

Solid hydrogen films in a nonequilibrium state

R.N.J. Conratt, U. Albrecht, S. Herminghaus and P. Leiderer

Fakultät für Physik, Universität Konstanz, D-7750 Konstanz, Germany

Molecular hydrogen can be prepared as a metastable quantum solid by quench-condensation onto a substrate at sufficiently low temperature. Using surface plasmon resonance and light scattering techniques we have investigated such H₂ films with thicknesses up to several 100 Å, condensed onto a silver surface at 1.5 K. Since solid H₂ does not wet silver these films are in an extreme nonequilibrium state. At a temperature of 2.3 K we observe a dramatic rearrangement of the film overstructure on scales of ~1 μm, which implies an extremely high mobility of the H₂ molecules at a temperature far below the desorption threshold (3.8 K).

1. Introduction

During the last decade, the physics of thin films has become a field of substantial interest. In particular, quantum systems like helium or hydrogen films have been studied, using methods like calorimetry [1], microbalances [2], neutron-scattering [3]. In the experiments reported here, we have used optically excited surface plasmons [4] to investigate the static and dynamic behavior of quench-condensed hydrogen films. Hydrogen shows triple-point wetting on silver and gold surfaces [2,5], which implies that far below the triple point temperature ($T_t=13.96\text{K}$), films with only a few monolayers thickness are thermodynamically stable. The preparation of relatively thick (ca. 10nm) films by quench-condensation hence leads to metastable phases with yet unknown morphology.

2. Experimental

Our experiments are performed in an optical He4 cryostat at temperatures ranging from 1.3 K to 9K. The substrate is a 50nm silver or gold film evaporated on a glass prism. At the vacuum-metal interface, surface plasmons can be resonantly excited by illuminating (HeNe-laser or a tungsten lamp with a monochromator) the metal film through the prism at a particular angle of incidence [6]. This resonance angle is affected by a thin dielectric film adsorbed onto the metal/vacuum interface. The shift in resonance angle is proportional to the thickness of a dielectric layer on the metal film and can thus be used to determine the adsorbed film thickness. In our experiments the

hydrogen film thickness was in the range between 0.1 and 100 nm and could be determined with a resolution of ~0.1 nm. On the metal-dielectric-interface, the plasmons also can decay into photons by scattering from inhomogeneities of the interface. The scattering gives information about the roughness of the hydrogen films on a μm-scale. Simultaneously, the presence of roughness results in a broadening of the surface plasmon resonance. Experiments were performed with H₂, HD and D₂.

3. Results

In Fig. 1a, typical traces of the surface plasmon resonance position for H₂ and D₂ (gauged as effective film thickness) are displayed. The films are prepared at temperatures below 2K. Afterwards, the temperature is slowly ramped up, with a typical rate in the range of 0.7 to 2 K/h. Thus, the abscissa is a temperature axis and a time axis simultaneously. For each system, there exists obviously a quite well defined temperature at which the film desorbs (3.8K for H₂, 5.3K for D₂.) Quite remarkably, one observes in addition a transition to some intermediate phase about 1 to 2 K below this temperature. In Figs 1b and 1c, corresponding traces of the plasmon resonance width and the scattered light intensity are shown. The plasmon width has been normalized to its value for the uncovered substrate and like the light scattering gives a qualitative measure of the roughness of the film, as mentioned above. Obviously, the film roughness changes strongly when the system goes into the intermediate state. The transition was found to be irreversible, which means that the observed

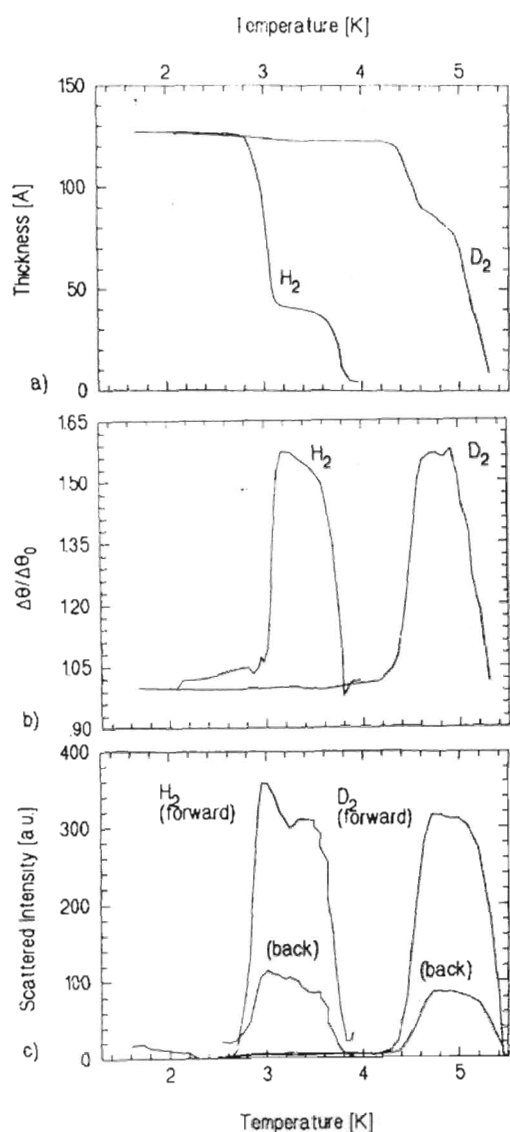


Fig 1a) Resonance Position (optical thickness),
 b) broadening of the plasmon resonance and
 c) scattered light intensity at two scattering
 vectors (forw.: $3.7 \mu\text{m}^{-1}$, backw.: $19.5 \mu\text{m}^{-1}$) for
 quench-condensed H_2 and D_2 films.

transition cannot be interpreted as a roughening transition. No significant changes in the annealing behaviour was observed on changing the heating-rate in the range from 0.7 K/h up to 2 K/h. Similar results were obtained for HD films, indicating that

ortho-para conversion has no influence on the effects.

Interpretation

We associate the observed transition with the incomplete wetting of hydrogen below T_t . A rearrangement of the adsorbed molecules should occur in films with a thickness larger than the equilibrium value[5], once the molecules become mobile. As a result we expect the formation of a thin film (only a few monolayers for the temperatures studied here) in coexistence with small crystallites. Assuming constant average coverage up to the desorption temperature we find that all the results presented in Fig 1 regarding the reduction in effective thickness and the increase in light scattering and the plasmon width of the transition are consistent with the appearance of crystals $\sim 400\text{nm}$ in height at an average spacing of $\sim 2\mu\text{m}$. Since the bulk mobility of hydrogen molecules in the investigated temperature range is small and the vapor pressure, on the other hand, is negligible below the desorption threshold, the material transport very likely occurs via surface diffusion. The sharp transitions in the structure of H_2 and D_2 films at 3.0K and 4.6K respectively, imply that surface diffusion sets in rather rapidly at these temperatures.

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