The Optimization of Cold Rubidium Atom Two-Photon Transition Excitation with an Erbium-Fiber Optical Frequency Comb

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Received: 11 January 2019; Accepted: 1 March 2019; Published: 5 March 2019

Abstract: We demonstrated the observation of cold rubidium atom two-photon transition excitation by a fiber optical frequency comb. In addition to this, we optimized the repetition rate of optical frequency comb to enhance two-photon intensity by controlling cavity length and pump source of optical comb. This technique can fine tune repetition rate to corresponding stepwise two-photon transition resonance frequency and improve the transition intensity by three times. This method is useful in Doppler laser cooling and detection of macromolecules.

Keywords: cold atom; two-photon transition; optical frequency comb

1. Introduction

With the development of frequency-stabilization techniques [1–3] and laser cooling and trapping techniques [4], two-photon spectroscopy has developed fast in recent years. For its high accuracy and stability, two-photon spectroscopy has many practical applications in many fields, such as fiber communication [5,6], precision measurement [7–15], detection of macromolecules [16], and Doppler laser cooling [17,18]. Among this, the Rubidium (Rb) 5S_{1/2}–5D_{5/2} two-photon transition (TPT) has unique advantages, such as narrow linewidth and low sensitivity to environment [19]. The relevant energy levels of Rb ladder type two-photon transition is shown in Figure 1. In addition, the 778 nm transition can be easily excited by frequency doubling the communication band lasers at the C band (an infrared communication band from 1530 to 1565 nm). For these reasons, it is suitable for optical communication frequency standard. The stability of the Rb two-photon frequency standard has reached $10^{-13}$ [20]. Because of many natural advantages of optical frequency comb (compact scheme, wide spectrum bandwidth and high peak intensity), the direct frequency comb spectroscopy (DFCS) on the TPTs is the research focus of two-photon spectroscopy [5–15]. Compared to a narrow linewidth continuous wave (CW) laser, the linewidth of each optical comb teeth is easier to be realized and adjusted to sub-hertz [21]. Moreover, for its higher instantaneous power and higher conversion efficiency in the deep ultraviolet (122 nm or 200 nm) [18], optical frequency comb has the potential to demonstrate the TPT excitations of the C, H, N, and O atoms in organic chemistry that are not available for CW laser [18]. The two-photon spectroscopy using optical frequency combs was first proposed for observation of the 1s-2s transition of hydrogen atoms [7]. In addition, a coherent control method, which uses optical frequency comb to directly excite TPT, was proposed in recent years [8]. This method, which is called Direct Frequency Comb Spectroscopy (DFCS), is widely used for TPT spectroscopy of molecules and ions [5,6,9–15]. In the previous work, we explore a simplified scheme to obtain the DFCS of Rb two-photon transition [5,6]. In contrast to the conventional scheme, this scheme utilizes an Erbium-fiber-based frequency comb to excite the two-photon transition. The natural advantages of Erbium (Er) optical frequency make the whole scheme more simplified and robust.
In order to implement accuracy and stability of the system, we eliminate the Doppler-broadening, which is the main line broadening. The laser cooling technologies, such as magneto optical trap [4] and molasses, can eliminate most of the Doppler-broadening background. However, the number of atoms in the magneto-optical trap is much lower than the number of atoms in a normal gas cell. Cold Rb TPT transition intensity is weak for establishing frequency standard or Doppler laser cooling. Therefore, following the previous research [5,6], we try to optimize repetition frequency of optical frequency comb to achieve higher transition intensity with the same average probe laser power. As the result of measurement, transition intensity is improved threefold.

2. Principle

Specific theoretical analysis is as follows. Each mode of optical frequency comb can be expressed as

\[ F_n = f_0 + nf_{\text{rep}} \]

where \( f_0 \) is the carrier-envelop offset frequency, \( f_{\text{rep}} \) is the pulse repetition rate, and \( n \) is an integer on the order of \( 10^6 \). The sum frequency of two comb modes is given by

\[ F_{\text{sum}(m+n)} = 2f_0 + (m + n)f_{\text{rep}} \]

Whenever the sum frequency of two modes coincides transition frequency, the optical comb excites the two-photon transition. A hundred thousand pair modes can satisfy the resonant condition at the same time. When two comb modes are near resonance with the 5S-5P and 5P-5D transitions, the intermediate 5P state enhances stepwise two-photon transitions (S-TPT). We utilize this phenomenon to enhance the two-photon transition signal intensity.

Due to the negligible Doppler broadening in cold atom experiments, the S-TPT excitation conditions are more critical. S-TPT can be achieved only when the 5S-5P and 5P-5D transition frequency difference is exactly an integral multiple of the repetition frequency. If \( f_{\text{rep}} \) deviates \( (f_{776nm} - f_{780nm})/n \), no S-TPT can be excited, because no other comb excites another single-photon transition when one comb frequency is aligned with a single-photon transition. To validate this feature, we consider all 14 S-TPT transition pathways for \( 5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5D_{5/2} \) of \( ^{87}\text{Rb} \). 14 S-TPT pathways and the TPT intensity of each TPT pathway are shown in Table 1.

Each transition path has a different single-photon transition frequency, as well as a different two-step transition frequency difference, which has a different repetition frequency resonance value. The results in Figure 2 show the simulation curves of the total TPT intensities produced by these 14 S-TPT pathways with repetition rates. It can be seen that the S-TPT intensity exhibits a periodicity of about 8 kHz with \( f_{\text{rep}} \), which corresponds to the range of variation of the repetition frequency from one resonance value to the next resonance value (\( \Delta f/n \rightarrow \Delta f/(n + 1) \)). Take the transition path \( 5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F = 3) \rightarrow 5D_{5/2}(F = 4) \) as an example. The two-step transition frequencies are \( f_{780nm} = 384,228,115,203.2 \text{ kHz} \), \( f_{776nm} = 386,341,017,529.38 \text{ kHz} \). The resonance frequency of the repetition
frequency corresponding to the point A in the figure is \( (f_{776\text{nm}} - f_{780\text{nm}})/16,307 = 129,570 \text{ kHz} \). The next repetition resonance frequency value is \( (f_{776\text{nm}} - f_{780\text{nm}})/16,306 = 129,578 \text{ kHz} \). The difference between the two values is about 8 kHz. The calculation results of other points are also about 8 kHz, with only slight differences. So, as a whole, the TPT intensities exhibit periodicity of 8 kHz with \( f_{\text{rep}} \).

**Table 1.** 14 two-photon transition (TPT) pathways and transition intensity of these pathways.

| Rb\(^{87}\) TPT Pathways | Intensity |
|--------------------------|-----------|
| \( 5S_{1/2}(F = 1) \rightarrow 5P_{3/2}(F = 0) \rightarrow 5D_{5/2}(F = 1) \) | 0.1666 |
| \( 5S_{1/2}(F = 1) \rightarrow 5P_{3/2}(F = 1) \rightarrow 5D_{5/2}(F = 1) \) | 0.125 |
| \( 5S_{1/2}(F = 1) \rightarrow 5P_{3/2}(F = 1) \rightarrow 5D_{5/2}(F = 2) \) | 0.2916 |
| \( 5S_{1/2}(F = 1) \rightarrow 5P_{3/2}(F = 2) \rightarrow 5D_{5/2}(F = 1) \) | 0.00817 |
| \( 5S_{1/2}(F = 1) \rightarrow 5P_{3/2}(F = 2) \rightarrow 5D_{5/2}(F = 2) \) | 0.0972 |
| \( 5S_{1/2}(F = 1) \rightarrow 5P_{3/2}(F = 2) \rightarrow 5D_{5/2}(F = 3) \) | 0.3111 |
| \( 5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F = 1) \rightarrow 5D_{5/2}(F = 1) \) | 0.015 |
| \( 5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F = 1) \rightarrow 5D_{5/2}(F = 2) \) | 0.035 |
| \( 5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F = 2) \rightarrow 5D_{5/2}(F = 1) \) | 0.0049 |
| \( 5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F = 2) \rightarrow 5D_{5/2}(F = 2) \) | 0.0583 |
| \( 5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F = 2) \rightarrow 5D_{5/2}(F = 3) \) | 0.1866 |
| \( 5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F = 3) \rightarrow 5D_{5/2}(F = 2) \) | 0.0666 |
| \( 5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F = 3) \rightarrow 5D_{5/2}(F = 3) \) | 0.0933 |
| \( 5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F = 3) \rightarrow 5D_{5/2}(F = 4) \) | 0.5999 |

**Figure 2.** Simulation results about relationship between repetition rate of comb and transition intensity. Dotted lines indicate simulation results. Vertical line indicates 14 stepwise two-photon transitions (S-TPT) pathways. The resonance frequency of the repetition frequency corresponding to the point A is \( (f_{776\text{nm}} - f_{780\text{nm}})/16,307 = 129,570 \text{ kHz} \).

Based on the above calculation and analysis, in order to further enhance the signal intensity of two-photon transition signal in cold atoms, we need to finely control \( f_{\text{rep}} \). The standard technique to change repetition rate of an Er fiber optical frequency comb is to change the laser cavity length by a piezoelectric transducer (PZT) or a translation stage. The relationship between \( f_{\text{rep}} \) and cavity length can be given as

\[
\frac{f_{\text{rep}}}{f_{\text{rep}} + 1} = \frac{f_{776\text{nm}}}{f_{780\text{nm}}}
\]

where \( f_{\text{rep}} \) is the repetition frequency of the comb, and \( f_{776\text{nm}} \) and \( f_{780\text{nm}} \) are the laser frequencies at which the transitions occur.
where $L$ is the original length, $l(t)$ is length of the PZT or the translation stage at time $t$, $c$ is the light velocity, and $k$ is the average refractive index of the cavity. The resolution of the laser’s phase shift is given as in Reference [22]:

$$\min[\Delta \theta_o(t)] = \frac{l_{res} \cdot t_{res}}{L + l(t) + l_{res}}$$

where $\Delta \theta_o(t)$ is the laser’s phase shift, $l_{res}$ (most larger than 0.5 nm) is the length-tuning resolution of the PZT or the translation stage, and $t_{res}$ (usually in sub-microsecond scale) is the response time. However, limited by length-tuning resolution and long response time, it is hard for the laser’s phase shift to be smaller than 40 fs [22]. The cavity-length-controlling technique can demonstrate a wide range adjustment of $f_{rep}$, but is not suitable for fine adjustment.

The other way to change the repetition rate of the fiber optical frequency comb is using pump power modulation. This is different with Ti as with the sapphire frequency comb, gain fiber is added into the fiber optical comb cavity. Changes in pump power affect the interaction between atoms in gain fiber that result in changes of $f_{rep}$ [23, 24]. Based on the experiment and analysis in Reference [22], we know changing the pump source affects $f_{rep}$ in a linear way. This method of adjustment is not limited by PZT length-tuning resolution and response time, and the laser’s phase resolution can increase by two orders of magnitude. By controlling the pump source, fine $f_{rep}$ adjustment can be achieved.

In summary, combined with cavity length controlling and pump source controlling, we can find the $f_{rep}$ corresponding S-TPT resonance to increase the two-photon transition intensity.

### 3. Experiment & Results

The experiment setup for our system is shown in Figure 3. An Erbium-fiber-based frequency comb with the 1556 nm center wavelength is used as the source to excite atoms. The Er frequency comb emits 100 fs pulses with 100 mW average power and a repetition rate of 129 MHz. The cavity length can be adjusted via a PZT and a translation stage in the cavity. A home-built two-stage erbium-doped fiber amplification module is used to amplify the output of 1556 nm mode-locked laser to 200 mw. In order to improve pulse peak power, the comb is compensated the pulse broadening in optical fiber with a pair of prisms made of silica. The properties of the optical pulse are adjusted to focus the power of the optical frequency comb spectrum to the vicinity of 1556 nm. Then we focus the beam into a periodically poled lithium niobate crystal (PPLN) by a lens to double the frequency of the comb. A 20 mW frequency comb with center wavelength of 778 nm is obtained in the end.

Secondly, the 778 nm frequency comb is directed into magneto optical trap (MOT) cell to probe the cold $^{87}$Rb atom cloud which is cooled and trapped in MOT. To eliminate the influence of magnetic fields in MOT, we design a timing control cycle. We switched off the magnetic fields in MOT when the 778 nm frequency comb is directed into MOT cell to excite the Rb atom for 2 ms. Then the magnetic fields are switched on and the atom cloud is captured and cooled in MOT for 8 ms, and the 778 nm frequency comb is switched off by Acousto-optical Modulators (AOM) at the same time. Repeating this 10 ms cycle, we demonstrate the excitation of the two-photon transition.

To verify the relationship between $f_{rep}$ and TPT intensity, we scan $f_{rep}$ and detect 420 nm fluorescence signal from cascade decay via the 6P-5S state with a photomultiplier (PMT), used in photon counting mode. First, in order to verify the simulation results in Figure 2, we fix the pump current at 92.2 mA in experiments and adjust the repetition frequency to observe the fluorescence signal at the same time. When we scan $f_{rep}$ 50 Hz, the comb teeth at 780 nm will scan 129 MHz. So, we can get the direct frequency comb spectroscopy which illustrates all 5S→5D transition lines by scanning $f_{rep}$ 50 Hz with controlling voltage on PZT. The intensity of each line depends on the center scanning frequency of $f_{rep}$. The peak intensity of the DFCS is recorded. Then, we change the center $f_{rep}$ scanning frequency by adjusting the translation stage in cavity and repeat the above steps.
The obtained result is shown in Figure 4. Blue dots show the peak intensity of the DFCS with different center \( f_{\text{rep}} \) scanning frequency. Red lines show simulation results. It can be seen that the experiment data and the simulation results are almost perfectly matched, which fully illustrates the influence of the repetition frequency on the two-photon intensity and verifies our theoretical analysis. Then, we verify the small change of repetition frequency caused by the pump power change. In ensuring the laser cavity length and average power fixed, we change the pump power and measure TPT intensity, and the results are shown in Figure 5. We can observe the small shift of the TPT peaks in the spectrum because the frequency of the two photon transitions is fixed. It can be seen that different pump powers affect the repetition frequency of the frequency comb.

Figure 3. Simplified block diagram for stabilization system. MLL: Mode-locked laser; EDF: Er-doped fiber; ISO: isolator; HNLF: High non-linear fiber; PPLN: Periodically Poled Lithium Niobate; PMT: photon-multiplier; MOT: magneto optical trap; QWP: quarter-wave plate; HWP: half-wave plate.

Figure 4. Relationship between repetition rate of comb and two-photon transition intensity. Red lines show simulation results. It can be seen that the experiment data and the simulation results are almost perfectly matched, which fully illustrates the influence of the repetition frequency on the two-photon intensity and verifies our theoretical analysis. Then, we verify the small change of repetition frequency caused by the pump power change. In ensuring the laser cavity length and average power fixed, we change the pump power and measure TPT intensity, and the results are shown in Figure 5. We can observe the small shift of the TPT peaks in the spectrum because the frequency of the two photon transitions is fixed. It can be seen that different pump powers affect the repetition frequency of the frequency comb.
Figure 4. Relationship between repetition rate of comb and two-photon transition intensity. Red lines show simulation results. Blue dots show experiment results. The dashed line shows the average peak intensity of direct frequency comb spectroscopy (DFCS) without optimization.

Figure 5. The small $f_{rep}$ change caused by the pump power change.

Finally, we improve the two-photon transition by adjusting the repetition frequency to correspond to the S-TPT resonance frequency. The PMT signal is delivered into a lock-in amplifier to generate an error signal. The error signal is fed back to tune the PZT and pump source to lock center scanning frequency of $f_{rep}$ to the corresponding $5S_{1/2}(F=2)$-$5P_{3/2}(F=3)$-$5D_{5/2}(F=4)$ S-TPT resonance frequency. Next, we fine scan $f_{rep}$ to get Rb TPT signals. Direct frequency comb spectroscopy of cold Rb TPTs is shown in Figure 6. The peak fluorescence intensity detected through PMT at the optimization spectrum in Figure 6 is about 1189 photons per millisecond when we tune $f_{rep}$ with PZT and pump power. If we only optimize DFCS with controlling PZT, the peak intensity is 903 per millisecond. As a comparison, the peak intensity of most DFCS (about 75% DFCS) without $f_{rep}$ optimization is around 400 photons per millisecond. The final TPT intensity can increase by up to three times before optimization. The experimental results are consistent with the simulation in Figure 4 which shows that the peak intensity (transition intensity after optimization) is 850 photons per millisecond and the bottom intensity (transition intensity without optimization) is 320 photons per millisecond. Due to weak Doppler Effect in MOT, the linewidth of the cooled Rb TPT resonance is improved. We achieve better linewidth in cooled atoms (about 900 kHz) compared with linewidth in thermal atoms in our previous work [6] (about 2 MHz). The residual linewidth is mainly due to the 600-kHz natural linewidth and the 300-kHz transit time broadening. The reduced linewidth is approximately equal to the Doppler broadening of the thermal atoms (about 1.2 MHz). This shows that we can ignore Doppler broadening in cold atoms.
4. Conclusions

In conclusion, we have demonstrated cold Rb atom two-photon transition excitation with an Er-fiber optical frequency comb. In addition, we optimized the repetition rate of optical frequency comb to enhance TPT intensity by controlling cavity length and pump source. The optimization method not only excites rubidium atoms to establish frequency standard, but also has applications for utilizing fiber optical comb to excite other atom TPTs. For example, there have been studies of Doppler cooling using fiber optical combs [25]. However, most of these studies are limited by the fiber optical combs power and have failed to expand in depth. The optimization method can greatly reduce the power required for Doppler cooling to solve power problem. It has many potential applications in Doppler laser cooling and detection of macromolecules [16].

Author Contributions: Data curation, J.L.; Investigation, H.L. and J.L.; Methodology, J.Z.; Writing, H.L. and J.Z.

Funding: This work was funded by the National Natural Science Foundation of China (NO. 61535001 & NO. 91836301) and the Program of International S&T Cooperation (No. 2016YFE0100200).

Conflicts of Interest: No conflict of interest.

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