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High-resolution diode-laser spectroscopy of calcium

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Abstract. Saturated-absorption signals on the calcium 657 nm transition are observed by direct absorption using diode lasers and a high flux atomic-beam cell. Linewidths as narrow as 65 kHz are observed with a high signal-to-noise ratio. Prospects for using this system as a compact wavelength/frequency reference are considered.

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The intercombination transitions of the alkaline-earth atoms have been recognized as important optical frequency/wavelength references. Prominent among these, in terms of development effort, is the calcium transition at 657 nm which has a 400 Hz natural linewidth. Important advances in research on this system have come from a number of laboratories including saturated-absorption signals [1], atomic-beam optical Ramsey fringes [2–6], resolution of photon recoil [7], observation of second-order Doppler perturbation of Ramsey fringes [8], and suppression of one recoil component [9]. Laser cooling of calcium has also been demonstrated and promises significant improvement in accuracy and precision in the future [10–12].

Recent experiments on very high resolution spectroscopy of laser-cooled magnesium atoms have provided beautiful results that now demonstrate some of this promise [13]. In addition, diode-laser spectroscopy of barium [14] and strontium [15] show optical resonances with the potential to be used as wavelength references. All of these alternative systems have inherent advantages and disadvantages that could favor one for specific applications.

The reasons to choose the calcium (657 nm) transition are mostly well known; some are fundamental and others are of a practical nature. These include relatively small field shifts, no ground-state hyperfine structure, potential for laser cooling, visible wavelength (hence, easy to use for interferometry etc.), recognition by standards community [16], projected accuracy much higher than I_2 stabilized lasers, and the expectation of an actual frequency measurement that will provide absolute calibration of frequency/wavelength [17]. A renewed interest in calcium is the result, at least in part, of its compatibility with existing semiconductor diode-laser technology. This technology will allow a significant reduction in size and cost that may lead to construction of a compact portable transfer standard. Not only are high performance diode-lasers possible at 657 nm [18], but high-power diode lasers can also be frequency-doubled efficiently to produce the 423 nm light required for laser cooling [19].

In striving for high accuracy and stability in optical references we need unperturbed transitions with very narrow linewidths and high signal-to-noise ratios. The quantum nature of atomic transitions results in a fundamental tradeoff between the linewidth and signal-to-noise ratio for spectroscopic measurements. This is readily apparent in trapped-ion experiments that use single ions for the highest accuracy [20]. With neutral atoms we can hope to achieve higher signal-to-noise ratios by using more atoms, but we must pay the very serious penalty of limited observation time, and hence linewidth. Even using laser cooling/trapping/fountain methods with neutrals has limitations that result from the perturbing effects of the trapping fields. These cannot be ignored and generally require that we turn off the trapping light. Present best-case neutral-atom fountains have unperturbed observation times of ≈ 1 s; whereas trapped-ion experiments have nearly unlimited observation times. We anticipate that to achieve the highest accuracy laser cooling will be required, but we report here about a system in which ultimate accuracy is traded for short term stability, compactness and the future potential for portability. Such a system could serve as a relatively

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simple transfer standard of optical wavelength/frequency.

1 Experimental system

Our simple diode-laser saturated-absorption system is diagrammed in Fig. 1. To conserve the optical power we use a minimum number of components in the optical beam path. In contrast to previous high resolution studies of calcium we have done the saturated-absorption in a high-flux hybrid beam-cell rather than in a traditional low-density atomic beam. The idea here is to take advantage of the low amplitude noise on the diode laser by detecting the absorption directly. The high-flux beam provides narrow resonances and high signal-to-noise ratios and avoids the problems associated with buffer gases. As usual, absorption has advantages and disadvantages compared to the common fluorescence detection systems. For fast calcium atoms the traditional fluorescence detection methods have detection efficiencies (signal-to-noise ratios) that are limited by the 20 cm long decay length of the 657 nm fluorescence. The signal-to-noise ratio of our absorption signal was limited by the diode's amplitude noise power which was within a factor of 5 of the shot-noise limit at the 1.5 MHz detection frequency we chose.

Our high-flux beam is generated from calcium metal that is heated in a stainless steel boat (internal dimensions $1 \times 1.1 \times 10\,cm^3$). The boat is capped with a collimating nozzle which is formed by a narrow channel (480 μm wide) that is cut through a 4.5 mm thick stainless steel plate. A wire-EDM (Electric Discharge Machine) was used to cut the continuous channel in a pattern that forms a rectangular zig-zag across the surface of the plate. The rectangular zig-zag pattern is contained in an area of 3 mm wide by 8 cm long and consists of 106 interconnected parallel slots. The result is a net nozzle area of $\approx 115\,mm^2$. When the calcium vapor pressure is low ($T \leq 800$ K) the nozzle should produce an atomic beam divergence (in the direction of the laser propagation) of about 12 degrees. With a single fill of calcium the beam-cell can be operated for about 15 h. Counter propagating laser beams intersect the atomic beam ≈3 cm downstream from the nozzle. The calcium beam then condenses on a water cooled surface ≈4 cm beyond the interaction region. Under normal operating conditions the source temperature is ≈900 K and produces a Doppler-broadened absorption of about 3%. In this case the calcium pressure in the nozzle is higher than necessary for effusive flow; hence we observe a Doppler width of about 700 MHz. The present beam-cell has a volume of about 4 l and is now pumped with a turbo pump, although ion pumping seems feasible. With some minor design modifications of the nozzle and thermal/vacuum enclosure the beam-cell should be portable and have a much longer calcium source lifetime.

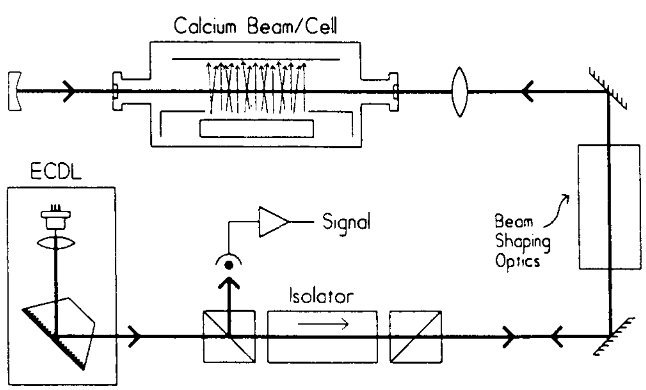

Fig. 1. Saturated-absorption spectrometer for calcium. The output from an extended-cavity diode laser passes through a two-stage optical isolator (isolation ≈60 dB), beam-shaping optics and then on to the calcium beam cell. The return beam is deflected by the isolator and detected with a photodiode

In this experiment the extended-cavity diode lasers [21, 22] use commercial index-guided lasers in conjunction with a special holographic optical selectors [23] for wavelength control. Although other good optical designs are possible, the holographic selectors are attractive because they provide both high diffraction efficiency (60–90%) and high resolving power (≈3600 lines/mm). In addition, and in contrast to traditional ruled gratings, the high diffraction efficiency is obtained when the laser polarization is parallel to the grating lines.

To improve the single-mode tuning range and stable output power of the extended-cavity lasers it is important to have good Anti-Reflectance (AR) coatings on the laser's output facets [24–26]. Unfortunately, almost all of the commercial laser manufacturers use optical coatings on the facets in order to insure long lifetimes, and to tailor the laser's facet reflectance for optimum output power. These coatings obviously constrain the performance of any additional AR coatings that we might add. But even without removing the manufacturer's coatings we can achieve good AR coatings with a number of different coating designs. For example, we commonly use a two-layer coating that consists of Al_2O_3 plus HfO_2 to achieve modal reflectance as low as 1×10^{-3}. With these coatings we obtain good operating conditions of extended-cavity red lasers with coarse tuning within ± 10 nm of the free-running laser wavelength. Starting with a 3 mW, single spatial-mode, commercial laser, the extended-cavity systems typically provides ≈1.5 mW of usable power in a single spectral mode. These extended-cavity lasers typically have a free-running fast linewidth of about 30 kHz for a 10 cm extended-cavity length. Linewidths are determined by direct beat note measurements.

To further narrow the laser's linewidth we use the Pound-Drever-Hall [27] method to lock one of our red lasers to a stable reference cavity. For fast electronic control of this laser's frequency an ADP electro-optic modulator is used inside the extended cavity. The use of the electro-optic modulator helps to suppress the coupling of frequency modulation to amplitude modulation in the diode laser system. When locked this laser has a fast-linewidth of about 500 Hz and a residual frequency jitter (due to vibrational perturbations of the reference cavity) of about 5 kHz [18]. The laser linewidth with respect to the reference cavity may be decreased by extending the servo bandwidth beyond the present 1 MHz. Presently, the laser's linewidths do not contribute significantly to our observed calcium transition width (≈ 65 kHz). Both the electronically stabilized laser and unlocked, but long (≈ 30 cm), extended cavity lasers give calcium resonance widths that are similar. The observed transition linewidth is presently dominated by transit and wavefront curvature effects. We also note that Simonsen [29] has reported an alternative laser narrowing scheme that combines optical-locking with a grating-tuned extended-cavity laser.

As is usual for diode lasers the spatial mode is both asymmetric and astigmatic. The mode also usually contains additional small aberrations that are caused by imperfections in the beam path (the holographic selector in particular). Some effort is then required to process this optical beam to obtain good quality optical wavefronts for saturation spectroscopy. At present we use beam-forming optics that includes: a cylindrical lens ($f \approx 4$ m) to correct astigmatism, two sets of anamorphic prisms to shape the beam, and two spherical lenses for collimation. The resulting beam then has a nearly rectangular shape 4 mm × 6 mm with a wavefront flatness of about 1 fringe.

2 Results

For typical operating conditions with the beam-cell system the measured calcium saturation-dip is about 1% of the laser power, while the Doppler background absorption is about 3%. Using optical heterodyne techniques [30] produces saturated-absorption signals with high signal-to-noise ratios as shown in Fig. 2. So far the narrowest lines we have measured with our beam-cell system are 65 kHz (FWHM) which corresponds to a resonance $Q \approx 10^{10}$. These signals are observed with about 1 mW of optical power and provide a signal-to-noise ratio of about 30 in a 40 kHz detection bandwidth.

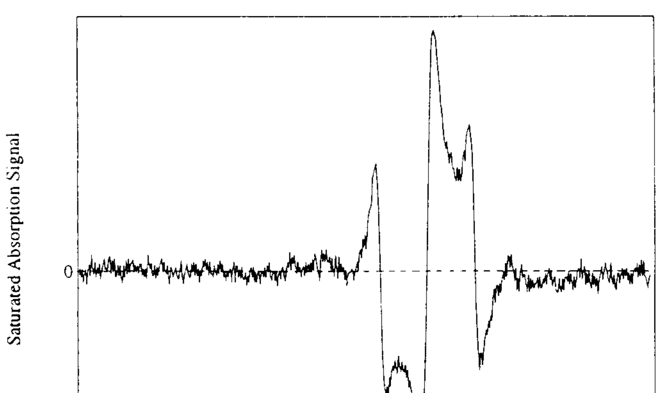

Fig. 2. Calcium saturated-absorption signal observed using the system diagrammed in Fig. 1. The lineshape results from the optical-heterodyne detection method using frequency modulation of the laser at 1.1 MHz. This example shows a calcium resonance with a 100 kHz width with a nearly flat background. A good signal-to-noise ratio is achieved even using a 60 kHz detection bandwidth

If optimally used, the noise-limited-stability of a diode laser locked to this resonance would be $\sigma_y(\tau) \approx 3 \times 10^{-14} \tau^{-1/2}$. This estimate may be reasonable for short and medium averaging-times ($100\ \mu s \leq \tau \leq 100$ s) but will not persist for long averaging-times which will be limited by systematic effects. The time scale at which long-term systematic perturbations become important has not yet been determined for this system. Based on experience with similar systems, we might expect the stability to degrade for averaging times larger than a few hundred seconds. This projected stability of the calcium beam-cell saturated-absorption resonance is excellent and comes from a system that is still far from optimized.

With better quality optical beams we can anticipate narrowing the single-zone saturated-absorption signal by a factor of 2 or 3. This improvement of the spatial mode will require higher quality windows and optics along the beam path as well as interferometric quality alignment in the interaction region. In the beam forming process a number of factors are traded against each other in order to achieve spectrally narrow saturation signals while maintaining a high signal-to-noise ratio. These tradeoffs include: limited optical power, poor initial spatial-mode quality, and the desire to have a large spot size (because of significant transit and wavefront-curvature broadening) [31, 32]. A preferred method may be to use the optical Ramsey-fringe technique proposed by Baklanov et al. [33] and pioneered by Bergquist et al. [3,34]. These methods have already demonstrated linewidths as narrow as a few kilohertz [3,7,8].

The most significant factors that limit the resolution and accuracy of non-cooled neutral-atom optical frequency/wavelength references are velocity dependent shifts such as residual Doppler, second-order Doppler, transit and wavefront curvature effects. Other factors that will contribute smaller uncertainties include photon recoil (23 kHz splitting), gas lens [35], coupling of laser AM and FM, quadratic Zeeman (10^8 Hz/T^2) and Stark [≈ 1 Hz/(V/cm)^2] shifts. Many of these effects are complicated by their dependence on the system geometry and atomic trajectories and will have to be evaluated in detail for any specific system realization. In particular the most troublesome shifts are caused by the wavefront curvature and second-order Doppler effects that are determined by averaging the atomic velocity distribution over the spatial distribution of the laser field. These effects will typically limit non-cooled optical frequency standards to an accuracy of about 1 kHz [6,31].

The second-order Doppler shift for hot ($T \approx 900$ K) calcium atoms is significant ($\approx 2 \times 10^{-12}$ for the most probable velocity of ≈ 740 m/s) and will have to be evaluated for any proposed standard. Fortunately, by using the optical transitions the atomic velocity distribution can be measured with some accuracy. The second-order Doppler shift can then be calculated and subtracted as is done for cesium atomic frequency standards. For these microwave primary standards uncertainties of a few percent of the second-order shift are achieved, but the uncertainties will likely be larger for the laser saturated-absorption case.

In evaluating the potential accuracy and precision of calcium standards there are still other unresolved questions, one of which is the possibility of pressure shifts of the 657 nm transition. While pushing towards the highest accuracy frequency standards it has been discovered that significant pressure shifts affect even trapped-ion systems with pressures as low as 10^{-8} Pa ($\Delta\upsilon/\upsilon \approx 2\times10^{-13}$ on an rf transition of 303 MHz in Be) [36]. Similarly, Cs-Cs shifts have been measured in Cs fountain system ($\Delta\upsilon/\upsilon \approx 10^{-12}$ for a cold Cs density of $\approx 3\times10^{9}\,\mathrm{cm}^{-3}$) [37]. In unpublished data, Barger measured pressure shifts of the 657 nm saturated-absorption transition due to argon and krypton collisions. The measured shifts were $\approx$68 kHz/Pa (9 MHz/Torr) at low pressures. Unfortunately, the corresponding pressure shifts are not known for calcium perturbed by calcium, or other atoms that might be found in a typical vacuum system. The density of calcium atoms in the interaction region of our high flux beam is about $10^{12}/\mathrm{cm}^3$ ($\approx 10^{-2}$ Pa) with a background gas pressure of about 7×10^{-5} Pa. If, for purposes of discussion, we assume a Ca-Ca pressure shift of the same magnitude as the measured argon shift, our present system would have a pressure shift of $\approx$1 kHz. Linewidths measured as a function of calcium beam flux and system background pressure from 10^{-3} Pa to 10^{-4} Pa show no indication of pressure broadening. This limited diagnostic gives some indication that pressure shifts are not playing a significant role at our present resolution (65 kHz), but these will have to be evaluated in future higher accuracy systems.

3 Summary and future prospects

Using diode lasers for high resolution saturation spectroscopy of calcium demonstrates the potential for a frequency/wavelength reference in the visible that could achieve very high precision with transportability. The calcium system should compete favorably with iodine stabilized lasers (scatter in frequency accuracy of good iodine cells is about 10 kHz) [38]. These initial results on calcium saturated-absorption in a beam-cell provides narrow linewidth signals with high signal-to-noise ratios. We can expect very high short-term stability from this system. Careful evaluation of all possible systematic effects, and intercomparisons of various system designs, will be required to determine the ultimate accuracy of the calcium saturated-absorption resonance.

It is clear that cold (low velocity) atoms will be required to achieve the ultimate resolution and accuracy. We are presently a long way from the 400 Hz natural-lifetime-limited resolution that is possible on the 657 nm transition, but we expect that this will be achieved even with an all-diode-laser system. The 423 nm cooling transition in calcium is a very fortuitous match for frequency-doubling high-power diode lasers. Using $KNbO_3$ in a ring build-up cavity and a 150 mW diode laser at 846 nm, we have produced a usable beam of 35 mW at 423 nm [19]. To improve the long term stability and tuning of the blue light, we have injection-locked the 150 mW diode laser using an extended-cavity diode laser (output power $\approx$23 mW) as the master oscillator. The injection locking narrows the high-power laser linewidth and allows continuous scans of about 7 GHz. The power and stability of the blue light which we have achieved should be adequate to test the fundamental resolution limits for calcium.

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