A numerical renormalization group study of laser induced freezing

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Abstract. – We study the phenomenon of laser induced freezing, within a numerical renormalization scheme which allows explicit comparison with a recent defect mediated melting theory. Precise values for the 'bare' dislocation fugacities and elastic moduli of the 2-d hard disk system are obtained from a constrained Monte Carlo simulation sampling only configurations *without* dislocations. These are used as inputs to appropriate renormalization flow equations to obtain the equilibrium phase diagram which shows excellent agreement with earlier simulation results. We show that the flow equations need to be correct at least up to third order in defect fugacity to reproduce meaningful results.

Introduction. – Re-entrant "laser-induced" freezing [1, 2] (RLIF) has received considerable attention [3–13] in recent times. A static interference pattern obtained by two crossed laser beams provides an external potential periodic in one dimension (1-d) which induces a system of dielectric colloidal particles, confined in two dimensions (2-d), to freeze. Surprisingly, a further increase in potential strength causes a *reentrant melting* [2] transition. Qualitatively, starting from a liquid phase, the external periodic potential immediately induces a density modulation, reducing fluctuations which leads to solidification. Further increase in potential confines the system to decoupled 1-d strips. The dimensional reduction now *increases* fluctuations remelting the system. The early mean field theories, namely, Landau theory [1] and density functional theory [3] predicted a change from a first order to continuous transition with increase in potential strength and failed to describe the reentrant behavior, a conclusion seemingly confirmed by early simulations [6]. Quite generally however, in 2-d, the effect of thermal fluctuations is substantial and mean field theory may obtain qualitatively wrong results especially for fluctuation driven transitions. Following the defect mediated disordering approach of Kosterlitz and Thouless [14](KT), Frey, Nelson and Radzihovsky [5](FNR)

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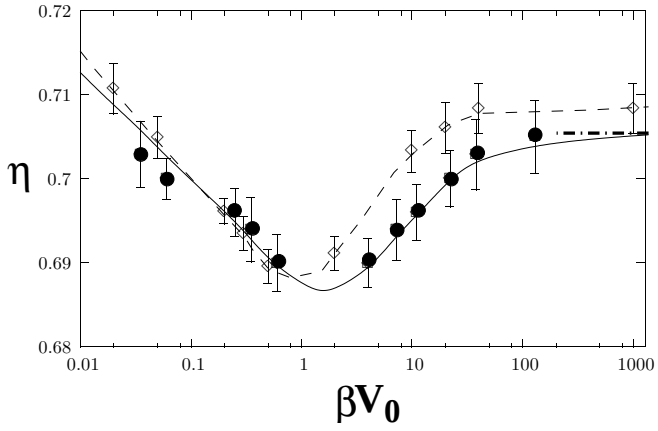

Fig. 1 – The phase diagram of the hard disk system in the presence of a 1-d, commensurate, periodic potential in the packing fraction (η) - potential strength (βV_0) plane. The lines in the figure are a guide to the eye. The dashed line denotes earlier Monte Carlo simulation results[7] and the solid line is calculated through our numerical renormalization group study. The dash-dotted line at $\eta \simeq .705$ denotes the calculated asymptotic phase transition point at $\beta V_0 = \infty$.

proposed a detailed theory for the reentrant transition based on the unbinding of dislocations with Burger's vector parallel to the line of potential minima. More accurate simulation studies [7–13] on systems of hard disks [7], soft disks [8], DLVO [9, 10] etc. confirmed the reentrant freezing-melting transition in agreement with experiments [2] and FNR theory. A systematic finite size scaling analysis [7] of simulation results for the 2-d hard disk system in a 1-d modulating potential showed, in fact, several universal features consistent with the predictions of FNR theory. Non universal predictions, namely the phase diagram, on the other hand, are difficult to compare because the FNR approach (like KT theory) is expressed in terms of the appropriate elastic moduli which are notoriously time-consuming to compute near a continuous phase transition. Diverging correlation lengths and times near the phase transition point further complicate an accurate evaluation of the non universal predictions of the theory.

In this Letter, we calculate the phase diagram of a 2-d hard disk system with a modulating potential (see Fig. 1) following a Monte Carlo renormalization approach proposed recently [15] for the XY-model. The twin problems of diverging length and time scales are eliminated by simulating a constrained system which *does not* undergo a phase transition! This is achieved by rejecting all Monte Carlo moves which tend to distort an unit cell in a way which changes the local connectivity [16]. The percentage of moves thus rejected is a measure of the dislocation fugacity [16]. This, together with the elastic constants of the dislocation free lattice obtained separately, are our inputs (bare values) to the renormalization flow equations [5] to find out the melting points and hence the phase diagram. Our results (Fig. 1) clearly show a modulated liquid (ML) $\rightarrow$ locked floating solid (LFS) $\rightarrow$ ML re-entrant transition with increase in the amplitude (V_0) of the potential. In general, we find, the predictions of FNR theory to be valid. The location of the phase transition as evaluated within this theory with our inputs, show *excellent* agreement with earlier simulations [7] throughout the $\eta - \beta V_0$ plane ($\beta = 1/k_B T$ with k_B = Boltzmann constant and T = temperature, η = packing fraction). We discuss our calculations in detail below.

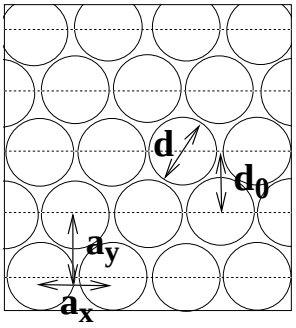

Fig. 2 – This cartoon shows a typical hard disk system. The dashed lines indicate minima of external modulating potential $\beta V(y) = \beta V_0 \cos(2\pi y/d_0)$. a_x is the lattice parameter and a_y indicate the average separation between two layers along y-direction perpendicular to a set of close-packed planes. For a perfect triangular lattice $a_y = \sqrt{3}a_x/2$. The modulating potential is commensurate with the lattice such that $d_0 = a_y$.

The bulk system of hard disks where particles i and j, in 2-d, interact via the potential $V_{ij} = 0$ for $|\mathbf{r}_{ij}| > \mathrm{d}$ and $V_{ij} = \infty$ for $|\mathbf{r}_{ij}| \leq \mathrm{d}$, where d is the hard disk diameter and $\mathbf{r}_{ij} = \mathbf{r}_j - \mathbf{r}_i$ the relative position vector of the particles, is known to melt [16, 18, 19] from a high density triangular lattice to an isotropic liquid with a narrow intervening hexatic phase [16, 17, 19]. Simulations [19], experimental [20] and theoretical [21] studies of hard disks show that for $\eta > .715$ the system exists as a triangular lattice which transforms to a liquid below $\eta = .706$. The small intervening region contains a hexatic phase predicted by the KTHNY theory [17] of 2-d melting. Apart from being easily accessible to theoretical treatment [22], experimental systems with nearly "hard" interactions viz. sterically stabilized colloids [20] are available. The hard disk free energy is entirely entropic in origin and the only thermodynamically relevant variable is the number density $\rho = N/V$ or the packing fraction $\eta = (\pi/4)\rho \mathrm{d}^2$.

Theory. – In presence of a periodic external potential, the only other energy scale present in the system is the relative potential [23] strength βV_0. A cartoon of the system considered for our study is given in Fig. 2. For a solid in presence of a modulating potential $\beta V(y)$ (Fig. 2) displacement mode u_y becomes massive, leaving massless u_x modes. After integrating out the u_y modes the free energy of the LFS may be expressed in terms of u_x and βV_0 dependent elastic moduli [5], namely, the Young's modulus K and shear modulus μ,

$$\mathcal{H}_{el} = \int dxdy \left[K \left(\frac{\partial u_x}{\partial x} \right)^2 + \mu \left(\frac{\partial u_x}{\partial y} \right)^2 \right] \tag{1}$$

Similar arguments [5] show that among the three sets of low energy dislocations available in the 2-d triangular lattice, only those (type I) with Burger's vector parallel to the line of potential minima survive at large βV_0. Dislocations with Burger's vector pointing along the other two possible close-packed directions (type II) in the 2-d triangular lattice have larger energies because surrounding atoms are forced to ride the crests of the periodic potential [5]. Within this set of assumptions, the system therefore shares the same symmetries as the XY model. Indeed, a simple rescaling of $x \to \sqrt{\mu}x$ and $y \to \sqrt{K}y$ leads this free energy to

the free energy of the XY-model with spin-wave stiffness $K_{xy} = \sqrt{K\mu}a^2/4\pi^2$ and spin angle $\theta = 2\pi u_x/a_x$. The corresponding theory for phase transitions can then be recast as a KT theory [14] and can be described in the framework of a two parameter renormalization flow for the spin-wave stiffness $K(l)$ and the fugacity of type I dislocations $y'(l)$, where l is a measure of length scale as $l = \ln(r/a_x)$, r being the size of the system. The flow equations can be expressed in terms of $x' = (\pi K_{xy} - 2)$ and $y' = 4\pi\ exp(-\beta E_c)$ where E_c is the core energy of type I dislocations which can be obtained from the dislocation probability [16]. Keeping upto next to leading order terms in y' the renormalization group flow equations [15, 24] are,

$$\begin{aligned} \frac{dx'}{dl} &= -y'^2 - y'^2 x' \\ \frac{dy'}{dl} &= -x'y' + \frac{5}{4}y'^3. \end{aligned} \tag{2}$$

Flows in l generated by the above equations starting from initial or "bare" values of x' and y' fall in two categories. If, as $l \to \infty$, y' diverges, the thermodynamic phase is disordered (i.e. ML), while on the other hand if y' vanishes, it is an ordered phase (a LFS) [5]. The two kinds of flows are demarcated by the *separatrix* which marks the phase transition point. For the linearized equations the separatrix is simply the straight line $y' = x'$, whereas for the full non-linear equations one needs to calculate this numerically.

Simulation results and Discussion. – We obtain the bare y' and x' from Monte Carlo simulations of $43 \times 50 = 2150$ hard disks and use them as initial values for numerically solving the Eqs. (2). The bare numbers are relatively insensitive to system size since our Monte Carlo simulation does not involve a diverging correlation length. This is achieved as follows [15,16]. We monitor individual random moves of a hard disk (after checking for overlaps with neighbors) and look for distortions of the neighboring unit cells. If in any of these unit cells the length of a next nearest neighbor bond tends to become smaller than a nearest neighbor bond, the move is rejected. The probabilities of such bond breaking moves are however computed by keeping track of the number of such rejected moves. One has to keep track of three possible distortions of the unit rhombus (see Fig. 3 inset) with measured probabilities $P_{mi}, i = 1, 3$. Each of these distortions involves two dislocation-antidislocation pairs which, we assume, occur independently. The probabilities for occurrence of the dislocation pairs themselves P_{di} (Fig. 3) which are proportional to the square of the fugacities, can then be computed easily eg. $P_{d1} = (P_{m2} + P_{m3} - P_{m1})$. Finally, we obtain the required dislocation fugacity y' by normalizing $(P_{di})^{1/2}$ to the known [16] dislocation fugacities for $\beta V_0 = 0$ at any η. The same restricted Monte Carlo simulation can be used to find out the stress tensor, and the elastic moduli from the stress-strain curve, following the method described by Farago *et. al.* [25]. The errors in 'bare' elastic moduli are at worst within a percent.

In Fig. 4 we have plotted the bare values of x' and y' for $\eta = .7029$ along with the separatrices for the linearized and the non-linear flow equations (Eq. 2). The line of initial conditions is seen to cross the non-linear separatrix twice (signifying re-entrant behaviour) while crossing the corresponding linearized separatrix only once at high potential strengths. The phase diagram (Fig. 1) is obtained by computing the points at which the line of initial conditions cut the non-linear separatrix using a simple interpolation scheme. It is interesting to note that within a linear theory the KT flow equations fail to predict a RLIF transition.

Further, comparing with previous computations [7] of the phase diagram for this system (also shown in Fig. 1) we find that our results agree at all values of η and βV_0. This validates both our method and the quantitative predictions of Ref. [5]. The effect of higher order terms

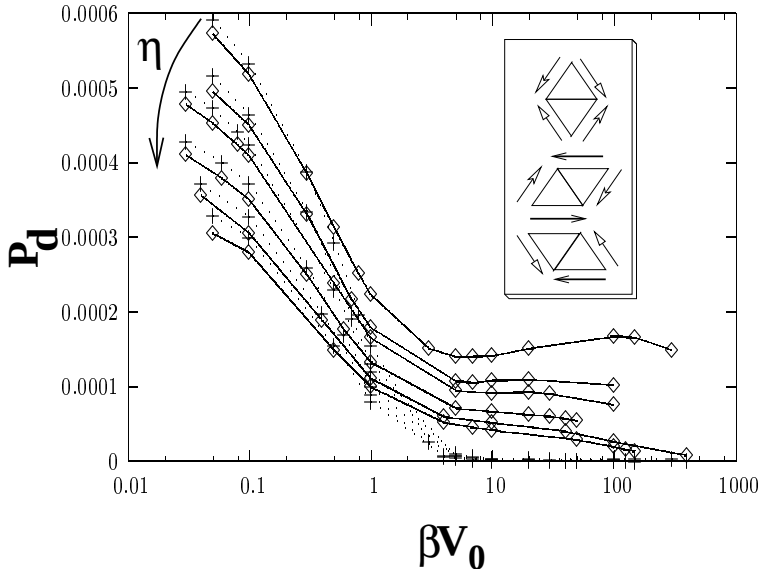

Fig. 3 – Inset shows the unit cell and three possible bond-breaking moves for a triangular solid in the orientation shown in Fig. 2. Arrows show the Burger's vectors for the associated dislocations. Dislocations with Burger's vector parallel to the horizontal direction are type I dislocations. The probabilities for these moves are P_{m1}(top), P_{m2} and P_{m3}. In the main plot the $\diamond$ symbols correspond to P_{d1}, the probability for type I dislocations and the $+$ symbols to $(P_{d2} + P_{d3})/2$ the probability for type II dislocations obtained from the P_{mi} (see text) for various η values, arrow denoting the direction of increasing $\eta(= .69, .69395, .696, .7, .7029, .705)$. These probabilities are plotted against the potential strength βV_0. Note that for $\beta V_0 > 1$, the probability for type I dislocations is larger than that of type II.

in determining non-universal quantities has been pointed out earlier [15] for the planar rotor model but in the present case their inclusion appears to be crucial. In view of the fact that the linear flow equation predicts a solid phase at all but the largest values of βV_0 we require at least upto next to leading order corrections in flow to obtain meaningful results. Even then, we expect our procedure to break down at high packing fractions in the $\beta V_0 \to 0$ limit where effects due to the cross-over from a KT to a KTHNY [17] transition at $\beta V_0 = 0$ become significant. Indeed, as is evident from Fig. 3 for $\beta V_0 < 1$ the dislocation probabilities of both type I and type II dislocations are similar and our process (which involves only type I dislocations) fails to produce melting as $\beta V_0 \to 0$ for $\eta \geq .705$ — the solid line in Fig. 1 being an extrapolation from our results for smaller η. This fact is also supported by Ref. [7] where it was shown that though at $\beta V_0 = 1000$ the scaling of susceptibility and order parameter cumulants gave good data collapse with values of critical exponents close to FNR predictions, at $\beta V_0 = .5$, on the other hand, ordinary critical scaling gave better data collapse than the KT scaling form, perhaps due to the above mentioned crossover effects. In the asymptotic limit of $\beta V_0 \to \infty$ the system freezes above $\eta \sim .705$ which was determined from a separate simulation in that limit. This number is very close to the earlier value $\eta \sim .71$ quoted in Ref. [7]. As expected, the freezing density in the $\beta V_0 \to \infty$ limit is lower than the value without the periodic potential.

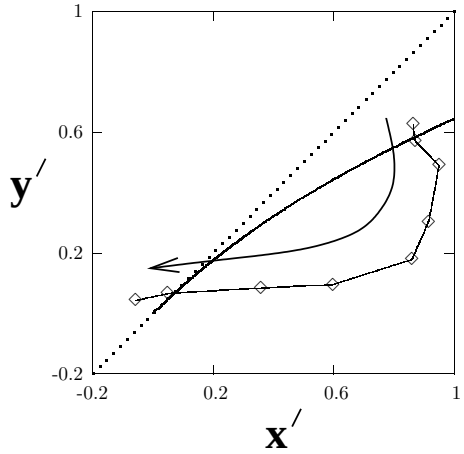

Fig. 4 – The initial values of x' and y' obtained from the elastic moduli and dislocation probability at $\eta = .7029$ plotted in $x' - y'$ plane. The line connecting the points is a guide to eye. The arrow shows the direction of increase in $\beta V_0 (= .01, .04, .1, .4, 1, 4, 10, 40, 100)$. The dotted line denotes the seperatrix ($y' = x'$) incorporating upto the leading order term in KT flow equations. The solid curve is the seperatrix when next to leading order terms are included. In $l \to \infty$ limit any initial value below the seperatrix flows to $y' = 0$ line whereas that above the seperatrix flows to $y' \to \infty$. The intersection points of the line of initial values with the seperatrix gives the phase transition points. The plot shows a freezing transition at $\beta V_0 = .035$ followed by a melting at $\beta V_0 = 38$.

Conclusion. – In conclusion, using FNR theory we calculate the phase diagram for a 2-d system of hard disks under a commensurate modulating potential to find extremely good agreement with earlier simulated results. We show that the re-entrance behavior is built into the 'bare' quantities themselves. To obtain the correct phase diagram, however, flow equations need to be correct at least upto next to leading order terms in the dislocation fugacity. Our results, especially for small potential strengths, is particularly sensitive to these terms. Cross-over effects from zero potential KTHNY melting transition are also substantial at small values of the potential.

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