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

Jörg Angrik, Masoud Sohaili, Jürgen Klier, Paul Leiderer*

University of Konstanz, Department of Physics, D-78457 Konstanz, Germany

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 (Phys. Rev. Lett. 88 (2002) 55702) 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.

© 2003 Elsevier Science B.V. All rights reserved.

Keywords: Triple-point dewetting; Molecular hydrogen; Solid films; Surface roughness

Triple-point dewetting of noble gases on solid substrates like Au (see, e.g., Ref. [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 roughnesses, 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 [3,4], which had an rms roughness of 1.3 nm and more.

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 analyser and is detected by a photodiode.

*Corresponding author.

E-mail addresses: joerg.angrik@uni-konstanz.de (J. Angrik), juergen.klier@uni-konstanz.de (J. Klier).

Fig. 1. Schematic of 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 the PCSA geometry (the letters indicate the order of the components polariser, compensator, sample and analyser).

Fig. 2. Relative H_2 film thickness on Si and Au. The data points refer to average values of several cooling and warming runs. On the left y-axis the thick wetted film, measured for $T > T_3$, is normalized to 1. On the right y-axis, the absolute H_2 thickness on Au, measured via the surface plasmon technique, is shown.

During measurements the optical components remain fixed. Instead of rotating compensator and analyser (like in ordinary null-ellipsometry) the intensity of the light passing the analyser 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 13–15 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. Since the van der Waals interaction of H_2 with Au and Si is quite similar [5], the film thicknesses in this regime is expected to agree within a few percent. Note that the natural oxide on Si with a thickness of about 2 nm also has a similar van der Waals interaction. This layer will therefore have a negligible influence on the H_2 thickness. 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 as expected for complete wetting of the liquid phase and (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 van der Waals interaction of H_2 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 or annealed in situ. Preliminary measurements have shown that (see also Basso et al. [6]) 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.

This work was supported by the DFG-Schwerpunkt "Wetting and Structure Formation at Interfaces" under Grant Le 315/20.

References

- [1] J. Krim, J.G. Dash, J. Suzanne, Phys. Rev. Lett. 52 (1984) 640.
- [2] F.T. Gittes, M. Schick, Phys. Rev. Lett. 30 (1984) 209.
- [3] J. Klier, C. Weichhard, P. Leiderer, Physica B 284 (2000) 391.
- [4] A. Esztermann, M. Heni, H. Löwen, J. Klier, M. Sohaili, P. Leiderer, Phys. Rev. Lett. 88 (2002) 055702.
- [5] G. Vidal, M.W. Cole, Surf. Sci. 110 (1981) 10.
- [6] H.C. Basso, W. Dietsch, H. Kinder, P. Leiderer, Kapitza Resistance of Laser-Annealed Surface, Proceedings of the Fourth International Conference on Phonon Scattering in Condensed Matter, Springer, Berlin, 1984, pp. 266–268.