Spatio-temporal correlations across the melting of 2D Wigner molecules

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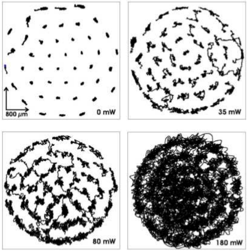
Theme:

- **Coulomb interacting particles in two dimensional confinements**
- **Static & Dynamic responses across 'melting'.**



Objective:

- **Explore spatio-temporal correlations when system size $\sim$ range of interaction**
- **Effects of 'disorder' (irregularity)**

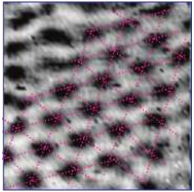
Biswarup Ash

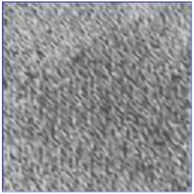
CQDISCOR2017, ICTS, 2^nd June, 2017

Crystal of Coulomb particles and its melting:

Wigner Crystal Melting
(1934)

Competetion between
PE & KE





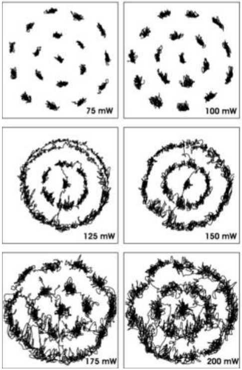
If the electrons had no kinetic energy, they would settle in configurations which correspond to the absolute minima of the potential energy. These are close-packed lattice configurations, with energies very near to that of the body-centered lattice....“ **(in 3D)**

Coulomb repulsion forces particles to stay as far as possible from each other, localizing them in a crystal. Kinetic Energy delocalizes them.

- KE $\sim k_B T$ (equipartition) $\Rightarrow$ Thermal / Classical melting
- KE $\sim$ Quantum (zero-point) fluctuations $\Rightarrow$ Quantum melting.

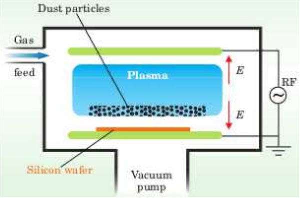
Experiments: Wigner Physics in confinements

- Realizing Wigner melting in bulk ?? Goldman *et al.* PRL (90); Yoon *et al.* PRL (99); Chen *et al.* Nat Phys.(06)
- **"Wigner molecules"** are promising !!

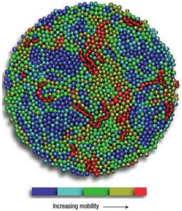


Dusty plasma
A. Melzer Group (2012)

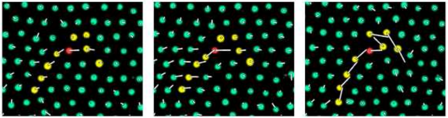
Dusty plasma setup



Dynamics across melting



air driven steel beads
(Keys *et. al.*; Nat.Phys. '07)

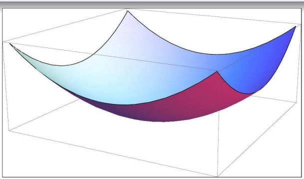


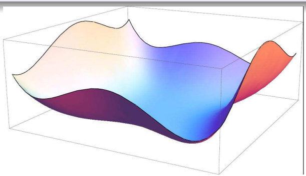
Relaxation of localized stress in 2*D* Colloid; (B. Meer, *et.al.*, PNAS'14)

Model, Method & Parameters :

Objective

- **Static & Dynamics for system size $\sim$ interaction range**
- **Effects of *inherent* 'disorder' (irregularity) on observables**





Hamiltonian for the model system

$$\mathcal{H} = \frac{q^2}{4\pi\epsilon}\sum_{i<j}^{N}\frac{1}{|\vec{r}_i - \vec{r}_j|} + \sum_{i}^{N} V_{\rm conf}(r_i); \quad r = |\vec{r}| = \sqrt{x^2+y^2}$$

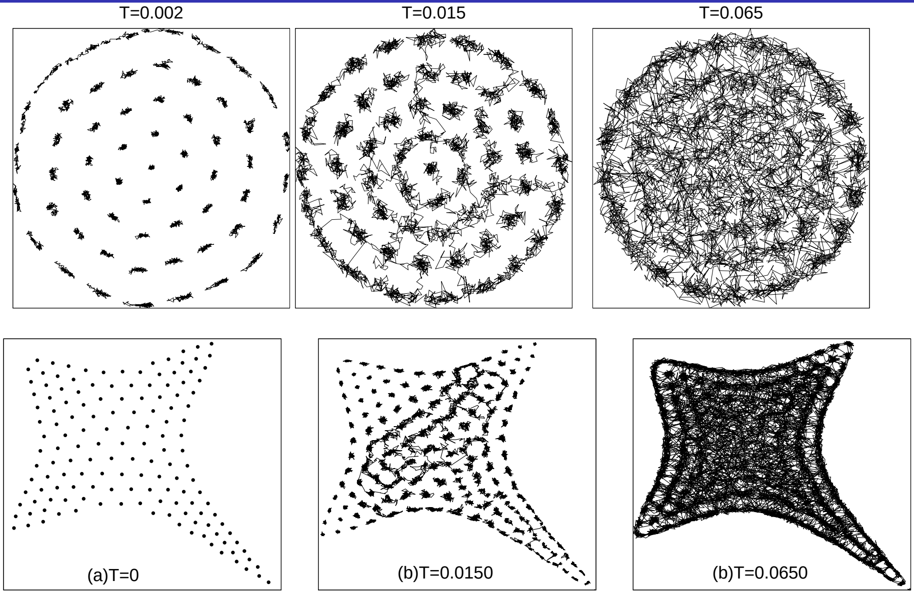
(a) Irregular: $V_{\rm conf}^{\rm Ir}(r) = a\{x^4/b + by^4 - 2\lambda x^2y^2 + \gamma(x-y)xyr\}$,

(b) Circular: $V_{\rm conf}^{\rm Cr}(r) = \alpha r^2$, with $\alpha = m\omega^2/2$.

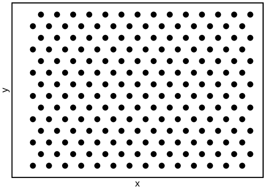
Computational Tools

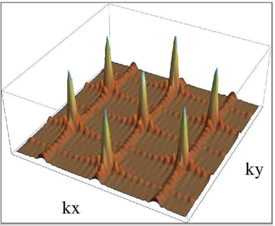
Molecular dynamics (MD) and **Classical (Metropolis) Monte Carlo (MC)** with **Simulated Annealing** at finite T. **Path Integral QMC** at low T; **Variation and Diffusion QMC** at $T = 0$.

Thermal melting of Wigner Molecules (WM)



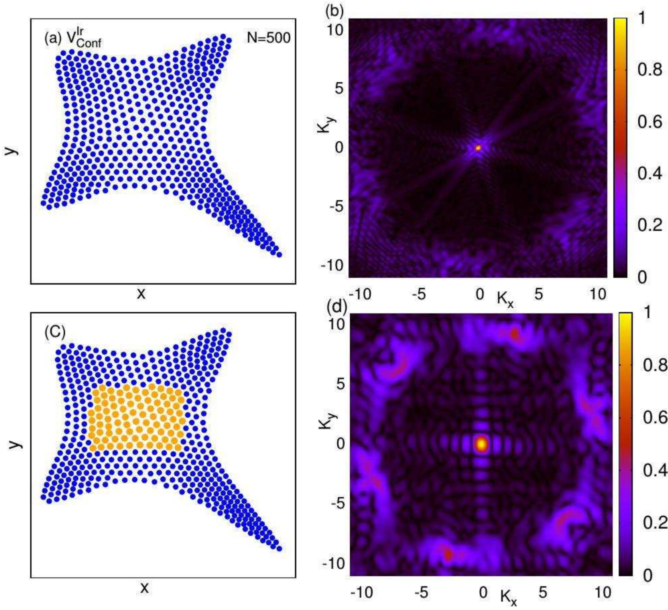
Absence of *Positional order* in irregular Wigner Molecule



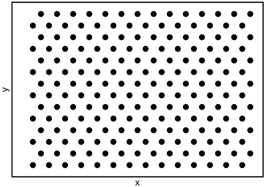


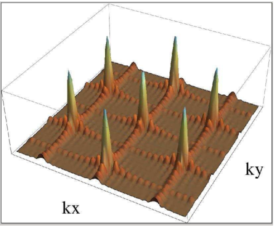
$$\rho_k = \frac{1}{N} \sum_{i=1}^{N} \exp(i\vec{k} \cdot \vec{r_i})$$

- Appreciable T-dependence of ρ_k **not** found.

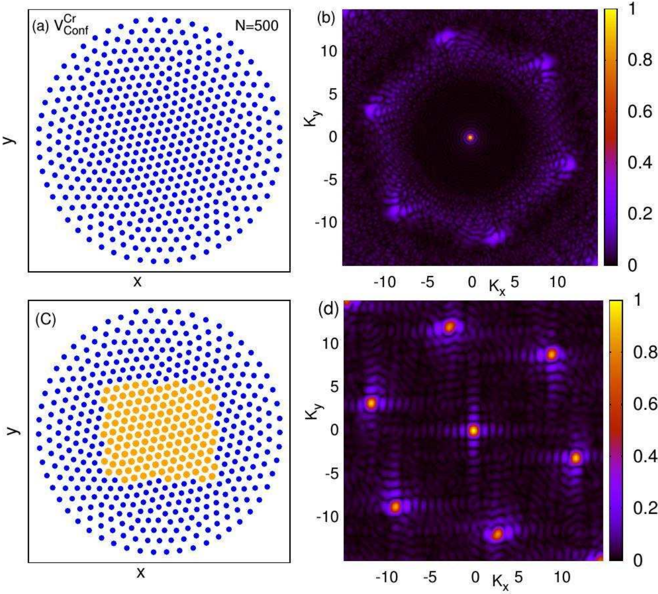


Positional order in Circular Wigner Molecule



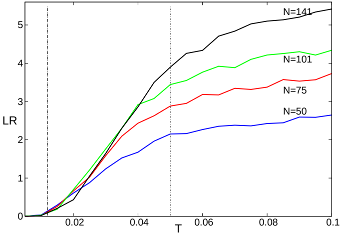


$$\rho_k = \frac{1}{N} \sum_{i=1}^{N} \exp(i\vec{k} \cdot \vec{r}_i)$$

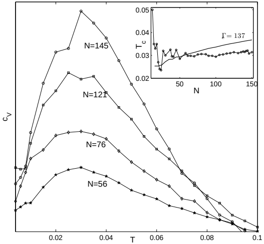


Static Correlations: EPJB **86**, 499, (2013), arXiv:1701.02338

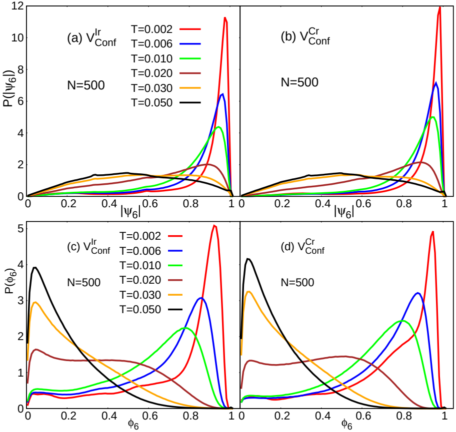
Lindemann: $\mathcal{L} = \frac{1}{N} \sum_{i=1}^{N} a_i^{-1} \sqrt{\langle |\vec{r}_i - \vec{r}_i^{\,0}|^2 \rangle}$



Specific Heat: $c_V = \frac{d\hat{E}}{dT} = T^{-2} \left[\langle E^2 \rangle - \langle E \rangle^2 \right]$



BOO: $\psi_6(i) = \sum_{k=1}^{N} \frac{1}{N_b} \sum_{l=1}^{N_b} e^{i6\theta_{ki}}$



$m_6(i)$: projection of $\psi_6(i)$ onto *mean local* orientation field.

$m_6(i) = \left| \psi_6^*(i) \, \frac{1}{N_b} \sum_{k=1}^{N_b} \psi_6(k) \right|$ Larsen & Grier, PRL '96

- Also studied $g(r), g_6(r)$, Generalized susceptibilities: χ_ψ, χ_ϕ

Take-home messages from static correlations & questions:

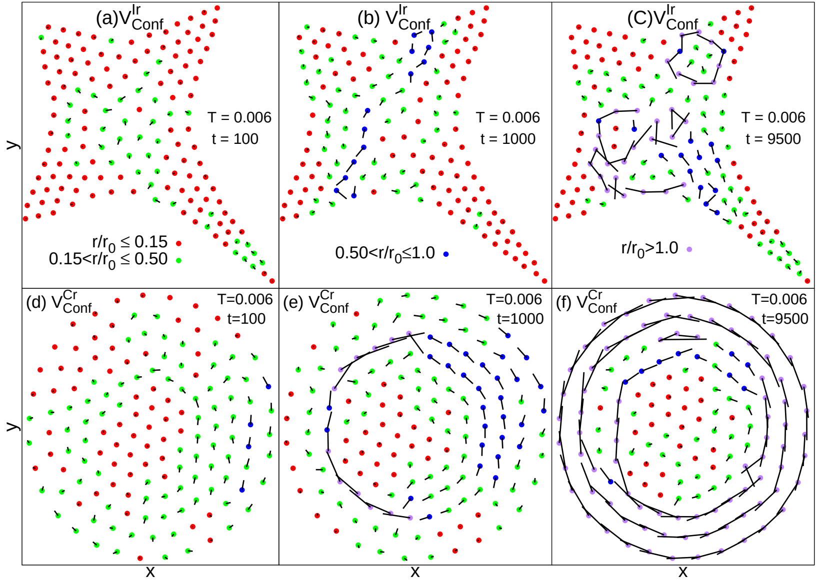
1. Crossover from 'solid'-like to 'liquid'-like behavior discerned from independent observables (unique T_x within tolerance).
2. No apparent distinction between T_X (within errorbars) in circular and irregular confinements.
3. Qualitative responses are more-or-less independent of N (for $100 \geq N \leq 100$) though there are differences in details.

- What can dynamics tell us about the 'solid' and 'liquid' in traps?
- Can motional signatures distinguish the crossover based on the *nature* of the confinement? (e.g., circular vs. irregular)
- Can we access generic signatures of disordered dynamics in traps?

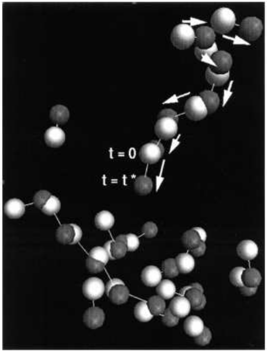
EPL, 114, 46001 (2016); arXiv:1701.02338; and unpublished

Displacements $[\Delta\vec{r}(t) = \{\vec{r}(t) - \vec{r}(0)\}]$ in 'solid'

- Spatially correlated inhomogeneous motion at large t even at low T in irregular traps.

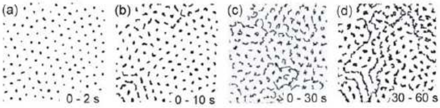


Spatially "correlated" displacements in literature...



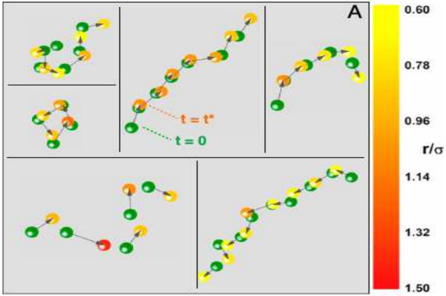
Lennard-Jones binary mixture

C.Donati *et. al.*; PRL(1998)



2D dusty plasma

C. Chan,*et.al.*, Contrib. Plasma Phys.(2009)



grain boundary in colloidal polycrystal

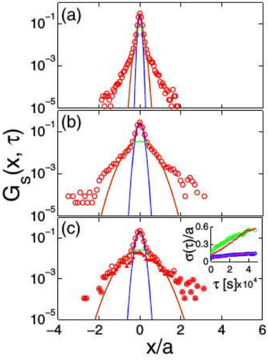
K.H.Nagamanasa, *et.al.*, PNAS (2011)

spatio-temporal density correlations:

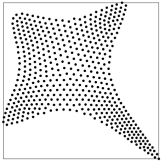
Dynamical (spatio-temporal) information best extracted from Van-Hove correlation function:

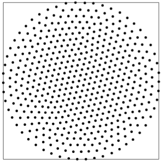
$$G(r,t) = \langle \sum_{i,j=1}^{N} \delta\left[r - |\vec{r}_i(t) - \vec{r}_j(0)|\right] \rangle$$

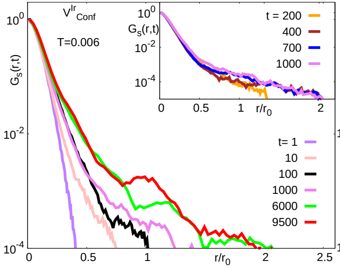
- Self-part $G_s(r,t)$ (when $i = j$): probability to move on an average a distance r in time t.

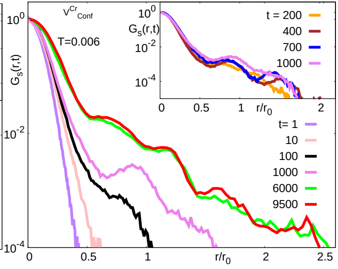


[Y. Gao *et. al.*; PRL'07]

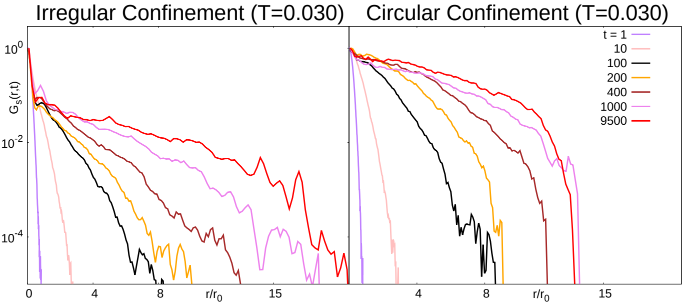


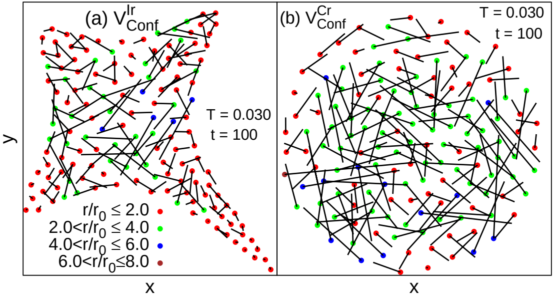






Results: $G_s(r,t)$ in 'liquid' (T=0.03) for IWM & CWM



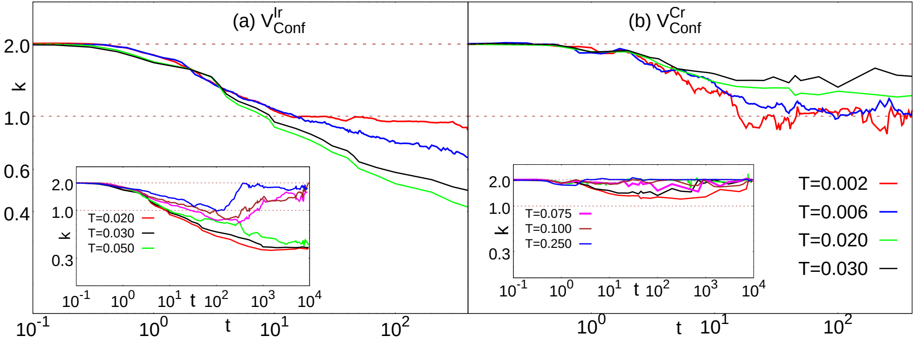


Stretched exponential decay of spatial correlation in IWM

Observation: • $G_s(r,t) \sim e^{-r^2/c}$ for small r $\forall t$. • $G_s(r,t)$ shows complex tail (large t).

Postulate: $G_s^{\rm small}(r,t) \sim e^{-r^2/c}$ for $r \le r_c$ and $G_s^{\rm large}(r,t) \sim e^{-lr^k}$ for $r > r_c$

• Optimal r_c and other parameters (**including** k) determined by minimizing **total** χ^2.



• Small t, All T: $k \simeq 2$, (Gaussian tail).

• Large t, Low T: (IWM + CWM) $k \sim 1$ (exponential tail) **[P. Chaudhuri *et al.*, PRL (2007)]**

• Large t, High T: • (CWM) $1 \ge k \le 2$, (**stretched Gaussian** tail); Expt: **[He *et al.* ACS Nano ('13)]**

• (IWM) $k < 1$, T-dependent **Stretched exponential** tail of spatial correlation!

Time scales: $\alpha-$ relaxation time from overlap Function

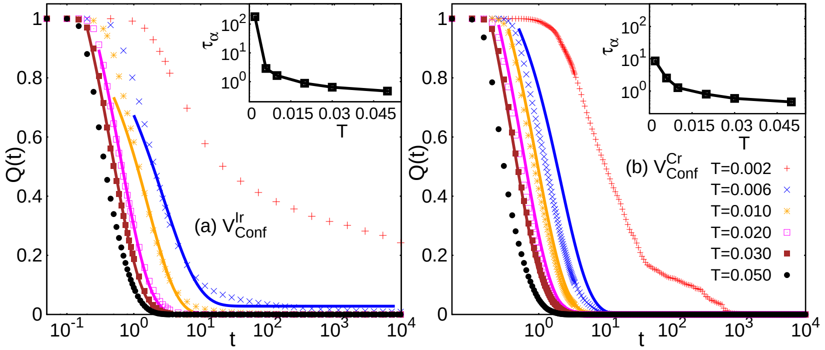
Overlap function

$$Q(t) = \frac{1}{N}\sum_{i=1}^{N} W(|\vec{r_i}(t) - \vec{r_i}(0)|) \quad \text{[Kob et al. ('12); Karmakar et al. ('14)]}$$

where $W(r_i) = 1$ if $r_i < r_{\text{cut}}$, & $W(r_i) = 0$ if $r_i > r_{\text{cut}}$ (satisfied once, only on first passage).

$\alpha-$ relaxation time (τ_α) from Overlap function: $Q(\tau_\alpha) = e^{-1}$

- similar to τ_α obtained from Intermediate Scattering Function.

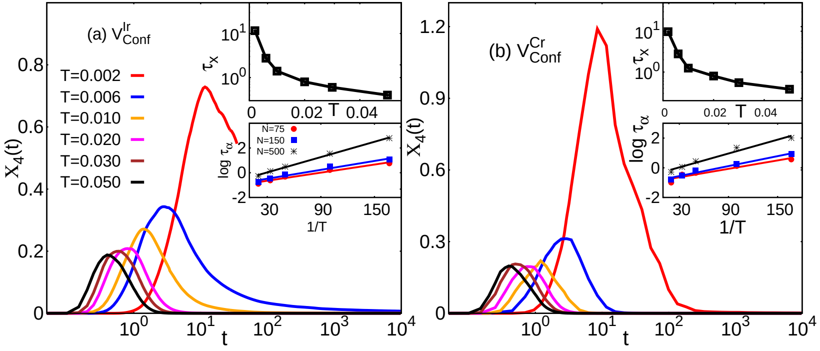


Time scales: from four-point density correlation $\chi_4(t)$

Dynamical four-point susceptibilities, $\chi_4(t)$, defined as:

$$\chi_4(t) = \frac{1}{N}[\langle Q^2(t)\rangle - \langle Q(t)\rangle^2]$$ [S.Karmakar et.al. PNAS, ('08)]

- $\chi_4(t)$ measures extent of dynamic heterogeneity (spatial correlations in particles' dynamics).
- $\tau_x(T)$ is the time-scale when dynamic heterogeneity is maximum at the given T.



- $\tau_x(T)$ similar for both confinements, while the nature of heterogeneity are different.

Time scales: Cage correlation Function

- When particles' cages rearrange, the system relaxes & particles diffuse.

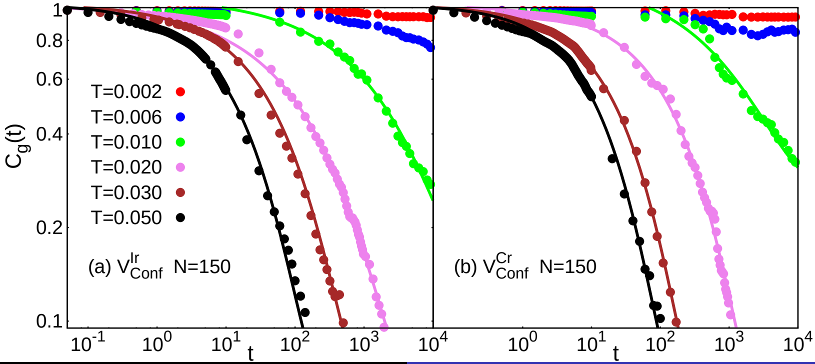
$\Rightarrow$ corresponding structural change characterized by a cage correlation (CC) function.

- Generalized neighbor list $\mathbf{L_i(t)}$ for particle i is a vector of length N $\mathbf{L_i(t)} = f(r_{ij})$.
- $f(r_{ij}) = 1$ if j is the nearest neighbor of i at time t **and** 0 otherwise. The CC function:

$$C_g(t) = \frac{\langle \mathbf{L_i(t)} \cdot \mathbf{L_i(0)} \rangle}{\langle \mathbf{L_i^2(0)} \rangle}$$

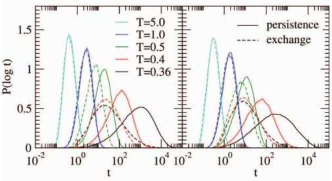
[E.Rabani et.al. PRL, ('99)]

- $C_g(t) \sim \exp[-(t/\tau_g)^c]$ with $c \sim 0.5$ for irregular, and ~ 0.6 for circular traps

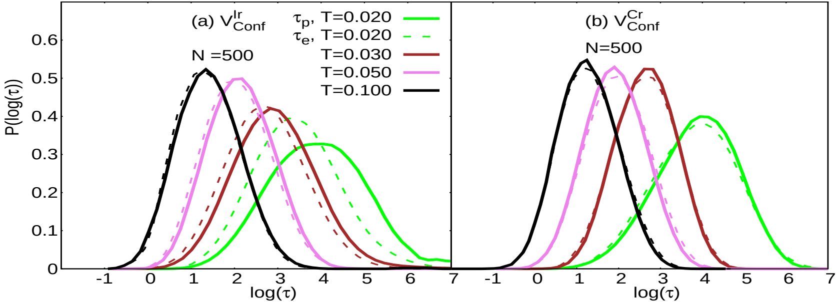


Time scales: Persistence and Exchange time

- Persistence time (τ_p, solid line): a particle displaced beyond a cut-off for the first time
- exchange time (τ_e, dotted line): time required for subsequent passage by cut-off distance.
  [Hedges *et. al.*, J. Chem. Phys.(2007)]



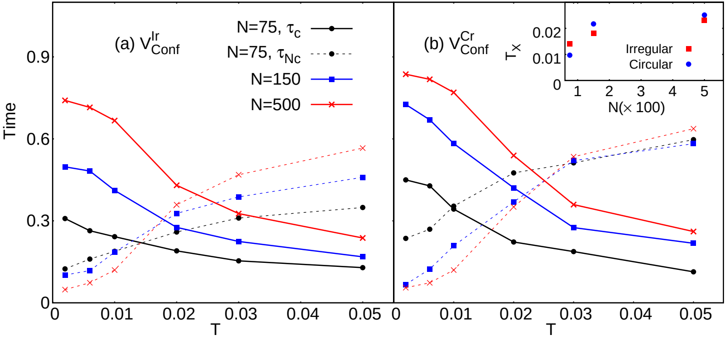
Glass-formers → **two time scales decouple at low T.**



- Are the two distributions of τ_e & τ_p statistically independent?
- Jackknife analysis and Ansari-Bradley test ⇒ Decoupling of distributions for $T \leq 0.020$ in IWM.
- Strong signature of glassy dyanmics, possibly absent in CWM.

Time scales: Caging time

A particle is caged if the relative fluctuation in the average distance w.r.t nearest neighbors is less than 10%. [J. Kim, C. Kim, and B. J. Sung, Phys. Rev. Lett. (2013)]

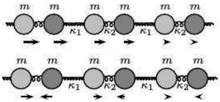


- T_X from the Crossing of average caging (τ_C) and non-caging (τ_{NC}) times is consistent with its extraction from statics.

Normal Mode (NM) Analysis: (ongoing research)

- Addresses dynamic responses: how each particle proposes to move in a given configuration (remember phonon in crystals!)

- **Normal Modes:** Construct $2N \times 2N$ Hessian matrix



$$A = \left(\frac{\partial^2 H}{\partial r_{i\alpha} \partial r_{j\beta}} \right)$$

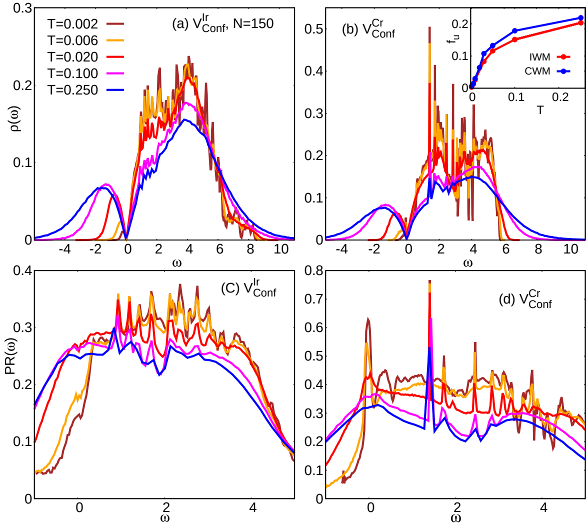
with INSTANTANEOUS configuration of N particles $\{(r_{1x}, r_{1y}), \cdots (r_{Nx}, r_{Ny}))\}$
$\Rightarrow$ results into Instantaneous Normal Modes (INM).

Eigenvalues of $A \rightarrow$ square of the eigen frequencies $\omega_n (n = 1, 2, 3, \cdots, 2N)$

Eigenvector $e_n \rightarrow$ oscillation pattern of the particles in mode number n.

INM: Density of States & Participation Ratio

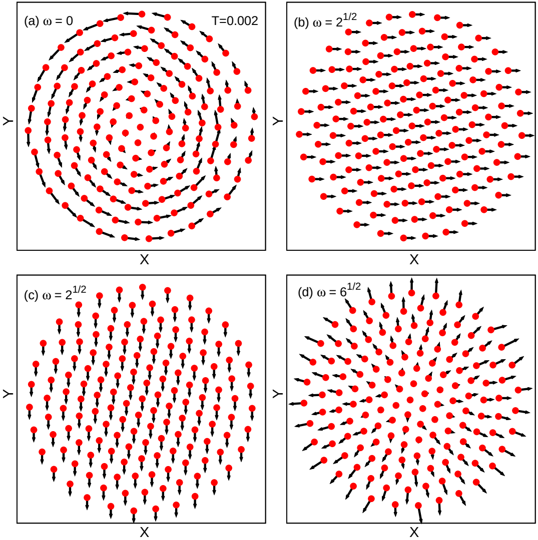
Density of states (DOS): Distribution of normal mode freq. with normalization $\int d\omega \rho(\omega) = 1.$



- Unstable modes ⇒ configurational transitions over potential hills.
- Some stable ($\omega > 0$) modes robust, features peak in $\rho(\omega)$ [at all T!].
- Large PR: large fraction of particles contribute to that mode.
- Intriguing correlation between robust modes and PR.
- Robust modes can occur from symmetry e.g. Circular.

How robust are the *robust* modes in irregular trap?

Normal mode pattern



- **CWM:** Symmetry dictated stable (robust to T) modes (N independent):

  (a) $\omega = 0$: rotation of the system as a whole

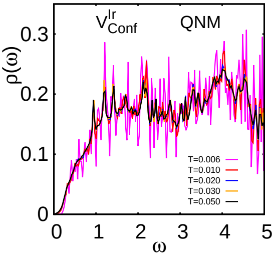
  (b,c) $\omega = \sqrt{2}$: sloshing mode (the center-of-mass mode, doubly degenerate in 2D)

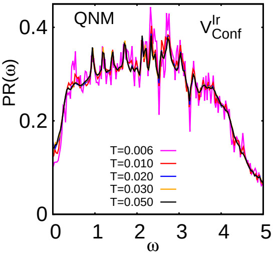
  (d) $\omega = \sqrt{6}$: breathing mode(BM)

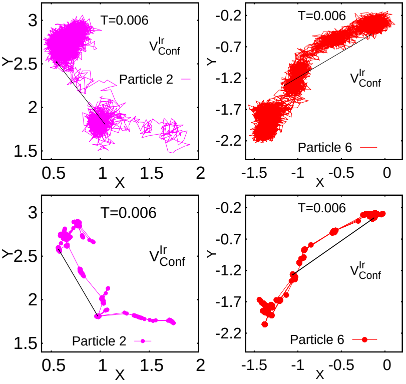
- **IWM:** Hard to associate particular pattern: mostly vortex-anti-vortex type

Quenched Normal Modes (QNM):

- Normal mode analysis with energy minimized configurations leading to Inherent Structures(IS) $\Rightarrow$ only stable modes !
- Possibility of observing signatures of glassy dynamics through QNM.



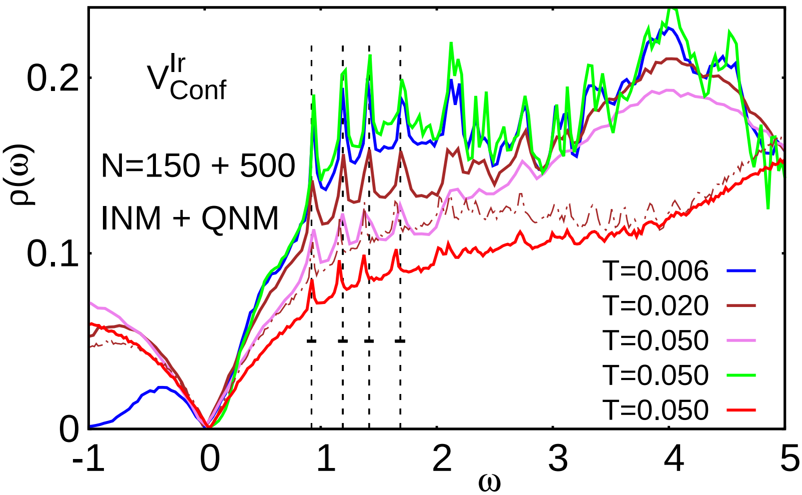




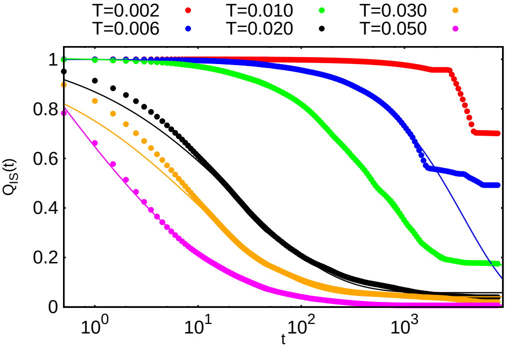
- Stable modes at same ω found from INM.

- Removes vibrational motion.
- Long displacement through transition between different ISs.

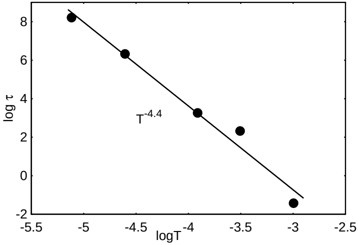
Normal modes

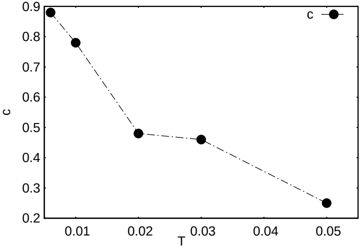


Overlap Function (in inherent structure (IS) space!)



$$Q_{IS}(t) \sim \exp[-(t/\tau)^c]$$

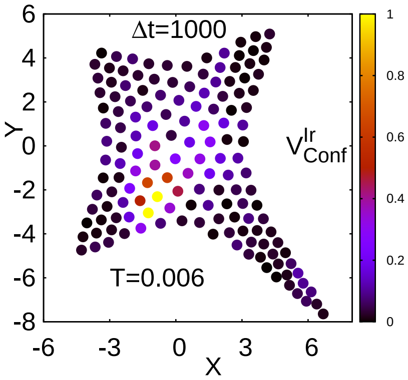




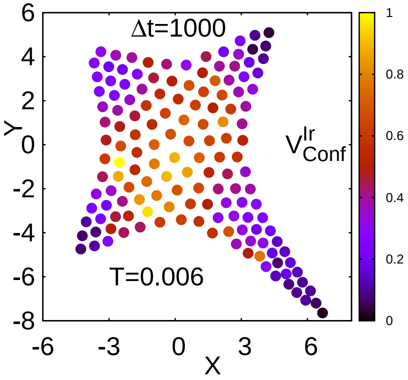
Region of propensity

- Can we construct long time displacements from normal modes at *initial* instant?
- Proposed for glassy systems (**A.W. Cooper et al. Nat. Phys. ('08)**).

Displacement of All particles



Eigen vector sum: $E^i_s = (N_e)^{-1} \sum_{n=1}^{N_e} |e^i_n|^2$



• Close connection between the dynamics of the particles and properties of the inherent structures sampled by them

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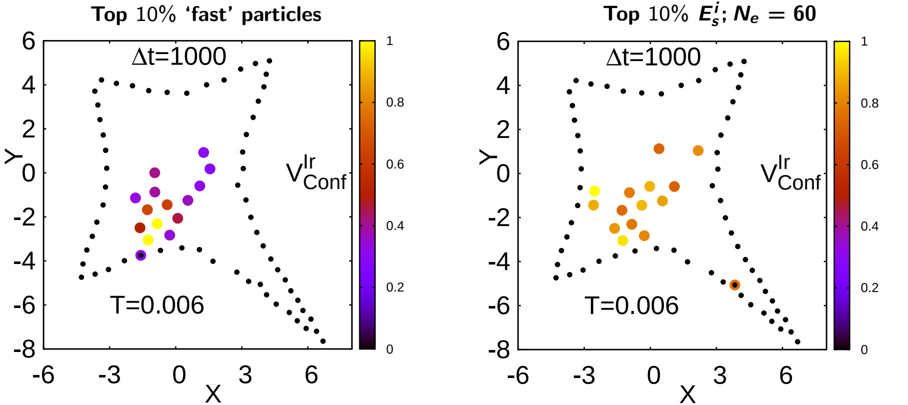

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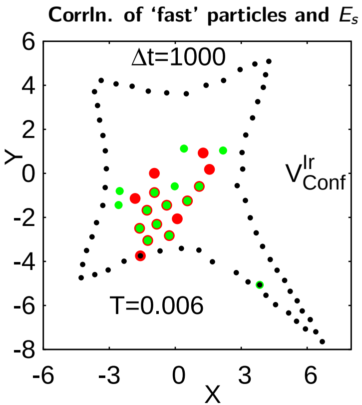
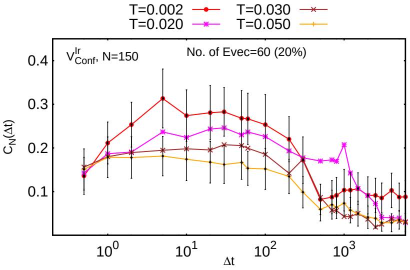
Quantifying propensity



$$C_N(\Delta t) = N_f^{-1} \sum_{i=1}^{N} \langle C_d(i, \Delta t) C_e(i, t_0) \rangle$$

$C_d(i, \Delta t) = 1$ only if i among top 10% 'fast' particles
$C_e(i, t_0) = 1$ if i = top 10% of highest Es_i

Here, N_f is the total number of top 10% fast particles



- Quantifying propensity for a wide parameter space is under current investigation !

Quantum Melting in confinements:

Hamiltonian (for Harmonic trap):

$$H = \sum_{i=1}^{N} \left[-\frac{n^2}{2} \nabla_i^2 + r_i^2 \right] + \sum_{i<j}^{N} \frac{1}{r_{ij}}.$$

$n = \sqrt{2} l_0^2 / r_0^2$, $l_0^2 = \hbar / m\omega_0$ ($E_0 = e^2/\epsilon r_0 = m\omega_0^2/2$) and $r_s = 1/n^2$.

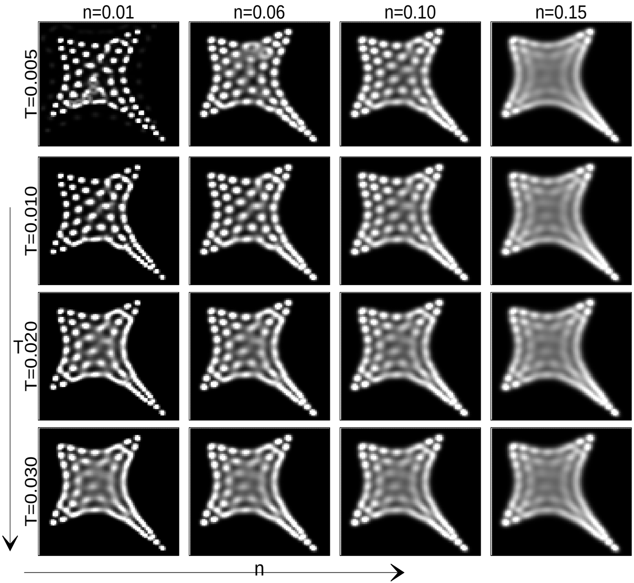
$n = 0 \Rightarrow$ classical, increase of n induces quantum fluctuations.

- Included: **Zero-point motion / quantum dynamics.**
- Quantum statistics: Boltzmannons (PIMC), Spin-$\frac{1}{2}$ Fermions (VMC + DMC)

• with Dyuti Bhattacharya, Filinov, and Bonitz; Eur. Phys. J. B, 89 (2016)

• With **Anurag Banerjee**, ongoing!

Density profile



- Thermal fluctuations → tortuous path of melting.
- Quantum fluctuations → largely by diffusion around equilibrium position.
- **Re-Entrant Behavior**

  → For the largest quantum fluctuation ($n = 0.15$), increasing T 'weakens liquidity'!

Acknowledging Collaborators

Students:
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Chandan Dasgupta
(IISc Bangalore)

A. V. Fillinov & M. Bonitz (Kiel U. Germany)

- Thanks : Dipak Dhar (TIFR + IISER Pune)

Conclusions

1. Spatio-temporal correlations chracterize ‘solid’ to ‘liquid’ crossover in Wigner molecules.
2. T_X is not sensitive to N or confinement geometry for $100 \leq N \leq 500$.
3. Intriguing motional signatures for confined Coulomb particles!
4. Multiple time-scales for relaxation identified.
   - Complex motion yields slow relaxations, akin to supercooled liquids.
5. Outlook:
   - "Glassiness" and the role of defects?
   - Classical vs. Quantum dynamics, observables?