

ULTRACOLD HYDROGEN

Principal Investigators

Professor Daniel Kleppner and Professor Thomas J. Greytak

Research Staff and Visiting Scientists: Dr Robert deCarvalho,
Dr. Lorenz Willmann (University of Groningen)

Graduate Students:

Nathan Brahms (Harvard University), Cort Johnson, Lia Matos, Bonna Newman, Julia Steinberger, Kendra Vant

Undergraduate Students:

Anat Berger, Annemarie Grandke, Alexander Ellis (visitor), Andrew Koller, Charles Sievers

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Research in the Ultracold Hydrogen Group centers on studies of the structure of atomic hydrogen, ultracold collisions, the properties of hydrogen as a quantum gas and ultrahigh precision spectroscopy. Principal activities during the past year include construction and demonstration of a mode-locked laser for optical frequency metrology of hydrogen, and construction of an apparatus designed to trap and cool hydrogen that employs a thermalizing agent—a second species of trapped atom—and uses buffer gas cooling techniques.

1) Octave-spanning frequency comb generator (in collaboration with O. Kuzucu, T. R. Schibli, J. Kim, E. P. Ippen, and F. X. Kaertner)

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We have developed a new type of femtosecond laser intended for frequency comb generation for our program of ultraprecise spectroscopy of trapped hydrogen. Such lasers have revolutionized frequency metrology in the last 4 years [1,2,3]. Typical setups employ commercially available Ti:sapphire lasers [4] that produce pulses in the 10-30 fs range and photonic crystal fibers that broaden the spectrum to cover an octave. This enables a self-referenced comb stabilization scheme in which the long wavelength part of the spectrum is doubled and brought to interfere with the short wavelength to allow direct carrier-envelope offset frequency detection and stabilization. However, an octave-spanning laser would provide important advantages in coherence and reliability.

We have developed Ti:sapphire lasers with 80 MHz and 150 MHz repetition rates that span an octave and could be scaled to GHz repetition rate. Broadband dispersion compensation is provided by novel double chirped mirror pairs [5,6] in combination with thin BaF₂-wedges. These constitutes a major improvement compared to previously demonstrated octave-spanning Ti:sapphire lasers which used double-chirped mirrors and prism pairs to produce intra-cavity continuum generation [7,8,9], as well as other high-repetition rate sources [10]. In particular, since the resonator dispersion in a prism-less cavity is much less sensitive to the alignment of the cavity, stable long-term mode-locking is obtained.

The laser is an astigmatically compensated x-fold cavity, with 80 MHz or 150 MHz repetition rate. The laser has a 2mm long Ti:sapphire crystal with an absorption of $\alpha=2.7 \text{ cm}^{-1}$ at 532 nm, pumped by a diode pumped Nd:vanadate laser (Millenia XS). The folding mirrors have 10 cm ROC, and the pump lens has a 60 mm focal length (figure 1a). All mirrors in the cavity, with exception of the end mirrors, are type I (gray) and type II (black) double chirped mirrors that generate a smooth group delay dispersion when used in pairs. Figure 1b shows reflectivity and group delay of the DCM pairs. The average group delay is smooth from 600 nm to 1200 nm. The reflectivity is on average 99.9% over the full octave. One of the cavity end mirrors is a silver mirror and the other is a broadband output coupler (OC), either ZnSe/MgF₂ (80 MHz repetition rate laser) or a broadband TiO₂/SiO₂ (150 MHz repetition rate laser) with 1% transmission. The output power in cw operation is typically 40 mW with 4.4 W pump power. In mode-locked operation, the average power is 90 mW. Limitations in output power are due to low output coupling and losses in the OC substrate as well as the silver mirror. Increased output coupling and elimination of the silver mirror is highly desirable to increase the laser efficiency.

Figure 2 shows the spectra in broadband operation for the 80 MHz and 150 MHz lasers. The octave is reached at about 25 dB below the average power level. The same plot shows the output coupler (OC) transmission curve. The width of this transmission curve is also a determining factor in the width of the output spectrum, significantly enhancing the spectral wings.

To employ these broadband lasers as frequency comb generators, it is necessary to demonstrate control over the carrier-envelope offset frequency, f_{ceo} , and the repetition rate, f_{rep} . We directly measure f_{ceo} by the 1f-2f technique described in [2]. Using a dichroic beamsplitter, the laser output is split into long and short wavelength components and recombined after insertion of a proper time delay stage (figure 3a). The beam is then focused into a 1mm BBO crystal cut for type I SHG of 1160 nm. The resulting output is sent through a 10 nm wide spectral filter centered at 580 nm. The delay stage is essential to compensate for different group delay of the 580 nm and 1160 nm radiation in the optical components (splitters, OC and BBO crystal). After projecting the doubled 1160 nm light and the fundamental into the same polarization to enable interference, the beat signal is detected by a photo multiplier tube (PMT). Figure 3b shows the detected f_{ceo} beat signal, with 10 kHz and 100 kHz measurement bandwidths. The signal-to-noise is 30 dB and 40 dB respectively. This signal is limited by the amount of power in the fundamental at 580 nm (it can be seen to be below the noise floor of the optical spectrum analyzer in the spectrum shown in fig 2a). An improved OC design, providing more bandwidth in the short wavelength part of the spectrum, as well as higher output coupling, is likely to improve signal-to-noise.

To demonstrate control over the comb we filter the component $f_{rep} + f_{ceo}$ at 130 MHz from the PMT signal and phase-lock it to a stable RF synthesizer using a phase-locked loop. To increase the locking range the 130 MHz component of the signal is properly filtered, amplified and sent to a 32-fold frequency divider and is then compared with the synthesizer signal in the digital phase detector. The properly filtered and amplified phase error signal controls the pump power via an acousto-optic modulator, which directly controls the carrier-envelope frequency of the mode-locked laser [11]. In lasers with intra-cavity prisms, the transducer which controls f_{ceo} is a piezo driven tilting end mirror [12]. In a prismless cavity we can use modulation of the pump power to control f_{ceo} over the required bandwidth.

Figure 4 shows the measured in-loop phase error spectral density at the output of the digital phase detector, from 50 mHz to 1 MHz. The integrated phase error is 1.4 rad relative to the 130 MHz signal. The overwhelming contribution to this phase noise error comes from the pump noise shown in relative units in figure 4. The spectral components from 1-10 KHz contribute 90 % of the phase error. Still, this comb can be used for metrology since no cycle slips occur. In the near future either an improved loop filter design may suppress this pump noise more strongly or a lower noise pump laser can be used to achieve lower residual carrier-envelope phase fluctuations.

Further details are presented in reference [13].

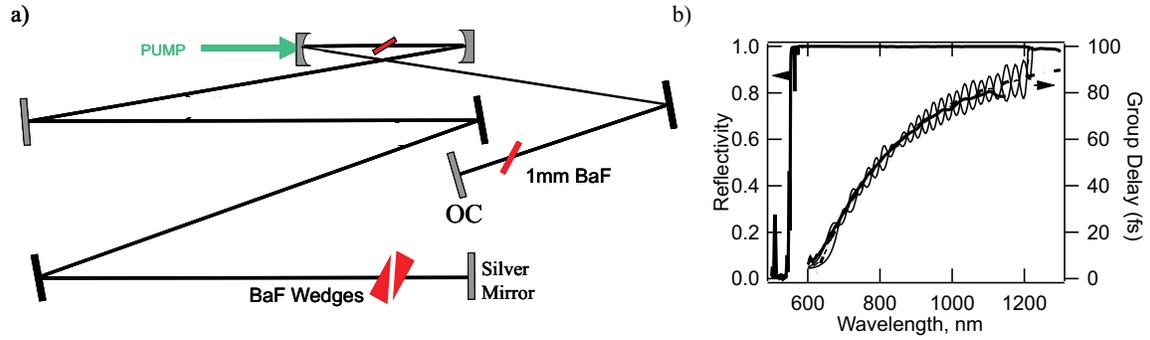


Figure 1: a) Schematic of octave spanning prismless laser. Green and blue mirrors are type I and II DCMs. The BaF₂ wedges are used for fine tuning of the dispersion; b) Reflectivity of the type I DCM with pump window shown as thick solid line with scale to the left. The group delay design is shown as thick dash-dotted. The individual group delays of type I and II DCMs are shown as thin lines and its average as a dashed line,

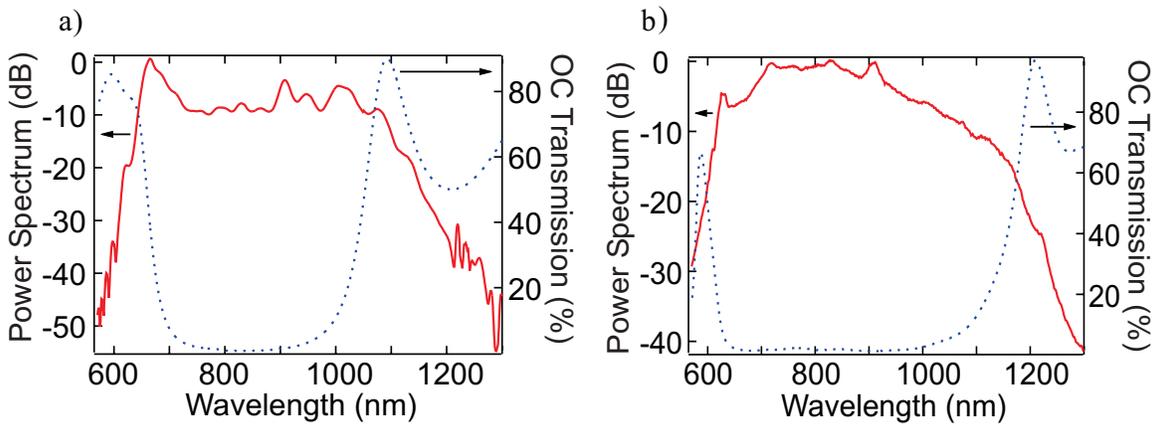


Figure 2: a) Measured output spectrum for 80 MHz repetition rate and (b) 150 MHz repetition rate lasers. For both, the octave is reached at approximately 20 dB below the average power level. The average mode-locked power is 90 mW for both lasers. Also shown are the respective OC transmission curves, as ZnSe/MgF₂ stack for the laser (left) and a SiO₂/TiO₂ for the laser (right).

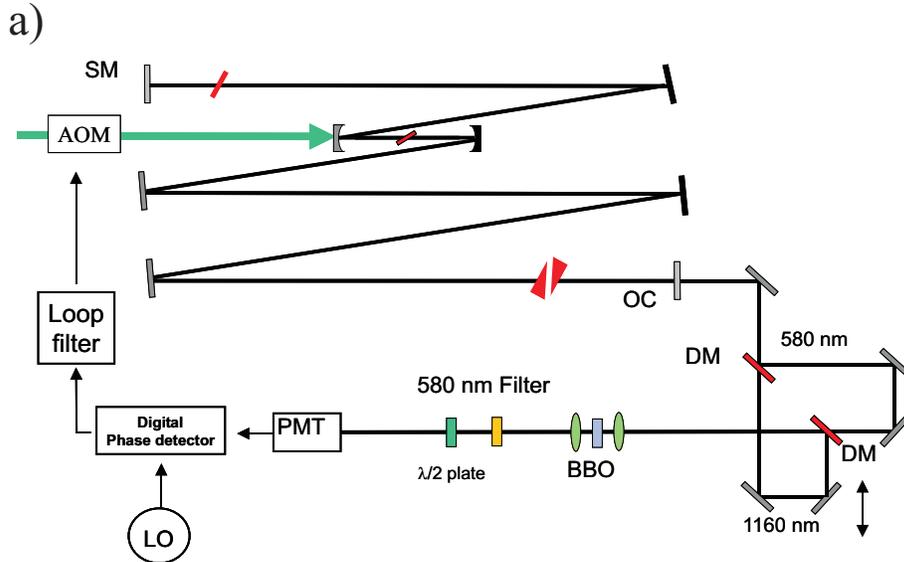


Figure 3a. Setup for f_{ceo} detection and lock.

b)

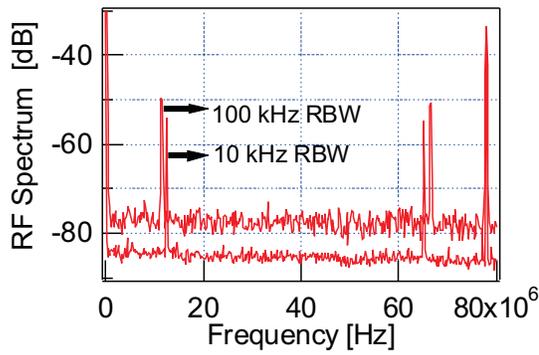


Figure 3b. Measured carrier-envelope beat signal from the 80 MHz laser.

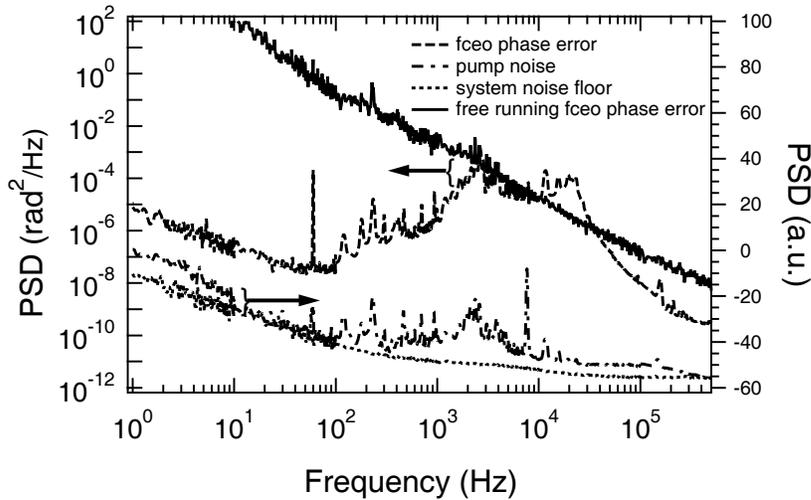


Figure 4. Spectral density of phase error signal at the output of digital phase detector for f_{ceo} lock (scale is on the left) and spectral density of pump noise (scale on right). It can be seen that overwhelming contribution to f_{ceo} noise comes from pump noise contributions. References

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2) Hydrogen-Lithium Buffer-Gas Loading

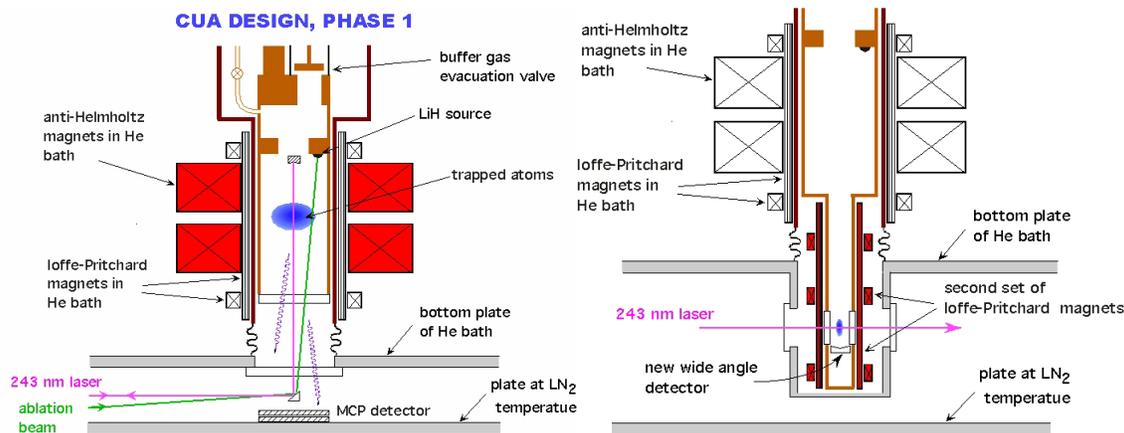
The search for Bose-Einstein condensation originated with studies of atomic hydrogen. Although BEC was eventually achieved with hydrogen [1], studies of hydrogen in the quantum regime, as well as other atomic studies of ultracold hydrogen, have been frustratingly difficult because of experimental limitations. The first step of hydrogen cooling is by thermalization on a superfluid helium film, in contrast to laser-cooling of alkali metal atoms. Subsequent cooling is by evaporation, a technique first developed for use in hydrogen [2]. Hydrogen has an anomalously small elastic scattering cross section in the S-wave regime, which severely limits the rate of evaporative cooling and the number of atoms that can be cooled. The low collision rate almost prevents achieving BEC.

In this CUA core project, we are pursuing an entirely new approach to cooling and trapping hydrogen. It is based on the buffer-gas loading technique of Doyle [3]. An essential feature is co-trapping Li with H. The H-Li elastic cross section is about 3000 times larger than the H-H cross section [4]. Collisions with lithium will vastly increase the rate at which hydrogen thermalizes during evaporation. This approach represents a radical departure from all previous cold-hydrogen experiments and requires the design and creation of a major apparatus. We describe here the design features and the state of construction.

Hydrogen trap Atomic hydrogen is initially confined in a spherical quadrupole trap. The trap has a diameter of 7 cm and a peak magnetic field 4.2 T. The magnet design is based on previous work by Doyle. This magnet has been constructed and tested.

Hydrogen and lithium source To provide atomic hydrogen for low temperature research, all previous studies have used some form of radiofrequency discharge to dissociate molecular hydrogen. However, such a discharge is difficult to implement in a buffer gas loading apparatus

and cannot be used to provide lithium atoms. Our approach is to dissociate solid LiH by laser ablation. Ablative sources have been used in various buffer-gas cooling experiments but never to provide atomic hydrogen. We have carried out a room temperature experiment on ablation of LiH, detecting the lithium spectroscopically. We observe large amounts of Li, which suggests that the ablative source will meet our requirements for simultaneously producing both H and Li.



(left). Diagram of the hydrogen buffer gas loading apparatus in its initial configuration, intended for the study of the loading and cooling process. (right) Diagram of the second phase of the apparatus in which the hydrogen is transferred to a relatively open Ioffe-Pritchard trap, and then transported to a separate experimental region where it can be studied with great flexibility.

Buffer-gas loading Hot H and Li from the ablative source are thermalized in the quadrupole magnet by collisions with a buffer gas of ^3He . The buffer gas is confined in a cell at a temperature of about 300 mK. Thermalization with the buffer gas causes the atoms to be trapped in the quadrupole, which forms a magnetic trap with a depth of about 2.8 K. A valve is then opened allowing the ^3He to escape, leaving behind the confined atoms which are subsequently cooled by evaporation.

The detailed analysis of buffer-gas loading requires numerical modeling of the diffusion and flow processes that govern the dynamics of the system. A crucial parameter in this modeling is the diffusion constant for H in ^3He at the temperatures of interest. This has been computed for us by Dalgarno [5]. Because of its relatively small magnetic moment, 1 Bohr magneton, and small mass, buffer-gas loading of hydrogen presents some special challenges. Nevertheless, the results of our modeling calculations are encouraging.

Superfluid-free cell A very important experimental feature of our approach is that there is no ^4He in the cell. The absence of a superfluid film permits great flexibility in incorporating windows, detectors, and other devices, such as the second trap shown in the above figure, right.

A two-trap cell Although the spherical quadrupole trap is ideal for the initial trapping and cooling of the gas, it is not well suited to precision studies. It has a zero in the magnetic field which permits slow escape of the atoms, and its geometry limits experimental access. To avoid these problems we plan to transfer the atoms to a second magnetic trap of the Ioffe-Pritchard type as shown in the figure above, right. Such a trap has no magnetic field zero. Since the atoms are already quite cold when introduced, the fields need not be extraordinarily high allowing a relatively open geometry for the coils.

More efficient H detection All our past studies of ultracold hydrogen have detected the gas by exciting the 1S-2S two-photon transition, then applying a small electric field, and detecting the Lyman-alpha radiation. Due to geometric limitations to the viewing angle and inefficiencies in

photon detection, our efficiency has been less than 10^{-5} . The two-trap design opens opportunities for major improvement. Some improvement can be achieved by better optical access. However, exciting the 2S atoms to a Rydberg state, which is then field-ionized, or directly photo ionizing the 2S atoms, would improve the detection efficiency by many orders of magnitude. Such an improvement would have a major impact on nearly every aspect of ultracold hydrogen research.

Goals The new source opens many research opportunities. By co-trapping H and Li we would have an opportunity to create LiH in the triplet state by two-step Raman excitation [6]. This molecule is of particular interest because it has a remarkable property: LiH possesses but a single vibrational state. We should be in an excellent position to create this unusual molecule and also to study LiD.

A longer term goal is high-precision spectroscopy of transitions such as 2S-10S. This would immediately improve our knowledge of some combination of the Rydberg constant, the Lamb shift and the proton radius. The 1S-2S transition, is also of great interest. This transition is currently the touchstone for high precision hydrogen spectroscopy [7] and is a potential candidate for an optical atomic clock. Resolution close to the natural linewidth, 1.3 Hz, should be possible using ultracold hydrogen. Pursuing this research requires high performance lasers and state of the art equipment for optical frequency metrology, as well as the new hydrogen apparatus. The laser systems are being developed under a separate program sponsored by NSF, ONR and a MURI program.

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3) Design of new measurement of the hydrogen 2S hyperfine structure

The theory of quantum electrodynamics (QED) has had great success in calculating features of the hydrogen spectrum. Nevertheless, current high precision experiments on hydrogen, deuterium, muonium and positronium, are all pushing calculations of QED theory to its limits.

The comparison of the 1S and 2S hyperfine splitting in hydrogen-like atoms is an attractive test of theorists' ability to calculate QED corrections to atomic spectra [1,2]. A quantity of particular interest is $D_{21} = 8 f_{\text{HFS}}(2\text{S}) - f_{\text{HFS}}(1\text{S})$, the weighted difference between the 1S and 2S hyperfine splittings f_{HFS} . This quantity primarily consists of radiative corrections and relativistic terms.

D_{21} can be calculated to high precision by purely QED methods [2]. Most calculations of hydrogen level structure include nuclear terms which ultimately require difficult quantum chromodynamics (QCD) calculations, but in the case of D_{21} the nuclear terms cancel.

The hydrogen hyperfine splitting in the 1S state is known to high accuracy [3]. The 2S hyperfine interval has been measured by using rf [4,5] and recently optical [6] techniques. All these experiments measured the hyperfine splitting at zero or very small magnetic field.

The MIT Ultracold Hydrogen Group has attempted to use our ultracold metastable spin-polarized 2S hydrogen sample to measure the 2S hyperfine splitting $f_{\text{HFS}}(2\text{S})$ by rf spectroscopy. The proposed experiment was to deduce D_{21} indirectly by rf spectroscopy of the 2S hyperfine spectrum at relatively high (810 Gauss) magnetic fields.

The 178 MHz 2S hyperfine splitting $f_{\text{HFS}}(2\text{S})$ is usually measured at low magnetic fields, as the transition frequency between the members of the upper $F = 1$ manifold and the lower $F = 0$ state. This type of experiment is not possible in our apparatus because our rf coils are not designed to deliver significant power above 100 MHz. Instead, our goal is to measure $f_{\text{HFS}}(2\text{S})$ indirectly, using the properties of the 2S hyperfine spectrum.

The hydrogen S-state manifolds contain special points, called field-independent points, where the transition frequency between two states is independent of the magnetic field to first order. The field-independent transition we are interested in is between the two uppermost states, at a magnetic field of 810 Gauss.

The hyperfine splitting frequency $f_{\text{HFS}}(2\text{S})$ can be calculated from the field-independent point transition frequency and a conversion factor. This conversion factor is a function of well understood quantities: the electron and proton Landé g factors and the proton-to-electron mass ratio. Thus by measuring the field-independent point transition frequency, we can indirectly measure $f_{\text{HFS}}(2\text{S})$. The uncertainty introduced by the conversion factor is far less than current experimental uncertainties.

The experimental strategy is to trap and cool 1S hydrogen atoms in a low magnetic field trap, then raise the magnetic field to 810 Gauss, changing the magnetic potential adiabatically. A population of 2S atoms in the highest hyperfine state is created by two-photon laser excitation. The transition within the 2S hyperfine manifold is excited using an rf coil wrapped around the cell. Then one or the other 2S hyperfine state is removed from the trap by excitation to the 3P state (using our new 656 nm diode laser system). The remaining 2S atoms are detected. As we sweep the rf frequency through the resonance, we expect to see either signal enhancement or depletion, depending on which 2S state we choose to keep in the trap.

Unfortunately, several factors contributed to making the 2S hyperfine splitting measurement impossible in our current apparatus. By developing new tools to measure the trap magnetic field, we found that our trapping field model is incorrect. Although we are ultimately able to create a high magnetic field trap, we do not know important characteristics such as trap frequency and vertical position. The trap frequency is important because a large trap frequency indicates a large confinement, and causes sample heating. The vertical position is important because unless the trap is at the same position as the laser beam focus, our signal amplitude is suboptimal.

To measure the 2S hyperfine splitting, we require both a cold sample and large signal amplitude at high magnetic field. We are not able to achieve these, most likely because our magnet system does not allow us to have both a low trap frequency and the correct vertical trap position. However, in the process of attempting to measure the 2S hyperfine splitting, we were able to develop tools that will be useful in future high precision experiments in our apparatus. These tools include using the 1S hyperfine spectrum as well as the 2S-3P transition properties to precisely measure our trap magnetic field. We also understand much better systematics of the 2S double-exponential lineshape (used to estimate the temperature of the sample).

Although ultimate goal of doing spectroscopy of the 2S manifold at the field-independent point is not possible in our current experiment, a better trapping magnet design could make it possible in the future.

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Publications

L. Matos and D. Kleppner, O. Kuzucu, T. R. Schibli, J. Kim, E. P. Ippen, and F. X. Kaertner, "Direct frequency comb generation from an octave-spanning, prismless Ti:sapphire laser", *Optics Letters*, **29**, 1623 (2004).

Theses

Charles Sievers, Frequency Stabilization for a 486 nm Dye-ring, MIT B.S. Thesis, 2004
Laser

Annemarie Grandke, Investigating Noise Reduction in an Octave-Spanning Ti:Sapphire Laser, MIT B.S. Thesis, 2004.