Anomalous transport and charge ordering in quantum dot arrays

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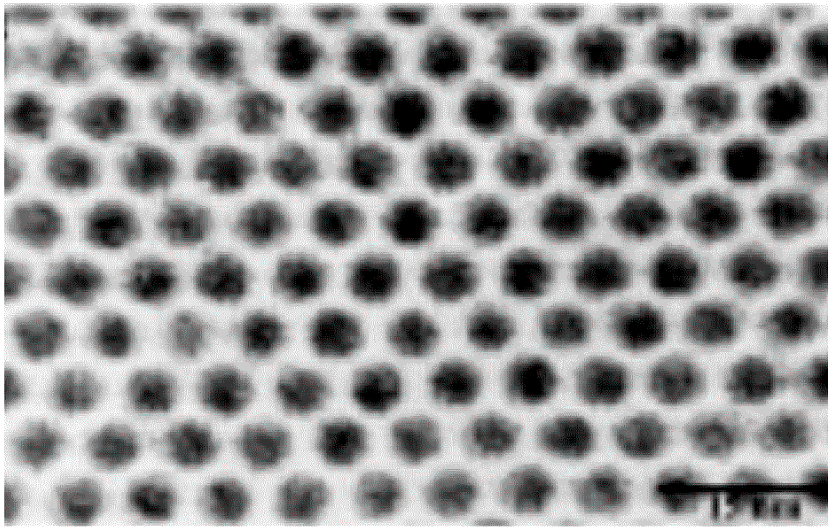
Quantum Dot Array



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

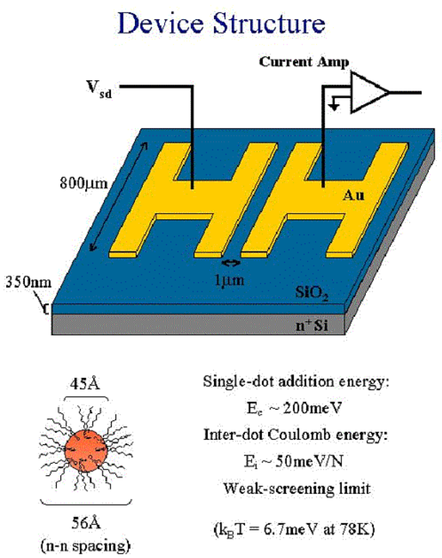
2. QD Array as ideal $2D$ $\triangle$ lattice

- Charge ordering types
- Phase diagram
- Transport

3. Transport at large bias

- Experimental results
- Transport model

Experimental Setup



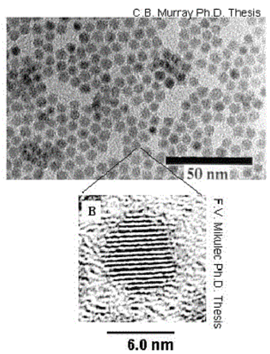


image from N.Y. Morgan, PhD Thesis (MIT, 2001)

- 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:

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

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

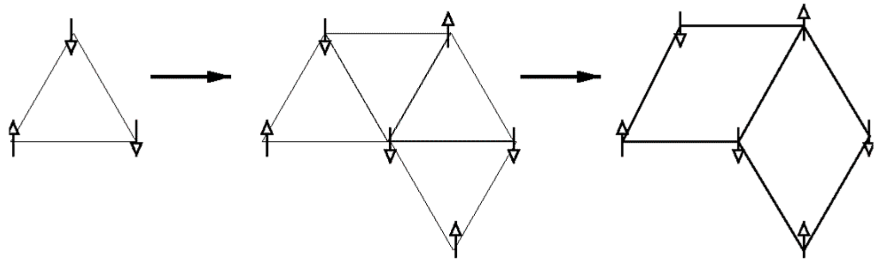
$d/a \sim 2 - 10$

- Incoherent hops between neighboring sites

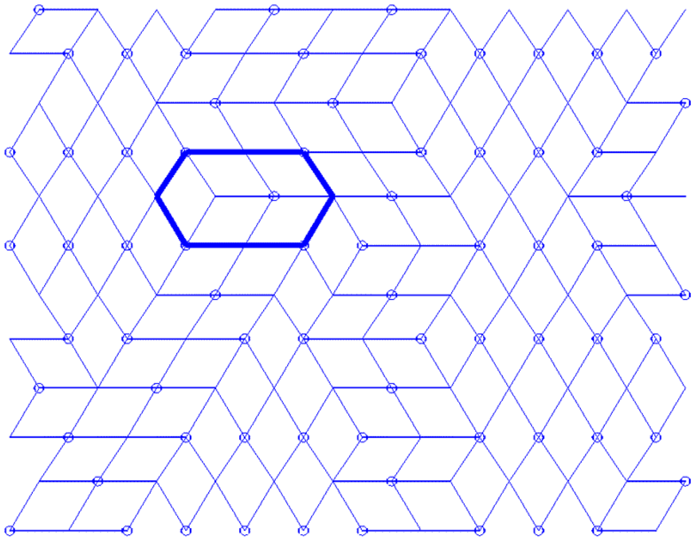
Map on long range $\triangle$IAFM

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

Frustrated Lattice

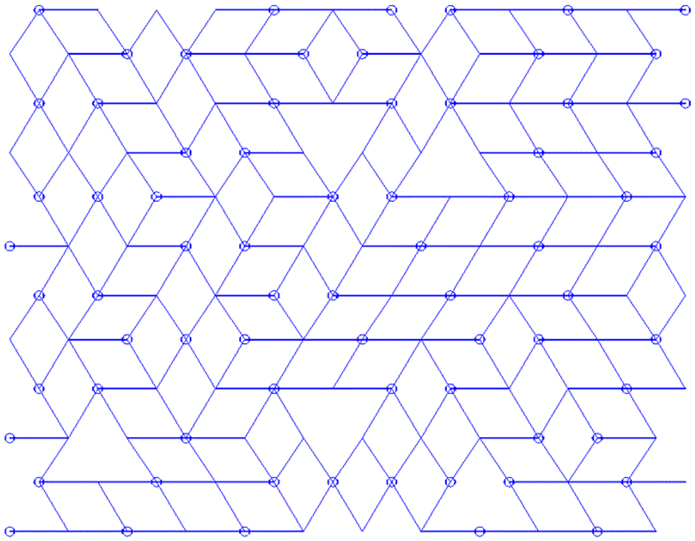


Height Field, $T = 0$



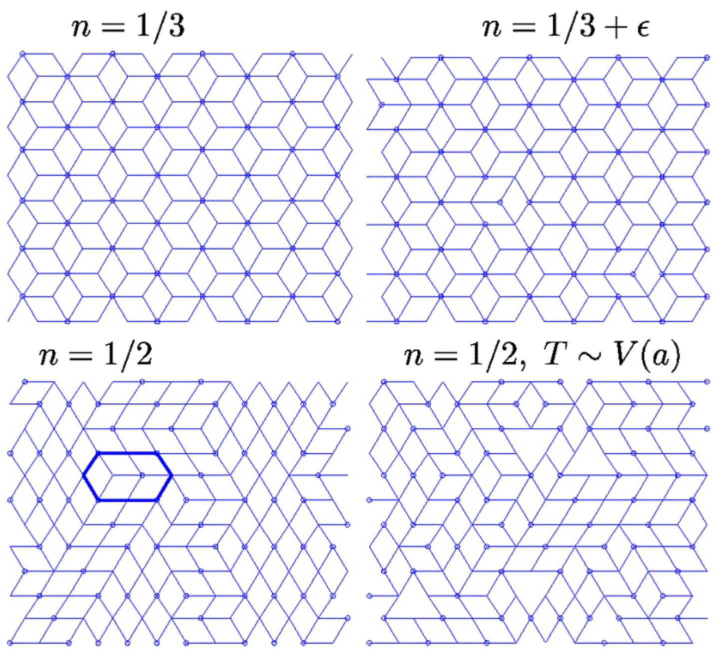
- Minimize # of frustrated bonds: pair elementary triangles $\rightarrow$ rhombi
- Covering by rhombi $\leftrightarrow$ Map to SOS
- Correlated hops

Height Field, $T > 0$



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

Charge Ordering via Height Field

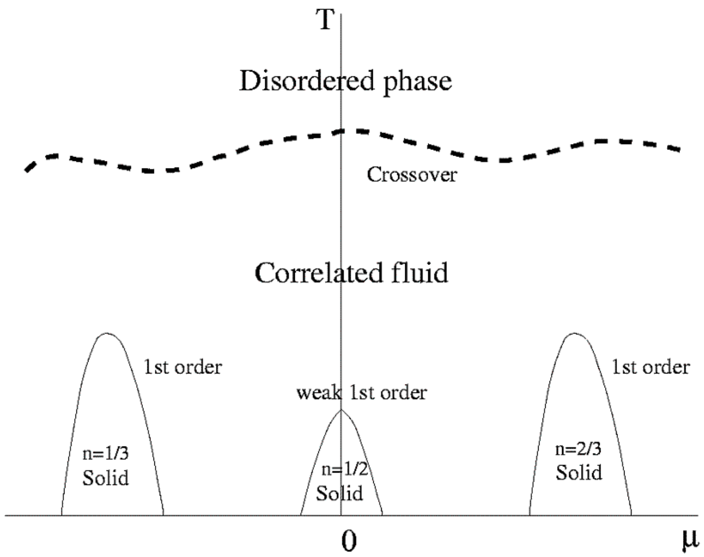


$\triangle$IAFM $\rightarrow$ Height Field $\rightarrow$ Roughening:

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

$\kappa_{\triangle\mathrm{IAFM}} = \pi/9$ 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* $\to$ *commensurate*: 1st order
- Height *globally not defined* at $T > T_{\mathrm{freezing}}$
- Unbound dislocations; no B.K.T. transition

Correlated Phase

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

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

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

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

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

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

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

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

- **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

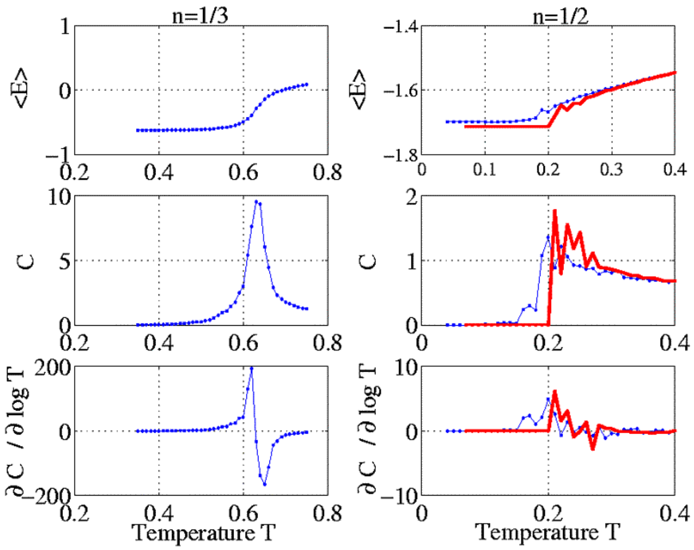
- T ~ Next Nearest Neighbor coupling: Commensurate phase, finite entropy
- Nearest Neighbor: extensive entropy, no freezing!
- Ground state: const average tilt of a height field
- Singularity in $\sigma(T)$ ~ singularity in $\langle E \rangle$
- $n = 1/3$: First order
  $\delta n_i = n_i - 1/9$, symmetry 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 = $C_{3v} \times$ translations

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

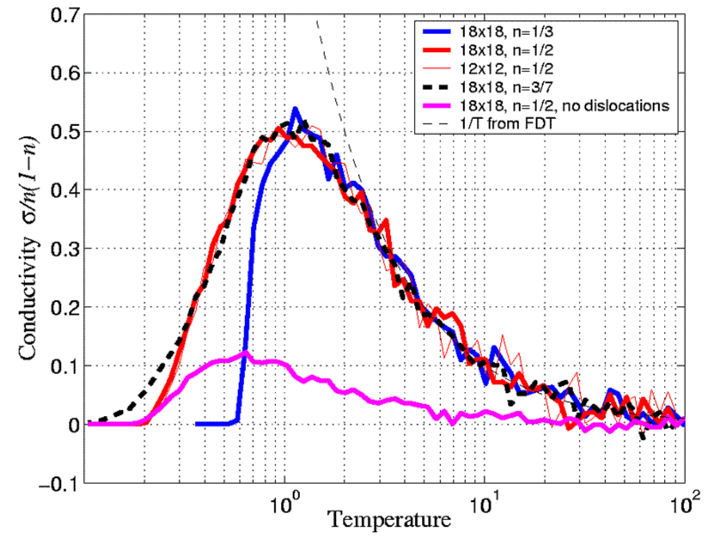
Freezing Phase Transitions



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
$\langle E \rangle = M_1$, $C = \frac{\partial \langle E \rangle}{\partial T} = \frac{M_2}{T^2}$, $\frac{\partial C}{\partial \ln T} = \frac{M_3 - 2TM_2}{T^3}$

Conductivity vs Temperature



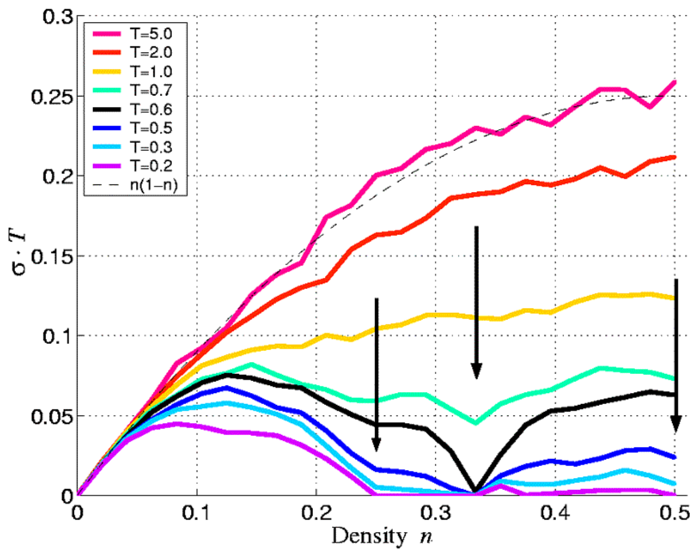
FDT: $\int dt \, \langle j_\mu(t) j_\nu(0) \rangle = 2\sigma_{\mu\nu} T$

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

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:

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

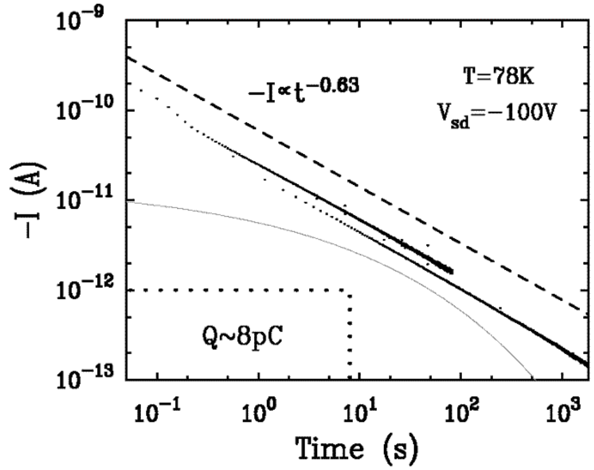
Conclusions so far...

- 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 ∼0.5 V / dot)

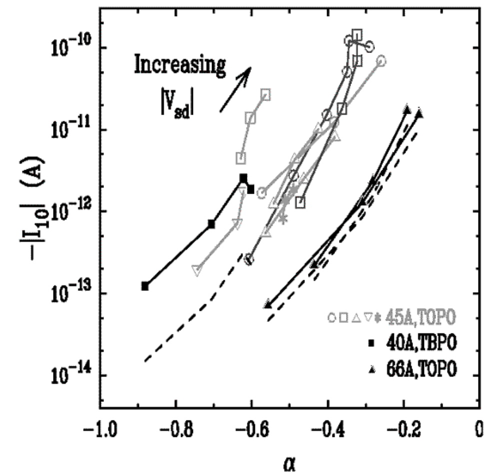
What's observed?

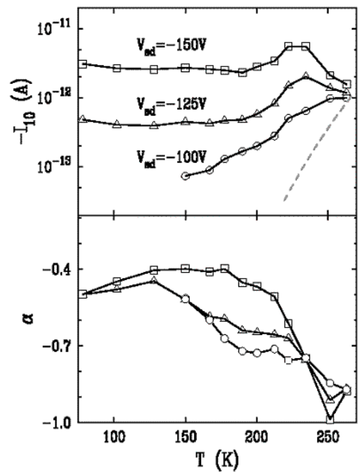


N.Y. Morgan *et al.*, cond-mat/0204560

- $I = I_0\, t^{-\alpha}$ 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 ⇒ smaller I_0, same α

Non-universal power law





α is non-universal!

Non-Stationary Response

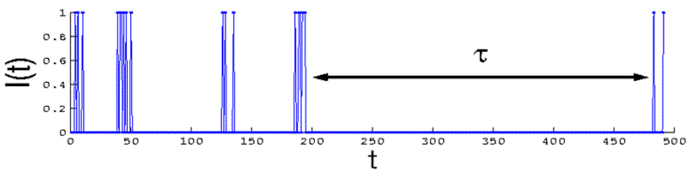
Non-stationary system
(charging? dots move? cool down?)

Stationary system (!)
(Anomalous diffusion)

... + any mix of the above?

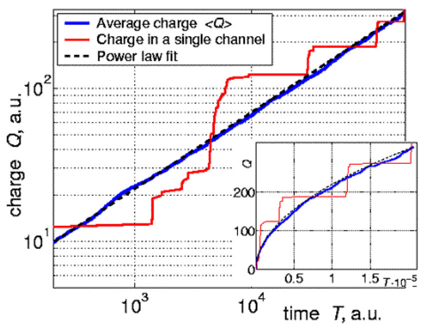
Phenomenology

- $N \gg 1$ independent conducting channels



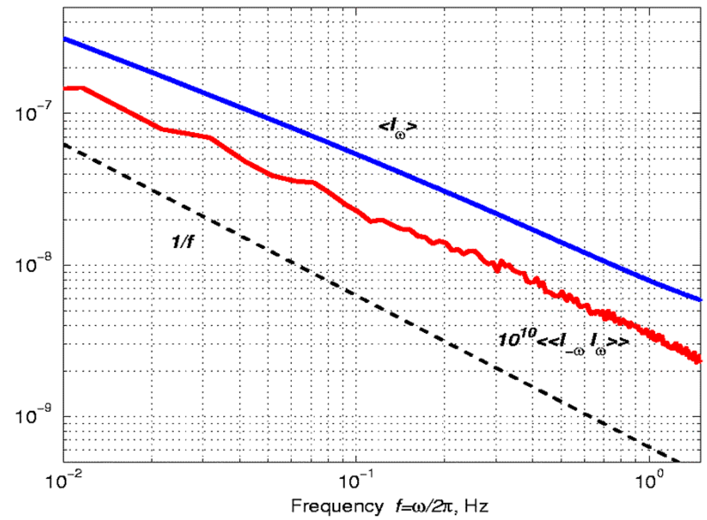
- Single channel: *Stationary* Lévy process

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



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

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

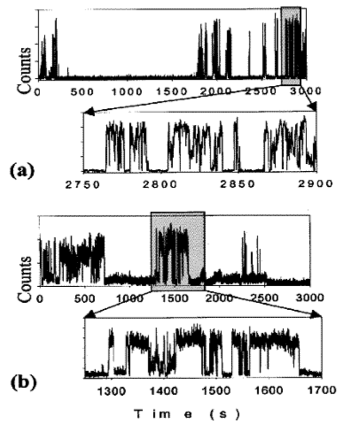


FIG. 1. Typical intensity time trace of CdSe(ZnS) QD fluorescence intermittency at (a) RT and (b) 10 K. Expanded view illustrates the similar nature of the blinking events at RT and 10 K but with different time scales.

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:

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

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):

$$P(t) = A t^{-\alpha}. \quad (2)$$

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

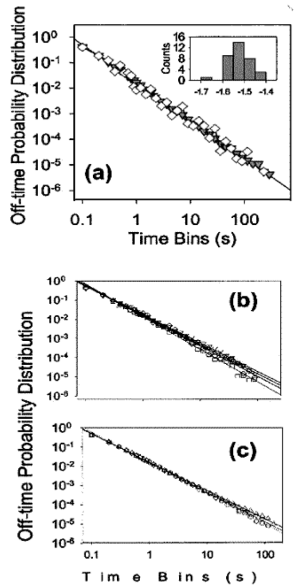


FIG. 2. (a) Normalized *off*-time probability distribution for one CdSe QD (◇) and average of 39 CdSe QD's (▼). Inset shows the distribution of fitting values for the power-law exponent in the 39 CdSe QD's. The straight line is a best fit to the average distribution with exponent ~ -1.5 (b) Average *off*-time 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...