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Spatial correlations and temporal heterogeneity of the slow dynamics of a colloidal fractal gel.

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► **To cite this version:**

Agnes Duri, Luca Cipelletti. Spatial correlations and temporal heterogeneity of the slow dynamics of a colloidal fractal gel.. HERCULES (Higher European Research Course for Users of Large Experimental Systems) European School, Feb 2007, Grenoble / Saclay, France. hal-02923795

HAL Id: hal-02923795

<https://hal.inrae.fr/hal-02923795>

Submitted on 27 Aug 2020

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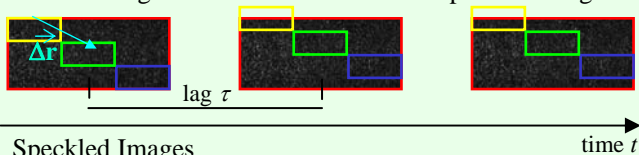
Experimental System

Polystyrene colloids ($d = 20 \text{ nm}$) suspended in a buoyancy-matching $\text{H}_2\text{O}/\text{D}_2\text{O}$ (45/55 vol/vol) $\Leftrightarrow \phi_{\text{PS}} = 6 \cdot 10^{-4}$
+ $\text{MgCl}_2 \Leftrightarrow C_{\text{MgCl}_2} = 10 \text{ mM} \Rightarrow$ **Fractal gel**

Experimental Set Up

“Multispeckle” Light Scattering experiments (Single Scattering)

- **N°1 Far field** – Several q ($4337 \text{ cm}^{-1} < q < 52177 \text{ cm}^{-1}$) – **Time Resolved Dynamics** – $V \Leftrightarrow$ All the scattering volume \Leftrightarrow All the speckled image
- **N°2 Imaging geometry** – Single q ($q = 10^4 \text{ cm}^{-1}$) – **Time and Space Resolved Dynamics** – $V \Leftrightarrow$ Portion of the scattering volume \Leftrightarrow Portion of the speckled image



Time (N° 1) and Space (N° 2) Resolved Degree of Correlation :

$$c_i(t, \tau, \vec{r}) = \frac{\langle I_p(t) I_p(t + \tau) \rangle_{p \in V(\vec{r})}}{\langle I_p(t) \rangle_{p \in V(\vec{r})} \langle I_p(t + \tau) \rangle_{p \in V(\vec{r})}} - 1$$

$I_p(t)$: intensity p-th pixel at time t

Spatially Resolved Intensity Correlation Function (N°1):

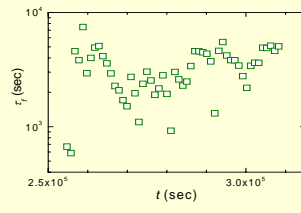
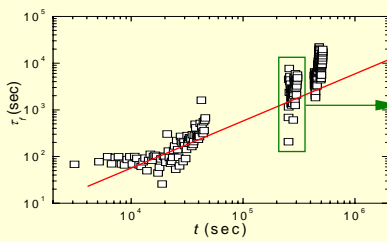
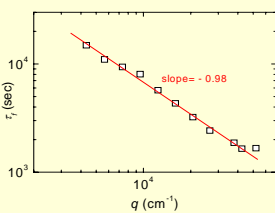
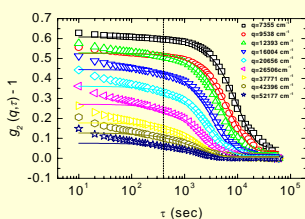
$$g_2(\tau, \vec{r}) - 1 = \langle c_i(t, \tau, \vec{r}) \rangle_t$$

Spatial Correlation of the Dynamics (N° 2):

$$X_{ci}(\tau, \vec{r}) = \left\langle \frac{\langle [c_i(t, \tau, \vec{r}) - \langle c_i(t, \tau, \vec{r}) \rangle_t] [c_i(t, \tau, \vec{r} + \Delta \vec{r}) - \langle c_i(t, \tau, \vec{r} + \Delta \vec{r}) \rangle_t] \rangle_t}{\sqrt{\sigma_{ci}^2(t, \tau, \vec{r}) \sigma_{ci}^2(t, \tau, \vec{r} + \Delta \vec{r})}} \right\rangle_{\vec{r}}$$

Experimental Results

Average Dynamics :



Fit by a “compressed” exponential

$$g_2(q, \tau) - 1 \propto \exp[-(t/\tau_f)^{3/2}]$$

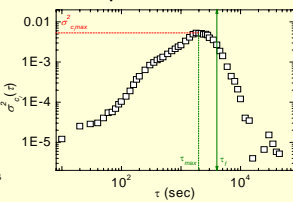
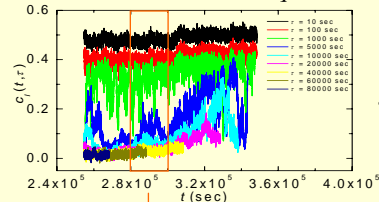
$\tau_f \propto q^{-1} \Rightarrow$ Drift motion

$\tau_f \rightarrow t \Rightarrow$ Aging behaviour

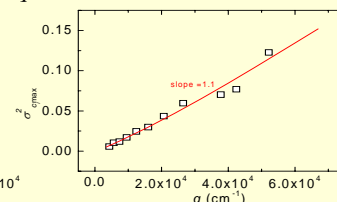
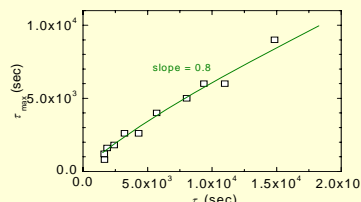
Large fluctuations of τ_f
 \Downarrow
Heterogeneous dynamics

Time Resolved Dynamics – Fluctuations Study:

$q = 20656 \text{ cm}^{-1} \Leftrightarrow 3 \mu\text{m}$



$4337 \text{ cm}^{-1} < q < 52177 \text{ cm}^{-1}$



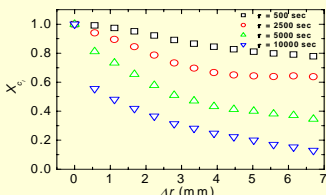
\rightarrow Statistics treatment

$\tau_{\text{max}} \propto \tau_f$

$\sigma_{c, \text{max}}^2 \propto q$

Time and Space Resolved Dynamics – Local dynamics Study:

Fluctuations max at large length scale



$\tau_f = 1000 \text{ sec}$

$\tau < \tau_f$:
Very long-ranged correlation
 \Rightarrow « solid-like » behavior

$\tau > \tau_f$:
Spatial correlation decay
 \Rightarrow « fluid-like » behavior

References :

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- [3] A. Duri and L. Cipelletti, *Europhys. Lett.*, 2006, **76**, 972-978