I. INTRODUCTION

Rheological electro- and magnetofluids are very flexible systems where long-range dipolar interactions lead to self-organized filamentation. Since its discovery in ferrofluids by de Gennes and Pincus [1], this phenomenon has been intensely studied in classical systems [2]. This is typically done using Monte Carlo simulations of hard- and soft-sphere dipolar particles [3] and comparison can subsequently be made to experiments with colloids that display many intriguing phases [4–6]. Important technological applications can be found in nanostructuring using atomic lithography [8] and in colloidal quantum dot photovoltaic devices [9]. However, as the systems become increasingly smaller one must start to consider also quantum aspects of the structure and the dynamics.

The explosive experimental development in ultracold atoms has lead to a wealth of new possibilities in terms of simulating the properties of condensed-matter systems with extensive control over both quantum statistics, interactions, and geometry [10–12]. Building on this success, a great deal of effort has been put into the cooling of Rydberg atoms, permanent magnetic dipole moment atoms, homo- and heteronuclear molecules, and samples of near-degenerate polar molecules can now be produced [13–15]. These systems provide access to strong dipole-dipole forces that are long-range and anisotropic [16]. In particular, in low-dimensional geometries it has been shown that the systems are more stable towards two-body chemical reaction loss [17] and to many-body instabilities [17].

Here we focus on a system consisting of polar molecules that are restricted to move in a stack of two-dimensional planes. This setup resembles the classical ferrofluids and colloidal systems with the major exception that for degenerate or near-degenerate ultracold molecules the system is intrinsically quantum and physics beyond the classical regime can be addressed. An external field can be used to align the dipoles [18], and we assume they are always perpendicular to the layer planes. Theoretically, a host of interesting few- [19–25] and many-body [26–27] states have been predicted in single- and multilayer systems. For very large dipole moments, a triangular crystal phase has been predicted [28] that is similar to the Wigner crystal of the electron gas [29]. All these predictions demonstrate the similarity of the layered dipolar case to systems with Coulomb interactions in quantum Hall, graphene, or semiconductor bilayers and to stripe formation in high-temperature superconductor materials [30].

In the current investigation, we are interested in the so-called dipolar chain liquid proposed in Ref. [26] for bosonic molecules and later also for fermionic molecules [31–33]. For perpendicularly oriented dipoles, the longest chain possible (one molecule in each layer) is always the most bound structure [26]. In the case of bosons, it was found that the longest chain will condense below a critical temperature [26] which depends slightly on whether internal vibrational modes of the chain are included [36]. This has been confirmed by a recent Monte Carlo study [37], where a lattice model was employed. Common to all the studies mentioned is that they neglect the repulsive intralayer interaction of the polar molecules. Here we include this effect in a parametric approach and considered the implications on thermodynamic quantities such as entropy and heat capacity. Thermodynamic variables have been addressed experimentally in the case of the unitary Fermi gas [35] and for two-dimensional bosonic systems [39]. In particular, density fluctuation measurements can determine compressibility [40] and heat capacity [41]. Since absorption imaging has been demonstrated also for polar molecules [42], these measurements should be possible for such systems as well.

Below we present results for the thermodynamic behavior of two- and three-chain systems for any number of layers. In the case of two chains, we determine the partition function analytically. Three chains requires, however, numerical calculation of the degeneracy of many-body states of given total energy. Our formalism is based on an effective harmonic approximation for the two-body interactions to reduce the Hamiltonian to an exactly solvable form. Naturally, this requires a careful choice of
II. METHOD

The system we consider is shown schematically in Fig. 1. In general, we have $N$ molecules of mass $m$ and dipole moment $D$ in a layered setup with equidistant layers separated by a distance $d$. The layers are assumed to be infinitely thin and allow no interlayer tunneling. This will be the case for a deep optical lattice potential. We assume that the dipoles are all oriented perpendicular to the layers, in which case we have the interlayer potential

$$V_a(x, y, n) = D^2 \frac{x^2 + y^2 - 2(nd)^2}{(x^2 + y^2 + (nd)^2)^{5/2}},$$

where $x, y$ are relative coordinates of two polar molecules that are restricted to move in planes located a distance $nd$ apart (i.e., adjacent planes have $n = 1$ and so forth). It is convenient to define the dipolar strength, $U = \frac{mD^2}{\hbar^2}$, and to measure all energies in units of $\frac{k_B^2}{m^2 d^2}$, and all temperatures in units of $k_B$, where $k_B$ is Boltzmann’s constant. We will also denote the relative radial distance between two molecules by $r = \sqrt{x^2 + y^2}$. The interaction between two molecules in the same layer is found by simply setting $n = 0$ in Eq. (1), i.e.

$$V_r(r) = D^2 \frac{1}{r^3},$$

a purely repulsive dipolar term.

The Hamiltonian for the $N$-body system is

$$H = -\frac{\hbar^2}{2m} \sum_{k=1}^{N} \left( \frac{\partial^2}{\partial x_k^2} + \frac{\partial^2}{\partial y_k^2} \right) + \frac{1}{2} \sum_{i \neq k} V_{ik}(x_{ik}, y_{ik}, n_{ik}),$$

where $(x_{ik}, y_{ik}) = (x_i - x_k, y_i - y_k)$ is the relative coordinate of the pair $(ik)$ with $i, k = 1, \ldots, N$, and the factor of $\frac{1}{2}$ is for double counting. $n_{ik}$ is an index that denotes the distance (in units of $d$) between the layers containing molecules $i$ and $k$. For $n_{ik} = 0$, $V_{ik} = V_r$ and for $n_{ik} > 0$, $V_{ik} = V_a$. For simplicity, we assume that there is no external confinement in the layer plane itself, but this is not essential and can be easily included. Of course, it implies that the center of mass motion of the total system must be treated separately.

To solve the $N$-body problem, we follow the strategy outlined in Ref. 43 of approximating the interaction terms in the Hamiltonian by quadratic forms for which the problem can be analytically solved. Using simplified models to describe the behavior of self-assembling systems that form complex structures has proven very successful [14] in spite of the diverse origin of the physical interactions. For the interlayer potential in Eq. (1), the harmonic approach has proven very accurate in reproducing energy, spatial structure, and wave function [22, 24, 25, 45]. The real dipole potential is thus replaced by

$$V_a^{\text{HO}}(r, n) = V_0 \left( \frac{r^2}{2(nd)^2} - 1 \right),$$

where $V_0 = V_0(D^2, m, d)$, and the corresponding frequency (which is denoted $a$ for attractive) is

$$\omega_a^2 = \frac{2V_0(D^2, m, d)}{m(nd)^2}.$$  

This procedure reduces all interlayer terms to harmonic oscillators that are carefully chosen to reproduce the two-body energies and the spatial structure of the state (more concretely the node of the real potential in Eq. (1) is reproduced).

The intralayer repulsion in Eq. (2) can also be approximated by a harmonic oscillator term. However, it must be the opposite sign compared to the interlayer term in order to describe a repulsive force, i.e. the term becomes

$$V_r^{\text{HO}}(r) = -\frac{1}{4} \omega_r^2 m r^2.$$
The problem is now to relate $\omega_r$ to the properties of the repulsive intralayer dipole-dipole potential. This has been discussed at length in Ref. 25.

In the current work, we are interested in the qualitative influence that the intralayer terms will have when inducing an interaction between the chains in the system. We therefore use a parametric approach and consider different values of $\omega_r$ without paying much attention to what the physical value may be. However, the systems we consider are in the self-bound regime, where the attractive interlayer terms must overcome the intralayer repulsion. If we fix $\omega_a$ for given $U$, then there will be a critical frequency, $\omega_c$, beyond which the $N$-body harmonic Hamiltonian will develop complex solutions. This is the signal of instability toward breaking of the chain system into smaller systems as discussed in Ref. 25.

Here we consider extremal values: $\omega_r = 0$ and $\omega_r = 0.99 \omega_c$. These choices will help determine what the effects of the chain-chain interaction is on thermodynamic observables within the harmonic approximation. It is then reasonable to expect that the real system will behave similarly in those limits at least on a qualitative level. The major point of the current works is thus to investigate how one can determine the role of chain structures in the system through thermodynamic measurements, specifically the changes expected due to quantum statistics and due to chain-chain interactions.

III. SINGLE CHAINS

We first consider the case of a single chain, which was the focus of Refs. 26 and 36. Since the geometry is layered and no tunneling is allowed, the single chain system is equivalent to a system of $N$ distinguishable particles. Solving the harmonic $N$-body Hamiltonian yields $\mathcal{D}(N-1)$ (where $\mathcal{D}$ is the spatial dimension, $\mathcal{D} = 2$ in the case of planes) vibrational degrees of freedom since the center of mass separates. We denote the mode frequencies by $\omega_i$, $i = 1, \ldots, \mathcal{D}(N-1)$. Each of these modes will contribute a zero point energy, $\hbar \omega_i$. This will be part of the overall ground state energy which is not our concern here as we focus on the thermal behavior for non-zero temperatures and will set the ground state energy to zero.

The partition function in the canonical ensemble, $Q(N,V,T)$, is

$$Q(N,V,T) = \sum_i g_i \exp(-\beta E_i),$$

where $g_i$ is the degeneracy of the $N$-body state of energy $E_i$, and $\beta = 1/(k_B T)$ is the inverse temperature. For the vibrational spectrum of a single chain, each mode is non-degenerate and the spectrum is equidistant. The partition function is therefore a geometric series and we have

$$q_{\text{vib}}(N,V,T) = \frac{\exp(-\Theta_i/(2T))}{1 - \exp(-\Theta_i/T)},$$

where $\Theta_i = \hbar \omega_i/k_B$. Eq. (8) is the partition function of one mode. A state with energy, $E_i$, involves all the modes, and since the energy is in the exponent of Eq. (7), we merely need to take the product over all individual modes

$$q_{\text{sep}}(N,V,T) = \prod_i \frac{\exp(-\Theta_i/(2T))}{1 - \exp(-\Theta_i/T)},$$

where $\Omega$ is the total number of vibrational degrees of freedom in the system, which as mentioned before is $\Omega = 2(N-1)$ when the center of mass has been factorized. As we will see below, in the case of multiple chains, the problem will factorize into a term related to single chain behavior, $q_{\text{sep}}$, and a term containing the chain-chain modes.

The center of mass motion can easily be taken into account. In the case where there is no external confinement in the layers, we simply multiply by the well-known non-interacting translational partition function

$$q_{\text{trans}} = \left[ \frac{mk_B T}{2\pi \hbar^2} \right]^{\mathcal{D}/2} V,$$

where $V$ is the volume and $D$ is the dimension. The total partition function is $Q = q_{\text{sep}}q_{\text{trans}}$. If, however, there is external confinement in terms of an in-plane quadratic field, the number of vibrational degrees of freedom is simply $\mathcal{D}N$ with $\mathcal{D}$ frequencies that are exactly given by the external confinement frequency. The partition function can be computed simply from Eq. (9). If the system isotropic is and the center of mass separates, then Eq. (9) can be simplified to

$$q_{\text{sep}}(N,V,T) = \left[ \prod_i \frac{\exp(-\Theta_i/(2T))}{1 - \exp(-\Theta_i/T)} \right]^{\mathcal{D}}.$$ (11)

Notice that our description of the single chain includes all possible internal vibrational modes similar to the approach of Ref. 36. However, we fix the potential to reproduce physical two-body features of the interlayer dipole-dipole interaction, whereas Ref. 36 uses an expansion of the dipolar potential around the origin to second order. Both approaches yield accurate results for energies of longer chains when compared to exact numerics 15.

IV. TWO AND THREE CHAINS

In order to explore the effect of the intralayer repulsion, we need to extend to at least two molecules per layer. The minimal model would thus be a two-chain system. Ideally we would like to consider the thermodynamics for any number of chains. However, the requirements of quantum statistics makes this an extremely complicated problem to solve due to the computational effort involved in determining the degeneracy of the many-body states. Fortunately, systems of ultracold molecules often have
very low density, and in this case we expect the dynamics of a few chains to capture the leading correlations in the system. The reasons for considering both two and three chain systems is that the dipolar interaction is expected to induced a triangular crystal in the limit of very strong dipole moment \cite{27}. A precursor of this transition should be present in a three chain model. For high density, one would need a different starting point, typically mean-field theory. We concentrate on this low-density limit from now on and calculate the properties of two- and three-chain systems.

When dealing with identical particles, one must take into account the appropriate symmetry requirements of the wave function of those particles. For identical fermions, the wave function should be antisymmetric, and for identical bosons, symmetric. The ground state of the harmonic solution is a symmetric Gaussian wave function, which could thus be a good approximation to a Bose system. Once we consider excited states for use in the partition function, the problem is much tougher, especially for many particles, as most excitations will, at first glance, appear neither symmetric nor antisymmetric. However, as we discuss below, in the case of two chains, the required symmetrization can be done analytically. For three chains one must resort to a numerical determination of the partition function \cite{46}.

A. Two Chains

For two chains (as shown in Fig. 1), molecules in different layers can be treated as distinguishable particles, while quantum statistics are important for molecules in the same layer. The total partition function can be written as a product of the separable single-chain term of Eq. (9) and an intralayer quantum statistically restricted term

\begin{equation}
Q = q_{sep} q_{f/b},
\end{equation}

where \(q_{f/b}\) is the boson/fermion intralayer part.

From diagonalization of the \(N\)-body Hamiltonian, we obtain \(2W - 1\) \cite{43} frequencies, where \(W\) is the number of layers, and the center of mass has been separated. Out of these frequencies, \(W - 1\) refer to the single chain modes. The remaining \(W\) frequencies describe the intralayer repulsion \cite{25}. Each of these \(W\) frequencies refer to a particular mode in the diagonalizing coordinates. When one writes these new coordinates in terms of the original laboratory coordinates \cite{43} introduced above, one sees that they are in fact antisymmetric in the coordinates of the identical molecules in each layer, i.e. they correspond to traditional relative coordinates, \(r_{1,k} - r_{2,k}\), where \(k = 1, \ldots, W\) is a layer index. This immediately implies that exciting any one of these \(W\) intralayer modes by an even number of quanta will produce a symmetric state under exchange of two particles in the layer, while an odd number of quanta produces an antisymmetric state. In turn, this can be used to construct the excitations that are allowed for given quantum statistics.

1. Bosons

Bosons require a symmetric wave function under interchange in each plane. In two dimensions, this can be accomplished if all quanta of excitation are either even or odd. It can also be done with a combination of even and odd numbers, as long as they are the same in each dimension for each quantum number, i.e. \(n_{1x}, n_{1y} = \text{odd}\), and \(n_{2x}, n_{2y} = \text{even}\). The partition function for bosons now becomes

\begin{equation}
q_b = \prod_k (2j + 1) \exp(-2j \Theta_k/T),
\end{equation}

where the product runs over all layers, \(k = 1, \ldots, W\), while the sum runs to infinity. This expression can be written

\begin{equation}
q_b = \prod_k \left[ \frac{\exp(-\Theta_k/T) + \exp(-3\Theta_k/T)}{1 - \exp(-2\Theta_k/T)^2} \right].
\end{equation}

2. Fermions

For fermions, we require antisymmetry with respect to particle exchange. This is achieved by enforcing that the total excitation in each direction is odd, i.e. \(n_{1x} + n_{1y} = \text{odd}\). We have

\begin{equation}
q_f = \prod_k (2j + 1) \exp(-(2j - 1) \Theta_k/T),
\end{equation}

which becomes

\begin{equation}
q_f = 2^W \prod_k \left[ \frac{\exp(-2\Theta_k/T)}{1 - \exp(-2\Theta_k/T)^2} \right].
\end{equation}

The prefactor \(2^W\) accounts for all possible interchanges between the two directions. For example, for a three layer system with two molecules in each layer, there will be three intralayer pairs that must be antisymmetrized. The lowest fermionic state has an additional degeneracy of eight. Using the notation \(|n_{1x} n_{2x} n_{3x}, n_{1y} n_{2y} n_{3y}\rangle\) for the quantum state in these three frequencies, the states are \(|111,000\rangle, |110,001\rangle, |101,010\rangle, \text{ and } |011,100\rangle\), and an additional four from interchange of \(x\) and \(y\). Every excited state get this additional degeneracy factor of \(2^W\).

3. Extensions

The symmetry considerations above can be straightforwardly extended to three dimensions. In addition, an
in-plane external confinement potential given by a harmonic oscillator does not change anything. The center of mass mode will be symmetric in all coordinates so it merely modifies the separable part of the partition function as discussed above. Thus, it does not modify the symmetry properties.

In the oscillator formalism, a constant can be added to the Hamiltonian in order to achieve some desired property of the energy of the system. Here we are interested in the behavior of the system as it is thermally excited, and not in the ground state energy. We therefore choose in the behavior of the system as discussed above. Thus, it does not modify the symmetry properties.

The energy, \( E \), is thus

\[
E = k_B T \ln Q - k_B T \sum_j \left( \frac{\Theta_j}{2T} + \ln(1 - \exp(-\Theta_j/T)) \right)
\]

where the sum is taken over all vibrational degrees of freedom in the system. If the system is isotropic, then the free energy is

\[
F = \mathcal{D} k_B T \sum_{j=1}^{W-1} \left( \frac{\Theta_j}{2T} + \ln(1 - \exp(-\Theta_j/T)) \right)
\]

The energy, \( E \), is

\[
E = k_B T \frac{\partial \ln Q}{\partial T}
\]

\[
E = \mathcal{D} k_B T \sum_{j=1}^{W-1} \left( \frac{\Theta_j}{2T} + \Theta_j \exp(-\Theta_j/T) \right)
\]

The constant volume heat capacity, \( C_V \), is simply the constant volume temperature derivative of the previous equation

\[
C_V = \frac{\partial E}{\partial T}
\]

\[
C_V = \mathcal{D} k_B T \sum_{j=1}^{W-1} \left[ \frac{\Theta_j}{T} \right]^2 \frac{\exp(-\Theta_j/T)}{[1 - \exp(-\Theta_j/T)]^2}
\]

The entropy, \( S \), is

\[
S = k_B \ln Q + k_B T \frac{\partial \ln Q}{\partial T} = \mathcal{D} k_B \sum_{j=1}^{W-1} \left( \frac{\Theta_j}{T} \exp(-\Theta_j/T) \right)
\]

\[
- \ln[1 - \exp(-\Theta_j/T)]
\]

\[
S = -k_B T \ln q_{sep}
\]

\[
E = k_B T \sum_{j=1}^{W} \left( \frac{\Theta_j}{T} \exp(-\Theta_j/T) \right)
\]

\[
+ \frac{4 \exp(-2\Theta_j/T)}{1 - \exp(-2\Theta_j/T)}
\]

\[
S = S_{sep} - A_0/T
\]

\[
E = E_{sep} + \sum_{j=1}^{W} k_B \Theta_j
\]

\[
\times \left[ \frac{\exp(-\Theta_j/T) + 3 \exp(-3\Theta_j/T)}{\exp(-\Theta_j/T) + \exp(-3\Theta_j/T)} \right]
\]

\[
\frac{4 \exp(-2\Theta_j/T)}{1 - \exp(-2\Theta_j/T)}
\]

\[
C_{V,b} = C_{V,sep} + \sum_{j=1}^{W} \frac{4k_B \Theta_j^2 \exp(-2\Theta_j/T)}{T^2}
\]

\[
\times \left[ \frac{1}{1 + \exp(-2\Theta_j/T)} \right]^2 + \frac{2}{1 - \exp(-2\Theta_j/T)} \right]
\]

For the fermion case, the free energy is

\[
F_f = k_B T \ln q_{sep} - W k_B T \ln 2
\]

\[
+ 2k_B T \sum_{i=1}^{W} \left( \frac{\Theta_i}{T} + \ln[1 - \exp(-2\Theta_i/T)] \right)
\]

The other observables proceed in the same manner as for bosons. The factor from the different permutations, \( 2^W \),
FIG. 2: Helmholtz free energies, $F$ (upper row), and energies, $E$ (lower row), divided by $Nk_B T$ for two bosonic (left column) and two fermionic (right column) chains. The different lines correspond to different values of the dipole strength, $U$, the number of layers, $W$, and the magnitude of the repulsive intralayer frequency, $\omega_r$, measured in percentage of the critical frequency at which the system breaks apart. The nomenclature is $(U, W, \omega_r \omega_{r,c})$.

contributes to the entropy, and as a constant factor to the free energy, but does not contribute to any observable related to any derivative of the partition function. The entropy is

$$S_f = S_{sep} + 2k_B \sum_k W \left( \frac{2\Theta_k \exp(-2\Theta_k/T)}{T[1 - \exp(-2\Theta_k/T)]} \ln[1 - \exp(-2\Theta_k/T)] \right) + Wk_B \ln 2,$$

and finally the heat capacity is

$$C_{V,f} = C_{V,sep} +$$

$$8k_B^2 \sum_k W \left( \frac{\Theta_k^2 \exp(-2\Theta_k/T)}{[1 - \exp(-2\Theta_k/T)]^2} \right).$$

B. Three Chains

The analytical results above for the partition function in the two-chain case can unfortunately not be extended to a larger number of chains. In this case one needs to return to the general partition function Eq. (7) and compute this numerically. As demonstrated in Ref. [46], this can be done quite effectively in the harmonic approximation for both bosonic and fermionic particles. The three-chain case has been computed numerically in order
to obtain $q_{f/b}$ from Eq. (12) that goes along with $q_{sep}$ to obtain Eq. (7).

V. RESULTS

We now present the results for both two and three chain systems for different values $U = 10$ and 20, for $\omega_r = 0$ and $\omega_r = 0.99\omega_c$, and for layer numbers $W = 6$ and 12. These values of $U$ are both in the regime where the harmonic approximation to $V$ of Eq. (1) is extremely accurate. We first discuss Helmholtz free energy and energy for two chains, omitting the case of three chains which is very similar. Next we present the entropies for two and three chains, and finally we show the heat capacities. We specialize to also discuss the distinct low temperature behavior of the heat capacity where quantum statistics is most pronounced.

A. Energy

In Fig. 2 we plot the Helmholtz free energy (upper row) and energy (lower row) for bosons (left column) and fermions (right column) for two-chain systems. An immediate striking difference between the two upper plots, is the behavior at zero temperature where bosons are smooth, whereas fermions have a kink. This is a consequence of the ground state degeneracy of the fermionic system and appears through the entropy term in $F = E - TS$. We will discuss this in more detail when we present entropies below. At slightly higher temperatures, $F$ agrees quite well for bosons and fermions, with fermions having lower overall values in all cases. In the case of $\omega_r = 0.99\omega_c$, the curves are considerably below the $\omega_r = 0$ case. This is a consequence of the fact that the mode frequencies are lowered as $\omega_r$ grows [25], allowing easier thermal excitation of the system. We also note that changing the layer number from 6 to 12 induces only slight quantitative changes. The energies in the lower
row of Fig. 2 show the same tendencies as $F$, and their close similarity attests to the fact that the entropy term is causing the differences in $F$ at low temperature.

B. Entropy

The entropies of boson (left column) and fermion (right column) two (upper row) and three (lower row) chain systems are shown in Fig. 3. For two fermionic chains we see that the entropy does not go to zero at zero temperature. This is caused by the fact that antisymmetry requires at least one quantum of excitation. For two-dimensional oscillators the first excited state is degenerate and this implies finite entropy at zero temperature. From Eq. (27), the zero temperature contribution is $k_B W \ln 2$, so the plot approaches $\ln 2$ for many layers (recall that $N$ is the total number of particles, while $W$ is the number of layers, i.e. $W/N = 1/2$). For three chains, the ground state is non-degenerate in two dimensions (the oscillator has a non-degenerate ground state and a twice degenerate first excited state). Thus the entropy goes to zero at zero temperature for three chains irrespective of quantum statistics.

We notice that the entropy starts to increase at larger temperatures for bosons than for fermions, implying that the activation energy gap is larger for bosons. This will become more pronounced when we consider heat capacities below and we postpone the discussion. In general, we see that for larger $\omega_r$, the entropy in general grows for all temperatures as the modes go down in energy as discussed above. Again we observe that the quantum statistics does not seem to play much role as the left and right columns in Fig. 3 are very similar.

C. Heat Capacity

The derivatives of the heat capacity makes it very sensitive to changes in behavior due to various modes that become activated in the system. This can be seen for instance in second order phase transitions where the heat capacity will become singular at the critical temperature. As we discussed in the introduction, the heat capacity is now a measurable quantity in ultracold atomic gases. We therefore anticipate that similar measurements should be possible also for experiments with polar molecules in the near future. Another good observable would be the compressibility of the system. The detailed balance between attractive and repulsive interactions should be visible there as well.

In Fig. 4 we show a plot similar to Fig. 3 for the heat capacities for the same temperature ranges employed in Figs. 2 and 3. Overall, we note the high temperature behavior of all cases which approaches the equipartition value of $2Nk_B$ characteristic of two-dimensional oscillators. Another general feature is that for larger $U$, the heat capacity decreases due to the larger cost of excitation in the system. When we compare fermions to bosons, we find that fermions in general approach the limit slightly slower for both two and three chain systems. There is a clear ‘shoulder’ at low temperature which seems to be more pronounced for bosons, but appears at lower temperatures for the fermions. Also, going from two to three chains seems to make the ‘shoulder’ more pronounced. This feature is clearly associated with the larger value of $\omega_r$, and also becomes more clear with increasing $W$. This implies that measurements of the heat capacity can potentially yield information about the role that chain-chain interaction plays in a dipolar chains liquid.

In order to further investigate the interesting features at low temperature in the heat capacities, we show a plot where the range of temperature is zoomed in around the low temperature region in Fig. 3. Here we see that there is striking differences between fermions and bosons, and between the two and three chain systems. Not unexpectedly, the effects of intralayer repulsion is most pronounced at low temperatures. The clear ‘shoulder’ seen for large $\omega_r$ and three fermions is the one we see at higher temperatures for bosons in Fig. 4. This can be understood from the fact that bosons have larger activation energies due to a gap in the low energy spectrum. For bosons one cannot generate a totally symmetric state by promoting one molecule from the ground to the first excited state, since the wave function would then necessarily become antisymmetric. Thus, the gap for bosons is twice the level spacing in the low-energy spectrum, whereas for fermions this is allowed and the gap is just the level spacing. This is true for the three chain systems as seen clearly in the lower row of Fig. 4. For two chains, we find that the gap is always the same (twice the level spacing) for both kinds of molecules, and the stronger increase of $C_V$ for bosons at low temperature is due to a larger density of states. Note also the difference for three fermions for the two different $\omega_r$ values, with the larger one giving a much smaller activation energy due to the lowering of the mode frequencies.

It is interesting to note that the heat capacity grows with $W$ when $\omega_r = 0$, while the opposite is true for large $\omega_r$. This is connected to the fact that in the former case, the longer chains have more (degenerate) modes to thermally excite, whereas in the latter case the repulsion is pushing all modes down toward zero but slower for longer chains where more attractive terms are present. This means that measurements of heat capacities for systems of different $W$ can help determine whether the repulsive intralayer interaction is important or not for given system density.

D. Relation to Current Experiments

For comparison to experiment it is convenient to recall our units for dipolar strength and for temperature. Here we use that 1 Debye equals $3.336 \times 10^{-30}$ C·m in SI.
FIG. 4: Heat capacity, $C_V$, divided by $Nk_B$ for systems of two (upper row) and three (lower row) chains of bosonic (left column) and fermionic (column) molecules. The nomenclature of the legends is the same as in Fig. 2.

Also we must remember that the dipole-dipole interaction in SI units has a factor of $\frac{1}{4\pi\epsilon_0}$, where $\epsilon_0$ is the permittivity of vacuum. The dimensionless dipolar strength can be written as

$$U = 0.015 \left( \frac{m}{u} \right) \left( \frac{D}{1 \text{ Debye}} \right)^2 \left( \frac{1 \mu\text{m}}{d} \right),$$

(30)

where $u$ is the atomic mass unit. Similarly, the unit of temperature that we use can be expressed in the form

$$\frac{\hbar^2}{md^2} k_B = 485 \left( \frac{u}{m} \right) \left( \frac{1 \mu\text{m}}{d} \right)^2 \text{nK},$$

(31)

The experiments most relevant to our setup are the ones being conducted at the moment at JILA [15]. The rotational and vibrational ground state molecules used are $^{40}\text{K}^{87}\text{Rb}$ which has a maximum dipole moment of $D = 0.566$ Debye. The experiment reported in Ref. [15] has studied chemical properties of a sample confined to a quasi-two-dimensional stack of about 23 layers and found profound effects due to the optical lattice potential on the loss rates. The dipoles are perpendicular to the planes and the applied field has a magnitude that induces a dipole moment of $D = 0.158$ Debye. With a lattice spacing of 532 nm, this amounts to $U = 1.15$ for maximum dipole moment, while for the experiment cited it is roughly $U = 0.09$. Our results are based on the strong-coupling harmonic approximation, and $U$ must therefore be large. The JILA experiments are therefore not in the regime of validity of our approach.

To get a stronger dipole moment, NaK molecules consisting of a Sodium and a Potassium atom can be used. They are predicted to have a maximum moment of about $D = 2.7$ Debye [47]. This has the advantage that several isotopes of Potassium are available for ultracold atomic experiments of both bosonic and fermionic character. Experiments on this system are currently under way [48]. Assuming that $d = 532$ nm, one could in principle reach values of $U \sim 13$ which is within the region where we expect our approximations to hold. Furthermore, the range of temperature shown in Figs. 2-4 above corresponds to
27 nK in this case. We thus expect that intralayer effects could be studied for system temperatures below about 270 nK. Reaching this scale should not be difficult experimentally.

VI. SUMMARY AND OUTLOOK

The physics of dipolar chain systems in multilayered geometries holds great potential for realization of exotic few- and many-body physics that is hard to access in other setups. Here we have considered the case of chains of dipolar molecules with dipole moment oriented perpendicular to the layer planes and studied the thermodynamics for both bosonic and fermionic molecules. In order to render the problem tractable, we approximate the real Hamiltonian by a harmonic form with suitably chosen parameters that reproduce realistic two-body properties of dipolar systems.

In contrast to previous studies of the dipolar chain liquid, we include the effect of interactions between the chains by introducing a repulsive harmonic term that parametrizes this effect. We have also explicitly taken care of the quantum statistical effects of having more than one molecule per layer. In the simplest non-trivial case, there are two chains in the system and for that we can determine the partition function and in turn all thermodynamic properties analytically within the harmonic approach. In the case of three chains, the degeneracies of the states could no longer be determined analytically and we resort to numerics.

Our findings indicate that heat capacity measurements for the system can probe the relative influence of excitation within single chains and the inter-chain dynamics. While we have only considered the two and three chain cases here, for a low density system we expect these to be the leading correlations in the system. A viable way to study our predictions could be through measurements at different densities. Starting from very low density, we would expect that the single chain modes are more important and will dominate the thermodynamics. However, as the density is increased, one should start to see effects of the inter-chain modes. In the case where the repulsive intralayer interaction is very strong we find a softening of
the modes of the system and thus an increase in the heat capacity that could be observable. Another observable that we expect will be strongly influenced by inter-chain dynamics is the compressibility. In the limit of very large density, we expect our approach to become less accurate and mean-field theory is probably a better starting point.

The melting of classical crystals including those with dipolar interactions have been extensively studied [49], and recently the corresponding quantum system in the case of a single layer with perpendicular dipoles was addressed [28]. The interest is focused on the ability of dipolar interactions have been extensively studied [49], and mean-field theory is probably a better starting point.

In the limit where the number of layers is large the two types of instability should merge. It is large the two types of instability should merge. It would be interesting to try to come up with an analytical expression for the dynamics in this limit. Here the harmonic approximation could be very useful.

Another question that arises for a system consisting of dipolar chains is whether one can develop an effective description that considers the chains themselves as the basic constituents. This is conceivable in the limit of low temperatures where the occupancy of higher modes is small. The quantum statistics of the chain would then depend on whether it has an even (bosonic) or odd (fermionic) number of molecules. At first glance, the former would constitute a (repulsive) Bose gas [54], while the latter would correspond to a Fermi liquid-like state [51]. The effective interaction between these entities could help discern potential instabilities in the system, such as the roton instability for bosons or the density-wave instability for fermions. In the limit where the number of layers is large the two types of instability should merge. It would be interesting to try to come up with an analytical expression for the dynamics in this limit. Here the harmonic approximation could be very useful.

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