Absolutely Direct Frequency Measurement of Two-Photon Transition Using Multi-Peak Fitting Approach *

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The optical frequency comb has been widely used in precision measurement. In this study, a multi-peak fitting approach is first proposed to fit the two-photon transition spectrum which overlaps with the neighboring transition in ^{87}Rb. The multi-peak fitting approach is used to eliminate the frequency shift affected by the neighboring transition. With locking the carrier envelope offset frequency at 1/4 repetition frequency, the transition frequency is measured to be 770569132739.9±5.8 kHz, which agrees well with the previous result recommended by Comité International des Poids et Mesures.

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Doppler-free two-photon transition (TPT) is one of the most important parts of laser spectroscopy. It is widely applied to measure the frequency of forbidden transition. The TPT spectra are first observed by using cw laser in a counterpropagating beam geometry.[1] The deficiencies are that the cw laser cannot directly measure the frequency of TPT and one cw laser is only suitable for one transition. The TPT frequency cannot be measured extensively until the invention of the femtosecond optical frequency comb (FOFC).[2–8]

As the particular frequency characteristic of the FOFC with the precise phase relation of a train of ultrashort pulses, the transition frequency can be measured directly and precisely,[9,10] and this process is called the direct frequency comb spectroscopy (DFCS). The TPT spectra are realized by scanning the repetition rate (f_r) or carrier envelope offset frequency (f_o) of the FOFC.[11] By precisely counting the changed frequency,[6,12] the frequency of TPT can be measured directly.

In the last decade, studies on the improvement of the signal-to-noise ratio (SNR) of the direct two-photon spectroscopy were carried out to enhance the accuracy of the measurements. A series of corresponding spectral control techniques were applied to enhance the TPT signal[13] or to eliminate the Doppler broadening background.[10,14] In addition, the effects on the TPT of laser properties have been studied in detail.[11,15] Furthermore, a variety of laser locking methods were proposed to measure the TPT frequencies.[16–20] In those works, most of the targeted transitions are far away from the neighboring transitions. The most common method to determine the central frequency is just to fit the spectrum with a single peak fitting function. The transition overlapping the neighboring transition is difficult to measure precisely. A kind of multi-peak method is used in measuring the molecule transitions.[21–23] However, this kind of multi-peak method has not been applied in the DFCS.

In this Letter, we propose a multi-peak fitting approach to reduce the shift caused by the neighboring transitions for measurement of the $5S$–$5P$–$5D$ TPT spectrum of ^{87}Rb by using an Er-doped fiber FOFC. This method provides the feasibility in the frequency measurement of the TPT overlapping neighboring transitions. Meanwhile, the method improves the measuring precision. To our knowledge, this is the first time to precisely measure the frequency of the neighboring overlapping TPT. In addition, the experimental process to ensure initial frequency of FOFC is overlooked by locking f_o at $f_r/4$. This novel locking approach simplifies the way to precisely measure the initial frequency of the FOFC.

An Er-doped fiber FOFC[24,25] was made to excite the rubidium 87 atoms from the $5S_{1/2}$ state to the $5D_{5/2}$ state. The transition process is shown in Fig. 1. There are two kinds of transition modes in the TPT transition. One is the resonant transition driven via the intermediate resonance.[10,26,27] The other one is the nonresonant transition,[10,28,29] in which pairs of the FOFC modes excite the atoms from the $5S_{1/2}$ state to the $5D_{5/2}$ state through virtual energy levels. The essential condition of nonresonant transition is that the total energy of a pair of frequency modes from the FOFC is the same as the energy of TPT, shown in Fig. 1.

A periodically poled lithium niobate crystal (PPLN)[30] is used to double the frequency of the comb

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laser. The power of the second harmonic pulse laser (SH) is about 23.36 mW. According to the f_r and f_o of the Er-doped FOFC, the nth mode frequency of the SH laser is $f_n = nf_r + 2f_o$, where f_n can be expressed as $(n+1/2)f_r$[31] by locking f_o at $f_r/4$. Subsequently, the pulse laser is divided into two beams with equal power in a counterpropagating beam geometry by a polarized beam splitter (PBS) and two mirrors. Two laser pulses temporally overlap at the center of the cell saturated with pure atomic vapor of ^{87}Rb. The temperature of the Rb cell is stabilized at 27±0.02℃. A shielding box is applied for magnetic shielding. The optical structure is shown in Fig. 1. In the Rb cell, the TPT can take place over the whole light path. The Doppler-free TPT only takes place at the temporally overlapping point.

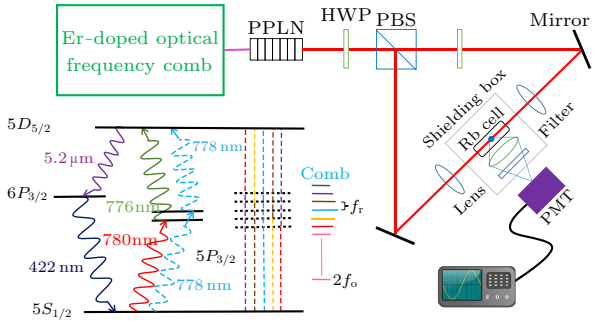


Fig. 1. The system of the direct frequency measurement of Rb consisting of the two-photon transition energy level structure of ^{87}Rb. PPLN, periodically poled lithium niobate crystal; HWP, half wave plate; PBS, polarized beam splitter; PMT, photomultiplier tubes; f_r, repetition rate; and f_o, carrier envelope offset frequency.

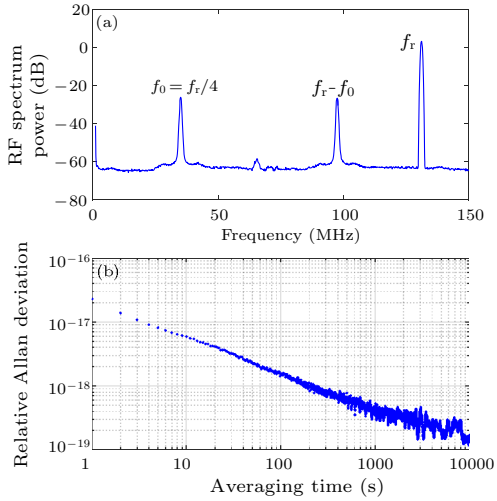


Fig. 2. (a) The frequency spectrum of f_o and f_r of the optical frequency comb. (b) Relative Allan deviation of f_{ceo} to $f_{1550} \approx n \times f_r$.

Here f_r can be stabilized with the hydrogen master clock by controlling the effective optical length of the optics parametric oscillator. The stability of f_r is in accordance with the hydrogen master clock. Here f_o is locked at $f_r/4$ to overlook the judging process of f_o which is quite tedious, and the frequency spectrum is shown in Fig. 2(a). The relative Allan deviations of locked f_o are shown in Fig. 2(b). In this way, the frequencies of all of the comb modes are ensured to be $(n+1/2)f_r$.

Scanning f_r, the TPT spectrum of ^{87}Rb is gained as shown in Fig. 3. In the figure, the TPT spectra of ^{87}Rb are displayed with an f_r period of 24 Hz, and the measured TPT signal is shown as the red dotted line. Compared with the solid lines in different colors, which stand for the center line in theory,[1,32] the dotted line shows the possible TPT. The transition strength shown by the peak value of the dotted line is generally consistent with the theoretical values shown as the heights of the solid lines. In this study, we measure the frequency of the $5S_{1/2}(F = 2)$–$5D_{5/2}(F'' = 4)$ TPT, as this transition has the strongest transition strength according to the transition rule. As shown in Fig. 3, the $5S_{1/2}(F = 2)$–$5D_{5/2}(F'' = 4)$ TPT spectrum overlaps with the $5S_{1/2}(F = 1)$-$5D_{3/2}(F'' = 3)$ TPT spectrum. When f_r is near 136.238701 MHz, the frequency interval is about 1.16 Hz, which is too close to accurately fit the spectrum, as the measured transition linewidth of the $5S_{1/2}(F = 2)$–$5D_{5/2}(F'' = 4)$ TPT is about 4.5 MHz (consisting of 0.6 MHz natural linewidth, 0.5 MHz transit broadening, and 3.4 MHz residual Doppler broadening). Increasing the cavity length of the FOFC by heating the temperature of the cavity, f_r is turned to 136.230540 MHz and the interval is increased to 2.025 Hz. This interval causes the overlap of the two transitions on a small scale.

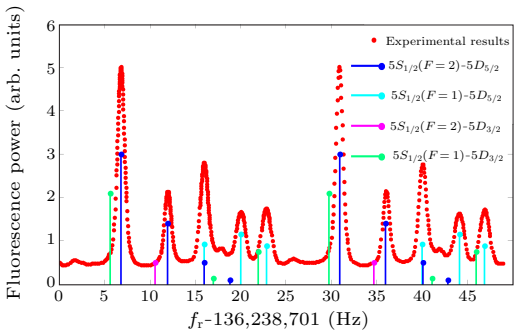


Fig. 3. The two-photon transition spectrum of ^{87}Rb. The value of f_r starts from 136.238701 MHz.

In theory,[4,10] the symmetry of the TPT spectrum determines that the spectrum can be fitted by using any kind of symmetric function without introducing a systematic error. According to Ref. [10] a simplified fitting function consisting of a sum of a single Gaussian and Lorentzian can be well used for every transition. However, the overlap between two TPT spectra makes this impossible. For the $5S_{1/2}(F = 2)$–$5D_{5/2}(F'' = 4)$ TPT, the $5S_{1/2}(F = 1)$–$5D_{3/2}(F'' = 3)$ TPT spectrum deforms the left part of the spectrum as shown in Fig. 3. In this case, the single peak fitting approach will be improper. To eliminate the effect of the overlap, a multi-peak fitting approach is proposed. The multi-peak fitting approach means that the fitting function has multiple

peaks standing for the corresponding transitions. In the experiment, the noise transition strength is much lower than the targeted transition, which means that the influence of the noise transition on the targeted transition is quite small. Therefore, the multi-peak function can be simply a sum of two pairs of the Gaussian function and the Lorentzian function. In the model, the widths of the Gaussian and the Lorentzian functions are given as free parameters and are not physically meaningful. Also, an approximate interval between the two peaks should be presented according to the theory.

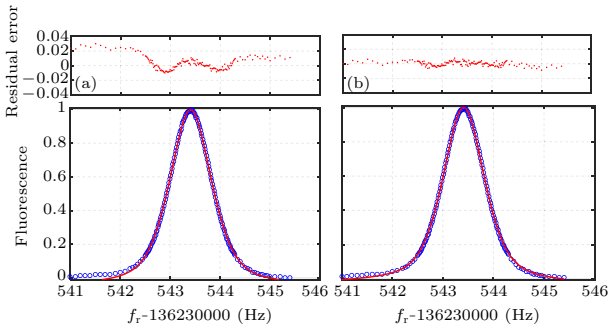


Fig. 4. Fluorescence decay signal (normalized) shown as open circles obtained by scanning f_r of the FOFC driving the $5S_{1/2}$–$5D_{5/2}$ transition: (a) The single peak fitting result, and (b) the multi-peak fitting result. The solid curve is the fitting result, and the fit residuals are shown on top.

To verify the correctness of the theory model, we fit the spectrum using the single peak function and the multi-peak function separately. Firstly, we use a single peak function to fit the transition spectrum, as shown in Fig. 4(a). The figure shows that the fitting degree becomes relatively worse near the noise transition. Then, the multi-peak function is applied and the fitting results are shown in Fig. 4(b). The residuals of the multi-peak fit are obviously better than the single peak fit. By fitting the spectrum with a multi-peak function, the central frequency of the spectra is determined to be 136230543.4221±0.0006 Hz. The frequency of $5S_{1/2}(F=2)$–$5D_{5/2}(F''=4)$ TPT is measured to be 770569132821.6±3.4 kHz with $f = Nf_r$. Compared with the fitting results of the single peak fit, the measured frequency difference between two kinds of spectra fit is about 28.3 kHz, which is larger than the uncertainty.

Before the absolute frequency of the targeted transition is determined, an imperative process to quantify and correct all of the possible systematic shifts should be completed. Normally, the systematic shifts consist of light shift, collision shift, second-order Doppler shift, black-body radiation shift and Zeeman shift. Light shift is the main systematic shift in this experiment. As the light is proportional to light power, light shift can be deduced with the measured frequencies under different light powers. The variation curve is shown in Fig. 5. The light shift at 23.36 mW is found to be 80±2.3 kHz. Different from the light shift, the other systematic shifts can be quantified according to the established works.

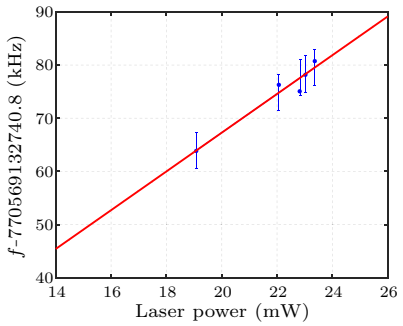


Fig. 5. The frequency calibration of the light shift of the ^{87}Rb $5S_{1/2}$–$5D_{5/2}$ transition.

The collision shift is caused by the collisions among the particles in the cell. In the experiment, the collision shift is mainly caused by the collision among Rb atoms, which is about −10 Hz.[33,34] The uncertainty can be ignored. According to the previous study,[35] the black-body radiation shift is about −179± 2 Hz. The second-order Doppler shift is −328.5± 0.3 Hz. The main shift due to the external magnetic field is the second-order Zeeman shift. As our Rb cell is put in a μ-metal magnetic shield, the residual magnetic field is smaller than 0.5 Gauss. The second-order Zeeman shift is about 2.2±0.1 kHz. As a result, the total frequency shift is about 81.7 kHz and the uncertainty is 2.4 kHz. The calibration of all of the possible frequency shifts leads to the absolute transition frequency of 770569132739.9 kHz of $5S_{1/2}(F=2)$–$5D_{5/2}(F''=4)$ in ^{87}Rb with the uncertainty of 5.8 kHz.

Table 1. The summary of the systematic frequency shifts.

Effect	Shifts at 770 THz
Light shift	80±2.3 kHz
Second-order Doppler shift	−328.5±0.3 Hz
Rb–Rb collisions shift	−10 Hz
Black-body radiation shift	−179±2 Hz
Magnetic shift	2.2±0.1 kHz
Total	81.7±2.4 kHz

Table 2. Obtained frequencies of ^{87}Rb $5S_{1/2}(F=2)$–$5D_{5/2}(F''=4)$. The frequency should be added on 770569132700 kHz.

	[a]Ref. [32]	[a]Ref. [36]	[a]Ref. [1]	[b]Ref. [9]	[b]Ref. [12]	[b]This work
^{87}Rb F=2–4	32.6(16)	43.4(12)	40.8(4)	48.8(16.8)	46(10)	39.9(5.8)

[a]Using continuous laser, and [b]using optical frequency comb.

Table 2 and Fig. 6 show the comparison between this work and the previous studies. Our result is in good agreement with the previous works, which means that the multi-peak fitting formula is reasonable. There are two kinds of laser sources for measuring the transition frequency from $5S_{1/2}(F=2)$–$5D_{5/2}(F''=4)$ of ^{87}Rb. One is the continuous laser,[1,32,36] and the other is the optical frequency comb, comparatively one work was a magneto-optical trap in the MOT[9] and the other was in the hot Rb cell.[12] The measured frequency of Ref. [1] is the recommended frequency by the Comité International des Poids et Mesures (CIPM). Compared with the results using the continuous laser, the uncertainty of our result has the same magnitudes. Compared with the results using the FOFC, our result is closer to the recommended value as shown in Fig. 6. This means that the multi-peak fitting approach improves the measuring accuracy. In addition, the uncertainty of our result is smaller than other results using FOFC. The good consistency between our works and previous works proves that the multi-peak fitting approach is reasonable.

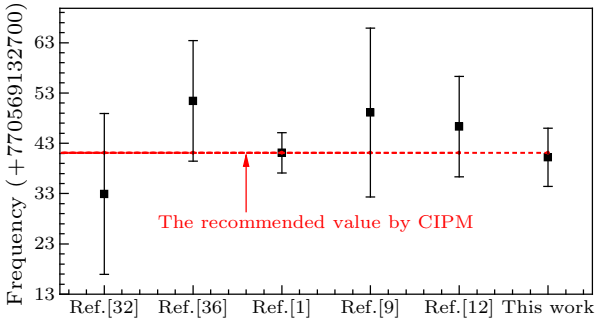


Fig. 6. The measured frequencies of $5S_{1/2}(F=2)$–$5D_{5/2}(F=4)$ of ^{87}Rb.

In summary, we have gained the two-photon transition spectrum of ^{87}Rb from $5S_{1/2}$ to $5D_{5/2}$ by scanning f_r of a femtosecond optical frequency comb. In addition, f_o is locked at $f_r/4$ to overlook the judging process of f_o. We use a multi-peak fitting approach to eliminate the error caused by the overlap with another TPT transition, and obtain the absolute transition frequency of 770569132739.9±5.8 kHz of $5S_{1/2}(F=2)$–$5D_{5/2}(F''=4)$ in ^{87}Rb, which is in good agreement with the CIPM data. The multi-peak fitting approach is a feasible way to eliminate the effect of the neighboring transition. This provides a good way for quick measurement in precision measurement experiments.

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