# The effect of roughness on the $T_3$-dewetting of molecular hydrogen

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**Abstract**

Triple-point dewetting is a well-known behaviour of molecular hydrogen and other van der Waals systems like noble gases on a solid substrate. Recent theoretical and experimental investigations (PRL **88**, 55702 (2002)) suggest that it is caused primarily by the roughness of the substrate. Strain induced due to the mismatch of the lattice constant of the substrate and the growing layers of the adsorbed materials is increased by the micro-roughness of the substrate which eventually leads to the growth of only a thin solid film of the adsorbate. The dominating role of the substrate roughness is demonstrated, e.g., by ellipsometric measurements on smooth Si surfaces (rms 0.15 nm), where a thicker solid hydrogen film than the 3 monolayers on "usual" substrates is observed. We present a way to modify and improve the surface quality of substrates for such wetting studies of solid van der Waals films.

*Key words:* triple-point dewetting; molecular hydrogen; solid films; surface roughness

Triple-point dewetting of noble gases on solid substrates like Au [1] is a well-known phenomenon which always happens below the bulk triple point $T_3$ of the adsorbed gas. One of the first theoretical approaches to the wetting-dewetting of solid substrates by noble atoms and molecules has been done by Gittes and Schick [2]. In this theory the substrate is considered as an ideal surface (i.e., atomically smooth and so having a well-known lattice parameter) and the thickness of the adsorbed film is related to the substrate-adsorbate interaction strength $R$. However, experiments performed so far show stronger non-wetting, even when the interaction strength is reduced, e.g., by preplating [3]. For $1.5 \leq R \leq 3$ complete wetting is expected in the solid phase [2], in contradiction to the experiment. But all practical substrates have a finite roughness and so one may ask about the role of the roughness profile of the applied substrate on the wetting property. In addition adsorption of impurity layers during the sample preparation and cooling down the cell may change the estimated values of the substrate-adsorbate interaction. Recently, a more realistic calculation has been done by Esztermann *et al.* [4], in which the roughness profile of the substrate is taken into account. The computed results of that investigation predict a significant variation of the film thickness on substrates with different roughness, i.e., the adsorbed film will be thicker on the smoother substrate, and for a certain range of substrate strength ($1.5 \leq R \leq 3$) it will diverge as the roughness goes to zero.

In this work we present results of adsorbed $H_2$ on a Si-wafer with an average roughness (rms) of 0.15 nm. Its surface is distinctly smoother than the previously used evaporated gold substrates on glass [3].

We use an ellipsometry setup, as shown in Fig. 1, to measure the $H_2$ film thickness on the Si substrate. The sample is mounted vertically in a Cu cell with windows for optical access to the sample surface. The whole assembly is placed inside an optical flow cryostat. A laser diode provides a very stable laser beam which passes through a polarizer and compensator before entering the cryostat. After reflection on the sample the beam leaves the cryostat, passes through an analyzer and is detected by a photodiode.

During measurements the optical components remain fixed. Instead of rotating compensator and ana-

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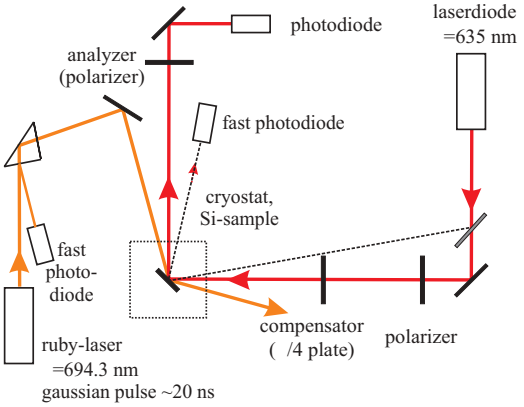


Fig. 1. Schematically shown is the annealing and ellipsometry setup. The surface can be annealed using pulses from a ruby laser, monitored by a fast photodiode. The ellipsometry setup is in PCSA geometry (the letters indicate the order of the components polarizer, compensator, sample and analyzer).

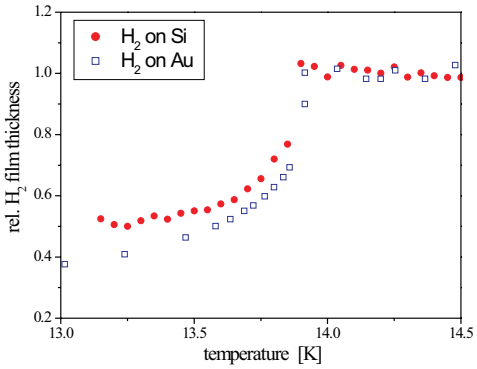


Fig. 2. Shown is the relative $H_2$ film thickness on Si and Au. The data points refer to average values of several cooling and warming runs. The thick wetted film, measured for $T > T_3$, is normalized to 1.

lyzer (like in ordinary null-ellipsometry) the intensity of the light passing the analyzer is measured. Since the adsorption of hydrogen molecules on the substrate changes the polarization of the reflected beam, the thickness of the adsorbed film can be monitored.

Usually a measuring run is started by doing an adsorption isotherm of $H_2$ at 15 K. After reaching saturated vapour pressure the temperature is scanned in the range of 13 K to 15 K. Thermodynamic equilibrium is tested by performing different ramping speeds. It turned out that for ramping speeds less than 50 mK/min equilibrium is ensured.

In Fig. 2 the $T_3$-dewetting of molecular $H_2$ on two different substrates, Si and Au, is shown. Note that the thickness of each film is normalized to its equilibrium film thickness in the complete wetting regime, i.e., at temperatures above 14 K. In Fig. 2 two different regions can be seen: i) for $T > T_3$ (13.95 K) the film thickness is large and nearly constant for both substrates

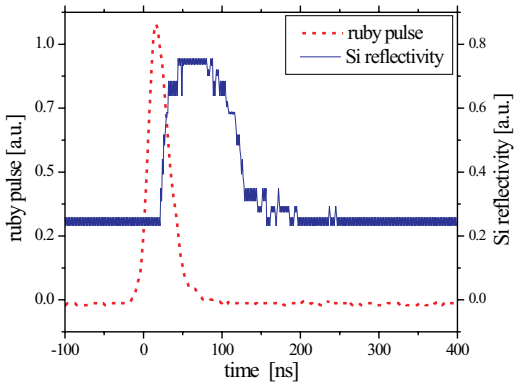


Fig. 3. *In-situ* annealing of a Si wafer. The dotted line shows a typical ruby laser pulse. By applying the ns-pulse the reflectivity (solid line) rises to the value of liquid Si which shows that the surface melts up (The reflectivity of Si increases by about a factor of 2 upon melting).

as expected for complete wetting of the liquid phase; ii) for $T < T_3$ triple point dewetting sets in being more pronounced for the relatively rough Au than for the smooth Si substrate. Since the $H_2$ van der Waals interaction with Au and Si, respectively, is similar, we interpret this result as a further confirmation that surface roughness contributes to the incomplete wetting of solid films below $T_3$.

For the results reported so far there might still be an impurity layer adsorbed on the Si surface. To overcome this problem we will in future measurements use Si substrates which are laser cleaned *in-situ*. In Fig. 3 it is demonstrated that the Si surface can reversibly be molten and recrystallized, which should result in an impurity-free substrate.

In conclusion, we have investigated and observed the effect of substrate roughness on $T_3$-dewetting. The results are consistent with theoretical estimations [4]. In addition, we have used the technique of laser annealing to prepare more ideal surfaces, on which future measurements will be done.

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