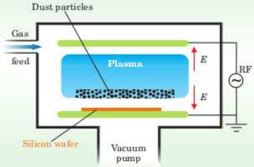
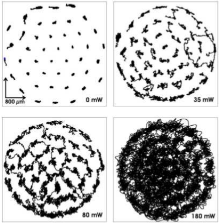
# Spatio-temporal correlations across the melting of 2D Wigner molecules

**Amit Ghosal**
IISER KOLKATA



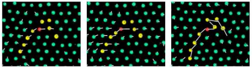

- Coulomb interacting particles in $2\mathrm{D}$ confinements.
- Static & Dynamic responses across 'melting'.
- Effect of 'Disorder/irregularity' on melting.




Melzer Group

## Computational Tools

- **Molecular dynamics** and **Classical (Metropolis) Monte Carlo** with **Simulated Annealing** at finite $T$.
- Path integral Quantum Monte Carlo (QMC) at low $T$; variational and diffusion QMC at $T = 0$.



B. Meer, *et.al.*, PNAS'14

# Crystal of Coulomb particles and its melting:

Wigner Crystal Melting (1934)

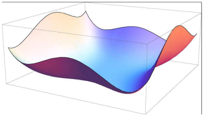
Competetion between PE & KE

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 **[Gann, Chakravarty & Chester, 1979]**
- KE $\sim$ Quantum (zero-point) fluctuations $\Rightarrow$ Quantum melting. **[Tanatar & Ceperley, 1989]**

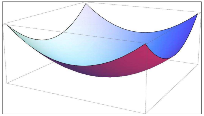
• In confinements, Wigner Crystal $\Rightarrow$ "Wigner Molecule"

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



**(a) Irregular:**

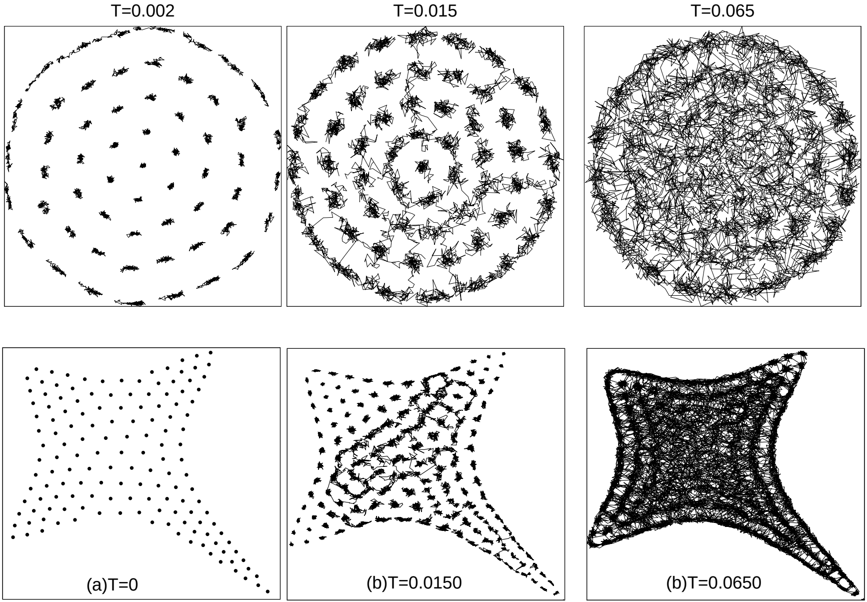
$$V_{\rm conf}^{\rm Ir}(r) = a\{x^4/b + by^4 - 2\lambda x^2y^2 + \gamma(x-y)xyr\}$$



**(b) Circular:**

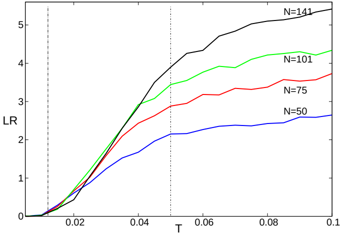
$$V_{\rm conf}^{\rm Cr}(r) = \alpha r^2, \text{ with } \alpha = m\omega^2/2$$

# Thermal melting of Wigner Molecules (WM)

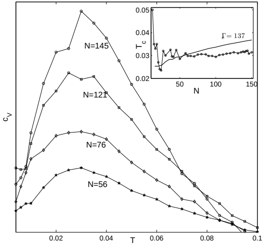


# Static Correlations: EPJB **86**, 499, (2013), arXiv:1701.02338

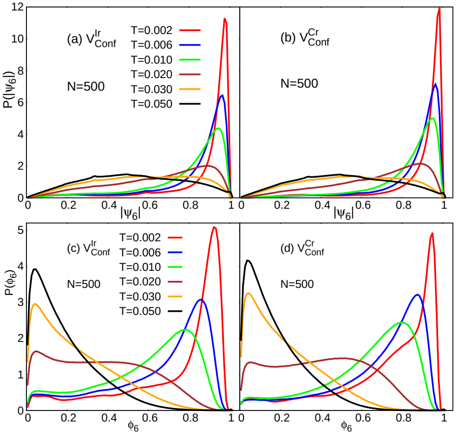
**Lindemann:** $\mathcal{L} = \frac{1}{N} \sum_{i=1}^{N} a_i^{-1} \sqrt{\langle |\vec{r}_i - \vec{r}_i^{\,0}|^2 \rangle}$




**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: $\chi_\psi, \chi_\phi$

# Take-home messages from static correlations & questions:

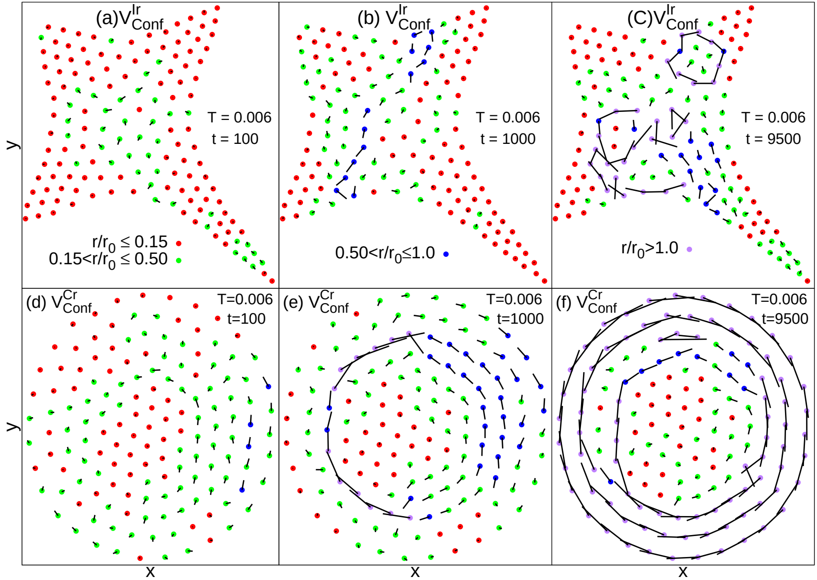
1. Crossover from 'solid'-like to 'liquid'-like behavior discerned from independent observables (unique $T_x$ within tolerance).
2. No apparent distinction between $T_X$ (within errorbars) in circular and irregular confinements.
3. Qualitative responses are more-or-less independent of $N$ (for $100 \geq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

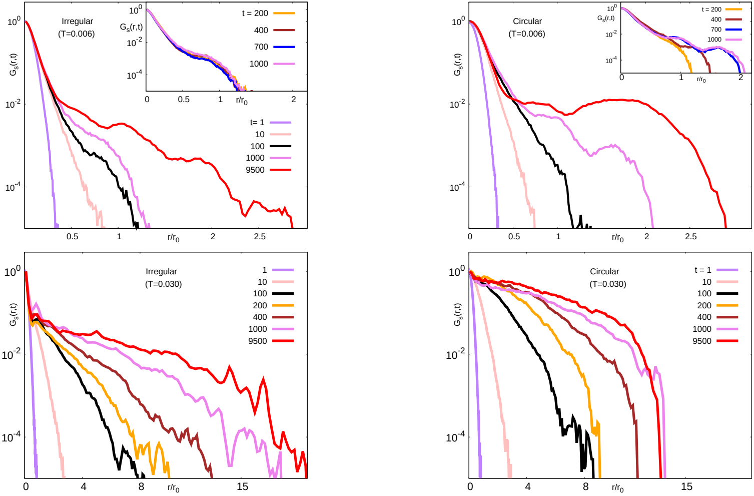


# spatio-temporal density correlations:

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 $t$.

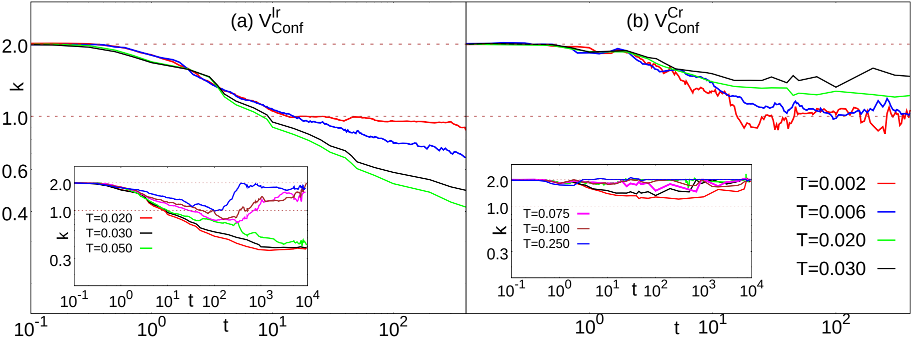


# *Stretched exponential* decay of spatial correlation in IWM

**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 $r \leq r_c$ 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** $\chi^2$.



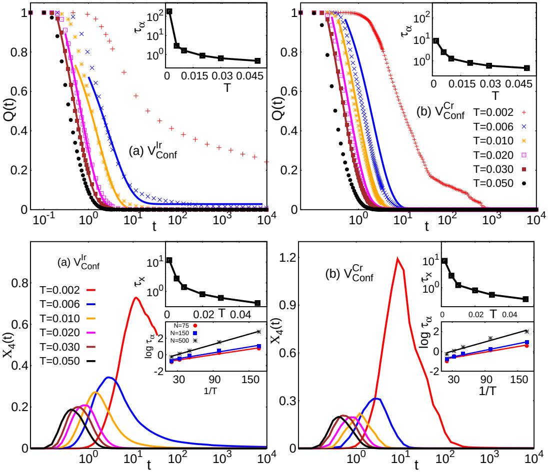

• Small $t$, All $T$: $k \simeq 2$, (Gaussian tail).

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

• Large $t$, High $T$: • (CWM) $1 \geq k \leq 2$, (**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




- $Q(t) = \frac{1}{N}\sum_{i=1}^{N} W(|\vec{r}_i(t) - \vec{r}_i(0)|)$

  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).

  [Kob et al. ('12); Karmakar et al. ('14)]

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

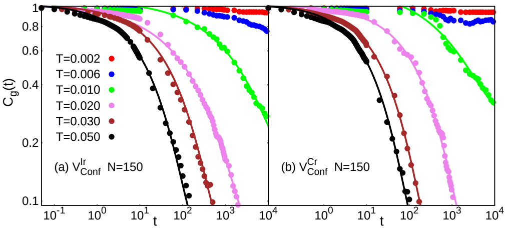
- $\chi_4(t) = \frac{1}{N}[\langle Q^2(t)\rangle - \langle Q(t)\rangle^2]$

  [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 $T$.

# Dynamical Correlations




- When particles' cages rearrange, the system relaxes & particles diffuse.
$\Rightarrow$ corresponding structural change characterized by a cage correlation (CC) function.

**Cage correlation function:**

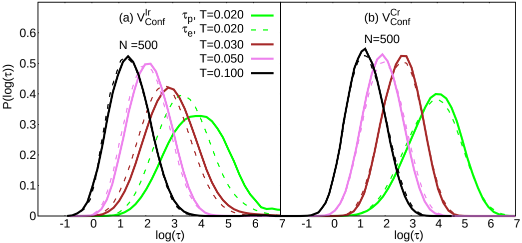
$$C_g(t) = \frac{\langle \mathbf{L^{(i)}(t) \cdot L^{(i)}(0)} \rangle}{\langle \mathbf{L^{(i)2}(0)} \rangle}$$ [Rabani et.al. PRL'99]

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

- Persistence time ($\tau_p$, solid line): a particle displaced beyond a cut-off for the first time.

- Exchange time ($\tau_e$, dotted line): time required for subsequent passage by cut-off distance. [Hedges *et. al.*, J. Chem. Phys.(2007)]

- Two distributions decouple for Irregular confinement but signature of decoupling is weaker for Circular confinement.



# 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 $r_s = 1/n^2$.

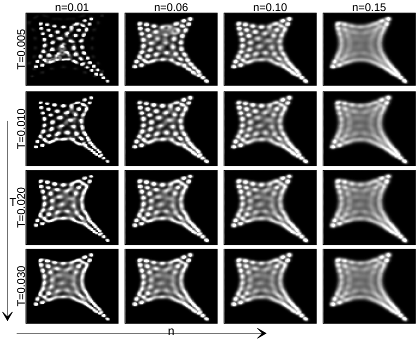
$n = 0 \Rightarrow$ classical, increase of $n$ induces quantum fluctuations.

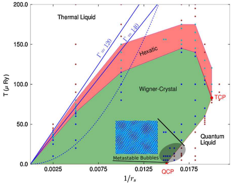
- Included: **Zero-point motion / quantum dynamics.**
- Quantum statistics: Boltzmannons (PIMC), Spin-$\frac{1}{2}$ Fermions (VMC + DMC)
- Thermal fluctuations $\rightarrow$ tortuous path of melting.
- Quantum fluctuations $\rightarrow$diffusion around equilibrium position.

A study similar to that on bulk systems.

**B. Clark, M. Casula, and D. Ceperley PRL (2009)**

[D. Bhattacharya et.al. EPJ.B, 89, 60 (2016)]



# Acknowledging Collaborators

Students:
(IISER K)


Biswarup Ash


Dyuti Bhattacharya


Anurag Banerjee

Collaborators:


Jaydeb Chakrabarti
(SNBNCBS Kolkata)


Chandan Dasgupta
(IISc Bangalore)

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

# 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?