Optical frequency synthesizer for precision spectroscopy of Rydberg states of Rb atoms

Naoto Watanabe*, Hikaru Tamura, Mitsuru Musha, and Ken’ichi Nakagawa

Institute for Laser Science, University of Electro-Communications, Chofu, Tokyo 182-8585, Japan

*E-mail: n_watanabe@ils.uec.ac.jp

Received May 9, 2017; accepted August 18, 2017; published online October 12, 2017

We have developed an optical frequency synthesizer for the precision spectroscopy of highly excited Rydberg states of Rb atoms. This synthesizer can generate a widely tunable 480 nm laser light with an optical power of 150 mW and an absolute frequency uncertainty of less than 100 kHz using a high-repetition-rate (325 MHz) Er fiber-based optical frequency comb and a tunable frequency-doubled diode laser at 960 nm. We demonstrate the precision two-photon spectroscopy of the Rydberg states of \(^{87}\)Rb atoms by observing the electromagnetically induced transparency in a vapor cell, and measure the absolute transition frequencies of \(^{87}\)Rb to \(nD\) (\(n = 53–92\)) and \(nS\) (\(n = 60–90\)) Rydberg states with an uncertainty of less than 250 kHz. It is the first direct frequency measurements of these transitions using an optical frequency comb. © 2017 The Japan Society of Applied Physics

1. Introduction

Optical frequency synthesizers have become invaluable tools for recent atomic physics experiments, as they provide various optical frequencies with a high absolute frequency accuracy in the optical region from near-infrared to visible.\(^{1−7}\) Such optical frequency synthesizers have been used for optical clocks.\(^{8}\) Recently, Rydberg atoms, highly excited states of atoms with a large principal quantum number, have become attractive because quantum entanglement between nearby atoms is relatively simple to realize by using the long-range interaction between Rydberg atoms.\(^{9}\)

To excite atoms into Rydberg states, multiple lasers with different wavelengths are usually required to achieve the transition by a multiphoton process. In the case of rubidium (Rb) Rydberg atoms, two-frequency-tunable lasers at 780 and 480 nm with an accuracy of less than 100 kHz \((\Delta \nu / \nu < 2 \times 10^{-10})\) are required for two-photon excitation. Although the 780 nm laser is stabilized on the \(5S_{1/2} \rightarrow 5P_{3/2}\) transition, the 480 nm laser is typically stabilized to an optical cavity.\(^{10,11}\) The stabilization method using an optical cavity is simple and useful. However, it is difficult to measure the absolute optical frequency by this method. On the other hand, an optical frequency synthesizer can generate an arbitrary optical frequency with a frequency accuracy equivalent to that of a global positioning system (GPS)-locked reference atomic clock. An optical frequency synthesizer based on an erbium (Er) fiber mode-locked laser is very useful because of its stable long-time operation, convenient operating wavelength of the Er-doped optical fiber amplifier (EDFA) and low cost. In many of these applications, Er fiber mode-locked lasers with low repetition rates from 50 to 200 MHz have been typically used because of the relative ease of experimental realization. However, the higher repetition frequency is desirable because of the relative ease of the determination of mode numbers with a typical commercial wavemeter. It is also expected to obtain a higher signal-to-noise ratio (SNR) in the beat measurements between the comb and other CW lasers owing to the higher optical power per comb mode. Recently, Er fiber mode-locked lasers with repetition rates of up to 300 MHz have been developed by several groups.\(^{12−17}\)

However, a self-referenced comb based on a high-repetition-rate laser requires a higher average power of more than 300 mW for the expanding spectrum. We have developed an optical frequency synthesizer based on an Er fiber mode-locked laser with a high repetition rate at 325 MHz,\(^{14}\) using a cascade of two EDFAs. In this system, the mode number is easily determined using a commercial wavemeter, and an arbitrary frequency can be generated over a broad-wavelength region owing to the high teeth power of the comb.

We also report on the precision spectroscopy of Rydberg states of \(^{87}\)Rb atoms using the developed optical frequency synthesizer. The high-resolution spectroscopy of Rydberg states is possible by observing the electromagnetically induced transparency (EIT) spectrum in an atomic vapor cell.\(^{18−23}\) In previous reports,\(^{22}\) these absolute transition frequencies of \(nS\) (\(n = 19–65\)) and \(nD\) (\(n = 19–57\)) \(^{87}\)Rb Rydberg states were measured using a wavemeter and a comb with an uncertainty of less than 800 kHz. The direct frequency measurements of the \(nP\) and \(nF\) Rydberg states of \(^{85}\)Rb have also been demonstrated using three laser frequencies, which are locked to the Rydberg transitions, and measured using a comb.\(^{23}\) However, to the best of our knowledge, the results reported here are the first direct frequency measurements of the \(nS\) (\(n = 60–90\)) and \(nD\) (\(n = 53–92\)) Rydberg states of \(^{87}\)Rb with a 3-fold reduction in uncertainty using the optical synthesizer. The present synthesizer can generate a high frequency-accurate and frequency-tunable 480 nm laser beam, and is useful for the high-resolution spectroscopy of the Rydberg states of Rb, which we use for our experiments\(^{24}\) regarding quantum simulation using cold Rydberg atoms.

2. Optical frequency comb

We developed an optical frequency comb based on a high-repetition-rate Er fiber mode-locked laser.\(^{14}\) Because the repetition frequency at 325 MHz is approximately \(10^6\) in the 1 mm wavelength region, the mode number of the comb can be determined using a commercial wavemeter with accuracy from \(10^{-6}\) to \(10^{-7}\). However, a higher peak power is required
for the spectrum broadening using a high nonlinearity fiber (HNLF), because the pulse energy decreases as the repetition frequency of the oscillator increases. To obtain sufficient pulse energy (>1 nJ) for the spectrum broadening, the output power of the oscillator at 120 mW is amplified up to 480 mW using a cascade of two EDFA. Figure 1(a) shows a schematic diagram of the self-referenced frequency comb. To obtain a higher gain power amplification, each Er-doped fiber has a high peak absorption of 80 dB/m at 1530 nm. The lengths of the first and second gain fibers are 720 and 1530 nm, respectively. The EDFA are pumped by 976 nm diode lasers from the output side via a wavelength division multiplexing (WDM) coupler. The pump power for the first EDFA was 950 mW. The pump source for the second EDFA is two pairs of 976 nm diode lasers to offer a pump power up to 1570 mW. The amplified light is sent to the HNLF, which is 500 mm long. The HNLF has a nonlinear coefficient of 21 W−1 m−1 and a zero-dispersion wavelength of 1549 nm. To achieve the higher peak power for the octave-spanning spectrum, the broadened pulse trains from the two EDFA are compressed by splicing a single-mode fiber (SMF) before the HNLF. Figure 1(b) shows the octave-spanning spectrum after passing through the HNLF. A carrier envelope offset (CEO) frequency is detected from the beat signal between the repetition frequency of the oscillator. Figure 1(c) shows the detected CEO beat signal with 100 kHz resolution bandwidth. (d) Energy diagram of 87Rb.

3. Optical frequency synthesizer

Next, we developed an optical frequency synthesizer for Rb Rydberg experiments based on the high-repetition-rate frequency comb described in Sect. 2. Figure 1(d) shows the energy-level diagram of 87Rb. Two lasers at 780 and 480 nm are used for two-photon excitation to the nS and nD Rydberg states of 87Rb. For the experiments using cold Rydberg atoms, each laser requires a frequency accuracy of less than 100 kHz and a wide frequency-tunable range. We generate an arbitrary optical frequency close to 480 nm using a 960 nm laser stabilized by the comb realized in Sect. 2. These optical frequency references are generated by broadening the spectrum of the comb from 1 to 2 µm, as shown in Fig. 1(b). In
general, each optical frequency reference is generated by splitting the oscillator output into multiple branches that are amplified and sent to HNLFs. To simplify the optical setup, the output spectrum from the HNLF is split into three wavelength regions at 1030, 1560, and 1920 nm using two long-pass dichroic mirrors (DMs). The second harmonics of the wavelength components of the comb around 1560 and 1920 nm were generated at 160 and 300 µW, respectively, by using PPLN crystals. The tooth powers at 780 and 960 nm were 130 and 60 nW with spectral bandwidths of 0.8 and 5 nm (FWHM), respectively. The SNRs of two beat signals between each comb spectrum and each extended-cavity diode laser (ECLD) were more than 25 dB over a bandwidth of 100 kHz. Thus, we obtain sufficient power in each comb mode to stabilize the optical frequency of the lasers at two wavelength regions because the teeth power of the high-repetition-rate comb is sufficient.

The 480 nm laser light was generated by a frequency-doubling of the 960 nm ECLD using a periodically poled KTP (PPKTP) crystal. The output light of 960 nm is amplified up to 1 W using a tapered amplifier (TA), and sent to a ring build-up cavity with an input coupling mirror reflectivity of 92% to enhance the fundamental light intensity. The build-up cavity is stabilized by the Pound–Drever–Hall method in which the current of the laser is modulated at 15 MHz. With this configuration, blue light is generated with a power of more than 150 mW. Wide scanning of the optical synthesizer is accomplished by changing the repetition rate of the comb because the beat note is maintained at the same frequency. The repetition frequency of the comb can remain locked over a scan range of 700 Hz corresponding to 1.3 GHz in the 480 nm wavelength region. In Fig. 2, the blue laser at 480 nm was scanned over more than 150 MHz with an approximately 400 kHz step size. Thus, we can generate an arbitrary optical frequency in the 480 nm region with an uncertainty of less than 100 kHz \((\Delta \nu / \nu < 2 \times 10^{-10})\). In next section, we describe an optical setup to observe the EIT spectrum of Rb Rydberg atoms using the optical synthesizer.

4. Absolute transition frequency measurements of Rb Rydberg states

As a demonstration of the precision frequency tuning and the frequency accuracy of our optical frequency synthesizer, we measured the absolute transition frequencies of \(^{87}\)Rb Rydberg states. These frequencies were determined by observing the EIT spectra of a probe laser transmission at 780 nm. We show this optical setup in Fig. 3. The probe laser beam with a power of approximately 4 µW and a beam waist of 30 µm propagates through a magnetically shielded Rb vapor cell of 10 cm length. To reduce the intensity fluctuation of the probe laser, the probe laser beam is split into two before and after transmission through the cell, and the difference between two signals is recorded by an oscilloscope. A coupling laser beam at 480 nm counter-propagates through the cell at a power of approximately 15 mW and a beam waist of 100 µm. The probe laser is frequency-stabilized to the \(5S_{1/2} F = 2\)–\(5P_{3/2} F = 3\) transition of \(^{87}\)Rb by modulation transfer spectroscopy, and its absolute optical frequency is measured by our optical frequency comb. The coupling laser frequency is scanned by changing the repetition rate of the comb. The scan range is approximately 40 MHz and the frequency step is approximately 100 kHz at 480 nm. A typical Rydberg EIT spectrum is shown in Fig. 4. We repeat each scan 10 times and average the recorded probe signals. As the coupling laser frequency is precisely generated by our optical synthesizer, we can obtain higher-resolution EIT spectra with a higher SNR. In Fig. 4, there is a small resonance peak shifted by approximately 12 MHz from the main resonance peak, which is assigned as the \(61D_{3/2}\). As the fine structure splitting of the \(61D\) state is approximately 50 MHz, this small peak is not the \(61D_{1/2}\) state. Moreover, there is no corresponding hyperfine splitting of the intermediate \(5P_{3/2}\) state. Thus, this small peak has not been assigned to any transition so far. There are also small peaks near other \(nS\) and \(nD\) states. The shifting frequency from the main peak varies by transition states. As this non-assigned small resonance peak is overlapped with the main peak, we determined the center frequency of the main peak by fitting the observed signal with two Lorentzian curves. The determined center frequencies with the two-curve fitting
were shifted by up to 500 kHz from those with a simple one-curve fitting. The measured transition frequencies between the $5P_{3/2}$ $F = 3$ and $nS$ or $nD$ Rydberg states are summarized in Table I. The center frequency of the EIT signal is slightly shifted owing to the Doppler mismatch effect\cite{18,22} if the probe laser frequency is detuned ($\Delta \nu_p$) from the intermediate state $5P_{3/2}$ $F = 3$, and this shift ($\Delta \nu_c$) is given by

$$\Delta \nu_c = -\nu_p/\nu_c \Delta \nu_p,$$

where $\nu_p$ and $\nu_c$ are the probe laser and the coupling laser frequencies, respectively. Figure 5 shows the Doppler mismatch effect within a linewidth frequency of the intermediate state. The absolute transition frequency of $^{87}$Rb from $5P_{3/2}$ to the intermediate state $5S_{1/2}$ $F = 2$ to $5P_{3/2}$ $F = 3$ has been precisely measured and reported to be $384.228 \pm 15$ THz.\cite{27} We measured the absolute frequencies of the probe laser and corrected the measured frequencies between the $5P_{3/2}$ $F = 3$ and $nS$ or $nD$ Rydberg states using Eq. (1). To evaluate the performance of our optical synthesizer system, we compare the measured frequencies with the theoretical ones calculated using the reported quantum defects.\cite{21} We also compare the presently measured frequencies with the reported frequencies.\cite{22} All newly measured frequencies are in good agreement with the calculated ones and also the previously measured ones within approximately 1 MHz. For the $58D_{3/2}$ excited state, the absolute transition frequency was independently measured before using an optical frequency comb system based on a mode-locked Ti:sapphire laser (GigaOpticas GigaJet-20). The new frequency for the $58D_{3/2}$ transition is also in good agreement with the previously measured one within 500 kHz. As a result, the frequency accuracy of the present optical frequency synthesizer is confirmed within 500 kHz. Additionally, we have confirmed that the transition frequencies for higher Rydberg states ($n > 58$) can be well predicted within approximately 1 MHz from the theoretical calculation using the reported quantum defects and ionization energy.\cite{22} The total frequency uncertainty in the present experiment condition is evaluated using the frequency uncertainty of the probe laser at 780 nm and the fitted center line, as shown in Table I. The frequency uncertainty of the coupling laser at 480 nm is neglected because this laser is locked to a tooth of the comb. In the present experiment, for experimental simplicity, we locked the probe laser to the absorption line of Rb instead of the frequency comb line. As the frequency drift of the probe

| Rydberg state | Transition frequencies (THz) | Difference (kHz) |
|---------------|-------------------------------|-----------------|
|               | This work | Expt.\cite{22} | Calc.\cite{22} | This work | Expt.\cite{22} | Calc.\cite{22} |
| $53D_{3/2}$   | 624.561 122 20(6) | 624.561 121 2(8) | 624.561 121 93 | 1000 | 270 |
| $58D_{3/2}$   | 624.769 175 78(8) | — | 624.769 175 86 | — | — | 80 |
| $59D_{3/2}$   | 624.804 427 02(4) | — | 624.804 426 71 | — | 310 |
| $60D_{3/2}$   | 624.837 889 95(5) | — | 624.837 889 95 | — | — | 0 |
| $61D_{3/2}$   | 624.869 684 80(2) | — | 624.869 684 43 | — | 360 |
| $72D_{3/2}$   | 625.135 158 78(6) | — | 625.135 157 69 | — | — | 1100 |
| $80D_{3/2}$   | 625.262 410 77(12) | — | 625.262 409 43 | — | 1340 |
| $85D_{3/2}$   | 625.324 084 06(14) | — | 625.324 082 90 | — | 1170 |
| $92D_{3/2}$   | 625.393 885 18(11) | — | 625.393 885 19 | — | — | 10 |
| $53S_{3/2}$   | 624.561 199 65(6) | 624.561 199 5(8) | 624.561 199 67 | 150 | — | — | 20 |
| $58S_{3/2}$   | 624.769 234 16(8) | — | 624.769 234 81 | — | — | 650 |
| $59S_{3/2}$   | 624.804 482 68(4) | — | 624.804 482 65 | — | 30 |
| $60S_{3/2}$   | 624.837 942 61(5) | — | 624.837 943 08 | — | — | 470 |
| $61S_{3/2}$   | 624.869 735 15(2) | — | 624.869 734 94 | — | 210 |
| $72S_{3/2}$   | 625.135 189 02(6) | — | 625.135 188 11 | — | 910 |
| $80S_{3/2}$   | 625.262 432 03(10) | — | 625.262 431 48 | — | 550 |
| $85S_{3/2}$   | 625.324 102 13(10) | — | 625.324 101 23 | — | 900 |
| $92S_{3/2}$   | 625.393 899 62(16) | — | 625.393 899 59 | — | 30 |
| $60D_{1/2}$   | 624.776 971 36(8) | 624.776 972 08(8) | 624.776 971 86 | — | 640 | — | 500 |
| $61D_{1/2}$   | 624.811 824 17(12) | 624.811 823 4(8) | 624.811 825 05 | 770 | 880 |
| $72D_{1/2}$   | 625.100 585 66(16) | — | 625.100 585 44 | — | 170 |
| $80D_{1/2}$   | 625.237 449 13(25) | — | 625.237 449 46 | — | — | 330 |
| $90D_{1/2}$   | 625.358 254 05(7) | — | 625.358 256 78 | — | — | 2730 |

![Fig. 5.](image-url) Center frequency shift between $5P_{3/2}$ $F = 3$ and $61D_{3/2}$ state due to the probe laser detuning $\Delta \nu_p$ from the intermediate state $5P_{3/2}$ $F = 3$. The frequency shift is given by the difference between the measured frequency and the calculated one.\cite{22} The dotted line represents the Doppler mismatch curve with $-\nu_p/\nu_c \Delta \nu_p \sim 1.6 \times \nu_p$.}
laser during the frequency measurement was measured to be less than 50 kHz, the corresponding frequency error in the EIT resonance peak was estimated to be 80 kHz owing to the Doppler mismatch. The uncertainty of each fitted center line became worse from 10 to 230 kHz by increasing the principal quantum number because of the decreasing SNR of the spectra. Thus, the total frequency uncertainty is estimated to be from 20 to 250 kHz. However, we also observed the dependence of frequency shift on probe power, as shown in Fig. 6. The frequency shifts of different detuning probe laser show similar curves after correcting the Doppler mismatch. A slope of the frequency shift changes at near 80 nW. This is caused by a spectral shape broadened asymmetically owing to the power broadening of unknown states and the determined state. Under our typical experimental condition, the probe power shift is less than 500 kHz. To improve the accuracy of transition frequencies under a pure condition, frequency shifts including the AC Stark shift, power broadening, and Zeeman shift should be carefully evaluated for each transition. As the main purpose of the present experiment is to characterize the frequency accuracy of the optical frequency synthesizer system, we did not go further in the evaluation of the AC Stark shift and other shifts. In future experiments, we will carefully evaluate these level shifts to reduce the total frequency uncertainty of our measurement of the Rydberg states.

5. Conclusions
We have developed an optical frequency synthesizer based on an Er fiber-based optical frequency comb with a repetition rate at 325 MHz, which can generate an arbitrary optical frequency around 480 nm with an uncertainty of less than 100 kHz, and demonstrated the high-resolution two-photon spectroscopy of Rb by using the optical synthesizer. To the best of our knowledge, we could determine the absolute transition frequencies of $^{87}$Rb atoms to $nS$ and $nD$ states ($n > 60$) with an uncertainty of less than 250 kHz from the direct frequency measurements with the optical frequency comb. We also observed the light shift of the transition frequencies owing to the variation of the probe laser power. The measured results are in good agreement with the calculated frequencies from previous data within approximately 1 MHz. Thus, the transition frequencies of Rydberg states can be well predicted within approximately 1 MHz from the theoretical calculation based on the reported quantum defects and ionization energy. In future experiments, we will employ the present optical frequency synthesizer for our quantum simulation experiment using the cold Rydberg atoms in the optical trap.

Acknowledgments
We would like to thank Zhigang Zhang of Peking University for technical advice and valuable suggestions about the high-repetition-rate Er fiber mode-locked laser, and Mark Sadgrove of Tohoku University for useful discussion. This work was supported by the Photon Frontier Network Program (MEXT).

1) Th. Udem, R. Holzwarth, and T. W. Hänsch, Nature 416, 233 (2002).
2) J. D. Jost, J. L. Hall, and J. Ye, Opt. Express 10, 515 (2002).
3) B. R. Washburn, R. W. Fox, N. R. Newbury, J. W. Nicholson, K. Feder, P. S. Westbrook, and C. G. Jørgensen, Opt. Express 12, 4999 (2004).
4) A. K. Mills, Y.-F. Chen, K. W. Madison, and D. J. Jones, J. Opt. Soc. Am. B 26, 1276 (2009).
5) T. Fordell, A. E. Wallin, T. Lindvall, M. Vainio, and M. Merimaa, Appl. Opt. 53, 7476 (2014).
6) W. Gunton, M. Semczuk, and K. W. Madison, Opt. Lett. 40, 4372 (2015).
7) H. Liu, M. Yin, D. Kong, Q. Xu, S. Zhang, and H. Chang, Appl. Phys. Lett. 107, 151104 (2015).
8) A. D. Ludlow, M. M. Boyd, and J. Ye, Rev. Mod. Phys. 87, 637 (2015).
9) M. Saffman, T. G. Walker, and K. Molmer, Rev. Mod. Phys. 82, 2313 (2010).
10) P. Bohlouli-Zanjani, K. Afroushese, and J. D. D. Martin, Rev. Sci. Instrum. 77, 093105 (2006).
11) S. Uetake, K. Matsubara, H. Ito, K. Hayasaka, and M. Hosokawa, Appl. Phys. B 97, 413 (2009).
12) J. L. Morse, J. W. Sickler, J. Chen, F. X. Kärtner, and E. P. Ippen, Proc. CLEO, 2009, CML1.
13) J.-L. Peng, T.-A. Liu, and R.-H. Shu, Proc. IFCS/ETF9, 2009, p. 344.
14) H. Qi, J. Zhang, G. Zhou, A. Wang, and Z. Zhang. Chin. Opt. Lett. 11, 061402 (2013).
15) J. Zhang, F. Niu, X. Chen, X. Gao, Y. Liu, A. Wang, and Z. Zhang, Proc. CLEO, 2014, SW3E1.
16) J. Zhang, Z. Kong, Y. Liu, A. Wang, and Z. Zhang, Photonics Res. 4, 27 (2016).
17) W. Du, H. Xia, H. Li, C. Liu, P. Wang, and Y. Liu, Appl. Opt. 56, 2504 (2017).
18) A. K. Mohapatra, T. R. Jackson, and C. S. Adams, Phys. Rev. Lett. 98, 113003 (2007).
19) R. P. Abel, A. K. Mohapatra, M. G. Bason, J. D. Pritchard, K. J. Weatherill, U. Raitzsch, and C. S. Adams, Phys. Rev. Lett. 94, 073107 (2009).
20) B. Sanguinetti, H. O. Majeed, M. L. Jones, and B. T. H. Varcoe, J. Phys. B 42, 165004 (2009).
21) L. A. M. Johnson, H. O. Majeed, B. Sanguinetti, T. Becker, and B. T. H. Varcoe, New J. Phys. 12, 063028 (2010).
22) M. Mack, F. Karlewski, H. Hattermann, S. Hoch, F. Jessen, D. Cano, and J. Fortagh, Phys. Rev. A 83, 052511 (2011).
23) L. A. M. Johnson, H. O. Majeed, and B. T. H. Varcoe, Appl. Phys. B 106, 257 (2012).
24) H. Tamura, T. Unakami, I. H. Ye, H. Miyamoto, and K. Nakagawa, Opt. Express 24, 18132 (2016).
25) J.-L. Peng, H. Ahn, R.-H. Shu, H.-C. Chui, and J. W. Nicholson, Appl. Phys. B 86, 49 (2007).
26) R. W. P. Drever, J. L. Hall, F. V. Kowalski, J. Hough, G. M. Ford, A. J. Munley, and H. Ward, Appl. Phys. B 31, 97 (1983).
27) J. Ye, S. Swartz, P. Jungner, and J. L. Hall, Opt. Lett. 21, 1280 (1996).