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Stability of solid T_2 films

M. Przyrembel, H. Fischer, A. Hermanni, E. Otten
Johannes-Gutenberg-Universität Mainz, 6500 Mainz, FRG

and

P. Leiderer
Universität Konstanz, 7750 Constance, FRG

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Films of molecular tritium, physisorbed on a cold substrate, have been investigated by means of ellipsometry. A significant effect of the radioactive decay processes on the desorption kinetics and the low temperature stability of these films is not observed.

The properties of physisorbed films are a field of intense current interest. Investigations reach from two-dimensional behavior in the submonolayer and monolayer regime up to thicker films, where the transition from 2D to 3D behavior is studied, and where phenomena like wetting, layering, surface roughening and surface melting have been observed [1]. In the case of very light adsorbates the behavior is expected to be modified by quantum effects, and this has been found indeed for the light noble gases and the hydrogen isotopes H_2 and D_2 [2]. For the heaviest hydrogen isotope, T_2, investigations are complicated due to the radioactive decay of this molecule. Thus to our knowledge there have been no studies of T_2 films up to now, except for very thick layers (>100 nm) and relatively high temperatures in the vicinity of the triple point at 20.6 K [3].

Our motivation for investigating solid T_2 films was twofold: (i) first, such films would provide valuable information about the systematics of quantum effects by comparing their properties to those of the lighter isotopes. For this purpose also neutron diffraction studies of T_2 are in progress [4]; (ii) a second, and more practical, reason is that a thin film of solid T_2 is projected as a source for an experiment at the University of Mainz in which the neutrino rest mass is to be investigated via the β-decay of tritium [5]. Since these measurements require long sampling times, it is essential that a film with a thickness of some ten monolayers, facing the vacuum of the β-spectrometer, has a lifetime on the order of at least 24 h.

As a first step for investigating the behavior of thin tritium films we chose to study their stability in a temperature range where the vapor pressure of the T_2 is in the high vacuum regime ($T<10^{-6}$ mbar). Although no publications exist, the behavior of such films is a matter of considerable controversy. An upper limit for the stability of a T_2 film is given by the lifetime of the nuclei, $t_{1/2}=12.3$ y. It has been argued, however, that the stability of the film is perhaps drastically reduced due to the energy released by the radioactive decay process.

In order to determine the stability of the T_2 films we have measured their thickness by means of ellipsometry. The configuration of the ellipsometer was PCSA (polarizer–compensator–sample–analyzer), where the compensater was held at a fixed position, and polarizer and analyzer were adjusted for minimum transmitted light intensity. As a light source we used a He–Ne laser, attenuated to 0.01 mW to avoid heating effects at the sample surface.

The experimental set-up is sketched in fig. 1. T_2 gas was obtained by heating a Ti pellet in the gas

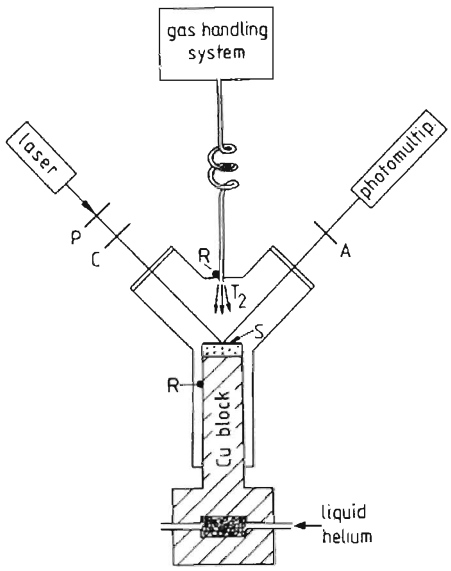

Fig. 1. Schematic experimental set-up. P: polarizer; C: compensator; S: sample; A: analyzer; R: thermometer and heater.

handling system, and was then purified by passing it through a LN_2 cold trap. A capillary connected the storage volume at room temperature with the sample cell. During growth of the film the nozzle in the cell was kept at a temperature of about 20 K to prevent it from blocking. The cold surface onto which the T_2 film was frozen had a temperature of 2.8 K and was located opposite to the nozzle at a distance of 1 cm. The T_2 molecules were thus quench-condensed and formed a probably strongly disordered film, as is known from X-ray diffraction and specific heat measurements of rare gas films [6] and from measurements with surface state electrons on solid H_2 films prepared in a similar way [7].

The substrate for the adsorbate was an aluminum film evaporated onto a glass plate, which was thermally coupled to a copper cold finger. The copper block was attached to the evaporation stage of a helium cryostat and thus was the coldest part in the sample cell. The rest of the cell walls, apart from the optical windows, were stainless steel; due to thermal radiation from surrounding baffles they were at a distinctly higher temperature than the copper block. The temperature of the substrate surface was also somewhat higher than that of the cold finger (by about 10^{-2} K) as a result of the finite thermal conductivity of the glass plate. This fact is important for the interpretation of the measurements, since under these conditions T_2 molecules desorbing from the film are collected mainly on the copper finger and only with a small probability are readsorbed on the sample surface investigated by the ellipsometer (i.e. the sample can be considered as a nearly open system).

In order to bring out possible peculiarities arising from the radioactive decay of T_2 the experiments were also carried out with H_2 and D_2 gas as adsorbates. All these ellipsometric measurements of the hydrogen isotopes are hampered by the fact that the refractive index of these substances is small (1.14, 1.20 and 1.21 for H_2, D_2 and T_2, respectively) and that the detecting laser beam has to pass a number of windows at low temperature which develop stress birefringence. Nevertheless a resolution in thickness of about 5 Å could be achieved, which roughly corresponds to 1.5 monolayers. Results for as-quenched films of H_2 at $T=2.8$ K are shown in fig. 2 for various thicknesses, obtained by subsequent condensation of given quantities of gas. (Due to the preferential direction of the gas molecules leaving the nozzle the film thickness was not uniform, but had its maximum in the centre of the glass substrate and was close to zero at the edge. This distribution was taken into account for the calculation of fig. 2.)

A comparison of the ellipsometrically determined film thickness with the total amounts of admitted gas

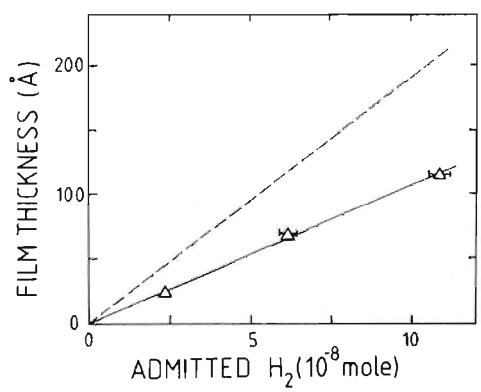

Fig. 2. Ellipsometrically determined thickness of a quench-condensed H_2 film at 2.5 K (measured at the centre of the sample) versus the admitted amount of H_2 gas. The dashed line represents the result expected for a sticking coefficient of 1.

shows that under the given conditions the sticking coefficient of hydrogen was considerably less than 1: Under the present conditions only a fraction of about 55% of the molecules condensed onto the surface which they first hit after leaving the nozzle. Similar results were obtained for T_2. This has important implications for the preparation of a solid T_2 film as a source for a neutrino rest mass experiment, because the "non-sticking" molecules will contaminate the spectrometer unless they are properly shielded during the evaporation process.

For a test of the stability of the adsorbed T_2, a film with a thickness of 310 monolayers, kept at a temperature of 2.8 K, was monitored as a function of time. No change in thickness was observed over 2 h within our accuracy of 1.5 monolayers. Since the film thickness was relatively large in this case, self-heating effects, if important, should have been even more serious than for the projected source with a thickness of some ten monolayers, where thermal excitations can be carried away more efficiently by the substrate. Defining τ as the time in which the film thickness has decreased by a factor of 2, we obtain $\tau > 500$ h. Thus thin T_2 films appear to be sufficiently stable at a temperature of 2.8 K.

A second criterion for the stability of T_2 films is their desorption behavior at higher temperatures, in particular in comparison to the lighter hydrogen isotopes. For this purpose we have plotted in fig. 3 results for the desorption rate of H_2, D_2 and T_2 as a function of temperature. In these measurements the films were prepared, as before, at $T = 2.8$ K, and then the temperature of the substrate was slowly increased. The evaporation rate, as determined from the temporal change of the film thickness, is at first below our experimental resolution. The temperature where appreciable desorption sets in (> 1 monolayer/s) is 4.5, 6 and 7.5 K for H_2, D_2, and T_2, respectively. The dashed curves in fig. 3 represent the behavior expected for the desorption coefficient on the basis of a model, which takes the finite temperature difference between sample and cold finger and the geometry of the sample cell into account [8]. (For an adsorbate facing UHV it is estimated that the curves would roughly be shifted to lower temperature by about 0.5 K.) The steep increase of the desorption rate reflects the exponential rise of the vapour pressure $p = p_0 \exp(-E/kT)$, where E is the

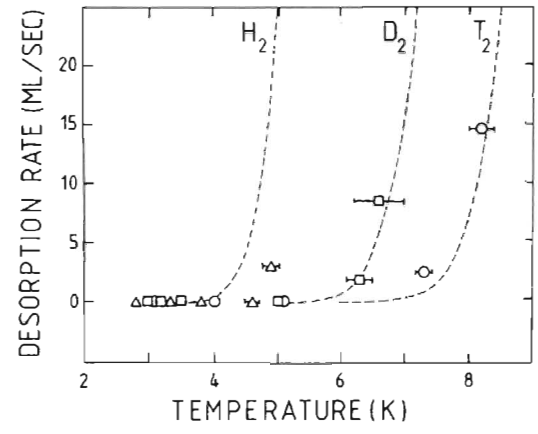

Fig. 3. Temperature dependence of the desorption rate (in monolayers/s) for H_2 (△), D_2 (□), and T_2 (○). The dashed curves indicate the behavior expected on the basis of the vapor pressure of the adsorbates (see text).

latent heat of sublimation per molecule [9]. The shift of the T_2 desorption curve to higher temperature as compared to H_2 and D_2 is completely in line with the behavior expected due to the higher sublimation energy of tritium.

In summary, we have studied adsorption and desorption of thin films of the hydrogen isotopes. Our results show no sign of any markedly anomalous behavior of the radioactive isotope T_2. Molecular tritium films are stable on long time scales and thus appear as a promising source for the projected neutrino experiment. Moreover, they present an interesting possibility to extend the studies of quantum adsorbates to a slightly more classical system.

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