

High-temperature calcium vapor cell for spectroscopy on the $4s^2S_0 - 4s4pP_1$ intercombination line

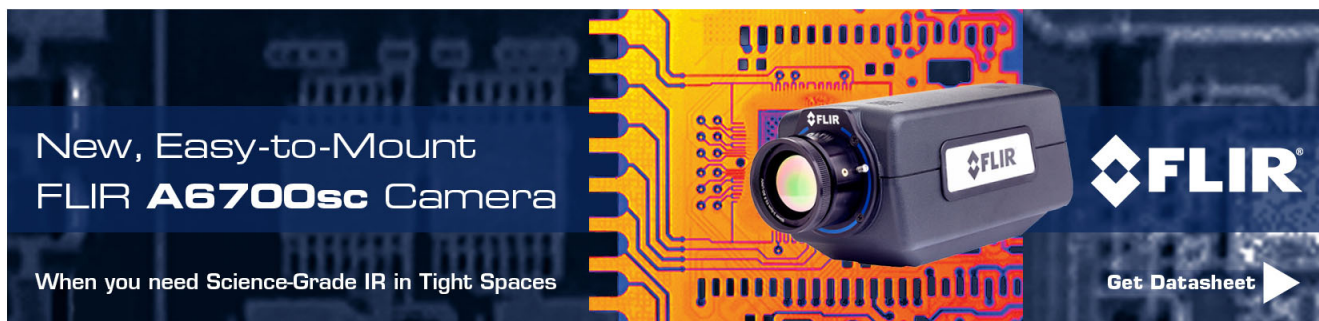
Christopher J. Erickson, Brian Neyenhuis, and Dallin S. Durfee

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High-temperature calcium vapor cell for spectroscopy on the $4s^2\ ^1S_0-4s4p\ ^3P_1$ intercombination line

Christopher J. Erickson, Brian Neyenhuis, and Dallin S. Durfee^{a)}
Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84602

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We have demonstrated a high-temperature vapor cell for absorption spectroscopy on the Ca intercombination line. The cell uses a dual-chamber design to achieve the high temperatures necessary for an optically dense vapor while avoiding the necessity of high-temperature vacuum valves and glass-to-metal seals. We have observed over 50% absorption in a single pass through the cell. Although pressure broadening in the cell prevented us from performing saturated-absorption spectroscopy, the broadening resulted in higher signal-to-noise ratios by allowing us to probe the atoms with intensities much greater than the $0.2\ \mu\text{W}/\text{cm}^2$ saturation intensity of the unbroadened transition. The techniques presented in this article could easily be applied to study other transitions in a variety of atomic species. © 2005 American Institute of Physics. [DOI: 10.1063/1.2148992]

I. INTRODUCTION

Vapor cells are often a very convenient source of atoms for spectroscopic measurements. Although they suffer from relatively large Doppler shifts compared to laser-cooled or beam sources, they are typically very simple and inexpensive devices and can often produce much larger optical densities compared to other more complicated schemes. Vapor cells are widely used, and have been for some time. Most alkali-atom-based laser-cooling experiments utilize vapor cells to lock the cooling and probing lasers to the atomic transition. These vapor cells typically consist of a glass vessel containing a small amount of alkali metal which is heated with a low-power heater or simply by contact with the room-temperature environment to produce an optically thick vapor.

Alkaline-earth spectroscopy has gained considerable interest in the past years, in part due to the use of the calcium $4s^2\ ^1S_0-4s4p\ ^3P_1$ intercombination transition to realize optical frequency standards. As such, there has been interest in creating simple sources of alkaline-earth vapors to use as references for these experiments. In our case we wanted to have a relatively simple calcium source for use in a laser lock for an excited-state laser-cooling scheme and in order to find the frequencies of the modes of a high-stability optical cavity relative to the calcium intercombination line.

The vapor pressures of the alkaline-earth elements are considerably lower than those of the alkali atoms. Furthermore, the narrowness of the intercombination line compared to the Doppler broadening of a near-room temperature vapor results in a very small effective absorption cross section, requiring high atomic densities in order to achieve appreciable absorption. As such, producing an optically dense vapor in a thermal cell requires vapors at temperatures which are incompatible with standard vacuum technology.

There are many ways to achieve higher optical densities, such as the use of an electric discharge.^{1,2} But these schemes

tend to be much more complicated than a simple vapor cell. Even with these techniques it can still be difficult to produce the needed densities for spectroscopy on the intercombination line. Before deciding to construct a high-temperature calcium vapor cell we tried to measure absorption on the intercombination line in a specially designed high-density discharge cell, but did not detect any absorption even with sensitivity below the 1% level. From measurements on the 423 nm resonance line we determined that this cell produced a ground-state density of 9×10^{11} atoms/cm³. Although the temperature of the vapor was not well known, it was definitely above room temperature. Using this low-temperature estimate we calculated that a single pass through the 10 cm discharge should result in less than 1.2% absorption on the intercombination line.

II. VAPOR CELL DESIGN

When designing a vapor cell there are several key factors to consider. The first is the length of the vapor cell. A longer cell will result in higher absorption at a given temperature. We decided that we wanted a relatively compact cell and decided to fix the length of the column of vapor to be about 10 cm. As discussed below, the total length of the vacuum chamber is a longer length of 51 cm to limit heat flow away from the vapor and to allow the ends of the chamber to be cool enough to mount to our optical table. To simplify the use of the cell we decided to design the cell to achieve our target of 50% absorption in a single pass through the cell, although the windows in the cell are large enough to accommodate multiple passes through the cell.

Given a cell length of $t=10$ cm and a target of 50% absorption, we can calculate the required density of atoms in the cell using Beer's law. Beer's law states that the fraction of light transmitted through the cell when our laser is on resonance should be equal to $\exp(-n\sigma_{\text{eff}}t)$ where n is the density of atoms and t is the length of the cell. The effective cross section σ_{eff} is lower than the natural absorption cross

^{a)}Electronic mail: dallin_durfee@byu.edu

section $\sigma_0=3\lambda^2/2\pi$ because due to Doppler broadening only a small fraction of the atoms will be in resonance with the laser light at any given time. When the laser is on resonance, only those atoms which have a Doppler shift less than about one natural linewidth will scatter significant amounts of light, and you would expect that the effective cross section should be lower than the natural cross section by a factor of the order of the ratio of the natural linewidth to the Doppler-broadened linewidth.

Because the Doppler broadening in our cell is much larger than both the natural linewidth Γ and the laser linewidth, the effective on-resonance cross section σ_{eff} is approximately equal to $\sigma_0\Gamma\lambda(m/32\pi k_B T)^{1/2}$ where λ is the resonant wavelength of the transition, m is the mass of an atom, k_B is Boltzmann's constant, and T is the temperature of the vapor. In this limit σ_{eff} does not depend on the laser linewidth. Although the effective cross section is temperature dependent, it is only mildly so—it varies by only 18% as the vapor goes from 500 to 800 °C. As such, to estimate the required temperature we can calculate the effective cross section at some reasonable temperature and then treat it as a constant. For a calcium vapor at 500 °C, the effective on-resonance cross section for the intercombination line is 8.8×10^{-20} m². With this cross section, to get 10% absorption in a column of a vapor 10 cm long requires a density of 1.2×10^{13} at./cm³. This is accomplished by a calcium vapor in equilibrium with solid calcium at 522 °C. For 50% absorption, a density of 7.9×10^{13} at./cm³ is needed, requiring a temperature of 587 °C.

The construction of a vapor cell for spectroscopy on the calcium intercombination transition is nontrivial due to the relatively high temperature required and the scarcity of vacuum components which can operate at these temperatures. Because the calcium will tend to accumulate at the coldest point in the cell, the coldest point will eventually determine the vapor pressure in the cell. In order to prevent fogging of the windows, the cell's windows should be kept considerably hotter than the temperature of the vapor. As such, producing a 587 °C vapor requires portions of the cell to be considerably hotter than 587 °C. A blown glass cell would become soft and collapse at the required temperatures. The most commonly used metal-to-metal or glass-to-metal seals can only be baked up to about 450 °C. The only inexpensive and highly reliable way to make metal-to-metal vacuum seals at these temperatures are to weld or braze components together. In order to evacuate the chamber, a high-temperature valve is also required. Commercially available high-temperature valves are relatively expensive and typically cannot be baked above 450 °C.

To get around the expense and limitations of available high-temperature vacuum components, we decided to use a two-chamber design. In this design the hot calcium vapor is contained within an inner chamber which is surrounded by an outer chamber. Because the vapor is contained in the inner chamber, the entire outer chamber does not have to be held at high temperatures. This allows simple commercially available seals and valves to be used on the outer chamber. And because the outer chamber is evacuated, the seals between the inner chamber and outer chamber do not have to be

vacuum tight in order to maintain vacuum in the inner chamber. The only requirement on the seals between the inner and outer chambers is that they leak calcium vapor at a sufficiently low rate to allow the cell to operate for a long time without maintenance.

A scheme similar to our two-chamber design is discussed in Ref. 3. In this design there was no seal between the inner and outer chambers, but long narrow tubes were used to limit calcium diffusion out of the hot inner chamber. Because the inner chamber was evacuated through these tubes, no high-temperature valve was required. Optical access to the vapor was also done through these tubes, making window seals unnecessary. The large diffusion rate of calcium out of the inner chamber resulted in rapid fogging of the outer-chamber windows. This was overcome in Ref. 3 using mirrors inside the outer vacuum to deflect the light to windows which were away from the direct line of sight of the inner cell. These mirrors were quickly coated with calcium, which reflected the light reasonably well. We chose not to implement this design for two reasons. First, we expected that calcium-coated mirrors would have a tendency to oxidize when the chamber was vented to reload it with calcium. Second, and more critical, the diffusion rate of calcium through the tubes was rather large—in the tests reported in Ref. 3 the cell ran for only three days, running at 460 °C using an unspecified amount of calcium. To avoid the trouble of continually reloading the cell we designed a cell which uses windows to seal the calcium into the inner chamber.

One of our goals for the cell was to maintain a vapor dense enough to achieve 10% absorption for over 1000 h, with an initial load of 5 g of calcium. This implies a maximum allowable leak rate which is equivalent to that of a hole with a cross section of about 1 mm². While this is a very large hole by vacuum standards, it does require some care to be taken. For example, the two 20-mm-diam windows in our design have a combined circumference of 13 cm. If the average distance between the edge of the windows and the mating surface is greater than 8 μm, the calcium leak rate will be too large. Looser requirements could be achieved simply by loading the cell with more calcium. But this would increase the cost of operating the cell and increase the risk that over time enough of the calcium leaking out of the inner chamber would condense on the outer-chamber windows to significantly reduce their transparency.

Our design is shown in Fig. 1. In this design the inner chamber is sandwiched between the two halves of the outer chamber. The inner chamber is 10 cm in length and 5.1 cm in diameter. This diameter was chosen to accommodate the mounting of 20-mm-diam windows on the ends of the inner chamber. Each half of the outer chamber is 20 cm long to allow the far ends of the cell to be relatively cool compared to the temperature of the inner chamber. The outer cell uses standard off-the-shelf metal-to-metal flanges with copper gaskets, and the windows on the outer cell are standard commercial vacuum windows.

To evacuate the cell, one half of the outer chamber is connected to a vacuum pump through a valve. The two halves of the outer chamber are connected by a 5/16 in. steel tube, allowing them to both be evacuated at the same time.

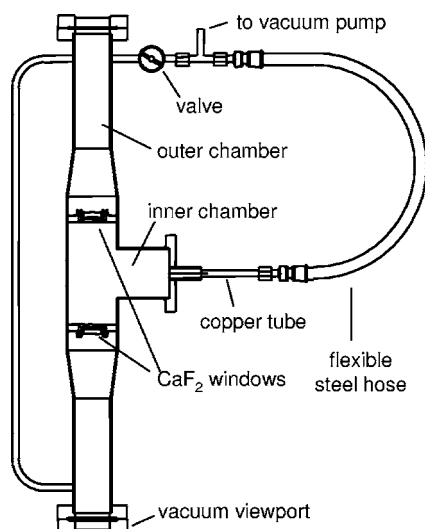


FIG. 1. Diagram of the high-temperature vapor cell. The diagram is to scale.

The inner chamber is connected to the outer chamber through a long flexible steel tube connected to a short piece of copper tubing which is brazed into a plate at the top of the inner chamber (see Fig. 1). After evacuating the inner cell through this copper tube, we crush the tube with a pinchoff tool commonly used by plumbers to rapidly seal water leaks in copper tubing. We found that on a clean piece of copper this tool creates a vacuum-tight seal as long as the tool remains on the tube. To create a permanent seal, before removing the tool we cut the tube off just above the pinch and fill the stub with silver solder. The copper tube is made long enough to allow us to do the pinchoff procedure several times before we have to braze a new tube into the cell.

Two windows separate the inner chamber from the two halves of the outer chamber. To create a tight-enough seal to contain the calcium vapor, copper gaskets are placed between the windows and a flat steel mating surface. Each window is then compressed by a metal ring held down with three bolts. The copper gaskets have beveled edges to create a knife-edge contact on each side of the gasket, similar to the gaskets used in the Noble-Kasevich seal,⁴ but with no attempt to obtain a vacuum-tight junction. Sapphire windows were first used, but even when held at a temperature over 100 °C hotter than the calcium vapor they quickly fogged over (possibly due to a chemical reaction). We eventually switched to calcium fluoride windows, which have not shown any significant fogging and which have a thermal-expansion coefficient which is a much better match to stainless steel and copper.

The cell is heated by two band heaters at the locations of the windows and by a high-temperature heat tape wrapped around a cylindrical aluminum sleeve which is slid over the copper tube. The maximum temperatures that the cell can maintain are set by the maximum-rated temperatures of the heaters. The band heaters can be heated as high as 800 °C. The heat tape is rated to go up to 740 °C, sufficient to produce atomic vapors with nearly 100% absorption. If not wrapped properly, however, heat tape can generate “hot spots” and fail. As such, we ran the heat tape conservatively, well below the maximum-rated temperature.

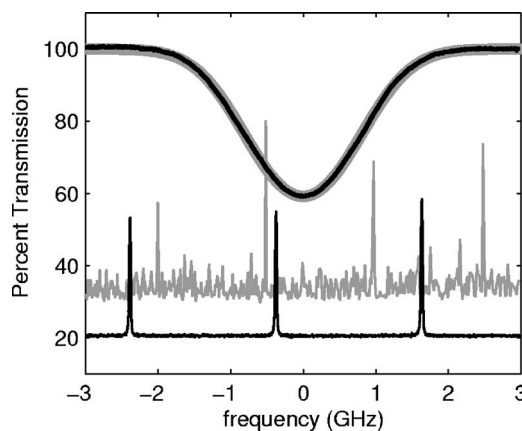


FIG. 2. Absorption spectra. The black line at the top of the figure is the measured transmission through the cavity as a function of the laser detuning from the center of the intercombination line. It is drawn over a thicker grey line which shows the calculated absorption for a 10-cm-long column of Ca vapor at a temperature of 750 °C (the temperature of the hottest parts of the inner cell) but with a density consistent with a solid source of calcium which is at 580 °C. Superposed on this graph are the transmission through two Fabry-Perot etalons (in arbitrary units with arbitrary offsets) as a function of the laser detuning. The black line with three sharp peaks at the bottom of the graph represents the transmission through the reference etalon which we used to measure how far our laser had scanned. Above the reference etalon trace in grey is the transmission through the ultrastable high-finesse cavity whose modes we measured relative to the absorption line using our vapor cell.

The inner chamber is wrapped in several layers of high-temperature fiberglass insulation and aluminum foil. The outer chamber is not insulated, resulting in large temperature gradients such that the far ends of the cell are at a temperature of less than 100 °C during operation of the cell. This allows us to seal the ends of the cell with standard vacuum view ports and makes it possible to mount the cell to our optics table without generating a significant heat load. Since standard vacuum seals are used, we were able to easily add a vacuum “T” to one side of the outer chamber in order to accommodate a vacuum gauge.

After loading calcium into the cell the cell is first heated using just the two band heaters, ensuring that the windows are the hottest parts of the inner chamber and thus preventing calcium buildup on the windows. Eventually, the calcium migrates to the colder copper tube and the vapor density drops. After this migration occurs we typically operate the cell with the band heaters fixed at 750 °C and use the heat tape to control the calcium vapor pressure.

III. RESULTS

A typical absorption spectrum from our cell is shown in Fig. 2. All of our absorption data was taken using a grating-stabilized diode laser of the design described in Ref. 5. A portion of the laser light was sent to a reference etalon to measure how far the laser was scanned when taking absorption spectra. To verify the calibration of the etalon’s free-spectral range, we scanned the laser frequency over a large range such that we could compare the etalon to a commercial wavelength meter. This measurement agreed with the manufacturer’s specified free-spectral range of 2 GHz to within 0.5%.

A beam splitter positioned just before the light entered the cell directed a portion of the beam to a photodiode so that we could record a laser intensity reference to normalize away changes in the laser intensity during the scan. Upon exiting the cell the light passed through a pair of interference filters to block the blackbody radiation from the cell. The light level was then detected on a photodiode. During each scan a digital oscilloscope recorded the light power before the cell, the light power after passing through the cell, and the amount of light passing through the reference etalon. To remove stray-light background levels, a second set of traces were recorded with the light blocked near the laser.

To process the data we first subtracted the background from the scans of the laser power before and after the cell. We then divided the absorption signal at each point in time by the intensity reference. Next, we calculated a second-order polynomial fit to the location in time of each peak measured on the reference etalon and used this fit to convert the time axis for each of our scans into a calibrated frequency axis corrected for the nonlinear scan of the laser's frequency in time (see Fig. 2). Finally, we corrected for etaloning effects due to the windows in the cell by fitting the very edges of the absorption signal, where essentially no absorption occurred, to a second order polynomial. We subtracted the polynomial from the absorption scan and then normalized the scan so that the leftmost edge was equal to 1.

To test the longevity of the cell we loaded the cell with only 0.5 g of calcium and ran the cell for over 60 h at high-enough temperatures to produce between 30% and 50% absorption. Because the rate at which calcium leaks out of the inner cell is proportional to the density of the vapor, we can conclude that the amount of calcium which leaked out of the inner chamber while we ran the cell is equivalent to the leakage that would occur while operating the cell for over 200 h at a lower temperature to produce 10% peak absorption, suggesting that we have surpassed our design goal of 1000 h with 5 g of calcium.

We have compared our absorption data to a model of the expected absorption profile. We first started with a model which had just one free parameter, the temperature of the vapor. Assuming that the temperature of the solid calcium (which acts as the source for the calcium vapor) was similar to the temperature of the vapor, we expected that this one parameter would describe both the Doppler width of the absorption curve as well as the density of the vapor (which, along with the Doppler width, determines the on-resonance absorption). Although this simplest model produced reasonably good results on our earliest data, after running the cell for several hours the measured absorption curves became noticeably wider than the curves predicted by our model.

The fact that our single-parameter model does not fit most of our data well is not surprising. One would expect that once all of the solid calcium had migrated into the copper tube, which was at a significantly lower temperature than the rest of the inner chamber, a somewhat more complicated two-parameter model would be needed. In this model the temperature of the solid calcium, which acts as the source of the vapor, and the effective temperature of the vapor were entered as separate free parameters. We also tried

fitting our data to a Voigt profile model which included a third free parameter to account for possible effects of pressure broadening.

Our two-parameter model fits the data reasonably well when we assume that the temperature of the vapor is close to the temperature of the windows (see Fig. 2). Although we do not know the precise temperature at the location of the solid calcium, from measurements at the top and bottom of the copper tube we have a range of possible temperatures. From the curve fits to our data it is apparent that the density of the vapor is consistent with this range of temperatures.

When we added pressure broadening to our model it was very obvious that the Voigt profile did not have the same shape as the data unless pressure broadening was assumed to be less than 100 MHz. This implies that most of the disagreement between our simple one-parameter model and the measured data is due to the higher temperature of the vapor rather than to the pressure broadening. Because 100 MHz is much smaller than the Doppler width, we can only ascertain an upper limit on pressure broadening through curve fitting. We have determined a lower limit on pressure broadening by observing an increase in the saturation intensity; at the cell temperatures used to take the data in Fig. 2 we found that using all of the power from our laser we could increase the intensity of our light to over 10 000 times the saturation intensity for a collision-free gas without seeing any significant reduction in absorption, implying a lower limit on the pressure broadening of several megahertz.

Because the pressure-broadened width is significantly smaller than the Doppler width, a simpler absorption model which does not include pressure broadening is still quite accurate. This makes data analysis much faster, avoiding repetitive calculations of the integral in the Voigt profile to optimize a curve fit. The simple model works because the area under the absorption curve of a pressure-broadened gas is independent of the amount of pressure broadening. This can be shown without any knowledge of what the atoms are colliding with, what the collision frequency is, or even whether the line shape is Lorentzian or if the impact approximation applies. We only need to assume that collisions only result in phase shifts with no direct excitation or deexcitation. From the atom's frame it is equivalent to think of the laser field's phase, rather than the atom's phase, as changing during the collision, resulting in a light field which is effectively frequency broadened. As such, pressure broadening can be thought of as the redistribution of optical power to different frequencies (from the perspective of the atom), with the total power in the light field remaining constant.

Pressure broadening in our cell is a disadvantage if precise location of the atomic transition frequency is required. But for our purpose, finding the frequencies of the modes of an ultrastable high-finesse cavity relative to the absorption line, we only needed to find the top of the absorption line to an accuracy of about 10 MHz. And because the saturation intensity of collisionless calcium vapor is just $0.2 \mu\text{W}/\text{cm}^2$, the increased saturation intensity due to collisions allows us to send much more light through the cell and make huge improvements in the signal-to-noise ratio. At these intensities our principal noise source was the digitization noise of the

digital oscilloscope which we used to acquire data, and with little effort we acquired clean-enough data to find the center of the Doppler-broadened line within the required 10 MHz. With our high-stability optical cavity we determined that the center of the line did not shift measurably over the range of temperatures used in the cell. For us the only disadvantage of pressure broadening was the fact that we were unable to reach the increased saturation intensity with our ~ 1 mW laser beam and were therefore unable to perform saturated-absorption spectroscopy.

IV. DISCUSSION

We have demonstrated a high-temperature two-chambered vapor cell for absorption spectroscopy on the calcium $4s^2\ ^1S_0-4s4p\ ^3P_1$ intercombination transition. We have achieved high absorption and a long lifetime using a fairly simple and very robust design, without needing to use expensive or complicated high-temperature seals or valves. Although the seals between the two chambers do not need to be vacuum tight, we have found that care must be taken in order

to keep the calcium loss rate reasonably small. Significant pressure broadening in the cell has been used to our advantage, allowing higher light intensities to be used, thereby improving the quality of our data. These techniques could easily be extended to perform spectroscopy on other transitions in different atoms which require similar temperatures.

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