High Resolution Spectroscopy of Calcium Atoms

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Abstract

The recent results on saturated absorption optical interference spectroscopy of calcium are presented. The photon recoil splitting of the Ca $^1S_0$ - $^3P_1$ intercombination line at 657 nm has been fully resolved. Linewidths as narrow as 3 kHz half width half maximum intensity (HWHM) are reported for radiation beams spatially separated by up to 3.5 cm. Second order Doppler is shown to be the present limitation to the accuracy of this technique. Methods are discussed which could lead to an optical wavelength/ frequency standard with an accuracy of better than 10^{-14}.

Introduction

Saturated absorption with spatially separated optical fields [1-5] permits extremely high resolution with good signal-to-noise ratio and can be accomplished with simple, easily achievable, optical systems. Use of this optical Ramsey interference method with highly stabilized dye lasers and long lived transitions [6-8], such as the $^1S_0$ - $^3P_1$, intercombination resonance lines of Ca and Mg can now lead to resolution of the photon recoil doublet. This partially removes the accuracy limitation previously imposed by unresolved recoil components in the visible spectrum. We summarize our investigations of the Ca $^1S_0$ - $^3P_1$ line at 657 nm using the above methods with which we have obtained linewidths as narrow as 3 kHz HWHM and completely resolved the recoil doublet which is split by 23.051 kHz. Also we discuss techniques which could lead to an optical wavelength/ frequency standard for this line with an accuracy of better than 10^{-14}. Fractional frequency stability for such a calcium stabilized dye laser oscillator is calculated to exceed 10^{-16}.

The Ca transition [6-7] is very suitable for measurements of the highest level of accuracy, such as those in metrology, in spectroscopy and in various relativity experiments. It has a long lifetime of 0.39 ms (natural linewidth of 410 Hz), small electromagnetic field shifts of about 10^8 Hz/T^2 (1 Hz/G^2) and 1 Hz/(V/cm)^2 for the $\Delta m_J = 0$ transition. Its transition probability allows an optimum laser power of less than 1 mW. ^{40}Ca possesses a nondegenerate ground state and has zero nuclear spin giving no hyperfine structure. A complete resolution of the recoil peaks and full knowledge of the distributed velocity contribution to the Ramsey fringes of each recoil component should permit precise definition of line center.

Basic Concepts

The theory of photon recoil effects in saturation spectroscopy has been discussed by Kol'chenko, et. al. [9] and more thoroughly by Bordé [2,10]. Conservation of 4-momentum (momentum and energy) requires that, in the absorption process, the atom absorbs the energy and momentum of the photon, thereby changing not only the atom's internal energy, but also its motional energy. Similarly, in the emission process the atom must change its kinetic and internal energy to provide for the emitted photon's energy and momentum. If there are no other energy sources available, then the kinetic energy of recoil must be provided by the absorbed or emitted photon. This requires that the frequency of the absorbed photon be blue shifted relative to the Doppler shifted natural resonance frequency of the atom and, correspondingly, that the emitted photon be red shifted. In linear spectroscopy only the absorption or emission process is observed, with the result that the frequency shift is not directly discernable (in fact it could be very easily included in the definition of the two level atom state energy difference). Saturation spectroscopy includes both emission and absorption aspects, thus, with sufficient resolution, the counterrunning probe wave will disclose the effect of the recoil frequency shift as a doublet Lamb dip structure, i.e., there will be two frequencies of anomalously high transmission for the probe wave. One occurs at the frequency where the atom's ground state density is reduced by the power wave interacting with the same set of absorbers. The second transmission peak comes at the frequency where both the power and probe waves interact with the same excited state velocity class absorbers. The two peaks are symmetrically displaced from the location of the ordinary Bohr frequency with the total frequency splitting being $\delta\nu = hk^2/4\pi^2 m$, where h is Planck's constant, k is the wave vector amplitude, and m is the atomic mass. For ^{40}Ca, this recoil splitting amounts to 23.051 kHz for the $^1S_0$ - $^3P_1$ intercombination line at 6573 Å.

Until now the only instance of photon recoil resolution in this optical region has been that of Hall, et al. [11], with the methane saturation absorption peaks at the longer 3.39 μm wavelength. The resolution represented in the methane experiment of $\sim 2 \times 10^{-11}$ came with continuous excitation (one zone) throughout the interaction region with many hours of integration time and high quality, large aperture optics. In fact, in most spectroscopy experiments, the radiation field extends more or less uniformly throughout the volume in which move the absorbers to be studied. Ramsey [12] was the first to point out that this may not be the most advantageous method in which to apply the oscillating driving field. He observed that useful spectroscopic information could be obtained if the amplitude and phase of the radiation field were non-uniform throughout the region. Of particular value is the arrangement that allows independent phase evolution of atoms and radiation field between interactions of atom and field. A simple scheme to produce this field on, field off, field on effect is to intercept a beam of atoms with spatially separated radiation fields. This gives an absorption profile dependent on the absorber's natural transition frequency with spectral width determined by the time between the radiation regions, rather than the transit time through each region. To help visualize this, we note that the interaction of absorber and field in the first radiation beam produces a coherent superposition of upper and lower states. This results in a dipole which oscillates at the natural resonance frequency in the field free region between the radiation zones. The effect of the second light field depends on the phase of the radiation relative to the absorber's oscillations, so that the absorbers passing through this field will either be further excited or returned to the ground state by stimulated emission. Thus, an interference results which produces line narrowing since the quantum absorption transition probability is dependent not only on the frequency of the driving field, but also on the phase evolution difference of quantum system and field between zones. The phase evolution difference is proportional to the interzone transit time.

For detailed discussions of saturated absorption optical Ramsey interference with separated oscillatory fields the reader is referred to theoretical discussions of Baklanov, et al. [1] and Bordé [2] and to the treatments of Bergquist, et al [3-5]. But for completeness in this paper let us remind the reader of the important features. For three equally spaced, parallel, standing wave radiation fields, transversely crossed by a monovelocity atomic beam, the line profile is a complex combination of linear and nonlinear terms. First there is a broad Doppler pedestal given by the first order linear absorption term. Superimposed on this is a Lorentzian Lamb dip associated with the third order nonlinear terms which carry only single zone resolution in either the preparation or probing process or both. Note, then, that the low resolution, nonlinear field-atom interactions can occur in any one of the three zones, or any two, or even all three to produce our "single zone" Lamb dip (the low resolution term from interactions in all three zones is a factor of two narrower than the other terms). Finally, superimposed on this is the sharp Ramsey fringe pattern produced by the previously described interference experienced by those atoms which nonlinearly interact with all three zones. The sharp interference fringe results only if the nonlinear, atom-field interactions have high resolution in both the preparation and probing process. Bordé [2] has shown that the fringe pattern or oscillatory part of the signal for equal relaxation constants $\gamma_{ij} = \gamma_{ii} = \gamma$ is an exponentially damped cosine proportional to

$$(1/v^2)\exp\left[-(\omega-\omega_o')^2\, a^2/v_r^2\right]\cos\left[(\omega-\omega_o')(2v_xL/a^2-\gamma)a^2/v_r^2\right]$$

where a is the laser mode radius, L the common zone separation, $v^2 = (v_r^2 + v_z^2)$, $v_r^2 = v_x^2 + v_y^2$, and $\omega_o' = \omega_o(1 - v^2/2c^2 \pm \hbar k/2mc)$ to a good approximation. In the expression for ω_o', we have included not only the shift in the natural resonance frequency due to the recoil terms, $\pm \hbar k/2mc$, but also due to the second order Doppler term, $-v^2/2c^2$. In the limit that the second order Doppler frequency shift is much smaller than the detuning frequency, $\Delta\omega = \omega - \omega_o$, at which $\Delta\omega(v_xL/v_r^2) = \pi$, we can ignore this shift, and velocity averaging for a Maxwell-Boltzmann beam velocity distribution will then produce a strongly damped cosine pattern consisting of a primary peak at line center plus one or two smaller side peaks, similar to those described by Ramsey for linear RF excitation with separated oscillatory fields. We will return to a fuller discussion of this point later. If the radiation standing waves are composed of equal intensity counter propagating waves, variation of the relative cavity phases across the three zones produces a variation of fringe intensity (from positive through zero to negative) but no asymmetry, and hence, no shift in the fringe center, a particularly appealing characteristic for accurate measurements. If the cavity phase condition is nonstationary then the fringes will wash away with averaging time. The opposition of two cat's-eye retroreflectors to produce three or more radiation zones intrinsically holds the cavity phase condition constant [3,4]. This is due to the cat's-eye's high insensitivity to thermal and mechanical fluctuations and to small angle variations of the input laser beam. Finally, if properly focused, the cat's-eyes give fringes of maximum positive amplitude for all velocity classes. We are unaware of any other arrangement which automatically (i.e., without servoing) holds constant the initial phase condition.

There are important advantages of the separated oscillatory field method as compared to the cw, single zone interrogation method. Higher resolution can be obtained without loss of signal-to-noise ratio, whereas in the cw case, higher resolution is inversely proportional to S/N ratio (best case) because increasingly fewer atoms contribute to the increasingly narrower resonance. Note the immediate consequence to frequency standards applications, since $\sigma_T \propto 1/(Q \cdot S/N)$. Of course, this also directly impacts spectroscopy with the possibility of real time data acquisition as opposed to many hours of averaging. Secondly, the wavefront flatness is required only over the mode diameter 2a, rather than three zone interaction length of 2L. And finally, power broadening and shift are minimized.

Experimental Apparatus

The very high resolution possible in this experiment consequently demands a dye laser spectrometer of unprecedented frequency characteristics. Figure 1 shows a block diagram representation which highlights some of the important features of our Ca dye laser spectrometer as well as the Ca beam and interrogation method. Much of our fast-stabilized dye laser system has been previously described [13]. The dye laser is stabilized to the midpoint of a transmission fringe in an external optical Fabry-Perot cavity of extremely high finesse. The cavity length is controlled to provide long term stability and frequency tuning capability. Two important improvements have been made in the system [6]. Firstly, the short term linewidth of our laser has been reduced to approximately 800 Hz rms with an improved high frequency servo amplifier with gain to 5 MHz [14]. Secondly, we have reduced the long term drift to less than 2 kHz/hr by controlling the servo cavity length by means of a first derivative line center lock to the few MHz wide saturated absorption line in an external calcium cell. In the present setup the line center lock was obtained by directly frequency modulating the laser at 300 kHz. This modulation frequency is chosen so that the FM sidebands are within the cell saturated absorption linewidth, to give the first derivative signal, but largely outside the narrower beam saturated absorption line [15]. The modulation index is chosen to essentially only produce the first order sidebands at 300 kHz, leaving the majority of the power in the carrier. A dc scan voltage introduced at the integrator sweeps the laser frequency over the atomic beam line. The dc scan is calibrated using 3.39 μm He-Ne lasers by measuring the beat frequency between a local oscillator locked to the servo cavity, and a methane stabilized laser. Poor finesse of the cavity mirrors at the 3.39 microns presently limits the calibration accuracy to about 10 percent.

Adding sidebands to the laser adds complexity to the correct line analysis. First, three equally spaced laser frequencies (our carrier plus the two sidebands) produce five equally separated saturated absorption lines on the beam Doppler profile (provided the beam Doppler is sufficiently large). These five features may or may not overlap depending on the ratio of the inverse single zone transit time to the modulation frequency. In our case this ratio is approximately one, which results in only

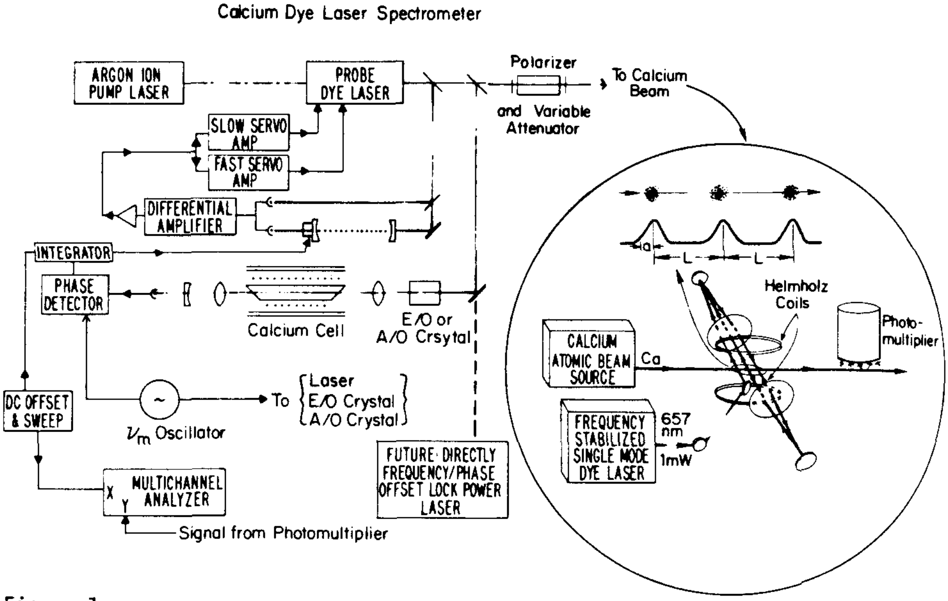

Figure 1

slightly overlapping lines. This presents a difficulty in correctly subtracting out the baseline. The second aspect is that there are three separate and distinct contributions to the central "resonant" saturated absorption line. There is the usual contribution from the $v_z \sim 0$ absorbers which are excited by the carrier and subsequently probed by the carrier. But there are two other class of $v_z \not\sim 0$ absorbers which also contribute to the central feature. One of these classes sees the blue shifted sideband as the pump and the red shifted one as the probe. In a symmetrical way, the oppositely signed v_z class sees the red shifted sideband as the pump and the blue shifted as the probe. Interestingly, these all contribute in a coherent way to the sharp Ramsey fringe structure for each recoil peak. Thus, there are now two possibilities to use the full beam Doppler to further enhance the signal-to-noise ratio while maintaining full resolution in the saturated absorption Ramsey fringe method. The first is to make the radiation zones narrow so that the acceptance angle $\sim \lambda/3a$ matches the beam Doppler (wavefront curvature and the corresponding confocal parameter are the limitations here); the second is to simply add sidebands to the laser to fully cover the beam Doppler [16].

We also show the possibility in Fig. 1 to either phase modulate with electro-optic crystal or to frequency modulate with an acousto-optic crystal, both of which are exterior to the laser. This allows the addition of sidebands to line center lock, but we now are able to choose whether or not to modulate that part of the laser light sent to the atomic beam. We have made preliminary investigations with both crystals. Perhaps the most elegant method [4] is to build a second low power reference laser which could be locked to one of the magnetically shifted, $\Delta m_J = 1$, ${}^1S_0$ - ${}^3P_1$ lines in the Ca absorption cell. The more powerful probe dye laser could be phase or frequency offset locked from the stable reference laser with a tunable RF oscillator which would provide a precise frequency scale. More generally, the reference laser could be stabilized to any of a number of possible atomic lines which are nearby the line(s) to be studied. For example, iodine nearly fills the entire visible spectrum with sharp transitions to which a laser could be locked. This would permit an extremely precise, stable, tunable dye laser spectrometer of general versatility.

In the circled inset in Fig. 1, we show a three dimensional representation of the atomic beam and interaction region. A Ca atomic beam from a resistively heated oven sequentially interacts with three equally spaced and parallel standing wave light beams from our stabilized dye laser. The atomic beam is collimated to give a Doppler beam width of approximately 1.7 MHz HWHM and has a density of about $10^8/cm^3$ at the laser excitation region. Typically, the laser power in the interaction region is on the order of 1 mW focused to a beam spot radius of approximately 0.15 cm. To form the three standing wave radiation fields, we use the duo cat's-eye reflector which was described above. The common spatial separation of the light fields is variable from 0 to ~ 3.5 cm. A transverse magnetic field of a few times 10^{-4} Tesla (a few Gauss) is superimposed on the interaction region which splits off the $m_j = \pm 1$ components. Furthermore, with light linearly polarized in the direction of the applied magnetic field, we excite only the $m_j = 0$ to $m_j = 0$ transition, which has no first order Zeeman effect. For signal detection we use a 5 cm diameter cathode photomultiplier located 20 cm downstream from the excitation region and 1 cm from the beam. With this arrangement we estimate that we collect about one percent of the total fluorescence photons. The signal is recorded on a multichannel analyzer with typical signal averaging times of a few minutes.

Results

The photon recoil doublet structure produces two partially overlapping fringe patterns as is shown in Fig. 2. The outer curve is the atomic beam fluorescence profile, with HWHM of 1.7 MHz, showing the single zone saturation dip with HWHM of 175 kHz. The two solid inner curves are the bottom of the single zone dip greatly expanded to show the observed Ramsey patterns for 2L = 3.5 and 7 cm, with the positions of the recoil components indicated by the vertical dashed lines. These curves were obtained by recording first derivative signals, with a narrowbanded S/N of about 10, and then integrating them with the multichannel analyzer to improve the apparent S/N. The separate fringe patterns for the two recoil peaks are shown as dashed curves above each experimental curve. The fringe intensity is a few percent of the total signal. The intensity ratio of the recoil peaks is approximately one, but the scatter in this ratio for our data is so large at present that no meaningful comparison can be made with the expected ratio of 0.998:1.0 [9].

Our measured value for the recoil splitting for a power density of about $10\ mW/cm^2$ is $\delta\nu = 23.6 \pm 2$ kHz, where the error of about 10 percent is mostly due to the poor absolute calibration of the dc frequency scan mentioned above. By holding this scan constant and obtaining relative measurements of the splitting versus power, we have obtained a possible indication of the contraction of the recoil splitting with power predicted by Bordé [2,10]. This is a light intensity shift of each recoil component toward line center caused by higher order coherent processes contributing

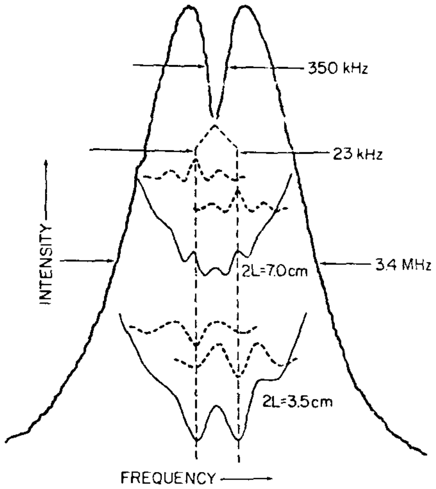

Figure 2 Observed fluorescence profiles. Outer curve: Atomic beam Doppler profile showing "single" zone saturation dip. Inner curve: Expansions of bottom of single zone dip (solid curves) showing Ramsey fringes for 2L = 3.5 and 7 cm. Fringe patterns for each photon recoil component shown with dashed curves. Note fringe sign inversion between the two resolution cases. See text for discussion.

extra intensity to the inner side of each line. Bordé calculates the shift to be, in first approximation, $\Delta = (\mu E/2h)^2/(f\cdot\delta v)$(Hz) for the case of continuous excitation and well resolved recoil peaks. In this expression μ is the electric dipole moment (for Ca, $\mu = 4.8 \times 10^{-20}$ esu-cm) and E is the electric field amplitude. For three-zone Ramsey excitation, this contraction should be reduced by a factor approximately equal to 2a/L, since the atom experiences the electric field for only the same fraction, 2a/L, of its time in the interaction region. With the electric field averaged over the Gaussian mode and diluted by 2a/L, the predicted contraction for 2L = 7 cm is $\Delta = -0.14$ kHz/(mW/cm^2). Experimentally, the recoil splitting versus power for 2L = 7 cm and power densities up to 17 mW/cm^2 shows a contraction of $\Delta = -0.2$ (± 0.2) kHz/(mW/cm^2). Although the experimental error at present is large, this result suggests agreement with Bordé's theory for continuous excitation attenuated in this way.

The power induced shifts for the two recoil peaks are equal, to first order, but opposite in sign, and thus the center of the line remains unshifted. It should be possible to line center lock to both peaks simultaneously, using modulation sidebands for instance, to give an average frequency for the centroid which is power independent to first order. Also, one might use identical circular polarizations to obtain the $m_J=0 \leftrightarrow m_J = \pm 1$ crossover resonance which has only one recoil peak, and hence no light-induced shift [2]. Thus, we do not believe this shift will be a serious problem for accurate frequency measurements.

The predominant limit to the accuracy of a frequency measurement for this line is the second order Doppler shift, 1.7 kHz for the most probable velocity. The second order Doppler shift has been an important limitation to all present frequency standards and requires careful velocity measurements to reduce this systematic offset. Similarly for calcium, a very careful determination of each velocity class contribution to the signal would be necessary to reduce this systematic offset below 10^{-14}.

It is an interesting feature, peculiar to the Ramsey interference method, that the line center position becomes highly sensitive to resolving power when the magnitude of the resolution, $\Delta v/v$, approaches that of the second order Doppler shift, $-\frac{1}{2}\, v^2/c^2$ [17]. This can be easily understood when one remembers that the resonant frequency of each recoil peak for a particular velocity class, v, is shifted by the second order Doppler term, $-\frac{1}{2}\, v^2/c^2$, but that the interfringe spacing is proportional to v/2L (actually, to $v_r/2L$, but $v_r \simeq v$ for saturation spectroscopy signals). We already have seen that velocity averaging washes out the fringe structure away from the recoil shifted line center, because each cosine, with period dependent on v, only contributes coherently at line center, but adds with random phase elsewhere, thus washing to zero in the wings. But, recall that this result obtains with fractional resolution much lower than the second order Doppler shift. When the resolution improves to where the interference fringe spacing begins to approach the same order of magnitude as the second order Doppler shift, then there is no frequency, or "line center", where all velocity contributions add coherently. Rather, there is an arbitrary frequency position, in general blue shifted from the recoil resonance frequency, which receives the majority coherent build up. This position depends critically on the interzone spacing and on the atomic beam velocity distribution. Contrast this to the single zone, cw excitation method where the second order Doppler shifted resonance is also highly sensitive to the velocity distribution but largeley independent of resolving power. However, even for sufficiently long lived systems, the single zone method has an ultimate resonance width limited by the quadratic Doppler broadening (for the multiple velocity case) independent of potentially better spectrometer resolving power, whereas the multiple zone interference method can yield artificially narrow resonances. Depending on the combination of velocity distribution and interzone spacing, it is possible to produce linewidths that are sub second order doppler and, perhaps, even subnatural in some cases.

Returning now to Fig. 2, note that the fringe pattern for each recoil peak in the high resolution case (2L = 7 cm) is inverted from that of the lower resolution case of 2L = 3.5 cm. We had earlier attributed this inversion to cavity phase shift between radiation zones. However, a velocity integration which includes the quadratic Doppler shift and no cavity phase shift gives inverted fringes for the 2L = 7 cm spacing. In fact, already at 3.5 cm there is a small asymmetry due to the quadratic effect.

It is clear that the task of determining the unshifted line center, or the accuracy and reproducibility has increased in complexity. But the method of optical Ramsey fringes applied to long lived atoms, such as Ca, is not hopeless, and there remain a number of potentially useful schemes. One could pulse the radiation field to select one, or at most a few, velocity classes in order to precisely determine the second order Doppler shift (also provides a means to determine the velocity distribution). Only those atoms with velocities, $v = nL/\tau$, where τ^{-1} is the pulse repetition rate and n is an integer, will interact with all radiation fields. However, this method is very expensive in signal-to-noise ratio since most atoms do not contribute to the signal. Additionally, the single velocity atomic beam signal is a slightly damped cosine function, which presents problems in determining and locking to line center.

Another alternative would be radiation pressure cooling of all velocity components with a broadband laser frequency red shifted from the natural resonance frequency of a suitable cooling line. For both Ca and Mg, the $^1S_0$ - $^1P_1$, line, at 4228 Å and 2853 Å, respectively, is a potential candidate. A possibly better cooling mechanism for Ca would be to simultaneously optically pump on the three $4^3P_{0,1,2}$ - 5^3S transitions near 600 nm. In either case, it should be possible to thermally cool Ca by a factor of 10^2 with ~ 5×10^4 scattering events in an approximately 1 cm interaction zone immediately outside the oven. The velocity would be reduced by a factor of 10 and the quadratic effect by a factor of 100. This would give a second order shift of only 17 Hz for the most probably velocity, permitting a clear resolution of the natural linewidth of 410 Hz. The three zone interaction region would follow the cooling region in a way such that the cooling process does not take place in the interaction region (a mirror with a hole in it would serve to bring in the longitudinal cooling beam). A common interzone spacing of 1.5 cm would fully resolve the recoil splitting and give a linewidth of 750 Hz HWHM. The fluorescence decay length is also reduced form approximately 32 cm to 3.2 cm, enhancing collection efficiency. As described in detail elsewhere, it would be possible to further improve the signal-to-noise ratio by photon amplification of the excited $^3P_1$ state to overcome detection and collection efficiencies [6]. The useful limit to S/N ratio could then be determined by the shot noise in atomic beam intensity. A conservative estimate of 10^4 for the interference fringe S/N ratio would give a pointing precision, or fractional frequency stability, of 3×10^{-16} at one second for our 750 Hz line. This precision should make it possible to study and correct systematic errors to better than 10^{-14} and result in a very high accuracy frequency standard in the visible spectrum.

In conclusion, we have for the first time obtained ultra high resolution with optical Ramsey interference fringes in a long lived atomic system. We have fully resolved the recoil splitting of the saturated absorption signal in the Ca, $^1S_0$ - $^3P_1$ intercombination line at 657 nm. We have introduced a locking scheme which greatly extends the useful stability of a dye laser spectrometer. Methods have been proposed which should permit a visible wavelength frequency standard, based on this Ca transition, which should exhibit a frequency stability of $3 \times 10^{-16} (\tau)^{-\frac{1}{2}}$ and a reproducibility exceeding 1×10^{-14}. This precision should allow improvements in experiments involving measurements of frequency offsets, such as gravitational red shift.

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