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Two-Photon Transitions of ^{85}Rb $5D_{5/2}$ State by Using an Optical Frequency Comb and a Continuous-Wave Laser *

Shu-Kai Cao(曹书凯), Peng-Rui Fan(范鹏瑞), Yi-Chi Zhang(张一驰), Li-Rong Wang(汪丽蓉)**, Lian-Tuan Xiao(肖连团), Suo-Tang Jia(贾锁堂)

State Key Laboratory of Quantum Optics and Quantum Optics Devices, Institute of Laser Spectroscopy, Shanxi University, Taiyuan 030006

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A high-resolution two-photon spectrum of $5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5D_{5/2}$ transitions in a thermal ^{85}Rb vapor cell is presented by using an optical frequency comb and a cw laser. The fluorescence of $6P_{3/2} \rightarrow 5S_{1/2}$ spontaneous emission is detected when the cw laser frequency is scanned from the $5S_{1/2}$ ground state to $5P_{3/2}$ hyperfine levels and the optical frequency comb repetition rate is fixed. The hyperfine splittings ($F_{\rm f} = 2$–5) of the $5D_{5/2}$ excited state are well resolved. The dependences of fluorescence intensities on the cw laser intensity and temperature of ^{85}Rb vapor cell are studied, respectively. The experimental results are in good agreement with the theoretical analyses.

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The two-photon Doppler-free spectroscopy is a useful method in determining fundamental constants of atoms and molecules, such as hyperfine structure constants,[1] Rydberg constants[2,3] and proton charge radius.[4,5] Optical frequency combs have been proven to be an excellent tool for studies of precise atomic and molecular spectroscopy and related constants.[6] Researchers used optical frequency combs to measure electron energy levels in atoms, to detect and control atomic energy level transitions in real time, to precisely determine atomic transition frequencies, as well as to investigate quantum coherence, atomic and molecular dynamics.[7]

Jason *et al.* measured the absolute frequencies and hyperfine coupling constants in ^{133}Cs vapor.[8] The ground state atoms were excited to the different hyperfine states by employing narrowband interference filters in the exciting-lasers pathways.[9] However, the narrowband interference filters attenuated the intensity of the exciting laser. In this Letter, we report a combined action of an optical frequency comb and a cw laser, each of them drives one step of the two-photon transitions of $5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5D_{5/2}$ state in the thermal ^{85}Rb vapor cell. The cw laser plays a role as a velocity-selective filter, instead of the traditional narrowband interference filters.

There are numerous applications of two-photon transitions of $5S_{1/2} \rightarrow 5D_{5/2}$ in Rb atoms. Firstly, the two-photon transitions of $5S_{1/2} \rightarrow 5D_{5/2}$ can be recommended as a secondary frequency reference due to the long natural lifetime of the $5D_{5/2}$ state (about 240 ns).[10] Secondly, the $5S_{1/2} \rightarrow 5D_{5/2}$ transitions are used to study the excitation transfer between the Rb $5D$ hyperfine structure states in collision with ground-state atoms.[11] Finally, two-photon transitions are the important optical frequency standards for telecommunication applications.[12]

In this experiment, a cw laser and an optical frequency comb are used to excite the $5S_{1/2} \rightarrow 5P_{3/2}$ transition at 780 nm and the $5P_{3/2} \rightarrow 5D_{5/2}$ transition at 776 nm. The fluorescence from the $6P_{3/2}$ state to the $5S_{1/2}$ ground state is obtained as a function of the cw laser frequency while the repetition rate of the optical frequency comb is fixed. We obtain a high-resolution two-photon spectrum of $5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5D_{5/2}$ transitions in a thermal ^{85}Rb vapor cell. The hyperfine splittings ($F_{\rm f} = 2$–5) of the $5D_{5/2}$ excited state are well resolved. In addition, the fluorescence intensity variations of $5S_{1/2}(F_{\rm g} = 3) \rightarrow 5D_{5/2}(F_{\rm f} = 5)$ and $5S_{1/2}(F_{\rm g} = 3) \rightarrow 5D_{5/2}(F_{\rm f} = 4)$ transitions with the vapor cell temperature and the cw laser intensity are demonstrated and discussed, respectively.

The relevant hyperfine structure energy levels of ^{85}Rb in this experiment are shown in Fig. 1(a). The excitation pathway from the $5S_{1/2}$ state to the $5D_{5/2}$ state is via the $5P_{3/2}$ state. The decay channel from the $5D_{5/2}$ state to the $5S_{1/2}$ ground state is through the $6P_{3/2}$ state. The fluorescence is detected from the $6P_{3/2}$ state to the $5S_{1/2}$ ground state.

Figure 1(b) is the schematic diagram of the experimental setup. A cw laser (DL PRO) with a linewidth of ~1 MHz is used to excite the $5S_{1/2} \rightarrow 5P_{3/2}$ transition at 780 nm. It is split into two beams by a polarization beam splitter (PBS). One is used to ob-

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**Corresponding author. Email: wlr@sxu.edu.cn

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tain saturated absorption spectroscopy (SAS) as a frequency reference. The other beam is focused into the ^{85}Rb vapor cell. A femtosecond optical frequency comb (Menlo systems FC1500) is employed to excite $5S_{1/2} \rightarrow 5P_{3/2}$ and $5P_{3/2} \rightarrow 5D_{5/2}$ transitions. The optical frequency comb is based on a mode-locked Erbium fiber laser, which has a repetition rate of ~250 MHz, a pulse width of <150 fs, and an offset frequency is tuned to 20 MHz. The repetition rate of the optical frequency comb is phase locked to a reference signal from a cesium atomic clock via a global positioning system. In addition, the carrier-offset-envelope frequency is phase locked to the same cesium atomic clock by the well-known f–2f self-referencing technique.

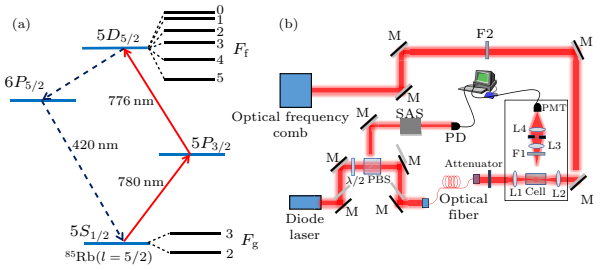


Fig. 1. (a) Energy levels related to the $5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5D_{5/2}$ two-photon transitions in ^{85}Rb. The solid lines are the excitation pathways, and the dashed lines represent the decay channels. (b) Experimental setup. M: reflector; PBS: polarizing beam splitter; SAS: saturated absorption spectroscopy; F2: the filter in 780nm; F1: the filter in 420 nm; L1, L2, L3 and L4: optical lenses; PMT: photomultiplier tube; and PD: photodiode.

The average output power of the optical frequency comb is ~70 mW when its wavelength range covers 600–900 nm. The power of fs laser is ~6.1 mW after passing through the filter at 780 nm and the spot diameter of the output laser is ~100 μm. To improve the efficiency of the two-photon transitions, the beam of cw laser must overlap with that of the optical frequency comb in the center of ^{85}Rb vapor cell. The cell has a 100 mm length and a 25 mm diameter. The cell is wrapped in μ metal and aluminum foil to isolate the external magnetic field and make thermal insulation, respectively.

The fluorescence at 420 nm from the $6P_{3/2}$ state to the $5S_{1/2}$ ground state is obtained as a function of the cw laser frequency when the repetition rate and offset frequency of the optical frequency comb are fixed. The photomultiplier tube (PMT) (Hamamatsu, CR131) is used to detect the fluorescence. To increase the fluorescence collection efficiency and to remove stray light, a lens group and a filter (center wavelength: 420 nm, bandwidth: 10 nm) are placed in front of the PMT. The signal from the PMT is recorded and analyzed by a computer.

In this experiment, the cw laser excites the first step $5S_{1/2} \rightarrow 5P_{3/2}$ transition and the fs laser excites the second step $5P_{3/2} \rightarrow 5D_{5/2}$ transition. Fluorescence at 420 nm is emitted from the $6P_{3/2} \rightarrow 5S_{1/2}$ spontaneous decay. The frequency of the optical frequency comb f is equal to $f_0 + nf_r$, where f_0 is the offset frequency, f_r is the pulse repetition rate, and n is an integer of the order of 10^6. The cw laser frequency is υ. If x-axis is defined to be the propagation direction of the beam and v_x is the x component of the atomic velocity, $|1\rangle$, $|2\rangle$ and $|3\rangle$ represent $5S_{1/2}$, $5P_{3/2}$ and $5D_{5/2}$, respectively. The resonance condition for the two-photon transitions is, according to Ref. [13],

$$\upsilon_{|1\rangle\rightarrow|3\rangle} = (f_0 + nf_r)(1 - v_x/c) + \upsilon(1 + v_x/c). \quad (1)$$

The maximum transition probability occurs when the resonance condition for the two-photon transitions is satisfied. Figure 2 shows the fluorescence intensity as a function of the cw laser frequency. The cw laser frequency is scanned over two Doppler-broadened $5S_{1/2} \rightarrow 5P_{3/2}$ hyperfine transitions of ^{85}Rb at 780 nm while the repetition rate of the optical frequency comb is fixed. The power of the fs laser and the cw laser are ~6.1 mW and 22.5 mW, respectively, before entering the cell. In the middle of the cell, the beam diameter of the fs laser and the cw laser are 100 μm and 300 μm, respectively. The temperature of ^{85}Rb cell is ~340 K. The upper curve in Fig. 2 is the saturated absorption signal for D_2 lines of ^{85}Rb. It is used to calibrate the cw laser frequency. The lower curve is the fluorescence signal at 420 nm as a function of the cw laser frequency. The interval of the fluorescence peak is 250 MHz, which is excited by different modes of the frequency comb. When the fs laser repetition period is shorter than the excited-state relaxation time, velocity-comb-like excited-state hyperfine level populations are observed. This is a direct result of the fs laser excitation considered in the frequency domain.[14]

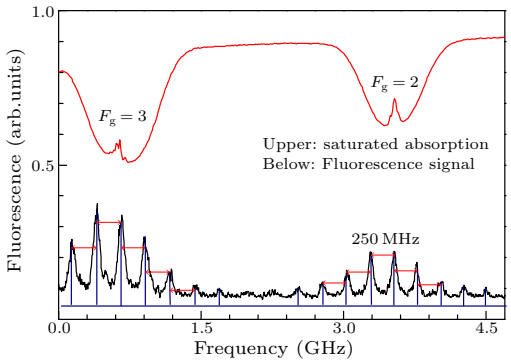


Fig. 2. Fluorescence from $6P_{3/2}$ to $5S_{1/2}$ as a function of the cw laser frequency for D_2 Doppler lines of ^{85}Rb. The upper curve is the saturated absorption signal of ^{85}Rb, which is detected simultaneously.

The hyperfine structure of the $5D_{5/2}$ state is studied by decreasing the scanning rate of the cw laser

frequency. The high-resolution two-photon spectrum of ^{85}Rb is obtained. Figure 3(a) is the scanning signal of the cw laser frequency with a scanning rate ∼0.3 MHz/s, and Fig. 3(b) illustrates the two-photon spectrum of ^{85}Rb for $5S_{1/2}(F_g = 3) \rightarrow 5D_{5/2}(F_f = 2$–$5)$ as a function of the cw laser frequency. The fluorescence signal of the $5S_{1/2}(F_g = 3) \rightarrow 5D_{5/2}(F_f = 1)$ transition is invisible due to the weaker signal amplitude than the noise level. The hyperfine splittings of the $5D_{5/2}$ excited state are well resolved. The typical full width at half maximum (FWHM) of fluorescence resonances for $5S_{1/2}(F_g = 3) \rightarrow 5D_{5/2}(F_f = 5)$ observed is ∼3.0 MHz. The value is larger than the natural linewidth, due to the other broadening mechanisms, such as residual Doppler broadening, transit time broadening and laser power broadening. The relative intensity ratio of hyperfine transition for $5S_{1/2}(F_g = 3) \rightarrow 5D_{5/2}(F_f = 2$–$5)$ is ∼0.60:1.00:2.15:3.40. The experimental results are consistent with the theoretical results.[15]

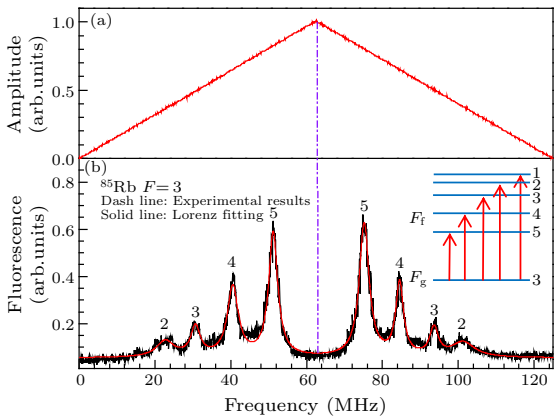


Fig. 3. (a) The scanning range of the cw laser frequency. (b) Hyperfine two-photon spectrum of ^{85}Rb for the $5S_{1/2}(F_g = 3) \rightarrow 5D_{5/2}(F_f = 2$–$5)$ transitions. The fluorescence signal of $5S_{1/2}(F_g = 3) \rightarrow 5D_{5/2}(F_f = 1)$ transition is annihilated in noise.

The temperature dependence on the peak of fluorescence intensity is measured between 323–383 K. The results are shown in Fig. 4(a) for the cw laser operated at 30 mW and the fs laser operated at 6.1 mW. The square dots represent the experimental results for the two-photon transition of $5S_{1/2}(F_g = 3) \rightarrow 5D_{5/2}(F_f = 5)$, the circular dots denote the results for the two-photon transition of $5S_{1/2}(F_g = 3) \rightarrow 5D_{5/2}(F_f = 4)$. In Ref. [15], the profile dominated by a spectral line is given by

$$G(\omega) \propto e^{\{-4\ln 2[(\omega-\omega_0)^2/\Delta\omega^2_{\text{Doppler}}]\}}, \tag{2}$$

and the Doppler-broadened linewidth is $\frac{2\omega_0}{c}\sqrt{\frac{2k_BT}{m}\ln 2}$, where T is the temperature of cell, m is the atomic mass, and k_B is Boltzmann's constant. The solid curve in Fig. 4(a) is a theoretical simulation with the experimental parameters by using Eq. (2). The fluorescence signal intensity increases in the temperature range of 323–370 K, then the signal gradually approaches saturation as the temperature is ∼370 K. At the higher temperature, the signal intensity does not increase due to the self-absorption of the $6P_{3/2} \rightarrow 5S_{1/2}$ transition. The results are in good agreement with Ref. [16]. The error is mainly attributed to the systematic error of multiple measurements.

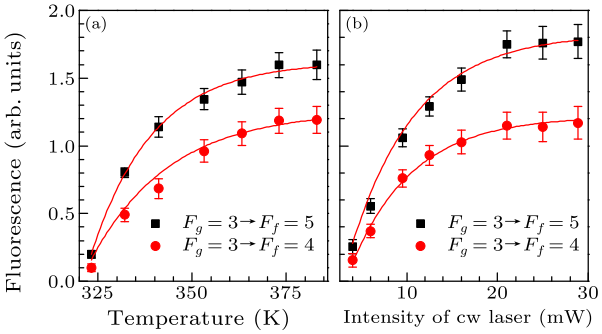


Fig. 4. (a) Fluorescence intensity as a function of temperature of the ^{85}Rb vapor cell. (b) Fluorescence intensity as a function of the cw laser intensity. The square dots represent the $5S_{1/2}(F_g = 3) \rightarrow 5D_{5/2}(F_f = 5)$ transitions, and the circular dots represent the $5S_{1/2}$ $(F_g = 3) \rightarrow 5D_{5/2}(F_f = 4)$ transition.

The population of the final state in the steady-state regime is obtained by solving the Bloch equation for one specific cw laser frequency integrated over the contribution of the whole velocity group within the Doppler profile,[17]

$$\rho_{33}^{(F,F',F'')}(\delta) = \frac{1}{(0.36\pi\Delta\omega^2_{\text{Doppler}})^{1/2}} \cdot \int_{-\infty}^{+\infty} \rho_{33}(\Delta,\delta)e^{-\Delta^2/0.36\Delta\omega^2_{\text{Doppler}}}d\Delta. \tag{3}$$

The strength of transition is determined by the corresponding Rabi frequency. As we know, the Rabi frequency equals to $\mu E/\hbar$, where μ is the electric-dipole moment for transition in the direction of the electric field. When the temperature of ^{85}Rb vapor cell is ∼340 K and the average power of the fs laser is ∼6.1 mW before entering the cell, the dependence of fluorescence intensity on the cw laser intensity is studied. The results for $5S_{1/2}(F_g = 3) \rightarrow 5D_{5/2}(F_f = 5)$ and $5S_{1/2}(F_g = 3) \rightarrow 5D_{5/2}(F_f = 4)$ hyperfine transitions are shown in Fig. 4(b). The fluorescence intensity increases with the cw laser intensity varying from 4.0 mW to 22.5 mW. A threshold-like behavior is observed along with the peak fluorescence intensity when the cw laser intensity exceeds 22.5 mW. The theoretical explanation can be performed by using the Bloch equations for a three-level cascade system as above.

In conclusion, the two-photon $5S_{1/2} \rightarrow 5D_{5/2}$ transitions have been studied by using the combined

action of a cw laser and a femtosecond optical frequency comb. The fluorescence signal of $5S_{1/2}(F_g = 3) \rightarrow 5D_{5/2}(F_f = 2\text{–}5)$ transition with the higher signal-to-noise ratio is obtained. The dependences of fluorescence intensity on the cw laser intensity and temperature of ^{85}Rb vapor cell are studied, respectively. The agreement between the measured results and the theoretical analyses is good. It is expected that this two-photon transition is recommended as a secondary frequency reference under the optimized experimental conditions.

References

[1] Gerginov V, Calkins K, Tanner C E, McFerran J J, Diddams S, Bartels A and Hollberg L 2006 *Phys. Rev.* A **73** 032504

[2] Mohr P J, Taylor B N and Newell D B 2008 *Rev. Mod. Phys.* **80** 633

[3] Leggett A J 2001 *Rev. Mod. Phys.* **73** 307

[4] Wynards R and Weyers S 2005 *Metrologia* **42** S64

[5] Budker D and Romalis M 2007 *Nat. Phys.* **3** 227

[6] Fischer M, Kolachevsky N, Zimmermann M, Holzwarth R et al 2004 *Phys. Rev. Lett.* **92** 230802

[7] Guéna J, Lintz M and Bouchiat M A 2005 *J. Opt. Soc. Am.* B **22** 21

[8] Stalnaker J E, Mbele V, Gerginov V, M Fortier T, Diddams S A, Hollberg L and Tanner C E 2010 *Phys. Rev.* A **81** 043840

[9] Aumiler D, Ban T, Skenderović H and Pichler G 2005 *Phys. Rev. Lett.* **95** 233001

[10] Snadden M J, Bell A S, Riis E and Ferguson A I 1996 *Opt. Commun.* **125** 70

[11] Wang L R, Zhang Y C, Xiang S S, Cao S K, Xiao L T and Jia S T 2015 *Chin. Phys.* B **24** 063201

[12] Bernard J E, Madej A A, Siemsen K J, Marmet L, Latrasse C, Touahri D, Poulin M, Allard M and Têtu M 2000 *Opt. Commun.* **173** 357

[13] Jin L, Zhang Y C, Xiang S S, Wang L R, Ma J, Zhao Y T, Xiao L T and Jia S T 2013 *Chin. Phys. Lett.* **30** 103201

[14] Barmes I, Witte S and Eikema K S E 2013 *Phys. Rev. Lett.* **111** 023007

[15] Stalnaker J E, Chen S L, Rowan M E, Nguyen K, Pradhananga T, Palm C A and Jackson K D F 2012 *Phys. Rev.* A **86** 033832

[16] Olson A J, Carlson E J and Mayer S K 2006 *Am. J. Phys.* **74** 218

[17] Ban T, Aumiler D, Skenderović H and Pichler G 2006 *Phys. Rev.* A **73** 043407