Optical excitation of molecules for Spin-Polarized Nuclear Fusion

T. Peter Rakitzis
Department of Physics, University of Crete and IESL-FORTH
Institute of Electronic Structure and Laser
Foundation for Research and Technology - Hellas
Fusion polarization dependence for (1) and (2)

(1) $^2\text{D} + ^3\text{T} \rightarrow ^5\text{He} \ (I=3/2) \rightarrow ^4\text{He} + \text{n}$

(2) $^2\text{D} + ^3\text{He} \rightarrow ^5\text{Li} \ (I=3/2) \rightarrow ^4\text{He} + \text{p}^+$

$\sim 20 \text{ keV}$

$I_D=1$ $\otimes$ $I_T=1/2$ $\Rightarrow$ $I_{\text{He}}=1/2$

$\sim 3 \text{ MeV}$

$I_{\text{He}}=3/2$

4/6 of unpolarized m-state combinations give $I=3/2$
2/6 of unpolarized m-state combinations give $I=1/2$
Fusion polarization dependence for (1) and (2)

(1) $^2\text{D} + ^3\text{T} \rightarrow ^5\text{He} \ (I=3/2) \rightarrow ^4\text{He} + \text{n}$

(2) $^2\text{D} + ^3\text{He} \rightarrow ^5\text{Li} \ (I=3/2) \rightarrow ^4\text{He} + \text{p}^+$

Only 4/6 of unpolarized m-state combinations can react.
Fusion polarization dependence for (1) and (2)

(1) $^2\text{D} + ^3\text{T} \rightarrow ^5\text{He} \ (I=3/2) \rightarrow ^4\text{He} + \text{n}$

(2) $^2\text{D} + ^3\text{He} \rightarrow ^5\text{Li} \ (I=3/2) \rightarrow ^4\text{He} + \text{p}^+$

100% of polarized reactants give $I=3/2$

(1) Polarization increases rate by 50%

(2) Directional neutron distribution $\sim \sin^2\theta$

(3) Together, reactor efficiency may improve by up to a factor of 2.
Open questions in Polarized Fusion

(1) Does the polarization survive the ~20 keV plasma?
(2) What is the polarization dependence of DD fusion?
(3) Can $10^{22}$ pol-D/s be produced, which is needed for fusion reactors (ITER)?
Standard Methods of SPH production

- Strong B-field/cryogenic cooling, DNP
- Stern-Gerlach separation
- Optical-pumping
Strong B-field/cryogenic cooling

$D_2$ cooled to 25 mK (2 weeks) with 15T B field:

- Polarization only $\sim 20\%$
- Takes too long
- Makes too little

Unclear how the $D_2$ “ice” can be introduced into a reactor, without depolarization
SPH production at BNL (Stern-Gerlach technique)

$10^{17}$/s $<< 10^{22}$/s

needed for a fusion reactor
Current Methods of SPH production

- **Optical-pumping** Limited to alkali atoms (and noble gases and SPH via Spin-exchange optical pumping)

\[
\begin{align*}
10^{17}/s & \ll 10^{22}/s \\
\text{needed for a fusion reactor}
\end{align*}
\]
Each technique has significant drawbacks.

Laser excitation of molecules offers advantages compared to traditional methods

Two new methods:

(1) UV photodissociation of Hydrogen Halides

(2) IR rovibration excitation of molecules
(1) Molecular Photodissociation

Adiabatic correlation of molecular electronic states to specific atomic m states


\[
AB(\Omega_g=0) + hf \rightarrow AB(\Omega_i=1) \rightarrow A(J_A,m_A) + B(J_B,m_B)
\]

Circularly Polarized

e.g. HI

\[
\begin{align*}
\Omega_i & \rightarrow |m_A,m_B\rangle \\
|+1\rangle_{A^1\Pi} & \xrightarrow{R \to \infty} |-\frac{1}{2},+\frac{3}{2}\rangle \\
|+1\rangle_{a^3\Pi} & \xrightarrow{R \to \infty} |+\frac{1}{2},+\frac{1}{2}\rangle \\
|0\rangle_{a^3\Pi} & \xrightarrow{R \to \infty} |\pm\frac{1}{2},\pm\frac{1}{2}\rangle
\end{align*}
\]
(1) Molecular Photodissociation

Adiabatic correlation of molecular electronic states to specific atomic m states


\[ \text{AB}(\Omega_g=0) + h\nu \rightarrow \text{AB}(\Omega_i=1) \rightarrow \text{A}(J_A, m_A) + \text{B}(J_B, m_B) \]

Circularly Polarized

e.g. HI

\[ \Omega_i \rightarrow |m_A, m_B\rangle \]

\[ |+1\rangle_{A^1 \Pi} \xrightarrow{R \to \infty} \left| -\frac{1}{2}, +\frac{3}{2}\right\rangle \]

\[ |+1\rangle_{a^3 \Pi} \xrightarrow{R \to \infty} \left| +\frac{1}{2}, +\frac{1}{2}\right\rangle \]

\[ |0\rangle_{a^3 \Pi} \xrightarrow{R \to \infty} \left| +\frac{1}{2}, \pm\frac{1}{2}\right\rangle \]
Production of highly-polarized atoms from molecular photodissociation

Advantages
• short timescales of production (100 fs dissociation)
• Transitions in UV, where powerful lasers exist (∼10^{22} phot/s)
• Efficient: 1 photon → 1 polarized H/D
• high-density (from stable molecules, e.g. DI, HBr, at high pressure)
Halogen atoms Polarization measured with Resonance Enhanced Multi-photon Ionization (REMPI)

I.P. \[ \rightarrow \] X$^+$

X(J' = 3/2)

m$_X' =$ -3/2  -1/2  +1/2  +3/2 

RCP (Δm=+1)

m$_X =$ -1/2  +1/2 

m-state selection

X(J=1/2) ground-state
HCl $\rightarrow$ H + Cl (J=1/2)

$\frac{I_{RCP}}{I_{LCP}} = 1.8 \pm 0.2$

$\Delta \beta_2 = -0.3 \pm 0.1$
HCl $\rightarrow$ H + Cl (J=3/2)

$$\frac{I_{RCP}}{I_{LCP}} = 1.4 \pm 0.2$$

$$\Delta \beta_2 = -1.0 \pm 0.1$$
Cl polarization from HCl photodissociation at 193 nm

(Diatomics understood)


Polarization parameters

Measure the SPH be seen **directly!**

- Quantization axis of SPH (z-axis)
- RCP or LCP probe light
- PMT
- Polarizer (parallel to z)
- Fluorescence Interaction region

Note: Quantization axis of excitation and detection are the same (z axis)
SPH fluorescence detection

H 2p\(^{(2P_{3/2})}\) \(\rightarrow\) H 1s \((2S_{1/2})\)

**Excitation** \((\Delta m_Z = +1)\)

**fluorescence allowed**

**fluorescence forbidden**

**Linearly Polarized Fluorescence** \((\Delta m_Z = 0)\)

Advantages:
1) Hyperfine resolution not necessary
2) Sensitive to SPH velocity (Doppler shift)
3) Detect on nanosecond timescale
4) Very sensitive

22.6 km/s

Probe wavelength (nm)
121.576 121.567 121.558

HBr

Signal intensity (arb. units)

LL
RL

22.6 km/s

(10 mTorr HBr)
19.3 km / s

(10 mTorr HCl)
Results

SPH detection
Infer from halide
Theory

SPH production at BNL (current density record)
SPH production and detection on Crete
## Comparison

<table>
<thead>
<tr>
<th></th>
<th>S-G (BNL)</th>
<th>Crete (lasers)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1) Density (SPH/cm³)</td>
<td>(10^{12})</td>
<td>(&gt;10^{18})</td>
</tr>
<tr>
<td>2) Production rate s(^{-1})</td>
<td>(10^{17})</td>
<td>(10^{19} - 10^{23})</td>
</tr>
<tr>
<td>3) Detection timescale</td>
<td>ms</td>
<td>ns</td>
</tr>
<tr>
<td>4) Velocity resolution</td>
<td>None</td>
<td>100 m/s</td>
</tr>
<tr>
<td>5) Polarization</td>
<td>0.92</td>
<td>0.45 (0.90)</td>
</tr>
</tbody>
</table>
“Single-molecule Stern-Gerlach Spin-Separator”

\[ L \approx 1 \text{ m} \]
\[ T \approx 1 \text{ ms} \]

Explains why density of polarized atoms can be many orders of magnitude higher than traditional S-G experiment

\[ L \approx 1 \text{ nm} \]
\[ T \approx 100 \text{ fs} \]

\( 10^{18} \text{ cm}^{-3} \) (1 ns) density estimated from H-Rb depolarization rate

H-halogen-atom depolarization unknown
How high can the SPH density reach?

A direct measurement is necessary, before applications can be considered.

Recently inspired by development of kJ-MJ laser pulses for laser fusion
Time-dependent magnetization measurements with pickup coil

Hyperfine beating in electron polarization and magnetization:

\[
\langle m_s(t) \rangle = \frac{1}{2} e^{-t/\tau_p} [1 - \alpha \sin^2(\omega t/2)]
\]

\[
\langle m_I(t) \rangle = \frac{1}{2} e^{-t/\tau_p} [\alpha \sin^2(\omega t/2)]
\]
Magnetization oscillation at hyperfine beat frequency

125 mbar DI / 875 mbar SF$_6$  

\[
\langle m_s(t) \rangle = \frac{1}{2} e^{-t/\tau_p} \left[ 1 - \alpha \sin^2 (\omega t / 2) \right]
\]

\[
\langle m_1(t) \rangle = \frac{1}{2} e^{-t/\tau_p} \left[ \alpha \sin^2 (\omega t / 2) \right]
\]

70 mbar HBr / 930 mbar SF$_6$

SPD depolarization rate vs. DI pressure

Depolarization saturating. Even higher densities possible?

\[
D^\uparrow + DI \xrightarrow{k_1}{k_1^{-1}} DI - D^\uparrow
\]
Complex formation

\[
DI - D^\uparrow \xrightarrow{k_d} DI - D
\]
Intramolecular depolarization

\[
DI - D^\uparrow + DI \xrightarrow{k_2} D^\uparrow + 2 DI
\]
Collisional dissociation DI

>10^{19} \text{ cm}^{-3} \text{ density (} \sim 10 \text{ ns lifetime)}

>100 \text{ time higher than expected}

\[
K = \frac{k_1 k_d [DI]}{k_1 + k_d + k_2 [DI]}
\]
SPD depolarization rate vs. $\text{SF}_6$ pressure: Evidence of DI-D complex destruction

$$DI-D^\uparrow + X \xrightarrow{k_2^X} D^\uparrow + DI + X$$

Collisional dissociation by inert gas X

$$K = \frac{k_1 k_d [\text{DI}]}{k_{-1} + k_d + k_2 [\text{DI}] + k_2^X [X]}$$
Signal vs. laser intensity and focusing
Summary I

- SPH densities of $10^{19} \text{ cm}^{-3}$ ($\sim 1 \text{ atm}$) depolarization rate flattens at high pressure.

- Future work: Go to higher pressures (5-10 atm), as even higher densities are likely possible.
Pump-probe polarized Fusion
(D-³He or D-T)

Fusion laser
2 MJ/shot
at NIF (Livermore, CA)

Fusion counts \(\approx 10^6 /\text{pulse}\)

\(\text{Phys. Rev. Lett. 118, 233401 (2017).}\)

Photodissociation laser

\([D\uparrow] > 10^{19} \text{ cm}^{-3}\)

\[ D(\theta, \phi) = \frac{\sigma_0}{3} [(2 + p) - (2p + p_{zz})P_2(\cos \theta)]/4\pi \]

Neutron angular distribution becomes anisotropic ONLY if polarization survives, \(p > 0\).

National Ignition Facility (NIF, Livermore CA)

2 MJ / (ns pulse)
Summary II

• UV photodissociation method good for generating high SPD densities for demonstration of polarized laser fusion (not for generating pellets for laser fusion).
Method 2: Pulsed IR rovibrational excitation of molecular beams

Polarizing molecular rotation (instead of electronic spin):

\( v'=1, J'=1 \) -> \( F'=2 \), \( F'=1 \), \( F'=0 \) (hyperfine states excited coherently)

\( v=0, J=0 \) -> \( F=1 \), \( F=0 \)

Pulsed Excitation of H$_2$

Always sum to 1

Rotational polarization of H$_2$(v=1,J=1)

Nuclear spin polarization of ortho-H$_2$
Pump-Probe (IR-UV) setup for measurement of hyperfine beating
Pulsed Excitation of HCl

H$^{35}$Cl(v=2,J=1)

$\langle M_{J_{HCl}} \rangle$

$\langle M_{J_{Cl}} \rangle$

Photodissociate here!

Nuclear spin polarization of $^{35}$Cl($^2P_{3/2}$)

Always sum to 1

Rotational polarization of H$^{35}$Cl(v=2,J=1)
Hyperfine beating in HD and D$_2$

HD($v=1, j=2, m=0$)

*Phys. Chem. Chem. Phys.*, 2009, 11, 142

Note: IR excitation not possible here, rovibrational excitation performed with 2-photon stimulated Raman pumping, of low efficiency.

D$_2$($v=1, j=2, m=0$)

*Phys. Chem. Chem. Phys.*, 2010, 12, 15689
Planning scaled-up experiments at Jefferson Lab

Gas inlet

Gas nozzle

IR Excitation beams
$10^{19} - 10^{22}$ photons/s
(W) - (kW)

HX, NH$_3$ isotopes

Location of maximum nuclear polarization

Freeze polarization in nuclei by:

1. Freezing molecules at surface, OR
2. Introduction a B-field, with $B > B_c$, OR
3. Photodissociate molecule

Planned spin-polarized production rate of $10^{19} - 10^{22}$ molecules/s
Fast neutral D beam for fusion reactor

D\textsuperscript{-} beam

Accelerated to desired energy (e.g. 300 keV)

\sim 10^{22} \text{ D/s}

532 nm photodetachment lasers
Proposal for $D^-$ beam production

\[ H^- + D \rightarrow H + D^- \]

Charge exchange efficiency demonstrated at 15%
Proposal for $D^-$ beam production

$H^- + D \rightarrow H + D^-$

Charge exchange efficiency demonstrated at 15%
High-power lasers and production rates

• Commercial appropriate IR and UV lasers exist with $10^{19}-10^{21}$ photon/s

• Assuming 1-10% total efficiency, $10^{18}-10^{20}$ D/s beams possible.

• Crucial to understand the efficiency of each step, in proof-of-principle experiment.

• Clearly (even if very efficient) $10^{22}$ D/s beams for ITER will require custom, industrial-scale lasers (100s of kW, which exist).
Conclusions

• Two new methods for production of spin-polarized molecules:

(1) UV photodissociation $\Rightarrow$ Demonstration of Polarized laser Fusion

(a) Large production rates of

(2) IR rovibrational excitation $\Rightarrow$ spin-polarized molecules

(b) Likely large production rates of spin-polarized D$^-$ and T$^-$ possible

All ideas are welcome
Acknowledgements

FORTH
Dimitris Sofikitis
Chrysovalandis Kannis

Alexander Andreev (Szeged, Berlin)

Juelich
Markus Büscher
Ralf Engels
Anna Hützen

Method 1

Jefferson Lab
Andy Sandorfi
Matt Poelker
Amy Sy

Method 2