Zeeman-Sisyphus deceleration of CaF molecules



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Motivation

Molecular properties







(1) times 🔒 Greater control of quantum state

Extend observation

Potential applications

Platform for quantum simulation of many body Hamiltonians [1] Precise tests of fundamental physics [2] Examine role of quantum effects in chemical reactions [3]

Direct laser cooling is proving to be a successful technique for producing cold molecules

Why Zeeman-Sisyphus?

For closed rotational transitions, diatomic molecules require a type II MOT $(N \ge N') \rightarrow$ weaker trapping forces and **lower MOT capture velocities,** ~24 m/s for CaF [4].

CaF are made in a cryogenic buffer gas source. Molecules exit the source at ~150 m/s, so must be slowed. Molecules can be slowed using chirped laser slowing [8].

Challenges

- Laser slowing requires $\sim 10^4$ photon scatters \rightarrow decay out of cooling cycle
- Range of transverse velocities from source \rightarrow loss from pluming
- Time dependent methods \rightarrow only address





Theory

Molecules pass through a spatially varying magnetic field. They are optically pumped into a weak field seeking (WFS) state in a region of high B field and a strong field seeking (SFS) state in a region of low B field \rightarrow Molecules continually climb up the potential hill.

$|\mathbf{e}\rangle$ Δ_{WS} Δ_{sw}

Requirements:

- A ground state split into wfs and sfs manifold.
- Excited state with negligible Zeeman splitting.

Energy lost per stage: $\Delta E_{stage} \sim 2\mu_B B_{max}$

Average deceleration force: $F = \frac{h(\Delta_{ws} - \Delta_{sw})}{r}$

For maximum deceleration, $\Delta_{sw} = 0$. However, this would prevent optical pumping. $\Delta_{sw} \neq 0$ also increases the transverse acceptance of the decelerator.

The ZS decelerator was first proposed in [5] and the concept has been subsequently demonstrated using superconducting magnetic coils [6,7]

Simulation results

Simulations done by Bethan Humphreys based on code by James Crilly, supervised by Dr David Carty and Dr Hannah Williams

portions of pulses

Zeeman-Sisyphus

- Requires fewer photons ullet
- Incorporates transverse guiding
- Time independent

Decelerator Design for CaF

To generate the spatially varying magnetic field, we use **80 permanent magnet stages**.



The magnetic field landscape is created using approximate Halbach cylinders made up of N52 NdFeB wedge magnets with residual magnetisation 1.44T

K = 6 stage

-2 -1 0 1 2 X (mm)

K = 2 stage

3D render of the Zeeman-Sisyphus decelerator

K **= 2 stages** generate a transversely uniform high magnetic field regions.

K **= 6 stages** are interleaved with *K* **= 2** stages to create zero field regions and provide transverse guiding.



Electronic ground state, $X^2\Sigma^+$ ($\nu = 0, N = 1$), has ~28 GHz/T splitting between SFS and WFS. First electronic excited state, $A^2 \Pi_{1/2}$ ($\nu = 0, J = 1/2$),

Simulated using a million molecules into decelerator, $\Delta_{sw} = -2.5 \text{ GHz}$, $\Delta_{ws} = 17 \text{ GHz}$



Reduction in mean forward velocity to **102 m/s**

0.28% of the million molecules entering the decelerator are decelerated **below 50 m/s**

For a pulse of 10¹¹ molecules from the source, expect $\sim 10^6$ out from the decelerator below 50m/s

Impact of Δ_{ws}

17 GHz chosen to maximise output at the slow tail

Detune above this: slower molecules never come into resonance and are not decelerated



Below this: now on resonance with slower molecules, which come to rest before exiting



Initial

200

250

150

z velocity (m/s)

has ~0.6 GHz/T splitting.

For CaF, with $B_{max} \sim 1 T$, $\Delta E_{stage} \sim h \times 28 GHz$

Measured B field in decelerator and calculated splitting of $X^2\Sigma^+$ state

Requires at least 60 deceleration stages to bring molecules down from 150 m/s to $v_{\rm capture} \sim$ 24 m/s.

Only scatters $\sim 10^2$ photons in this process, much fewer than the 10^4 required for traditional laser slowing \rightarrow promising for molecules with poor branching ratios

Experimental Progress

We have built a buffer gas source for CaF and have seen our first molecular signal.

Currently characterising and improving the source, then will assemble the decelerator.





Zeeman-Sisyphus deceleration for CaH

This simulation used a 1.8m long decelerator, with B_{max} = 1 T, molecular constants from [10, 11]

- Reduction in mean forward velocity from **145** to **51 m/s**
- 6% of the million molecules slowed below 50 m/s
- Reduction in mean forward velocity from **145** to **51 m/s**

References

[6] Augenbraun B.L. et al. *Phys. Rev. Lett.* **127** 263002 (2021)

Slowed

50

100

(x5)

[1] Blackmore J.A., et al. *Quant. Sci. And Tech.* **4** 1 (2018) [2] DeMille D., et al. Science 357 990 (2017) [3] Krems R.V. Phys. Chem. Chem. Phys. 10 (2008) [4] Xu S., et al. New J. Phys. 23 063062 (2021) [11] Frum C.I. et al. *Astrophys. J.* **408** (1993) [5] Fitch N.J. et al. *ChemPhysChem* **17** 22 (2016)

[7] Sawaoka H. et al. *Phys. Rev. A* **107** 022810 (2023) [8] Truppe S. et. al. New J. Phys. **19** 022001 (2017) [9] Devlin J. et al. J. Mol. Spect. **317** (2015) [10] Chen J. et al. *Phys. Rev. A* **73** 012502 (2006)

particles alised)

0.2



Frequency control

Need two frequencies for the optical pumping 19.5GHz apart at 606.1nm, each with ~250mW power, without any intermediate frequencies

Achievable with a 9.5GHz electro-optical modulator (Qubig).

However, the EOM's frequency can only be adjusted ±50MHz. Add in AOM(s) operating at 250MHz and/or

110MHz to give finer frequency control.





