Normal mode analysis of Coulomb particles in irregular traps

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Objective:

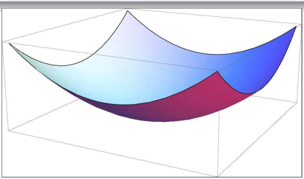
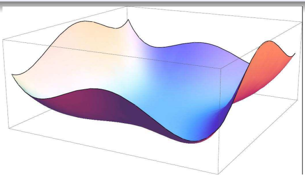
- Insights on dynamics in systems with Coulomb interacting particles & disorder.
- Analysis of normal modes (Quenched + Instantaneous).
- Characterization: Density of modes, participation ratio & spectral statictics.
- **How low lying inherent structures control long-time dynamic heterogeneity?**

ICTS, ISPCM 2018

Model, Method & Parameters :

Study

- **Static & Dynamics of Coulomb particles in irregular (& regular!) confinements.**

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: $V^{\rm Ir}_{\rm conf}(r) = a\{x^4/b + by^4 - 2\lambda x^2 y^2 + \gamma(x-y)xyr\}$, [Bohigas et.al, Phys. Rep. **223**, 43 (93)]

Broken spatial symmetry of $V^{\rm Ir}_{\rm conf}(r)$, together with chaotic motion of single particle in it, are taken as *signatures of disorder*

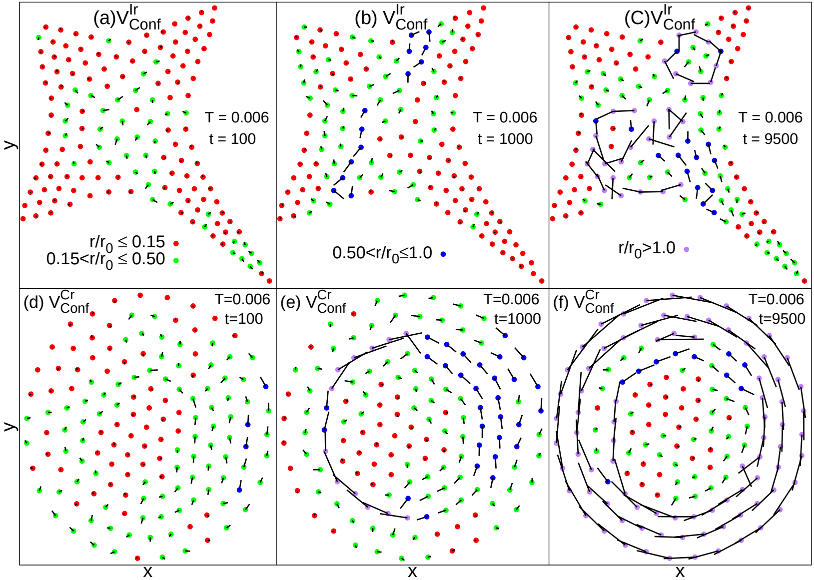
(b) Circular: $V^{\rm Cr}_{\rm conf}(r) = \alpha r^2$, with $\alpha = m\omega^2/2$.

Computational Tools

Molecular dynamics (MD) and **Classical (Metropolis) Monte Carlo (MC)** with **Simulated Annealing** at finite T.

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

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



Take-home messages from spatio-temporal correlations:

1. Crossover from 'solid'-like to 'liquid'-like behavior discerned studying independent observables (unique T_x found within tolerance).
2. No apparent distinction between T_X (within errorbars) in circular & irregular confinements.
3. Qualitative responses are more-or-less independent of N (for $100 \leq N \leq 500$) though there are differences in details.

- Intriguing motional signatures (e.g. dynamic heterogeneity) found.
- Nature of dynamic heterogeneity distinguishes the thermal crossover based on the type of confinement (e.g., circular vs. irregular)
- Multiple time-scales for relaxation identified.
  - Complex motion yields slow relaxations, akin to supercooled liquids.
- **Access generic signatures of disordered dynamics in traps?**

• **EPJB 86, 499, (2013);** • **EPL 114, 46001 (2016);** • **Phys. Rev. E, 96, 042105 (2017)**

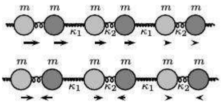
Normal Mode (NM) Analysis

Goal:

- **Develop deeper insight into the dynamics of Coulomb particles in traps.**

- 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 $\{(r_{1x}, r_{1y}), \cdots (r_{Nx}, r_{Ny})\}$

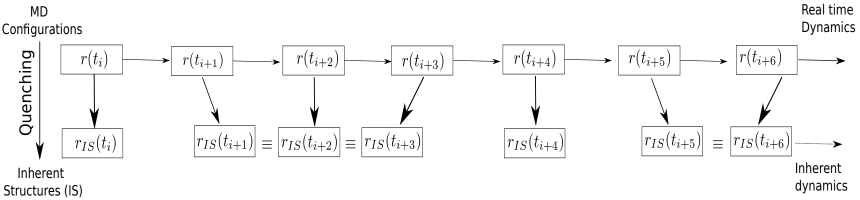
$\Rightarrow$ results into Instantaneous Normal Modes (INM).

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

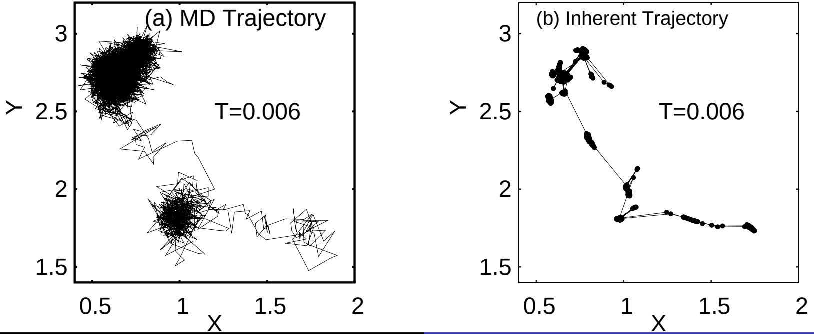
Eigenvector $e_n \rightarrow$ oscillation pattern of the particles in mode number n.

Quenched Normal Mode (QNM) Analysis

- Procedure to obtain the inherent structures from instantaneous configurations



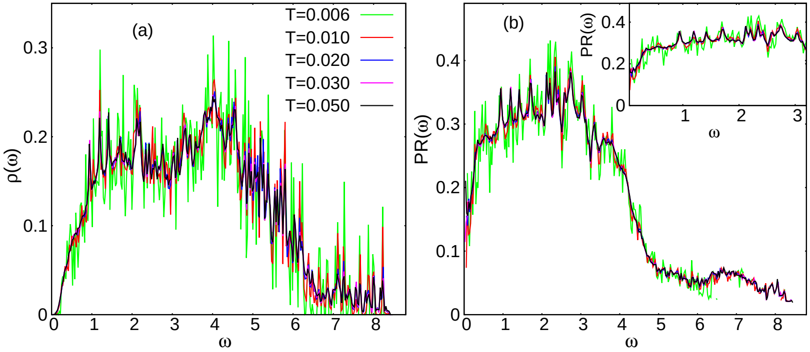
- Quenching effects on actual dynamics of particles $\rightarrow$ get rid of small amplitude vibrations.



Analysis of Quenched Normal Modes (QNM)

QNM: Energy minimized configurations starting from equilibrium configurations at a T.

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



Participation ratio $\mathrm{PR}(\omega_m) = \left[N \sum_{i=1}^{N} \left(\vec{e}_i(m) \cdot \vec{e}_i(m) \right)^2 \right]^{-1}$

- **PR**$(\omega_m) < 0.05$: small (very low and high ω_m) $\Rightarrow$ Localized modes.
- **PR**$(\omega_m) > 0.35$: Large (intermediate ω_m) $\Rightarrow$ Delocalized modes.

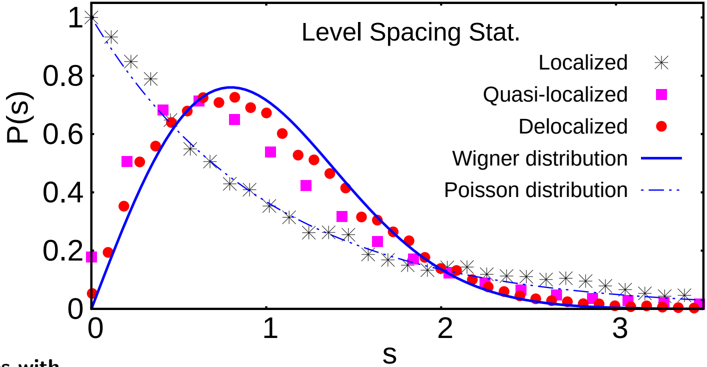
quasi-localized modes for $0.05 <$ **PR** < 0.35 ?

Analysis of QNM: Spectral Statistics

Localized, **quasi-localized** and **delocalized** modes.

Distribution of spacings s between the eigenvalues (λ) of the Hessian matrix.

$s_i = (\lambda_{i+1} - \lambda_i)/\Delta$; Δ is the mean-level spacing.



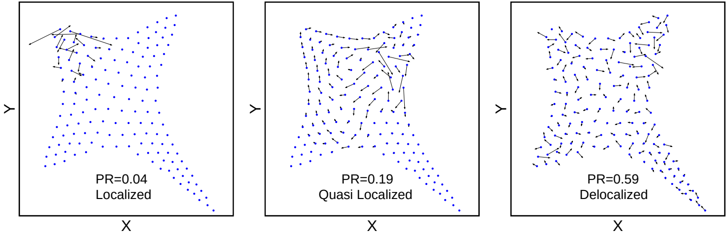
For modes with

- **PR**$<$ 0.05 Poisson distribution $\Rightarrow$ Localized modes.
- **PR**$>$ 0.35 Wigner distribution $\Rightarrow$ Delocalized modes.
- 0.05 $<$**PR**$<$ 0.35 $\Rightarrow$ $P(s)$ intermediate between Poisson and Wigner distribution
  $\Rightarrow$ Quasi-localized modes (established through Brody function test).

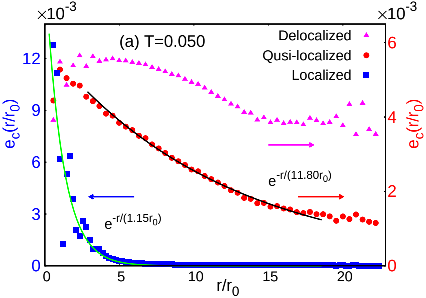
Analysis of QNM: Distribution of polarization vector

Localized, **quasi-localized** and **delocalized**

Representative modes:



- Magnitude of polarization vector $n(\vec{r}_i) = |\vec{e}(\vec{r}_i)|$; **measure correlation** $\langle e_c(\vec{r}) = n(\vec{r}_i)n(\vec{r}_i + \vec{r})\rangle$

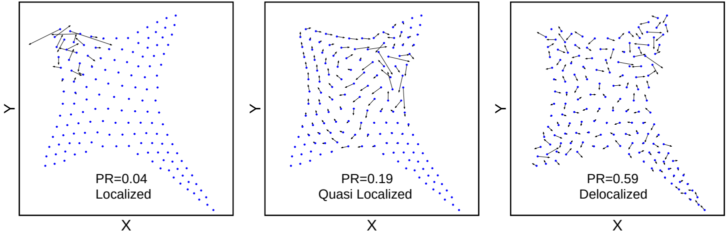


- Delocalized modes: Weak r dependence.
- Localized modes: Sharp fall. $e_c \to 0$ by $r \sim r_0$.
- **Quasi-localized** modes: $e_c(r) \propto \exp[-r/\xi_{\rm ql}]$; $\xi_{\rm ql} \sim 11r_0$.

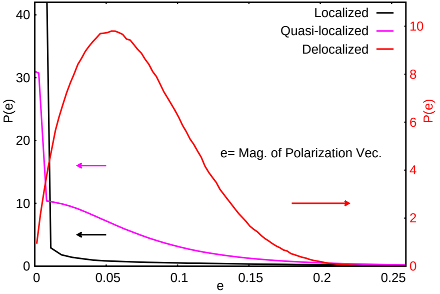
Analysis of QNM: Distribution of polarization vector

Localized, **quasi-localized** and **delocalized**

Representative modes:



- **Distribution of the magnitude (e) of polarization vectors.**



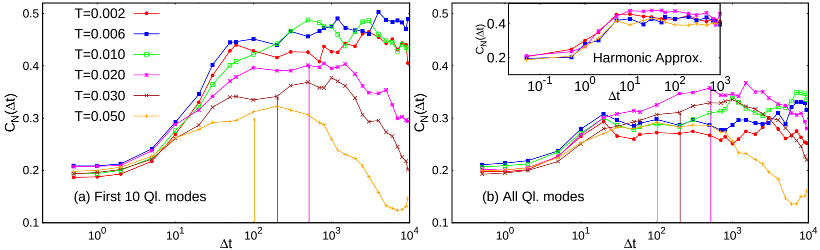
- Localized modes $\Rightarrow$ Sharp peak at $e \sim 0$.
- Delocalized modes $\Rightarrow$ Broad distribution.
- Quasi-localized modes $\Rightarrow$ Peak at zero $+$ Long tail.

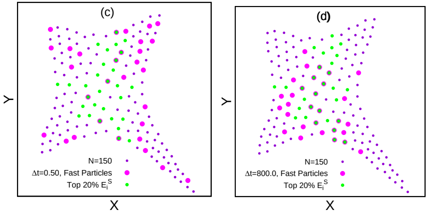
Identify 'fast' particles at long time from quasi-loc modes

$C_d(i, \Delta t) = 1$ only if i among top 20% particles with largest displacement during time $(t_0, t_0 + \Delta t)$;

$C_e(i, t_0) = 1$ if i among top 20% of highest E_i; Here, $E_i = \frac{1}{N_e} \sum_{m=1}^{N_e} |e_i(m)|^2$ with QNMs at t_0; $N_e \ll 2N$ (typically).

Define correlation: $\boldsymbol{C_N(\Delta t) = \sum_{i=1}^{N} \langle C_d(i, \Delta t) C_e(i, t_0) \rangle}$



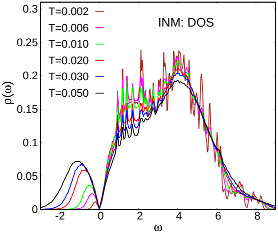


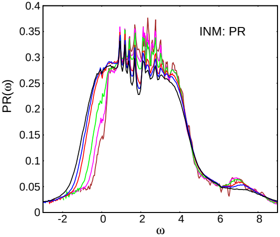
- Only few QL modes identify 'fast' particles.
- Harmonic approx. good for small t, but, fails to characterize structural relaxation, which requires jumps over barrier beyond HA.
- $C_N(\Delta t)$ attains maximum for $\Delta t \sim \tau_\alpha$.
- Explains structural origin of heterogeneous and slow dynamics in supercooled liquids.

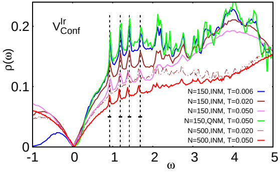
[Widmer-Cooper *et al.* Nat. Phys. ('08); JCP ('09)]

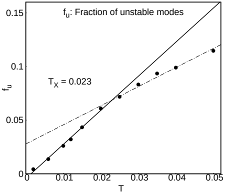
Analysis of Instantaneou Normal Modes (INM)

Hessian matrix evealuated from *instantaneous* equilibrium configurations !!









- Unstable mode (negative $\lambda \rightarrow$ imaginary ω) $\Rightarrow$ configurational transitions over potential hills.
- Some stable ($\omega > 0$) modes are robust, features peak in $\rho(\omega)$. [insensitive to T or N !!].
- Robust mode occur at similar ω in INM & QNM for different Ns!
- Identify T_X from T-dependence of fraction of unstable modes !!

Conclusions

- **Comprehension of classical *Coulomb-glass* from NM analysis**

1. Intriguing motional signatures for confined and long-range (Coulomb) interacting particles!
2. Glassiness clarified through Normal mode analysis.
3. Quasi-localized modes appraised via participation ratio and spectral analysis. Length-scale associated with quasi-localized mode extracted by cluster analysis.
4. Instantaneous Normal modes identify the crossover temperature (T_X) for solid- to liquid-like behavior.
5. **Role of low lying quasi-localized modes elucidated in dictating the long-time dynamic heterogeneity**.

Quasi-localized mode: Cluster analysis

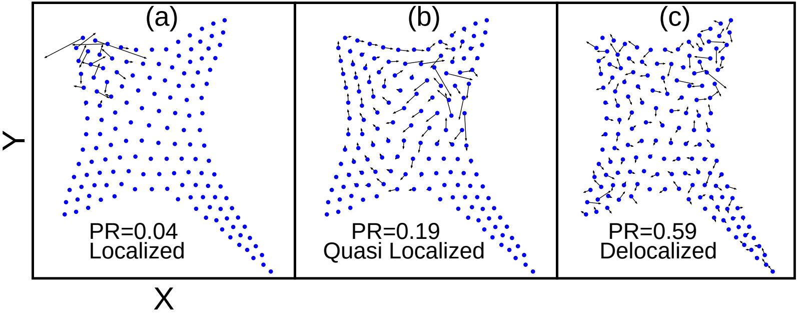
Quasi-localized modes:

- They lie in between localized and delocalized modes.
- Particles with significant magnitude of polarization vector are spatially clustered.

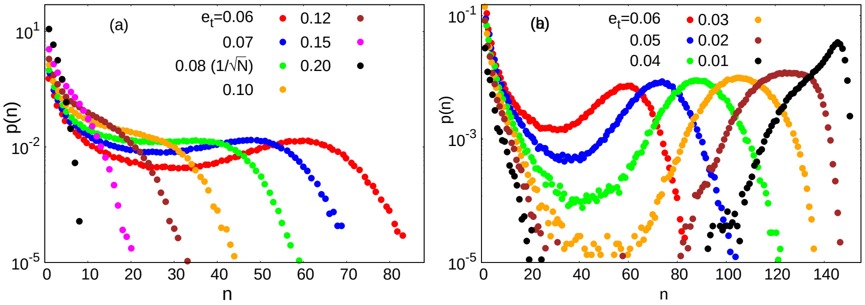
Can we make a statistical estimate of size of such clusters?

Two parameters: (1) Cut-off on magnitude of a polarization vector for it to be part of a cluster.

(2) Parameter to decide whether two particles are members of a gievn cluster.
$\sim$ average interparticle spacing



Probability of finding cluster of n-particles: $P(n)$



- As the cut-off (on the magnitude of polarization vector, to be considered a part of cluster) is increased, $P(n)$ changes gradually from a bimodal curve to a monotonic and nearly exponential.
- $P(n)$ found to be insensitive to T.
- Bimodality first occur for cut-off on polarization vector $e_c \sim \frac{1}{\sqrt{N}} \sim$ 0.07-0.08 $\sim$ 10%-Lindemann ratio, yielding a typical cluster size $\sim 8r_0 \sim \xi_{\rm ql}$.

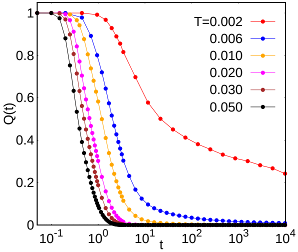
Overlap function and structural relaxation time

$$Q(t) = \frac{1}{N}\sum_{i=1}^{N} W(|\vec{r}_i(t) - \vec{r}_i(0)|);\ \text{where}\ W(r_i) = 1\ \text{if}\ r_i < r_{\text{cut}},\ \&\ W(r_i) = 0\ \text{if}\ r_i > r_{\text{cut}}$$

[Karmakar et al. ('14)]

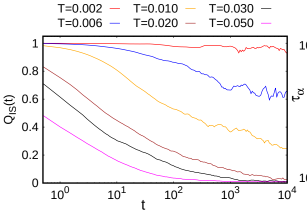
From MD configurations: [Ash, Chakrabarti & AG, PRE, **96**, 42105]

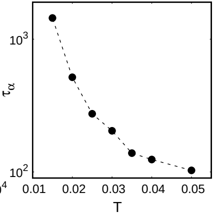
- Small time decay due to vibrational motion, while long time decay corresponds to structural relaxation.
- The two step decay makes the extraction of structural relaxation time (τ_α) difficult, even erroneous!



Inherent structure space:

- Structural relaxation through transition from one inherent structure to another.
- Estimate (τ_α) from the area under the $Q_{IS}(t)$ vs. t curve for all T (provided $Q_{IS}(t) \to 0$ at long time for that T).
- τ_α increases rapidly as T is decreased.





Comparing trajectories from MD & Harmonic approx.

Comparing particle trajectories from full numerical simulations (molecular dynamics), with those from harmonic approximation: