Chapter 2. Basic Atomic Physics

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2.1 The Diamagnetic Rydberg Atom

Sponsors

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Project Staff

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A highly excited hydrogen atom in a strong magnetic field, the so-called "diamagnetic hydrogen atom," is among the simplest nonseparable systems in quantum mechanics. Understanding it can provide a key to the more general aspects of nonseparable systems. The problem is also attracting attention in the context of nonlinear dynamics because its classical behavior displays a transition from orderly to disorderly motion as the energy is increased in a fixed magnetic field. One can study the quantum structure of the system in this regime both theoretically and experimentally. Thus, the diamagnetic hydrogen atom provides an ideal testing ground for studying the relation between quantum structure and disorderly classical motion, a subject sometimes called "quantum chaos."

We have developed techniques for carrying out high resolution laser spectroscopy on the lithium atom in a strong magnetic field. (As we shall demonstrate, the differences between lithium and hydrogen are essentially negligible.)

The experiment uses a lithium atomic beam which is excited by two c.w. lasers. The first laser excites the atoms from the 2S state to the 3S state by a two-photon transition and the second laser excites the atoms to Rydberg states. The excited atoms are detected by electric field ionization. We typically operate in magnetic fields near 6T. We can determine the energy within 10^{-3} cm⁻¹, and the magnetic field within 5 gauss.

The Hamiltonian for the diamagnetic hydrogen atom, in atomic units, is

$$H = \frac{p^2}{2} - \frac{1}{r} + \frac{1}{2}L_zB + \frac{1}{8}B^2\rho^2, \qquad (1)$$

There are no general solutions to this problem, and perturbation theory is not applicable in the positive energy regime. Our experimental results² have

¹ Department of Physics, Wellesley College, Wellesley, Massachusetts.

² C. lu, G.R. Welch, M.M. Kash, L. Hsu, and D. Kleppner, *Phys. Rev. Lett.* 63: 1133 (1989).



Figure 1. Comparison between the experimental spectrum of lithium at B = 6.113 T ($L_z = 0$, odd parity) with the calculated spectrum of hydrogen, from reference 1.

helped to stimulate theoretical advances, and these two efforts have now been joined to provide a comparison of experiment and theory at an unprecedented level of detail. Figure 1 displays a high resolution spectrum of lithium in a field of 6 tesla in an energy range near the ionization limit, a region that until recently was essentially unexplored. The results of a recent theoretical calculation by Delande and Gay³ are displayed for They employ the complex comparison. coordinate-rotation method and a matrix diagonalization using a Sturmian-type basis. Figure 2 shows a detailed comparison between our experiment and their calculation near 7 cm⁻¹ at 6.131 T. This is well above the ionization limit in this field, 2.81 cm⁻¹. The agreement is, for all present purposes, perfect.



Figure 2. Detailed comparison of experiment and theory, from reference 1.

³ D. Delande, A. Bommier, and J.C. Gay, *Phys. Rev. Lett.* 66: 141 (1991); C. Iu, G.R. Welch, M.M. Kash, D. Kleppner, D. Delande, and J.C. Gay, *Phys. Rev. Lett.* 66: 145 (1991).

This result represents a twofold achievement. By confirming the validity of the calculation, our experiment has opened the way to the use of calculational methods for studying the atom-field system in broad regimes. Thus, we have a powerful new tool for studying the system. Furthermore, the agreement between theory and experiment confirms the reliability of the experiment. It assures, for instance, that lithium is a suitable test atom for the diamagnetic hydrogen problem and that potentially worrisome experimental effects, for instance the effects of small stray electric fields, are not, in fact, important.

In addition to our experimental work, we have been investigating the theoretical energy level structure in a bound state regime where numerical calculations are relatively straight forward. The goal of this effort is to shed light on the origin of periodic structures in the spectrum that we have discovered in the positive energy regime.² We have carried out calculations for energy levels below -35 cm^{-1} from 0 to 6 tesla using an IBM RT computer. The calculations were carried out by diagonalizing 1,241 λ type bases.⁴ The results of the calculation were verified by experiment at a field of 6T. A typical result is shown in figure 3.

The important physical features revealed in figure 3 are these. In the low field region the principal quantum number n is "good," though the angular quantum number I is destroyed by the magnetic field. The levels follow quadratic trajectories in the field that are governed by an approximate sym-



Figure 3. Diamagnetic structure of lithium from 0 to 6 T.

⁴ C. Iu, Ph.D. diss. Dept. of Physics, MIT, 1990.



Figure 4. One-dimensional Stark splitting near 6 T, n = 70. The electric field is approximately 80 mV/cm.

metry, the λ symmetry.⁵ As the field is increased, levels from different n-manifolds begin to interact and the levels start to repel each other. One expects that as the field is increased the level interactions would become so strong that the λ would be completely destroyed. symmetry However, in figure 3, we can see the levels reconstitute themselves after passing through a region of strong level repulsion, as manifested by a structure of successive narrowly avoided crossings. At large field, these levels form approximately parallel structures with spacings equal to a sequence of (Dashed lines are drawn along Rydberg levels. these lines to help their identification.) These levels appear to correspond to the Rydberg series of the lowest Landau level near the ionization limit seen in reference 1, except that they lie at much lower energy.

In addition to the study of level structure, we have studied the Stark splitting of the one-dimensional hydrogen atom that is created by magnetic confinement of the electron transverse to the magnetic field. Figure 4 shows an example of the Stark splitting. The levels correspond to the Rydberg level of n = 70. The electric field was about 80 mV/cm.

The predicted Stark splitting is 0.050 cm^{-1} , and the measured splitting from figure 4 is 0.047 cm^{-1} . This observation of the one-dimensional Stark splitting further confirms the regular behavior of the quantum system.

Publications

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Thesis

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⁵ D. Delande and J.C. Gay, Comment At. Mol. Phys. 19: 35 (1986).

2.2 Millimeter-Wave Frequency Measurement of the Rydberg Constant

Sponsors

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Project Staff

Pin P. Chang, Scott Paine, Robert Lutwak, Professor Daniel Kleppner

The Rydberg constant R_{∞} determines the wavelengths of the spectrum of hydrogen. More fundamentally, it relates the atomic and practical length scales. It is among the most accurately known fundamental constants: Recent experiments have determined R_{∞} to nearly one part in 10^{10} using optical spectroscopy.⁶ These measurements are approaching the practical limits of optical wavelength metrology. Future progress will have to come from frequency measurements, making use of the modern definition of length in terms of time intervals and a defined speed of light c.

We are attempting to advance the precision of R_{∞} by measuring it in frequency units. The specific quantity we are measuring is cR_{∞} , which might be called the "Rydberg frequency." Our approach is based on millimeter wave spectroscopy on transitions between adjacent Rydberg states of hydrogen, around n = 30. Because the frequency of millimeter wave radiation can be measured to the full precision of modern atomic clocks, the experiment is not limited by metrological standards.

The goals of our experiment are three-fold: First is the reevaluation of R_{∞} itself. Second is the measurement of the Lamb shift. Our measurements involve high angular momentum states for which the Lamb shift is extremely small. A comparison of our results with optical measurements can yield an improved value of the Lamb shift. Third is the precise frequency calibration of the spectrum of hydrogen in order to provide independent confirmation of the accuracy of optical frequency metrology as this technique starts to advance.

Our experiment works as follows. Hydrogen or deuterium in an atomic beam is excited by twophoton absorption to the state n = 29, m = 0. A crossed electric and magnetic field scheme7 then transfers the atoms to the longer-lived n = 29, |m| = 28 "circular" state. The atoms then pass into an interaction region where a resonance transition takes place on the $n = 29 \rightarrow n = 30$ transition. The atoms interact with the millimeter wave radiation at two locations in a Ramsey separated oscillatory fields geometry. Finally, the atoms are state-analyzed in a selective electric field ionization detector capable of differentiating between the n = 29 and n = 30 circular states. The resonance signal is obtained by counting the atoms in each state as the millimeter wave frequency is tuned across the $n = 29 \rightarrow n = 30$ transition.

Figure 5 illustrates the main features of the atomic beam apparatus. Atomic hydrogen is produced by dissociating H₂ in a radio frequency discharge. To minimize the frequency width of the resonance transition, the interaction time is prolonged by cooling the atomic beam. This is accomplished by flowing the hydrogen through an aluminum thermalizing channel whose temperature can be as low as 10K. The atoms are excited optically in a crossed electric and magnetic field region. The magnetic field is produced by permanent magnets. The electric field is produced by an arrangement of strip electrodes that allows the field magnitude and direction to be switched in order to boost the atoms to the circular state. A pulsed electric field ionization detector is available to monitor the excited atoms shortly after they are produced during laser setup and tuning.

The interaction region is designed to allow application of a carefully controlled electric field which defines the quantization axis for the Rydberg states. It is shielded both electrically and magnetically and cooled by a liquid helium flow system to reduce the effects of blackbody radiation.

The final detectors employ a spatially-resolved electric field ionization method. Here, the atoms enter a region of increasing electric field (1kV/cm-2kV/cm) produced by a ramped field plate. The two detectors are spaced such that the n = 30 atoms, which ionize in lower field, are picked up in the first detector, and n = 29 are picked up in the second. A quadrupole mass analyzer at the far end of the apparatus is used to

⁶ M.G. Boshier et al., *Phys. Rev. A* 40: 6169 (1989); P. Zhao et al., *Phys. Rev. A* 39: 2888 (1989); F. Biraben et al., *Phys. Rev. Lett.* 62: 621 (1989).

⁷ D. Delande and J. C. Gay, *Europhys. Lett.* 5: 303 (1988).



Figure 5. Schematic of the atomic beam apparatus. Insets detail the dissociation, production, and detection of circular states of atomic hydrogen.

monitor the intensity and dissociation fraction of the atomic beam.

A schematic of the laser system is shown in figure 6. Two independent systems produce tunable UV light near 365 nm by sum frequency mixing the fundamental output of a pulsed Nd:YAG laser with tunable yellow dye lasers pumped by the second harmonic of the YAG. One of the UV beams is frequency tripled in Kr gas to produce 121 nm light to drive the $1s \rightarrow 2p$ transition; the second UV beam drives the $2p \rightarrow n = 29$ transition. The dye lasers (figure 7) are required to have narrow spectral linewidth and are thus designed to operate in a single longitudinal mode. A feedback system⁸ keeps the laser cavity mode centered on the frequency band selected by the grating and tuning mirror.

The millimeter wave optical system is shown in figure 8. Its function is to place two beam waists of appropriate size with adjustable power, polarization, and relative phase on the atomic beam.

The apparatus is substantially complete. Improvements are now under way, and we expect to commence millimeter wave spectroscopy shortly.

⁸ Raymond et al., Opt. Lett. 14: 1116 (1989).



Figure 6. Schematic of the optical system employed in exciting Rydberg states of atomic hydrogen.



Figure 7. Detail of pulsed tunable dye laser. Laser operates in a single longitudinal cavity mode for narrow line width.



Figure 8. Layout of quasi-optical system for the 256 GHz mm-wave radiation.

2.3 Precision Mass Spectroscopy of Ions

Sponsors

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Project Staff

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In 1990 we initiated a program to substantially improve our precision mass measurement experiment. These improvements should allow us to reach a precision of about 10^{-11} in our mass measurements of individual atomic and molecular ions, the next step toward our ultimate goal of a few parts in 10^{12} . This capability will allow us to do a variety of experiments which address issues in both fundamental and applied physics, including:

- The ³H⁺ ³He⁺ mass difference, important in ongoing experiments to determine the electron neutrino rest mass;
- 2. Determination of excitation and binding energies of atomic and molecular ions by weighing the small decrease in energy, $\Delta m = E_{bind}/c^2$;
- 3. Determination of Avogadro's number N_A , by weighing γ -rays—its accurate determination would permit the replacement of the "artifact" mass standard by an atomic mass standard; and
- Improvement of many traditional applications of mass spectroscopy by orders of magnitude improvement in both accuracy and sensitivity.

Our experimental approach is to measure ion cyclotron resonance on a single molecular or atomic ion in a Penning trap, a highly uniform magnetic field with axial confinement provided by weaker electric fields. We monitor the ion's oscillation along the magnetic field lines by detecting the currents induced in the trap electrodes. Working with only a single ion is essential because space charge from other ions leads to undesired frequency shifts. This work in trapping and precision resonance draws on techniques developed by Hans Dehmelt at the University of Washington and Norman Ramsey at Harvard, for which they shared in the 1989 Nobel Prize.

We have developed techniques for driving, cooling, and measuring the frequencies of all three normal modes of Penning trap motion. Thus we can manipulate the ion position reproducibly to within 30 microns of the center of the trap, correcting for electrostatic shifts in the cyclotron frequency to great accuracy. We use a π -pulse method to coherently swap the phase and action

of the cyclotron with the axial modes.⁹ Therefore, although we detect only the axial motion directly, we can determine cyclotron frequency by measuring the phase accumulated in the cyclotron motion in a known time interval (figure 9).

In the past year we have built an entirely new Penning trap and detector, including a higher-Q resonant circuit and quieter RF SQUID; all of which should improve our signal to noise ratio by a factor of two. We have also replaced the DC electric field supply, added provisions for rapidly cycling between two ion species, and added a pressure regulator to the liquid helium bath of our superconducting magnet to help stabilize the field.



Figure 9. For each plotted point, we perform the following experiment: The initially cold ion is pulsed into a cyclotron orbit of known initial phase, and then allowed to evolve "in the dark" for an indicated amount of time, t. Then a pulse is applied which exchanges cyclotron and axial motions, bringing the ion's cyclotron action and phase into the axial mode. As the ion's axial motion rings down, its phase is detected. The appropriate multiple of 360° is added, and a line is fitted to the points. The slope of the line is the frequency difference between the frequency generator and the trap cyclotron frequency.

⁹ E.A. Cornell, R.M. Weisskoff et al., Phys. Rev. A 41: 312 (1990).

In addition, we are building ion optics and an external ion source to allow us to make the ions in a discharge at room temperature and then load them into the trap. This will eliminate the problem of residual neutral gas in the trap when using volatile species such as hydrogen and helium.

We also performed a mass "comparison" between N^{\ddagger} and itself, to check for any unsuspected effects of loading new ions.¹⁰ The results, shown in figure 10, indicate that any systematic shifts due to our ion-loading process are below the uncertainty of our previous measurement of N^{\ddagger} - CO(< 4 x 10⁻¹⁰).

With all the improvements to the system, we foresee being able to resolve the existing 10 eV discrepancy between measurements of $m(^{3}H) - m(^{3}He)$ in the next year. In addition, we plan to demonstrate a classical squeezing technique which should reduce the thermal fluctuations of our measurement by a factor of three to five. After that, we should continue development of techniques to measure two ions of different mass simultaneously. The two-ion technique in combination with the various improvements made over the last year will lead to precision in the range of 10^{-11} .



Figure 10. We loaded the same type of ion three times and measured the cyclotron frequency. The lines are a single fit to three parameters: slope, intercept, and splitting. The lower line is the fit for the open squares (ions numbers 1 and 3), and the upper line is the fit for the filled squares (ion number 2). The error bar for the splitting (not shown) is 1.8 mHz (compared to 1.1 mHz splitting). The slope is due to the trap moving in the linear magnetic field gradient as the liquid nitrogen boils off.

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2.4 Atom Interferometry

Sponsors

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Project Staff

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Using fabricated transmission gratings as optical elements for matter waves, we are constructing an atom interferometer which will physically separate atom waves before recombining them. Atom interferometers will be useful in studies of atomic properties, tests of basic quantum physics, for metrology, as a rotation sensor, and perhaps ultimately as devices to make ultra-small structures using atom holograms.

During 1990 our atom interferometer evolved from a rough and ready state to an essentially complete device. Major effort was spent on a new detector and on developing procedures to increase detector sensitivity, source modifications which have given about ten times the previous signal with more reliability, a computer data acquisition and analysis system and appropriate software, rebuilding components to reduce the vibrational noise level, and a simulation program to calculate the expected interference pattern and signal. We also worked in collaboration with the MIT Submicron Structures Laboratory to produce atom gratings with higher transmission, better dimensional stability, and less distortion. We tried several times to observe atom fringes, but were frustrated by grating problems each time. The improvements in signal to noise

¹⁰ E.A. Cornell, *Mass Spectroscopy Using Single Ion Cyclotron Resonance*, Ph.D. diss., Dept. of Physics, MIT, 1990.

and grating transmission resulted in much better atom diffraction patterns (see figure 11).

Our interferometer consists of three 0.2 μ m-period diffraction gratings equally spaced ~0.65 m apart in our atomic beam machine. The maximum sepa-

ration of the atom waves will be $\sim 60 \ \mu m$. The first two gratings separate and redirect the atomic beam forming a standing wave interference pattern in the atomic flux at the third grating, which acts like a mask to sample this pattern. Figure 12 shows the design of the interferometer.



Figure 11. Diffraction of atoms from fabricated grating.



Figure 12. Our current atom interferometer with laser interferometer stabilization system (not to scale).

The mechanical vibrations of our machine present a principal technical obstacle because they might blur the interference pattern. There are two types of required limits on vibrations. First, the three gratings must move relative to each other by less than $\sim 1/4$ period (50 nm) during the time the

final grating samples the intensity at a given position. Thus, the rms amplitude of relative vibrations integrated over all frequencies greater than the reciprocal of the detector integration time must be less than ~50 nm. The second requirement is related to the motion of the gratings due to acceleration of, or rotation about, the center of mass of the grating system during the 1.3 ms it takes for the atoms to traverse the interferometer. This means that below ~900 Hz the rms acceleration must be less than 10^{-2} ms⁻², and the rms angular velocity must be held below 10^{-4} radians per second.

Because each grating/slit assembly in the interferometer is in neither the near nor the far field of the others, it is not possible to derive an analytic expression for the interference signal. We have advanced the state-of-the-art of interferometer calculations by devising a way to cast the multiple grating problem as a convolution problem, enabling us to use Fast Fourier Transforms. A tenminute run on a CRAY computer can simulate the interferometer with an incoherent source possessing the actual velocity profile. Figure 13 shows the results of a typical calculation.

These numerical simulations have allowed us to investigate several important issues in interferometer design. We have investigated the rate at which fringe contrast degrades with mis-spacing of the three gratings and due to the spread of initial velocities (and corresponding change of deBroglie wavelengths) in the source beam. We have also examined the possibilities of constructing interferometers with varying degrees of beam collimation, and we plan to study the effects of source coherence (the collimator does not really have a blackbody source behind it).

When we have successfully demonstrated this interferometer, our first experimental objective is to make improved measurements of the polarizability of sodium and the Aharonov-Casher effect.

Publications

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Figure 13. (Top) Predicted intensity distribution at position of third grating. Atoms diffracted by different routes wind up in the major bumps shown. When different routes diffract into the same bumps, interference results, which appears as solid black. (Bottom) Detail of intensity on right side of first order bump (the one used in our interferometer). The interference pattern has the period of one grating period—hence the total transmitted intensity is a periodic function of the third grating's position.

Sub-Poissonian Statistics in the Transfer of Momentum from Light to Atoms." *Phys. Rev. Lett.* 65: 1555 (1990).

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2.5 Cooling and Trapping Neutral Atoms

Sponsor

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Project Staff

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Our current objective is to produce an intense slow atom beam and trap it in a magnetic trap. The atoms will be trapped by the forces of an inhomogeneous magnetic field on the magnetic moment of the atom. We have in mind several schemes capable of cooling the atoms to microkelvin temperatures, which will greatly increase the density in the bottom of the trap.

Dense samples of atoms cooled to microkelvin temperatures promise to open up new and exciting areas of physics. The lower interaction rate of the atoms due to their reduced thermal motion, together with the possibility of indefinitely long interaction times, make samples of trapped atoms ideal for high resolution spectroscopy and for use as atomic frequency standards.¹¹ High density samples of ultra-cold atoms will also make it possible to study interatomic collisions in a new regime characterized by a long deBroglie wavelength, the importance of weak forces, and collision durations which are longer than radiative lifetimes so that absorption, stimulated and spontaneous radiative transitions take place during a collision. This opens the possibility of controling the outcome of collisions by weak laser irradiation. High density samples of atoms at low temperature atoms are also well suited in the search for quantum collective phenomena such as Bose-Einstein condensation, and a predicted increase or decrease of the radiative decay in a dense gas of Bose and Fermi particles respectively.12

In 1990 we have made progress in our development of a continuous source of slow atoms which will be used for the study of cold collisions and to load future magnetic traps. In addition, we have completed the modeling of rf and optical absorption spectra of cold atoms in a magnetic trap.

2.5.1 Slow Atom Source

We are working on a simple, intense, continuous source of slow atoms that separates the atoms from the laser light used to slow them. Separating the atoms is a crucial requirement for further experiments because the atoms are perturbed by the intense light used to slow them. There are several promising schemes for achieving these goals, and we have successfully demonstrated the first steps in two of these schemes.

One technique uses a continuous "Zeeman slower"-a spatially varying magnetic field used to compensate the changing Doppler shifts of the atoms in the slowing process.13 Our present setup is very compact, 25 cm in length, and slows atoms with thermal velocities below 600 meters/second. By recording the Doppler profile with a second probe laser, we have observed a high flux of slow atoms with velocities of 100 to 200 meters/second. We are now designing a new magnetic field configuration which should allow us to achieve considerably smaller final velocities.

The second step in this scheme for a slow atom source is the separation of the cold atoms from the counterpropagating slowing laser beam. This will be done by using light pressure forces from the side. The deflecting laser must have two frequencies to circumvent optical pumping to hyperfine levels not excited by the laser. Furthermore, the laser frequencies have to be chirped to achieve large deflection angles, because of the changing Doppler shift of the deflected atoms. The setup for generating the frequency chirped light has been built and tested and consists of electro-optical and acousto-optical modulators and various rf generators.

Our second approach toward an intense slow atomic beam is based on our spontaneous light

¹¹ D.E. Pritchard, "Trapping and Cooling Neutral Atoms," in *Electronic and Atomic Collisions*, eds. D.C. Lorents, W.E. Meyerhof, and J.R. Peterson (New York: Elsevier B.V., 1986).

¹² K. Helmerson, M. Xiao, and D. Pritchard, "Radiative Decay of Densely Confined Atoms," International Quantum Electronics Conference, Anaheim, California, May 21-25, 1990.

¹³ J.V. Prodan, W.D. Phillips, and H. Metcalf, "Laser Production of a Very Slow Monoenergetic Atomic Beam," Phys. Rev. Lett. 49: 1148 (1982).

force trap.¹⁴ Slow atoms are collected from the thermal Maxwell-Boltzman distribution and are cooled in the intersection volume of six laser beams. A weak magnetic quadrupole field of 10 Gauss per cm induces an imbalance in the scattering forces from the six laser beams, resulting in restoring forces. About 10^6 atoms were captured from sodium vapor at a pressure of 10^{-9} Torr and trapped for about one second. We are now trying to increase the number of trapped atoms and to extract them from the trap, either by imbalance in the intensities of counterpropagating laser beams or by frequency modulation techniques.

2.5.2 Magnetic Trap for Neutral Atoms

In the last year we have performed numerical simulations to extract quantitative details form rf and optical spectra obtained in our pioneering experiments on sodium atoms in a superconducting magnetic trap. For the first time, absorption and rf spectra were obtained for trapped atoms.¹⁵ Our models allow us to derive the density and energy distribution of trapped atoms, thus making spectroscopy a powerful technique to characterize samples of magnetically trapped atoms in future experiments.

In addition to spectroscopy, we have demonstrated Doppler cooling of the magnetically confined atoms. Since Doppler cooling was applied in only one dimension, we had to take into account the coupling of all the motions inside the trap to describe quantitatively the time dependence of the cooling process. Our analysis of the absorption spectra obtained from Doppler cooled atoms (figure 14) shows that we were able to cool the thermal motion of the trapped atoms to a few millikelvin.

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¹⁴ E.L. Raab, M. Prentiss, A. Cable, S. Chu and D.E. Pritchard, "Trapping of Neutral Sodium Atoms with Radiation Pressure," *Phys. Rev. Lett.* 59: 2631-2634 (1987).

¹⁵ V.S. Bagnato, G.P. Lafyatis, A.G. Martin, E.L. Raab, R.N. Ahmad-Bitar, and D.E. Pritchard, "Continuous Stopping and Trapping of Neutral Atoms," *Phys. Rev. Lett.* 58: 2194-2197 (1987); A.G. Martin, K. Helmerson, V.S. Bagnato, G.P. Lafyatis, and D.E. Pritchard, "rf Spectroscopy of Trapped Neutral Atoms," *Phys. Rev. Lett.* 61: 2431-2434 (1988).



Figure 14. Absorption spectrum of Doppler cooled atoms and calculated lineshapes for temperatures of 2.5, 5.0 and 10 mK.