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Final Report

Melting Transition in the Two-Dimensional Gaussian Core Model

Introduction

Many important phenomenas of two dimensional colloidal crystals like the freezing transition [1, 2], the interaction between dislocations [3], colloidal gels [4], the glass transition [5] and the coexistence between liquid and solid phases [6] were studied both, in computer simulations and experiments [7] in recent years. Still, one aspect of two dimensional colloidal crystals (and two dimensional crystals in general) that is still not fully understood is the melting transition on an atomistic level [8].

Purpose of the visit

The purpose of my visit was to study the melting transition in a two dimensional colloidal crystals with Monte Carlo simulations. The basis of my work was a theoretical description of this melting process elaborated by Kosterlitz, Thouless, Halperin, Nelson, and Young [12]. Their theory suggests that the melting transition in two dimensions is driven by the unbinding of dislocations. The theory also predict an additional intermediate phase, namely the hexatic phase. I studied the KTHNY scenario in the Gaussian core model. This model consists of purely repulsive particles with a pair interaction of Gaussian shape [9, 10]. Here, it serves as a general model for soft colloidal particles. It is a realistic model for the interaction between polymers in a solution [11]. The Gaussian Core model exhibits an interesting feature called reentrant melting. At low densities and low temperature the system is in the fluid phases. After compressing the system it becomes solid, but after further compression the system becomes fluid again. My goal was to study both, the low and high density melting in terms of KTHNY theory and to elaborate possible differences between the two scenarios.

Description of the work carried out during the visit

An early study of the melting transition of the Gaussian Core model in two dimensions was done in 1981 by Stillinger and Weber [13]. In this work they

studied the pressure as a function of the temperature at a given density. As expected for a first order phase transition they observed a sharp kink at coexistence and concluded that there is no indication for a hexatic phase or any other regime than a first order phase transition. I did similar calculations at various densities. I was able to reproduce the results from [13], but also found that the kink broadens at higher densities and hysteresis can be observed.

For further studies of this transition I followed the path of Frenkel and coworkers [14].They calculated the phase diagram for a Lennard Jones system from the free energy and then identified the hexatic phase with the Lamé coefficients λ and μ. In order to calculate the phase diagram in the Gaussian Core model I had to compare the Helmholtz free energies of the liquid and the solid phases at given temperatures and densities [15]. The same technique was used by Prestipino and coworkers [9, 16] in order to study the phase behaviour of the Gaussian Core model in three dimensions. For the liquid phase thermodynamic integration allows one to calculate the free energy difference between two given states. The free energy of a reference state was calculated using the Widom insertion method [15] which uses the ideal gas as a reference. In order to calculate the free energy of the solid phase one cannot use the ideal gas as a reference because there is no reversible path from the liquid to the solid. For the solid phase I employed the Frenkel-Ladd method [15] with a fixed center of mass and the Einstein crystal as a reference.

Main results

I was able to draw a precise phase diagram of the 2d Gaussian Core model from the comparison of the free energies of the undercooled liquid and the overheated solid. This is the main result from the simulations I did during my visit. Still, further investigations are necessary to finally identify the hexatic phase and to conclude whether the KTHNY-scenario is applicable for the Gaussian Core model.

Concluding remarks and further collaborations

During my stay at Columbia University I implemented the thermodynamic integration simulation, the Widom insertion and the Frenkel-Ladd simulation. All of the above would have not been such a success without the great support of Prof. Reichman. Aside from the productive time at Columbia University, I established many links to scientists at Columbia University for further collaborations.

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