Resolution of photon-recoil structure of the 6573-Å calcium line in an atomic beam with optical Ramsey fringes

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The photon-recoil components of the Ca 6573-Å line have been resolved using the three-zone optical Ramsey interference technique with an atomic beam. Linewidths as narrow as 3 kHz HWHM (line Q of 8×10^{10}) are reported for zone separations up to 7 cm. An indication of the light-shift-induced contraction of the recoil splitting predicted by C.J. Bordé is obtained. Techniques are discussed which should lead to an optical wavelength/frequency standard with an accuracy of better than 10^{-14}.

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Unresolved photon-recoil components can limit the accuracy of high-resolution measurements in saturated absorption spectroscopy to the order of $10^{-12} - 10^{-13}$ in the visible spectrum. These components can now be resolved for atoms with long-lived transitions, such as ${}^1S_0$-${}^3P_1$ of calcium and magnesium, by using long-excitation-region atomic beams excited with highly stabilized dye lasers.[1-3] The necessary optical resolution can be obtained using saturated absorption with spatially separated optical fields[4-8] (three-zone optical Ramsey interference technique). We report here our investigations of the Ca intercombination resonance line ${}^1S_0$-${}^3P_1$(6573 Å), with which we have obtained linewidths as narrow as 3 kHz half-width–half-maximum intensity (HWHM), completely resolved the 23-kHz separation photon-recoil doublet, and obtained an indication of the contraction of the recoil splitting with laser power predicted by Bordé.[5] Also, we discuss techniques for this line which should lead to a measurement accuracy of better than 10^{-14}.

This Ca transition[2] is very suitable for measurements of the highest level of accuracy, such as those in the wavelength/frequency standards field and in various relativity experiments. It has a long lifetime of 0.39 msec (natural linewidth of 410 Hz) and small magnetic and electric field shifts of about 10^8 Hz/T^2 (1 Hz/G^2) (for the $m_j = 0$ to $m_j = 0$ component) and 1 Hz (V/cm)$^{-2}$, respectively, its transition probability is high enough that the optimum laser power is less than 1 mW, and complete resolution of the recoil peaks should lead to symmetric line profiles.

Kol'chenko *et al.*[9] and Bordé[10] have discussed the theory of photon recoil in saturated absorption. They predict this effect splits the saturation peak into a doublet with frequency separation.

$$\delta\nu = \hbar k^2/2\pi M \qquad (= 23 \text{ kHz for Ca 6573 Å}),$$

where k is the wave vector and M is the atomic mass. Also, Bordé predicts a light-shift-induced contraction of the separation, as discussed below. Until now, the only instance of photon-recoil resolution in the optical region has been that of Hall *et al.*[11] with the methane saturated absorption peaks at 3.39 μm.

Our fast-stabilized dye laser system has been previously described.[12] The frequency is stabilized to an external optical cavity, the length of which is controlled to provide long-term stability and frequency-tuning capability. Two improvements have been made in the system. First, use of an improved high-frequency servo amplifier[13] with gain to 5 MHz has reduced the short-term rms noise (5-MHz bandwidth) to about 800 Hz from the previous 5 kHz. Second, long-term drift has been reduced to less than 2 kHz/h through use of the following technique.[14] The servo cavity length is controlled by using a first-derivative line center lock to the saturated absorption peak (a few MHz wide) in a calcium absorption cell. A dc scan voltage introduced into the servo loop sweeps the frequency over the sharper atomic beam line. The 300-kHz modulation frequency is chosen so that the FM sidebands are within the cell linewidth to give the first-derivative signal but completely outside the much narrower beam saturated absorption linewidth. The dc scan is calibrated using 3.39-μm 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 3.39 μm presently limits the calibration accuracy to about 10%.

The atomic beam is collimated to give a Doppler beamwidth of about 1.7 MHz HWHM and has a density of about 10^8/cm^3 at the laser excitation region. The laser beam with about 1 mW power and a 0.5-cm spot diameter is incident normal to the atomic beam. We use the two opposing cat's eyes reflector technique[6] to form the three standing-wave zones with relative phases which are constant in time and determined by geometry and lens aberrations. A transverse magnetic field of a few times 10^{-4} T (a few Gauss) is superimposed on the interaction region so that with linearly polarized light we excite only the field-independent (to first order) $m_j = 0$ to $m_j = 0$ transition. For signal detection we use a 5-cm-diam cathode photomultiplier located 20 cm downstream from the excitation region and 1 cm from the beam. With this arrangement we collect approximately 10^{-2} of the total fluorescence photons. The signal is recorded on a

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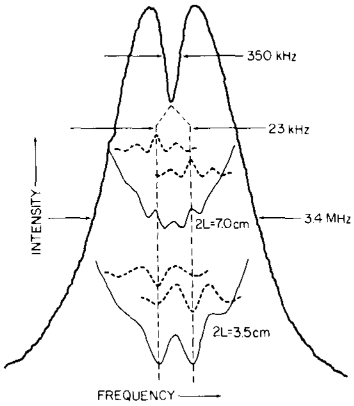

FIG. 1. Observed fluorescence profiles. Outer curve: atomic beam profile showing single-zone saturation dip. Inner curves: expansions of bottom of single-zone dip (solid curves) showing Ramsey fringes for $2L = 3.5$ and 7 cm, with separate fringe patterns (dashed curves) for each photon-recoil component.

multichannel analyzer with signal averaging times of a few minutes.

The Ramsey fringe technique has several advantages[6,7] over the case of continuous excitation throughout the interaction length. As this length is increased, higher resolution is obtained without serious loss in S/N. For equal-intensity counter-running components of the standing waves, variation of the relative 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 particular appealing characteristic for accurate measurements. Also, with this technique wavefront flatness is required only over the laser mode diameter $2w_0$ rather than the three-zone interaction length $2L$.

For detailed discussions of three-zone optical Ramsey interference, the reader is referred to the theoretical discussions of Baklanov *et al.*[4] and Bordé[5] and to the treatments of Bergquist *et al.*[6–8] For three standing-wave radiation zones, the line profile basically consists of a broad Lorentzian peak, corresponding to one-zone saturation, with a superimposed sharp fringe pattern produced by the light-wave phases experienced by the atoms as they go through the zones. Velocity averaging produces a damped cosine pattern consisting of a primary peak at line center, plus one or two smaller side peaks, similar to those described by Ramsey[15] for linear rf excitation with separated oscillating fields. For three-zone saturated absorption, Bordé[5] shows that the fringe profile, for equal zone seprations L and equal relaxation constants $\gamma = \gamma_a = \gamma_b$, is proportional to

$$\exp[-(\omega-\omega_0)^2 w_0^2/v_r^2]\cos[(\omega-\omega_0)(2v_x L/w_0^2-\gamma)w_0^2/v_r^2],$$

where w_0 is the laser mode radius and v is the atom velocity.

With two photon-recoil components, we observe two overlapping fringe patterns, as shown in Fig. 1. 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 are obtained by recording first-derivative signals, with 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 fringe sign reversal accompanying a relative phase change of π across the three radiation zones is clearly demonstrated in the two sets of curves. The intensity ratios of the recoil peaks are 0.6 for $2L = 3.5$ cm and 1 for $2L = 7$ cm; however, the scatter in this ratio for all our data is so large at present that no meaningful comparison can be made with the expected ratio.[9,10]

The fringe width and primary-secondary peak spacing are to first order proportional to $\alpha/2L$, where α is the most probable velocity (8×10^4 cm/sec for our calcium beam). The values of the proportionality constants for saturated absorption obtained from our experiment and from Bordé's theory[5] for a single velocity class (with relaxation constants $= 0.1 v_x/w_0$ and Rabi frequencies $\Omega^{\pm} = \mu E/2\hbar = (0.01/2\pi)v_x/w_0$), along with Ramsey's[15] values for optimum linear excitation, are given in Table I.

The experimental results fit the theoretical values reasonably well, especially since population decay between excitation and observation regions leads to preferential observation of velocities higher than α.

Our measured value for the recoil splitting is $\Delta\nu = 23.6 \pm 2$ kHz, where the error of about 10% 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 an indication of the contraction of the recoil splitting with power predicted by Bordé. This is a light shift of each recoil component toward line center caused by higher-order coherent processes contributing extra intensity to the inner side of each line. Bordé calculates the shift to be, in first approximation, $\Delta = (\mu E/2\hbar)^2/4\delta\nu$ (Hz) for continuous excitation. For three-zone excitation, since the atom experiences the electric field only a fraction $2w_0/L$ of the flight time, the shift should be reduced by a factor approximately equal to $2w_0/L$ ($=\frac{1}{7}$ in our case for $2L = 7$ cm). With the electric field averaged over the Gaussian mode and $\mu = 4.8\times10^{-20}$ esu cm, this predicted contraction for $2L = 7$ cm is $\Delta = -0.14$ kHz (mW/cm^2)$^{-1}$.

We have measured the recoil splitting versus power for $2L = 7$ cm and power densities up to 17 mW/cm^2 (Rabi frequency of about 38 kHz for our geometry) and find a contraction of $\Delta = -0.2\ (\pm 0.2)$ kHz (mW/cm^2)$^{-1}$. Al-

TABLE I. Proportionality constants relating HWHM and fringe spacing to $\alpha/2L$.

	Saturated absorption			Linear excitation
	Experiment		Bordé	Ramsey
HWHM	0.25	±0.03	0.29	0.325
Fringe spacing	1.1	±0.1	1.07	1.22

though the experimental error at present is quite large, this result shows that Bordé's theory, which assumes continuous excitation, predicts too large a shift for separated fields.

The shifts for the two peaks are equal, to first order, but opposite in sign and thus the centroid remains unshifted. Use of techniques such as, for instance, line-center locking to both peaks simultaneously using modulation sidebands should give an average frequency for the centroid which is power independent to first order. Furthermore, it may be possible to circumvent this problem with a technique proposed by Bordé, the use of circular polarization to obtain the $J = 0$ crossover resonance which should have only one recoil peak, and hence, no light-induced shift. Thus, we do not believe this shift will be a serious problem for accurate frequency measurements.

The predominant remaining limit on accuracy of frequency measurement is the second-order Doppler shift, 1.7 kHz for the most probable velocity. Use of various techniques [16–18] for velocity measurement should reduce this systematic offset to below 10^{-14}.

It is of interest to estimate the limit of pointing precision which could be obtained with this Ca line with optimized techniques. A large gain in signal should be obtained by using additional lasers to simultaneously optically pump [19] the three $4\ ^3P_{0,1,2}$-$5\ ^3S$ transitions over 1-cm of the beam downstream from the excitation region. Since the ^3S lifetime is about 10^{-8} sec, for high-enough pump power each $^3P_1$ atom entering the pump region would be repeatedly cycled between the two levels to give a "photon amplification" of about 10^3. Also, nearly all the resulting fluorescence would be emitted over the 1-cm length instead of over the present 30-cm $^3P_1$ decay length; this should make it possible to improve the fluorescence collecting efficiency by a factor of about 20. With these large gains in detection efficiency, the useful S/N ratio should be determined by the shot noise in the atomic beam intensity. Optimization of the atomic beam geometry should result in about 10^{12} atoms/sec with velocity within the acceptance angle for saturated absorption, giving a shot-noise-limited S/N of about 10^6. For a fringe height 1% of the signal and a lifetime-limited fringe width of 400 Hz, this would give a pointing precision of better than $\Delta\nu/\nu = 10^{-15}$ for 1-sec intergration.

This high pointing precision should make it possible to correct systematic errors to better than 10^{-14} and result in a very-high-accuracy frequency standard in the visible spectrum. The high precision should also make possible improvements in experiments involving measurements of frequency offsets, such as the gravitational red shift.

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