Aging Wiener-Khinchin Theorem

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Niemann, Kantz, EB PRL (2013)
Sadegh, EB, Krapf NJP (2014)
Leibovich, EB PRL (2015)
Leibovich, Dechant, Lutz, EB PRE (2016)
Leibovich, EB PRE (2017)

Gdansk

Outline

- $1/f^{\beta}$ noise and the low frequency cutoff paradox (Mandelbrot).
- Aging Wiener-Khinchin theorem.
- Noise on the nano-scale: the conditional spectrum.
- Sample fluctuations for blinking quantum dot model.

$1/f^\beta$ noise and the infrared catastrophe

- Low frequency $1/f^\beta$ power spectrum is widely observed with $0 < \beta < 2$ (1925-2018).

- If $\beta \geq 1$ the apparent total energy is infinite

$$\int_0^\infty S(f)df = \infty \quad \text{if} \quad \beta \geq 1.$$

- The total energy cannot be infinite if the underlying process is bounded.

Power spectral density

- Sample spectrum

$$I_t(\omega) = \int_0^t I(t') \exp(-i\omega t') dt'$$

$$S_t(\omega) = \frac{|I_t(\omega)|^2}{t}$$

- If the process is stationary use Wiener-Khinchin theorem.

$$\boxed{\langle S(\omega) \rangle = \int_{-\infty}^{\infty} e^{i\omega\tau} \langle I(t+\tau) I(t) \rangle d\tau}$$

- The power spectrum is then normalizable.

Low frequency paradox (physics)

- If $I(t) = x_0 \cos(\omega_0 t)$, i.e., one mode.

$$S(\omega) \propto |x_0|^2 \delta(\omega - \omega_0).$$

- $|x_0|^2$ is proportional to the energy of the mode.
- Sum over all the modes cannot be infinite.
- $1/f$ noise implies non countable normal modes.

Low frequency paradox (math)

$$\int_{-\infty}^{\infty} S_t(\omega) d\omega = \frac{1}{t} \int_{-\infty}^{\infty} d\omega \int_0^t dt_1 \exp(-i\omega t_1) I(t_1)$$

$$\times \int_0^t dt_2 \exp(i\omega t_2) I(t_2) =$$

$$\underbrace{\frac{2\pi}{t} \int_0^t I^2(t_1) dt_1}_{\text{Variance}} < 2\pi (I_{max})^2$$

- The total energy of *bounded* stationary or non-stationary process, is finite.
- So how do we observe non-integrable $1/f$ noise?

Possible solution

- Total energy must be finite

$$S_t(\omega) \sim \frac{\omega^{-\beta}}{t^z}$$

- z is called the aging exponent

$$\int_{1/t}^{\infty} S_t(\omega) d\omega \propto t^{-z} t^{-1+\beta} = \text{const.}$$

- A relation between the exponents, based on the finite value of the total power

$$z = \beta - 1.$$

Stationary or Non-stationary that is the question

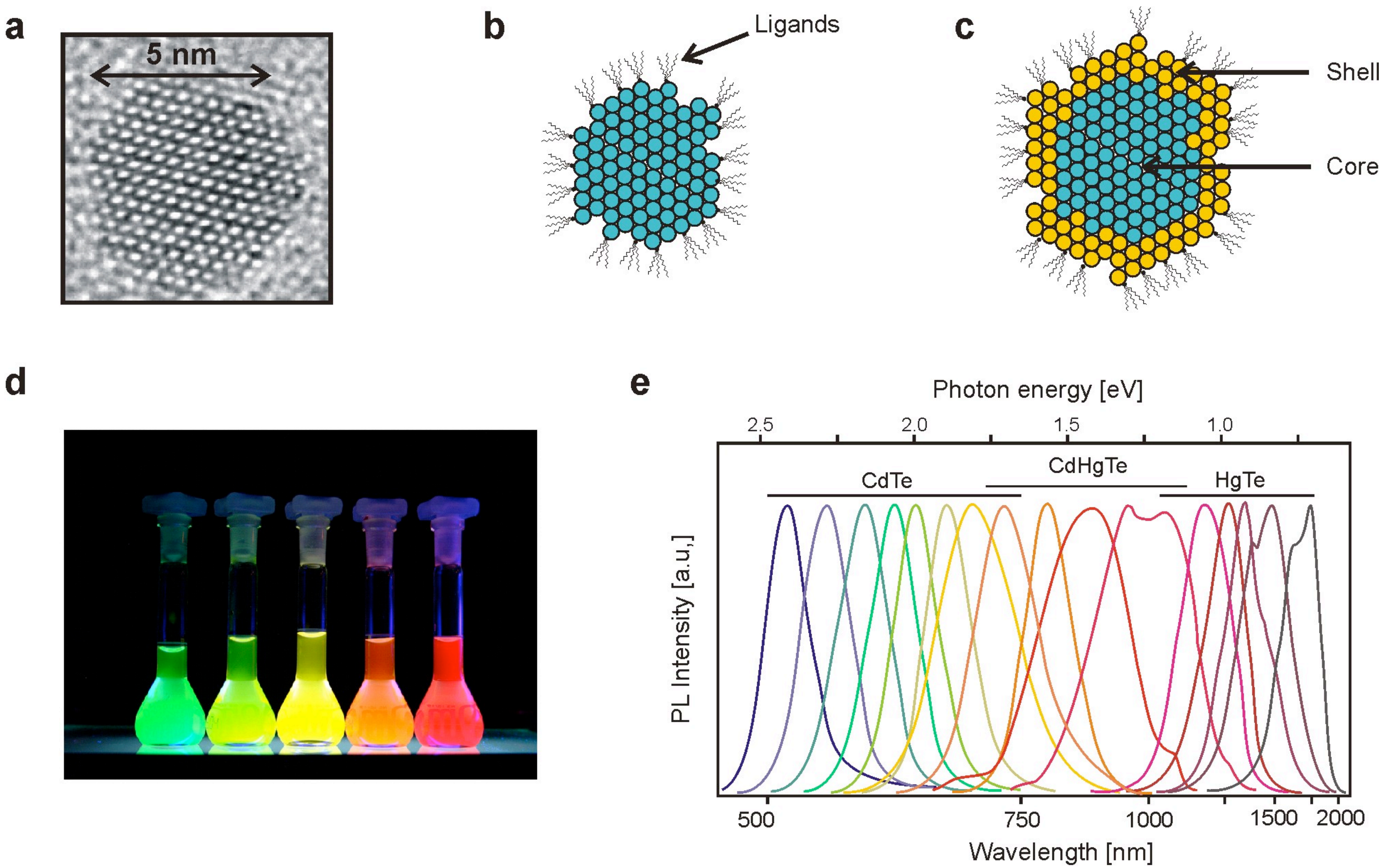
• **A blind extrapolation of $f^{-\beta}$ $(\beta \geq 1)$ to $f = 0$ incorrectly suggests that the total energy is infinite (infrared catastrophe) ... If $\beta \geq 1$ one needs a non-Wienerian spectral theory to account for $f^{-\beta}$ noise.** *Mandelbrot IEEE* 1967.

Non-stationary: **Manneville 1980** Intermittency, and **Bouchaud, Cugliandolo, Kurchan, Mezard 1997** Spin Glasses.

• **As is customary in statistical physics, we shall henceforth assume that the noise process is stationary, for simplicity, and in the absence of overwhelming evidence to the contrary.** *Dutta and Horn RMP 1981*.

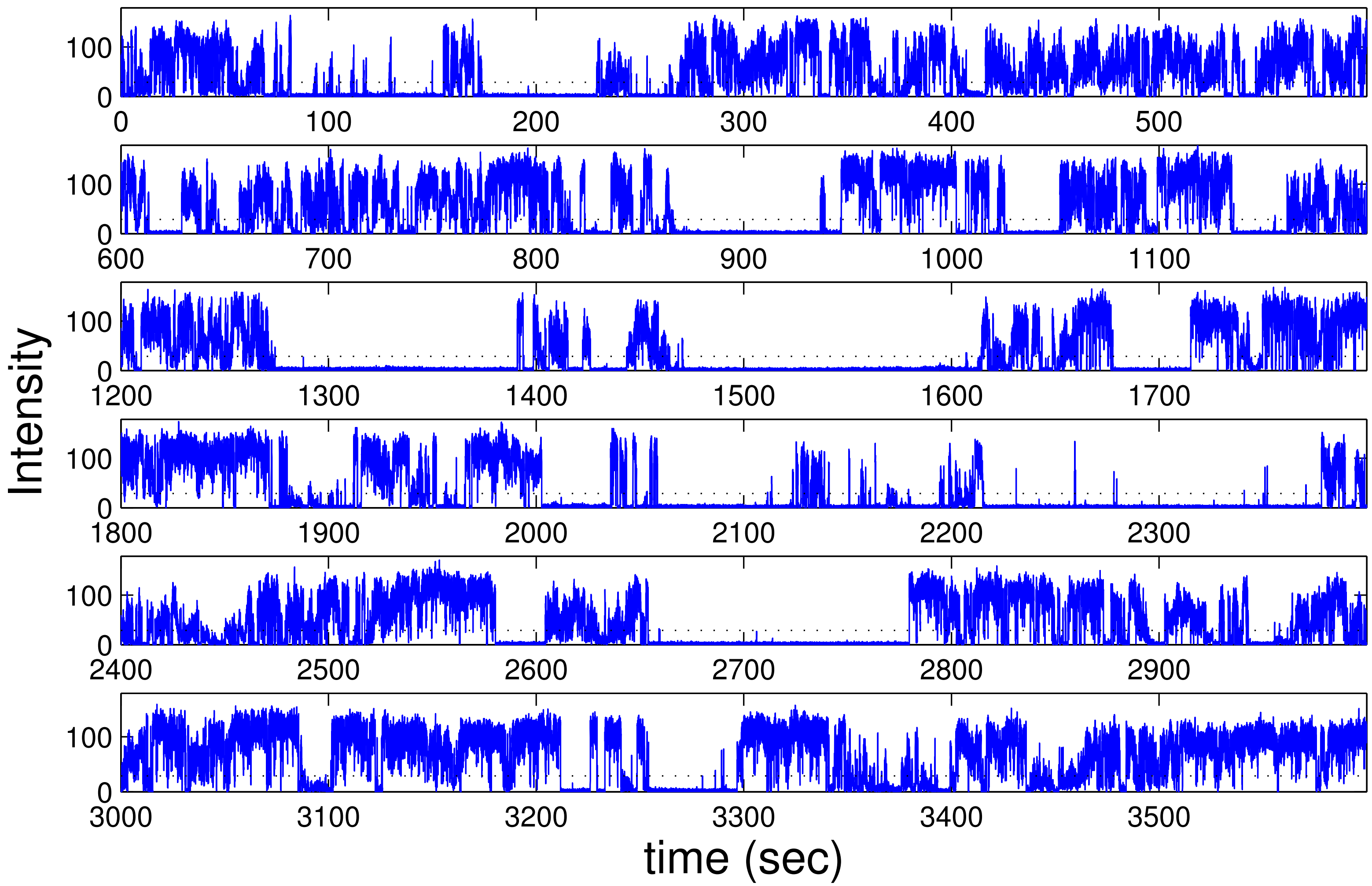
The statistical properties of 1/f noise in physical sources are fully consistent with the assumption of stationarity. **Stoisiek and Wolf 1975**

Quantum Dots

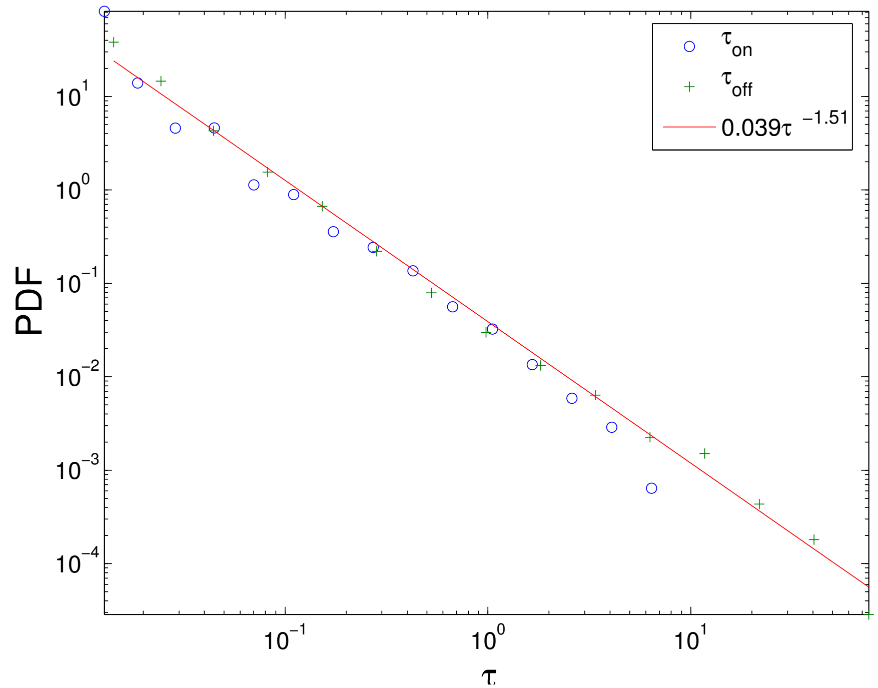


Stefani, Hoogenboom, Barkai **Physics Today** 62, 34 (2009).

Blinking Nano Crystals (coated CdSe)



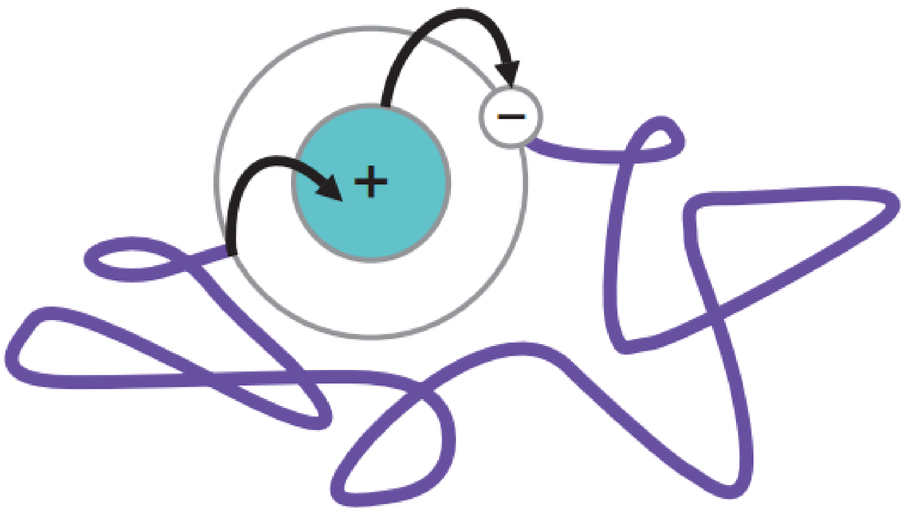
Power Law Distribution of on and Off times



Power law waiting time $\psi(\tau) \sim \tau^{-(1+\alpha)}$.

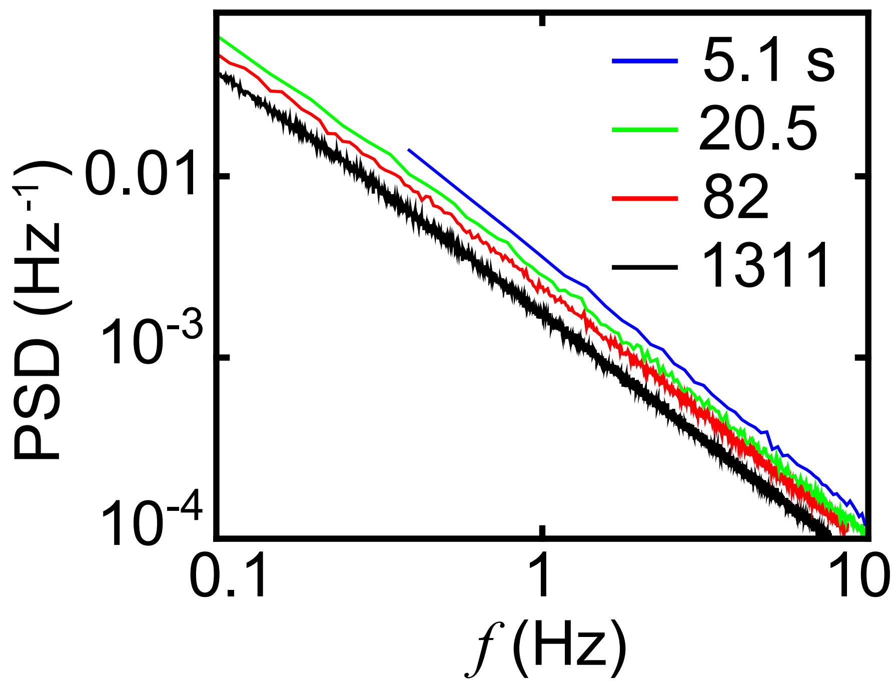
Averaged time in States On and Off is infinite $\langle \tau \rangle = \infty$.

Physical explanation for blinking



Stefani, Hoogenboom, Barkai **Physics Today** 62, 34 (2009).

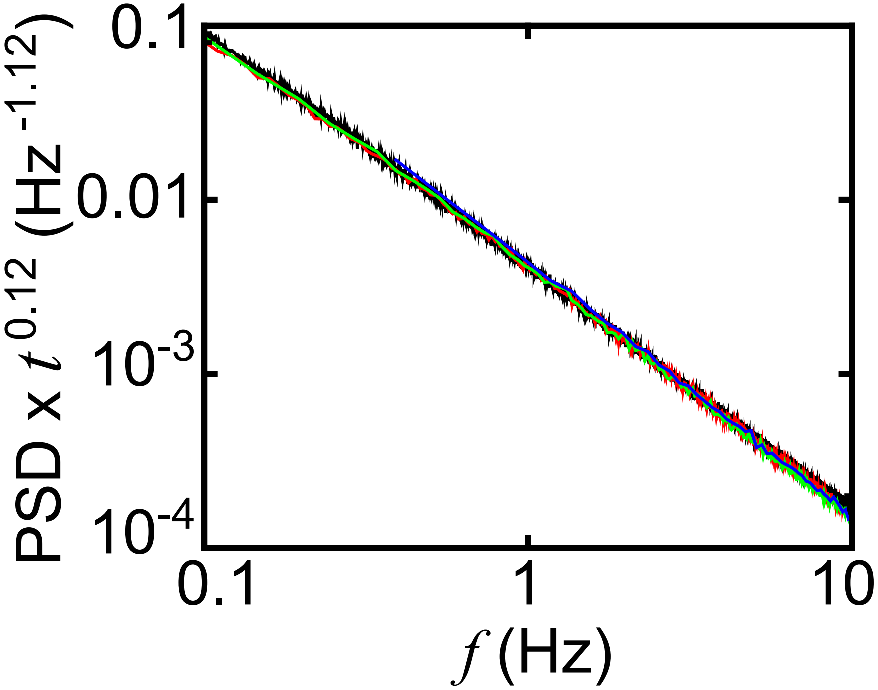
Measured Power Spectrum Ages



Sadegh, EB, Krapf **NJP** (2014)

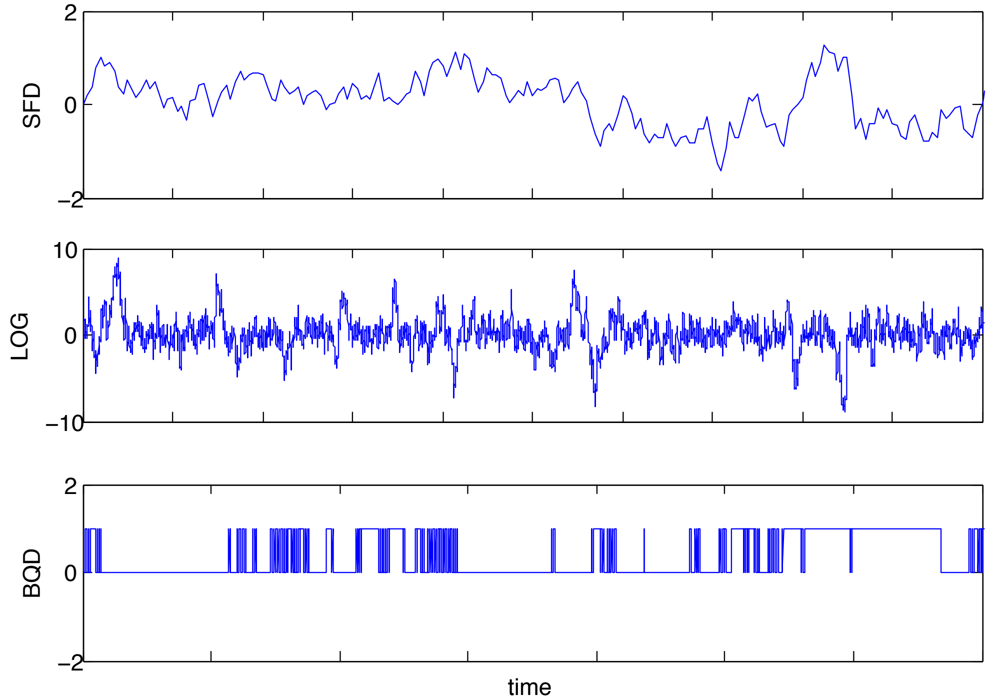
Two other recent experiments show similar behavior!

Power Spectrum $\times t^z$



Sadegh, EB, Krapf **NJP** (2014) Experiment

Mechanisms of $1/f$ noise



Long range correlations induced by many body effects, power law intermittency, burstines in amplitude, multiplicative noise, distributed kinetics, yield $1/f$ noise.

Aging Wiener Khinchin Theorem

$$t_m \langle S_{t_m}(\omega) \rangle = \int_0^{t_m} dt_1 \int_0^{t_m} dt_2 e^{i\omega(t_2 - t_1)} \langle I(t_1) I(t_2) \rangle. \tag{1}$$

$$\langle S_{t_m}(\omega) \rangle = \frac{2}{t_m} \int_0^{t_m} d\tau (t_m - \tau) \langle C_{TA}(t_m, \tau) \rangle \cos(\omega\tau) \,. \tag{2}$$

$$C_{TA}(t_m, \tau) = \frac{1}{t_m - \tau} \int_0^{t_m - \tau} dt_1 I(t_1) I(t_1 + \tau). \tag{3}$$

$$\langle C_{TA}(t_m, \tau) \rangle = (t_m)^\gamma \varphi_{TA}(\tau / t_m) \,, \tag{4}$$

$$\langle S_{t_m}(\omega)\rangle = 2(t_m)^{1+\gamma}\int_0^1 d\tilde{\tau}\,(1-\tilde{\tau})\,\varphi_{TA}\,(\tilde{\tau})\cos\left(\omega t_m \tilde{\tau}\right). \tag{5}$$

$$\langle I(t+\tau)I(t)\rangle = t^{\gamma}\phi_{EN}(\tau/t). \tag{6}$$

$$\varphi_{TA}(x) = x^{\gamma} y(x)\int_{y(x)}^{\infty}\frac{\phi_{EA}(z)}{z^{2+\gamma}}dz \qquad y(x) = 1/(1-x) \tag{7}$$

$$\langle S_{t_m}(\omega)\rangle = 2t_m\int_0^1 \phi_{EA}\left(\frac{x}{1-x}\right)\frac{\tilde{\omega}x\sin(\tilde{\omega}x)+\cos(\tilde{\omega}x)-1}{(\tilde{\omega}x)^2}dx \tag{8}$$

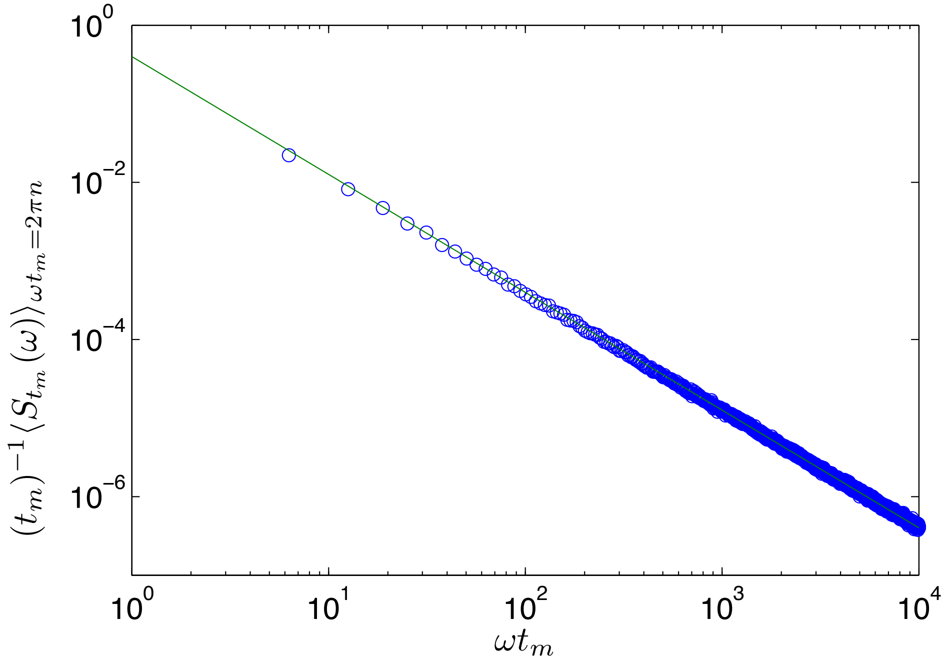
$$\langle S_{t_m}(\omega)\rangle =$$

$$\frac{2(t_m)^{\gamma+1}}{2+\gamma}\int_0^1 (1-x)^{\gamma}\phi_{EN}\left(\frac{x}{1-x}\right){}_1F_2\left(1+\frac{\gamma}{2};\frac{1}{2},2+\frac{\gamma}{2};-\left(\frac{\tilde{\omega}x}{2}\right)^2\right)dx.$$

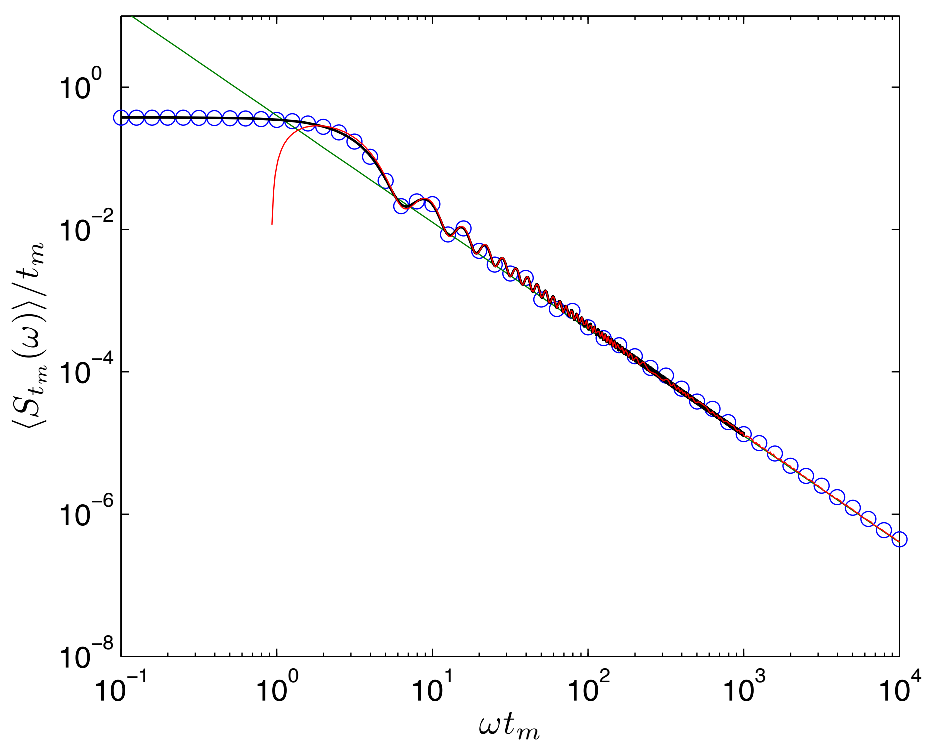
Leibovich, EB **PRL** (2015)

Leibovich, Dechant, Lutz EB **PRE** (2017)

Blinking dot power spectrum on natural frequencies



Blinking dot power spectrum beyond the natural



Model		γ	ν
Unilayer Parisi's Tree	$0<\alpha<1$	0	$\alpha-1$
Blinking Quantum Dot	$0<\alpha<1$	0	$1-\alpha$
Laser-Cooled Atoms	$1<\alpha<3$	$2-\alpha$	$2-\alpha$
Single-File Diffusion		$1/2$	$1/2$
Generalized Elastic Model	$0<\alpha<1$	α	α
Coupled Oscillators		$[-0.58,-0.4]$	-0.14 ± 0.03 0
1D Growing Interfaces		≈ 0.33	
RC transmission Line	$1<\alpha<2$	$\alpha-1$	$\alpha-1$

Table 1: The aging behavior of several models, where the correlation function is given in terms of $\langle I(t)I(t+\tau)\rangle \sim t^{\gamma}\phi_{EA}(\tau/t)$ and $\phi_{EA}(x)\propto A_{EA}-B_{EA}x^{\nu}$ when $x\ll 1$.

$$\langle S_{t_m}(\omega)\rangle \sim \frac{2\Gamma\left(1+\nu\right)\sin\left(\frac{\pi\nu}{2}\right)B_{EA}}{\left(\gamma-\nu+1\right)\left(t_m\right)^{\nu-\gamma}\omega^{1+\nu}}.$$

Hold your horses

- Measurement of most cond-mat $1/f$ noise sources do not exhibit aging.
- No signature of non-stationary noise.
- Fundamental difference between measurements of macroscopic source of noise and single molecule measurements.
- $\sum S_i \simeq N\langle S\rangle$ is time independent.
- When the number N of fluctuating subunits, *on the time scale of the the measurement time* is increasing with measurement time.

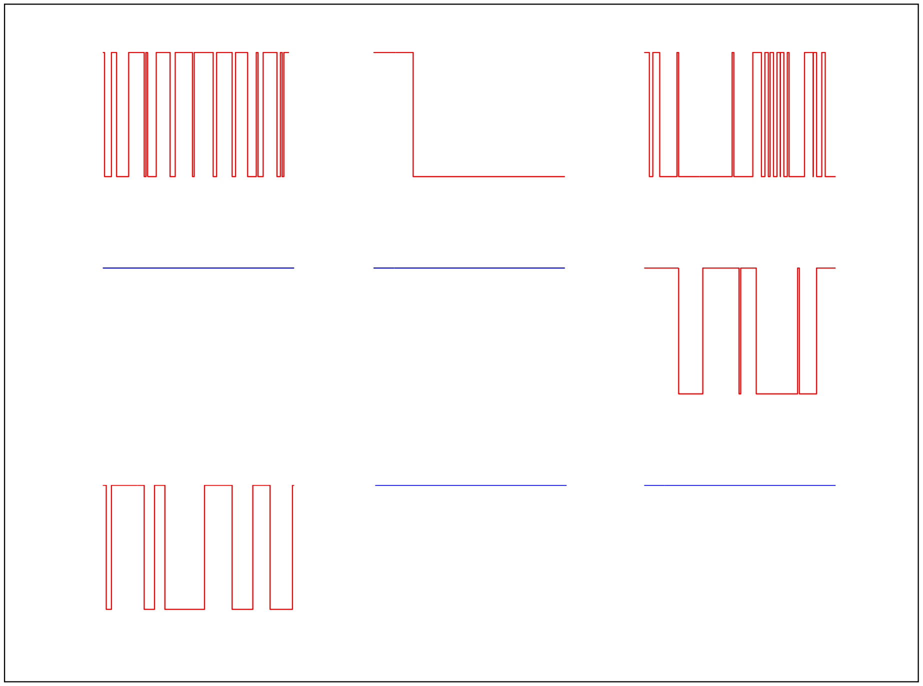
Leibovich EB **PRE** (2017)

Distributed Kinetics: Popular Old Model

- $S(\omega)$ is time independent. $S(\omega) = \int p(\tau) \frac{\tau}{1+\omega^2\tau^2} d\tau$
- Here exponential decay of the correlation function, and distributed kinetics yields $1/f$ noise.
- In experiment no Lorentzian spectrum for single QDs.
- Take $p(\tau) = 1/\tau$.
- But $1/\tau$ is non-normalized.
- So maybe add a cutoff.
- What is the upper limit of the integral?
- Should we put the age of universe (as suggested)?
- Then $S(\omega)$ depends on age of universe? but measurement time is much less then that.
- Answer: upper limit of integral is measurement time.

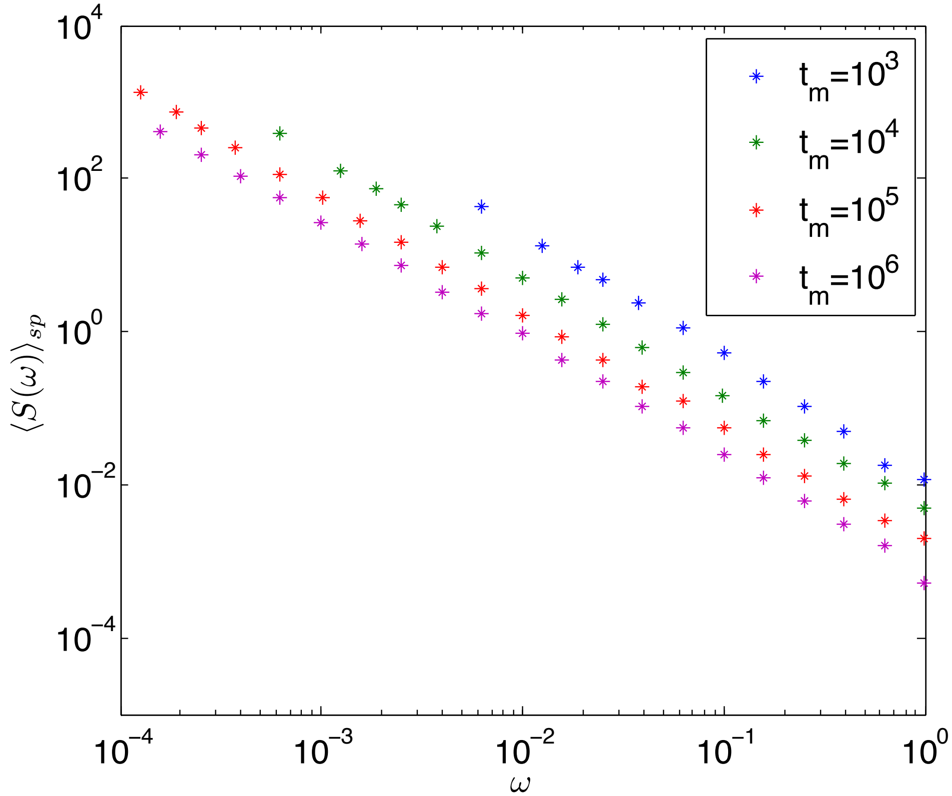
- Then $S(\omega)$ depends on measurement time.
- And exactly in the way we claim $\beta - 1 = z$.
- How many fluctuating objects?
- $N \sim t^z$
- So $N\langle S\rangle$ is time independent and also independent of the non-relevant age of the universe. As observed in many macroscopic experiments.

Distributed Kinetics Model



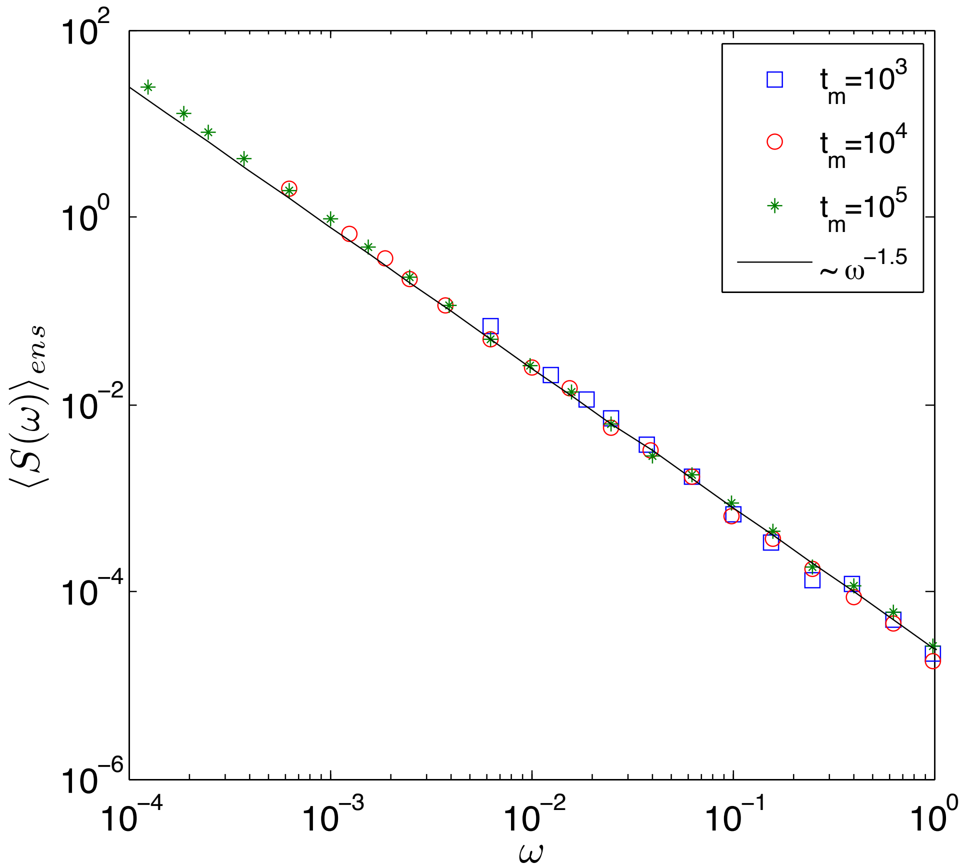
Number of fluctuators increases with measurement time

Aging PSD in Single Molecule Measurements



Distributed kinetics model.

No Aging in Macroscopic Measurements



Distributed kinetics model.

Ergodicity and the sample power spectrum

Non-stationarity means that underlying process is non-ergodic.

Sample spectrum is **not** equal to its ensemble average.

The amplitude of $1/f$ noise is random, varies from one measurement to the other.

This is related to non-stationarity and non-ergodicity.

Niemann, Kantz, EB **PRL** (2013) Theory

Fluctuations of the power spectrum

- The average decreases with measurement time

$$\langle S_t(\omega)\rangle \simeq \frac{C}{t^{1-\alpha}}\frac{1}{\omega^{2-\alpha}}$$

- The value of $S_t(\omega)$ is fluctuating

$$\left(\frac{S_t\left(\omega_1\right)}{\langle S_t\left(\omega_1\right)\rangle},\cdots,\frac{S_t\left(\omega_n\right)}{\langle S_t\left(\omega_n\right)\rangle}\right)\to Y_\alpha\left(\xi_1,\cdots,\xi_n\right)$$

- Y_α has a normalized Mittag-Leffler distribution.
- The ξ_i are independent exponential random variable with mean 1.
- The whole spectrum has a common random prefactor Y_α.

Motivation of the result

$$\int_0^t d\tau \exp(i\omega\tau) I(\tau) \simeq \sum_{j=1}^{n(t)} d_j(\omega)$$

$$d_j(\omega) \simeq i\chi_j \exp(i\omega T_j) \frac{1 - \exp(i\omega\tau_j)}{\omega}$$

- With this approximation

$$S_t(\omega) \simeq \frac{1}{t} \sum_{k,l=1}^{n(t)} d_k(\omega) d_l(-\omega)$$

- Assuming that $n(t)$ is independent of waiting times

$$\langle S_t(\omega)\rangle \simeq \frac{\langle n(t)\rangle}{t}\langle d_l(\omega)d_l(-\omega)\rangle,$$

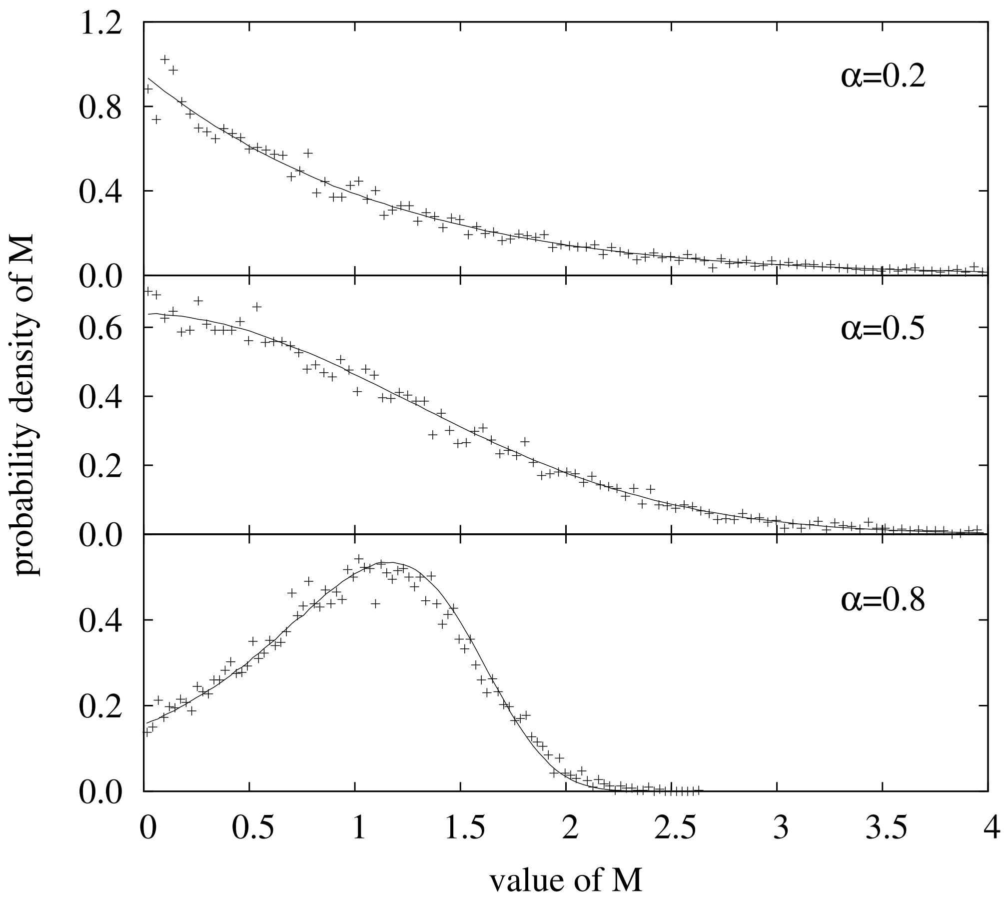
$$\langle S_t(\omega)\rangle \simeq (I_0)^2\ \frac{\langle n(t)\rangle}{t}\ \frac{2-\hat{\psi}(i\omega)-\hat{\psi}(-i\omega)}{\omega^2}$$

- The spectrum is time dependent since $\langle n(t)\rangle \sim t^{\alpha}$.
- We get $\beta = 2-\alpha$ and $z = 1-\alpha$ so $\beta = z+1$.
- Repeat this for $\langle S_t(\omega_1)\cdots S_t(\omega_2)\rangle$.
- The Mittag-Leffler distribution describes number of renewals $n/\langle n(t)\rangle$.
- To isolate the Mittag Leffler fluctuations

$$M = \frac{1}{N}\sum_{j=1}^{N}\frac{S_t(\omega_j)}{\langle S_t(\omega_j)\rangle}$$

with large N.

Mittag Leffler distribution



Niemann, Kantz, EB **PRL** (2013) MMNP (2016) Theory

Summary

- Critical exponents describe the spectrum of blinking dots and models of $1/f$ noise.
- Power spectrum ages, the longer the measurement time the noise is reduced.
- This solves the low frequency paradox, the total power is a constant.
- Fundamental difference between measurement of noise on the single molecule level, if compared with macroscopical measurements.
- Non-stationarity implies non-ergodicity, which was quantified.
- Aging Wiener Khinchin theorem: a tool for the calculation of power spectrum.
- A wide variety of mechanism responsible for $1/f$ noise: intermittency, single file diffusion and large amplitude burstiness. The unifying theme are scale invariant correlation functions.

Ref. and Thanks

- M. Niemann, H. Kantz, E. Barkai *Fluctuations of 1/f noise and the low frequency cutoff paradox* **Phys. Rev. Lett. 110**, 140603 (2013).
- S. Sadegh, E. Barkai, and D. Krapf $1/f$ *noise for intermittent quantum dots exhibits non-stationarity and critical exponents* **New. J. of Physics 16** (2014) 113054.
- N. Leibovich, and E. Barkai *Aging Wiener-Khinchin Theorem* **Phys. Rev. Lett. 115**, 080602 (2015).
- N. Leibovich, A. Dechant, E. Lutz, and E. Barkai *Aging Wiener-Khinchin theorem and critical exponents of* $1/f^{\beta}$ *noise* **Phys. Rev. E. 94**, 052130 (2016).
- N. Leibovich, and E. Barkai *Conditional* $1/f^{\alpha}$ *noise: from single particles to macroscopic measurements* **PRE** (2017).

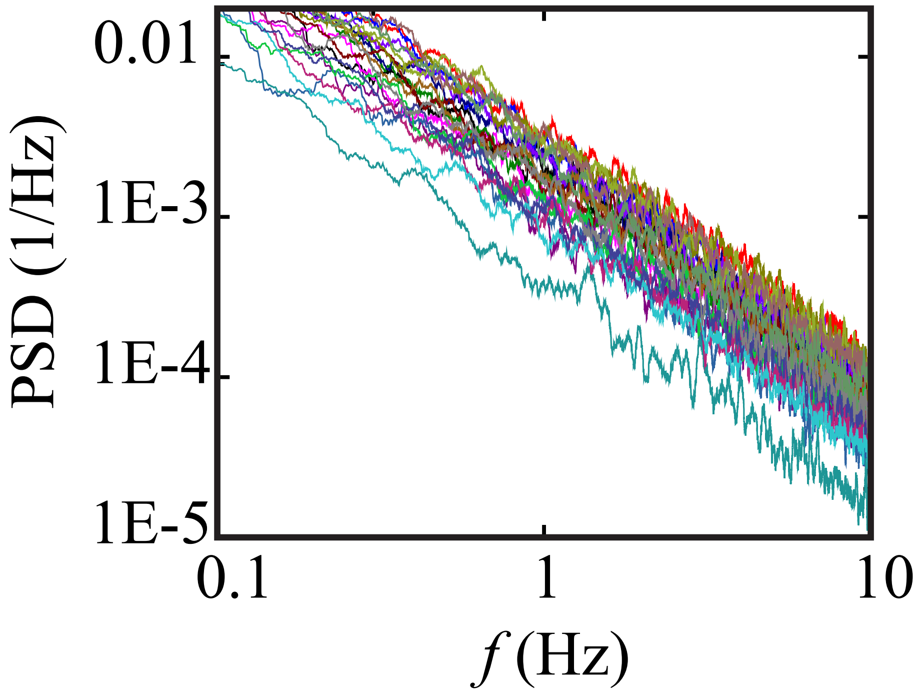
REF

- N. Leibovich, and E. Barkai $1/f^\beta$ *noise for scale-invariant processes: How long you wait matters* **EPJ B** (2017) 90: 229. Topical issue dedicated to Continuous Time Random Walk still trendy: Fifty-year history, current state, and outlook - edited by Ryszard Kutner and Jaume Masoliver.

- M. Niemann, E. Barkai, and H. Kantz *Renewal theory for a system with internal states* **Mathematical Modelling of Natural Phenomena** 11 3 (2016) 191-239.

- Nava Leibovich, and excellent PhD student, searching for a post-doc position.

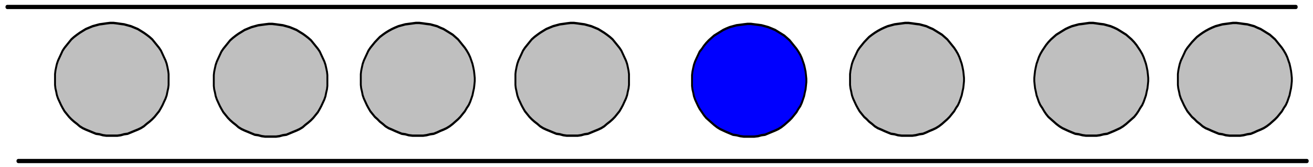
Fluctuations of power spectrum



Amplitude of $1/f$ noise is random, varies from one measurement to the other.

This is related to non-stationarity and non-ergodicity.

Single File Diffusion

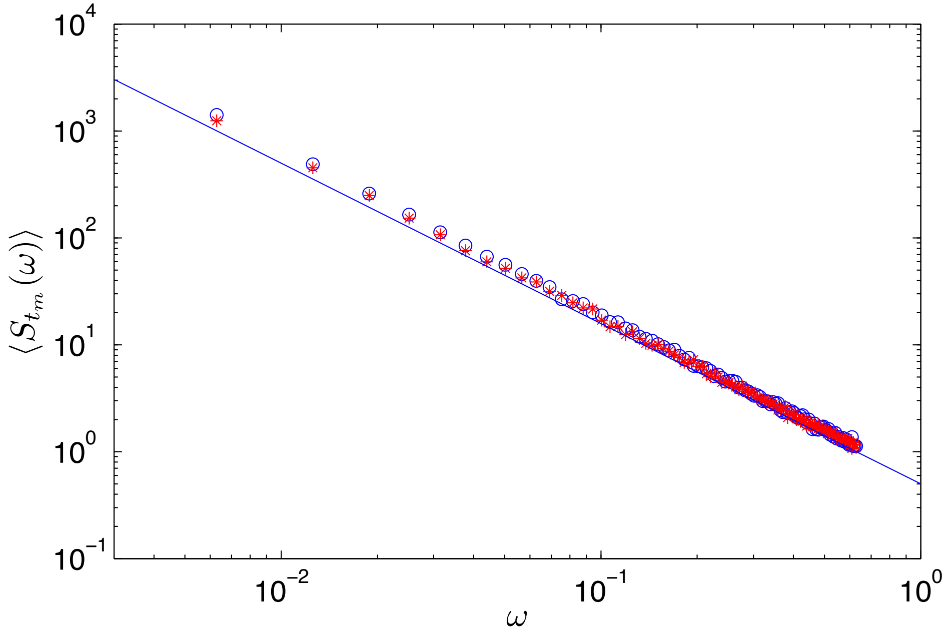


$$\langle x(t+\tau)x(t)\rangle \sim \frac{1}{\rho}\sqrt{\frac{D}{\pi}}t^{1/2}\left(\sqrt{1+\frac{\tau}{t}}+1-\sqrt{\frac{\tau}{t}}\right).$$

$$\boxed{\langle x^2\rangle \sim t^{1/2}}$$

- Hard core point particles, free diffusion between collision events, infinite system, fixed density ρ.
- Harris (65)... Leibovich-Barkai (2013).

PSD single file diffusion



The meaning of the PS

$\langle I \rangle =$ Const as for the WK spectrum.

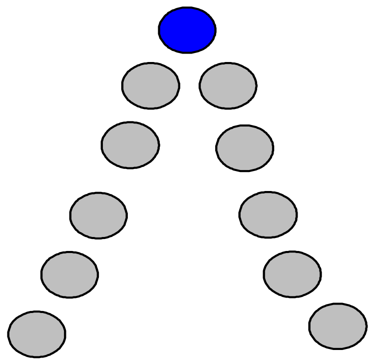
$\langle I(t)I(t+\tau)\rangle = t^{\gamma}\phi_{EN}(\tau/t)$:- scale invariant correlation function.

This gives the nearly standard relation:

$$\int_{-\infty}^{\infty} S_{t_m}(\omega)d\omega = \frac{2\pi\langle I^2\rangle}{1+\gamma}$$

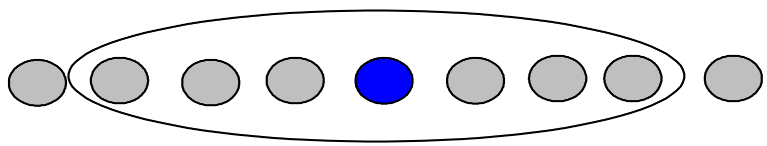
And since $S_{t_m}(\omega) \geq 0$ it is reasonably justified to call it a PS.

Single File Diffusion



$$(x_T)^2 \sim \frac{Dt}{N}$$

Aslangul EPL 1986



$$N \sim \rho\, L(t) \sim \rho\, (Dt)^{1/2}$$

$$(x_T)^2 \sim \frac{(Dt)^{1/2}}{\rho}$$

Harris 65, Levitt 73

Models

		β $S \sim \omega^{-\beta}$	z $S \sim t_m^{-}$
single file diffusion		$3/2$	0
blinking quantum dot - finite mean 'on' time	$0 < \alpha < 1$	α	$1 - \alpha$
blinking quantum dot - infinite mean	$0 < \alpha < 1$	$2 - \alpha$	$1 - \alpha$
logarithmic potential	$1 < \alpha < 2$	$3 - \alpha$	0
	$2 < \alpha < 3$		

Table 2: Summary of the critical exponents for the three systems discussed in sections VI-VIII.

Fluctuations of the power spectrum

- Power spectrum ergodic processes, we expect

$$PDF[S_t(\omega)] \to \delta[S_t(\omega) - \langle S_t(\omega)\rangle]$$

  This is correct, after binning/smoothing.

- For non ergodic process, this will not happen

$$S_t(\omega) \sim \langle S_t(\omega)\rangle C$$

- Where C is random

$$C = Y_\alpha \xi$$

  .

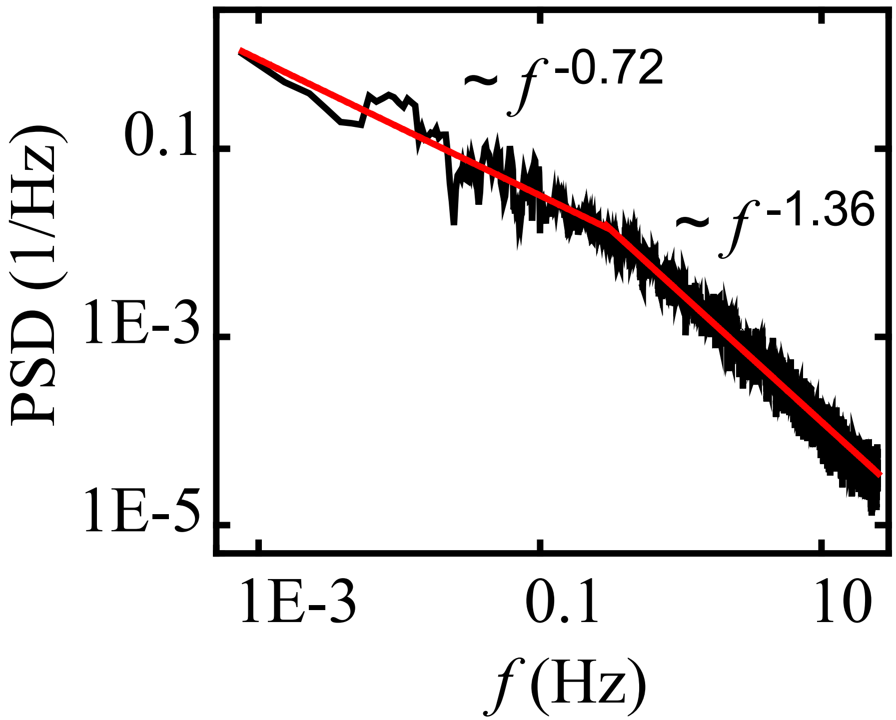
- Y_α (ξ) is a ML (exp) RV.

$$Y_\alpha \to n/\langle n\rangle$$

- If I know the fluctuations of number of transitions, I have the fluctuations of the sample spectrum.

- Even better find PDF of $(S(\omega_1),.....S(\omega_n))$.

Transition frequency



Power spectrum exhibits a transition at $f_c = 0.1$Hz.

This is related to cutoff on the on times.

Things to do

Do we believe in conservation of energy (total power cannot be infinite), a mathematical theorem (bound on total power), on the one hand, and the experimental fact that noise of **many** different sources exhibits non integrable $1/f$ noise.

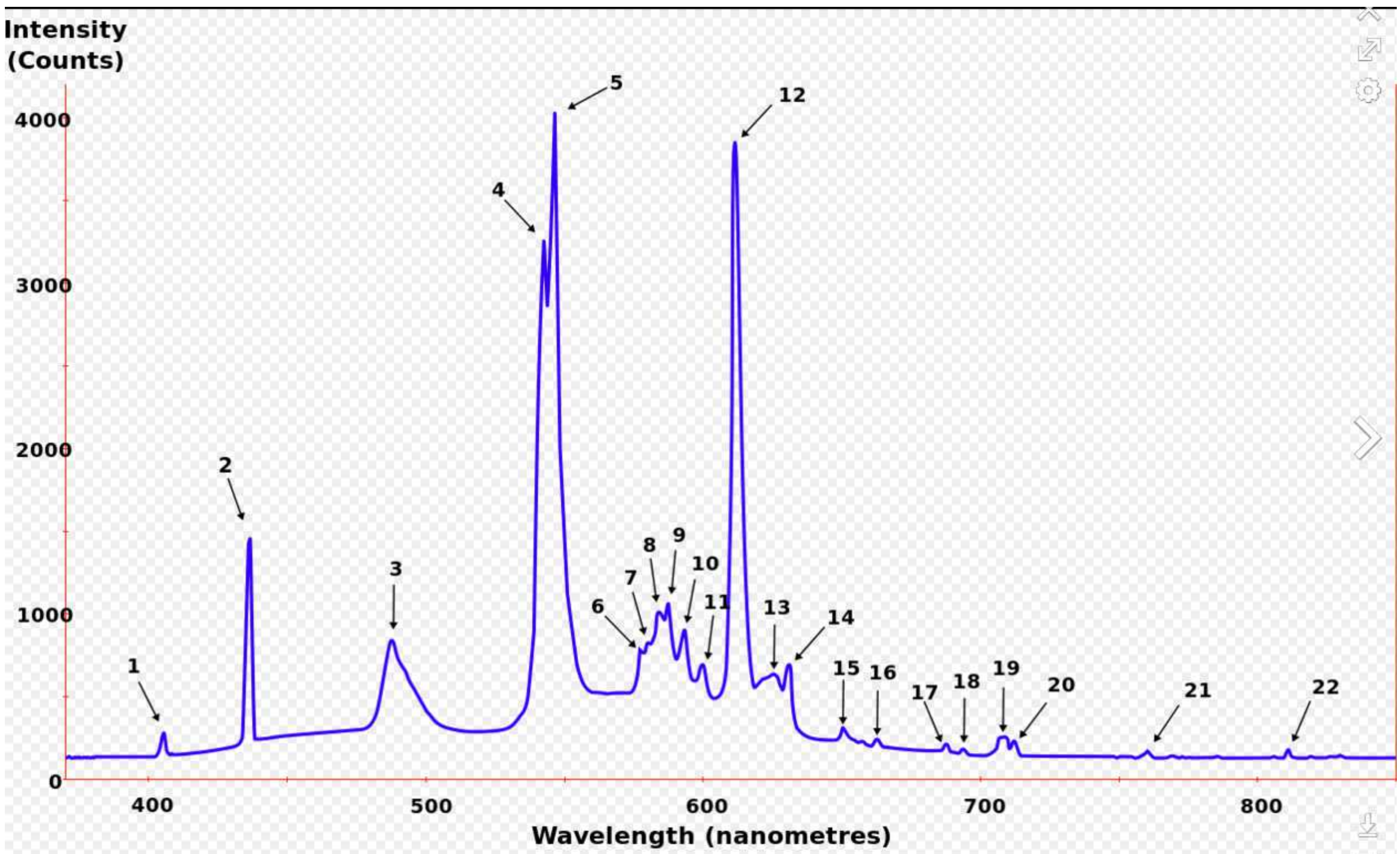
Better, believe in the basic laws of math and physics and the experiments, but throw away the notions of stationary process and ergodicity.

Better explain why N increases with t and why it exactly cancels the aging.

At start list of systems where this phenomena is found.

Add some detail on the derivation, multipoint correlation functions. Show that you actually worked hard on the theory.

Wiki


Intensity
(Counts)
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1000
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400
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Wavelength (nanometres)
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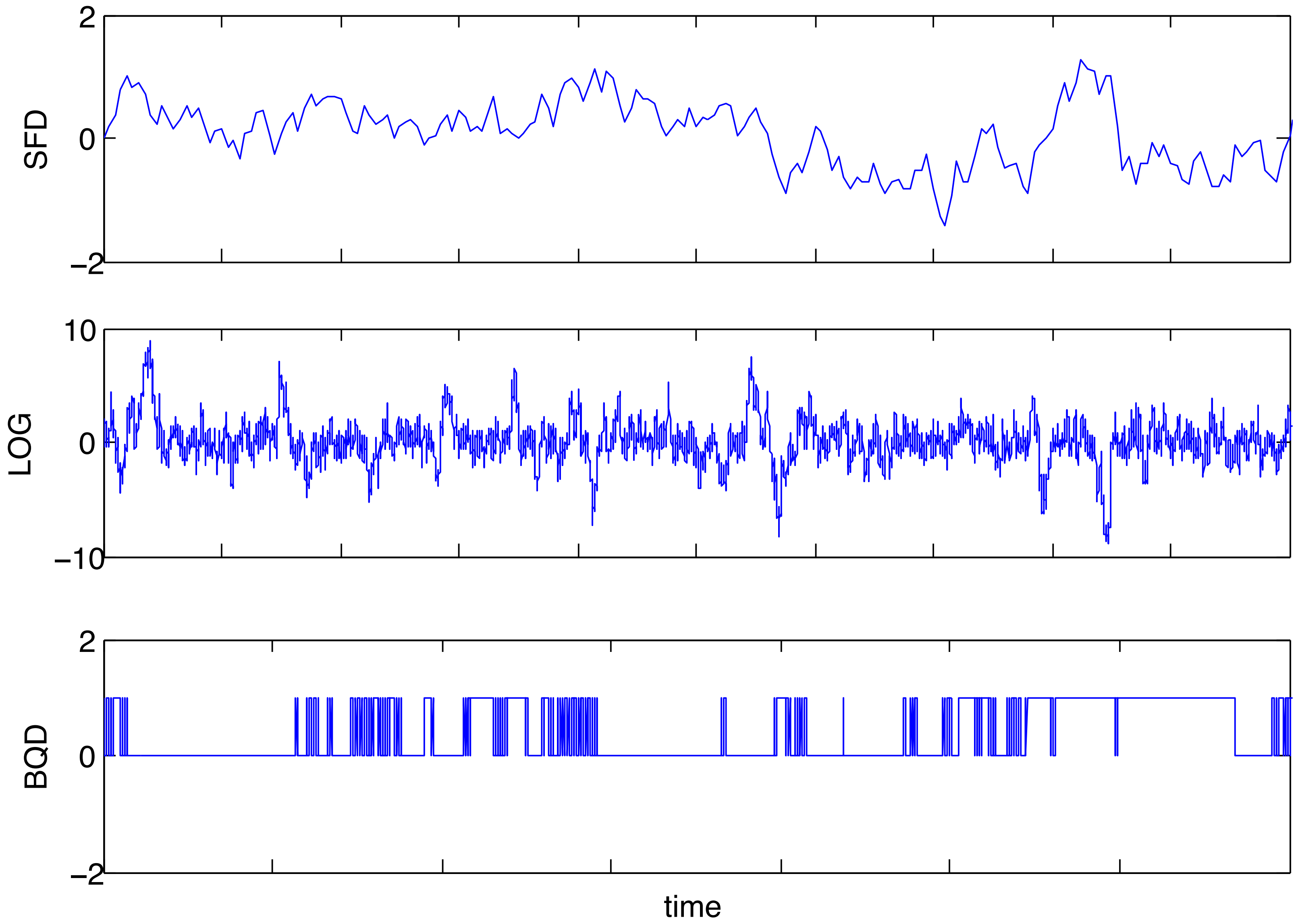
2
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time

Define the two state model better.

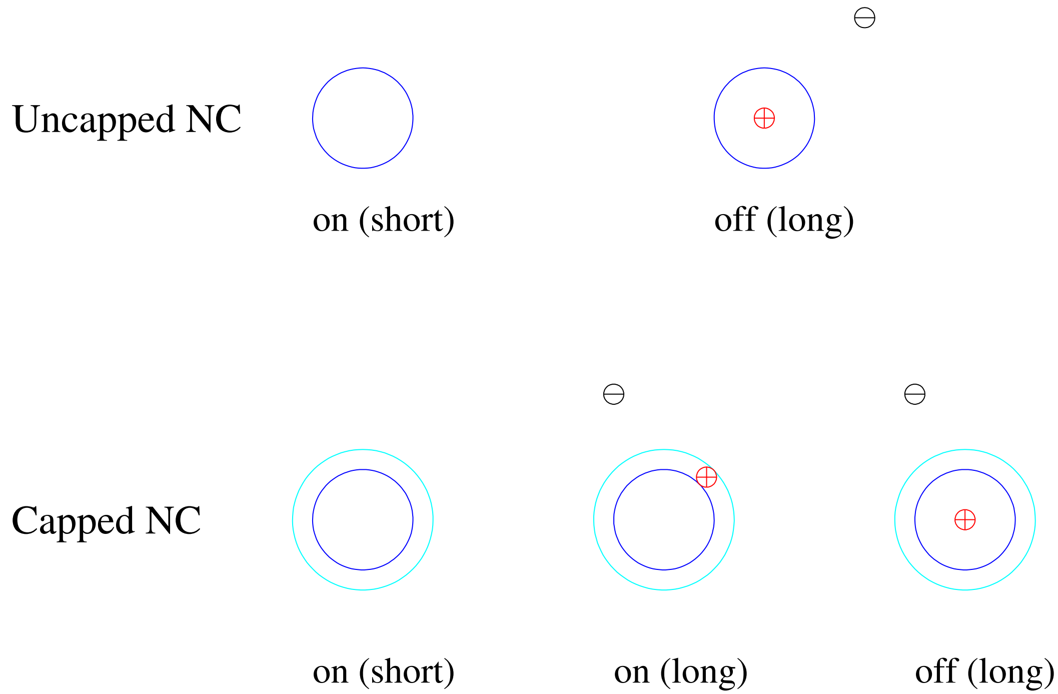
Infinite energy or infinite Power?

History

Wikipedia: In applied mathematics, the Wiener Khinchin theorem, also known as the Wiener Khintchine theorem and sometimes as the Wiener?ÄìKhinchin?ÄìEinstein theorem or the Khinchin?ÄìKolmogorov theorem, states that the autocorrelation function of a wide-sense-stationary random process has a spectral decomposition given by the power spectrum of that process

From the web: This is the Einstein-Wiener-Khinchin theorem (proved by Wiener, and independently by Khinchin, in the early 1930?Äôs, but ?Äî as only recently recognized ?Äî stated by Einstein in 1914). MIT OpenCourseWare Power Spectral Density Chapter 10.

Group	Material	Nu.	Radii	T	α_{on}	α_{off}
Dahan	CdSe-ZnS	215	1.8nm	300 K	$0.58(0.17)$	$0.48(0.15)$
Orrit	CdS		2.85	1.2	EXP	$0.65(0.2)$
Bawendi	CdTe....	200	1.5	$10-300$	$0.5(0.1)$	$0.5(0.1)$
Kuno	CdSe-ZnS	300	2.7	300	$0.8-1.0$	0.5
Cichos	Si			300	$0.8-1.0$	0.5
Ha	CdSe(coat)			300	Exp?	1



Efros, Orrit, Onsager, Hong-Noolandi

$$r_{Ons} = \frac{e^2}{k_b T \epsilon} \simeq 7\text{nm}$$

Critical exponents of nano-crystals

		Theory	Experiment	Simulation
$\beta_>$	$S \sim f^{-\beta}$	$\beta_> = 2 - \alpha$	1.4	1.3
$\beta_<$		$\beta_< = \alpha$	0.7	0.6
z	$S \sim t^{-z}$	$z = 1 - \alpha$	0.12	0.3
γ	$f_c \sim 1/t^{\gamma}$	$\gamma = 1$	0.8	0.8
ω	$S_t(0) \sim t^{\omega}$	$\omega = 1$	0.8	0.8

Aging exponent is lower than expected.

Scaling relations between exponents hold $\gamma(\beta_> - 1) \simeq z$, $\omega = \gamma$ within errors.