Laboratoire des Colloïdes Verres et Nanomatériaux

Spatial Correlations and Temporal Heterogeneity of the Slow Dynamics of a Colloidal fractal gel

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Experimental System

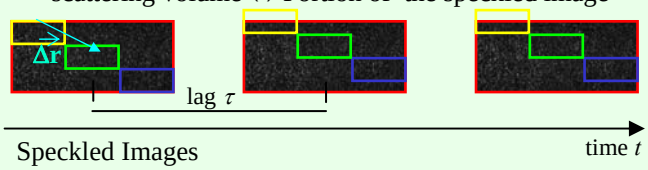
Polystyrene colloids (d = 20 nm) suspended in a buoyancy-matching $\mathbf{H_2O/D_2O}$ (45/55 vol/vol) $\Leftrightarrow \phi_{PS} = 6\ 10^{-4}$

+ $\mathbf{MgCl_2} \Leftrightarrow C_{MgCl_2}$=10 mM $\Rightarrow$ **Fractal gel**

Experimental Set Up

"Multispeckle" Light Scattering experiments (Single Scattering)

- **N°1** **Far field** – Several q **(** 4337 cm^{-1} < q < 52177 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 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):

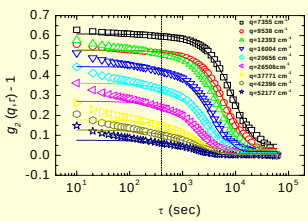
$$g_2(\tau,\vec{r}) - 1 = \langle c_I(t,\tau,\vec{r})\rangle_t$$

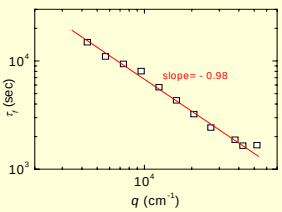
Spatial Correlation of the Dynamics (N° 2):

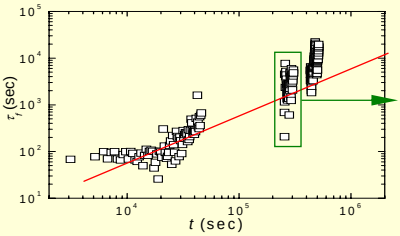
$$X_{cI}(\tau,\vec{r}) = \left\langle \frac{\left\langle \left[c_I(t,\tau,\vec{r}) - \langle c_I(t,\tau,\vec{r})\rangle_t\right]\left[c_I(t,\tau,\vec{r}+\Delta\vec{r}) - \langle c_I(t,\tau,\vec{r}+\Delta\vec{r})\rangle_t\right]\right\rangle_t}{\sqrt{\sigma^2_{cI}(t,\tau,\vec{r})\,\sigma^2_{cI}(t,\tau,\vec{r}+\Delta\vec{r})}} \right\rangle_{\vec{r}}$$

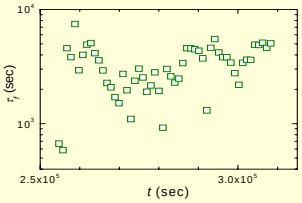
Experimental Results

Average Dynamics :









Fit by a "compressed" exponential

$g_2(q,\tau)$-1 ∝ exp[-(τ/ τ_f) $^{3/2}$]

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

$\tau_f \nearrow t$ ⇒ **Aging behaviour**

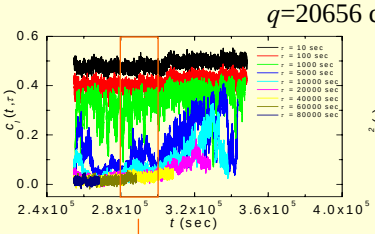
Large fluctuations of τ_f

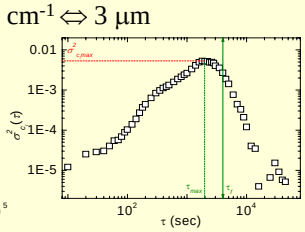
⇓

Heterogeneous dynamics

Time Resolved Dynamics – Fluctuations Study:

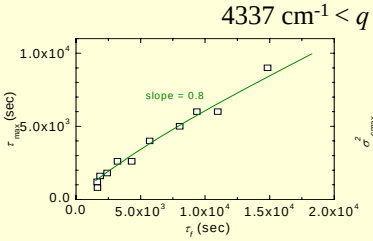
q=20656 cm^{-1} $\Leftrightarrow$ 3 μm



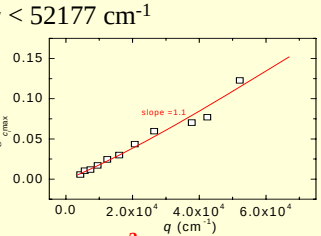


Statistics treatment

4337 cm^{-1} < q < 52177 cm^{-1}



$\tau_{max} \propto \tau_f$

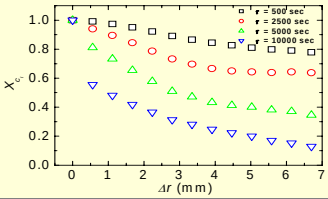


$\sigma^2_{cImax} \propto q$

⇓

Fluctuations max at large length scale

Time and Space Resolved Dynamics – Local dynamics Study:



τ_f = 1000 sec

$\tau < \tau_f$:

Very long-ranged correlation

⇒ ***« solid-like » behavior***

$\tau > \tau_f$:

Spatial correlation decay

⇒ ***« fluid-like » behavior***

References :

[1] L. Cipelletti, H. Manley, R.C. Ball, D. Weitz, *J. Phys. Rev. Lett.*, 2000, **84**, 10

[2] L. Cipelletti, H. Bissig, Trappe V, P. Ballesta, S. Mazoyer, *J. Phys.: Condens. Matt.*, 2003, **15**, S257- S262

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