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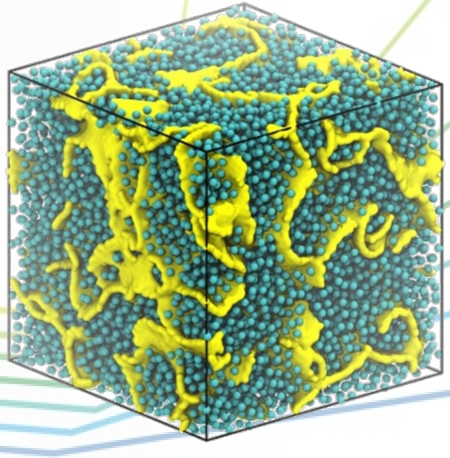
Apr. 17, 2026



Theory, Molecular Modeling, and Neutron Scattering of Fluid Transport in Nanoporous Materials

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https://benoitcoasne.github.io/

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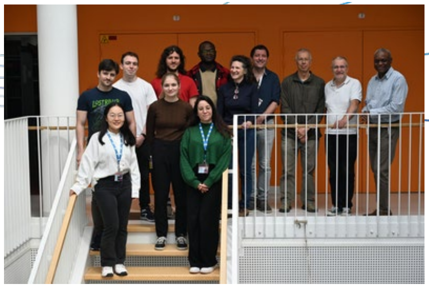
ILL Theory Group



Provide theoretical support & develop theory projects on neutron-related subjects

Affiliates, Long Term Visitors and Emeritus (7)

LEPETIT Marie-Bernadette*	Electronic Structure/Magnetism
COASNE Benoit**	Soft Matter/Chemical Physics
ZIMAN Timothy (Emer.)	Condensed Matter
GASPARD Jean-Pierre (Emer.)	Chemical Physics
BICOUT Dominique	Biophysics
FIELD Martin	Biochemistry
ZHITOMIRSKY Mike	Condensed Matter

Postdocs (4)

DENG Shiyu	Electron. Structure/Magnetism
DE IZARRA Ambroise	Soft Matter
COSTE Amaury (LIPhy/CEA)	Soft Matter
MURUGESAN Ramasamy (Néel)	Electron. Str./ Magnetism

PhD students and Interns (8)

DIDIER Loriane (PhD, LIPhy)	Soft Matter
KENGNI-ZANGUIM Brice (PhD, Néel)	Magnetism & IA
MATA-INFANTE Ferran (MSc)	Soft Matter
MECOLI Victor (PhD)	Magnetism
PERSENDA Gautier (PHELMA)	Biophysics
AKERLIND Gunnar (MsC)	Soft Matter
Elliott Perryman (PhD)	IA
EBE Mohamed (PHELMA)	Biophysics

-- 16/20 articles/year
-- ILL Groupe Leader* + ILL College 2 Secretary**
-- Organization Committees ICNS 2027
-- Chair MATERIAUX2026

ILL Theory Group

Connected to the experimental ILL Groups (non exhaustive)

Lattice Dynamics in Multiferroic Materials
S. Deng,
M. B. Lepetit

Emergent Polar Metal Phase in a Van der Waals Mott Magnet
S. Deng, A. Wildes,
S. S. Saxena

Acoustic Stimulation of Fluid in Nanoporous Materials
L. Didier, M. Plazanet,
B. Coasne

Local dynamics in lipid bilayers
D. J. Bicout,
J. Peters

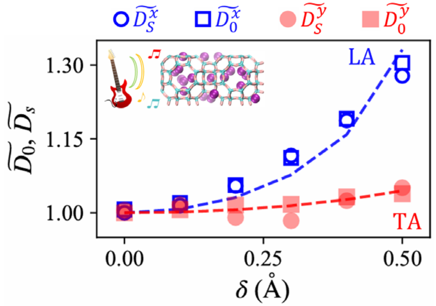
Autonomous experiment
D. E. Perryman, M-B Lepetit, D. Mazzone et M. Boehm

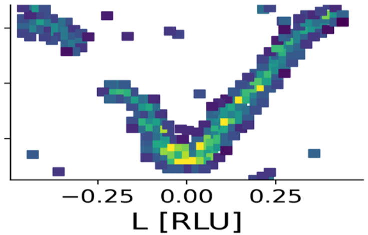
TOF Resolution
V. Mecoli, H. Jacobsen, E. Rebolini,
T. Weber, J. Ollivier, M-B Lepetit

Solid State Physics, Magnetism

Soft Matter, Chemical and Biophysics

AI and Data Analysis





ILL Theory Group

Connected to the CS group

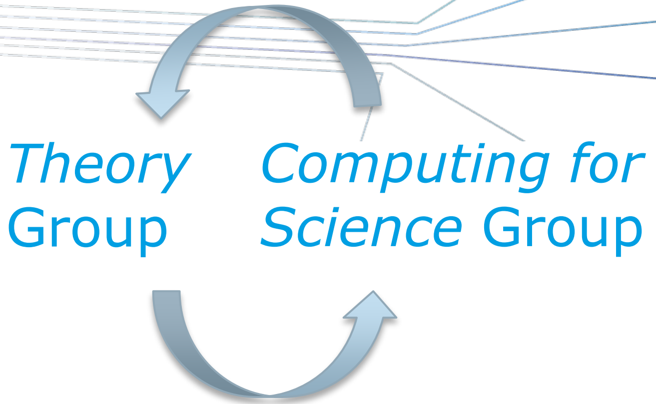
- Similar background in simulation and theory despite pursuing different objectives

- Common task to provide support to Science [User And ILL]

- Shared use of ILL HPC Cluster, [molecular packages, numerical codes, data management strategy]

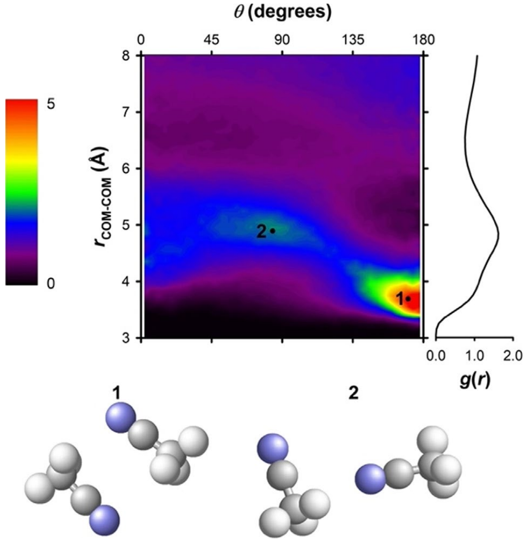
- Link molecular modelling and more formal approaches (Statistical Phys., Solid State Phys.)

- Shared Position between numerical/theoretical groups [Ex: Battery Hub, Quantum/Magnetism] ?



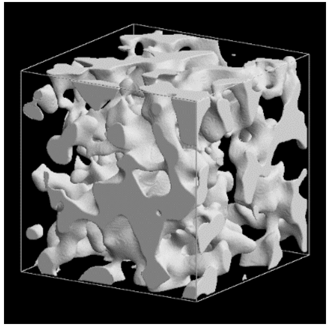
Theory Group

Computing for Science Group



Ex:
Cohen,
Plazanet, Rols,
Voneshen,
Fourkas,
Coasne, *J. Mol. Liq.* 2022

Fluid in Nanoporous Materials



Courtesy of Levitz

Large surface areas (ex. ashes ~m^2/g) but larger surfaces can be reached ~1000 m^2/g

Nanoporous solids (with one dimension ~ *nm*) are of particular interest because D ~ ξ

New thermodynamic and transport phenomena

Adsorption/transport interplay
Complex hydrodynamics

e.g. slippage, interfacial transport, and non-viscous effects

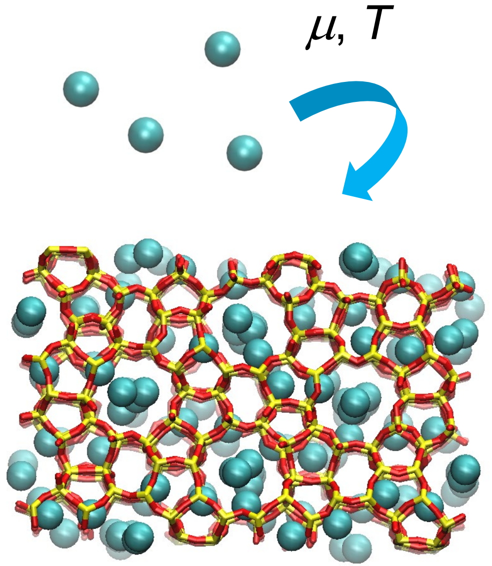
Bridging molecular dynamics/macroscopic transport using statistical mechanics?

- Self-diffusion. Available microscopic formalisms (intermittent brownian motion, free volume theory, surface diffusion, etc.)

- Permeability. De Gennes narrowing and the link between structure and collective diffusion

Methane in Zeolite

A Lennard-Jones fluid in flexible silicalite-1 (pores ~5-8 Å)



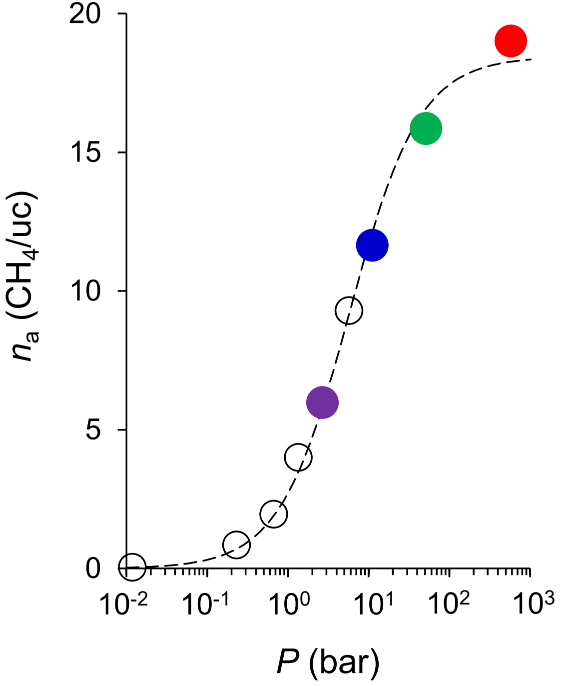
● Fluid molecule (CH_4)

Silica tetrahedron (SiO_4)

Grand Canonical
Monte Carlo

⇒

Type 1
adsorptionisotherm
(Langmuir model)



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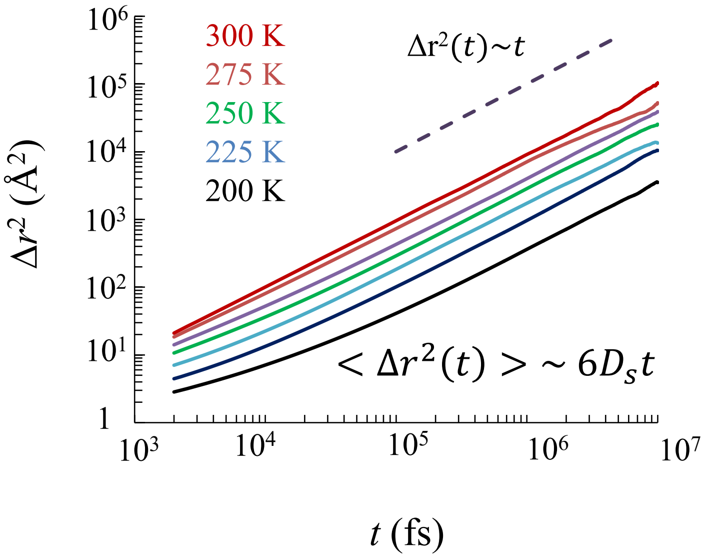
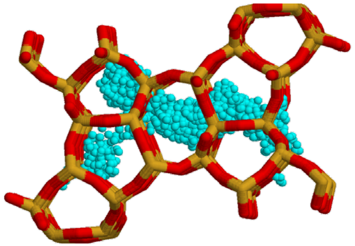
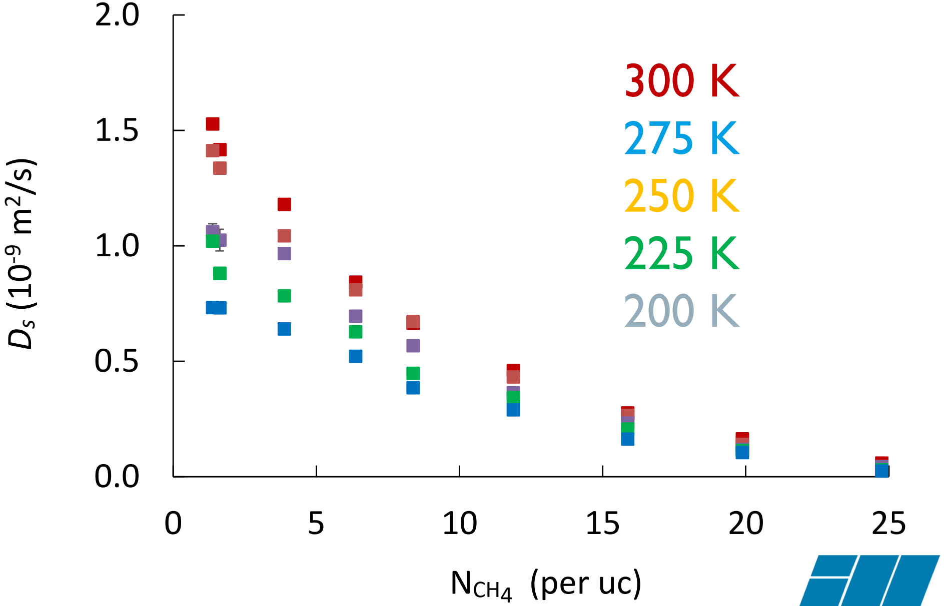
Self-Diffusion

Molecular Dynamics Simulation

Kellouai et al. 2024, 2025





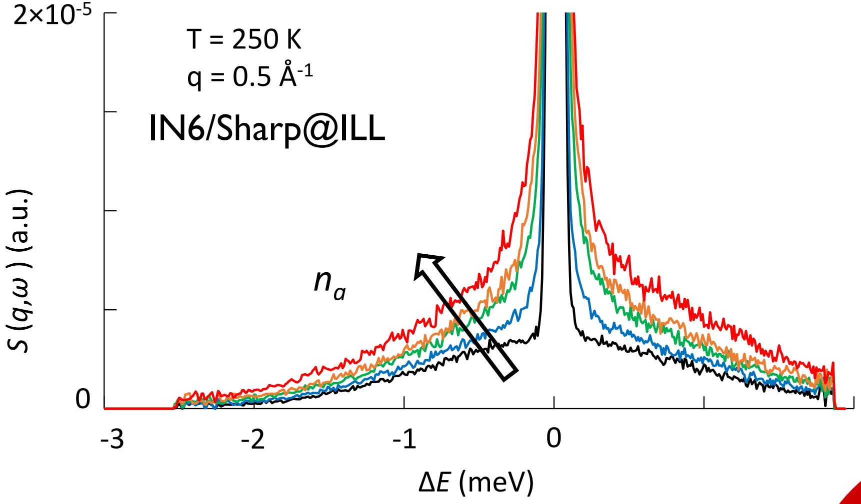


Self-Diffusion

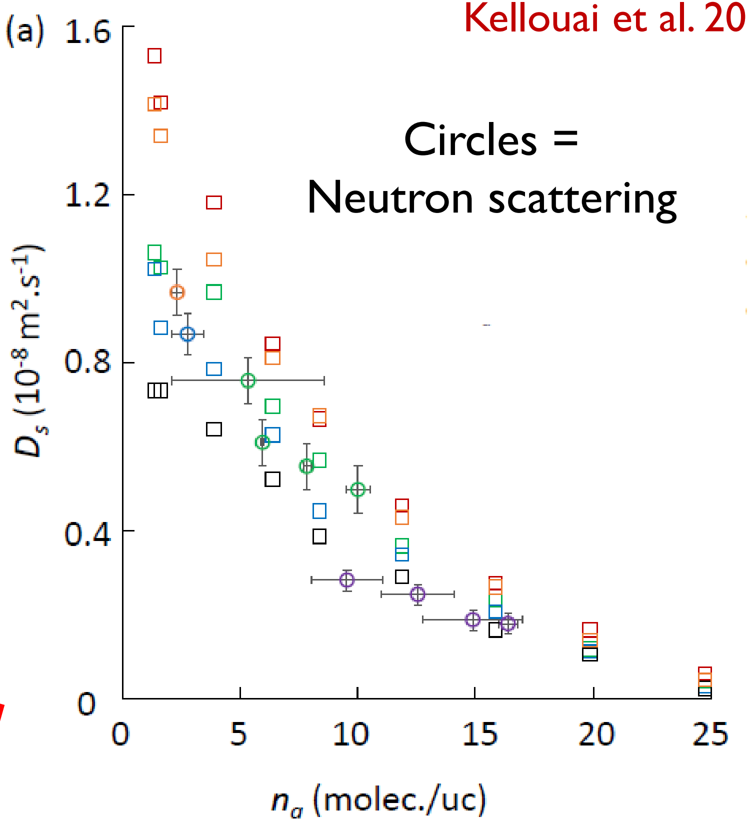
Quasi Elastic Neutron Scattering

W. Kellouai, P. Judeinstein, M. Plazanet,
S. Baudoin, Q. Berrod, J. M. Zanotti

Kellouai et al. 2024, 2025



n_a ↗ ⇨ narrowing ⇨ ↘

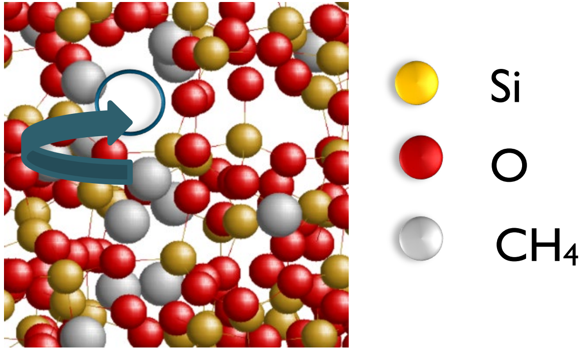


Circles =
Neutron scattering

Free Volume Theory

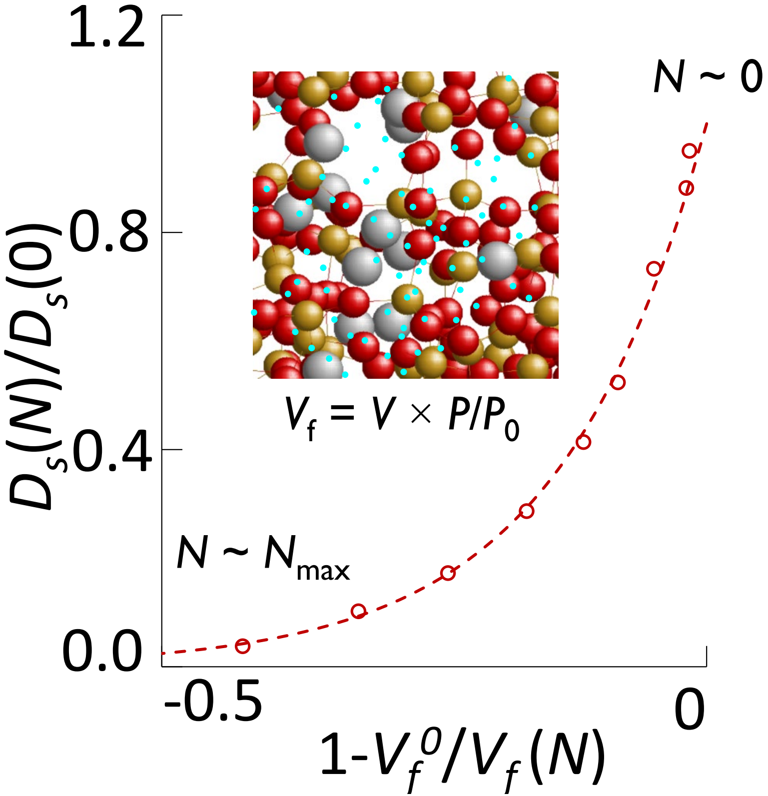
Physical Model for Self-Diffusion

Kellouai et al. 2024, 2025



$$D_s(N) \sim D_s(0)\exp\left[-\gamma \frac{V_{mol}(N)}{V_f(N)}\right]$$

$$V_{mol}(N) = V_f^0 - V_f(N) \sim N$$

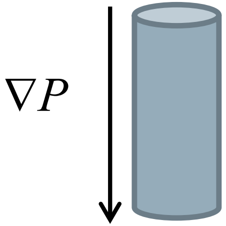


Permeability and Hydrodynamics Breakdown

Darcy's Law

$$v = -\frac{k}{\eta}\nabla P$$

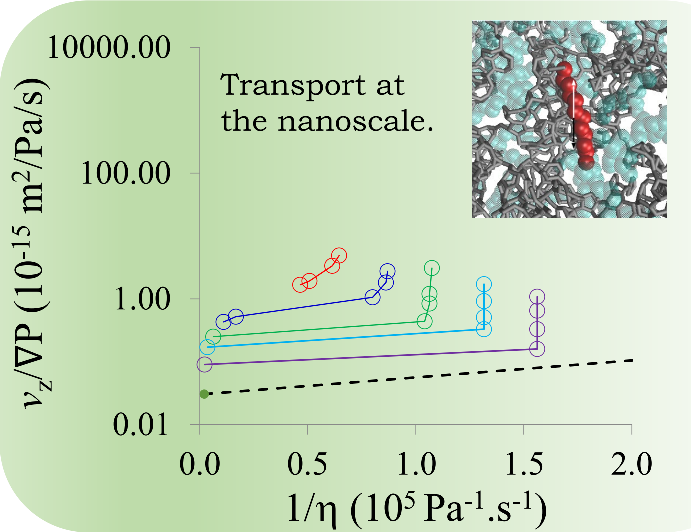
k permeability,
η viscosity



$$\boxed{\rho \frac{\partial \boldsymbol{v}}{\partial t}} = -\nabla \mathrm{P} + \boxed{\eta \nabla^2 \boldsymbol{v}} - \zeta \boldsymbol{v}$$

$$\eta = \frac{1}{V k_B T}\int \sigma_{xy}(t)\sigma_{xy}(0)\mathrm{d}t \sim \exp[-t/t_\mathrm{R}] \Rightarrow t_\mathrm{R} \sim 1\ \mathrm{ps}$$

$$\langle v_k(0)\, v_{-k}(t)\rangle \sim \exp\left[\left(-\frac{\eta k^2}{\rho} - \zeta\right)t\right] \Rightarrow \tau \sim \frac{\rho}{\eta k^2}$$



Hydrodynamic regime $\rho/\eta k^2 >> t_\mathrm{R}$
Breakdown $k \sim 1/L$ (L = 1 nm)

Falk, Bocquet, Coasne, Nature Comm. 2016

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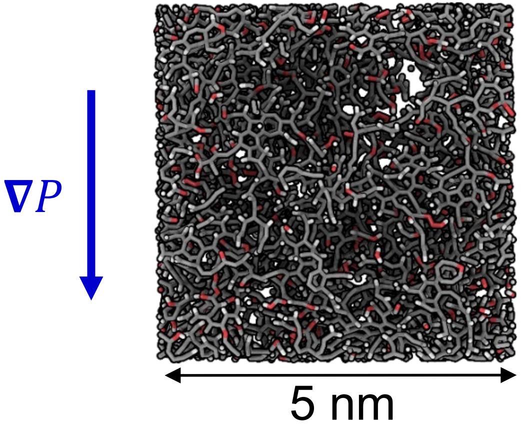
Permeability ~ Collective Diffusion

Darcy's Law

- Consider a fluid confined in a nanoporous material
- Flow induced by a pressure gradient ∇P applied



Falk, Coasne, Bocquet, et al. Nature Comm. 2015

Bousige, et al. Nature Materials 2016

Darcy	**Onsager**
$\mathbf{J} = -\rho K \nabla P$	$\mathbf{J} = -\rho \frac{D_0}{k_B T} \nabla \mu$

Gibbs-Duhem: $\mathrm{dP} = \rho \mathrm{d}\mu$

$$\Rightarrow \quad K = \frac{D_0}{\rho k_B T}$$

Schlaich, Barrat, Coasne, Chem. Rev. 2024

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De Gennes Narrowing

Kellouai, Barrat, Coasne et al. 2024

Microscopic Theory for Collective Dynamics

$$\mathbf{J} = -\rho \frac{D_0}{k_B T} \boldsymbol{\nabla}\mu$$ **Onsager**

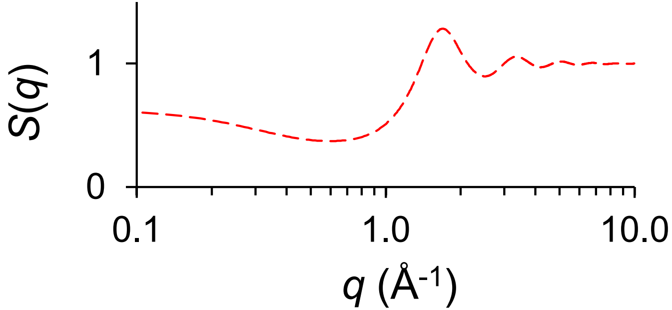
$$\frac{\partial \rho(\mathbf{r},t)}{\partial t} + \nabla \cdot \mathbf{J}(\mathbf{r},t) = 0$$ **Mass conservation**

$$\frac{\partial \rho(\mathbf{r},t)}{\partial t} - \frac{\bar{\rho} D_0}{k_B T} \nabla^2 \mu(\mathbf{r},t) = 0 \quad \Longleftrightarrow \quad \frac{\partial \rho(\mathbf{q},t)}{\partial t} + \frac{\mathbf{q}^2 \bar{\rho} D_0}{k_B T} \frac{\delta F[\bar{\rho}]}{\delta \rho(\mathbf{q},t)} = 0$$

$$F[\bar{\rho}] = \frac{1}{2} \frac{\mathrm{k}_B T}{\rho V} \frac{\sum_{\mathbf{q}} \rho(\mathbf{q}) \rho^*(\mathbf{q})}{S(\mathbf{q})}$$ with $\mathrm{S}(\mathbf{q}) = \langle \rho(\mathbf{q}) \rho^*(\mathbf{q}) \rangle$

$$\boxed{F_{\mathrm{coh}}(\mathbf{q},t) = \langle \rho(\mathbf{q},t) \rho^*(\mathbf{q},0) \rangle = \exp[-t/\tau_0(q)]}$$

with $\tau_0(q) = \dfrac{S(\mathbf{q})}{D_0 \mathbf{q}^2}$



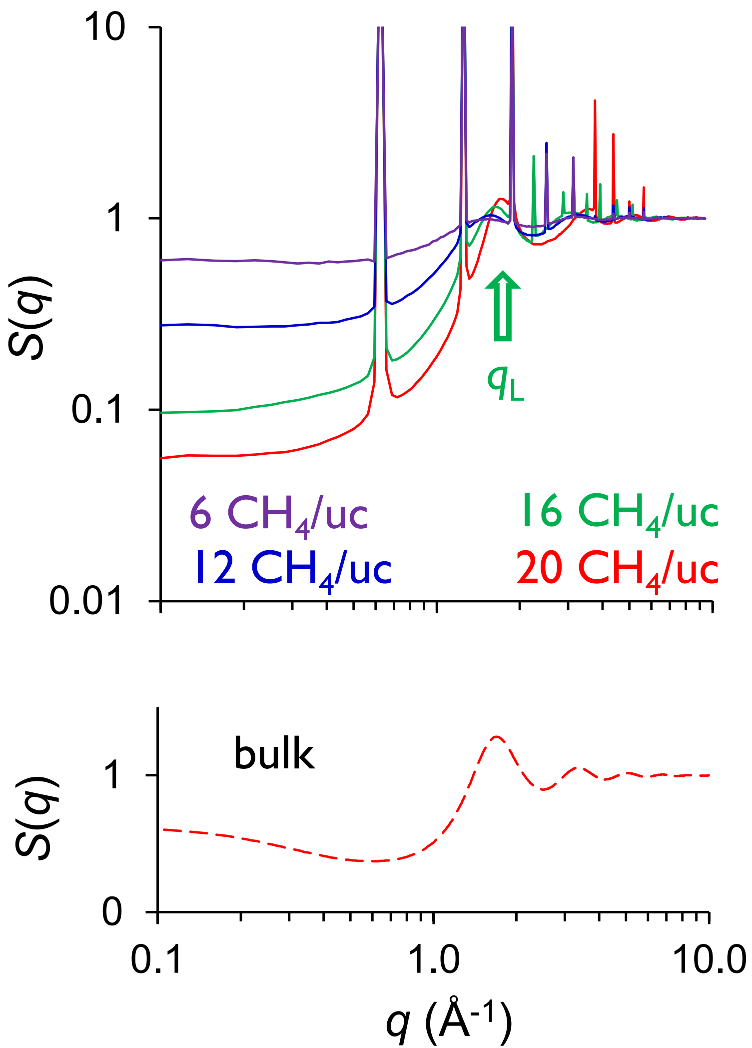
Fluid Structure Factor $S(q)$

$$S(\mathbf{q}) = \langle \rho(\mathbf{q})\rho^*(\mathbf{q}) \rangle$$

$$\rho(\mathbf{q}, t) = \int \rho(\mathbf{r}, t) \exp[i\mathbf{q} \cdot \mathbf{r}(t)] \, \mathrm{d}^3\mathbf{r}$$

$$= \sum_{i=1,N} \exp[i\mathbf{q} \cdot \mathbf{r}_i(t)]$$

- Strong Bragg peaks imposed bythe zeolite structure
- Main fluid correlation peak $\mathbf{q}_L$ at a similar wave vector than the bulk fluid
- Fluid correlation peak is narrower than the bulk as the bulk fluid as the fluid phase is more ordered (i.e. coherent ordering over larger distances)



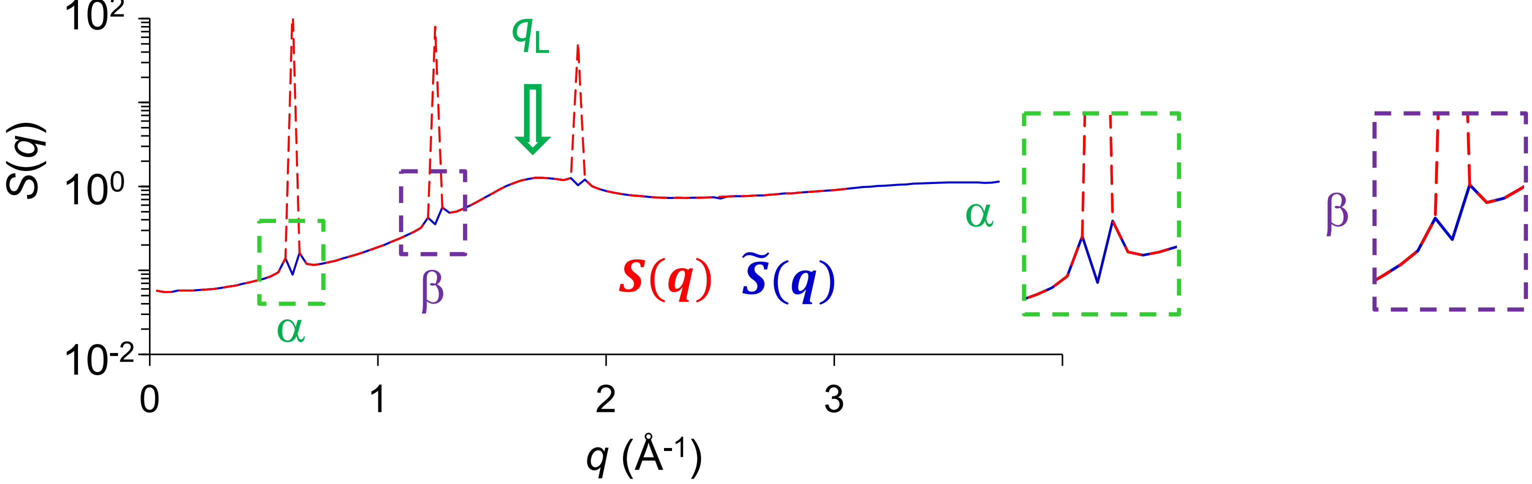
Fluctuations in *S*(*q*)

- These peaks are structural, i.e. static, features which are not involved in the dynamics
- Correction must be perfomed (correction not needed for q ≠ q_{Bragg})
- Surprisingly, the fluid is less correlated at these Bragg peaks

$$\tilde{\boldsymbol{S}}(\mathbf{q}) = \langle \delta\rho(\mathbf{q})\delta\rho^*(\mathbf{q})\rangle$$

where $\delta\rho(\mathbf{q}) = \rho(\boldsymbol{q}) - \langle\rho(\mathbf{q})\rangle$

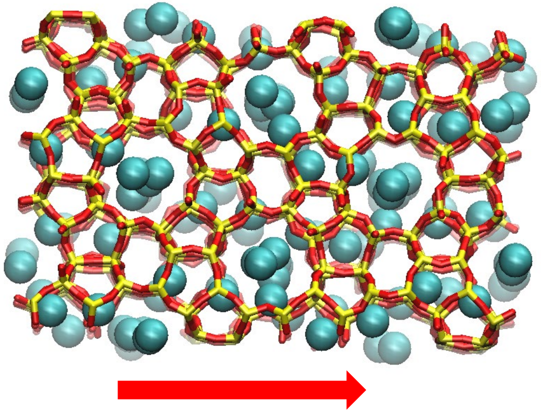
$$S(\mathbf{q}) = \langle \rho(\mathbf{q})\rho^*(\mathbf{q})\rangle$$



Collective Diffusion D_0

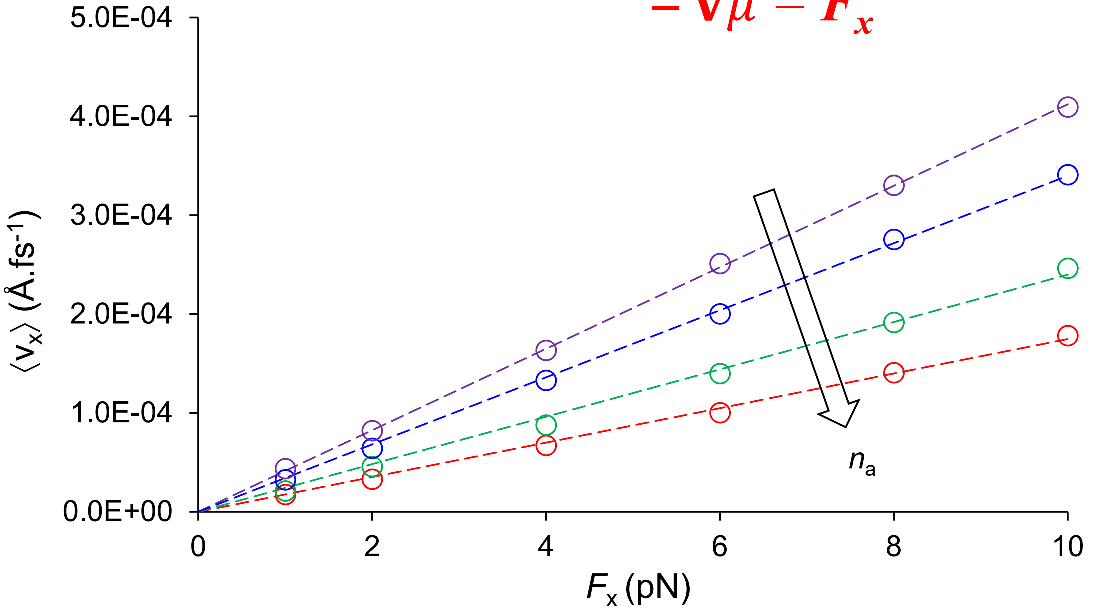
Non-equilibrium Molecular Dynamics

$$\mathbf{J} = -\rho \frac{D_0}{k_B T} \boldsymbol{\nabla}\mu \ = \rho \langle v_x \rangle$$



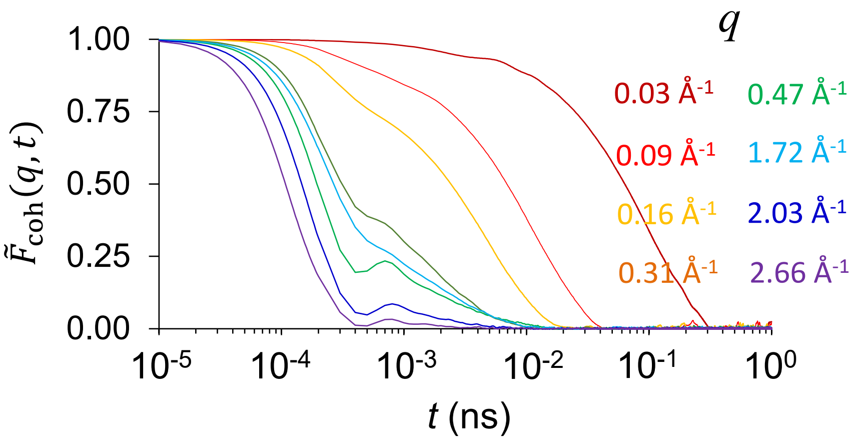
$$- \boldsymbol{\nabla}\mu = \boldsymbol{F_x}$$

- Linear regime verified
- Induced flow rate remains much smaller than the thermal motion
- D_0 decreases upon Increasing the loading n_a due to steric repulsion

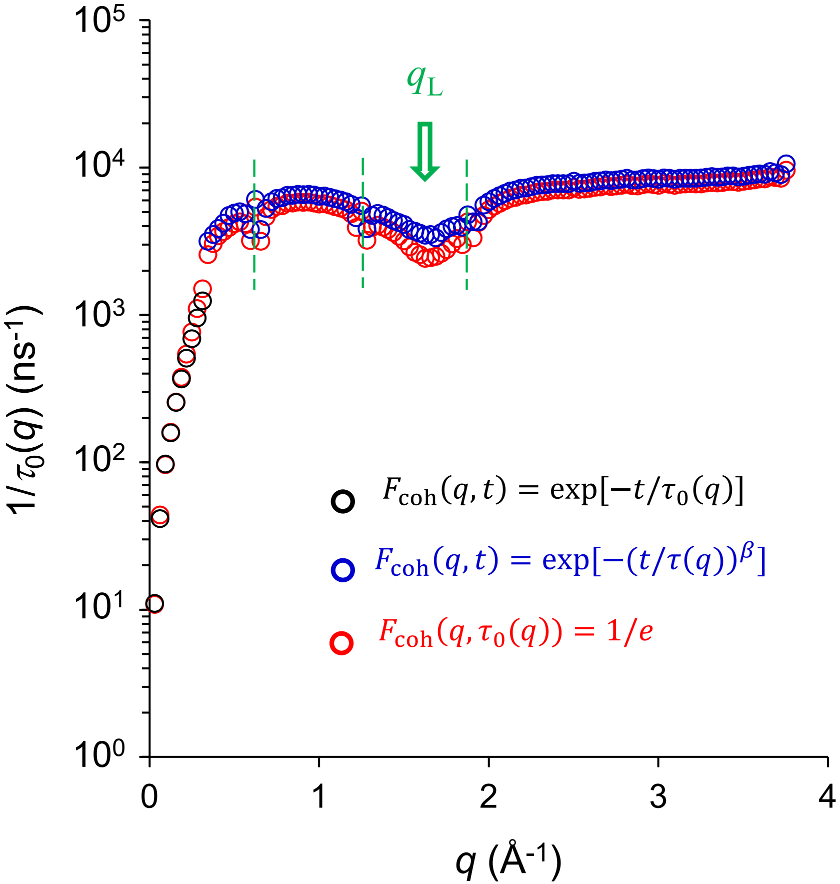


Coherent Scattering Functions

$$F_{\mathrm{coh}}(\mathbf{q},t) \sim \langle \rho(\mathbf{q},t)\rho^*(\mathbf{q},0)\rangle$$



- In qualitative agreement with De Gennes narrowing, marked variations are observed at wavevectors *q* where features are observed in *S*(*q*)



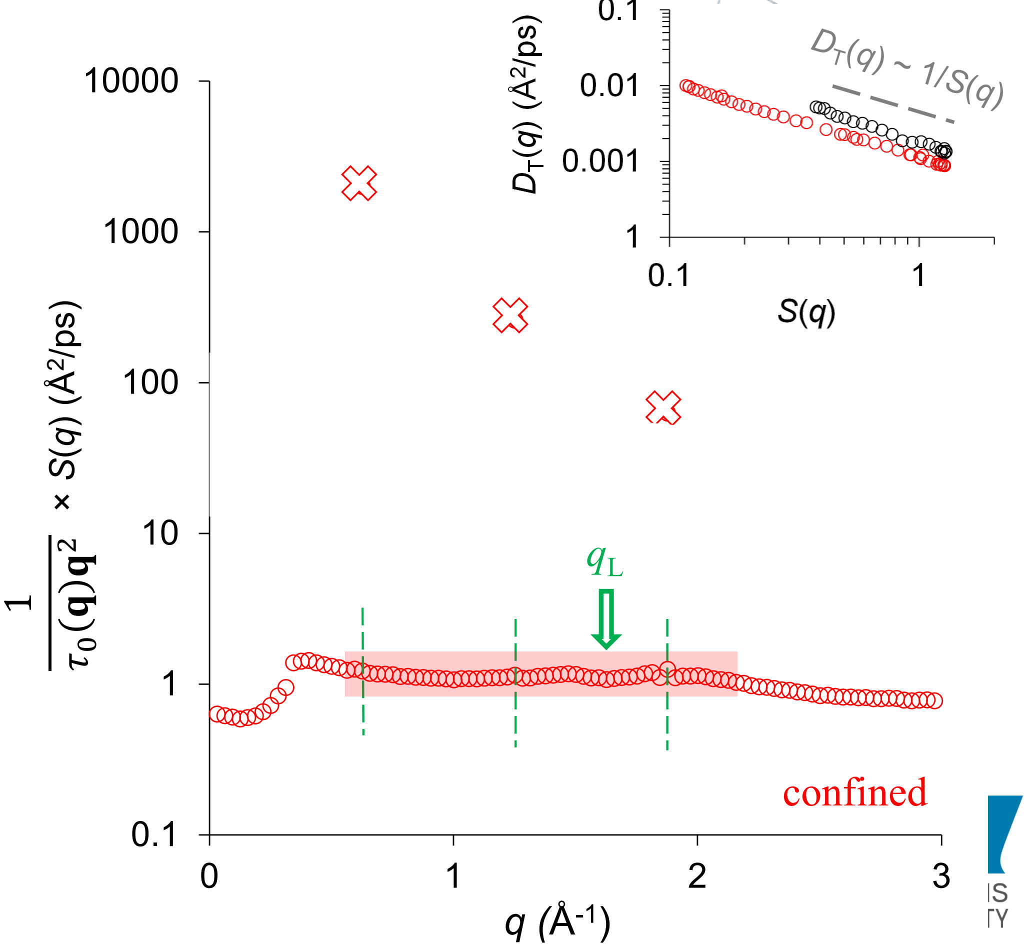
De Gennes Narrowing

$$\boxed{\frac{1}{\tau_0(q)q^2} = \frac{D_0}{S(q)}}$$

- De Gennes narrowing observed around the correlation peak $D_T(q)S(q) \sim$ constant

- Features are observed in the collective diffusivity and not in the self-diffusivity

- Correction for the static part of the structure factor is essential (see big red crosses)



Conclusions

A. Schlaich, J. L. Barrat, B. Coasne,
Theory and Modeling of Transport for Simple Fluids in Nanoporous Materials
Chem. Rev. **125**, 2561 (2025)

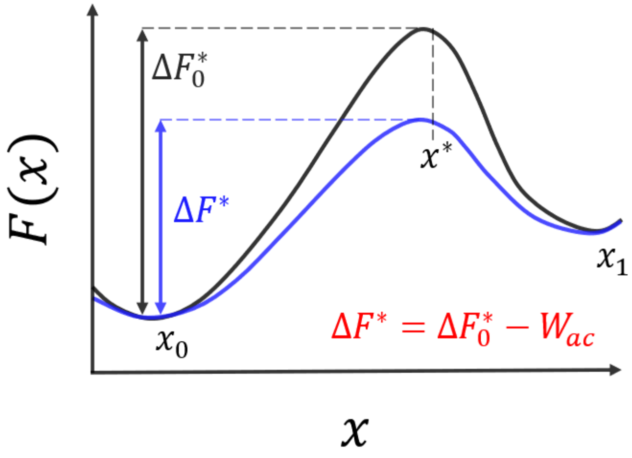
- Simple physical microscopic model – relying on available parameters to simple experiments – are available to describe diffusion and permeability of fluids in nanoporous media

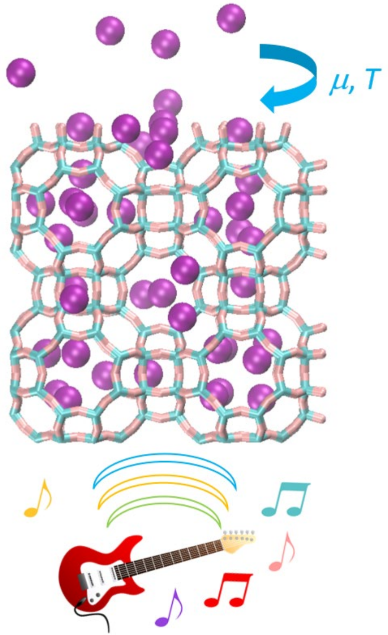
- Despite the simplicity of our system, we expect these predictions to hold when more complex systems are considered
(molecules with intramolecular modes, disordered materials, etc.)

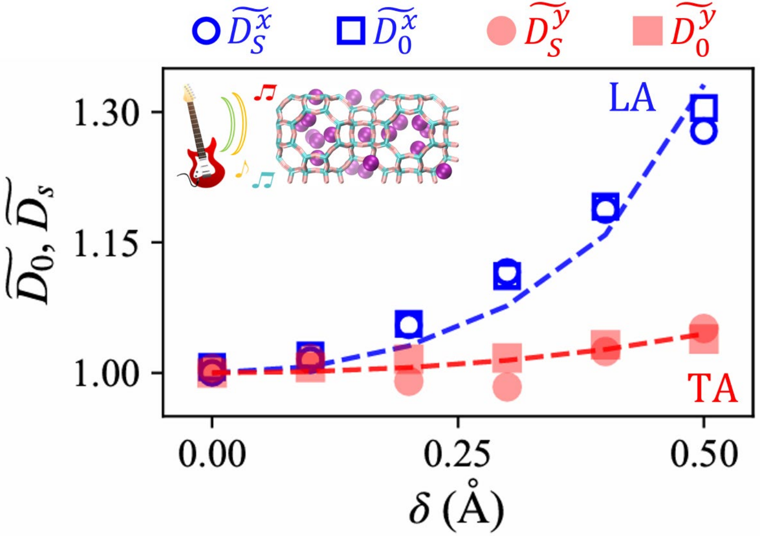
- These powerful frameworks allow predicting and rationalizing macroscopic transport in nanoporous media from a molecular physics perspective (statistical mechanics)

Perspective (1)

Explore acoustic stimulation of the adsorption and transport of fluids confined in nanoporous materials.







Strong acoustic-driven increase in self and collective diffusion as the work of the acoustic force decreases the free energy barriers

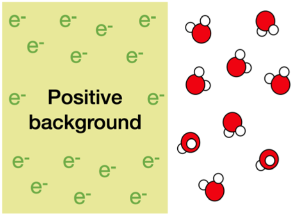
$$D_0(\delta) \sim \exp\left[-\beta(\Delta F_0^* - F_{ac}\delta)\right]$$

$$\sim D_0(0)\exp\left[\beta F_{ac}\delta\right]$$

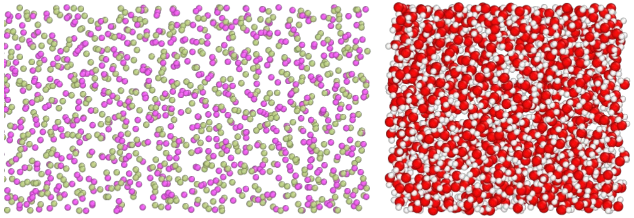
Perspective (2)

Quantum (electronic) effects in confining metal surfaces lead to electrostatic screening → impact on the thermodynamics and dynamics of interfacial fluids at metallic surfaces

Physical problem

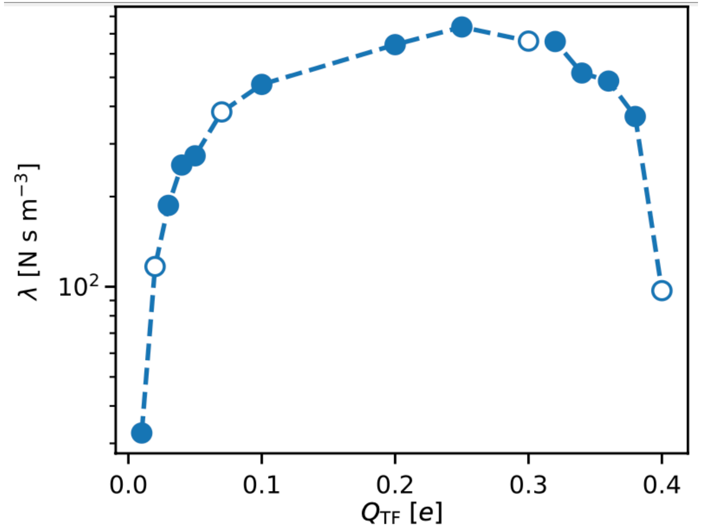


Effective modeling using *Virtual Thomas-Fermi fluids*



- We report a dynamic intwertwining: solid and liquid dynamics are coupled through friction, strongly affected by metallicity (Figure)

- This coupled dynamics can be envisaged for energy harvesting by generating electric currents through liquid flowing. This behavior is reported in our molecular simulation is in agreement with theoretical predictions.



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