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The formation of H_2: experiments on an amorphous silicate surface

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2 LASSIE

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Index

A brief timeline: experiments

$H_{gas} + H_{grain} \rightarrow H_2\,(v'', J'')_{gas}$ + K.E. + grain heating

- HD on polycrystalline silicates (Pirronello et al. 1997): efficient up to $T_{dust} < 10$ K.
- HD on amorphous carbon (Pirronello et al. 1999).
- HD on amorphous water ice (Manico et al. 2001).
- HD on amorphous ice at 8K (Hornekaer et al. 2003).
- HD on amorphous silicate (Perets, 2007): efficient up to $T_{dust} < 14$ K.
- HD formed on graphite held at 15 K (Latimer et al 2008).
- D_2 formed on silicate held at 10 K efficient to $T_{dust} < 70$ K. (Lemaire & Vidali 2010).

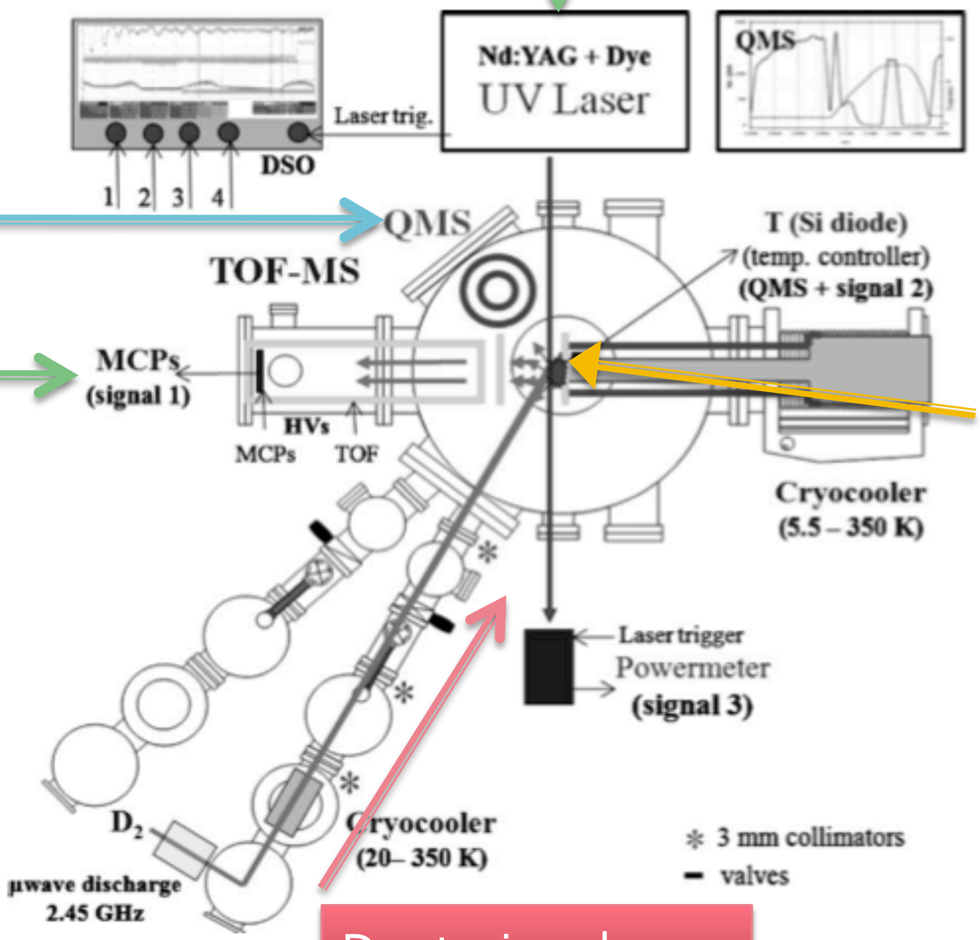
The experimental setup

UV Laser (220 nm)

QMS

REMPI-TOF

Silicate sample



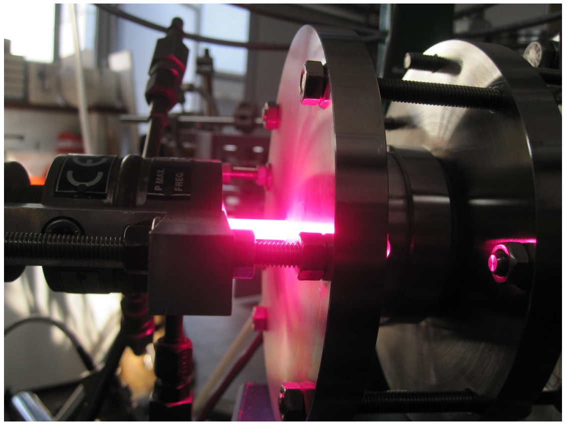
Deuterium beam

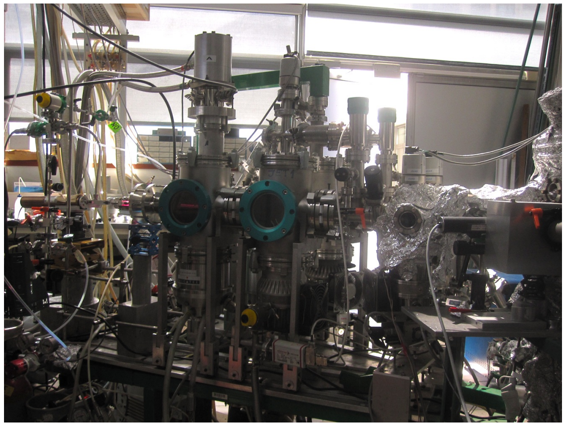
View from top

(Lemaire & Vidali, 2010)

The atomic beam

- We start with D_2.
- A microwave discharge gives a dissociation rate up to 70%.
- D atoms are sent towards our sample via a triple differentially pumped beam and reach the UHV chamber at 10^{-10} mbar.
- The surface is seen at an angle of ~41° to the beam, allowing detection of 41/180 of the molecules and a density of ~2.9×10^7 molecules cm^{-3} in all states.
- T_{beam} = 20 - 300 K

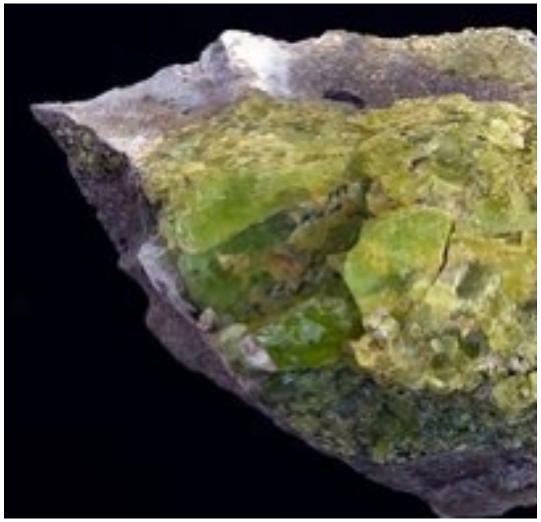




FORMOLISM @ Cergy-Pontoise

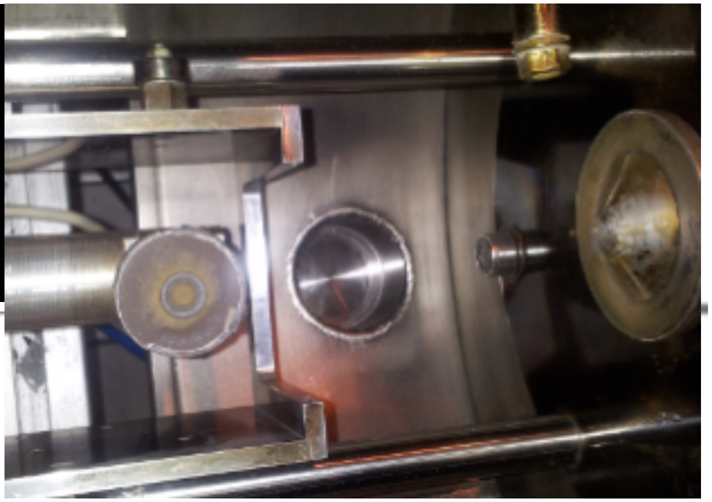
The sample

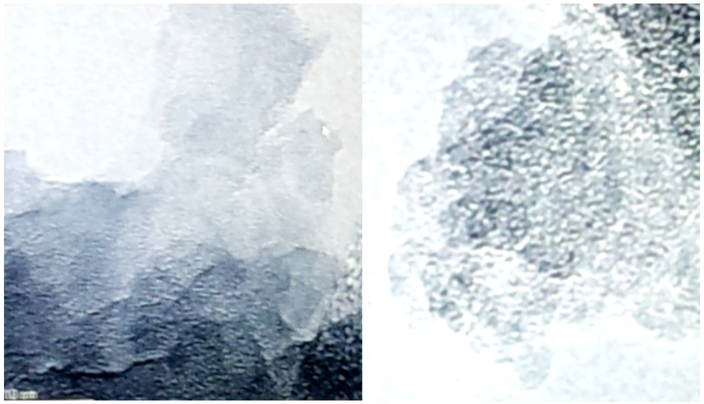
- We use an amorphous silicate $(Fe_x, Mg_{1-x})_2SiO_4$. A "rough" surface prepared from naturally occurring San Carlos Olivine.
- Chosen because it has deeper adsorption sites than crystalline silicates (Perets et al. 2005), which may enhance formation.
- T_{sample} = 5 – 300 K.

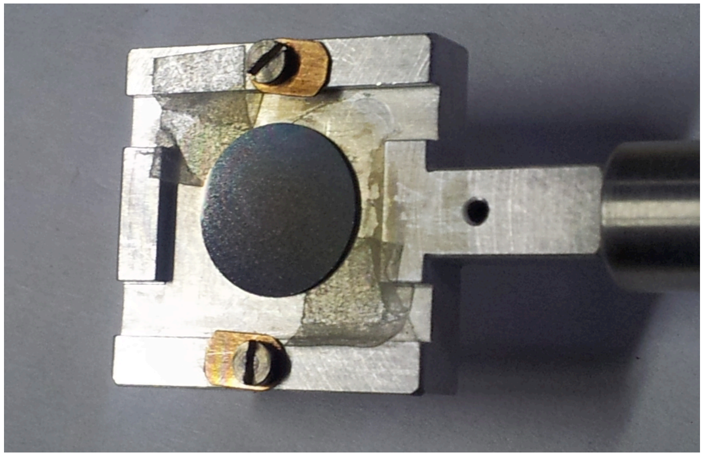


Mg Fe silicate (Arizona, San Carlos Apache Reservation)

The sample



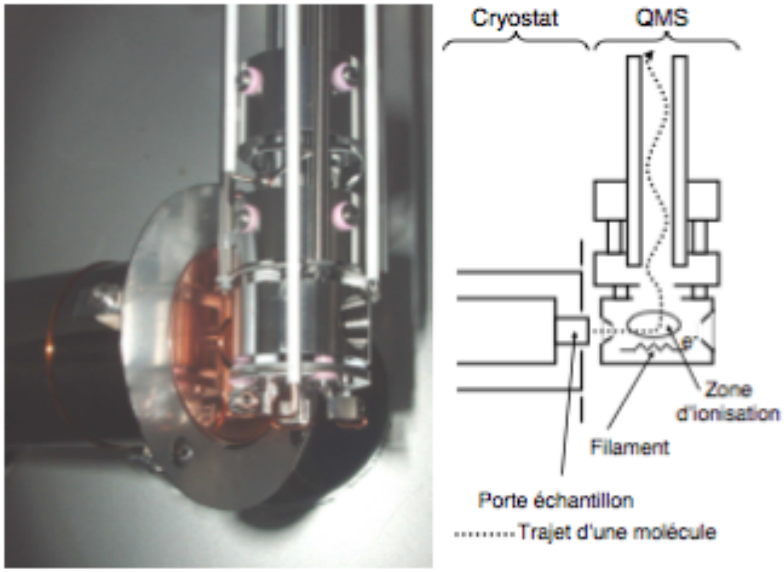




- Also, Forsterite (Mg_2SiO_4) and Fayallite (Fe_2SiO_4) samples have been prepared by laser ablation on Si disks in collaboration with the Jena astrophysical institute (Germany).
- We will have one amorphous and one crystalline sample of each type (annealed to > 750 C).
- Diagnostics: TEM, EDX, IR.
- The goal is to study the chemisorption effects due to surface morphology and composition on H_2 formation.

The detectors: QMS (Quadrupole mass spectrometer)

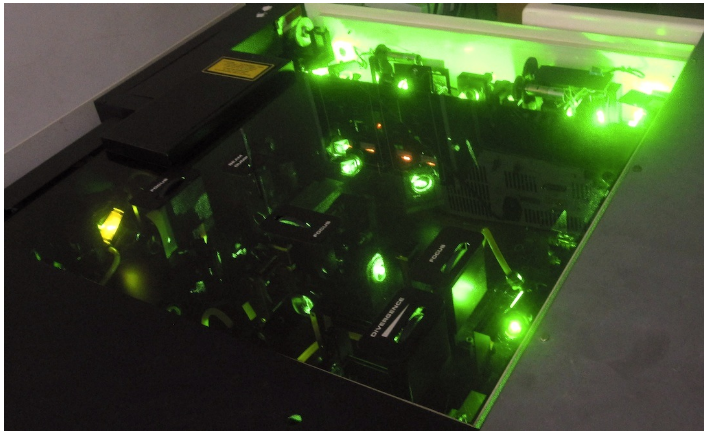
- This QMS can measure masses between 1 and 50 amu.
- It gives a good idea on the absolute sticking of atoms and molecules, but as it only distinguishes across masses it's not a reliable method to study formation.
- We use the King and Wells Method, in which the QMS measures the average density of particles in the gas phase in the main chamber.



Amiaud, 2006

The detectors: REMPI-TOF (Resonantly Enhanced Multiphoton Ionization)

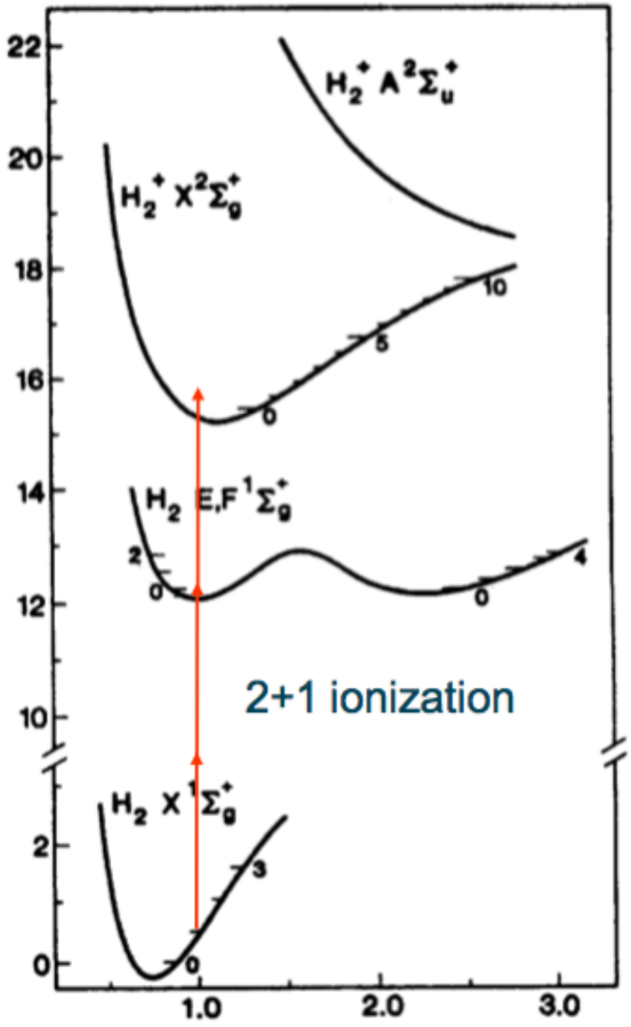
- REMPI is a quantum selective detector.
- REMPI (2+1) works with 3 photons at 220 nm. 2 are used used to excite and one to ionize formed D_2 at v''=4, J''=2.
- 1 mJ pulsed laser (20 Hz).
- The laser is calibrated in an annex setup at higher D_2 pressure.



Nd YAG laser @ Cergy

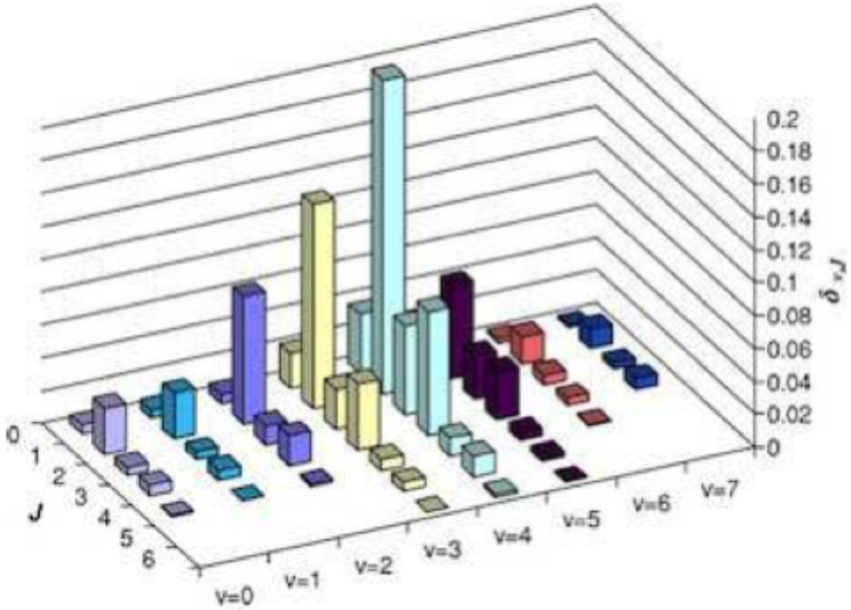
Internal excitation of nascent H_2

- REMPI (2+1): H_2^+ (E, F state)
- Beam is cooled down so that D atoms are in rovibrationally grounded, and only newly recombined D_2^* is detected.



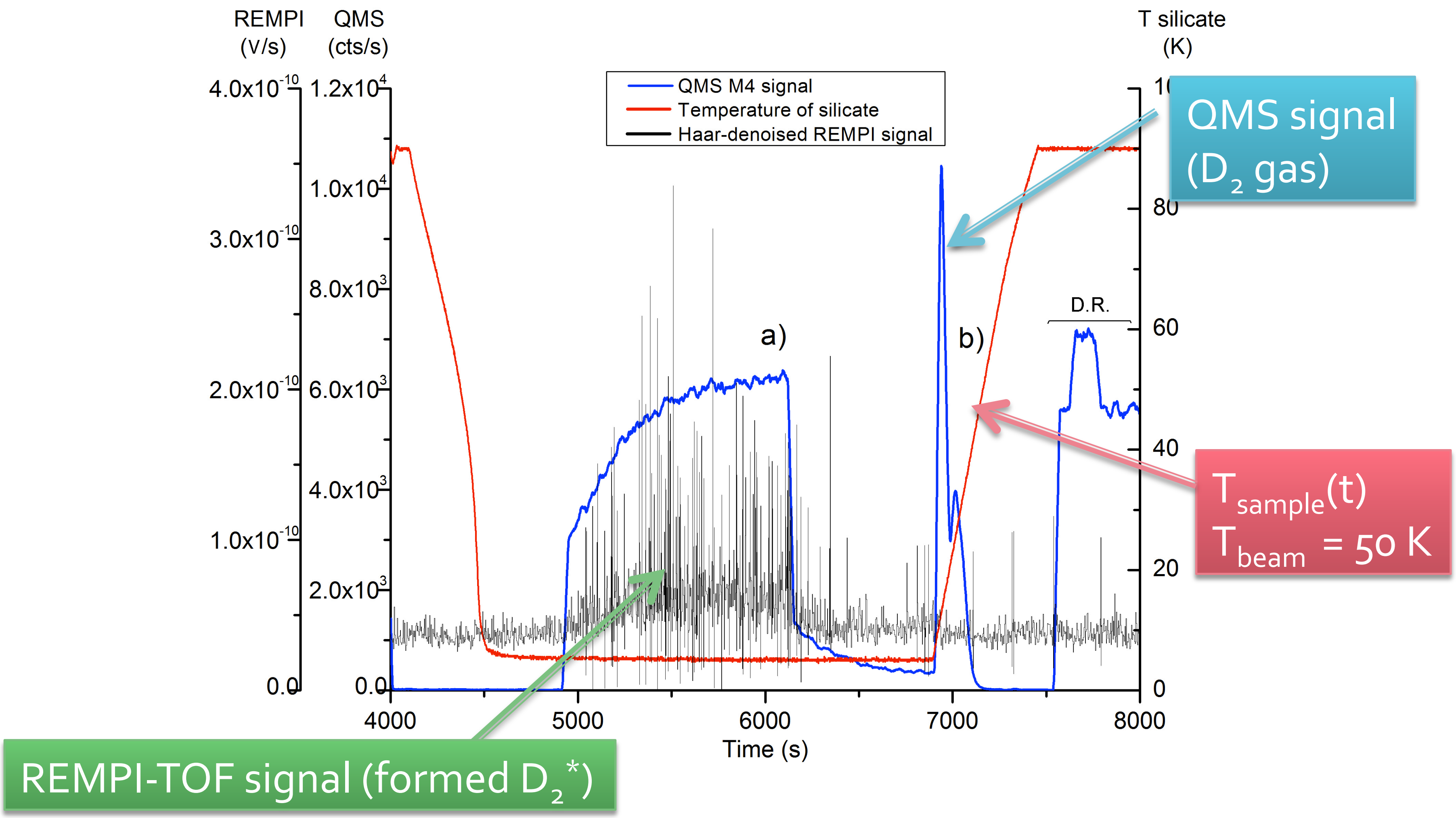
Internal excitation of nascent H_2

- For REMPI, the photon wavelength is tuned so that it ionizes only the v″=4, J″=2 level, predicted to be the most populated rovibrational level after formation.
- It's also a convenient state because of its Frank Condon factor.



Rovibrational distribution of molecular hydrogen after formation extrapolated from HD at 15 K (Creighan 2006, Islam 2010).

Experiments: T_{beam} effect on formation



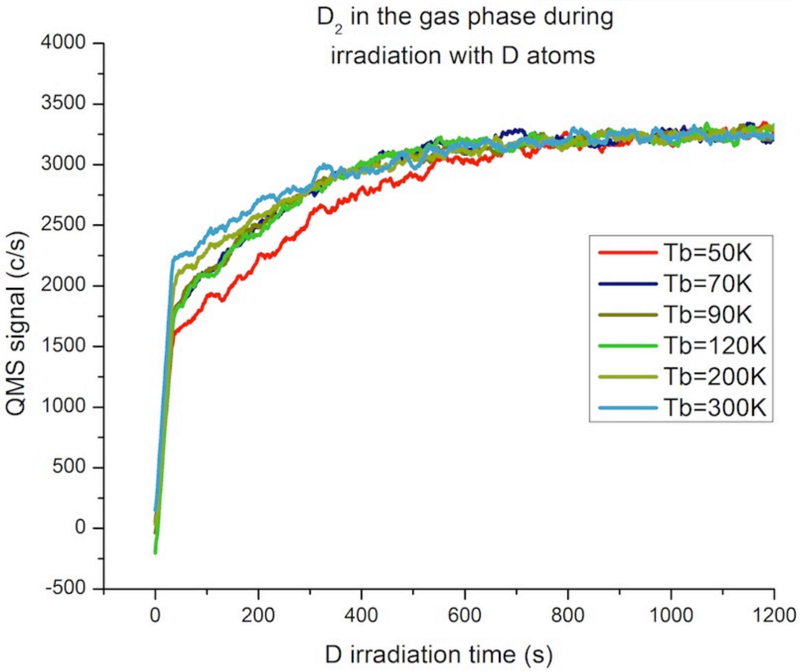
QMS signal (D_2 gas)

T_{sample}(t)
T_{beam} = 50 K

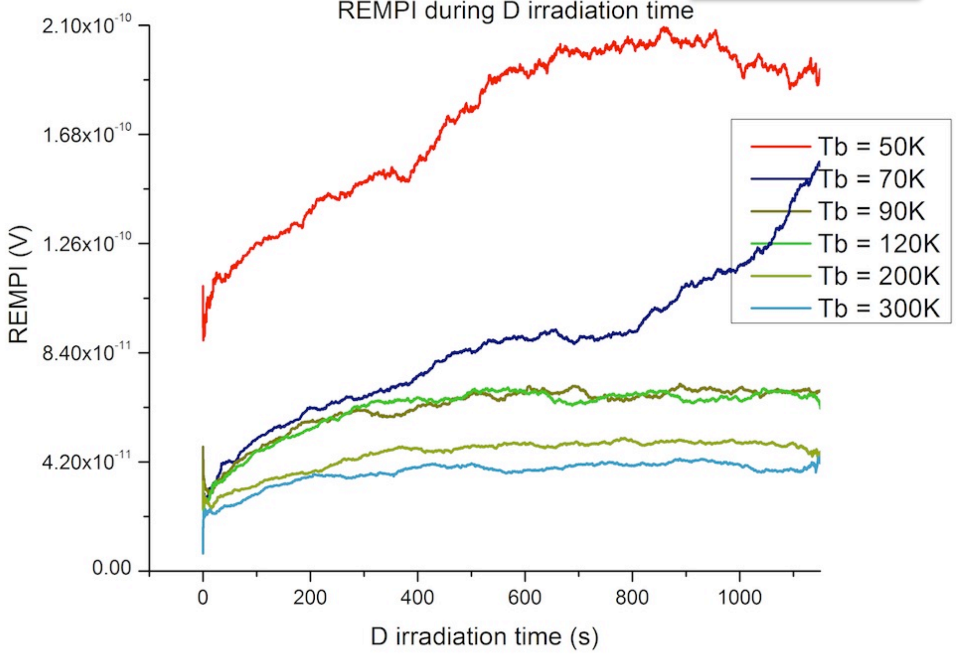
REMPI-TOF signal (formed D_2^*)

Experiments: T_{beam} effect on formation

REMPI signal

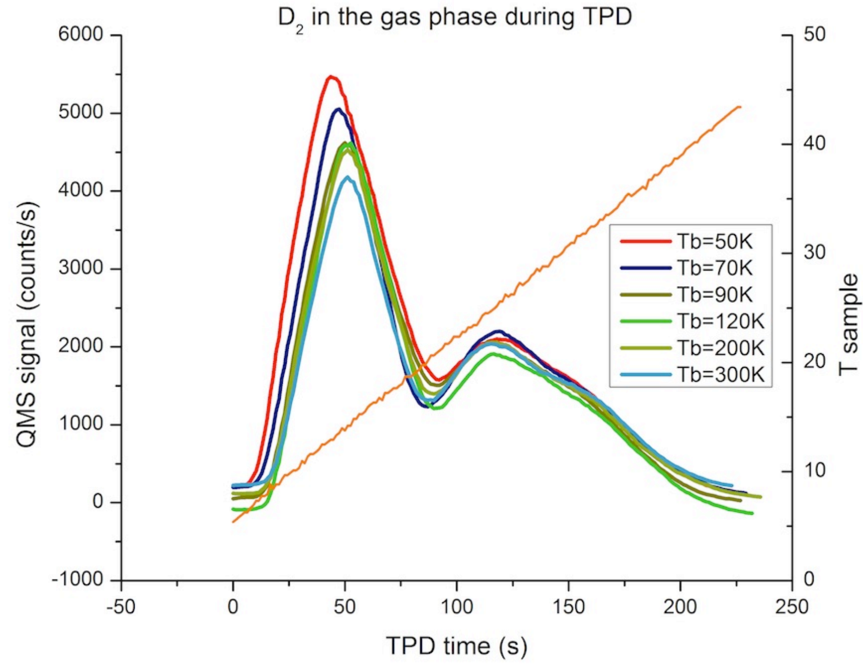


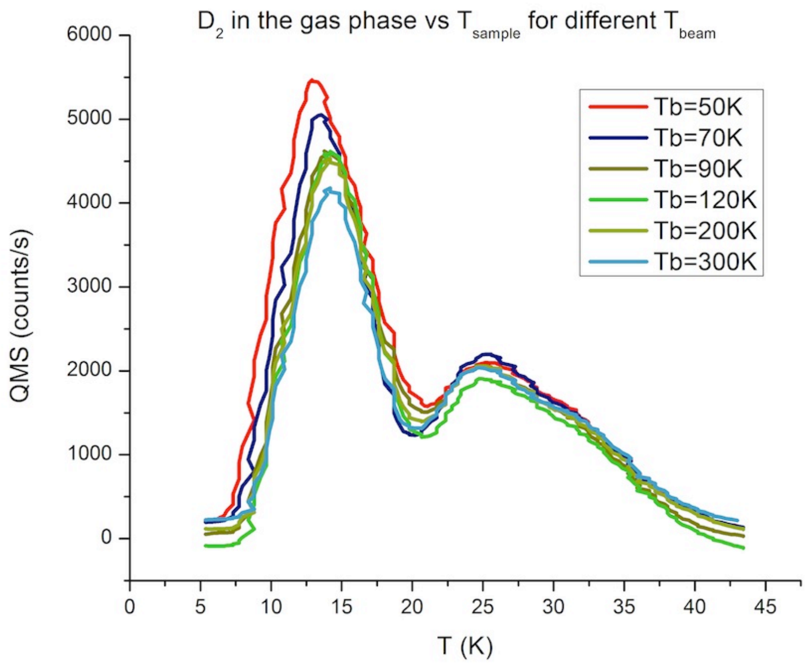
QMS signal



- QMS: largest D sticking from Tb = 50K, but slowest increase until recombination/sticking steady state. Growth curve may be dominated by flux of undissociated D_2 (greatest at lower T_b).
- REMPI: recombination during this time enhanced for Tb = 50K. Unlike the QMS, it discriminates the formed D_2 from undissociated D_2.

Experiments: T_{beam} effect on formation

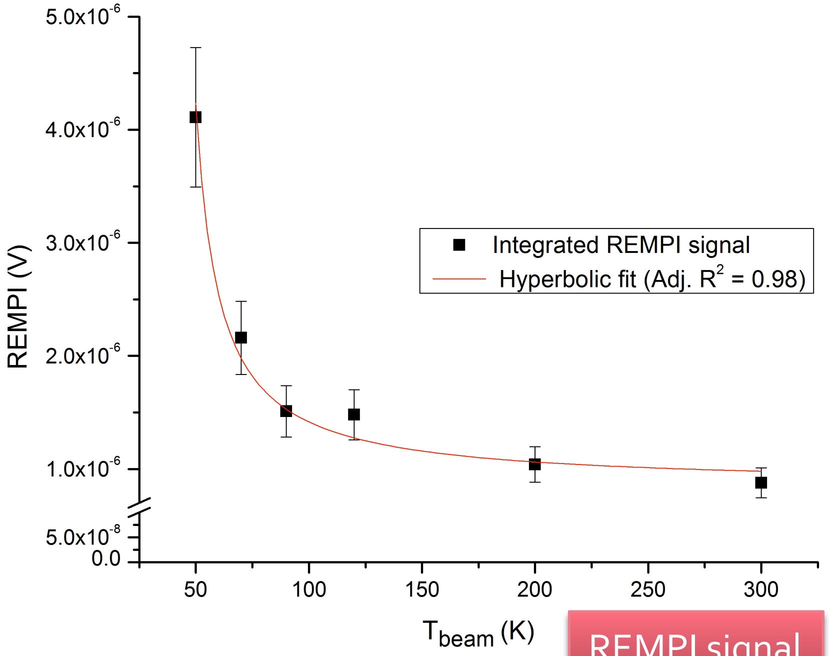

QMS signal


QMS signal

- QMS: TPD curves showing the largest sticking of D2 (recombined and undissociated) was at Tb = 50K. TPD is desorbing the particles that were more strongly bounded to the surface, and it confirms what D atoms have the largest sticking on silicate for colder beam, i.e. Tb = 50K.
- QMS: TPD curves vs T sample can be analyzed by desorption models.

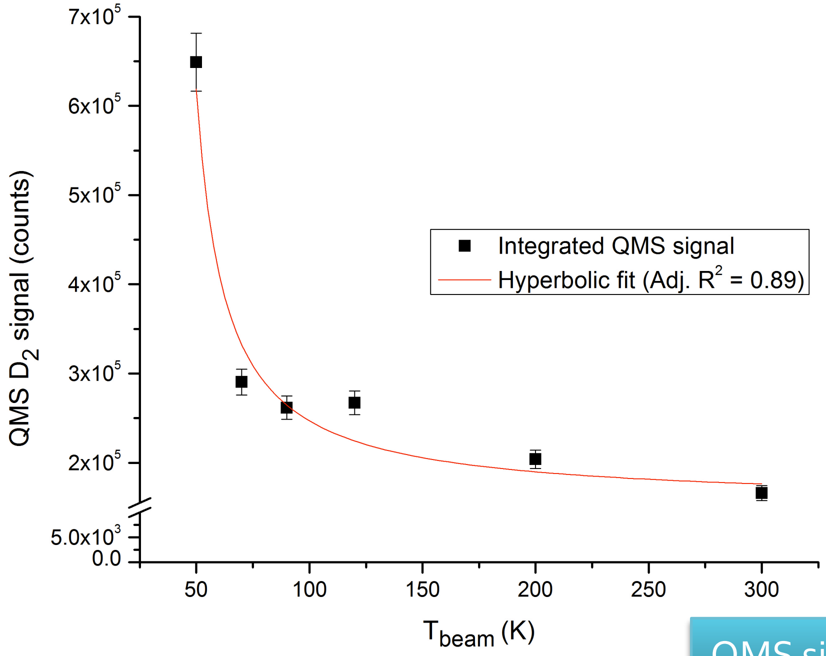
Experiments: T_{beam} effect on formation

D irradiation interval



REMPI signal

TPD interval

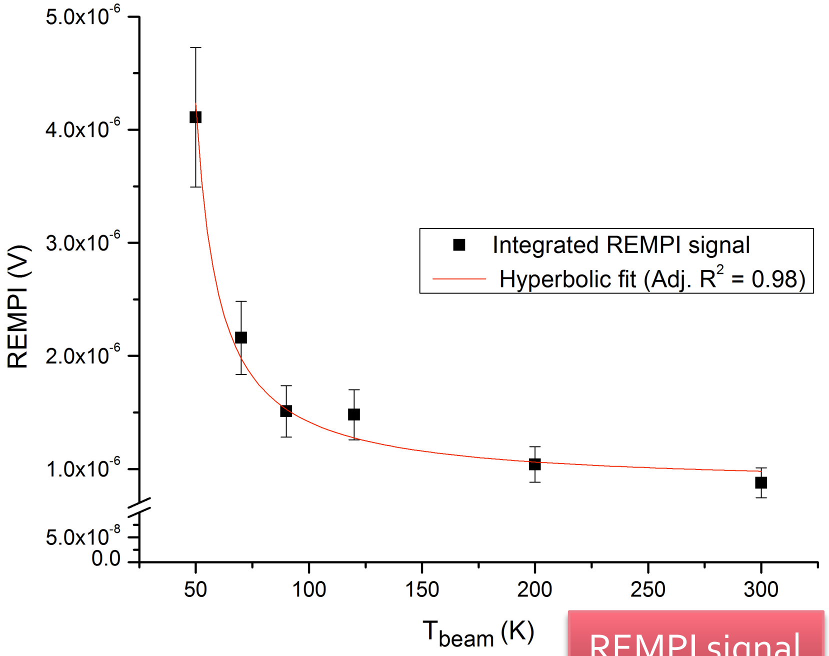


QMS signal

- REMPI area during D irradiation: Lower beam temperatures enhance formation.
- QMS area during TPD: Lower beam temperatures enhance sticking of D atoms on the surface.
- **We confirm the positive correlation between sticking and D_2 recombination for T_{sample} = 5K and T_{beam} = 50-300K, enhanced at lower gas temperatures.**

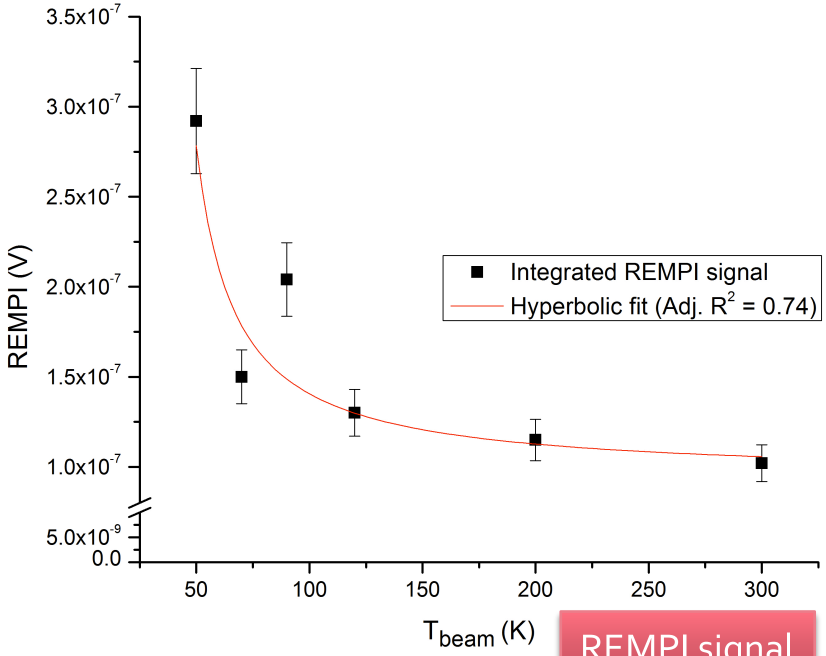
Experiments: T_{beam} effect on formation

D irradiation interval



REMPI signal

TPD interval

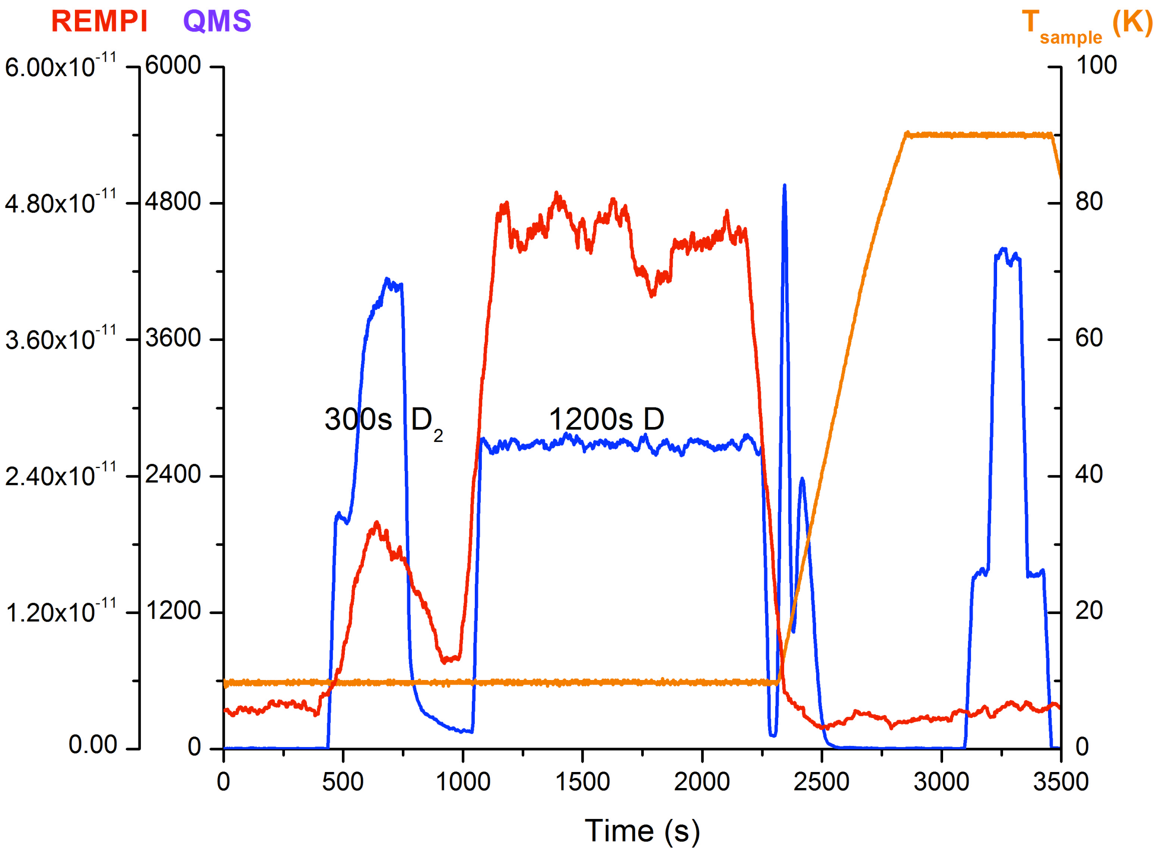


REMPI signal

- REMPI area during TPD: formation is activated by sample heating. The heating ramp helps surface D atoms overcome the diffusion barrier, allowing them to recombine
- Notice the REMPI signal is an order of magnitude weaker here than during D irradiation.

Experiments: D_2 pre-coverage effect on formation

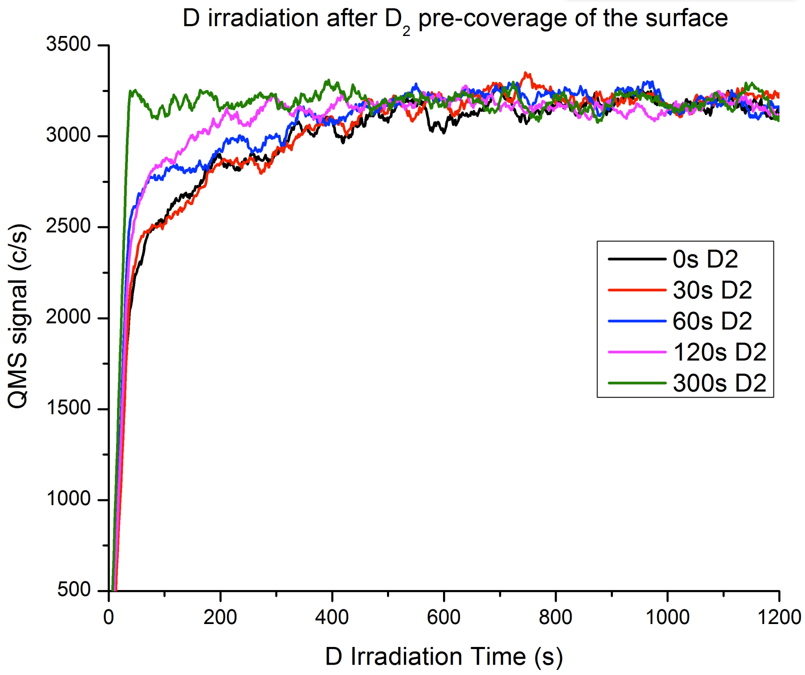
-D2 or H2 on the surface can modify the process of accretion or diffusion (Govers et al., 1980; Tielens, 2005).

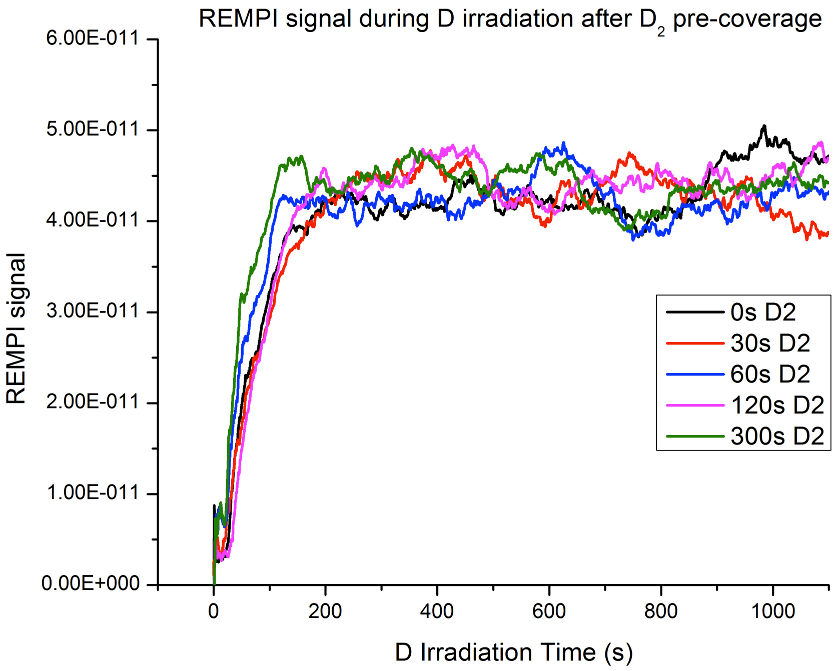


Experiments: D_2 pre-coverage effect on formation

QMS signal

REMPI signal

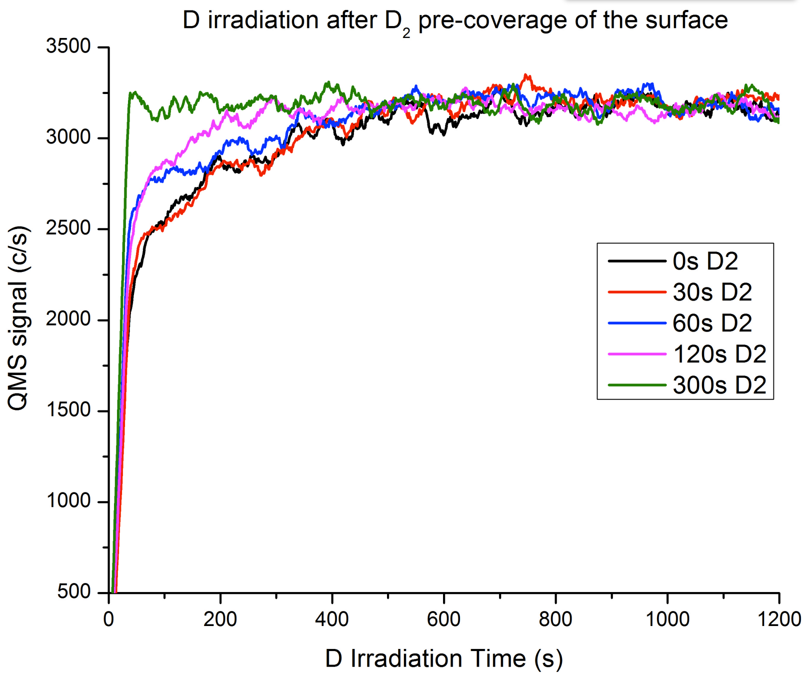

D irradiation after D_2 pre-coverage of the surface


REMPI signal during D irradiation after D_2 pre-coverage

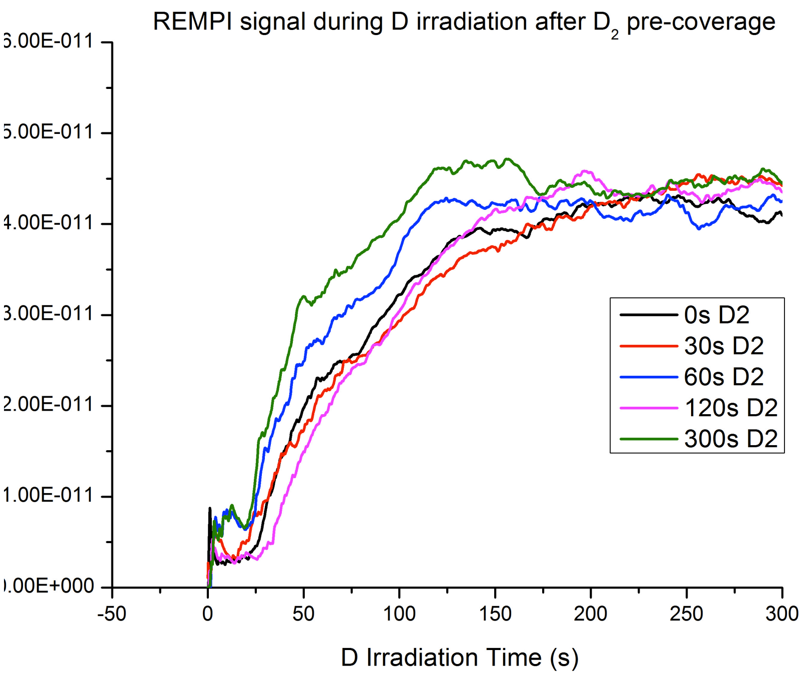
- QMS: growth curve depends on pre-coverage of D2. Surface pre-coverage with 300s D2 reaches a recombination steady state faster.
- REMPI: the REMPI signal confirms this being greatest for 300s of D2 pre-coverage.

Experiments: D_2 pre-coverage effect on formation

QMS signal



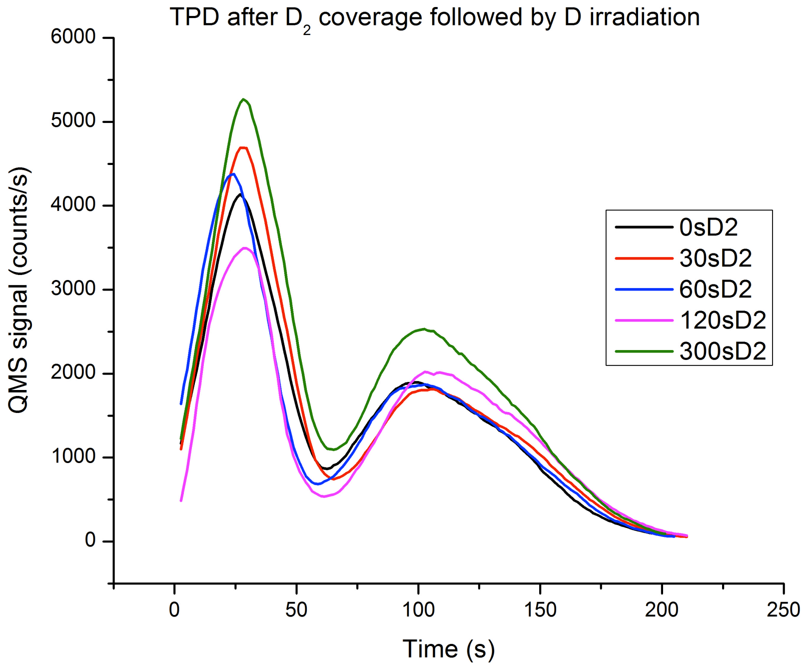
REMPI signal



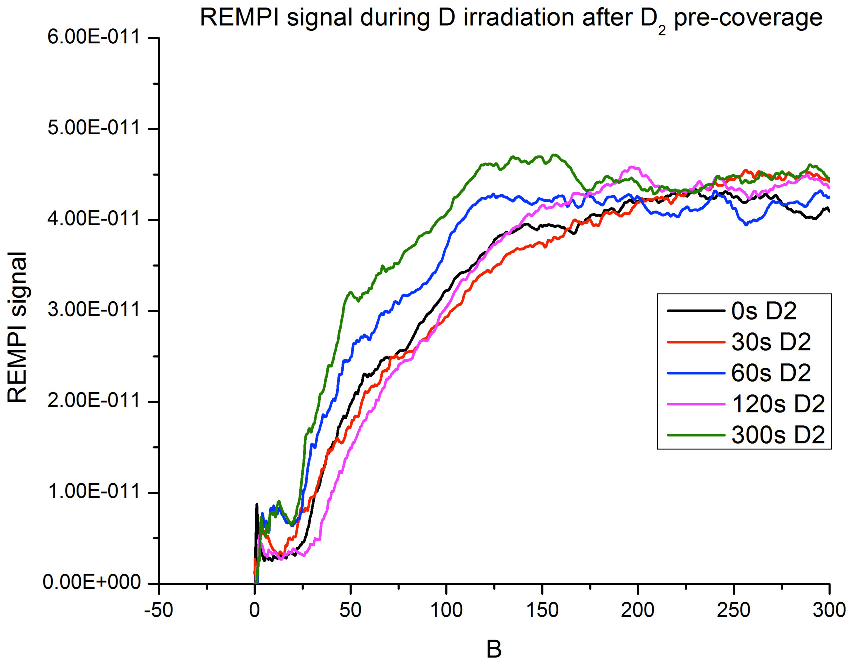
- REMPI: The effect of D_2 coverage seems to be most prominent during the first 170s of irradiation with D atoms.
- After this, the REMPI curves converge with a more homogenous coverage (both D and D2).

Experiments: D_2 pre-coverage effect on formation

QMS signal



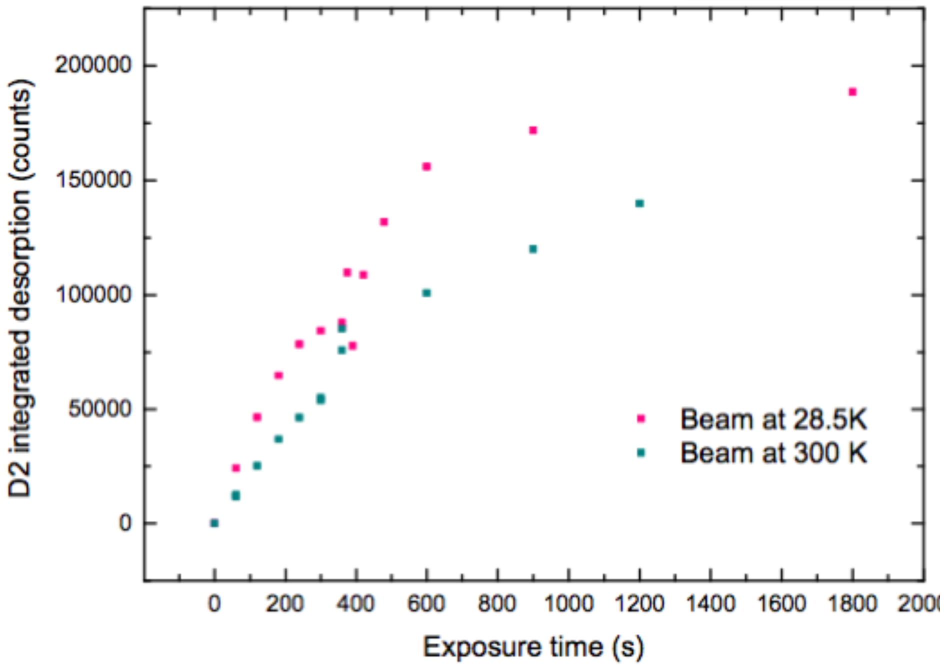
REMPI signal



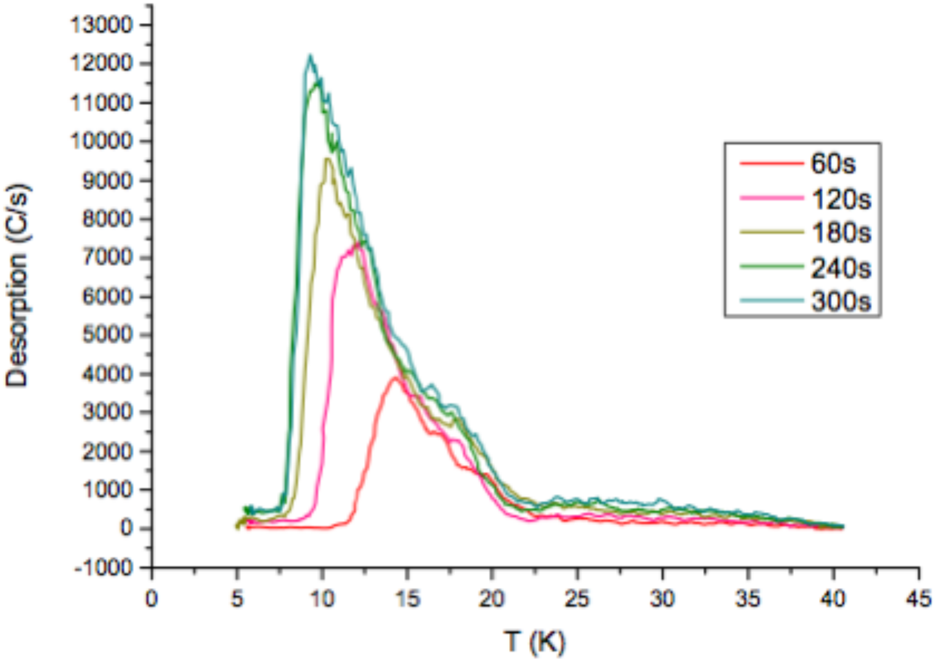
- QMS: TPD curves show that coverage was non-linear with respect to irradiation dose on the surface. This is mostly affected by uneven waiting after D2 dosage before D irradiation. D2 is very volatile on the surface at 5K.
- REMPI: The effect of D_2 coverage on REMPI is correlated to the TPD areas.

Experiments: D_2 pre-coverage effect on formation

QMS signal



QMS signal



- Previous work on the sticking of D2 molecules found that the evaporation flux at a surface temperature of 5.5K is not negligible.
- It is impossible to build a D_2 multilayer even at 5K.

Conclusions: formation mechanisms

- These new experiments probe T_{dust} = 5K (fixed) and T_{beam} = 50-300K ranges.
- We have focused on the effects of dynamical coverage of the surface on formation. We found that:
  - D_2 coverage enhances formation so D_2 formation is self-regulating.
  - Lower T_{beam} enhances formation as longer residence time enhances sticking.
- Hybrid mechanisms depend on the current coverage with D and D_2 on the surface.
  - When the grain is bare, at low coverage, few D atoms chemisorb on the surface. Initial irradiation causes a burst in formation for all Tbeam. Eley-Rideal dominates.
  - Greater surface coverage with D and D2 can lower barriers against diffusion, i.e. strong silicate binding sites are filled, so D atoms become more mobile and able to recombine. Langmuir-Hinshelwood dominates.

Conclusions: formation mechanisms

- Experimentally, the QMS is a detector that will give us only a partial view of the formation process.
- The QMS signal enlightens the absolute sticking and its evolution.
- However, since it only distinguishes across masses, and given the limitation in our dissociation source, it doesn't uncouple the signal D_2 due to recombination or from D_2 from the jet.

- REMPI-TOF has the great advantage of a quantum selective detector, profiting from the ro-vibrational excitation of newly formed D_2 on the dust.
- Its main disadvantages are the time-consuming wavelength tuning, variations of laser power from heating of the optics, and random drifts in the detector.