H_2 Formation in the ISM: Deuterium atom physisorption and recombination on cold silicate dust grains

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I. Introduction

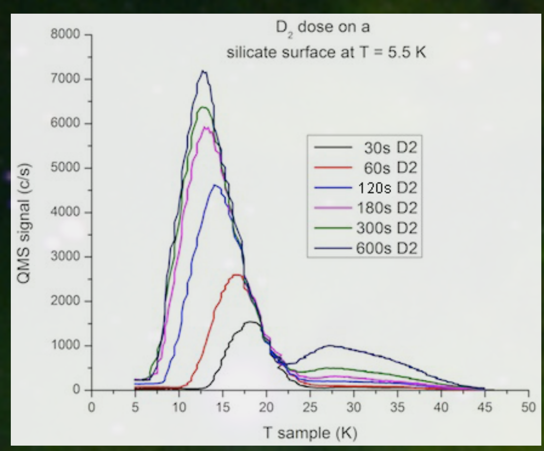
H_2 is the most abundant molecule in the universe and a significant fraction of its baryonic mass. H_2 formation occurs in the gas phase at high redshift and on grains in the local universe. The exothermicity of this reaction affects the heating and cooling balance in clouds, constraining collapse timescales and thereby affecting the local ISM chemistry and dynamics. We study this physical phenomenon in the laboratory by simulating ISM conditions such as dust and gas at cryogenic temperatures. Multiple techniques from surface science and non-linear optics are employed to enlighten the physical conditions that give rise to H_2 formation.

II. The Experiments

FORMOLISM (Formation of Molecules in the ISM) is an experimental setup developed at the Université de Cergy-Pontoise (NW of Paris). This setup includes a triply differentially pumped D_2 jet dissociated by a microwave discharge into mostly D atoms which are then cooled down to 50K. A UV laser is aligned so that the focal waist is 4mm away from the silicate and ionizes the formation-pumped D_2 molecules via REMPI (2+1). The resulting D_2* ions are extracted by a TOF spectrometer.

IV. Results & Discussion

The TPD of D_2 doses gives us an energetic distribution of the D_2 pre-coverage (fig. 3). We use the number of counts at these TPD peaks as an estimate of the total number of particles on the surface after the D_2 dose and after irradiation with D atoms on this dose. We assume that a superposition of TPDs would be the result of no reactivity between the D and the pre-covered surface. We compare this theoretical TPD to the actual TPD obtained after D_2 + D/D_2 irradiation. This gives us an idea of the destruction due to the exothermicity of the D_2 recombination process (4.48 eV) and a comparison to the formation efficiency on a bare silicate surface.



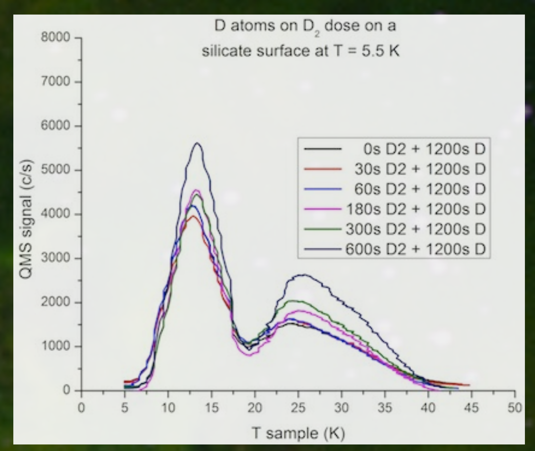


Figure 3. (Left) QMS signal during the TPD of D_2 doses at T_s = 5.5K. TPDs of order n = 1 with a peak binding energy distribution E_{bin} = 51 to 36 meV for 30 s to 600 s D_2.

Figure 4. (Right) QMS signal during TPD after irradiation of D_2 doses with D atoms, n = 1 order with peak E_{bin} = 35 meV.

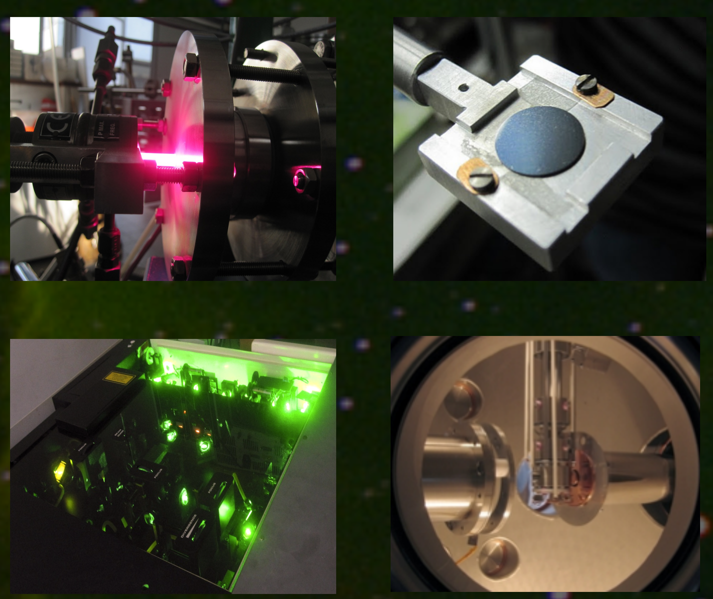


Figure 1 (CW). Microwave discharge, silicate sample, QMS and cryo-cooler, UV laser.

Abstract

We investigate the effect of a silicate surface pre-covered by D_2 molecules at T_s = 5.5, 10, and 16K on D_2 formation. We employ the UHV *FORMOLISM* setup that allows us to emulate a variety of astrophysically relevant ranges (5K < T_{dust} < 90K and 50K < T_{gas} < 300K). A QMS monitors the dynamic sticking on the dust surface while a REMPI-TOF spectrometer detects the newly formed D_2 molecules (excited at v" = 4, J" = 2). We find a positive correlation between D_2 sub-monolayer doses on an amorphous silicate surface and D_2 formation, which is enhanced up to 50% towards pre-saturation of the surface with D_2 molecules.

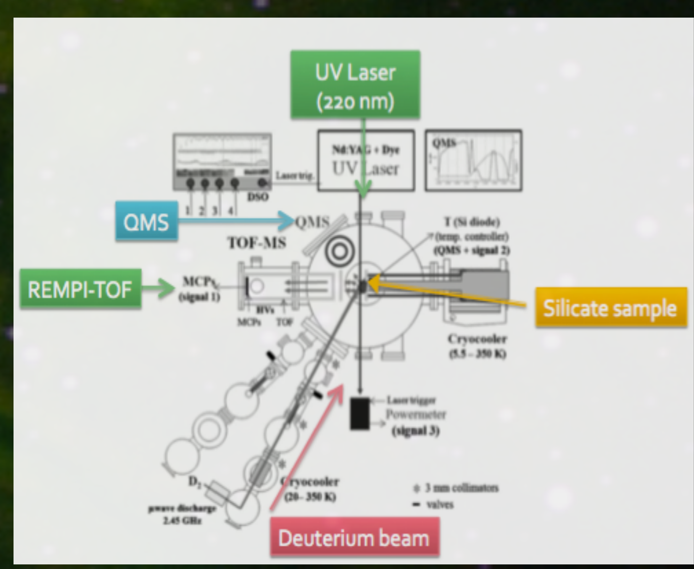


Figure 5. FORMOLISM design: D beam, QMS and REMPI detectors.

Methods: We devise a series of experiments planned to understand the effect of D_2 sub-monolayer coverage on D_2 formation. We begin by irradiating our silicate as soon as it reaches 5.5K with a dose of D_2 molecules followed by irradiation with a jet of D atoms (T_{beam} = 90K). Both irradiations are followed by a TPD to 90K at 10K/min. The dissociation rate of the microwave discharge, the flux, and the UV laser power are permanently recorded. A calibration with the same doses of D_2 to characterize the D_2 pre-coverage are done separately right after each atomic exposure experiment.

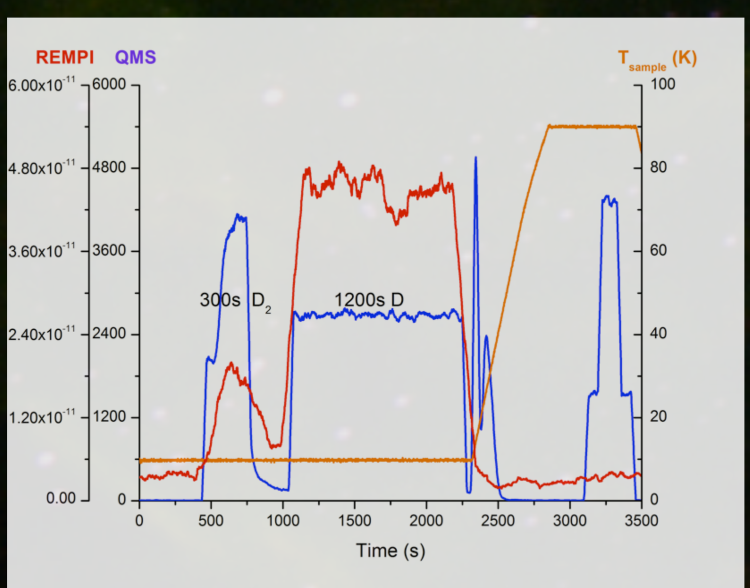


Figure 2. Data for one sample experiment carried out at a fixed beam temperature of 90K.

We begin with a 300s D_2 dose at T_{beam} = 90K on a silicate that is cooled down to $T_{surface}$ = 10K (notice how at this silicate temperature the dose saturates around 120s).

We close the valve and wait for the evaporation to reach approximately 0 counts/s and then proceed to irradiate this pre-coverage with cold atoms (D/D_2 irradiation) for 1200s at a dissociation rate of 70%.

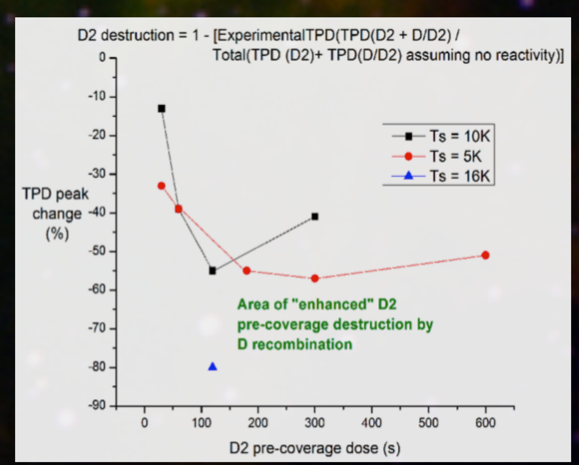
III. The Data

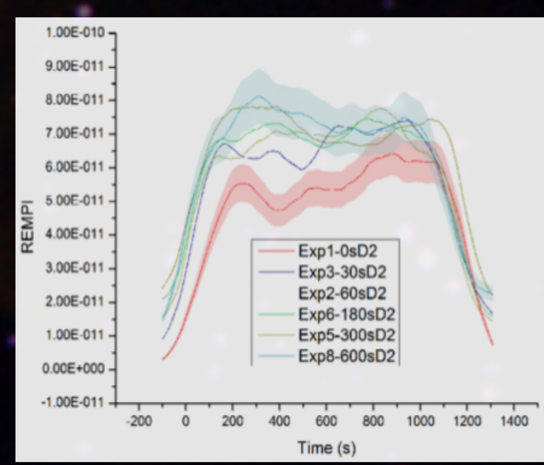
The reduction of the QMS signals and REMPI-TOF signals was done at specific intervals. Interval a) corresponds to the D_2 dose, interval b) corresponds to the D irradiation done after a certain evaporation wait, interval c) is the TPD right after a) and b), but is also done separately after a), to calibrate the doses of D_2 molecules on the bare silicate and characterize the new binding sites available for D_2 recombination. The REMPI signal has a quadratic dependence on the number of photons (laser power) and an approximate linear dependence on the flux of D atoms; the original signal was carefully normalized by fluctuations on both parameters. We smoothed the original signal very strongly in order to visualize its positive fluctuations over the noise. The REMPI signal has a larger error bar (10%) than the QMS signal (2%).

QMS and REMPI signals: The TPD analysis shows us that the percent destruction due to D_2 recombination increases with greater coverage but decreases slightly for maximum coverage for both T_s = 5.5K and T_s = 10K. Towards this coverage the thermal accommodation of the impinging D atoms is diminished by the surface saturation with D_2 molecules. In some silicate areas, there is thermal accommodation of the impinging D atoms on the silicate surface by the D_2 molecules but on others the D_2 will encounter mostly D_2 and not the silicate surface. Towards saturation most binding sites may be occupied by the monolayer of D_2.

Figure 6. Destruction of D_2 pre-coverage by D_2 recombination on D_2 pre-coverage doses.

Figure 7. REMPI-TOF signal during D exposure at different D_2 coverages.





V. Conclusions

We have empirically found that D_2 molecular coverage on a silicate enhances D_2 formation. We detect an increase of about 50% in the REMPI signal between 0s and 600s D_2 at T_s = 5.5K. This is in agreement with the findings from the TPD destruction analysis at T_s = 5.5K (up to 57%). At T_s = 10K the REMPI signal shows an increase of about 33% in the formation for different D_2 pre-coverage doses between 0s and 300s. In addition, formation is about 30% (+/- 10%) larger at T_s = 5.5K than the REMPI average at T_s = 10K. These results can constrain H_2 formation rates in ISM environments where dust and gas are thermally decoupled and physisorption is the dominant formation mechanism, such as proto-stellar cores (PSCs), PDR/XDR regions and dark clouds.

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