Precision Frequency Measurement of ^{87}Rb $5S_{1/2}$ $(F = 2)\rightarrow 5D_{5/2}$ $(F'' = 4)$ Two-Photon Transition through a Fiber-Based Optical Frequency Comb *

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The absolute frequency of ^{87}Rb $5S_{1/2}$ $(F = 2)\rightarrow 5D_{5/2}$ $(F'' = 4)$ two-photon transition at 778 nm is measured in an accuracy of 44 kHz. A home-made erbium-doped fiber laser frequency comb with frequency stability of 5.0×10^{-13}@1 s is employed for the light source. By using a periodically poled lithium niobate, the femtosecond pulse operating in 1556 nm is frequency-doubled to 778 nm to obtain the direct two-photon transition spectroscopy of thermal rubidium vapor. Through sweeping the carrier envelope offset frequency ($f_{\rm ceo}$), the $5S_{1/2}$ $(F = 2)\rightarrow 5D_{5/2}$ $(F'' = 4)$ two-photon transition line is clearly resolved and its absolute frequency is determined via the peak-finding of the fitting curve. After the frequency correction, the measured result agrees well with the previous experiment on this transition. The entire system configuration is compact and robust, providing a potential candidate of optical frequency standard for telecommunication applications.

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Two-photon excitation spectroscopy is now a well-established technique for studying high-energy levels of atoms and molecules based on the nature that the Doppler shift caused by the velocity of an atom can be compensated by an opposite shift from a counter-propagating beam.[1–4] As a result, atoms of all velocities are able to contribute to the Doppler-free signals even without the use of laser cooling and trapping techniques.[5] As an example, the Rb $5S$–$5D$ two-photon transitions are of great importance. Owing to the high vapor pressure of the atomic rubidium and the narrow line width (∼660 kHz) of the $5D$ state, these two-photon transitions appear to be a low-cost frequency standard with good short-term stability in the visible domain (∼778 nm), which was recommended for the realization of the metric length unit meter by the Comité International des Poids et Mesures.[6]

In early studies, the Rb two-photon transition has been extensively investigated by using continuous wave[7] and mode-locked lasers.[8] Since the invention of the femtosecond frequency comb at the beginning of this century, there has been a giant leap forward in this field. An optical frequency comb permits a precise connection between the microwave and optical frequencies, thus it serves as a frequency ruler to simply measure the absolute frequency of the laser locked to these transitions.[9] Due to the spectral versatility, the direct frequency-comb spectroscopy (DFCS) has been proposed to perform simultaneous precision measurement of multiple Rb two-photon transitions with a single experimental system.[10,11] In this scheme, pairs of modes from the laser frequency comb combine to the same total energy, thus all the frequency components contribute to a large two-photon transition amplitude. In the following decade, studies on the improvement of the signal-to-noise ratio (SNR) of the direct two-photon spectroscopy were carried out. Dudovich *et al.* demonstrated the enhanced two-photon transition signal beyond the level by a transform limited pulse by controlling over the spectral phase of the pulse.[12] The polarization pulse shaping method was proposed to control the angular distribution of the final state with high SNR.[13] In 2013, Barmes *et al.* demonstrated a simple and flexible split-pulse technique to eliminate the Doppler background of the two-photon signal.[14] Actually, most of these researches studies have been performed through Ti:sapphire femtosecond optical frequency combs. Since fiber lasers offer several clear advantages over solid-state systems like compact design, thermal management, minimal alignment and long-term stability, the fiber-based optical frequency comb has become a valuable option for Rb two-photon transition spectroscopy. In 2013, Wu *et al.* first reported the erbium fiber laser-based direct frequency comb spectroscopy of Rb two-photon transitions.[15] Subsequently, they built an optical comb frequency standard based on these transitions.[16] Although the

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accuracy of several kilohertz has been achieved, the employment of frequency modulation and an acousto-optical modulator leads to high complexity of the measurement system.

In this Letter, we demonstrate an alternative method to determine the absolute frequency for Rb two-photon transition. The relatively simple experimental configuration uses a frequency-doubled self-referenced Er-fiber optical frequency comb to excite the thermal rubidium atoms from the ground state to the $5D_{5/2}$ state. By sweeping the carrier-envelope offset frequency, direct two-photon spectroscopy with high signal-to-noise ratio can be obtained, allowing us to determine precisely the absolute frequency, as well as the light shift and the pressure shift of the transition. Our results are consistent with the previous measurements, paving the way for establishing a Rb-based optical frequency comb working at telecommunication band with high precision and stability.

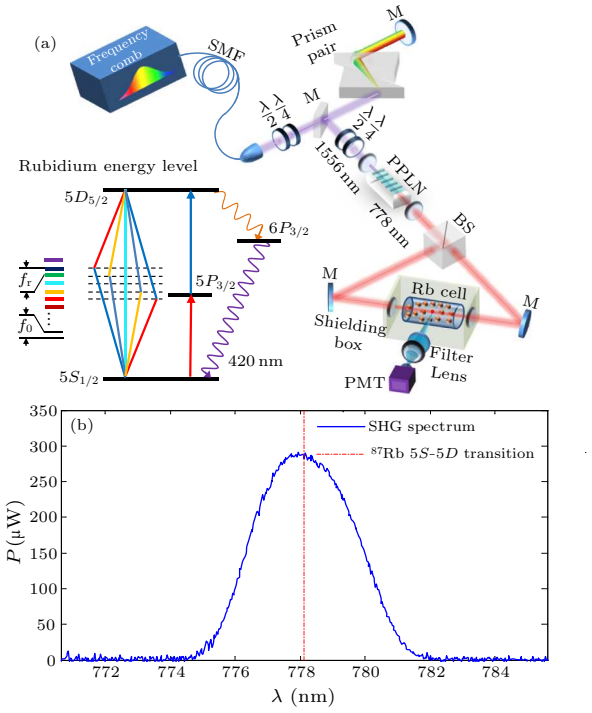

Fig. 1. The $5S$–$5D$ two-photon spectroscopy in thermal rubidium vapor by using the fiber-based optical frequency comb: (a) experimental setup. SMF, single-mode fiber; PPLN, periodically poled lithium niobate; BS, beam splitter; M, mirror. (b) Spectrum of the frequency-doubled laser frequency comb at 778 nm.

The diagram illustrating the basic experimental apparatus is presented in Fig. 1(a), which also contains the low-lying energy levels for atomic ^{87}Rb. The excitation pathways from the ground level $5S_{1/2}$ to the $5D_{5/2}$ level may proceed through either direct two-photon excitation or a stepwise excitation via the $5P_{3/2}$ state. For the former case, atomic rubidium must simultaneously absorb two photons with frequencies ν_1 and ν_2 to be excited to the $5D_{5/2}$ state through a virtual level which is indicated by the horizontal broken line. The sum frequency of the photon pair should satisfy the two-photon resonance condition $\nu_1+\nu_2=\nu_t$, where the ν_t corresponds to the resonance frequency of the $5S_{1/2}\rightarrow 5D_{5/2}$ hyperfine transition. The latter case is a stepwise process with the lower transition to the $5P_{3/2}$ state, which is the well-known D2 line at 780 nm. Before the excited atom decays to the ground state, it can be further excited to the upper state $5D_{5/2}$ through absorbing another resonant photon at 776 nm.[17] In our present experiment, detection of atoms promoted to the $5D_{5/2}$ levels is made by monitoring the $5D_{5/2}$–$6P_{3/2}$–$5S_{1/2}$ cascaded fluorescence emission around 420 nm.

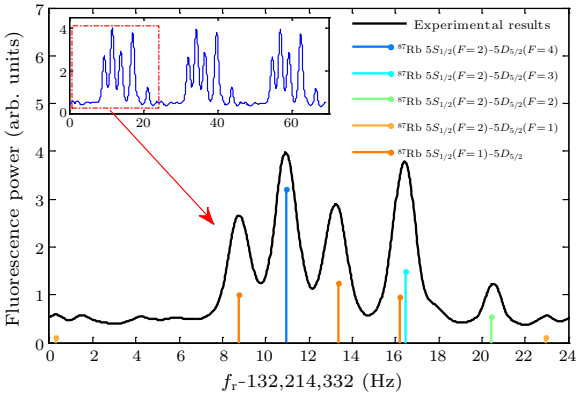

Fig. 2. Experimental fluorescence from the $6P\rightarrow 5S$ decay as a function of sweeping f_r. The vertical colored lines denote the theoretical relative probability and center positions of the two-photon transition lines.

An erbium-doped fiber laser frequency comb with central wavelength of 1556 nm and average output power of ~170 mW has been used as the light source. Firstly, the femtosecond pulse was coupled into free space through a collimator. After the polarization adjustment, a pair of silicon prisms was utilized to compress the pulse duration, thus a near transform-limited pulse (~80 fs) has been achieved. Then, the pulses were sent to a periodically poled lithium niobate (PPLN) for the second harmonic generation (SHG) with up to ~30 mW of output power. Through careful adjustment of the incident spot size and precision temperature control of the PPLN crystal, the frequency-doubled femtosecond pulses had a center wavelength of ~778 nm with the full width at half-maximum (FWHM) spectral width of about 4 nm, as shown in Fig. 1(b), covering the $5S_{1/2}$–$5D_{5/2}$ two-photon transitions of interest. The laser frequency comb was further split into two channels by using a 50:50 non-polarizing beam splitter. The counter-propagating beams were then focused by lenses with focal length of 25 cm to a spot size of ~400 μm inside the pure ^{87}Rb isotope vapor cell. Actually, focusing the beams to a small waist size will not only enhance the two-photon excitation rate, but also induce the broadening of the resonance

due to the finite transit-time of the atoms through the beams. The transit-time broadening for a waist of such size is about 67.3 kHz for room-temperature Rb atoms. Considering the Landé g factors between the S and the D states, a shielding box was used to zero out of the earth's magnetic field in the region of the vapor cell. Furthermore, the temperature of the cold finger of the Rb cell was varied by a thermoelectric cooler to evaluate the systematic shifts arising from Rb–Rb collision pressure shifts. The blue signal fluorescence was collected at a right angle to the counter-propagating laser beams by a short focal length lens ($f \sim 3$ cm) and spectrally separated from the scattered laser light and other fluorescence by a 420-nm filter with 10-nm bandwidth. The filtered fluorescence was detected by a blue-sensitive photomultiplier tube (PMT). The output from the detector was pre-amplified, filtered, and then sent to the digital acquisition board for analysis. The repetition rate $f_{\rm r}$ and the carrier envelope offset frequency $f_{\rm ceo}$ of our erbium-doped fiber frequency comb are about 132.210 MHz and 30 MHz, respectively. Both the parameters have been phase locked to a commercial hydrogen maser and can be swept precisely to produce the direct frequency comb two-photon transition spectrum of ^{87}Rb.

An experimental direct frequency comb spectroscopy of ^{87}Rb $5S_{1/2}$–$5D_{5/2}$ two-photon transition acquired with a frequency scan of repetition rate $f_{\rm r}$ is shown in Fig. 2. In the experiment, the carrier envelope offset frequency was stabilized to 35.791 MHz. The spectrum consists of several narrow Doppler-free peaks and a constant Doppler-broadened background. Actually, the narrow peaks are produced by the direct two-photon excitation process from the counter-propagating pulses. Regarding the 778 nm optical frequency comb, the optical frequency of the comb mode can be written as

$$\nu_N = N \cdot f_{\rm r} + 2f_{\rm ceo}, \tag{1}$$

where N is an integer mode number with a typical value of 10^6, and $f_{\rm ceo}$ has been doubled due to the fact that the 778 nm laser frequency comb is the second harmonic generation of the original frequency comb at 1.5 μm. If the sum frequency of the light from two counter-propagating comb lines labeled by N and M satisfies the condition of $\nu_{\rm t} = (N+M)f_{\rm r}+4f_{\rm ceo}$, there will be hundreds of thousands of comb pairs $(N+k, M-k)$ that possess the same sum frequency and thus could contribute to a sizable Doppler-free two-photon transition amplitude. By comparison, the number of comb modes involved in the stepwise 5S–5P–5D excitations is relatively small and its influence on the spectrum is hardly observed, resulting in the constant Doppler-broadened background.[18] As a result, it is appropriate to assume that no intermediate levels are populated in our case. According to the non-resonant absorption condition, the two-photon transition spectrum will repeat its pattern each time when the following equation is satisfied,

$$\begin{aligned}\nu_{\rm t} &= \nu_M + \nu_N = (M+N)\cdot f_{\rm r} + 4\cdot f_{\rm ceo} \\ &= (M+N-k)\cdot(f_{\rm r}+\Delta f_{\rm r}) + 4\cdot f_{\rm ceo},\end{aligned} \tag{2}$$

where k is an arbitrary integer, and $\Delta f_{\rm r}$ is the variation of the repetition rate. If we take literature values of the $5S_{1/2}$ $(F=2)$–$5D_{5/2}$ $(F''=4)$ two-photon transition to be 770, 569, 132, 748 kHz, we may figure out that the repeating pattern occurs at intervals of about 22.685 Hz, corresponding to a change in the optical frequency of the comb of 66.106 MHz (i.e., half the repetition rate).

Due to the periodicity of the direct two-photon transition spectroscopy through sweeping $f_{\rm r}$, some transitions are so close to adjacent ones that the fluorescence curve tends to be overlapped. To overcome this problem, we may perform the direct frequency comb spectroscopy of the ^{87}Rb two-photon transition through sweeping the carrier envelope offset frequency while keeping the repetition rate of pulse fixed. A scan over the $5S_{1/2}$ $(F=2)$–$5D_{5/2}$ $(F''=4)$ transition is achieved and the experimental result is presented in Fig. 3.

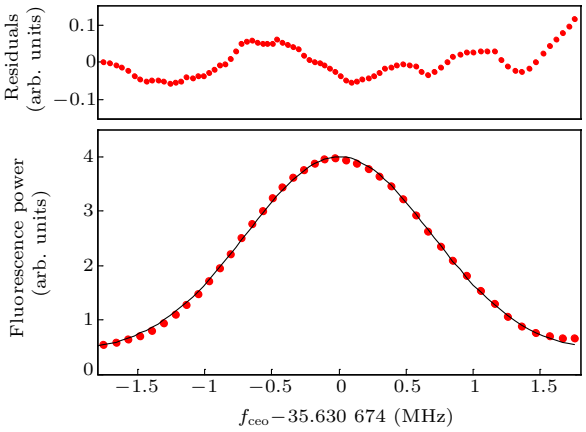

Fig. 3. The typical recording of the $5S_{1/2}$ $(F=2)$–$5D_{5/2}$ $(F''=4)$ transition as a function of the carrier envelope offset frequency. The excellent SNR allows determination of the line center to about 1:200 of the measured linewidth. This data was taken at a temperature of 149.8 K and was used in evaluating systematic effects related to the temperature dependence.

Such a kind of spectra shown in Fig. 3 can be understood and calculated by using standard two-photon transition probabilities derivable from the second-order time-dependent perturbation theory. Considering that there are a number of comb mode pairs contributing to the Doppler-free two-photon transition amplitude and assuming that no intermediate levels $(v_{\rm i})$ has been populated, the direct frequency comb two-photon transition spectrum can be

expressed as[14]

$$|a_{\rm f}^{(2)}|^2 \propto \sum_{n_1,n_2}\left\{\frac{I(\nu_{n_1})\cdot I(\nu_{n_2})}{(\nu_{n_1}-\nu_i)^2[(\nu_{\rm t}-\nu_{n_1}-\nu_{n_2})^2+(\gamma_{\rm f}/2)^2]}\right\} * \exp\left[-\left(\frac{c}{u}\right)^2\left(\frac{\nu_{\rm t}-\nu_{n_1}-\nu_{n_2}}{\nu_{n_1}-\nu_{n_2}}\right)^2\right], \quad (3)$$

where * means the evolution, n_1 and n_2 represent the indexes of the comb mode pair participating in the excitation with frequencies v_{n_1} and v_{n_2}, intensities $I(v_{n_1})$ and $I(v_{n_2})$; $v_{\rm t}$ and $v_{\rm i}$ refer to the transition frequency from the ground state to the final $5D_{5/2}$ level and the intermediate $5P_{3/2}$ level, respectively; $\gamma_{\rm f}$ denotes the homogeneous linewidth of the final state, and u is the most probable velocity of the ^{87}Rb atom. From Eq. (3), it can be seen that for each pair of modes, n_1 and n_2, and a given value of the repetition rate $f_{\rm r}$, there will be unique carrier envelope offset frequency $f_{\rm ceo}$ to obtain the maximum transition probability as

$$\nu_{\rm t}-\nu_{n_1}-\nu_{n_2}=\nu_{\rm t}-(n_1+n_2)\cdot f_{\rm r}-4\cdot f_{\rm ceo}=0. \quad (4)$$

As a result, it is possible to determine the line center through the curve fitting of the experimental data set, and the numerical calculations as well as the fit residuals are shown in Fig. 3. Due to the residual Doppler broadening effect, the measured transition linewidth is about 2 MHz. In our experiment, the signal strength of the measured transition above the Doppler broadening background is about 4 V while the shot noise of our detector and data acquisition system is less than 10 mV. The high quality SNR permits the determination of the transition frequency better than 1/200 of the measured linewidth.

Before determination of the absolute frequency of the transition, the possible systematic errors have to be investigated and corrected. To begin with, earth's magnetic field was cancelled by wrapping three layers of mu-metal sheet around the rubidium vapor cell, thus the magnetic (Zeeman) shift can be ignored. Moreover, considering the pulse energy of ~100 pJ with peak intensity of 100 W·cm^{-2}, the strong field effect such as multiphoton ionization can be negligible as well.

The light shift (the ac Stark shift) which scales linearly with the average power of the laser has been investigated by changing the average laser intensity, and the measured line center of the two-photon transition is shown in Fig. 4(a). The dashed line indicates the recommended value from CIPM.[19] The saturated vapor pressure is estimated to be 5.919 mTorr, and a linear relationship between the input intensity and the measured frequency can be obtained with a slope efficiency of about −1.828±1.1 kHz/mW.

The pressure shift, originating from the collision between the Rb atoms, plays the key role in the accurate determination of the transition frequency. An experimental measured pressure shift data is shown in Fig. 4(b) by varying the cold-finger temperature of the rubidium vapor cell. In this case, the average power of the laser frequency comb is fixed at 20 mW. The pressure shift is found to be −66.58 kHz/mTorr by extrapolation of the experimental data.

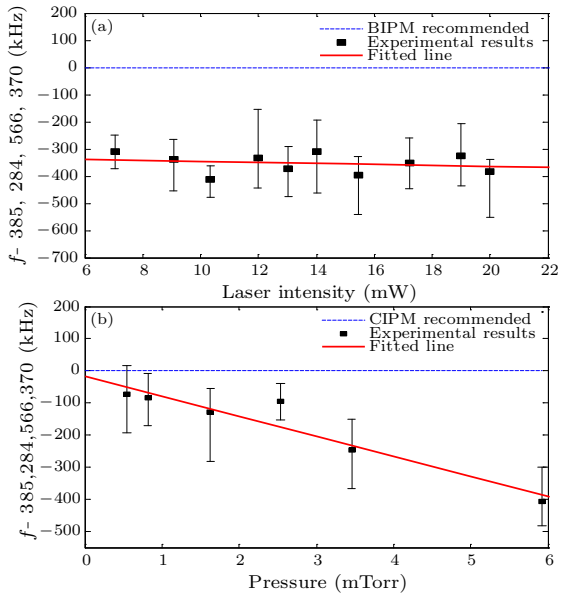

Fig. 4. Frequency shift of the $5S_{1/2}$ ($F=2$)–$5D_{5/2}$ ($F''=4$) two-photon transition versus: (a) the average intensity of the laser frequency comb; and (b) the pressure in the rubidium cell. The dashed line indicates the recommended value by CIPM.

According to the previous study on the blackbody-radiation-induced dynamic Stark shifts of energy levels of alkali atoms,[20] a T^4-dependent energy level shift leads to a shift of −665.8 Hz for the $5S_{1/2}$–$5D_{5/2}$ two-photon transitions at 385 THz. The line-shape distortion caused by the tails of neighboring transitions results in a frequency shift of the line center, which is evaluated to be 1.5 kHz owing to the Doppler background of neighboring transitions.

All these systematic shifts are summarized in Table 1, and the transition frequencies are corrected to be 385, 284, 566, 354 (44) kHz, which are of the same order of magnitude of the previous studies of this transition.[21]

Table 1. Summary of the systematic frequency shifts.

Effect	Shift at 778 nm
Light shift	−36.6±22 kHz
Pressure shift	−370.5±17 kHz
Blackbody radiation	−665.8±40 Hz
Neighboring transitions	±1.5 kHz
Electronics	±4 kHz
Total	−407.7±44 kHz

In summary, we have demonstrated the rubid-

ium $5S$–$5D$ two-photon absorption excited by an Er-doped fiber laser frequency comb operating at the optical communication bands. Through the second harmonic generation, a 778 nm laser frequency comb with average power of 20 mW is obtained. By using this frequency-doubled laser frequency comb, we have measured the absolute frequency of the ^{87}Rb $5S_{1/2}$ $(F = 2)$–$5D_{5/2}$ $(F'' = 4)$ two-photon transitions to an accuracy of 44 kHz. So far, the limitation of the accuracy at present is dominated by the light shift and the pressure shift which have been analyzed in detail. The linewidth of the DFCS is about ∼2 MHz, which is larger than the natural linewidth of the $5D$ level. This can be attributed to the spectral coverage (∼4 nm) of the frequency-doubled femtosecond laser comb at 778 nm, and the residual Doppler-shift cannot be canceled completely. Therefore, the accuracy can be further improved by tailoring the spectrum distribution of the laser frequency comb by using a spatial light modulator. The developed system can also be extended to other narrower two-photon transition measurement applications and can benefit the high-resolution spectroscopy measurement in thermal atomic vapor.

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