Anomalous transport and charge ordering in quantum dot arrays

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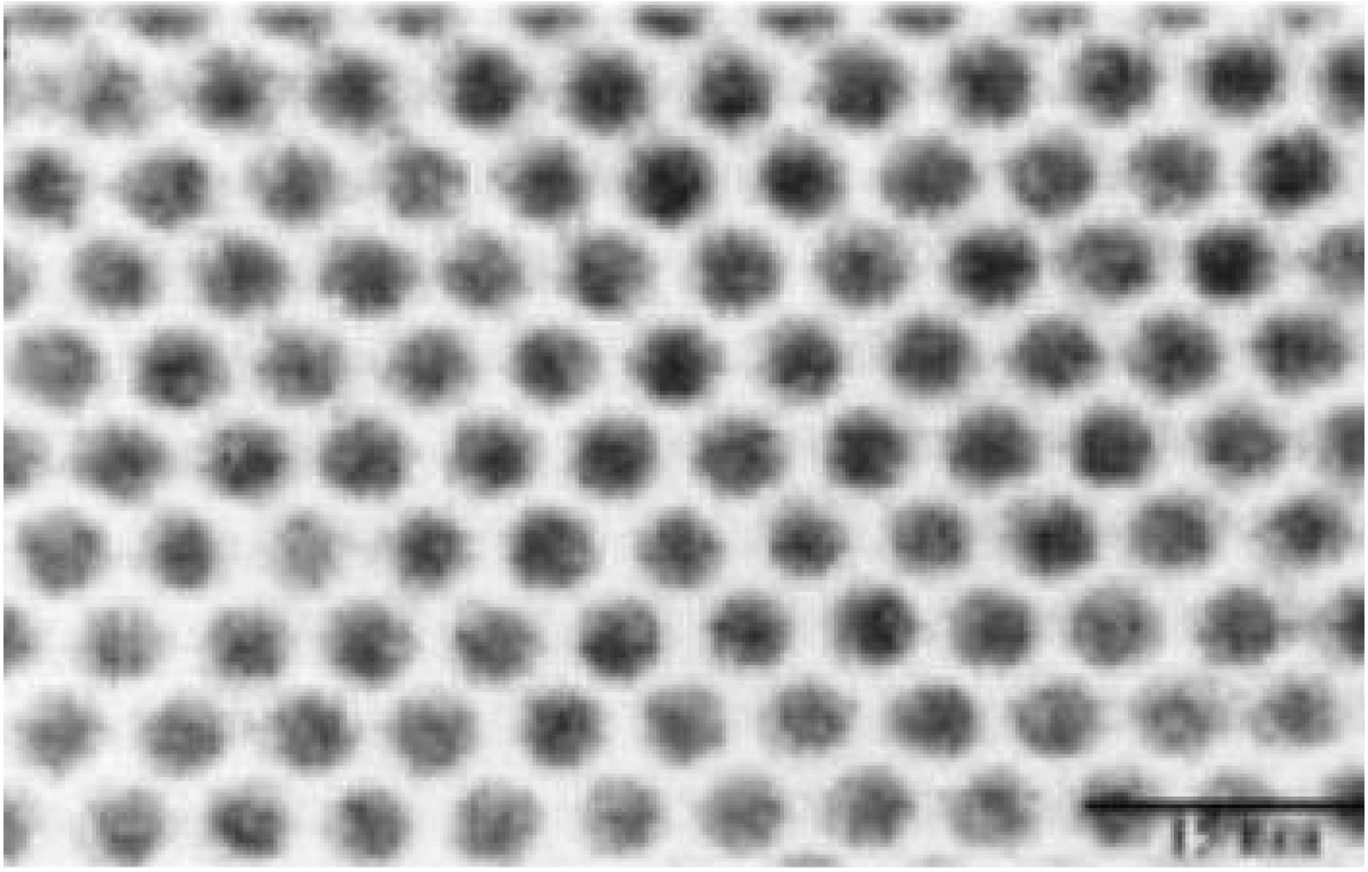
Quantum Dot Array



courtesy Bawendi group, MIT Chemistry Dept

Motivation

– **Novel self-assembled tunable system**

– **Rich physics at all scales**

- Single dot properties
  fluorescence intermittency, Lévy statistics

- Charge ordering in dot arrays
  Coulomb + geometric frustration

– **Transport measurements**

- Charge ordering

- Dynamics

Outline

1. Experimental setup

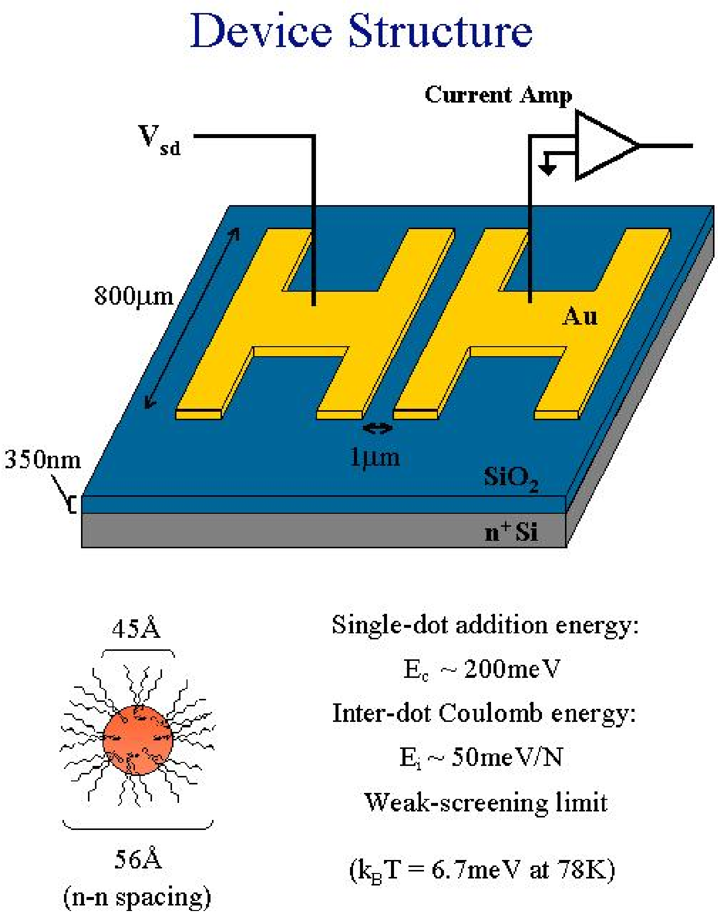
2. QD Array as ideal $2D$ $\triangle$ lattice

- Charge ordering types
- Phase diagram
- Transport

3. Transport at large bias

- Experimental results
- Transport model

Experimental Setup



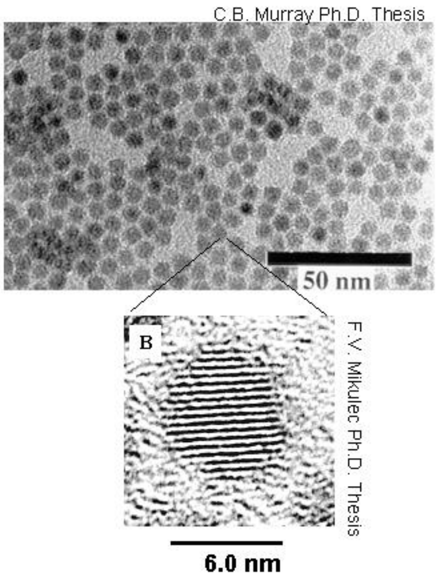


image from N.Y. Morgan, PhD Thesis (MIT, 2001)

- CdSe dots 2-15 nm diam (5% rms), 1nm capping
- $2D$ closely packed triangular arrays
- 1-2 μm gap between source and drain
- back gate controls electron density

Ideal $2D$ triangular lattice

Geometric frustration + Coulomb $\Rightarrow$

Classical $\triangle$IAFM w/ long range interactions

The Model

- $2D$ triangular lattice
- Large $E_c \;\Rightarrow\; q = 0, 1$ site occupancy
- Classical Hamiltonian with long range interaction:

$$\mathcal{H} = \frac{1}{2}\sum_{i,j} V(\vec{r}_{ij}) q_i q_j + \sum_{\vec{r}_i} (V_{\rm gate} + \phi_{\rm ext}(\vec{r}_i)) q_i \ ,$$

$$V(\vec{r}_{ij} \neq 0) = \left(\frac{1}{\epsilon |\vec{r}_{ij}|} - \frac{1}{\epsilon \sqrt{(\vec{r}_{ij})^2 + d^2}} \right) e^{-\gamma |\vec{r}_{ij}|} \ , \ \gamma^{-1} = 2d \ .$$

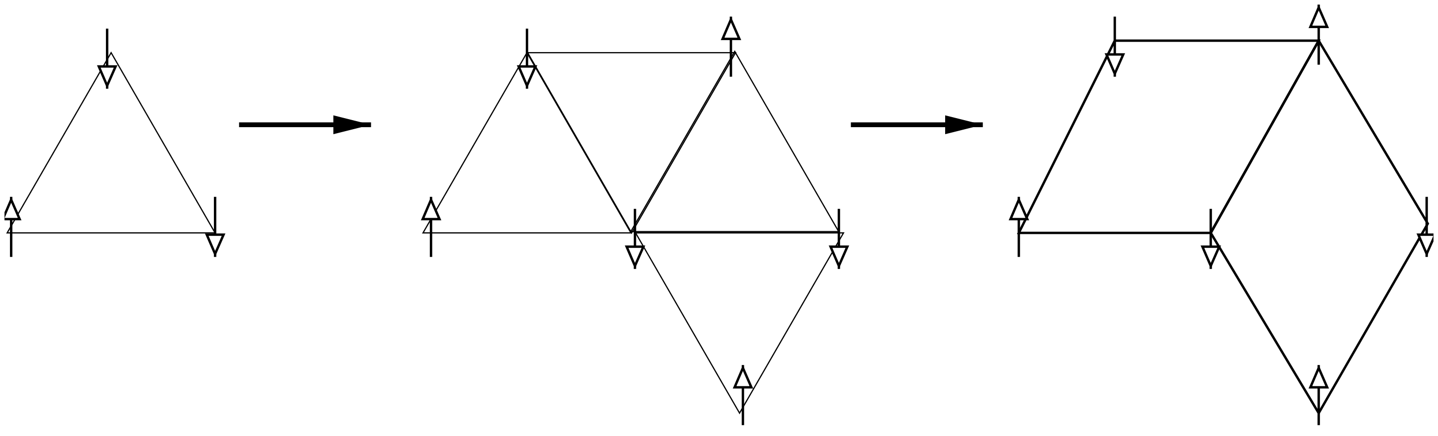
$d/a \sim 2 - 10$

- Incoherent hops between neighboring sites

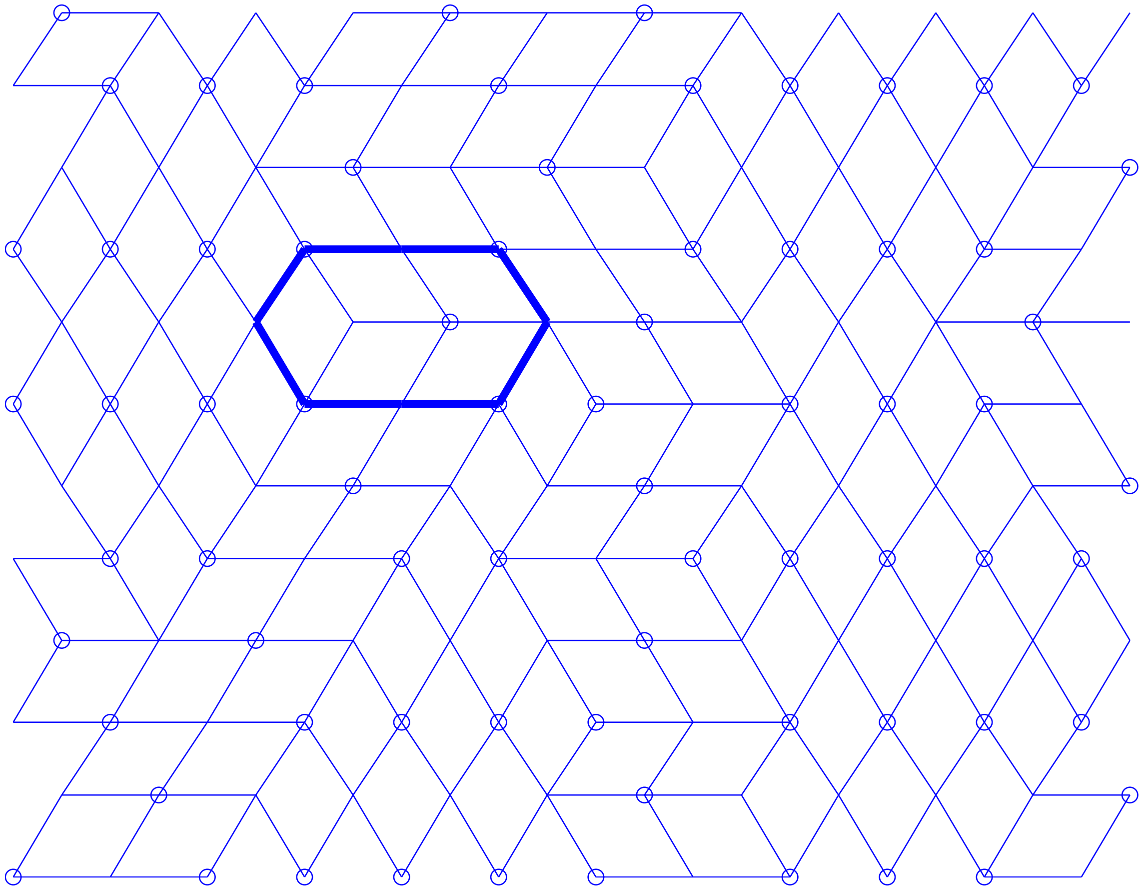
Map on long range △IAFM

Charge model	Spin model
$q = 1$	$s = \uparrow$
$q = 0$	$s = \downarrow$
V_{ij}	J_{ij}, AF sign
V_{gate}	H_{ext}

Frustrated Lattice



Height Field, $T = 0$

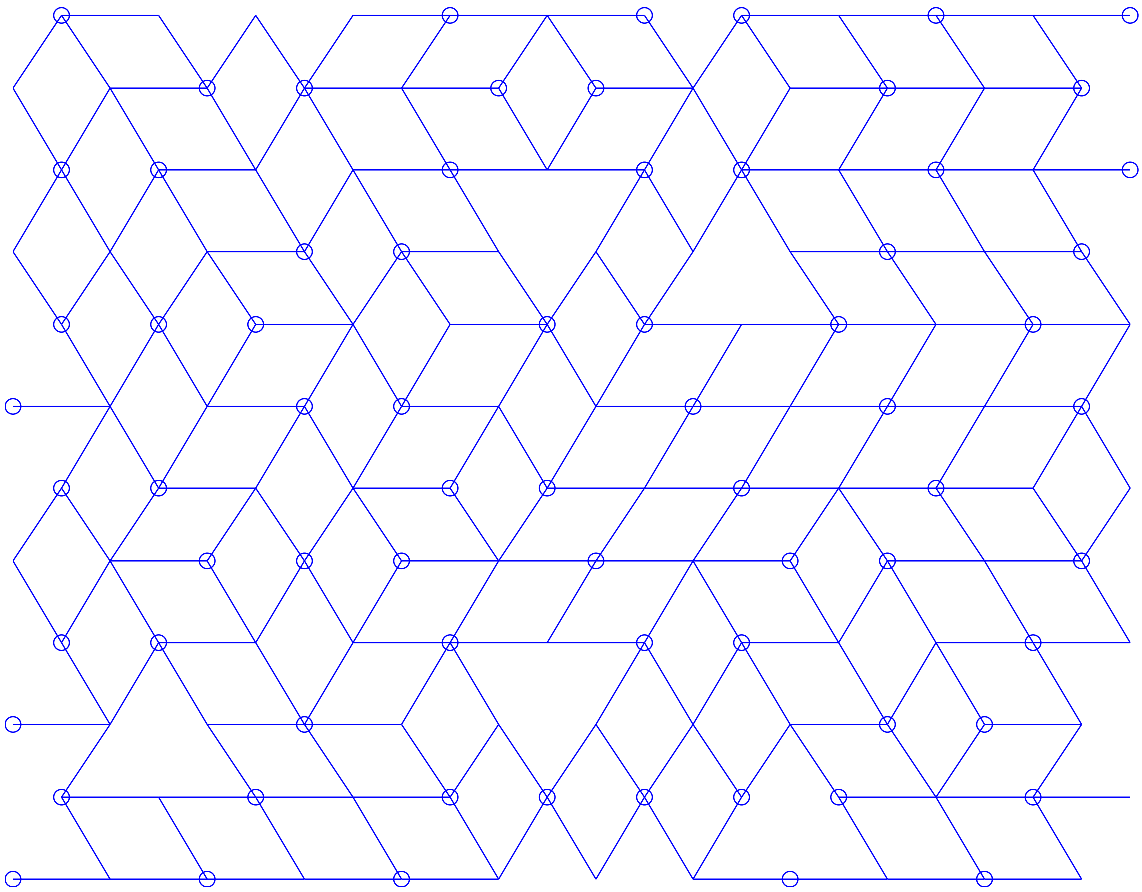


- Minimize # of frustrated bonds:
pair elementary triangles $\to$ rhombi

- Covering by rhombi $\leftrightarrow$ Map to SOS

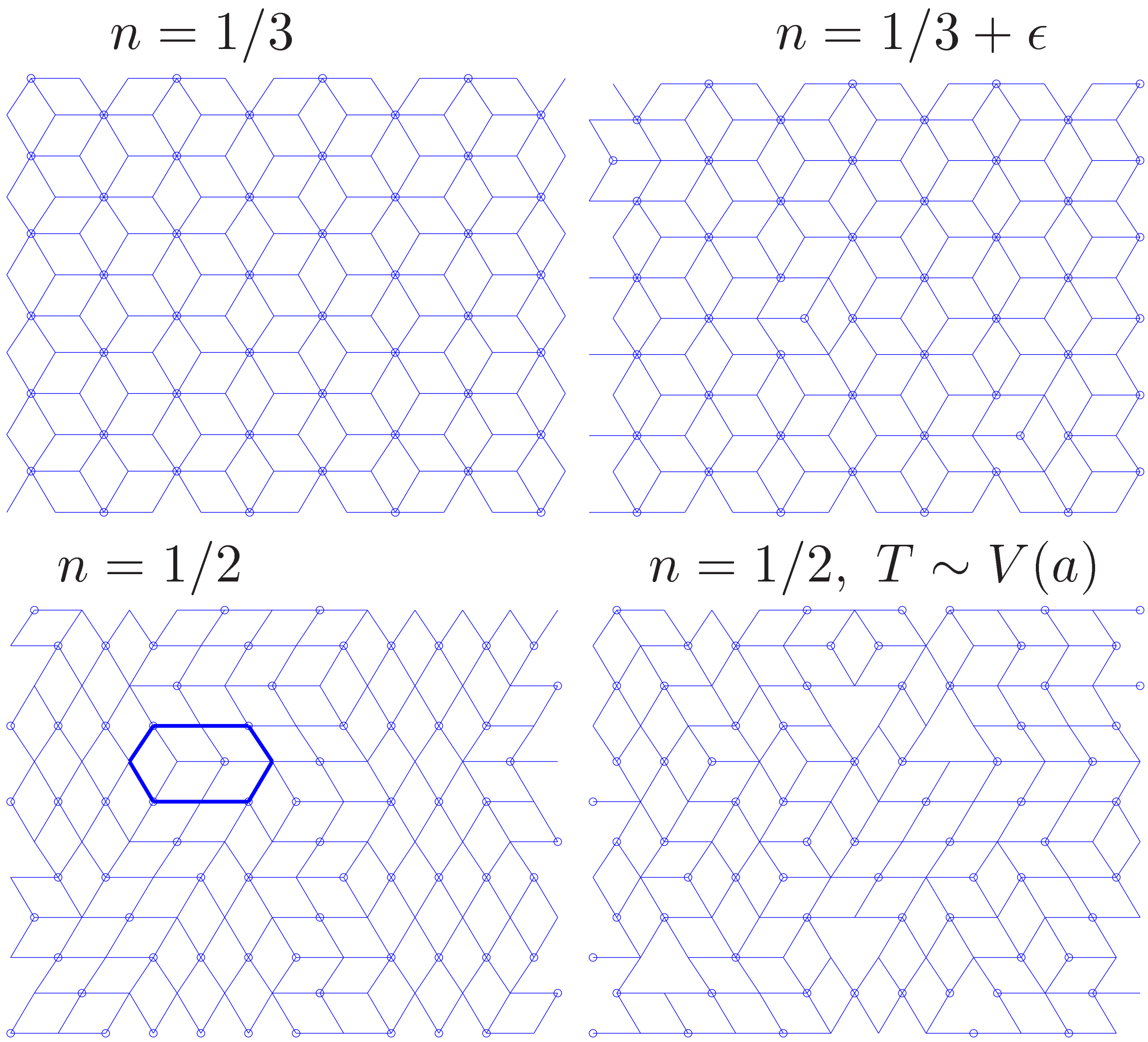
- Correlated hops

Height Field, $T > 0$



- Dislocations at $T > 0$ (large triangles): topological defect pairs

Charge Ordering via Height Field

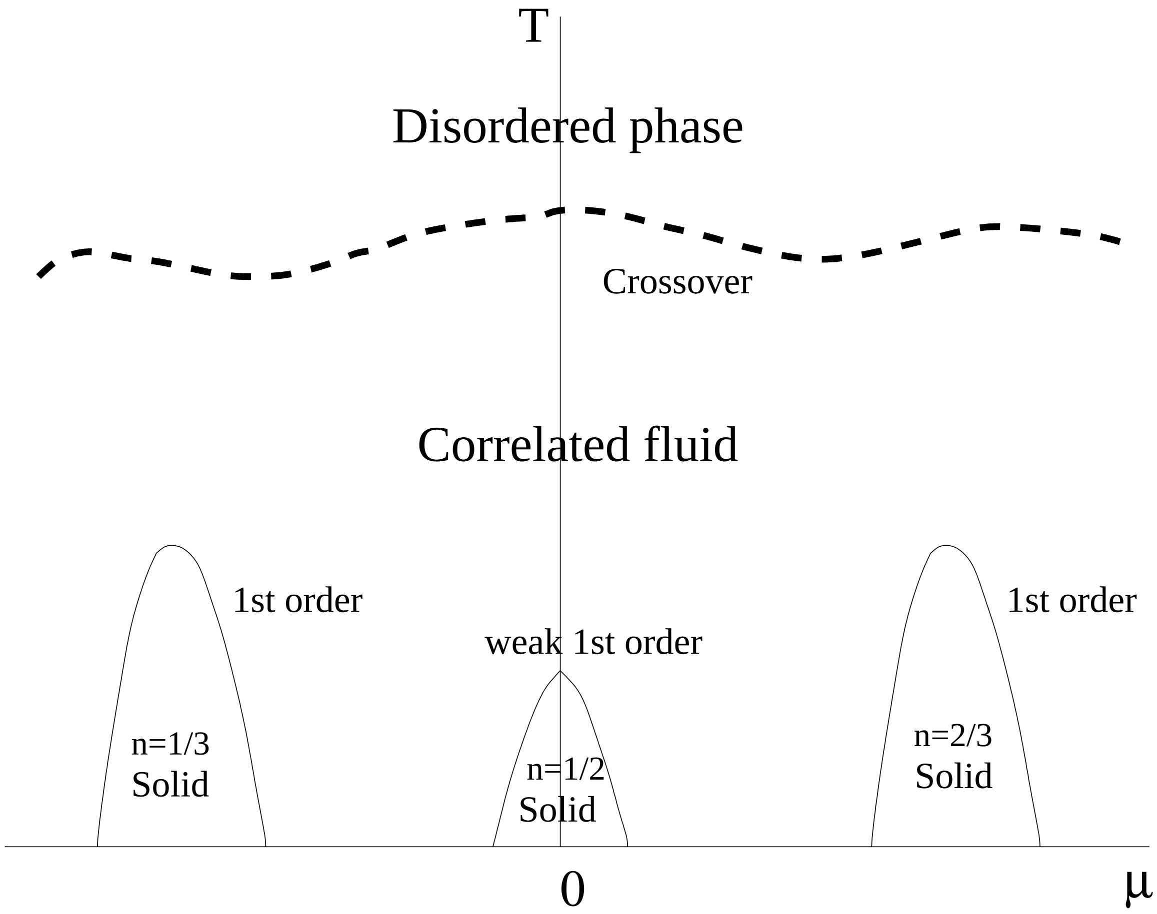


$\triangle$IAFM $\to$ Height Field $\to$ Roughening:

H.W.J. Blöte and H.J. Hilhorst, J.Phys. A**15**, L631 (1982);

B.Nienhuis, H.J.Hilhorst, H.W.J.Blöte, J.Phys.A**17**, 3559 (1984)

Phase Diagram



- Correlated phase: fluctuating height surface
- Freezing into commensurate states
- No low temperature $2D$ XY phase

Nearest Neighbor vs. Long Range

Nearest Neighbor:

- Height defined only at $T = 0$
- Height surface is *rough*: [H.W.J. Blöte et al]

$$\left\langle (h(\vec{r}_i) - h(\vec{r}_j))^2 \right\rangle = \frac{1}{\pi\kappa} \ln \frac{r_{ij}}{r_0} \,,$$

κ =*rigidity*, $\quad \mathcal{F}_h = \int d^2\mathbf{r}\, \frac{\kappa}{2} \left(\nabla_{\mathbf{r}} h\right)^2$

$\kappa_{\triangle\mathrm{IAFM}} = \pi/9$ at $n = 1/2$
[J. Stephenson, J. Math. Phys. A**11**, 413 (1970)]

Long Range $V(\mathbf{r})$:

- Height defined at $0 < T < T_{\mathrm{freezing}}$ (commensurate ph.)
- Freezing *rough* $\rightarrow$ *commensurate*: 1st order
- Height $\textit{globally not defined}$ at $T > T_{\mathrm{freezing}}$
- Unbound dislocations; no B.K.T. transition

Correlated Phase

- **No Berezinskii-Kosterlitz-Thouless Transition**

Rigidity $\kappa < \kappa_f = \frac{\pi}{8} < \kappa_{\rm KT} = \frac{2\pi}{9}$,

$$b \; = \; [\text{cubic cell diagonal}]/3 \; \equiv \; 1 \; ,$$

$$\kappa_{\rm KT} = \frac{8\pi}{b_\triangle^2} \; , \quad b_\triangle = 6b$$

- **Total Free Energy :** $\mathcal{F} = \mathcal{F}_h + \mathcal{F}_n + \mathcal{F}_{\rm int}$,

$$\mathcal{F}_h = \int d^2\mathbf{r} \left(\frac{\kappa}{2} (\nabla_{\mathbf{r}} h)^2 \; + \; g \cos\frac{2\pi h}{b} \; + \; f\, V_{\rm gate} \cos\frac{\pi h}{b} \right) \; ,$$

$$\mathcal{F}_n = \frac{1}{2} \int d^2\mathbf{r} d^2\mathbf{r}'\, n_{\mathbf{r}} U_{\mathbf{r}-\mathbf{r}'} n_{\mathbf{r}'} + \int d^2\mathbf{r}\, n_{\mathbf{r}} \Phi^{\rm ext}(\mathbf{r}) \; ,$$

$$\mathcal{F}_{\rm int} = \lambda \int d^2\mathbf{r}\; n_{\mathbf{r}} \; \cos\frac{\pi h_{\mathbf{r}}}{b} \; .$$

- **Measure κ directly using MC dynamics**

Monte-Carlo Dynamics

- **Boltzmann Hopping Probabilities**

Nonconserving dynamics (fixed $V_{\rm gate}$)

$$W_i/\overline{W}_i = e^{-2\Phi_i/T}\ ,\ W_i + \overline{W}_i = 1$$

Conserving dynamics (fixed n)

$$W_{i\to j}/W_{i\to i} = e^{(\Phi_i - \Phi_j)/T}\ ,\ W_{i\to j} + W_{i\to i} = 1$$

$$\Phi_i = \sum_{r_j \neq r_i} V(\vec{r}_{ij}) q_j + V_{\rm gate} + \phi(\vec{r}_i)$$

FREEZING: 1st order

- $T \sim$ Next Nearest Neighbor coupling: Commensurate phase, finite entropy

- Nearest Neighbor: extensive entropy, no freezing!

- Ground state: const average tilt of a height field

- Singularity in $\sigma(T) \sim$ singularity in $\langle E \rangle$

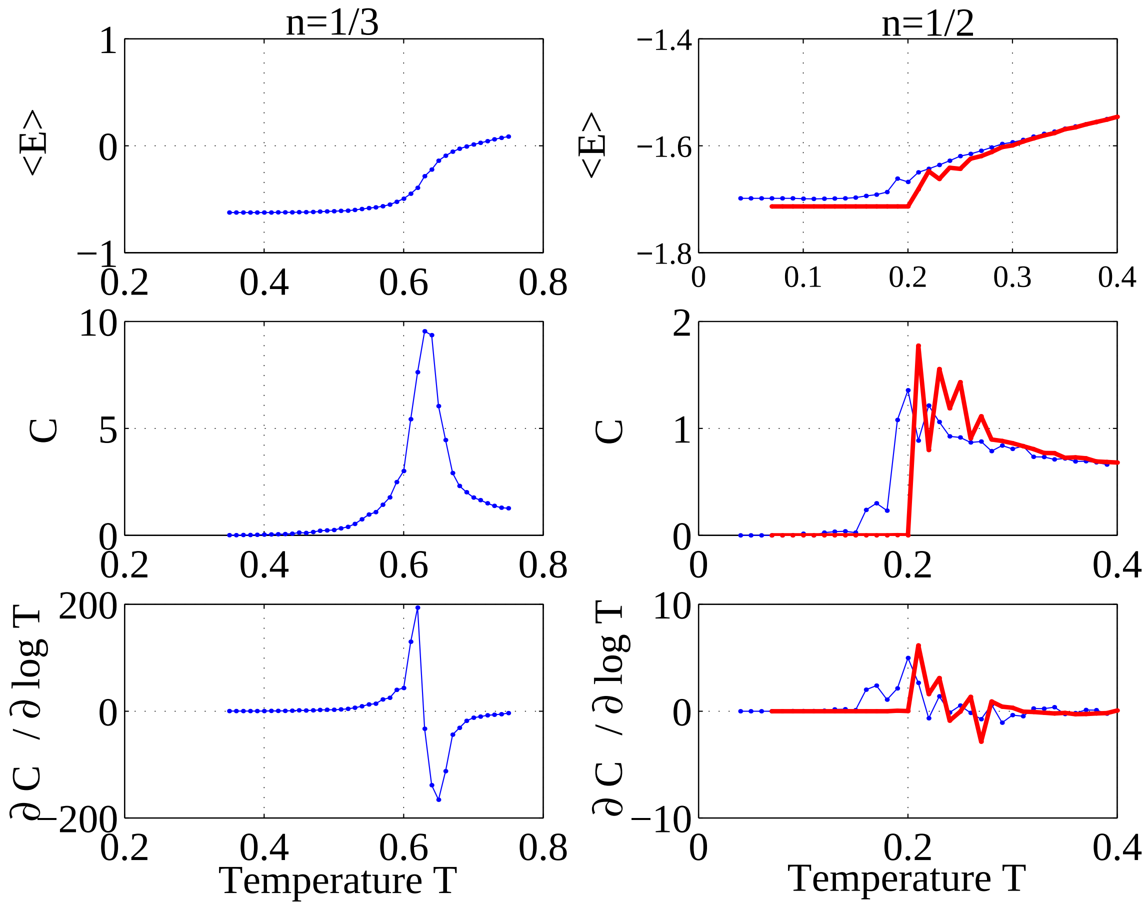
- $n = 1/3$: First order
$\delta n_i = n_i - 1/9$, symmetry S_3
$\varphi_1 = \frac{\delta n_1 - \delta n_2}{\sqrt{2}}, \quad \varphi_2 = \frac{\delta n_1 + \delta n_2 - 2\delta n_3}{\sqrt{6}}$

$$\mathcal{F}_{1/3} = A_2 \left(\varphi_1^2 + \varphi_2^2\right) + A_3\, \varphi_2(\varphi_2^2 - 3\varphi_1^2) + A_4 \left(\varphi_1^2 + \varphi_2^2\right)^2$$

- $n = 1/2$: First order
$s_i = 2n_i - 1$ on i-th edge of a fixed $\triangle$
symmetry: P6mm = $\mathrm{C_{3v}} \times$ translations

$$\mathcal{F}_{1/2} = B_2 \left(s_1^2 + s_2^2\right) + B_3 \cdot s_1 s_2 s_3 + B_{41} \left(s_1^4 + s_2^4 + s_3^4\right) + B_{42} \left(s_1^2 + s_2^2\right)^2$$

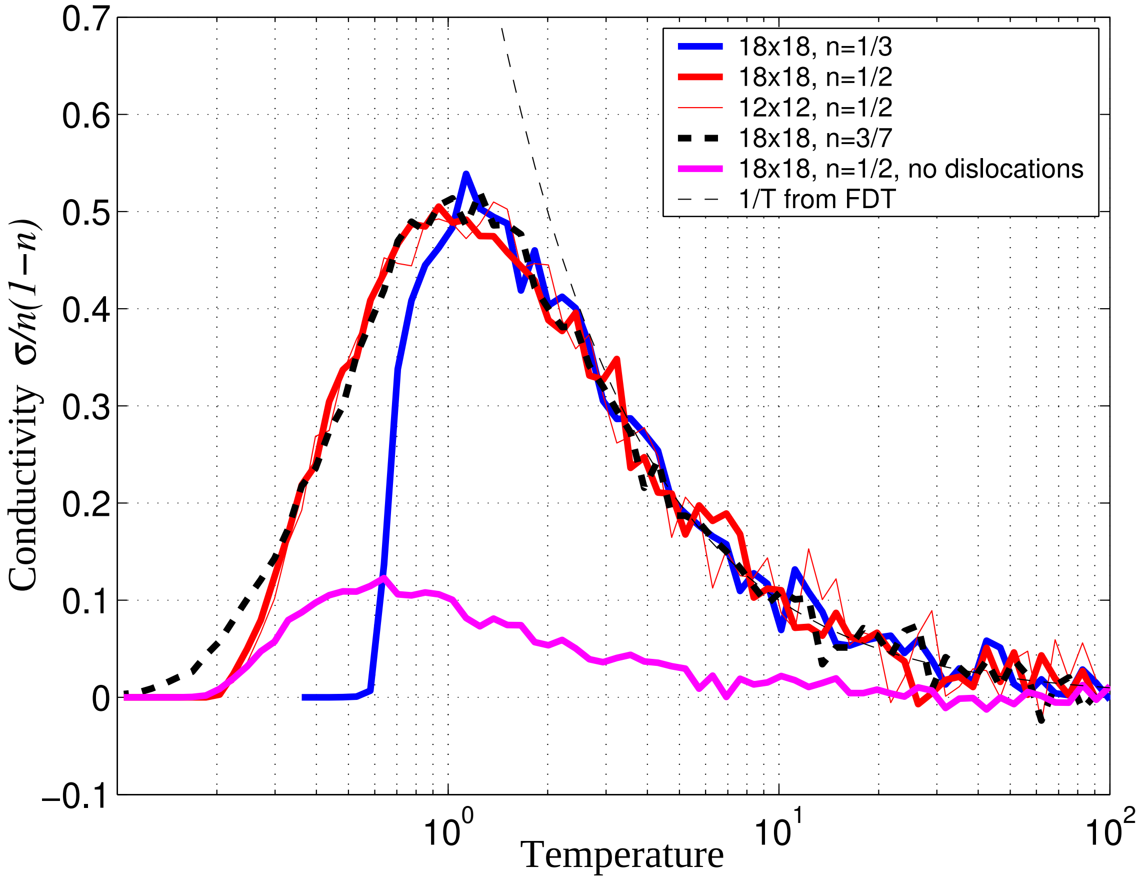
Freezing Phase Transitions



Moments of $p(E)$: $M_1 = \langle E \rangle$,
$M_2 = \left\langle (E - \langle E \rangle)^2 \right\rangle, \; M_3 = \left\langle (E - \langle E \rangle)^3 \right\rangle$

At equilibrium
$\langle E \rangle = M_1, \; C = \frac{\partial \langle E \rangle}{\partial T} = \frac{M_2}{T^2}, \; \frac{\partial C}{\partial \ln T} = \frac{M_3 - 2T M_2}{T^3}$

Conductivity vs Temperature



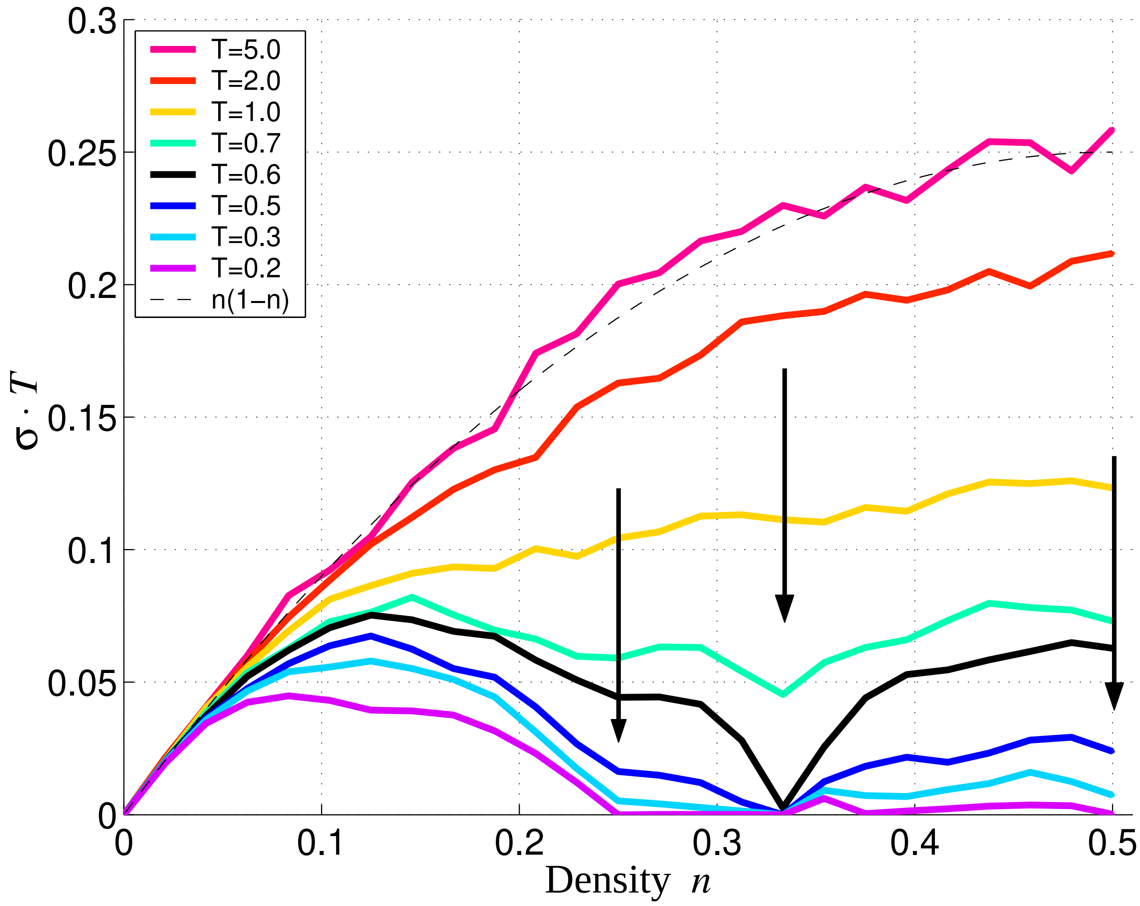
FDT: $\int dt\, \langle j_\mu(t) j_\nu(0)\rangle = 2\sigma_{\mu\nu} T$

$$\left\langle j^2 \right\rangle = \frac{4}{6} \cdot 2n(1-n) \cdot (\delta s)^2 \cdot \left(a \cos\frac{\pi}{6} \right)^2 \cdot w$$

High temperature limit for the conductivity:

$$\sigma = a^2\, \frac{n(1-n)}{T}\ .$$

Conductivity vs Density



Singularities correspond to charge ordering at

$$n = 1/4,\ 1/3,\ 1/2$$

High temperature limit for the conductivity:

$$\sigma \cdot T = n(1-n)$$

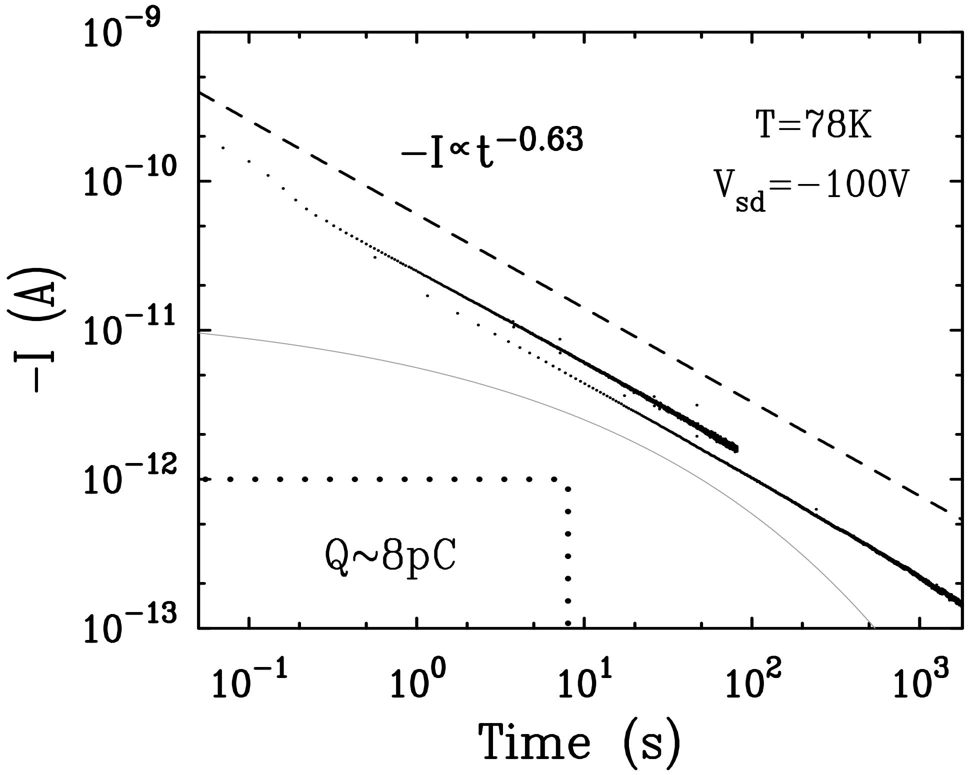
Conclusions so far...

- Correlated phase: collective behavior in a classical frustrated system
- No dislocations unbinding transition
- Freezing via the first order transition
- Conductivity as a probe of charge ordering

In Reality...

...Anomalous transport at a large bias
(up to $\sim$0.5 V / dot)

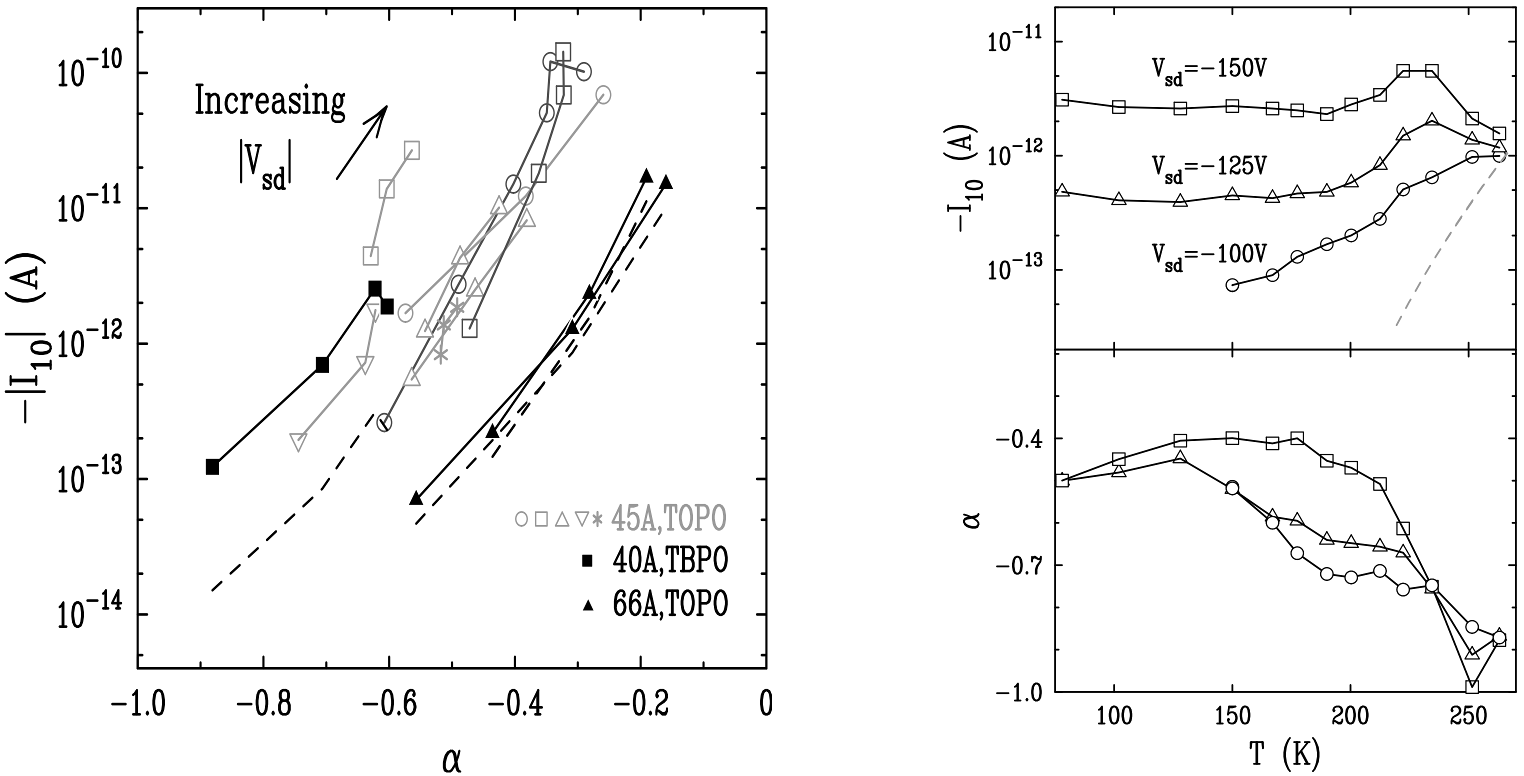
What's observed?



N.Y. Morgan *et al.*, `cond-mat/0204560`

- $I = I_0\, t^{-\alpha}$ for 5 decades in time (up to $2 \cdot 10^4$ s)
- $0 < \alpha < 1$, $\int_0^\infty dt\, I(t) = \infty$
- Memory effect: repeat $\Rightarrow$ smaller I_0, same α

Non-universal power law



α is non-universal!

Non-Stationary Response

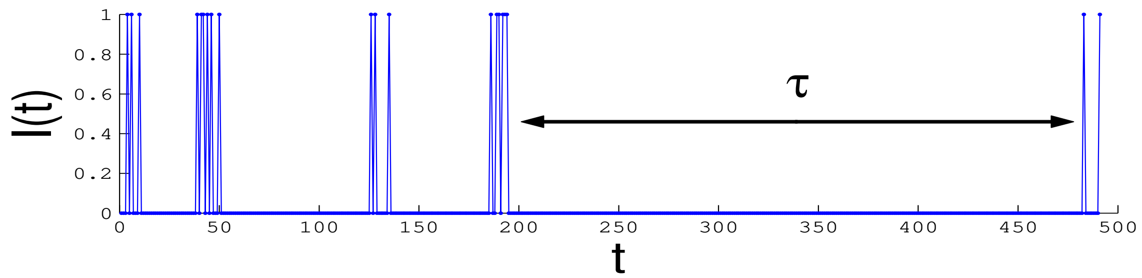
Non-stationary system
(charging? dots move? cool down?)

Stationary system (!)
(Anomalous diffusion)

… + any mix of the above?

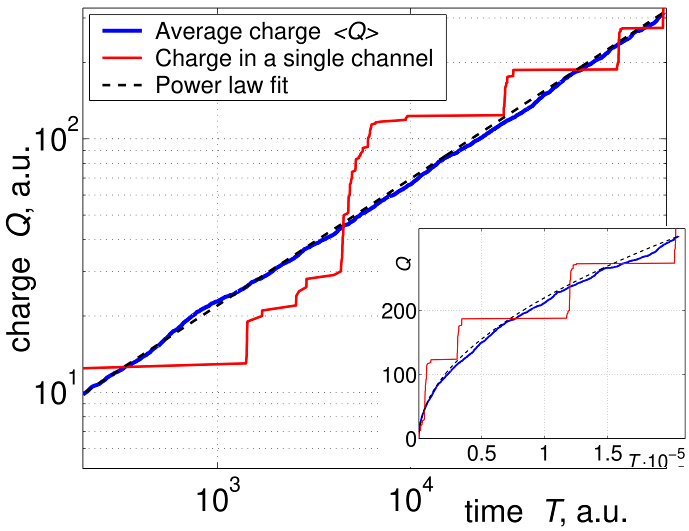
Phenomenology

- $N \gg 1$ independent conducting channels



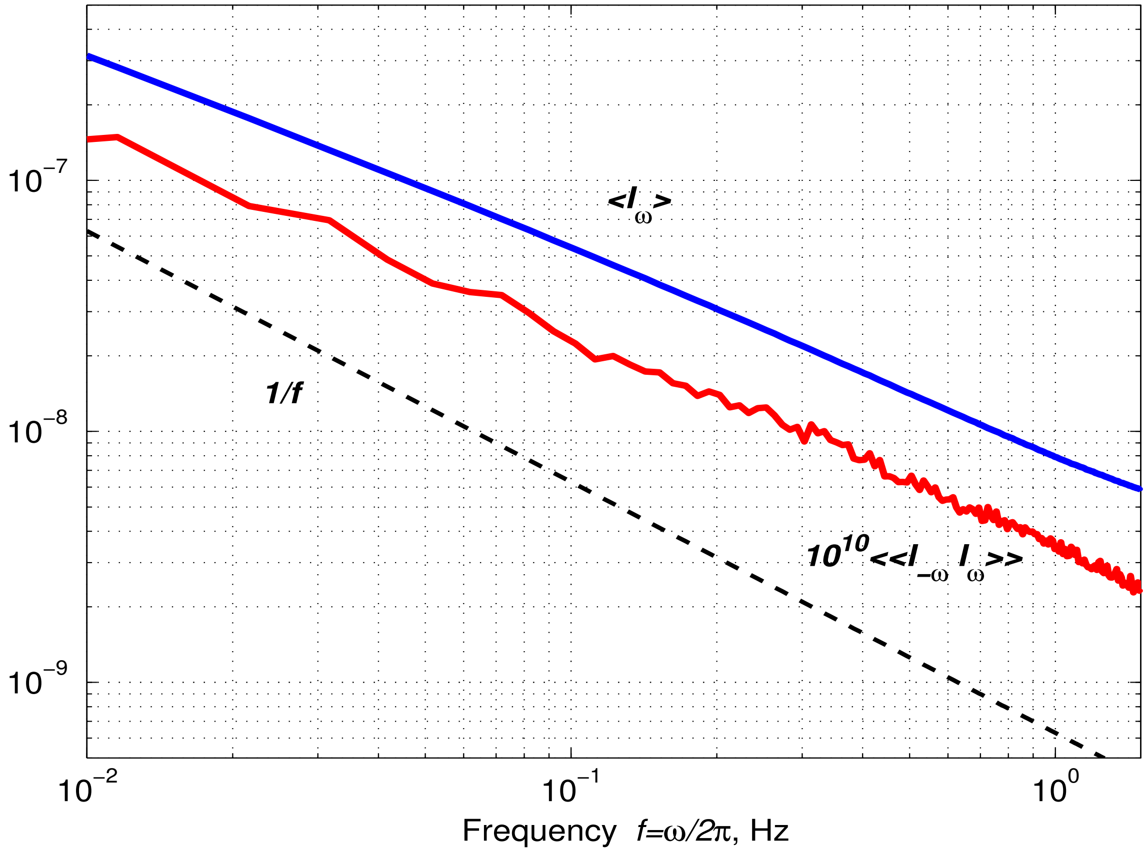
- Single channel: *Stationary* Lévy process

$$p(\tau) = \frac{a}{\tau^{1+\mu}}\ , \quad 0 < \mu < 1\ ; \quad \overline{\tau} = \infty$$



- $N \gg 1$: $\quad \langle I(t) \rangle \sim t^{-\alpha}\ ,\ \alpha = 1 - \mu\ ; \quad \langle Q(t) \rangle \sim t^{\mu}$

NOISE



M. Drndic, D.N. et al., unpublished

Power law noise spectrum (Lévy statistics):

$$\langle I_\omega \rangle \propto \omega^{-\mu} \ , \quad \langle\langle I_{-\omega} I_\omega \rangle\rangle \propto \omega^{-\mu} \ , \quad \omega T \gg 1$$

Fluorescence intermittency

On and Off times – according to the Lévy process,

$p(\tau) = \frac{a}{\tau^{1+\mu}}\ , \quad \mu \sim 0.5$

K. T. Shimizu *et al.*, Phys. Rev. B **63**, 205316 (2001);
Y. Jung, E. Barkai, R.J. Silbey `cond-mat/0204378`;
X. Brokmann *et al.*, `cond-mat/0211171`

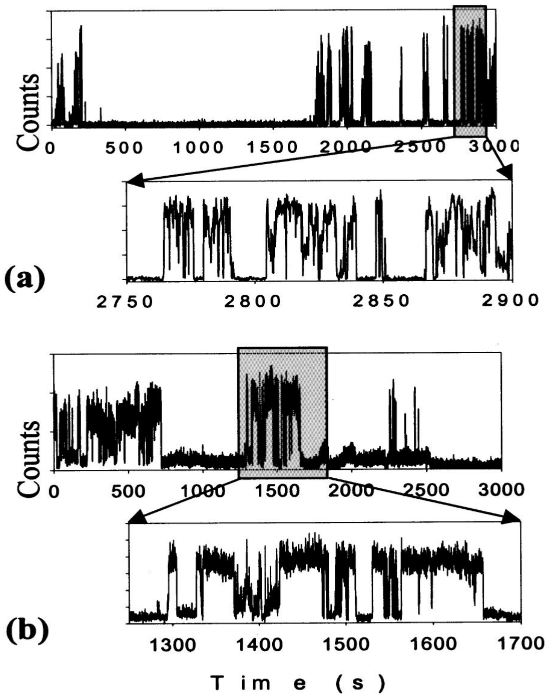


FIG. 1. Typical intensity time trace of CdSe(ZnS) QD fluorescence intermittency at (a) RT and (b) 10 K. Expanded view illustrates the similar nature of the blinking events at RT and 10 K but with different time scales.

Figs. 1(a) and 1(b), respectively. A small section of the time trace is expanded to show the self-similarity and complexity of the traces on different time scales. We define the *on* time (or *off* time) as the interval of time when no signal falls below (or surpasses) a chosen threshold intensity value. The probability distribution is given by the histogram of *on* or *off* events of length t:

$$P(t)=\sum_t [\text{events of length } t]. \tag{1}$$

The *off*-time probability distribution for a single CdSe/ZnS QD at room temperature is shown in Fig. 2(a). The distribution follows a pure power law for the time regime of our experiments ($\sim 10^3$ s):

$$P(t)=At^{-\alpha}. \tag{2}$$

Most individual dots also follow a power-law probability distribution with the same value in the power-law exponents ($\alpha \approx 1.5 \pm 0.1$). A histogram of α values for individual QD's is shown in the inset of Fig. 2(a). The universality of this

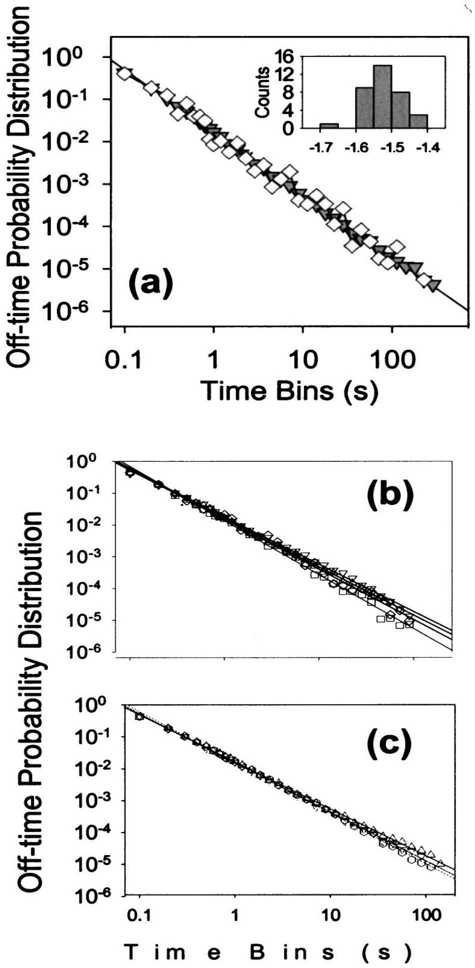


FIG. 2. (a) Normalized *off*-time probability distribution for one CdSe QD (◇) and average of 39 CdSe QD's (▼). Inset shows the distribution of fitting values for the power-law exponent in the 39 CdSe QD's. The straight line is a best fit to the average distribution with exponent ~ -1.5 (b) Average *off*-time probability distribution for 25-Å radius CdSe(ZnS) QD at 300 K (▽), 10 K (△), 30 K (◇), and 70 K (□). The α values are 1.41, 1.51, 1.37, and 1.45, respectively. (c) Average *off*-time probability distributions for 39 CdSe(ZnS) QD of radius 15 Å, (▽) and 25-Å radius CdSe(ZnS) QD (◇) and 25-Å radius CdSe QD (△) at RT. The α values are 1.54, 1.59, and 1.47, respectively.

statistical behavior indicates that the blinking statistics for the *off* times are insensitive to the different environments of each dot. Kuno *et al.*[8] first observed this behavior at room temperature using conventional confocal microscopy. Due to higher signal-to-noise ratio, confocal microscopy provides a time resolution as high as 200 μs; however, the same probability distribution is also observed with 100-ms integration times. Initial experiments at RT show that the same blinking

Summary

• Novel self-assembled system with rich physics

• Transport and noise as a probe

• Non-stationary response in a statistically stationary system due to Lévy statistics

• Excited states of the dot:
optical vs. transport measurements?

• More questions than answers...