$C_0$ speckle correlation and near-field interactions in
strongly scattering media

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We study the LDOS statistics in a strongly disordered system

\[
\rho(\omega) = \sum_k \delta(\omega - \omega_k)
\]

\[
\rho(r_0, \omega) = \sum_k \|E_k(r_0, \omega)\|^2 \delta(\omega - \omega_k)
\]
We study the LDOS statistics in a strongly disordered system.

**Density of States (DOS)**

\[ \rho(\omega) = \sum_k \delta(\omega - \omega_k) \]

**Local Density of States (LDOS)**

\[ \rho(r_0, \omega) = \sum_k \|E_k(r_0, \omega)\|^2 \delta(\omega - \omega_k) \]

**Green function**

\[ E(r) \propto \hat{G}(r, r_0, \omega) p \]

**LDOS in terms of Green function**

\[ \rho(r_0, \omega) \propto \mathcal{I} \left[ \text{Tr} \hat{G}(r_0, r_0, \omega) \right] \]
In vacuum

\[ \Gamma_0 \propto \mathcal{I} \left[ \text{Tr} \, G_0(r_0, r_0, \omega) \right] \]


**Relationship 1**

**LDOS = Spontaneous decay rate**

**The Purcell effect**

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**In vacuum**

\[ \Gamma_0 \propto I \left[ \text{Tr} \ G_0(r_0, r_0, \omega) \right] \]

**Emitter (molecule)**

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**With an environment**

\[ \Gamma \propto I \left[ \text{Tr} \ G(r_0, r_0, \omega) \right] \]

**Emitter (molecule)**

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Scattering medium

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Relationship 1  \[ \text{LDOS} = \text{Spontaneous decay rate} \]

Artistic view

Picture ©Enrique Sahagun
Relationship 1  \[ \text{LDOS} = \text{Spontaneous decay rate} \]
Experiments: direct access to the LDOS

Observations of decay rates fluctuations

- Spontaneous emission of a nanoscopic emitter in a strongly scattering disordered medium
  

- Observation of Spatial Fluctuations of the Local Density of States in Random Photonic Media
  

- Fluctuations of the local density of states probe localized surface plasmons on disordered metal films
  
LDOS fluctuations \( \propto \) the \( C_0 \) correlation

\[
\frac{\langle I(u) I(u') \rangle}{\langle I(u) \rangle \langle I(u') \rangle} = C_1 (u \cdot u') + C_2 (u \cdot u') + C_3 (u \cdot u')
\]

short range \hspace{1cm} long range \hspace{1cm} long range

\[
\left\langle \frac{\rho^2}{\langle \rho \rangle^2} \right\rangle - 1 = C_0
\]

Relationship 2: LDOS fluctuations \( \propto \) C₀ speckle correlation

\[
\frac{\langle I(u) I(u') \rangle}{\langle I(u) \rangle \langle I(u') \rangle} = C_1 (u \cdot u') + C_2 (u \cdot u') + C_3 (u \cdot u')
\]
short range \hspace{2cm} long range \hspace{2cm} long range

\[
\frac{\langle I(u) I(u') \rangle}{\langle I(u) \rangle \langle I(u') \rangle} = C_0 \quad \text{infinite range} \quad \text{non-universal} \quad \text{short range}
\]

\[
\frac{\langle \rho^2 \rangle}{\langle \rho \rangle^2} - 1 = C_0
\]

In this study...

...we analyse the statistical distributions of the LDOS (or decay rates)

- **Single vs multiple** scattering
- **Near-field** interactions
- Impact on $C_0$ correlation
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...we analyse the statistical distributions of the LDOS (or decay rates)

- Single vs multiple scattering
- Near-field interactions
- Impact on $C_0$ correlation

Important to address fundamental questions as

- how to improve fluorescence lifetime imaging techniques,
- how to control light emission and propagation by multiple scattering?
System of punctual resonant scatterers

- **Polarisability** (can be a two-level atom far from saturation)
  \[ \alpha(\omega) = \frac{-3\pi\gamma}{k^3(\omega - \omega_0 + i\gamma/2)} \]

- **Scattering cross-section**
  \[ \sigma(\omega) = \frac{k^4}{6\pi} |\alpha(\omega)|^2 \propto \lambda^2 \]

- **Scattering mean-free path**
  \[ \ell_B = [\rho\sigma(\omega)]^{-1} \]
Computation of the LDOS (or of the decay rate)

Electric field on the scatterer $j$ for all source dipole orientations

\[
E_j = G_0(r_j, r_0, \omega) + \alpha(\omega) k^2 \sum_{n=1 \atop n \neq j}^{N} G_0(r_j, r_n, \omega) E_n
\]

- Emitter contribution
- Scatterers contribution
Computation of the LDOS (or of the decay rate)

Electric field on the scatterer $j$ for all source dipole orientations

$$E_j = \sum_{n=1}^{N} G_0(r_j, r_n, \omega) + \alpha(\omega) k^2 \sum_{n \neq j}^{N} G_0(r_j, r_n, \omega) E_n$$

Emitter contribution

Scatterers contribution

Electric field at the source position

$$E(r_0) = \sum_{n=1}^{N} G_0(r_0, r_n, \omega) + \alpha(\omega) k^2 \sum_{n=1}^{N} G_0(r_0, r_n, \omega) E_n$$

$$S(r_0, r_0, \omega)$$
Computation of the LDOS (or of the decay rate)

Electric field on the scatterer $j$ for all source dipole orientations

\[ E_j = \underbrace{G_0(r_j, r_0, \omega)}_{\text{Emitter contribution}} + \alpha(\omega) k^2 \sum_{n=1}^{N} \underbrace{G_0(r_j, r_n, \omega) E_n}_{\text{Scatterers contribution}} \]

Electric field at the source position

\[ E(r_0) = G_0(r_0, r_0, \omega) + \alpha(\omega) k^2 \sum_{n=1}^{N} G_0(r_0, r_n, \omega) E_n \]

Normalised decay rate

\[
\frac{\rho}{\rho_0} = 1 + \frac{6\pi}{k} \mathcal{I} \left[ \operatorname{Tr} S(r_0, r_0, \omega) \right]
\]
Typical distribution

- **Wavelength** $\lambda = 630\,\text{nm}$
- **Cluster size** $R = 1.2\,\mu\text{m}$
- **Exclusion volume**
  $R_0 = 50\,\text{nm}$
- **Minimum distance between scatterers**
  $d_0 = 7.5\,\text{nm}$
- **Scattering mean-free path**
  $\ell_B = 1.9\,\mu\text{m}$
Regime 1: Multiple scattering

\[ \rho < \rho_0 \]

\[ P(\rho/\rho_0) \]

\[ 10^{-10} \]

\[ 10^0 \]

\[ 10^1 \]

\[ 10^2 \]

\[ \rho/\rho_0 \]

\( \rho < \rho_0 \): collective interactions (multiple scattering)

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Regime 1  Collective interactions

Distributions

Regime 1: Collective interactions

Distributions

Probability of having lifetimes larger than in vacuum (inhibition of the LDOS)

⇒ Interference effects (multiple interactions, finite-size effects)

Regime 1: Effective medium theory
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- Lorentz-Lorenz theory

\[ \epsilon_{\text{eff}}(\omega) = \frac{3 + 2\rho \alpha(\omega)}{3 - \rho \alpha(\omega)} \]

→ Independant scattering
Regime 1  Effective medium theory

- Lorentz-Lorenz theory

\[ \varepsilon_{\text{eff}}(\omega) = \frac{3 + 2\rho\alpha(\omega)}{3 - \rho\alpha(\omega)} \]

→ Independant scattering

- Local effective permittivity

\[ \varepsilon_{\text{eff}}(\omega) \]

→ Recurrent scattering
Regime 1  Effective medium theory

- **Lorentz-Lorenz theory**
  \[
  \varepsilon_{\text{eff}}(\omega) = \frac{3 + 2\rho\alpha(\omega)}{3 - \rho\alpha(\omega)}
  \]
  $\rightarrow$ Independant scattering

- **Local effective permittivity**
  \[
  \varepsilon_{\text{eff}}(\omega)
  \]
  $\rightarrow$ Recurrent scattering

- **Non-local effective permittivity**
  \[
  \varepsilon_{\text{eff}}(r, \omega)
  \]
  $\rightarrow$ Correlations of the disorder
Regime 1: Average LDOS

Stronger scattering

Independant scattering
**Regime 1:** Average LDOS

**Numerical results**

**Stronger scattering**

- $k_0 \ell_B = 10$

- $k_0 \ell_B = 2$

**Indepandant scattering**

**Recurrent scattering**
Regime 2 Single scattering in near-field

\[ \propto \rho^{-3/2} \]

1. \( \rho < \rho_0 \): collective interactions (multiple scattering)

2. \( \propto \rho^{-3/2} \): near-field interaction with one scatterer

Single interaction in near-field

\[ \rho \propto \frac{1}{r^3} \frac{1}{r^3} = \frac{1}{r^6} \]

\[ P(r) \propto r^2 \quad \Rightarrow \quad P(\rho) \propto \rho^{-3/2} \]

Regime 3: Multiple scattering in near-field

1. $\rho < \rho_0$: collective interactions (multiple scattering)

2. $\propto \rho^{-3/2}$: near-field interaction with one scatterer

3. $\rho > \rho_{\text{cut-off}}$: near-field interaction with more than one scatterer

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Regime 3 Influence of correlations of disorder

$P\left(\frac{\rho}{\rho_0}\right)$

$f$ is an effective volume fraction (correlation parameter)

$d_0$

Sensitivity to correlations of disorder (non-universal fluctuations)

Regime 3: Influence of correlations of disorder

Distributions

\[ P(\rho/\rho_0) \]

\[ \frac{\langle \rho^2 \rangle}{\langle \rho \rangle^2} - 1 \]

\[ C_0 = \frac{\langle \rho^2 \rangle}{\langle \rho \rangle^2} - 1 \]

Correlation parameter \( f \)

LDOS in a strongly scattering medium: two important messages!

Numerical identification of **different regimes:**
- Multiple scattering (collectives interactions)
- Near-field interactions with one or more scatterers

Decay rate or LDOS fluctuations \((C_0)\) are strongly affected by **near-field interactions** and can be seen as a probe of the local environment of the emitter.