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Solubility of hydrogen in liquid helium - development of a measurement apparatus

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Introduction

LHe-Transportdewar
[Fa. Cryotherm]

Former assumption

LHe generally has a high degree of purity

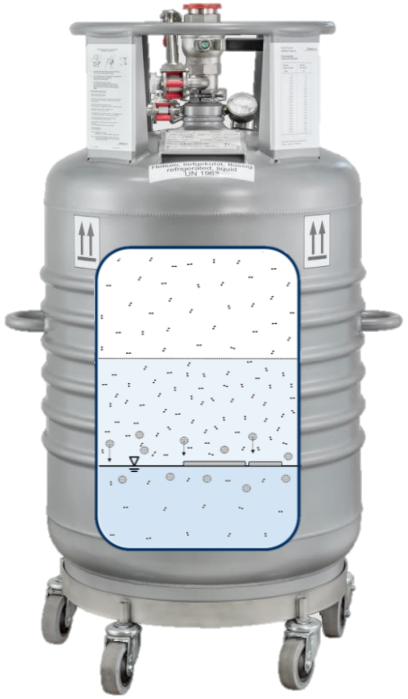
However, it has been known for some years:

LHe contains solutes and/or suspended solids

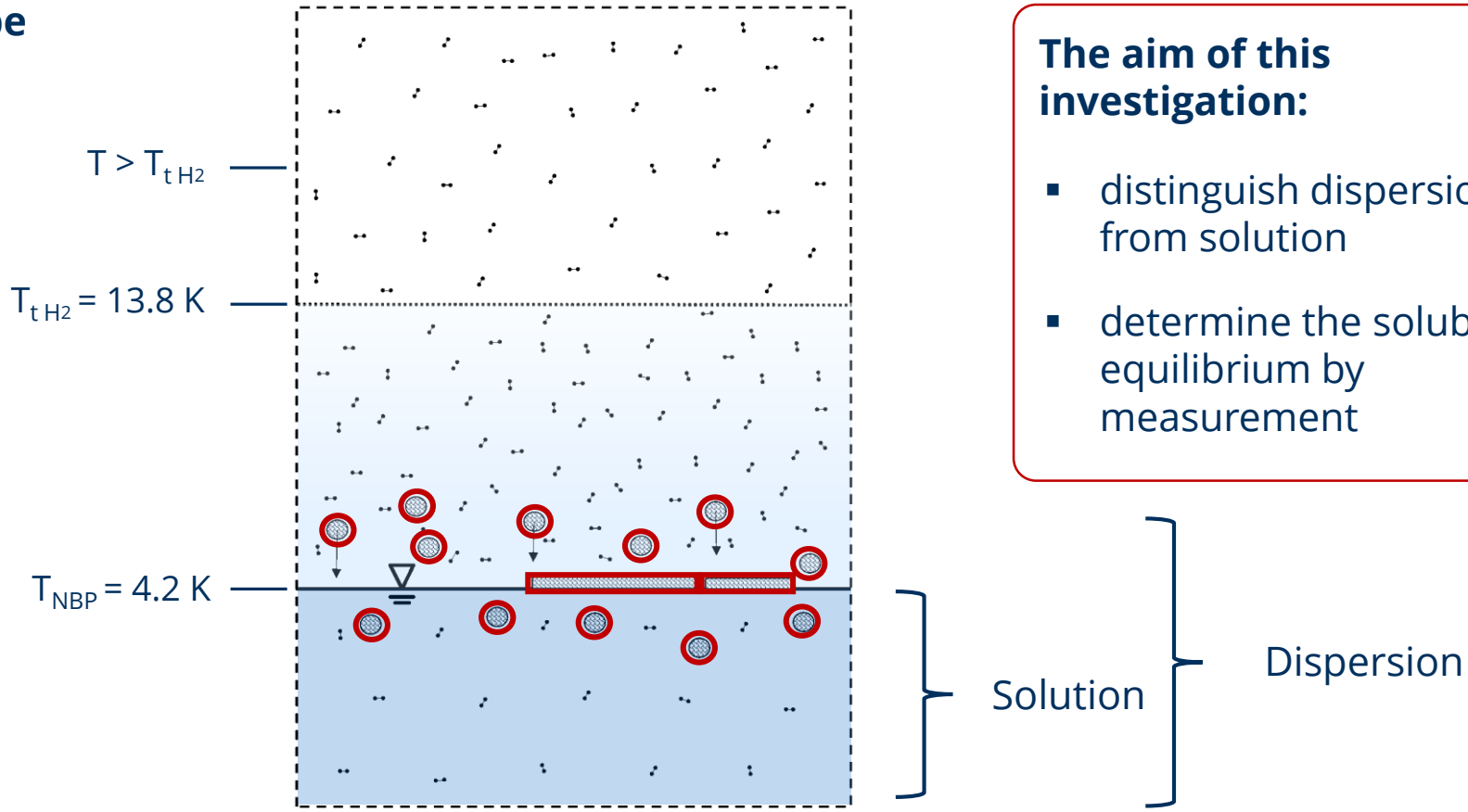
Under suspicion:

The binary system helium / hydrogen

Different equilibria are to be expected



LHe-Transportdewar
[Fa. Cryotherm]



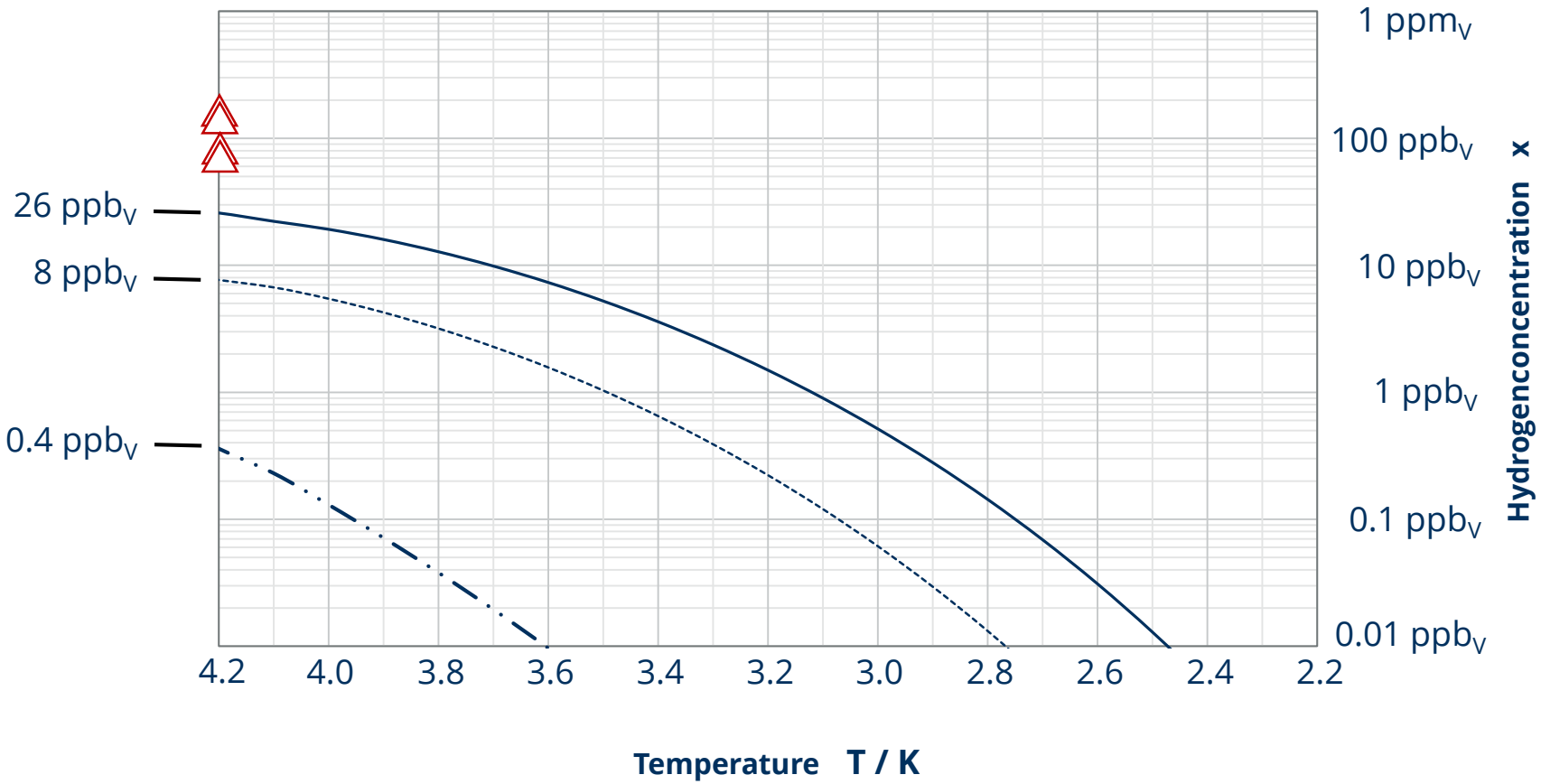
Hypothetical macroscopic behavior of the He- H_2 system at low pressures and low hydrogen concentrations

The aim of this investigation:

- distinguish dispersion from solution
- determine the solubility equilibrium by measurement

The binary system helium / hydrogen

Estimation of solubility using regular solution theory (RST) [1]:



Ideal solubility extended by change of heat capacities

Ideal solubility

RST according to Jewell et al. [2]

Measured liquid phase values achieved for various Dewar:

- △ 127 ppb$_V$
- △ 71 ppb$_V$
- △ 83 ppb$_V$
- △ 113 ppb$_V$

(σ_s=15...20%,
+ unknown systematic error)

Development of a measurement apparatus

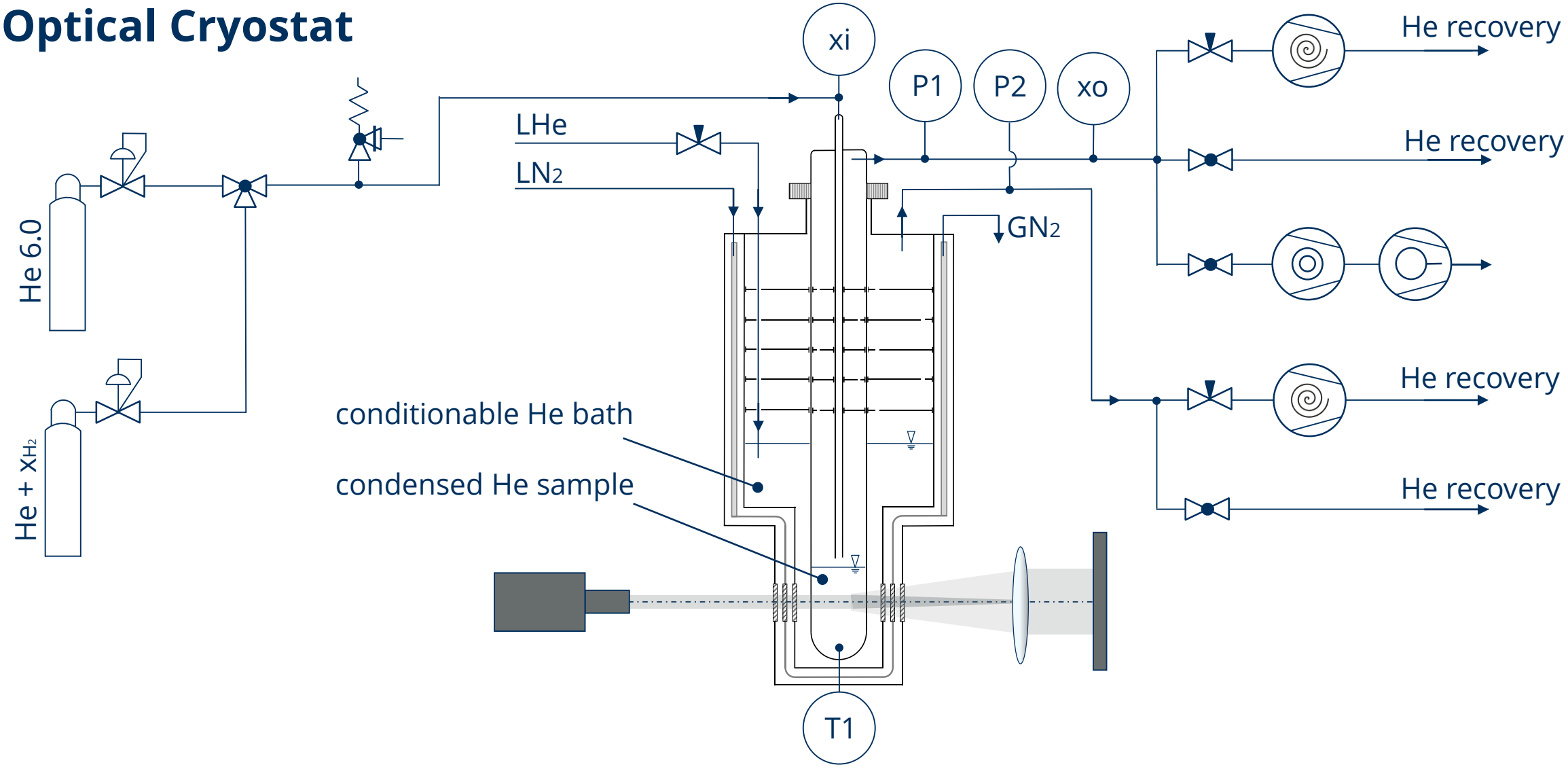
Three functions to fulfill:

1. Provision of a contaminated and condensed helium sample
2. Possibility of conditioning the sample and measuring the relevant parameters (pressure / temperature / H_2 concentration)
3. Detection of exceeding or falling below the solubility limit on an optical path

Challenges in three disciplines have to be tackled:

1. Cryostat design	2. Analytics	3. Solubility limit detection
– Optical cryostat	– Gaschromatographie	– Laser diffraction method

Optical Cryostat



Gaschromatography

Specialised gaschromatograph:

Peak Performer 1 RCP 910-100

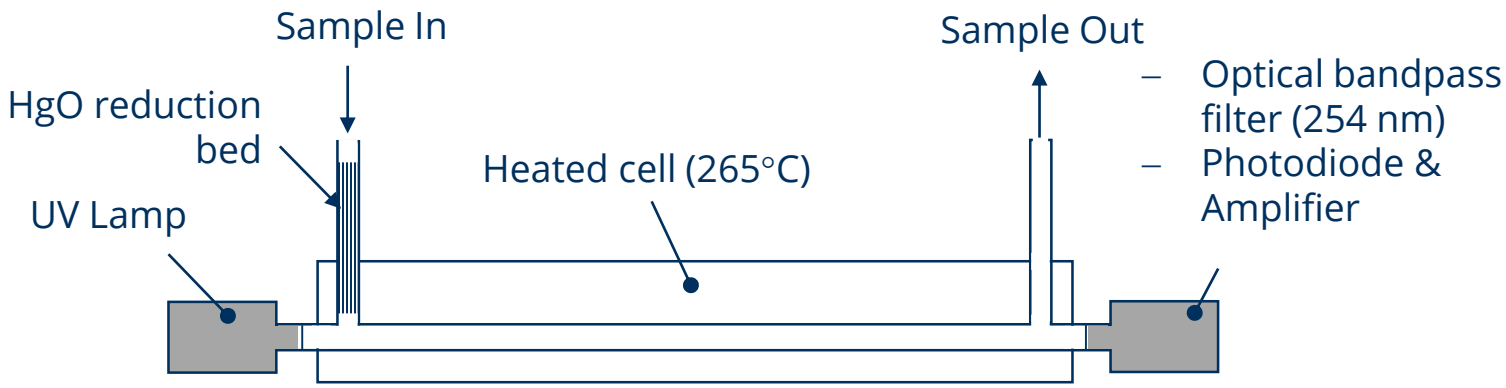
Peaklaboratories Inc., California, USA

- Exclusively for trace levels of H_2 and CO
- Reducing Compound Photometer (RCP)

Gaschromatographic principle:

- Two columns in series: *Unibead 1S* and *Molesieve 13X* @ 105°C
- 1 mL sample loop (1 … 100 ppb_v H_2)
- Specified detection limit: 0.8 ppb_v for H_2
  0.3 ppb_v for CO

Peak Performer 1
[Fa. Peaklaboratories, USA]

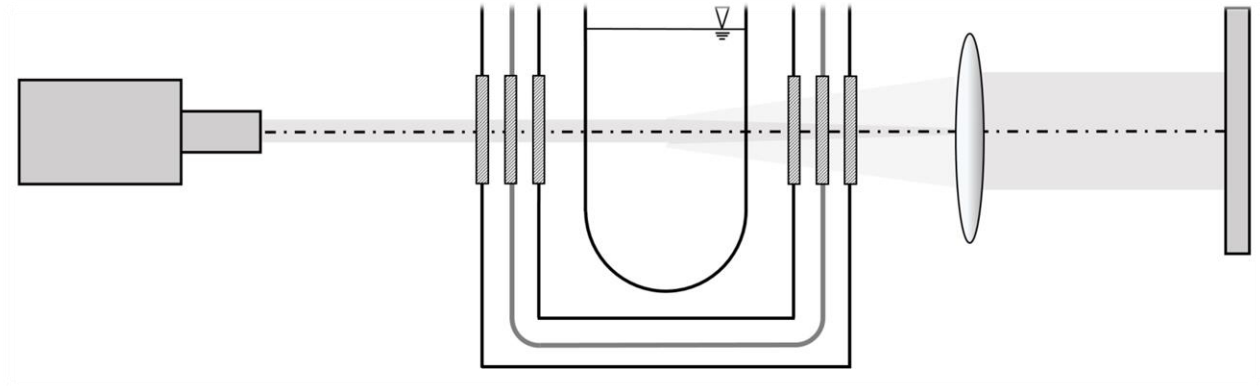


Scheme of Reducing Compound Photometer (RCP) used as detector

Saturation limit detection

Detection of precipitation and solubilisation of H_2 by means of laser diffraction method

Investigation and selection still ongoing



Questions arise:

- Is that feasible?
- Has anyone done something similar before?

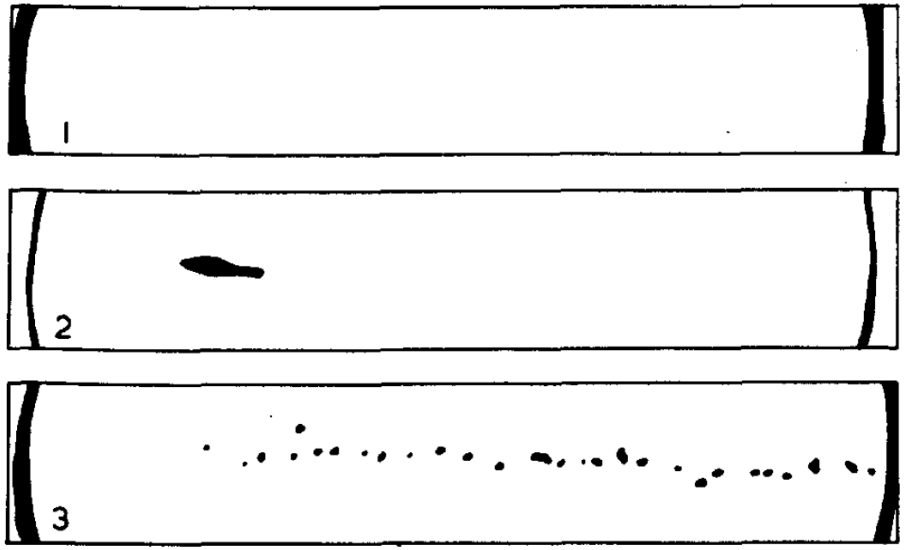
Saturation limit detection

Abrikosova et al. [2] in 1971:

Motivation: In course of investigations on electrical breakdown parameters

Abrikosova et al. found that the measurement results could not only depend on the expected ionization of the helium atoms. Sub micron solid impurities were hypothesized.

- Laser scattering method with pulsed ruby laser ($2 \cdot 10^9$ W·cm^{-2})
- Investigation of purified and unpurified liquid helium



– Before the disturbance of the helium
– Laser flash
– Immidiately after the disturbance of the helium

Abrikosova et al.: Evidently sufficent submicroscopic solid particles appear after disturbance . Rapidly-settling particles can be seen in a laser beam

Saturation limit detection

Bewley et al. [3]:

Motivation: Using solid hydrogen particles as tracer
in turbulent liquid helium for laser doppler velocimetry (LDV)

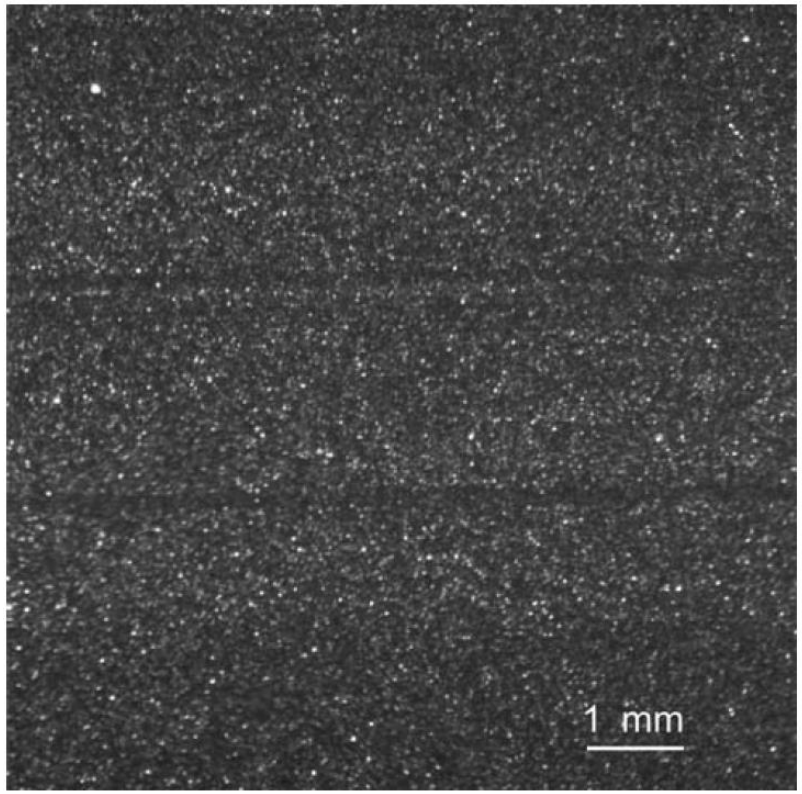
- Argon Laser (≈ 100 $W \cdot cm^{-2}$) and CMOS Camera
- Introducing of a H_2:He – mixture directly into the liquid helium (Amount of mixture in the deciliter range)

→ 1H_2 : 5He particlesize > 30 µm
particle size shrinks as the H_2 is diluting

→ 1H_2 : 10He 0.013 cm^3 solid hydrogen: 6 ppm_v (± 30 %), polydisperse Dispersion

→ 1H_2 : 500He No optical resolution of particles

- Estimation of characteristic particle size: d_c = 1.8 µm (± 30 %)



Bewley et al.: image of hydrogen particles taken about 1 min after being generated in liquid helium at 4.2 K using hydrogen gas diluted mixture 1H2 : 10He

Conclusion

- H_2-contaminated LHe is a known problem in many cryogenic laboratories
- The thermodynamic behavior of this binary mixture over a certain concentration and temperature range is poorly understood
- The measurement-based determination of the solubility limit of H_2 in LHe is the aim of this work and part of the project HyLiqHe
- A cryostat for condensing and conditioning an H_2-contaminated helium sample has been designed and is currently under assembly
- Adequate analytics is used
- A laser diffraction method is being considered for the detection of precipitation or solubilisation of H_2

Thank you for your attention

[1] Will J and Haberstroh C 2020 Hydrogen contamination in liquid helium *IOP Conf. Ser. Mater. Sci. Eng.* **755** pp 01211
[2] Abrikosova II and Shal´nikov AI 1971 The purity of liquid helium *Cryogenics* **11** pp 137 - 138
[3] Bewley GP, Sreenivasan KR and Lathrop DP 2008 Particles for tracing turbulent liquid helium *Experiments in Fluids* **44** pp 887 – 896