Stabilizing diode laser to 1 Hz-level Allan deviation with atomic spectroscopy for Rb four-level active optical frequency standard

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Abstract

We achieve a compact ultra-stable 420 nm blue diode laser system by immediately stabilizing the laser on the hyperfine transition line of Rb atom. The Allan deviation of the residual error signal reaches 1 Hz-level Allan deviation within 1 s averaging time, and the fractional frequency Allan deviation is $1.4 \times 10^{-15} / \sqrt{\tau}$, which shows the best result of frequency-stabilized lasers based on the atomic spectroscopy without Pound–Drever–Hall (PDH) system. The signal-to-noise ratio of the atomic spectroscopy is evaluated to be 3,000,000 from the Allan deviation formula, which is the highest record, to the best of our knowledge. The frequency noise suppression characterization is demonstrated and the maximal noise suppression can be near 40 dB at 6 Hz. As a good candidate of pumping source, the ultra-stable 420 nm diode laser is successfully used in our Rb four-level active optical frequency standard system. The method can be easily extended to other wavelengths ultra-stable lasers with a Allan deviation of 10^{-15} level retaining an atomic reference with low cost and low complexity while in the absence of an expensive and complicated PDH system.

1 Introduction

Ultra-stable laser, in most cases, is a crucial factor that determines the final performance in atomic molecular optical (AMO) physics [1–3]. Especially, in these fields such as optical frequency standard [4–6], high-resolution optical sensing, coherent optical communications [7, 8], and fundamental physics [9, 10], a compact ultra-stable laser source is extremely demanding. Actually, a high-stability laser for 0.1–10 s averaging time can be achieved by locking the laser frequency to a super cavity by employing the well known Pound–Drever–Hall (PDH) technique [11]. This technique has been extensively applied in achieving narrow-linewidth and high-stability laser since it was first introduced. Several latest results on PDH frequency stabilization are reported [12–14], showing that the narrowest laser linewidth can be sub-10 mHz with the help of cryogenic silicon single-crystal optical cavity. We know that the fast frequency noise of lasers can be suppressed by this technique, but the slow

frequency noise is hard to be reduced due to the slow temperature drift of the cavity. Additionally, the whole experimental setup is very large and expensive as it needs rigorous control of cavity's temperature and vacuum degree. Moreover, this kind of PDH system cannot be easily transported from one lab to another; thus, it is suitable for staying in one place.

Nevertheless, atoms or molecules can provide the absolute quantum reference and the Allan deviation of stabilization on atoms or molecules satisfies the rule of $\tau^{-1/2}$. Furthermore, the experimental scheme of stabilization on atoms or molecules is more compact and insensitive to the environment compared with the PDH stabilization scheme. Saturated absorption spectroscopy (SAS) [15] and polarization spectroscopy (PS) [16] methods are very commonly used in the laser frequency stabilization for their compact structures. Numerous papers on SAS and PS have been published in the last decades [17–19]. It is convenient to stabilize the laser frequency by SAS and PS, but it is difficult to achieve 10^{-14} or better stability due to their common residual Doppler background of the obtained error signal. However, modulation transfer spectroscopy (MTS) [20–22], which is insensitive to the background absorption and directly provides a dispersion-like signal with a zero-crossing at line center, is an excellent method for achieving a high-stability stable laser. For example, there is an experimental paper to

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prove that the MTS method is better than polarization spectroscopy method [23]. Recently, Moritz Mihm et al. present a technique for simultaneous laser frequency stabilization on transitions of multiple atomic species by MTS method with a single optical setup [24]. Several groups have achieved the compact I_2 molecular optical frequency standard with a stability of 10^{-15} level using the MTS method [25–32]. In practical application, iodine frequency reference based on modulation transfer spectroscopy on a sounding rocket was reported [33]. Frequency-stabilized lasers working on Rb [34], K [35], He [36] based on MTS method have also been reported. In particular, with more and more theoretical and experimental studies on Rb 420 nm $5S_{1/2}$ to $6P_{3/2}$ transition [37–42], it is well known that the natural linewidth of Rb 420 nm transition is narrow. Therefore, frequency-stabilized laser working on Rb 420 nm transition based on MTS method is also a promising alternative for compact frequency standard for various applications [43].

In this paper, based on our preliminary experiment with a Allan deviation of $1.2 \times 10^{-14}/\sqrt{\tau}$ [43], a Rb-referenced 420 nm blue diode laser system integrated in a 15-l box with a Allan deviation of $1.4 \times 10^{-15}/\sqrt{\tau}$ and a signal-to-noise ratio of 3,000,000 is achieved by optimizing the frequencies and gains of the servo loop circuit and the performance of diode laser. The laser source employed here is a 420 nm interference filter external cavity diode laser (ECDL) of which the Lorentz-fit linewidth is less than 30 kHz and the tuning range is more than 15 GHz [44]. By feeding the error signal back to three different frequency response ports: slow PZT port, slow diode current port, fast diode current port, the frequency noise of the 420 nm laser ranging from low frequency to high frequency is suppressed effectively. The Allan deviation of the residual error signal is $1.4 \times 10^{-15}/\sqrt{\tau}$ within 0.3 s averaging time, showing a great superiority among all the compact ultra-stable lasers in the absence of expansive and complicated PDH locking. Compared with 532 nm I_2 -stabilized YAG laser [25–32], the 420 nm Rb-referenced diode laser occupies more advantages like that the Rb cell can be made smaller for its better signal-to-noise ratio and the diode laser has more extensive application in the AMO physics field compared with YAG laser. The ultra-stable diode laser is a good candidate of the pumping source for the Rb four-level active optical frequency standard. Furthermore, it is expected to be recommended as an absolute wavelength standard, and the method can be applicable for other wavelengths, such as the absolute quantum frequency reference of Cs atom 459 nm $6S_{1/2}$ to $7P_{1/2}$ transition, which is applied in the Cs four-level active optical clock [45–47].

2 Experiment setup

2.1 Ultra-stable laser system

A photograph and a schematic of the experimental setup payload of blue diode laser stabilized on Rb 420 nm transition line are shown in Figs. 1 and 2, respectively. See Fig. 1a. The output light passes through an isolator, laser beam expander, and a combination of a half-waveplate ($\lambda/2$) and a polarization beam splitter (PBS1). Afterwards, it is divided

Fig. 1 **a** Diagram of the experimental setup of blue diode laser stabilized on Rb 420 nm transition line. The black dotted frame is the ultra-stable laser system. ECDL external cavity diode laser, ISO isolator, Exp expander, $\lambda/2$ half-waveplate, PBS polarization beam splitter, EOM electro-optic modulator, SIG signal source, M mirror, PD photodetector, PID proportional integral derivative controller, Low-pass low-pass filter, T controller temperature controller. The red dotted frame is the 1367 nm Rb active optical frequency standard system. **b** The related Rb atomic energy level

Fig. 2 **a** A photograph of the experimental setup payload of blue diode laser stabilized on Rb 420 nm transition line. **b** The inner structure of the interference filter ECDL

into two beams with proper light power ratio by HWP and PBS2. One light beam is used as a pump laser and the other is used as a probe laser. The pump one is phase modulated by an electro-optic modulator (EOM) at the modulation frequency of 1.3 MHz and interacts with Rb atoms in a vapor cell restored in a magnetic shielding. The vapor cell is a natural Rb cell (^{85}Rb , 78%, and ^{87}Rb , 22%). An HWP before the EOM is used to minimize residual amplitude modulation (RAM) [48] of the EOM by adjusting the light polarization to match the main axial of the crystal. The temperature of Rb cell is controlled precisely by a temperature controller, the temperature control accuracy is within 0.01°C . The counter-propagate probe laser passes through the Rb cell and is focused to the photodetector (PD1). The beat note between the light carrier and modulation sidebands is detected by PD1. The detected signal is filtered by a low-pass filter and amplified and then mixed with a sinusoidal signal having the same frequency as the driving signal of EOM. The output signal from the mixer is used as an error signal. It is sent to the PID circuit to control the PZT and current of the diode laser.

The ultra-stable 420 nm laser can be used as a pumping source for Rb four-level active optical frequency standard, which pumps the Rb atoms from $5S_{1/2}$ state to $6P_{3/2}$ state. The atoms will decay to $6S_{1/2}$ state and it will form the population inversion between $6S_{1/2}$ state and $5P_{1/2}$ state. Under the effect of a cavity, the 1367 nm active optical frequency standard signal will be detected by PD2. The related Rb atomic energy level is shown in Fig. 1b.

See Fig. 2a. The volume of the whole system is about 15 l. See Fig. 2b. The laser is an interference filter ECDL with a linewidth of less than 30 kHz and a tuning range of more than 15 GHz. The tuning coefficient of piezovoltage we measured is 30 MHz/V and the response bandwidth is 800 Hz. While acting on the diode's injection current, the tuning coefficient is about 150 MHz/mA and the response bandwidth is about 3 kHz. We design a high-pass filter circuit directly connected to the laser diode which is used

to reduce the high-frequency noise. The cutoff frequency of the filter is about 20 kHz.

2.2 Narrow-linewidth 420 nm interference filter external cavity diode laser

The laser used in this setup is an interference filter external cavity diode laser (ECDL) which stands out for its robustness and good tunability. The external cavity with 7 cm length provides an intrinsic narrow fast laser linewidth of less than 30 kHz. The output laser power can be 20 mW at the working wavelength and the tunability is more than 15 GHz without mode hopping. The inner structure of the laser is presented in Fig. 2b. The light emitted from the diode is collimated by an objective lens with short focal length of 4.51 mm and high numerical aperture of 0.55. Then, the light transmits the interference filter with a pass band of 0.5 nm at a nominal wavelength of 420.3 nm and a lens focuses the light on the mirror with a reflection of 50%. A piezoelectric transducer (PZT) is attached to the mirror to fine tune the external cavity length. There is a second lens used to collimate the laser beam. The temperature of the laser diode is controlled by a thermoelectric cooler (TEC) and works at room temperature. All the optical elements set on an invar pedestal are stored in a box made of aluminum.

2.3 Active optical frequency standard

The active optical frequency standard was first introduced in 2005 [49, 50]. The outstanding superiority of active optical frequency standard is that it can break through the limitation of atomic natural linewidth in passive optical frequency standards and can achieve mHz-level-linewidth laser output. Afterwards, a variety of world-class groups have started to investigate the active optical frequency standards [51–56]. The experimental scheme of Rb four-level active optical frequency standard is shown in Fig. 1a. The ultra-stable 420 nm laser is used as a pump source, which pumps Rb atoms from $5S_{1/2}$ state to $6P_{3/2}$ state. The atoms will decay to the $6S_{1/2}$ state from $6P_{3/2}$ state, and the population inversion between $6S_{1/2}$ state and $5P_{3/2}$ state will occur owing to the longer lifetime of the $6S_{1/2}$ state [37–39, 57]. Under the effect of the bad cavity [50], the 1367 nm active optical signal output will be realized. The right cavity mirror is a plane convex lens, which is anti-reflective for 420 nm pump laser and all reflective for 1367 nm laser. The left cavity mirror is a plane mirror, which is all reflective for 420 nm pump laser and partial reflective for 1367 nm laser. The maximal output power of 1367 nm active optical signal can reach about 200 μW when the 420 nm pumping laser is locked.

3 Results

3.1 Rb atomic spectroscopy

To diagnose the properties of the diode laser, we use it to perform SAS on the $5S_{1/2} \rightarrow 6P_{3/2}$ transition of Rb atom. The length and diameter of the Rb cell are 5 cm and 2 cm, respectively. The powers of pump and probe laser are 1 mW and 0.2 mW, respectively. The Rb cell works at 165°C. When a triangular wave signal of 800 mV and 10 Hz is added to the PZT port of the diode laser, we can obviously observe four groups of hyperfine transitions of ^{87}Rb and ^{85}Rb as shown in Fig. 3. From left to right, the SAS signals are corresponding to hyperfine transitions from ^{87}Rb , $F = 1$, ^{85}Rb , $F = 2$, ^{85}Rb , $F = 3$ and ^{87}Rb , $F = 2$. Their details of hyperfine transition peaks are shown in four inserted diagrams. All the components including crossovers are observed. The FWHM of each peak can be indicated by the frequency interval of two hyperfine levels. The laser can be locked on any hyperfine transition. In this paper, we measure the FWHM of ^{85}Rb , $F = 3 \rightarrow F' = 4$ hyperfine transition signal which is 3.2 MHz by the frequency interval of 39.6 MHz between $F' = 4$ and $F' = 3$ sub-levels, and lock the laser on ^{85}Rb , $F = 3 \rightarrow F' = 4$ hyperfine transition by MTS method as the signal-to-noise ratio of this closed transition is better.

3.2 Frequency noise power spectral density

To check the locking performance, the frequency noise of the MTS signal of ^{85}Rb , $F = 3 \rightarrow F' = 4$ hyperfine transition at Fourier frequencies up to 1 MHz in three different cases of free running, locking with intermediate gain and locking

with optimal gain is measured. The frequency fluctuation of MTS signal and the responding frequency noise power spectral density (PSD) in different cases are shown in Figs. 4 and 5. From Fig. 4, we can see that the frequency fluctuation when the laser is free running is about 2 MHz. While locking with an intermediate gain, the frequency fluctuation range is about several hundreds of Hz and it is only several tens of Hz under the optimal gain condition. Similarly, the frequency noise PSD (Hz^2/Hz) of the three conditions is shown in Fig. 5, which is converted from intensity noise PSD (dBm/Hz) [58] by taking into account the slope of MTS signal. It can be seen that the frequency noise is effectively

Fig. 4 Locking performance of the laser system. Comparison of frequency fluctuation of MTS signal in free running (top), locking with intermediate gain (middle) and optimal gain cases (bottom)

Fig. 3 Saturated absorption spectroscopy on the $5S_{1/2} \rightarrow 6P_{3/2}$ transition of Rb atom. It shows the SAS signals of ^{87}Rb , $F = 1$, ^{85}Rb , $F = 2$, ^{85}Rb , $F = 3$ and ^{87}Rb , $F = 2$ from left to right. The insert diagrams are their respective details

Fig. 5 Frequency noise PSD of MTS signal. Comparison of frequency fluctuation of MTS signal in free running (top), locking with intermediate gain (middle) and optimal gain cases (bottom). The blue straight line is β -separation line

suppressed within 6 kHz Fourier frequency when the laser is locked. The frequency noise at 10 Hz can be suppressed by near 40 dB. As seen in Fig. 5, when the laser is locked with optimal gain, the bump at 6 kHz is obviously suppressed near 10 dB, the frequency noise amplitude of the laser is further reduced and the locking bandwidth is extended to about 150 kHz compared to that with intermediate gain. But there is still a bump at about 200 kHz, that means the locking bandwidth needs to improve further. Next step, we will increase the locking bandwidth to eliminate the two bumps. The peaks at 50 and 100 Hz arise from the industrial frequency noise. The β -separation line [59–61], defined as $S_{\beta\nu}(f) = 8f \ln 2 / \pi^2$, is also drawn.

3.3 Allan deviation

The frequency fluctuation of the residual error signal after locking is shown in Fig. 6a. The Allan deviation of the residual error signal is obtained as shown in Fig. 6b. The error signal is applied to slow PZT, slow current and fast current ports in order to correct for frequency deviations on both fast and slow time scales. The error signal from the PID circuit is proportional to the frequency fluctuation of the laser. In the case of only one frequency standard system, stability measured with a self-comparison is a common approach to evaluate the frequency standard in frequency metrology field that converts the voltage fluctuation of the residual error signal to the frequency fluctuation and measuring the Allan deviation of the frequency fluctuation [62, 63]. In this way, the Allan deviation result shorter than 1 second averaging

time is credible although the long-term result cannot truthfully reflect the performance of the system.

As shown in Fig. 6b, the obtained Allan deviation of fractional frequency reaches $1.4 \times 10^{-15} / \sqrt{\tau}$ within 0.3 s averaging time, which is trustworthy to some extent. It can be found that the Allan deviation result shorter than 1 s this work is better than that of 532 nm I_2 optical frequency standard. The Allan deviation of the residual error signal reaches about 1.4 Hz from 1 to 40 s averaging time, which shows a great superiority among all compact ultra-stable lasers without using an expensive and complicated PDH locking system. In our future work, we will build the second identical system to reevaluate the long-term frequency stability of the system by beat note between them. According to the formula of $\sigma_y(\tau) = \frac{\Delta\nu}{KS/N\nu_0} \frac{1}{\sqrt{\tau}}$, where K is a constant of the order unity, here K is simplified as 1, the signal-to-noise ratio of the system can be estimated to be about 3,000,000 under the condition of $\Delta\nu = 3$ MHz, $\nu_0 = 7.1 \times 10^{14}$ Hz and $\sigma_y(1s) = 1.4 \times 10^{-15}$, where $\Delta\nu$ is the linewidth of clock spectroscopy, $\sigma_y(\tau)$ is the Allan deviation. In the Rb active optical frequency standard system, the ultra-stable 420 nm laser is a good candidate of the pumping source and very helpful for the final frequency stability of the active optical frequency standard. Furthermore, the ultra-stable 420 nm laser system can be recommended as the absolute wavelength standard like I_2 -stabilized YAG laser because of its more compact and better short-term frequency stability characterization.

3.4 Pumping source for Rb active optical frequency standard

The active optical frequency standard was first introduced in 2005 [49, 50]. The outstanding superiority is that it can break through the limitation of atomic natural linewidth in passive optical frequency standards and can achieve mHz-level-linewidth laser output. The suppressed cavity pulling effect, that means, the output frequency does not follow the cavity mode changing exactly, but in a way of dramatically reduced frequency shift when the cavity mode is changing, is the main characteristic of the active optical frequency standard [45–47, 64–67]. Afterwards, a variety of world-class groups have started to investigate the active optical frequency standards [51–54].

Here, we use the ultra-stable 420 nm laser as a pumping source and first experimentally achieve the output of 1367 nm Rb four-level active optical frequency standard signal. The experimental scheme is shown in Fig. 1a. The 420 nm pumping laser pumps Rb atoms from $5S_{1/2}$ state to $6P_{3/2}$ state. The atoms will decay to the $6S_{1/2}$ state from $6P_{3/2}$ state, and the population inversion between $6S_{1/2}$ state and $5P_{3/2}$ state will occur owing to the longer lifetime of the

Fig. 6 **a** Frequency fluctuation of the residual error signal after locking. **b** Allan deviation of Rb-referenced 420 nm laser as a function of averaging time. The green solid line indicates that the fractional frequency stability is $1.4 \times 10^{-15} / \sqrt{\tau}$. The pink curve shows the 532 nm I_2 result [29] and the blue one shows the result in 2017 [30]

$6S_{1/2}$ state. Under the effect of the bad cavity, the 1367 nm stimulated emission will be realized. The cavity is composed of a plane mirror and a concave mirror with a radius of curvature of 50 cm. The concave mirror has a high reflectivity of nearly 99% for 1367 nm laser, and the plane mirror has a reflectivity of 80% for 1367 nm laser. The cavity length is 15 cm and the free spectral range is 1 GHz. A 5-cm long Rb cell with diameter of 19 mm is put in the cavity and it works at 115°C temperature. The requirement for the cell windows flatness is high. Optical loss impairs the transmitted power and hence increases the threshold of the Rb active optical frequency standard. So to achieve 1367 nm output signal, the optical loss must be as low as possible. The optical loss of the cell windows can be reduced by improving the window flatness and optical coating of the end windows. In this system, the Rb cell (TRIAD TECHNOLOGY, TT-RB87-19×50-Q-BA) window is wedge window instead of flat window, the window angle is 11°, the window flatness is $\lambda/4$, and the window thickness is 3 mm; the optical loss of the cell windows is about 20%.

As the first step of the experiment, we use the 420 nm frequency-stabilized diode laser with 4 mW as the pumping source, and achieve the 1367 nm active optical frequency standard signal output as shown in Fig. 7. The spot diameter of 420 nm laser beam is about 3 mm. The 1367 nm active optical frequency standard output signal corresponds to the saturated absorption peak of $^{87}\text{Rb } 5S_{1/2}, F = 2 \rightarrow 6P_{3/2}, F' = 3$ hyperfine transition. When the pumping laser is stabilized on the hyperfine transition line of $5S_{1/2} F = 2 \rightarrow 6P_{3/2} F' = 3$ of ^{87}Rb , the relationship between the 1367 nm active optical frequency standard output signal and the 420 nm pumping power is shown in Fig. 8. It can be seen that the power threshold of 420 nm pumping laser is 0.5 mW. The power of 1367 nm active optical output signal increases with the increasing 420 nm pumping power.

Fig. 7 1367 nm active optical frequency standard signal (red curve) and 420 nm $^{87}\text{Rb } 5S_{1/2}, F = 2$ saturated absorption spectroscopy (blue curve)

Fig. 8 1367 nm active optical frequency standard signal output power while changing 420 nm pumping power when the cell works at 115°C

The ultra-stable pumping source will be theoretically benefit to the performance of the 1367 nm active optical frequency standard signal. In the scheme of four-level active optical frequency standard, the performance (including frequency stability) of pumping source will affect the characterization of active optical frequency standard signal (linewidth and frequency stability). Therefore, for achieving the high-performance 1367 nm Rb four-level active optical frequency standard, it is necessary to improve the frequency stability of 420 nm laser at first. Limited by the only one available pumping laser, the comparison of the active optical signal performance induced by the pumping source with different frequency stabilities cannot be achieved at present. In the future work, we will build the second system and make the comparison.

4 Discussion

In conclusion, compared with previous system, a compact ultra-stable Rb-referenced 420 nm diode laser system with some improvements is achieved. Without the temperature control and vibration isolation of the whole experimental system, first, it shows a sufficiently good short-term performance with a Allan deviation of 1 Hz level within 1 s averaging time, and a fractional frequency Allan deviation of $1.4 \times 10^{-15} / \sqrt{\tau}$ by increasing the locking bandwidth, which shows the best result of frequency-stabilized lasers based on the atomic spectroscopy without PDH system. Second, the signal-to-noise ratio is improved to be about 3,000,000 by increasing the diameter of the probe and pump beams, which is extraordinarily impressive results among all the reports in the absence of a PDH stabilization system, to the best of our knowledge. The frequency noise characterization of the diode laser is also demonstrated and the maximal

noise suppression can be near 40 dB at 6 Hz. In addition, we have successfully used the ultra-stable 420 nm laser as a pumping source for the Rb four-level active optical frequency standard system, and first experimentally obtained the 1367 nm active optical signal output. The method can be easily extended to other wavelengths ultra-stable lasers with absolute quantum frequency reference in the absence of PDH system.

The performance of the ultra-stable laser, we believe, can be pushed forward. First, the spot diameter of the diode laser can be increased from 3 mm currently used to 5 mm or larger, thus improving the signal-to-noise ratio of the atomic spectroscopy and narrowing the linewidth limited by the transit time. Second, the wider locking bandwidth of the whole system enables the reduction of high frequency noise of the laser. By this way, the short-term stability will be polished up by several factors. Third, the laser power fluctuation also causes changes in the laser frequency. Therefore, the long-term frequency stability will be put forward to a better level by adding the power stabilization. Fourth, within the temperature control and vibration isolation, the system's frequency stability with a further improvement is expected. Last, the whole system's size will be reduced to 3 l by the integration and engineering technology.

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