

Hyperfine structure and absolute frequency of the $^{87}\text{Rb } 5P_{3/2}$ state

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We have constructed two highly stable and reproducible $^{87}\text{Rb } D_2$ -saturated-absorption spectrometers at 780 nm, using dither/third-harmonic lock-in detection and radio-frequency sideband techniques, respectively. We achieved ± 3 -kHz reproducibility and agreement between these two independent systems. Heterodyne measurements of the hyperfine splittings of the $5P_{3/2}$ state give its magnetic dipole (A) and electric quadrupole (B) hyperfine constants with a 10-fold reduction in uncertainty. © 1996 Optical Society of America

In the pursuit of simple, low-cost, yet high-quality visible optical frequency sources, we recently constructed an optical chain to measure the absolute frequency of molecular I_2 hyperfine resonances in the 532-nm region¹ (doubled YAG). A saturated-absorption spectrometer of the $^{87}\text{Rb } D_2$ line at 780 nm was used as a reference transfer standard.² The accuracy of our initial chain result was limited by the ± 60 -kHz uncertainty of the $^{87}\text{Rb } D_2$ line, which was in part associated with its broad natural resonance line width of 6-MHz FWHM. To improve the utility of this reference standard we studied its systematics and made corresponding corrections. A dramatic improvement on the available frequency stability and reproducibility has been achieved, and we are now able to lock onto the line centers with an accuracy of $\sim 1/2000$ of the linewidth.

Degradation of the $^{87}\text{Rb } D_2$ profiles occurs for the hyperfine-structure components with strong cycling character (e.g., $F'' = 2 \rightarrow F' = 3$, $F'' = 1 \rightarrow F' = 0$). With optical intensities below 0.2 mW/cm^2 we observed sign reversals on the saturated-absorption peaks owing to velocity-selective optical pumping process.³ With careful experimentation on the spectrometer, we were able to find a comfortable optical intensity range in which the observed line shapes can be well accounted for by a simple FM or third-harmonic formula. We stress here that the optimal intensity depends on the choice of the beam size. We also minimized optical power shift on the lines by dividing the power into pump and probe beams in an appropriate ratio, balancing various effects of light pressure⁴ and optical pumping.

Heterodyne experiments between two $^{87}\text{Rb } D_2$ spectrometers offer a powerful check on their systematics as well as a natural determination of all possible hyperfine component intervals. We built two such systems based on two different signal-recovery modulation schemes. JILA-built Ti:sapphire lasers were used for the sole reason of participating in the power-demanding optical frequency chain. Figure 1(a) shows the dither/third-harmonic lock-in detection system. The 6-MHz peak-to-peak optical frequency dither at 5 kHz was provided by a double-passed acousto-optic modulator (AOM) fed from an

FM-dithered 80-MHz voltage-controlled oscillator. To ensure a stable center frequency, we tightly phase locked this oscillator (after a digital phase division by 10) to an 8-MHz precision FM waveform generated by a direct-digital-synthesis (DDS) frequency synthesizer. To achieve a better rejection of offsets by neighboring lines and Doppler background we recovered the signal at the third harmonic, using a lock-in synchronized to three times the 5-kHz dither. The residual AM at 15 kHz was carefully monitored on our probe detector. (The 5- and 10-kHz signals were heavily notched out, to near the shot-noise level.) By adjusting the Bragg angle of the AOM, we reduced the 15-kHz AM to be below -60 dB relative to our signal. A broadband intensity stabilizer following the spatial mode filter provided another >10 -dB suppression. We chopped the saturating beam at 610 Hz and demodulated the 15-kHz lock-in output additionally by a 610-Hz lock-in to capture the part of the Rb signal that was changed by the chopped saturating beam.

Figure 1(b) shows our FM system. The pump beam is chopped and frequency offset by an 80-MHz AOM, and 4-MHz FM sidebands with modulation index of 0.8 are imposed on the probe beam. A resonant 4-MHz photodetector detects the AM signal converted from FM by the Rb atoms, and its signal is subsequently demodulated by a double-balanced mixer (DBM). In both spectrometers the pump and probe beams propagate accurately antiparallel and are spatially overlapped to avoid residual Doppler shifts and broadening. Polarizers guarantee parallel linear polarizations in the Rb cells. Magnetic fields in the cells are reduced to be $<2 \mu\text{T}$ by two layers of magnetic shields. The ratios of pump to probe powers were kept near 4. In the 3- f system the beam diameter was ~ 3.5 mm, and we typically used $35\text{-}\mu\text{W}$ pump power (0.37 mW/cm^2); in the FM system they were 1 mm and $7 \mu\text{W}$, respectively (0.89 mW/cm^2). The 3- f spectrometer showed small power shifts (see Fig. 2, $\pm 20\%$): e component, $+50 \text{ Hz}/\mu\text{W}$; d component, $+1 \text{ Hz}/\mu\text{W}$; d/f crossover, $+70 \text{ Hz}/\mu\text{W}$. High-quality Rb cells were made at JILA with the standard cleaning and baking procedure.⁵ The Rb was purified several times by distillation before being transferred into the cells. The pressure at tip-off was $<130 \mu\text{Pa}$. The resid-

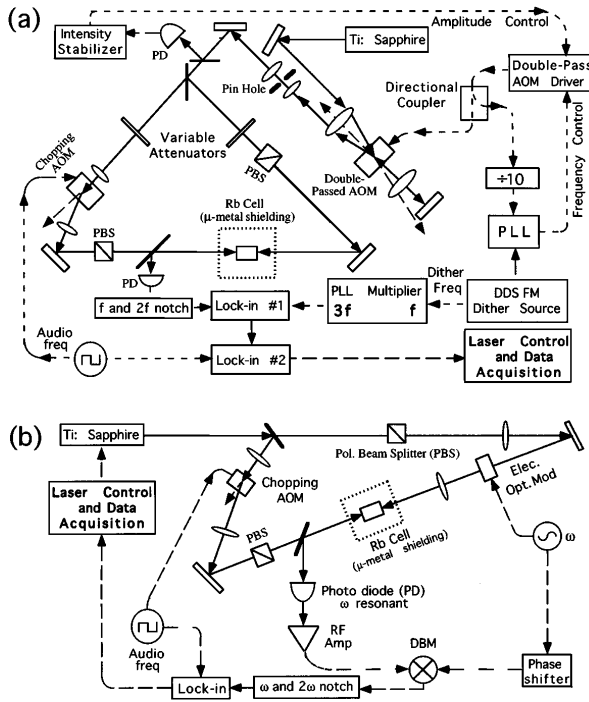


Fig. 1. Experimental setup for our Rb D_2 spectrometers. Both use an AOM to chop and frequency offset the saturating beam. The third-harmonic dither lock-in detection approach is shown in (a), and the FM technique is shown in (b). PLL's, phase-locked loops.

ual background contamination can be estimated by measurement of the linewidths of the Rb transitions. Extrapolating to zero power results in a measured linewidth in both spectrometers of ~ 7 MHz, compared with ~ 6 -MHz natural linewidth. This extra 1 MHz includes all experimental parameters and in part is due to vibration of the laser prestabilizing reference cavities as well as to any possible gas contamination in the cell. Various cells placed in one spectrometer produced frequency shifts of less than a few kilohertz. It is satisfying that the lasers are locked with an offset of < 3 kHz from the unperturbed line center of a free atom.

Figure 2 shows a typical scan for the $^{87}\text{Rb } 5S_{1/2} (F'' = 2) \rightarrow 5P_{3/2} (F' = 1, 2, 3)$ saturated-absorption profiles in both spectrometers. Assignment of the hyperfine components is indicated. Excellent S/N results (normalized for 1-Hz bandwidth) of 7200:1 in the FM and 2600:1 in the $3f$ systems were achieved. Increased laser noise at the relatively low modulation frequency of 5 kHz in the $3f$ system is mainly responsible for the reduced S/N. FM⁶ and Wahlquist third-derivative line-shape formulas were used to fit the correspondingly scanned data. The latter third-derivative line shape⁷ is derived from

$$\text{Signal}(3\omega) = \text{Sign}(H_\delta) \left(\frac{2}{H_\omega} \right)^2 \left[\frac{\sqrt{2\gamma - \mu}}{2\sqrt{\mu - 2}(\mu - \gamma)} - 2\sqrt{2\gamma - \mu} \left(\frac{\mu - 1}{\sqrt{\mu - 2}} - \sqrt{\mu} \right) \right], \quad (1)$$

where H_δ is the frequency detuning from the line center and H_ω is the modulation amplitude. The

dimensionless parameters are defined as follows:

$$\gamma = 1 + \beta^2 + \alpha^2, \quad \mu = \gamma + \sqrt{\gamma^2 - 4\alpha^2}, \quad (2)$$

with $\alpha = (H_\delta/H_\omega)$ and $\beta = 1/2 H_{1/2}/H_\omega$, $H_{1/2}$ being the FWHM of the resonance line. The fit residuals, displayed in Fig. 2 with 10-fold magnification, show basically structureless noise, indicating that the line shape of our spectrometers is well understood.

Figure 3 shows the recorded long-time beat between the FM and the third-harmonic-detection stabilized lasers, both locked on the df -crossover component. The corresponding Allan variance of $\sigma_y = 3.8 \times 10^{-12}/\sqrt{\tau}$ matches that calculated from the S/N data of the $3f$ spectrometer, and it has the white frequency noise characteristics. This noise is roughly half that of the 633-nm He-Ne/I₂ system. At 1-s averaging we obtained a frequency noise of ± 1.4 kHz. More significantly, the Allan variance continues to

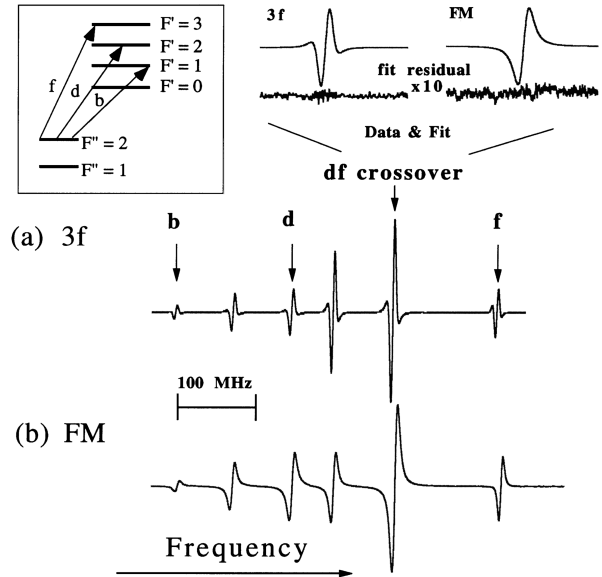


Fig. 2. Frequency scan over the hyperfine components in ^{87}Rb in both spectrometers. Theoretical fits for the df crossover and their residuals (10 \times) are shown. Assignment of the components is indicated.

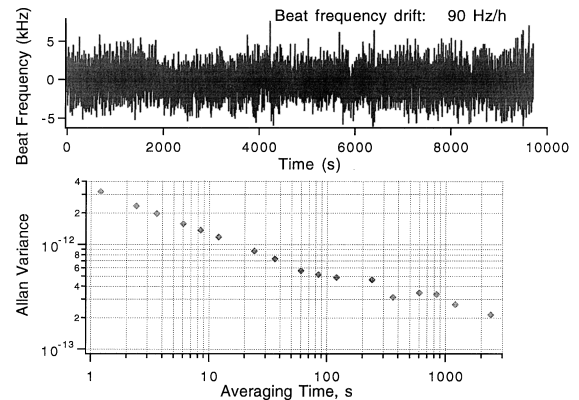


Fig. 3. Time record of the beat frequency between the two spectrometer-stabilized laser systems. Allan variance is calculated from these data. The offset of 5 kHz between the two systems has been suppressed.

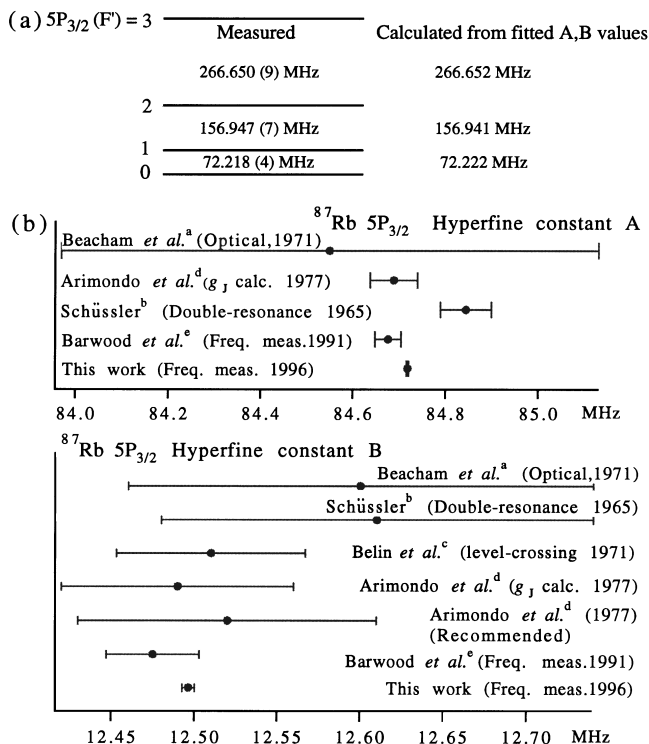


Fig. 4. (a) Measured values of the hyperfine splittings of the $^{87}\text{Rb } 5P_{3/2}$ state; (b) comparison between A and B values determined here and some previous results. The key to references is as follows: ^a, Ref. 10; ^b, Ref. 11; ^c, Ref. 12; ^d, Ref. 8; ^e, Ref. 2. The Schüssler measurement of the constant A is also the recommended value given in Ref. 8.

decrease below 2×10^{-13} with increasing averaging time to >2500 s, indicating the relative absence of changing systematic offsets. This is clearly verified by the beat frequency's slow drift of 90 Hz/h. By flipping the lasers back and forth between several pairs of transitions one can determine the offsets between the two systems. In the FM spectrometer, frequency pulling effects by neighboring lines are exacerbated by their broader features, so appropriate corrections were subsequently calculated and applied. Transitions originating from both $F'' = 1$ and $F'' = 2$ components of the ground state $5S_{1/2}$ were used, and data for hyperfine splittings were accumulated over several months with a reproducibility of approximately ± 3 kHz. The scale of this frequency noise and reproducibility is set by the linewidth of 6 MHz.

Figure 4(a) summarizes our result for the hyperfine splittings of $5P_{3/2}$ state. Using least squares with the hyperfine Hamiltonian⁸ and our hyperfine interval data, we determine the magnetic dipole constant A and the electric quadrupole constant B to be $A = 84.7185(20)$ MHz and $B = 12.4965(37)$ MHz. These results agree with Refs. 2 and 8 but are more accurate by at least a factor of 10. Figure 4(b) shows the comparison among those various re-

sults. As a reality check, we also determine the coefficient A for the $5S_{1/2}$ ground state from its splittings. Our value of 3417.346 (3) MHz is in satisfactory agreement with that of 3417.3413 MHz given by high-precision microwave measurement.^{8,9}

Recently the two-photon Rb transition at 778 nm was established (± 5 kHz) as a secondary frequency standard.¹³ With the help of a Schottky diode, we also measured¹⁴ the direct frequency gap between the $^{87}\text{Rb } D_2$ line $5S_{1/2} \rightarrow 5P_{3/2}(F'' = 2 \rightarrow F' = 2/F'' = 2 \rightarrow F' = 3$ crossover resonance) and the two-photon transition of $^{85}\text{Rb } 5S_{1/2} \rightarrow 5D_{3/2}(F'' = 2 \rightarrow F' = 4)$ to be $-1\,014\,234.4856$ MHz ± 2.3 kHz, thereby establishing the absolute frequency of this Rb D_2 line as $384\,227\,981.8773$ MHz ± 5.5 kHz.

In summary, these two highly stable Rb D_2 spectrometers are able to operate at ± 3 -kHz frequency reproducibility, representing line-position uncertainties of $<1/2000$ linewidth. Hyperfine constants of the $5P_{3/2}$ state were determined with precision improved by a factor of 10. We expect to realize inexpensive, compact, and yet excellent 780-nm secondary optical frequency standards using our spectrometers with diode lasers.

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References

1. P. Jungner, S. Swartz, M. Eickhoff, J. Ye, J. L. Hall, and S. Waltman, *IEEE Trans. Instrum. Meas.* **44**, 151 (1995).
2. G. P. Barwood, P. Gill, and W. R. C. Rowley, *Appl. Phys. B* **53**, 142 (1991); *Proc. SPIE* **1837**, 262 (1992).
3. O. Schmidt, K.-M. Knaak, R. Wynands, and D. Meschede, *Appl. Phys. B* **59**, 167 (1994).
4. R. Grimm and J. Mlynek, *Appl. Phys. B* **49**, 179 (1989).
5. J. M. Chartier, S. Picard-Fredin, and A. Chartier, *CCDM/92-2/Rapport BIPM 92/4*.
6. J. L. Hall, L. Hollberg, T. Baer, and H. G. Robinson, *Appl. Phys. Lett.* **39**, 680 (1981).
7. H. Wahlquist, *J. Chem. Phys.* **35**, 1708 (1961).
8. E. Arimondo, M. Inguscio, and P. Violino, *Rev. Mod. Phys.* **49**, 31 (1977), and references therein.
9. All errors are quoted on a 1σ basis.
10. J. R. Beacham and K. L. Andrews, *J. Opt. Soc. Am.* **61**, 231 (1971).
11. H. A. Schüssler, *Z. Phys.* **182**, 289 (1965).
12. G. Belin and S. Svanberg, *Phys. Scr.* **4**, 269 (1971).
13. F. Nez, F. Biraben, R. Felder, and Y. Millerioux, *Opt. Commun.* **102**, 1643 (1993).
14. J. L. Hall, J. Ye, L.-S. Ma, S. Swartz, P. Jungner, and S. Waltman, in *5th Symposium on Frequency Standards and Metrology*, J. C. Bergquist, ed. (World Scientific, Singapore, 1995).