotropic pseudo-potential at short range could of course introduce additional rotation coupling. The dipolar coupling matrix elements are seen to be controlled by the density and the square of the dipole moment. We therefore define a ‘‘dipolar coupling’’ parameter  $c_d$  as the ratio between the prefactor of the dipolar matrix elements and the rotational constant  $B$ , specifically,  $c_d = \frac{\rho d^2}{4\pi\epsilon_0 B}$ .

From eq. (14) for  $V_{LM}^{L'M'}$  we see that  $V_{LM}^{L'M'} = 0$  whenever  $|M| > 1$  or  $|M'| > 1$ , regardless of the value of  $L$ . Thus the molecular translations and rotations are coupled by the dipole-dipole interaction only for  $M \in \{-1, 0, 1\}$ . From a numerical point of view, this significantly reduces the size of the matrix to be diagonalized to solve the eigenvalue problem. We shall refer to the mixed rotational and translational excitations  $\omega_{lm}(\mathbf{k})$  as rotation-phonon modes.

### A. Results for $g = 0$

We first examine the excitations when the short range interaction is absent, i.e., when  $g = 0$  and only the long range dipole-dipole interaction is present.

Fig. 1 shows the energy  $\omega_1(\mathbf{k})$  of the lowest modes, in units of the rotational constant  $B$ . In the absence of dipole-dipole coupling,  $\omega_1(\mathbf{k})$  would correspond to the  $L = 0$  rotational state for a given  $\mathbf{k}$ . In Fig. 1,  $\omega_1(\mathbf{k})$  is plotted as function of the reduced wave number  $k/k_0$ , where  $k_0 = \sqrt{2mB/\hbar^2}$ , for two directions of  $\mathbf{k}$ , namely parallel and perpendicular to the polarization direction. The lower dispersion curve shows the parallel and the upper dispersion curve the perpendicular mode. In these calculations the field strength  $\epsilon/B$  was set to 0.05, resulting in a weak polarization for which the field-averaged single molecule dipole moment  $\langle \mathbf{d} \rangle$  amounts to only 0.0081, i.e. 0.81% of the bare dipole moment value. The strength of the dipolar coupling  $c_d$  was then increased from  $10^{-6}$  to  $10^{-1}$ , in steps of factors of 10. We also plot for comparison the energy dispersion for aligned dipoles without

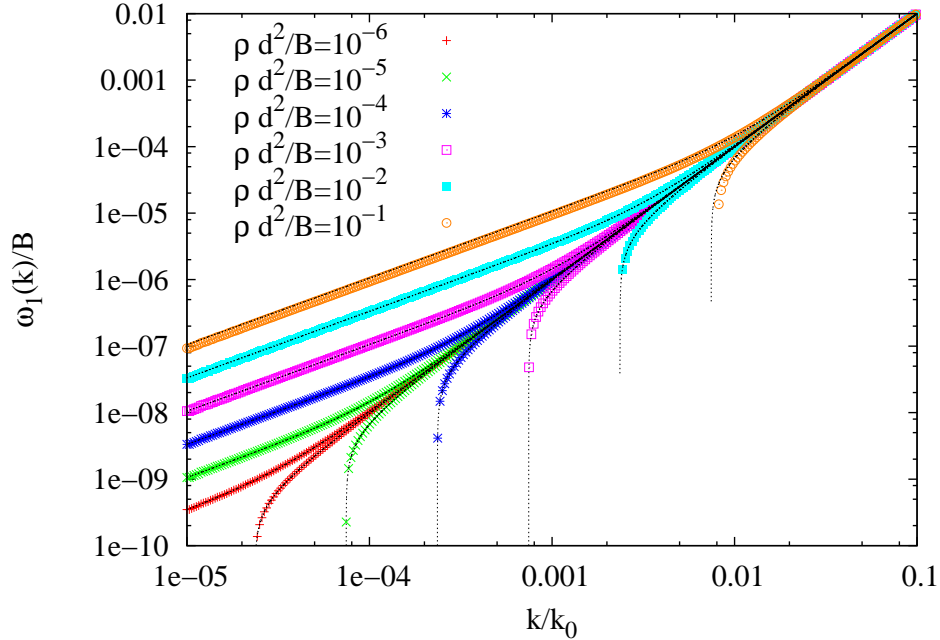


FIG. 1. Energy spectrum  $\omega_1(\mathbf{k})$  of a homogeneous Bose gas of rotating dipoles in a weak electric field, evaluated for dipolar coupling strengths  $c_d = \rho d^2/4\pi\epsilon_0 B = 10^{-6}, 10^{-5}, \dots, 10^{-1}$ , in a field of strength  $\varepsilon/B = 0.05$  and no short range repulsion ( $g = 0$ ). For each value of  $c_d$ , we show two curves, a lower and a higher dispersion branch which correspond to a wave vector  $\mathbf{k}$  oriented perpendicular and parallel to the polarization ( $z$ ) direction, respectively. For all coupling values, the perpendicular mode becomes imaginary below some finite wave number, thus destabilizing the system. The dotted lines show as reference the spectra of non-rotating dipoles polarized in the  $z$ -direction with an average dipole moment  $\langle \mathbf{d} \rangle$ , also in the absence of short range interactions (see eq. (4)).

rotational degrees of freedom,  $\epsilon(\mathbf{k})$ , using  $\langle \mathbf{d} \rangle$  as the value of the dipole moment in eq.(4). We see that on the scale shown here the excitations  $\omega_1(\mathbf{k})$  and  $\epsilon(\mathbf{k})$  are indistinguishable for dipolar coupling strengths up to  $c_d = 10^{-2}$ . Only for the largest value of the dipolar coupling,  $c_d = 10^{-1}$ , do we see small deviations between the two excitation energies. We note that this corresponds to a very high dipolar coupling: for a molecule with  $B = 0.01\text{cm}^{-1}$  and  $d = 5$  Debye, at a density of  $\rho = 10^{14}\text{cm}^{-3}$ , the corresponding value of  $c_d$  is still only  $\approx 10^{-3}$ .

The most striking feature of Fig. 1 is the behavior of the perpendicular dispersion curves,

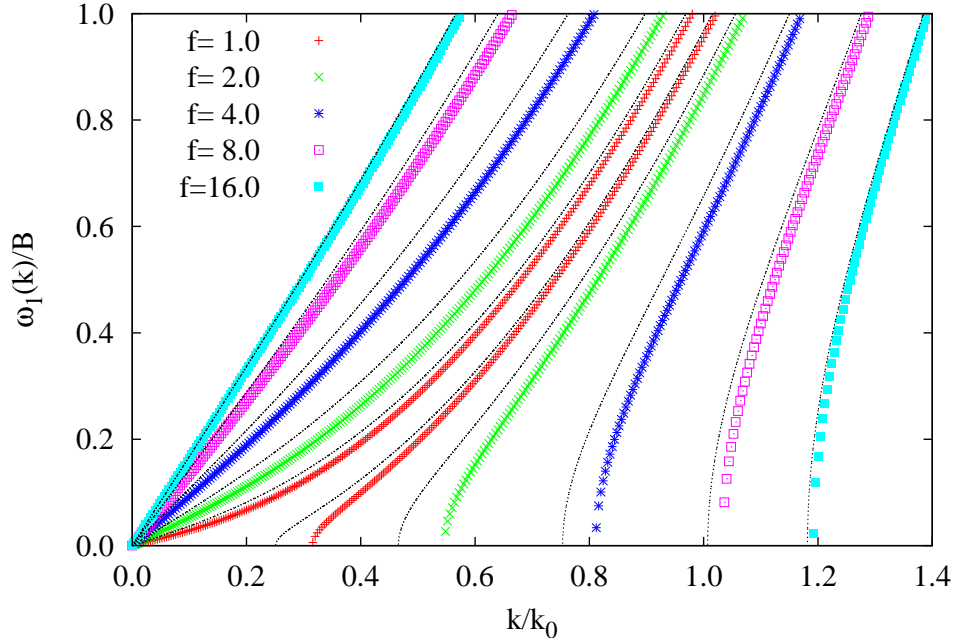


FIG. 2. Energy spectrum  $\omega_1(\mathbf{k})$  of a homogeneous Bose gas of rotating dipoles for large dipolar coupling  $c_d = 0.3$ , in electric fields of strength  $\varepsilon/B = 1; 2; 4; 8; 16$ . For each value of field strength  $\varepsilon/B$ , the lower and higher dispersion curves correspond to a wave vector  $\mathbf{k}$  perpendicular and parallel to the polarization ( $\mathbf{z}$ ) direction, respectively. Dotted lines show the corresponding excitation spectra  $\epsilon(\mathbf{k})$ , eq. (4), for non-rotating dipoles polarized in the  $z$ -direction with dipole moment  $\langle \mathbf{d} \rangle$ . As the field strength is increased,  $\langle \mathbf{d} \rangle \rightarrow d$  and  $\omega_1(\mathbf{k})$  approaches  $\epsilon(\mathbf{k})$ .

i.e., the excitations with  $\mathbf{k} \perp \mathbf{z}$ . For all values of dipolar coupling  $c_d$  in Fig. 1,  $\omega_1(\mathbf{k})$  is seen to become imaginary for sufficiently low perpendicular wave number, regardless of how small the dipolar coupling  $c_d$  becomes and even at a weak average polarization of less than 1%. Hence, taking into account rotational degrees of freedom leads to the same conclusion as found for a fully polarized gas, namely that without a short range repulsion the DMBG is not stable in the homogeneous limit. Thus within the mean field approximation, the rotational zero-point motion of the interacting molecules does not stabilize a homogeneous gas of rotating and interacting dipoles.

We now make an closer examination of the effects of neglecting rotational degrees of freedom in the mean field analysis for the case of large dipolar coupling, while varying the external field. Fig. 2 shows the lowest excitation energy  $\omega_1(\mathbf{k})$  for a very large dipolar

coupling  $c_d = 0.3$ . The field strength is varied as  $\varepsilon/B = 1; 2; 4; 8; 16$ , with respective induced dipole moments  $\langle \mathbf{d} \rangle/d = 0.16; 0.29; 0.48; 0.64; 0.75$ , relative to the bare  $d$ . As before, we show excitations for both parallel and perpendicular dispersion (upper and lower curves, respectively). In all cases, the system again exhibits unstable low momentum modes in the perpendicular direction. As before, we compare these spectra with those of non-rotating dipoles polarized in the  $z$ -direction,  $\epsilon(\mathbf{k})$ , eq. (4), with dipole strength  $d$  given by the average value  $\langle \mathbf{d} \rangle$  (dotted lines). For low relative polarization, i.e.,  $\langle \mathbf{d} \rangle/d \ll 1$ , we see that neglecting molecular rotations leads to significant deviations from the full  $\omega_1(\mathbf{k})$ . But on increasing the relative polarization towards unity (i.e., towards fully aligned dipoles), this deviation shrinks and  $\epsilon(\mathbf{k})$  approaches  $\omega_1(\mathbf{k})$ , as expected, indicating that the two mean field results are consistent.

Finally, we note that regardless of the choice of system parameters, the rotation-phonon energies  $\omega_1(\mathbf{k})$  are found to lie always below the excitations of the fully polarized gas,  $\epsilon(\mathbf{k})$ .

## B. Results for $g > 0$

We have seen above that in the absence of short range repulsion, i.e., when  $g = 0$ , an arbitrarily small external field leads to a collapse of the homogeneous DMBG, even when rotational degrees of freedom are taken into account. We now stabilize the DMBG by adding a short range interaction of strength  $g$ , until the imaginary part of the spectrum at small values of momentum vanishes, and compare again with approximation of non-rotating dipoles  $\epsilon(\mathbf{k})$ , eq. (4). We define a repulsion parameter  $\gamma \equiv \rho g/B$ , which is the ratio between the mean field of the short range repulsive interaction and the molecular rotational constant. In Fig. 3, we show the dispersion of the lowest mode  $\omega_1(\mathbf{k})$  for perpendicular and parallel wave vectors  $\mathbf{k}$ , in units of  $B$ . As before the dipolar coupling was set to a very large value of  $c_d = 0.3$ . The external field strength here is  $\varepsilon/B = 5 \times 10^{-3}$ , resulting in the weak relative polarization  $\langle \mathbf{d} \rangle/d = 0.81 \times 10^{-3}$ . The repulsion parameter in the four panels varies as  $\gamma = 0; 0.015; 0.030; 0.035$ . The plot shows that a short range repulsive interaction can shift the instability down to smaller values of  $k$  and for a sufficiently large value of  $g$  the instability is entirely removed. Furthermore, just as is seen with  $g = 0$ , the mixed rotation-phonon excitation  $\omega_1(\mathbf{k})$  is found to lie consistently below the excitation  $\epsilon(\mathbf{k})$  in the full polarization approximation. This implies that a larger value of  $\gamma$  and hence of the short range interaction

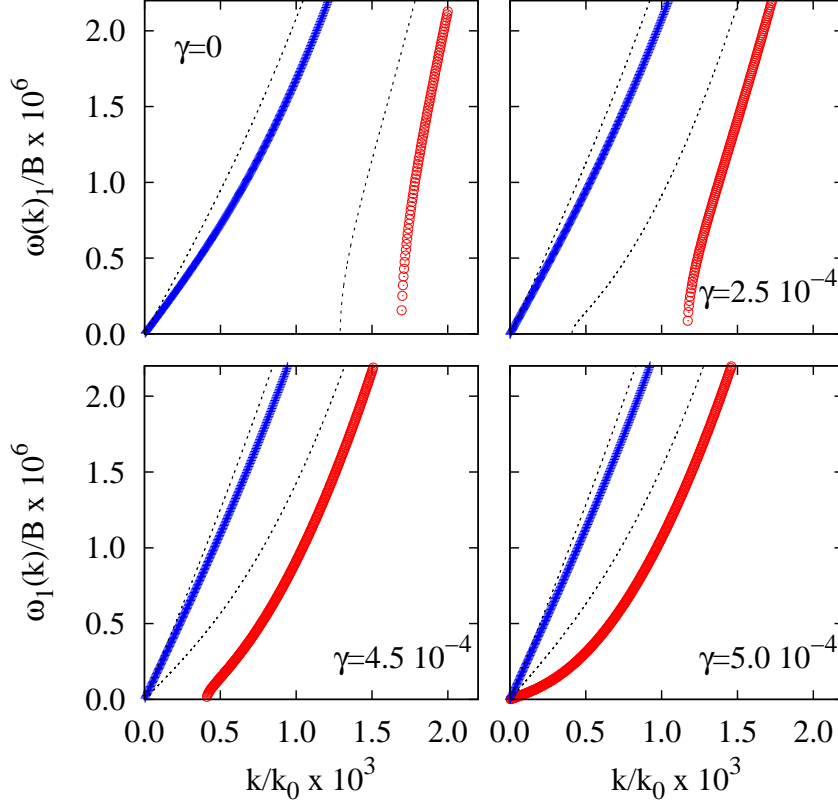


FIG. 3. Energy spectrum  $\omega_1(\mathbf{k})$  of a homogeneous Bose gas of rotating dipoles for dipolar coupling strength  $c_d = 0.3$ , in a weak electric field of strength  $\varepsilon/B = 5 \times 10^{-3}$ , for four different short range repulsion parameters  $\gamma \equiv \frac{gg}{B} = 0; 0.015; 0.030; 0.035$ . The dispersion relations for perpendicular and parallel wave vector  $\mathbf{k}$  are shown in red and blue, respectively. Dotted lines show the corresponding spectra  $\epsilon(k)$ , eq. (4), of non-rotating dipoles polarized in the  $z$ -direction, with average dipole moment  $\langle \mathbf{d} \rangle$  and the same values of  $g$ .

strength  $g$  is required to stabilize a DMBG with active rotational degrees of freedom.

The inequality  $\omega_1(\mathbf{k}) < \epsilon(\mathbf{k})$  evident in Figs. 2 and 3 has another consequence. Fitting the excitation energy  $\omega_1(\mathbf{k})$  to the form of  $\epsilon(\mathbf{k})$ , using  $\langle d \rangle$  as a fit parameter is inconsistent with the anisotropy of the excitations. For example for perpendicular  $\mathbf{k}$ , we could indeed obtain an excellent fit with an effective  $\langle d \rangle$  value that is larger than the true value. However, according to eq. (4), such a larger value of  $\langle d \rangle$  increases  $\epsilon(\mathbf{k})$  for *parallel*  $\mathbf{k}$ , moving away from  $\omega_1(\mathbf{k})$ . Conversely, we can choose to fit the parallel dispersion relation (giving an effective lower  $\langle d \rangle$ ), but then the perpendicular dispersion is not fit well. Thus it is not possible to fit  $\epsilon(\mathbf{k})$  to  $\omega_1(\mathbf{k})$  consistently for all directions of  $\mathbf{k}$  with a single fit parameter  $\langle d \rangle$ .



## V. CONCLUSIONS

In this work we have calculated the excitation energies of a dilute gas of translating and rotating dipolar molecules in a mean field approximation. We analyzed the excitation spectrum by constructing the Bogoliubov-deGennes (BdG) equations for the coupled translational and rotational excitations from the linearized Gross-Pitaevskii mean field equation for a homogeneous dipolar gas. We calculated the rotation-phonon excitations for molecules interacting with both the long range dipole-dipole potential and a repulsive short range potential, modeled here as a simple isotropic pseudo-potential.

Analysis of the unpolarized case in the absence of an external field, showed that only the  $L = 1$  rotational states of the molecules are coupled by the dipole-dipole interaction. Adding a finite short range interaction is seen to modify only the  $L = 0$  spectrum, when this short range interaction is approximated by an isotropic pseudopotential. Consequently all states with  $L \geq 2$  are unaffected by both short and long range interactions. The interesting case is  $L = 1$  for which the BdG equations have an analytic solution with one singly degenerate ( $M = 0$ ) and one doubly degenerate ( $M = \pm 1$ ) energy level. In the  $k = 0$  limit where the excitations are purely rotational, the resulting pure dipolar splitting of the  $L = 1$  rotation states is found to be proportional to the density  $\rho$ . For realistic molecular dipoles and densities, the relevant energy splitting is in the range of radio frequencies. This splitting might be of experimental relevance since it could be used as a probe of the density of a dipolar molecular Bose gas.

We also considered the polarized case where an external field leads to a partial alignment of the molecules, resulting in a non-vanishing average dipole moment  $\langle \mathbf{d} \rangle$  parallel to the field. Here we found that the BdG excitations of the homogeneous Bose gas of rotating dipoles lie below the corresponding fully polarized excitations  $\epsilon(\mathbf{k})$  for dipoles oriented along  $\mathbf{z}$  with effective dipole moment  $\langle \mathbf{d} \rangle$ . Just as for fixed dipoles, in the absence of short range repulsion, we found that an arbitrarily small external field acting on the dipoles will lead to a collapse of the system that is driven by the long wavelength phonon excitations in the direction perpendicular to the field. Thus we conclude that the rotational zero-point motion is not sufficient to stabilize the pure dipolar system against collapse. However, adding a finite short range interaction was seen to move the onset of instability down to smaller wavenumbers and to remove it entirely when the short range interaction strength, measured here by  $g$ , is

large enough. Thus, as in the full polarization approximation,  $g$  can be tuned to stabilize the DMBG. However we expect that such tuning will be very system specific and non-trivial for molecules, unlike the case of atoms where  $g$  may be generically tuned with a magnetic field.

Additional factors contribute to the stabilization of a DMBG in situations other than the homogenous setting considered here. It is well known that cold atomic gases with purely attractive interactions may be stabilized by spatial confinement up to a critical occupation number<sup>45</sup>. Similarly, for cold atoms with magnetic dipole moments it has been confirmed experimentally that they can be stabilized by appropriate trap geometries, with an oblate trap geometry typically imparting more stability<sup>3</sup>. The dipolar interaction drives dipoles towards head-to-tail orientations, which would lead to an elongation of the density distribution, and eventually to a mean attraction so strong as to lead to a collapse. A sufficiently strong trap in the polarization direction will prevent this elongation and thus can protect a dipolar gas from the associated collapse<sup>21</sup>. With new experiments on quantum gases of RbCs molecules coming online<sup>46</sup>, further extension of the current work to analysis of *inhomogeneous*, i.e., trapped, DMBGs of unpolarized and polarized molecules is worthwhile. It is straightforward to derive the corresponding GP equation and BdG equations using the procedures outlined here, although solving them numerically will be computationally more expensive than for the homogeneous situation for which solutions were derived in this work. Finally, it is important to test the predictions made here using the mean field approach by comparison with methods that go beyond the restriction of the mean field approximation. We have recently developed a path integral ground state Monte Carlo code for molecules possessing both rotational and translational degrees of freedom that allows analysis beyond the mean field approximation<sup>47</sup>. Within this approach excitations may be accessed by evaluation of imaginary time correlation functions. An alternative approach to go beyond mean field is to employ quantum many-body methods. A promising option here is the hypernetted-chain Euler-Lagrange method, which has recently been shown by one of us to be capable of describing ground and excited states of polarized dipolar quantum gases in the strong correlation regime<sup>26</sup>. Generalization of this approach to include rotational degrees of freedom will be made in future work.

## ACKNOWLEDGMENTS

KBW thanks the Physics Department of the Johannes Kepler University of Linz, Austria, for a visiting research fellowship. REZ was supported by the Austrian Science fund (FWF) project P23535. We thank Roman Krems for useful discussions.

## Appendix A: Dipole-Dipole Interaction

Since the dipole-dipole interaction  $V$ , eq. (1), enters the GP equation as a convolution product, we need to calculate the Fourier transform of  $V$ . We express the scalar product in terms of spherical harmonics,  $\Omega_i \cdot \hat{\mathbf{r}} = \frac{4\pi}{3} \sum_m Y_{1m}(\Omega_i) Y_{1m}^*(\Omega_r)$ , and obtain

$$V = \frac{1}{r^3} \frac{4\pi}{3} \sum_m Y_{1m}(\Omega_1) Y_{1m}^*(\Omega_2) - \frac{3}{r^3} \left( \frac{4\pi}{3} \right)^2 \sum_m Y_{1m}(\Omega_1) Y_{1m}^*(\Omega_r) \sum_{m'} Y_{1m'}^*(\Omega_2) Y_{1m'}(\Omega_r) \quad (\text{A1})$$

The Fourier transform of the first term alone is not defined (the integral over  $\frac{1}{r^3}$  diverges), but the divergent term will be cancelled by parts of the second term. We Fourier transform the second term, by expanding the plane wave in spherical harmonics

$$\begin{aligned} V_{\mathbf{k}}^{(2)}(\Omega_1, \Omega_2) &= -3 \left( \frac{4\pi}{3} \right)^2 \int d^3r e^{-i\mathbf{k}\mathbf{r}} \frac{1}{r^3} \sum_m Y_{1m}(\Omega_1) Y_{1m}^*(\Omega_r) \sum_{m'} Y_{1m'}^*(\Omega_2) Y_{1m'}(\Omega_r) \\ &= -3 \left( \frac{4\pi}{3} \right)^2 4\pi \sum_{\lambda, m''} \sum_{mm'} (-i)^\lambda C_\lambda Y_{\lambda m''}(\Omega_k) Y_{1m}(\Omega_1) Y_{1m'}(\Omega_2) \begin{bmatrix} \lambda & 1 & 1 \\ m'' & m & -m' \end{bmatrix} (-1)^{m'} \end{aligned}$$

where  $C_\lambda \equiv \int dx \frac{j_\lambda(x)}{x}$  and the bracket denotes an integral over three (complex conjugates of) spherical harmonics

$$\begin{aligned} \begin{bmatrix} \ell_1 & \ell_2 & \ell_3 \\ m_1 & m_2 & -m_3 \end{bmatrix} &\equiv \int d\Omega Y_{\ell_1, m_1}(\Omega) Y_{\ell_2, m_2}(\Omega) Y_{\ell_3, m_3}^*(\Omega) \\ &= \sqrt{\frac{(2\ell_1 + 1)(2\ell_2 + 1)(2\ell_3 + 1)}{4\pi}} \begin{pmatrix} \ell_1 & \ell_2 & \ell_3 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} \ell_1 & \ell_2 & \ell_3 \\ m_1 & m_2 & -m_3 \end{pmatrix} (-1)^{m_3} \end{aligned}$$

Due to selection rules, only  $\lambda = 0, 2$  contribute. For the  $\lambda = 0$  term we get  $-4\pi C_0 P_1(\Omega_1 \cdot \Omega_2)$ , which indeed cancels the Fourier transform of the first term in eq. (A1). Hence, the Fourier transform of  $V$  is ( $C_2 = \frac{1}{3}$ )

$$V_{\mathbf{k}}(\Omega_1, \Omega_2) = \frac{d^2}{4\pi\epsilon_0} \frac{(4\pi)^3}{9} \sum_{mm'm''} Y_{2m''}(\Omega_k) Y_{1m}(\Omega_1) Y_{1m'}^*(\Omega_2) \begin{bmatrix} 2 & 1 & 1 \\ m'' & m & -m' \end{bmatrix} \quad (\text{A2})$$

$V_{\mathbf{k}}(\Omega_1, \Omega_2)$  depends on the direction of  $\mathbf{k}$  (but not the magnitude  $k$ ) and the two orientations  $\Omega_1$  and  $\Omega_2$ .

A special case of (A2) is the Fourier transform of the interaction between *aligned* dipoles  $\Omega_1 = \Omega_2 = \mathbf{e}_z$

$$V_{\mathbf{k}} = \frac{d^2}{4\pi\epsilon_0} \frac{4\pi}{3} (3 \cos^2 \theta_k - 1) \quad (\text{A3})$$

that can also be obtained directly, without a detour via non-aligned dipoles<sup>37</sup>.

- 
- <sup>1</sup> Axel Griesmaier, Jörg Werner, Sven Hensler, Jürgen Stuhler, and Tilman Pfau. Bose-Einstein condensation of chromium. *Phys. Rev. Lett.*, 94:160401, 2005.
  - <sup>2</sup> Axel Griesmaier, Jürgen Stuhler, Tobias Koch, Marco Fattori, Tilman Pfau, and Stefano Giovanazzi. Comparing contact and dipolar interactions in a bose-einstein condensate. *Phys. Rev. Lett.*, 97:250402, 2006.
  - <sup>3</sup> T. Lahaye, T. Koch, B. Fröhlich, M. Fattori, J. Metz, A. Griesmaier, S. Giovanazzi, and T. Pfau. Strong dipolar effects in a quantum ferrofluid. *Nature*, 448:672, 2007.
  - <sup>4</sup> T. Koch, T. Lahaye, J. Metz, B. Fröhlich, A. Griesmaier, and T. Pfau. Stabilization of a purely dipolar quantum gas against collapse. *Nature Physics*, 4:218, 2008.
  - <sup>5</sup> Jeremy M. Sage, Sunil Sainis, Thomas Bergeman, and David DeMille. Optical production of ultracold polar molecules. *Phys. Rev. Lett.*, 94:203001, 2005.
  - <sup>6</sup> K. Winkler, F. Lang, G. Thalhammer, P. v. d. Straten, R. Grimm, and J. Hecker Denschlag. Coherent optical transfer of Feshbach molecules to a lower vibrational state. *Phys. Rev. Lett.*, 98:043201, 2007.
  - <sup>7</sup> F. Lang, K. Winkler, C. Strauss, R. Grimm, and J. Hecker Denschlag. Ultracold triplet molecules in the rovibrational ground state. *Phys. Rev. Lett.*, 101:133005, 2008.
  - <sup>8</sup> Johann G. Danzl, Elmar Haller, Mattias Gustavsson, Manfred J. Mark, Russell Hart, Nadia Bouloufa, Olivier Dulieu, Helmut Ritsch, and Hanns-Christoph Nägerl. Quantum gas of deeply bound ground state molecules. *Science*, 321:1062, 2008.
  - <sup>9</sup> S. Ospelkaus, A. Pe'er, K.-K. Ni, J. J. Zirbel, B. Neyenhuis, S. Kotochigova, P. S. Julienne, J. Ye, and D. S. Jin. Efficient state transfer in an ultracold dense gas of heteronuclear molecules. *Nature Physics*, 4:622, 2008.

- <sup>10</sup> K.-K. Ni, S. Ospelkaus, M. H. G. de Miranda, A. Pe'er, B. Neyenhuis, J. J. Zirbel, S. Kotochigova, P. S. Julienne, D. S. Jin, and J. Ye. A high phase-space-density gas of polar molecules. *Science*, 322:231, 2008.
- <sup>11</sup> A.-C. Voigt, M. Taglieber, L. Costa, T. Aoki, W. Wieser, T. W. Hänsch, and K. Dieckmann. Ultracold heteronuclear fermi-fermi molecules. *Phys. Rev. Lett.*, 102:020405, 2009.
- <sup>12</sup> Johann G. Danzl, Manfred J. Mark, Elmar Haller, Mattias Gustavsson, Russell Hart, Jesus Aldegunde, Jeremy M. Hutson, and Hanns-Christoph Nägerl. An ultracold high-density sample of rovibronic ground-state molecules in an optical lattice. *Nat. Phys.*, 6:265, 2010.
- <sup>13</sup> K.-K. Ni, S. Ospelkaus, D. Wang, G. Quemener, B. Neyenhuis, M. H. G. de Miranda, J. L. Bohn, J. Ye, and D. S. Jin. Dipolar collisions of polar molecules in the quantum regime. *Science*, 464:1324, 2010.
- <sup>14</sup> Krzysztof Goral, Kazimierz Rzazewski, and Tilman Pfau. Bose-einstein condensation with magnetic dipole-dipole forces. *Phys. Rev. A*, 61:051601, 2000.
- <sup>15</sup> Krzysztof Goral and L. Santos. Ground state and elementary excitations of single and binary bose-einstein condensates of trapped dipolar gases. *Phys. Rev. A*, 66:023613, 2002.
- <sup>16</sup> D. H. J. O'Dell, S. Giovanazzi, and G. Kurizki. Rotons in gaseous bose-einstein condensates irradiated by a laser. *Phys. Rev. Lett.*, 90:110402/1–4, 2003.
- <sup>17</sup> L. Santos, G.V. Shlyapnikov, and M. Lewenstein. Roton-maxon spectrum and stability of trapped dipolar bose-einstein condensates. *Phys. Rev. Lett.*, 90:250403/1–4, 2003.
- <sup>18</sup> S. Ronen, D. C. E. Bortolotti, D. Blume, and J. L. Bohn. Dipolar bose-einstein condensates with dipole-dependent scattering length. *Phys. Rev. A*, 74:033611, 2006.
- <sup>19</sup> S. Ronen, D. C. E. Bortolotti, , and J. L. Bohn. Bogoliubov modes of a dipolar condensate in a cylindrical trap. *Phys. Rev. A*, 74:013623, 2006.
- <sup>20</sup> L. Santos and T. Pfau. Spin-3 chromium bose-einstein condensates. *Phys. Rev. Lett.*, 96:190404, 2006.
- <sup>21</sup> S. Ronen, D. C. E. Bortolotti, , and J. L. Bohn. Radial and angular rotons in trapped dipolar gases. *Phys. Rev. Lett.*, 98:030406, 2007.
- <sup>22</sup> Ryan M. Wilson Shai Ronen, John L. Bohn, and Han Pu. Manifestations of the roton mode in dipolar bose-einstein condensates. *Phys. Rev. Lett.*, 100:245302/1–4, 2008.
- <sup>23</sup> H. P. Buchler, E. Demler, M. Lukin, A. Micheli, N. Prokofiev G. Pupillo, and P. Zoller. Strongly correlated 2d quantum phases with cold polar molecules: Controlling the shape of the interaction

- potential. *Phys. Rev. Lett.*, 98:060404/1–4, 2007.
- <sup>24</sup> G. E. Astrakharchik, J. Boronat, I. L. Kurbakov, and Yu. E. Lozovik. Quantum phase transition in a two-dimensional system of dipoles. *Phys. Rev. Lett.*, 98:060405, 2007.
- <sup>25</sup> F. Mazzanti, R. E. Zillich, G. Astrakharchik, and J. Boronat. Dynamics of a two-dimensional system of quantum dipoles. *Phys. Rev. Lett.*, 101:110405, 2009.
- <sup>26</sup> D. Hufnagl, E. Krotscheck, and R. E. Zillich. Polarized dipolar Bose gas with strong interactions. *J. of Low Temp. Phys.*, 158:85–91, 2010.
- <sup>27</sup> B. Capogrosso-Sansone, C. Trefzger, M. Lewenstein, P. Zoller, and G. Pupillo. Quantum phases of cold polar molecules in 2d optical lattices. *Phys. Rev. Lett.*, 104:125301, 2010.
- <sup>28</sup> L. Pollet, J. D. Picon, H. P. Büchler, , and M. Troyer. Supersolid phase with cold polar molecules on a triangular lattice. *Phys. Rev. Lett.*, 104:125302, 2010.
- <sup>29</sup> Franco Dalfovo, Stefano Giorgini, Lev P. Pitaevskii, and Sandro Stringari. Theory of Bose-Einstein condensation in trapped gases. *Rev. Mod. Phys.*, 71:463, 1999.
- <sup>30</sup> Jens O. Andersen. Theory of the weakly interacting Bose gas. *Rev. Mod. Phys.*, 76:599, 2004.
- <sup>31</sup> Anna Posazhennikova. Weakly interacting, dilute Bose gases in 2D. *Rev. Mod. Phys.*, 78:1111, 2006.
- <sup>32</sup> Anthony J. Leggett. Bose-Einstein condensation in the alkali gases: Some fundamental concepts. *Rev. Mod. Phys.*, 73:307, 2001.
- <sup>33</sup> S. Yi and L. You. Trapped atomic condensates with anisotropic interactions. *Phys. Rev. A*, 61:041604, 2000.
- <sup>34</sup> A. Derevianko. Anisotropic pseudopotential for polarized dilute quantum gases. *Phys. Rev. A*, 67:033607, 2003.
- <sup>35</sup> C. Ticknor. Energy dependence of scattering ground-state polar molecules. *Phys. Rev. A*, 76:052703, 2007.
- <sup>36</sup> J. L. Bohn, M. Cavagnero, and C. Ticknor. Quasi-universal dipolar scattering in cold and ultracold gases. *New J. Phys.*, 11:055039, 2009.
- <sup>37</sup> T. Lahaye, C. Menotti, L. Santos, M. Lewenstein, and T. Pfau. The physics of dipolar bosonic quantum gases. *Rep. Prog. Phys.*, 72:126401, 2009.
- <sup>38</sup> B. Friedrich and D.R. Herschbach. On the possibility of orienting rotationally cooled polar molecules in an electric field. *Z. Phys. D*, 18:153, 1991.
- <sup>39</sup> N. N. Bogoliubov. *J. Phys. (USSR)*, 11:23, 1947.

- <sup>40</sup> L. P. Pitaevskii and S. Stringari. *Bose-Einstein Condensation*. Oxford University Press, 2003.
- <sup>41</sup> R. E. Zillich and K. B. Whaley. Rotational spectra of methane and deuterated methane in helium. *J. Chem. Phys.*, 132:174501, 2010.
- <sup>42</sup> P. W. Anderson. Pressure broadening in the microwave and infra-red regions. *Phys. Rev.*, 76:647, 1949.
- <sup>43</sup> C. H. Townes and A. L. Schawlow. *Microwave Spectroscopy* Dover Publications, 1975.
- <sup>44</sup> J. H. Van Vleck. The dipolar broadening of magnetic resonance lines in crystals. *Phys. Rev.*, 74:1168, 1948.
- <sup>45</sup> J. M. Gerton, D. Strekalov, I. Prodan, and R. G. Hulet. Direct observation of growth and collapse of a Bose-Einstein condensate with attractive interactions. *Nature*, 408:692, 2000.
- <sup>46</sup> R. Grimm, private communication.
- <sup>47</sup> B. P. Abolins, R. E. Zillich, K. B. Whaley. ground state Monte Carlo approach for study of dipolar systems with rotational degrees of freedom submitted to *J. Low Temp. Phys.*, 2011.