

A Summary of Experimental Results on H₂ Formation on Dust Grain Analogues

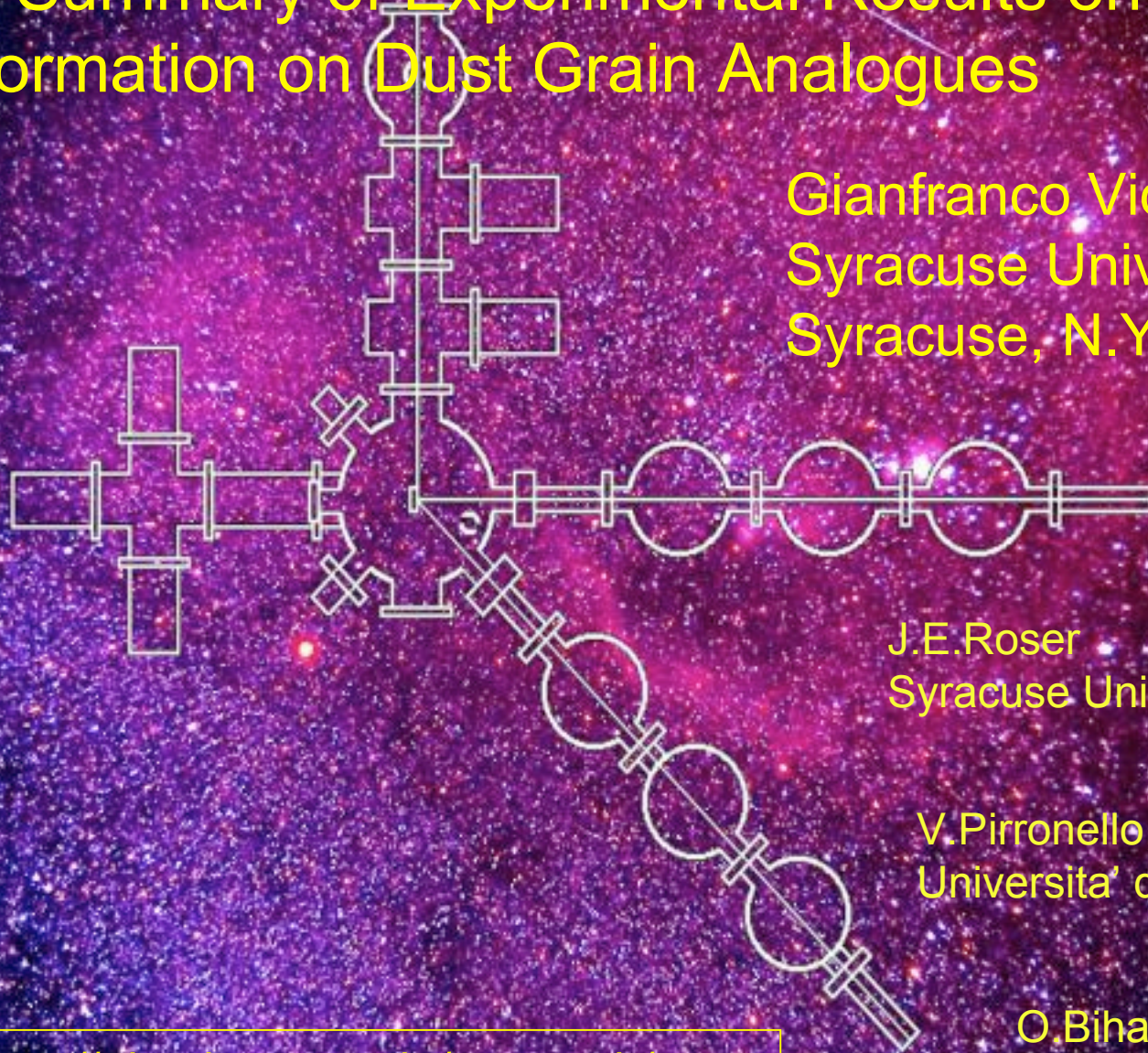
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<http://physics.syr.edu/research/astro>



Formation of H₂ in the ISM

H+H→H₂ not in the gas phase

$$d n(\text{H}_2) / dt = R n n(\text{H}) - \beta n(\text{H}_2)$$

$$R = 3 \cdot 10^{-17} \text{ cm}^3 \text{ s}^{-1} \quad \beta = 5 \cdot 10^{-11} \text{ s}^{-1}$$

(Jura '75, Habbard et al. 2004)

H+H→H₂ on dust grains (polycrystalline ice) -
Salpeter, Hollenbach ~1970

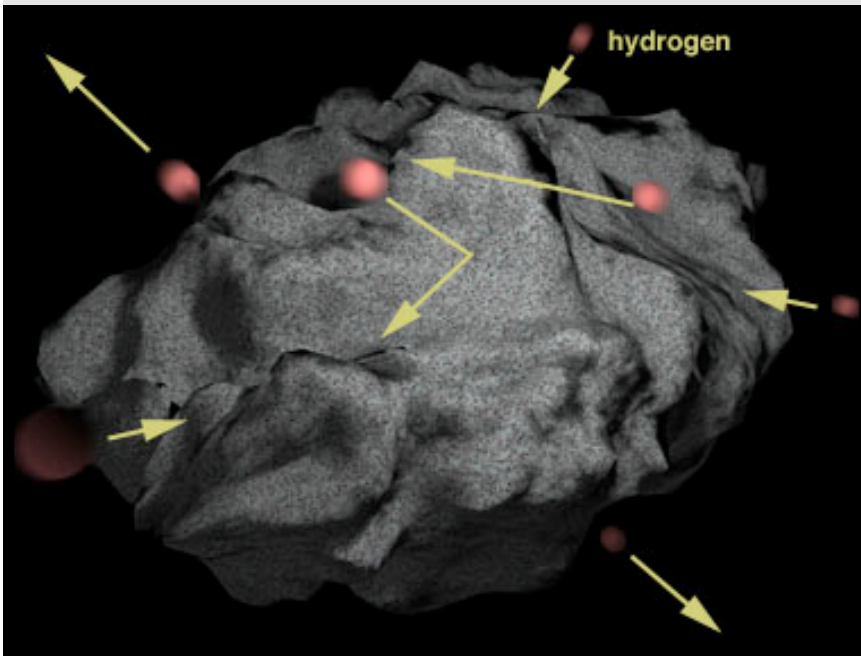
$$\text{Model: } R_{\text{H}_2} = 1/2 (n_{\text{H}} v_{\text{H}} \sigma \xi) n_{\text{g}} \text{ (cm}^{-3} \text{ s}^{-1}\text{)}$$

H+H→H₂ on amorphous ice - Smoluchowski,
1979

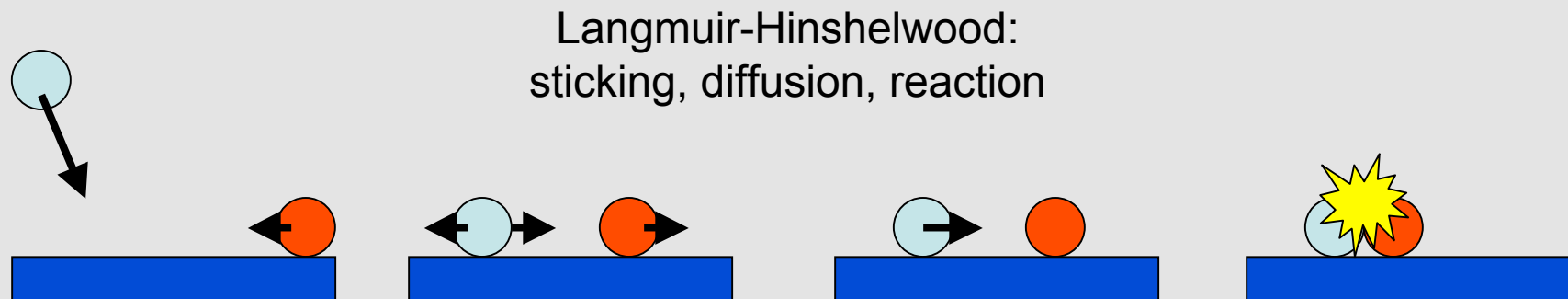
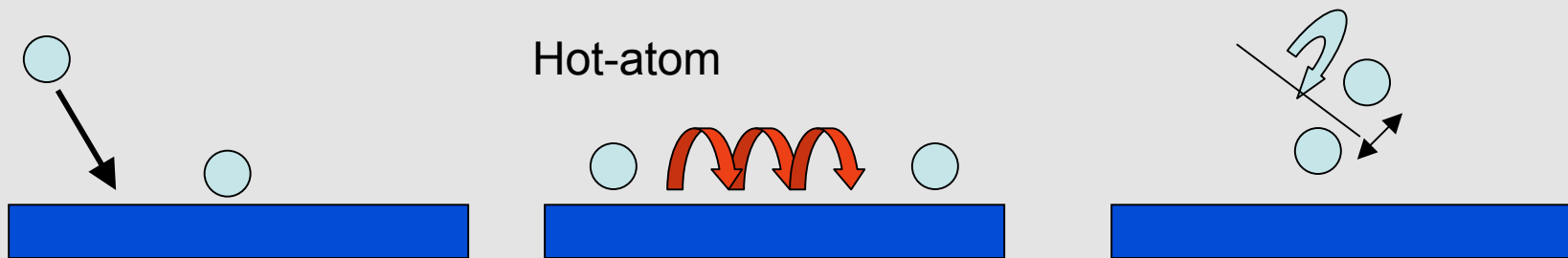
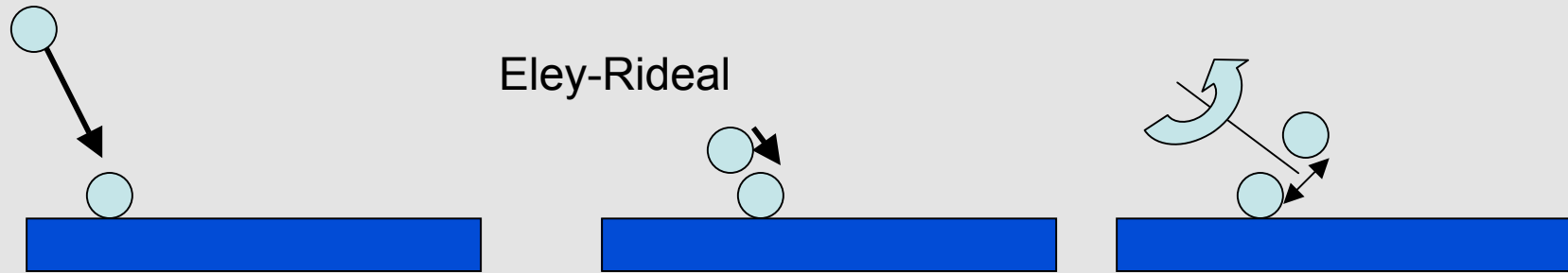
- * How does H₂ form on grains?
- * Under what conditions (temperature, grain morphology, etc.)?
- * With what rates?
- * What happens to the energy released in the reaction?



We initiated a
research program at
Syracuse University
in 1996 to answer
these questions



Mechanisms of reaction



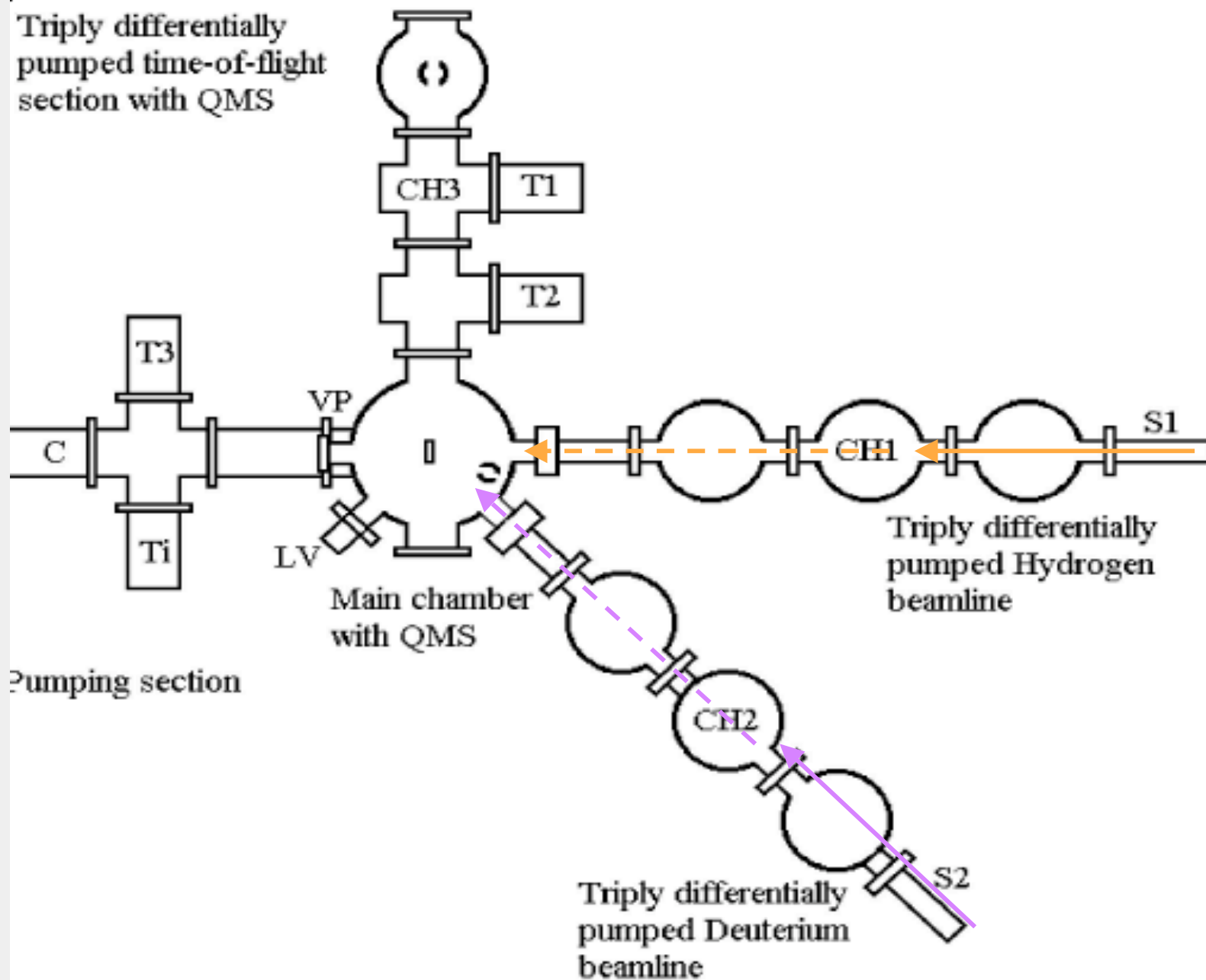
Themes being addressed by laboratory research

- **Efficiency & Mechanism** of formation of H₂ in diffuse clouds - grains are bare (T~15-25 K) (**Syracuse University, the very first results in 1997**)
- **Efficiency & Mechanism** of formation of H₂ in dense clouds - grains are coated with ices (mostly H₂O) (T ~ 10-20 K) (**Syracuse University - 2001-2003, Denmark - 2003**)
- **Partition of energy** released in the H₂ bond formation (**Syracuse University - 2003, Denmark - 2003, UCL, Cergy-Pontoise**)

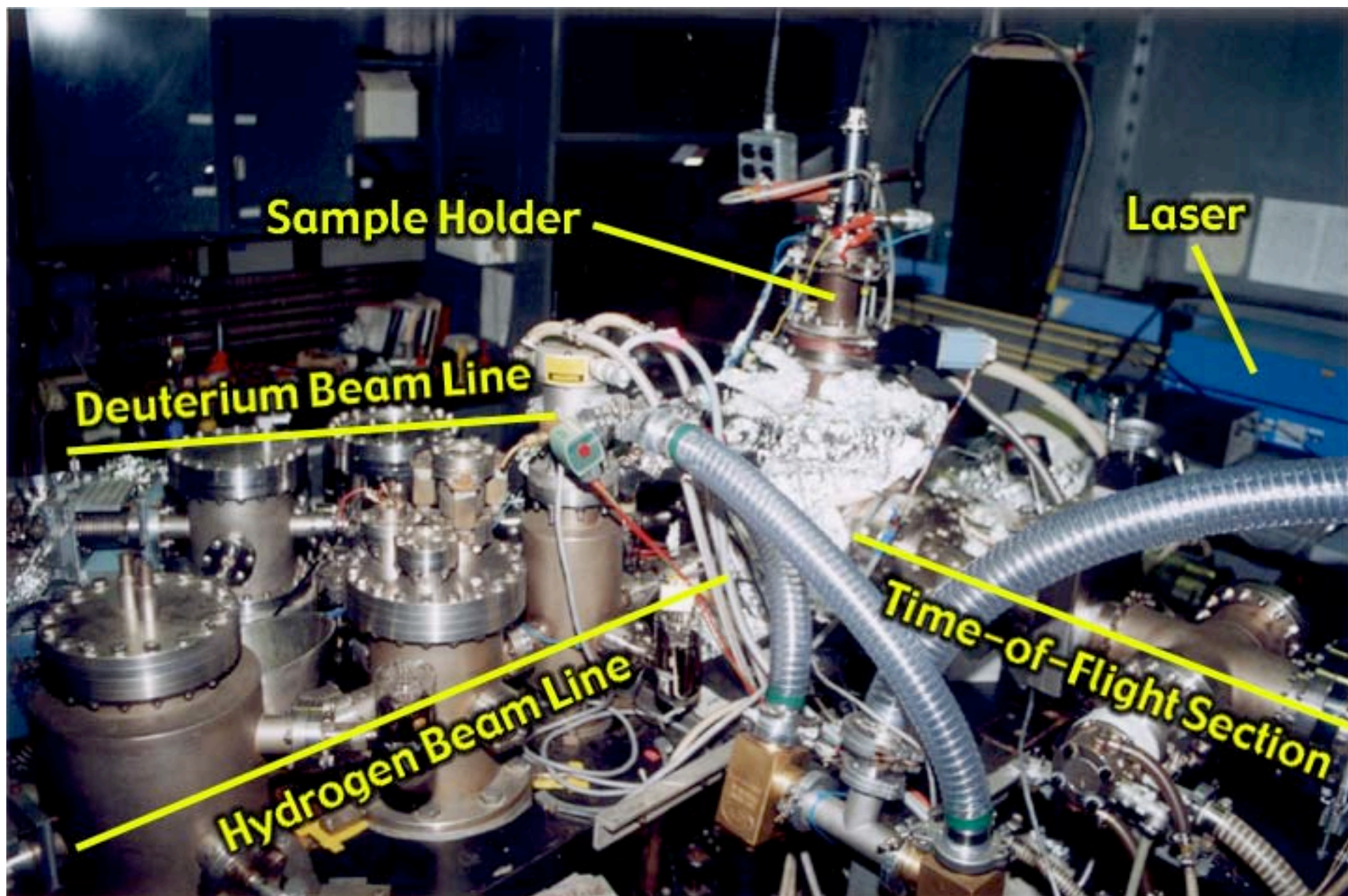
Application of surface science techniques to astrophysical problems

- In 1996, at **Syracuse University** we started a program of measurement of **hydrogen recombination and hydrogenation/oxidation** reactions on surfaces of dust grain analogues
- Experimental Conditions
 - Low background pressure** (10^{-10} torr)
 - Low sample temperature** (5-40 K)
 - Two atomic beams**
 - Low kinetic energy of H atoms:** ~ 200 -300 K
 - Very low flux of H atoms** $< 10^{12}$ atoms/cm²/sec

Experimental Apparatus at S.U.



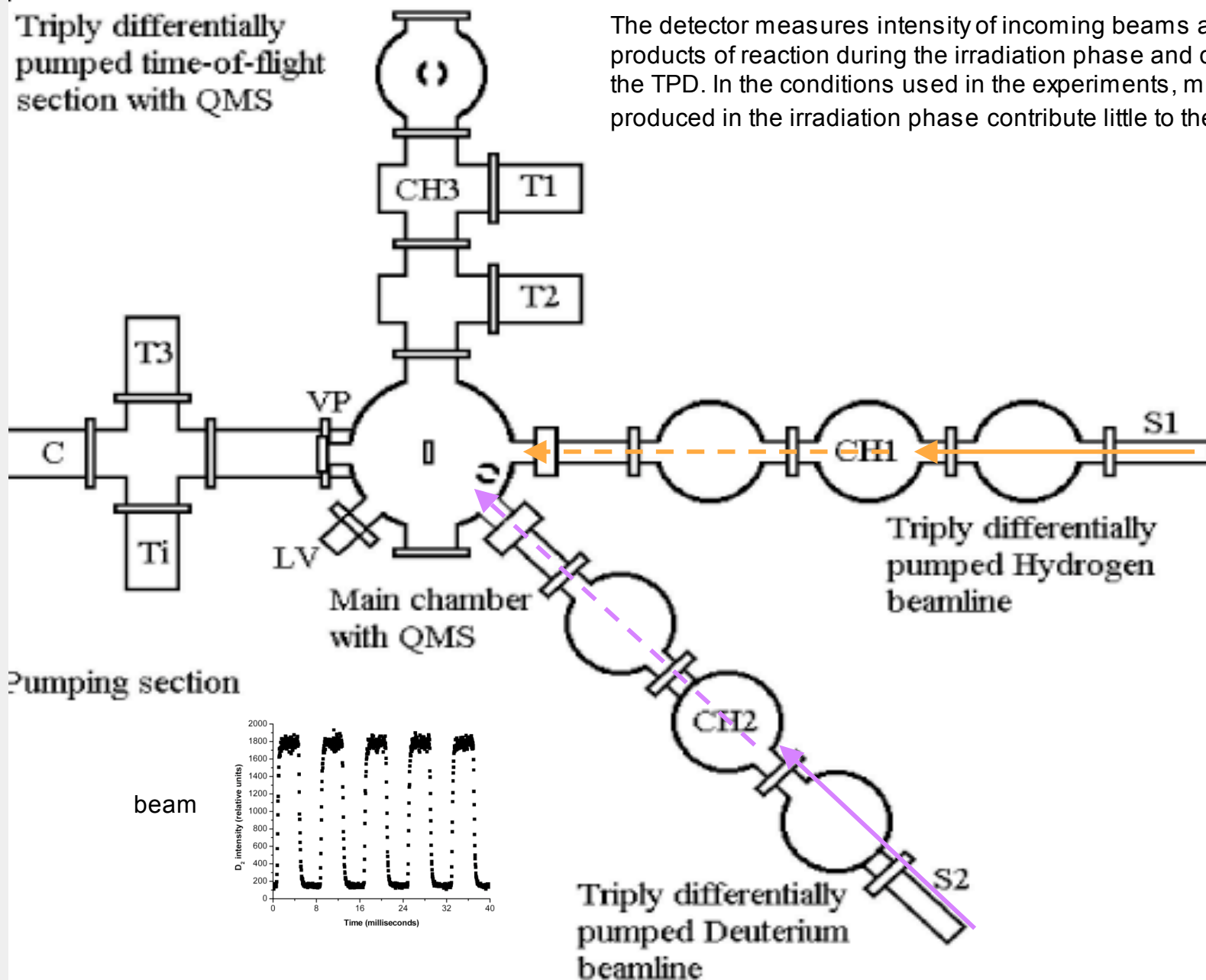
Apparatus at S.U. to study molecule formation on dust grain analogues



Experimental Apparatus at S.U.

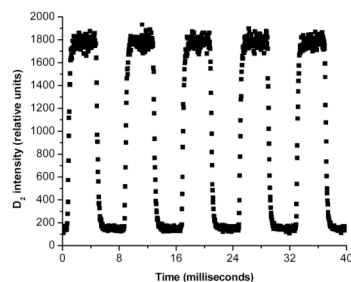
Triply differentially pumped time-of-flight section with QMS

The detector measures intensity of incoming beams and the products of reaction during the irradiation phase and during the TPD. In the conditions used in the experiments, molecules produced in the irradiation phase contribute little to the total yield



pumping section

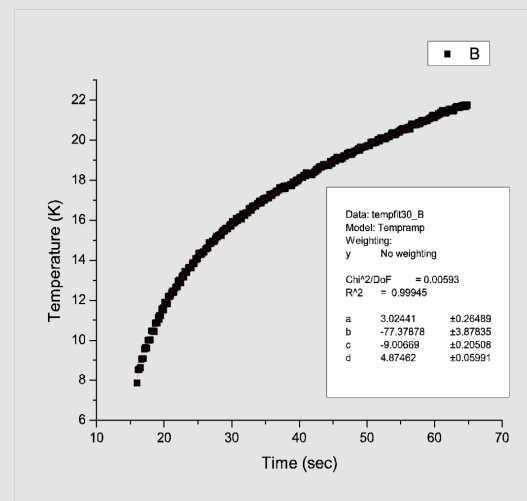
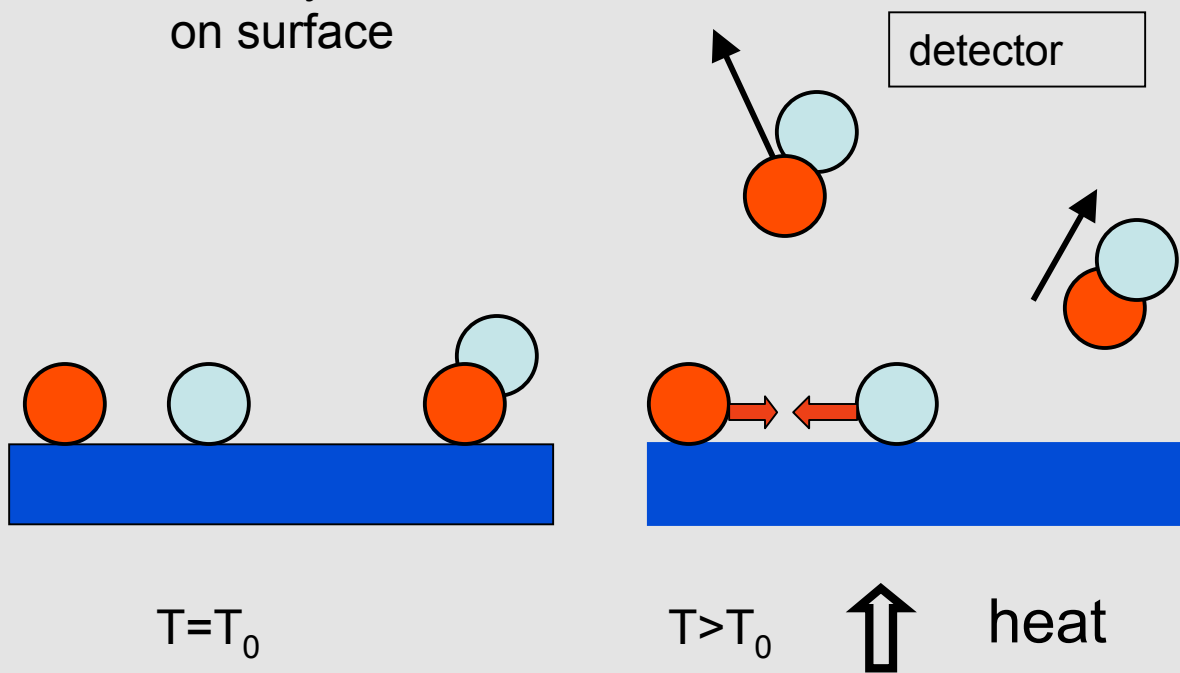
beam



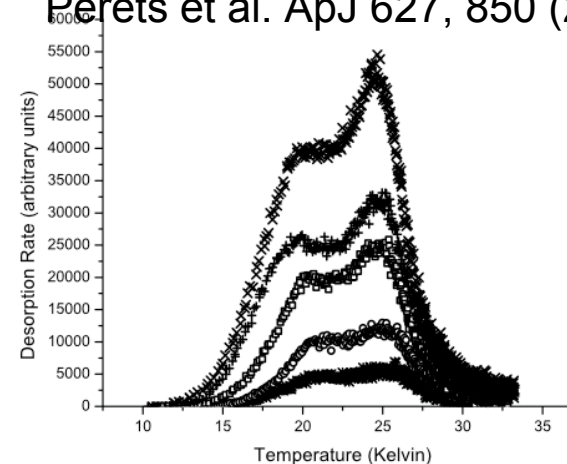
Triply differentially pumped Deuterium beamline

Temperature Programmed Desorption

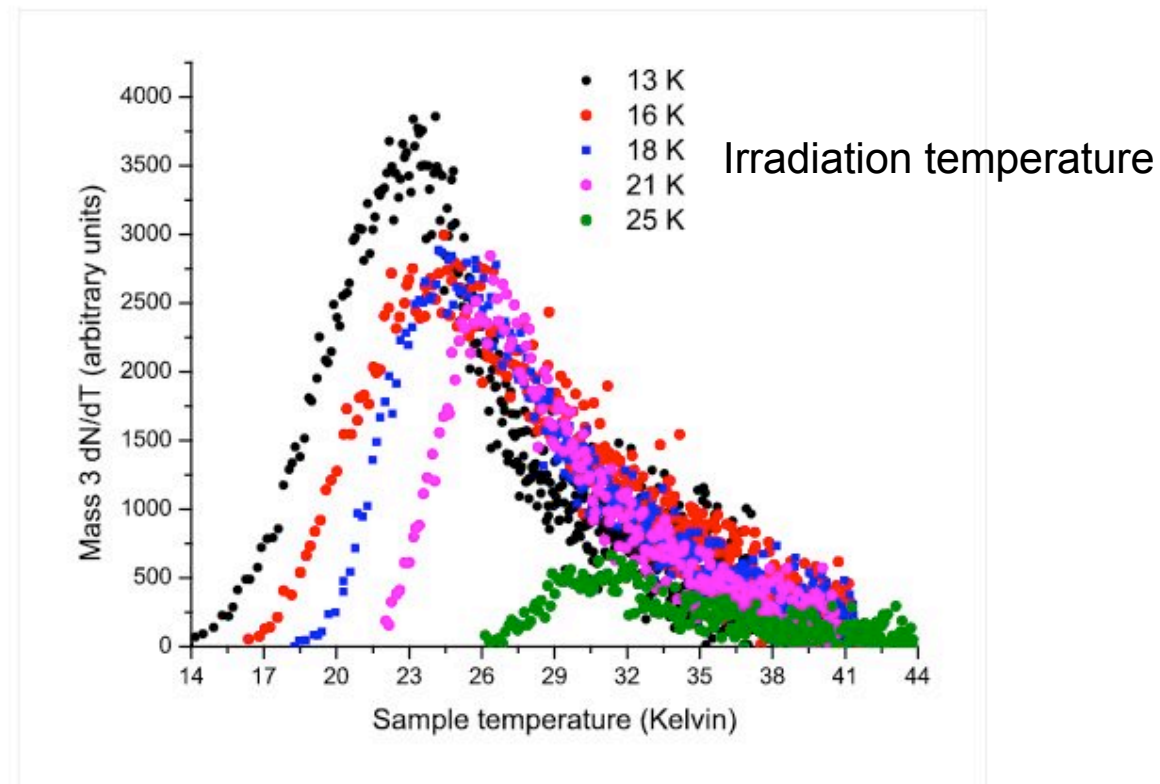
1. To set atoms in motion or
2. To desorb molecules already formed on surface



Perets et al. ApJ 627, 850 (2005)



Example of TPDs after irradiation at different sample temperature

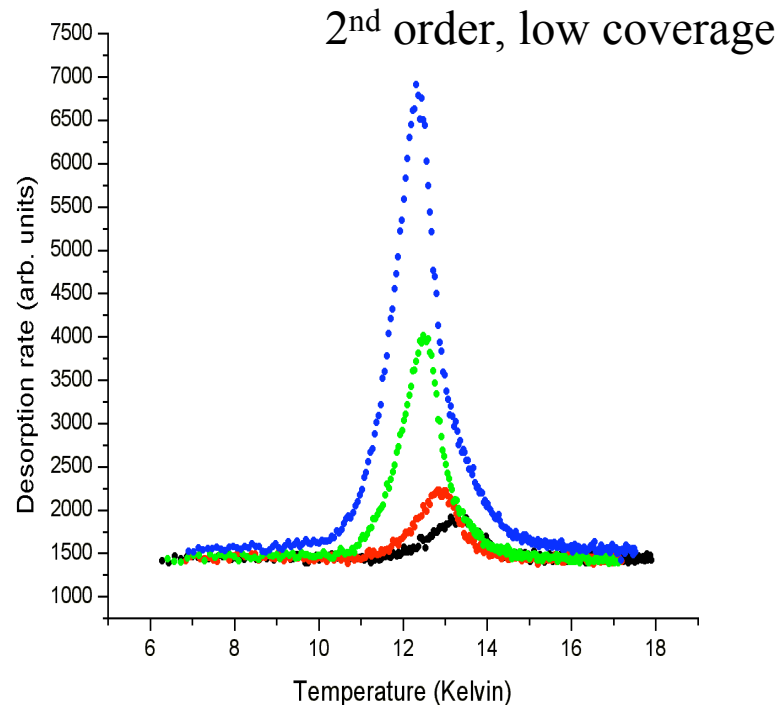


HD formation on amorphous silicates

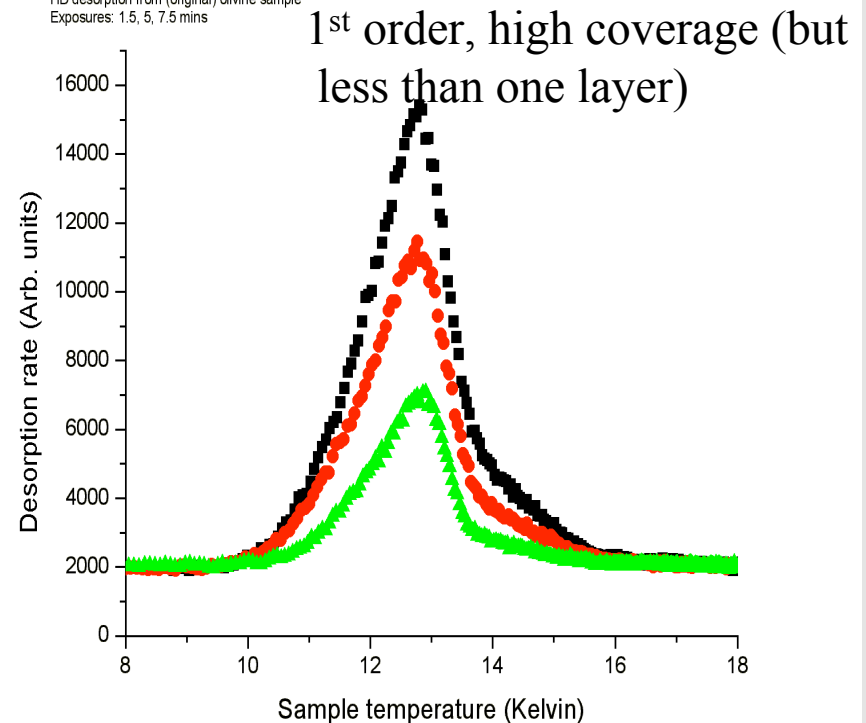
First Results of H+D \rightarrow HD Reaction on Surfaces of Dust Grain Analogues in Simulated ISM Conditions

- Thermal desorption trace: HD from a **polished polycrystalline olivine** as a function of exposure (sub-monolayer coverage) - Pirronello et al. Ap.J. 483, L131 (1997)

HD desorption from (original) olivine sample
Exposures: 5, 2.5, 0.5, 0.25 mins



HD desorption from (original) olivine sample
Exposures: 1.5, 5, 7.5 mins



- Evidence of formation of HD during the TPD at low coverage
- This implies that there is thermally activated mobility

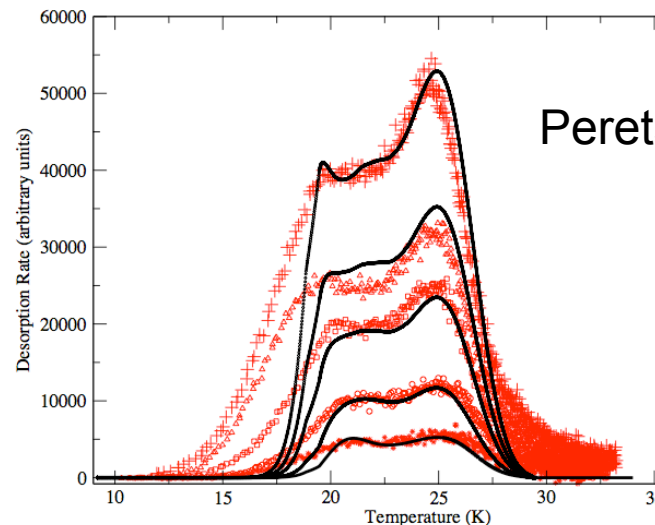
Hornekaer et al. (2003): evidence for instantaneous formation of HD on amorphous ice at ~8K

Experiment at SU:

TPD traces on low density amorphous ice:

Five different coverages

Left peak shows second order kinetics, right peak first order kinetics



This indicates that on this type of ice, there is evidence of thermally activated diffusion

Studies of formation of HD in/on amorphous water ice

Comparison of HD desorption after:

- deposition of H and D at ~10K

and after

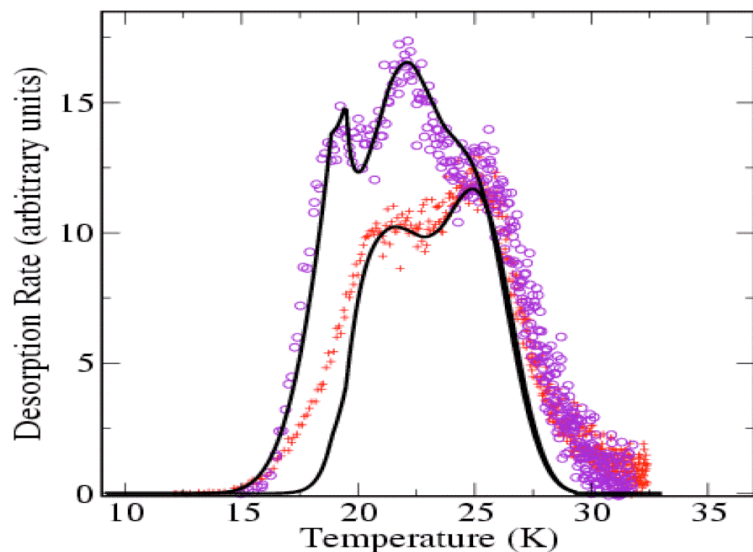
- deposition of HD at 10K



They are different !,
indicating that two different
processes are taking place



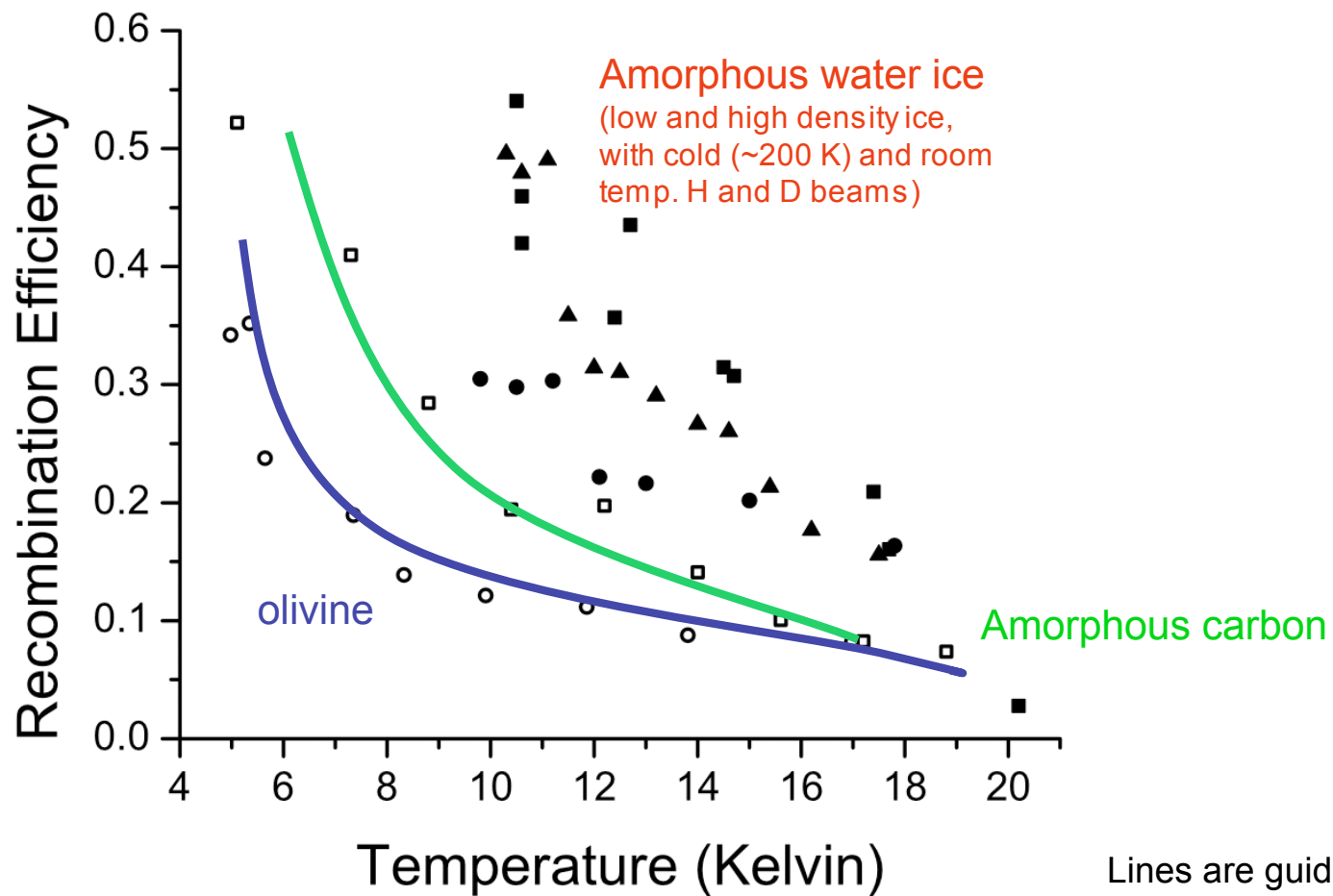
Evidence that HD doesn't form quickly, but thermal activation
is necessary to enable diffusion!



Perets et al. ApJ 627, 850 (2005)

H+D irradiation vs. HD irradiation

H₂ recombination efficiency on dust grain analogues (data from S.U.)



The recombination efficiency is defined here as half the ratio of the number of molecules detected and the number of atoms striking the sample

Lines are guide to the eye

Analysis of Thermal Desorption Experiments: Rate equations

$$d N_H/dt = F (1 - N_H - N_{H_2}) - p_H n_H - 2 \alpha N_H^2$$

$$d N_{H_2}/dt = \alpha \mu N_H^2 - p_{H_2} N_{H_2}$$

$p_H = \nu \exp(-E_H/kT)$ - desorption rate

$\alpha = \nu \exp(-E_d/kT)$ - diffusion+recomb. rate

Attempt frequency

Recombination rate

Biham et al. MNRAS, 296, 869 (1998)

Katz et al. ApJ 522, 305 (1999)

Biham et al. ApJ 553, 595 (2001)

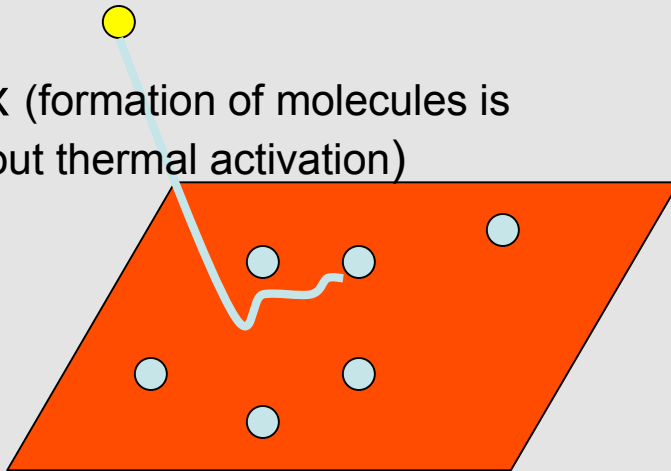
- $R(t) = (1 - \mu) \alpha N_H^2 + p_{H_2} N_{H_2}$

H_2 production rate

- $R(t) = (1 - \mu) \alpha N_H^2 + p_{H_2} N_{H_2}$ (single grain)
- Steady state conditions ($dN_{H, H_2}/dt=0$):
 - when coverage is high ($\alpha \gg p_H^2/F$; $p_H = 1/t_H$)
 - $R_{H_2} = 1/2 (n_H v_H \sigma \xi) n_g$ (rate/unit volume)
 - Indep. of H coverage - linear in flux
 - When coverage is low ($\alpha \ll p_H^2/F$)
 - $R_{H_2} = 1/2 (n_H v_H \sigma \xi t_H)^2 n_g \alpha \gamma$
 - Phenomenologically derived by Pirronello et al. ApJ 483, L131 (1997)
 - Quadratic in H coverage

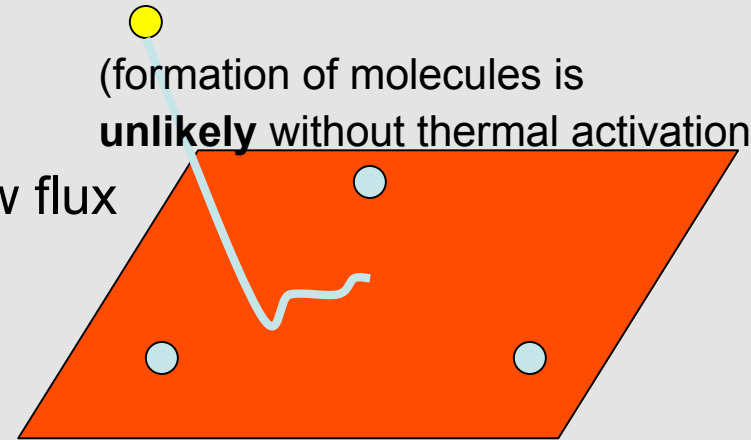
Effect of flux on the lab measurements

High flux (formation of molecules is likely without thermal activation)



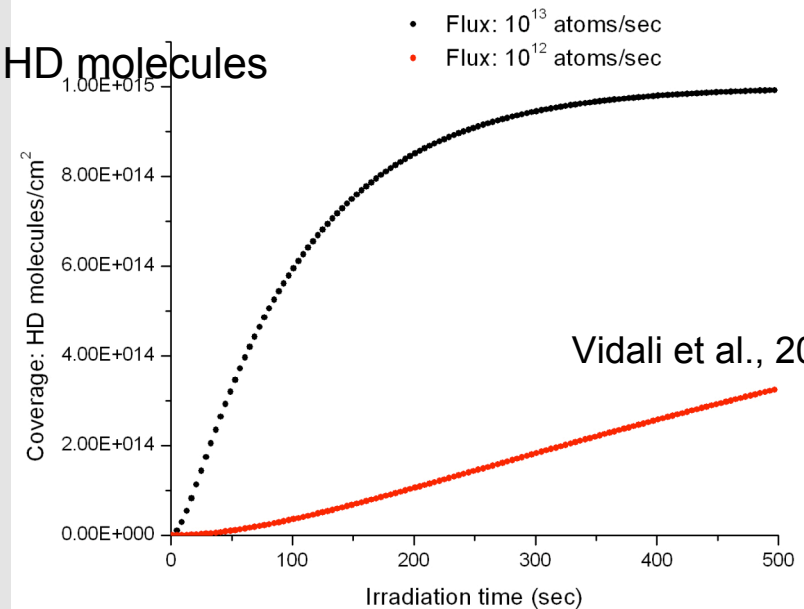
(formation of molecules is **unlikely** without thermal activation)

Low flux



Hot-atom mechanism (a possible explanation for the formation of HD in the experiments at high coverage on ice of Hornekaer et al., (2003) and the experiments at high coverage on olivine, Pirronello et al. (1997)) : atoms lands on surface and travel tens of Angstrom before coming to rest (as per MD simulations – Buch & Zhang Ap.J. '91; Takahashi & Uehara, Ap.J. 2001)

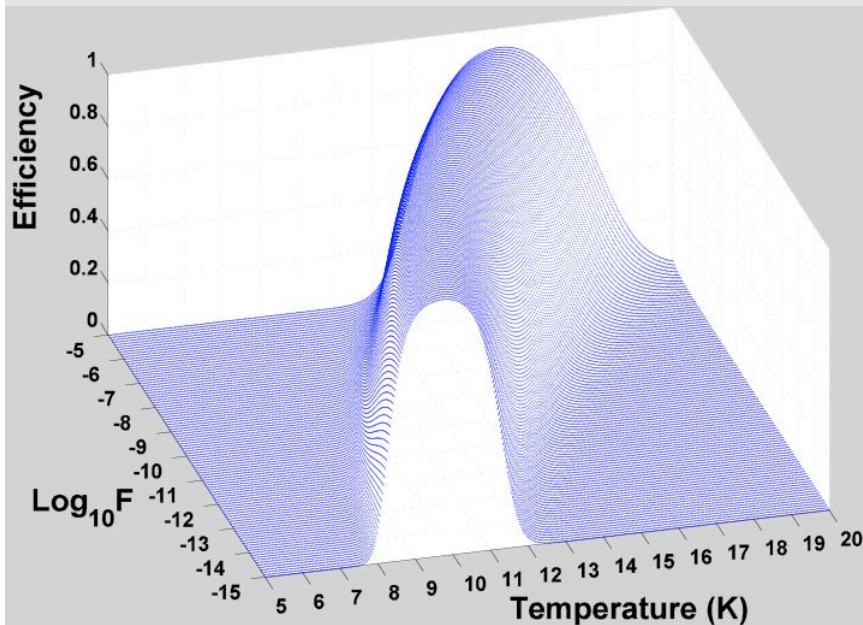
HD molecules



Typical irradiation time: ~ a few mins

From the laboratory to the ISM

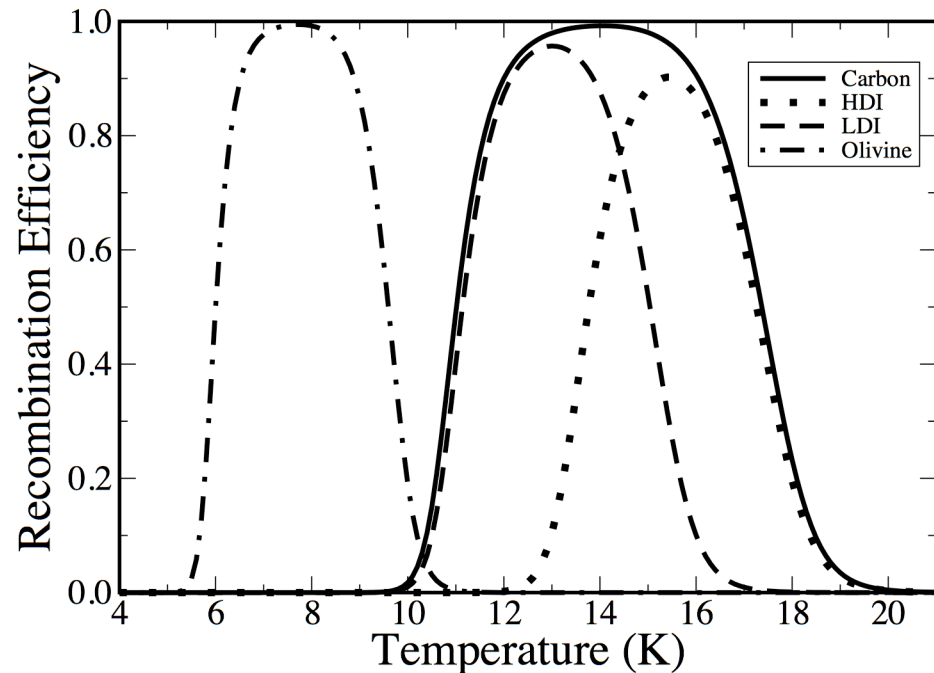
Simulations of recombination efficiency (probability of molecule formation when two atoms are on the surface) under ISM conditions



Katz et al. 1999

Simulations are based on laboratory data taken at S.U.

HDI, LDI= high and low density amorphous water ice



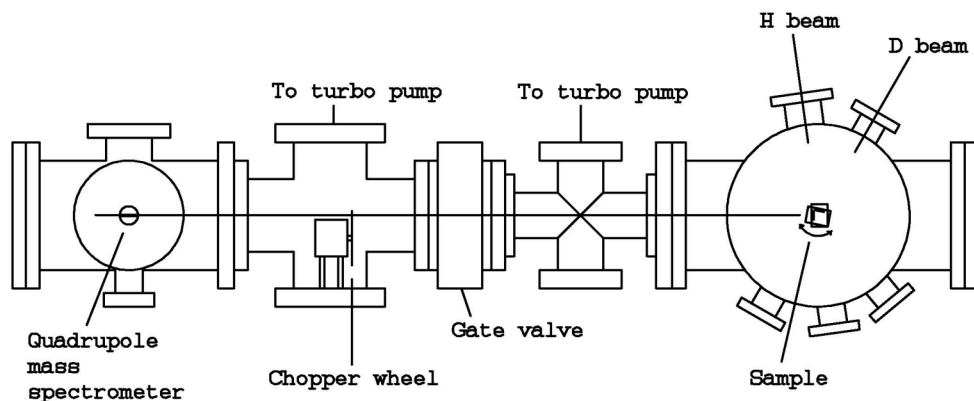
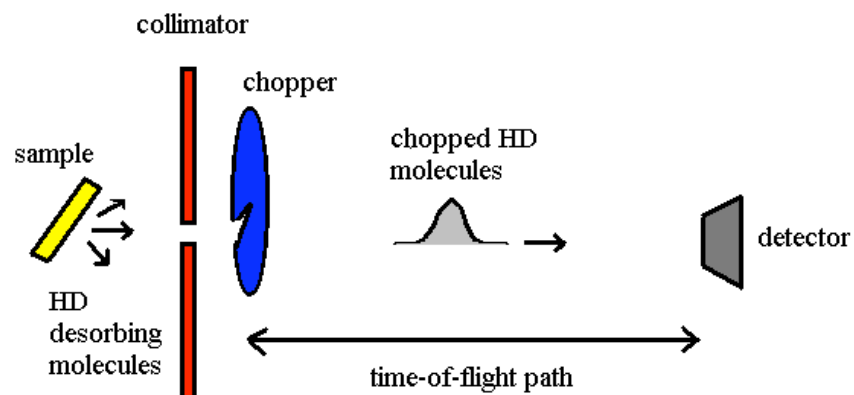
$n \sim 10 \text{ cm}^{-3}$ $T_{\text{gas}} \sim 100 \text{ K}$

Figure courtesy of Perets and Lipshtat

Measurement of kinetic energy

- Time-of-flight detection to measure the **translational** energy of molecules

Schematics of time-of-flight measurements



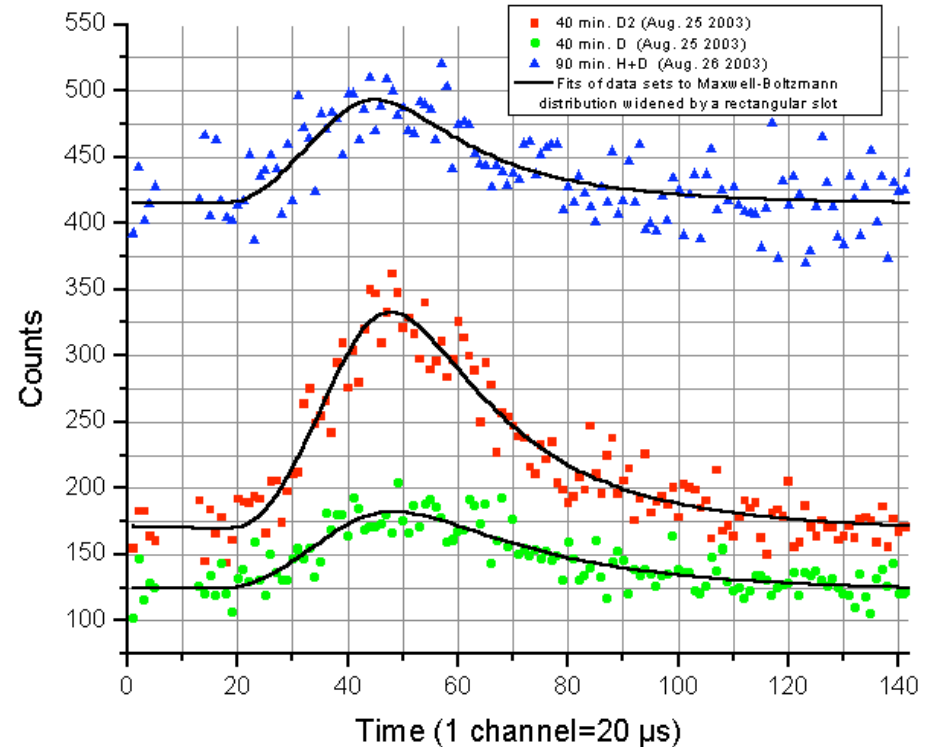
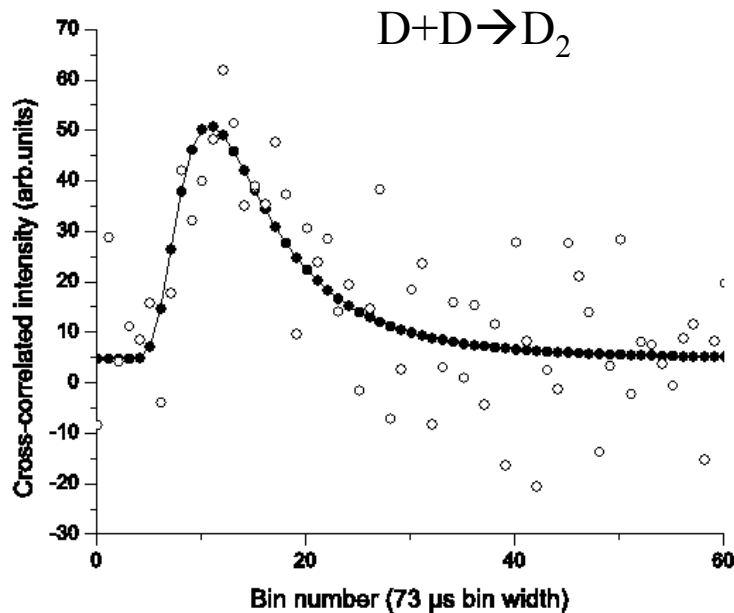
Time-of-flight data

$H+H \rightarrow H_2$ on amorphous ice

$$f(v)dv \sim v^2 e^{-mv^2/2kT} dv$$

$$f(t)dt \sim t^{-4} e^{-mL^2/2kTt^2} dt$$

$$E_{\text{kinetic}} = 2kT \quad T \sim 16 - 22 \text{ K}$$



Roser et al. ApJ 596, L55 (2003);
 J.Geophys.Res. (2004) J.Geophys.Res.
 Hornekaer et al. Science (2003)

Summary of results from experiments

- Efficiency of formation of H₂ on various dust grain analogues as a function of sample temperature under simulated ISM conditions
- Information obtained:
 - Mechanism: Langmuir-Hinshelwood, hot atom
 - Efficiency: high for amorphous carbon and amorphous water ices but in a rather narrow range of grain temperature
 - Energetics: diffusion and desorption energies
 - Kinetic energy of just-formed molecules
- Current and future directions:
 - More powerful theoretical tools (rate equations, master equation) to relate experimental quantities to conditions in ISM
 - Experiments probing H₂ formation on higher temperature surfaces

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Syracuse University: G.V., Joe Roser
University of Catania: Valerio Pirronello
Giulio Manico',
Giuseppe Raguni

University of Cagliari: Emmanuele Congiu
Hebrew University: Ofer Biham, Hagai Perets



Recent publications

- H.Perets, O.Biham, G.Manico', V.Pirronello, J.E.Roser, S.Swords, and G.Vidali: "Molecular Hydrogen Formation on Ice under Interstellar Conditions", *Astrophys.J.* 627, 850 (2005)
- G.Vidali, J.E.Roser, G.Manico', and V.Pirronello: "Laboratory Studies of Formation of Molecules on Dust Grain Analogues under ISM Conditions", *J. Geophys. Res.* 109, E07S14 (2004).
- G.Vidali, J.E.Roser, G.Manico', and V.Pirronello: "Laboratory Studies of Formation of Molecules on Dust Grain Analogues under ISM Conditions", *J. Geophysics Res.* 109, E07S14 (2004).
- V.Pirronello, G.Manico', J.E.Roser and G.Vidali: "H₂ Formation on Dust Grains", invited review in *Astrophysics of Dust*, ed. by A.Witt et al. (2003)p.529
- J.E.Roser, S.Swords, G.Vidali, G.Manico', and V.Pirronello: "Measurement of the kinetic energy of hydrogen molecules desorbing from amorphous water ice", *Astrophys.J.*, (2003) 595, L55.
- J.Roser, G.Manico', V.Pirronello, and G.Vidali: "Formation of Molecular Hydrogen on Amorphous Water Ice: Influence of Morphology and Ultraviolet Exposure" *Astrophys.J.* 581, 276 (2002).
- J. Roser, G.Vidali, G. Manico', and V. Pirronello: "Formation of Carbon Dioxide by Surface Reactions on Ices in the Interstellar Medium" *Astrophys.J.* 555, L61 (2001).
- O. Biham, I. Furman, V. Pirronello, and G. Vidali: "Master Equation for Hydrogen Recombination on Grain Surfaces", *Astrophys.J.* 553, 595 (2001).
- G. Manico', G. Raguni', V. Pirronello, J.E. Roser, and G. Vidali: "Laboratory Measurements of Molecular Hydrogen Formation on Amorphous Water", *Astrophys.J.* 548 L253 (2001).
- N.Katz, I.Furman, O.Biham, V.Pirronello, and G.Vidali : "Molecular Hydrogen Formation on Astrophysically Relevant Surfaces", *Astrophys.J.*, 522, 305 (1999).
- V.Pirronello, C.Liu, J.Roser, and G.Vidali:"Measurement of Molecular Hydrogen Formation on Carbonaceous Grains", 1999, *Astron.&Astrophys.* 344, 681.
- O.Biham, I.Furman, N.Katz, V.Pirronello, and G.Vidali: "H₂ Formation on Interstellar Grains in Different Physical Regimes", *MNRAS*, 296, 869 (1998).
- V.Pirronello, O.Biham, C.Liu, L.Shen, and G.Vidali: "Efficiency of Molecular Hydrogen Formation on Silicates", *Astrophys. J.* 483, L131 (1997).
- V.Pirronello, C.Liu, L.Shen, and G.Vidali: "Laboratory Synthesis of Molecular Hydrogen on Surfaces of Astrophysical Interest", *Astrophys. J.* 475, L69 (1997)