Many-body formation and dissociation of a dipolar chain crystal

Jhih-Shih You\textsuperscript{1,2,3} and Daw-Wei Wang\textsuperscript{1,2}

\textsuperscript{1} Physics Department and Frontier Research Center on Fundamental and Applied Sciences of Matter, National Tsing-Hua University, Hsinchu, Taiwan
\textsuperscript{2} Physics Division, National Center for Theoretical Sciences, Hsinchu, Taiwan
\textsuperscript{3} Department of Physics, University of California, San Diego, CA 92093, USA
E-mail: jhihshihyou@gmail.com

Received 30 March 2014, revised 26 May 2014
Accepted for publication 4 June 2014
Published 28 July 2014
New Journal of Physics \textbf{16} (2014) 073041
doi:10.1088/1367-2630/16/7/073041

Abstract

We propose an experimental scheme to effectively assemble chains of dipolar gases with a uniform length in a multi-layer system. The obtained dipolar chains can form a chain crystal with the system temperature easily controlled by the initial lattice potential and the external field strength during processing. When the density of chains increases, we further observe a second order quantum phase transition for the chain crystal to be dissociated toward layers of 2D crystal, where the quantum fluctuation dominates the classical energy and the compressibility diverges at the phase boundary. The experimental implication of such a dipolar chain crystal and its quantum phase transition is also discussed.

Keywords: dipolar chain, crystal, quantum phase transition

1. Introduction

Recently, tremendous interest in exotic strongly correlated physics has arisen from the anisotropic long-ranged dipolar interactions, which can be realized in ultracold polar molecules \cite{1,2,3}, magnetic atoms of large electronic spin \cite{4,5,6,7} and highly excited Rydberg atoms \cite{8,9}. These systems provide a new avenue for ultracold physics beyond those with short-range and isotropic interactions of ordinary atomic gases, offering novel quantum phases that cannot be
achieved even in traditional condensed matter systems [10, 11]. One of the most interesting systems along this line is the self-assembled dipolar chain liquid in an array of one- or two-dimensional structure [12–21], where a strong optical lattice potential suppresses the possible collapse instability on the attractive interaction side [22, 23]. The stabilized composite particles made of dipolar atoms/molecules in such multi-component systems hybridize the few-body and many-body physics and can be a quantum counterpart of colloid fluid, which has been extensively studied in soft-matter physics [24–28].

It is known that the energy of a single dipolar chain is lower than that of unbound free dipoles even in small dipole moment limit [12]. However, the collision rate for so many particles to form such a bound state can still be low in the current experimental parameter regime. How to develop an experimentally realistic scheme to increase the collision rate is therefore an important issue for realizing a dipolar chain liquid. Besides, most studies in the literature do not include the interaction between chains, so the general many-body properties of such interesting systems are still unclear [12–21]. Only a few works have been done in 1D Luttinger liquid [18] or in 2D BEC–BCS crossover systems [29].

In this paper we first propose a scheme to experimentally generate a dipolar chain liquid by manipulating a 3D optical lattice and the DC external field (see figures 1(a)-(d)), and investigate its many-body properties in the regime of the crystal phase (i.e. strongly interacting regime). In order to have a chain liquid (or crystal) of almost the same length, one can first prepare dipolar gases (for example polar molecules [30–32]) in a Mott insulator state with one particle per site in a 3D optical lattice. An external DC electric field is then adiabatically applied to reduce the spatial fluctuations further and to form a dipolar chain through the strong dipolar interaction.

Figure 1. Three stages to form a dipolar chain crystal. (a) Prepare polar molecules in a Mott insulator state in a 3D optical lattice. (b) Polarize the dipole moment by an external electric or magnetic field to form chains. (c) Adiabatically switch off the in-plane optical lattice to form a chain liquid or a chain crystal. (d) Three stages in terms of external parameters (lattice strength versus external field). The dashed circle indicates the regime of large spatial fluctuation, which is avoided completely in this work.
Finally, the in-plane optical lattice is adiabatically reduced to zero, leading to a chain liquid (or crystal) in the free 2D space. Evasion from the large fluctuation regime (i.e. weak lattice and weak field, see figure 1(d)) ensures that a strong collision rate between particles of different layers can form a chain.

To investigate the many-body properties, we consider the regime where these chains form a crystal phase with a small spatial fluctuation. By explicitly calculating the phonon spectrum and the quantum zero point energy, we can obtain the system entropy and monitor how the system temperature changes during the formation of chains. Furthermore, we calculate the system free energy and observe a quantum dissociation phase transition from a classical chain crystal to layers of crystal, i.e. the binding particles within a chain become unbound, as inter-layer distance is larger than a critical value. This dissociation transition is driven by the many-body quantum fluctuation effects (i.e. not existing in a single dipolar chain), and leads to a divergent compressibility along the chain at the phase boundary. Such a geometrically induced quantum phase transition is a feature of a dipolar chain liquid/crystal, resulting from the interplay of anisotropic dipolar interaction with anisotropic trapping potential. We further discuss how such a quantum phase transition can be observed in the present experimental parameter regime.

The paper is organized as follows. In section 2 we first present the system Hamiltonian and its harmonic approximation within the small fluctuation limits. In section 3 we show the phonon excitation spectrum for various parameter regimes. In section 4 we present how the temperature changes during the adiabatic (constant entropy) process, as shown in figure 1(d). In section 5, we further calculate some many-body properties of the system, including entropy, free energy, system pressure, and compressibility, from these elementary excitations. Using these results, we demonstrate the quantum phase transition from a chain crystal to layers of crystal. We then discuss the experimentally relevant issues in section 6 and summarize our results in section 7.

2. System Hamiltonian and harmonic approximation

A complete theoretical study on the formation and on the many-body properties of composite particles, such as dipolar chains, is very challenging. To simplify the calculation, in this paper, we will limit our calculation to the regime of small spatial fluctuations (i.e. spatial variation is smaller than average inter-particle distance), resulting from either a strong lattice potential and/or strong dipole interaction (see figure 1(d)). In other words, throughout this paper, we always assume the spatial fluctuation is so small that dipoles are always in a crystal phase rather than in a liquid state, although most of our results should also apply to the liquid phase at least qualitatively.

We first consider dipolar atoms/molecules loaded in a stack of 2D pancake layers, where an additional in-plane triangular optical lattice is applied (see figure 1(a)). From a theoretical point of view, such initial preparation can simplify the calculation greatly, since the average positions of the particles will not be changed during the whole process of manipulating the external field and in-plane lattice potential (the particle density is assumed fixed). Extension to other types of in-plane lattice potential is in principle possible, but the theoretical calculation becomes much more complicated, because the equilibrium position of dipolar particles can be changed as the in-plane lattice is reduced, in order to form a self-assembled dipolar crystal. Since the thermodynamic properties are independent of the initial parameter during the
adiabatic process, we believe the details of the initial preparation should not affect the final conclusion of our results.

We start from a system where polar molecules (or other dipolar particles) are initially prepared in a Mott insulator state with one particle per site, as described in figure 1(a); the system Hamiltonian including the dipolar interaction can be written as

\[ H = \sum_n \frac{p_n^2}{2m} + \frac{1}{2} \sum_{n_1=n_2=(n_1,n_2)\neq (m_1,m_2)} V_{ij}(R_n - R_m) + \frac{1}{2} \sum_{n \neq m, n \neq m'} V_{\perp}(R_n - R_m) + \sum_{n} V_{\text{lat}}(R_n) \]  

(1)

where \( R_n = n_1 a \hat{a}_1 + n_2 a \hat{a}_2 + n_3 d \hat{z} + r_n \) is the position of particles at the lattice coordinate \( n \equiv (n_1, n_2, n_3) \). \( \hat{a}_1 = (1, 0) \) and \( \hat{a}_2 = (1/2, \sqrt{3}/2) \) are the two unit vectors for the in-plane triangular lattice and \( \hat{z} \) is the unit vector perpendicular to the plane. \( a \) and \( d \) are their lattice constants respectively, leading to the two energy units defined as \( E_0 \equiv \hbar^2/ma^2 \) and \( E_0 \equiv \hbar^2/md^2 = \frac{1}{2} \hbar E_R \), where \( E_R \) is the recoil energy. \( r_n \) is the in-plane spatial variation from the lattice center, and \( V_{\perp}(R_n - R_m) \) are the intra-plane and inter-plane interaction respectively. Finally, \( V_{\text{lat}} \) is the in-plane lattice potential. For simplicity, here we have neglected the fluctuation in the perpendicular direction (\( \hat{z} \)) by assuming a very strong 1D optical lattice.

Since we are interested in the regime of small spatial fluctuations, we can expand the above Hamiltonian to the quadratic order about the equilibrium lattice positions, leading to

\[ H = H_{\text{ph}} + E_0 + E_{\perp}, \]

where the last two terms are the in-plane and out-of-plane classical energies (see appendix A for explicit expressions), and the quadratic Hamiltonian \( (H_{\text{ph}}) \) can be easily diagonalized. The phonon excitation can be expressed as

\[ \hbar \Omega_{\kappa,\sigma}(\kappa) = \sqrt{\left(\hbar \omega\right)^2 + U_{\kappa,\sigma} E_0 \lambda_{k,\sigma}} \]

(2)

where \( U_{\kappa,\sigma} \equiv D^2/\kappa^3 \) measures the strength of dipolar interaction with the electric dipole moment, \( D \), and \( \lambda_{k,\sigma} \) is the eigenvalue of the interaction matrix element of the quadratic order (see appendix A for explicit expressions). \( \omega \) is the effective harmonic frequency inside each optical lattice, obtained by expanding \( V_{\text{lat}}(R_n) \) with respect to the lattice point to the quadratic order.

3. Phonon spectrum of dipolar chain crystal in 2D free space

In figure 2, we show the calculated in-plane phonon excitation dispersion for a dipolar chain crystal system in free space (i.e. no in-plane lattice potential, \( \omega = 0 \)). We consider ten layers \( (M = 10) \) with three different lattice constants (i.e. three different densities): \( a/d = 0.7, 1.6, \) and \( 3 \). The high symmetry points \( \Gamma, X \) and \( M \) in the reciprocal lattice are depicted in the inset of figure 2(a). Note that smaller \( a/d \) leads to the single layer limit with tenfold degeneracy in each 2D plane mode. In this limit, the properties of a chain are poorly defined, because any in-plane excitation of finite momentum can be larger than the internal excitation energy of a chain, leading to a strong in-plane correlation. On the other hand, larger \( a/d \) in the dilute limit is close to the single chain limit, where the discrete energy band in figure 2(c) shows the eigenmodes for a quantized vibration of chains. Note that the in-plane coupling does broaden the eigenmodes of the chain vibration to form a band, while a single chain is still well defined since the band width is smaller than the energy gap in a chain; i.e., when applying an in-plane local measurement to
identify a single chain, the energy perturbation can be (in principle) so small that no internal degree of the chain is excited.

In figure 2(d), we further show structure of the density of states (DOS) for various $a/d$ values. It is easy to see that when $a/d$ is smaller (higher density limit) there are mainly two acoustic phonon bands, dominated by the excitation energy near the X point. These two energy bands indicate two temperature scales when thermal excitations are occupied from the low temperature limit. This may be related to the two-stage melting process (solid–hexatic–liquid) for a 2D triangular lattice, which has been extensively discussed in the literature [33–37].

When $a/d$ is larger, the excitations along the chain direction become comparable to the in-plane ones, lifting the degeneracy in the DOS by showing more bands for each mode along the chain. When $a/d \sim 1.6$, the DOS starts to have gaps opened at finite energy. The presence of narrow bands with gaps between them shows the possibility to identify a single mode excitation along the chain direction in such a strongly interacting chain crystal. Therefore, when a weakly coupled 2D crystal (small $a/d$) is adiabatically changed to a weakly coupled chain crystal (large $a/d$), we expect a significant change in the system properties (see below).

From the phonon excitations, we can then calculate the spatial fluctuations of each dipolar particle due to quantum zero point energy and/or thermal energy. They have to be smaller than the inter-particle distance during the whole process (see figure 1(d)) in order to justify the calculation of phonon excitations. Our calculation shows that this criterion is well fulfilled in all

**Figure 2.** (a)–(c) The typical phonon dispersion of a self-assembled dipolar chain crystal along high symmetry points in momentum space for different in-plane densities ($a/d = 0.7$, $a/d = 1.6$, and $a/d = 3$ respectively). Here we consider $M=10$ layers in a uniform space ($\hbar \omega = 0$) with dipolar interaction $U_{dd}/E_0 = 1$. (d) The density of states for the three cases.
the parameter range we considered here and we therefore will not mention this again throughout this paper.

4. Entropy and temperature change during the chain formation

The change of the phonon spectrum (see figure 2) leads to a significant change of thermodynamic properties. One of the most important questions is how the system temperature changes during the whole process of figure 1, if it is kept adiabatic (constant entropy). The system entropy can be easily calculated from the phonon excitation spectrum through the partition function, $\mathcal{Z} \equiv \text{Tr} e^{-\beta H}$, so that $S/k_B = \ln \mathcal{Z} + E_{\text{tot}}/k_B T$, where the quantum zero point energy of the phonon fluctuations has been included in the total energy, $E_{\text{tot}}$.

In figure 3, we show how the entropy changes as a function of temperature for various parameters. There are two general features. First, for a fixed lattice potential, systems of a stronger dipole interaction always have lower entropies (i.e. entropy decreases from stage 1 to stage 2). This can be easily understood, since the dipole interaction will freeze the relative motion between dipoles to form a crystal phase with less spatial variation. Second, for a given dipolar interaction, systems of a weak in-plane lattice potential always have larger entropy (i.e. entropy increases from stage 2 to stage 3), since the spatial variation increases when the optical lattice potential decreases. As a result, the final temperature can be either increased (heating) or decreased (cooling), determined by the competition of these two processes.

However, as mentioned above, the phonon spectrum of a free standing dipolar chain (i.e. without in-plane optical lattice) strongly depends on the in-plane density of chains. For systems of higher density (smaller $a/d$, see dash–dotted lines of figure 3(a)), the entropy increases as $T^2$ in the low temperature limit, contributed from the linear dispersion of the in-plane phonon excitations as shown in a single layer crystal limit (figure 2(a)). In the dilute limit with a discrete energy band (see figure 2(c)), on the other hand, the low temperature behavior changes more dramatically (see figure 3(b)). Such different low temperature behavior of entropy leads to a significant results of the temperature change during the formation of chain crystal: if dipoles are
initially prepared in a dilute limit, the entropy of the dipolar chain crystal without an optical lattice (stage 3, dash–dotted lines) is larger than the entropy of the initial state for particles confined in the optical lattice without (or with weak) dipole moment (stage 1, solid lines), making the whole process a cooling process. This effect, however, can be opposite (heating) if the in-plane density of chains is larger, as shown in figure 3(a).

As for the adiabatic process conserving the system entropy, in figure 4(a) we show how the system temperature changes during the three stages to form a dipolar chain crystal. When the DC field is gradually increased from $U_{dd}/E_0 = 0$ (stage 1) to $U_{dd}/E_0 = 30$ (stage 2), the level spacing is enhanced and the rise of temperature is followed to keep the same number of energy levels. The temperature then decreases when the in-plane lattice potential is reduced to zero as shown above. In figure 4(b), we show how the final temperature depends on the lattice potential and/or dipolar interaction strength of the intermediate stage (stage 2). If the initial lattice potential is strong enough in stage 1, the final state temperature can be lower than the initial temperature, leading to a polarization cooling, as also observed in a dipolar system confined in a harmonic potential [38].

The adiabatic process mentioned above is experimentally achievable because the elementary excitations have a gap, $\Delta$, which is defined by equation (2) at the $\Gamma$ point. When the energy gap dominates the dissipation due to ramping the parameters, i.e. $|d\Delta/dt| \ll \Delta^2$ (see [39] for example), the system entropy can be expected to be conserved. From stage 1 to stage 2, the gap is mostly dominated by the lattice confinement frequency, $\omega$, which is a few kilohertz in most experiments inside the Mott insulator regime. From stage 2 to 3, however, the gap is dominated by the dipolar interaction, $U_{dd}$, through the bound state of the chains (see figure 2(c)). For typical polar molecules with a dipole moment of a few Debye, we can easily have $U_{dd}$ around a few kilohertz too. As a result, we expect that the system entropy could be almost conserved during the whole process if the ramping time scale is longer than a few milliseconds. Finally, the long-range feature of dipolar interaction also makes the system relax quickly to the equilibrium.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure4}
\caption{(a) Change of the system temperature during the adiabatic process (stage 1 to stage 3 of figure 1) with lattice constant $a/d = 0.6$, initial temperature $T_i = 5.7E_0$, and layer number $M=10$. Different initial lattice strengths can lead to cooling (blue circle, $\hbar \omega/E_0 = 30$) or heating (red triangle, $\hbar \omega/E_0 = 10$) in the final stage. (b) The density plot of the final temperature (for the same initial temperature) as a function of the lattice strength and dipolar interaction strength of stage 2 in figure 1(d). The solid white line indicates the parameters to have a final state temperature the same as the initial temperature.}
\end{figure}
5. Quantum dissociation phase transition of a chain crystal

It is known that the ground state of a multi-layer system of one particle per layer should be a bound state because of the attractive interaction between dipolar atoms/molecules [40–42], while mutual repulsive interaction between dipoles in the same layer certainly increases the energy cost and may lead to a dissociation transition. Similar situations also appear in other systems of composite particles, for example, transition between a Wannier exciton (bound state) and electron–hole plasma (unbound state) [43–46], or crossover physics between BEC (bound state) and BCS (Cooper pair) for fermions in free space [47–51] or in bilayer systems [29, 52] etc.

To investigate the many-body problem between the bound and unbound states, or the possible dissociation transition of a dipolar chain crystal, we first calculate the Helmholtz free energy, \( F(T, N) = - k_B T \ln Z(N, T) \), and then the ‘pressure’ along the z direction, \( P_z = - \frac{\partial F}{\partial V} \). (Here \( V = A M d \) is the volume, with \( A \) and \( M \) being the layer area and the number of layers respectively. Both \( A \) and \( M \) are fixed for the differential.) The isothermal compressibility along the chain direction can be also defined to be \( \beta_{zz}^T \equiv - V^{-1} \frac{\partial V}{\partial p} \), which reflects the response to the variation of the inter-layer spacing along the chain direction.

In figure 5(a), we show the Helmholtz free energy calculated at zero temperature (i.e. equivalent to the total energy). It is easy to see that in the short inter-layer distance limit the free energy is dominated by the classical energy (i.e. \( E_\perp + E_\parallel \)), which is purely attractive and scaled as \( -(1/d)^3 \). When the inter-layer distance is larger, however, the quantum zero point energy due to phonon fluctuations becomes dominant, because it scales as the const \( + d^{-5/2} \) in the large \( d/a \) limit (the constant term results from the in-plane quantum fluctuation). The competition between the classical and the quantum zero point energies makes such anisotropic systems highly non-trivial: the pressure along the z direction reaches its maximum at \( d = d_c \) and becomes negative at shorter distances. The maximum of pressure indicates the divergence of compressibility, \( \beta_{zz} \), at \( d = d_c \). This quantum dissociation phase transition is second order, with a typical mean-field scaling exponent.

When the dipolar chain crystal becomes dissociated due to quantum fluctuation, the ground state of the system can be regarded as a collection of 2D dipolar crystals, one in each layer, self-assembled by the repulsive intra-layer interaction, as extensively studied before. The strong phonon fluctuations within each layer make the interlayer coherence short ranged, very different from a regular dipolar chain liquid/crystal. Note that the dissociation phase transition above is a pure ‘many-body’ effect due to the interaction between particles in the chain crystal. Such a transition does not exist if considering a single chain only, which always shows the ground state at zero temperature, as studied before [12, 13, 15]. Besides, even near the phase transition boundary, the spatial fluctuation of all dipolar particles about its average position can still be small, so the calculation of the phonon spectrum in this paper is still well justified. In figure 5(d), we further show the quantum phase diagram as a function of inter-layer distance and dipole interaction strength. Results for more than ten layers are very similar, since the many-body properties are mainly determined by dipoles in the bulk. Finally, due to the anisotropic nature of dipolar interaction, the in-plane pressure and the in-plane compressibility can be very different from the results perpendicular to the layer (i.e. along the chain direction). According to our calculation, in-plane pressure (compressibility) is monotonically decreasing (increasing) as a function of inter-chain distance, \( a/d \), and therefore we will not show it here.
Before concluding, we discuss the experimentally relevant issues here. We first consider polar molecules such as LiCs as an example. For LiCs [53], the typical energy scale, \( E_0 = \hbar^2/4md^2 \sim 257 \text{ Hz} \) for an optical lattice of lattice spacing \( \lambda/2 = 530 \text{ nm} \) (recoil energy \( E_R \sim 1.27 \text{ kHz} \)). The corresponding dimensionless strength of the dipole interaction \( U_{\text{dd}}/E_0 = mD^2/\hbar^2d \) can be as much as 15.8, when the electric dipole moment \( D = 2 \text{ Debye} \) (the permanent dipole moment of this molecule in the ground state is about \( D \sim 0.5 \text{ Debye} \)). As a result, we can easily estimate the critical temperature of a single layer dipolar crystal to be \( T_D \sim \sqrt{3/4}\beta_0 \) or \( T_D \sim 1.25/21/4 \) nK for \( \hbar \omega = 2 \sqrt{V_0 E_R} = 15E_0 \), or about \( 3.5E_0 \sim 32 \text{ nK} \) with a lattice strength \( V_0 \sim 12 \text{ kHz} \) (where \( \hbar \omega = 30E_0 \)).

For magnetic atoms such as Dy [56], the energy scale \( E_0 \sim 221 \text{ Hz} \), but the interaction energy, \( U_{\text{dd}}/E_0 = 0.04 \), is very small even for a fully polarized magnetic moment, \( \mu = 10 \mu_n \). As a result, we do not expect a promising result of chaining or crystallization for magnetic dipolar atoms, although certain long-ranged interaction effects should be still observable.

For the measurement of dipolar chains, spectroscopic techniques, such as lattice shaking [57, 58], should be available for measuring the bound state energy. Besides, since the effective

---

**Figure 5.** (a) Helmholtz free energy (F) per particle, (b) system pressure \( (P_z) \) along the normal direction, and (c) isothermal compressibility \( (\beta_T) \) as a function of inter-layer distance, \( d/\Lambda \). Results of two different dipole interaction energies are shown together for comparison. The two vertical thin dashed lines indicate the position of the phase boundary, defined by \( d/\Lambda \), where \( \beta_T \) diverges. (d) Phase diagram of a dipolar chain crystal as a function of inter-layer distance (\( d/\Lambda \)) and dipolar interaction \( (U_{\text{dd}}/E_0) \). All the results are calculated for a system of ten layers at zero temperature. Results for more layers are almost the same.

6. Experimental parameters and measurement
mass of a chain is much larger than the original molecular mass, in situ measurement and/or the change of dynamical properties can be direct evidence of the composite particles (i.e. chaining). Time-of-flight measurement should provide direct evidence of the crystal order, and the interference should be averaged out when the chains are dissociated and the inter-layer correlation disappears. Rf spectroscopy [59] and Bragg spectroscopy [60, 61] etc. could provide additional information on the phonon dispersion and density of states of the chain crystal.

In order to observe the quantum dissociation phase transition discussed in this paper, we propose that one can also measure the response of the system with respect to the perturbation of confinement potential along the chain direction (perpendicular to the layer). Since the isothermal compressibility along the chain direction should diverge near the phase boundary, it is natural to expect a highly non-trivial quadrupole collective motion along the chain direction, if the inter-layer single-particle tunneling amplitude can be tuned from zero to a finite but small value (making the dynamics along the chain direction available). Furthermore, according to the fluctuation–dissipation theorem, the isothermal compressibility is proportional to the number fluctuations within a grand canonical ensemble [62], i.e.

\[ \langle \Delta N^2_{\text{tot}} \rangle = \langle N^2_{\text{tot}} \rangle - \langle N_{\text{tot}} \rangle^2 = \frac{\langle N_{\text{tot}} \rangle^2}{V} \times k_B T \kappa_T; \]  

we may expect a large number fluctuation in each layer when the parameters are tuned near the phase boundary ($\kappa_T \to \pm \infty$). This can be easily observed from in situ measurement or interference pattern when a small but finite inter-layer tunneling is turned on.

Another quantity closely related to the quantum dissociation is the shear modulus. It is tempting to claim that the shear modulus could become zero at the phase transition boundary. However, a significant difference concerns the in-plane crystal structure: when the inter-layer distance is changed to calculate compressibility, the triangular lattice structure is unchanged. However, when calculating the shear modulus, the relative displacement between layers makes the equilibrium position of dipolar particles different from the triangular one, leading to additional energy cost for in-plane phone excitation. Further effort by taking account of quantum zero point energy needs much more complicated calculation, which cannot be carried out in the present work. As a result, even though shear modulus is closely related to the compressibility we have calculated here, their relationship is not trivial at all. We believe that results obtained in this paper have shown very important many-body effects on the chain crystal.

Finally, we emphasize that such quantum dissociation should exist even in a dipolar chain liquid phase, because strong intra-layer quantum fluctuations are more important. Finite temperature calculation should also reveal the nontrivial change of the critical temperature when the chains of the crystal melt. These liquid phases (either normal or quantum) cannot be correctly described in the harmonic expansion of this paper, because the spatial fluctuations can become of the same order as the inter-particle distance. Full quantum mechanical simulation (real space quantum Monte Carlo, for example) will be needed to investigate the full phase diagram.

7. Summary

In summary, we have proposed an experimental scheme to effectively assemble dipolar gases (especially polar molecules) to form dipolar chains, and calculate the evolution of temperature of the whole adiabatic manipulation. We also calculate the collective modes (phonon
excitations) of a dipolar crystal, and demonstrate a quantum dissociation phase transition between two distinct phases: dipolar chain crystal and layers of dipolar crystal. The quantum zero point energy leads to a divergence of the compressibility near the phase boundary. The formation of dipolar chains, phonon excitations of the chain crystal and the dissociation phase transition should be easily observable within present experimental techniques, opening a new area for strongly interacting dipolar gases.

Acknowledgments

We thank C-H Chen, E Demler, K-K Ni, Y-Y Tian, and J Ye for fruitful discussion. This work was supported by the National Center for Theoretical Sciences and the National Sciences Council in Taiwan. JS You also acknowledges the support from NSC grant no. 102–2917-I-007–032.

Appendix A. Hamiltonian

The Hamiltonian including the dipolar interaction and optical lattice can be described by

\[
H = \sum_n \frac{P_n^2}{2m} + \frac{1}{2} \sum_{n_1 \neq n_2 \neq n_3} \sum_{\mathbf{R}_{n_1} - \mathbf{R}_{n_2}} V_{ij} \left( \mathbf{R}_{n_1} - \mathbf{R}_{n_2} \right) + \frac{1}{2} \sum_{m_1 \neq m_2 \neq m_3} \sum_{\mathbf{R}_{m_1} - \mathbf{R}_{m_2}} V_{ij} \left( \mathbf{R}_{m_1} - \mathbf{R}_{m_2} \right) + \sum_n V_{i0} \left( \mathbf{R}_n \right),
\]

(A.1)

where the in-plane interaction matrix element is

\[
V_{ij} \left( \mathbf{R}_{n_1} - \mathbf{R}_{m_1} + r_{n_1} - r_{m_1} \right) = \frac{D^2}{\left| (n_1 - m_1)a_1 + (n_2 - m_2)a_2 + \Delta r_{nn}^\parallel d \right|^3}
\]

\[
= \frac{D^2/d^3}{l_{n,m}^3} \times \left[ 1 - \frac{3}{2l_{n,m}^2} \Delta r_{nn}^\parallel 2 + \frac{15}{2} \frac{1}{l_{n,m}^4} \right]
\]

\[\times \left( l_{n,m} \cdot \Delta r_{nn}^\parallel \right)^2 + \cdots \]

(A.2)

and the inter-layer interaction matrix element is

\[
V_{ij} \left( \mathbf{R}_{n_1} - \mathbf{R}_{m_2} + r_n - r_m \right) = \frac{D^2/d^3}{\left| l_{n,m} + \Delta r_{nn} + h_{nm} \right|^3}
\]

\[\times \left[ 1 - \frac{3h_{nm}^2}{l_{n,m} + \Delta r_{nn}^\parallel + h_{nm}^2} \right] \]

\[\times \left[ \frac{2h_{nm}^2 + l_{n,m}^2}{l_{n,m}^2 + h_{nm}^2} \right]^{5/2} + \frac{3}{2 \left( l_{n,m}^2 + h_{nm}^2 \right)^{7/2}} \Delta r_{nn}^\parallel 2
\]

\[\times \left[ l_{n,m} \cdot \Delta r_{nn}^\parallel \right] + \cdots \]

(A.3)

Here we have defined the relative coordinate (normalized to the inter-layer distance, d):

\[ l_{n,m} = \frac{\left( \mathbf{R}_{n_1} - \mathbf{R}_{m_2} \right)}{d} = \frac{(n_1 - m_1)a_1 + (n_2 - m_2)a_2}{d} \quad \text{with} \quad a_1 = a(1, 0), \ a_2 = a(1/2, \sqrt{3}/2), \ h_{n,m} = (n_3 - m_3), \ \Delta r_{nn}^\parallel \equiv \left( r_{n_1} - r_{n_2} \right)/d \quad \text{and} \quad \Delta r_{nn}^\parallel \equiv \left( r_n - r_m \right)/d \quad (for
\[ n_3 \neq m_3 \). We focus on deviations of molecules from their equilibrium positions, so the terms linearly proportional to \( \Delta r_{\parallel, m}^n \) must be summed to zero.

By keeping the Hamiltonian to quadratic order in \( r_n \) and using the Fourier transformation in the \( x-y \) plane \( r_n/d = \frac{i}{\sqrt{N_{3D}}} \sum_k u_k(n) e^{ikw' \cdot d} \) with \( N_{3D} = N_{3D} M \), we have

\[
H = H_{\text{ph}} + E_{\parallel} + E_{\perp},
\]

where

\[
E_{\parallel} = \frac{D^2}{2d^3} \sum_{n,m,p,m',p'} \frac{1}{l_{n,m}^3},
\]

\[
E_{\perp} = \frac{D^2}{2d^3} \sum_{n,m,p,m',p'} \frac{-2h_{n,m}^2 + l_{n,m}^2}{(l_{n,m}^2 + h_{n,m}^2)^{3/2}}
\]

and

\[
H_{\text{ph}} = \frac{1}{2m} \sum_n p_n^2 + \frac{ma^2}{2} \sum_n r_n^2 + \frac{D^2}{2d^3} \sum_k u_k^\dagger V_k u_k
\]

with the following dimensionless interaction matrix in the basis of lattice fluctuations:

\[
V_k = \begin{bmatrix}
V_{kx} & V_{ky} \\
V_{ky} & V_{ky}
\end{bmatrix},
\]

and

\[
u_k^\dagger = [u_{x, k}^*(1), \ldots, u_{x, k}^*(M), u_{y, k}^*(1), \ldots, u_{y, k}^*(M)].
\]

The diagonal elements of \( V_{kx} \), \( V_{ky} \) and \( V_{ky} \) are shown below:

\[
V_{kx}(n_3, n_3) = \sum_{p \neq 0} \frac{\sin^2(k \cdot l_p/2)}{|l_p|^5} \left( -6 + \frac{30(p_1a_1^x + p_2a_1^y)^2}{|l_p|^2} \right)
\]

\[
+ \sum_{m_3, m_3 \neq n_3} \sum_p \left[ \frac{12h_{n,m}^2 - 3|l_p|^2}{(h_{n,m}^2 + |l_p|^2)^{7/2}} - \frac{90h_{n,m}^2 - 15|l_p|^2}{(h_{n,m}^2 + |l_p|^2)^{9/2}}(p_1a_1^x + p_2a_1^y)^2 \right],
\]

\[
V_{ky}(n_3, n_3) = \sum_{p \neq 0} \frac{\sin^2(k \cdot l_p/2)}{|l_p|^5} \left( -6 + \frac{30(p_1a_1^y + p_2a_1^y)^2}{|l_p|^2} \right)
\]

\[
+ \sum_{m_3, m_3 \neq n_3} \sum_p \left[ \frac{12h_{n,m}^2 - 3|l_p|^2}{(h_{n,m}^2 + |l_p|^2)^{7/2}} - \frac{90h_{n,m}^2 - 15|l_p|^2}{(h_{n,m}^2 + |l_p|^2)^{9/2}}(p_1a_1^y + p_2a_1^y)^2 \right],
\]
\[ V_k^{xy}(n_3, n_3) = \sum_{p \neq 0} 30 \left( p_1 a_1^x + p_2 a_1^y \right) \left( p_1 a_1^y + p_2 a_1^y \right) \sin^2 \left( \frac{k \cdot l_p}{2} \right) \left| l_p \right|^2 \]

\[ + \sum_{m \neq n} \sum_p \left[ \frac{-90h_{n,m}^2 + 15\left| l_p \right|^2}{\left( h_{n,m}^2 + \left| l_p \right|^2 \right)^{2/3}} \left( p_1 a_1^x + p_2 a_1^y \right) \left( p_1 a_1^y + p_2 a_1^y \right) \left| l_p \right|^2 \right] \]

\[ = V_k^{xy}(n_3, n_3). \]  

(A.12)

For the off-diagonal elements of \( V_k^{xx} \), \( V_k^{xy} \) and \( V_k^{yy} \), we have

\[ V_k^{xx}(n_3, m_3) = -\sum_p \left[ \frac{12h_{n,m}^2 - 3\left| l_p \right|^2}{\left( h_{n,m}^2 + \left| l_p \right|^2 \right)^{3/2}} - \frac{90h_{n,m}^2 - 15\left| l_p \right|^2}{\left( h_{n,m}^2 + \left| l_p \right|^2 \right)^{3/2}} \left( p_1 a_1^x + p_2 a_1^y \right)^2 \right] \times \cos (k \cdot l_p), \]  

(A.13)

\[ V_k^{xy}(n_3, m_3) = -\sum_p \left[ \frac{12h_{n,m}^2 - 3\left| l_p \right|^2}{\left( h_{n,m}^2 + \left| l_p \right|^2 \right)^{3/2}} - \frac{90h_{n,m}^2 - 15\left| l_p \right|^2}{\left( h_{n,m}^2 + \left| l_p \right|^2 \right)^{3/2}} \left( p_1 a_1^y + p_2 a_1^y \right)^2 \right] \times \cos (k \cdot l_p), \]  

(A.14)

\[ V_k^{yy}(n_3, m_3) = V_{xy}(n_3, m_3) = \sum_p \left[ \frac{90h_{n,m}^2 - 15\left| l_p \right|^2}{\left( h_{n,m}^2 + \left| l_p \right|^2 \right)^{3/2}} \left( p_1 a_1^x + p_2 a_1^y \right) \left( p_1 a_1^y + p_2 a_1^y \right) \left| l_p \right|^2 \right] \times \cos (k \cdot l_p). \]  

(A.15)

References

[1] Ni K-K et al 2008 Science 322 231
[2] Deiglmayr J, Grochola A, Repp M, Mörtlbauer K, Glück C, Lange J, Dulieu O, Wester R and Weidemüller M 2008 Phys. Rev. Lett. 101 133004
[3] Aikawa K, Akamatsu D, Hayashi M, Oasa K, Kobayashi J, Naidon P, Kishimoto T, Ueda M and Inouye S 2010 Phys. Rev. Lett. 105 203001
[4] Griesmaier A, Werner J, Hensler S, Stuhler J and Pfau T 2005 Phys. Rev. Lett. 94 160401
[5] Beaufils Q, Chicireanu R, Zanon T, Laburthe-Tolra B, Marechal E, Vernac L, Keller J C and Gorceix O 2008 Phys. Rev. A 77 061601(R)
[6] Aikawa K, Frisch A, Mark M, Baier S, Rietzler A, Grimm R and Ferlaino F 2012 Phys. Rev. Lett. 108 210401
[7] Lu M, Burdick N Q and Lev B L 2012 Phys. Rev. Lett. 108 215301
[8] Saffman M, Walker T G and Molmer K 2010 Rev. Mod. Phys. 82 2313
[9] Schaub P et al 2012 Nature 491 87
[10] Lahaye T, Menotti C, Santos L, Lewenstein M and Pfau T 2009 Rep. Prog. Phys. 72 126401
[11] Baranov M A, Dalmonte M, Pupillo G and Zoller P 2012 Chem. Rev. 112 5012
[12] Wang D-W, Lukin M D and Demler E 2006 Phys. Rev. Lett. 97 180413
[13] Zhu K-Y, Tan L, Gao X and Wang D-W 2008 Chin. Phys. Lett. 25 48
[14] Lu X, Wu C-Q, Micheli A and Pupillo G 2008 Phys. Rev. B 78 024108
[15] Klawunn M, Duhme J and Santos L 2010 Phys. Rev. A 81 013604
[16] Potter A C, Berg E, Wang D-W, Halperin B I and Demler E 2010 Phys. Rev. Lett. 105 220406
[17] Capogrosso-Sansone B and Kuklov A B 2011 J. Low Temp. Phys. 165 212
[18] Wang D-W, Lukin M D and Demler E 2006 Phys. Rev. Lett. 97 180413
[19] Zhu K-Y, Tan L, Gao X and Wang D-W 2008 Chin. Phys. Lett. 25 48
[20] Lu X, Wu C-Q, Micheli A and Pupillo G 2008 Phys. Rev. B 78 024108
[21] Klawunn M, Duhme J and Santos L 2010 Phys. Rev. A 81 013604
[22] Potter A C, Berg E, Wang D-W, Halperin B I and Demler E 2010 Phys. Rev. Lett. 105 220406
[23] Capogrosso-Sansone B and Kuklov A B 2011 J. Low Temp. Phys. 165 212
[24] Wunsch B, Pekker D and Wang D-W 2012 Phys. Rev. A 85 013603
[25] Klawunn M, Pikovski A and Santos L 2010 Phys. Rev. A 82 044701
[26] Zimmermann R, Kilimann K, Erné B H and Philipse A P 2003 Nature Materials 2 88
[27] Zinner N T, Wunsch B, Pekker D and Wang D-W 2012 Phys. Rev. A 85 013603
[28] Danzl J G, Mark M J, Haller E, Gustavsson M, Hart R, Aldegunde J, Hutson J M and Nägerl H C 2010 Nature Phys. 6 265
[29] Engelbrecht J R, Randeria M and Sáde Melo C A R 1997 Phys. Rev. B 55 15153
[30] Forchel A, Laurich B, Wagner J, Schmid W and Reinecke T L 1982 Phys. Rev. B 25 2730
[31] Leggett A J 1980 Modern Trends in the Theory of Condensed Matter ed A Pekalski and R Przystawa (Berlin: Springer)
[32] Leggett A J 1980 Modern Trends in the Theory of Condensed Matter ed A Pekalski and R Przystawa (Berlin: Springer)
[33] Young A P 1979 Phys. Rev. B 19 1855
[34] Nelson D R and Halperin B R 1979 Phys. Rev. B 19 2457
[35] Halperin B I and D R Nelson D R 1978 Phys. Rev. Lett. 41 121
[36] Nelson D R and Halperin B R 1979 Phys. Rev. B 19 2457
[37] Halperin B I and D R Nelson D R 1978 Phys. Rev. Lett. 41 121
[38] Strandburg K J 1988 Rev. Mod. Phys. 60 161
[39] Tian Y-Y and Wang D-W 2010 Europhys. Lett. 91 66006
[40] Shih S-M and Wang D-W 2009 Phys. Rev. A 79 065603
[41] Armstrong J R, Zinner N T, Fedorov D V and Jensen A S 2010 Europhys. Lett. 91 16001
[42] Klawunn M, Pikovski A and Santos L 2010 Phys. Rev. A 82 044701
[43] Zimmermann R, Kilimann K, Kraeft W D, Kremp D and Röpke G 1978 Phys. Status Solidi B 90 175
[44] Forchel A, Laurich B, Wagner J, Schmid W and Reinecke T L 1982 Phys. Rev. B 25 2730
[45] Zimmermann R and Stolz H 1985 Phys. Status Solidi B 131 151
[46] Sarma S D and Wang D-W 2000 Phys. Rev. Lett. 84 2010
[47] Leggett A J 1980 Modern Trends in the Theory of Condensed Matter ed A Pekalski and R Przystawa (Berlin: Springer)
[48] Nozières P and Schmitt-Rink S 1985 J. Low Temp. Phys. 59 195
[49] Engelbrecht J R, Randeria M and Sáde Melo C A R 1997 Phys. Rev. B 55 15153
[50] Bloch I, Dalibard J and Zwerger W 2008 Rev. Mod. Phys. 80 885
[51] Giorgini S, Pitaevskii L P and Stringari S 2008 Rev. Mod. Phys. 80 1215
[52] Pikovski A, Klawunn M, Shlyapnikov G V and Santos L 2010 Phys. Rev. Lett. 105 215302
[53] Deiglmayr J, Grochola A, Repp M, Dulieu O, Wester R and Weidemuller M 2010 Phys. Rev. A 82 032503
[54] Kalia R K and Vashishta P 1981 J. Phys. C 14 643
[55] Büchler H P, Demler E, Lukin M, Micheli A, Prokof’ev N, Pupillo G and Zoller P 2007 Phys. Rev. Lett. 98 060404
[56] Lu M, Youn S H and Lev B L 2010 Phys. Rev. Lett. 104 063001
[57] Stöferle T, Moritz H, Schori C, Khl M and Esslinger T 2004 Phys. Rev. Lett. 92 130403
[58] Schori C, Stöferle T, Moritz H, Khl M and Esslinger T 2004 Phys. Rev. Lett. 93 240402
[59] Stewart J T, Gaebler J P and Jin D S 2008 Nature 454 744
[60] Stenger J, Inouye S, Chikkatur A P, Stamper-Kurn D M, Pritchard D E and Ketterle W 1999 Phys. Rev. Lett. 82 4569
[61] Stamper-Kurn D M, Chikkatur A P, Görlitz A G, Inouye S, Gupta S, Pritchard D E and Ketterle W 1999 Phys. Rev. Lett. 83 2876
[62] Pathria R K and Beale P D 2011 Statistical Mechanics 3rd edn (Amsterdam: Elsevier)