

Precise measurements of UV atomic lines: Hyperfine structure and isotope shifts in the 398.8 nm line of Yb

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Abstract. – We demonstrate a technique for frequency measurements of UV transitions with sub-MHz precision. The frequency is measured using a ring-cavity resonator whose length is calibrated against a reference laser locked to the D_2 line of ^{87}Rb . We have used this to measure the 398.8 nm $^1S_0 \leftrightarrow ^1P_1$ line of atomic Yb. We report isotope shifts of all the seven stable isotopes, including the rarest isotope ^{168}Yb . We have been able to resolve the overlapping ^{173}Yb ($F = 3/2$) and ^{172}Yb transitions for the first time. We also obtain high-precision measurements of the excited-state hyperfine structure in the odd isotopes, ^{171}Yb and ^{173}Yb . The measurements resolve several discrepancies among earlier measurements.

Precise measurements of the frequencies of atomic transitions are an important tool in expanding our knowledge of physics. For example, precise measurement of the D_1 line in Cs [1] combined with an atom-interferometric measurement of the photon recoil shift [2] could lead to a more accurate determination of the fine-structure constant α . In addition, hyperfine-structure and isotope-shift measurements in atomic lines can help in fine-tuning the atomic wave function, particularly due to contributions from nuclear interactions. This is important when comparing theoretical calculations with experimental data in atomic studies of parity violation [3]. The most precise optical frequency measurements to date have been done using the recently developed frequency-comb method with mode-locked lasers [1], with errors below 100 kHz being reported. However, to the best of our knowledge, this technique has not yet been applied to UV spectroscopy, which relies on older and less-accurate techniques.

In this letter, we present the most comprehensive study of the 398.8 nm $^1S_0 \leftrightarrow ^1P_1$ line of atomic Yb. Yb ($Z = 70$) is an attractive candidate for studying atomic parity violation [4] and the search for a permanent electric-dipole moment in atoms [5]. Laser-cooled Yb has also been proposed for frequency-standards applications [6]. The $^1S_0 \leftrightarrow ^1P_1$ line is widely used in laser-cooling experiments [5, 7]. Over the years, there has been much interest in this line, and its isotopic and hyperfine components have been measured using a variety of techniques —level-crossing and anti-crossing spectroscopy [8–10], Fabry-Perot cavity [11], saturated-absorption spectroscopy [12], photon-burst spectroscopy [13], and most recently

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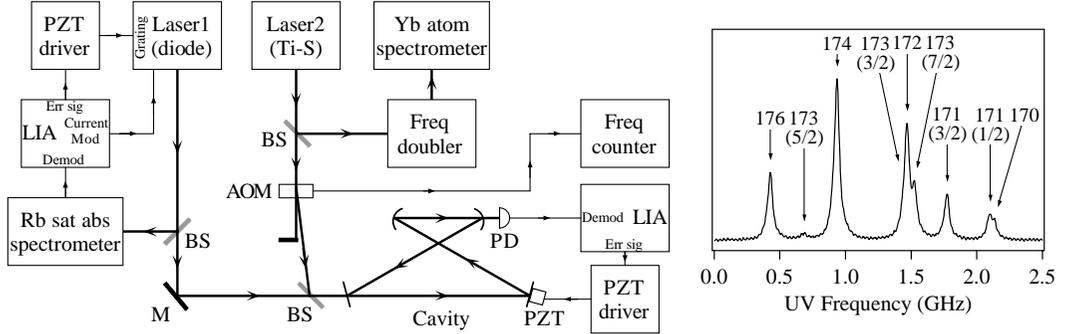


Fig. 1 – Schematic of the experiment. The trace on the right is the fluorescence signal showing the different Yb transitions. Two peaks contain overlapping transitions and need to be separated by multi-peak fitting.

using optical double-resonance spectroscopy with cold atoms in a magneto-optic trap [7]. However, all these measurements have errors of several MHz and show wide discrepancies with each other. We report the first sub-MHz measurement of the isotope shifts of all seven stable isotopes, including the rarest isotope ^{168}Yb (natural abundance = 0.13%). We have been able to resolve the overlapping ^{173}Yb ($F = 3/2$) and ^{172}Yb transitions for the first time. We also report high-precision measurements of excited-state hyperfine structure in the odd isotopes, ^{171}Yb and ^{173}Yb .

Our novel technique for the frequency measurement uses the fact that the *absolute* frequency of the D_2 line ($5S_{1/2} \leftrightarrow 5P_{3/2}$ transition) in ^{87}Rb has been measured with an accuracy of 10 kHz [14]. A stabilized diode laser locked to this line is used as a frequency reference along with a ring-cavity resonator whose length is locked to the reference laser. For a given cavity length, an unknown laser on an atomic transition has a small frequency offset from the nearest cavity resonance. This offset is combined with the cavity mode number to obtain a precise value for the *absolute* frequency of the unknown laser. We have earlier used this technique to make measurements of hyperfine intervals in the D_2 line of ^{85}Rb with 30 kHz precision [15], where we have also highlighted the advantages of the Rb-stabilized ring cavity. In this work, we apply the technique for UV measurements using a frequency-doubled IR laser to access the UV lines. By measuring the frequency of the IR laser and not the UV laser, we avoid several complications associated with UV spectroscopy. The technique is uniquely suited for measuring hyperfine intervals and isotope shifts since several sources of systematic error cancel in such measurements.

The schematic of the experiment is shown in fig. 1. Laser1 is a frequency-stabilized diode laser that acts as the frequency reference for the cavity. It is locked to the D_2 line of ^{87}Rb using saturated-absorption spectroscopy in a vapor cell. Laser2 is a tunable Ti-sapphire laser (Coherent 899-21) whose output is fed into an external frequency doubler (Laser Analytical Systems LAS100) to access the 398.8 nm Yb line. The doubler uses a patented Δ -cavity design that allows it to track large-frequency scans of the Ti-S laser (up to 30 GHz) without mode hops. The Ti-S laser is frequency-stabilized to a linewidth of 500 kHz. A part of its output is tapped off before the doubler and coupled into the ring-cavity resonator for the frequency measurement. Yb spectroscopy is done inside a vacuum chamber maintained at a pressure below 10^{-9} torr. The Yb atomic beam is produced by heating a quartz ampoule containing metallic Yb to a temperature of 400°C. To reduce Doppler broadening, the laser beam intersects the atomic beam at right angles and the fluorescence from the atoms is detected

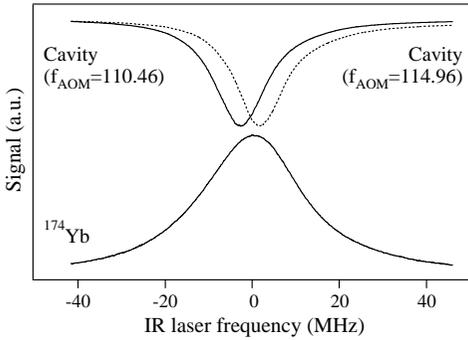


Fig. 2

Fig. 2 – Determination of the frequency offset. The lower trace is the fluorescence signal as the laser is scanned across the ^{174}Yb transition. The upper traces are the cavity-reflection signal for two values of the AOM frequency: 110.46 MHz for the solid line and 114.96 MHz for the dotted line. A fit to the separation between the fluorescence and cavity peaks *vs.* the AOM frequency yields the exact frequency that brings the cavity on resonance, which in this case is 112.64 MHz.

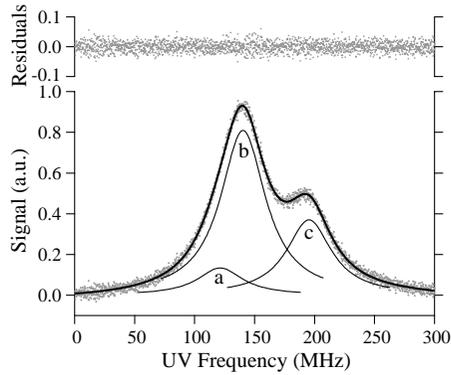


Fig. 3

Fig. 3 – Three-peak fitting to extract overlapping transitions. The fluorescence signal (shown as gray circles) is fitted to three Lorentzians with the same linewidths. The three peaks correspond to ^{173}Yb ($F = 3/2$) labeled as a, ^{172}Yb labeled as b, and ^{173}Yb ($F = 7/2$) labeled as c. The thick line is the sum of the three peaks and fits the measured data very well as seen from the structureless residuals shown on top.

through a narrow slit. A typical Yb spectrum is shown in fig. 1b. With 20 mW of UV light in a beam of diameter 1 cm, we achieve a linewidth of 45 MHz, or about 50% larger than the natural linewidth of 28 MHz.

The frequency measurement proceeds as follows. The outputs of Laser1 (reference) and Laser2 (Ti-S) are fed into the ring cavity. The cavity length is adjusted using a piezo-mounted mirror to bring it into resonance with the wavelength of Laser1. The cavity is then locked to this length in a feedback loop. However, Laser2 will still be offset from the cavity resonance. This offset is accounted for by shifting the frequency of the laser using an acousto-optic modulator (AOM) before it enters the cavity. The signal from a scan of Laser2 is shown in fig. 2. The upper trace shows the reflected signal from the cavity, which goes to a minimum as the cavity comes into resonance. The lower trace is the fluorescence signal corresponding to the ^{174}Yb transition. The cavity resonance slightly to the left of the ^{174}Yb peak (solid line) is for an AOM frequency of 110.46 MHz, while the one to the right (dotted line) is for a frequency of 114.96 MHz. Thus, by changing the AOM frequency, we can move the cavity resonance across the ^{174}Yb peak. The peak centers are determined to an accuracy of 50 kHz by fitting a Lorentzian lineshape, and we verify that there is no significant deviation from this lineshape from the featureless residuals. A straight-line fit to the peak separation *vs.* the AOM frequency gives the AOM frequency that brings the cavity into resonance (with a typical accuracy of 100 kHz). Once the exact cavity length (or mode number) is known, the absolute frequency of Laser1 is used to determine the absolute frequency of Laser2.

The measurements rely on the fact that the cavity mode number is known exactly. For this, we measure the cavity free-spectral range (fsr) very precisely, in the following manner. We first lock the cavity with the reference laser on the $F = 2 \rightarrow F' = (2, 3)$ transition in

^{87}Rb and measure the AOM offset for the ^{174}Yb transition. We then shift the reference laser to the $F = 1 \rightarrow F' = (1, 2)$ transition, which is exactly 6622.887 MHz higher [14, 16]. This shift causes the cavity mode number to increase by almost exactly 5, since the fsr is about 1326 MHz. The cavity is locked to the new frequency and the AOM offset for the same ^{174}Yb transition is measured. The difference between the two AOM offsets along with the change in the reference frequency gives exactly 5 times the cavity fsr. Using this method, we determine the fsr with a precision of 30 kHz. To determine the mode number, we measure the frequency of the ^{174}Yb transition to an accuracy of 20 MHz using a home-built wavemeter [17]. Thus, there is a *unique* mode number that matches the cavity resonance condition and the measured fsr. Indeed, the next-nearest mode that satisfies the resonance condition has an fsr differing by 625 kHz, or about 20 times the error in the determination of the fsr. Similarly, a change in the mode number by one causes the frequency of the ^{174}Yb transition to change by 60 MHz, or about 3 times the error with which the frequency is known.

The Yb spectrum has two peaks where there is significant overlap between neighboring transitions. One of these consists of the ^{171}Yb ($F = 1/2$) and the ^{170}Yb transitions. This does not present any problem in our technique because the two transitions are separated by about 40 MHz and the spectrum shows two clear maxima. A two-Lorentzian fitting algorithm uniquely extracts the locations of the two transitions. However, the other multiple peak consists of the ^{173}Yb ($F = 3/2$), ^{172}Yb , and ^{173}Yb ($F = 7/2$) transitions, all lying within 50 MHz of each other. As seen from fig. 3, the first two transitions have a separation less than the natural linewidth and are completely merged. When we fit a single Lorentzian to these two overlapping transitions, we find its linewidth to be 1.4 times larger than the linewidth of the neighboring ^{173}Yb ($F = 7/2$) transition, and indeed the linewidth of all the other peaks in the spectrum. This gives us confidence that the increased linewidth is a result of the convolution of two individual Lorentzians. Therefore, we fit two Lorentzians to the overlapping peaks, and a third Lorentzian to the neighboring peak, with the constraint that the linewidth of all three peaks is the same. The algorithm then returns three peaks having linewidths similar to what we obtain for other well-resolved peaks. As seen from fig. 3, the fit residuals are very small and their structureless noise shows that there is no ambiguity in the fitting. After fitting to about 30 spectra, we obtain an average value of 17.64 ± 0.90 MHz for the separation between the ^{173}Yb ($F = 3/2$) and the ^{172}Yb line centers. We believe this is the first direct measurement of this separation.

The first source of systematic error we consider arises due to improper perpendicular alignment of the laser beam with the Yb atomic beam. A misalignment angle of 10 mrad can cause a Doppler shift of about 7 MHz. To minimize this, we have repeated each measurement with a counter-propagating laser beam. Since the shift in this case is of opposite sign, the error cancels when we take an average of the two values. Indeed, the difference between the two values gives an estimate of the misalignment angle, which in our case is less than 2 mrad. In any case, the error cancels in the determination of the isotope shifts (relative to ^{174}Yb) since all isotopes experience the same shift. Of course, there is a small differential Doppler shift due to the fact that the different isotopes leave the oven with slightly different velocities. But even for a large misalignment angle of 10 mrad the differential shift is only 120 kHz, which is negligible at our level of precision.

There are two classes of systematic error inherent to our technique. The first depends on variations of the reference laser. The possible causes are shifts in the laser lock point due to residual Doppler profile or optical-pumping effects in the Rb saturated-absorption spectrometer which change the lineshape of the peaks. We have tried to minimize this by using third-harmonic locking and careful control of pump and probe intensities in the spectrometer. Collisional shifts in the Rb vapor cell are estimated to be less than 10 kHz. The magnetic

TABLE I – Listed are the various transitions of the 398.8 nm $^1S_0 \leftrightarrow ^1P_1$ line in Yb. The shifts from the ^{174}Yb transition measured in this work are compared to values reported in earlier work.

Isotope	Shift from ^{174}Yb (MHz)				
	This work	Ref. [7]	Ref. [13]	Ref. [11]	Ref. [19]
^{176}Yb	-509.98 ± 0.75	-507.2 ± 2.5		-509.4 ± 4.0	-469.2 ± 2.7
^{173}Yb ($F = 5/2$)	-254.67 ± 0.63				
^{173}Yb ($F = 3/2$)	516.26 ± 0.90				
^{172}Yb	533.90 ± 0.70	527.8 ± 2.8		529.9 ± 4.0	530.20 ± 7.80
^{173}Yb ($F = 7/2$)	589.00 ± 0.45	578.1 ± 5.8			
^{171}Yb ($F = 3/2$)	833.24 ± 0.75	832.5 ± 5.6	834.4 ± 4.0		
^{171}Yb ($F = 1/2$)	1152.86 ± 0.60	1151.4 ± 5.6	1136.2 ± 5.8		
^{170}Yb	1192.48 ± 0.90	1175.7 ± 8.1	1172.5 ± 5.7	1195.0 ± 10.8	1158.9 ± 11.4
^{168}Yb	1886.57 ± 1.00		1870.2 ± 5.2		
^{173}Yb (centroid)	291.61 ± 0.35			291.2 ± 10.0	
^{171}Yb (centroid)	939.78 ± 0.54	938.8 ± 4.2	935.0 ± 3.3	943.7 ± 7.0	

field in the vicinity of the cell is less than 0.1 mT. Such magnetic fields cause broadening of the lines but do not shift the line center. As a further check on these errors, we have repeated the experiments with the vapor cell at different locations, and using vapor cells from different manufacturers. In our earlier work [15], we have shown that systematic shifts in the reference frequency are below 30 kHz. Finally, we have measured the same Yb transition using different hyperfine transitions to lock the reference laser. The reference frequency changes by known amounts (up to several GHz), and we have verified that the measured frequencies are consistent within the error bars. Changing the lock point of the reference laser also changes the cavity mode and the AOM offset needed to bring the Yb transition into resonance with the cavity. This checks for systematic errors that might arise from variations in the direction of the beam entering the cavity when the AOM frequency is varied.

The second class of systematic errors in our technique is wavelength dependent. Since our cavity is comparing the *wavelengths* of the two lasers, we have to convert these to frequencies using the refractive index of air [18]. Any error in the refractive index would reflect as a systematic shift in the measured frequency. From the reliability of the refractive index formulae, we estimate that this could cause a shift as large as 3 MHz in the measured frequencies. The only way to reduce this is to use an evacuated cavity, which we plan to do in the future. Wavelength-dependent systematic errors can also arise from varying phase shifts in the cavity mirrors, but we expect these to be negligible at the MHz level. It is important to note that both classes of systematic errors do not affect the determination of *frequency differences* of the unknown laser, up to several tens of GHz. For example, in the measurement of the Yb line, these errors would cause all the frequencies to shift, but the *differences* listed in table I will not change down to the kHz level.

The measured frequencies of the various transitions in the $^1S_0 \leftrightarrow ^1P_1$ line of Yb are listed in table I. Each value is an average of 6 individual measurements and the error quoted is the statistical error in the average. Even though we measure the absolute frequency of each transition, we have listed the values as shifts from the frequency of the ^{174}Yb transition. This is because, as mentioned earlier, the measured frequencies may have large systematic errors due to the uncertainty in the refractive index, but the frequency differences have negligible error. Furthermore, this way of presenting the data allows direct comparison with previous

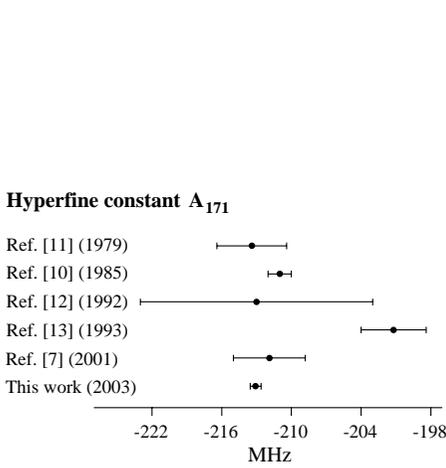


Fig. 4

Fig. 4 – Hyperfine structure in ^{171}Yb . The figure shows a comparison of our value of the hyperfine constant A_{171} with earlier values.

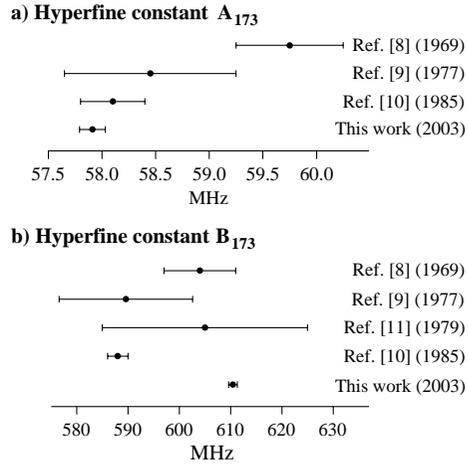


Fig. 5

Fig. 5 – Hyperfine structure in ^{173}Yb . The figure shows a comparison of our values of the hyperfine constants with earlier values: of A_{173} in a) and of B_{173} in b).

work where only the shifts were measured. It is clear from the several blank entries for the previous work that our work is the most comprehensive measurement of this line to date. In addition, some of the previous results have non-overlapping error bars to the extent of 10σ and our work resolves these discrepancies. For ^{176}Yb , it is now accepted that the low value obtained by Chaiko [19] is incorrect. For ^{171}Yb ($F = 1/2$), our value is in agreement with the recent value of 1151.4 MHz reported by Loftus *et al.* [7] but inconsistent with the value of 1136.2 MHz reported by Deilamian *et al.* [13]. Indeed, the main result in the work of Deilamian *et al.* is to resolve the overlapping ^{171}Yb ($F = 1/2$) and ^{170}Yb transitions. The separation they measure is 36.3 ± 2.5 MHz, which is in good agreement with our value of 39.5 ± 0.4 MHz. For ^{170}Yb , the most discrepant value is the result of Chaiko, where we have already seen that the value for ^{176}Yb is incorrect. The most recent result for ^{170}Yb is consistent only at the 2σ level, suggesting the need for further high-precision measurements to verify our results. All the other shifts reported by us agree with previous work.

We have used the data in table I to obtain the hyperfine-coupling constants in the $6s6p\ ^1P_1$ state of the odd isotopes, ^{171}Yb and ^{173}Yb . For ^{171}Yb , the measured $\{3/2 - 1/2\}$ interval is used to calculate the magnetic-dipole coupling constant A , yielding a value of $A_{171} = -213.08(47)$ MHz. This value is compared with earlier values in fig. 4. Our value is consistent but has significantly smaller error. For ^{173}Yb , there are three hyperfine levels and the measured intervals are used to calculate the magnetic-dipole coupling constant A and the electric-quadrupole coupling constant B . We obtain values of $A_{173} = 57.91(12)$ MHz and $B_{173} = 610.47(84)$ MHz. These values are again compared to earlier results in figs. 5a and b, respectively. While our value of A_{173} is consistent, the value of B_{173} is higher than the most recent value reported by Liening [10] from a level-crossing experiment. However, our value is in agreement with the other values. In addition, we have calculated the value of g_J from the value of $A_{171}/g_J = 206.0(16)$ reported by Budick and Snir [8] using a level-crossing experiment. We obtain a value of $g_J = 1.034(7)$, which is consistent with the currently accepted

value of 1.035(5) [11], giving further confidence in our results.

In conclusion, we have demonstrated a novel technique for measuring the frequencies of UV transitions using a Rb-stabilized ring-cavity resonator. The UV lines are accessed using a frequency-doubled IR laser. By measuring the frequency of the IR laser instead of the UV laser, we simplify the measurement process and avoid several sources of systematic errors. We have used this technique to measure various transitions in the 398.8 nm line in Yb. We obtain sub-MHz accuracy in the determination of isotope shifts for all isotopes (including the rarest isotope ^{168}Yb), representing an order of magnitude or more improvement in precision. We have also determined hyperfine structure for the odd isotopes with high precision. Our technique actually gives the *absolute* frequencies of the different transitions. However, we have quoted only the shifts from the ^{174}Yb transition since uncertainties in the refractive index of air in the cavity can cause an error as large as 3 MHz. In future, we plan to evacuate the cavity which will eliminate the need for refractive index correction. In addition, we plan to use a frequency-doubled *diode laser* for the UV spectroscopy. The output of the diode laser can be easily frequency modulated by modulating the injection current. The error signal generated can be fed back to lock the AOM frequency to a given peak, which will give us much higher accuracy in the frequency measurement. Using such a current-modulated diode laser, we have already demonstrated a precision of 30 kHz in measuring the 780 nm D_2 line of ^{85}Rb [15], and we hope to achieve similar precision in the measurement of UV lines.

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REFERENCES

- [1] UDEM T., REICHERT J., HOLZWARH R. and HÄNSCH T., *Phys. Rev. Lett.*, **82** (1999) 3568.
- [2] WEISS D. S., YOUNG B. C. and CHU S., *Phys. Rev. Lett.*, **70** (1993) 2706.
- [3] WOOD C. S., BENNETT S. C., CHO D., MASTERSON B. P., ROBERTS J. L., TANNER C. E. and WIEMAN C. E., *Science*, **275** (1997) 1759.
- [4] DEMILLE D., *Phys. Rev. Lett.*, **74** (1995) 4165.
- [5] HONDA K., TAKAHASHI Y., KUWAMOTO T., FUJIMOTO M., TOYODA K., ISHIKAWA K. and YABUZAKI T., *Phys. Rev. A*, **59** (1999) R934.
- [6] HALL J. L., ZHU M. and BUCH P., *J. Opt. Soc. Am. B*, **6** (1989) 2194.
- [7] LOFTUS T., BOCHINSKI J. R. and MOSSBERG T. W., *Phys. Rev. A*, **63** (2001) 023402.
- [8] BUDICK B. and SNIR J., *Phys. Rev.*, **178** (1969) 18.
- [9] BAUMANN M., LIENING H. and LINDEL H., *Phys. Lett. A*, **59** (1977) 433.
- [10] LIENING H., *Z. Phys. A*, **320** (1985) 363.
- [11] GRUNDEVIK P., GUSTAVSON M., ROSÉN A. and RYDBERG S., *Z. Phys. A*, **292** (1979) 307.
- [12] BERENDS R. W. and MALEKI L., *J. Opt. Soc. Am. B*, **9** (1992) 332.
- [13] DEILAMIAN K., GILLASPY J. D. and KELLEHER J. D., *J. Opt. Soc. Am. B*, **10** (1993) 789.
- [14] YE J., SWARTZ S., JUNGNER P. and HALL J. L., *Opt. Lett.*, **21** (1996) 1280.
- [15] BANERJEE A., DAS D. and NATARAJAN V., to be published in *Opt. Lett.* (2003).
- [16] ARIMONDO E., INGUSCIO M. and VIOLINO P., *Rev. Mod. Phys.*, **49** (1977) 31.
- [17] BANERJEE A., RAPOL U. D., WASAN A. and NATARAJAN V., *Appl. Phys. Lett.*, **79** (2001) 2139.
- [18] BIRCH K. P. and DOWNS M. J., *Metrologia*, **30** (1993) 155; **31** (1994) 315.
- [19] CHAIKO Y., *Opt. Spectrosc.*, **20** (1966) 424.