

Dipolar Bilayer with Antiparallel Polarization – a Self-Bound Liquid

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Dipolar bilayers with antiparallel polarization, i.e. opposite polarization in the two layers, exhibit liquid-like rather than gas-like behavior. In particular, even without external pressure a self-bound liquid droplet of constant density will form. We investigate the symmetric case of two identical layers, corresponding to a two-component Bose system with equal partial densities. The zero-temperature equation of state $E(\rho)/N$, where ρ is the total density, has a minimum, with an equilibrium density that can be adjusted by the distance d between the layers (decreasing with increasing d). The attraction necessary for a self-bound liquid comes from the interlayer dipole-dipole interaction that leads to a mediated intralayer attraction. We investigate the regime of negative pressure towards the spinodal instability, where the bilayer is unstable against infinitesimal fluctuations of the total density, confirmed by calculations of the speed of sound of total density fluctuations.

I. INTRODUCTION

Experiments with Bose gases of atoms with large magnetic moments ($^{52}\text{Cr}^{1,2}$, $^{164}\text{Dy}^3$, $^{168}\text{Er}^4$) are fueling the interest to understand the effects of the dipole-dipole interaction (DDI) on stability, shape, and dynamics of dipolar Bose condensates (reviewed in Refs. 5–7). The strength of the DDI is commonly characterized by the dipole length $r_D = mD^2/(4\pi\epsilon_0\hbar^2)$, where m is the mass of the dipolar atom or molecule, and D is its dipole moment. The value of r_D can be compared with the average inter-particle spacing, $r_s \sim \rho^{-1/m}$, where ρ is the number density of the condensate and m the dimensionality. For $r_D \ll r_s$, the DDI is weak; in general, other contributions to the interaction, such as the s -wave scattering length a , will dominate (except if a is tuned to a sufficiently small value²). For $r_D \gtrsim r_s$, the DDI will be the dominant interaction. The magnetic DDI is usually negligible, only for the handful of atoms mentioned above, its effect has been observed, as it is difficult to increase the density such that $r_D \gtrsim r_s$. Compared with the magnetic dipole moment of atoms, the electric dipole moment of heteronuclear molecules can be orders of magnitude larger, leading to large values for r_D (e.g. $r_D = 5 \times 10^5 \text{Å}$ for a fully polarized NaCs). Association of two atoms using a Feshbach resonance and transfer to the rovibrational ground state has been achieved for example for $^7\text{Li}^{133}\text{Cs}^8$, $^{40}\text{K}^{87}\text{Rb}^9$, $^{41}\text{K}^{87}\text{Rb}^{10}$ and $^{85}\text{Rb}^{133}\text{Cs}^{11,12}$. But it is quite a challenge to produce a degenerate quantum gas of dipolar molecules.

The anisotropy of the DDI leads to a measurable anisotropy of the speed of sound¹³, but also an anisotropic superfluid response¹⁴ has been predicted. The attractive part of the DDI can give rise to roton or roton-like excitations in a dipolar Bose gas layer^{15–19}. An anisotropic 2D quantum gas can be realized by tilting the polarization dipoles in a deep 1D trap, and a stripe phase can form spontaneously^{20,21}. For $r_D \gg r_s$, dipoles will crystallize without imposing an optical lattice^{22–25}. Fermionic dipoles in 1D, polarized along the length of the system, can be self-bound as has been shown us-

ing mean field theory²⁶. A bilayered dipolar Bose gas can dimerize if the polarization direction in the two layers is the same²⁷. Also glassy behavior has been studied for antiparallel polarization in two layers, i.e. when dipoles are perpendicular to the layer, but the orientation of the dipoles in one layer is opposite to that in the other layer²⁸. Like the DDI corresponding to parallel dipoles, the DDI corresponding to antiparallel dipoles can be realized by static electric and/or microwave fields. For example, one can apply a static electric field \mathbf{E} and excite the molecules in one layer to rotational states $(J, M) = (1, \pm 1)$, which have an effective polarization parallel to \mathbf{E} . The molecules in the other layer, preferably a different species with different rotational constant, can be excited to rotational states $(J, M) = (1, 0)$, which are effectively polarized opposite to \mathbf{E} . Thus molecules in the same layer are subject to a repulsive DDI at long range, and molecules in different layers to an attractive DDI. Details about DDI interaction in static electric field can be found in Ref. 29, where also alternative approaches using microwave fields are worked out.

In this work we study such a bilayer of bosonic dipoles with antiparallel polarization. The key result is that it is a self-bound liquid: unlike a gas, that expands if there are no walls or a trap potential that exerts external pressure, a liquid is bound together by the interaction between its particles – hence the interaction must be at least partially attractive. The DDI is clearly partially attractive, depending on the orientations of the dipoles relative to the distance vector between them. However this is not enough to ensure a self-bound liquid: as mentioned above, dipolar bilayers with the same polarization in both layers undergo dimerization below a certain layer distance²⁷; yet, the system as a whole is not self-bound. We show that the liquid nature is a consequence of the particular form of the interlayer DDI for opposite polarization, which is attractive for finite separation (the 2D separation projected on the bilayer planes). This leads to cohesion due to “dipole bridges” that effectively act as a glue to bind all particles together. As required for a liquid, the densities in the bilayers adjust to an equilibrium density ρ_{eq} in the absence of external pressure. ρ_{eq}

can be adjusted over a very wide range by the distance d between the layers.

For our calculations we use a variational many-body theory, the hypernetted-chain Euler-Lagrange method (HNC-EL), which includes optimized pair correlations. The HNC-EL method has been demonstrated to give accurate results even for such dense and strongly correlated quantum liquids as superfluid ^4He . For comparison and validation, we use path integral Monte Carlo (PIMC) simulations.

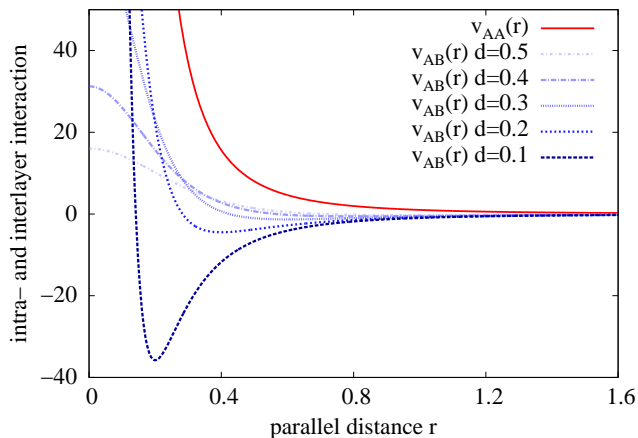


FIG. 1. (Color online) Intralayer interaction $v_{AA}(r) = v_{BB}(r)$ (full line) and interlayer interaction $v_{AB}(r)$ (dashed lines) for $d = 0.1, \dots, 0.5$, as function of the parallel distance, i.e. the distance projected on the layer planes.

II. METHODOLOGY

A 1D optical lattice slices a BEC into quasi-2D layers separated by a distance d . Since the dipole length r_D can easily exceed the typical d value of about 500nm, the DDI interaction between dipoles in different layers can lead to appreciable coupling. We consider here two translationally invariant layers A and B , approximate each layer as two-dimensional, and assume no tunneling occurs. With these simplifications we get two coupled 2D systems, i.e. a binary Bose mixture. For simplicity we assume the particles in the two layers have the same mass and dipole moment. As common in this field, we use dimensionless quantities, where length is given in units of the dipole length r_D and energy in units of the dipole energy $E_D = \hbar^2/(mr_D^2)$. Other dimensionless quantities follow from r_D and E_D , e.g. the number density is given in units of r_D^{-2} etc. The Hamiltonian in dipole units is

$$H = -\frac{1}{2} \sum_{\alpha,i} \nabla_{i,\alpha}^2 + \frac{1}{2} \sum_{\alpha,\beta} \sum'_{i,j} v_{\alpha,\beta}(|\mathbf{r}_{i,\alpha} - \mathbf{r}_{j,\beta}|).$$

α and β index the layer, $\alpha, \beta \in \{A, B\}$, and i the particles within a layer. The primed sum indicates that for

$\alpha = \beta$ we only sum over $i \neq j$. Note that all coordinates $\mathbf{r}_{i,\alpha}$ are 2D projections on the layer plane; the z -coordinate is either 0 or d in the layer A or B , and this d -dependence is integrated into the interaction $v_{\alpha,\beta}$. $v_{\alpha,\beta}(|\mathbf{r}_{i,\alpha} - \mathbf{r}_{j,\beta}|)$ is the DDI, in units of E_D , between dipole i at $\mathbf{r}_{i,\alpha}$ in layer α and dipole j at $\mathbf{r}_{j,\beta}$ in layer β . We neglect short-ranged interactions compared to the DDI. The intralayer interaction ($\alpha = \beta$) is purely repulsive, $v_{\alpha,\alpha}(r) = 1/r^3$. The interlayer interaction, $\alpha \neq \beta$, is $v_{AB}(r) = (2d^2 - r^2)/(d^2 + r^2)^{5/2}$, which is repulsive for small r , but attractive for large r , and has a minimum at $r_{\min} = 2d$. We show $v_{AA}(r) = v_{BB}(r)$ in Fig. 1 with a full line and, for layer distances $d = 0.1, \dots, 0.5$, $v_{AB}(r)$ with dashed lines. Although longer-ranged, $v_{AB}(r)$ looks qualitatively similar to the interaction between two neutral atoms: an attractive well, followed by repulsion for small r . Note that, since the average interlayer interaction vanishes, $\int d^2r v_{AB}(r) = 0$, the coupling between layers in the ground state would vanish in a mean field approximation and the ground state energy would just be the sum of the energies of each layer.

For the many-body ground state we use the variational Jastrow-Feenberg ansatz³⁰ consisting of a product of pair correlation functions for a multi-component Bose system,

$$\Psi_0 = \exp \left[\frac{1}{4} \sum_{\alpha,\beta} \sum'_{i,j} u_{\alpha,\beta}(|\mathbf{r}_{i,\alpha} - \mathbf{r}_{j,\beta}|) \right] \quad (1)$$

Higher order correlations $u_{\alpha,\beta,\gamma}(\mathbf{r}_{i,\alpha}, \mathbf{r}_{j,\beta}, \mathbf{r}_{k,\gamma})$ could be included, as is routinely done for single-component calculations. Past experience has shown that triplet correlations improve the ground state energy, leading to results very close to exact QMC simulations, but they do not change the qualitative picture. We therefore restrict ourselves to pair correlations, but check the results against PIMC simulations (details about the PIMC simulations are given in the appendix). The expression for the expectation value of the energy, $E \equiv \langle \Psi_0 | H | \Psi_0 \rangle / \langle \Psi_0 | \Psi_0 \rangle$, contains $u_{\alpha,\beta}(r)$, but also the pair distribution function $g_{\alpha,\beta}(r)$, defined as

$$g_{\alpha,\beta}(|\mathbf{r}_\alpha - \mathbf{r}_\beta|) = \frac{N_\alpha(N_\beta - \delta_{\alpha\beta})}{\rho_\alpha \rho_\beta} \int d\tau_{\alpha,\beta} |\Psi_0|^2$$

where the integral is over all particles except one in layer α and one in layer β , and $\rho_\alpha = N_\alpha/V$ is the partial density of component α . $u_{\alpha,\beta}(r)$ and $g_{\alpha,\beta}(r)$ are of course related, via the exact hypernetted-chain equations³¹,

$$g_{\alpha,\beta}(r) = e^{u_{\alpha,\beta}(r) + N_{\alpha,\beta}(r) + E_{\alpha,\beta}(r)}.$$

The sum of all so-called “nodal diagrams”, $N_{\alpha,\beta}(r)$, is itself related to $g_{\alpha,\beta}(r)$ but the multi-component generalization of the Ornstein-Zernicke relation³¹, which would provide closure to the equations, where it not for the sum of all “elementary diagrams”, $E_{\alpha,\beta}(r)$. The latter can only be computed approximately, e.g. by truncating the sum. Here, we simply neglect them completely; what we said about neglecting triplet correlations also applies to neglecting elementary diagrams.

The pair distributions $g_{\alpha,\beta}(r)$ (and thus the energy and other quantities of interest) are determined from Ritz' variational principle, i.e. from the Euler-Lagrange equations, $\delta E/\delta g_{\alpha,\beta}(r) = 0$. These coupled, nonlinear equations are generically called hypernetted-chain Euler-Lagrange (HNC-EL) equations, and called HNC-EL/0 if the elementary diagrams are neglected as in the present work. Details about the HNC-EL method can be found in reviews^{32,33}. Particularly for Bose mixtures the HNC-EL/0 equations for an arbitrary number of components can be cast into the following form (bold-faced capital letters denote matrices), see Refs. 34–37:

$$\begin{aligned} \mathbf{W}(k) &= -\frac{1}{2} \left[\mathbf{S}(k)\mathbf{T}(k) + \mathbf{T}(k)\mathbf{S}(k) - 3\mathbf{T}(k) \right. \\ &\quad \left. + \mathbf{S}^{-1}(k)\mathbf{T}(k)\mathbf{S}^{-1}(k) \right] \\ V_{\alpha,\beta}^{ph}(r) &= g_{\alpha,\beta}(r)v_{\alpha,\beta}(r) + \frac{\hbar^2}{2m_{\alpha,\beta}} \left| \nabla \sqrt{g_{\alpha,\beta}(r)} \right|^2 \\ &\quad + (g_{\alpha,\beta}(r) - 1)W_{\alpha,\beta}(r) \\ \mathbf{V}^{ph}(k) &= \mathbf{S}^{-1}(k)\mathbf{T}(k)\mathbf{S}^{-1}(k) - \mathbf{T}(k) \end{aligned}$$

where $m_{\alpha,\beta}$ is the reduced mass (for the symmetric bilayer, $\hbar^2/2m_\alpha = \hbar^2/2m_\beta = 1/2$ in dipole units). $S_{\alpha,\beta}(k)$ is the static structure function

$$S_{\alpha,\beta}(k) = \delta_{\alpha\beta} + \sqrt{\rho_\alpha \rho_\beta} \text{FT}[g_{\alpha,\beta} - 1]$$

where FT denotes Fourier transformation. The kinetic energy matrix, $T_{\alpha,\beta}(k) = \delta_{\alpha\beta}(\hbar^2 k^2/4m_{\alpha,\beta})$, becomes $T_{\alpha,\beta}(k) = \delta_{\alpha\beta} \frac{k^2}{2}$ in our case. The HNC-EL/0 equations can be solved iteratively. Usually the convergence is stable and fast, but close to an instability like the spinodal point discussed below, we use linear mixing between iterations to ensure convergence.

III. RESULTS

We calculated the ground state energy per particle, $E(\rho)/N$, as function of total density $\rho = \rho_A + \rho_B$ for different layer distances d . The interlayer DDI scales with d^{-3} , therefore the energy per particle, E/N , varies over a wide range, as can be seen in the top panel of Fig. 2 that shows $E(\rho)/N$ for four values of d . A key result is that $E(\rho)/N$ has a minimum at a certain equilibrium density $\rho_{\text{eq}}(d)$, where the pressure p vanishes: without an externally applied pressure provided e.g. by a radial trap potential, the total density of the bilayer system will adjust itself to $\rho_{\text{eq}}(d)$. Rather than expanding like a gas, a dipolar bilayer system with antiparallel polarization is a *self-bound liquid*. Despite the purely repulsive intralayer interaction, the partly attractive interlayer interaction provides the “glue” that binds the system to a liquid. The phenomenon of an effective intralayer attraction, mediated by particles in the other layer, is discussed in more detail below.

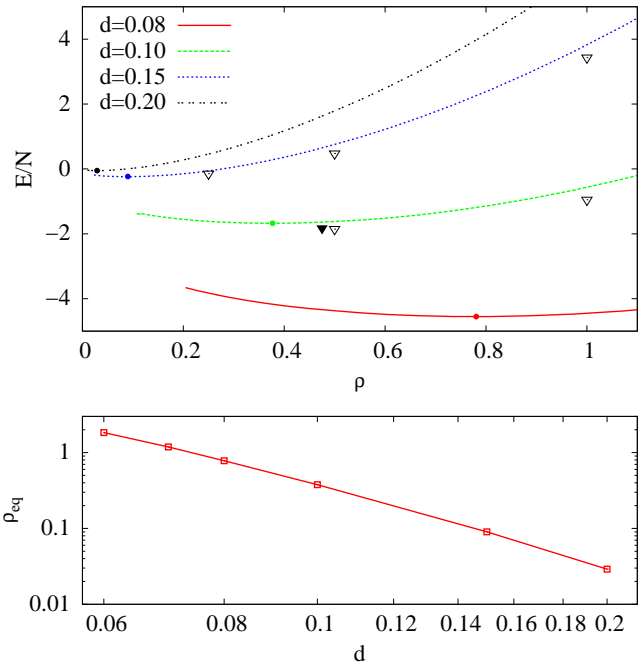


FIG. 2. (Color online) Top panel: Ground state energy per particle E/N versus the total density ρ , for several layer distances d . The results from HNC-EL/0 are shown as lines (small circles indicating the equilibrium density ρ_{eq}), the open symbols are from corresponding PIMC simulations. The filled symbol indicates E/N and ρ_{eq} estimated from a PIMC simulation of a self-bound two-dimensional droplet of 50 dipoles in each layer, see appendix. Bottom panel: equilibrium density ρ_{eq} versus d .

During the iterative numerical optimization, convergence becomes very sensitive as the density ρ or the distance d between layers A and B is decreased, until the HNC-EL/0 equations eventually fail to converge. Past experience with HNC-EL/0 is that a numerical instability usually has a physical reason. Indeed, as we will show below, there is a spinodal point at a d -dependent critical density ρ_{sp} where the homogeneous phase assumed in our calculation becomes unstable against phase separation by nucleation of two-dimensional droplets (“puddles”). Thus the equation of state $E(\rho)/N$ for a *homogeneous* phase can be calculated only for densities $\rho > \rho_{\text{sp}}$. This can be seen in the upper panel of Fig. 2 where each curve for $E(\rho)/N$ has an end point. The end point is close to the critical density ρ_{sp} (reaching $\rho = \rho_{\text{sp}}$ is impossible since already an infinitesimal fluctuation can nucleate a finite density wave, creating e.g. a bubble).

Also shown in the top panel of Fig. 2 are the energies obtained with PIMC simulations, see appendix. The temperature for the PIMC simulations is set to $T = 0.5$ ($T = 0.25$ for the smallest ρ), which is low enough that the thermal effect on E/N is smaller than the symbol size. The open triangles are bulk simulations of $N_A = N_B = 50$ dipoles with periodic boundary condi-

tions. The HNC-EL/0 results are upper bounds on E/N , consistent with a variational approach. The overall dependence of E/N on ρ and d is reproduced quite well with the HNC-EL/0 method, which is orders of magnitude faster than PIMC simulations. The black triangle shows the energy from a PIMC simulation of $N_A = N_B = 50$ dipoles and layer separation $d = 0.1$ *without* periodic boundary conditions. Due to the liquid nature of the bilayer, the dipoles in the simulation indeed coalesce into a droplet of finite density, given by $\rho_{\text{eq}}(d)$ apart from corrections due to the surface line tension. The density corresponding to the filled triangle is obtained from the radial density profile $\rho(r)$ at $r = 0$ (see Fig. 7 in the appendix) where r is defined relative to the center of mass of the droplet. Thermal evaporation, that can occur in the absence of periodic boundary conditions, was suppressed by choosing a much lower temperature of $T = 0.0625$. Although this simulation of a finite cluster is not equivalent to bulk PIMC or HNC-EL/0 calculations, the central density of the droplet is indeed close to $\rho_{\text{eq}}(d)$ from HNC-EL/0.

The equilibrium density ρ_{eq} as function of d is shown in the bottom panel of Fig. 2. $\rho_{\text{eq}}(d)$ decreases rapidly with increasing d . For smaller d , the decrease is approximately $\rho_{\text{eq}} \sim d^{-3}$ and for larger d it is closer to $\rho_{\text{eq}} \sim d^{-4}$. Based purely on the interlayer DDI $v_{AB}(r)$, one would expect a scaling of ρ_{eq} with the inverse square of $r_{\text{min}} = 2d$, leading to a scaling d^{-2} . The deviation from d^{-2} is due to the kinetic energy. Only for very small d (very deep $v_{AB}(r)$), this simple picture can be expected to be valid, and indeed the ρ_{eq} -curve becomes less steep for smaller d in the double logarithmic representation in Fig. 2. We note that in the regime of very small d , and thus of extremely strong interlayer correlations, the HNC-EL method would not be reliable anymore.

In Fig. 3 we show the intralayer and interlayer pair distributions, $g_{AA}(r)$ and $g_{AB}(r)$, in the top and middle panel for progressively smaller layer distance d up to the smallest numerically stable value $d = 0.063$, for $\rho = 1$. The growth of a strong correlation peak in $g_{AB}(r)$ as d is decreased is a direct consequence of the increasingly deep attractive well of $v_{AB}(r)$ around $r_{\text{min}} = 2d$ (see Fig. 1). The peak in $g_{AB}(r)$ is located at a distance r_m which is a somewhat larger than $2d$ due to zero-point motion. But also $g_{AA}(r)$ develops additional correlations, seen as a shoulder in the top panel. The additional correlations are best seen in the difference between $g_{AA}(r)$ and the uncoupled ($d \rightarrow \infty$) limit $g_{AA}^\infty(r)$, $\Delta g_{AA}(r) = g_{AA}(r) - g_{AA}^\infty(r)$, shown in the bottom panel. The additional positive correlation between dipoles in the *same* layer is mediated by dipoles in the *other* layer: the attraction between a dipole in layer A and a dipole in layer B, that leads to a peak at distance r_m , induces an effective attraction between the dipole in A and another dipole in A, leading to peak at about twice the distance, $2r_m$. The inset in the middle panel shows the positions of the peaks of $g_{AB}(r)$ and $\Delta g_{AA}(r)$ (indicated by circles in the plots of $g_{AB}(r)$ and $\Delta g_{AA}(r)$) as function of distance d . Indeed the peaks

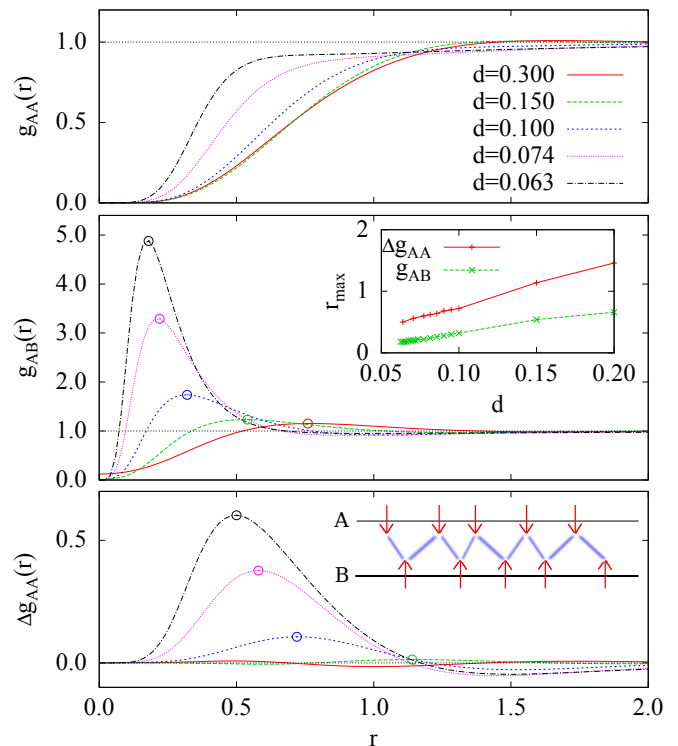


FIG. 3. (Color online) Top panel: Intralayer pair distributions $g_{AA}(r)$ at density $\rho_A = \rho_B = 0.5$ for progressively smaller layer distance d . Middle panel: Corresponding interlayer pair distributions $g_{AB}(r)$, with circle indicating the maxima of $g_{AB}(r)$. Bottom panel: incremental intralayer pair distributions $\Delta g_{AA}(r) = g_{AA}(r) - g_{AA}^\infty(r)$, i.e. the change from uncoupled layers. The inset in the middle panel shows the positions of the maxima of $g_{AA}(r)$ and $g_{AB}(r)$, respectively, as function of d . The inset in the bottom panel sketches the attractive forces between dipoles in different layers.

of $\Delta g_{AA}(r)$ are located at about twice the distance of the peaks of $g_{AB}(r)$. This effective intralayer attraction, induced by the real interlayer attraction, is illustrated by a simple 1D sketch in the inset in the bottom panel, which also illustrates a preference for a certain interparticle spacing, i.e. density, where “dipole bridges” (blue lines in the sketch) can form. The present 2D situation is more complicated than the simple 1D sketch, but our results for the pair correlations demonstrate this picture is approximately valid.

The identification of the low density instability with a spinodal point can be proven by calculating the long wavelength modes. For two coupled layers, there are two excitation modes, $\epsilon_{1,2}$ for any given wave number k , a density mode and a concentration mode. In the density oscillations, the dipoles in different layers move in phase, and in a concentration oscillation they move out of phase. In the long wavelength limit, $k \rightarrow 0$, each mode can be characterized by the speed of a density or concentration fluctuation, c_1 and c_2 , respectively. At the spinodal point, the speed of sound c_1 vanishes which means that

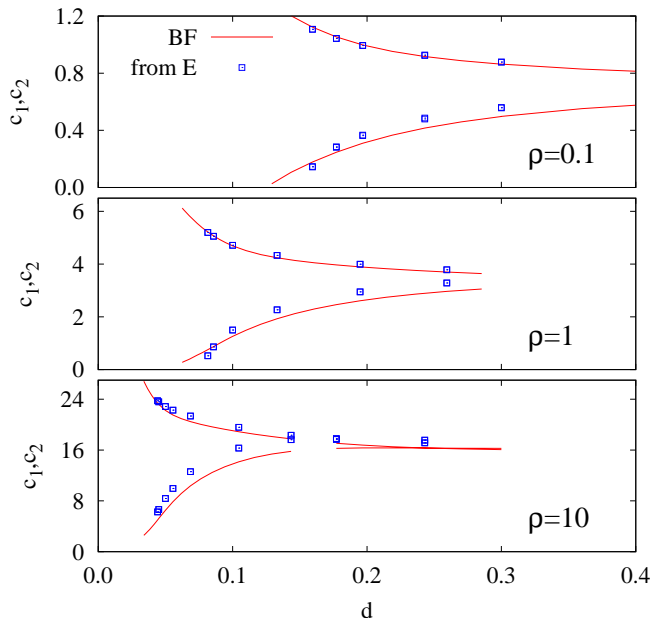


FIG. 4. (Color online) Speed of density fluctuations c_1 and speed of concentration fluctuations c_2 , as function of layer distance d for three densities, $\rho = 0.1; 1; 10$ (top, middle, and bottom panels). Full lines show the Bijl-Feynman approximation, eq. (2), and symbols are the thermodynamic estimates, eq. (3).

the system becomes unstable against infinitesimal $k \rightarrow 0$ fluctuations of the total density, triggering the spinodal decomposition: the system spatially separates a high and low density phase, such as droplets surrounded by vacuum (at $T = 0$). The easiest way to calculate c_1 and c_2 is the Bijl-Feynman approximation (BFA) for the excitation energies $\epsilon_i(k)$. In the case of a single component, the BFA excitation energy $\epsilon(k)$ follows from $\frac{k^2}{2} = \epsilon(k)S(k)$, which is of course trivial to solve; we get the well-known Bijl-Feynman approximation $\epsilon(k) = \frac{k^2}{2S(k)}$.³⁸ In the case of a multi-component Bose system we need to solve the generalized eigenvalue problem $\frac{k^2}{2}\vec{\phi} = \epsilon_i(k)\mathbf{S}(k)\vec{\phi}$, where $\mathbf{S}(k)$ is the static structure matrix introduced above. For strong correlations, the BFA gives only a rough idea of the true excitation structure, e.g. the BFA for the roton energy of superfluid ^4He is off by a factor of two. However, it describes the low momentum limit of the dispersion relation very well, which is what we need for c_i . For a symmetric bilayer, the eigenvalues are

$$\epsilon_{1,2}(k) = \frac{k^2}{2}(S_{AA}(k) \pm S_{AB}(k))^{-1}$$

with the associated eigenvectors $\vec{\phi}_1 \sim (1, 1)$ and $\vec{\phi}_2 \sim (1, -1)$. $\vec{\phi}_1$ describes fluctuations of the total density where particles in different layers move in phase, and $\vec{\phi}_2$ describes concentration fluctuations, where particles in different layers move out of phase, and the total density

is constant. For small k , $\epsilon_1(k) < \epsilon_2(k)$, i.e. the density mode has lower energy than the concentration mode. For $k \rightarrow 0$ we get

$$c_{1,2} = \frac{1}{2}(S'_{AA} \pm S'_{AB})^{-1} \quad (2)$$

with $S'_{\alpha,\beta} = dS_{\alpha,\beta}(k)/dk|_{k=0}$. For single-component Bose systems, it is known that the long-wavelength limit of $S(k)$ obtained with HNC-EL is biased by the approximation made for elementary diagrams (omitted here altogether). This leads to an inconsistency between the speed of sound c obtained from the HNC-EL approximation for $S(k)$ and the thermodynamic relation between c and the energy, $c^2 = \frac{\partial}{\partial \rho} \rho^2 \frac{\partial E}{\partial \rho N}$ (in dipole units). Therefore we need to assess the reliability of our results for c_1 and c_2 in the present case of a multi-component Bose system. We compare the BFA values obtained from $S_{\alpha,\beta}(k)$, eq. (2), with the generalization of the thermodynamic relation between $c_{1,2}$ and the energy to binary systems:

$$c_{1,2} = \sqrt{\frac{\rho}{2}(e_{AA} \mp e_{AB})} \quad (3)$$

where $e_{\alpha\beta}$ is the second derivative of E/N with respect to ρ_α and ρ_β .³⁹

In Fig. 4 the results for c_1 and c_2 obtained with eq. (2) and eq. (3), respectively, are shown as function of layer distance d for densities $\rho = 0.1; 1; 10$. As the coupling between layers is increased by reducing d , c_1 and c_2 behave differently. The speed of concentration fluctuations c_2 increase (without actually diverging) while the quantity of main interest, the speed of density fluctuations c_1 , decreases to zero, in agreement with the interpretation of the instability as a spinodal point. The critical distance where c_1 vanishes is lower for higher ρ . Thus increasing the density for a given d makes the system more stable. The BFA for $c_{1,2}$ and their thermodynamic estimates agree qualitatively, but differ especially for the interesting regime near the spinodal point where $c_1 \rightarrow 0$. The Bijl-Feynman values for c_1 appear to go to zero linearly and at slightly smaller d , while the thermodynamic estimates approach zero more steeply, possibly in a non-analytic fashion. Unfortunately, these uncertainties preclude a meaningful analysis of critical exponents for $c_1(d)$ or $c_1(\rho)$. Monte Carlo simulations, including a finite size scaling analysis, may shed more light on this question, but would certainly require very large simulations, which is beyond the scope of PIMC simulations performed for this paper.

IV. CONCLUSION

We have shown that a dipolar bilayer with antiparallel polarization in the two layers constitute a *self-bound liquid*, evidenced by a minimum of $E(\rho)/N$ at a finite density ρ_{eq} . This means the dipoles in the two layer relax to a stable equilibrium density and require no external pressure provided by a radial trapping potential. This makes

is possible to study homogeneous quantum phases, because – apart from a narrow surface region – the density is essentially constant and given by ρ_{eq} . The value of ρ_{eq} can be controlled by the distance d between the layers (measured in dipolar units), where ρ_{eq} decreases with increasing d . The intra- and interlayer pair distribution functions, $g_{AA}(r)$ and $g_{AB}(r)$, exhibit distinct peaks that show that the liquid can be understood as being held together by a network of “dipole bridges”: the attraction between dipoles in different layers mediates an effective attraction between dipoles in the same layer that overcomes the repulsion of the intralayer dipole-dipole interaction. Such situations where an inter-species attraction wins against an intra-species repulsion are common for self-bound systems. For example, in an ionic crystal the arrangement of having oppositely charged ions as close as possible (next neighbors) and equally charged ions further away (next-next neighbors) leads of course to stable crystals.

For comparison and validation of our HNC-EL/0 results we performed exact PIMC simulations, and found good agreement with our HNC-EL/0 results. Furthermore, and as expected for a liquid, the equation of state $E(\rho)/N$ ends at a critical density ρ_{sp} (*spinodal point*) below which the bilayer becomes unstable against infinitesimal long-wavelength perturbations and breaks into droplets. This is evidenced by a vanishing speed c_1 of total density fluctuations, which we determined from our HNC-EL/0 results using two independent methods, which agree quite well and only deviated from each other very close to the spinodal point. Finite size PIMC simulations confirm that the system indeed coalesces into a droplet with a flat density profile given by the equilibrium density.

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Appendix A: PIMC simulations

An important question is how accurate are our ground state results obtained with the variational HNC-EL/0 method, where we neglect elementary diagrams and higher than two-body correlations in the ansatz for the wave function, eq. (1). We performed path integral Monte Carlo (PIMC) simulations to assess the quality of our HNC-EL/0 results and found good agreement between HNC-EL/0 energies and PIMC energies at low temperature. In this appendix we present additional comparisons of the static structure matrix, as well as results for finite systems, where self-bound “puddles”, i.e. 2D droplets are formed because of the spinodal instabil-

ity. All simulations were done with 50 particles per layer; bulk simulations used quadratic simulation boxes with periodic boundary conditions with a box size adjusted to achieve a given density. For antiparallel bilayers, cut-off corrections to the dipole-dipole interaction cancel each other and therefore are not needed.

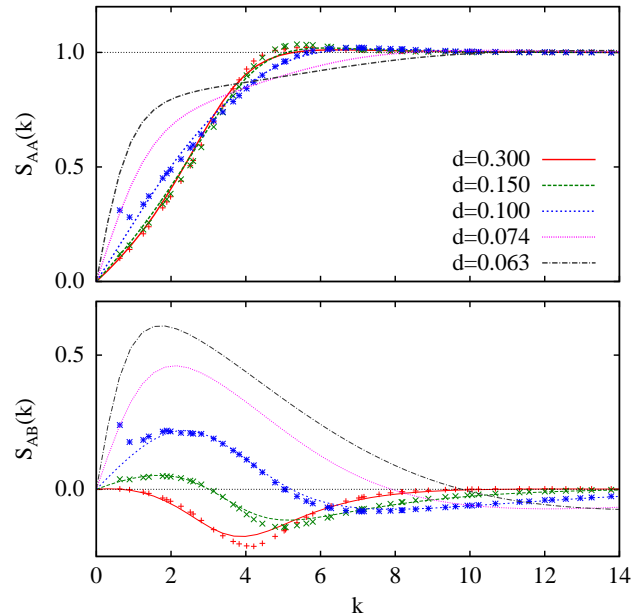


FIG. 5. (Color online) Static structure functions $S_{AA}(k)$ (upper panel) and $S_{AB}(k)$ (lower panel) obtained with HNC-EL/0 (lines) for a total density $\rho = 1$ and progressively smaller layer distances d as indicated in the upper panel. The symbols show $S_{AA}(k)$ and $S_{AB}(k)$ obtained by PIMC simulations at $T = 0.5$ for distances down to $d = 0.1$.

For predicting the spinodal instability, but also for calculating excitation properties, an important quantity is the static structure matrix, $S_{\alpha\beta}(k)$. In Fig. 5 we compare $S_{AA}(k)$ and $S_{AB}(k)$ obtained with HNC-EL/0 (lines) and PIMC (symbols), at a total density of $\rho = 1$. The temperature in the PIMC simulation was $T = 0.5$ which was low enough that $S_{\alpha\beta}(k)$ did not change upon lowering the temperature further. We see that the HNC-EL/0 approximation works well, considering that intralayer correlations are quite strong. For $d = 0.3$ and $d = 0.15$ PIMC simulations predict slightly more pronounced peaks and troughs, but HNC-EL/0 calculations are faster by several orders of magnitude. For $d = 0.1$, the agreement is also very good, except for the smallest k value possible in a simulation box of side length 10, $k = 2\pi/10 \approx 0.63$. In the PIMC results, both $S_{AA}(k)$ and $S_{AB}(k)$ turn up sharply for this smallest k value. When we reduce d even more, this apparent peak at $k = 0$ grows very large. This peak has a very simple reason: as we approach the spinodal point by reducing d , we enter the metastable regime of the phase diagram, where E/N as function of total density has a negative slope. In this regime a finite perturbation can lead to a collapse and the system

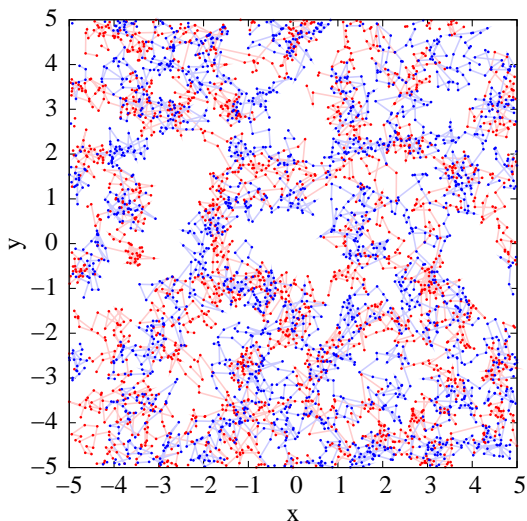


FIG. 6. (Color online) PIMC simulation snapshot for $\rho = 1$ and $d = 0.1$ ($T = 0.5$). Red and blue chains are the dipoles in layer A and B.

phase separates. Since PIMC has no trial wave function that could prevent phase separation, this collapse indeed happens as we go further below the equilibrium density. In fact, Monte Carlo snapshots such as in Fig. 6 show density fluctuations already for $d = 0.1$ that resemble

small “bubbles”, i.e. voids in the liquid dipolar bilayer. In Fig. 6 red and blue dots, connected by lines, are the beads of the discretized imaginary time paths sampled in PIMC; each bead is a particle at a discrete time step. For even lower d or lower total densities we observe a clear decomposition into a droplet and a low-density gas, as discussed in the next paragraph. A large peak at $k = 0$ can therefore be seen as a zero momentum Bragg peak due to phase separation.

As final confirmation of the liquid nature of dipolar bilayers with antiparallel polarization, we show the results of a PIMC simulation without periodic boundary conditions. Since there is no radial trapping potential, a two-dimensional gas would of course spread out indefinitely. A liquid on the other hand will coalesce into a droplet of finite density. For a droplet large enough that effects of surface line tension are negligible, the density inside the droplet is given by the equilibrium density ρ_{eq} , i.e. the density of a bulk system at zero pressure. In Fig. 7 we show the radial density profile $\rho(r)$ for 50 dipoles in each layer separated by $d = 0.1$, where r is measured relative to the center of mass. In order to prevent evaporation we set the temperature to $T = 0.0625$. $\rho(r)$ is approximately constant for $r \lesssim 4$ and quickly falls to zero for larger r . This is the behavior expected for the density profile of a self-bound liquid, and very different from the density profile of a quantum gas in a trap. As discussed above and shown in Fig. 2, indeed $\rho(r = 0) \approx \rho_{\text{eq}}$ even for such a small droplet.

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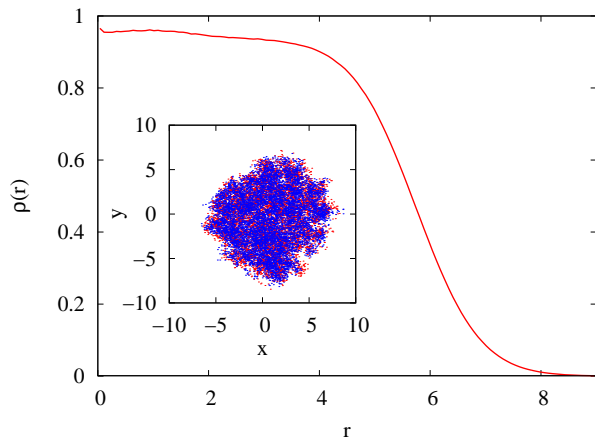


FIG. 7. (Color online) Density profile $\rho(r)$ for a self-bound droplet of 50 dipoles in each layer. Inside the droplet the density is approximately constant, with a value close to the equilibrium density ρ_{eq} of a bulk system at zero pressure. The distance is $d = 0.1$ and the temperature was set to $T = 0.0625$, which is low enough to prevent evaporation. The inset shows a snapshot of the simulation, with red and blue indicating the dipoles in the layers A and B, respectively, at the imaginary time steps of the paths of PIMC.

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