

# Measurement of isotope shifts and hyperfine splittings of ytterbium by means of acousto-optic modulation

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The isotope and hyperfine shifts for the Yb  $^1S_0(6s^2) \rightarrow ^3P_1(6s6p)$  transition were determined with an acousto-optic modulator used to frequency shift part of a laser beam. The frequency-shifted and -unshifted laser beams were superimposed and excited an atomic beam. The laser was scanned across the transition while fluorescence produced by the radiative decay of the excited state was detected by a photomultiplier. Each isotope generated two peaks in the spectrum separated by the acousto-optic shift, which permitted the frequency to be calibrated. This relatively simple method yields results that agree well with the most accurate existing data, which were obtained by measurement of frequency shifts with a Fabry-Perot étalon whose length was stabilized with a helium-neon laser locked to an iodine line.

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## 1. INTRODUCTION

Isotope shifts and hyperfine structure are commonly studied with lasers used to excite atomic beams.<sup>1</sup> The fluorescence produced from the radiative decay of the beam atoms is detected while the laser wavelength is scanned across the transition. The change in laser frequency is measured when part of the laser is passed through a Fabry-Perot étalon. A maximum in the laser transmission occurs whenever the frequency changes by an amount equal to the cavity's free spectral range  $c/2L$ , where  $c$  is the speed of light and  $L$  is the cavity's optical length. The present investigation uses an acousto-optic modulator to frequency shift a laser by a specified amount. The frequency-shifted and -unshifted laser beams are then superimposed and excite an atomic beam. Hence each isotopic transition gives rise to two peaks in the fluorescence spectrum that are separated by the acousto-optic frequency. This method permits a more accurate determination of frequency shifts than is possible with interferometers and has the additional advantage of using a much simpler apparatus. Acousto-optic modulators are used in many experiments such as laser cooling and precision measurements of Stark shifts.<sup>2-5</sup>

Ytterbium is important in a range of applications, including efforts to improve frequency standards and atomic traps.<sup>6-9</sup> It has naturally occurring isotopes with atomic mass units 168 (0.13%), 170 (3.05%), 171 (14.3%), 172 (21.9%), 173 (16.12%), 174 (31.8%), and 176 (12.7%). The nuclear spin for the even isotopes is zero, and isotopes 171 and 173 have spins of 1/2 and 5/2, respectively. Ytterbium's relatively low melting point of 819 K facilitates the generation of an atomic beam. Furthermore, it has several transitions at optical wavelengths that are accessible to cw lasers.<sup>10-12</sup>

The transition between the ground state  $^1S_0(4f^{14}6s^2)$  and the first excited state  $^3P_1(4f^{14}6s6p)$  at 555.6 nm was studied in several different experiments.<sup>1,13-15</sup> All of them determined isotope shifts and hyperfine structure with a Fabry-Perot étalon. For precise frequency cali-

bration the optical length of the interferometer must be accurately known. In addition, the cavity length should be stabilized to prevent changes in the free spectral range caused by fluctuations in temperature or air pressure. In this paper we discuss the simpler method of using an acousto-optic modulator in Section 2. The results are compared with previous measurements in Section 3.

## 2. EXPERIMENTAL PROCEDURE

The apparatus is illustrated in Fig. 1. An atomic beam was used rather than a cell to eliminate collisions that can broaden transitions, obscuring individual lines of the various isotopes. The atomic beam was generated with an oven that was heated to  $\sim 700$  K. The atoms were collimated by a series of slits, producing a beam with a divergence of  $\sim 2$  mrad. The apparatus was enclosed in a vacuum chamber that was pumped by a diffusion pump to a pressure of  $2 \times 10^{-7}$  Torr.

The laser light was supplied by a ring dye laser (Coherent 699), which was pumped by 6 W of 514-nm light supplied by an argon-ion laser. The manufacturer-quoted linewidth of the dye laser was 0.5 MHz. The laser readily supplied several hundred milliwatts of light with Rhodamine 110 used as the laser dye. Part of the dye laser beam passed through an acousto-optic modulator. The latter consisted of a tellurium oxide crystal that has a diffraction efficiency higher than 50% for shifting light at 590 nm by 220–320 MHz. The modulation frequency  $\nu_{AO}$  was supplied by a frequency synthesizer with an accuracy of three parts in  $10^6$ . This signal was then amplified to a power of 1 W. The frequency-shifted laser beams were spatially deflected a few milliradians, and the trajectory of the unshifted beam at frequency  $\nu$  was unaffected. The modulator position was adjusted to maximize the intensity of the diffracted laser beam at the frequency  $\nu - \nu_{AO}$ .

Two 0.1-cm-wide slits located 160 cm apart, which were positioned on either side of the vacuum chamber, were used to superimpose the shifted and unshifted laser

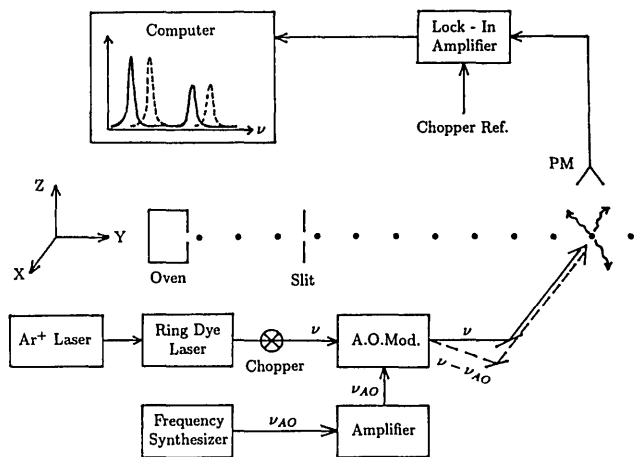


Fig. 1. Apparatus. See text for a detailed discussion. A.O. Mod., acousto-optic modulator.

beams. Each laser beam was centered on the two slits to within half a slit width. Hence the collimation of the two beams was the same to better than 0.3 mrad. The lasers then orthogonally intersected the atomic beam, eliminating first-order Doppler shifts.

Fluorescence produced by the radiative decay of the excited state back to the ground state was detected by a photomultiplier. The region in which the laser and the atomic beams intersected was carefully shielded to eliminate scattered light. The signal-to-noise ratio of the measured fluorescent light was in excess of 1000. The photomultiplier signal was sent to a lock-in amplifier whose reference signal was provided by a chopper that modulated the laser beam at a frequency of 1 kHz. The lock-in amplifier digitized the demodulated signal at a rate of 128 Hz, while the laser was scanned across the resonance at a speed of  $\sim 50$  MHz/s. Each scan of 7 GHz, as is shown in Fig. 2(a), therefore lasted  $\sim 2.5$  min. The data were then stored on disk and transferred to a computer for analysis.

Figure 2 shows typical data taken with an acousto-optic modulation frequency of 300.000 MHz. Eight peaks were readily resolved by both the frequency-shifted and -unshifted laser beams. The peak that is due to isotope 168, shown in Fig. 2(c), was observed only for the unshifted laser, which had a higher power than the frequency-shifted laser beam. All lines were clearly resolved, as is emphasized in Fig. 2(b), which shows the transitions for isotopes 170 and 173, which have the smallest frequency separation. We obtained the line centers by simply finding the locations of the intensity maxima. A more complicated fit of the spectrum to a summation of Lorentzian line shapes was unnecessary, since neighboring lines had insufficient overlap. For the data shown in Fig. 2(b) the line centers determined from a Lorentzian fit were within 0.5 MHz of the positions of the peak maxima.

The data shown in Fig. 2 were taken with neutral-density filters used to attenuate the laser beams to a power of  $\sim 1$  mW. This reduced the power broadening, giving a linewidth (FWHM) of  $\sim 7$  MHz. Data were taken with the unshifted laser beam power varied by a factor of 10 while the power of the shifted laser beam was held constant. No difference in results for isotope pre-

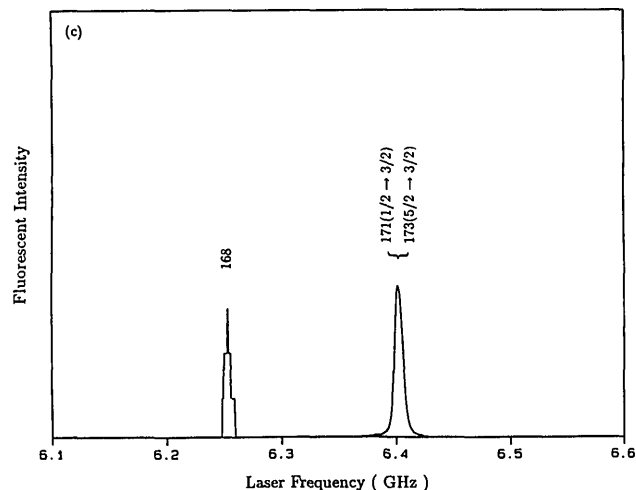
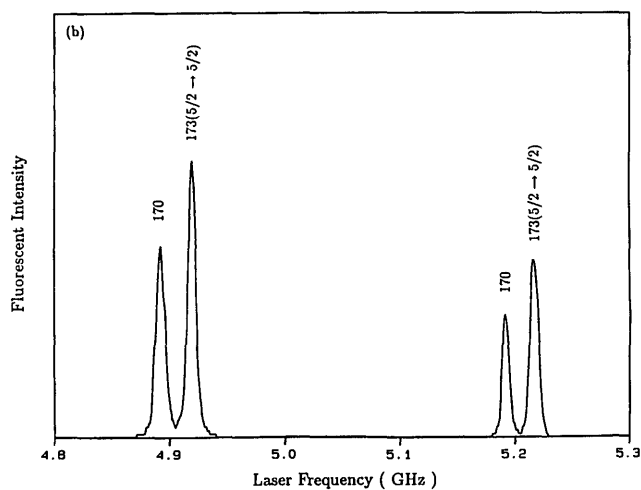
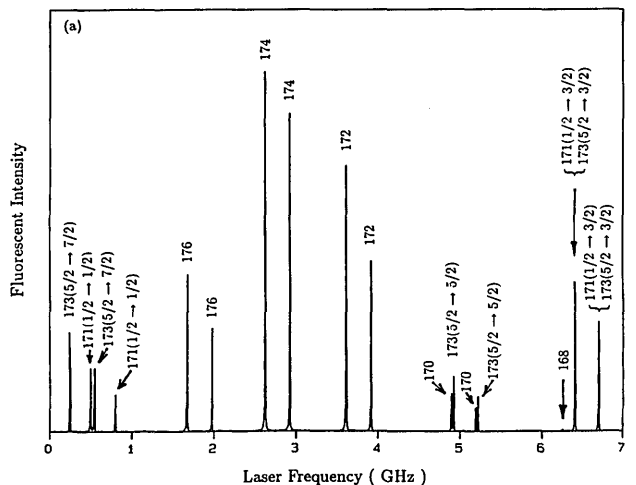


Fig. 2. Frequency scan of laser across the  $\text{Yb } ^1S_0(6s^2) \rightarrow ^3P_1(6s6p)$  transition. (a) 7-GHz scan with the frequency-shifted and -unshifted lasers used to excite an atomic beam. Each isotope or hyperfine peak was resolved, as is shown (b), where the vertical scale has been changed by a factor of 5. The transition for  $^{168}\text{Yb}$  was excited by the unshifted laser, as is shown in (c), where the vertical scale has been magnified by a factor of 60. The vertical scale used for the peak that is due to  $^{171}\text{Yb}$  and  $^{173}\text{Yb}$  in (c) is the same as in (a).

**Table 1. Isotope Shifts and Hyperfine Splittings of the 5556.5-Å Transition of Yb I**

Spectral Component	Position Relative to $^{176}\text{Yb}$ (MHz)				
	This Work	Ref. 1	Ref. 15	Ref. 14	Ref. 13
173(5/2 $\rightarrow$ 7/2)	-1432.6(12)	-1431.73(50)	-1432.5(43)	-1460(6)	-1451(6)
171(1/2 $\rightarrow$ 1/2)	-1177.3(11)	-1177.23(50)	-1176.6(35)	-1187(3)	-1184(6)
176	0	0	0	0	0
174	954.2(9)	954.76(50)	953.0(13)	947(2)	947(2)
172	1954.8(16)	1955.04(50)	1954.0(26)	1940(9)	1940(9)
170	3241.5(28)	3241.51(50)	3239.6(40)	3229(18)	3232(6)
173(5/2 $\rightarrow$ 5/2)	3267.1(28)	3266.49(50)	3264.3(98)	3247(9)	3253(6)
168	4611.9(44)	4610.14(50)	4609.5(54)	4581(15)	
171(1/2 $\rightarrow$ 3/2)	4761.8(37)	4759.83(50)	4759.5(143)	4761(9)	4761(9)
173(5/2 $\rightarrow$ 3/2)	4761.8(37)	4760.42(50)	4759.5(143)	4749(12)	4700(6)

viously shifts obtained using the frequency-shifted and -unshifted lasers was observed.

The calibration of the laser frequency was accomplished as follows. Each transition was excited by the frequency-shifted and -unshifted lasers, producing two fluorescent peaks. The scan time between these two peaks corresponds to the acousto-optic frequency if the two laser beams are perfectly superimposed and the laser has negligible frequency jitter. This occurs even if the intersection angle between the laser and the atomic beams is not exactly 90°, since the signals produced by the two lasers will be equally Doppler shifted. In our experiment the two laser paths overlapped to within 0.3 mrad, as was discussed above. No change in the direction of either laser beam was detected during the wavelength scan. Hence, for a Doppler width of ~500 MHz, the maximum difference in residual first-order Doppler shifts is 0.15 MHz. This represents one part in 2000 when compared with the acousto-optic frequency of 300 MHz. An improved frequency calibration with a higher acousto-optic frequency was not possible because of the 300-MHz bandwidth of the amplifier.

The spectra in Fig. 2(a) show eight pairs of peaks arising from transitions that were excited by both frequency-shifted and -unshifted lasers. The number of data points separating each pair of peaks was found. The eight pair separations showed random differences that were due to the frequency jitter of the laser but gave no indications that the scan was nonlinear. The frequency interval represented by a single data point was therefore found by division of the acousto-optic frequency by the average of the eight pair separations. Each scan was separately calibrated, since the time for the laser to be tuned through 7 GHz varied randomly from scan to scan by as much as 0.1%. The isotope and the hyperfine splittings were obtained by measuring the various line centers relative to the line generated by  $^{176}\text{Yb}$ . Table 1 gives the averaged results of more than 40 wavelength scans along with the statistical uncertainties, which are enclosed in parentheses.

### 3. RESULTS AND CONCLUSIONS

Our results agree well with those obtained by four other groups, which are cited in Table 1. The experiments of Chaiko<sup>13</sup> and Miller and Ross<sup>14</sup> both used liquid-nitrogen-cooled hollow-cathode discharges to excite enriched samples of ytterbium. Chaiko then measured the spec-

trum of emitted light with a Fabry-Perot interferometer, which he scanned by varying the air pressure between the two mirrors. Miller and Ross examined the fluorescence with a Fabry-Perot interferometer in conjunction with a spectrograph. The lines observed in these two experiments were thermally broadened, and their accuracy was therefore considerably less than that achieved in later experiments that used laser excitation of atomic beams.

Both the groups of Clark *et al.*<sup>1</sup> and Jin *et al.*<sup>15</sup> used a laser with a linewidth of ~5 MHz to excite an atomic beam. The laser frequency was monitored with a confocal Fabry-Perot cavity having a free spectral range of close to 300 MHz. Jin *et al.* claimed an accuracy of 0.1–0.2% for the even isotope shifts and 0.3–0.4% for the hyperfine structure of the odd isotopes. Our table lists the uncertainties found with their lower error estimates. Clark *et al.* actively stabilized the length of their étalon with a helium-neon laser that was locked to an iodine reference line. The Fabry-Perot cavity was housed in an evacuated chamber to greatly reduce pressure- and temperature-dependent fluctuations. These effects can alter the index of refraction and hence change the cavity's free spectral range. These refinements permitted the achievement of an accuracy of 0.5 MHz for the various isotopic shifts and hyperfine splittings. Clark *et al.* also resolved the  $F = 1/2 \rightarrow 3/2$   $^{171}\text{Yb}$  and  $F = 5/2 \rightarrow 3/2$   $^{173}\text{Yb}$  transitions by using a magnetic field that affected the two isotopes differently. The line centers were measured as a function of the magnetic field, and the data were extrapolated to zero field to find the peak positions.

A number of ways exist to improve the accuracy of the present experiment. A dye laser with less frequency jitter and less variation in the time required for each laser scan would produce more accurate results. The calibration of the laser frequency can also be improved. As was mentioned, a higher acousto-optic frequency would decrease the relative importance of any different residual Doppler shifts of the fluorescent peaks resulting from imperfect overlap of the frequency-shifted and -unshifted laser beams. A second possibility is that passing the two laser beams through a single-mode optical fiber could improve their collimation. This approach was used by Cable *et al.*<sup>16</sup> in their study of sodium atoms in a magneto-optical trap. A third approach would be to reduce the Doppler width substantially by use of well-developed laser cooling and trapping methods.<sup>17</sup>

A different method for determining isotope shifts and hyperfine structure is to use electro-optic modula-

tion of a laser beam. This generates frequency sidebands that are not spatially deflected as in the case of acousto-optic modulation and would therefore improve the frequency calibration. Unfortunately an electro-optic modulator was not available for this experiment. This method was used by Deilamian *et al.*<sup>11</sup> to study the Yb  $^1S_0(6s^2) \rightarrow ^1P_1(6s6p)$  transition at 398.8 nm. Electro-optic modulation frequencies can be higher than acousto-optic modulation frequencies and would be especially useful for measuring large frequency shifts as occur in fine structure splittings.

In conclusion, the isotope and hyperfine shifts of the Yb  $^1S_0(6s^2) \rightarrow ^3P_1(6s6p)$  transition found with acousto-optic modulation agree with the most accurate results determined with a Fabry-Perot étalon used for frequency calibration. The apparatus is relatively simple, and frequency shifts can be precisely and easily specified. This method is especially useful for calibrating small frequency shifts for which interferometers would have an inordinately long length. This would permit precise measurement of linewidths and facilitate the study of broadening mechanisms that are due to collisions and power broadening as well as the determination of radiative lifetimes.<sup>18</sup> Hence, acousto-optic and electro-optic modulators are likely to play an increasingly important role in precision spectroscopy.

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## REFERENCES

1. D. L. Clark, M. E. Cage, D. A. Lewis, and G. A. Greenlees, "Optical isotopic shifts and hyperfine splittings for Yb," *Phys. Rev. A* **20**, 239–253 (1979).
2. L. S. Goldner, C. Gerz, R. J. C. Spreeuw, S. L. Rolston, C. I. Westbrook, and W. D. Phillips, "Momentum transfer in laser-cooled cesium by adiabatic passage in a light field," *Phys. Rev. Lett.* **72**, 997–1000 (1994).
3. C. E. Tanner and C. Wieman, "Precision measurement of the Stark shift in the  $6S_{1/2} \rightarrow 6P_{3/2}$  cesium transition using a frequency-stabilized laser diode," *Phys. Rev. A* **38**, 162–165 (1988).
4. W. A. van Wijngaarden, E. A. Hessels, J. Li, and N. E. Rotbery, "Precision measurement of Stark shifts for  $6P_{3/2} \rightarrow nS_{1/2n} = 10-13$  transitions in cesium," *Phys. Rev. A* **49**, R2220–R2223 (1994).
5. M. Kasevich and S. Chu, "Measurement of the gravitational acceleration of an atom with a light-pulse atom interferometer," *Appl. Phys. B* **54**, 321–332 (1992).
6. R. Grimm and J. Mlynek, "Observation of light-pressure-induced dispersion in Yb vapor," *Phys. Rev. Lett.* **61**, 2308–2311 (1988).
7. G. D. Sprouse, J. Das, T. Lauritsen, J. Schecker, A. Berger, J. Billowes, C. H. Holbrow, H. E. Mahnke, and S. E. Rolston, "Laser spectroscopy of light Yb isotopes on-line in a cooled gas cell," *Phys. Rev. Lett.* **63**, 1463–1466 (1989).
8. D. J. Seidel, A. Williams, R. W. Berends, and L. Maleki, "The development of a ytterbium ion frequency standard," in *Proceedings of the IEEE Frequency Control Symposium* (Institute of Electric and Electronics Engineers, New York, 1992), p. 70–75.
9. A. S. Bell, H. A. Klein, A. P. Levick, C. Tamm, and D. Schnier, "Laser cooling of trapped ytterbium ions using a four-level-excitation scheme," *Phys. Rev. A* **44**, R20–R23 (1991).
10. R. W. Berends and L. Maleki, "Hyperfine structure and isotope shifts of transitions in neutral and singly ionized ytterbium," *J. Opt. Soc. Am. B* **9**, 332–338 (1992).
11. K. Deilamian, J. D. Gillaspay, and D. E. Kelleher, "Isotope shifts and hyperfine splittings of the 398.8-nm Yb I line," *J. Opt. Soc. Am. B* **10**, 789–793 (1993).
12. G. D. Alkhazov, A. E. Barzakh, V. P. Denisov, K. A. Mezilev, Yu. N. Novikov, V. N. Panteleev, A. V. Popov, E. P. Sudentas, V. S. Letokhov, V. I. Mishin, V. N. Fedoseev, S. V. Andreev, D. S. Vedeneev, and A. D. Zyuzikov, "Measurement of isotopic shifts and hyperfine structure of neutron-deficient Yb isotopes using a new highly sensitive method of atomic spectroscopy," *Bull. Russ. Acad. Sci.* **56**, 1712–1716 (1992).
13. Y. Chaiko, "Isotope shifts of resonance lines in the spectrum of ytterbium," *Opt. Spectrosc.* **20**, 424–428 (1970).
14. G. E. Miller and J. Ross, "Isotope shifts in the arc spectra of dysprosium, erbium and ytterbium," *J. Opt. Soc. Am.* **66**, 585–589 (1976).
15. W. G. Jin, T. Horiguchi, M. Wakasugi, T. Hasegawa, and W. Yang, "Systematic study of isotopic shifts and hyperfine structures in Yb I by atomic-beam laser spectroscopy," *J. Phys. Soc. Jpn.* **60**, 2896–2906 (1991).
16. A. Cable, M. Prentiss, and N. P. Bigelow, "Observations of sodium atoms in a magnetic molasses trap loaded by a continuous uncooled source," *Opt. Lett.* **15**, 507–509 (1990).
17. M. Kasevich, K. Moler, E. Rijs, E. Sunderman, D. Weiss, and S. Chu, "Applications of laser cooling and trapping," presented at the Twelfth International Conference on Atomic Physics, Ann Arbor, Mich., 1990.
18. W. A. van Wijngaarden and J. Li, "Use of acousto-optic modulators for precision spectroscopy," presented at the Conference of the Canadian Association of Physicists, Regina, Saskatchewan, Canada, 1994.