Metastability exchange optical pumping in $^3$He gas up to 30 mT: Efficiency measurements and evidence of laser-induced relaxation

PhD defence Marion Batz, July 8th, 2011
Applications of hyperpolarised $^3$He

- hyperpolarisation = nuclear polarisation $M$ enhanced relative to thermal equilibrium ($\propto B/T$, $\sim 10^{-9}$ @ mT, 300 K)

$^3$He: $l = \frac{1}{2}$

$$M = \frac{\frac{N^+}{g} - \frac{N^-}{g}}{N_g}$$
Applications of hyperpolarised $^3$He

- **hyperpolarisation** = nuclear polarisation $M$ **enhanced** relative to thermal equilibrium ($\propto B/T$, $\sim 10^{-9}$ @ mT, 300 K)
- **hyperpolarised $^3$He**: versatile **tool** in different fields of **fundamental physics** and **biomedical science**
- Examples of **applications**:
  - **spin filters** for polarising neutrons
  - **scattering targets** for investigations of the **structure of nucleons**
  - **nuclear precession magnetometers** e.g., to probe **fundamental symmetries** (Lorentz and CPT violation), to search for **permanent EDM**
  - **investigations of nonlinear NMR dynamics** in **hyperpolarised liquid $^3$He**
  - **magnetic resonance imaging (MRI)** of the lung in humans and in animals with inhaled gas

$^3$He: $I = \frac{1}{2}$

![Hyperpolarised 3He](image1)

- **healthy**
- **lung transplant patient – 3D**
- **asthma**

Duke
Mainz
Virginia Univ.
Motivation

- **Goal**: contribute to understanding of current limitations of $^3$He MEOP to ultimately overcome them and achieve highest possible polarisation.

- Especially for applications in fundamental physics: high nuclear polarisation $M$ is crucial.
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- Especially for applications in fundamental physics: high nuclear polarisation $M$ is crucial: figures of merit vary non-linearly with $M$!

  - **Spin filters for neutrons:**
    \[
    \text{transmission} \propto \cosh(\Omega M) \\
    \text{neutron polarisation} \propto \tanh(\Omega M)
    \]
    ($\Omega$: filter opacity)

  - **Scattering targets:**
    \[
    \text{figure of merit} \propto M^2 \\
    \text{measurement time} \propto 1/M^2
    \]

    e.g., reduction of beam time by a factor of 2 by increasing $M$ from 0.5 to 0.7 (to obtain data with given statistical uncertainty)

  Lelièvre-Berna *et al.* (2007)

  Krimmer *et al.* (2009)
**Motivation**

- **Goal:** contribute to *understanding of current limitations of $^3$He MEOP* to ultimately overcome them and achieve highest possible polarisation.

- Especially for applications in fundamental physics: high nuclear polarisation $M$ is crucial: figures of merit vary *non-linearly* with $M$!

  - **Spin filters for neutrons:**
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    - e.g., reduction of beam time by a factor of 2 by increasing $M$ from 0.5 to 0.7 (to obtain data with given statistical uncertainty)

- For applications in MRI:
  - increase *polarisation and production rate* for higher throughput.
Specificities of $^3\text{He}$ Metastability Exchange Optical Pumping (MEOP)

MEOP in moderate magnetic fields $B \leq 30$ mT

Experimental Setup and measurement of nuclear polarisation

Effect of magnetic field on plasma and OP performances

Global angular momentum budget approach

Laser-induced relaxation

Discussion: Physical process possibly involved?

Summary and Outlook
Specificities of OP in $^3$He:

- OP *not* performed from ground state: populate metastable $^2^3S$ state by rf discharge ($n_m/N_g \approx 1$ ppm)

- OP on $^2^3S$-$^2^3P$ transition (1083 nm): usually on $C_8$ or $C_9$ line; strong hyperfine interaction in $^2^3S$ and $^2^3P$: nuclear and electronic spin states simultaneously oriented

- Transfer of *nuclear* orientation from $^2^3S$ to ground state by metastability exchange collisions
Metastability exchange (ME) collisions

- involve 1 ground state (g.s.) atom + 1 $2^3S_1$ atom
- swap spin states of atoms
- preserve total spin
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Consequences:
- ground state:
  evolution of g.s. nuclear polarisation is governed by $2^3S$ polarisation $M^* = 2 <I_z>* / \hbar$

- metastable state:
  spin temperature distribution enforced in $2^3S$ (without OP) → used for absolute measurement of $M$ by 2 complementary light absorption rates, measured by weak probe laser beams

Rate $\gamma_e$ 
$3.8 \times 10^6$ s$^{-1}$ / mbar

Nuclear spin in g.s. and total spin in $2^3S$ strongly coupled

$M^* > M$ with OP

\[
\frac{dM}{dt} = \gamma_e \left( \frac{2 \langle I_z \rangle^*}{\hbar} - M \right) - \Gamma_R M
\]

metastability exchange relaxation
MEOP in moderate magnetic fields (≤ 30 mT)

Effects of moderate magnetic field:

- Structure of sublevels in $2^3S$ and $2^3P$ and transitions unaffected
- In $2^3S$ and $2^3P$: hf-coupling unaffected below 0.1 T ($A_{\text{HFS}} \approx 4400$ MHz)
MEOP in moderate magnetic fields (≤ 30 mT)

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- In $2^3S$ and $2^3P$: hf-coupling unaffected below 0.1 T ($A_{\text{HFS}} \approx 4400 \text{ MHz}$)
- Above 10 mT: In higher exited states: hf-decoupling ($A_{\text{HFS}} (3^1D): 136 \text{ MHz}$)

Angular momentum loss reduced in the cascade

Expected to yield higher OP performances

\[ R = \frac{\text{circular polarisation of 668 nm-line (3}^1\text{D}_2 \rightarrow 2^1\text{P}_1)}{\text{g.s. nuclear polarisation}} \]
Structure of talk

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- MEOP in moderate magnetic fields $B \leq 30$ mT

- Experimental Setup and measurement of nuclear polarisation
- Effect of magnetic field on plasma and OP performances
- Global angular momentum budget approach
- Laser-induced relaxation

- Discussion: Physical process possibly involved?
- Summary and Outlook
Experimental setup

Plasma in cell produced by rf discharge:

Magnetic field (0-30 mT) produced by solenoid:
Al bore cylinder and edge flasks with water cooling (constructed at Mainz University)
uniform B over OP cells (30 cm):
\[ T_{f,magn} \approx 435 \text{ h (@ 1 mbar)} \]

Electrode configuration optimised to obtain large range of plasma conditions

Pump laser:
broadband fibre laser (5 W, 1.7 GHz FWHM)

Probe laser:
fibre-coupled single-frequency DBR diode
- Double pass configuration with back-reflecting high quality mirror to increase absorption
- Gaussian pump beam, circularly polarised, expanded to 1.4 cm FWHM diameter
- Weak probe beam: two adjacent beams with opposite circular polarisations
- Double modulation scheme for high SNR
Measurement of nuclear polarisation

ME collisions tend to enforce a **spin-temperature distribution** in $2^3S$, ruled by $1^1S$ nuclear polarisation $M$:

$$a \frac{(m_F + 1)}{a \ (m_F)} = e^{\beta} = \frac{N_+}{N_-} = \frac{1 + M}{1 - M}$$

1/β: spin temperature

$a_i$: relative population of $2^3S$ sublevel $A_i$

![Diagram](image)

<table>
<thead>
<tr>
<th>$m_F$</th>
<th>$2^3S_1$, $F=1/2$</th>
<th>$2^3S_0$, $F=1/2$</th>
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<td>-1/2</td>
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<td>$A_1$</td>
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<tr>
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</tr>
<tr>
<td>1</td>
<td>$x^3$</td>
<td>$x^3$</td>
</tr>
</tbody>
</table>

**Two absorption rates measured:**

- $\sigma^+$
- $\sigma^-$

→ **populations** and $n_m$

Infer $M$ from the two selectively probed populations

**Systematic tests:** Method is reliable for the study of OP dynamics even with redistribution of populations by OP light
Important measured quantities

Nuclear polarisation AND pump transmission continuously monitored

Typical SNR: pump: 5-35 depending on $\tau_{\text{LIA}}$
probe: 500-1000 in presence of pump laser; 10000 without pump laser

Absorbed pump laser power can be inferred at all times:
$W_{\text{abs}} = (1 - T_p) W_{\text{inc}}$

Monoexponential decay: $\Gamma_R = \text{cst.} = \Gamma_D$
$\Gamma_D^{-1}: 1 \rightarrow 1000 \text{ s, depending on plasma conditions}$
Effect of magnetic field on plasma

1 mT results

\[ n_m^S(0) \left[ 10^{16} \text{ at/m}^3 \right] \]

\[ \Gamma_D \left[ \text{s}^{-1} \right] \]

\[ N_g \approx 10^{22} \text{ at/m}^3 @ 1 \text{ mbar} \]
Effect of magnetic field on plasma

Significant increase / decrease of $\Gamma_D$ at fixed $n_m$ in $B = 30$ mT

$N_g \approx 10^{22}$ at/m$^3$ @ 1 mbar
Effect of magnetic field on OP performances

OP at fixed pump laser power

$M_{eq}$ vs $n_m^S(M=0) \times 10^{16} \text{ at/m}^3$

- OP 1.66 W
- C8
- C9
- Open: 1 mT
- 0.63 mbar
- 2.45 mbar
Effect of magnetic field on OP performances

Steady state polarisation in 30 mT:
NOT significantly improved
Effect of magnetic field on OP performances

- Steady state polarisation in 30 mT: NOT significantly improved
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Rate of change $dM/dt$ at $M=0$ as function of absorbed pump laser power at $M=0$: identical behaviour at $B=1$ and 30 mT
Effect of magnetic field on OP performances

- Steady state polarisation in 30 mT: NOT significantly improved
- Rate of change $dM/dt$ at $M=0$ as function of absorbed pump laser power at $M=0$: identical behaviour at $B=1$ and 30 mT
- NEW different way to investigate involved MEOP processes required

although decay rates $\Gamma_D$ are modified: APPARENT PARADOX
**Global angular momentum budget approach - I**

**global budget: growth rate = gain - loss**

**ME collisions: angular momentum conserved**
Global angular momentum budget approach - II

Balance of angular momentum for ground state atoms

\[
\frac{dM}{dt} = 2\eta \frac{W_{abs}}{\hbar \omega} \frac{1}{N_g V_c} - \Gamma_R \left( M \right)
\]

- \( \eta \): photon efficiency
- \( \Gamma_R \): global polarisation loss rate \((M \neq 0)\)

may vary with \( M \) and MEOP conditions

- deposited orientation per absorbed photon
- number of absorbed photons per unit time
- number of atoms

measured quantities
Photon efficiency - I

Definition of photon efficiency (PE) $\eta$: net change of atomic angular momentum projection $m_F$ in the $2^3S$ state, upon absorption and emission of a photon

$\eta$ (at given transition) depends on the degree of collisional mixing in $2^3P$:
Photon efficiency - I

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\( \eta \) (at given transition) depends on the degree of collisional mixing in \( 2^3P \):

Kastler OP regime
no collisional redistribution
low pressure

\[ \eta^{K_{C8}} \approx 0.9 \]
Photon efficiency - I

Definition of photon efficiency (PE) $\eta$: net change of atomic angular momentum projection $m_F$ in the $2^3S$ state, upon absorption and emission of a photon

$\eta$ (at given transition) depends on the degree of collisional mixing in $2^3P$:

<table>
<thead>
<tr>
<th>Kastler OP regime</th>
<th>Dehmelt OP regime</th>
</tr>
</thead>
<tbody>
<tr>
<td>no collisional redistribution</td>
<td>full collisional redistribution between all 18 sublevels in $2^3P$</td>
</tr>
<tr>
<td>low pressure</td>
<td>high pressure</td>
</tr>
</tbody>
</table>

Emissied/scattered light globally unpolarized

$\eta^K_{C8} \approx 0.9$

$\eta^D_{C8} = 0.5$
Photon efficiency - II

Dependencies

<table>
<thead>
<tr>
<th></th>
<th>$\eta_{C_8}$</th>
<th>$\eta_{C_9}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$M$</td>
<td>no</td>
<td>yes</td>
</tr>
<tr>
<td>$W$</td>
<td>no</td>
<td>yes</td>
</tr>
<tr>
<td>pressure</td>
<td>yes</td>
<td>yes</td>
</tr>
</tbody>
</table>

$M=0, B=0, W_{inc} \rightarrow 0$

Radiative lifetime of $2^3P$

$\tau_P [s]$ vs $M$ for $C_9$ and $C_8$

- $C_9(M=0)$
- $C_9(M=0.5)$
- $C_9(M=0.8)$
- $C_9(M=0.995)$
- $C_8$ (all $M$)

Dehmelt
- $1.25E-7$ to $5E-7$ s
- 2.45 mbar

Kastler
- 0.63 mbar
Inferring total polarisation loss rates $\Gamma_R$

$C_9$: photon efficiency NOT constant

$\rightarrow$ use MEOP model to compute $\eta$ and infer polarisation loss rates

$C_8$: direct use of balance of angular momentum with measured $\eta$

- at $M=0$: relaxation loss vanishes: $-\Gamma_R M = 0$
  photon efficiency $\eta$ can be directly measured

- at $M_{eq}$: rate of change $dM/dt = 0$

- during polarisation build-up, total polarisation loss rates $\Gamma_R$ can be inferred dynamically from the full equation at all times

\[ \frac{dM}{dt} = 2\eta \frac{W_{abs}}{\hbar \omega} \frac{1}{N_g V_c} - \Gamma_R M \]

\[ \eta = \frac{1}{2} \frac{dM}{dt} (0) \frac{N_g V_c \hbar \omega}{W_{abs} (0)} \]

\[ \Gamma_R (M_{eq}) = \frac{W_{abs} (M_{eq})}{W_{abs} (0)} \frac{dM}{dt} (0) \frac{M_{eq}}{M_{eq}} \]

faster relaxation \quad lower $M_{eq}$ \quad smaller $dM/dt$
Laser-induced relaxation - I (C₈ transition)

C₈ data: angular momentum budget approach

\[ \Gamma_R [s^{-1}] \]

\[ \Gamma_D \]

\[ W_{abs} [W] \]

\[ W_{inc} [W] \]

\[ W_{abs} (M_{eq}) [W] \]
Compilation of $\text{C}_8$ data, $B=1$ mT: $\Gamma_L$ inferred from directly measured quantities only, using angular momentum budget approach.

Consistent set of data obtained when plotted as function of $W_{\text{abs}}$:

$$\Gamma_L \propto W_{\text{abs}}$$

Laser-induced relaxation rates exceed those measured in the plasma by up to two orders in magnitude in our experimental conditions.

Additional laser-induced loss rate

$$\Gamma_L = \Gamma_R - \Gamma_D$$
Systematic discrepancies (observed for C₈ and C₉ pumping) \( \rightarrow \) polarisation losses are STRONGLY underestimated during build-ups

IMPOSSIBLE to further increase steady-state polarisation (\( \rightarrow \) plateau) by further increasing pump laser power

\[
M_{eq} \propto \eta \frac{W_{abs}}{\Gamma_R}
\]
Laser-induced relaxation – III ($C_9$ transition)

$\Gamma_L$ inferred using OP model (data at $M_{eq}$ only)
Laser-induced relaxation – III (C$_9$ transition)

- $\Gamma_L$ inferred using OP model (data at $M_{eq}$ only)
- For identical plasma conditions: comparable effect both for C$_8$ and C$_9$ pumping lines
Laser-induced relaxation – III (C\textsubscript{9} transition)

- \( \Gamma_L \) inferred using OP model (data at \( M_{eq} \) only)
- For identical plasma conditions: comparable effect both for C\textsubscript{8} and C\textsubscript{9} pumping lines
- Strong discharge: good agreement with commonly observed proportional behaviour of \( \Gamma_L \) with respect to \( W_{abs} \)
- \( \Gamma_L \) still exceeds \( \Gamma_D \) by up to one order in magnitude

\[ \begin{aligned}
\text{2.45 mbar, } B=1 \text{ mT}
\end{aligned} \]
Laser-induced relaxation – IV ($B = 1$ and $30$ mT)

- Come back to astonishing observations at $B=30$ mT:
  - NO increase of $M_{eq}$ and
  - NO change of $dM/dt$
Laser-induced relaxation – IV ($B = 1$ and 30 mT)

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Laser-induced relaxation – IV ($B = 1$ and $30 \text{ mT}$)

- Come back to astonishing observations at $B=30 \text{ mT}$: NO increase of $M_{eq}$ and NO change of $dM/dt$ although decay rates $\Gamma_D$ are reduced.

- Same consistent behaviour of $\Gamma_L$ observed (build-up dynamics and at $M_{eq}$), independently of magnetic field $B$.

- Clarification: only at very small incident pump laser powers, $\Gamma_D$ basically determines obtainable $M_{eq}$; at higher $W_{inc}$ (= relevant cases in practice), $\Gamma_D$ is NOT the pertinent parameter limiting $M_{eq}$; but $\Gamma_L$. 

\[ \begin{align*}
\Gamma_L [s^{-1}] & \quad \Gamma_D (1 \text{ mT}) \\
W_{abs} [W] & \quad \Gamma_D (30 \text{ mT}) \\
2.45 \text{ mbar, OP C8} & \\
\text{filled: } 30 \text{ mT} & \quad \text{open: } 1 \text{ mT}
\end{align*} \]
Laser-induced relaxation - VI (different works)

\[ \Gamma_L \text{ [s}^{-1}] \]

low \( B \) (1-3 mT)

- ○ 2.45 mbar

this work

- \( \Gamma_D \)

\[ \frac{W_{abs}}{V_c} \text{ [W/cm}^3] \]

30 cm x 6 cm
Laser-induced relaxation - VI (different works)

\[ I_L [s^{-1}] \]

\[ W_{abs} / V_c [W/cm^3] \]

Abboud 2005

Abboud 2005


5 cm x 5 cm
Laser-induced relaxation - VI (different works)

**low \( B \): consistent overall qualitative behaviour:**

\[ W_{\text{abs}} \text{ per unit volume} \propto \text{laser-induced relaxation} \]

**low \( B \), high pressure data:**
- extend range of \( W_{\text{abs}} / V_c \) considerably
- in good agreement with all other data

\[ 1E-7 \quad 1E-6 \quad 1E-5 \quad 1E-4 \quad 0.01 \quad 0.1 \]
\[ 1E-4 \]
\[ \Gamma_L [s^{-1}] \]
\[ W_{\text{abs}} / V_c [W/cm^3] \]

Glowacz 2011
strong dc
weak dc

In collaboration with:

Laser-induced relaxation - VI (different works)

Ubiquitous phenomenon: observation of similar effects in other MEOP experiments as well.

- high $B$: at fixed $W_{abs} / V_c$
  - $\Gamma_L$ smaller than in low $B$
  - high $M_{eq}$ values can be recovered
- non negligible: $\Gamma_L$ at least of order $\Gamma_D$ as well
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- Specificities of $^3$He Metastability Exchange Optical Pumping (MEOP)
- MEOP in moderate magnetic fields $B \leq 30$ mT

- Experimental Setup and measurement of nuclear polarisation
- Effect of magnetic field on plasma and OP performances
- Global angular momentum budget approach
- Laser-induced relaxation

- Discussion: Physical process possibly involved?
- Summary and Outlook
Discussion – I (radiation trapping)

Physical processes possibly causing OP-enhanced relaxation - I

Radiation trapping: light resulting from spontaneous emission from the $2^3P$ state (not $\sigma^+$ polarised) might be absorbed by metastable atoms in $2^3S$ before exiting the cell

Main expected features of additional reabsorption-induced loss rates:

✔ should scale with $W_{abs}/V_c$

✗ should strongly decrease at high $M$

✗ very small quantitative impact on $M_{eq}$ values expected

✗ should scale as $n_m/p_3$

{Eckert et al. (1992)}
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Eckert et al. (1992)

\[
\frac{\Gamma}{W_{abs}} V_c \quad \text{[10}^{\text{-3} \text{ s}^{-1} \text{ W}^{-1}]}
\]

Ratio of polarisation loss rate to absorbed pump power, scaled to cell volume at $M_{eq}$ as function of metastable density:

- data are clearly not proportional to $n_m$; in spite of scatter: mild increase with $n_m$
- observed to weakly decrease with gas pressure, excludes $1/p_3$ scaling
**Discussion – II (plasma “poisoning” e.g., by He₂*)**

Physical processes possibly causing OP-enhanced relaxation - II

- **Light-enhanced creation of long-lived relaxing species through 2³P state:** e.g., metastable molecules He₂⁺ formed in 3-body collisions

  \[ \text{O} + \text{O} + \text{He} \rightarrow \text{He}_2 + \text{O} \]

  \[ \sigma \text{ from } 2^3P = 100 \times \sigma \text{ from } 2^3S \]

- **Relaxation through spin exchange?**
  and fast **dissipation** of nuclear angular momentum in **molecular internal degrees of freedom** (numerous rotational states)
Light-enhanced creation of long-lived relaxing species through $2^3P$ state: e.g., metastable molecules $\text{He}_2^*$ formed in 3-body collisions

$\sigma$ from $2^3P = 100 \times \sigma$ from $2^3S$  

Relaxation through spin exchange?
and fast dissipation of nuclear angular momentum in molecular internal degrees of freedom (numerous rotational states)

To account for our observations: $I_L$ should be proportional to molecular density:

- formation scales with $W_{\text{abs}}/V_c$ (through $2^3P$ state density)
- formation scales with $p_3^2$ (through $1^1S$ state density)
- slower decay at higher pressure: diffusion rate $\propto 1/p_3$
- should decrease with increasing $n_m$ (Penning collisions)

Measurements of laser-induced enhancement of molecular density: grossly insufficient to explain huge loss rates observed in low field and low pressure MEOP
Summary

- Observations in $B \leq 30 \text{ mT}$: polarisation decay rates $\Gamma_D$ modified in plasma, but OP performances not improved at high laser powers $W_{inc}$

  → apparent paradox clarified:
  - at low $W_{inc}$: $\Gamma_D$ basically determines obtainable steady state polarisation $M_{eq}$
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    - at higher $W_{inc}$: $\Gamma_D$ is **NOT** the pertinent parameter limiting $M_{eq}$

- Clear evidence of additional OP-induced relaxation effects:
  - NEW approach: balance of gains and losses in terms of angular momentum
  - efficiency of MEOP process measured in terms of absorbed pump laser power
Observations in $B \leq 30$ mT: polarisation decay rates $\Gamma_D$ modified in plasma, but OP performances not improved at high laser powers $W_{inc}$

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Clear evidence of additional OP-induced relaxation effects:
NEW approach: balance of gains and losses in terms of angular momentum
→ efficiency of MEOP process measured in terms of absorbed pump laser power

Main observed features of additional loss rates $\Gamma_L$:
- **linear scaling** with $W_{abs}/V_c$
- exceed decay rates $\Gamma_D$ by up to **two orders in magnitude**
Perspectives

- **Physical processes** causing OP-enhanced relaxation effects remain to be elucidated.
- **Further investigations** needed: e.g., at higher pressure and higher magnetic field: sublevel structure and transitions change.
- **Online monitoring of absorbed pump laser power** via pump transmission coefficient is essential.
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- **Online monitoring of absorbed pump laser power** via pump transmission coefficient is essential.

- **Reducing and ultimately eliminating** this source of relaxation would increase MEOP performances, possibility to increase polarisation and production rates of polariser units (low and high magnetic field).

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Low B polariser, Mainz

High B polariser, Kraków, G. Collier (2011)
He plasma gallery (from Grenoble, Mainz, Paris, Vancouver)

Merci à tous ! Herzlichen Dank !
References

[1] Final revised version of PhD thesis manuscript will be available online at http://tel.archives.ouvertes.fr and http://archimedes.uni-mainz.de (from October 2011 on)
[16] B. Głowacz, ongoing PhD thesis, LKB and Jagiellonian University Kraków ($\Gamma_L$ high pressure and low field, measurements of laser-induced enhancement of molecular density)
[17] G. Collier, ongoing PhD thesis, Jagiellonian University Kraków ($\Gamma_L$ high field and high pressure)