STIRAP Preparation of a Coherent Superposition of ThO $H^3\Delta_1$ States

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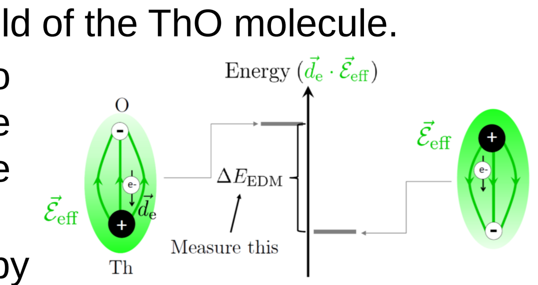
Abstract: The ACME Collaboration recently reported an order of magnitude improved limit on the electric dipole moment of the electron (eEDM) (ACME collaboration, Science 343 (2014), 269-272), setting more stringent constraints on many time reversal (T) violating extensions to the Standard Model. The experiment was performed using spin precession measurements in a molecular beam of thorium oxide. We report here on a new method of preparing the coherent spin superposition state that represents the initial state of the spin precession measurement using STImulated Raman Adiabatic Passage (STIRAP). We demonstrate a transfer efficiency of 75% giving a twelve-fold increase in signal. We discuss the particularities and challenges of implementing STIRAP in the ACME measurement and the methods we have used to overcome them.

For more information, **electronedm.info**

Measurement Scheme

- The electron electric dipole moment (eEDM) is aligned with the spin and interacts with the giant (~84 GV/cm) effective internal electric field of the ThO molecule.
- We measure the energy splitting between two electrons oriented oppositely with respect to the molecular electric field by performing a Ramsey-type phase measurement to observe molecule precession.
- The statistical sensitivity of the measurement is given by



$$\delta d_e = \frac{\hbar}{2\mathcal{C}\tau\mathcal{E}_{\text{eff}}\sqrt{\dot{n}T}}$$

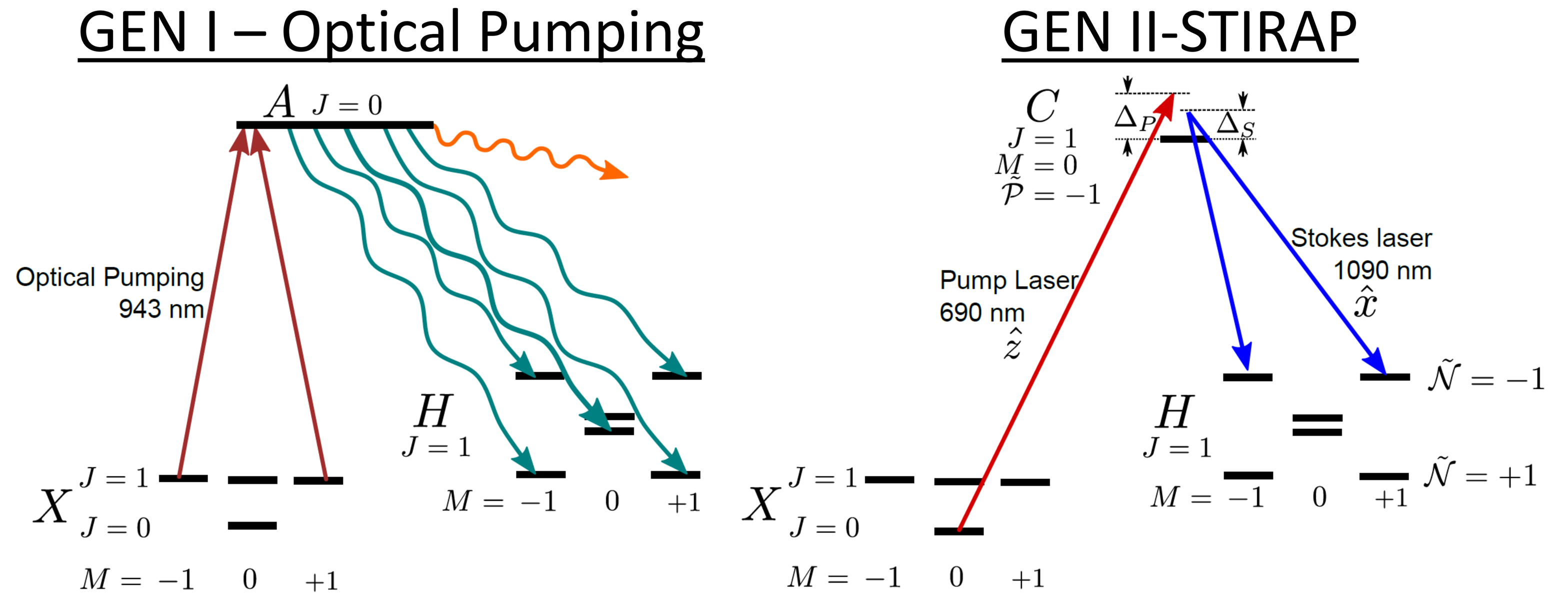
τ = coherence time
$\dot{n}T$ = number of molecules
$\mathcal{E}_{\text{eff}}$ = effective electric field

- STIRAP increases the usable molecule flux by improving the EDM state preparation efficiency.

Efficient State Preparation

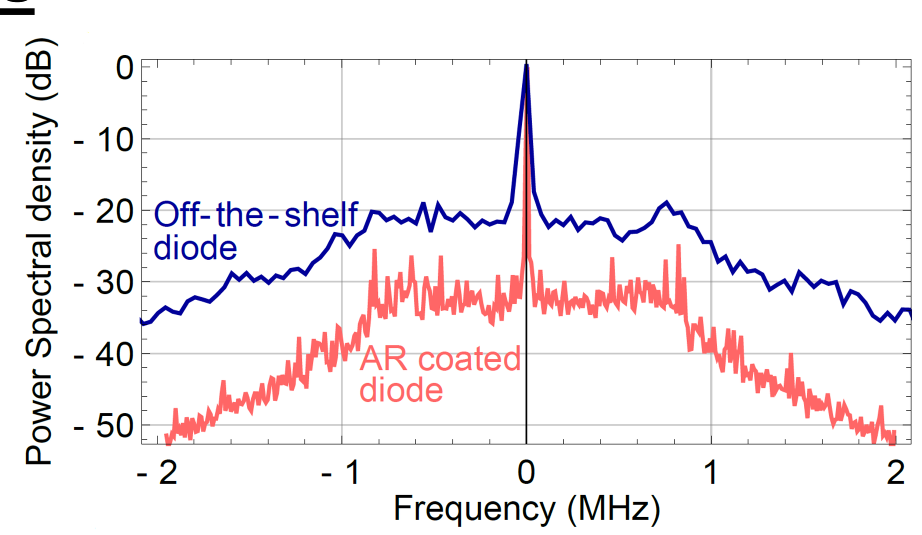
- In Gen I, state preparation using incoherent optical pumping was ~6% efficient.
- STIRAP transfers population into the desired superposition of state ~75% efficiency.

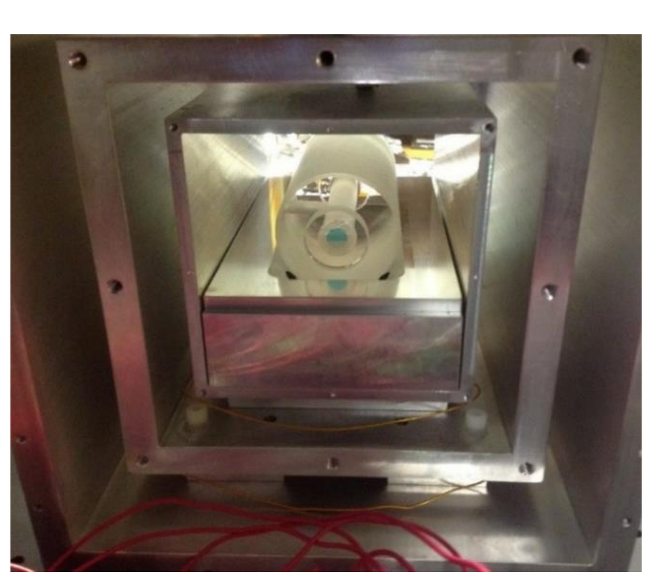
GEN I – Optical Pumping | GEN II-STIRAP

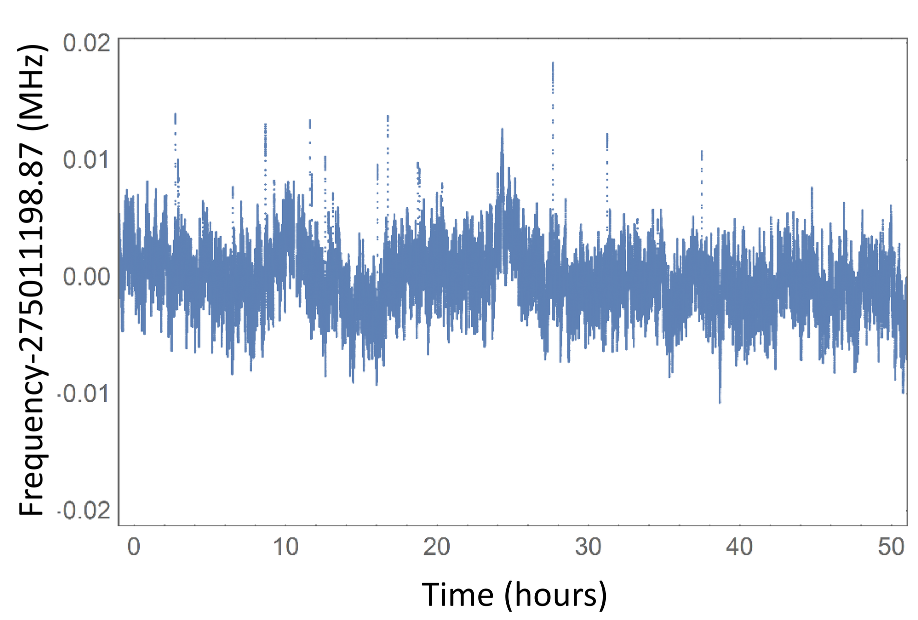


Narrow, Stable Lasers

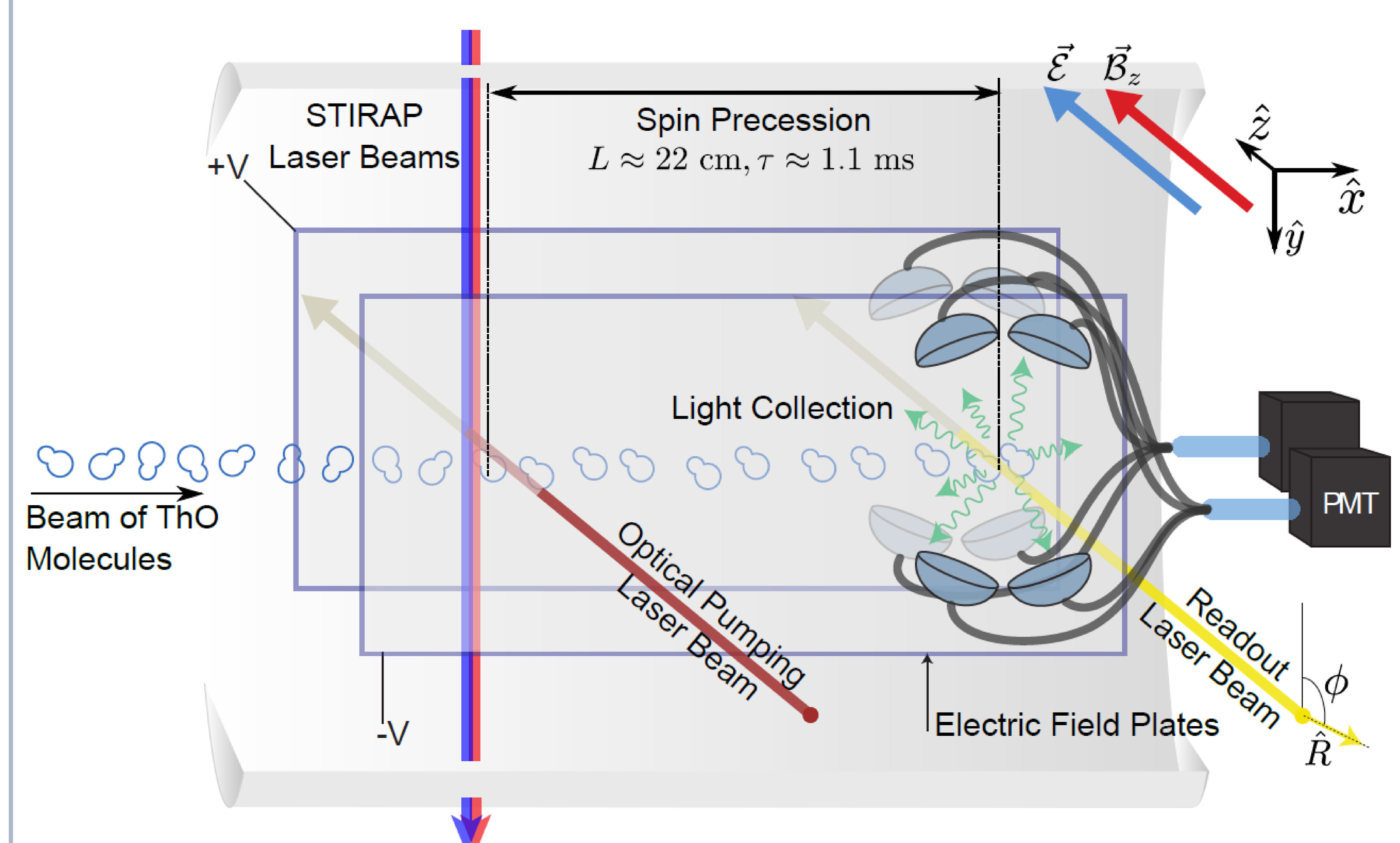
- Lasers locked to ULE cavities for linewidth reduction and frequency comb used for absolute frequency reference.
- Linewidths of ~100 Hz with noise pedestals suppressed by 30 dB.
- Long term stability better than ~10 kHz for days.



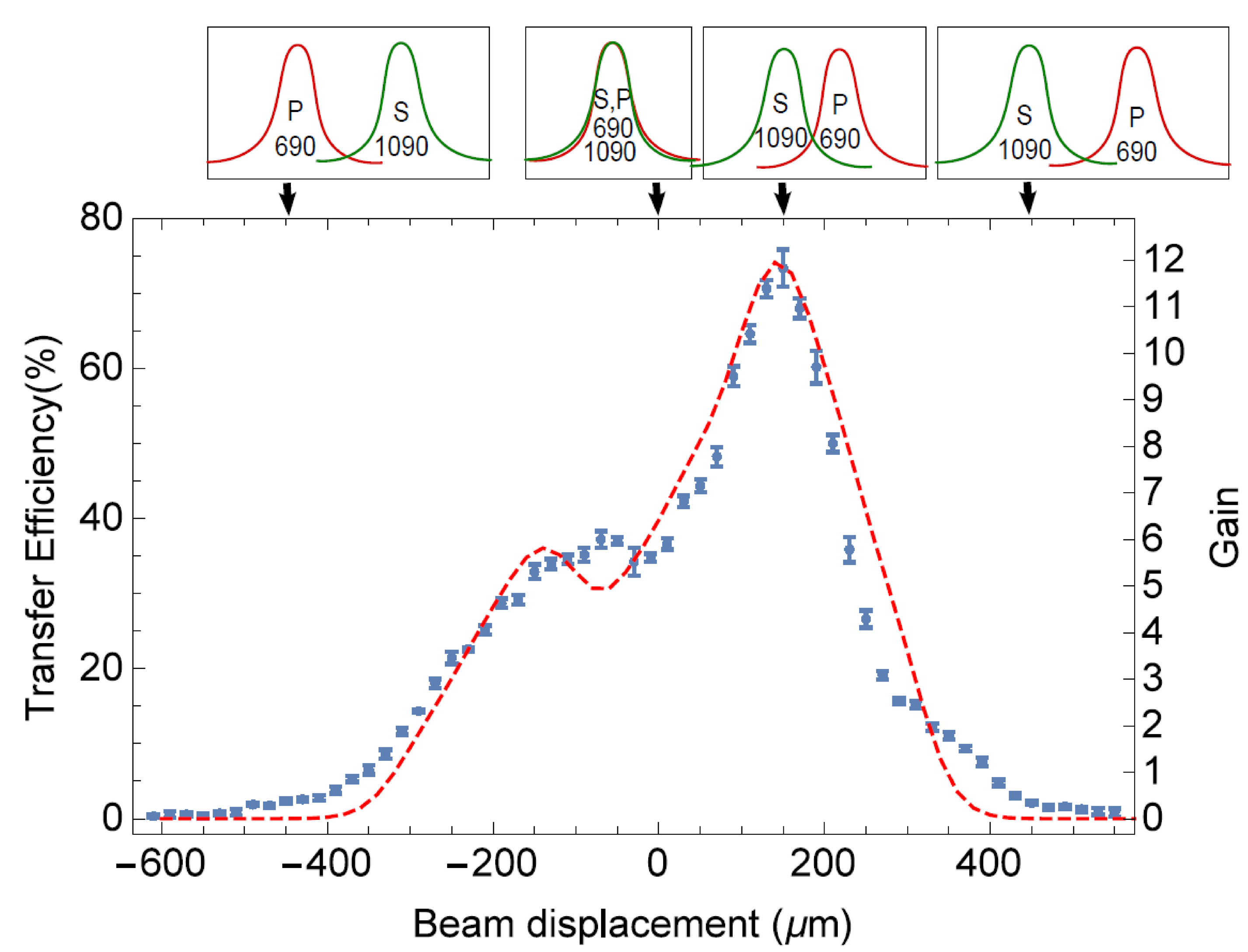




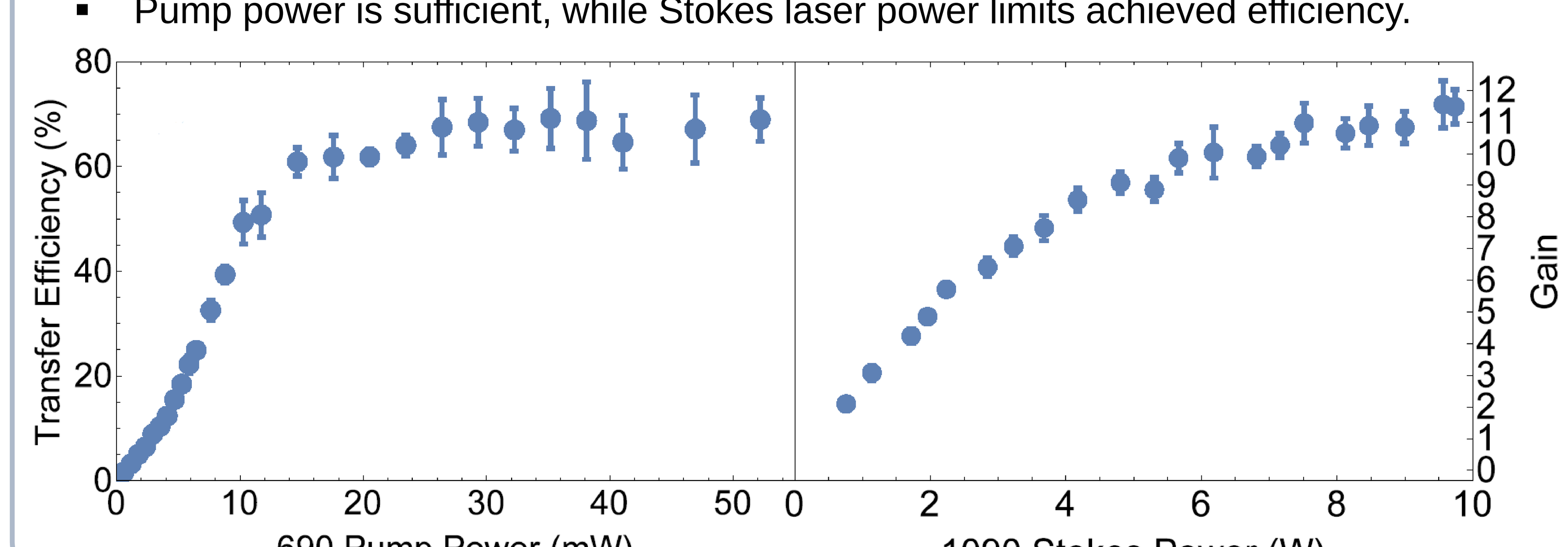
STIRAP Implementation



STIRAP Signature and Saturation Behavior

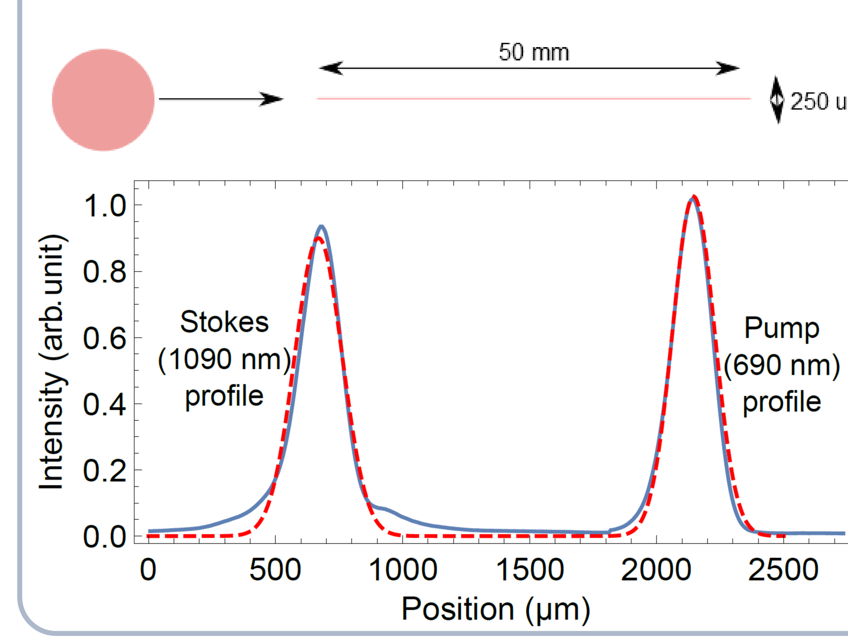


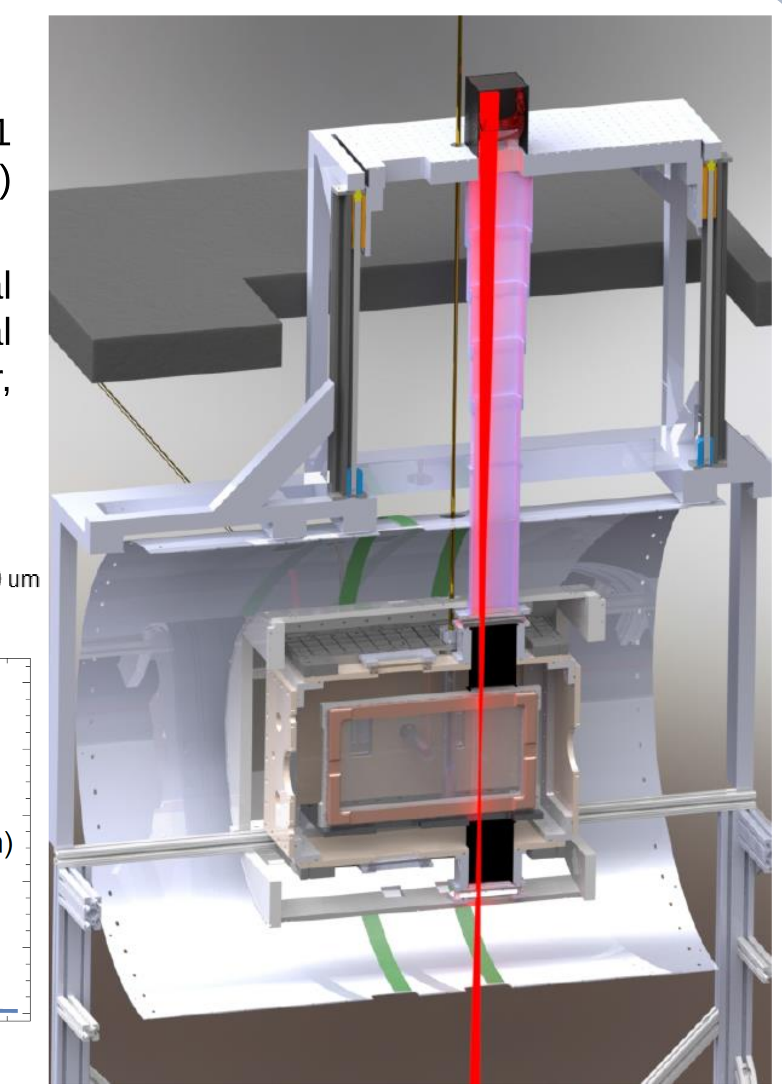
- Pump power is sufficient, while Stokes laser power limits achieved efficiency.



Laser Beamshaping

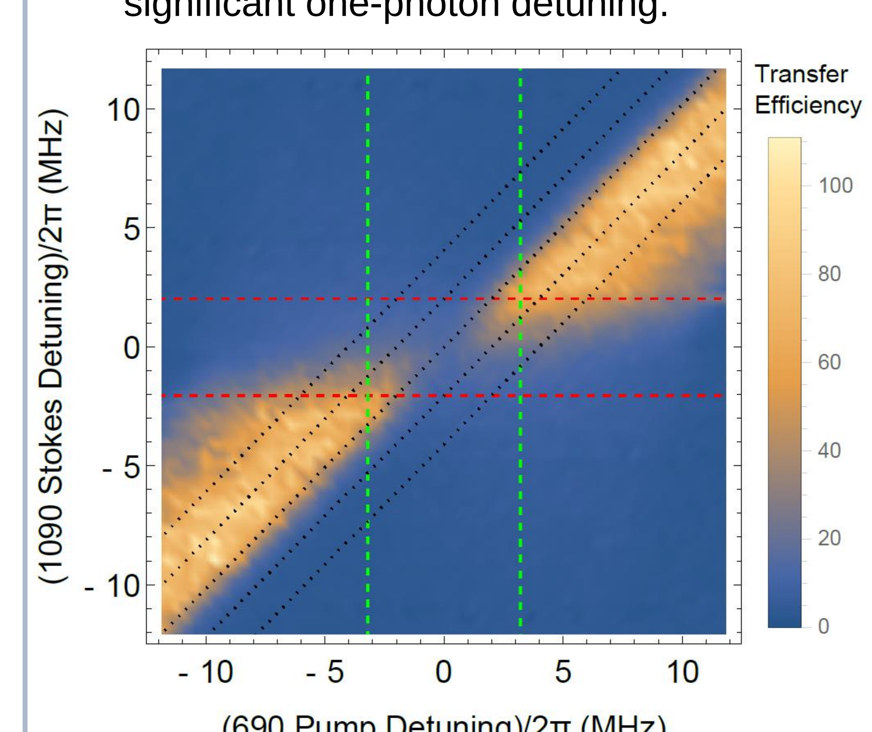
- Small H-C transition dipole moment (0.01 ea_0) and limited power (10 W at 1090 nm) constrain the Stokes intensity.
- Beamshaping difficult with limited optical access due to multiple experimental systems (field plates, vacuum chamber, magnetic shields, coils etc.)

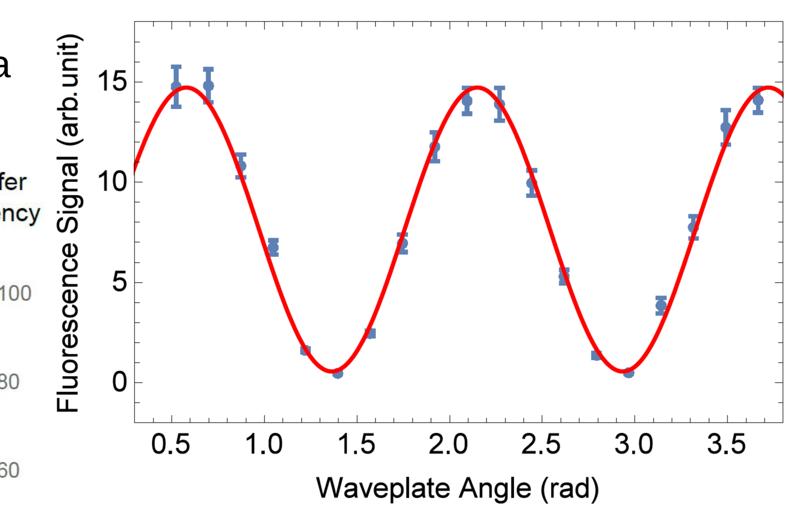




Coherent State Preparation

- We find it advantageous to run with a significant one-photon detuning.





- Spin coherence fringes show coherent state preparation.
- Contrast ~93%, comparable to GEN I levels.

Conclusion

We have demonstrated a factor of twelve increase in signal through STIRAP coherent state preparation. Out STIRAP implementation has been demonstrated to be robust and is already fully implemented in the main apparatus. We are confident that STIRAP and the other improvements will allow us to reduce the upper limit on the electron's electric dipole moment, or detect a non-zero value within the next order of magnitude.

References

ACME Papers:

1. **STIRAP and Other Results:** "Stimulated Raman adiabatic passage preparation of a coherent superposition of ThO H states for an improved electron electric-dipole-moment measurement." C. D. Panda et al. *Phys. Rev. A* **93**, 052110 (2016)
2. **GEN I Experimental Result:** "Order of Magnitude Smaller Limit on the Electric Dipole Moment of the Electron." ACME Collaboration. *Science* **343**, p. 269-272 (2014)

Effective E-field Calculations:

1. Skripnikov et al. *J. Chem. Phys.* **142** 024301 (2015)
2. T. Fleig et al. *J. Mol. Spectrosc.* **300** 16 (2014)