Quelques perspectives dans l'étude expérimentale du transport colloidal en milieu poreux complexe

Pierre Levitz, Laurent Michot and Natalie Malikova

PHENIX, Sorbonne Université – CNRS, 75005 Paris, France

pierre.levitz@sorbonne-universite.fr

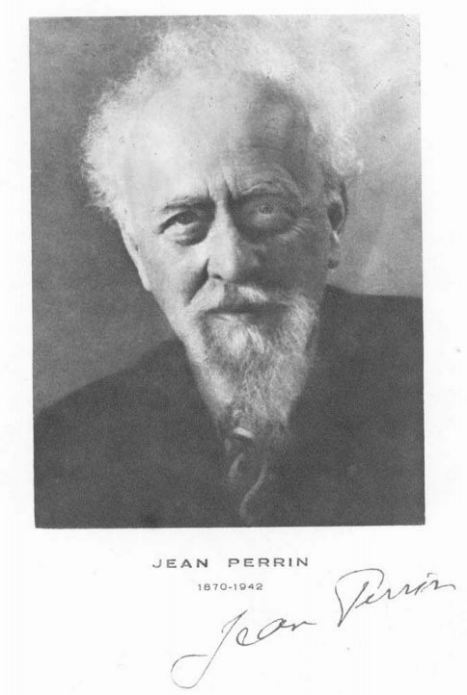
Colloidal transport in porous materials

- Filtration (transport, glogging)
- Chromatography
- Micro and nano fluidic
- Aquifer
- Porous rocks (sandstone,dolomite,……)
- Soil decontamination by colloids injection

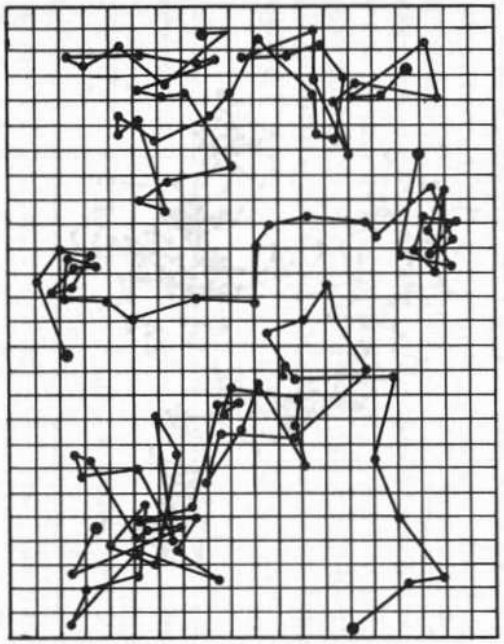
Direct observation of colloidal transport:

imaging colloidal trajectories

Not a new idea! (1907)



Microphotographie de la répartition en hauteur de sphérules de résines en suspension dans l'eau. Photographie reproduisant un cliché de Perrin, réalisée par le palais de la Découverte pour la présentation de ses expériences sur le mouvement brownien.



Dessin représentant le mouvement brownien de trois particules distinctes. Au moyen d'un projecteur, l'image de la solution est projetée sur un écran dépoli et quadrillé. A intervalles réguliers, les positions successives d'une particule sont notées par des points, ce qui permet de calculer le déplacement moyen des particules. Jean Perrin, *Mouvement brownien et realité moléculaire*, in *Annales de chimie et de physique*, sér. 8, vol. 18, 1909, p. 81.

$$D_{self} = \frac{k_B T}{6\pi\eta R}$$

Self-Limited Accumulation of Colloids in Porous Media

G. Gerber,[1,2] M. Bensouda,[1] D. A. Weitz,[2] and P. Coussot[1]
[1]*Université Paris-Est, Laboratoire Navier (ENPC-IFSTTAR-CNRS), Champs-sur-Marne 77420, France*
[2]*Experimental Soft Condensed Matter Group, School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA*

(Received 10 April 2019)

As a model porous medium we use monodisperse borosilicate beads (negatively charged, 63 μm average diameter) randomly packed (porosity $\phi_0 = 0.38$) in a square glass capillary (width $w = 1.0$ mm), and we invade this medium with positively charged, fluorescent, monodisperse (diameter $d_p = 1.0\ \mu$m) latex colloids. The suspensions are made at constant dilute concentration $C_0 = 20\ \text{mg mL}^{-1}$ in a mix of deionized water and DMSO to match the refractive index of the beads [28]. This allows us to visualize the particles inside the porous medium by

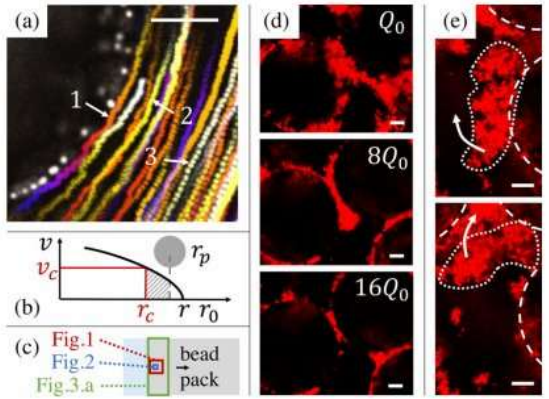


FIG. 2. Local transport and adsorption mechanisms. All scale bars are 10 μm. (a) Time projection of confocal imaging, with the suspension flowing upwards. $I = 10^{-6}$ M. Steady particles appear as white dots, while moving particles are represented by their trajectories. Selected trajectories: particle 1 along wall, 2 close to wall, 3 far from wall. (b) Schematic velocity profile in a pore of initial and current radii r_0 and r. Dashed area shows positions favorable to adsorption: $[r_c : r - r_p]$. (c) Schematics of the three scales used in Figs. 1, 2 and 3(a). (d) Selected area under steady state for different flow rates $[Q_0; 8Q_0; 16Q_0]$, at $I = 10^{-1}$ M. (e) Cluster formed on a surface at $I = 10^{-1}$ M (top), detaching and moving to a more stable position (bottom). Dashed lines highlight bead surfaces.

Very nice experiment but:
Need for relative light transparency of the pore network
Need to observe a large number of trajectories in order to perform in a post processing analysis, an ensemble average.

Some needs in case of completely opaque

and/or multidiffusif materials

- No direct visualization of colloidal trajectories.
- Need for a temporal resolution.
- Need to perform an experimental ensemble average.
- Need to extract meaningful "descriptors" either at macroscopic or at mesoscopic length scales

In this talk:

- Macroscopic experiments: Breakthrough curves
- Use of the Magnetic resonance imaging
- X-ray photon correlation spectroscopy (XPCS)
- Part 4 not available for actual publication

Part 1:Macroscopic experiments: Breakthrough curves

Chemosphere 81 (2010) 387–393

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Transport of metal oxide nanoparticles in saturated porous media

Tal Ben-Moshe, Ishai Dror *, Brian Berkowitz

Department of Environmental Sciences and Energy Research, Weizmann Institute of Science, Rehovot 76100, Israel

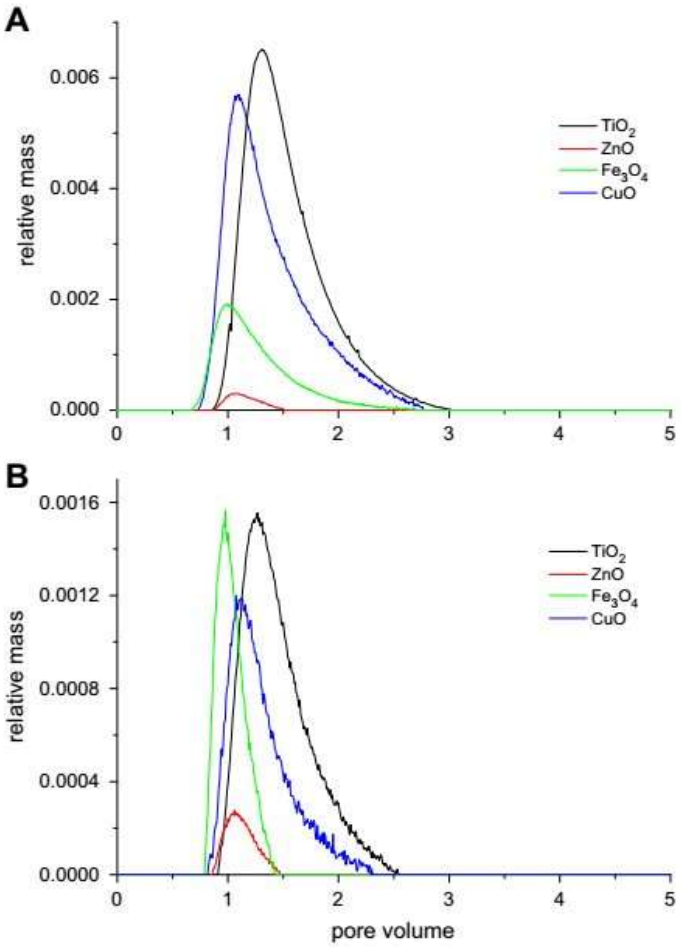


Fig. 4. Breakthrough curves for metal oxide nanoparticles at pH 7 with (A) 0.0 M and (B) 0.1 M NaCl.

$$\frac{\partial}{\partial t}C = D\frac{\partial^2 C}{\partial x^2} - v_p\frac{\partial C}{\partial x} - kC$$

Part 2: A power tool _ the Magnetic Resonance Imaging

Int. J. Environ. Sci. Technol. (2015) 12:3373–3384
DOI 10.1007/s13762-015-0767-4

ORIGINAL PAPER

Characterization of nanoparticle transport through quartz and dolomite gravels by magnetic resonance imaging

S. Lakshmanan · W. M. Holmes · W. T. Sloan · V. R. Phoenix

$$\frac{S}{S_0} = \exp\left(\frac{-t_e}{T_{2,i}}\right)$$

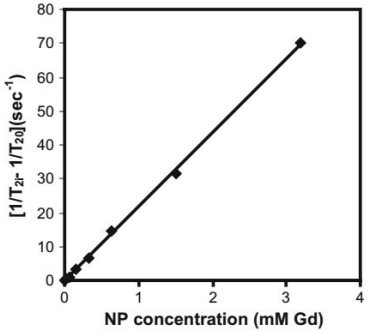


Fig. 1 Variation in the change of transverse relaxation rate with respect to *Gd NP* concentration (NP concentration shown as mM of Gd)

$$[C] = \frac{1}{R}\left[\frac{1}{T_{2,i}} - \frac{1}{T_{2,0}}\right]$$

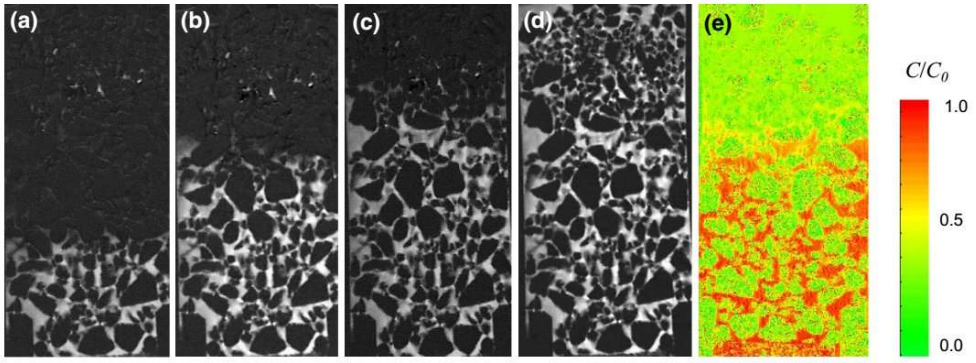


Fig. 3 T_2-weighted images of *Gd NP* in dolomite gravel at 35, 45, 65 and 75 min denoted by (**a**), (**b**), (**c**) and (**d**), respectively; NP transport is shown by bright regions. Figure 3**e** shows calibrated image of *Gd NP* concentration at 55 min

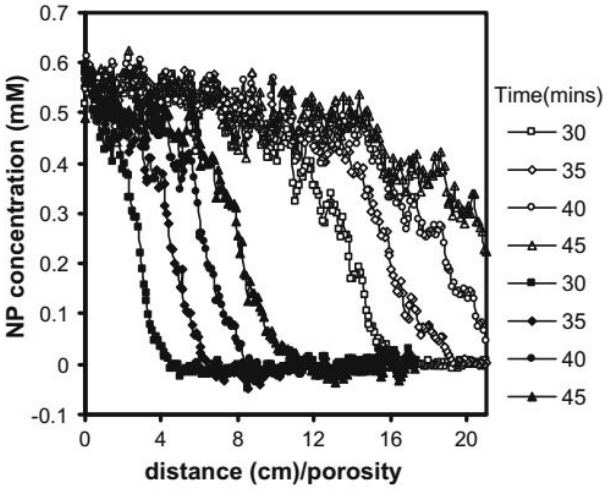


Fig. 5 *Gd NP* concentration at various time intervals for rose quartz (*open symbol*) and dolomite (*closed symbol*), normalized to porosity

Comparison exp-model

$$\frac{\partial}{\partial t}C = D\frac{\partial^2 C}{\partial x^2} - v_p\frac{\partial C}{\partial x} - kC \tag{4}$$

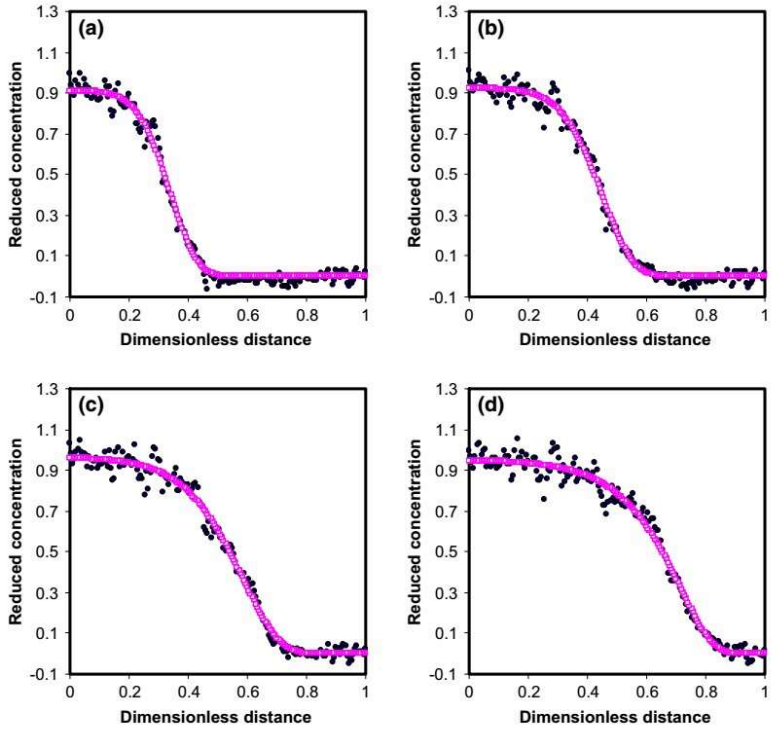
where C is the concentration, t is time, x is distance from the inlet, v_p average particle velocity, D dispersion coefficient and k is the decay coefficient. We assume a semi-infinite domain with constant concentration C_0 at the inlet.

$$C(x,t) = C_0 \text{ when } x = 0, t \geq 0 \tag{5a}$$

$$C(x,t) = C_0 \text{ when } 0 < x < \infty, t = 0 \tag{5b}$$

$$\frac{dC(x,t)}{dx} = 0 \text{ when } x = \infty \tag{5c}$$

Fig. 7 *Gd NP* concentration profile of dolomite gravel experiment at **a** 40 min, **b** 45 min, **c** 50 min, **d** 55 min. Experimental data (*closed symbol*) and CXTFIT data (*open symbol*)



See also Lehoux, S. Rodts et al, Phys. Rev E,94, 053107(20216)

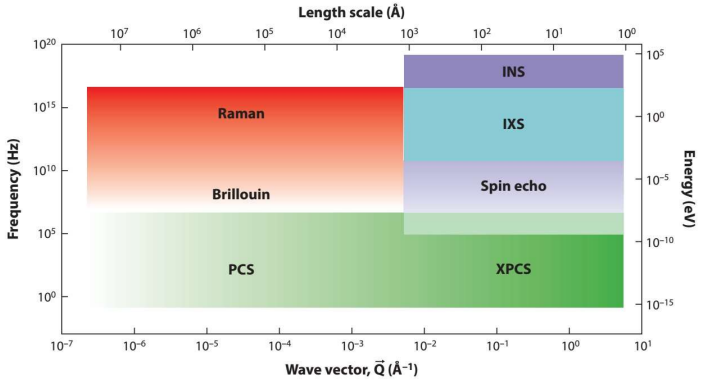
Using X rays beams to probe colloidal dynamics

Time correlation of 2D images

- **2D scattering images (XPCS)**

Part 3 : XPCS _ X ray photon Correlation Spectroscopy

"Extension" in the X-rays domain of the DLS (homodyne detection): Time resolved 2D scattering technique



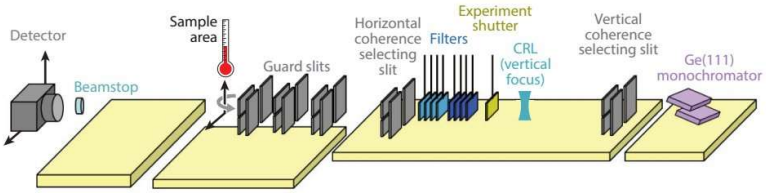


Figure 4

Schematic of the small-angle XPCS setup in enclosure 8-ID-I at the Advanced Photon Source. From right to left, 8-ID-I contains a Ge(111) monochromator, transverse coherence length–selecting slits, a compound refractive lens (CRL) for 1D vertical focusing at the sample position, a sample platform capable of supporting a wide variety of sample environments, a 4-m-long exit flight path, and a variety of area detectors. The monochromator is 65 m from the source, and the sample area is 68 m from the source. The detector and beamstop are 4 m downstream of the sample area.

No multidiffusive scattering. Large q range. Use of the Sieger theorem

$$g_2(Q,\tau) = \frac{\langle I(Q,t)I(Q,t+\tau)\rangle}{\langle I(Q)\rangle^2} = \beta(Q)g_1^2(Q,\tau) + 1,$$

Intermediate scattering function (ISF)

$$g_1(\vec{q},\tau) = \left\langle \frac{1}{N}\sum_{i=1}^{N}\sum_{j=1}^{N}\exp(i\vec{q}(\vec{r}_j(t_0+\tau) - \vec{r}_i(t_0)) \right\rangle_{t_0}$$

One the first application:

Probing the dynamics of very concentrated colloidal suspensions:

THESE

Présentée par

Aymeric ROBERT

Pour obtenir le grade de

DOCTEUR DE L'UNIVERSITE JOSEPH FOURIER

discipline : Physique

DYNAMIC BEHAVIOR OF CHARGE STABILIZED COLLOIDAL SUSPENSIONS

le 21 septembre 2001

THE JOURNAL OF CHEMICAL PHYSICS **137**, 114504 (2012)

Structure and short-time dynamics in concentrated suspensions of charged colloids

Fabian Westermeier,[1,a)] Birgit Fischer,[1] Wojciech Roseker,[1] Gerhard Grübel,[1] Gerhard Nägele,[2] and Marco Heinen[3,b)]

[1]*Hasylab, Deutsches Elektronen-Synchrotron, Notkestrasse 85, D-22607 Hamburg, Germany*

[2]*Institute of Complex Systems (ICS-3), Forschungszentrum Jülich, D-52425 Jülich, Germany*

[3]*Institut für Theoretische Physik II, Weiche Materie, Heinrich-Heine-Universität Düsseldorf, D-40225 Düsseldorf, Germany*

$$D_m(Q) = -\lim_{\tau \to 0} \frac{1}{Q^2} \frac{\partial}{\partial \tau} \ln g_1(Q, \tau) = D_0 \frac{H_m(Q)}{S_m(Q)},$$

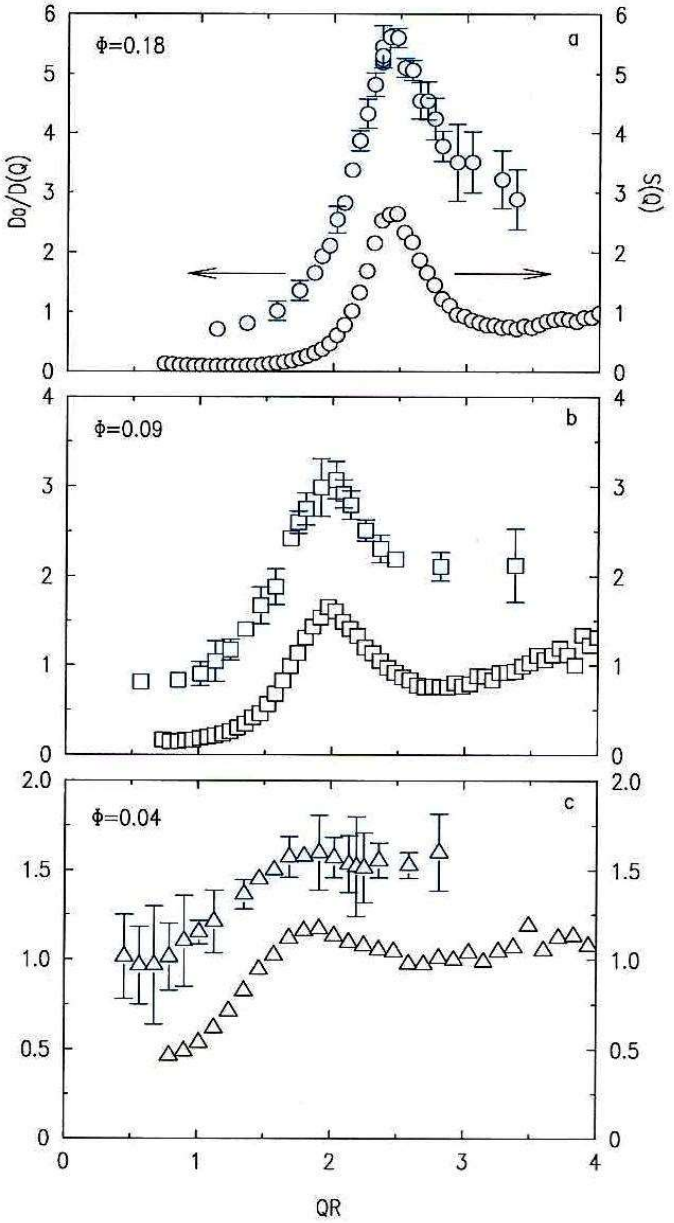


Figure 4.15: Static structure factor (○) for the colloidal suspensions of volume fraction $\phi = 0.18$ (a), 0.09 (b) and 0.04 (a). The blue symbols shows the Q–dependence of the normalized inverse effective diffusion coefficient $D_0/D(Q)$.

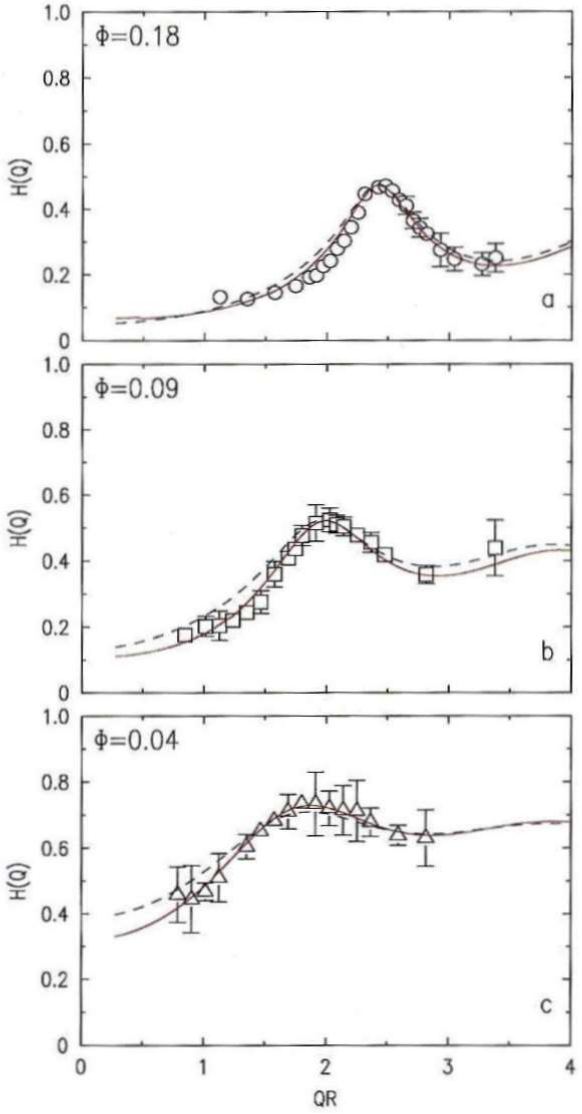


Figure 4.20: Hydrodynamic functions compared to two model calculations. The dashed lines are the result of the fit of eq. 4.3.8 (small Q approximation) to the data using the measured static structure factor as an input and with $\tilde{D}$ being the only fit parameter. The solid lines are the results of the fit of eq. 4.3.2 ($\delta\gamma$–expansion) using the measured static structure factor as an input and with α being the scaling parameter (see text).

Extremely Slow Diffusion of Gold Nanoparticles under Confinement in Mesoporous Silica

Ramona Mhanna, Michael Giroux, Kenneth J. T. Livi, Chao Wang, Andrei Fluerasu, Lutz Wiegart, Yugang Zhang, Mark Sutton, and Robert L. Leheny*

Cite This: *J. Phys. Chem. C* 2022, 126, 3614–3622

Read Online

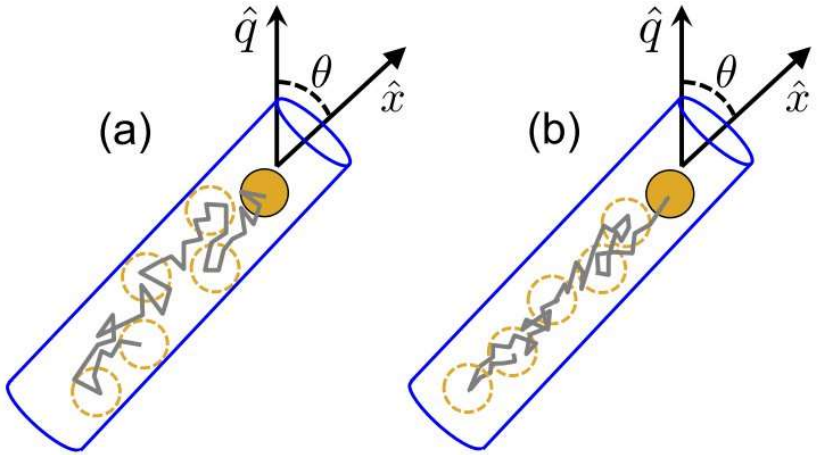
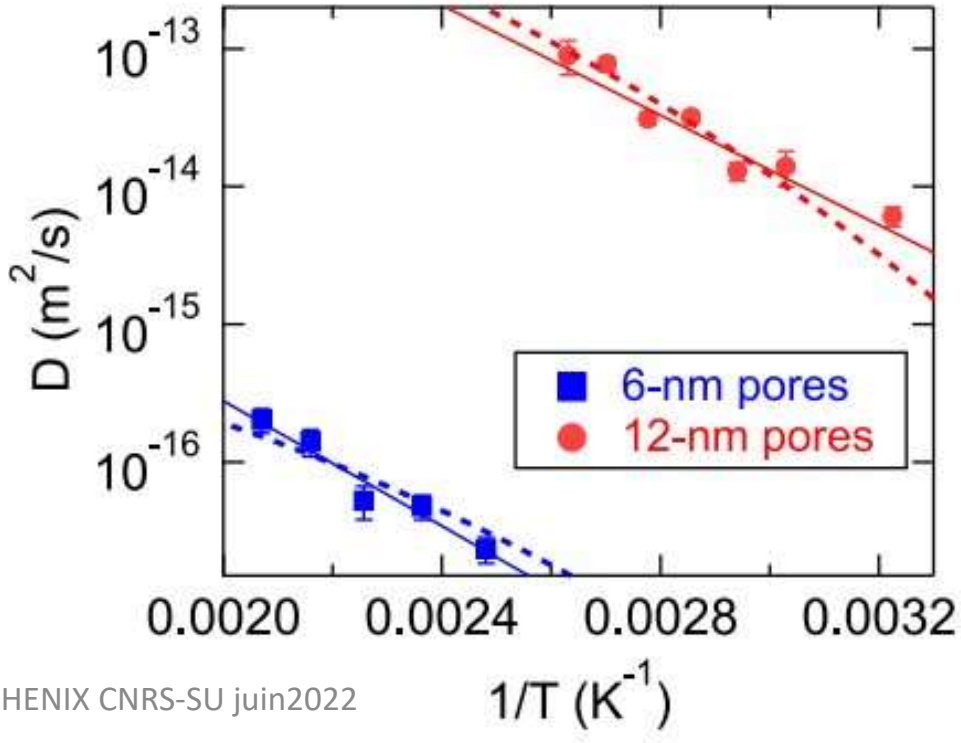


Figure 3. Schematics depicting different scenarios for a nanoparticle diffusing in glycerol within an SBA-15 pore. (a) Particle's trajectory is punctuated by repeated adsorption to the pore walls. (b) Particle is restricted to the central region of the pore because of an immobile layer of glycerol adjacent to the walls.

$$g_2(q,t) = 1 + \beta(X^2|g_{1,\mathrm{Au}}(q,t)|^2 + 2X(1-X)\mathrm{Re}[g_{1,\mathrm{Au}}(q,t)g^*_{1,\mathrm{SBA}}(q,t)] + (1-X)^2|g_{1,\mathrm{SBA}}(q,t)|^2)$$



To sum-up for XPCS

* Time correlation of 2D scattering images.

* Origin of 2D image constrast: Atomic X-ray scattering section

* Need to have a highly coherent X ray (lateral and longitudinal)

  (to get β close to 1)

* Can be performed on synchrotron beam lines (ID2, ID10 for ESRF)

* Need to check for radiation damages

Part 4

Not actually available