Laboratoire d'Étude du Rayonnement et de la Matière en Astrophysique

Gas temperature dependence sticking coefficient of hydrogen on amorphous water ice and silicate surfaces of interstellar interest

Henda CHAABOUNI

François Dulieu, Saoud Baouche, Elie Matar,
Emanuele Congiu, Lisseth Gavilan, and Jean-Louis Lemaire

LERMA-LAMAp, UMR 8112 du CNRS , Observatoire de Paris et Université de Cergy Pontoise
95031 Cergy-Pontoise, France

Hervé Bergeron

Université Paris-Sud, ISMO, UMR 8214 du CNRS, Bat. 351, 91405 Orsay, France

GDR–ARCHES Adsorption, Reactivité et Contrôle de l'Hydrogène En Interaction avec des Surfaces
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Outline of the talk

- **Introduction**

  Hydrogen in the ISM and Sticking process

- **Experimental setup and procedures**

  Sticking coefficient measurements

- **Results (Experiments + Model)**

  Sticking coefficient of D_2 and H_2 molecules

  Sticking coefficients of D and H atoms

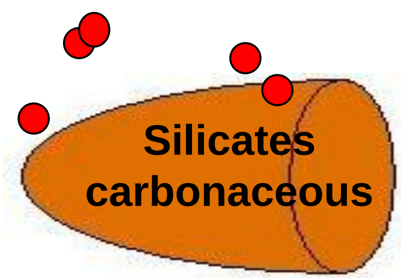
- **Conclusions**

Hydrogen in the ISM

ISM
- Gas (99% by mass) →
  - **hydrogen (75%)**
  - helium (20%)
  - and other elements (5%)
- Dust (1% by mass)

Diffuse clouds

$H \gg H_2$



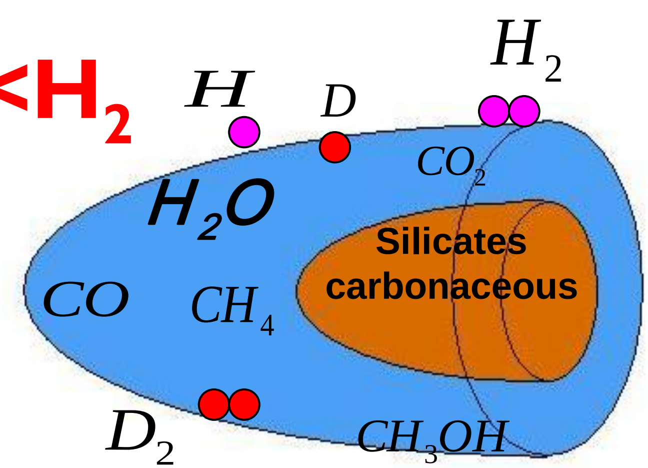
Dust : Silicates & or carbonacous materials

Atomic gas: density ~ 50 cm^{-3}

T_{Gas} = 50 –100 K, T_{grain} ~ 10-20 K

Dense clouds

$H \ll H_2$

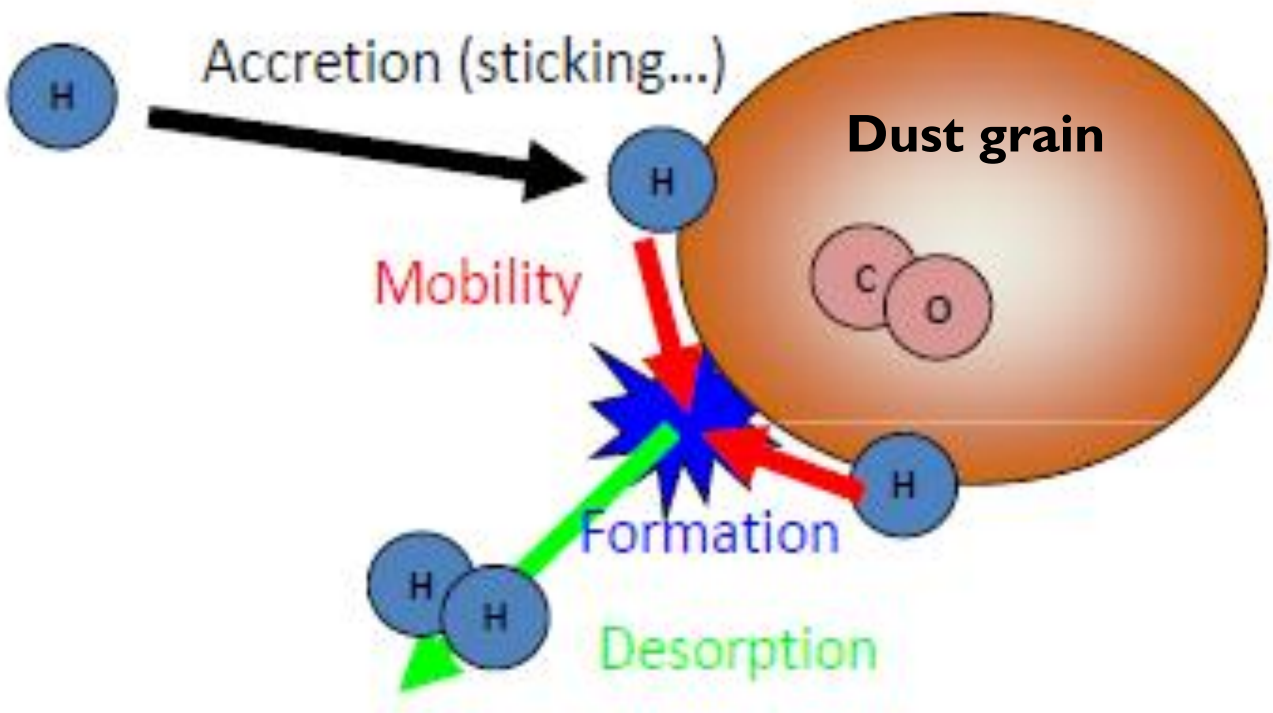


Dust + icy mantles (**amorphous H_2O**)

Molecular gas: density ~ $10^2 – 10^6$ cm^{-3}

T_{Gas} ~ T_{grain} = 10 K

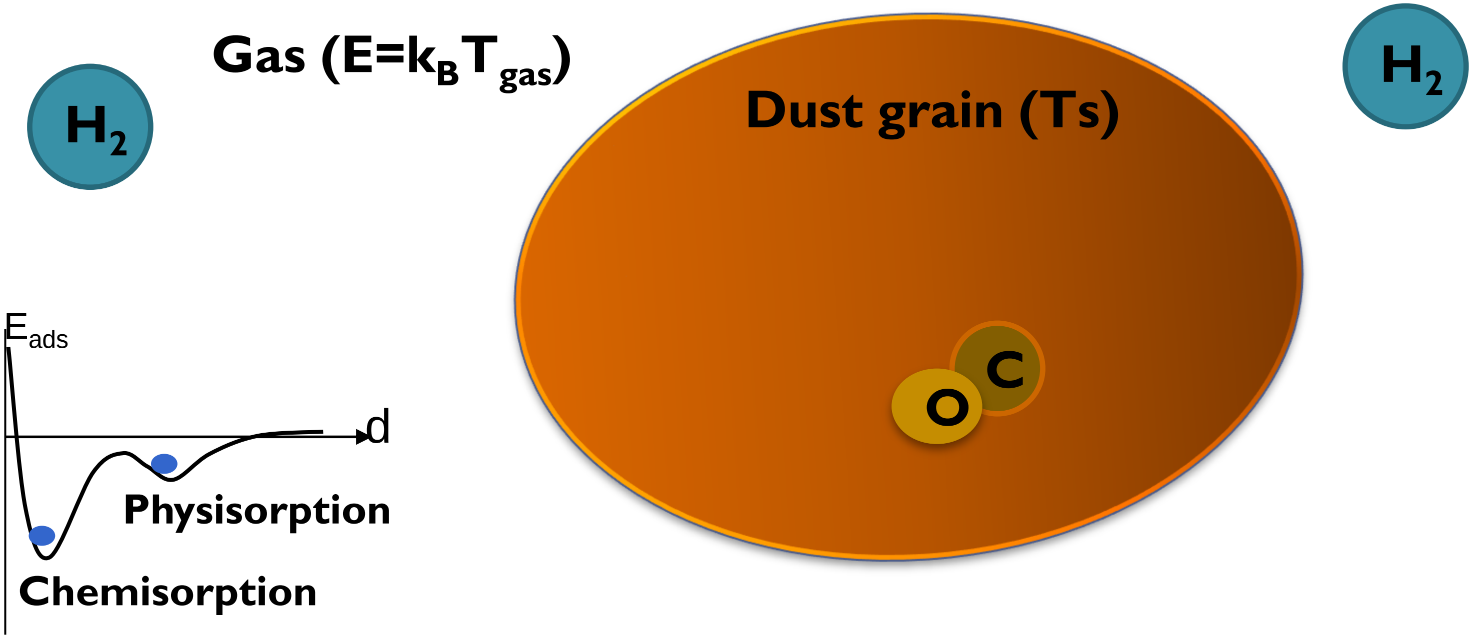
Gas-surface reactions



Processes of the surface chemistry on the dust grains:

- **Sticking**
- **Diffusion (Mobility)**
- **Formation**
- **Desorption**

Sticking Process



- The sticking coefficient S is the probability for a particle (atom or molecule) coming from the gas phase to thermally equilibrate with the grain.

- The sticking coefficient of light particles depends primarily on the gas temperature and less on the grain temperature.

Theoretical studies

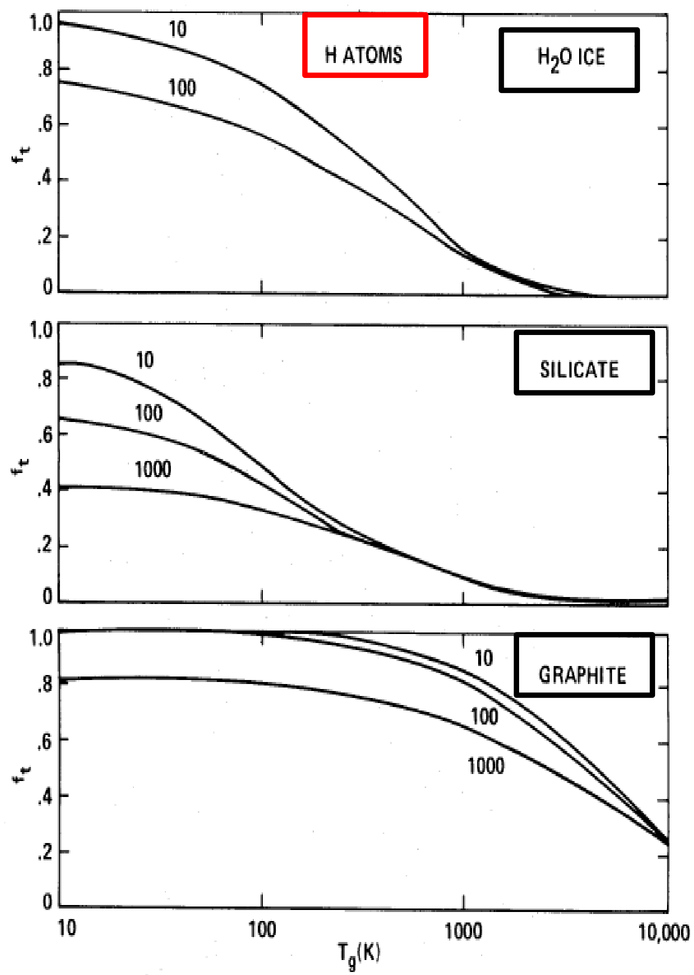
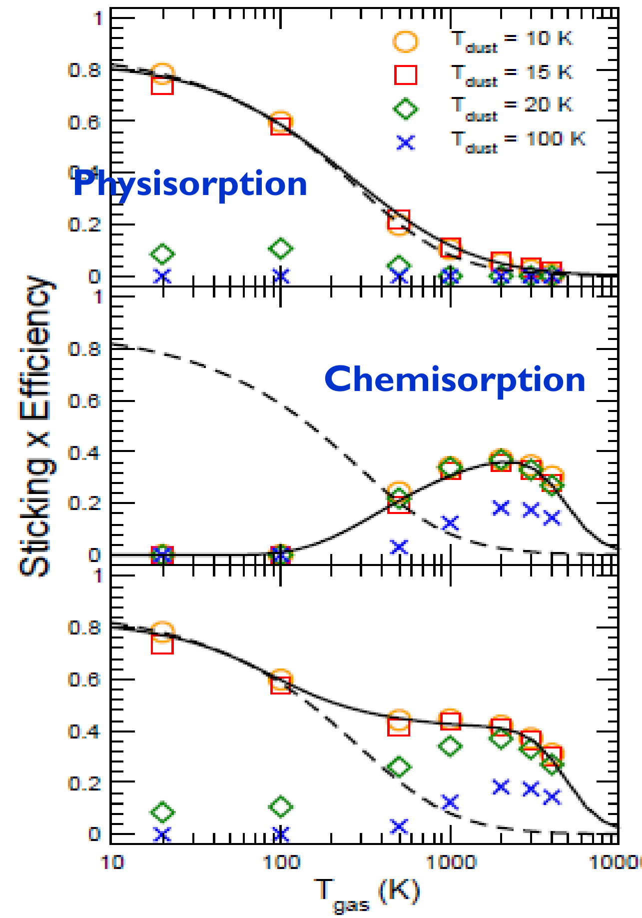


FIG. 5.—Trapping fraction f_t for atomic hydrogen incident on ice, silicate, and graphite surfaces vs. gas temperature. The curves are labeled by the surface temperature in kelvins.

(Burke & Hollenbach. 1983)

Graphite (Post choc regions)



Sticking probability of **H atoms** on graphite surface in post shock regions

(Cuppen et al. 2010)

Theoretical studies

Hexagonal Water Ice

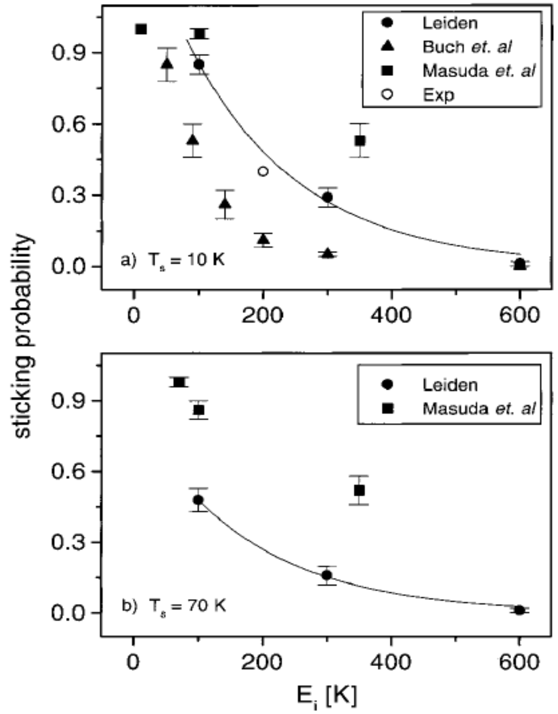


Figure 2. The sticking probability of hydrogen atoms to crystalline ice as a function of E_i for (a) T_s = 10 K and (b) T_s = 70 K, together with the previous results on sticking of hydrogen to amorphous ice.[22,42] The solid line is an exponential decay fit of our results. An experimental data point 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.

Sticking probability of **H atoms** on hexagonal crystalline water ice using **Classical Ttrajectory Calculations CT**

(Al-Halabi et al, 2002)

Clusters of Amorphous Water Ice at 10 K

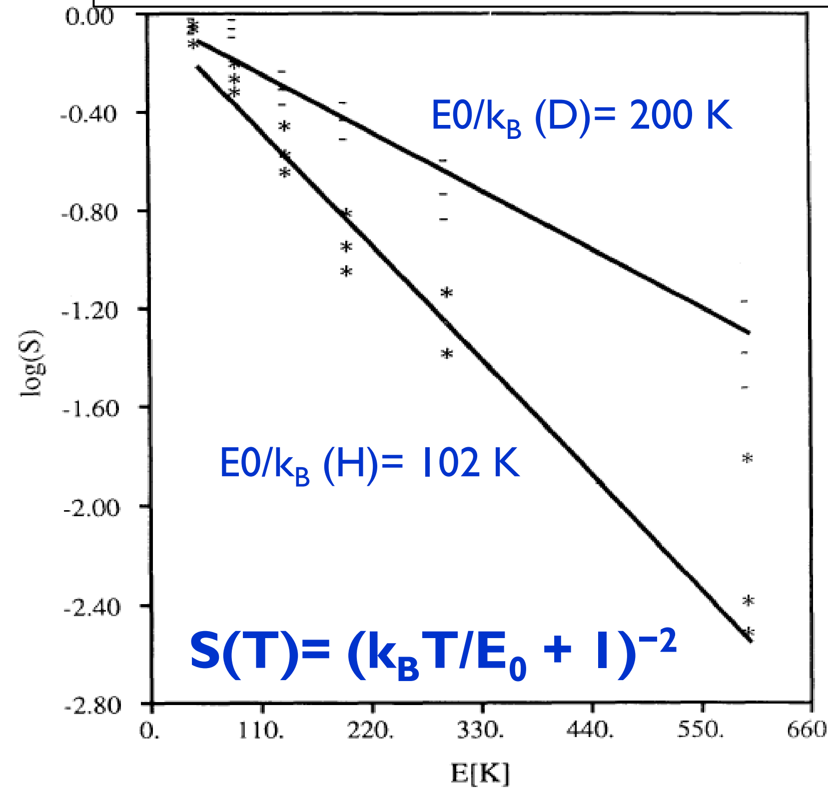


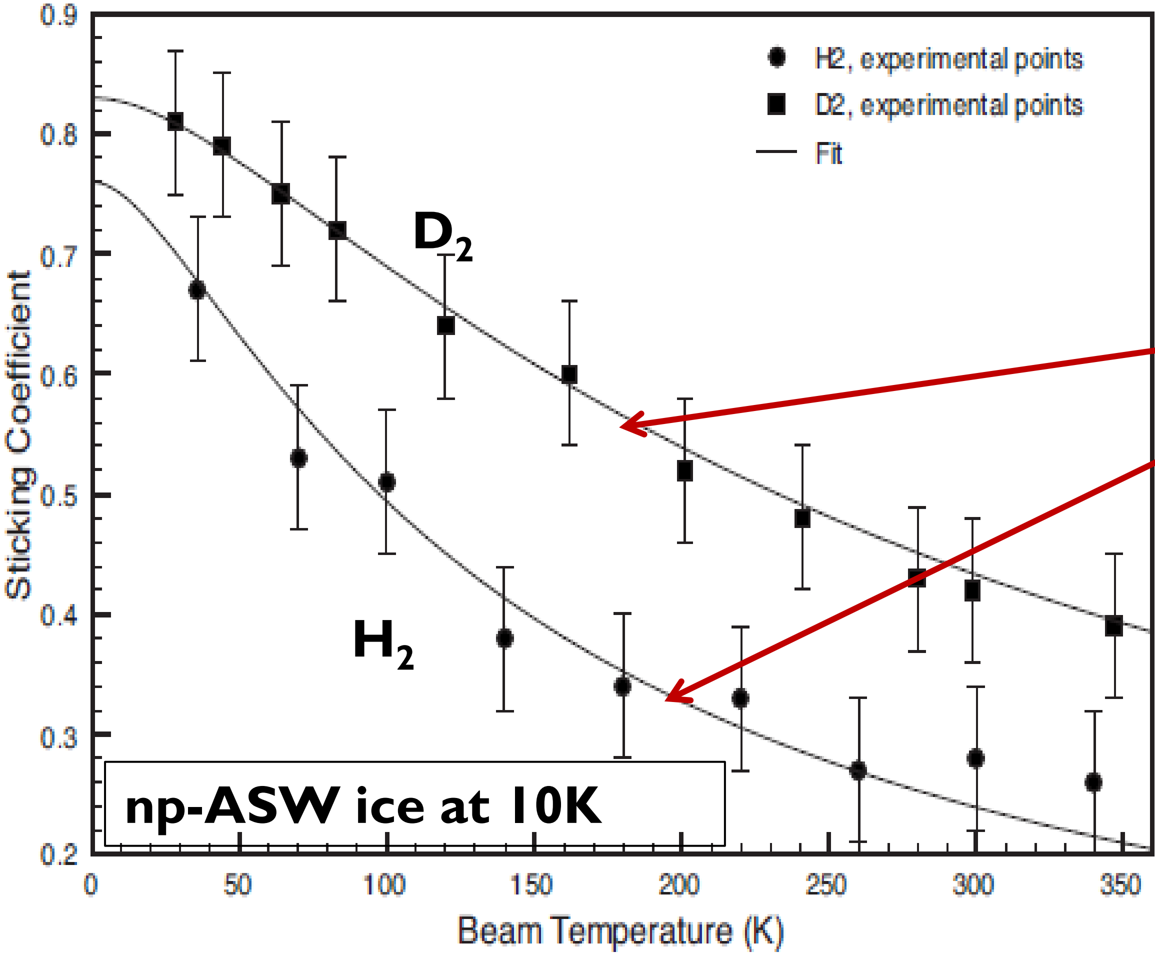
FIG. 3.—The sticking probability S_E, and the exponential fits eq. (4). *Top line:* fit for the D atom; bottom line: fit for the H atom. *Dashes*: numerical values for D; asterisks: numerical values for H. At each E the three numerical values correspond to S_{min}, S_E, and S_{max}, using 70% confidence level.

Sticking efficiency of **H and D atoms** on clusters of amorphous water ice using **Molecular Dynamical Simulations MD**

(Buch and Zhang, 1991)

(Experimental + theoretical) studies

Experiments: Sticking coefficient of H_2 and D_2 on **non porous-ASW ice** prepared at 120 K and cooled down to 10 K as a function of the gas temperature



A **physical Model** is developped to fit the experimental data

$$S(T) = S_0 \frac{\left(1 + \frac{\beta T}{T_0}\right)}{\left(1 + \frac{T}{T_0}\right)^{\beta}}$$

(Matar et al . 2010, JCP)

The Model

H. Bergeron , ISMO, Université Paris Sud Orsay *(Matar et al. 2010)*

Assumptions

- **Amorphous structure of the surface**: a sum of independent cells sufficiently small
- **Cell-dependent critical velocity *c(m, Cell)***

  v (particle) < c(m, Cell) inelastic collision ⇒ particle stuck

  v (particle) > c(m, Cell) particle rebounds in the gas phase

- **A probability law specifies the distribution of cell critical velocities**

Thermal sticking coefficient of a gas at temperature T

$$S(T) = S_0 \frac{\left(1 + \frac{\beta T}{T_0}\right)}{\left(1 + \frac{T}{T_0}\right)^{\beta}}$$

T_0, S_0 : sticking parameters of hydrogen on grain surfaces

S_0: sticking coefficient of particles at zero temperature

T_0 verifies $k_B T_0 = 1/2\, m c_0^2$, m (mass of particle)

β parameter reflects the geometry of the incident beam

β=2.22 (effusive beam) and β=2.5 (isotropic distribution of velocity)

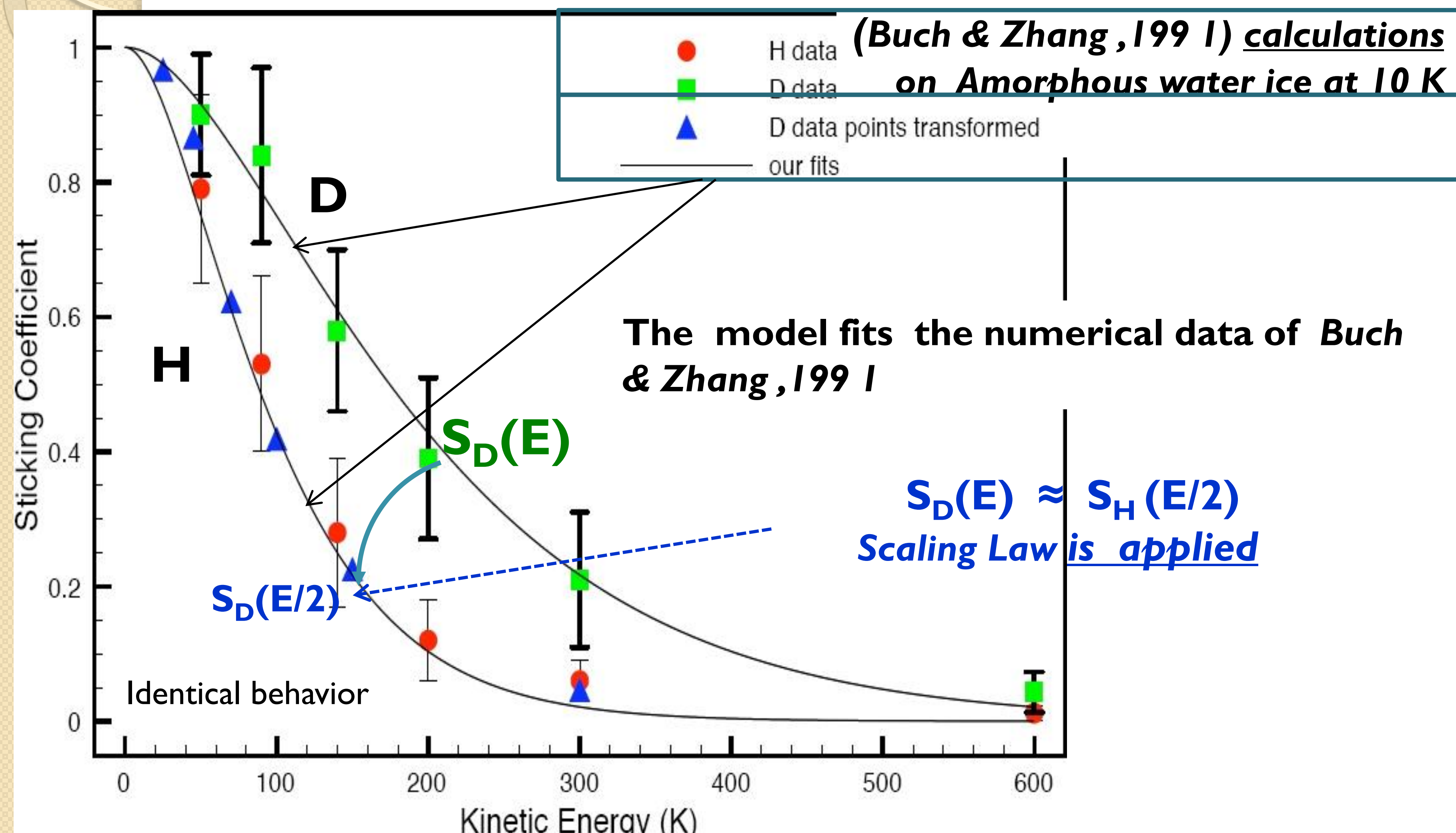
The linear mass dependence of T_0 is a critical prediction for the Model

$m(D_2) = 2 \times m(H_2) \Rightarrow T_0(D_2) = 2 \times T_0(H_2)$

Application of the Model

for D and H atoms

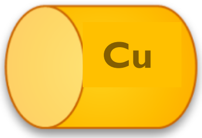
Mass dependence $m_D = 2^* m_H$



The model fits the numerical data of *Buch & Zhang ,199 1*

Sticking experiments on silicates

Substrate : Olivine type Silicates



Sample holder

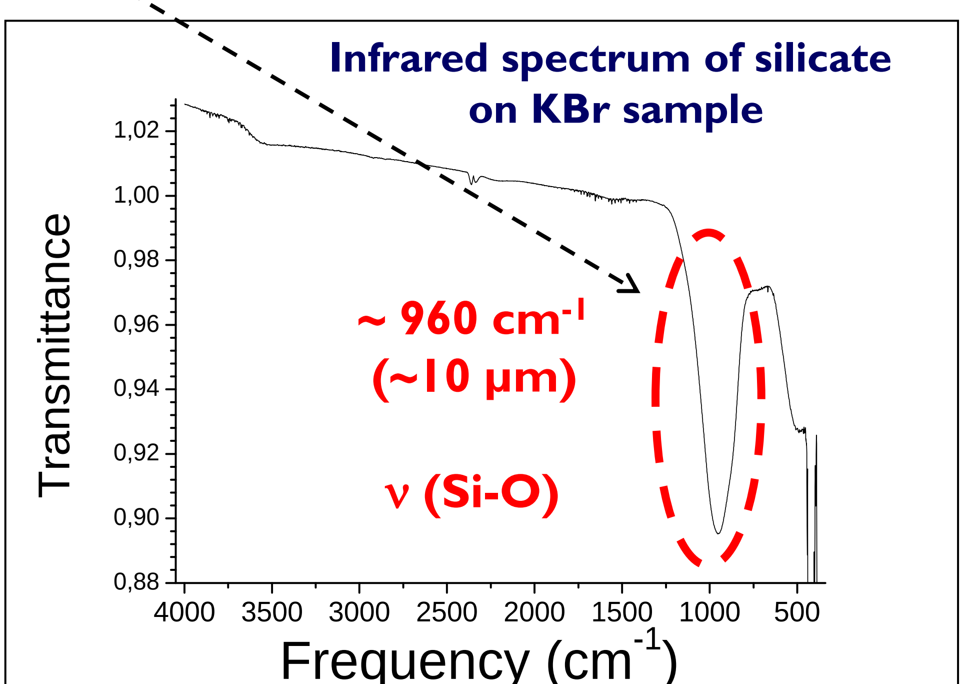
Gold-plated copper block
Covered with silicates

Thickness : ~ 100 nm fully covering the gold surface

$(Fe_x,Mg_{1-x})_2SiO_4$, where $0 < x < 1$.

Preparation: Electron-beam evaporation of San Carlos olivine in a high vacuum chamber

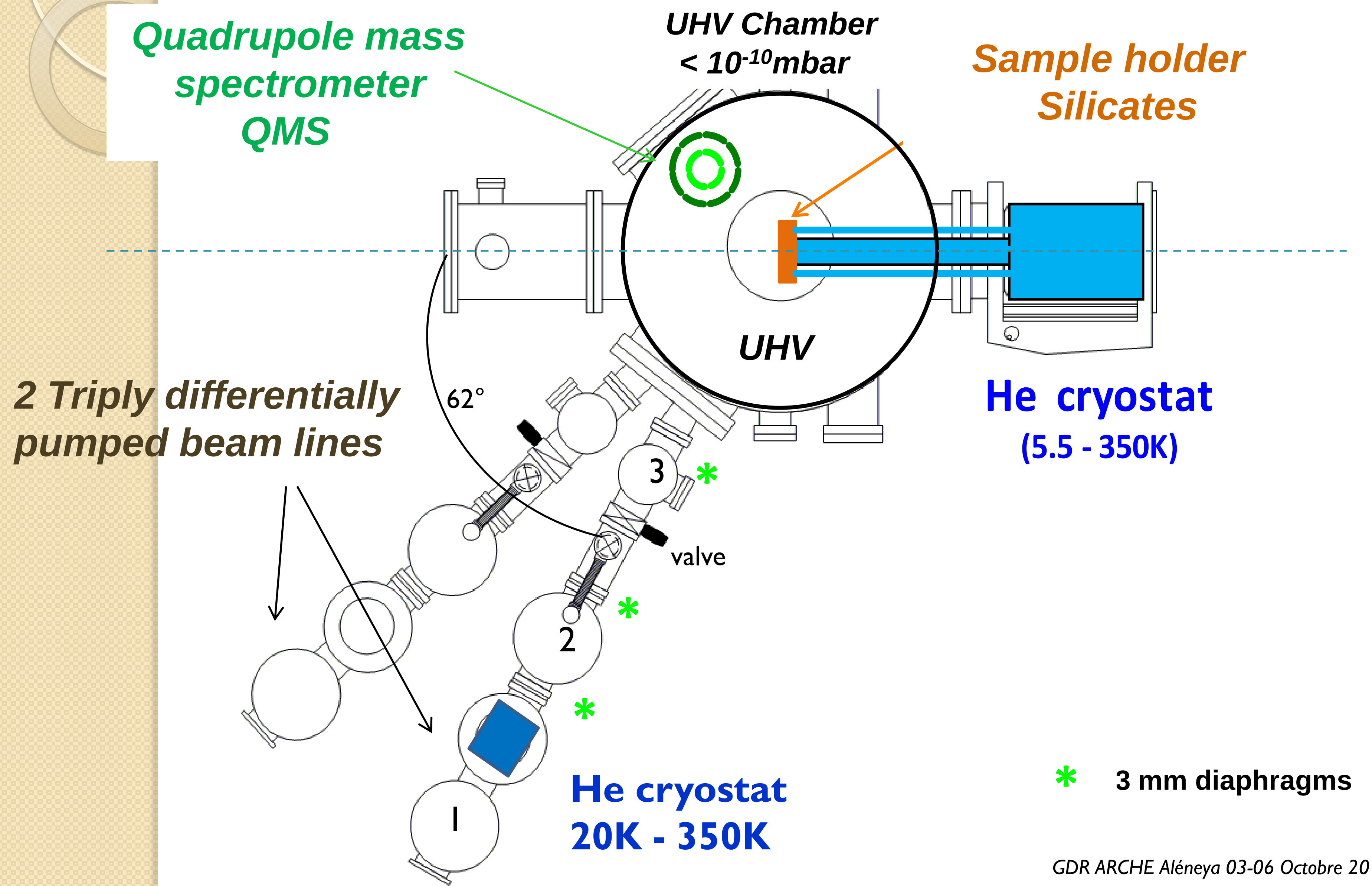
Structure: Amorphous



D'Hendecourt's group,
Astrophysical Laboratory
IAS Orsay

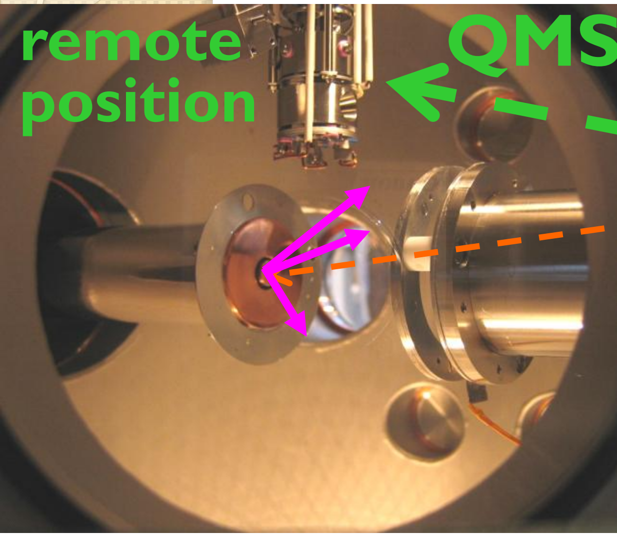
Experimental setup

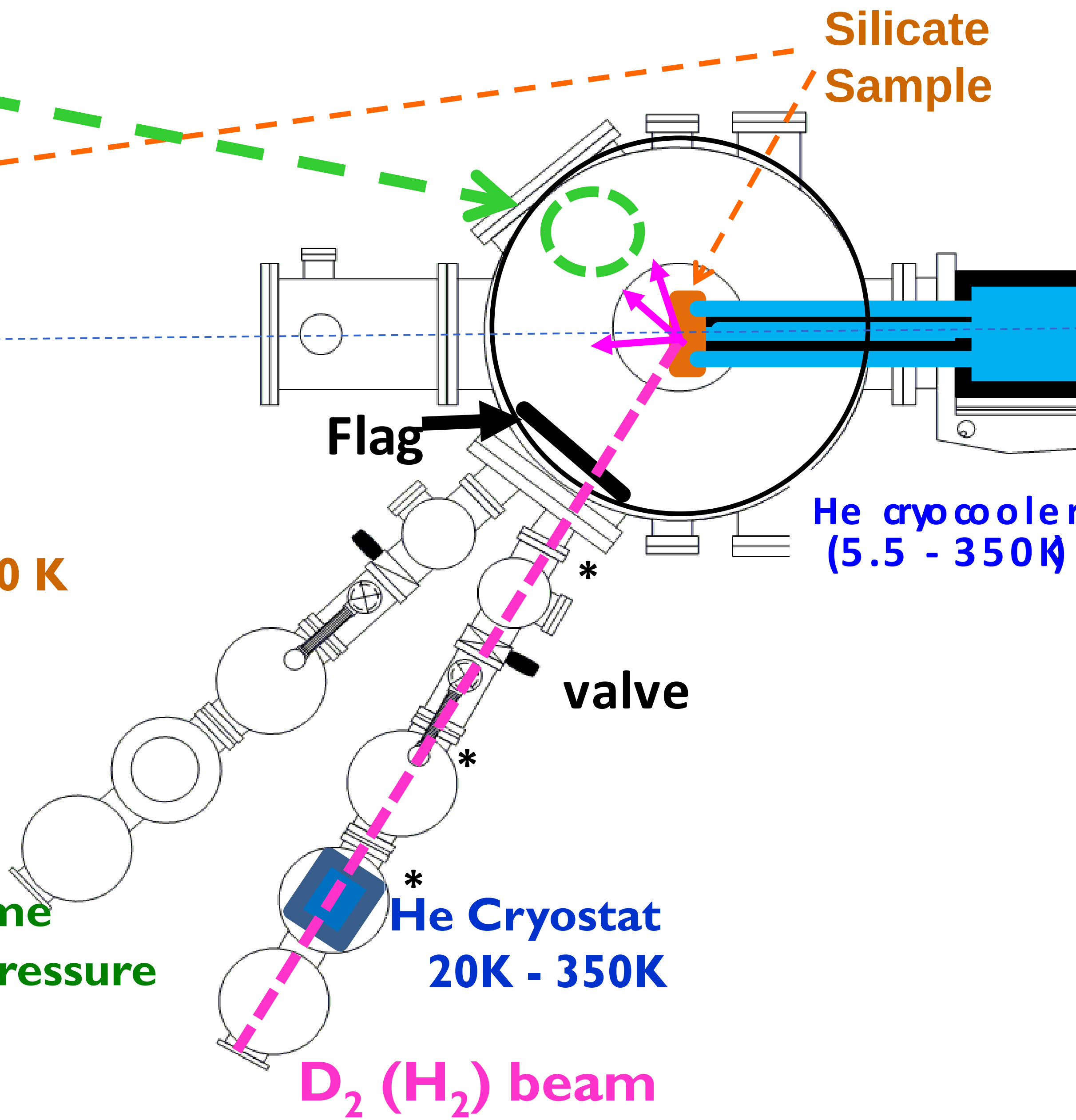
FORMOLISM (FORmation of MOLecules in the InterStellar Medium)



Experimental procedures

King and Wells method (1972)





0 - UHV (< 10^{-10} mbar)

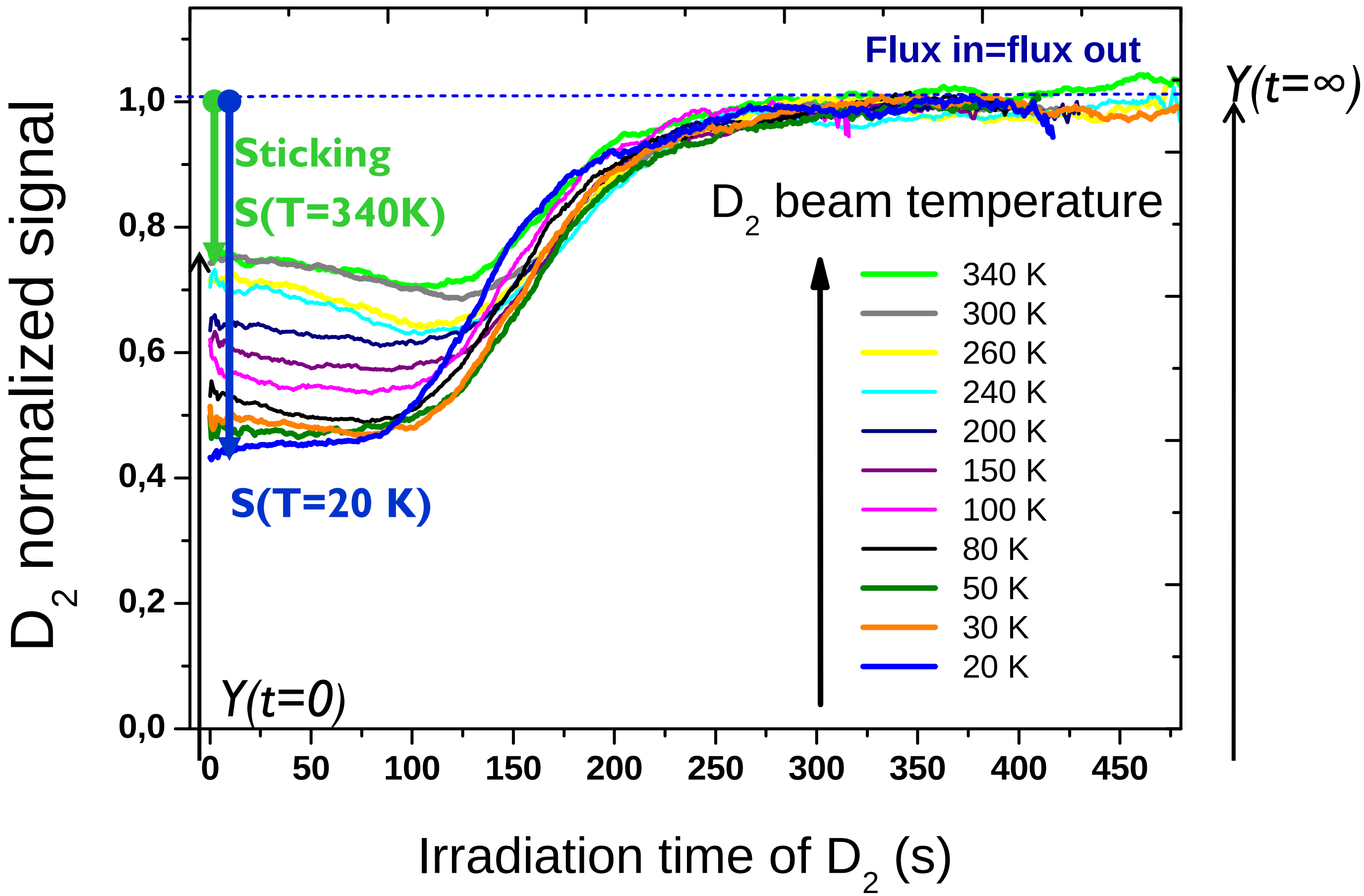
1- Substrate of Silicate at 10 K

2- Exposure of D_2 (H_2) beam (T_{gas}=20 K–350 K)

3- Monitoring in the real time the background partial pressure of D_2 (H_2)

Sticking measurements of molecules

Sticking coefficient of D_2 on silicates at 10K

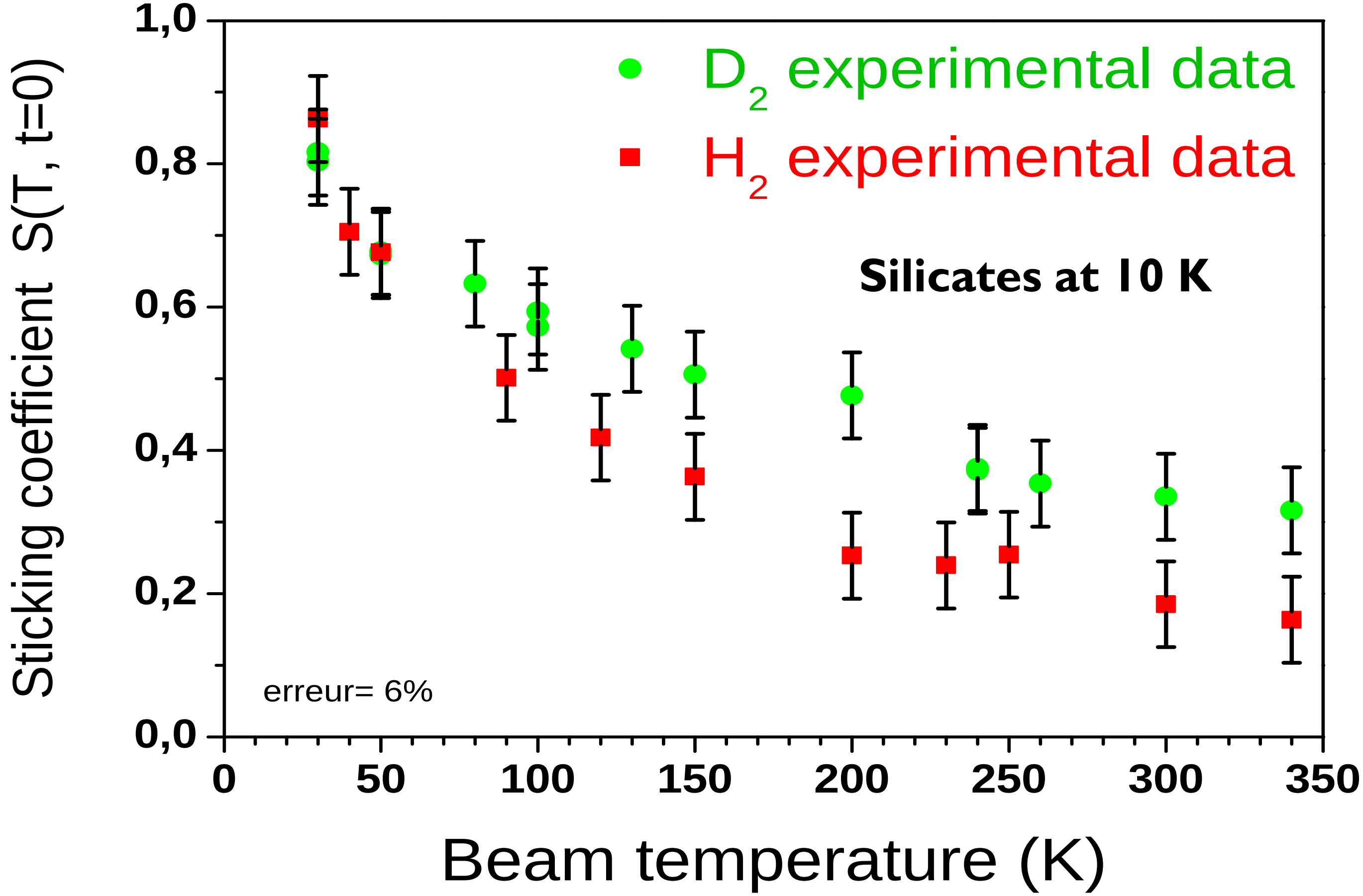


Absolute sticking coefficient S(T)

$$S(T) = S(T, t=0) = \frac{Y(T,\infty) - Y(T, t=0)}{Y(T,\infty)}$$

RESULTS: Sticking coefficient of D_2 and H_2 on Silicates

(Experiments)

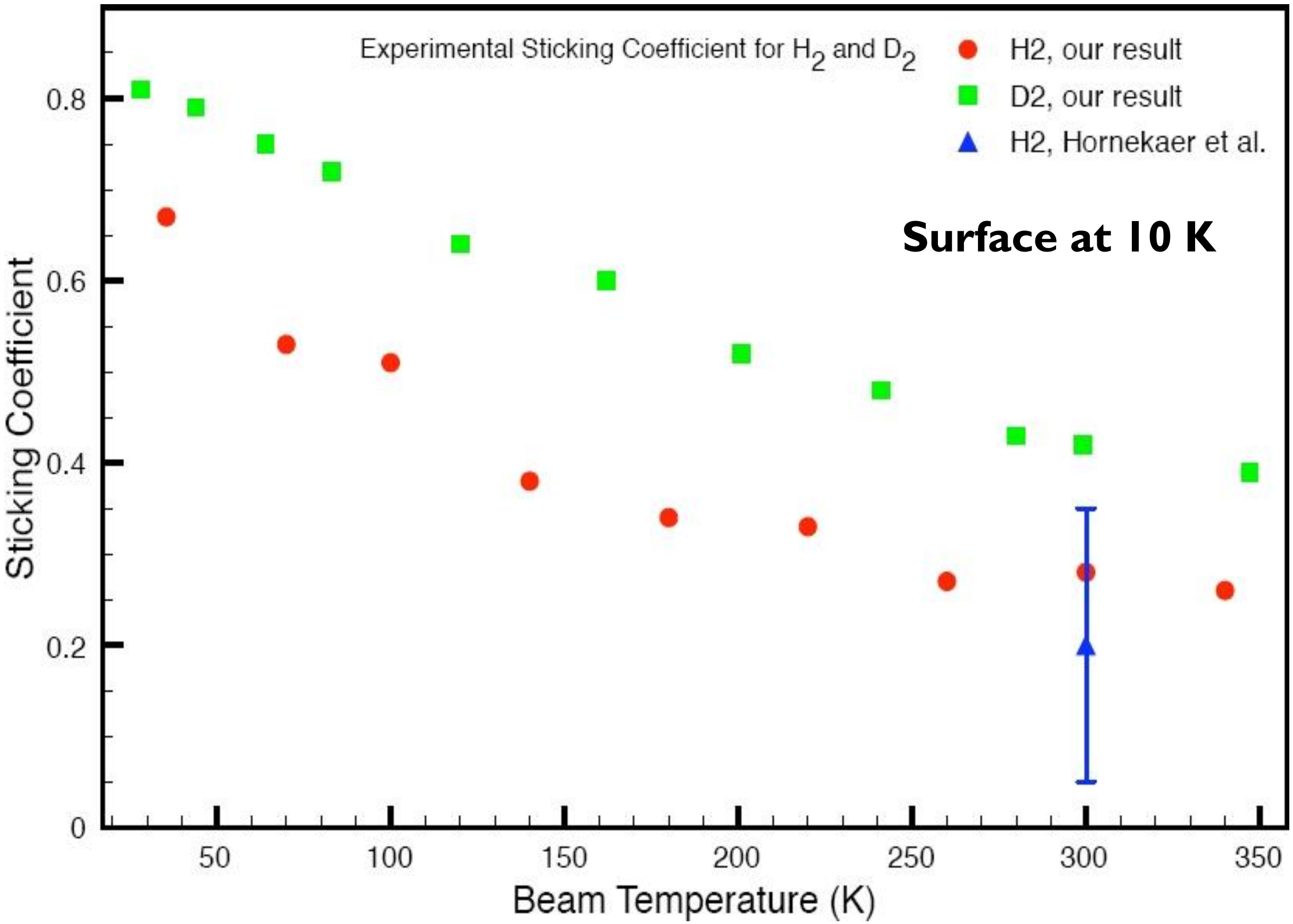


-The sticking coefficient decreases with the gas temperature

-The sticking coefficient is dependent on the mass of the impactor

(Chaabouni et al. 2011, submitted to A&A)

Sticking coefficient of D_2 and H_2 on np-ASW ice (Experiments)

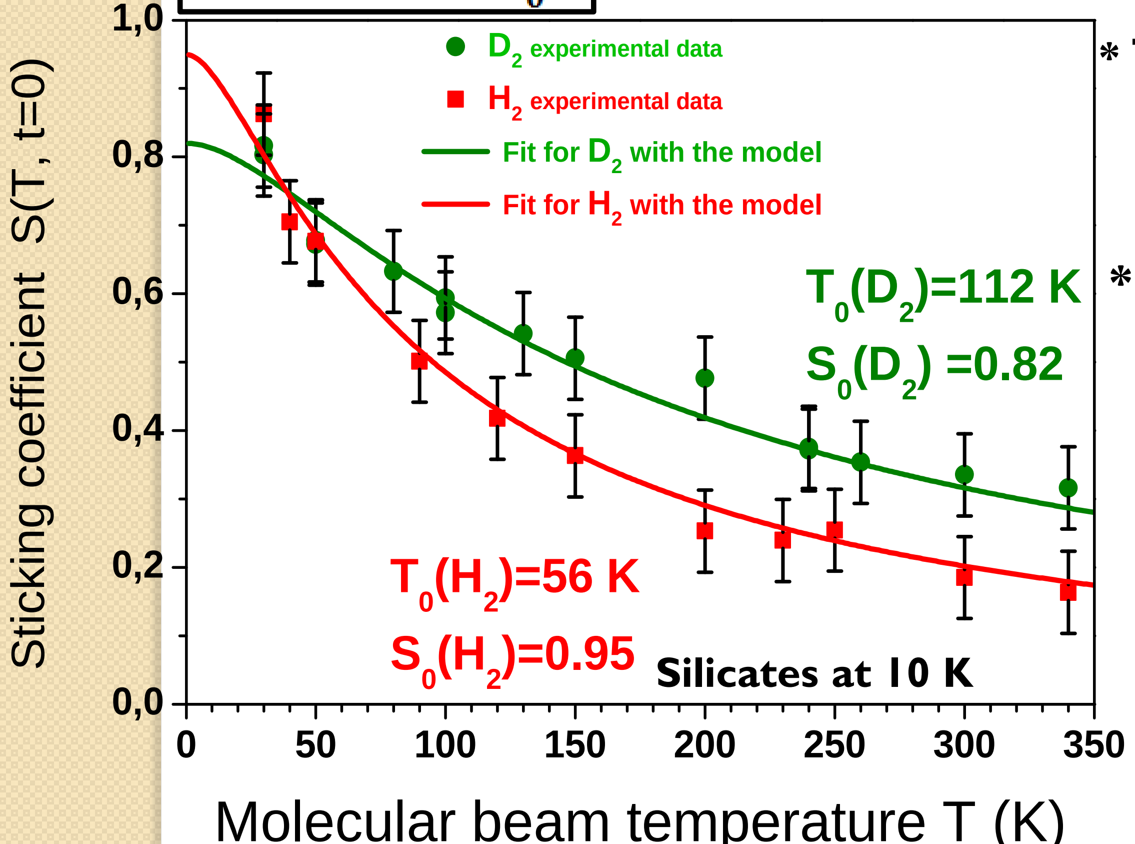


(Matar et al. 2010, JCP)

RESULTS: Sticking coefficient of D_2 and H_2 on Silicates (Model)

$$S(T) = S_0 \frac{\left(1 + \frac{\beta T}{T_0}\right)}{\left(1 + \frac{T}{T_0}\right)^{\beta}}$$

β=2.22 (our effusive beam (θ=62°))



* **The model fits the experimental data on silicates at 10 K for T > 50 K**

* T_0 satisfies the **Temperature dependence relation:**

$$T_0(D_2) = 2 \times T_0(H_2)$$

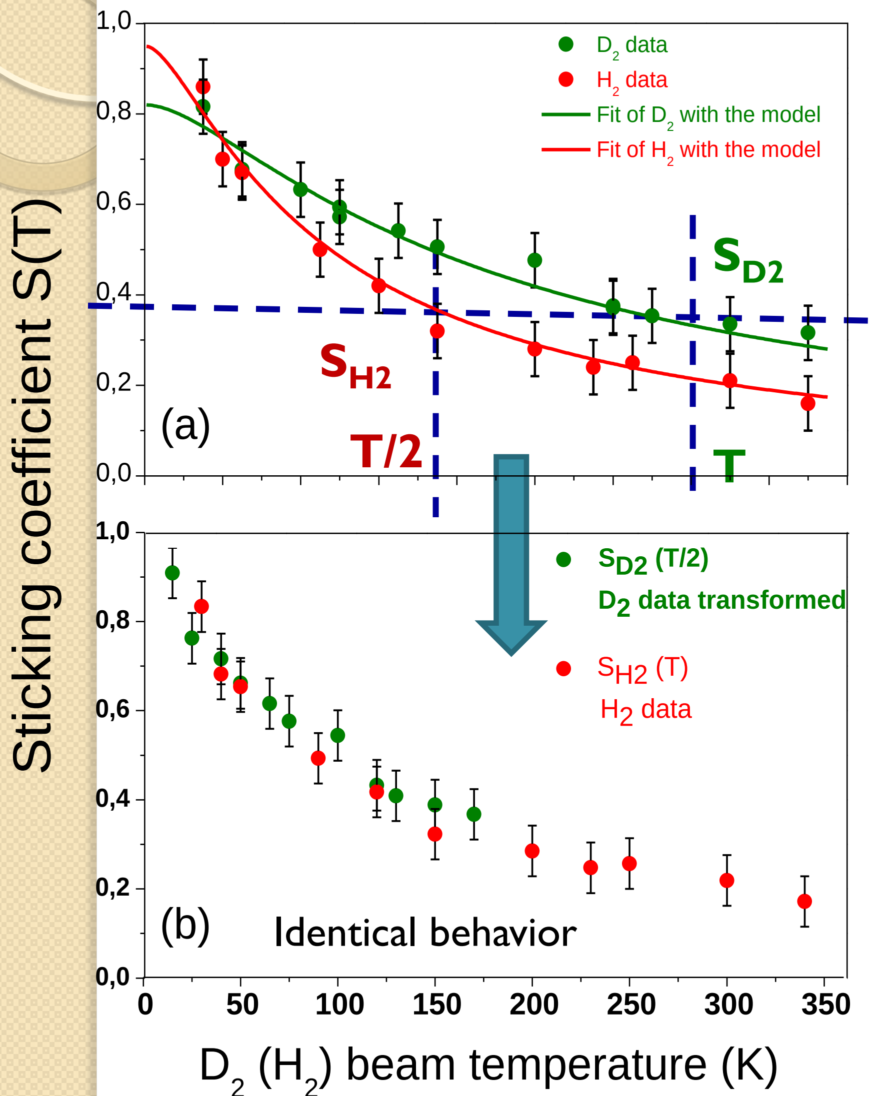
Mass dependence relation:

$$m(D_2) = 2 \times m(H_2)$$

(Chaabouni et al. 2011, submitted to A&A)

RESULTS: Sticking of D_2 and H_2 on Silicates at 10 K

Isotopic effect



Model: Mass dependence

$m(D_2) = 2\ m(H_2)$

Scaling law

(Renormalization-dilation transformation)

$$S_{H_2}\left(\frac{T}{2}\right) = \frac{S_0(H_2)}{S_0(D_2)} \times S_{D_2}(T)$$

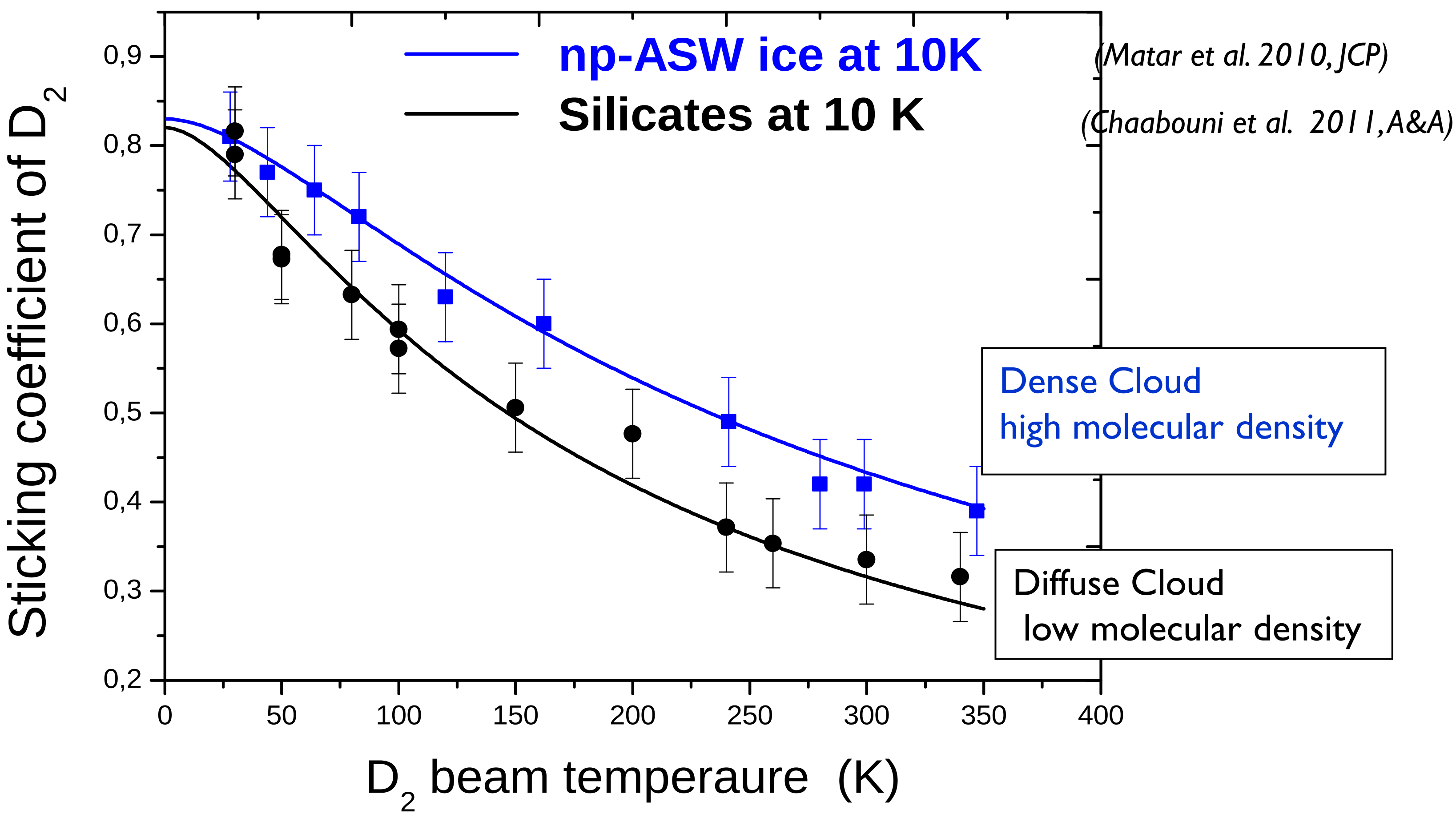
$S_0(D_2)/\ S_0(H_2) = 0.86$ (Silicates at 10K)

$\Rightarrow$ The scaling law is verified

$\Rightarrow$ **Isotopic effect between D_2 and H_2 is explained by the the factor 2**

(Chaabouni et al. 2011, submitted to A&A)

Grain surface dependence of the sticking coefficient



Dense Cloud
high molecular density

Diffuse Cloud
low molecular density

Sticking coefficient of hydrogen on silicates is lower than that on ASW ice

⇒ - Rigidity of silicates (binding energy of the solid network)

- Low transfer momentum energy

Sticking of atoms

Previous experimental studies

Interaction between H or D with silicates surfaces

(Pirronello et al. 1997; Perets et al. 2007; Vidali et al. 2007, 2009)

→ **No experimental estimates for the sticking of atoms**
No isotopic effects are usually considered

Experimental Difficulties

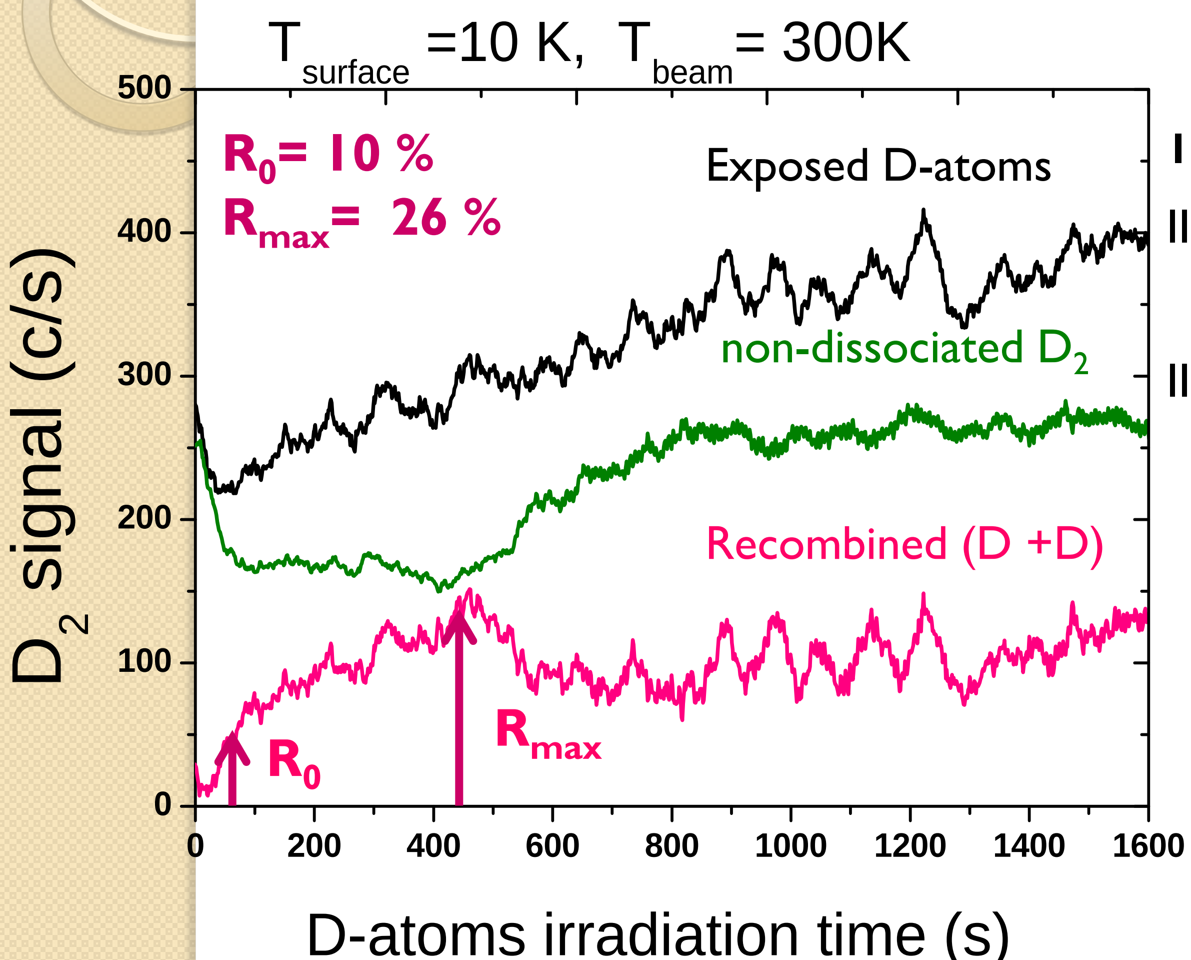
Sticking coefficient of hydrogen atoms cannot be directly measured as in the case of molecules (H_2 and D_2)

→ **Dynamic recombination of H atoms**
(Schutte et al. 1976, Govers et al. 1980, 2005, Lemaire et al. 2010)

Sticking coefficient measurement of H(D) atoms → **Extrapolation from hydrogen recombination efficiency**

Sticking measurements of atoms

Experiments: Sticking coefficient of D atoms



King and Wells Method (1972)

I- Silicates surface at10K

II- Exposure of D atoms

dissociation rate of (D_2) is 65%

III- Monitoring the D_2 signal

↓

Recombination efficiency

$$R = \frac{recombined\ (D+D)}{exposed\ (D+D)}$$

↓

10 % <S(D)< 26%

S(D) = 0.18 ± 0.08

T_{beam}= 300K

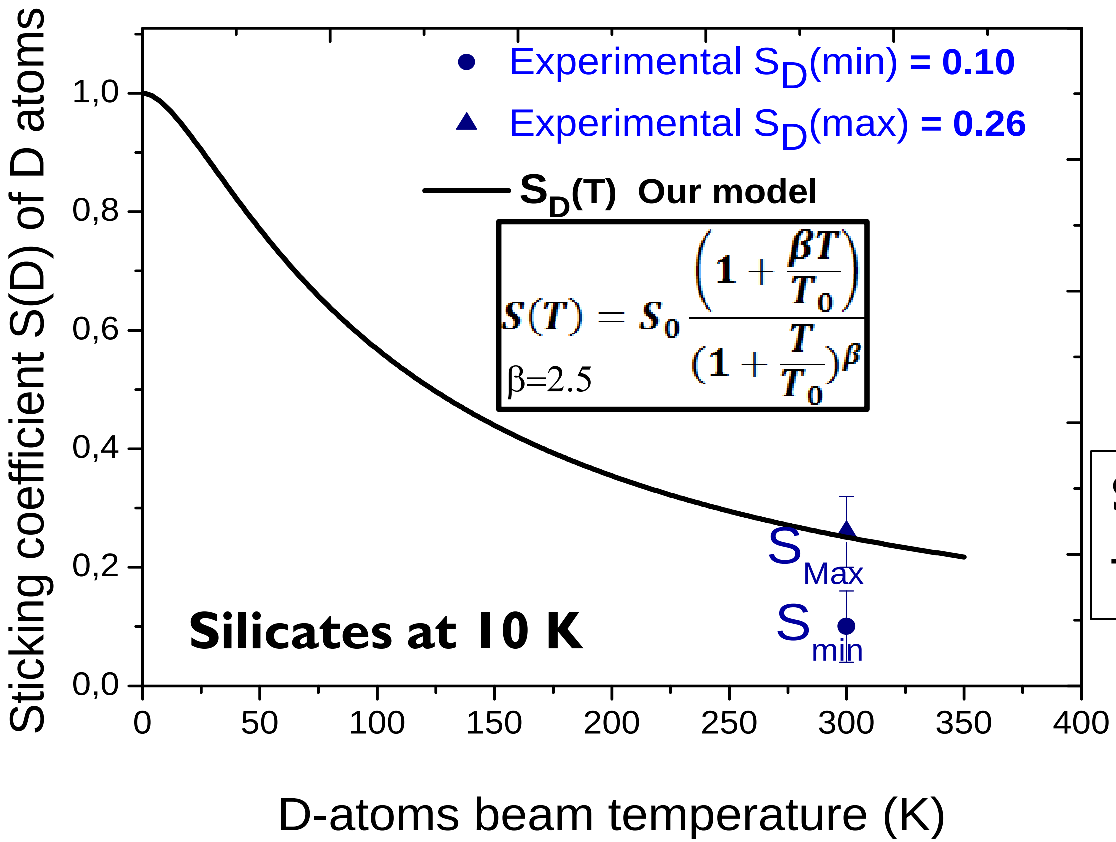
(Amiaud et al. 2007)

Sticking measurements of atoms

MODEL: Sticking coefficient of D atoms

Assuming **S_0(D) =1** (fits of atomic data by Buch & Zhang (1991) on ASW ice)

$T_0(D_2)$ / T_0(D) = 1.67 (for np-ASW ice and silicates)



$T_0(D_2)$ = 112 K (silicates)

T_0(D) = 50 K

S(D) = 0.26 (Model)

T_{beam}= 300 K

Correlation between experiments and model

Astrochemical Implications

$$S(T) = S_0 \frac{\left(1 + \frac{\beta T}{T_0}\right)}{\left(1 + \frac{T}{T_0}\right)^{\beta}}$$

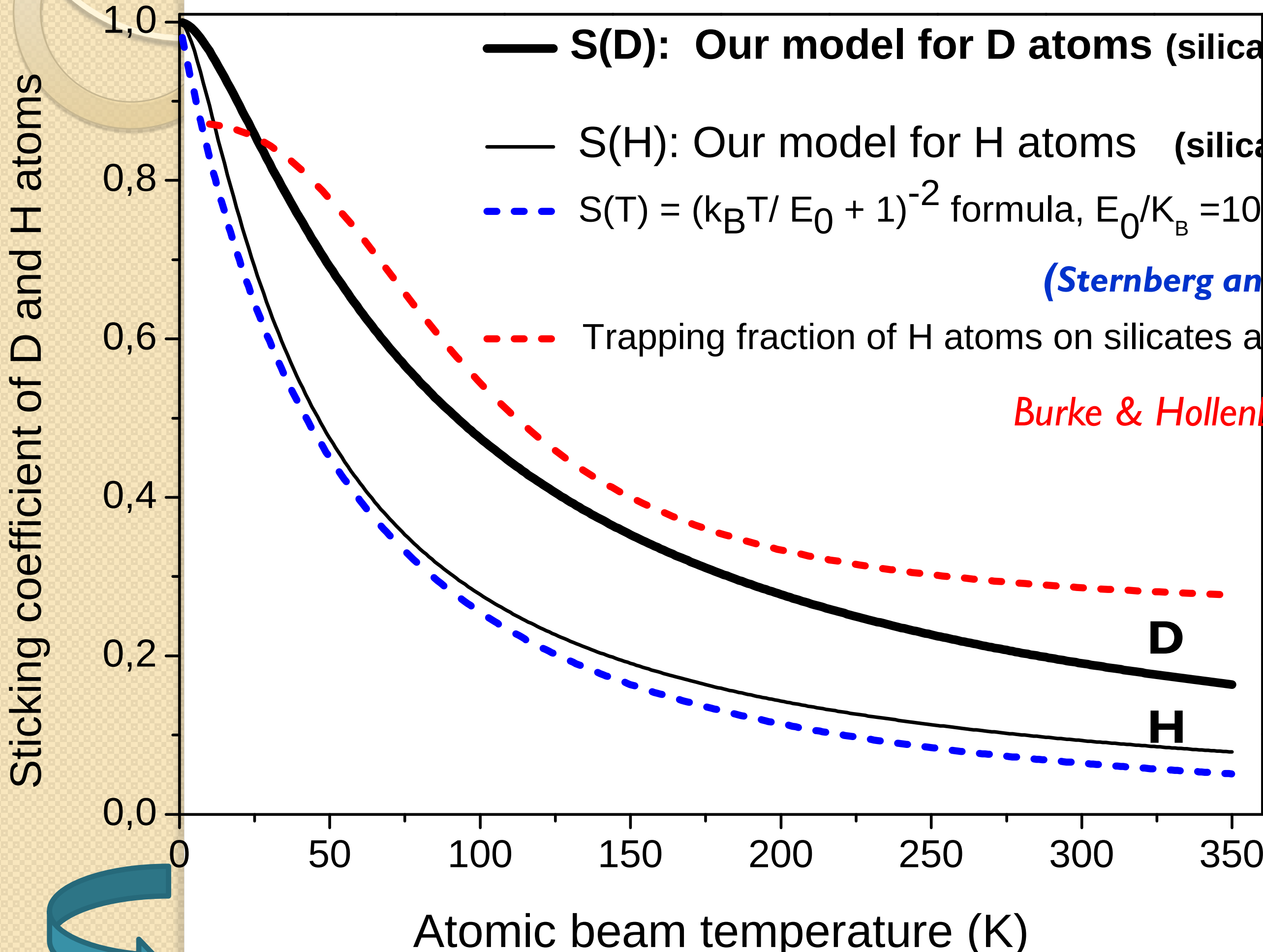
Sticking coefficient law

Table 1. Sticking parameters S_0 and T_0 for H_2, D_2, H, D and HD on np-ASW ice (Matar et al. 2010) and on silicate surfaces held at 10 K.

Substrate	Species	S_0	T_0 (K)	References	
NP-ASW ice at 10 K	H	1	52	Buch & Zhang (1991)	T0(D) = 2×T0(H)
	D	1	104	Buch & Zhang (1991)	$T_0(D_2)$ / T_0(D) = 1.67
	H_2	0.76	87	Matar et al. (2010)	**(Fit)**
	D_2	0.80	174	Matar et al. (2010)	**(Fit)**
	HD	0.83	130.5	Prediction	T_0 (HD)= 3/2 $T_0(H_2)$
Silicate at 10 K	H	1	25	Extrapolation	**T0(D) = 2×T0(H).**
	D	1	50	Extrapolation	**$T_0(D_2)$ / T_0(D) = 1.67**
	H_2	0.95	56	This study **(Fit)**	
	D_2	0.82	112	This study **(Fit)**	
	HD	0.87	84	Prediction	**T_0 (HD)= 3/2 $T_0(H_2)$**

(Chaabouni et al. 2011, submitted to A&A)

Astrochemical Implications



$S(T) = (k_B T/E0 + 1)^{-2}$ match our results on silicates if **$E0/k_B$ (H) = 80 K**

The Isotopic properties of the sticking have an incidence on the observed HD molecules

Conclusions

EXPERIMENTS

- **The sticking coefficient of D_2 (H_2) on np-ASW ice and silicates surface at 10 K decreases with the gas temperature.**

MODEL

- **Depends on the mass of the gas light species**
- **Fits efficiently the experimental data for H_2 and D_2 on np-ASW ice and silicates surface at 10 K**
- **Provides an analytical formula S(T) for the thermal sticking coefficient of hydrogen species on dust grains**
- **Explains the isotopic effect between D_2 and H_2, and D and H atoms**
- **Provides parameters S_0 and T_0 for D_2, H_2, HD, H and D atoms on both silicates and ASW ice. These are very valuable for astrochemical models.**