Static and dynamic properties of strongly coupled quasi-2D Yukawa plasma layers

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ABSTRACT

Complex plasma systems have been studied for a long time. In this thesis we focus on a quasi-2D layer system. In fact, most experimental studies of complex plasmas are based on 2D systems, because it is easy to use camera to record the inplane movement of particles. Unfortunately, due to the finite confining strength, the system is not a strictly 2D layer, it is a quasi-2D layer. We firstly studied the density profile of such a quasi-2D system by density functional theory (DFT). From the density profile research result, we found that the system can form a trilayer structure with proper parameters. Then we studied the dynamical properties of a trilayer system, and for simplicity, we only studied an ideal three layer model, both in liquid and lattice case. In lattice case, we firstly searched the stable lattice structure at different inter-layer distance. Then we used lattice sites summation to construct the dynamical matrix and solve the dispersion relation. For liquid case, we did the theoretical prediction for the collective dispersion by quasi localized charge approximation (QLCA), then we extracted the collective mode information from the molecular dynamics(MD) simulation. The QLCA and MD results were compared and discussed. The reason for the previous gap discrepancy problem is discovered.

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TABLE OF CONTENTS

ACKNOWLE	DGEMENTS	iii
LIST OF FIG	URES	vi
CHAPTER		
I. Intro	$\operatorname{duction}$	1
$\begin{array}{c} 1.1 \\ 1.2 \end{array}$	Strongly coupled plasma	$\begin{array}{c} 1 \\ 4 \end{array}$
II. Densi	ity Profile of Quasi-2D layer	5
2.1 2.2 2.3 2.4 2.5	Background System description System description Mean field theory Mean field theory Density functional theory with HNC approximation Density functional theory with HNC approximation 2.4.1 Direct correlation function for a uniform system with HNC 2.4.2 Density profile comparison between MD and HNC Peaks in liquid profile and lattice structure	5 7 12 13 17 22
111. Trilay 3.1 3.2 IV. QLCA	Yer lattice system: structure and dynamcis Stable lattice structure at different interlayer distance Lattice dispersion and eigenvectors 3.2.1 Dispersion for trilayer triangle lattice 3.2.2 Dispersion for trilayer square lattice 3.2.3 Eigenvector analysis A and MD comparison for trilayer liquid system	27 27 34 35 40 42 51
$\begin{array}{c} 4.1 \\ 4.2 \end{array}$	QLCA prediction	51 62

	4.2.1	Current correlation function and VAF 62
	4.2.2	Extracting collective mode information 66
4.3	QLCA a	nd MD result comparison
	4.3.1	More about the current correlation function 73
4.4	Current of	correlation function at small d
	4.4.1	When $k \to 0$ and $d \to 0$ in a trilayer system 83
4.5	Current of	correlation function in trilayer lattice
	4.5.1	Particle motion under harmonic oscillation model 88
	4.5.2	1D chain lattice $\dots \dots 90$
	4.5.3	Current correlation function in 1D chain lattice 93
	4.5.4	Disordered trilayer lattice
4.6	N depend	lence of the envelope $\ldots \ldots \ldots$
4.7	Why the	envelope equals the VAF?
4.8	More abo	out VAF 108
V. Sumn	nary	
APPENDICE	\mathbf{S}	
BIBLIOGRA	PHY	

LIST OF FIGURES

Figure

1.1	Temperature-density phase diagram(From Zoltan)	2
1.2	Schematic graph for dusty plasma equipment	3
2.1	Density profile comparison between the MD simulation and the MF approximation. $\Gamma = 4$, $\kappa = 0.4$, t=0.1	9
2.2	Density profile comparison between the MD simulation and the MF approximation, $\Gamma = 8$, $\kappa = 0.4$, t=0.2	10
2.3	Density profile comparison between the MD simulation and the MF approximation, $\Gamma = 16$, $\kappa = 0.4$, t=0.1	10
2.4	Density profile comparison between the MD simulation and the MF approximation, $\Gamma = 64$, $\kappa = 0.4$, t=0.2	11
2.5	Density profile comparison between the MD simulation and the MF approximation, $\Gamma = 64$, $\kappa = 0.4$, t=0.4	11
2.6	c(r) with different density. In the legend box, the first(blue) line is Yukawa potential $\kappa = 0.4$, From the second to the last line, they are calculated direct correlation functions with $\Gamma = 64$, $\kappa = 0.4$. The density for each line is 0.001, 0.01, 0.1, 1, 10, in the unit of $1/a^3$.	14
2.7	$g(r)$ with different density. In the legend box, from the first to the last line, they are calculated pair distribution functions with $\Gamma = 64$, $\kappa = 0.4$. The density for each line is 0.001, 0.01, 0.1, 1, 10, in the unit of $1/a^3$.	15

2.8	$c(r)$ with different Γ . In the legend box, the first(blue) line is Yukawa potential $\kappa = 0.4$, From the second to the last line, they are calculated direct correlation functions with density 0.02(in unit of $1/a^3$), $\kappa = 0.4$. The Γ for each line is 2, 16, 32, 64,	16
		10
2.9	$g(r)$ with different Γ . In the legend box, from the first to the last line, they are calculated pair distribution functions with density $0.02(\text{in unit of } 1/a^3), \kappa = 0.4$. The Γ for each line is 2, 16, 32, 64, 128	16
2.10	density profile comparison between HNC and MD	17
2.11	density profile comparison between HNC and MD	17
2.12	density profile comparison between HNC and MD $\ . \ . \ . \ .$.	18
2.13	density profile comparison between HNC and MD $\ . \ . \ . \ .$.	18
2.14	density profile comparison between HNC and MD $\ . \ . \ . \ .$.	19
2.15	density profile comparison between HNC and MD $\ . \ . \ . \ .$.	19
2.16	density profile comparison between HNC and MD $\ . \ . \ . \ .$.	20
2.17	density profile comparison between HNC and MD $\ . \ . \ . \ .$.	20
2.18	density profile comparison between HNC and MD $\ . \ . \ . \ .$.	21
2.19	density profile with different Γ	22
2.20	density profile with different Γ	23
2.21	density profile with different Γ	24
2.22	density profile with different Γ	25
2.23	Number of layers with different trapping strength, $\Gamma=512,\kappa=0.4$	26
2.24	Number of layers with different trapping strength, $\Gamma=724,\kappa=1.0$	26
3.1	Rhombic unit cell(left), rectangle unit cell(right)	28
3.2	Unshifted and shifted lattice structure, both top and side view	28

3.3	phase diagram with different interlayer distance	29
3.4	phase diagram with different interlayer distance $\ldots \ldots \ldots$	29
3.5	phase diagram with different interlayer distance $\ldots \ldots \ldots$	30
3.6	phase diagram map plot with different interlayer distance and different κ . The red zone is triangle, the dark blue zone is the shifted rhombic, the green zone is rectangle, the light blue zone is square.	30
3.7	Original lattice summation energy from different layers combination	32
3.8	Correlation energy from different layers combination	32
3.9	MD snapshot for particle distribution in a trilayer system with interlayer distance $d = 0.9$	33
3.10	staggered trilayer triangle lattice structure	35
3.11	trilayer triangular lattice structure	36
3.12	Reciprocal lattice: red line is for the whole projected single layer lattice, black line is for each sublattice as well as the total trilayer	36
3.13	Lattice dispersion for trilayer triangle lattice	37
3.14	Lattice dispersion for single triangle lattice	38
3.15	Lattice dispersion for single triangle lattice	38
3.16	Lattice dispersion for trilayer triangle lattice, interlayer distance is $0.2a$	39
3.17	Lattice dispersion for trilayer triangle lattice, interlayer distance is $3.0a$	39
3.18	Gap formation	40
3.19	top view of overlapping trilayer square lattice structure \ldots .	41
3.20	Lattice dispersion for trilayer square lattice, interlayer distance is $1.5a$	41

3.21	Lattice dispersion for trilayer square lattice, interlayer distance is $1.5a \dots \dots$	42
3.22	Displacement visualization of the eigenvectors, interlayer dis- tance is $1.5a$, φ is the propagation angle in k space respect to k_x axis, the length of the arrows means its magnitude, the black, red, green corresponds to layer 1-middle,2-upper,3-lower	43
3.23	Displacement visualization of the eigenvectors, interlayer dis- tance is $1.5a$, φ is the propagation angle in k space respect to k_x axis, the length of the arrows means its magnitude, the black, red, green corresponds to layer 1-middle,2-upper,3-lower	43
3.24	Displacement visualization of the eigenvectors, interlayer dis- tance is $1.5a$, φ is the propagation angle in k space respect to k_x axis, the length of the arrows means its magnitude, the black, red, green corresponds to layer 1-middle,2-upper,3-lower	44
3.25	Displacement visualization of the eigenvectors, interlayer dis- tance is $1.5a$, φ is the propagation angle in k space respect to k_x axis, the length of the arrows means its magnitude, the black, red, green corresponds to layer 1-middle,2-upper,3-lower	44
3.26	Displacement visualization of the complex eigenvecotors	47
3.27	Displacement visualization of the complex eigenvecotors	48
3.28	Displacement visualization of the complex eigenvecotors	49
3.29	Displacement visualization of the complex eigenvecotors	50
4.1	Inter/intra-layer pair distribution functions for $\Gamma = 160$, (a) $d = 3.0$, (b) $d = 1.5$, (c) $= 0.5$, (d) $d = 0.2$, note that the $g_{23}(r \rightarrow 0) > 1$ at larger d value and $g_{23}(r \rightarrow 0) \rightarrow 0$ at smaller d value . In (a), g_{11} and g_{22} nearly overlap; In (d), g_{11} and g_{12} nearly overlap	55
4.2	The same as Fig. 4.1 for $\Gamma = 10$	56
4.3	Six types of eigenvector, the length of each arrow represents the displacement of each layer, from 1 to 6, they are: AT,AL,MT,ML,ST,S	SL 57
4.4	QLCA dispersion of six modes for $d = 0.2 \dots \dots \dots \dots$	60
4.5	QLCA dispersion of six modes for $d = 1.0 \dots \dots \dots \dots$	60

4.6	QLCA dispersion of six modes for $d = 3.0 \dots \dots \dots \dots$	61
4.7	Avoided crossing between A and M modes dispersion curves for $\Gamma=160, d=0.5$	61
4.8	S mode dispersion curves for Γ =160, $d = 0.5$, each mode is always along a continuous line. The real crossing indicates S mode doesn't entangle with other two modes	62
4.9	peaks profile in longitudinal current correlation function, Γ =160, d=3.0, $ka = 0.32032$, the letter A, M, S label the corresponding mode	66
4.10	peaks profile in longitudinal current correlation function, $\Gamma=160$, d=0.2, $ka = 0.32032$,	68
4.11	peaks profile in transverse current correlation function, $\Gamma=160$, d=0.2, $ka = 0.32032$,	68
4.12	A mode dispersion comparison between MD and QLCA $\Gamma=10$, d=0.2, 0.5, 1.0	69
4.13	A mode dispersion comparison between MD and QLCA $\Gamma=10$, d = 1.5, 2.0, 3.0	70
4.14	A mode dispersion comparison between MD and QLCA Γ =160, d = 0.2, 0.5, 1.0	70
4.15	A mode dispersion comparison between MD and QLCA Γ =160, d = 1.5, 2.0, 3.0	71
4.16	M mode dispersion comparison between MD and QLCA Γ =160, d = 1.5, 2.0, 3.0	71
4.17	S mode dispersion comparison between MD and QLCA Γ =160, d = 1.5, 2.0, 3.0	72
4.18	M mode dispersion comparison between MD and QLCA $\Gamma=10$, d = 1.5, 2.0, 3.0	72
4.19	S mode dispersion comparison between MD and QLCA $\Gamma=10$, d = 1.5, 2.0, 3.0	73
4.20	T current correlation function Γ =160, $d = 1.5$	74

4.21	Current correlation function comparison between the liquid and lattice phase for $d = 1.5$. Left: T_{11} from liquid system with $\Gamma = 160$; Right: T_{11} from trilayer overlapping square lattice system.	75
4.22	Current correlation function comparison between the liquid and lattice phase for $d = 1.5$. Left: T_{11} from liquid system with $\Gamma = 160$; Right: L_{11} from trilayer overlapping square lattice system.	75
4.23	Current correlation function comparison between the liquid and lattice phase for $d = 1.5$. Left: T_{22} from liquid system with $\Gamma = 160$; Right: T_{22} from trilayer overlapping square lattice system.	76
4.24	particle snapshot for trilayer liquid system at $\Gamma=160,\;d=1.5$	76
4.25	T current correlation function Γ =160, $d = 0.2$	77
4.26	L_{11}, L_{12} at d=0.2, Γ =160	78
4.27	L_{22}, L_{23} at d=0.2, Γ =160	78
4.28	T_{11}, T_{12} at d=0.2, Γ =160	79
4.29	T_{22}, T_{23} at d=0.2, Γ =160	79
4.30	L_{11}, L_{12} at d=0.2, Γ =10	80
4.31	L_{22}, L_{23} at d=0.2, Γ =10	80
4.32	T_{11}, T_{12} at d=0.2, Γ =10	81
4.33	T_{22}, T_{23} at d=0.2, Γ =10	81
4.34	envelope in L at low Γ , $\Gamma = 10$, the envelope is k dependent	85
4.35	a plot of equation 4.40, the legend box is the k value for each line, its tendency coincides with our MD data	85
4.36	projected top view of an trilayer liquid at $\Gamma = 160$, d=0.2, the red circle is middle layer, the green triangle is the top layer, the blue square is bottom layer.	86
4.37	L current correlation function intensity map for an ordered trilayer lattice at $\Gamma = 1000$, d=0	87

4.38	T current correlation function intensity map for an ordered tri- layer lattice at $\Gamma = 1000$, d=0. Notice: T in our simulation is calculated by choosing k_y and v_x .	88
4.39	1D chain lattice, each unit cell contains just one particle \ldots .	90
4.40	disperion for 1D chain with one particle in each unit cell, black dash line is the Brillouin zone boundary	91
4.41	1D chain lattice, each unit cell contains three particle \ldots .	92
4.42	disperion for 1D chain with three particle in each unit cell, black dash line is the Brillouin zone boundary	92
4.43	$k_1 = -k_2$, they correspond to the same frequency (yellow dash line)	94
4.44	$k_1 + k_2 = 0, \ k_1 + k'_2 = \frac{2\pi}{3}, \ k_1 + k''_2 = \frac{4\pi}{3}$, they correspond to the same frequency (yellow dash line)	96
4.45	Top view of a trilayer lattice, along the black and red lines, you can only see one type of particles	98
4.46	peak comparison between T_{11} and T_{12} from a trilayer staggered triangle lattice, d=0, $\Gamma = 1000$, both are real part. $ka = 0.54912$. Left: T_{11} ; Right: T_{12}	99
4.47	peak comparison between real part and imaginary part of T_{12} from a trilayer staggered triangle lattice, d=0, $\Gamma = 1000$, $ka = 0.54912$. Left: real; Right: imaginary	99
4.48	comparison of the real part(left) and imaginary part(right) of L_{12} , full profile picture, the simulation data is from disordered trilayer system. $\Gamma = 1000$, d=0. In the legend box, it is the ka value for each line	102
4.49	The same as Fig 4.48 but a zoomed in profile picture	102
4.50	comparison of the real part of $L_{11}(\text{left})$ and $L_{12}(\text{right})$, full profile picture. the simulation data is from disordered trilayer system. $\Gamma = 1000$, d=0. In the legend box, it is the ka value for each line.	103
4.51	the comparison between envelope(left) and VAF(right). The envelope is obtained from $T_{11} - T_{12}$. In the legend box, it is the ka value of each line	104

4.52	N dependence of the envelope's amplitude for trilayer triangular lattice, $d=0$, $\Gamma = 1000$
4.53	N dependence of the envelope's amplitude for trilayer triangular lattice, d=0, $\Gamma = 1000$
4.54	the VAF for single layer lattice at $\Gamma = 1000$, the Fourier transformation is done without any window function
A.1	density profile with some stationary point

CHAPTER I

Introduction

1.1 Strongly coupled plasma

Plasma is a system with mobile charged particles. One component plasma(OCP) is a simple model, where each charged particle carries charge Q, with a neutralizing and uniform background. If the background is polarizable, due to the Debey screening, the particle interaction becomes Yukawa potential.

Suppose each particle carries charge q, a is the Wigner-Seitz radius, defined by $\pi a^2 n_s = 1(2D)$ or $\frac{4\pi}{3}a^3 n_s = 1(3D)$. We define coupling parameter as

$$\Gamma = \beta q^2 / a \tag{1.1}$$

which represents the ratio between the charge-charge interaction and the kinetic energy. The strongly coupled domain *Ichimaru* (1982) is characterized by $\Gamma \gg 1$. Fig. 1.1 shows the temperature-density phase diagram.

Dusty plasma *Shukla and Eliasson* (2009); *Morfill and Ivlev* (2009) is a good example of strongly coupled system. It was studied in astrophysics *Goertz* (1989), such as the planet ring. Later, people also discovered the dusty plasma in chip fabrication process, where the dusty plasma is contamination *Selwyn et al.* (1989). The dusty plasma is also created in laboratory. The Coulomb crystallization of



Figure 1.1: Temperature-density phase diagram(From Zoltan)



Figure 1.2: Schematic graph for dusty plasma equipment

dusty plasma was predicted by *Ikezi* (1986). In 1994, the dusty plasma crystal was observed by *Chu and I* (1994); *Hayashi and Tachibana* (1994); *Thomas et al.* (1994).

Because we can visualize the trajectory of dusty particles, it provides a very good way to study the dynamics of strongly coupled system.

Fig 1.2 is the schematic graph for dusty plasma equipment. In a chamber, there is argon gas flowing through it. The argon gas is discharged by high frequency(13.56MHz) AC electric field between the two planar electrodes. When we drop some dusty particles into the chamber, due to the smaller mass and higher mobility of the electrons, the dusty particle can absorb a large amount of electrons(thousands of electrons). Then the negatively charged dusty particles, together with the positive argon ions background, forms a new plasma system. The dusty particles' position can be visualized by a camera.

The dynamics of 2D Wigner crystal was studied in *Bonsall and Maradudin* (1977). Bilayer system was studied as early as 1981*Das Sarma and Madhukar* (1981). A multilayer system is a good model to study multi-component system.

1.2 Molecular dynamics simulation

Molecular dynamics simulation is a good way to study bulk fluid system. The system is initialized with N particles' positions and velocities. Divide the time into discrete steps, we can update the position and velocity values step by step. After enough time steps, we do statistical measurement on some physical variables we are interested. People usually use Verlet algorithm to implement the MD simulation.

$$\mathbf{r}_{i}(t+\Delta t) = \mathbf{r}_{i}(t) + \Delta t \dot{\mathbf{r}}_{i}(t) + \frac{\Delta t^{2}}{2} \ddot{\mathbf{r}}_{i}(t) + O(\Delta t^{3})$$
(1.2a)

$$\mathbf{r}_{i}(t - \Delta t) = \mathbf{r}_{i}(t) - \Delta t \dot{\mathbf{r}}_{i}(t) + \frac{\Delta t^{2}}{2} \ddot{\mathbf{r}}_{i}(t) - O(\Delta t^{3})$$
(1.2b)

Add the two equations in Eq 1.2, we can have

$$\mathbf{r}_{i}(t+\Delta t) \approx -\mathbf{r}_{i}(t-\Delta t) + 2\mathbf{r}_{i}(t) + \frac{\Delta t^{2}}{m}\mathbf{F}_{i}(t)$$
(1.3)

 $\mathbf{F}_i(t)$ is the total force acting on particle i at time t. When we predict a particle's trajectory, we don't need its velocity information(except the first step). The error for $\mathbf{r}_i(t)$ is in the order of Δt^4 . If we subtract the two equation in Eq 1.2, we can have

$$\dot{\mathbf{r}}_i(t) \approx \frac{1}{2\Delta t} [\mathbf{r}_i(t + \Delta t) - \mathbf{r}_i(t - \Delta t)]$$
(1.4)

The error for $\dot{\mathbf{r}}_i(t)$ is Δt^2 .

The particles' positions are in a cubic box, with periodic condition. This brings us some limitation when we measure the current correlation function. If the size of the box is L, the smallest k we can have is proportional to $\frac{1}{L}$. Because the box is parallel to the X or Y axis, we can only measure the dispersion propagating along the k_x and k_y direction rather than an arbitrary angle.

In this thesis, all the MD simulation work was done by Peter Hartmann.

CHAPTER II

Density Profile of Quasi-2D layer

2.1 Background

Dusty plasma has been studied experimentally for a long time, mostly in a 2D system. Because we can use a camera to track the movement of each particle. The 2D system is formed by some confinement along the Z direction. Unfortunately, no matter how strong the confinement is, the particles still have some freedom to oscillate along Z direction. So the system cannot be exactly 2D, we call it a quasi-2D system. Then an interesting problem arises, with finite confinement, what is the density profile along the Z direction? In this chapter, we will discuss about how to solve the density profile. The density profile under spherical confinement has been studied in papers (*Wrighton et al.*, 2009; *Bruhn et al.*, 2011; *Bonitz et al.*, 2006). *Totsuji et al.* (2001) studied the radial density distribution for a 2D Yukawa lattice.

2.2 System description

We consider a quasi-2D system of charged particles, interacting through a Yukawa potential, infinite in the x and y directions where the particles can move freely, and confined by a harmonic trapping potential in the z direction. Let n_s be the projected surface density of the quasi 2D layer, a characteristic parameter for a given system, then *a* is the Wigner-Seitz radius, $\pi a^2 n_s = 1$. We now introduce the customary (nominal) coupling parameter as

$$\Gamma = \beta q^2 / a \tag{2.1}$$

q is the charge carried by each particle. Eq 2.1 is exact for Coulomb interaction. For Yukawa interaction, the effective coupling $\Gamma_{\text{eff}}(\kappa) < \Gamma$. The strongly coupled domain is characterized by $\Gamma \gg 1$.

The Yukawa interaction potential and the harmonic trapping potential are, respectively

$$\varphi(r) = q^2 \frac{e^{-\kappa r}}{r}$$
, and $V(z) = \frac{m\omega^2 z^2}{2}$ (2.2)

With the 2D plasma frequency $\omega_p^2 = 2\pi q^2 n_s/(ma)$ we can as well define the parameter t characterizing the strength of the trapping potential, as

$$t^{2} = \frac{\omega^{2}}{\omega_{p}^{2}} = \frac{m\omega^{2}a}{2\pi q^{2}n_{s}} = \frac{m\omega^{2}a^{3}}{2q^{2}}$$
(2.3)

The behavior of a system with a given Yukawa potential (i. e. κ) is completely determined by these two parameters. Both the equilibrium properties and the dynamics of the system have been studied by Molecular Dynamics (MD) simulation and by theoretical analysis. Here we report on the equilibrium studies. The MD simulations trance the trajectories of 10 000 particles in an external trapping potential defined by V(z) and periodic boundary conditions in x and y in a cubic simulation domain. Initial positions are assigned to the particles based on a simple initial barometric estimate. Inter-particle forces are summed for all particles within a radius of $R \approx 44a$ for each particle. Before conducting any measurements the system is given enough time (approximately of 4 000 plasma oscillation cycles) to reach equilibrium during the initial thermalization phase using the velocity back-scaling thermostat. This is verified by observing the temperature stability after the thermaization is turned off. During the measurements data is collected and averaged over approx. 2000 plasma oscillation cycles (100000 time-steps).

2.3 Mean field theory

If we consider one test particle in the layer, the total potential it feels is U(z), according to the canonical ensemble, the density distribution is $n(z) \propto e^{-\beta U(z)}$

$$n(z) = \frac{N}{\pi R^2} \frac{e^{-\beta U(z)}}{\int_{-\infty}^{\infty} e^{-\beta U(z)} dz}$$
(2.4)

where the N is the total number of particles, the R is radius of the surface area we consider.

$$U(z) = \frac{mw^2 z^2}{2} + \int n(\mathbf{r}') \frac{e^{-\kappa(|\mathbf{r}-\mathbf{r}'|)}}{|\mathbf{r}-\mathbf{r}'|} d\mathbf{r}'$$
(2.5)

From Eq.2.5 we can see that U(z) is a functional of n(z), combine Eq 2.4 and Eq 2.5, we can solve n(z) iteratively. The iterative steps are in the following way:

- 1. Choose an initial density profile $n_0(z)$, normally we use the density profile where particles have no interaction, it is simply Gaussian.
- 2. Substitute the $n_0(z)$ into Eq 2.5, we get a potential profile $U_0(z)$.
- 3. Substitute $U_0(z)$ into Eq 2.4, we can get a new density profile $n_1(z)$.
- 4. Go back to the first step utill the density profile converges. In order to get converged solution, the density profile is updated in such a way $n(z) = \alpha n_0(z) + (1 \alpha)n_1(z), 0 < \alpha < 1$, we choose $\alpha = 0.98$

Before we process, we rescale some variables, $\tilde{r} = r/a$, $\beta U = \Gamma \tilde{U}$, $\kappa a = \tilde{\kappa}$, $\tilde{n}(\tilde{z}) = a^3 n(z)$

$$\tilde{n}(\tilde{z}) = a^{3}n(z)$$

$$= a^{3}\frac{N}{\pi\tilde{R}^{2}a^{2}}\frac{e^{-\Gamma\tilde{U}(\tilde{z})}}{\int_{-\infty}^{\infty}e^{-\Gamma\tilde{U}(\tilde{z})}d\tilde{z}a}$$

$$= n_{s}\frac{e^{-\Gamma\tilde{U}(\tilde{z})}}{\int_{-\infty}^{\infty}e^{-\Gamma\tilde{U}(\tilde{z})}d\tilde{z}}$$
(2.6)

where we used $n_s = \frac{N}{\pi \tilde{R}^2} = \frac{1}{\pi a^2}$.

$$\beta U(z) = \Gamma \tilde{U}(\tilde{z}) = \Gamma(\frac{m\omega^2 a^3}{2q^2} \tilde{z}^2 + \int \tilde{n}(\tilde{\mathbf{r}}') \frac{e^{-\tilde{\kappa}(|\tilde{\mathbf{r}} - \tilde{\mathbf{r}}'|)}}{|\tilde{\mathbf{r}} - \tilde{\mathbf{r}}'|} d\tilde{\mathbf{r}}')$$
(2.7)

From Eq 2.7 we have

$$\tilde{U}(\tilde{z}) = t^2 \tilde{z}^2 + \int \tilde{n}(\tilde{\mathbf{r}}') \frac{e^{-\tilde{\kappa}(|\tilde{\mathbf{r}} - \tilde{\mathbf{r}}'|)}}{|\tilde{\mathbf{r}} - \tilde{\mathbf{r}}'|} d\tilde{\mathbf{r}}'$$
(2.8)

Because the quasi-2D layer has cylindrical symmetry, we can do the angular integral analytically.

$$\tilde{U}(\tilde{z}) = t^2 \tilde{z}^2 + 2\pi n_s \frac{\int_0^{\tilde{R}} \tilde{\rho} d\tilde{\rho} \int_{-\infty}^{\infty} e^{-\Gamma \tilde{U}(\tilde{z})} \frac{e^{-\tilde{\kappa}(\sqrt{(\tilde{z}-\tilde{z}')^2 + \tilde{\rho}^2})}}{\sqrt{(\tilde{z}-\tilde{z}')^2 + \tilde{\rho}^2}} d\tilde{z}'}{\int_{-\infty}^{\infty} e^{-\Gamma \tilde{U}(\tilde{z})} d\tilde{z}}$$
(2.9)

Set $\sqrt{(\tilde{z} - \tilde{z}')^2 + \tilde{\rho}^2} = x$,

$$\int_{0}^{\tilde{R}} \tilde{\rho} d\tilde{\rho} \int_{-\infty}^{\infty} e^{-\Gamma \tilde{U}(\tilde{z})} \frac{e^{-\tilde{\kappa}(\sqrt{(\tilde{z}-\tilde{z}')^{2}+\tilde{\rho}^{2}})}}{\sqrt{(\tilde{z}-\tilde{z}')^{2}+\tilde{\rho}^{2}}} d\tilde{z}' \\
= \frac{1}{2} \int_{0}^{\tilde{R}} d((\tilde{z}-\tilde{z}')^{2}+\tilde{\rho}^{2}) \int_{-\infty}^{\infty} e^{-\Gamma \tilde{U}(\tilde{z})} \frac{e^{-\tilde{\kappa}(\sqrt{(\tilde{z}-\tilde{z}')^{2}+\tilde{\rho}^{2}})}}{\sqrt{(\tilde{z}-\tilde{z}')^{2}+\tilde{\rho}^{2}}} d\tilde{z}' \\
= \frac{1}{2} \int_{|\tilde{z}-\tilde{z}'|}^{\sqrt{(\tilde{z}-\tilde{z}')^{2}+\tilde{R}^{2}}} d(x^{2}) \int_{-\infty}^{\infty} e^{-\Gamma \tilde{U}(\tilde{z})} \frac{e^{-\tilde{\kappa}x}}{x} d\tilde{z}' \\
= \int_{|\tilde{z}-\tilde{z}'|}^{\sqrt{(\tilde{z}-\tilde{z}')^{2}+\tilde{R}^{2}}} e^{-\tilde{\kappa}x} d(x) \int_{-\infty}^{\infty} e^{-\Gamma \tilde{U}(\tilde{z})} d\tilde{z}' \\
= \int_{-\infty}^{\infty} e^{-\Gamma \tilde{U}(\tilde{z})} \frac{1}{\tilde{\kappa}} (e^{-\tilde{\kappa}|\tilde{z}-\tilde{z}'|} - e^{-\tilde{\kappa}\sqrt{(\tilde{z}-\tilde{z}')^{2}+\tilde{R}^{2}}}) d\tilde{z}'$$
(2.10)

Now let's compare the density profile from the MFT and the MD simulation. In the following text, we will omit the wave sign, but keep in mind, they are always rescaled variables.



Figure 2.1: Density profile comparison between the MD simulation and the MF approximation. $\Gamma=4,\,\kappa=0.4,\,t{=}0.1$



Figure 2.2: Density profile comparison between the MD simulation and the MF approximation, $\Gamma = 8$, $\kappa = 0.4$, t=0.2



Figure 2.3: Density profile comparison between the MD simulation and the MF approximation, $\Gamma = 16$, $\kappa = 0.4$, t=0.1



Figure 2.4: Density profile comparison between the MD simulation and the MF approximation, $\Gamma = 64$, $\kappa = 0.4$, t=0.2



Figure 2.5: Density profile comparison between the MD simulation and the MF approximation, $\Gamma = 64$, $\kappa = 0.4$, t=0.4

From the Fig 2.1 to Fig 2.3, we can see that MF result has a good agreement with the MD result when Γ is small or t is small. At larger Γ , from Fig 2.4 and 2.5, the MF reslt is completely wrong. In fact the MF can only lead to monotonic density profile on one side of z axis. The mathematical proof is in appendix A. Why MF fails in high Γ or high particle density cases? If we look at Eq 2.7, when we calculate the Yukawa interaction potential, $|\tilde{\mathbf{r}} - \tilde{\mathbf{r}'}|$ can be close to zero. However, in a real physical system, the probability that two particles can touch each other is almost zero, normally, in liquid, we have pair distribution function where $g(r \to 0) \to 0$. Only at idea gas limit, two particles can be infinitely close, then MF can give precise description.

2.4 Density functional theory with HNC approximation

In order to solve the density profile profile at strongly coupled phase, we need improve our model. Based on Hansen's book (*Hansen and McDonald*, 2013)(Eq 4.3.16), we have

$$n(\mathbf{r}) \propto \exp\left[-\beta V(\mathbf{r}) + \int \Delta n(\mathbf{r}') c_0(|\mathbf{r} - \mathbf{r}'|; n_0) \, \mathrm{d}\mathbf{r}'\right].$$
(2.11)

where n_0 is density of a uniform reference system. $\Delta n(\mathbf{r}) = n(\mathbf{r}) - n_0$, the $c_0(r; n_0)$ is the direct correlation function for this reference system. Eq 2.11 is derived from density functional method by minimizing the grand potential energy. It can also be found in book *Henderson* (1992). Becasue $\int c_0(|\mathbf{r} - \mathbf{r}'|; n_0)n_0 d\mathbf{r}'$ is constant for a given reference system. It doesn't affect the density profile. Then we can rewrite Eq 2.11 as

$$n(\mathbf{r}) \propto \exp\left[-\beta V(\mathbf{r}) + \int n(\mathbf{r}')c_0(|\mathbf{r} - \mathbf{r}'|; n_0) \,\mathrm{d}\mathbf{r}'\right]$$
(2.12)

Here the $n(\mathbf{r})$ is only z dependent, we will write it as n(z). Formally, Eq 2.12 is the same as the formula in MF method. We just define a new total potential

$$U_{new}(z) = \frac{mw^2 z^2}{2} - \frac{1}{\beta} \int n(\mathbf{r}') c_0(|\mathbf{r} - \mathbf{r}'|; n_0) \, \mathrm{d}\mathbf{r}'$$
(2.13)

Compared with Eq 2.5, the new density functional method just replaces the Yukawa potential by $-\frac{1}{\beta}c_0(|\mathbf{r} - \mathbf{r}'|; n_0)$. We can use the same algorithm in MF to get the density profile. The only information we need to proceed the calculation is the direct correlation function $c_0(|\mathbf{r} - \mathbf{r}'|; n_0)$.

2.4.1 Direct correlation function for a uniform system with HNC

In this section, we will try to obtain the direct correlation function for a uniform system with hyper-netted chain(HNC) approximation *Rowlinson* (1965). According to the Ornsteiin-Zernike(OZ) equation

$$h(r) = c(r) + n_0 \int c(|\mathbf{r} - \mathbf{r}'|)h(\mathbf{r}') \, \mathrm{d}\mathbf{r}'$$
(2.14)

This is an integrl equation associating two functions h(r) and c(r). It shows that the pair correlation function can be constructed by the direct correlation function The h(r) is the pair correlation function. It is also related to the pair distribution function g(r)

$$h(r) = g(r) - 1 \tag{2.15}$$

In order to sovel c(r), we have to get another realtionship between c(r) and g(r), which is the HNC closure

$$g(r) = \exp[-\beta\varphi(\mathbf{r}) + h(\mathbf{r}) - c(\mathbf{r})]$$
(2.16)

where the $\varphi(\mathbf{r})$ is the particle interaction potential, it is Yukawa in this chapter. Combine Eq 2.14 and Eq 2.16, we can solve both g(r) and c(r) simultaneously.

As we all know, when $r \to 0$, $g \to 1$, $h \to 0$, it means $-\beta \varphi(\mathbf{r}) = c(\mathbf{r})$, at idea gas limit, $g(r) \equiv 1$, it goes back to the MF approximation.

When solve the g(r) and c(r) in real and Fourier space iteratively, the algorithm

is based on Springer et al. (1973).



Figure 2.6: c(r) with different density. In the legend box, the first(blue) line is Yukawa potential $\kappa = 0.4$, From the second to the last line, they are calculated direct correlation functions with $\Gamma = 64$, $\kappa = 0.4$. The density for each line is 0.001, 0.01, 0.1, 1, 10, in the unit of $1/a^3$.



Figure 2.7: g(r) with different density. In the legend box, from the first to the last line, they are calculated pair distribution functions with $\Gamma = 64$, $\kappa = 0.4$. The density for each line is 0.001, 0.01, 0.1, 1, 10, in the unit of $1/a^3$.



Figure 2.8: c(r) with different Γ . In the legend box, the first(blue) line is Yukawa potential $\kappa = 0.4$, From the second to the last line, they are calculated direct correlation functions with density 0.02(in unit of $1/a^3$), $\kappa = 0.4$. The Γ for each line is 2, 16, 32, 64, 128.



Figure 2.9: g(r) with different Γ . In the legend box, from the first to the last line, they are calculated pair distribution functions with density 0.02(in unit of $1/a^3$), $\kappa = 0.4$. The Γ for each line is 2, 16, 32, 64, 128.

2.4.2 Density profile comparison between MD and HNC



Figure 2.10: density profile comparison between HNC and MD



Figure 2.11: density profile comparison between HNC and MD



Figure 2.12: density profile comparison between HNC and MD



Figure 2.13: density profile comparison between HNC and MD



Figure 2.14: density profile comparison between HNC and MD



Figure 2.15: density profile comparison between HNC and MD



Figure 2.16: density profile comparison between HNC and MD



Figure 2.17: density profile comparison between HNC and MD



Figure 2.18: density profile comparison between HNC and MD

From the figures above, we can see there is good agreement between HNC and MD result for $\Gamma=16$, 32, 64. Due to the new version of density functional method, we no longer have just monotonic profile. For $\Gamma = 128$, there is significant discrepancy, but the main tendency does match.

In the DFT calculation, the largest Γ values we can use is about 300, bigger Γ can't lead to convergence of integral equation Eq 2.12. From Fig 2.18, we can see that at $\Gamma = 512$, the system is already like a multilayer crystal, our calculated density profile doesn't match the MD result perfectly, but it still can predict the number of peaks and the position of the peaks.

In 2014, Matheus Girotto et al. (2014) published a paper with the same topic, they did apply the HNC approximation, but in a different manner, They calculated the correlation energy based on local density approximation (LDA). In that paper, the κ is 2, the comparison provided is either in lower gamma or lower trapping strength, which means the Γ_{eff} is not very strong.

2.5 Peaks in liquid profile and lattice structure

In last section, we see that when t is small and gamma is large, the density profile develops some small fluctuations, at very large Γ , it becomes sharp peak. These fluctuation in the density profile is easy to understand between a single and double-peak profile. Because the total potential energy contains two parts: the charge-charge interaction potential and trapping potential. The former one requires all the particles spread as far as possible, the latter one forces all the particles to condense around the z = 0 region. Under the competition of the two mechanism, the double-peak profile can happen.



Figure 2.19: density profile with different Γ


Figure 2.20: density profile with different Γ



Figure 2.21: density profile with different Γ



Figure 2.22: density profile with different Γ

From Fig 2.19 to Fig 2.22, we can see that the small fluctuations and the sharp peaks have the same position. At very large Γ , the system has already crystallized, each peak represents a single layer of lattice. Based on this, we can know that the fluctuation peaks in a liquid density profile just comes from the melting of a multi-layer lattice. It is easy to predict the number of peaks in a given density profile. In a lattice system, the average interparticle distance in each layer should be comparable with the interlayer distance.

Assume there are N layers, each layer is a square lattice, then the square side length $x_N = \sqrt{\pi N}$, the inter-layer distance is $d_N = \frac{w}{N-1}$, where the w is the distance between the two outermost peaks. Starting from N=2, we compare x_N and d_N , when $x_N > d_N$, we stop, the N where we stop is the predicted the number of layers (see Fig 2.23 and Fig 2.24). In (*Totsuji et al.*, 1997), they calculated the total energy of the system with different number of layers, including the particle interaction and the trapping energy. For a given trapping potential, we can always find an optimal N. The essential of the two methods are the same.

к=0.4	г=512	3	3	3	4	4	5	5	6	6	7	peak num
		0.34	0.3	0.22	0.2	0.14	0.12	0.09	0.08	0.07	0.06	t
N	$\sqrt{N\pi}$	3.5	4.2	6	7	10	12	15	17.5	19	22	layer width
2	2.5066	3.5	4.2	6	7	10	12	15	17.5	19	22	
3	3.0700	1.75	2.1	3	3.5	5	6	7.5	8.75	9.5	11	
4	3.5449	1.1667	1.4	2	2.3333	3.3333	4	5	5.8333	6.3333	7.3333	layer width
5	3.9633	0.875	1.05	1.5	1.75	2.5	3	3.75	4.375	4.75	5.5	N-1
6	4.3416	0.7	0.84	1.2	1.4	2	2.4	3	3.5	3.8	4.4	
7	4.6895	0.5833	0.7	1	1.1667	1.6667	2	2.5	2.9167	3.1667	3.6667	
8	5.0133	0.5	0.6	0.8571	1	1.4286	1.7143	2.1429	2.5	2.7143	3.1429	

Figure 2.23: Number of layers with different trapping strength, $\Gamma = 512$, $\kappa = 0.4$

к=1	г=724	3	3	4	4	5	6	6	6	7	7	peak num
		0.165	0.135	0.068	0.055	0.032	0.026	0.021	0.019	0.016	0.015	t
N	$\sqrt{N\pi}$	3.5	4.2	8	9	12	16	18.5	20	22	23	layer width
2	2.5066	3.5	4.2	8	9	12	16	18.5	20	22	23	
3	3.0700	1.75	2.1	4	4.5	6	8	9.25	10	11	11.5	
4	3.5449	1.1667	1.4	2.6667	3	4	5.3333	6.1667	6.6667	7.3333	7.6667	laver width
5	3.9633	0.875	1.05	2	2.25	3	4	4.625	5	5.5	5.75	M 1
6	4.3416	0.7	0.84	1.6	1.8	2.4	3.2	3.7	4	4.4	4.6	N = 1
7	4.6895	0.5833	0.7	1.3333	1.5	2	2.6667	3.0833	3.3333	3.6667	3.8333	
8	5.0133	0.5	0.6	1.1429	1.2857	1.7143	2.2857	2.6429	2.8571	3.1429	3.2857	

Figure 2.24: Number of layers with different trapping strength, $\Gamma = 724$, $\kappa = 1.0$

Experimental result Teng et al. (2003) shows the similar density profile in a quasi-2D system. Bonitz's (Chapter 10.7 in *Bonitz* (2010)) MD simulation work for both harmonic and soft-box trap also shows the similar density profile when varies the trapping strength. At high Γ , such shell formation in density profile is quite general, not only for planar system here, but also for spherical and cylindrical system.

CHAPTER III

Trilayer lattice system: structure and dynamcis

3.1 Stable lattice structure at different interlayer distance

As we have seen in the last chapter, with proper confining strength and coupling parameter, we can create a trilayer structure. Some simulation works (*Klumov* and Morfill, 2008) have confirmed it. Here we will try to study the properties of a trilayer system. For simplicity, we assume each particle only has X-Y displacement. We firstly study the stable lattice structure with different interlayer distance. We assume each layer has the same kind of lattice structure, due to the symmetry, the unit cell is ether rhombic or rectangle, here we can tune two parameters, rhombic angle and the aspect ratio. Originally, we put both the top layer and bottom layer at the geometric center of the middle layer(unshifted), then we can shift the top and bottom layer along a symmetric axis with opposite directions, hence we have another tuned parameter, shift value.

We define the Wigner-Seitz radius based on $\pi a^2 n_s = 1$, a is the w-s radius, n_s is the projected surface density. In this chapter, all the interlayer distance d values are in the unit of a if there is no further notification.

For a given lattice, we calculate the Yukawa interaction energy from all the other neighbor sites within a given radius. The total energy is $E_{total} = 3E_1 + 2E_2 + E_3$, E_1 is the energy within each layer, E_2 is the energy between the nearest



Figure 3.1: Rhombic unit cell(left), rectangle unit cell(right)



Figure 3.2: Unshifted and shifted lattice structure, both top and side view

layers, E_3 is the energy between the top and bottom layers.

We set a value for the interlayer distance. Then compare the energy from different types of lattice, the one with lowest energy is the optimal lattice.



Figure 3.3: phase diagram with different interlayer distance



Figure 3.4: phase diagram with different interlayer distance



Figure 3.5: phase diagram with different interlayer distance



Figure 3.6: phase diagram map plot with different interlayer distance and different κ . The red zone is triangle, the dark blue zone is the shifted rhombic, the green zone is rectangle, the light blue zone is square.

According to the phase diagram in Fig 3.3 to Fig 3.4, at low kappa, the phase

transition is: staggered triangle \rightarrow shifted rhombic \rightarrow square \rightarrow unshifted rhombic \rightarrow staggered triangle; at high kappa, the phase transition is : staggered triangle \rightarrow shifted rhombic \rightarrow unshifted rectangular \rightarrow square \rightarrow unshifted rhombic \rightarrow staggered triangle; Based on the well-known result that the stable state of a single layer Yukawa lattice is hexagonal, and a single layer hexagonal lattice can be decomposed into three hexagonal lattices, so when the interlayer distance is small, we should always get a staggered triangle lattice. At very large interlayer distance, since the coupling between layers is very weak, we can treat each of them like a single layer, so it also must be a triangular lattice. Lowen studied the layered structure transition under slit confinement for Coulomb *Oguz et al.* (2009a) and Yukawa *Oguz et al.* (2009b) system, experiment on colloids system *Oguz et al.* (2012) was also explored. The trilayer square lattice phase was confirmed in these works.

Whether the lattice distribution is shifted or unshifted can be characterized by the correlation energy, which is the difference between total energy and the Hartree energy. The Hartree part E_H can be calculated analytically.

$$E_H = 2\pi n \int_0^\infty \frac{e^{-\kappa\sqrt{r^2+d^2}}}{\sqrt{r^2+d^2}} r dr$$

= $\frac{2n\pi}{3\kappa} e^{-\kappa d}$ (3.1)

rescale the energy in the unit of $\frac{1}{a}$, we will get $E_H = \frac{2}{3\tilde{\kappa}}e^{-\tilde{\kappa}\tilde{d}}$.



Figure 3.7: Original lattice summation energy from different layers combination



Figure 3.8: Correlation energy from different layers combination

Form Fig 3.7 and Fig 3.8, we can see that when the correlation energy of E_3 becomes positive, it corresponds to the unshifted lattice. This makes sense as positive correlation energy means attraction between particles.



Figure 3.9: MD snapshot for particle distribution in a trilayer system with interlayer distance d = 0.9

In the previous section, we have found the optimal lattice at a certain interlayer distance. But whether it is stable, we still don't know, the collective mode dispersion can provide useful evidence to check the stability. In next section, we will study the in-plane collective mode dispersion. In fact, the real lattice structure should be based on the MD result rather than our theoretical prediction. Because we made some assumption. For example, we assumed the Bravais lattice unit cell, we assumed there is the same structure in both middle layer and top/bottom layers. From Fig 3.9 we can see that we get a weird structure like snakes, this is why in Fig 3.3, we get very messy phase transition behavior, since our model violates the reality, our prediction is not reliable in the region 0.5 < d < 1. We will not do the dispersion calculation for this region in the next section.

3.2 Lattice dispersion and eigenvectors

In order to obtain the collective mode dispersion, we need the dynamical matrix $M_{\mu\nu}^{ij}$, here, i, j is 1(middle),2(top),3(bottom); μ , ν is x, y. So the dynamical matrix is a 6×6 matrix.

$$M_{\mu\nu}^{ij}(\vec{k}) = \delta_{ij} \sum_{m=1,2,3} S_{\mu\nu}^{im}(0) - S_{\mu\nu}^{ij}(\vec{k})$$
(3.2)

$$S_{\mu\nu}^{ij}(\vec{k}) = \sum_{n} \frac{\partial^2 \varphi^{ij}(\vec{r_n})}{\partial \mu \partial \nu} e^{i\vec{k}\vec{r_n}}$$
(3.3)

 \sum_{n} means the lattice summation from all the sites within a given radius. The

n structure of dynamical matrix is in the form $\begin{pmatrix} M^{11} & M^{12} & M^{13} \\ M^{21} & M^{22} & M^{23} \\ M^{31} & M^{32} & M^{33} \end{pmatrix}$ where each subblock is $\begin{pmatrix} M^{ij}_{xx} & M^{ij}_{xy} \\ M^{ij}_{yx} & M^{ij}_{yy} \end{pmatrix}$ The dynamical matrix is always Hermitian. It always has

block is $\begin{pmatrix} M_{xx}^{ij} & M_{xy}^{ij} \\ M_{yx}^{ij} & M_{yy}^{ij} \end{pmatrix}$ The dynamical matrix is always Hermitian. It always has real eigenvalues. If our lattice is stable, all the eigenvalues must be positive, or else, some eigenvalue in some k range may be negative. Here we show the dispersion result from two types of lattice: staggered triangle, overlapped square. Note: All the dispersion graphs are generated with very fine cut of k value, the step for k is 0.0002, so we can clearly see how the lines direct to at a cross point.

Since the matrix has six dimensions, its eigen problem can only be solved by numerical calculation. Here we use the Eigen C++ library to do the computation.

When we display the six dispersion lines. We distinguish it by line continuity. We start to generate the w(k) values from k = 0.0001, we assign w(k) label from 1 to 6 based on the value of w(k). For each label, we make sure the eigenvectors from w(k) to $w(k + \Delta k)$ has the least change. More specifically, suppose we have six eigenvectors $\vec{v_n}$ at k and another six eigenvectors $\vec{v'_n}$ at $k + \Delta k$, if $\vec{v_i} \cdot \vec{v'_j} \approx 1$, we know \vec{v}_i and \vec{v}'_j belong to the same dispersion curve.

3.2.1 Dispersion for trilayer triangle lattice

Let's firstly study the dispersion for a trilayer triangular lattice, which is the stable structure when interlayer distance is close to zero or very large. Fig 3.10 shows the lattice structure. From its top view, each unit cell contains three particles (one from each layer). In Fig 3.11, each black triangle represents a unit cell. Its lattice structure is the same as that from each sublayer. But if you treat all the particles in the same symbol, the trilayer lattice becomes a single layer triangular lattice, which is different from the one of each sublayer, there is $\frac{\pi}{3}$ rotaion difference. Fig 3.12 shows the reciprocal lattice of total and sub lattices.



Figure 3.10: staggered trilayer triangle lattice structure



Figure 3.11: trilayer triangular lattice structure



Figure 3.12: Reciprocal lattice: red line is for the whole projected single layer lattice, black line is for each sublattice as well as the total trilayer



Figure 3.13: Lattice dispersion for trilayer triangle lattice

Fig 3.13 is the dispersion for different principle angles. Generally, for two arbitrary angles \vec{k} , if $\theta_1 + \theta_2 = \pi$ or 2π , their dispersion plots are the same. Based on Fig 3.10, because all the partiles are the same, we know that the dispersion from θ and $\theta + \frac{\pi}{3}$ are the same. Fig 3.14 and 3.15 shows the lattice dispersion for a single triangle lattice *Sullivan et al.* (2006), which is part of that calculated as trilayer.



Figure 3.14: Lattice dispersion for single triangle lattice



Figure 3.15: Lattice dispersion for single triangle lattice

When we increase the interlayer distance a little, the lattice structure retains. Fig 3.16 shows the lattice dispersion for interlayer distance is 0.2a. Fig 3.17 shows the lattice dispersion for interlayer distance is 3.0a.



Figure 3.16: Lattice dispersion for trilayer triangle lattice, interlayer distance is 0.2a



Figure 3.17: Lattice dispersion for trilayer triangle lattice, interlayer distance is 3.0a

Since we solve the eigenvalue of dynamical matrix to get the dispersion curve, originally, the eigenvalue is proportional to the w_p^2 , if the lattice is stable, all the eigenvalues should be positive. In the numerical calculation, if eigenvalue λ is negative, we display $-\sqrt{-\lambda}$. We can see that the w value is always positive for both d = 0.2a and d = 3.0a, which means the lattice is stable. This matches our lattice prediction.

We can see that when some two lines tend to cross, they form a gap. Fig 3.18 shows more detail about the gap formation. We will discuss the gap formation mathematically later. When the d is large enough, it converges to dispersion curve of a single layer with lower number density.



Figure 3.18: Gap formation

3.2.2 Dispersion for trilayer square lattice

At some intermediate value for d, the system prefers a square lattice(Fig 3.19). Here we show one example for d = 1.5a. Fig 3.20 to 3.21 show their dispersion curves at different principle angles. In the square lattice, the angle 0 and $\frac{\pi}{2}$ are equivalent. At every gap formed by two crossing lines, the eigenvector type exchanges rather than follows a continuous line.



Figure 3.19: top view of overlapping trilayer square lattice structure



Figure 3.20: Lattice dispersion for trilayer square lattice, interlayer distance is 1.5a



Figure 3.21: Lattice dispersion for trilayer square lattice, interlayer distance is 1.5a

3.2.3 Eigenvector analysis

Since for square lattice we have real symmetric dynamical matrix, all the eigenvectors are real, we can easily visualize it by 2D vectors. Suppose we get an eigenvector with six elements $(x_1, y_1, x_2, y_2, x_3, y_3)$, then we break it into three vectors (x_1, y_1) , (x_2, y_2) , (x_3, y_3) , we use an arrow to represent each vector. If all the tree vectors are in the same directon, it is labelled as 'A'(All), if the middle layer does move in one dirction and the other two layers are in the opposite direction, we label it as 'M'(Move). Fig 3.22 to Fig 3.25 show the eigenvector visualization for the square lattice. When $\varphi = 0$, compare the plot from ka = 0.2001 and ka = 0.3501, we can clearly see that subplot 2 and subplot 5 exchange pattern. Generelly, for acoustic mode at $k \to 0$, the slope of longitudinal(L) mode is always greater that of transverse(T) mode. For ka = 0.2001 and $\varphi = 0$ case, it is along the k_x direction, the vertical(1) displacement is T, the horizontal(2) displacement is L. For $\varphi = 90$, the k_y direction, we can see the subplot 1 and subplot 2 exchange order, which makes sense.



Figure 3.22: Displacement visualization of the eigenvectors, interlayer distance is 1.5a, φ is the propagation angle in k space respect to k_x axis, the length of the arrows means its magnitude, the black, red, green corresponds to layer 1-middle,2-upper,3-lower



Figure 3.23: Displacement visualization of the eigenvectors, interlayer distance is 1.5a, φ is the propagation angle in k space respect to k_x axis, the length of the arrows means its magnitude, the black, red, green corresponds to layer 1-middle,2-upper,3-lower



Figure 3.24: Displacement visualization of the eigenvectors, interlayer distance is 1.5a, φ is the propagation angle in k space respect to k_x axis, the length of the arrows means its magnitude, the black, red, green corresponds to layer 1-middle,2-upper,3-lower



Figure 3.25: Displacement visualization of the eigenvectors, interlayer distance is 1.5a, φ is the propagation angle in k space respect to k_x axis, the length of the arrows means its magnitude, the black, red, green corresponds to layer 1-middle,2-upper,3-lower

Generally, with finite interlayer distance, whenever two dispersion curves form a gap, they exchange eigenvector type. Or in other words, The slope of the dispersion curve from a given polarization type won't change sign around a cross point. This can be seen based on the matrix structure, for example, in square lattice, along $\varphi = 0^{\circ}$ or 90° direction, all the M_{xy} elements are zero. The dynamical matrix can be decomposed into two 3×3 sub-matrix(XX and YY). Each sub-

matrix has a structure as $\begin{pmatrix} A & B & B \\ B & F & C \\ B & C & F \end{pmatrix}$ The eigenvalues and eigenvectors will be

discussed in the next chapter.

For staggered hexagonal lattice, the dynamical matrix is complex, its eigenvectors are also complex. We can't simply use 2D vector to display the displacement. Here we use elliptical plot to display the complex eigenvectors. For a given eigenvector $(x_1, y_1, x_2, y_2, x_3, y_3)$, here all the numbers are complex, when display (x_1, y_1) , we get $\frac{x_1}{y_1} = re^{i\theta}$, where r and θ are real numbers. The visualization is based on the parametric equations

$$\begin{cases} \xi = r \sin(wt + \theta) \\ \eta = \sin(wt) \end{cases}$$
(3.4)

because sometimes r can be close to zero or infinity, practically, we plot it based on

$$\begin{cases} \xi = s \frac{r}{\sqrt{1+r^2}} \sin(wt + \theta) \\ \eta = s \frac{1}{\sqrt{1+r^2}} \sin(wt) \end{cases}$$
(3.5)

where $s = \sqrt{|x_1|^2 + |y_1|^2}$, Eq 3.5 only gives us the obit. However, the obit from (x_2, y_2) and (x_3, y_3) are always the same, in order to distinguish them, we will

define the phase difference between layers.

$$\begin{cases} \frac{x_2}{x_1} = \lambda_2 e^{i\delta_2} \\ \frac{x_3}{x_1} = \lambda_3 e^{i\delta_3} \end{cases} \begin{cases} \frac{y_2}{y_1} = \rho_2 e^{i\varepsilon_2} \\ \frac{x_3}{x_1} = \rho_3 e^{i\varepsilon_3} \end{cases}$$
(3.6)

we use δ and ε to characterize the phase difference. In each sub-eigenvector (x_1, y_1) , (x_2, y_2) and (x_3, y_3) , its θ , δ and ε can uniquely distinguish it from others. Here we give an example for the phase values. When d=0.2, $\varphi = 0$, we have the following conclusion for δ and ε .

- 1. θ_2 and θ_3 : one of them is $\frac{\pi}{2}$, the other one is $-\frac{\pi}{2}$
- 2. $\delta_2 \delta_3$ and $\varepsilon_2 \varepsilon_3$ are always 0 or π . When $\delta_2 \delta_3$ is 0, $\varepsilon_2 \varepsilon_3$ must be π ; When $\delta_2 - \delta_3$ is π , $\varepsilon_2 - \varepsilon_3$ must be 0.
- When δ₂ δ₃ or ε₂ ε₃ is 0, δ₂ or ε₂ must be either 0 or π, 0 means in phase, π means out of phase.

In the elliptical plot legend, it is in a form of $\begin{pmatrix} 1-R\\ 2-R,Y+\\ 3-L,Y+ \end{pmatrix}$, the number means

the layer label. The second column is letter R or L, it represents right rotating orbit or left rotating orbit. If θ is positive, it is L, or else it is R. After the comma, it is letter X or Y, if $\delta_2 - \delta_3$ is 0, it is X, if $\varepsilon_2 - \varepsilon_3$ is 0, it is Y. The last sign is either + or -, if δ_2 or ε_2 is 0, it is +; if δ_2 or ε_2 is π , it is -.

These conclusions above are only true for k_x direction. For other k direction, we will get different conclusions, but θ , δ and ε still can distinguish each eigenvector uniquely.



Figure 3.26: Displacement visualization of the complex eigenvecotors



Figure 3.27: Displacement visualization of the complex eigenvectors



Figure 3.28: Displacement visualization of the complex eigenvecotors



 $ka = 1.9001, \phi = 0, d = 0.2, \kappa = 0.4$

Figure 3.29: Displacement visualization of the complex eigenvectors

From Fig 3.26 and Fig 3.29 we can find that only at k=0 or k at the boundary of BZ, we can see obit with middle layer being almost zero, which represents S mode.

CHAPTER IV

QLCA and MD comparison for trilayer liquid system

4.1 QLCA prediction

In the last Chapter, we have studied the dispersion relation for a trilayer lattice. In this chapter, we will calculate the dispersion for a trilayer liquid system by Quasilocalized Charge Approximation(QLCA), where we need to borrow pair distribution data from MD simulation. The QLCA was firstly proposed by Kalman(Kalman and Golden, 1990; Kalman et al., 2005; Golden and Kalman, 2000).

The QLCA is proposed to calculate the collective mode dispersion of Yukawa or Coulomb plasma at high Γ liquid phase. In lattice phase, each particle is just oscillating with small amplitude around a fixed site, when we calculate the phonon dispersion from the dynamical matrix, which is based on the lattice site summation. In liquid phase, a particle has no fixed average position, it can diffuse to anywhere in the system. In a shorter time scale, a particle oscillates in a local potential well, this is the quasilocalization.

All the content in this chapter is for Yukawa system with $\kappa = 0.4$. The collective modes of the system are the solution of the characteristic equation

 $||\omega^2 \delta_{\mu\nu} \delta^{ij} - C^{ij}_{\mu\nu}|| = 0$, where i, j indicate the layer and μ, ν the 2D spatial coordinate. The middle layer is identified by i = 1, the top layer by i = 2, and bottom layer j = 3. This leads to a 6×6 dynamical matrix whose elements are given by

$$C^{ij}_{\mu\nu}(\vec{k}) = \delta_{ij} \sum_{m=1,2,3} S^{im}_{\mu\nu}(0) - S^{ij}_{\mu\nu}(\vec{k})$$
(4.1)

In lattice

$$S^{ij}_{\mu\nu}(\vec{k}) = \frac{1}{m} \sum_{n} \frac{\partial^2 \varphi^{ij}(\vec{r}_n)}{\partial r_\mu \partial r_\nu} e^{i\vec{k}\vec{r}_n}, \qquad (4.2)$$

In QLCA, we replace the lattice site summation by an integral based on $g(\vec{r})$,

$$S^{ij}_{\mu\nu}(\vec{k}) = \frac{n}{m} \int \psi^{ij}_{\mu\nu}(\vec{r}) e^{i\vec{k}\vec{r}} g(\vec{r}) d\vec{r}, \qquad (4.3)$$

where

$$q^2 \psi^{ij}_{\mu\nu}(r) = \frac{\partial^2 \varphi^{ij}(r)}{\partial r_\mu \partial r_\nu},\tag{4.4}$$

 $n = \frac{n_s}{3}$ is the surface density of a single layer, $r^2 = \rho^2 + d^2$, $\rho^2 = x^2 + y^2$, and d the inter-layer distance between layer *i* and *j*. Rescaling the matrix leads to

$$S_{\mu\nu}^{ij}(k) = \frac{2\pi n_s q^2}{ma} \frac{a}{6\pi} \frac{1}{a^3} a^2 \int_0^\infty \int_0^{2\pi} \psi_{\mu\nu}^{ij}(\rho, d, \theta) e^{ik\rho\cos\theta} g_{ij}(\rho) \rho d\rho d\theta$$

$$= \frac{\omega_p^2}{6\pi} \int_0^\infty \int_0^{2\pi} \psi_{\mu\nu}^{ij}(\rho, d, \theta) e^{ik\rho\cos\theta} g_{ij}(\rho) \rho d\rho d\theta$$
(4.5)

Keep in mind, in the following formulas, all the length variables are in the unit of a. Since the final result will be scaled by plasma frequency, we will ignore w_p^2 in the future. Let's firstly calculate the angular integral part,

$$f^{ij}_{\mu\nu}(k\rho,r) = \frac{1}{6\pi} \int_0^{2\pi} \psi^{ij}_{\mu\nu}(\rho,r,\theta) e^{ik\rho\cos\theta} \mathrm{d}\theta$$
(4.6)

$$\psi_{xx}^{ij} = \frac{e^{-\kappa r}}{r^5} [x^2(\kappa^2 r^2 + 3\kappa r + 3) - (1 + \kappa r)r^2]$$
(4.7a)

$$\psi_{yy}^{ij} = \frac{e^{-\kappa r}}{r^5} [y^2(\kappa^2 r^2 + 3\kappa r + 3) - (1 + \kappa r)r^2]$$
(4.7b)

$$\psi_{xy}^{ij} = \frac{e^{-\kappa r}}{r^5} [xy(\kappa^2 r^2 + 3\kappa r + 3)]$$
(4.7c)

 $x = \rho \cos \theta, y = \rho \sin \theta$, It is easy to see that the matrix element with ψ_{xy} integral is always zero. Define $A = \kappa^2 r^2 + 3\kappa r + 3$ and $B = 1 + \kappa r$, then

$$\begin{cases} f_{xx}^{ij} = \frac{1}{6\pi} \frac{e^{-\kappa r}}{r^5} \int_0^{2\pi} (A\rho^2 \cos^2\theta - Br^2) e^{ik\rho\cos\theta} d\theta \\ f_{yy}^{ij} = \frac{1}{6\pi} \frac{e^{-\kappa r}}{r^5} \int_0^{2\pi} (A\rho^2 \sin^2\theta - Br^2) e^{ik\rho\cos\theta} d\theta \end{cases}$$
(4.8)

The final result is

$$\begin{cases} f_{xx}^{ij} &= \frac{e^{-\kappa r}}{3r^5} [(\frac{J_1}{k\rho} - J_2)A\rho^2 - Br^2 J_0] \\ f_{yy}^{ij} &= \frac{e^{-\kappa r}}{3r^5} [\frac{J_1}{k\rho}A\rho^2 - Br^2 J_0] \end{cases}$$
(4.9)

J is the Bessel function of $k\rho$, at $k \to 0$ limit,

$$f_{xx}^{ij}(k \to 0) = f_{yy}^{ij}(k \to 0) = \frac{e^{-\kappa r}}{3r^5} (\frac{1}{2}A\rho^2 - Br^2)$$
(4.10)

, this means the transverse and longitudinal modes are degenerated at $k \to 0$, the last step is to calculate the integral $S^{ij}_{\mu\nu} = \int_0^\infty f_{\mu\nu} g_{ij}(\rho) \rho d\rho$, $g_{ij}(\rho)$ is from MD simulation. The dynamical matrix is a real symmetric matrix, it always has real eigenvalues. The structure of dynamical matrix is in the form $\begin{pmatrix} C^{11} & C^{12} & C^{13} \\ C^{21} & C^{22} & C^{23} \\ C^{31} & C^{32} & C^{33} \end{pmatrix}$ where each sub-block is $\begin{pmatrix} C_{xx}^{ij} & 0 \\ 0 & C_{yy}^{ij} \end{pmatrix}$ Because the middle layer has different environment from top or bottom layer, we should keep in mind that $g_{11} \neq g_{22}$ except two limit cases where d = 0 or ∞ . Fig 4.1 to Fig 4.2 are some examples of the pair distribution function.



Figure 4.1: Inter/intra-layer pair distribution functions for $\Gamma = 160$, (a) d = 3.0, (b) d = 1.5, (c) = 0.5, (d) d = 0.2, note that the $g_{23}(r \to 0) > 1$ at larger d value and $g_{23}(r \to 0) \to 0$ at smaller d value . In (a), g_{11} and g_{22} nearly overlap; In (d), g_{11} and g_{12} nearly overlap



Figure 4.2: The same as Fig. 4.1 for $\Gamma = 10$

When we plot the dispersion curve, we plot the lines by there eigenvector type rather than the line continuity. The same as that in the previous chapter. Fig 4.3 illustrates the six types of eigenvectors.



Figure 4.3: Six types of eigenvector, the length of each arrow represents the displacement of each layer, from 1 to 6, they are: AT,AL,MT,ML,ST,SL

In fact, since all C_{xy} terms are zero, the dynamical matrix can be decoupled into two 3×3 matrix, representing XX and YY subspace. The displacement pattern of the eigenvector can be seen based on the matrix structure, each sub-matrix has a

structure as $\begin{pmatrix} A & B & B \\ B & F & D \\ B & D & F \end{pmatrix}$ The eigenvalues of C-matrix are $\begin{cases} \alpha_1 = F - D \\ \alpha_2 = \frac{A + D + F - \sqrt{\Delta}}{2} \\ \alpha_3 = \frac{A + D + F + \sqrt{\Delta}}{2} \end{cases}$ (4.11)

their corresponding eigenvectors are

$$\begin{cases} v_1 = (0, -1, 1) \\ v_2 = (-\frac{-A+D+F+\sqrt{\Delta}}{2B}, 1, 1) \\ v_3 = (-\frac{-A+D+F-\sqrt{\Delta}}{2B}, 1, 1) \end{cases}$$
(4.12)

where $\Delta = (F + D - A)^2 + 8B^2$.

Each eigenvalue represents a mode, a mode's property is determined by its eigenvector. (0,-1,1) means the middle layer doesn't move while top and bottom layer move in the opposite directions, this is labelled as S mode. Since $-A + D + F + \sqrt{\Delta}$ is always positive and $-A + D + F - \sqrt{\Delta}$ is always negative, the displacements of the middle layer in v_2 and v_3 always have different signs. When the displacement of all the three layers have same sign, it is labelled as A mode. When the displacement of the middle layer has different sign with the top or bottom ones, it is labelled as M mode. The eigenvectors are visualized in Fig 4.3. The S mode displacement is independent of k, while A and M mode displacement type with M mode. Based on the C-matrix elements in the appendix, it is easy to show $A + D + F = \sqrt{\Delta}$ and B < 0 when k=0, which means α_2 is acoustic mode at small k region. The first element in v_2 is positive, it indicates α_2 is A mode. A mode is acoustic, this is physically reasonable. Accordingly $\alpha_3 = -3B$ when k=0.

The two optical eigenvalues at k=0 are $\alpha_1 = F - D = -B - 2D$ and $\alpha_3 = -3B$. How close the two gap values are is determined by the difference between g_{12} and g_{23} . If $g_{12}=g_{23}$, the S and M gap values are the same.

In the $d \to 0$ limit, according to the MD simulations $g_{11} = g_{22/33} = g_{12} = g_{23}$. Then the eigenvalues become (A - B, A - B, A + 2B). Suppose (x_1, x_2, x_3) is the eigenvector, A-B gives constraint $x_1 + x_2 + x_3 = 0$, A+2B gives constraint $x_1 = x_2 = x_3$, so we can choose (1, 1, 1) as the eigenvector for A+2B, (0, -1, 1), (-2, 1, 1) for A-B. Obviously, A+2B represents the acoustic mode, while A-B represents the S and M modes. Recall the Eq 4.1, $A = 3C^{11}(0) - C^{11}(k)$, $B = -C^{12}(k)$, since
$g_{11} = g_{12}$, we have $C^{11}(k) = C^{12}(k)$, then we have

$$\begin{cases}
A - B = 3C^{11}(0) \\
A + 2B = 3[C^{11}(0) - C^{11}(k)]
\end{cases}$$
(4.13)

The optical mode A-B is independent of k. Because $3C^{11} = C^{total}$ and we use $C^{total}(0) - C^{total}(k) = \omega^2$ to get the dispersion curve for a projected single layer, the acoustic curves in trilayer QLCA is exactly the same as the result in single layer. However, the gap mode $3C^{11}(0)$ is just the Einstein frequency of the projected single layer. This has been noticed in *Golden and Kalman* (2003).

Fig 4.4 to 4.6 are the QLCA dispersion of the six modes. When Γ is higher, the dispersion curves have deeper fluctuation, this is because at high Γ , the system behaves more like a lattice. A very important property is that the M and A modes can't cross each other. At the 'crossing' point, it forms a gap and exchanges eigenvector type. But the S mode can cross other lines. Fig 4.8 shows a more clear view of the crossing situation.



Figure 4.4: QLCA dispersion of six modes for d = 0.2



Figure 4.5: QLCA dispersion of six modes for d = 1.0



Figure 4.6: QLCA dispersion of six modes for d = 3.0



Figure 4.7: Avoided crossing between A and M modes dispersion curves for $\Gamma{=}160,\,d=0.5$



Figure 4.8: S mode dispersion curves for Γ =160, d = 0.5, each mode is always along a continuous line. The real crossing indicates S mode doesn't entangle with other two modes.

4.2 Extracting collective mode information from MD data

In last section, we finished the QLCA calculation, now we should compare the calculation with the MD simulation result.

4.2.1 Current correlation function and VAF

We determine the collective mode dispersion by the current correlation function. Set $\vec{v}_i(t)$, $\vec{r}_i(t)$ as the velocity and position of i^{th} particle at time t. Then the current is defined as(*Hansen and McDonald*, 2013)

$$\vec{j}(\vec{r},t) = \sum_{i=1}^{N} \vec{v}_i(t) \delta[\vec{r} - \vec{r}_i(t)]$$
(4.14)

where N is the number of particles. Its Fourier component is

$$\vec{j}(\vec{k},t) = \sum_{i=1}^{N} \vec{v}_i(t) e^{-i\vec{k}\cdot\vec{r}_i(t)}$$
(4.15)

The longitudinal current correlation function is

$$C_l(\vec{k},t) = \frac{1}{N} \langle [\vec{k} \cdot \vec{j}(\vec{k},t)] [\vec{k} \cdot \vec{j}(-\vec{k},0)] \rangle$$
(4.16)

The transverse current correlation function is

$$C_t(\vec{k},t) = \frac{1}{N} \langle [\vec{k} \times \vec{j}(\vec{k},t)] [\vec{k} \times \vec{j}(-\vec{k},0)] \rangle$$

$$(4.17)$$

In a liquid system, due to the isotropic property, we can simply set the \vec{k} in front of \vec{j} as unit vector along k_x direction, then we get

$$C_{l}(k,t) = \frac{1}{N} \langle j_{x}(k,t)j_{x}(-k,0) \rangle$$
(4.18a)
$$C_{k}(k,t) = \frac{1}{N} \langle j_{x}(k,t)j_{x}(-k,0) \rangle$$
(4.18b)

$$C_t(k,t) = \frac{1}{N} \langle j_y(k,t) j_y(-k,0) \rangle$$
(4.18b)

After the time Fourier transform, the current correlation function is

$$C(\vec{k}, w) = \lim_{T \to \infty} \frac{1}{2\pi T} \int_0^T C(\vec{k}, t) e^{iwt} dt$$
(4.19)

Then we look at the time Fourier transformed correlation function $C(\vec{k}, w)$, based on the peak position we determine the dispersion relation.

In Eq 4.16 and 4.17, the angle brackets mean the ensemble average. Assume the system is ergodic, we can use the time average to calculate the ensemble average. Practically, we do this current calculation in a different way because it is convenient to do numerically.

Let us use C_l as an example to show the derivation. The angle brackets mean

$$C_l(k,t) = \lim_{T_1 \to \infty} \frac{1}{N} \frac{1}{T_1} \int_0^{T_1} j_x(k,t+t') j_x(-k,t') dt'$$
(4.20)

Do the time Fourier transform

$$C_{l}(k,w) = \lim_{\substack{T_{1} \to \infty \\ T \to \infty}} \frac{1}{N} \frac{1}{2\pi T_{1}T} \int_{0}^{T_{1}} \int_{0}^{T} j_{x}(k,t+t') j_{x}(-k,t') e^{iwt} dt dt'$$

$$= \lim_{\substack{T_{1} \to \infty \\ T \to \infty}} \frac{1}{N} \frac{1}{2\pi T_{1}T} \int_{0}^{T_{1}} \int_{0}^{T} j_{x}(k,t+t') j_{x}(-k,t') e^{iw(t+t')} e^{iw(-t')} dt dt'$$

$$= \frac{2\pi}{N} j_{x}(k,w) j_{x}(-k,-w)$$
(4.21)

Generally, we do the MD measurement such as

$$C_l^{AB}(k,w) = \frac{2\pi}{\sqrt{N_A N_B}} j_x^A(k,w) j_x^B(-k,-w)$$
(4.22)

A,B represent the species. Since $j_x(k,w) = j_x^*(-k,-w)$, Eq 4.22 is always real for $C_l^{AA}(k,w)$, but it is complex for $C_l^{AB}(k,w)$. In order to reduce the random error and get high quality profile of $C_l^{AB}(k,w)$, we repeat the measurement for Eq 4.22 multiple times, then calculate its arithmetic average. Another useful term is density-density correlation function, it is defined as

$$F(\vec{k},t) = \frac{1}{N} \langle \rho_{\vec{k}}(t) \rho_{-\vec{k}}(0) \rangle \tag{4.23}$$

where

$$\rho_{\vec{k}}(t) = \sum_{i=1}^{N} e^{-i\vec{k}\cdot\vec{r}_i(t)}$$
(4.24)

The Fourier form of Eq 4.23 is

$$S(\vec{k}, w) = \lim_{T \to \infty} \frac{1}{2\pi T} \int_0^T F(\vec{k}, t) e^{iwt} dt$$
 (4.25)

Based on the definition above, according to Eq (7.4.28) in Hansen and Mc-

Donald (2013), we should have

$$C_l(k,w) = \frac{w^2}{k^2} S(k,w)$$
(4.26)

In the following text, we also use L(w, k), T(w, k) to represent $C_l(k, w)$ and $C_t(k, w)$.

Let us also look at the velocity auto-correlation function(VAF). It describes the behavior of a single particle. Peter used the definition like

$$Z(t) = \frac{\langle \vec{v}(t) \cdot \vec{v}(0) \rangle}{\langle |\vec{v}(0)|^2 \rangle}$$
(4.27)

Because $\langle |\vec{v}(0)|^2 \rangle \propto k_B T$, it is constant for any particle in an equilibrium system, we can simply just analyze the behavior of $\langle \vec{v}(t) \cdot \vec{v}(0) \rangle$.

$$Z(t) = \langle v_x(t)v_x(0) \rangle + \langle v_y(t)v_y(0) \rangle$$
(4.28)

In liquid or disordered lattice layer system, the system is isotropic, it is safe to say x component is equivalent to the y component(Not true for an ordered lattice). We can simply just analyze x component. Follow the derivation in Eq 4.21, we can have

$$Z(w) = v_x(w)v_x(-w) \tag{4.29}$$

Do the average among different particles, we have

$$Z(w) = \frac{1}{N} \sum_{i} v_x^i(w) v_x^i(-w)$$
(4.30)

Since $v_x(w) = v_x^*(-w)$, Z(w) is also real.

4.2.2 Extracting collective mode information

For a trilayer system, we should have four distinct current correlation functions: $C_{11}, C_{22}, C_{12}, C_{23}$. According to the eigenvector type in QLCA calculation, we can predict the peak profile in the current correlation functions as following:

 $C_{11} \rightarrow$ two positive peaks

 $C_{12} \rightarrow$ one positive peak, one negative peak

 $C_{22} \rightarrow$ three positive peaks

 $C_{23} \rightarrow$ two positive peaks, one negative peak

This can be confirmed by the MD result(Fig 4.9).



Figure 4.9: peaks profile in longitudinal current correlation function, Γ =160, d=3.0, ka = 0.32032, the letter A, M, S label the corresponding mode

Since there are multiple peaks, how do we distinguish them? Based on the physical meaning of the eigenvectors, we can know: C_{11} contains A, M modes; C_{12} contains A, S, M modes; C_{12} contains A, M modes, the positive one is A and the negative one is M; C_{23} contains A, S, M modes, the negative one is S. Since the

position of each peak is independent, sometimes two of them may overlap, this is why in Fig 4.9 we choose a special k value in order to show all the possible peaks. Moreover, sometimes the S, M mode peak is very weak compared with A mode, we will not try to locate the peak position in the original current correlation function picture such as Fig 4.9. We will do the following combination:

$$C_{11}$$
- $C_{12} \to M \mod e$

$$C_{11} + C_{12} \to A \mod C_{11}$$

$$C_{22}$$
- $C_{23} \to S \mod e$

After such a summation or subtraction, we expect to see a single peak representing the collective mode, whose amplitude will be enhanced by twice. We do the Gaussian fit to determine the peak position and use its half height width as the error bar range. Here we show some comparison between the MD and QLCA. For A mode, we always get good agreement between MD and QLCA. For the gap modes M and S, only at larger inter-layer distance can we get good agreement.

4.3 QLCA and MD result comparison

Let us look at Fig 4.9 again, this is picture for d = 3.0, we can see the A,M,S mode peaks, if we set $d \rightarrow 0$, C_{11} , C_{22} should be equivalent, C_{12} , C_{23} , C_{33} should also be equivalent. This means the M, S mode peaks should disappear. Only A mode will remain. This analysis is confirmed by the simulation data, in Fig 4.10, we do see only one strong peak, the A mode. There is some weak peak, we will discuss it in the next section. This type of profile also happens in transverse current correlation function(Fig 4.11). So we will display the comparison of A mode for small d and A, S, M modes for large d.



Figure 4.10: peaks profile in longitudinal current correlation function, Γ =160, d=0.2, ka = 0.32032,



Figure 4.11: peaks profile in transverse current correlation function, $\Gamma=160$, d=0.2, ka = 0.32032,

Fig 4.12 to 4.19 are the dispersion comparison between MD and QLCA, the red line is the QLCA, the blue square is MD. Each column corresponds to some d value. The first row is the longitudinal and the second row is transverse. In each subplot, Y axis is $\omega/\omega_{\rm p}$, X axis is ka. The blue square is the peak value from the Gaussian fit on the current correlation function. The error bar is the half height width from the Gaussian fit. We can see that we get very good agreement between MD and QLCA.



Figure 4.12: A mode dispersion comparison between MD and QLCA Γ =10, d = 0.2, 0.5, 1.0



Figure 4.13: A mode dispersion comparison between MD and QLCA $\Gamma{=}10,\,d=1.5,2.0,3.0$



Figure 4.14: A mode dispersion comparison between MD and QLCA $\Gamma{=}160,\,d=0.2,0.5,1.0$



Figure 4.15: A mode dispersion comparison between MD and QLCA $\Gamma{=}160,\,d=1.5,2.0,3.0$



Figure 4.16: M mode dispersion comparison between MD and QLCA $\Gamma{=}160,\,d=1.5,2.0,3.0$



Figure 4.17: S mode dispersion comparison between MD and QLCA $\Gamma{=}160,\,d=1.5,2.0,3.0$



Figure 4.18: M mode dispersion comparison between MD and QLCA $\Gamma{=}10,\,d=1.5,2.0,3.0$



Figure 4.19: S mode dispersion comparison between MD and QLCA Γ =10, d = 1.5, 2.0, 3.0

From these dispersion comparison above, we can see that QLCA result always matches the MD result in lower k range except for T current correlation functions, since weakly coupled liquid system can't support transverse mode, this cut off has been discussed in *Schmidt et al.* (1997); *Hartmann et al.* (2007) At higher k range, there is some deviation but the QLCA still can match the MD tendency.

When d is small, if we still follow the procedure above, the extracted MD disperison curve will have an obvious discrepancy compared with the QLCA prediction for S and M mode, which is similar to the previous result of bilayer system *Kalman et al.* (1999); *Donkó et al.* (2003b,a).

4.3.1 More about the current correlation function

In Fig 4.16, you can see there is an abrupt jump in the dispersion extracted from the MD simulation. Here we will explain it.

We assumed the current correlation function profile in Fig 4.9, but when d=1.5, there is a little difference. From Fig 4.20, we can see it contains more than the

A,S,M modes as we assumed.



Figure 4.20: T current correlation function $\Gamma=160, d=1.5$

Fortunately, we have found some clue for the extra peaks in the current correlation function. Recall the lattice structure for a trilayer system at d=1.5, we know it is an overlapping square lattice. Let's compare the current correlation function in a density map plot. From Fig 4.21 to Fig 4.23, in the left panel, there is a thin black line sketching the extra peaks, you can find it in the right panel at the same position.(Notice: the lattice simulation is based on a lattice with 45° rotation from Fig 3.19) So the extra peaks can be found in the lattice dispersion. A simple answer to this phenomenon is that the trilayer liquid system at $\Gamma = 160$ is similar to a lattice system. This can be seen from the particle snapshot in Fig 4.24.



Figure 4.21: Current correlation function comparison between the liquid and lattice phase for d = 1.5. Left: T_{11} from liquid system with $\Gamma = 160$; Right: T_{11} from trilayer overlapping square lattice system.



Figure 4.22: Current correlation function comparison between the liquid and lattice phase for d = 1.5. Left: T_{11} from liquid system with $\Gamma = 160$; Right: L_{11} from trilayer overlapping square lattice system.



Figure 4.23: Current correlation function comparison between the liquid and lattice phase for d = 1.5. Left: T_{22} from liquid system with $\Gamma = 160$; Right: T_{22} from trilayer overlapping square lattice system.



Figure 4.24: particle snapshot for trilayer liquid system at $\Gamma = 160, d = 1.5$

Furthermore, you can also see a gap in the AT dispersion curve in Fig 4.14, we have found the reason. Let's look at the T correlation function for a given kavalue. In Fig 4.25, we can't see a strong A mode peak at high ka value(In Fig 4.11, we do see the A mode peak). When we do the Gaussian fit for $T_{11} - T_{12}$, we will get the higher peak value of the envelope, which is not what we want. Some more accurate numerical method will be explored in the future.



Figure 4.25: T current correlation function $\Gamma=160, d=0.2$

4.4 Current correlation function at small d

When the interlayer distance is small, we see some weak peak. What are they? Let's firstly take a careful look at it.



Figure 4.26: L_{11}, L_{12} at d=0.2, Γ =160



Figure 4.27: L_{22}, L_{23} at d=0.2, Γ =160



Figure 4.28: T_{11}, T_{12} at d=0.2, Γ =160



Figure 4.29: T_{22}, T_{23} at d=0.2, Γ =160



Figure 4.30: L_{11}, L_{12} at d=0.2, Γ =10



Figure 4.31: L_{22}, L_{23} at d=0.2, Γ =10



Figure 4.32: T_{11}, T_{12} at d=0.2, Γ =10



Figure 4.33: T_{22}, T_{23} at d=0.2, Γ =10

Fig 4.27 to Fig 4.33 are the profile of MD current correlation function. Each picture contains three subplots. The first and second ones are either C_{11} and

 C_{22} or C_{22} and C_{23} . The third subplot is just the difference between the first two subplot, that is to say, $C_{11} - C_{12}$ or $C_{22} - C_{23}$. Each subplot contains six lines, each line corresponds to a certain ka value(See the legend box). In the third subplot, the A mode peak has been cancelled. The X axis is ka, the Y axis is in arbitrary unit.

We can see that no matter which combination we choose(T or L, 11-12 or 22-23), the profile (Fig 4.27 to Fig 4.33) in the third subplot is always nearly the same(Here the strongest A mode peak is cancelled). Genreally, L/T contains an A mode peak and an envelope. There are two properties about the envelope:

- 1. C_{ii} always contains a positive envelope, while C_{ij} always contains a negative envelope
- 2. the envelope's amplitude in C_{ii} is always almost twice of that from C_{ij}

At lower gamma, it shows a central diffusive peak and another single peak; at higher gamma, it shows two peaks. In fact, these peaks don't seem to be collective mode, they are more like the peaks in the velocity auto-correlation function(VAF) Hansen et al. (1975); Schmidt et al. (1997); Hartmann et al. (2005). Normally, at $k \rightarrow 0$ limit, a collective mode peak should be very sharp, like a Dirac function rather than a broad envelope. The two properties is compatible to the previous single 2D layer result Kalman et al. (2004). We can see only one single collective peak without only any envelope. Since

$$C_{total} = \frac{1}{3} [C_{11} + C_{22} + C_{33} + 2Re(C_{12} + C_{13} + C_{23})]$$
(4.31)

On the right side, all the envelope profiles perfectly cancel out, only a single peak remains. Up to now, we may post a conjecture that the envelope is just VAF. We will continue to explore the formation of this envelope.

4.4.1 When $k \to 0$ and $d \to 0$ in a trilayer system

Let's try to analyze the trilayer system as a 2D single layer. According equation 4.15,

$$\vec{j}(k=0,t) = \sum_{m=1}^{N} \vec{v}_m(t)$$
 (4.32)

The summation is among all the particles, because the momentum is conserved, $\vec{j}(k=0,t)$ is constant. We will see a Dirac peak at w = 0 in C(k=0,w), which coincides with the fact that ω and k are both zero in acoustic mode in a single layer. Numerically we should only see some noise.

Now, we randomly label the particles into three groups, the C(k = 0, w) shows some pattern. The only reason is that total momentum in each subgroup is not constant due to fluctuation. Assume the momentum fluctuation is $\vec{\xi}^{A,B,C}(t)$, their summation must be zero,

$$\vec{\xi}^{A}(t) + \vec{\xi}^{B}(t) + \vec{\xi}^{C}(t) = 0$$
(4.33)

For simplicity, here we just consider its x component. Its Fourier transformed form is

$$\xi_x^A(w) + \xi_x^B(w) + \xi_x^C(w) = 0 \tag{4.34}$$

Multiply $\xi_x^A(-w)$ on both sides, we can get

$$\xi_x^A(w)\xi_x^A(-w) + \xi_x^B(w)\xi_x^A(-w) + \xi_x^C(w)\xi_x^A(-w) = 0$$
(4.35)

then we can have

$$C_l^{AA}(w) + C_l^{AB}(w) + C_l^{AC}(w) = 0$$
(4.36)

Because $C_l^{AB}(w)$ and $C_l^{AC}(w)$ are equivalent, then we can have

$$C_l^{AA}(w) = -\frac{1}{2}C_l^{AB}(w)$$
(4.37)

This is the ratio we see in the MD data. We also know that the profile of $C_l^{AA}(w)$ is similar to Z(w), but we can't prove that $\xi_x^A(w)\xi_x^A(-w) \propto v_x(w)v_x(-w)$ strictly. We can only use an phenomenological model to explain it. Consider just one particle, it can collide with the group A,B,C as medium for them to exchange momentum, under this picture, $v_x(t) = \xi_x(t)$, the envelope is exactly VAF. At low Γ limit, the system behaves like ideal gas, particles have little correlation. The momentum fluctuation in each group should behave similar to a single particle.

Based on the formula (8.2.18) and (7.5.17) in book*Hansen and McDonald* (2013), we have

$$Z(w) = w^{2} \lim_{k \to 0} \frac{S_{s}(k, w)}{k^{2}}$$
(4.38)

$$S_s(k,w) = \sqrt{\frac{\beta m}{2\pi k^2}} e^{-\frac{\beta m w^2}{2k^2}}$$
(4.39)

Here the Eq 4.39 is only true at ideal gas limit, which means very low Γ . So we know that

$$Z(w) \sim \lim_{k \to 0} \frac{w^2}{k^3} e^{-\frac{w^2}{2k^2}}$$
(4.40)

this is what we observed in MD data(see picture 4.34 and 4.35), it confirms the envelope is highly close to VAF. On the other hand, because $C_t(k, w)$ is not proportional to the S(k, w), in Fig 4.32 and Fig 4.33 we can't see a decay in the envelope at $k \to 0$ limit. Of course ideal gas limit is for very low Γ , in our simulation, Γ is about 10, so equation 4.39 can't predict the peak at finite frequency in VAF, we will explain it later.



Figure 4.34: envelope in L at low Γ , $\Gamma = 10$, the envelope is k dependent



Figure 4.35: a plot of equation 4.40, the legend box is the k value for each line, its tendency coincides with our MD data

4.5 Current correlation function in trilayer lattice

In last chapter, we studied the trilayer lattice dispersion. Especially for d=0, it is a staggered hexagonal lattice, we do get six different modes. But why we can not observe it in the MD simulation with small d? Let's look at a snapshot of from MD when d=0 and high Γ value(see Fig 4.36). We can see the particles from the three layers are buckled each other with random distribution, it is not a good periodic structure. In order to compare our lattice dispersion calculation, we design a new MD simulation. We initialize a single layer hexagonal lattice, then label the lattice sites according to the Fig 3.10. The system is under a very high Γ , which means the kinetic energy of each particle is very small, each particle can only do small amplitude oscillation near a fixed site, its mean position will not change during the simulation process.



Figure 4.36: projected top view of an trilayer liquid at $\Gamma = 160$, d=0.2, the red circle is middle layer, the green triangle is the top layer, the blue square is bottom layer.



Figure 4.37: L current correlation function intensity map for an ordered trilayer lattice at $\Gamma=1000,$ d=0



Figure 4.38: T current correlation function intensity map for an ordered trilayer lattice at $\Gamma = 1000$, d=0. Notice: T in our simulation is calculated by choosing k_y and v_x .

From Fig 4.37 and Fit 4.38, we can see MD result does match the calculated lattice dispersion perfectly. On one hand, it confirms the lattice calculation, on the other hand, it shows that even at small interlayer distance, we can see the collective mode peak in the current correlation function. It is the disordered particle distribution that hides the collective mode and leads to the envelope.

4.5.1 Particle motion under harmonic oscillation model

At high Γ value, the system can be treated as a perfect lattice approximately. Since d=0, the total system is just a single layer triangle lattice. In this single layer lattice, we assume each particle's displacement is $\vec{v}(\vec{r},t) \propto e^{i(\vec{k}_1 \cdot \vec{r} - w(\vec{k}_1)t)}$, here both the \vec{r} and \vec{k}_1 are in the global coordinates, not in the local coordinate of each sublayer. Normally, \vec{r} is time dependent, because we study lattice, then \vec{r} is independent of time. In the following text, we will write $w(\vec{k}_1)$ as w_1 for simplicity. The pair (k_1, w_1) is a point on the lattice dispersion curve. Because there are more than one modes , we need to add a coefficient $p(w_1)$, here the $p(w_1)$ is not necessarily density of states(DOS)(at least we haven't proved it so far), it is just the Fourier component of the velocity. The complete version of the displacement is

$$\vec{v}(\vec{r},t) = \vec{v}_0 \sum_{w_1} e^{i(\vec{k}_1 \cdot \vec{r} - w_1 t)} p(w_1)$$
(4.41)

Since every particle in the lattice is equivalent, if we set $\vec{r} = 0$, we can get

$$\vec{v}(\vec{r}=0,t) = \vec{v}_0 \sum_{w_1} e^{-iw_1 t} p(w_1)$$
 (4.42)

According to Z(w) = v(w)v(-w), the VAF is just $p(w_1)p(-w_1)$. Current is

$$\vec{j}(\vec{k},t) = \sum_{i=m}^{N} \vec{v}_{m}(t) e^{-i\vec{k}\cdot\vec{r}_{m}(t)}$$

$$= \vec{v}_{0} \sum_{w_{1}} \sum_{m=1}^{N} e^{i(\vec{k}_{1}\cdot\vec{r}_{m}-w_{1}t)} e^{-i\vec{k}\cdot\vec{r}_{m}} p(w_{1})$$

$$= \vec{v}_{0} \sum_{w_{1}} \sum_{m=1}^{N} e^{i((\vec{k}_{1}-\vec{k})\cdot\vec{r}_{m})} e^{-iw_{1}t} p(w_{1})$$

$$= \vec{v}_{0} \sum_{w_{1}} f(\vec{k}_{1}-\vec{k}) e^{-iw_{1}t} p(w_{1})$$
(4.43)

where

$$f(\vec{k_1} - \vec{k}) = \sum_{m=1}^{N} e^{i((\vec{k_1} - \vec{k}) \cdot \vec{r_m})}$$
(4.44)

The Eq 4.44 is just the atomic form factor. The time Fourier form of current is

$$\vec{j}(\vec{k},w) = \frac{1}{2\pi} \int \vec{j}(\vec{k},t) e^{iwt} dt$$

$$= \vec{v}_0 \sum_{w_1} f(\vec{k}_1 - \vec{k}) \delta(w - w_1) p(w_1)$$
(4.45)

Because $C_{AB}(k, w) = j^A_\mu(k, w) j^B_\mu(-k, -w)$, μ is x or y. A,B are the layer label.

$$C_{AB}(k,w) = f^{A}(\vec{k_{1}} - \vec{k})f^{B}(\vec{k_{2}} + \vec{k})(\sum_{w_{1}} \delta(w - w_{1})p(w_{1}))(\sum_{w_{2}} \delta(w + w_{2})p(w_{2})) \quad (4.46)$$

Up to now, we finished the general derivation for all the correlation functions in a lattice system. In the following sections, we will apply it to the trilayer lattice, either ordered or disordered. Eq 4.46 is the key equation. We will use it frequently later. Eq 4.46 is derived in frequency domain, recently, *Raj and Eapen* (2019) also showed a derivation in time domain.

4.5.2 1D chain lattice



Figure 4.39: 1D chain lattice, each unit cell contains just one particle



Figure 4.40: disperion for 1D chain with one particle in each unit cell, black dash line is the Brillouin zone boundary

In Fig 4.39, it is 1D chain, each unit cell contains just one particle, the interparticle distance is a. Fig 4.40 is an assumed dispersion. Since it is 1D and only has one particle in each unit cell, we should have only one dispersion curve, it is acoustic.

Now if we choose a bigger unit cell that contains three particles (see Fig 4.41). Keep in mind, we never change the chain lattice itself. If we do the lattice dispersion calculation, we must get three dispersion curves. One is acoustic, the other two are optical. It looks like the profile in Fig 4.42. The lattice period for the new unit cell is 3a, so the new Brillouin zone is smaller, just $\frac{1}{3}$ of that from one particle unit cell. Since we need to solve a 3 by 3 matrix, each curve is colored based on its eigenvector type. The acoustic will not change. But we do get two more optical curves. They don't carry new information, In Fig 4.40, we see that the period is $\frac{2\pi}{a}$, we just shift the blue curves to the left or right with $\frac{1}{3}$ of the $\frac{2\pi}{a}$, we will get the other two optical mode. The Brillouin zone for a bigger unit cell is smaller.

In Fig 4.42, if we just look at the zone between the black dash lines, we lost some acoustic information beyond the black dash line, which will be compensated by the optical curves between the black dash lines. You can just fold the acoustic line into the first Brillouin zone to get the optical modes.



Figure 4.41: 1D chain lattice, each unit cell contains three particle



Figure 4.42: disperion for 1D chain with three particle in each unit cell, black dash line is the Brillouin zone boundary

4.5.3 Current correlation function in 1D chain lattice

Suppose we did the MD simulation for such a 1D chain lattice and measure the current correlation function. What will we see?

Firstly, let us measure it as a single chain. We can only measure C_{AA} , rewrite the Eq 4.46 as

$$C_{AA}(k,w) = f^{A}(\vec{k_{1}} - \vec{k})f^{A}(\vec{k_{2}} + \vec{k})(\sum_{w_{1}} \delta(w - w_{1})p(w_{1}))(\sum_{w_{2}} \delta(w + w_{2})p(w_{2})) \quad (4.47)$$

If we are considering a single layer system. Since the particles distribute uniformly in the space we have

$$\frac{1}{N}\sum_{m=1}^{N}e^{i\vec{k}\cdot\vec{r}_{m}} = \delta(\vec{k})$$
(4.48)

For a given \vec{k} , we just chose

$$\begin{cases} \vec{k_1} - \vec{k} = 0 \\ \vec{k_2} + \vec{k} = 0 \end{cases}$$
(4.49)

then $f^A(\vec{k_1} - \vec{k})f^A(\vec{k_2} + \vec{k})$ is N^2 , it shows a very strong peak at frequency w_1 . $f^A(\vec{k_1} - \vec{k})$ or $f^A(\vec{k_2} + \vec{k})$ can be nonzero if and only if $\vec{k_1} - \vec{k} = 0$ or $\vec{k_2} + \vec{k} = 0$. If one of conditions in Eq 4.49 is not satisfied, the current correlation function will contains a Dirac function term(Eq 4.48), it is always zero. This is why we can only one peak in the current correlation function in a single layer system. Eq 4.49 indicates $k_1 = -k_2$, it is just two points when a horizontal line crosses the dispersion curve(see Fig 4.43). k_1 , k_2 both correspond to the same frequency value $w_1 = w_2$, $C_{AB}(k, w) = N^2 p(w_1) p(-w_1)$,(assume there are N particles in the chain) we will see a very strong sharp peak at w_1 , where (w_1, k) is a point on the lattice dispersion curve. This strictly indicates the dispersion curve extracted from MD current correlation function in a single layer system is exactly the same as the calculated lattice dispersion for a given \vec{k} direction.



Figure 4.43: $k_1 = -k_2$, they correspond to the same frequency (yellow dash line)

If we treat the 1D chain as an ordered trilayer system. As shown in Fig 4.41, each layer contains particles with label A or B or C. The position of the particles with each label is: $r_A = 3na$, $r_B = (3n + 1)a$, $r_C = (3n + 2)a$. Because

$$f^{A}(k_{1}-k) = \sum_{n=1}^{N} e^{i((k_{1}-k)\cdot 3na)}$$

$$f^{B}(k_{2}+k) = \sum_{n=1}^{N} e^{i((k_{2}+k)\cdot 3na)} e^{i((k_{2}+k)a)}$$

,

, in order to let f^A and f^B have nonzero value, we set $k_1 - k = 0$, due to the factor $\alpha = e^{i((k_2+k)a)}$, the choice of $k_2 + k$ matters, if $k_2 + k$ is 0, the factor α is 1, we get a positive peak. If $(k_2 + k)3a = 2\pi$, the factor α is $e^{i\frac{2\pi}{3}} = -\frac{1}{2} + \frac{\sqrt{3}}{2}i$, if we display the real part, we get a negative peak and with one half amplitude. When $(k_2 + k)3a = 4\pi$, we have $e^{i\frac{4\pi}{3}} = -\frac{1}{2} - \frac{\sqrt{3}}{2}i$. Combine $k_1 - k$ and $k_2 + k$,
the three choices correspond to the result $k_1 + k_2 = 0, \frac{2\pi}{3a}, \frac{4\pi}{3a}$. Here we predict the peak profile in current correlation function just based on the fact $r_A = 3na$, $r_B = (3n+1)a, r_C = (3n+2)a$, we don't even need the dispersion information in Fig 4.42. In the next paragraph, I will show the result $k_1 + k_2 = 0, \frac{2\pi}{3a}, \frac{4\pi}{3a}$ can be reflected in the Fig 4.42.

Recall the dispersion curve(see Fig 4.44), we should get three curves, one acoustic, anther two can be obtained by shifting or folding. The first Brillouin zone range for one particle unit cell is $\frac{2\pi}{a}$, shift the acoustic line with $\frac{1}{3}\frac{2\pi}{a}$, we get the red line(see Fig 4.44), shift the acoustic line with $-\frac{1}{3}\frac{2\pi}{a}$, we get the green line, when a yellow dash line crosses all the dispersion curves, for a given k_1 , we have three choices k_2 , k'_2 , k''_2 , all of them correspond to the same frequency. we can see the optical modes obey $k_1 + k'_2 = \frac{2\pi}{3a}$, $k_1 + k''_2 = \frac{4\pi}{3a}$, and $k_1 + k_2 = 0$ means the two points are on the same acoustic line. This matches our conclusion from last paragraph.



Figure 4.44: $k_1 + k_2 = 0$, $k_1 + k'_2 = \frac{2\pi}{3}$, $k_1 + k''_2 = \frac{4\pi}{3}$, they correspond to the same frequency (yellow dash line)

Based on the analysis above, in a trilayer chain, we should have the four properties for the current correlation function:

- 1. C_{AA} should have three positive peaks.
- 2. the real part of C_{AB} should have one positive peak(acoustic), two negative peaks(optical). The imaginary part of C_{AB} should have just two peaks(optical), one is positive(represent the red line in Fig 4.44), one is negative(represent the green line in Fig 4.44)
- 3. in the real part of C_{AB} , the amplitude of the two optical mode peaks is $-\frac{1}{2}$ of that from C_{AA}

4. in the imaginary part of C_{AB} , the absolute value of amplitude of the two optical mode peaks is $\sqrt{3}$ time of that from the real part of C_{AB}

I will show the MD data to confirm the four properties later.

4.5.3.1 Connection between 1D trilayer and 2D trilayer

So far, we are done for 1D chain. For our 2D trilayer system, because we study L or T, we always just consider the projection of one direction, once you do the projection, it is reduced to the 1D chain analysis above.

In 1D case, we only have one k direction, but in our 2D hexagonal lattice trilayer, we have 4 principle directions, we can only observed folding effect for $\phi = 30^{\circ}, 90^{\circ}$ (see Fig 3.13). Since we used the fact that spatial position can be expressed in such way that $r_A = 3na$, $r_B = (3n+1)a$, $r_C = (3n+2)a$, if you look at the particles along 0° (orthogonal to 90°) and 120° (orthogonal to 30°) direction (red and black lines in Fig 4.45), each line covers only one type of particles, it means we can project all the particles into a 1D chain in Fig 4.41. But for the other two principle directions, this is not satisfied. The formation of purely gaped modes along 0° and 60° is a problem to be solved in the future.



Figure 4.45: Top view of a trilayer lattice, along the black and red lines, you can only see one type of particles

4.5.3.2 Conformation from the MD result

Here we show the two MD result figures to confirm the peak amplitude properties we have predicted.



Figure 4.46: peak comparison between T_{11} and T_{12} from a trilayer staggered triangle lattice, d=0, $\Gamma = 1000$, both are real part. ka = 0.54912. Left: T_{11} ; Right: T_{12}



Figure 4.47: peak comparison between real part and imaginary part of T_{12} from a trilayer staggered triangle lattice, d=0, $\Gamma = 1000$, ka = 0.54912. Left: real; Right: imaginary

In Fig 4.46, in the left panel, the amplitude of the second and third peak is about $1.14e^7$ and $1.36e^7$, in the right panel, the absolute value of the amplitude of second and third peak is about $0.57e^7$ and $0.66e^7$, $\frac{0.57}{1.14} = 0.5$, $\frac{0.66}{1.36} = 0.485$, this is very close to theoretical ratio $\frac{1}{2}$.

In Fig 4.47, in the left panel, the amplitude of the second and third peak is about $0.57e^7$ and $0.66e^7$, in the right panel, the absolute value of the amplitude of second and third peak is about $0.985e^7$ and $1.15e^7$, $\frac{0.985}{0.57} = 1.728$, $\frac{1.15}{0.66} = 1.742$, this is very close to theoretical ratio $\sqrt{3} = 1.732$. In the right panel, the acoustic peak is invisible, which also matches our prediction.

In the second property, we predicted the red optical line shows a positive peak in imaginary part of C_{AB} , the green optical line shows a negative peak in imaginary part of C_{AB} , now if you compare Fig 4.38 and Fig 4.44, you can understand why in the right panel of Fig 4.47, the lower frequency peak is positive and the higher frequency peak is negative.

From Fig 4.46 and 4.47 we can see the T current correlations accurately match the four properties we predicted.

4.5.4 Disordered trilayer lattice

In last sectioon, I have explained the peak formation for an ordered trilayer lattice.

If we are considering a disordered trilayer system, for the same reason, if Eq 4.49 is satisfied, we will get a strong positive peak, which is the acoustic one, no matter in C^{AA} or C^{AB} . This can be seen from Fig 4.10.

But the Eq 4.48 should be modified.

$$\sum_{m=1}^{N/3} e^{i\vec{k}\cdot\vec{r}_m^A} + \sum_{m=1}^{N/3} e^{i\vec{k}\cdot\vec{r}_m^B} + \sum_{m=1}^{N/3} e^{i\vec{k}\cdot\vec{r}_m^C} = N\delta(\vec{k})$$
(4.50)

This means that even though the condition in Eq 4.49 is satisfied, $f^A(\vec{k}_1 - \vec{k})$ or $f^B(\vec{k}_2 + \vec{k})$ is not $\delta(\vec{k})$ any more. If k is nonzero,

$$f^{A}(\vec{k}) + f^{B}(\vec{k}) + f^{C}(\vec{k}) = 0$$
(4.51)

the three parts in complex plane are just three equal length vectors with 120° angle. For a given k, if w doesn't match the lattice dispersion with k, then

$$C_{AB}(k,w) = f^A(\vec{k}_1 - \vec{k})f^B(-\vec{k}_1 + \vec{k})p(w)p(-w)$$
(4.52)

where $(\vec{k_1}, w)$ is a point on the lattice dispersion curve. If we set $f^A = e^{i\theta}$, then $f^B = e^{i(\theta + \frac{2}{3}\pi)}$ or $f^B = e^{i(\theta - \frac{2}{3})\pi}$ with equal weight. Then $|f^A|^2 = 1$, $Re(f^A(f^B)^*) = \frac{1}{2}Re(e^{i\frac{2}{3}\pi} + e^{-i\frac{2}{3}\pi}) = -\frac{1}{2}$, this explains the ratio of the envelope's amplitude. For the imaginary part, $Im(f^A(f^B)^*) = \frac{1}{2}Im(e^{i\frac{2}{3}\pi} + e^{-i\frac{2}{3}\pi}) = 0$.

Fig 4.48 and Fig 4.49 are the L_{12} plot from the the disordered trilayer lattice. The left panel is the real part of L_{12} , the right panel is the imaginary part of L_{12} . The two panels are in the same coordinate scale. From Fig 4.48, we can see the real part of L_{12} contains positive acoustic peak and negative envelope, while the imaginary part is negligible, just some noise. The Fig 4.49 is a zoomed picture.

Fig 4.50 is the comparison between the real part of L_{11} and L_{12} , they do have the same acoustic peak, but L_{11} contains positive envelope, L_{12} contains negative envelope, the amplitude ratio is nearly one half.



Figure 4.48: comparison of the real part(left) and imaginary part(right) of L_{12} , full profile picture, the simulation data is from disordered trilayer system. $\Gamma = 1000$, d=0. In the legend box, it is the ka value for each line.



Figure 4.49: The same as Fig 4.48 but a zoomed in profile picture.



Figure 4.50: comparison of the real part of $L_{11}(\text{left})$ and $L_{12}(\text{right})$, full profile picture. the simulation data is from disordered trilayer system. $\Gamma = 1000$, d=0. In the legend box, it is the ka value for each line.

Since Eq 4.51 is independent of k, this is why the envelope profile is independent of k. Up to now, at high Γ limit, we proved that the envelope is exactly the VAF. Fig 4.51 can perfectly confirm this result(Both two panels have been normalized).



Figure 4.51: the comparison between envelope (left) and VAF(right). The envelope is obtained from $T_{11} - T_{12}$. In the legend box, it is the ka value of each line.

4.6 N dependence of the envelope

We have done the MD simulation with different numbers of particles in the system to check the N dependence of the envelope's amplitude.



Figure 4.52: N dependence of the envelope's amplitude for trilayer triangular lattice, d=0, $\Gamma = 1000$



Figure 4.53: N dependence of the envelope's amplitude for trilayer triangular lattice, d=0, $\Gamma = 1000$

Fig 4.52 and Fig 4.53 clearly show that the envelope's amplitude is independent of N. Here we try to give a speculative explanation for it. Since we already know that the envelope is VAF and it is independent of k, we only analysis this problem at k = 0.

For a trilayer system with small d, each layer contains N particles. When k is zero, the

$$\vec{j}(k=0,t) = \sum_i^{3N} \vec{v}_i(t)$$

, it is simply the summation of the all the particles' velocity. Since the momentum is conserved, the total velocity summation should be constant. But the velocity summation from each layer has some fluctuation. Assume the momentum fluctuation is $\vec{\xi}^{A,B,C}(t)$, their summation must be zero,

$$\vec{\xi}^{A}(t) + \vec{\xi}^{B}(t) + \vec{\xi}^{C}(t) = 0$$
(4.53)

at a certain time, the length of the three vectors are not necessarily the same, but the average in a long time range must be the same, the angle between two vectors must be 120°. Since the system is disordered, for simplicity, we can only consider x component. We already know that $|\xi_x^A(w)|^2$ is the envelope. In order to study the N dependence of the envelope. We just need to study the N dependence of $\xi_x^A(t)$.

In the system, all the particles are equivalent. If we consider a single particle #i, we record time series data of velocity: $v_i(t_1)$, $v_i(t_2)$, $v_i(t_3)$,..., $v_i(t_n)$, the velocity has a certain distribution. Its average must be zero, set its variance as σ^2 . Since

$$\xi^A(t) = \sum_i^N v_i(t)$$

, the average of $\xi^A(t)$ is still zero, but its variance is $N\sigma^2$. Based on $\operatorname{Var}(X) = \overline{X^2} - \overline{X}^2 = \overline{X^2}$, this indicates the $\overline{\xi^A(t)^2} = N\sigma^2$ and $|\xi^A_x(w)|^2 \propto N$. Since we calculate current correlation function with $L(w,k) = \frac{1}{N}j(w,k)j(-w,-k)$ in the simulation, when k=0, the envelope must be independent of N, this is a general

conclusion regardless of the Γ value. $L(w, k = 0) \propto \sigma^2 \propto k_B T$, the envelope is due to the momentum fluctuation, which is related to the temperature. This has been confirmed by Fig 4.49, in which we can see the amplitude of the envelope is approximately in the same order of the noise. As we have proved in the previous section, the acoustic peak amplitude is proportional to N at crystal limit(high Γ).

4.7 Why the envelope equals the VAF?

In previous section, I have proved the envelope is the VAF at crystal limit. In fact, it is very easy to show this is always true regardless of Γ .

One particle's velocity can be written as

$$v(t) = \sum_{w} e^{-iwt} p(w) \tag{4.54}$$

The VAF is just Z(w) = p(w)p(-w). Since every particle is equivalent, the Fourier spectrum of every particle is the same. But obviously the velocity of each particle at a given time is not the same. This is because v(t) has a different phase θ for each particle,

$$v_n(t) = \sum_w e^{-iwt + \theta_n} p(w)$$

$$\xi^A(t) = \sum_n^N v_n(t)$$

should also proportional to

,

$$\sum_w e^{-iwt} p(w)$$

, this is why when k=0, the envelope is just the VAF.

4.8 More about VAF

From these VAF profile at different Γ , we can see a peak at about $w = 0.8w_p$ for all Γ , there is a lower frequency peak only for high Γ . The peaks in the VAF mean higher density of states. Generally, density of states is proportional to $\frac{dk}{dw}$, which means a stationary point on the dispersion corresponds to some peak in the VAF. Previous works *Hansen et al.* (1975); *Schmidt et al.* (1997) thought higher frequency peaks is from longitudinal mode, the lower frequency peak is from the transverse mode. But based on our new result, this conclusion might be wrong. It seems that the two peaks correspond to the two stationary points only on the longitudinal mode. Look at the first column in Fig 4.14, we can see the two stationary points in AL mode are at about $w = 0.8w_p$ and $w = 0.3w_p$, this matches the profile in Fig 4.26(the third subplot). On the other hand, the stationary point in AT mode is at about $w = 0.6w_p$, which can't be found in Fig 4.26.



Figure 4.54: the VAF for single layer lattice at $\Gamma = 1000$, the Fourier transformation is done without any window function.

From Fig 4.54 we can see there are two spikes on the right peak. One is $0.8w_p$, the other one is $0.88w_p$, the precisely matches the stationary points in the single layer triangle lattice dispersion(see Fig 3.14). We can also clearly see the peak amplitude is linear to the frequency. Its mechanism needs more research work in the future.

CHAPTER V

Summary

In this thesis, I fully studied the quasi-2D strongly coupled Yukawa system. The density profile under trap is studied by density functional method, with both MF and HNC approximations. A trilayer Yukawa lattice is studied to determine its stable lattice structure and its phonon dispersion. Then the MD result at $\Gamma = 1000$ perfectly matches the calculated lattice dispersion. For a trilayer liquid system, its collective mode is studied by the QLCA method, and then compared with the MD result. We get a very good agreement when interlayer distance is large. At short interlayer distance, the previous gap discrepancy is clarified. We found there is no optical collective mode in the current correlation function, we can only see a VAF profile. We gave a brief speculative explanation on the formation of the envelope and its connection to the VAF. So far, only the theoretical calculation and simulation have been done. We hope some experiment can be done for the trilayer system in the near future.

Future work direction:

- 1. Explore the peak formation of the VAF profile, we hope to understand whether it is from L mode or both L and T modes
- Try to give strict proof to the linear relationship of the peak amplitude of VAF peaks. Further more, we also can explore it in 3D system.

- 3. At intermediate d value(0.5 < d < 1.5), how does the current correlation function profile transform from the envelope type to the collective type?
- 4. In the trilayer liquid system with $\Gamma = 160$, d=1.5, the current correlation function has some extra peaks that can be found in the lattice current correlation function, We want to know how are they related quantitatively.
- 5. In the trilayer lattice(d=0) dispersion curves, we see the folding effect along some direction and purely optical modes along some other direction, we want to understand the physical meaning of those purely optical modes. Since the system is the same as a 2D layer, we hope to generate the optical mode just by the two acoustic modes in the 2D layer.

APPENDICES

APPENDIX A

The mathematical proof for the monotonic density profile in mean field theory

According to the screened Poisson equation, we have

$$[\Delta - \kappa^2]\varphi(\mathbf{r}) = -\frac{\rho(\mathbf{r})}{\epsilon_0}q \qquad (A.1)$$

where the φ is the total Yukuwa potential at a given position in sapce, q is the charge. Since the system is cylindrical symmetric. $\Delta = \frac{\partial^2}{\partial z^2}$. The Total potential is

$$U(z) = wz^2 + \varphi(z)q \tag{A.2}$$

Then density profile $\rho(z) \propto e^{-\beta U(z)}$, for simplicity, we ignore some coefficients and write down

$$\rho(z) = e^{-(wz^2 + \varphi(z))} \tag{A.3}$$



Figure A.1: density profile with some stationary point

Assume there a density profile like that in Fig A.1, there is local maximum point at z_0 , at z = 0, it is a local minimum point, and $\rho(z_0) > \rho(0)$.

$$\rho'(z) = -e^{-(wz^2 + \varphi(z))} [\varphi'(z) + 2wz]$$
(A.4a)

$$\rho''(z) = -e^{-(wz^2 + \varphi(z))} \{ [\varphi'(z) + 2wz]^2 - \varphi''(z) - 2w \}$$
(A.4b)

Based on the math property of a stationary point, we have $\rho'(z_0) = 0$ and $\rho''(z_0) < 0$, it leads to $\varphi''(z_0) + 2w > 0$, because $\left[\frac{\partial^2}{\partial z^2} - \kappa^2\right]\varphi(z) \propto -\rho(z)$, we finally have

$$\rho(z_0) < 2w + \kappa^2 \varphi(z_0) \tag{A.5}$$

similarly, at z = 0, we have $\rho'(0) = 0$ and $\rho''(0) > 0$, we will get

$$\rho(0) > 2w + \kappa^2 \varphi(0) \tag{A.6}$$

Because we have assumed $\rho(z_0) > \rho(0)$, then $U(z_0) < U(0)$, which means $wz_0^2 + \varphi(z_0) < \varphi(0)$. We will get

$$\rho(0) > 2w + \kappa^2 \varphi(0) > 2w + \kappa^2 \varphi(z_0) > \rho(z_0)$$
(A.7)

This conflicts to our assumption $\rho(z_0) > \rho(0)$. We can also assume there is a local maximum at z = 0 and there is a local minimum at z_0 . We will get the similar conflict, here we skip the detail. It means the density profile can't have a stationary point, the density profile in mean field theory is always monotonic.

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