\(1/f\) Noise on the nano-scale
Aging Wiener-Khinchin approach

Eli Barkai

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

Pohang (2016)
Outline

- $1/f^\beta$ noise and the low frequency cutoff paradox (Mandelbrot).
- Blinking quantum dots.
- Noise on the nano-scale: the conditional spectrum.
- Aging Wiener-Khinchin theorem.
**$1/f^\beta$ noise and the infrared catastrophe**

- Low frequency $1/f^\beta$ power spectrum is widely observed with $0 < \beta < 2$ (1925-2017).

- 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.

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

- The power spectrum is then normalizable.

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• 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)

\[
\int_{-\infty}^{\infty} S_t(\omega) d\omega = \frac{1}{t} \int_{-\infty}^{\infty} d\omega \int_{0}^{t} dt_1 \exp(-i\omega t_1) I(t_1)
\]

\[
\times \int_{0}^{t} dt_2 \exp(i\omega t_2) I(t_2) =
\]

\[
\frac{2\pi}{t} \int_{0}^{t} I^2(t_1) dt_1 < 2\pi (I_{max})^2
\]

\[\text{Variance}\]

- The total energy of \textit{bounded} stationary or non-stationary process, is finite.

- So how do we observe non-integrable \(1/f\) noise?

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Possible solution

- **Think!** Total energy must be finite

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

- \( z \) is called the aging exponent

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

- Hence we find a relation between the exponents, based on the finite value of the total power

  \[ z = \beta - 1. \]
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.


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

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Blinking Nano Crystals (coated CdSe)
Power law waiting time $\psi(\tau) \sim \tau^{-(1+\alpha)}$.

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

$\langle S(\omega, t) \rangle \sim C \omega^{-2+\alpha} / t^{1-\alpha}$

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Physical explanation for blinking


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Measured Power Spectrum Ages

Sadegh, EB, Krapf NJP (2014)

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

• $\sum S_i \sim N\langle S \rangle$ is time independent.

• When the number $N$ of fluctuating subunits, on the time scale of the measurement time is increasing with measurement time.
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.

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• 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.
Number of fluctuators increases with measurement time
Distributed kinetics model.
No Aging in Macroscopic Measurements

Distributed kinetics model.

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Long range correlations induced by many body effects, power law intermittency, burstiness in amplitude, yield $1/f$ noise.
Aging Wiener Khinchin Theorem

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

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

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

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

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

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

\[ \varphi_{TA}(x) = x^\gamma y(x) \int_0^\infty \frac{\phi_{EA}(z)}{z^{2+\gamma}} dz. \] \tag{7} 

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

\[ \frac{2(t_m)^{\gamma+1}}{2 + \gamma} \int_0^1 (1 - x)^\gamma \phi_{EN} \left( \frac{x}{1 - x} \right) \binom{1}{1} F_2 \left( 1 + \frac{\gamma}{2}; \frac{1}{2}, 2 + \frac{\gamma}{2}; -\left( \frac{\tilde{\omega} x}{2} \right)^2 \right) dx. \]
Blinking dot power spectrum on natural frequencies

\[ \left( t_m \right)^{-1} \langle S_{t_m}(\omega) \rangle_{\omega t_m = 2\pi n} \]
Blinking dot power spectrum beyond the natural
Single File Diffusion

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

\[ \langle x^2 \rangle \sim t^{1/2} \]

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

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PSD single file diffusion

\[ \langle S_{tm}(\omega) \rangle \]
<table>
<thead>
<tr>
<th>Model</th>
<th>$0 &lt; \alpha &lt; 1$</th>
<th>$\gamma$</th>
<th>$\nu$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unilayer Parisi’s Tree</td>
<td>0</td>
<td>0</td>
<td>$\alpha - 1$</td>
</tr>
<tr>
<td>Blinking Quantum Dot</td>
<td>0</td>
<td>0</td>
<td>$2\alpha - 2$</td>
</tr>
<tr>
<td>Laser-Cooled Atoms</td>
<td>$1 &lt; \alpha &lt; 3$</td>
<td>$2 - \alpha$</td>
<td>$2 - \alpha$</td>
</tr>
<tr>
<td>Single-File Diffusion</td>
<td></td>
<td>$1/2$</td>
<td>$1/2$</td>
</tr>
<tr>
<td>Generalized Elastic Model</td>
<td>$0 &lt; \alpha &lt; 1$</td>
<td>$\alpha$</td>
<td>$\alpha$</td>
</tr>
<tr>
<td>Coupled Oscillators</td>
<td></td>
<td>$[-0.58, -0.4]$</td>
<td>$-0.14 \pm 0.03$</td>
</tr>
<tr>
<td>1D Growing Interfaces</td>
<td></td>
<td>$\approx 0.33$</td>
<td></td>
</tr>
<tr>
<td>RC transmission Line</td>
<td>$1 &lt; \alpha &lt; 2$</td>
<td>$\alpha - 1$</td>
<td>$\alpha - 1$</td>
</tr>
</tbody>
</table>

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 $x \ll 1$.

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


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The meaning of the PS

\[ \langle I \rangle = \text{Const as for the WK spectrum.} \]

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

This gives the nearly standard relation:

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

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

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Single File Diffusion

\[ (x_T)^2 \sim \frac{D_t}{N} \]

Aslangul EPL 1986

\[ N \sim \rho \ L(t) \sim \rho \ (D_t)^{1/2} \]

\[ (x_T)^2 \sim \frac{(D_t)^{1/2}}{\rho} \]

Harris 65, Levitt 73
Models

<table>
<thead>
<tr>
<th>System</th>
<th>$\beta$ S $\sim$ $\omega^{-\beta}$</th>
<th>$z$ S $\sim t_m^{-z}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>single file diffusion</td>
<td>3/2</td>
<td>0</td>
</tr>
<tr>
<td>blinking quantum dot - finite mean 'on' time</td>
<td>$0 &lt; \alpha &lt; 1$</td>
<td>$\alpha$</td>
</tr>
<tr>
<td>blinking quantum dot - infinite mean</td>
<td>$0 &lt; \alpha &lt; 1$</td>
<td>$2 - \alpha$</td>
</tr>
<tr>
<td>logarithmic potential</td>
<td>$1 &lt; \alpha &lt; 2$</td>
<td>$3 - \alpha$</td>
</tr>
<tr>
<td></td>
<td>$2 &lt; \alpha &lt; 3$</td>
<td>0</td>
</tr>
</tbody>
</table>

Table 2: Summary of the critical exponents for the three systems discussed in sections VI-VIII.
Amplitude of $1/f$ noise is random, varies from one measurement to the other.

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

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Fluctuations of the power spectrum

- Power spectrum ergodic processes, we expect

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

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

\[ C = Y_\alpha \xi \]

- \( Y_\alpha (\xi) \) is a ML (exp) RV.

\[ Y_\alpha \rightarrow n/\langle n \rangle \]
- If I know the fluctuations of number of transitions, I have the fluctuations of the sample spectrum.

- Even better find PDF of \((S(\omega_1), \ldots, S(\omega_n))\).
Power spectrum exhibits a transition at $f_c = 0.1$ Hz.
This is related to cutoff on the on times.
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 nonintegrable $1/f$ 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.
Define the two state model better.

Infinite energy or infinite Power?
Wikipedia: In applied mathematics, the Wiener-Khinchin theorem, also known as the Wiener Khintchine theorem and sometimes as the Wiener-Khinchin-Einstein theorem or the Khinchin-Kolmogorov 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's, but as only recently recognized stated by Einstein in 1914). MIT OpenCourseWare Power Spectral Density Chapter 10.
<table>
<thead>
<tr>
<th>Group</th>
<th>Material</th>
<th>Nu.</th>
<th>Radii</th>
<th>( T )</th>
<th>( \alpha_{on} )</th>
<th>( \alpha_{off} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dahan</td>
<td>CdSe-ZnS</td>
<td>215</td>
<td>1.8,nm</td>
<td>300 K</td>
<td>0.58(0.17)</td>
<td>0.48(0.15)</td>
</tr>
<tr>
<td>Orrit</td>
<td>CdS</td>
<td></td>
<td>2.85</td>
<td></td>
<td>EXP</td>
<td>0.65(0.2)</td>
</tr>
<tr>
<td>Bawendi</td>
<td>CdTe....</td>
<td>200</td>
<td>1.5</td>
<td>10 – 300</td>
<td>0.5(0.1)</td>
<td>0.5(0.1)</td>
</tr>
<tr>
<td>Kuno</td>
<td>CdSe-ZnS</td>
<td>300</td>
<td>2.7</td>
<td>300</td>
<td>0.8 – 1.0</td>
<td>0.5</td>
</tr>
<tr>
<td>Cichos</td>
<td>Si</td>
<td></td>
<td></td>
<td></td>
<td>0.8 – 1.0</td>
<td>0.5</td>
</tr>
<tr>
<td>Ha</td>
<td>CdSe(coat)</td>
<td></td>
<td></td>
<td>300</td>
<td>Exp?</td>
<td>1</td>
</tr>
</tbody>
</table>
Efros, Orrit, Onsager, Hong-Noolandi

\[ r_{Ons} = \frac{e^2}{k_b T \epsilon} \sim 7 \text{nm} \]
Critical exponents of nano-crystals

<table>
<thead>
<tr>
<th></th>
<th>Theory</th>
<th>Experiment</th>
<th>Simulation</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\beta_&gt;$</td>
<td>$S \sim f^{-\beta}$</td>
<td>$\beta_&gt; = 2 - \alpha$</td>
<td>1.4</td>
</tr>
<tr>
<td>$\beta_&lt;$</td>
<td>$\beta_&lt; = \alpha$</td>
<td>0.7</td>
<td>0.6</td>
</tr>
<tr>
<td>$z$</td>
<td>$S \sim t^{-z}$</td>
<td>$z = 1 - \alpha$</td>
<td>0.12</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>$f_c \sim 1/t^\gamma$</td>
<td>$\gamma = 1$</td>
<td>0.8</td>
</tr>
<tr>
<td>$\omega$</td>
<td>$S_t(0) \sim t^\omega$</td>
<td>$\omega = 1$</td>
<td>0.8</td>
</tr>
</tbody>
</table>

Aging exponent is lower than expected.

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