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Quantum Fluids of Self-Assembled Chains of Polar Molecules at Finite Temperature *

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The finite temperature properties of self-assembled dipole chains of polar molecules in strongly confined pancake traps are investigated. The single-chain vibrations at finite temperature, which become important for long chains in a strongly interacting regime, are found to lower the transition temperature and to shift the chain distribution by less than 10%. We also propose experimental parameters to observe such quantum phase transition.

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In ultra-cold atomic physics, systems with dipolar interaction have recently attracted considerable attention theoretically and experimentally.[1] Manifestations of dipolar interaction have been observed in the density profiles of the chromium condensate.[2] The dipole–dipole interaction has long-range and anisotropy characteristics, which can be easily tuned by external magnetic or electric fields.[3] Systems of dipolar atoms will become an important area to investigate the properties of atomic quantum gases and are considered to provide new schemes for quantum computing.[4]

In particular, when loaded into optical lattice potential, the dipolar interaction may have great influence on the properties of ultra-cold atoms.[5−10] Using the Bogoliubov approach and mean-field decoupling approximation, the superfluid-Mott-insulator phase transition of dipolar bosons in optical lattice was analysed,[6] and the results have shown that the phase transition can be tuned by the dipole–dipole interaction as well as other effects. Due to the dipole–dipole interaction, the nonlinear dynamics of a dipolar Bose–Einstein condensate in an optical lattice is also investigated.[7] Barnett *et al.*[8] considered the polar molecules in an optical lattice prepared in a mixture of different rotational states, they found that novel Mott insulating and superfluid phases can be realized as a result of such an interaction. More recently, the quantum phase transition of cold polar molecules trapped in a 2D double well potential, the phase diagrams of polarized dipolar bosons in 2D square and 3D cubic optical lattice were studied, and a rich variety of quantum phases appear due to the dipole–dipole interaction.[9,10]

An important aspect of dipole systems is the attractive part of the interaction for dipoles aligned head to tail, which can lead to the system collapse.[11] To circumvent this problem, Wang *et al.*[12] considered a system of polar molecules in a 3D trap divided into a stack of two-dimensional pancakes by applying a strong 1D optical lattice.[13] When dipolar moments are polarized in the direction perpendicular to the planes of the traps and are sufficiently strong, the system is stable against collapse but attractive interaction between molecules in different layers leads to the formation of extended chains of molecules, analogously to the chaining phenomenon in classical rheological electro- and magneto-fluids.

In this Letter, we further explore the chaining properties. Based on an approach of single chain excitation, the effect of excitations on transition temperature and the dipole chain distribution are investigated. It is found that excitation only gets noticeable when the interaction becomes strong and dipole chains turn long. In these excitations favoured regime, transition temperature is lowered by a few percentages. These results further advances the study of quantum self-assembled liquids of polar molecules in optical lattices.

Our model starts with a special regime, where chains have been formed and dipole interaction dominates the dipole chain liquid system. We consider eigen vibration modes for a single chain. The Hamiltonian for M dipoles in successive layers is given by[12]

$$\begin{aligned} H_M &= H_{\rm CM} + H_{\rm rel}, \\ H_{\rm CM} &= \frac{P^2}{2Mm} + \frac{1}{2}Mm\omega^2 R^2, \quad [R,P] = i\hbar, \\ H_{\rm rel} &= \sum_{j=1}^{M}\Big[\frac{q_j^2}{2m} + \frac{m\omega^2 s_j^2}{2}\Big] \\ &\quad + \frac{1}{2}\sum_{i\neq j}^{M} V_{|i-j|}(|s_i - s_j|); \\ &\quad [s_j, q_l] = i\hbar\delta_{jl}(1 - M^{-1}), \end{aligned} \tag{1}$$

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where $H_{\rm CM}$ and $H_{\rm rel}$ are the c.m. motion and relative motion inside the chain, respectively; $V_j(r) = D^2(r^2-2j^2d^2)/(r^2+j^2d^2)^{5/2}$ denotes the usual dipole–dipole interaction potential with D being the electric dipole moment; ω is the in-plane trapping frequency; and m is the mass of a single molecule. New variables are defined as $q_j \equiv p_j - P/M$ and $s_j \equiv r_j - R$, where $P \equiv \sum_{j=1}^{M} p_j$ and $R \equiv \dfrac{1}{M}\sum_{j=1}^{M} r_j$ are the total momentum and the c.m. position of the chain. In the strongly interacting regime, most dipoles are tightly bonded, which results in a perpendicular dipole chain, with a thin length $r_j \ll d$. With the above assumption, the dipole interaction part of the relative motion Hamiltonian with parabolic expansion reduces to

$$H_{dd} = \frac{D^2}{d^3}\Big\{\sum_{i<j}^{M}\frac{-2}{|i-j|^3}\frac{6}{|i-j|^5} \cdot \frac{s_i^2+s_j^2-2s_is_j\langle s_i,s_j\rangle}{d^2}\Big\}, \tag{2}$$

where $H_{\rm rel}$ can be expressed in the $x-y$ coordinates $H_{\rm rel} = H_x + H_y + H_0$, because $x-y$ motion could be separated. $H_0(M) = -\dfrac{2D^2}{d^3}\sum_{\alpha=1}^{M}\dfrac{M-\alpha}{\alpha^3}$ is the chain length dependent constant. Since the harmonic potential is just for CM motion and not important for excitation, we ignore it in the following derivation. For simplicity, we introduce a layer distance parameter $t = |i-j|$, where $t = 1$ denotes the nearest layers and $t = 2$ means the interaction between dipoles in the next nearest layers, and so on. For all interactions considered, the separated Hamiltonian, in the language of vector and matrices, yields

$$H_x = \frac{1}{2m}p^Tp + \frac{6D^2}{d^5}x^TBx, \tag{3}$$

with the vectors $p = (p_1, p_2, \cdots, p_M)^T$ and $x = (x_1, x_2, \cdots, x_M)^T$, and the matrix $B = B_{t=1} + B_{t=2} + \cdots$. The matrix B contains all the information of dipole–dipole interaction inside a chain. Considering the nearest layers, $B_{t=1}$ is triangular; and for the next nearest layers, $B_{t=2}$ is pentangular. The exact form for matrix B follows

$$\begin{aligned} B_1 &= [-1][\mathrm{diag}(1,2,\cdots,2,1)][-1],\\ B_2 &= \frac{1}{2^5}[-1][0][\mathrm{diag}(1,1,2,\cdots,2,1,1)][0][-1]. \end{aligned} \tag{4}$$

Note that B_t decreases as $1/m^5$, hence involving a high order correction will only make B two lines wider. Numerical check of the eigen value of B shows that it is almost enough to consider layer interaction to the second nearest, since further will not result in more precision.

We proceed by exploring the eigen vibration modes for a single chain. By diagonalizing the matrix B, we are able to transform the dipole chain to a system of harmonic oscillators.[14] Here we introduce a linear transformation $p = Q\tilde{p}$, $x = R\tilde{x}$. $\tilde{p}, \tilde{x}$ are the respective eigen vectors. The transforming matrices Q, R abbey $Q^TQ = I$, $R^TBR = \mathrm{diag}(\lambda_1, \lambda_2, \cdots, \lambda_M)$. The eigen value λ_i of H_x denotes the corresponding eigen energy of ith vibration modes. The commutation relation of position and momentum vectors $[\tilde{x_i}, \tilde{p_j}] = \delta_{ij}$ and $[x_i, p_j] = \delta_{ij}$ will leads to $Q^{-1} = R^T$. Within this methodology, H_x becomes diagonalized. One would obtain

$$H_x = \frac{1}{2m}\tilde{p}^Tp + \frac{6D^2}{d^5}\tilde{x}^T[\lambda]x. \tag{5}$$

It is worthy to point out that when the plane trapping potential is included, all the eigen energy will jump a certain level equivalent to ω. Ignoring the tiny jump will lead to gapless excitation spectrum. Next we calculate the eigen vectors for related eigen values. Numerical results show the similarity between single chain vibration and phonon modes in usual 1D solid state physics. For the lowest energy modes, only centre of motion exists. When vibration energy is increased, more modes will become noticeable, which is accompanied by the corresponding phonon energy. Wave packets, which are related to the open boundary condition of single chain, appear in the highest energy regime.

For the system of many harmonic oscillators, zero point energy is associated with ground state, then the binding energy for a dipole chain can be written as

$$-E_{b,M} = 2\sum_{\alpha=1}^{M}\frac{1}{2}\hbar\omega_\alpha + H_0(M), \tag{6}$$

where $\omega_\alpha^2 = (12D^2/md^5)\lambda_\alpha$, $\lambda_\alpha = \mathrm{eigen}(B)_\alpha$. Note that the factor 2 comes from x and y degrees of freedom. After some straightforward algebra, the binding energy is simplified to

$$-E_{b,M} = \frac{2D^2}{d^3}\sum_{\alpha=1}^{M}\Big[\sqrt{\frac{3}{U_0}\lambda_\alpha} - \frac{M-\alpha}{\alpha^3}\Big], \tag{7}$$

with $U_0 = mD^2/\hbar^2d$, the dimensionless strength of the interaction. The critical binding value for M-dipole chain can be obtained by taking $E_{b,M} = 0$. When this criterion is applied to $M = 2, \cdots, 81$ dipoles, the numerical results show that the critical range for dipoles is $3.5 \le U_0^* \le 6$. This results is larger than the result obtained by variational method.[12]

From the eigen values of oscillating frequency, one may derive the corresponding wavefunction for the relative motion of a single chain:

$$\phi_0 = \Pi_\alpha\Big(\frac{m\omega_\alpha}{\pi\hbar}\Big)^{1/4}\exp\Big[-\frac{m\omega_\alpha}{2\hbar}(\tilde{x}_\alpha^2 + \tilde{y}_\alpha^2)\Big]. \tag{8}$$

In order to obtain the width for a single chain, we need to transform the eigen vectors back to the real space with the transformation $\tilde{x}_\alpha = \sum_{j=1}^{M} R_{\alpha,j}x_j$, one

would find the Gaussian half width of a dipole chain

$$r_j = \sqrt{\frac{\hbar}{m\tilde{\omega}_j}} = \frac{d}{(12U_0)^{1/4}\sqrt{\Sigma_{\alpha=2}^{M}\sqrt{\lambda_\alpha}R_{\alpha,j}^2}}. \quad (9)$$

In the following, the effect of excitation can be discussed by using the above results. The dispersion law for a single chain is important since it gives us a clear picture of energy spectrum. To match this interest, for finite long chains, we scale modes index to the momentum within the first period $k \in [0, \pi/d]$. While for infinite long chain, we derive some analytical results by introducing Fourier transformation (with $M \longrightarrow \infty$)[15]

$$p_j = \frac{1}{M}\sum_k \hat{p}_k e^{ikjd}, \quad x_j = \frac{1}{M}\sum_k \hat{x}_k e^{ikjd}, \quad (10)$$

where $\hat{p}_k, \hat{x}_k$ are operators ($\hat{x}_k^+ = \hat{x}_{-k}, \hat{p}_k^+ = \hat{p}_{-k}$), which follow the commutation relation

$$[\hat{x}_k, \hat{p}_{k'}] = i\delta_{k,k'}. \quad (11)$$

With this treatment, the Hamiltonian (with the second order dipole interaction) in momentum space yields

$$H_x = \sum_j \frac{1}{M}\sum_k\sum_{k'}\Big\{\frac{1}{2m}\hat{p}_k^+\hat{p}_{k'}e^{i(k+k')jd} + \frac{6D^2}{md^5}\hat{x}_k^+\hat{x}_{k'}e^{i(k+k')jd}(e^{ikd}-1)(e^{ik'd}-1) + \cdot\Big\}. \quad (12)$$

After some algebra, the Hamiltonian would be simplified to be $H_x = \sum_k\{p_k^2/2m + (6D^2/d^5)x_k^2\omega^2(k)\}$, with the dispersion relation

$$\omega(k) = \frac{2c}{d}\sqrt{\sin^2\Big(\frac{kd}{2}\Big) + \frac{1}{2^5}\sin^2(kd) + \cdots}, \quad (13)$$

where $c = \sqrt{12D^2/md^3}$ is the phonon velocity. At low energy regime, one may found $\omega = ck$, the usual phonon dispersion. A comparison between the finite long chain and infinite long chain reveals that when $M > 20$, it is reliable to consider it infinitely long.

Now we use canonical ensemble to calculate T_c. When the interactions between chains are neglected, the total energy of a dipole chain liquid is a sum of energies of individual chains. With the bending modes, we can write the single chain energy as a sum of the centre of mass and binding energies

$$\varepsilon_{i_x,i_y;k_x,k_y} = \omega(i_x + i_y + 1) - E_b + (\omega_{k_x} + \omega_{k_y}), \quad (14)$$

where i_x and i_y are quantum numbers for c.m. motion and k_x and k_y quantum numbers that denotes a specific vibration state. With the constraint of total numbers of dipoles $N_{\rm tot}$, and the help of Lagrange multiplier method and the partition function, the number of M-chain yields

$$N_M(\mu) = (L-M+1)\sum_{i_x=1}^{\infty}\sum_{i_y=1}^{\infty}\sum_{k_x=0}^{\pi/d}\sum_{k_y=0}^{\pi/d} \cdot \frac{1}{z_M^{-1}\exp\Big\{\beta\Big[\omega(i_x+i_y)+(\omega_{k_x}+\omega_{k_y})\Big]\Big\}-1}. \quad (15)$$

With the chemical potential μ, the maximum chain length L, $\beta = 1/k_BT$ and $z_M^{-1} = \exp[\beta(E_b + M\mu - \hbar\omega)]$. One may simplify the above summation with the dispersion for infinite long chain for all length chains, then N_M is expressed with the constant $\gamma = \pi/2M$, which is momentum difference between neighbouring modes.

$$N_M(\mu) = (L-M+1)\sum_{j_x=0}^{M-1}\sum_{j_y=0}^{M-1}\sum_{q=1}^{\infty} \cdot \frac{1}{z_M^{-1}\exp\Big\{\beta\Big[\omega q + \frac{2c}{d}(\sin\gamma j_x + \sin\gamma j_y)\Big]\Big\}-1}. \quad (16)$$

Transition occurs when the centre of motion of the longest chain is frozen, which means $\mu_c = L^{-1}[\omega - E_b(L)]$, $N_{\rm tot} = \Sigma_{M=1,M_{\rm min}}^{L} M N_M(\mu_c)$.

Figures 1 and 2 show that when excitation of single is considered, the transition temperature is decreased at a given interaction strength and dipole chain length. This is obvious since excitations will compete with chaining hence lower transition temperature. One step further, we find excitation becomes important for stronger interaction and long chains. With the consideration of chain vibration, at a given temperature, stronger interaction (which is attractive) will lead to a system of longer chains, thus excitation becomes noticeable. For a system of unbind dipoles (relates to high temperature or weaker dipole interaction), the main point is the negative interaction energy which leads to chaining effect, where excitation is overlapped by high temperature fluctuations.

Excitation will also affect the chain distribution at a given temperature. Figures 3 and 4 show this effect. In Fig. 3, we show that the calculated number of chains, N_M as a function of their length for different temperature with considering the excitations. Here we choose $U_0 = 6.0$, which is large enough to allow formation of chains of all length. At high temperature (dotted line), the entropy is more important, so short chains and unbound molecules dominate.[14] As the temperature is reduced further, a strong enhancement of the population of longer chains is found (solid line), which means Bose–Einstein condensate of the longest chains. Three-dimensional plot of single chain distribution as a function vibration mode is shown in Fig. 4. It reveals an exponentially decaying behaviour, in other words, at a temperature near T_c, chains only favour the lowest energy modes, which will not cause a chain to break.

The critical value proposed here could be achieved in experiments with the typical polar molecules (with

electric dipole moment about 1 Debye) trapped in the optical lattice of Nd:TAG laser, larger value of u_0 could also be obtained by turning the incident angle of the two laser beams.

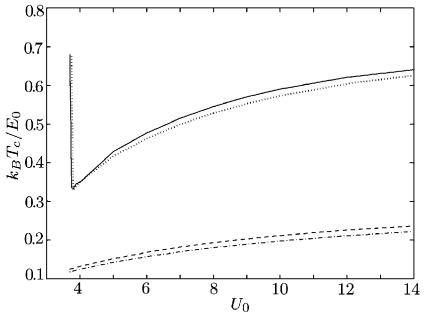

Fig. 1. T_c as function of U_0 for different chain length L. Solid, dashed, dash-dotted, and dotted lines are for chain length $L = 81$ (considering excitation), $L = 81$ (without considering excitation), $L = 20$ (considering excitation), $L = 20$ (without considering excitation), respectively. As the dipole interaction strength is increased, the difference between considering single chain excitation and not considering excitation grows (from direct summation of four quantum numbers i_x, i_y, k_x, k_y.)

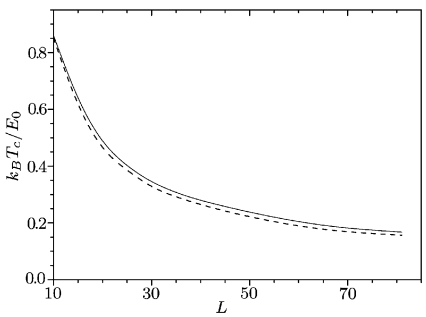

Fig. 2. T_c as function of L for $U_0 = 6$, solid line is without considering the excitation, while the dashed line for considering the excitations, this justifies the interpretation, only excitation in long chains are important.

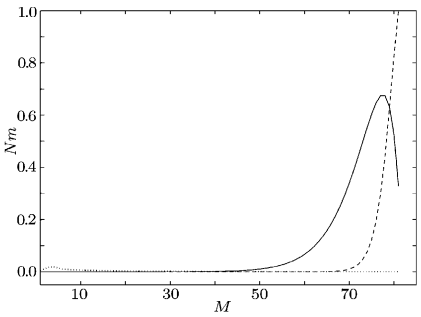

Fig. 3. The number of chains, N_m, as a function of the chain length M. Other parameters are $L = 81$, $N_{\rm tol} = 10^5$, $U_0 = 6.0$, and N_M has been normalized with dividing its maximum number. Solid, dashed, and dotted line are for temperatures $T = 0.5E_0$, $T = 0.2214E_0$, and $T = 1.5E_0$, respectively.

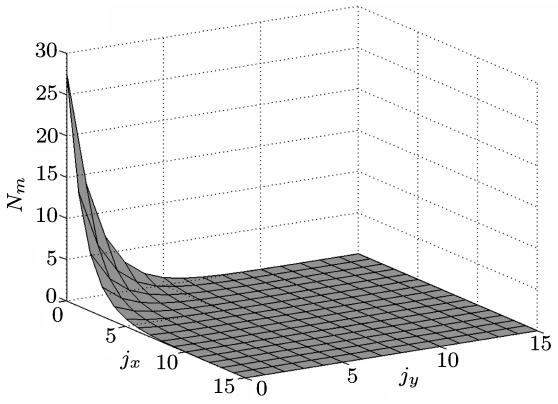

Fig. 4. Three-dimensional distribution for the number of chains, N_m, with length $M = 78$. Near transition temperature, most chains are in the lowest energy modes ($j_{x,y} \leq 5$). Parameters: $U_0 = 6$, $N_{\rm tot} = 1.0 \times 10^5$, $L = 81$, $T = 0.5E_0$.

In conclusion, with single chain excitation approach, we have further explored the effect of excitons on chaining, which becomes important especially for strong interacting dipoles at low temperature. The modified transition temperature, as functions of dipole strength and chain length, is found to be lower than that without considering single excitation. Excitons will also shift the chain distribution at a given temperature. We discuss the experimental parameters to observe such quantum phase transitions. An emergency area of interest is the analysis of Wigner crystal to glass transition[3,16] on account of its mathematical difficulties, a field where only experimental observation and quantum Monte Carlo simulations would work. The methodology and the results shown here not only provide a viewpoint to study strong interacting dipoles in optical lattices, but also provide a semi-soluble way to directly simulate such a transition at even stronger interacting regime with convincing reliability.

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