Absolute frequency measurement of rubidium 5S-7S two-photon transitions with a femtosecond laser comb

Hsiang-Chen Chui, Ming-Sheng Ko, Yi-Wei Liu, and Jow-Tsong Shy

Department of Physics, National Tsing Hua University, Hsinchu 300, Taiwan

Jin-Long Peng and Hyeyoung Ahn

Center for Measurement Standards, Industrial Technology Research Institute, Hsinchu 300, Taiwan

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The absolute frequencies of rubidium 5S-7S two-photon transitions at 760 nm are measured to an accuracy of 20 kHz with an optical frequency comb based on a mode-locked femtosecond Ti:sapphire laser. The rubidium 5S-7S two-photon transitions are potential candidates for frequency standards and serve as important optical frequency standards for telecommunication applications. The accuracy of the hyperfine constant of the $7S_{1/2}$ state is improved by a factor of 5 in comparison with previous results. © 2005 Optical Society of America

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The 760-nm Rb 5S-7S two-photon transitions, which provide a frequency standard for a frequencydoubled 1520-nm laser in telecommunication applications, has been observed with a 760-nm diode laser¹ and a frequency-doubled 1520-nm diode laser with a periodically poled lithium niobate (PPLN) waveguide.² The characteristic that such S-S transitions are insensitive to a magnetic field is particularly interesting as a frequency standard, in comparison with the 778-nm Rb 5S-5D transition, which has been recommended as the realization of the SI length unit meter.³ Meanwhile, half of the frequency of the Rb 5S-7S two-photon transition is only 30 GHz away from the P(5) transition of the simplest heteronuclear diatomic molecule HD.⁴

An optical frequency comb based on a femtosecond Ti:sapphire laser has been developed as a powerful tool that directly links the microwave frequency standard to an optical region.⁵ As a frequency ruler, the accuracy of such a frequency comb could reach as high as one part in 10^{15} in measuring an unknown optical frequency. In this Letter we report the frequency measurement of Rb 5S-7S transitions with an optical frequency comb.⁶

The optical frequency comb, shown in Fig. 1, was built by the Center for Measurement Standards, Taiwan.' It has a repetition frequency of 1 GHz and a pulse width of ~ 50 fs. It can deliver an average power of more than 700 mW. A commercial photonic crystal fiber with a core diameter of 1.8 µm and a zero-dispersion wavelength at 710 nm was used to expand the spectrum to cover an octave. A broadband frequency comb from 450 to 1100 nm with an average power of 200 mW was generated. The repetition frequency of the comb was phase locked to a 1-GHz signal from a synthesizer by tuning the laser cavity. The f-2f technique was used to detect the offset frequency.⁵ The typical signal-to-noise ratio of the offset beat signal was 30 dB in a 100-kHz resolution bandwidth. The offset frequency was phase locked to another synthesizer by controlling the pump power with an acousto-optic modulator. All the synthesizers, spectrum analyzers, and counters in this experiment were referenced to a stable microwave source. which consists of a Rb clock and a low-noise ovencontrolled quartz oscillator for improving short-term stability. For long-term stability the frequency of the Rb clock is calibrated with a global positioning system receiver. This microwave source has a stability of better than 2×10^{-12} for an integration time longer than 1 s. The uncertainty of the frequency calibration within 1 day of average is less than 10^{-12} . The stabilized repetition frequency and offset frequency have residual peak-to-peak fluctuations of <2 and 30 mHz, respectively. This contributes frequency fluctuations of < 0.8 kHz of the frequency comb near 760 nm.

The experimental setup of the Rb 5S-7S twophoton spectrometer is based on a 1520-nm externalcavity diode laser and a PPLN waveguide frequency doubler.² The Rb cell is heated to 130°C (vapor pressure of 1.07 mTorr), and the laser power is 7 mW in



Fig. 1. Optical frequency comb system of the Center for Measurement Standards, Taiwan: DM, dichroic mirror; APDs, avalanche photodiodes; SHG, second-harmonic generation; AOM, acousto-optic modulator; PZT, piezoelectric transducer; PCF, photonic crystal fiber; HWP, half-wave plate; PLL, phase-locked loop; LBO, lithium triborate; GPS, global positioning system.



Fig. 2. Experimental setup for the beat note frequency measurement: PBS, polarizing beam splitter.



Fig. 3. Beat signal between comb lines at 760 nm and the Rb-stabilized laser. The resolution bandwidth is 100 kHz.

the cell. One milliwatt of the 760-nm laser beam is picked up with a glass plate to perform the absolutefrequency measurement.

The setup for measuring the beat note of the 760-nm laser (UNKNOWN) and the optical frequency comb is shown in Fig. 2. A half-wave plate is used to adjust the polarization of the UNKNOWN. The comb lines are filtered with a 760-nm bandpass filter with a 10-nm bandwidth. The optical power of the 760-nm comb lines is ~ 1 mW. The UNKNOWN and the 760-nm comb lines combine at a polarizing beam splitter. A polarizer is used to project the polarizations of the UNKNOWN and comb lines in the same direction. The beat note is then detected with an avalanche photodiode. The beat signal first passes through a tunable rf bandpass filter and is then amplified to ~ -10 dBm. The beat signal, as shown in Fig. 3, has a linewidth of 8 MHz, which is contributed mainly by the frequency modulation of the diode laser. Finally, the beat note is counted by a universal counter (Agilent 53132A).

The Allan deviation of the beat frequency is better than 2×10^{-11} at a 10-s integration time (as shown in Fig. 4). The measured frequency of each transition is the average of four different data sets taken within 40 days. Figure 5 shows the distribution of an individual data set with good reproducibility. All the data sets agree with each other to within an uncertainty of 20 kHz.

An alternative method that frequency offset locks the diode laser on one of the optical frequency comb components rather than the Rb transition is employed to check the measurement. With this method we are able to scan the diode laser through the transition with high-precision frequency control, and then the center frequency of the transition can be found. This method has the advantage that it does not modulate the laser frequency, therefore providing a sharper beat note signal. However, it requires a longer measuring time and a more stable environment. The data set in Fig. 5, which is the result with this method, agrees with the other results.

Since the accuracy of our measurements could reach 20 kHz, several systematic effects should be taken into account to deduce the transition frequency. The calculated light shift (ac Stark shift) is -40 Hz/mW,¹ corresponding to -280 Hz at 7 mW, which is ten times smaller than 5S-5D. In our experimental scheme, because of the limitation of the available laser power and the accuracy of the frequency measurement, it is too small to be resolved.

The pressure shift is estimated to be -40 kHz/mTorr by extrapolation of the experimental data of the Rb–Rb collision in n > 10 states.⁸ With absolute frequencies measured under different temperatures (shown in Fig. 6) the measured pressure shift was found to be -51.7(5.0) kHz/mTorr, which is larger than the estimation mentioned above.

Two-photon spectroscopy eliminates the first-order Doppler broadening, but not the second order. For a temperature of 130°C we estimate a second-order Doppler shift of -240.6 Hz at 394 THz. The blackbody-radiation-induced dynamic Stark shifts of energy levels of hydrogen and different alkali atoms including Rb at 300 K have been calculated.⁹ Considering a T^4 dependence, such shifts at 130 °C are



Fig. 4. Allan deviation of the beat frequency between our optical frequency comb and the Rb-stabilized laser.



Fig. 5. Measured frequency of the 87 Rb F = 1-1 two-photon transition within 40 days.



Fig. 6. Frequency shift of the two-photon line center versus the pressure in the Rb cell. The 760-nm laser is stabilized on the ⁸⁷Rb F = 2-2 two-photon transition.

Table 1. Corrections of Systematic Effects

Effect	Shift at 760 nm
Light shift	-280 ± 4 Hz
Pressure shift	-55.3 ± 5.3 kHz
Blackbody radiation	-665.8 ± 4 Hz
Second-order Doppler shift	$-240.6 \pm 0.1 \text{ Hz}$
Neighboring transitions	< 10 Hz
Electronics	$\pm 500 \text{ Hz}$
Total	-56 ± 6 kHz

Table 2. Measured Laser Absolute Frequencies of Rb $5S_{1/2} \rightarrow 7S_{1/2}$ Two-Photon Transitions and the Results After the Corrections of Systematic Effects

Transition	Laser Frequency (kHz)	Note
⁸⁷ Rb	394 397 384 383(20)	Measured
F = 2 - 2	394 397 384 439(20)(6)	Corrected
⁸⁵ Rb	394 397 906 927(20)	Measured
F = 3 - 3	394 397 906 983(20)(6)	Corrected
⁸⁵ Rb	394 399 282 765(20)	Measured
F = 2 - 2	394 399 282 821(20)(6)	Corrected
⁸⁷ Rb	$394\;400\;481\;965(20)$	Measured
F = 1 - 1	$394\;400\;482\;021(20)(6)$	Corrected

Table 3. Hyperfine Constant A of the Rb $7S_{1/2}$ Statein Megahertz

Isotope	This Work	Ref. 10	Ref. 11
⁸⁷ Rb	319.759(28)	319.7(1)	318.1(32)
⁸⁵ Rb	94.658(19)	94.7(1)	94.00(64)

-9 Hz for $5S_{1/2}$ and -1340.5 Hz for $7S_{1/2}$, which leads to a shift of -665.8 Hz for the $5S_{1/2} \rightarrow 7S_{1/2}$ twophoton transitions at 394 THz. The line-shape distortion caused by the tails of neighboring transitions results in a frequency shift of the line center. These frequency shifts are evaluated by theoretical simulation of the line shape. The largest shift (⁸⁷Rb F= 2–2 transition), owing to the Doppler background of neighboring transitions, is +10 Hz. The stabilized laser frequency could be shifted approximately ± 500 Hz at 394 THz, owing to the offset at the output of the lock-in amplifier. The derivative-like error signal generated by FM spectroscopy has a slope of 500 Hz/mV (at 394 THz), and the offset error is assumed to have a maximum of ± 2 mV. The shifts caused by various systematic effects are summarized in Table 1. The frequency shift is contributed mainly by the pressure shift.

The measured laser absolute frequencies of Rb $5S_{1/2} \rightarrow 7S_{1/2}$ two-photon transitions and the results after the corrections of systematic effects are summarized in Table 2. The isotope shift of the Rb 5S-7S transition is calculated to be 131.567(73) MHz from this work. This result is in agreement with a previous result by use of the fringe interpolation method¹ but is more precise. The hyperfine constant of the 7S state is also derived from this measurement (see Table 3). Compared with previous experiments, the results are improved by a factor of 5.

In summary, the absolute frequencies of Rb twophoton transitions have been measured to an accuracy of 20 kHz with an optical frequency comb. These transitions provide four frequency markers at both 760 and 1520 nm. The limitation of the accuracy (20 kHz, 5×10^{-11}) is dominated by the observed transition linewidth (statistic) that resulted from the jitter of the diode laser and pressure (systematic) of the Rb two-photon spectrometer, rather than the optical frequency comb (0.8 kHz, 2×10^{-12}). Therefore the accuracy can be further improved by increasing the laser stability and lowering the pressure in the Rb cell.

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