Vacuum 233 (2025) 113931

Contents lists available at ScienceDirect

Vacuum

journal homepage: www.elsevier.com/locate/vacuum

Short communication

Estimation of the thermal radiation induced desorption yield for hydrogen and helium at liquid helium temperatures

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ARTICLE INFO

Handling Editor: Oleg Malyshev

Keywords:
Thermal radiation induced desorption
Cryogenic vacuum
Pressure limit
XHV

ABSTRACT

Infrared radiation induced desorption of adsorbed molecules from cryogenic surfaces can lead to potentially detrimental pressure limitation in cryogenic UHV/XHV systems. For a sub-monolayer coverage, we determined the corresponding yields for hydrogen and helium exposed to room temperature radiation to be in the order of $1{\cdot}10^{-4}$ and 0.02 $m^2(W{\cdot}s)^{-1}$ respectively. We used these yields to estimate the radiation induced pressure in the PUMA experiment, presently under construction at CERN.

1. Introduction

In a cryo-pumped UHV/XHV system, whose cryogenic surfaces are ideally shielded against incident thermal radiation, an adsorption-desorption equilibrium prevails, which can be well-described for a large number of gas-solid systems by the Dubinin–Radushkevich–Kaganer (DRK) isotherm [1,2]. In its inverted form, i.e. converted to p, it reads as follows:

$$p_{DRK} = p_0 \; exp\left\{ -\frac{1}{k_B T}\sqrt{-\frac{1}{D} ln\left(\frac{\theta}{\theta_m}\right)} \right\} \quad (1)$$

Wit θ being the actual coverage, p_{DRK} the actual pressure calculated from this model, p_0 the saturated vapour pressure, k_B the Boltzmann constant and T the absolute temperature. θ_m is the monolayer capacity of the surface and D is a parameter related to the binding energy. The latter two parameters need to be determined empirically for a given surface.

This model has been tested down to about 10^{-10} Pa by measurements for hydrogen and helium at temperatures below 10 K [3,4].

Significant deviations from the predicted DRK equilibrium pressure can be caused by thermal radiation. Antimatter experiments require local residual gas pressures far in the XHV range. In particular, the PUMA experiment that is presently being constructed requires local pressures of 10^{-15} Pa in its antiproton storage trap. Reliable data about the thermal radiation induced desorption are not available for hydrogen and helium in the sub-monolayer regime. We determine the thermal radiation induced desorption yield for hydrogen by measurements and for helium from the data available in Ref. [4].

Benvenuti et al. investigated in Ref. [5] the influence of thermal radiation on the saturated vapour pressures between 2.3 and 4.2 K and determined a "desorption efficiency". Above one monolayer the thermal radiation induced pressure is proportional to the absorbed heat, but independent of the coverage and the material. The authors concluded that the photons are first absorbed, and their energy is then transmitted by phonon interaction to the adsorbed molecules. Below one monolayer, the pressures were offset by a temperature independent, but coverage dependent thermal radiation induced pressure value [5, Fig 7]. The absorbed heat, however, was not indicated for this condition.

1.1. Thermal radiation induced desorption of hydrogen at 4.2 K

In one of our cryogenic experiments (Fig. 1) designed for electron stimulated desorption from cryogenic surfaces ("ESD-setup") we observe a proportional increase of H_2 equilibrium pressure with surface coverage in the sub-monolayer range (Fig. 2). A particularity of this experiment is that the cryogenic surfaces of the setup are exposed to a high thermal load due to room temperature radiation. A copper tube at 293 K is directly opposed to a sample at 4.2 K (ID = 52 mm, L = 155 mm), which is immersed in a liquid helium (LHe) bath. This coverage dependent offset is due to the thermal radiation. From the helium boil-off we measured a thermal radiation dominated heat load of 0.8 W.

We determine the thermal radiation-induced desorption yield under the following assumptions:

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https://doi.org/10.1016/j.vacuum.2024.113931
Received 8 September 2024; Received in revised form 1 December 2024; Accepted 4 December 2024
Available online 5 December 2024

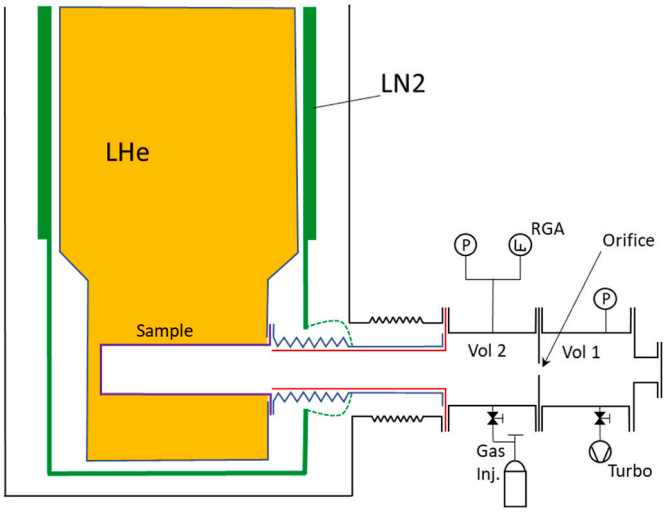

Fig. 1. Cryo-ESD setup. For clarity, elements not relevant for this report are not shown in this figure (e.g. electron source, support mechanical feed-throughs, …).

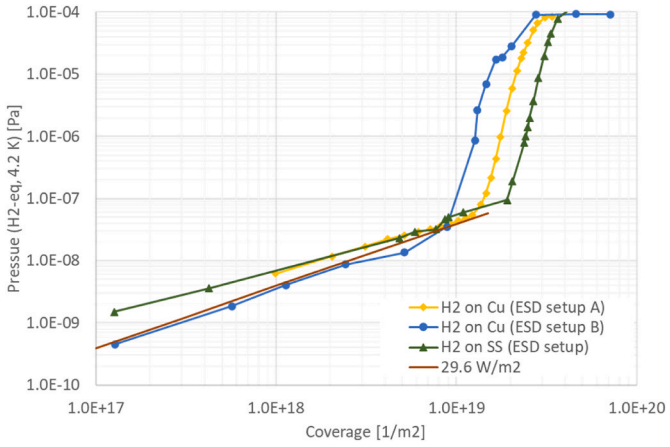

Fig. 2. Adsorption isotherms on copper and stainless-steel samples. The straight line (brown) is the fit to the linear part from which η_{th} in equation (2) was determined. It corresponds to the calculated pressure increase due to thermal radiation for a heat load of 0.8 W (29.6 W m^{-2}). The background pressure is subtracted. The measurements of yellow and green data sets were taken after extended periods of several hours, where the sample was at T < 20K before starting the first injections. The difference in slope is caused by a cumulated coverage from outgassing from RT parts, while zero initial coverage was assumed before the first injection.

- The radiative heat is equally absorbed over the whole cryogenic surface at 4.2 K.
- The desorbed molecules are leaving the surface with the kinetic energy of the cryogenic surface (4.2 K) and are immediately re-adsorbed with a sticking factor of 1.
- In equilibrium, desorption rate $\eta_{th} q\, \theta$ equals the impingement rate $p_R \bar{c}/(4 k_B T)$, resulting in the establishment of a time-independent, but coverage-dependent thermal radiation-induced equilibrium pressure which can be expressed as

$$p_R = \eta_{th}\, q\, \theta\, 4\, \bar{c}^{-1}\, k_B T, \tag{2}$$

where p_R being the local thermal radiation induced equilibrium pressure in Pa, η_{th} the thermal radiation induced desorption yield in m^2(W·s)$^{-1}$, q the surface specific absorbed radiative heat flux per unit area in W·m^{-2}, θ the surface coverage in m^{-2}, $\bar{c}$ the average speed of the molecules in m·s^{-1}, k_B the Boltzmann constant and T the temperature of the cryogenic surface in K.

Since there was no external pumping during the measurements, the yield, η_{th}, can be calculated from equation (2).

The local radiation induced pressure is about $4{\cdot}10^{-8}$ Pa for a coverage of $1{\cdot}10^{19}$ m^{-2}. Note that without thermal radiation the DRK prediction for the sub-monolayer coverage and the given temperature would be far below measurable values.

From the abovementioned absorbed heat (0.8 W) over the sample surface (0.027 m^2), the pressure ($4{\cdot}10^{-8}$ Pa), surface coverage ($1{\cdot}10^{19}$ m^{-2}) and the average molecular speed of hydrogen at 4.2 K (211 m s^{-1}) we estimate a desorption yield of:

$$\eta_{th\_H2} \approx 1.2{\cdot}10^{-4}\ \mathrm{m^2(W\,s)^{-1}}$$

Our experiment did not allow dedicated studies. In order to test this value for consistency with other experiments, we estimate hereafter the thermal radiation induced and coverage dependent pressures in other published data.

1.2. Compatibility with published data

As mentioned earlier, Benvenuti et al. [5] observed a temperature independent and approximately linear radiation-induced pressure at coverages below one monolayer (≈$3{\cdot}10^{19}$ m^{-2}) [5, Fig. 7]. The associated absorbed heat is not given for the condition shown, such that the photon-induced desorption yield cannot be inferred directly from this figure. However, equation (2) allows us to estimate the surface specific heat load from the radiation-induced pressure and θ given in that figure: $p_R = 5{\cdot}10^{-9}$ Pa for a coverage of $3{\cdot}10^{19}$ m^{-2}. We estimate radiative heat in this experiment ([5, Fig. 7], "Model B″ cryopump, stainless steel surface) to be in the order of 1.7 W m^{-2}.

Similarly, [5, Fig. 10] shows an adsorption isotherm with a thermally better performing "Model E″ cryopump on a silver coated surface: $p_R = 4{\cdot}10^{-11}$ Pa for a coverage of $1{\cdot}10^{19}$ m^{-2}. The estimated radiative heat for this experiment then is 0.041 W m^{-2}.

Both values lie within the range of the lowest heat loads measured on the respective cryopumps mentioned in the publication. From this point of view, applying the desorption yield determined in the ESD-setup seems consistent.

Further, for higher coverages above 2 monolayers (i.e. > $6{\cdot}10^{19}$ m^{-2}) the authors of [5] could determine an approximately substrate and coverage independent "desorption efficiency", defined as the (desorption rate · sublimation energy) divided by the absorbed heat flux. This dimensionless coefficient is [5, Fig. 14]:

$$\eta_{th\_sat} = 5{\cdot}10^{-6} \ldots 1{\cdot}10^{-5}\ \text{(for } \theta > 2 \text{ monolayers, on bare substrates)}$$

1.3. The corresponding thermal radiation induced pressure can be calculated as follows

$$p_{R\_sat} = \eta_{th\_sat}\, \epsilon^{-1}\, q\, 4\, \bar{c}^{-1}\, k_B T \tag{3}$$

With $\epsilon = 1.33{\cdot}10^{-21}$ [J] being the sublimation energy of a H_2-molecule from the bulk.

For copper, the associated surface specific absorbed heat is 4.6 [W·m^{-2}] (from [5, Fig. 14]). The molecular speed at 2.3 K is 156 m s^{-1}. This results in a thermal radiation induced pressure of

$$p_{R\_sat\_Cu} = 2{\cdot}10^{-8}\ \mathrm{Pa} \quad \text{from (3)}$$

This value lies within the error bars of the measured values. Note that this calculation assumes that the molecules leave with the kinetic energy of the substrate temperature and are re-adsorbed with a sticking factor of 1 on the same surface. In this particular experiment at 2.3 K, the equilibrium pressure is largely dominated by the photon-induced desorption. This pressure rise is approximately coverage independent

above 2 monolayers.

Up to one monolayer we can use the yield determined in the ESD-setup and plug it into equation (2). Calculating the radiation-induced pressure for one monolayer according to (2), p_{R_Cu} is of the same order as the one for high coverages (and coverage independent).

$p_{R\_Cu} = 1.35 \cdot 10^{-8}$ Pa (from (2) at 1 monolayer)

These results indicate that the two equations (2) and (3) give consistent estimates of the thermal radiation induced pressure.

E. Wallén measured adsorption H_2 isotherms at liquid helium temperatures [6]. He gave rather detailed information about the dimensions of the setup and the measurement procedure. From the described measurement stabilization times, we can assume a cryostat autonomy of around 24 h. The corresponding heat load is 0.14 W, which also include conductive heat and direct radiation into the LHe bath. The radiative heat load onto the sample's inner surface is lower and is more realistically in the order of 0.05 W. Fig. 3 shows Wallén's H_2 isotherms on copper at 4.2 K compared to our isotherm data taken on the ESD setup. While the effect of thermal radiation is clearly visible for our ESD setup, the influence on Wallén's setup is less pronounced due to the smaller heat load and lies near the measurement limit of that setup.

1.4. Helium

Helium has a lower adsorption energy than hydrogen and it is therefore expected to be desorbed with a higher yield. Indeed, the helium isotherms on stainless steel in [4, Fig. 4] show a strong linear dependence of the equilibrium pressure with coverage at low temperatures. This linear trend is not compatible with the DRK model. Fig. 4 shows the calculated helium adsorption isotherms by combining equations (1) and (2):

$$p(\theta) = p_R(\theta) + p_{DRK}(\theta) \quad (4)$$

using the DRK parameters determined by Erik Wallén [4, Fig. 6]. The trends shown in [4, Fig. 4] can be well reproduced with a thermal desorption yield $\eta_{th\_He} \approx 0.02$ $m^2(W{\cdot}s)^{-1}$. Note that the model assumes that the helium atoms are desorbed with a kinetic energy that correspond to the sample surface temperature and are re-adsorbed with a sticking factor of 1 on the same surface. Because the thermal energy of the photons is much higher than the adsorption energy and the surface temperature, it is likely that the kinetic energies of the photon induced desorbed He-atoms are higher than assumed. If this is the case, applying the thermal transpiration to the measurements at different sample temperatures, would lead to a systematic and apparent temperature dependent local pressure in the range where the pressure is dominated by photon induced desorption. This fact may contribute to the apparent systematically higher thermal photon induced pressures at higher substrate temperatures seen in [4, Fig. 4].

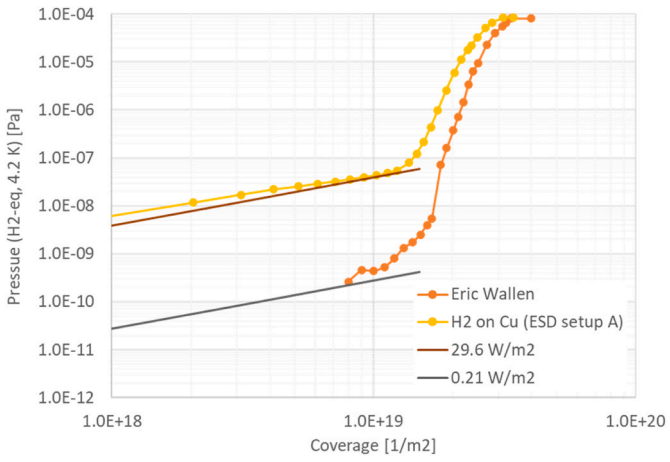

Fig. 3. H_2 isotherms measured on the ESD-setup and by Wallén in [6, Fig. 2]. The straight lines are the equilibrium pressures calculated using (2) with surface specific heat loads of 0.8 W (on a sample surface of 0.027 m^2) and 0.05 W (on a sample surface of 0.238 m^2).

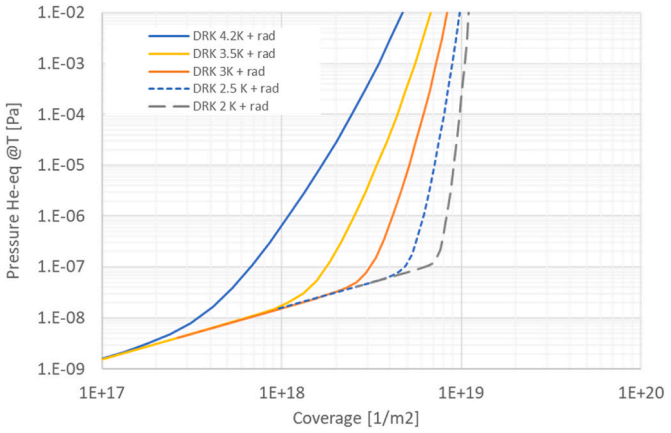

Fig. 4. Adsorption isotherm for helium calculated with the DRK model (D = $2.92 \cdot 10^4$ eV^{-2}, $\theta_m = 1.27 \cdot 10^{19}$ m^{-2}), *including thermal radiation induced desorption. Radiative heat 0.05 W, cold sample surface 0.1* m^2, $\eta_{th\_He} \approx 0.02$ $m^2(W{\cdot}s)^{-1}$.

1.5. Projection for the PUMA experiment

At the beginning of the PUMA project, the pressure evolution inside the antiproton storage trap has been simulated using the DRK isotherm parameters from Ref. [3] for hydrogen and from Ref. [4] for helium. Knowing that thermal radiation may have a detrimental influence on the equilibrium pressure, several measures to minimise the thermal radiation into the storage trap were taken. Those are a cold shutter at 50 K at the cryostat entry, a collimator tube at the entry to the trap, which also minimise the gas conductance, and carbon coating of cold parts in front of the trap to increase the effective cold pumping surface and absorb thermal radiation.

The residual radiative heat load into the trap with the shutter open is in the order of 5 μW. This estimate assumes a diffuse (isotropic) black body radiation at 293 K from the open shutter aperture (≈1 cm^2) into the aperture at the trap entry (also ≈ 1 cm^2) at a distance of 0.6 m. With the shutter closed the radiative heat load would be further reduced to a few tens of nW.

Applying the thermal radiation induced desorption yields as determined above for hydrogen and helium, we can now estimate their influence on the adsorption isotherm for the case of the shutter being open. The effective adsorption surface is around 0.1 m^2. Fig. 5 shows the calculated equilibrium pressure using (4) for a heat load of 5 μW, as estimated for the PUMA trap. To remain inside the target pressure of 10^{-15} Pa, the radiation induced hydrogen partial pressure would allow a coverage of around $2 \cdot 10^{17}$ m^{-2} (shutter open). This corresponds to about 0.01 monolayers.

For helium at 4.2 K the pressure at around 10^{-15} Pa is already dominated by the DRK prediction and the radiation has only a minor additional effect on the pressure. The corresponding coverage given by the DRK model is around 10^{15} m^{-2}.

2. Summary and conclusion

In summary, we estimated the thermal radiation induced desorption rate for hydrogen. In the sub-monolayer regime this yield is of the order of $1 \cdot 10^{-4}$ $m^2(W{\cdot}s)^{-1}$. For helium, we determined this yield from data in Ref. [4] to be in the order of 0.02 $m^2(W{\cdot}s)^{-1}$. Both yields are only valid for radiation from room temperature and are subject to rather high uncertainties. Nevertheless, they allow approximative quantifications of

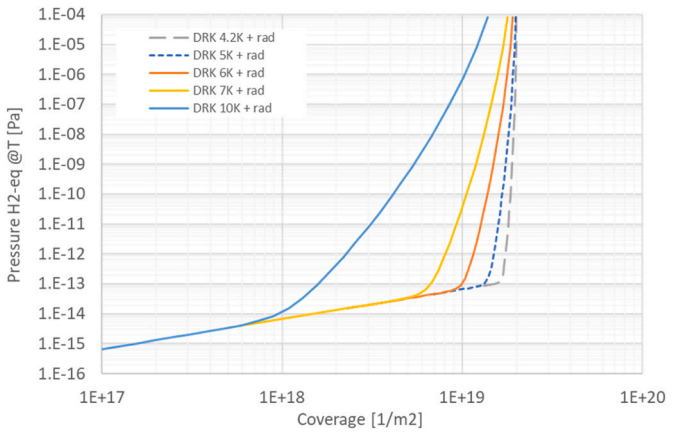

Fig. 5. Calculated adsorption isotherms for H_2 including a surface specific thermal radiation of $5{\cdot}10^{-5}$ W m^{-2}, using the DRK parameters from Ref. [3] and a thermal radiation desorption yield of $1.2{\cdot}10^{-4}$ m^2(W·s)$^{-1}$.

the influence of thermal radiation in a cryogenic vacuum system. For example, lifetime degradations over time in antimatter experiments with a direct view to room temperature parts may be caused by the accumulation of H_2 molecules on the cryogenic surface and thermal radiation induced desorption.

Our setup was not designed to study precisely and in detail the thermal radiation induced desorption. In view of the possible implication for future XHV cryogenic experiments, we recommend preparing a dedicated setup to measure this effect.

CRediT authorship contribution statement

Berthold Jenninger: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Elena Bez:** Writing – review & editing, Visualization, Software, Methodology, Investigation, Formal analysis, Data curation. **Anke Stöltzel:** Writing – review & editing, Visualization, Software, Formal analysis. **Fleur Rooijakkers:** Visualization, Software, Methodology, Formal analysis, Data curation. **Eino Tiirinen:** Writing – review & editing, Visualization, Software, Methodology, Formal analysis, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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