Multiple scattering of light from fractal aggregates

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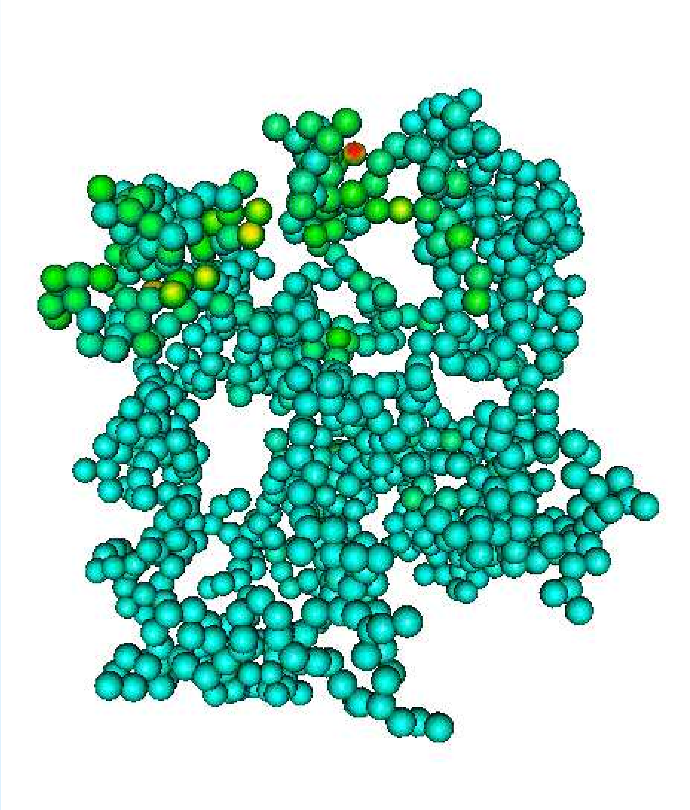
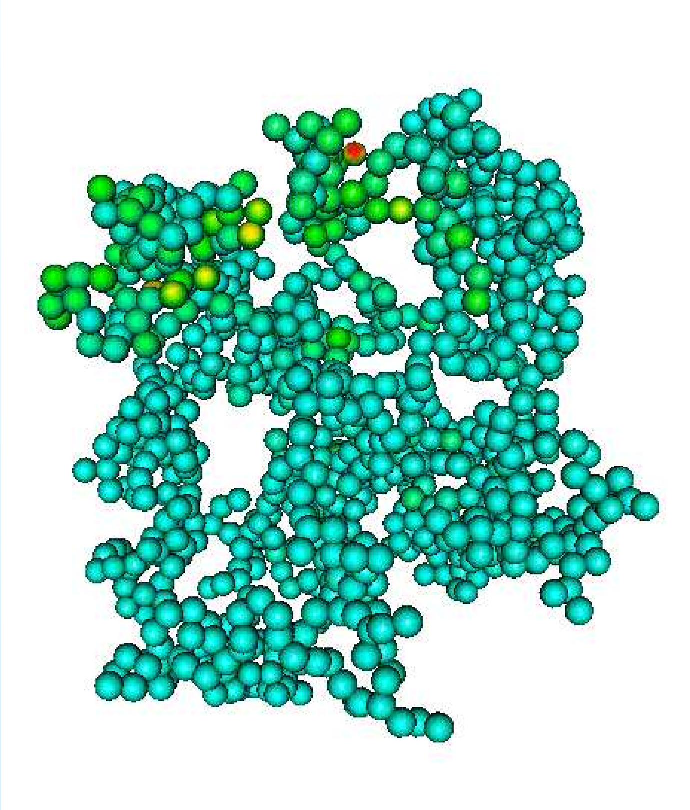
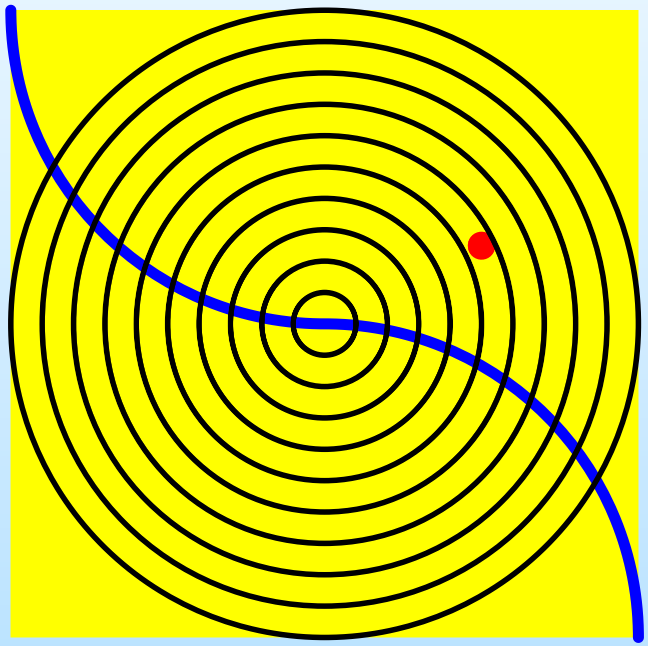
The System



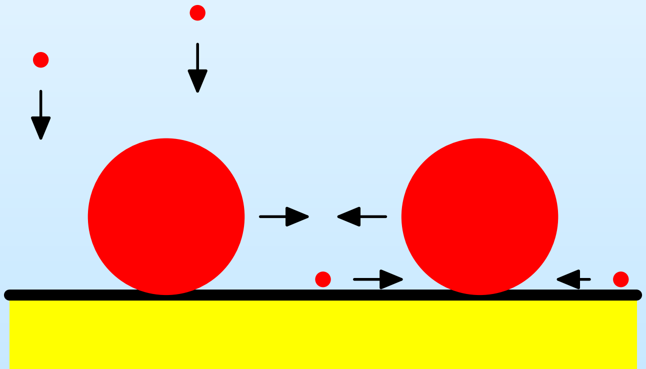
The System



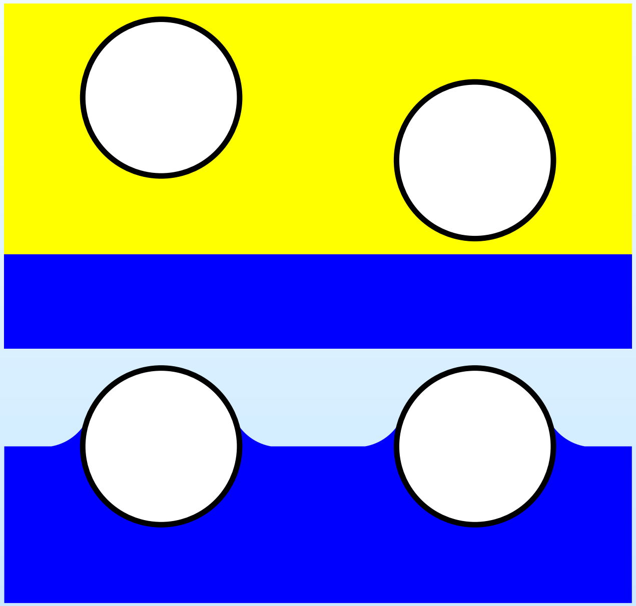


- Scale invariance: $M(r) \propto r^{d_f}$
- Two particle correlations: $C(r) \propto r^{d_f - d}$

Fractal aggregation at surfaces



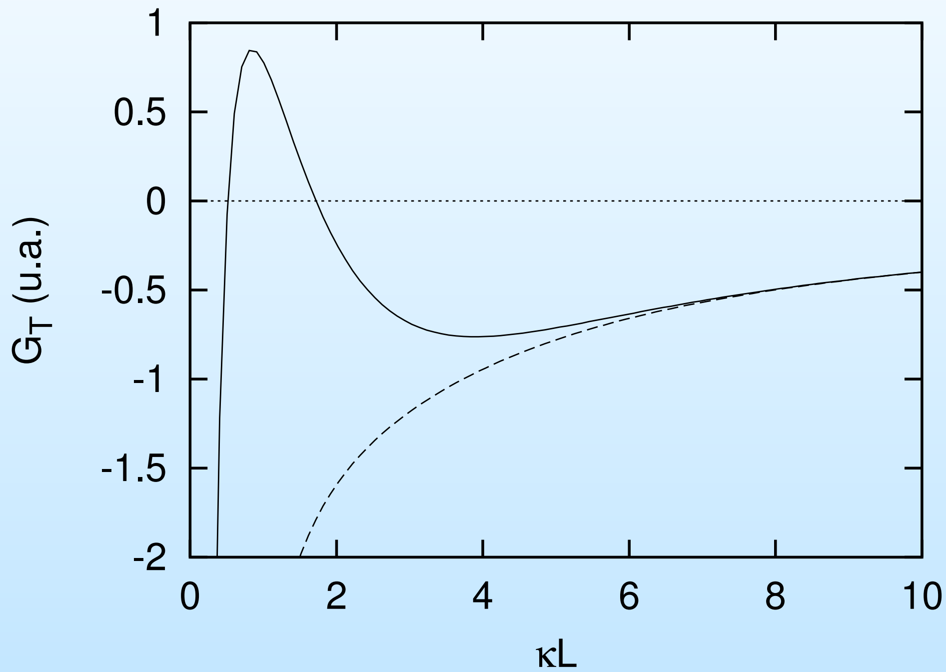
MBE on cold substrates



Latex spheres on water

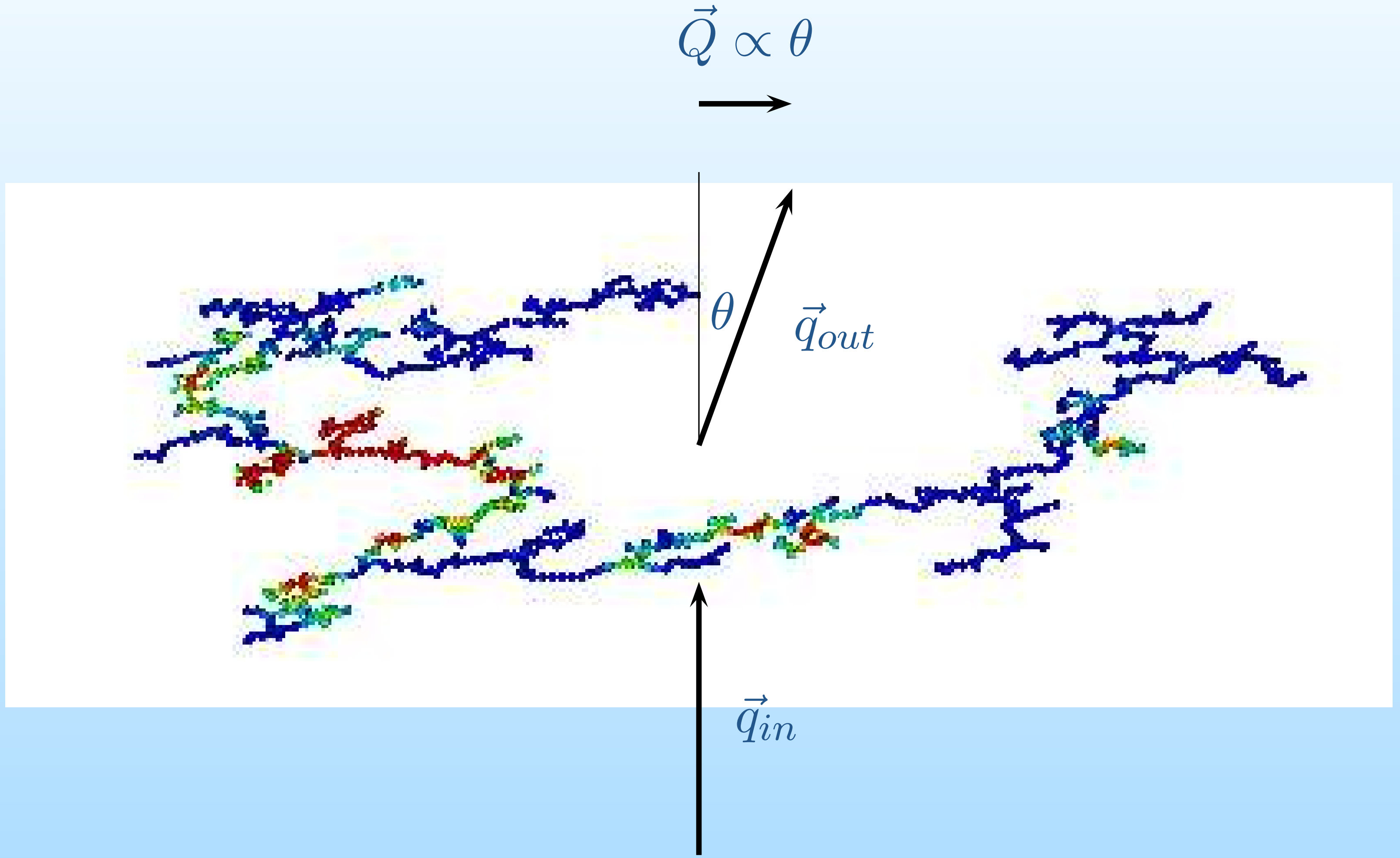
Interactions: screened Coulomb, depletion, fluctuation induced, surface induced,. . .

Interactions

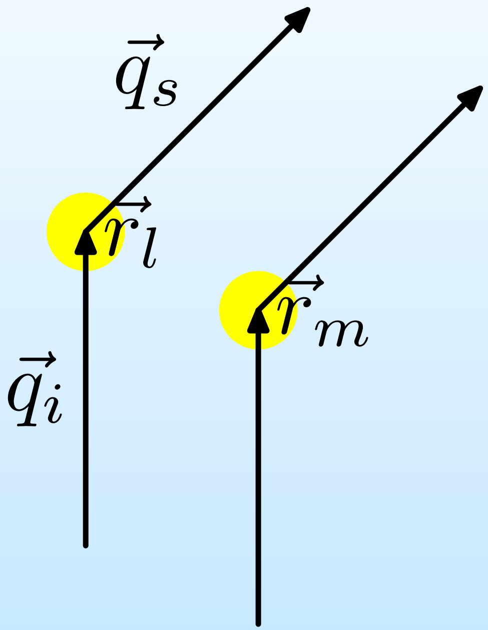


Controllable interaction: may be attractive, repulsive with a well, with two wells. . . ⇒ dispersion or different aggregation regimes

Scattering



Single scattering



$$\vec{E} \propto \sum_l e^{i\vec{q}_i \cdot \vec{r}_l} e^{i\vec{q}_s \cdot (\vec{r} - \vec{r}_l)} \propto \sum_l e^{-i\vec{Q} \cdot \vec{r}_l}$$

$\vec{Q} \equiv \vec{q}_s - \vec{q}_i \sim \theta$ (scattering wavevector)

$$\frac{d\sigma}{d\Omega} \propto \vec{S} \propto |E^2| \propto \sum_{lm} e^{-i[\vec{Q} \cdot (\vec{r}_l - \vec{r}_m)]}$$

$$= S(\vec{Q}) \equiv \mathcal{FT}[C(\vec{R})]$$

$$C(\vec{R}) \equiv \langle \rho(\vec{r}) \rho(\vec{r} + \vec{R}) \rangle$$

$$\rho(\vec{r}) = \sum_l \delta(\vec{r} - \vec{r}_l)$$

Scattering from a fractal

Correlation $C(\vec{r}) \propto r^{d_f - d}$
Differential scattering cross section

$$\begin{aligned}\frac{d\sigma}{d\Omega} &\propto S(\vec{Q}) \\ &= \int d^d r\, e^{i\vec{Q}\cdot\vec{r}} C(\vec{r}) \\ &\propto Q^{-d_f}\end{aligned}$$

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vs. Multiple Scattering

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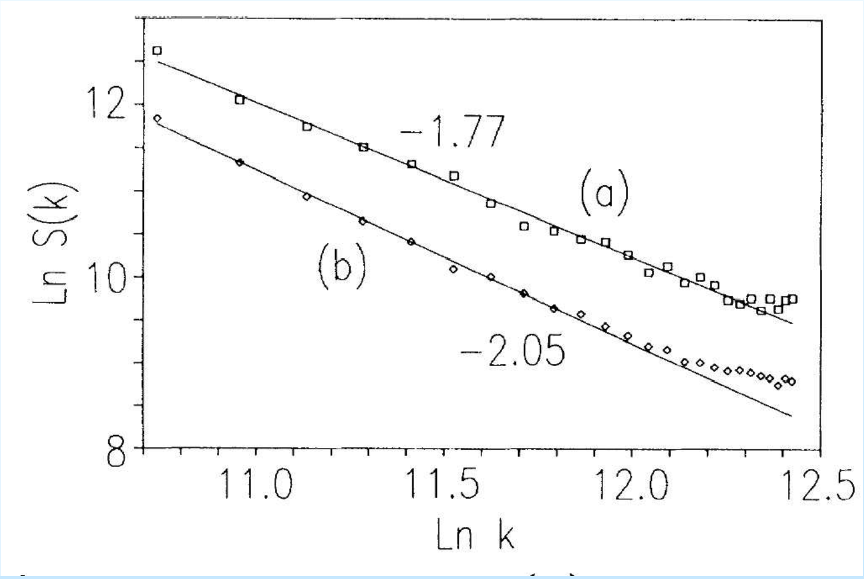
vs. Multiple Scattering

Weitz et al., PRL **54**, 1416 (1985) (scales)
Wilcoxon et al., PRL **58**, 1051 (1987) (doesn't)
Chen et al., PRB **37**, 5232 (1988) (does)

Wilcoxon et al., PRA **39**, 2675 (1989) (doesn't)

Experiments

- Weitz et al., PRL **54**, 1416 (1985) (scales)



d_f depends on kinetics

Experiments

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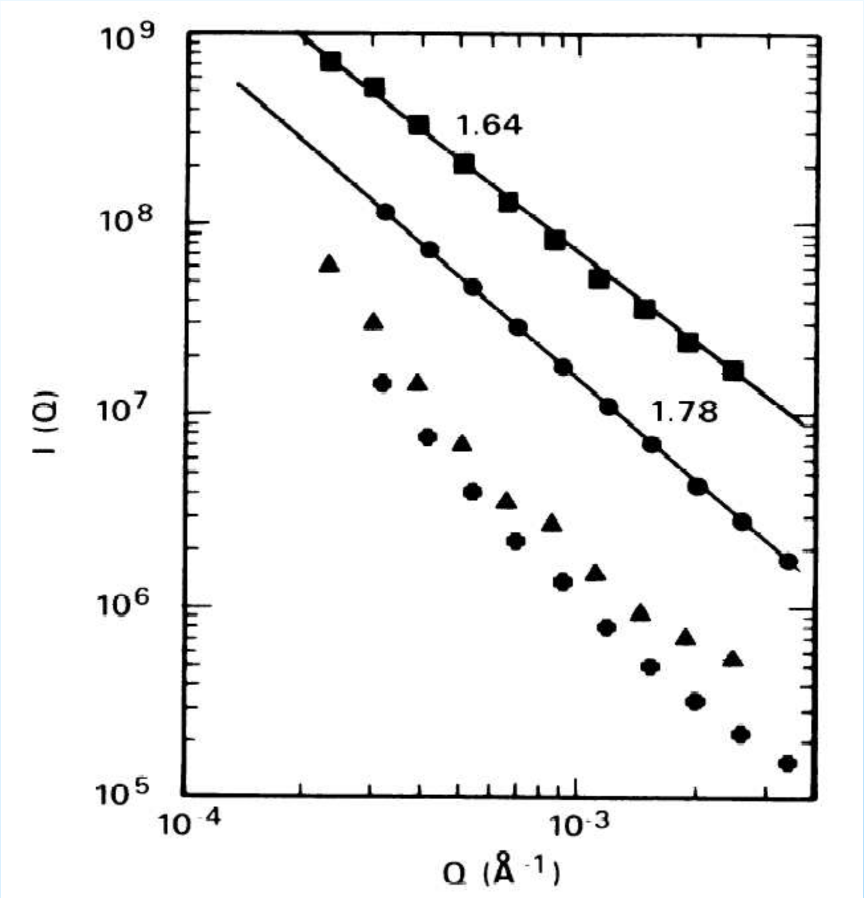
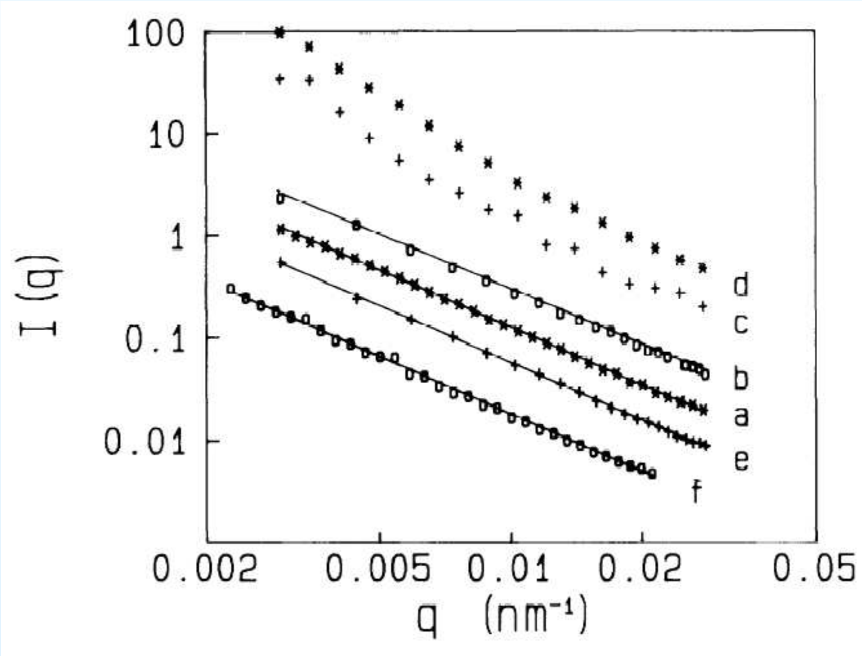
- Wilcoxon et al., PRL **58**, 1051 (1987) (doesn't)



d_f vs. wavelength? $\lambda = 632$nm, 457nm.

Experiments

- Weitz et al., PRL **54**, 1416 (1985) (scales)
- Wilcoxon et al., PRL **58**, 1051 (1987) (doesn't)
- Weitz et al. PRL **58**, 1052 (1988) (does)



scaling vs. dilution procedure

Experiments

- Weitz et al., PRL **54**, 1416 (1985) (scales)
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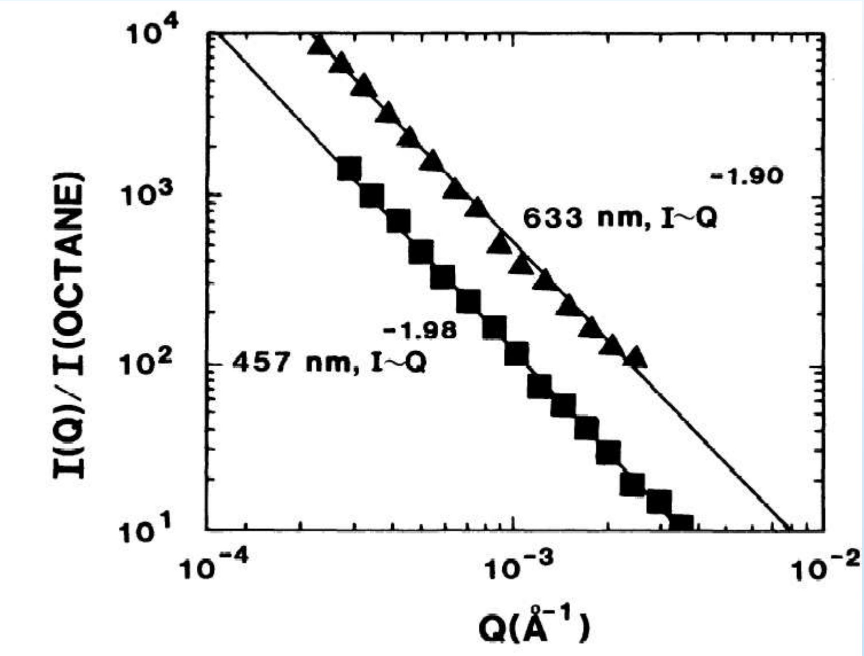

Multiple scattering: numerical calculation

$$\vec{p}_i e^{i\vec{q}_{in}\cdot\vec{r}_i} = \gamma \left(\vec{E}_0 e^{i\vec{q}_{in}\cdot\vec{r}_i} + \sum_j \overleftrightarrow{T}_{ij} \cdot \vec{p}_j e^{i\vec{q}_{in}\cdot\vec{r}_j} \right)$$

Scattered field

$$\vec{E}^{out}(\vec{Q}) \propto \vec{p}^{\,T}(\vec{Q}) = \sum_i \vec{p}_i^{\,T} e^{-i\vec{Q}\cdot\vec{r}_i}$$

Scattering cross section

$$\frac{d\sigma}{d\Omega} \propto |\vec{p}^{\,T}(\vec{Q})|^2$$

Does it scale at resonance?

Difficulties

- Many body system (N)
- Long range interactions (N^2)

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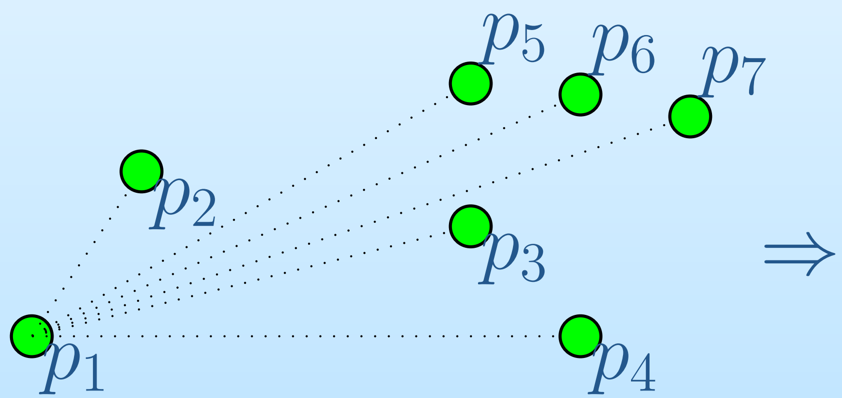
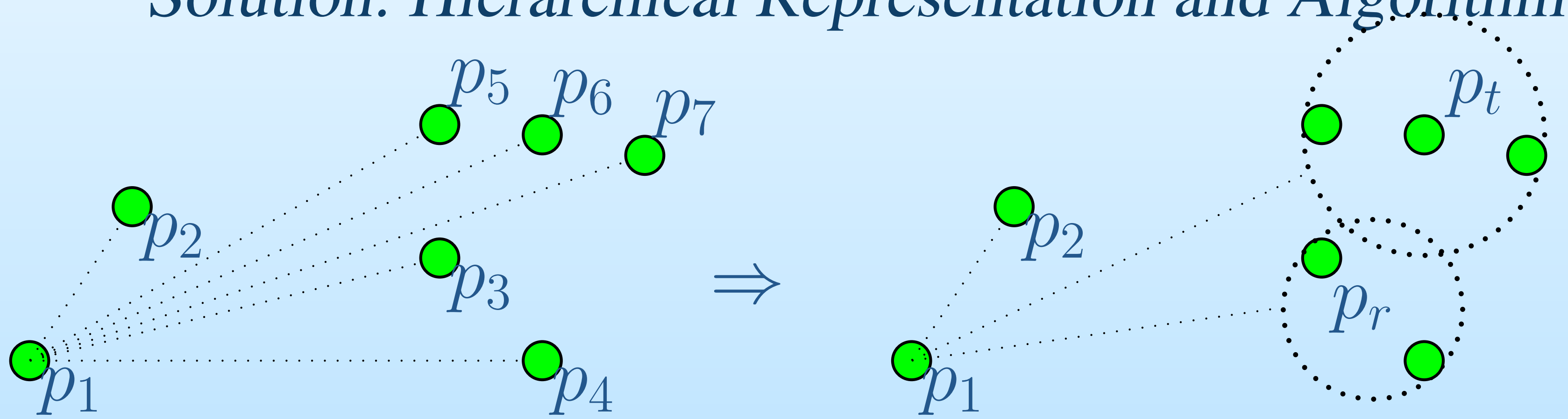


Difficulties

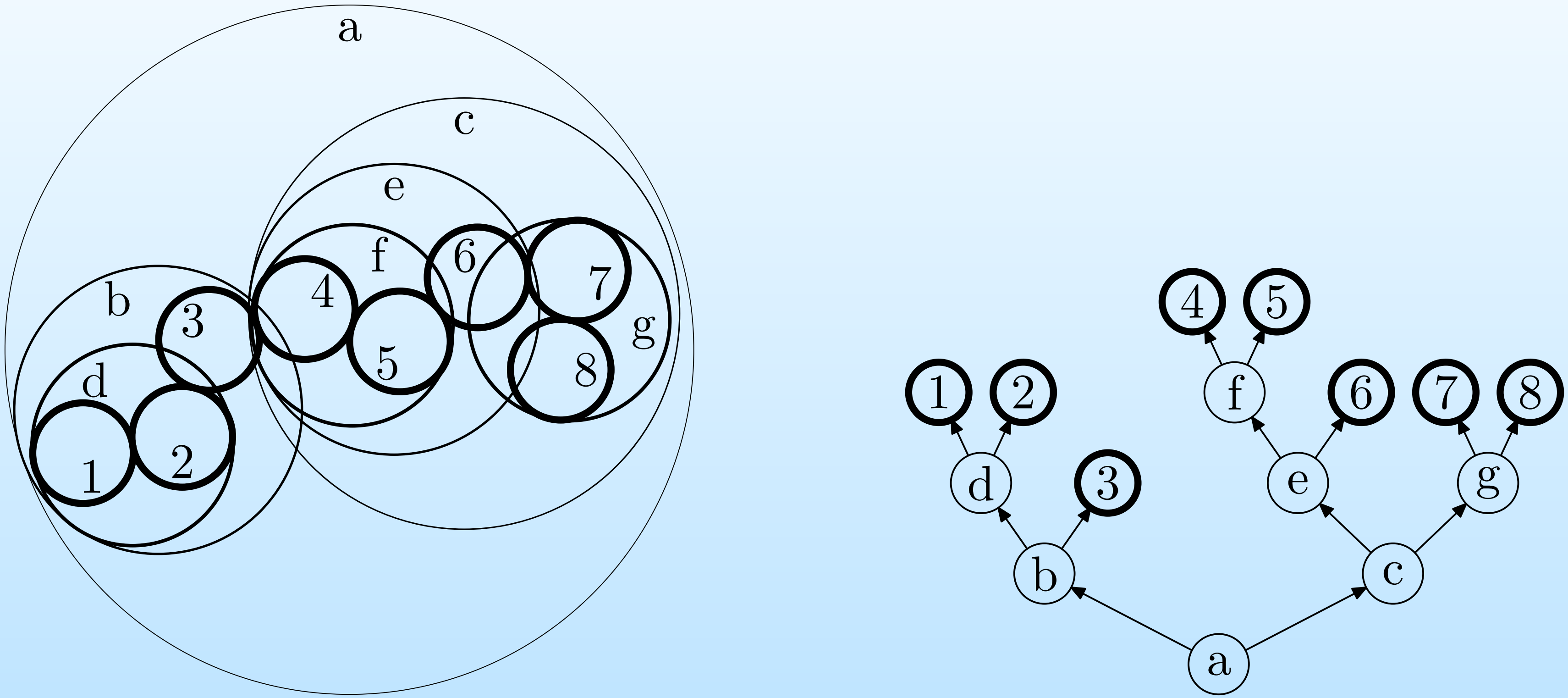
- Many body system (N)
- Long range interactions (N^2)

Solution: Hierarchical Representation and Algorithm



Interaction with a *pseudo-particle* replaces many inter-particle interactions.

Hierarchical Representation



Test System

- Diffusion limited cluster-cluster colloidal aggregate.
- 2D.
- Scalar approximation.
- Long wavelength external field.
- Non-retarded dipole-dipole interactions.

Hierarchy preparation

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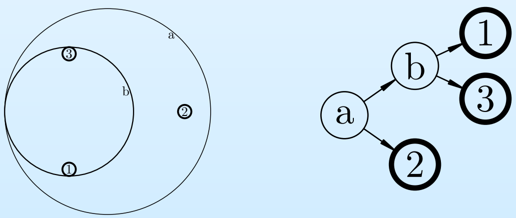
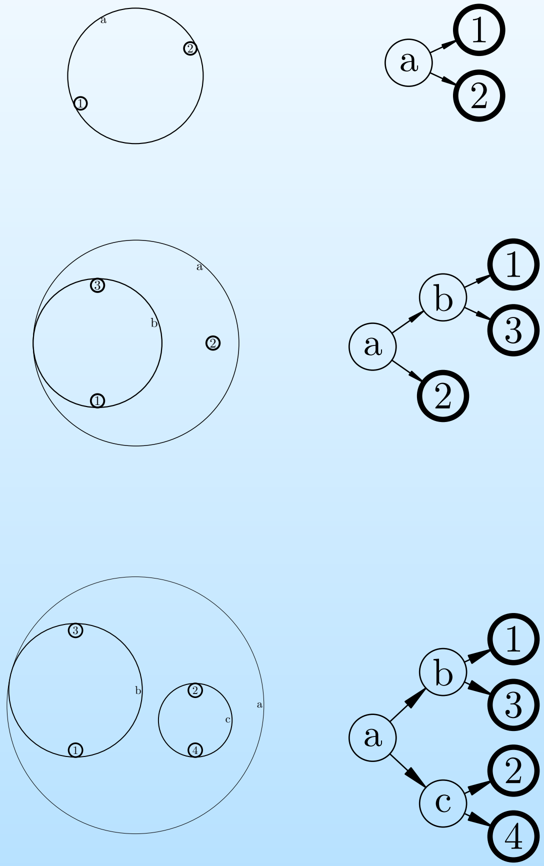
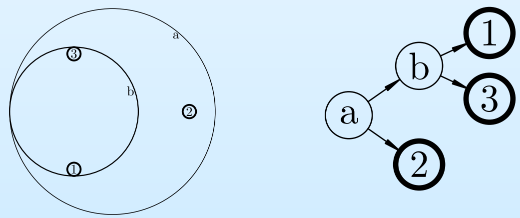
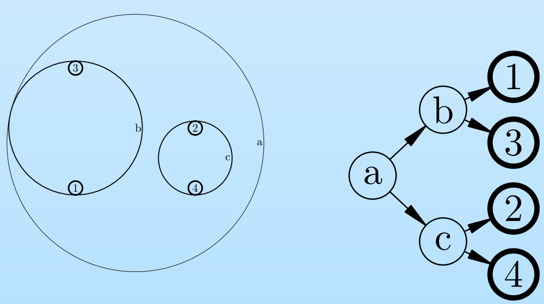


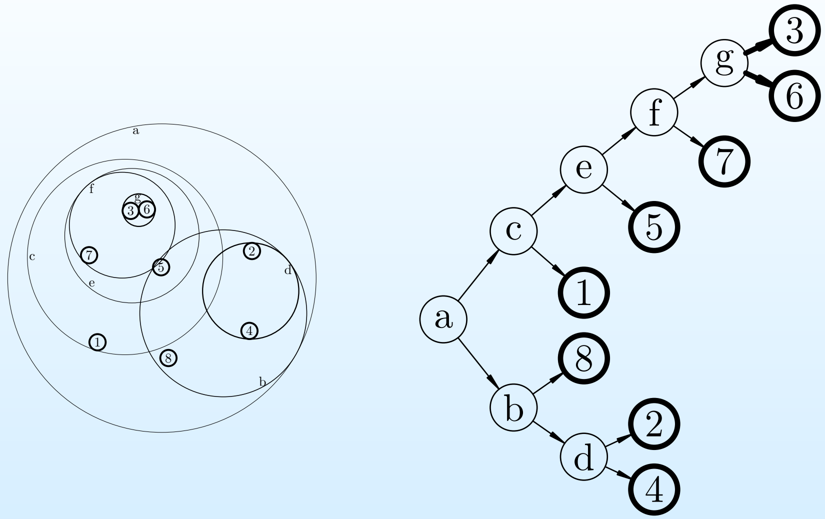
Hierarchy preparation



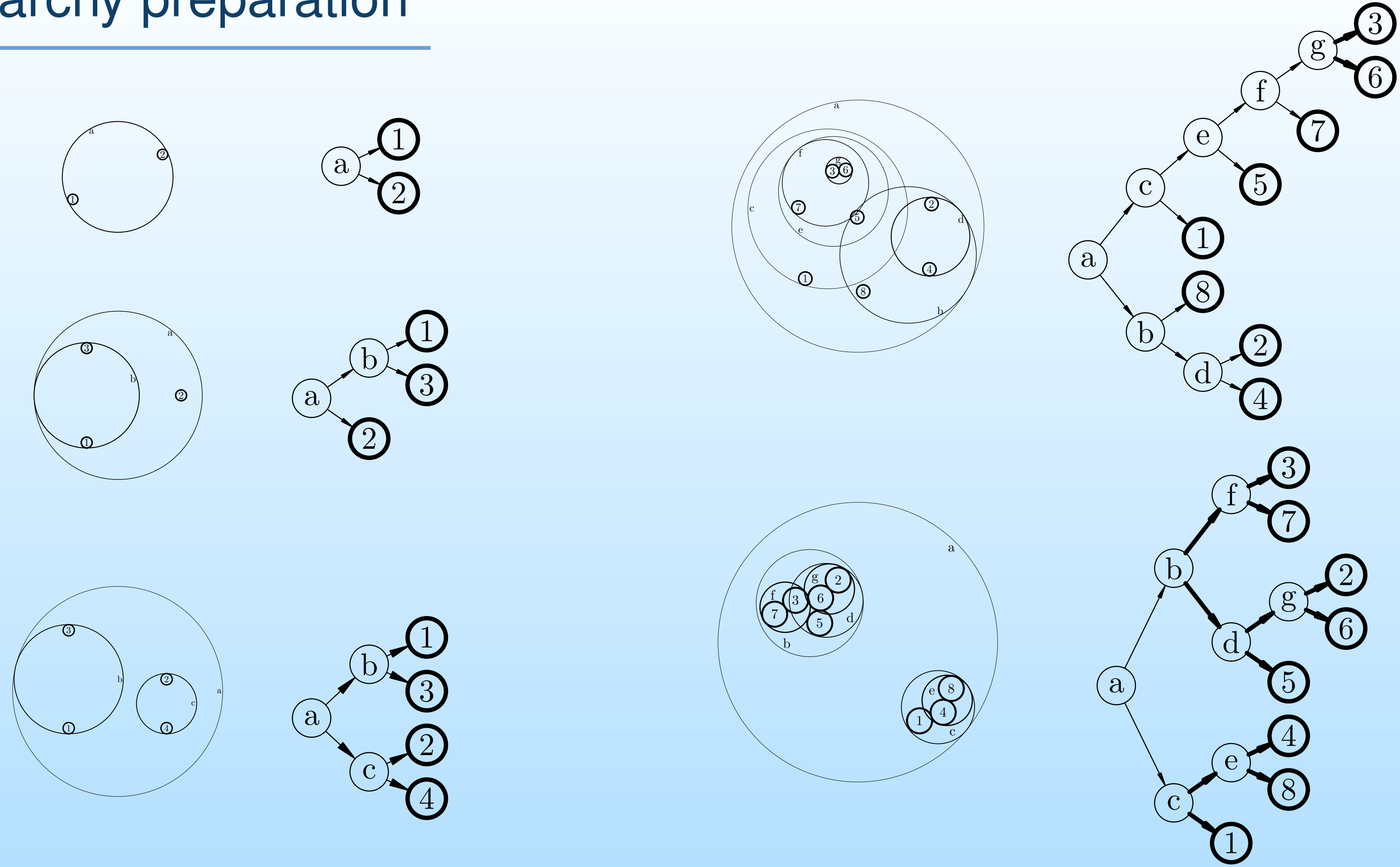
Hierarchy preparation



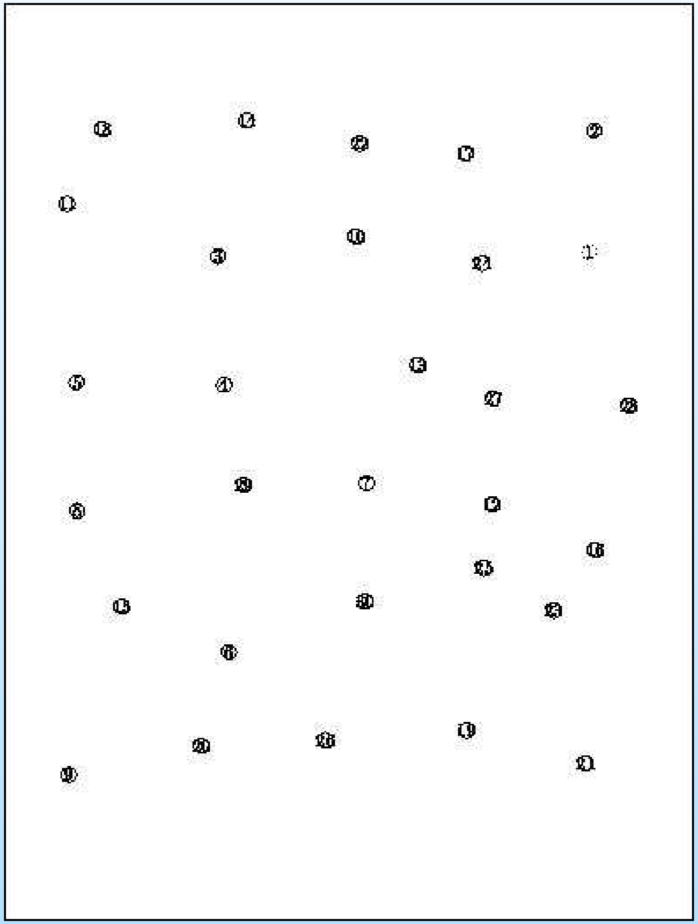




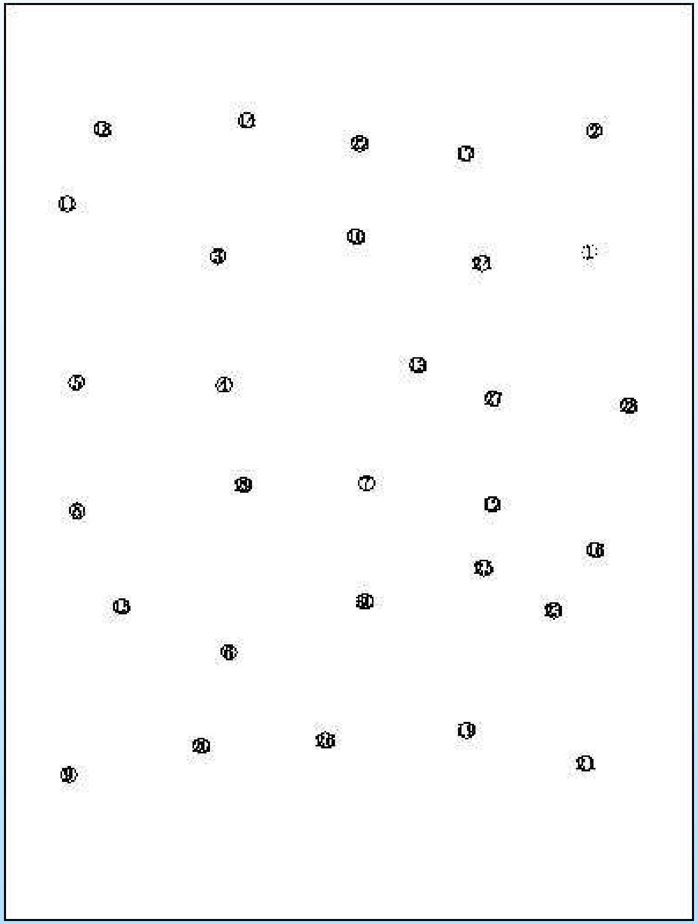
Hierarchy preparation

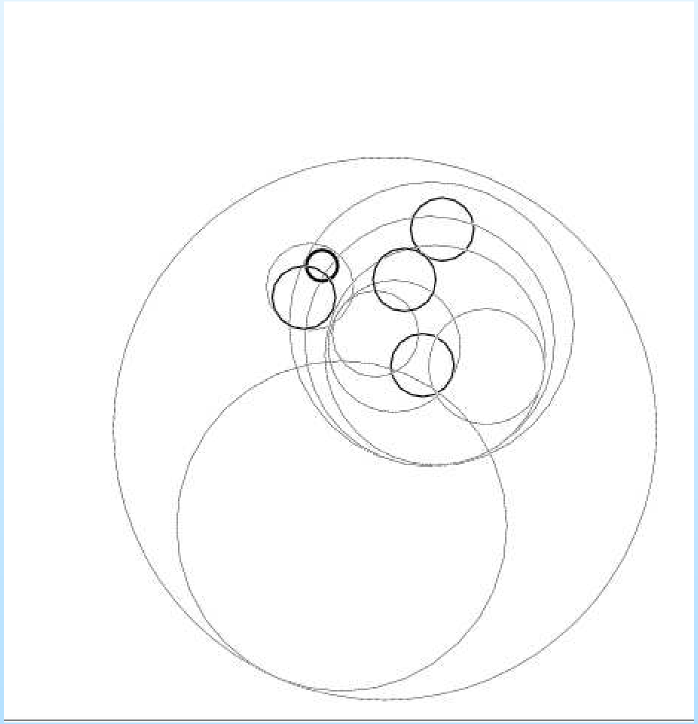


Cluster Generation



Cluster Generation





Polarizing field

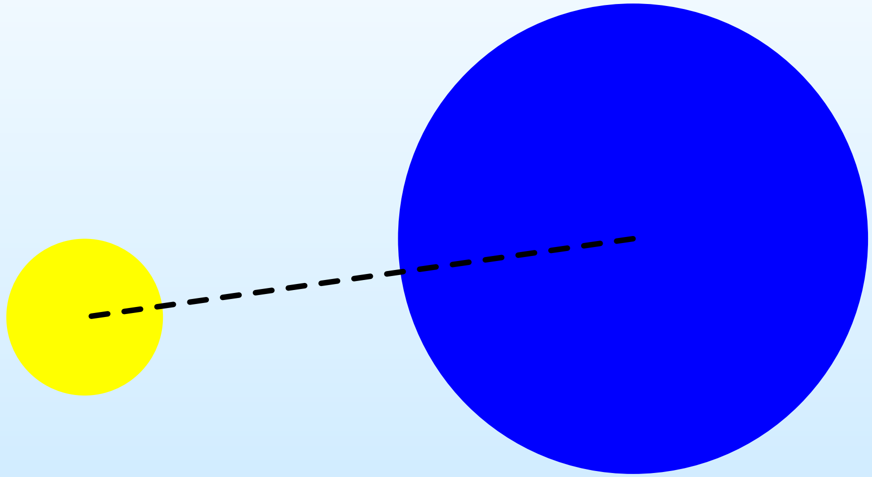
$$\vec{p}_i = \gamma \left(\vec{E}_0 + \sum_j \overleftrightarrow{T}_{ij} \cdot \vec{p}_j \right)$$
$$= \gamma \left(\vec{E}_0 + \vec{E}_{ir} \right)$$

$\vec{E}_{ir}$ is the field at i due to the whole system, represented by its root r.

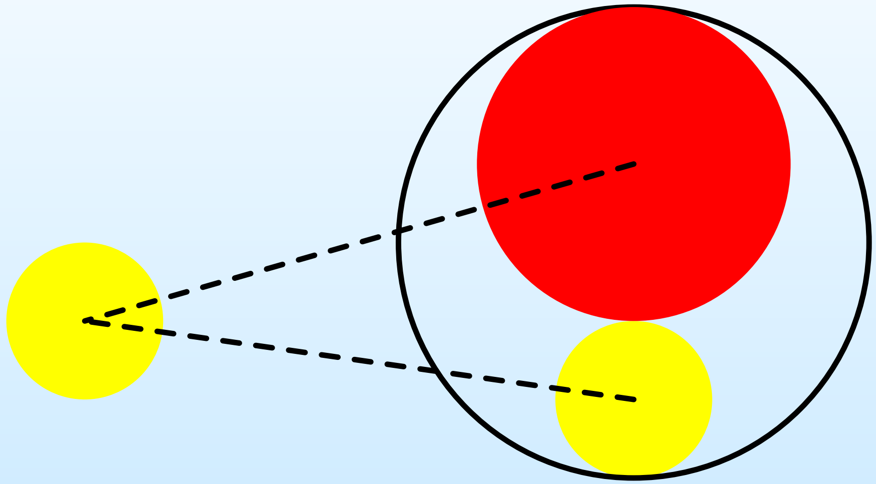
Recursive evaluation

$$\vec{E}_{i\zeta} = \begin{cases} \overleftrightarrow{T}_{i\zeta} \cdot \vec{p}_\zeta & \text{if } R_\zeta \text{ is } \textit{small} \\ E_{i\zeta_d} + E_{i\zeta_s} & \text{otherwise} \end{cases}$$

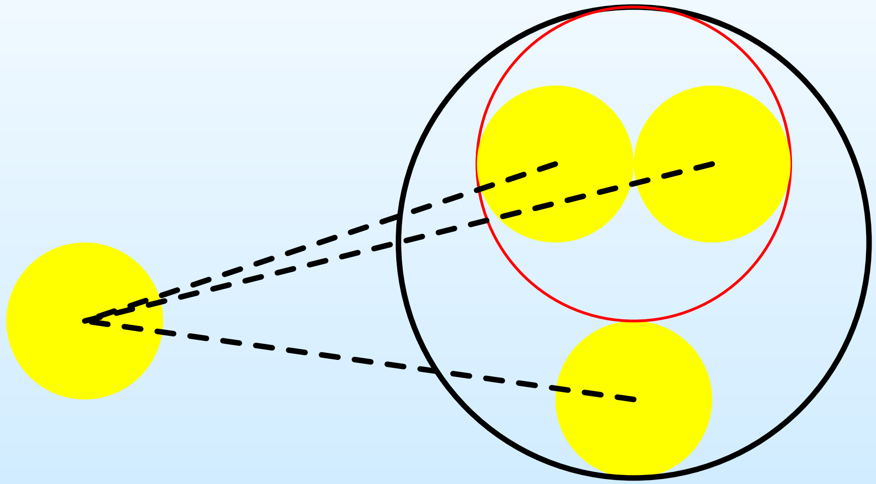
Recursive evaluation



Recursive evaluation



Recursive evaluation



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Induced polarization

Iterative Solution (small γ)

$$\vec{p}_i^{\,n+1} = \gamma(\vec{E}_0 + \vec{E}_{ir}^{\,n})$$

Full Solution (arbitrary γ)

$$\mathbf{Mp} \equiv \left(\frac{1}{\gamma}\mathbf{1} - \mathbf{T}\right)\mathbf{p} = \mathbf{E_0}$$

$$\mathbf{M}'\mathbf{p}' \equiv \left[\begin{pmatrix} \mathbf{M} & \mathbf{0} \\ \mathbf{0} & \mathbf{1} \end{pmatrix} - \begin{pmatrix} \mathbf{0} \\ \mathbf{C} \end{pmatrix}\right]\mathbf{p}' = \begin{pmatrix} \mathbf{E}_0 \\ \mathbf{0} \end{pmatrix} \equiv (\mathbf{E}_0)',$$

Make $\mathbf{M}'$ sparse, by replacing many interactions with far away particles with a single interaction with the containing group.

Susceptibility

$$\vec{P} = \vec{p}_r \equiv \sum_i \vec{p}_i \equiv V \overleftrightarrow{\chi} \vec{E}_0$$

Spectral variable: $u \equiv 1/(1 - \epsilon/\epsilon_h)$, $1/\gamma = (1 - 3u)/R_0^3$

$$|p\rangle = \left[(1/\gamma)\mathbf{1} - \mathbf{T}\right]^{-1} |E_0\rangle$$

$$P = \sqrt{N}\langle 0|p\rangle$$

$$\chi = \frac{3}{4\pi}\langle 0| \left[(1 - 3u)\mathbf{1} - R_0^3\mathbf{T}\right]^{-1} |0\rangle = \frac{3}{4\pi}\sum_n \frac{\langle 0|t_n\rangle\langle t_n|0\rangle}{1 - 3u - R_0^3 t_n}$$

Normal mode analysis

- Spectral variable u,

$$u = (\gamma - 1)/3\gamma.$$

- Total dipole P,

$$P = \sum_i p_i$$

- Spectral function $g(s)$,

$$P(u) = \frac{NR^3E}{3} \int_0^1 ds \frac{g(s)}{s-u}.$$

- $g(s)$ depends **only on geometry** and not on material properties.
- u depends on material properties and on frequency. For simple metals, $u = \omega^2/\omega_p^2$.

Strategy

- $g(s)$ is related to a projected density of states,

$$g(s) \propto \mathrm{Im}\langle 0|\hat{G}(s)|0\rangle.$$

  where

$$\hat{G}(s) = \left[s - \left(\hat{T} - \frac{1}{3} \right) \right]$$

  plays the role of a Green's function,

$$|u\rangle = (1, 1, 1, \ldots)/\sqrt{N}.$$

- Thus, $g(s)$ might be calculated with the recursive Haydock method (R. Haydock, Solid State Physics **35**, 1980):

Haydock's method

- Generate orthogonal basis so that the interaction $\hat{T}$ becomes tridiagonal $|u_k\rangle \rightarrow |u_{k+1}\rangle = \hat{T}|u_k\rangle$ + orthonormalization,

$$|u_{k+1}\rangle = \hat{T}|u_k\rangle - a_k|u_k\rangle - b_k^2|u_{k-1}\rangle$$

with diagonal elements

$$a_k = \frac{\langle u_k|\hat{T}|u_k\rangle}{\langle u_k|u_k\rangle},$$

and superdiagonal elements

$$b_k^2 = \frac{\langle u_k|u_k\rangle}{\langle u_{k-1}|u_{k-1}\rangle}.$$

Continued fraction

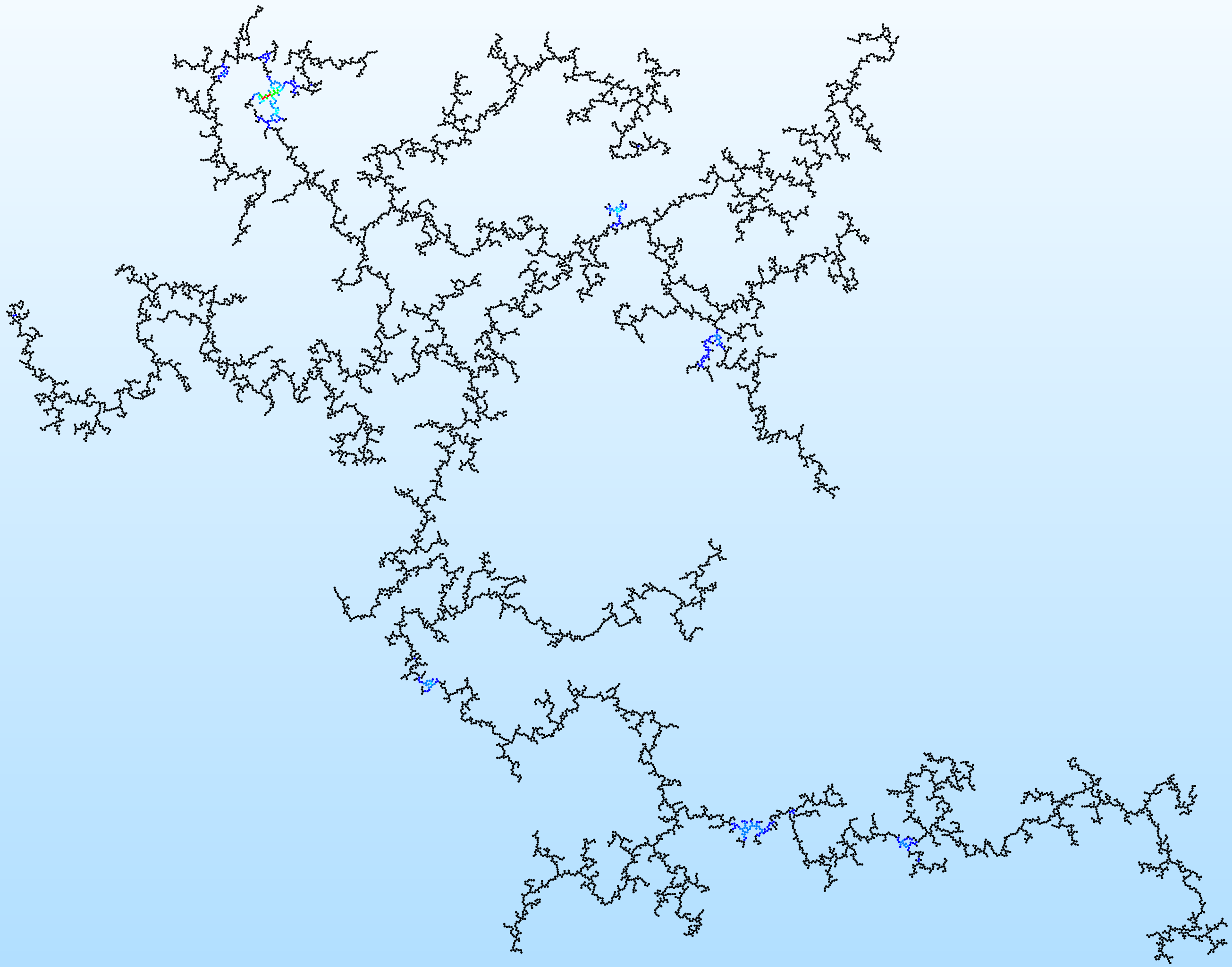
- Employ hierarchical representation to calculate $\langle u_k|\hat{T}|u_k\rangle$.
- From tridiagonal matrix, generate continued fraction for the Green's function,

$$
\begin{aligned}
P(u) &= NR^3E\langle u_0|\left(1-3\,u-\hat{T}\,R^3\right)^{-1}|u_0\rangle \\
&= \left\{\frac{NR^3E}{1-3u-a_0-\frac{b_1^2}{1-3u-a_1-\frac{b_2^2}{1-3u-a_2-\dots}}}\right\},
\end{aligned}
$$

- and finally,

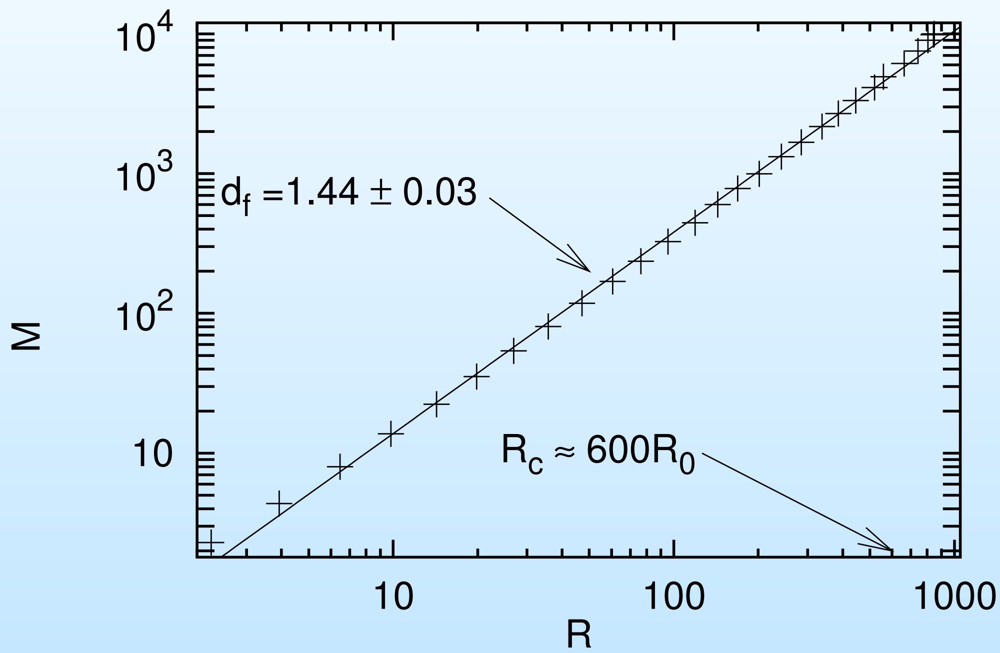
$$
g(s)=\frac{3}{\pi NR^3E}\,\mathrm{Im}\{P(s+i0^+)\}.
$$

2D-DLCA

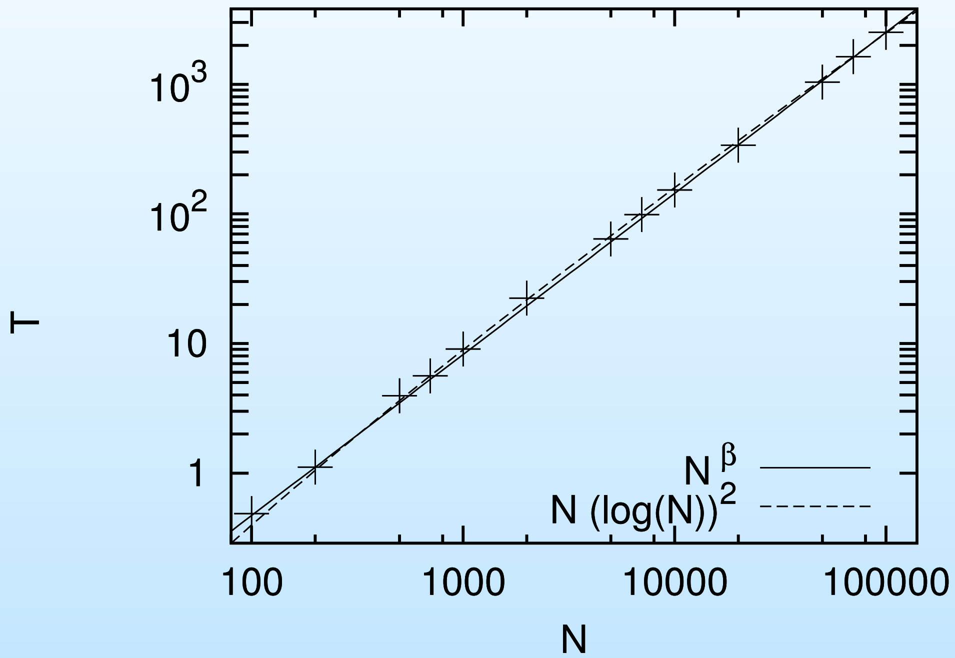


10^4 parts.

Fractal dimension



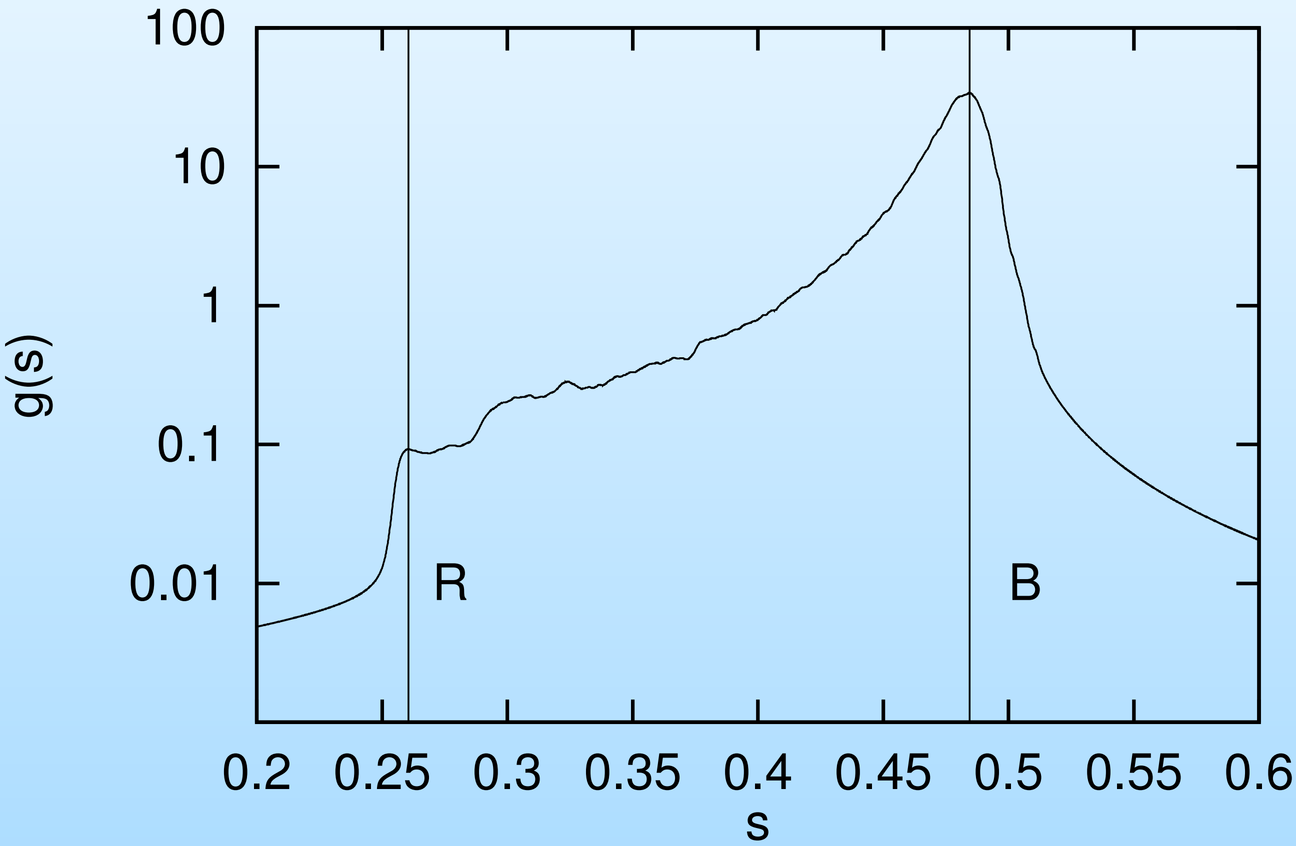
Time vs. size



Almost linear. . .

Spectral function

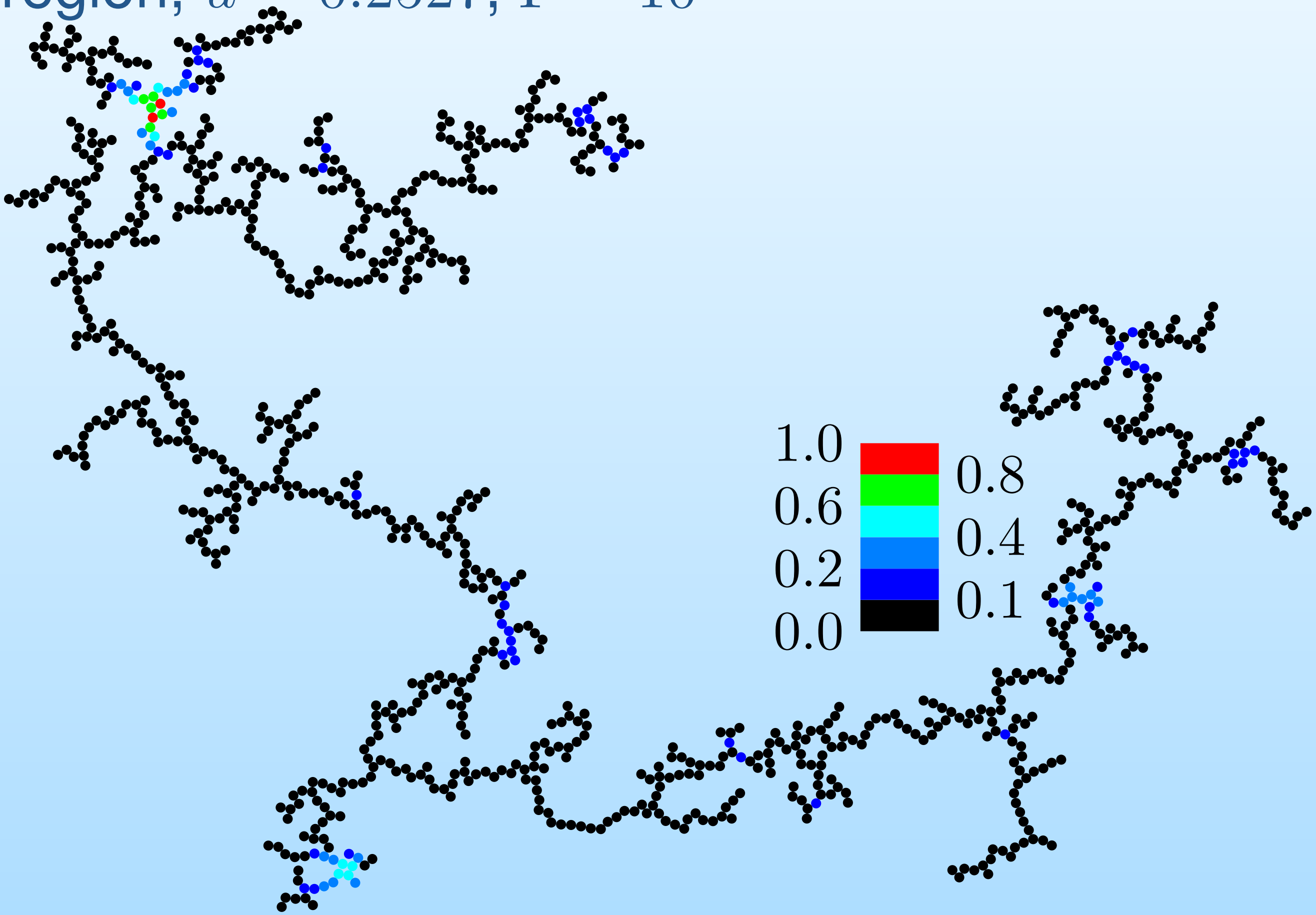
Spectral function g(s) for an ensemble of 10^4 particle DLCA cluster. Scalar model ($\vec{E}$ $\perp$plane)



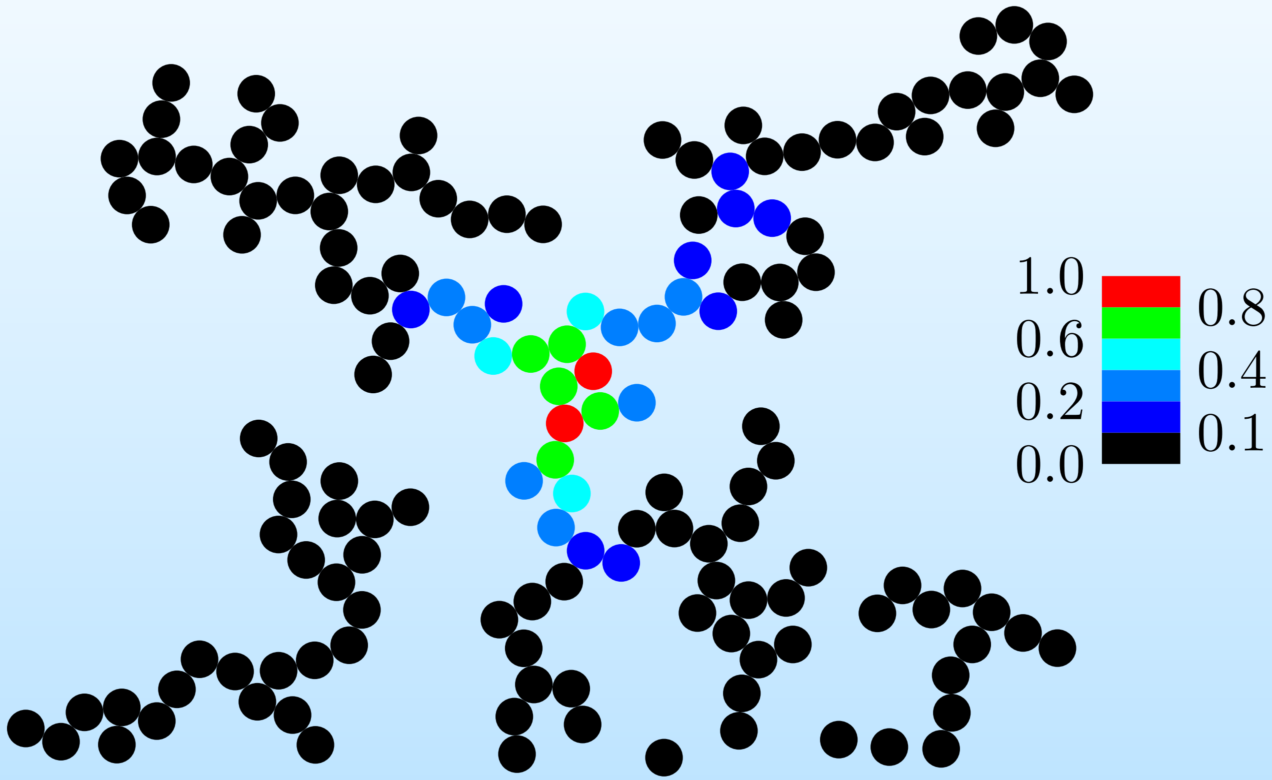
go shift vs. phase

Hot Spots

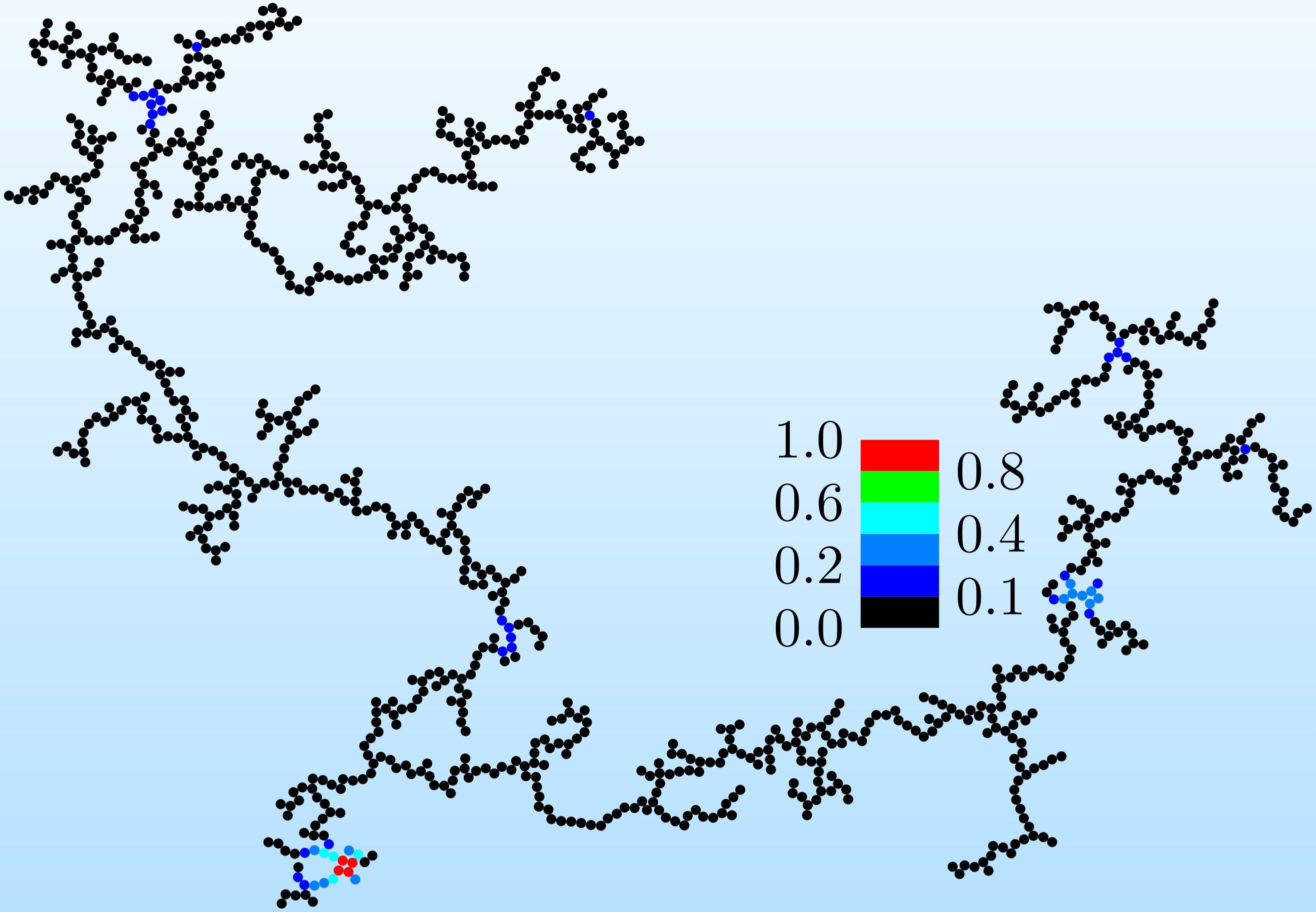
Local dipoles for a 10^3 particle DLCA aggregate at resonance in the red region, $u = 0.2527$, $\Gamma = 10^{-4}$



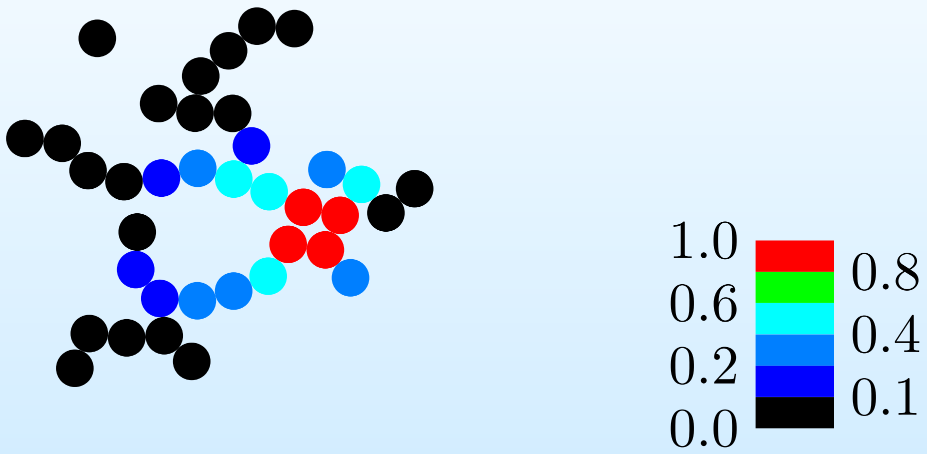
Detail for $u = 0.2527$



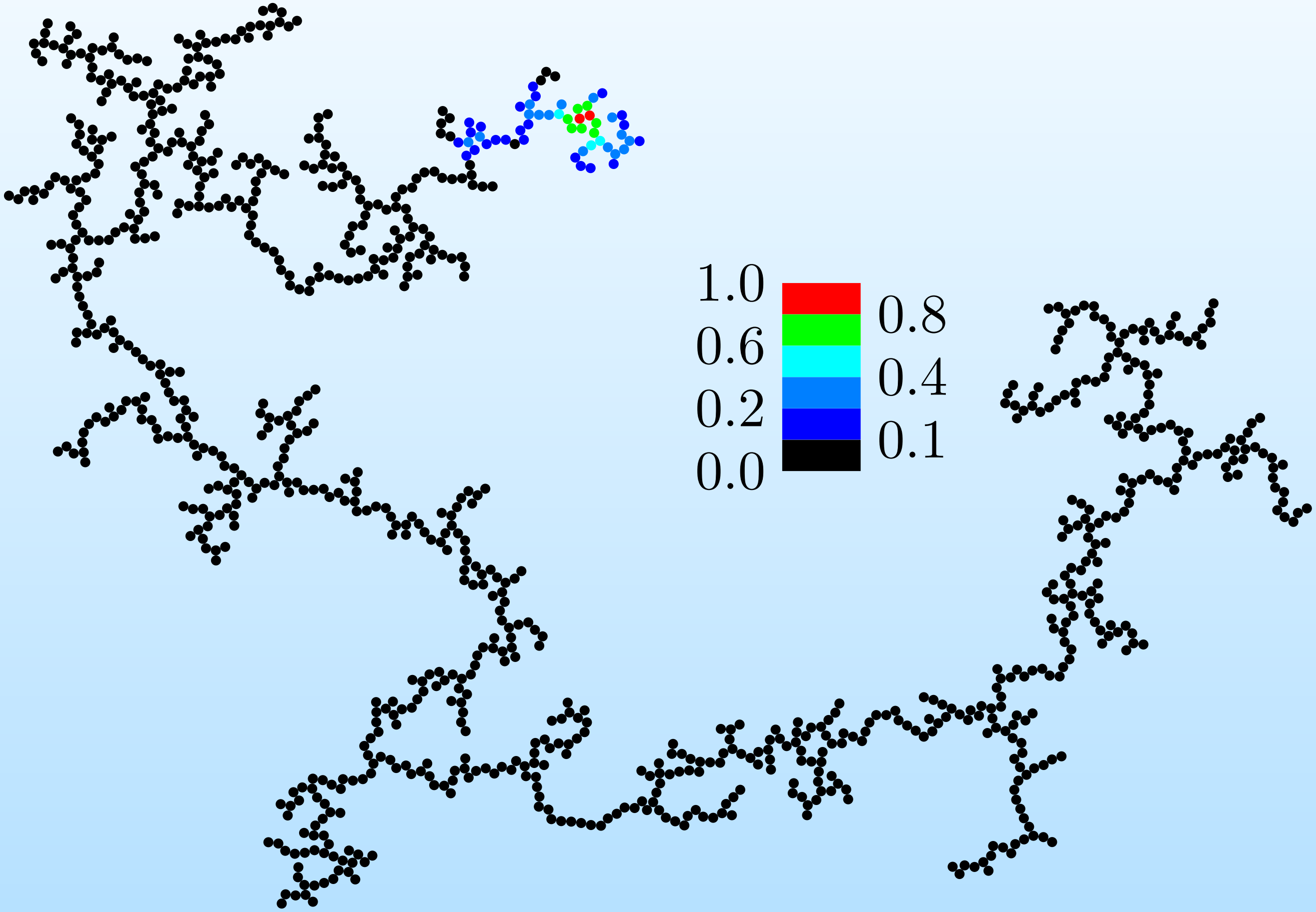
Next resonance, $u = .25303$



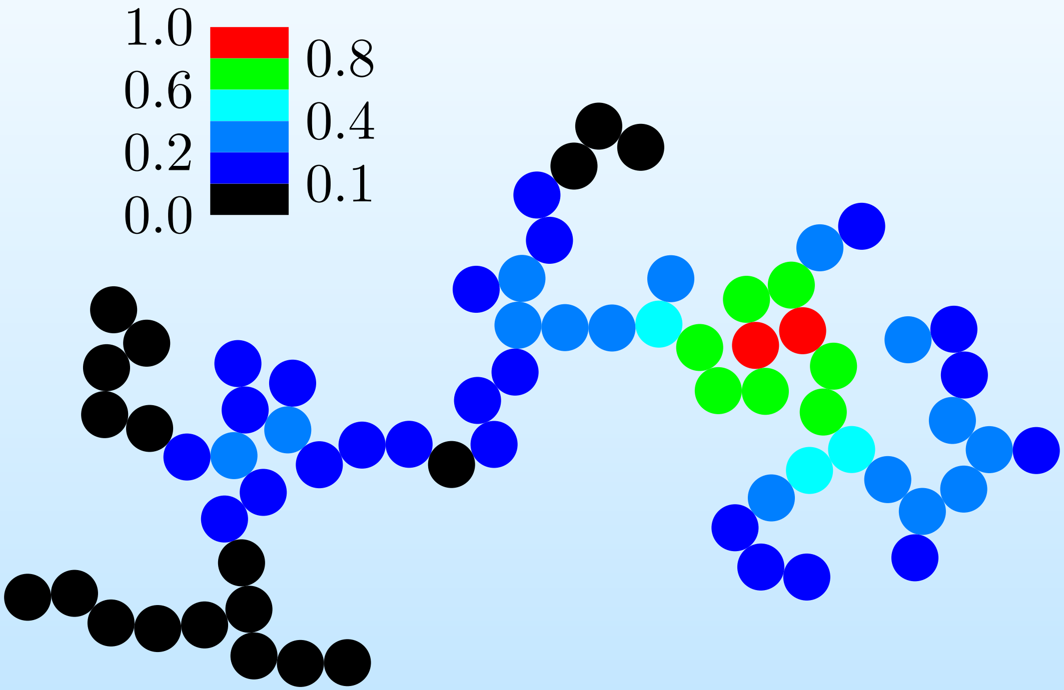
Detail for $u = .25303$



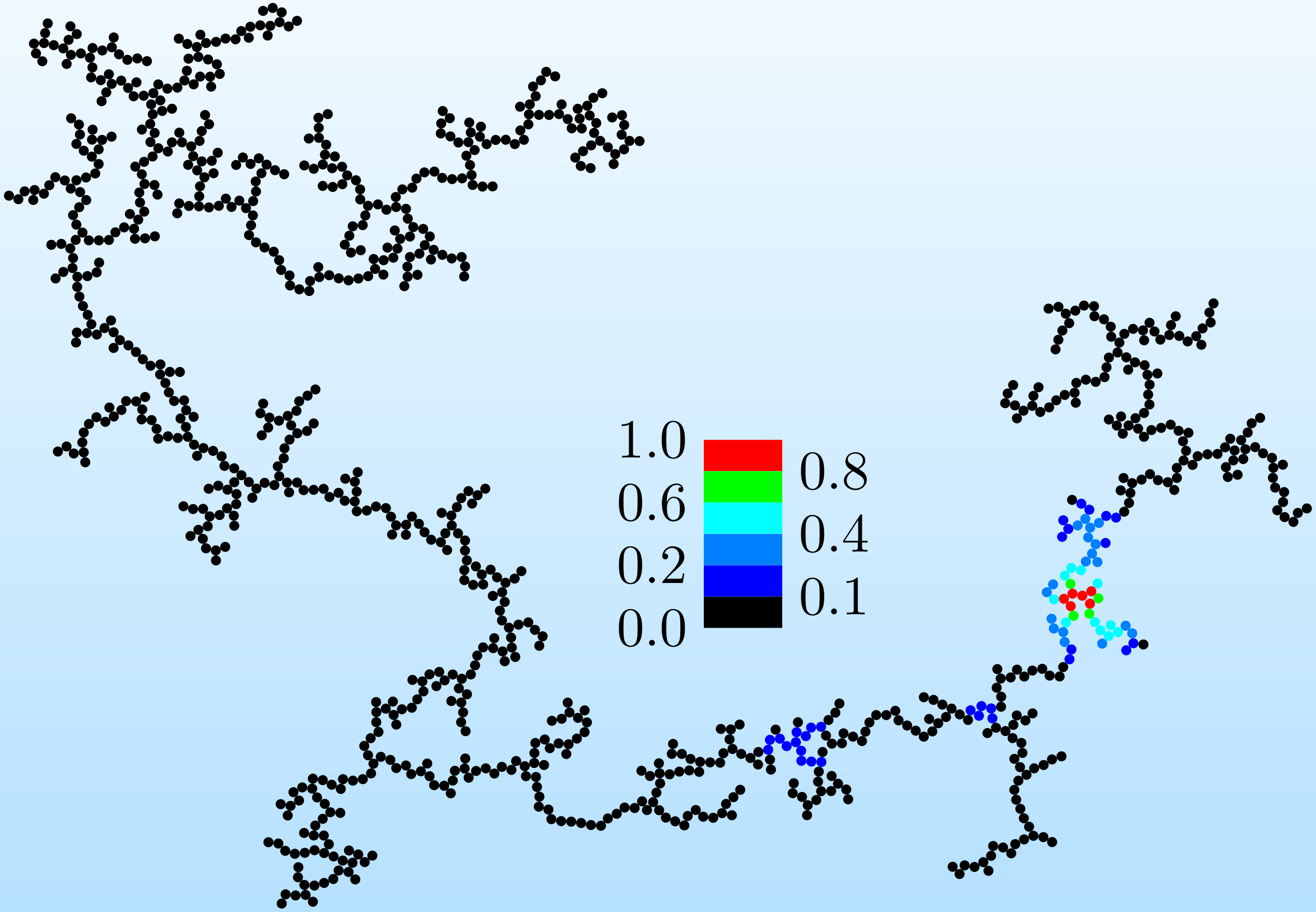
Blue resonance, $u = 0.4887$



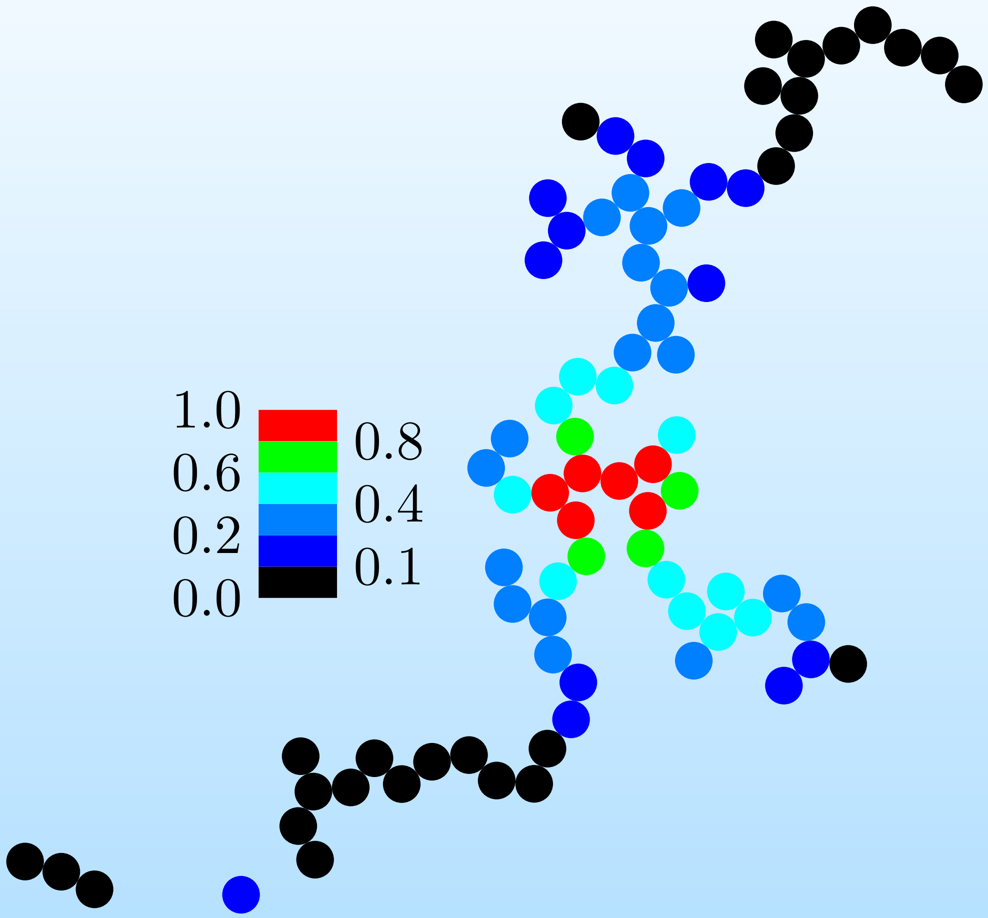
Detail for $u = 0.4887$



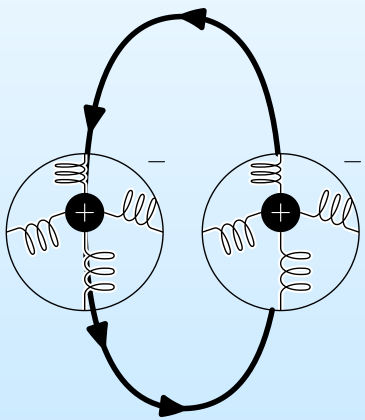
Next resonance, $u = .4865$



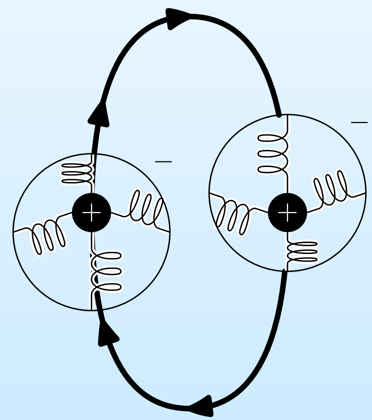
Detail for $u = .4865$



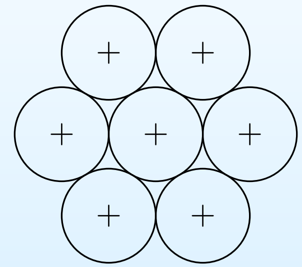
Shift vs. Phase



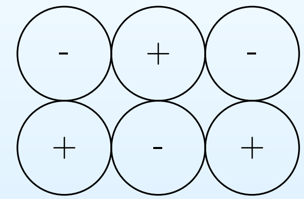
Blue



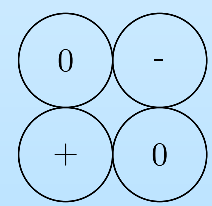
Red



$s = 0.50$



$s = 0.24$



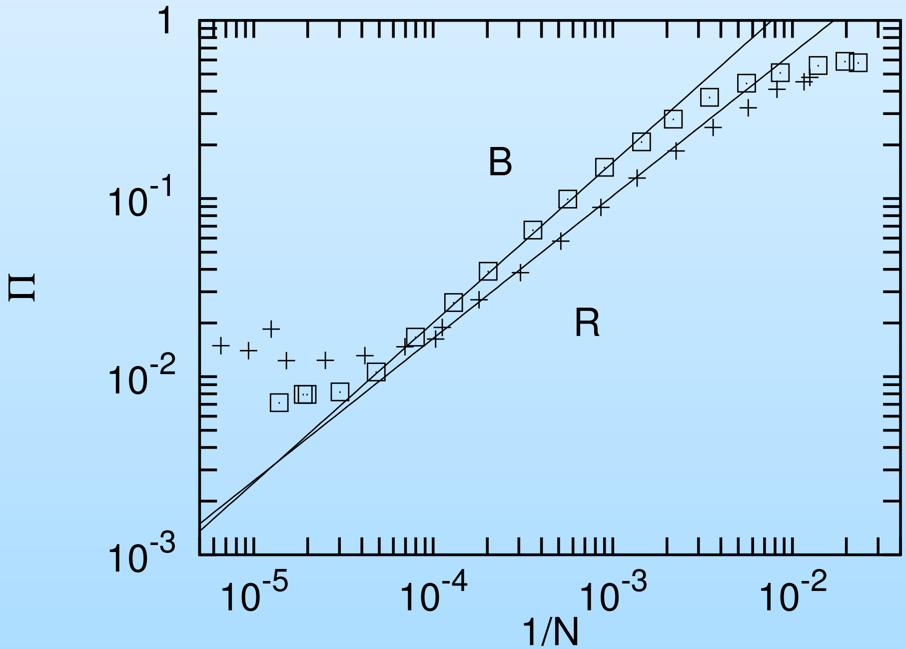
$s = 0.32$

Go $g(s)$

Localization

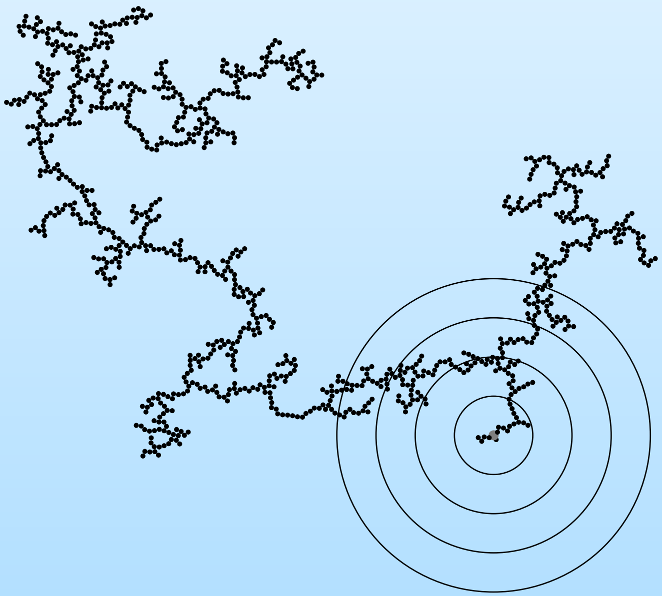
Characterized by the participation ratio

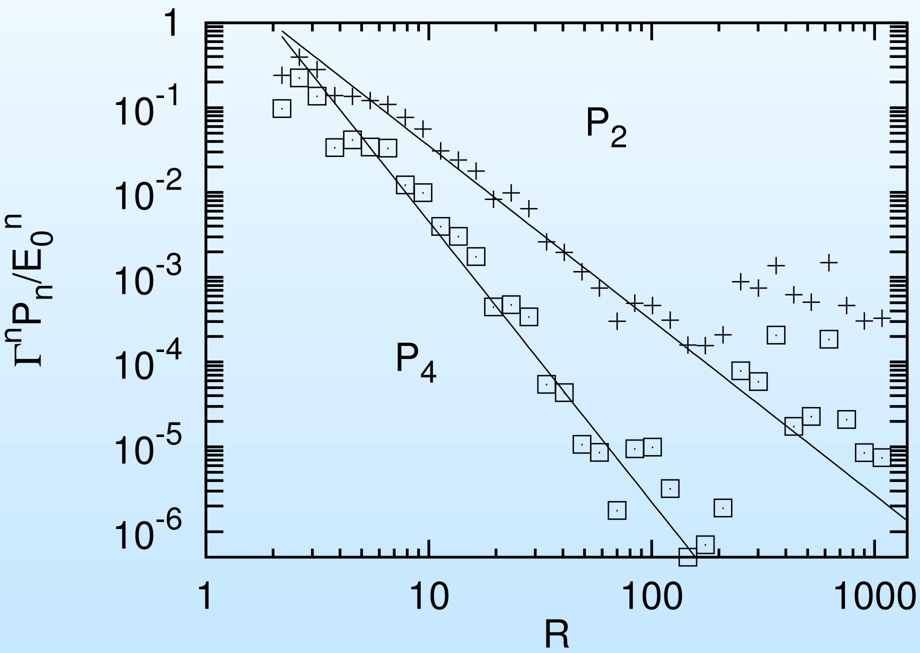
$$PR = \frac{1}{N}\frac{\left(\sum |p_i|^2\right)^2}{\left(\sum |p_i|^4\right)} \rightarrow \begin{cases} 1 & \text{Extended} \\ 1/N & \text{Localized} \end{cases}$$



Critical states
$PR \propto N^{-\beta}$
$\beta = 0.9$ (B),
$\beta = 0.8$ (R)

Decay of Polarization

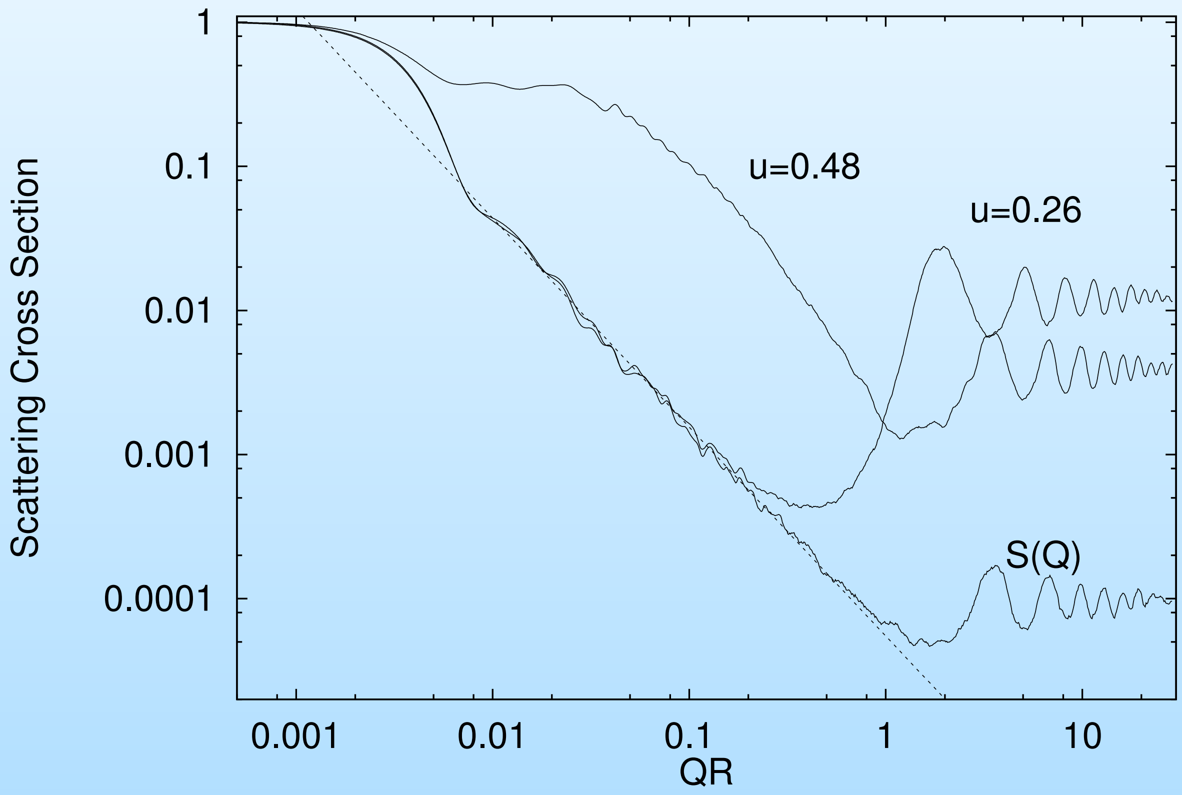




$$\left. \begin{aligned} P_2 &\propto R^{-2.06} \\ P_4 &\propto R^{-3.31} \end{aligned} \right\} \Longrightarrow \Pi \propto 1/N$$

Scattering cross section

10^4 particle DLCA cluster, $\Gamma = 10^{-4}$



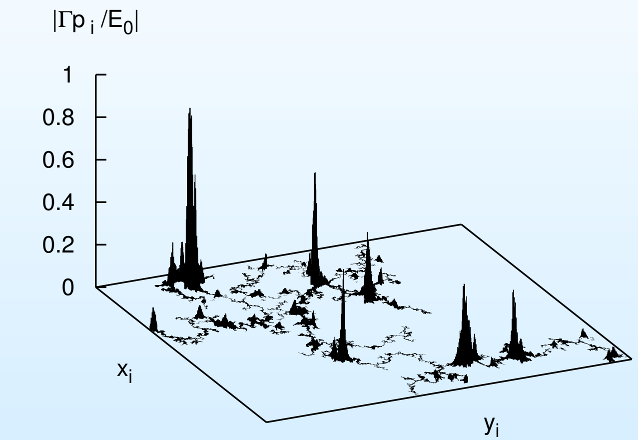
Scaling?

- The cross section seems to scale for red resonances
- and not to scale for blue resonances,

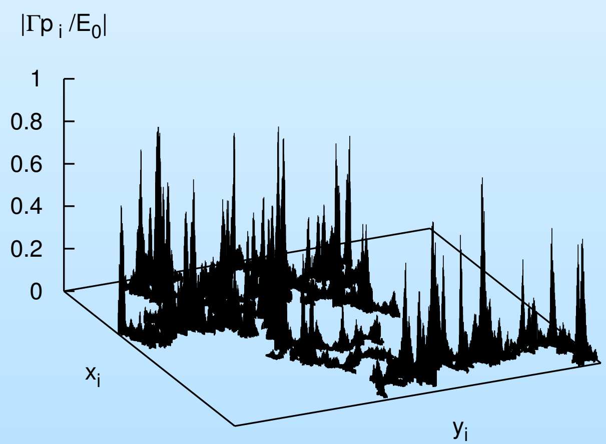
Scaling?

- The cross section seems to scale for red resonances
- and not to scale for blue resonances,
- BUT
- finite disipation leads to simultaneous excitation of neighboring modes.
- Hot spots of a spectral region have a given local geometry and form a fractal with the same dimension d_f as the full system.
- Thus, scaling ought to be recovered for an infinite system or for a larger Γ!

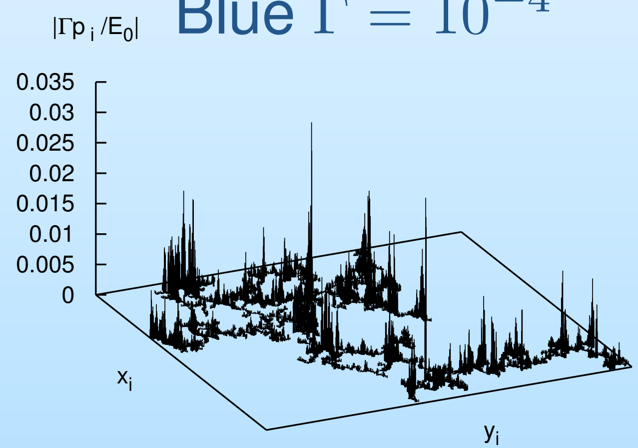
Induced Dipole Moments



Blue $\Gamma = 10^{-4}$



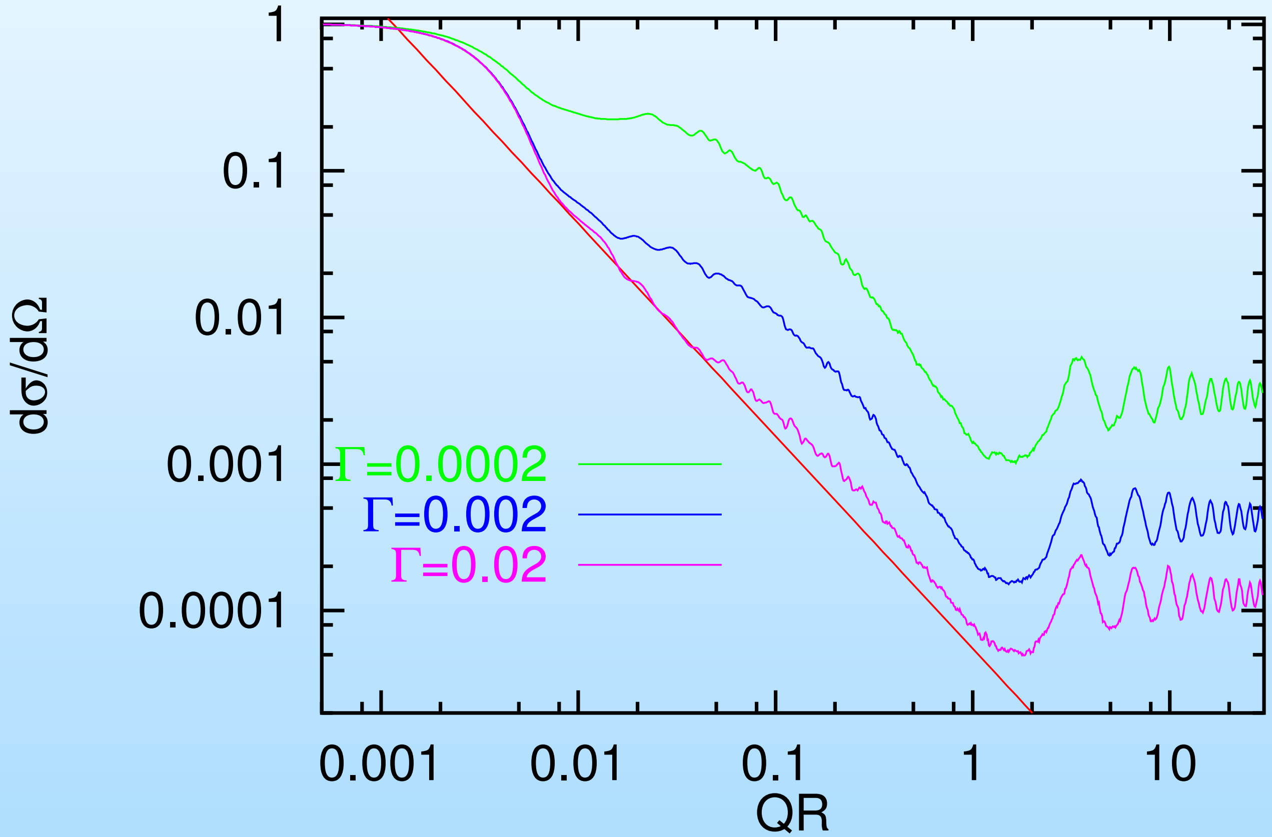
Blue $\Gamma = 2 \times 10^{-3}$



Red $\Gamma = 10^{-4}$

Scaling Recovered

10^4 particle DLCA cluster, $u = 0.48$



Hot Spot Scaling

- Number of hot spots $N_h \approx$ number of excited modes,

$$N_h \propto g\, N\, \Gamma.$$

- Hot spot distance scale,

$$N_h(r) \propto \left(\frac{r}{L_h}\right)^{d_f}.$$

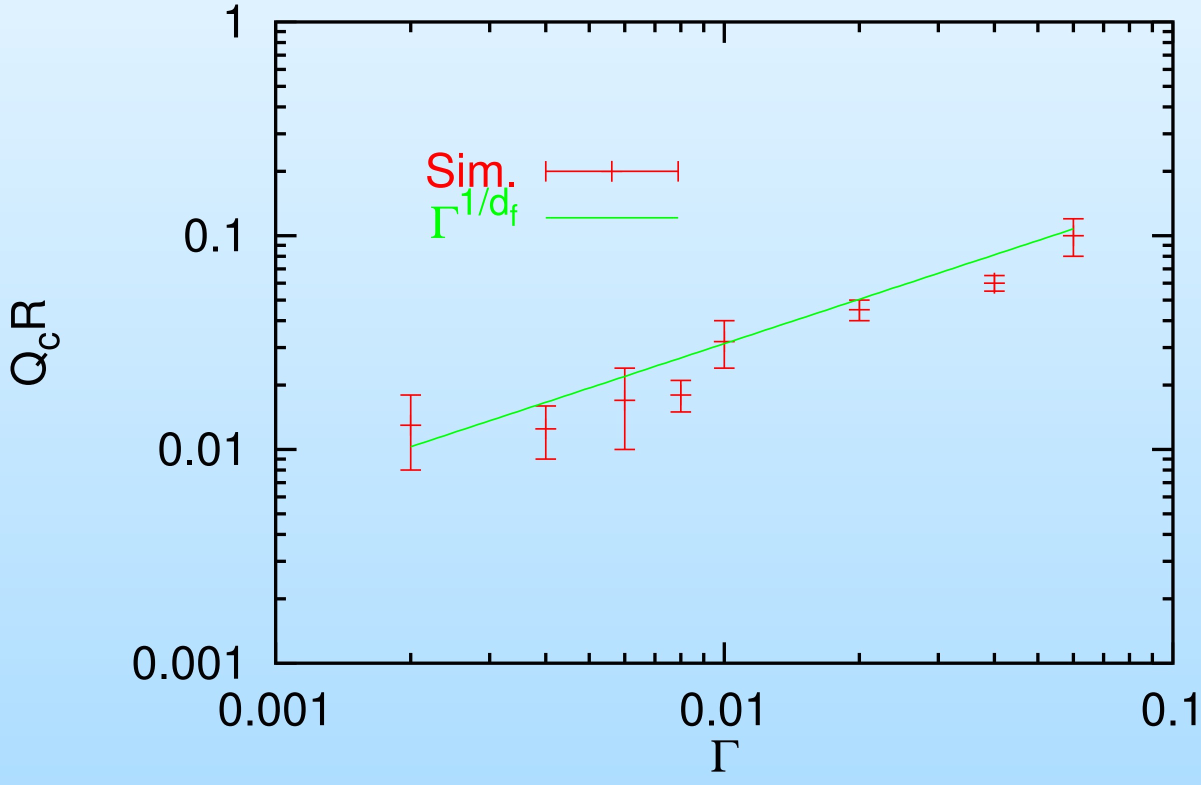
- Number of particles $N(r) \propto \left(\frac{r}{R}\right)^{d_f}$,

$$\Longrightarrow Q_h R \propto \left(g(s)\Gamma\right)^{1/d_f},$$

- No scaling beyond $Q_h \equiv 1/L_h$.

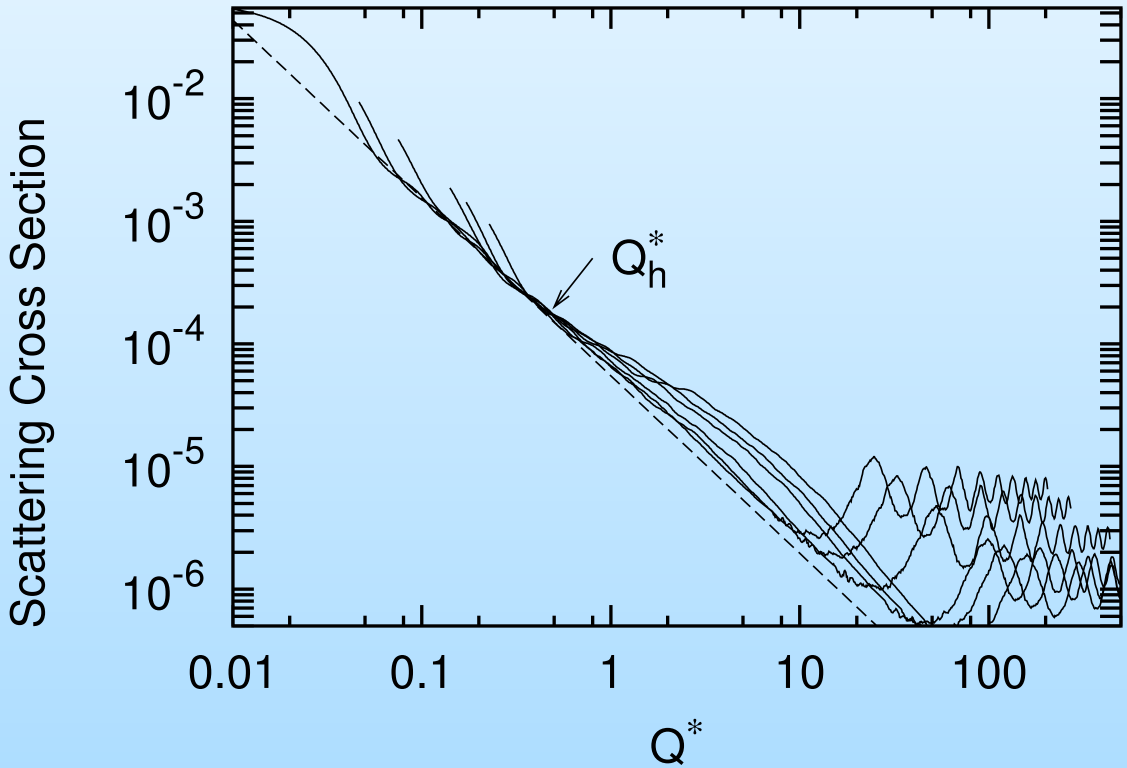
Q_h vs Γ?

$$Q_h R \propto (g(s)\Gamma)^{1/d_f}$$



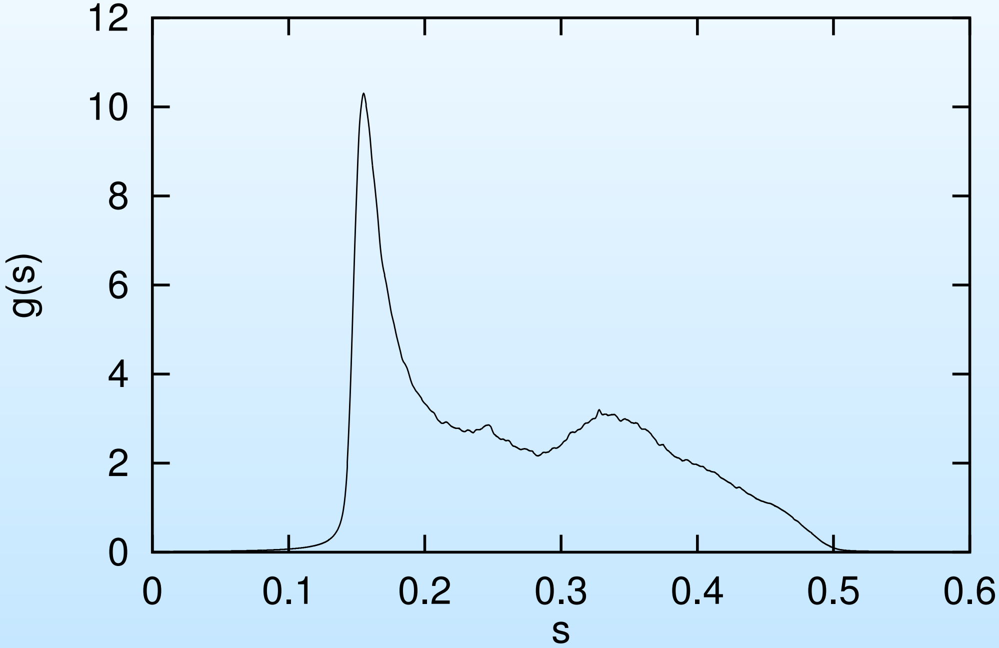
Q_h vs Γ?

$$Q_h R \propto (g(s)\Gamma)^{1/d_f}$$

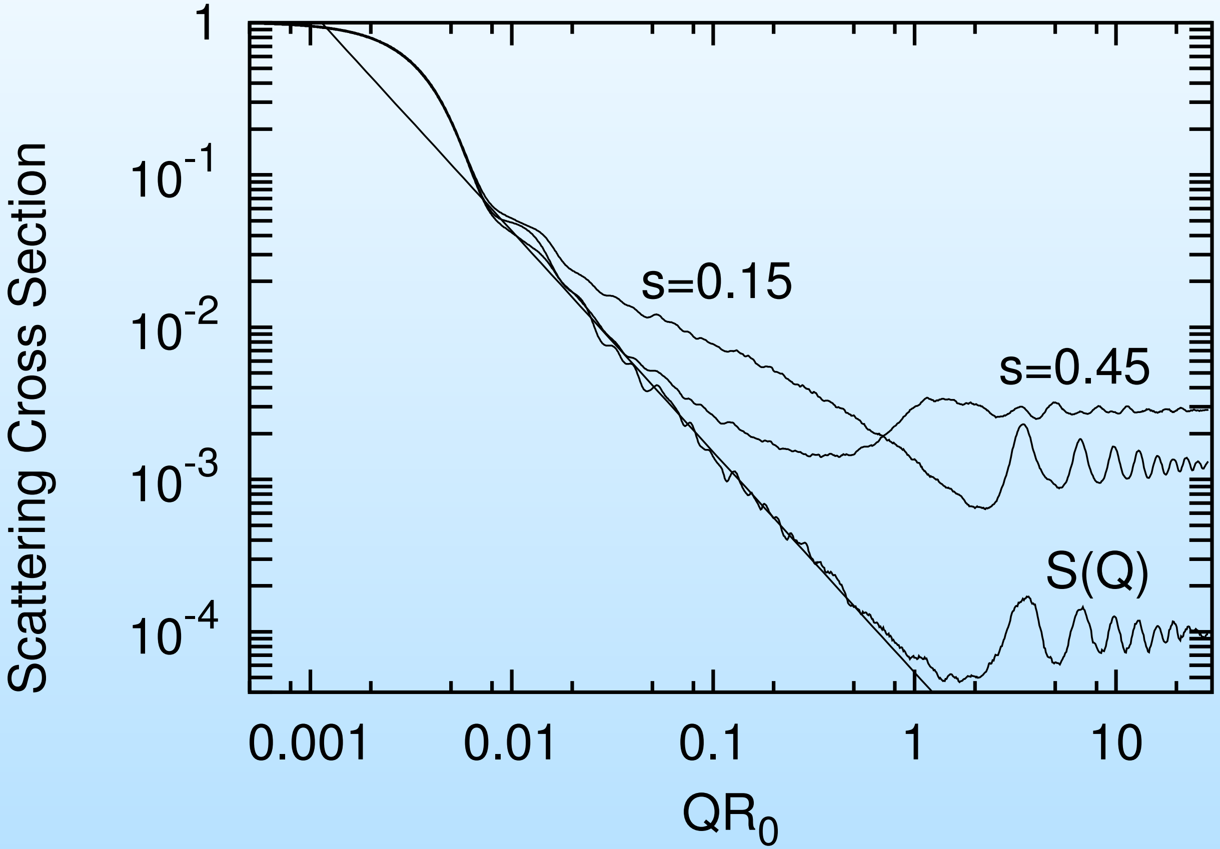


$$\Gamma = 10^{-3}\text{–}10^{-1}$$

Spectral Function for Transverse Polarization

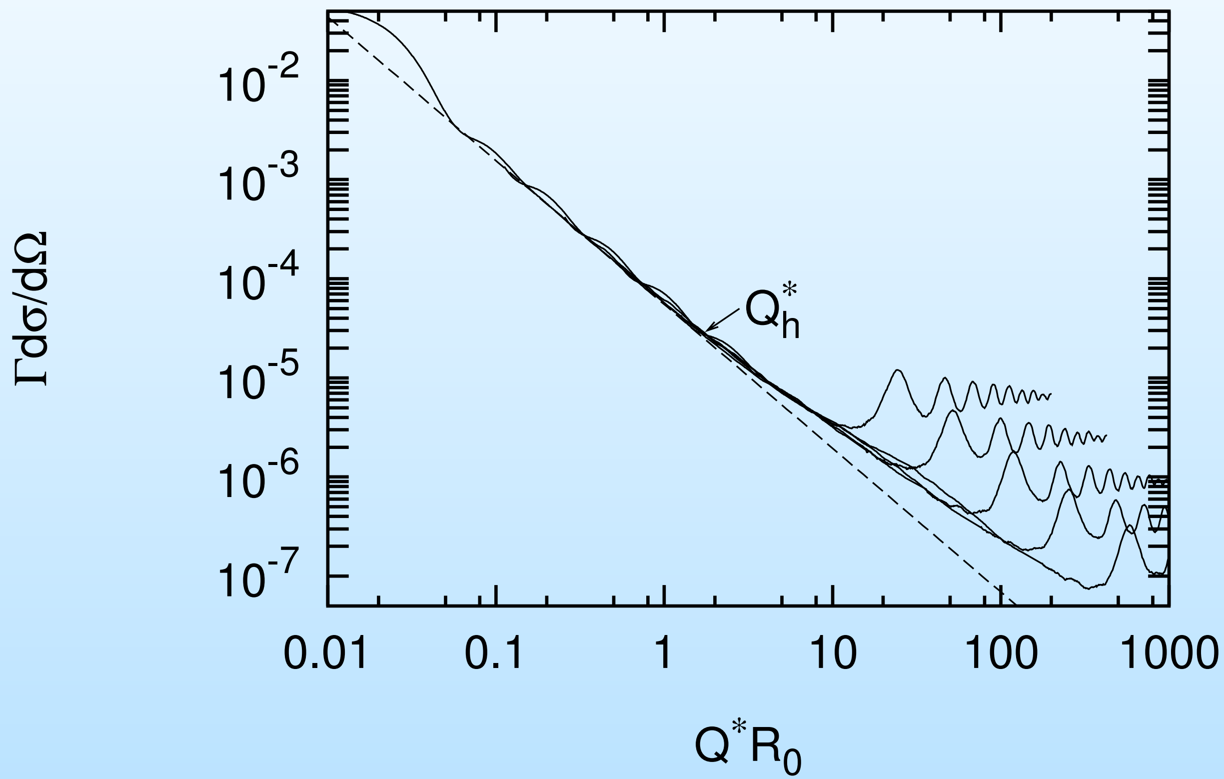


Scattering Cross Section



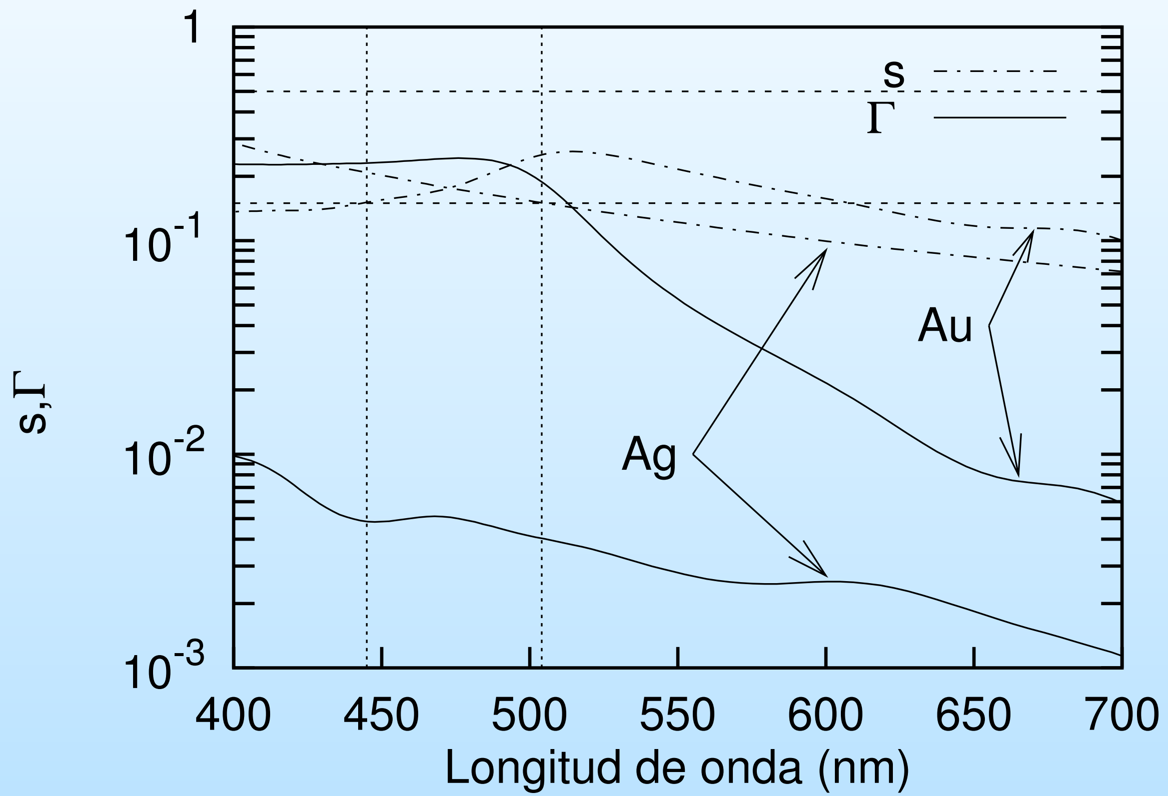
Scaling for transverse polarization

$Q^* \equiv Q/\Gamma^{1/d_f}$



$\Gamma = 10^{-3}$–10^{-1}

Applications (3D)



Band: $s = 0.15$–0.5 for 3D DLCA ($d_f \approx 1.78$)

Conclusions

- The hierarchical algorithm allowed the numerical study of scattering from ensembles of large colloidal aggregates.
- For 2D CDLA clusters, the spectral function extends from $s \approx 1/4$ to $s \approx 1/2$ and shows significative structures (scalar model).
- From a local analysis we found at the red end of the spectrum (R) the polarization is antiferromangnetic like, while at the blue end (B) it is ferromagnetic like.
- Normal modes consists of intense 'hot spots' whose position varies abruptly with frequency.
- They are not extended nor exponentially localized.

Conclusions

- We found power law scaling at the R region $d\sigma/d\Omega \propto Q^{-d_f}$,
- but **no scaling** at the blue (B) end of the spectrum,
- since the minimum distance L_h gets close to the system size.
- However, scaling is displayed for larger systems or larger widths Γ for which multiple hot spots are excited.
- Assuming excited hot spots form a fractal with the same dimension as the system aggregate, we obtained a power law $Q_h \propto \Gamma^{(1/d_f)}$, confirmed by simulations.
- Thus, experiments may show scaling or not in the multiple scattering regime, depending on frequency, aggregate size, and dissipation factors.
- Main results are confirmed with full transverse vectorial calculations.