Status of the Distillation Apparatus

S. Bouchigny
IPN Orsay – I3HP

- Introduction to solid HD target Polarization
- Distillation apparatus at Orsay
- First test run and results
- Improvement of the Apparatus

Static Polarization of HD targets

Solid HD at low temperature (Honig, 1967)

Polarized HD Molecule

High relaxation time at ~1K temperature

Lattice of the HD crystal

Good for DNP.
Problematic for Static polarization (target unpolarizable)
Static Polarization of HD targets

(Honig, 1967)

Polarized HD Molecule

Spin spin interaction

Lattice of the HD crystal

Target can be polarized

Temperature and field: ~10 mK - ~15 T

ortho-H2 → 6.4 days → para-H2

Note: The diagram illustrates the polarization process of HD targets, highlighting the role of spin spin interaction and the lattice of the HD crystal. The target can be polarized under specific temperature and field conditions.
Static Polarization of HD targets

(Honig, 1967)

Polarized HD Molecule

High relaxation time at ~1K temperature

Lattice of the HD crystal

para-H2

18.2 days decay

para-D2

ortho-D2
### Static Polarization of HD targets

**The three steps for static polarization**

<table>
<thead>
<tr>
<th>I</th>
<th>II</th>
<th>III</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dilution refrigerator</td>
<td>Transport cryostat</td>
<td>In Beam cryostat</td>
</tr>
<tr>
<td>10 mK</td>
<td>2 – 4 K</td>
<td>0.2 - 1.5 K</td>
</tr>
<tr>
<td>13-15 Tesla</td>
<td>1 - 2 T</td>
<td>1.5 Tesla</td>
</tr>
</tbody>
</table>

**Static polarization + Aging (oH₂ → pH₂)**

**Transportation**

**On site exploitation**

**Initial concentration Needed**

<p>| | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂: 0.1 0.2 %</td>
<td>D₂: &lt; 0.01%</td>
<td>Need to purify HD</td>
</tr>
</tbody>
</table>

Maximum purity of commercial HD ~0.6 % for both H₂ and D₂

### Dynamic Polarization of HD targets

**Adding impurities: free electrons.** For B=2.5 T and T = 1 K, e⁻ polarization = 92%

![Dynamic Polarization Diagram](image)

**Initial concentration Needed**

<p>| | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂: &lt; 0.05 %</td>
<td>D₂: &lt; 0.05%</td>
<td>Need to purify HD</td>
</tr>
</tbody>
</table>
**Rectification techniques**

Rectification

![Diagram](image1)

Separation using relative volatility of elements

Favorable for H₂ HD D₂

![Graph](image2)

Vapor pressure ratio α

**Orsay Still**

![Diagram](image3)

Three extraction points

Three temperature probes

To the mass spectrometer or extraction tanks

Stainless steel column with Stedman packing
Quadrupole Mass Spectrometer

Extraction tanks

Manifold of the still

Three Extraction Valves

Still

Quadrupole Mass Spectrometer

Extraction tanks
Efficiency of the column is given by:

Number of Theoretical Stages: \( \text{NTS} \)

\[
\text{NTS} = \frac{\ln(x_c(1-x_b)/x_b(1-x_c))}{\ln(\alpha)} - 1 \quad \text{(Fenske Relation)}
\]

Control of the distillation

\( \text{Pc} = \text{Power in the condenser} \)

\( \text{Pb} = \text{Power in the boiler} \)

\( \text{Pc + Pb control the operating temperature} \)

Control the reflux of liquid and vapor

---

Sample: \([\text{H}_2] = 0.5\% \quad [\text{D}_2] = 0.6\%\)

---

Working at lowest boiling rate and lowest temperature

Concentration of H\(_2\) \(\times 32\)
### Still at Work

#### III Extracting the HD. Initial sample: $[\text{H}_2] = 0.5\%$ $[\text{D}_2] = 0.6\%$

![Graph showing %H2 and %D2 over time from 23-avr to 23-mai](image)

<table>
<thead>
<tr>
<th></th>
<th>H2 rich</th>
<th>H2 poor</th>
<th>D2 rich</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.44 moles extracted: $[\text{H}_2] = 2.46%$ $[\text{D}_2] = 0.157%$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>3.5 moles extracted: $[\text{H}_2] = 0.08%$ $[\text{D}_2] = 0.49%$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>1.5 moles extracted: $[\text{H}_2] = 0.02%$ $[\text{D}_2] = 2.52%$</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

#### IV Double Distillation

![Graph showing %H2 and %D2 over time from 0 to 10 days](image)

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>334 mmoles extracted: $[\text{H}_2] &lt; 0.02%$ $[\text{D}_2] = 0.17%$</td>
</tr>
</tbody>
</table>

DNP sample
Relaxation Time of HD Sample

Relaxation time was measured in Bochum in July 2005 under radiation (E. Radke et al., Bochum).

- **High relaxation time without aging**
  - No lost of relaxation time

Optimum radiation dose in 7 days

Improvement of the Still

Increasing the efficiency of the column:

- Typical results:
  - NTS = 4
  - NTS = 0.3
  - NTS = 0

- No distillation between the bottom and the middle part of the distillator

- Need to change the design of the column
- Test of classical plates in the bottom part to retain more liquid in the column
Improvement of the Still

Increasing the efficiency of the column:

- Typical results:
  - NTS = 4.6
  - NTS = 1.35
  - NTS = 0

- Distillation between the bottom and the middle part of the still but less efficient on top.

- + 20% efficiency

Second version

Improvement of the Apparatus

Installation of an NMR apparatus in Orsay using a variable temperature cryostat.

- Cryostat can run from 1.5 K to 30 K
- Field up to 2 Tesla
- We made a new insert equipped with NMR coils contains 200 mmoles of HD.

NMR System is ready.
Summary and Outlooks

- More than 1 month of aging saved for static polarization
- Promising results on relaxation time for Dynamic polarization.
- Test of new configuration of the column packings gives +20% efficiency
- Systematic measurements of relaxation time vs. initial concentration of H₂ and D₂ ready to start next week.

Pure HD Target HYDILE

Relaxation time

\[
\begin{align*}
[H_2] &= 0.26 \pm 0.02 \\
[D_2] &= 0.23 \pm 0.02
\end{align*}
\]
HD solid state polarized targets offer:

**High Dilution Factor**: All nucleus are polarizable

**Long relaxation time**: Nuclear spin – lattice coupling switch off

**How to polarized HD target?**

**Static polarization.**

**Dynamic Nuclear Polarisation**

**What quality of HD do we need?**

### Concentration Measurements

**MKS Microvision Plus Quadrupole Mass Spectrometer**

- Analyse Mass from 1 to 6
- Measure [H2] down to $2 \times 10^{-4}$
- Measure [D2] down to $10^{-5}$