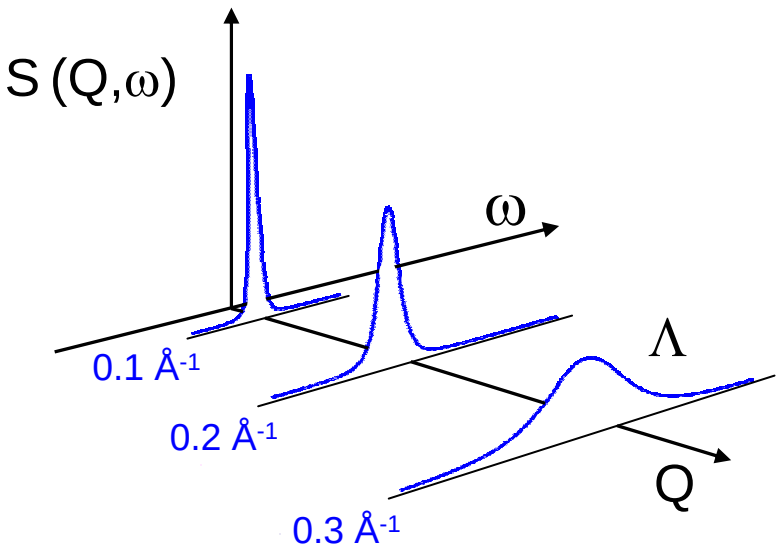
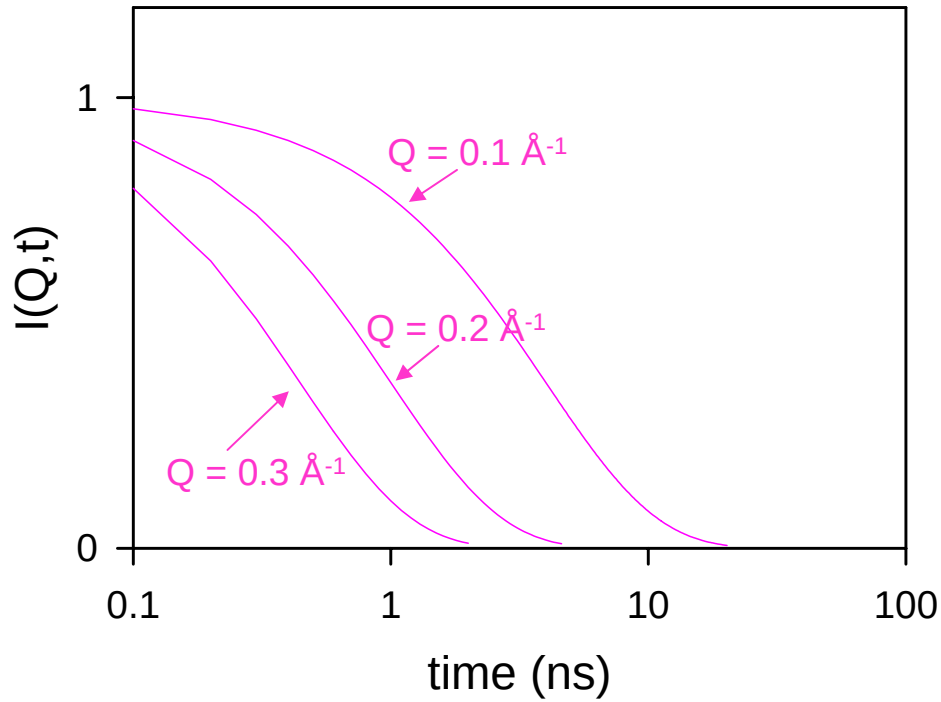
Diffusion studies by QENS measurements approaching the ‘ideal‘ situation

Hervé Jobic

Institut de Recherches sur la Catalyse, CNRS, 2 avenue Albert Einstein, 69626 Villeurbanne

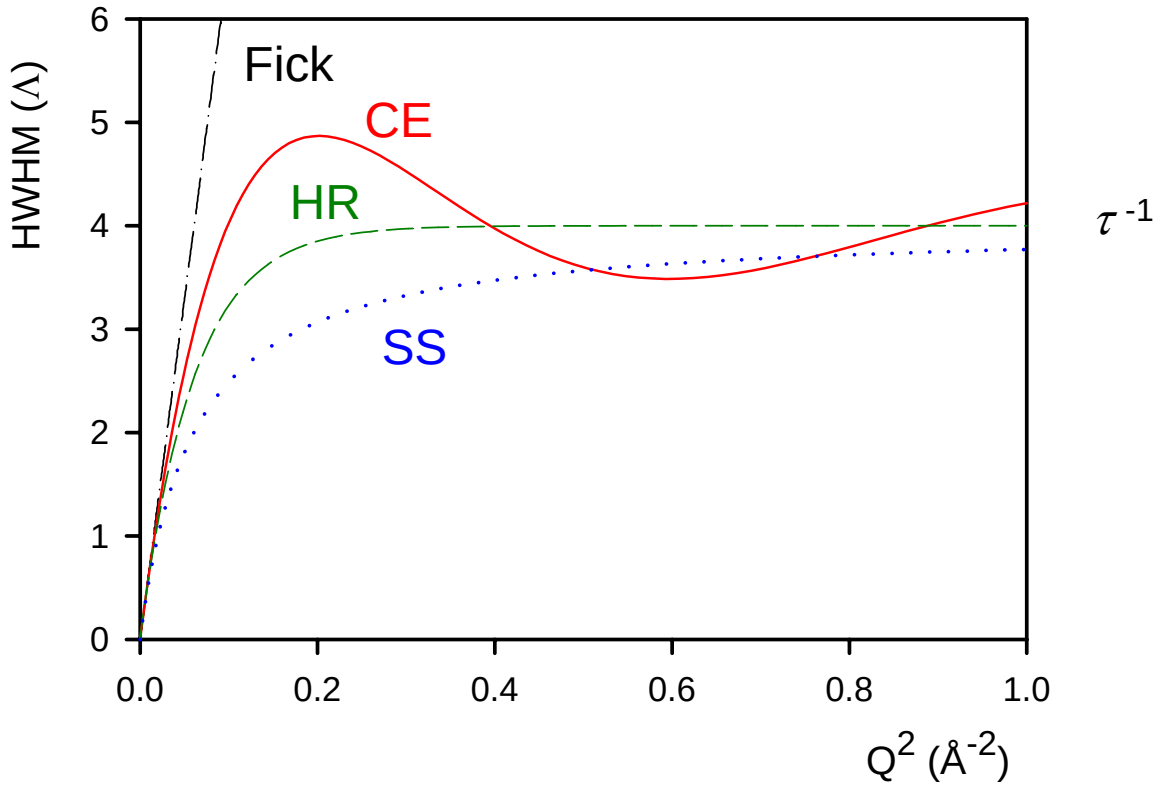




Quasi-elastic scattering:
QENS
TOF - BS

Neutron spin echo
NSE

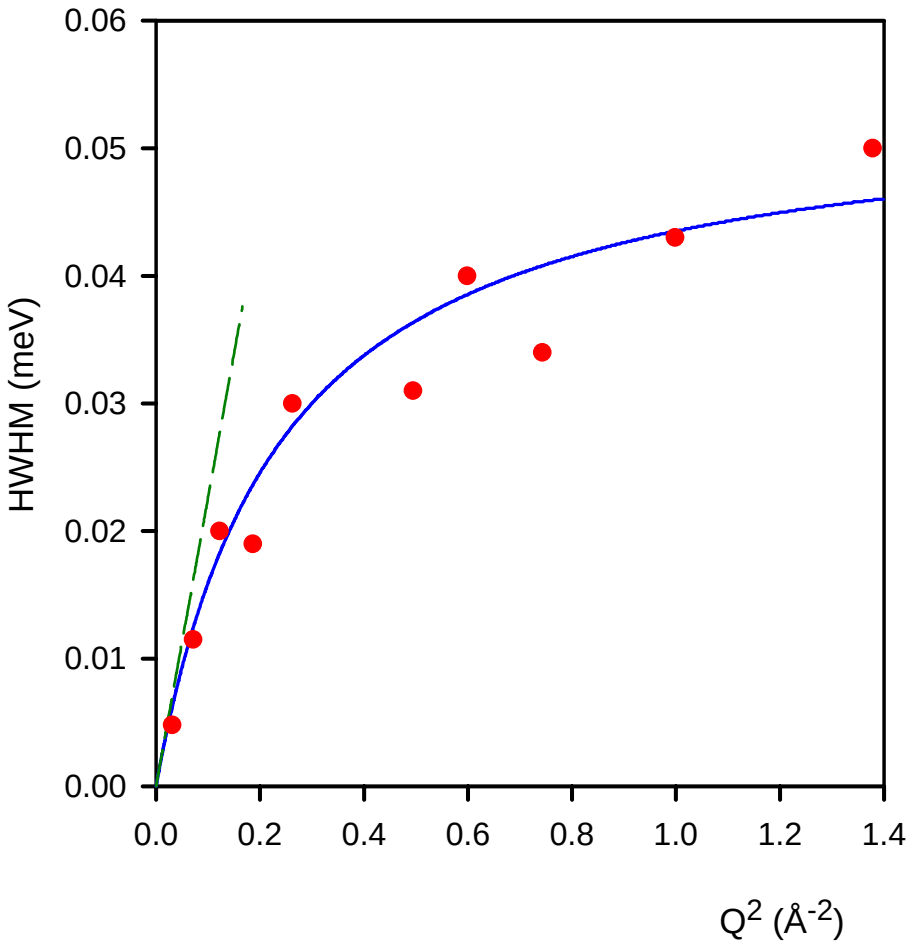
Fick: DQ^2



$\langle r^2 \rangle^{1/2}$ = 10 Å
$\tau \equiv$
} same D

NH_3 / silicalite

4.3 molecules / u.c. T = 360 K



SS model

$\langle d \rangle^{1/2}$ = 5 Å

τ = 12 ps

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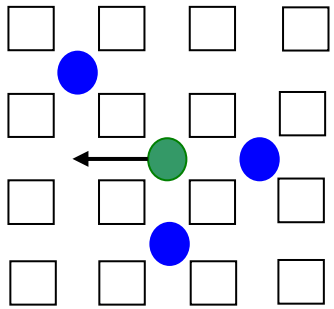
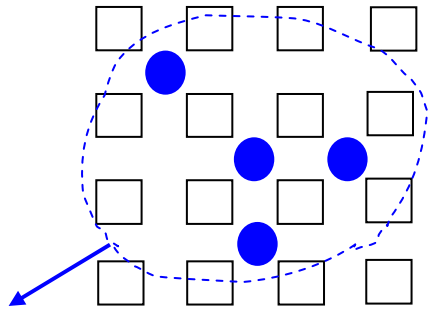




$$\frac{\partial G_S(\mathbf{r},t)}{\partial t} = D_s \nabla^2 G_S(\mathbf{r},t)$$

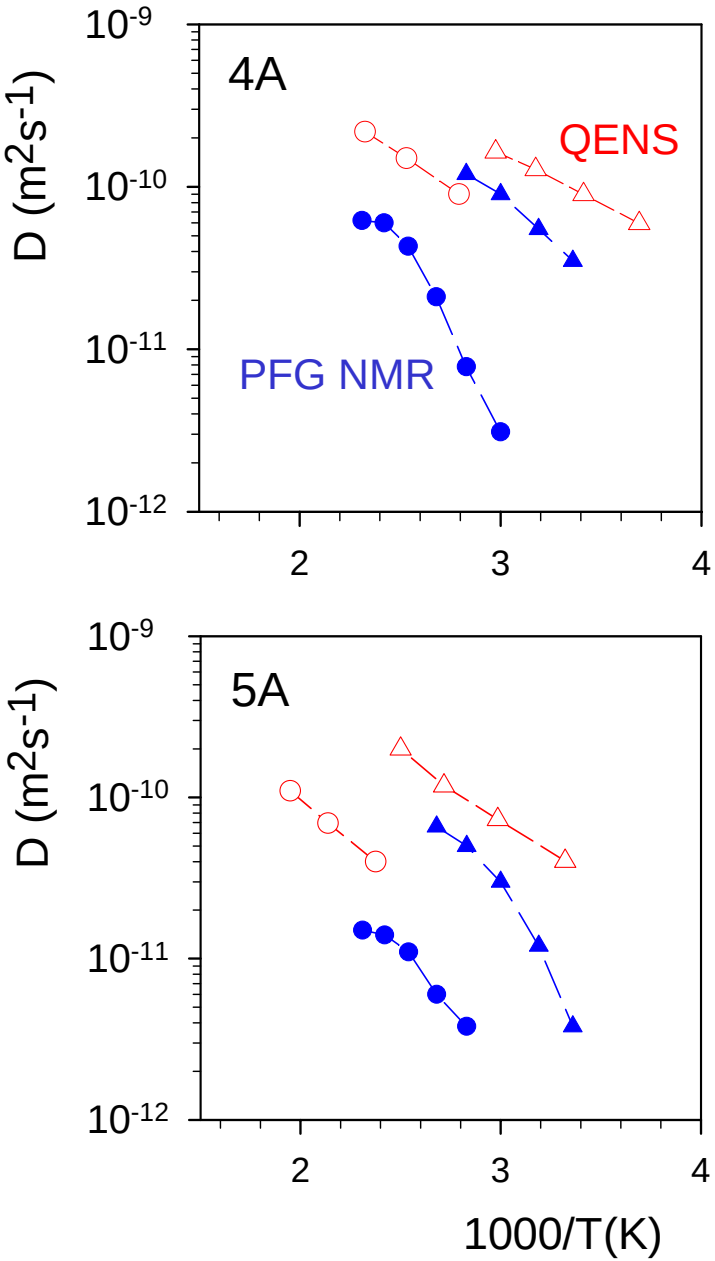
hydrogenated molecules

$$\frac{\partial \rho(\mathbf{r},t)}{\partial t} = D_t \nabla^2 \rho(\mathbf{r},t)$$

deuterated molecules
+ O_2, N_2, CO_2, SF_6…

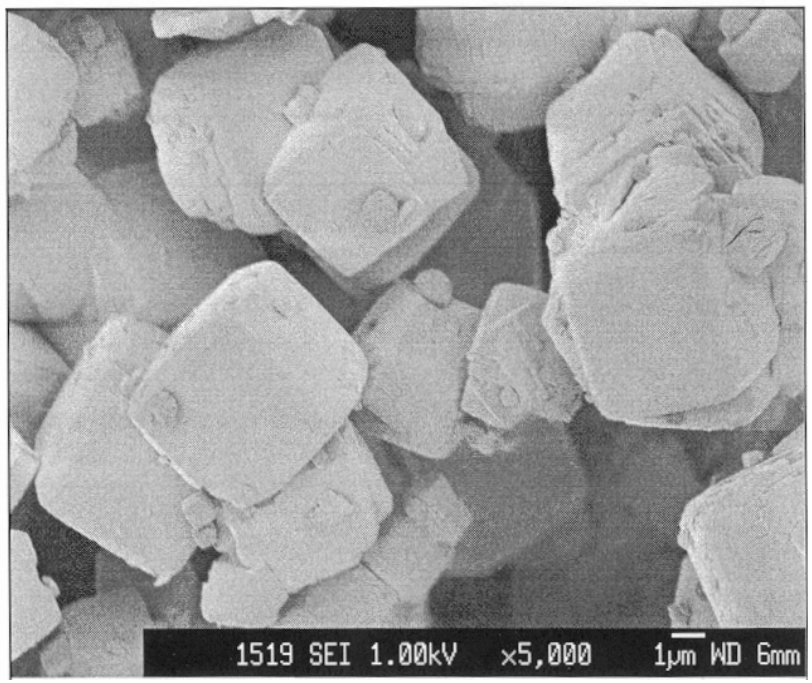
$$D_t = D_0 \frac{d \ln p}{d \ln c} = D_0 \Gamma$$

H_2O in type A zeolites



circles: 5 H_2O per α-cage

triangles: 15 H_2O per α-cage



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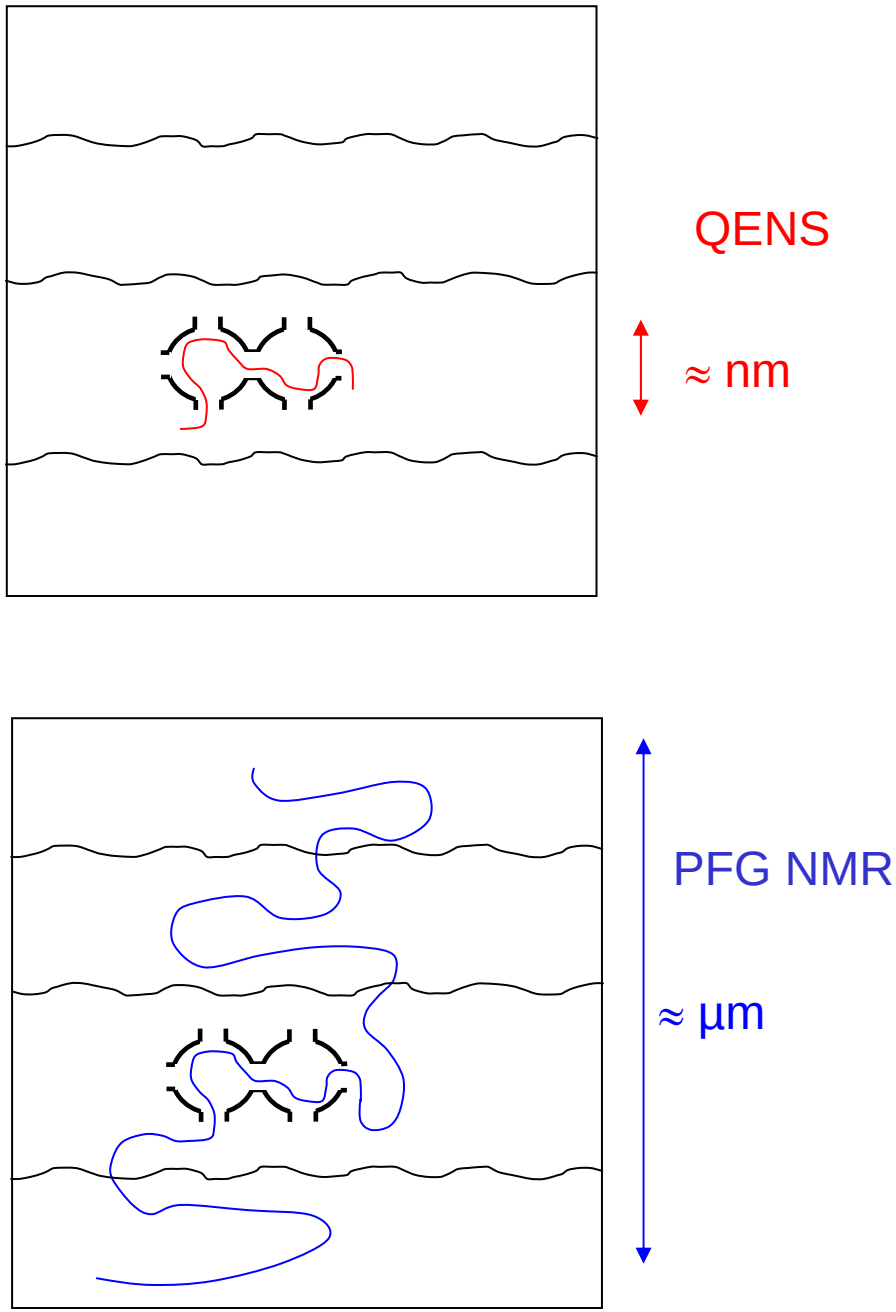
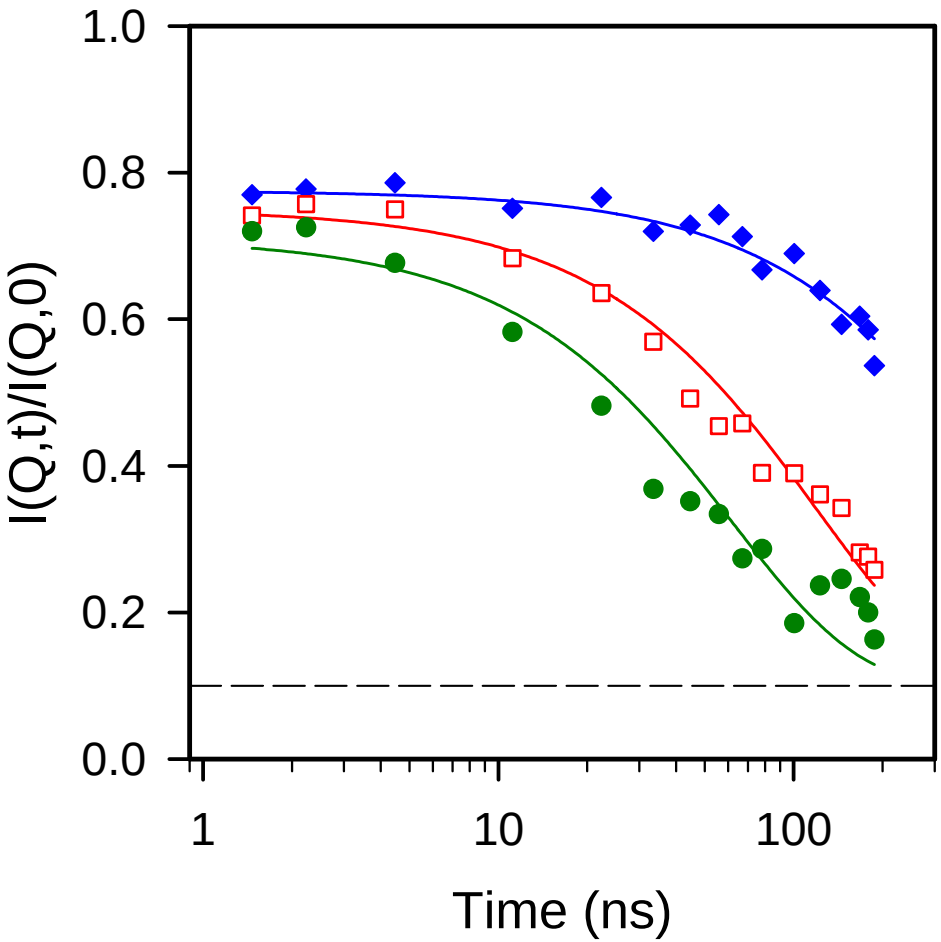

QENS
≈ nm
PFG NMR
≈ µm

n-alkanes in NaCaA (5A)

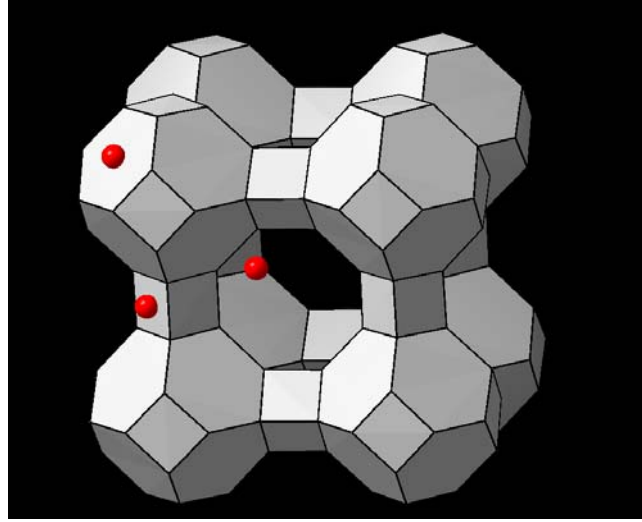
(T = 475 K, Q = 0.2 Å^{-1})



octane

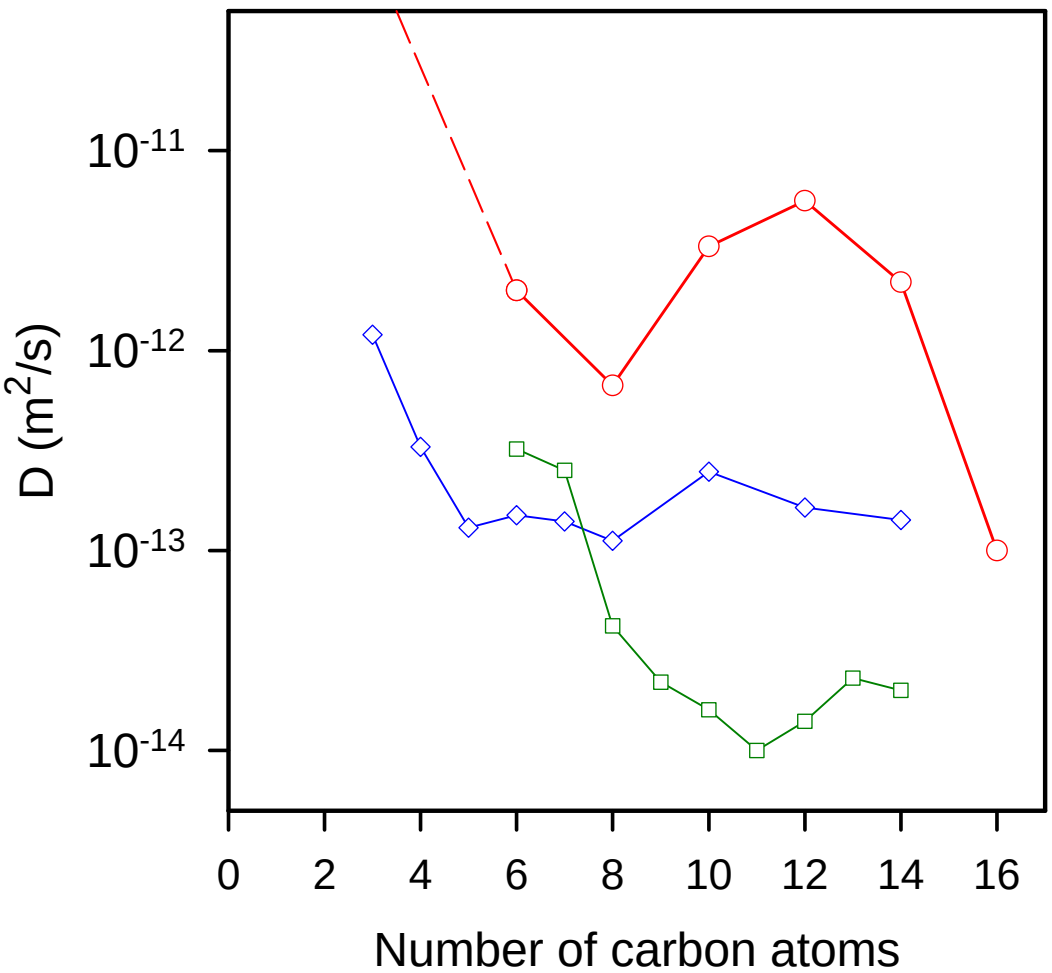
decane

dodecane



12 C / α-cage

n-alkanes in 5A

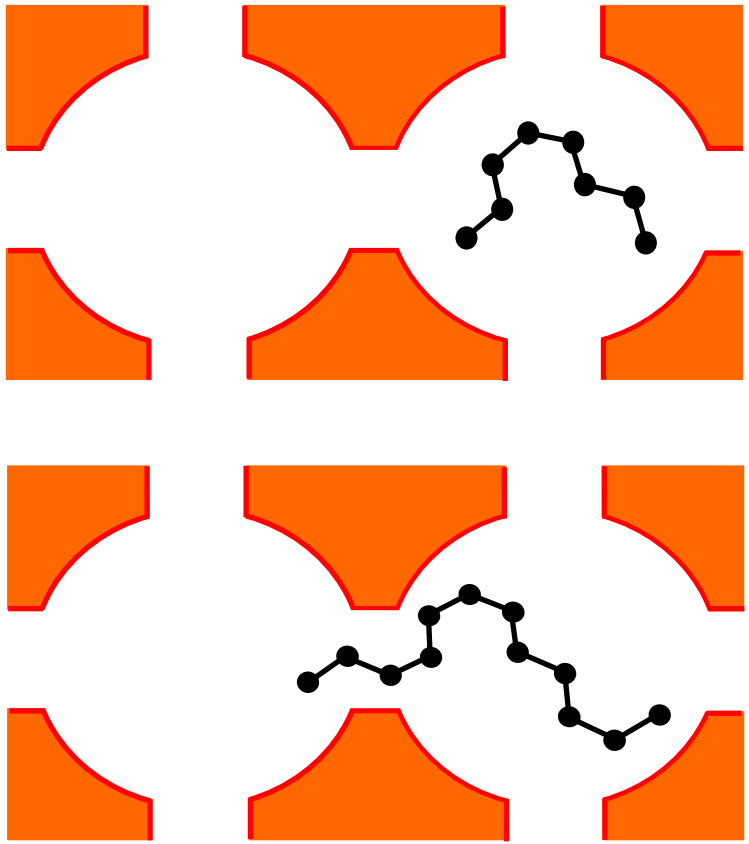


NSE (Dt)
475 K

PFG NMR (Ds)
423 K

ZLC (Do)
473 K

‘Window effect’



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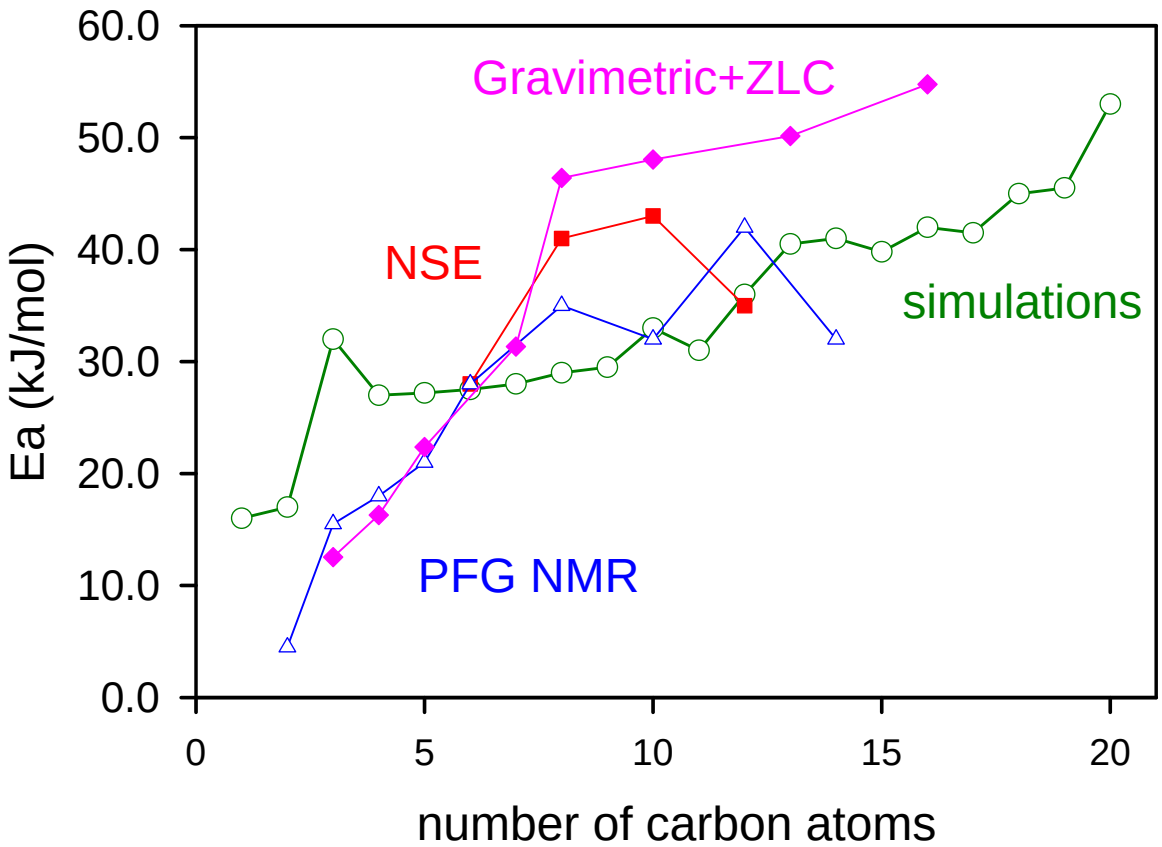
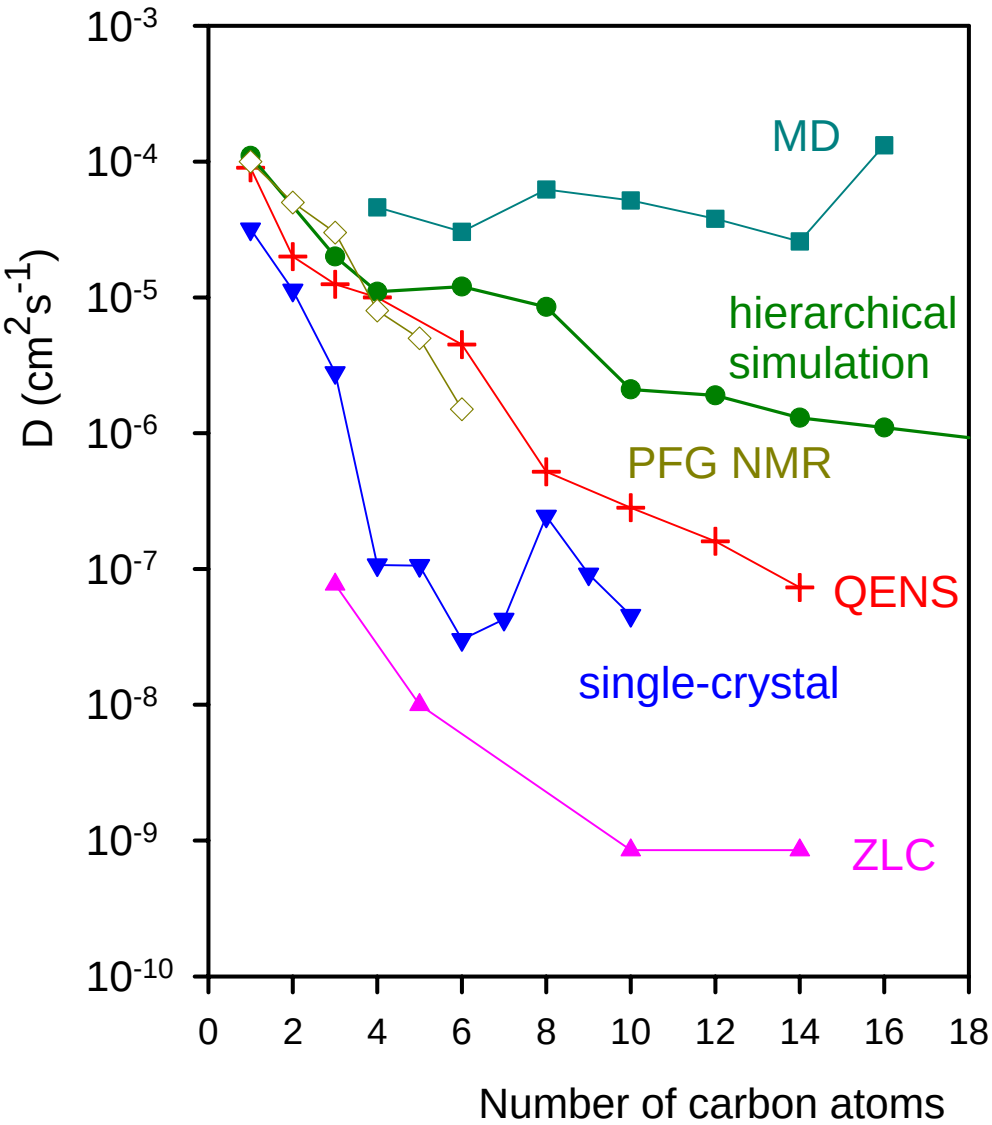
Ea *n*-alkanes in 5A



Ds *n*-alkanes / MFI
(T = 300 K)



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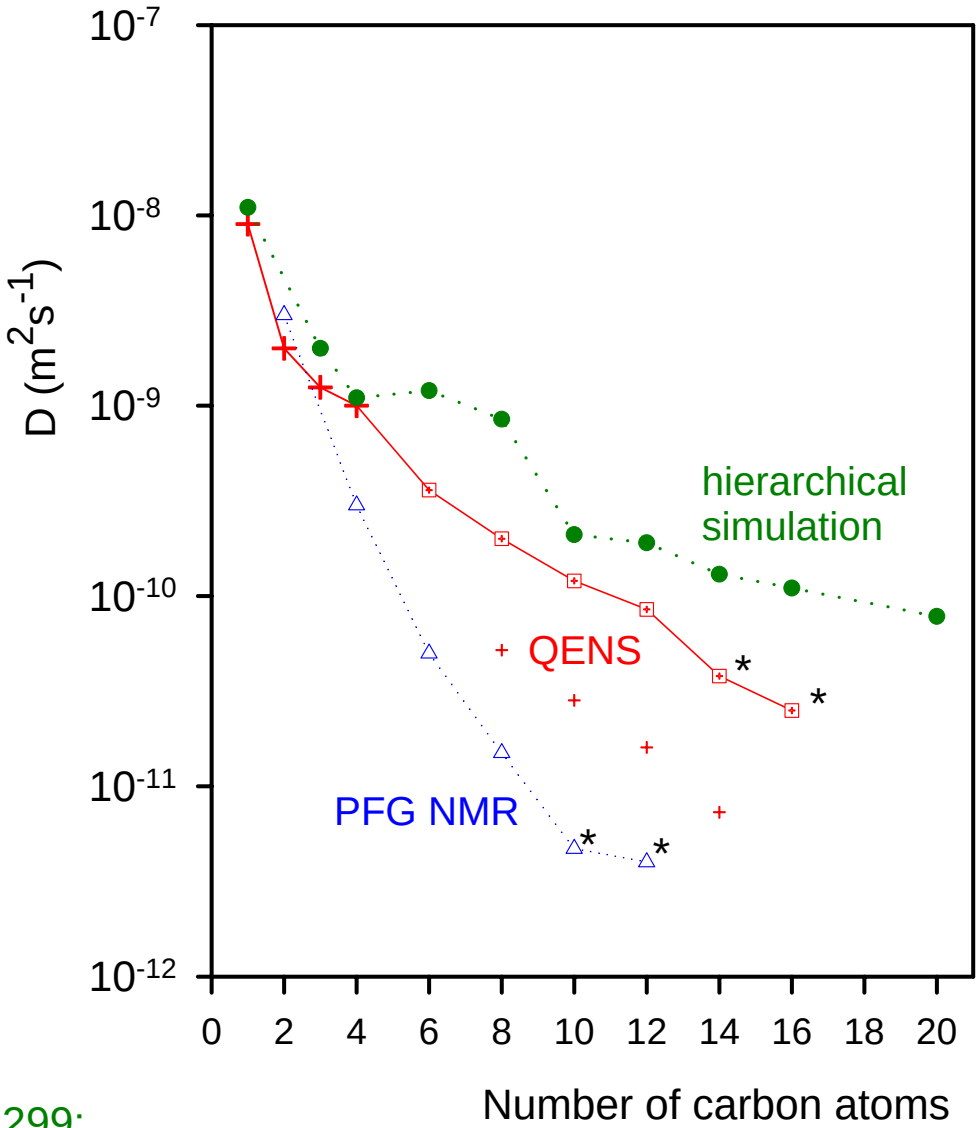
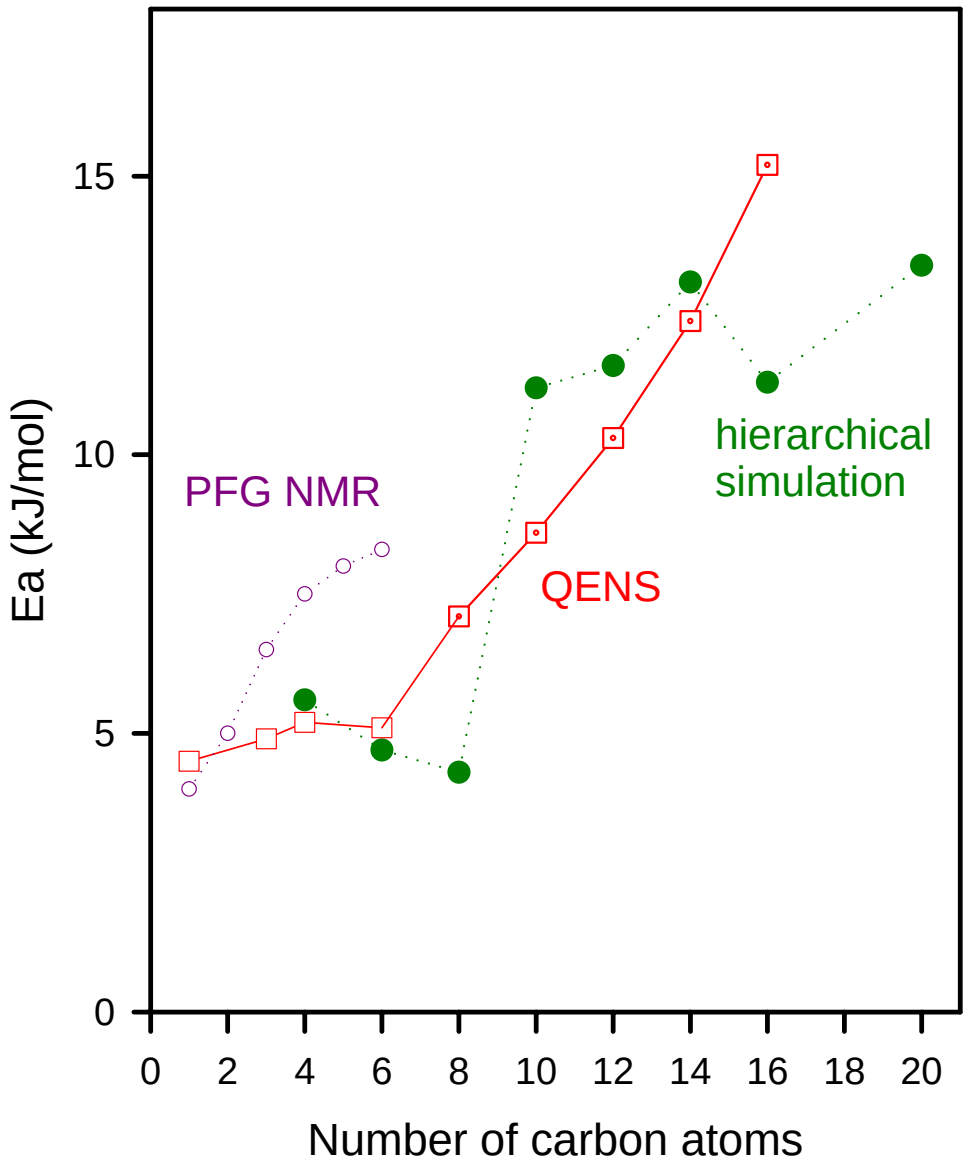
Ds *n*-alkanes / silicalite (T = 300 K)

50x160 µm

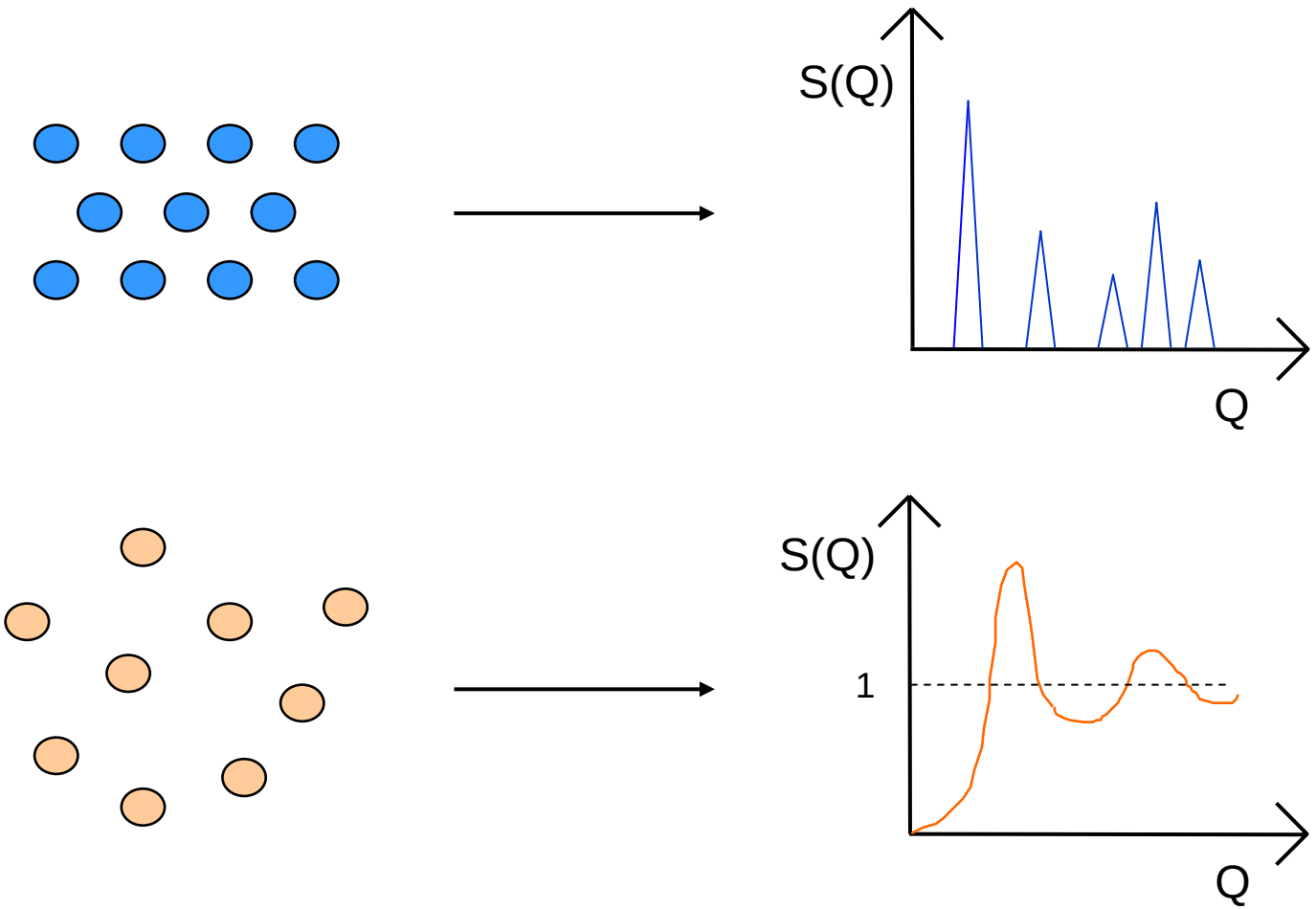


Microp .Mesop. Mater. 90 (2006) 299;
J.Phys. Chem. B 110 (2006) 1964

Ea *n*-alkanes / silicalite



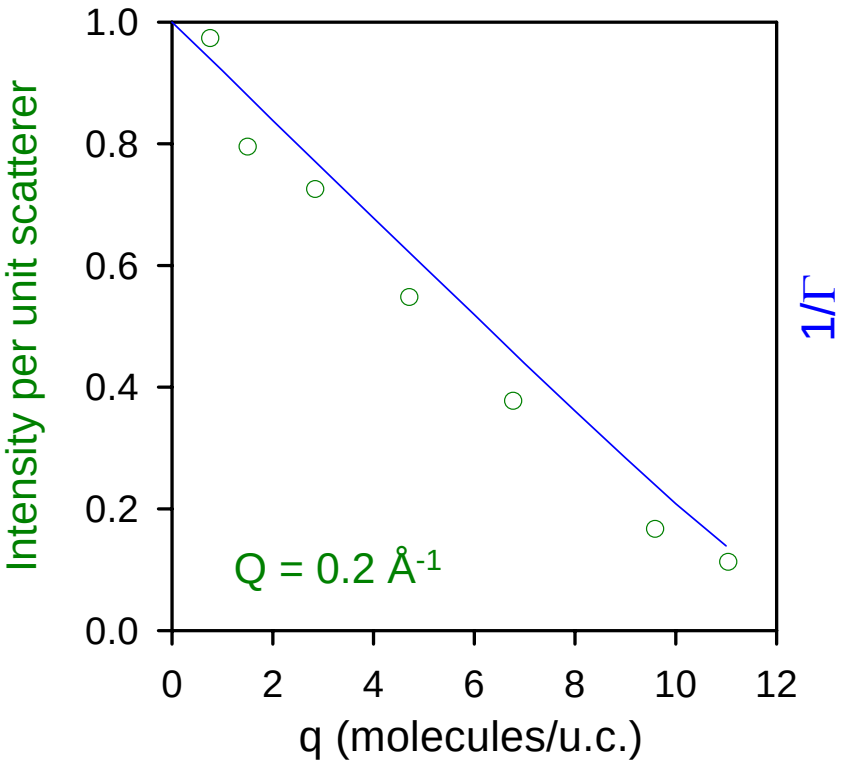
Coherent scattering

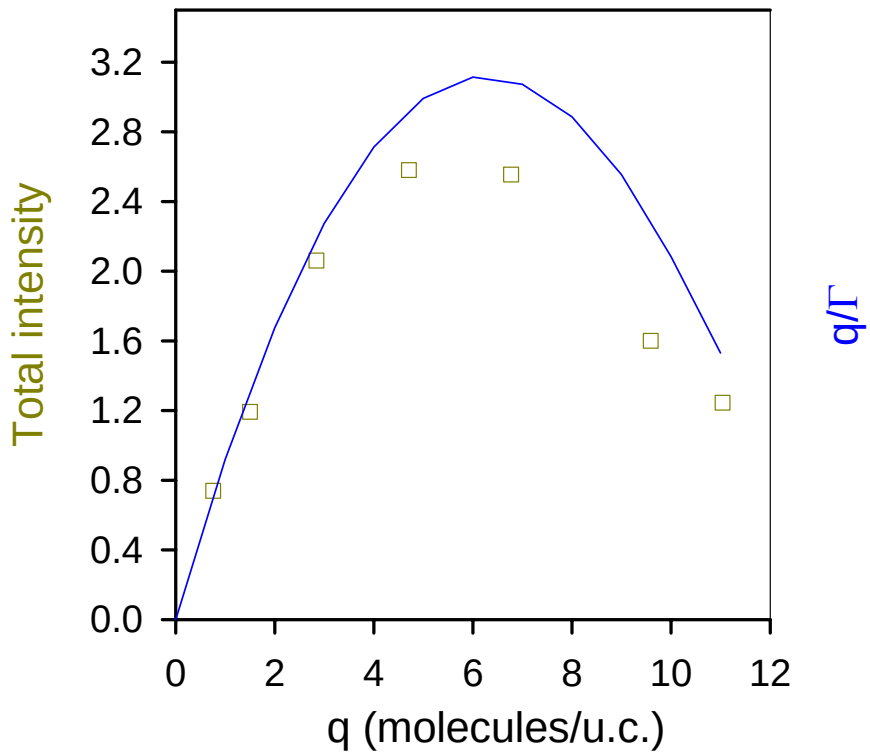


$$S(Q) = 1 + \int g(r) \exp(i\mathbf{Q}.\mathbf{r})\, d\mathbf{r}$$

$$S(Q) = \int S(Q,\omega)\, d\omega$$

C_2D_6 in silicalite @ 300K

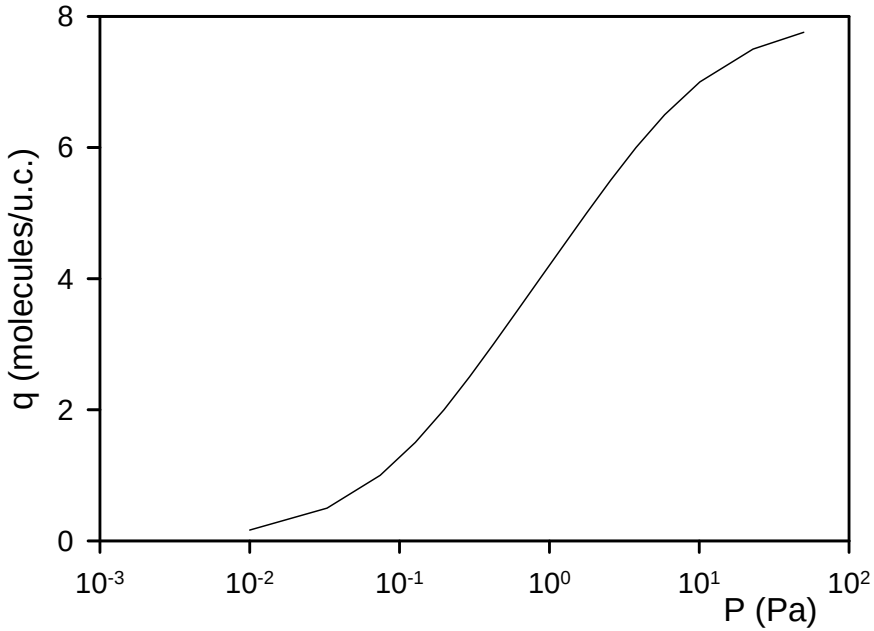


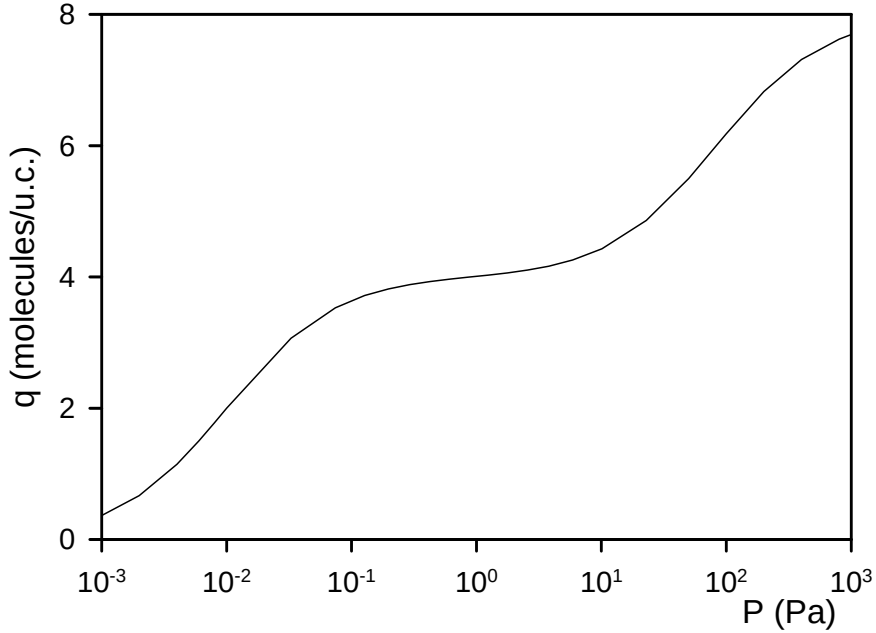


$$S(Q)_{Q\to 0} = \frac{< N^2 > - < N >^2}{< N >} = \rho k_B T \kappa_T$$

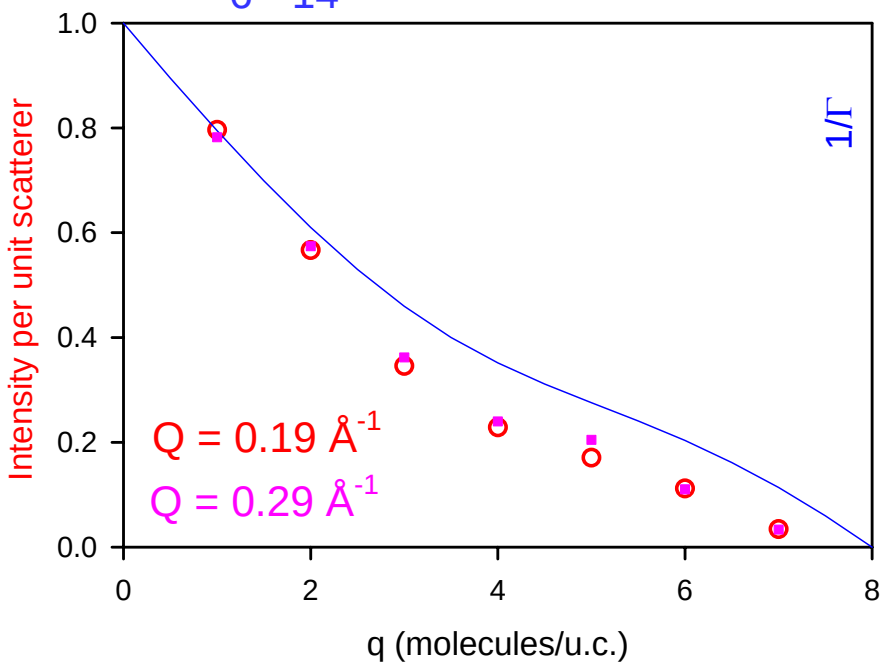
$$\kappa_T = \frac{1}{\rho k_B T}\left(\frac{d \ln q}{d \ln p}\right)$$

$$S(Q)_{Q\to 0} = \frac{d \ln q}{d \ln p} = \frac{1}{\Gamma}$$

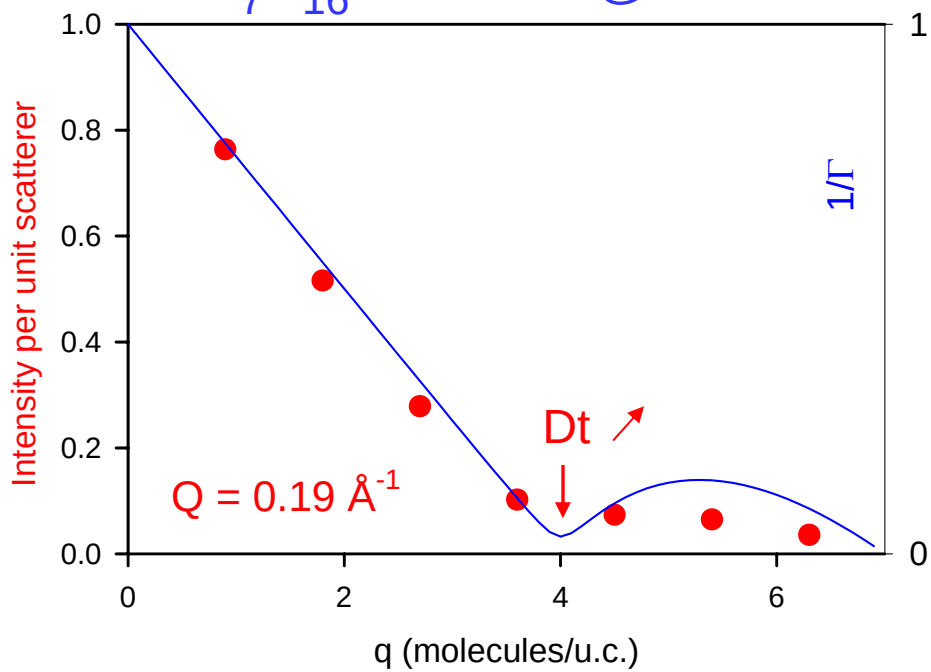




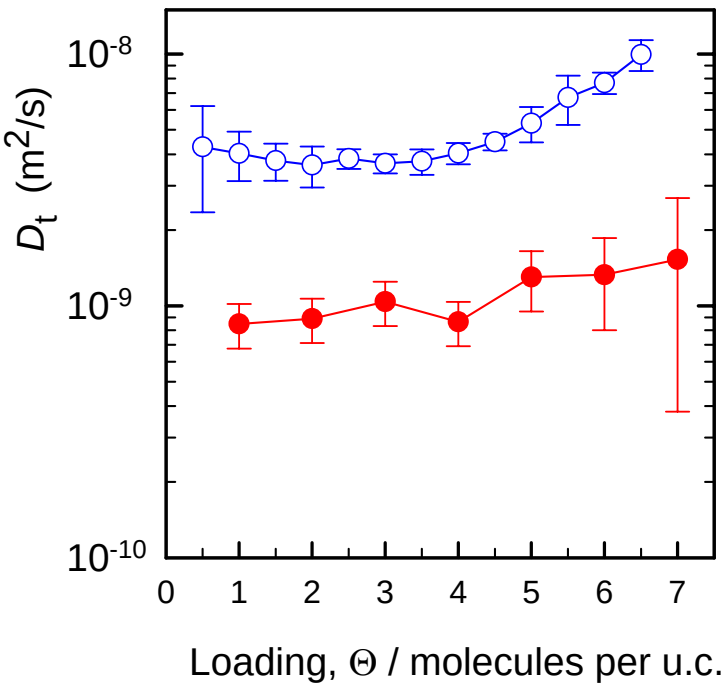
n-C_6D_{14} / silicalite @ 300K



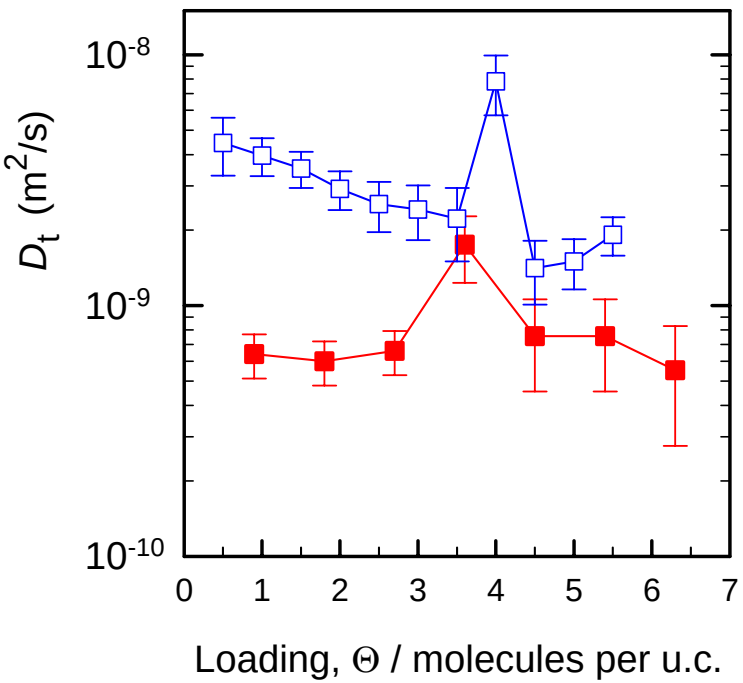
n-C_7D_{16} / silicalite @ 300K



D_t C_6/silicalite @ 300K



D_t C_7/silicalite @ 300K



MD

QENS

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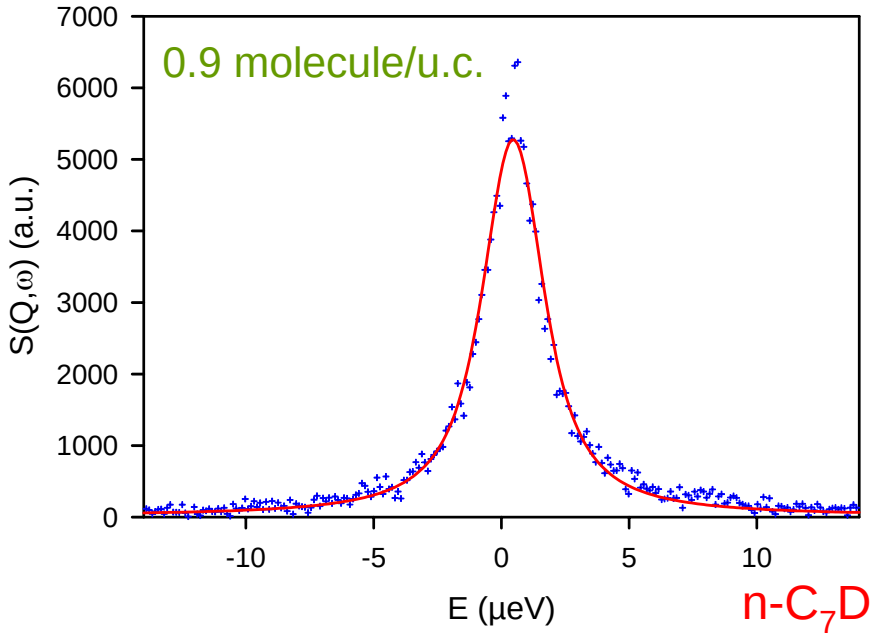
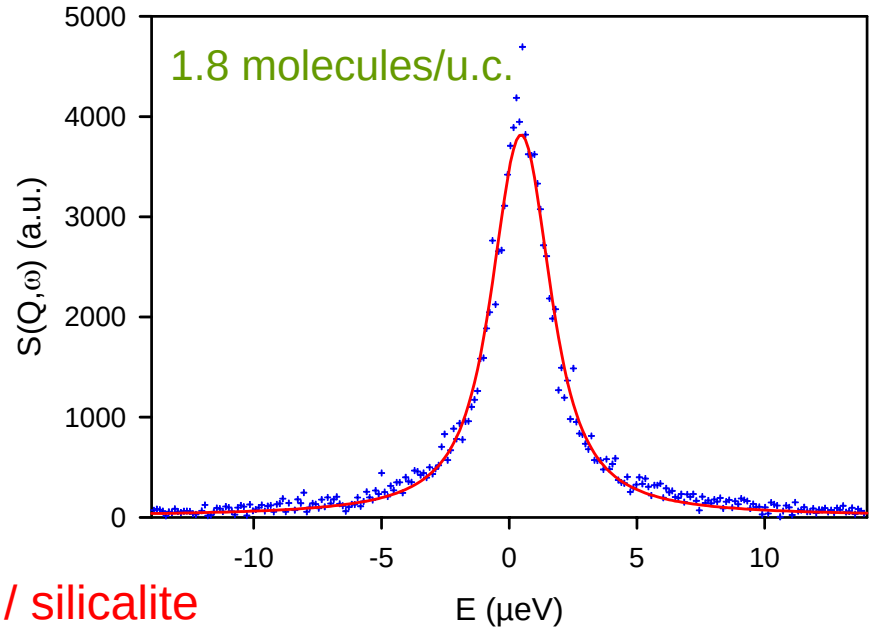
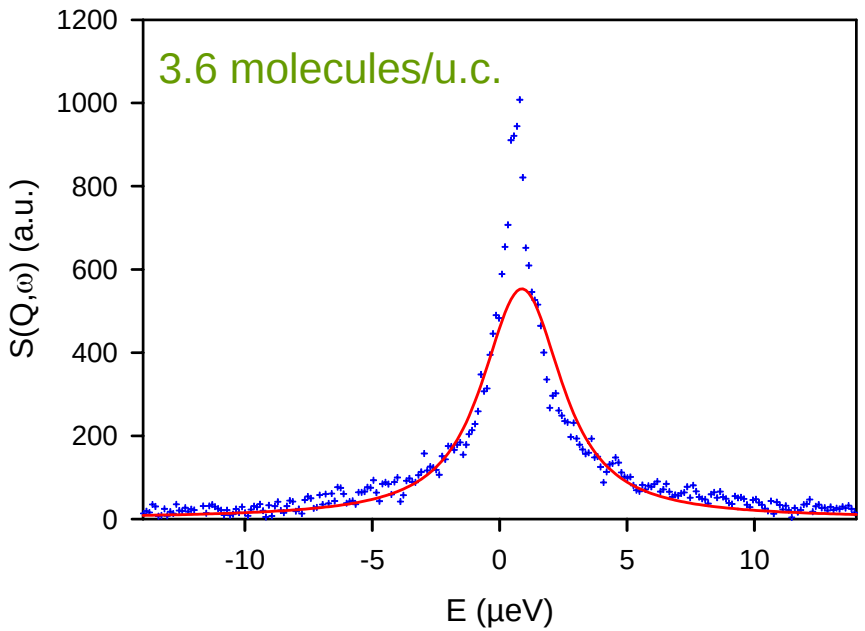
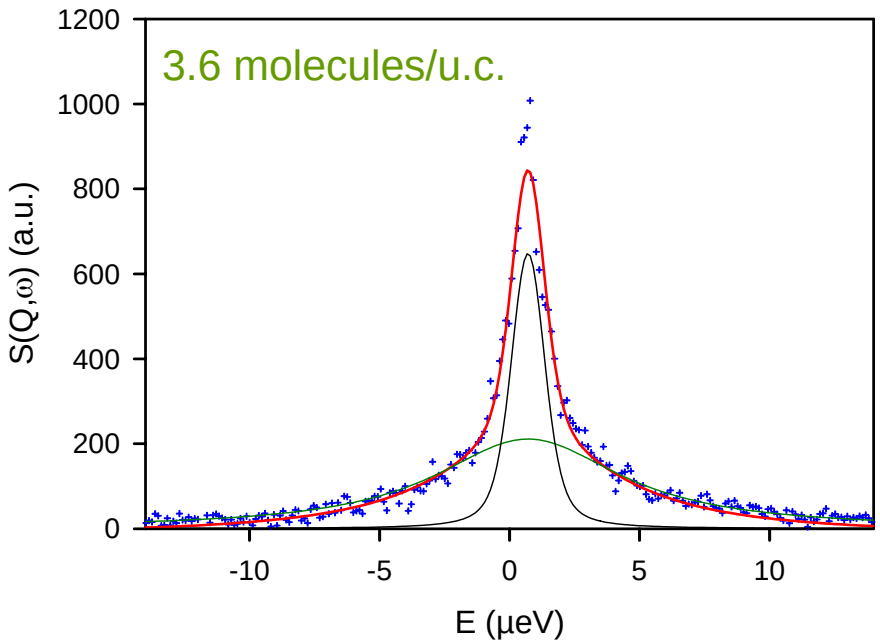




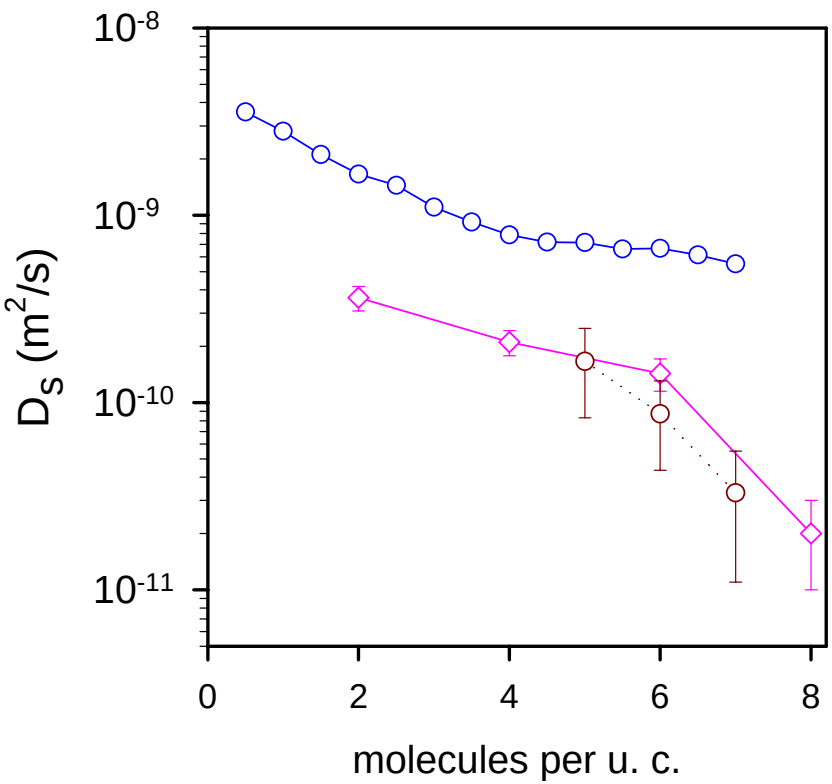
n-C_7D_{16} / silicalite

Q = 0.19 $Å^{-1}$

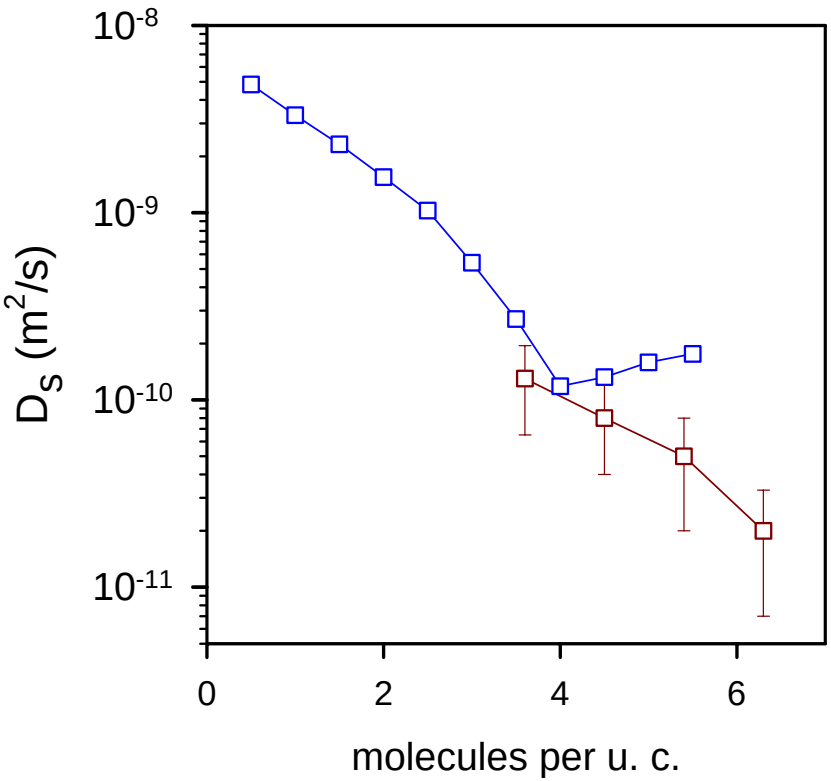




D_s C_6/silicalite @ 300K



D_s C_7/silicalite @ 300K

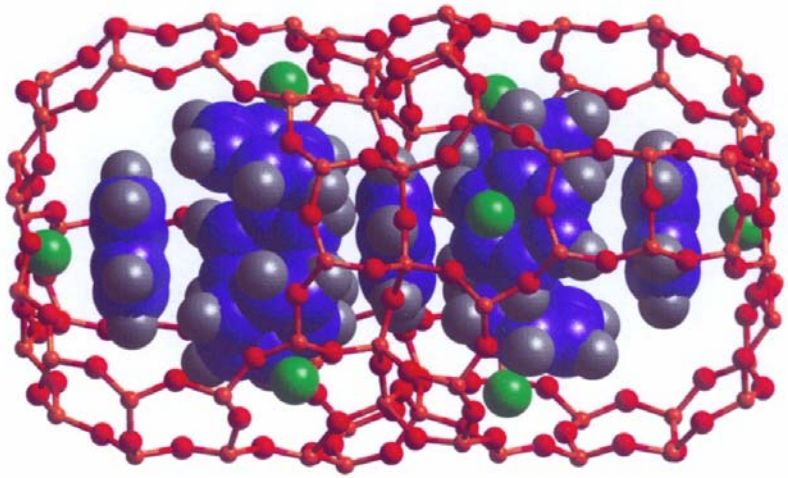


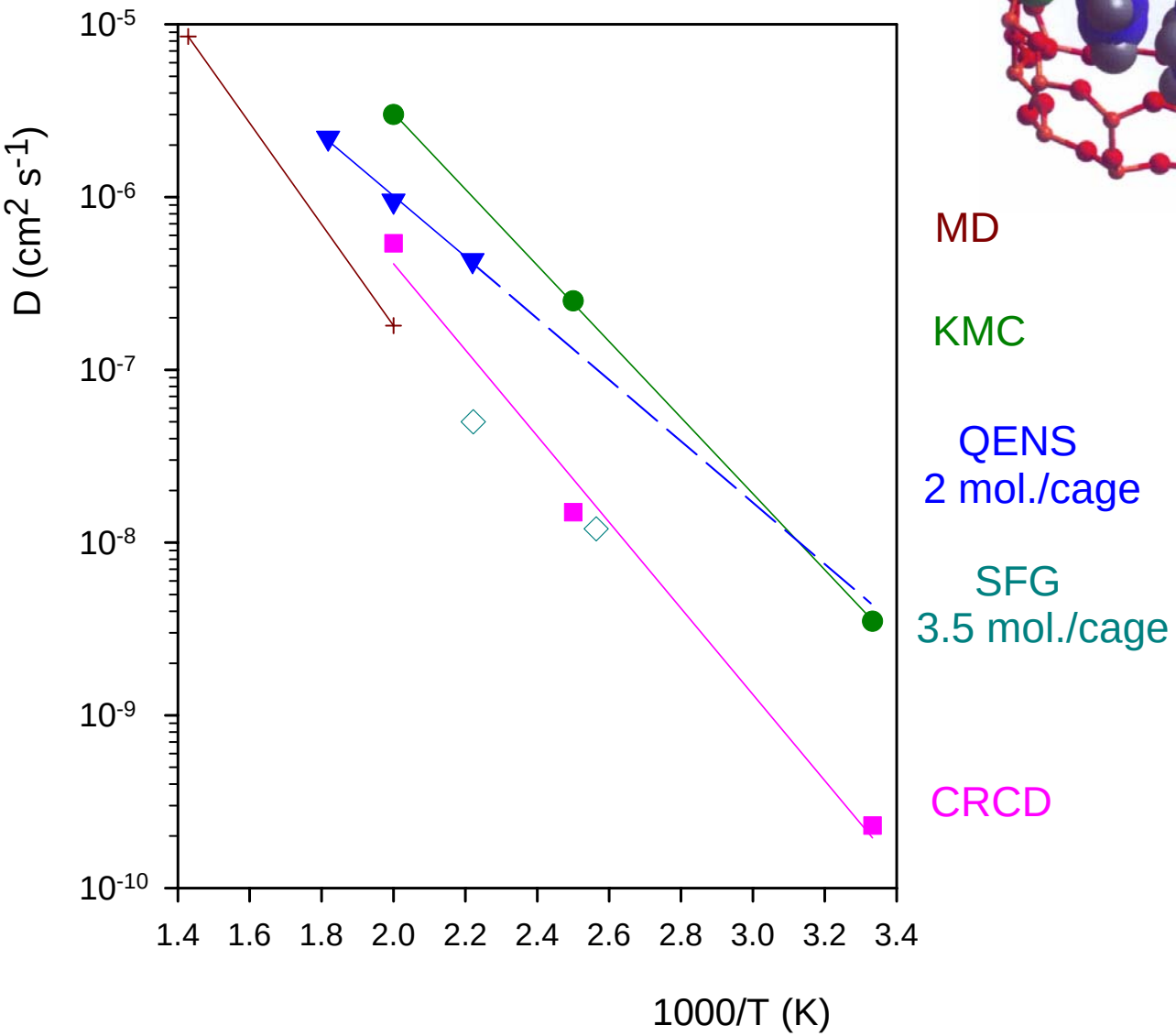
MD

inc QENS

coh QENS

Ds Benzene/NaY





MD

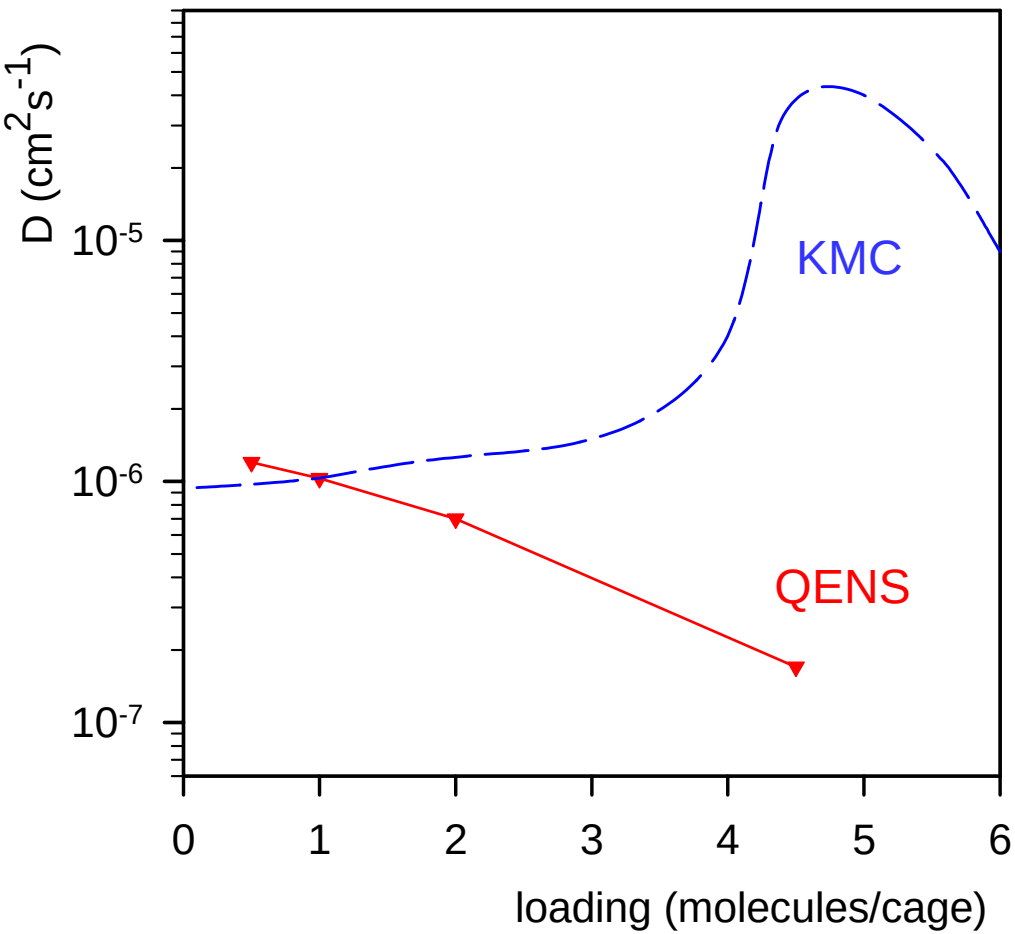
KMC

QENS
2 mol./cage

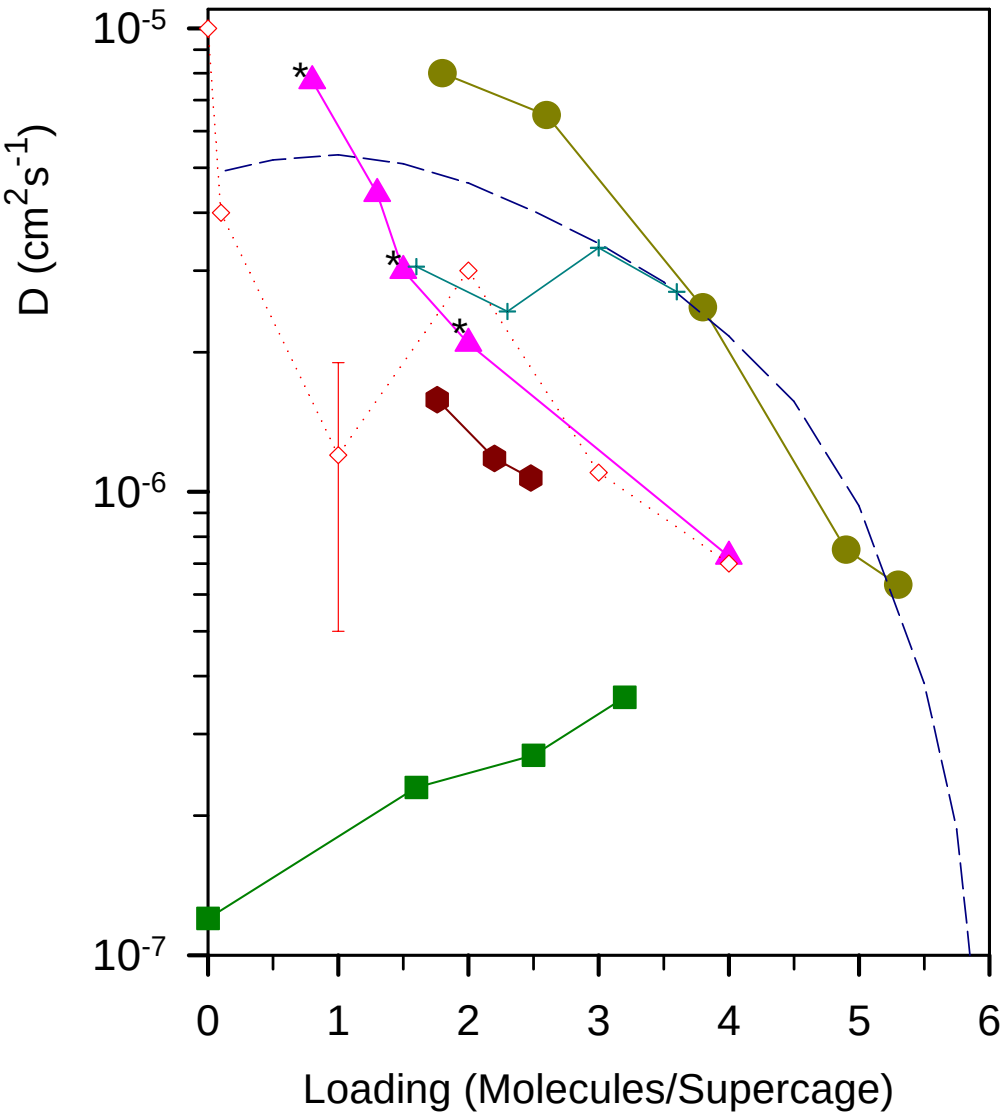
SFG
3.5 mol./cage

CRCD

Ds Benzene/NaY @ 480 K



Ds Benzene / NaX (T = 468 K)



PFG NMR

QENS

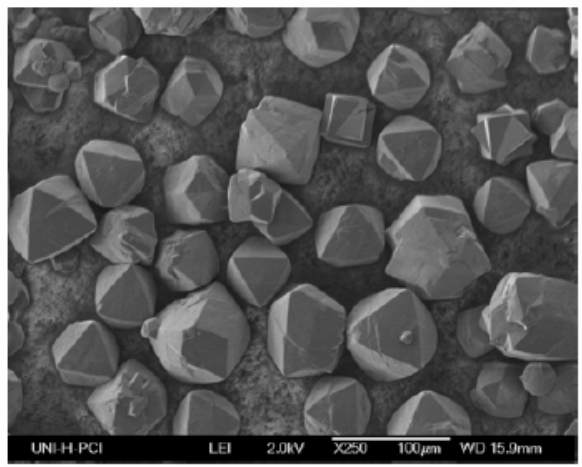
TFR

KMC (/10.)

MD

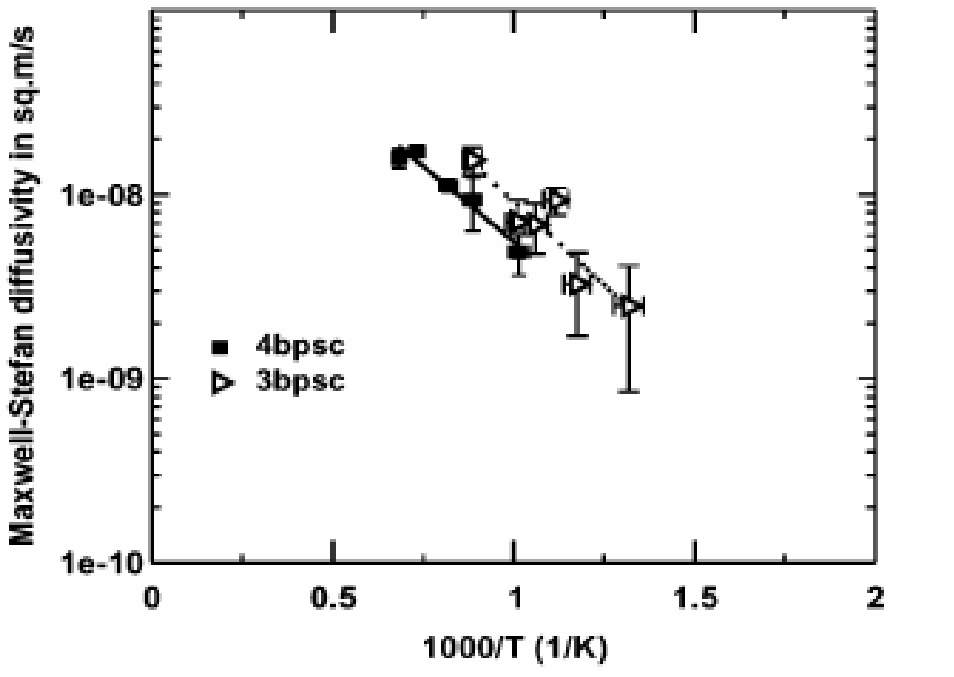
FR

TZLC



~50 µm

Do Benzene/NaX



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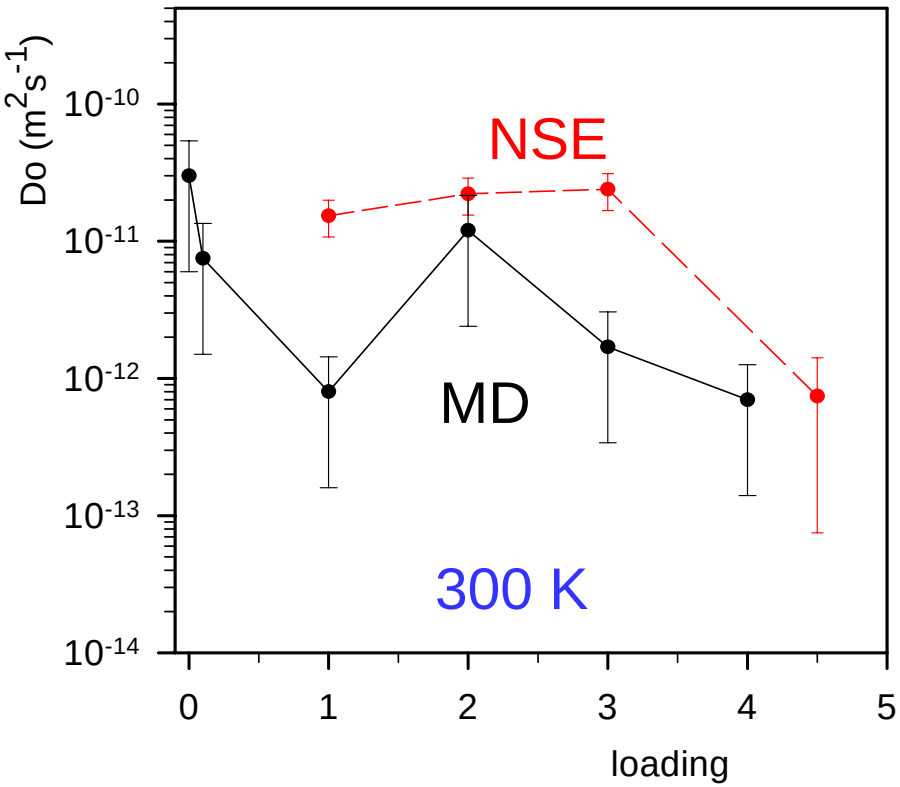

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CONCLUSIONS

Neutron scattering: structure & dynamics molecules adsorbed in zeolites.

one can work with small crystals (< μm)

Diffusion: t : 1 ps – 1 μs
d: 0.1 – 100 Å

Neutron diffusivities are insensitive to internal barriers and approach the simulations performed on ideal structures