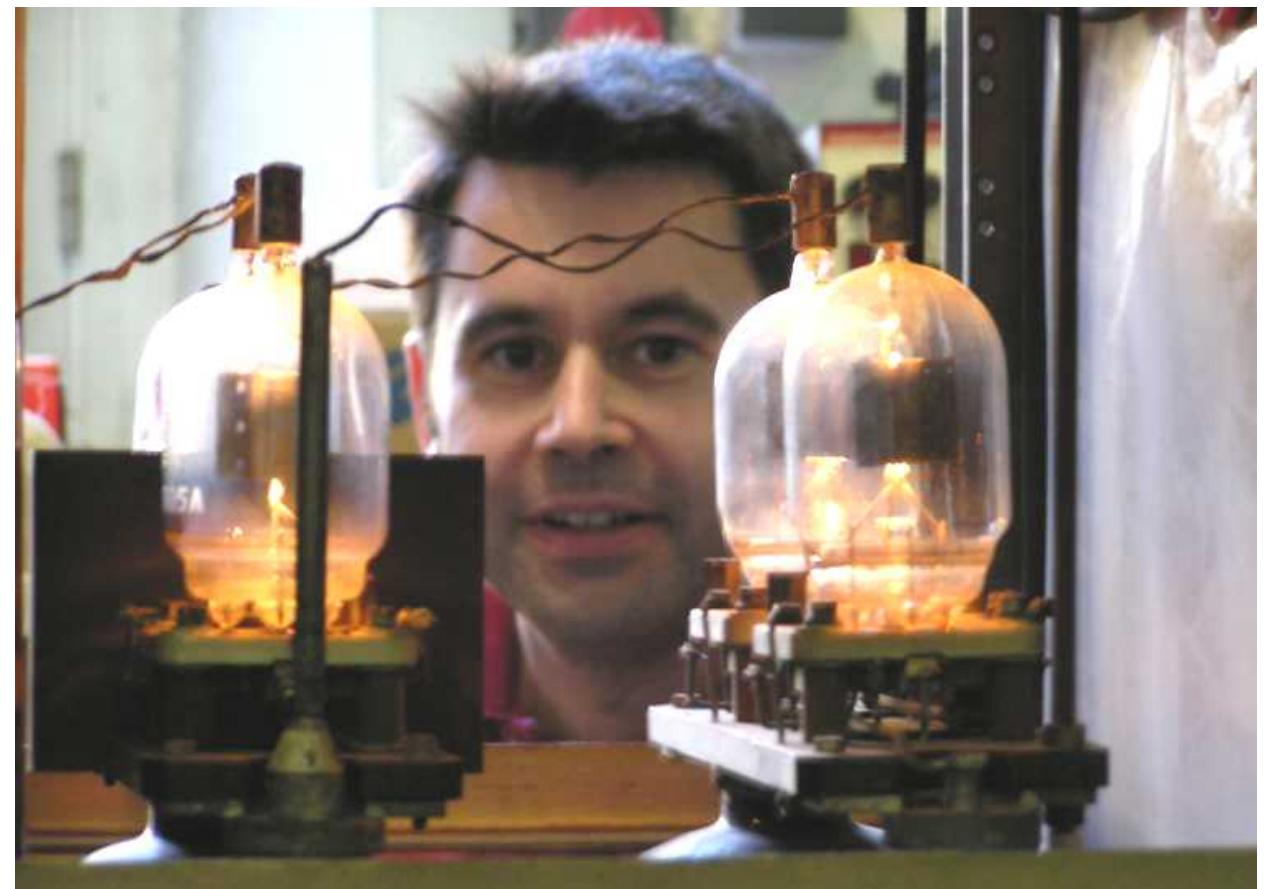


Experimental study of H₂ formation on grains overview and recent results

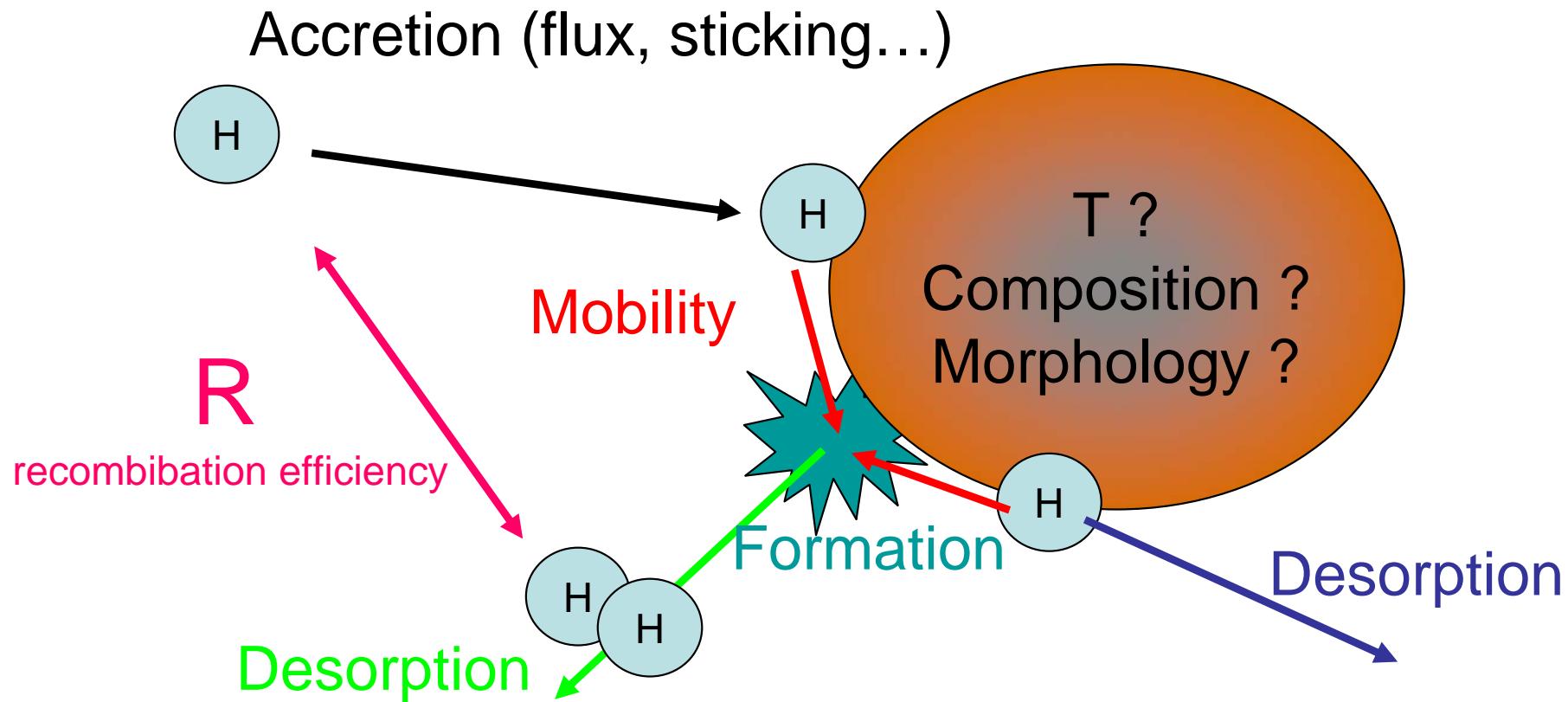
F. Dulieu

Experimental study of H₂ formation on grains overview and recent results

J. H. Fillion ! →

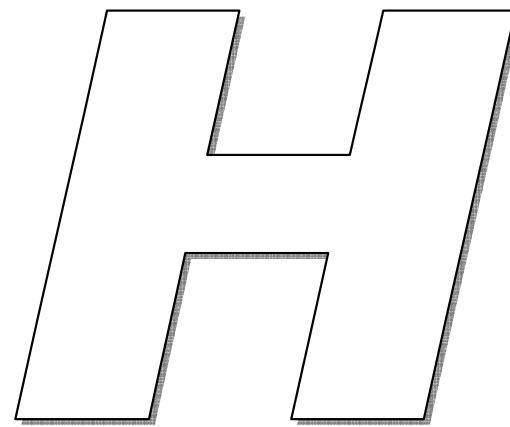


H_2 formation on grains ?



H_2 formation in ISM

Diffuse clouds

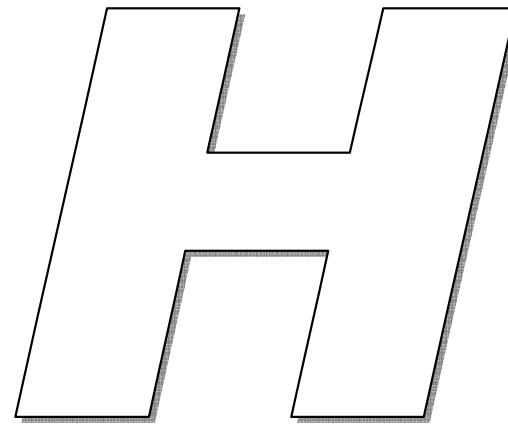


Dense clouds



H_2 formation in ISM

Diffuse clouds



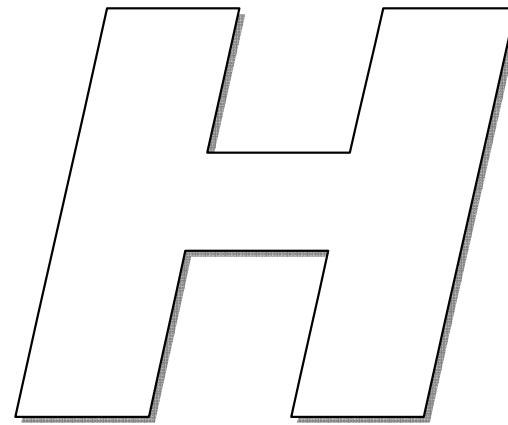
Dense clouds



- High destruction rate (UV...)
- Low destruction rate (cosmic...)

H_2 formation in ISM

Diffuse clouds



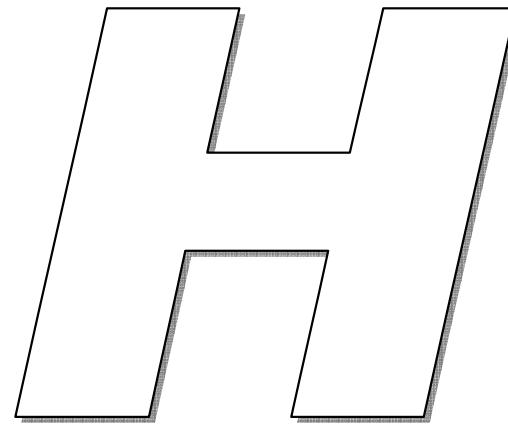
Dense clouds



- High destruction rate (UV...)
- Low destruction rate (cosmic...)
 $R \sim 0.3 !$

H_2 formation in ISM

Diffuse clouds



Dense clouds

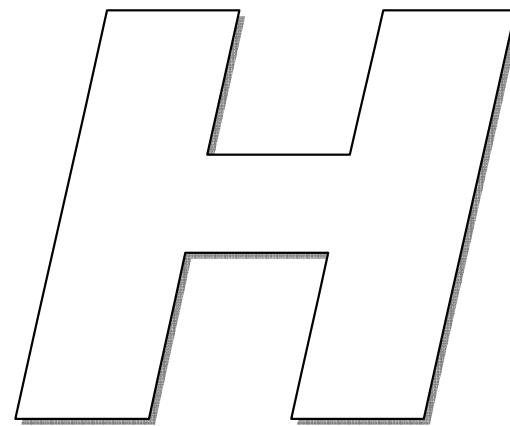


- High destruction rate (UV...)
- Bare grains
- $T > 20 \text{ K} \rightarrow 1 \text{ Chemisorbed}$

- Low destruction rate (cosmic...)
- Ice mantles (H_2O , CO...)
- $10 \text{ K} \rightarrow 2 \text{ Physisorbed atoms}$
- **Presence of H_2 adsorbed**

New experiments on H₂ formation

Diffuse clouds



Dense clouds



- UCL:
Graphite, excited molecules
Creighan et al (2006)
- U. Aarhus :
Graphite, chemisorbed sites
Hornekaer, poster 14

- UCP
Water ice surfaces
Amiaud et al (2006)

previous work on water ice surfaces

Experiments

- Govers et al (1980)
- Vidali, Pirronello, Biham et al (5 refs)
- Hornekaer et al (2003)

Calculations

- Hollenbach and Salpeter (1971)
- Buch et al (1991)
- Takahashi et al (2000)
- Al Halabi et al (2002) →

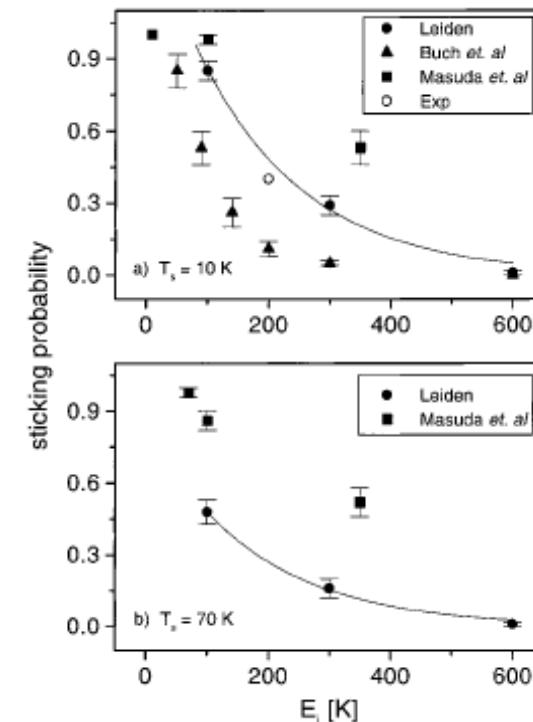
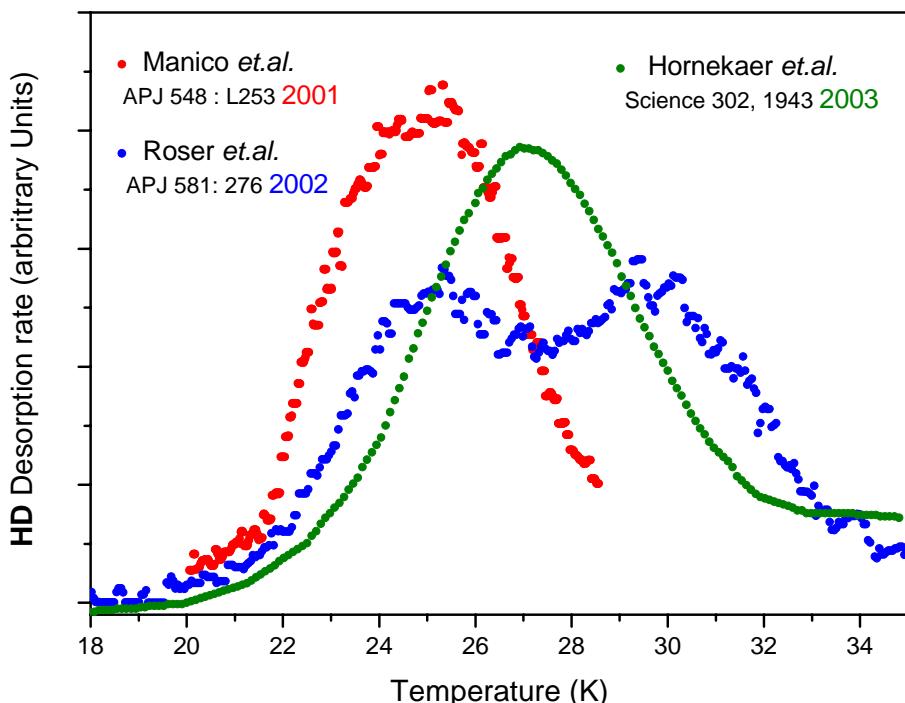
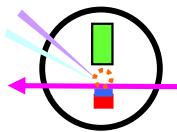


Figure 2. The sticking probability of hydrogen atoms to crystalline ice as a function of E_i for (a) $T_s = 10$ K and (b) $T_s = 70$ K, together with the previous results on sticking of hydrogen to amorphous ice.^{22,42} The solid line is an exponential decay fit of our results. An experimental data point for molecular hydrogen formation on amorphous ice is also shown at low T_s .⁴⁶ See the text for the connection of H_2 formation with the sticking of hydrogen atoms.

UCP experimental strategy

- Compare D₂ vs D+D, during and after irradiation (thermal ramp = TPD)
- Use ice as template of physisorption study (Porosity and adsorption sites can be changed)
- Characterize as much as possible different experimental parameters
- And not H+D studies (Vidali, Pirronello, Biham et al, Hornekaer et al)

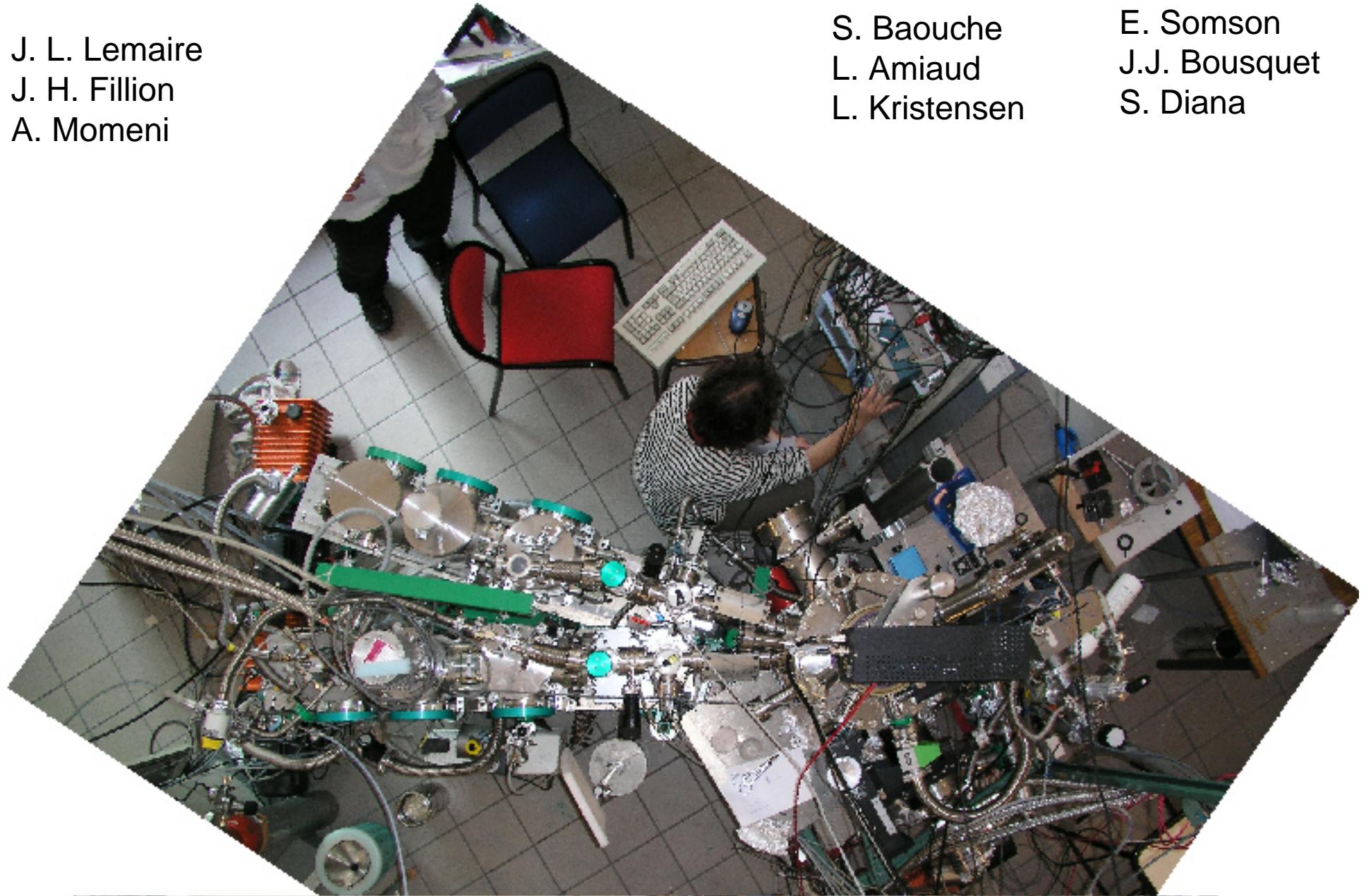


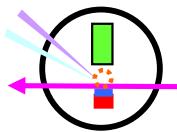
UCP

J. L. Lemaire
J. H. Fillion
A. Momeni

S. Baouche
L. Amiaud
L. Kristensen

E. Somson
J.J. Bousquet
S. Diana



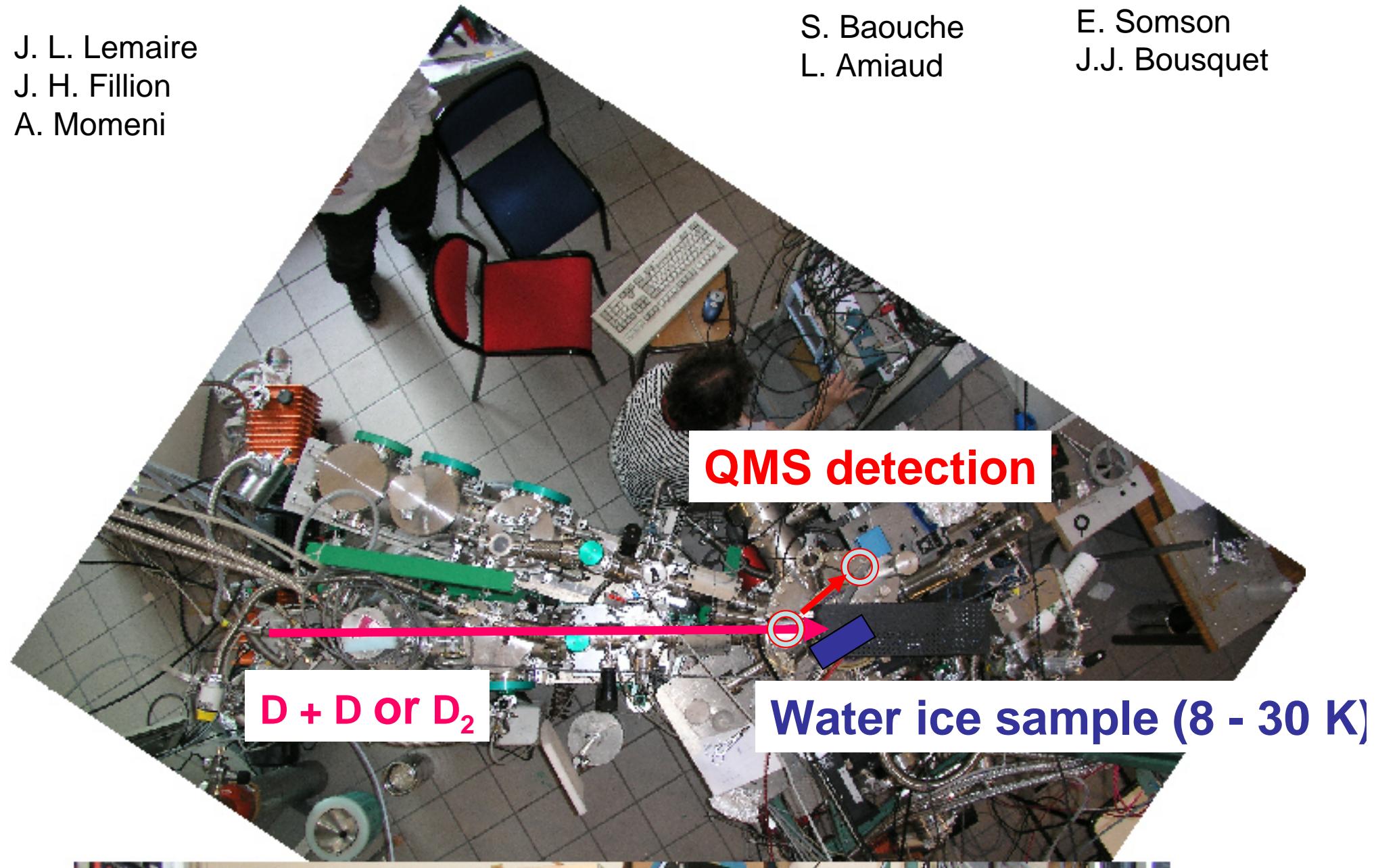


FORMOLISM

J. L. Lemaire
J. H. Fillion
A. Momeni

S. Baouche
L. Amiaud

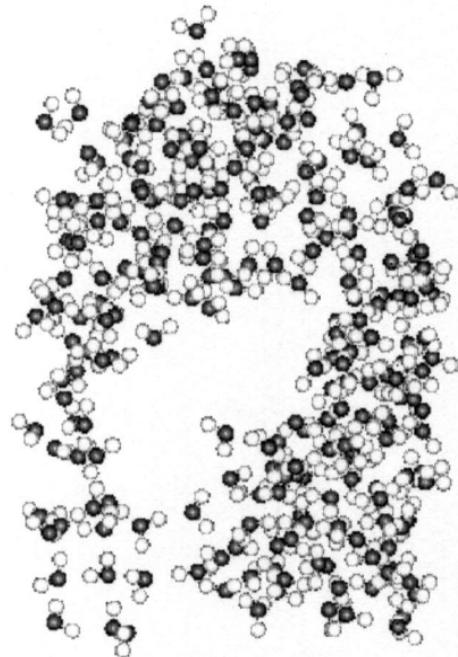
E. Somson
J.J. Bousquet





Ice samples

Growth at 10 K



© Guillot & Guissani 2004

High effective surface area
Proportionnal to thickness

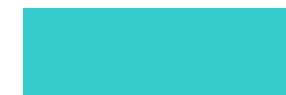
X ML

Growth at 120 K



© Kimmel et al 2001

Surface area ~ geometric area

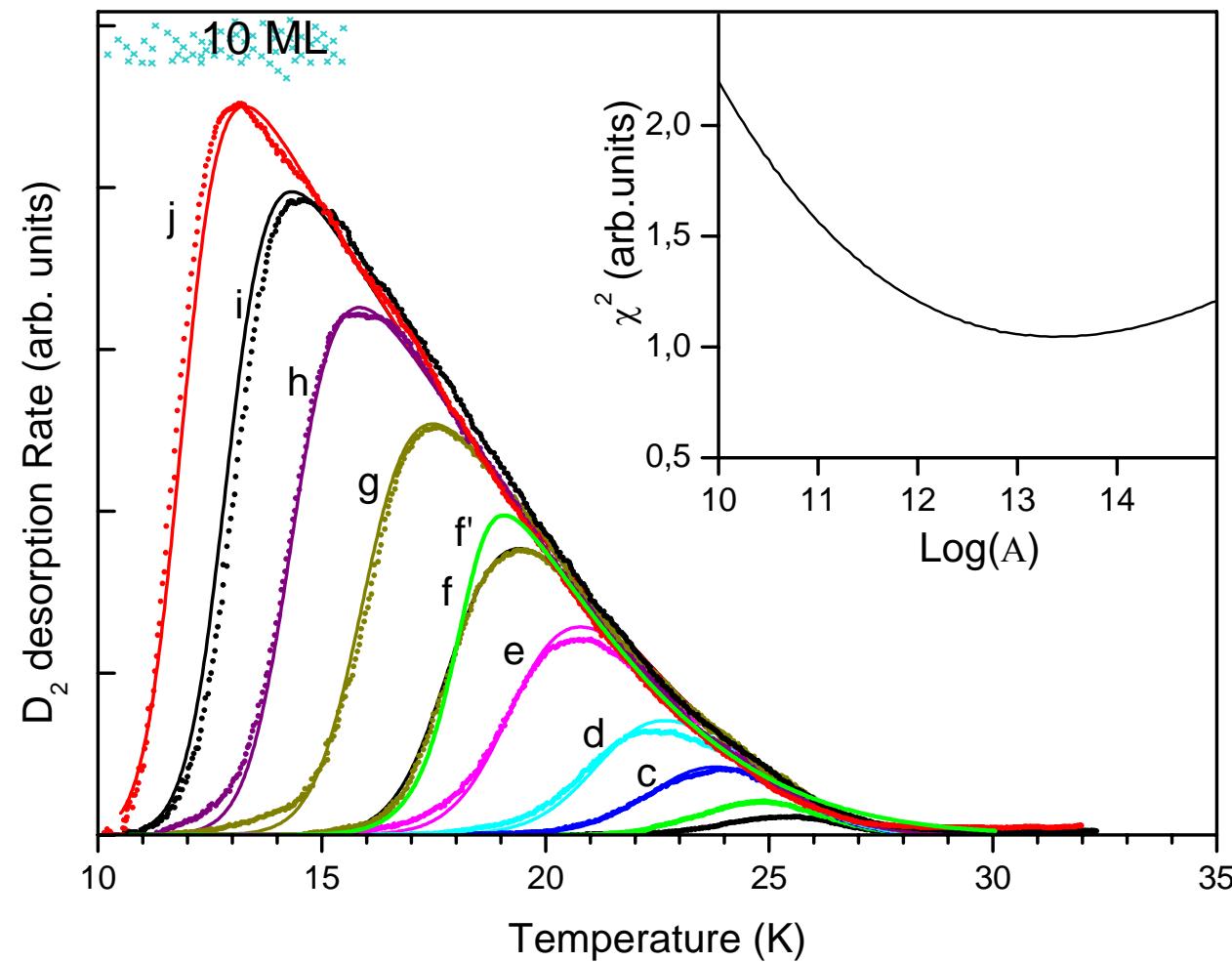


	Porous Ice	Non Porous Ice
D_2	I	II
$D + D$	IV	III

During and after irradiation !

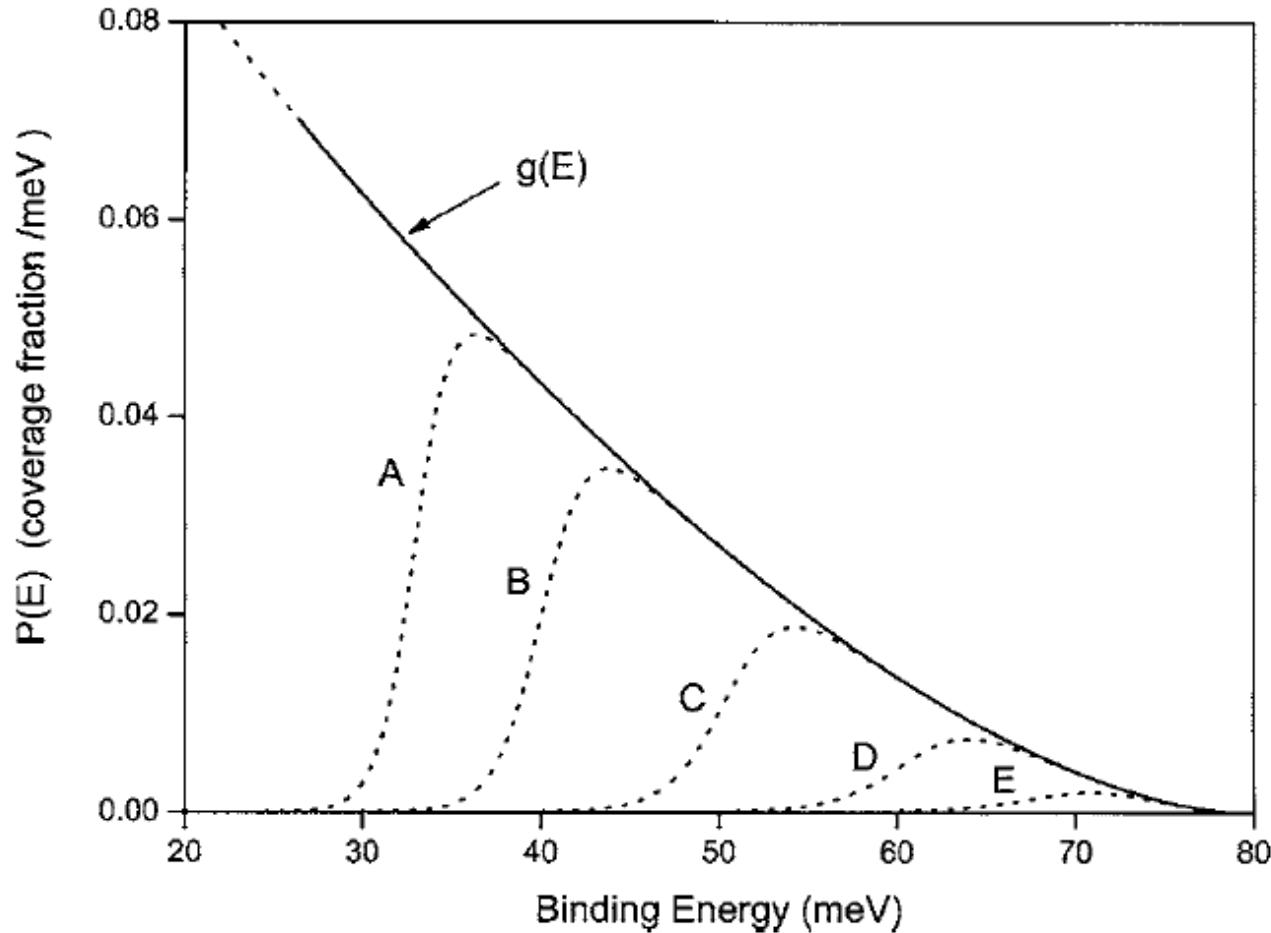
D_2 desorption from porous ASW

Figure 4



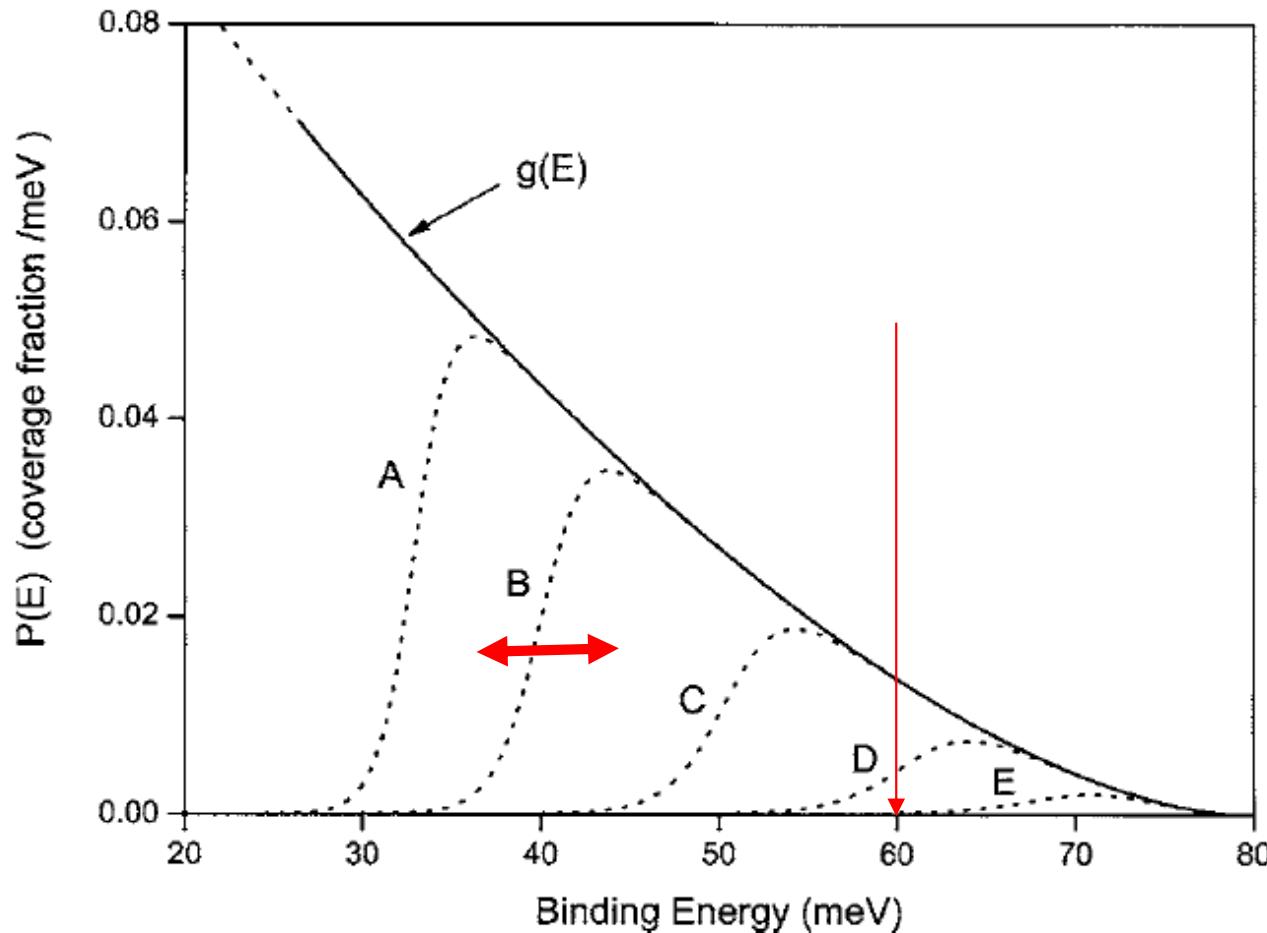
- Profiles depend on coverage
- Filling behaviour (Kimmel et al 2001)

New model → energy distribution



- Assume a distribution of adsorption sites $g(E)$
- Assume Thermal equilibrium (Fermi Dirac statistic)
- 4 fitting parameters

New model → energy distribution



- Assume a distribution of adsorption sites $g(E)$
- Assume Thermal equilibrium (Fermi Dirac statistics)
- 4 fitting parameters

D₂

Residence time on a site vs adsorption energy (E_a)

T = 10 K				
E _a (meV)	30	35	40	60
t _{res} (day)	0.01	4.8	1567	> 10 ¹³

$$P_{des} = v_o e^{-E_a/kT}$$
$$t_{res} = \frac{1}{P_{des}} = \frac{1}{v_o} e^{E_a/kT}$$

Residence time on a site vs adsorption energy (E_a)

		T = 10 K		
E_a (meV)	30	35	40	60
t_{res} (day)	0.01	4.8	1567	$> 10^{13}$

$$p_{des} = A e^{-E_a/kT} \quad t_{res} = \frac{1}{p_{des}} = \frac{1}{A} e^{E_a/kT}$$

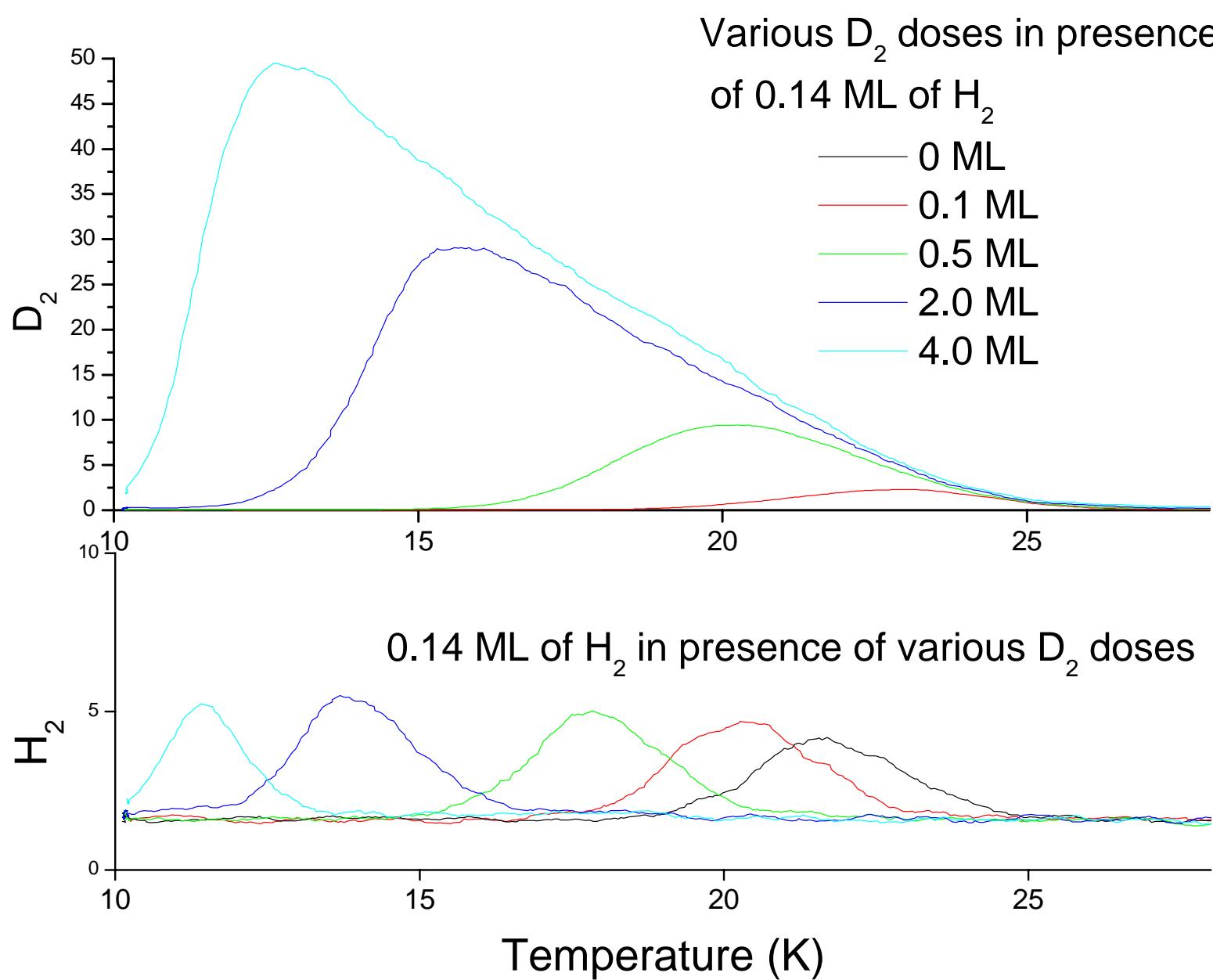


There is a non negligible amount of H₂ adsorbed on ice mantles

D_2

10 ML

Isotopic segregation effect



D_2

10 ML

Isotopic segregation effect

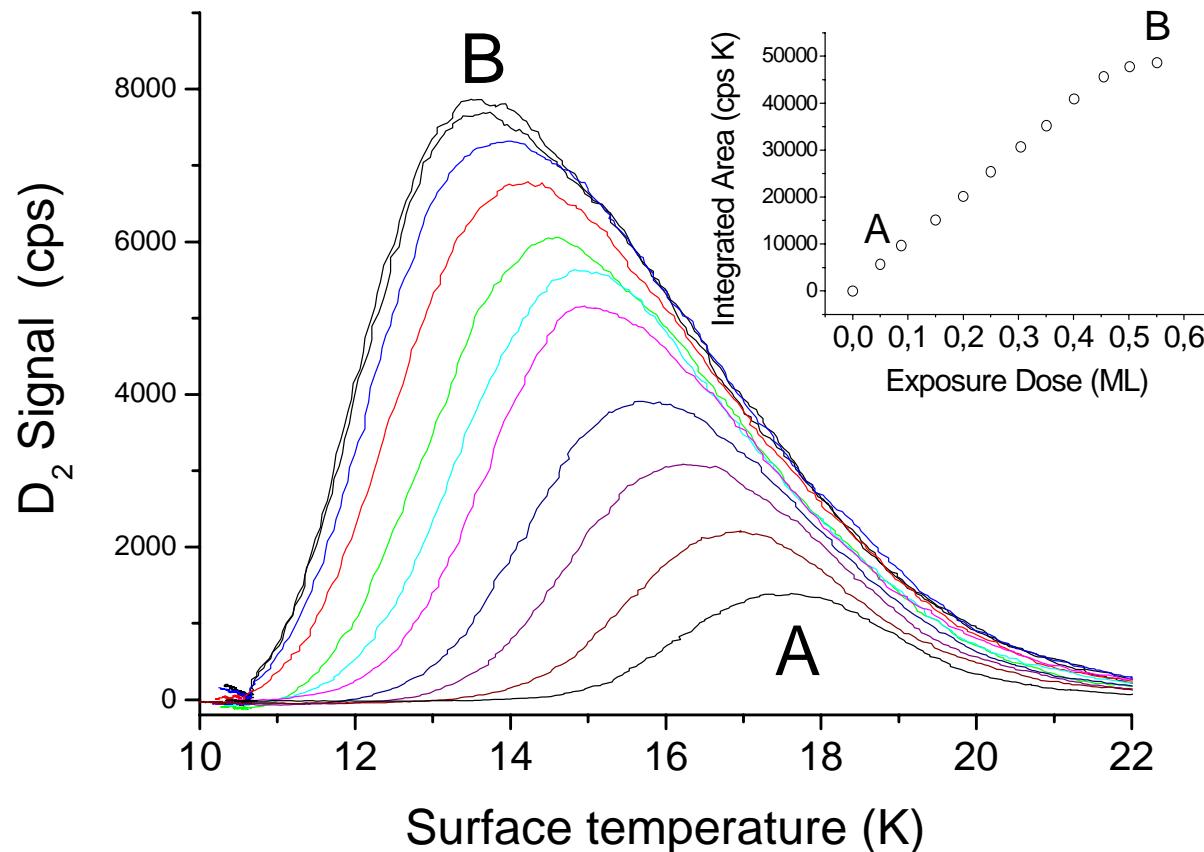
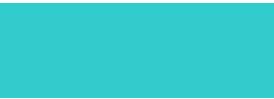
- Enrichment of deuterated molecules on ice mantles (see poster 18)
- TPD profiles of H_2 , HD and D (see below) are very sensitive to the presence D_2 (even 10%)

Summary I

D_2 interaction with porous ASW

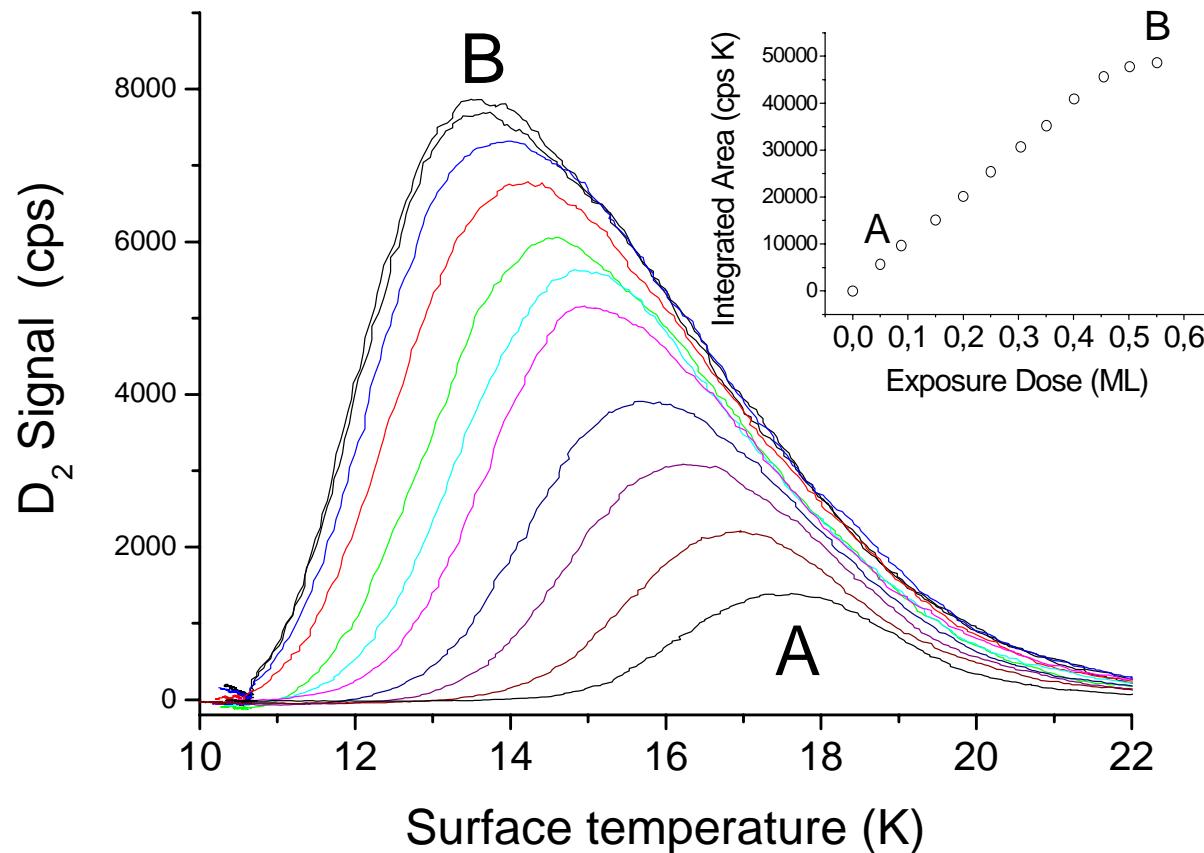
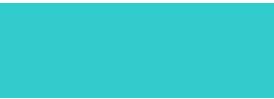
- Interaction is coverage dependant
- There is a very large energy distribution (in agreement with calculations by Hixson et al 1992)
- There is a strong isotopic effect
- We have modeled it → tool for ice characterization
- Our model permit us to study the effect of ice thickness, non uniform ice layer and non uniform deposition of D_2 , that dramatically affect the shapes of the TPD curves

D_2 desorption from Non porous ASW



Similar behaviour than porous ASW,
not the same energy distribution

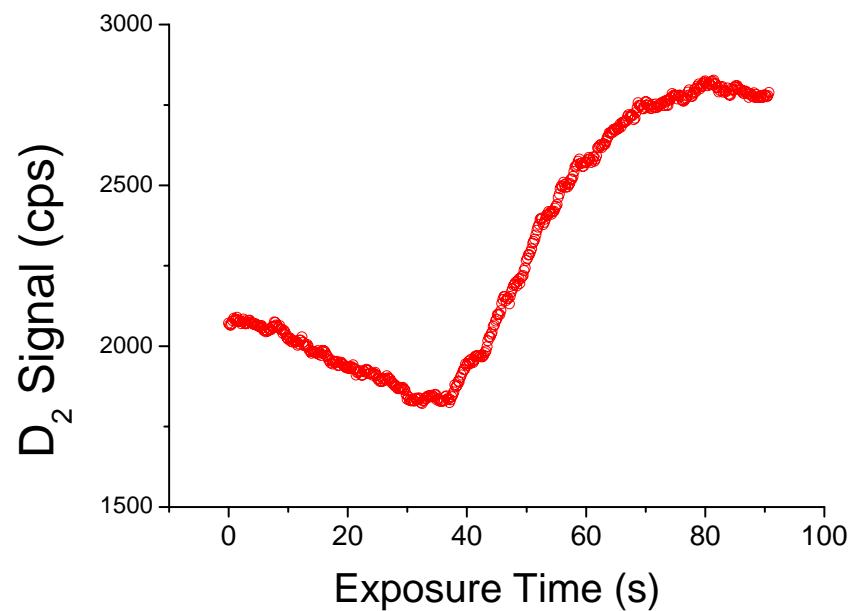
D_2 desorption from Non porous ASW



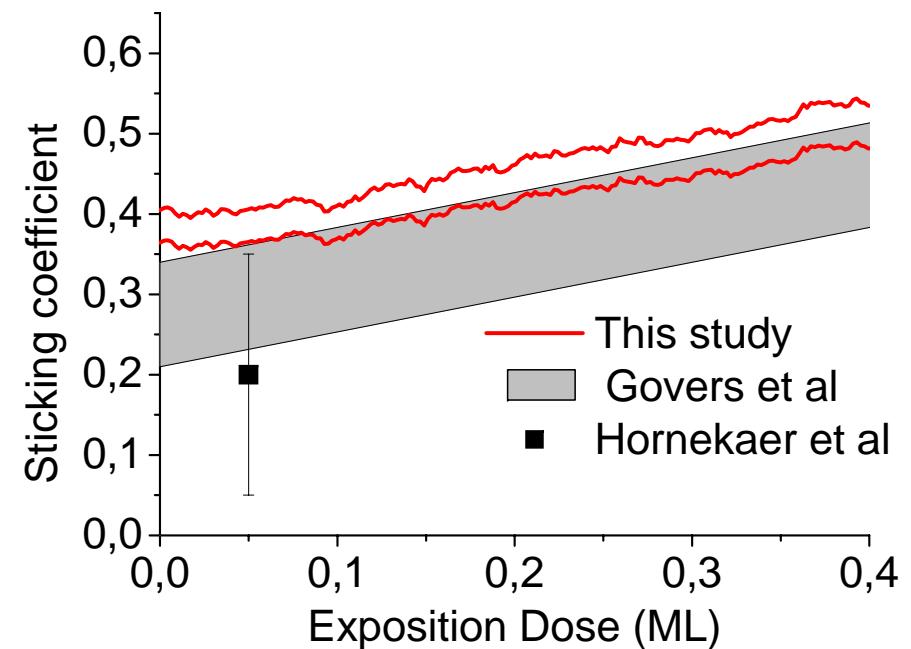
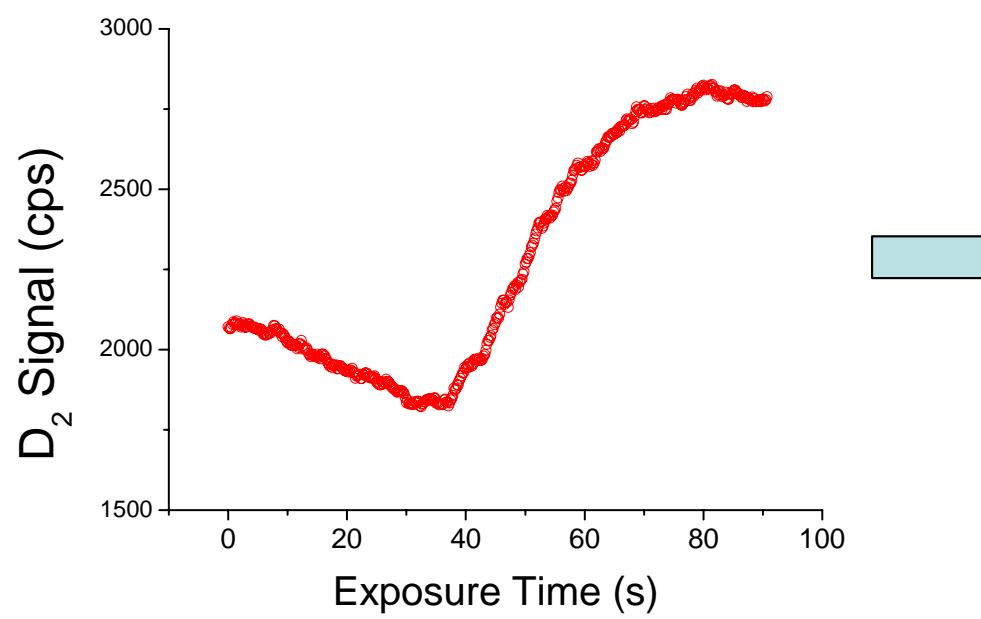
Similar behaviour than porous ASW,
not the same energy distribution



D_2 during irradiation



D_2 during irradiation

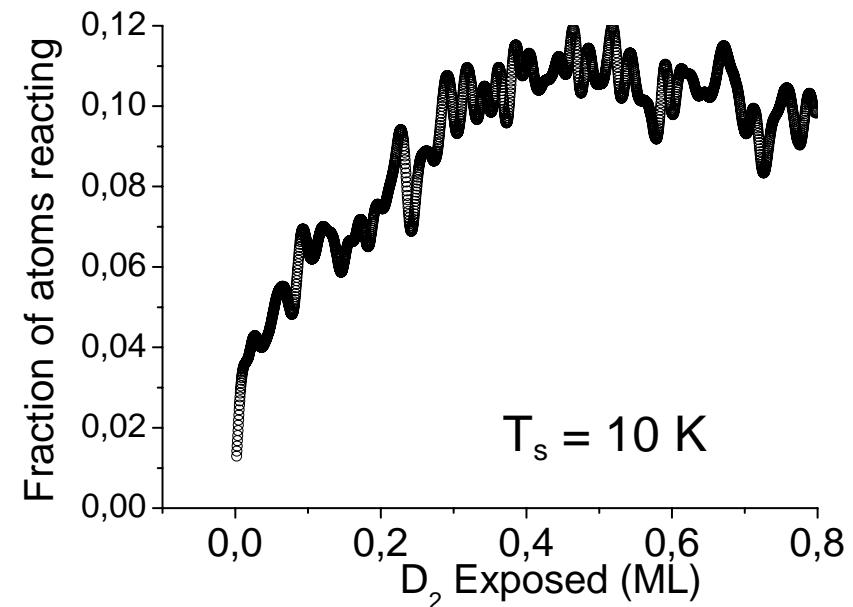
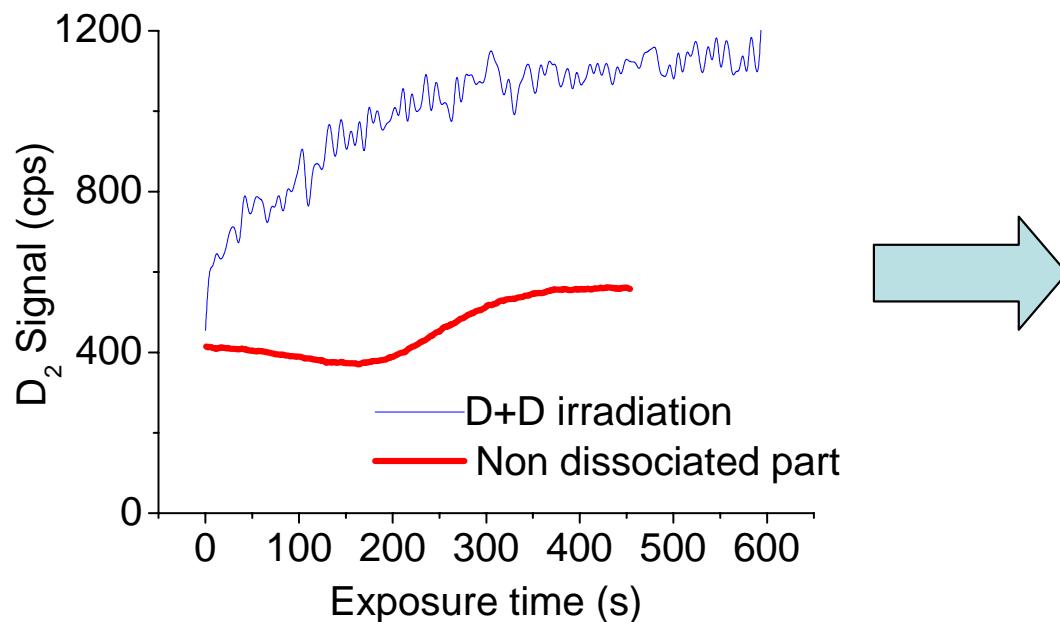


D_2 accommodates more efficiently when some D_2 is already adsorbed

Summary II: D_2 interaction with non porous ice

- Follow the « filling behaviour »
- Distribution of adsorption energy is significantly shifted to lower binding energies
- Sticking coefficient is dependant on the presence of already adsorbed molecules

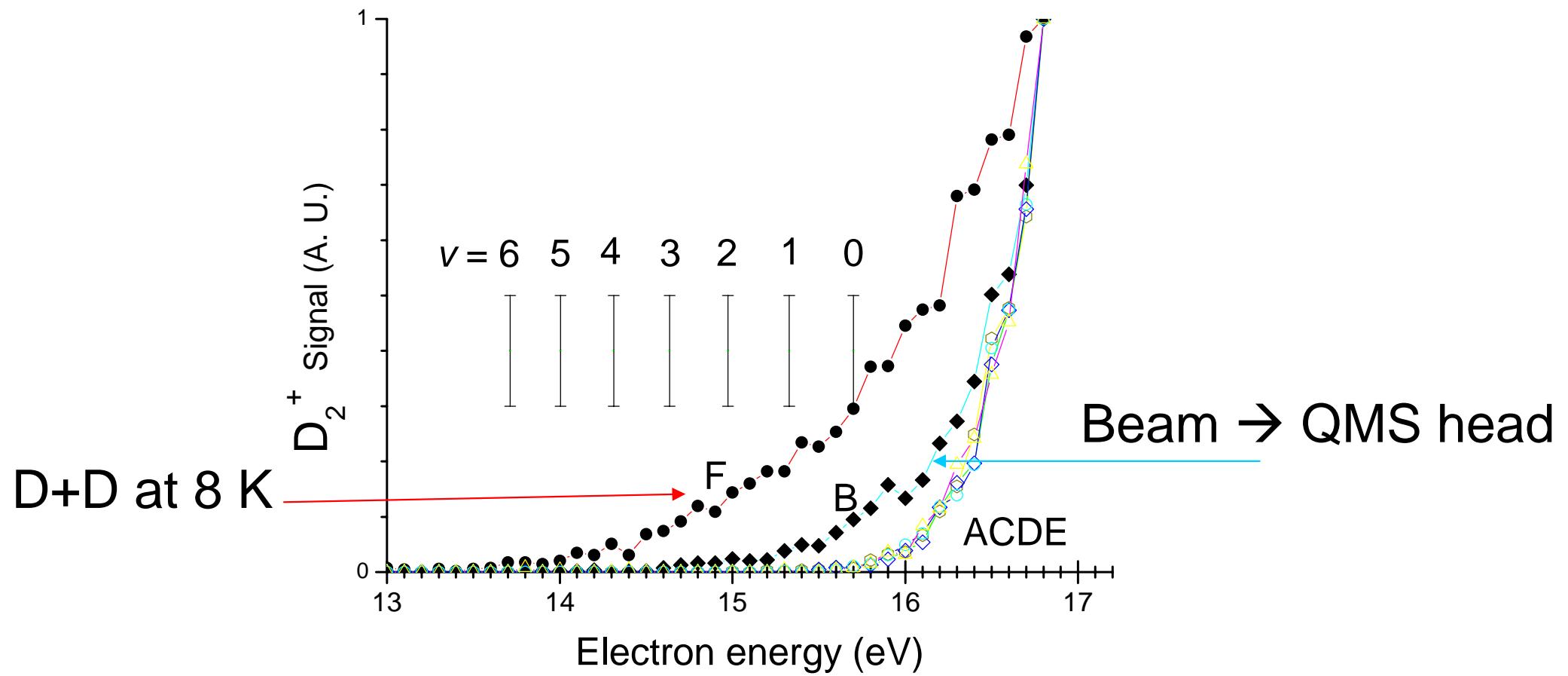
D + D during irradiation



- Prompt reaction occurs
- Recombination efficiency depends on sticking that is enhanced by adsorbed molecules
- $E_a(D) \sim 0.6 E_a(D_2)$ (as expected from polarisability)

D + D

Excited molecules



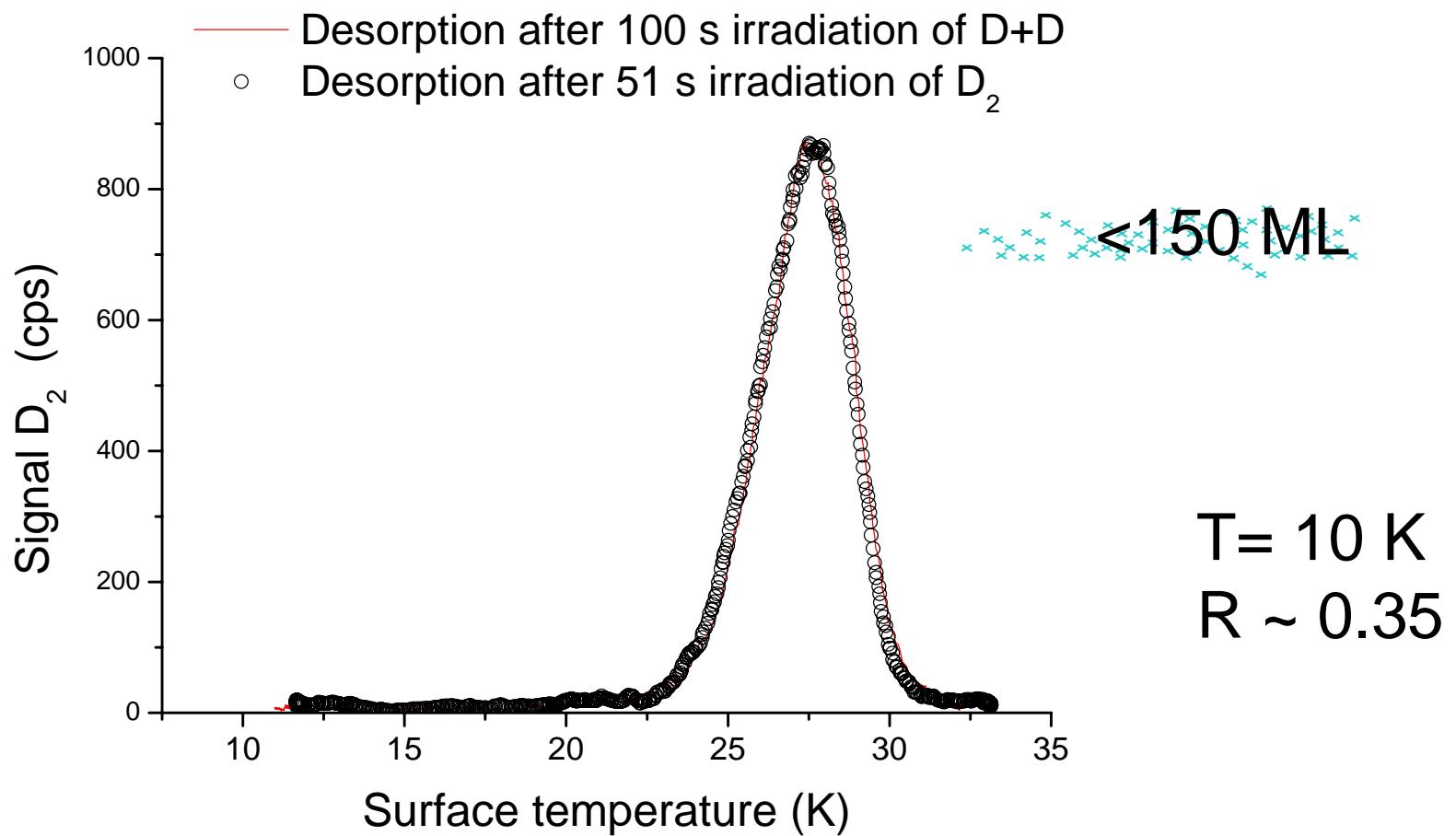
Up to $v = 4$ is detected

Summary III

D₂ formation on Non porous ice

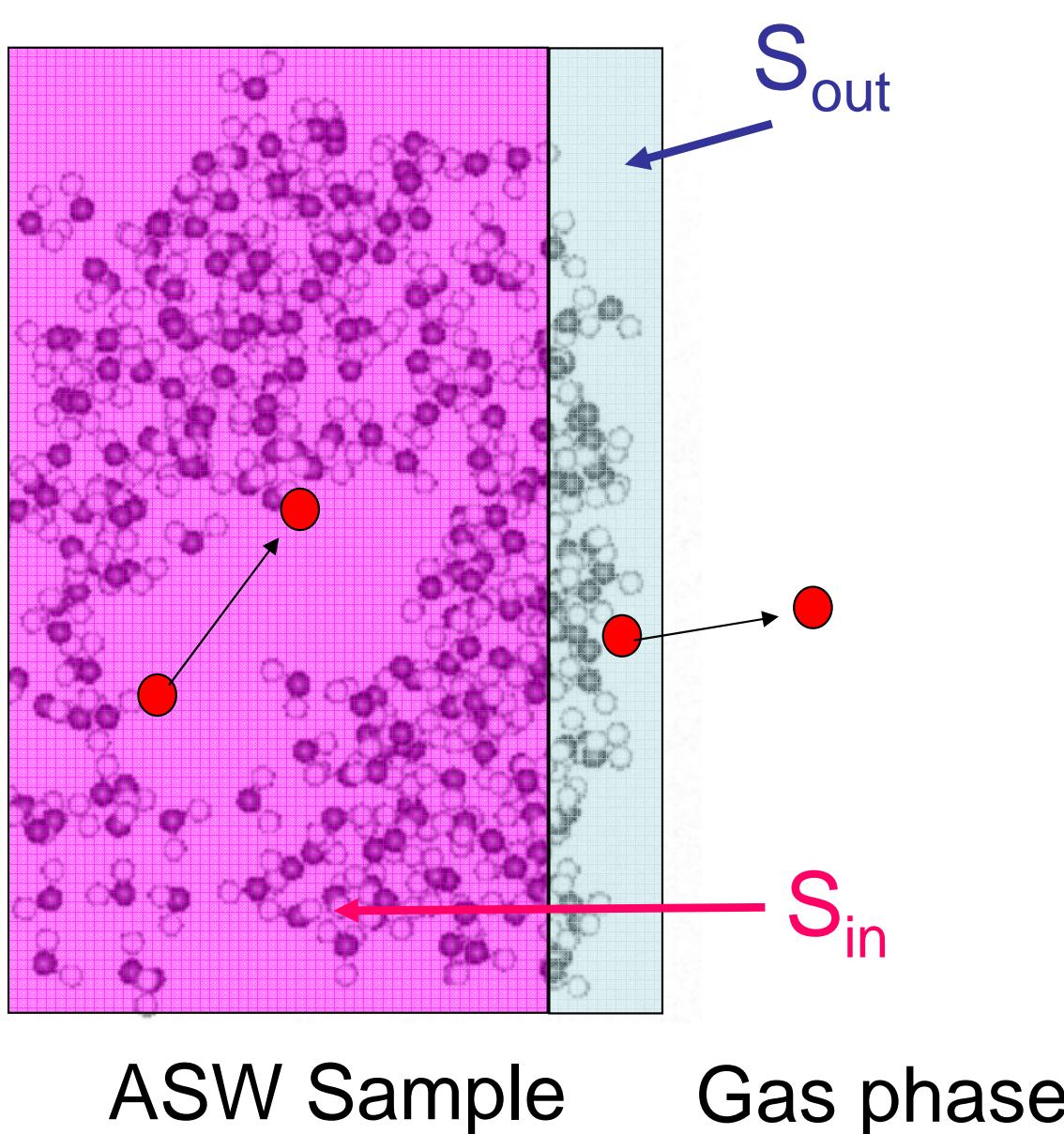
- Prompt reaction that leads to excited molecules at 10 K
- Ea(D₂) > Ea(D) > Ediff(D) in agreement with calculations
- Molecules already adsorbed enhance the recombination efficiency (see also Govers et al)

Formation on porous water ice surface



- TPD profiles provide no information about formation
- No track of formation between 10 K and 20 K
(in case of porous surfaces !)

Effect of porous structure



$$P_{des} = \frac{S_{out}}{S_{out} + S_{in}} v_0 e^{-E_a/kT}$$

Has been verified for molecules
(see also Hornekaer et al 2005)

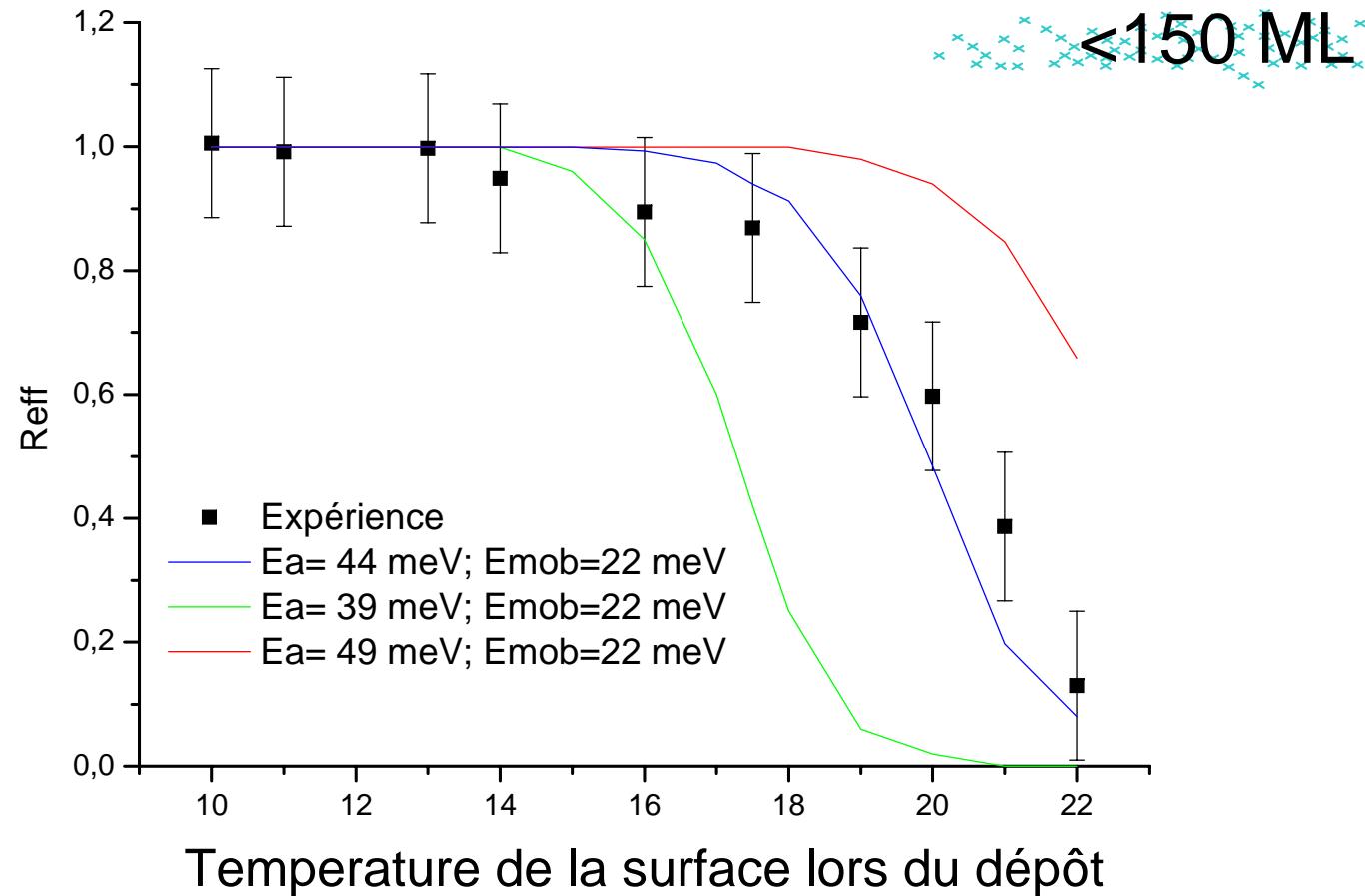
$$Ea(D2) > Ea(D) > Ediff(D)$$

Molecules are mobile enough to enter
the porous network
They react and are recaptured by the
porous structure

It happens during the irradiation

D+D

R vs T during D+D irradiation



Model: Katz et al, $\mu = 100\%$, $A = 10^{13} / \text{s}$
→ $\text{Emob} = 22 \text{ meV}$; 3 ms between 2 hops at 10 K.

but solution is not unique

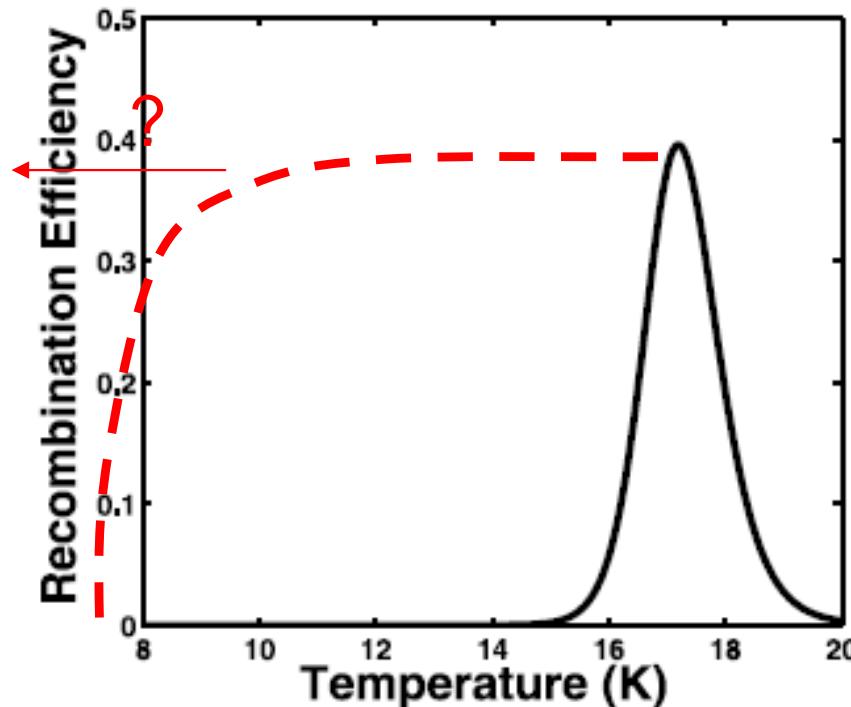
Outline of D+D experiments on porous amorphous ice

- Kinetics of formation is blurred by porous structure of the ice, TPDs reveal only molecular desorption
- $Ea(D_2) > Ea(D) > Ediff(D)$
- at 10 K diffusion is fast enough to allow atoms to enter the porous network and to react but the molecules formed are recaptured by the porous network
- The recombination efficiency is 0.35 at 10 K and is mostly governed by the sticking probability

Question to elucidate:

- What about the effect of adsorbed molecules ?
- Diffusion energy, Adsorption energy should have a distribution (see Buch et al 1991) need another approach

Astrophysical relevance ?



© Perrets et al 2005

FIG. 10.—Recombination efficiency of molecular hydrogen at steady state on HDI vs. the temperature T (K), using the parameters obtained from experimental measurements of HD desorption curves. The flux is the same as in Fig. 8.

H chemistry is effective at 10 K
Presence of H_2 (and isotopes) could play a role

Future work

- IR characterization of H₂ / H interaction with ASW
- Energetic balance (REMPI + imaging)
- Interaction with other samples (carbonaceous)
- Formation of water

Thank you !