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Observation of Rb two-photon absorption directly excited by an erbium-fiber-laser-based optical frequency comb via spectral control

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We present the observation of the Rb two-photon spectrum directly excited by an Er:fiber laser comb in a hot atomic vapor system. The influences of the atomic velocity distribution on direct and sequential two-photon transitions are discussed respectively and a combination of corresponding spectral control techniques is developed to eliminate the Doppler broadening background. A direct frequency comb spectroscopy is obtained with high-resolution two-photon transition lines. These techniques would benefit spectroscopy measurement in hot atomic vapor systems, and pave the way for establishing a high-stability optical frequency comb in 1.5 μm optical communication bands based on the Rb two-photon transitions.

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Direct frequency comb spectroscopy (DFCS) of two-photon transitions (TPTs) has proved to be a powerful tool for several branches of science, e.g., atomic spectroscopy and time domain ultrafast dynamics [1]. Several techniques of quantum coherence control, including varying the spectral phases [2–4], tailoring the comb spectrum [5,6], and introducing chirp on the comb [7], have been proposed to guide the TPT quantum system toward a desired final state. Rubidium $5S$-$5D$ TPTs are of particular interest for their high transition intensity and excellent frequency properties, and relevant optical frequency standards perform a high stability on the order of $10^{-13}\,\tau^{-1}$ for integration time τ up to 1000 s [8,9].

Two types of TPTs, direct TPTs (D-TPTs) excited by two photons together and sequential TPTs (S-TPTs) which correspond to the stepwise transitions via the intermediate state, can both contribute to the total TPT intensity. In most of the existing TPT DFCS observation experiments [1,10–12], only one type of TPT plays the dominant role, while the other type is neglected or artificially avoided. In these experiments, D-TPTs were canceled out because of destructive interference [1], or the S-TPTs were excluded since the spectrum was so narrow that it did not cover the intermediate state [10,12]; even the comb lines near the intermediate state are blocked to avoid the S-TPTs [11].

This Rapid Communication focuses on observation of the high-resolution Rb $5S$-$5D$ TPT spectrum via a frequency-doubled Er:fiber laser comb operating at $\sim$1.5 μm, which would pave the way to further establish a high-stability optical frequency comb (OFC) for optical communication. And differently from the studies mentioned above, this Rapid Communication deals with the situation that both S-TPTs and D-TPTs are involved in the TPT excitation in a hot Rb vapor system. It is necessary and of great meaning because it helps to make full use of the frequency-doubled OFC power, improve the signal-to-noise ratio (SNR) of the TPT signals, and would further benefit the establishment of OFC in fiber communication bands.

As shown in Fig. 1(a), the two components of comb lines involved in the $5S_{1/2}$-$5P_{3/2}$ (780 nm) and $5P_{3/2}$-$5D_{5/2}$ (776 nm) transitions are incorporated in our experiment, which enables the occurrence of S-TPTs via the $5P_{3/2}$ state. Compared to other works with these spectral parts blocked [11,13], this is a more general situation and could enhance the total TPT intensity [5]. With the spectrum parts <776 nm and >780 nm blocked to avoid destructive interference of D-TPTs, the S-TPT and D-TPT could contribute comparably to the total TPT intensity [5]. Also shown in Fig. 1(a) is the general TPT excitation setup, where the Rb cell is illuminated by counterpropagating ultrashort pulses, which temporally overlap at the cell center. In our experiment, the cell is filled with pure ^{87}Rb isotope and heated to 60 °C. The OFC centered at 778 nm is generated by frequency-doubling an Er:fiber-based OFC with $\sim$144.5 MHz repetition frequency ($f_{\rm rep}$) and frequency-stabilized carrier-envelop offset frequency ($f_{\rm ceo}$). The generated pulses have $\sim$100 fs pulse duration and 20 mW average power. A simplified frame is shown in Fig. 1(b), and more details can be found in Ref. [13].

The dynamics and evolution of the quantum system can be described by optical Bloch equations for a three-level system interacting with femtosecond pulses. Here we label the fundamental, intermediate, and upper states as $|1\rangle$, $|2\rangle$, and $|3\rangle$, respectively. The Hamiltonian of the system is $H = H_0 + H_{\rm int}$, where H_0 is the Hamiltonian of the free atom. And $H_{\rm int}$ represents the interaction of the atom with the laser electric field $E(t)$; that is,

$$H_{\rm int} = -\mu_{12}E(t)|1\rangle\langle 2| - \mu_{23}E(t)|2\rangle\langle 3| + \text{H.c.}, \quad (1)$$

where $\mu_{i,j}$ denotes the dipole moment of the electrically allowed i-j transition. For the excitation scheme in Fig. 1(a), the electric field of the pulse train can be written as

$$E_1(t) = \left[\sum_{n=1}^{\infty} \epsilon(t - nT_r)e^{in\Phi_{R0}}\right] e^{i\omega_0 t}, \quad (2)$$

where $\epsilon(t - nT_r)$ is the electric field envelope of the nth pulse of the train, T_r is repetition period of the pulses, Φ_{R0} is the round-trip phase during a repetition period, and ω_0 is the laser's central frequency corresponding to 778 nm. By utilizing the efficient iterative numerical scheme proposed by D. Felinto

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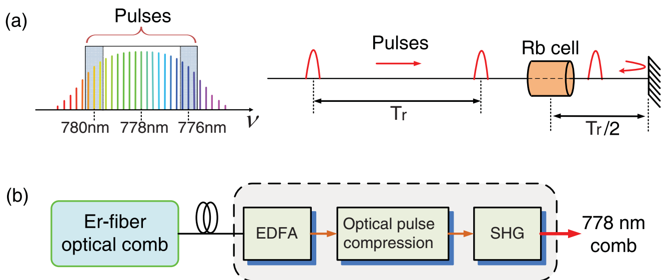

FIG. 1. (Color online) (a) Spectral components of the comb involved in the TPT excitation (left) and the regular experiment scheme (right) for TPT excitation using ultrashort pulses. (b) The optical control chain for second harmonic generation of the Er:fiber optical comb. EDFA, erbium-doped fiber amplifier; SHG, second harmonic generation.

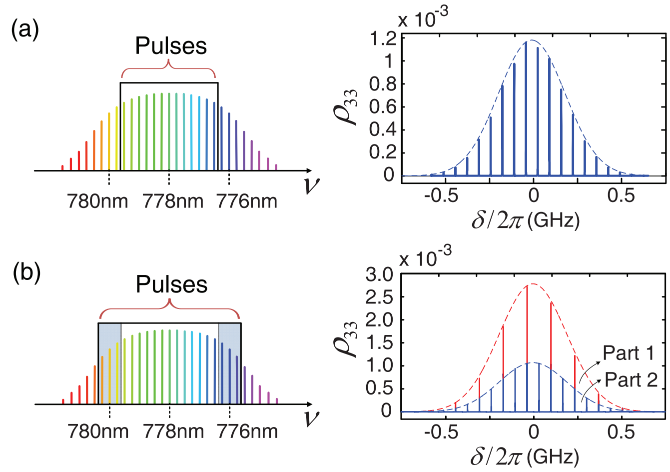

FIG. 2. (Color online) Two cases of pulses, (a) one excluding the comb lines around 780 nm and 776 nm and (b) the other incorporating these parts, and the corresponding contributions of the velocity groups of atoms to the TPTs. Part 1 and part 2 denote the contribution to the S-TPTs and D-TPTs, respectively.

et al. [14], we are able to simulate the temporal evolution of various elements of the atomic density matrix in the rotating-wave approximation.

D-TPTs and S-TPTs have different properties and excitation conditions. In D-TPTs, an Rb atom absorbs two photons at the same time, and is excited directly from the $5S$ state to the $5D$ state. This type of TPT requires the two photons, which come from one single unidirectional beam or two counterpropagating beams, should reach the atom "simultaneously," and from the point of the atom, the sum frequency of two corresponding comb lines (with frequencies ω_{L1} and ω_{L2}) should be equal to the $5S$-$5D$ transition frequency ω_t; i.e., $\omega_{L1} + \omega_{L2} = \omega_t$. In contrast, the S-TPT is a stepwise process. This means that two pulses which reach the atom at different times could participate in the two-step S-TPTs, provided that the atom has not decayed to the ground state before the second step. Our system can easily meet this requirement since the $\sim$7 ns repetition period of pulses is shorter than the 26 ns lifetime of the $5P_{3/2}$ state. And according to the S-TPT excitation condition, only the two comb lines (from either one copropagating comb or two counterpropagating combs) which are respectively resonant with the $5S$-$5P$ and $5P$-$5D$ transitions can act as a pair to excite the S-TPT of an Rb atom.

In this work, D-TPTs and S-TPTs can be excited by either one unidirectional beam or two counterpropagating beams. Besides, the velocity distribution of the hot atomic vapors will cause different influences on S-TPTs and D-TPTs, making the transition system more complex. All these factors would result in various effects on the TPT spectrum and should be well considered and analyzed to achieve an excellent DFCS of Rb TPTs.

In the case of D-TPTs excited by one unidirectional beam, the frequency of the "symmetric line" of the comb modes, i.e., each of the comb modes or the center of two adjacent modes, should be equal to half of the $5S$-$5D$ transition frequency. Nevertheless, for a moving atom in the hot vapor system, the whole comb and the "symmetric lines" are Doppler shifted due to its velocity. Hence, for a certain OFC, some velocity groups of Rb atoms satisfy the excitation condition, while others do not. Figure 2(a) illustrates the contribution of different atomic velocity groups to the ^{87}Rb $5S_{1/2}$ $(F = 2)$ to $5D_{5/2}$ $(F = 4)$ D-TPTs, where the atomic velocity is assumed to match the Gaussian distribution (the dash line), ρ_{33} is the $5D$ state population, and $\delta/2\pi$ is the corresponding Doppler shift of the velocity group. As $f_{\rm rep}$ ($\sim$144.5 MHz) is relatively small compared with the width ($\sim$0.5 GHz) of the Doppler broadening profile, when the comb is being scanned, there are always some comb-like velocity groups contributing to the D-TPTs and resulting in an almost constant Doppler broadening background [14]. Note that the interval between the comb-like lines corresponds to a Doppler frequency shift of $f_{\rm rep}/2$.

For the S-TPTs excited by one unidirectional beam, two of the comb modes are required to be resonant with the $5S$-$5P$ and $5P$-$5D$ transitions, respectively. Nevertheless, the Doppler shift effect of the mode pair should be taken into account in the hot vapor system. The result is, when $f_{\rm rep}$ is a subharmonic of the frequency difference $\Delta f_{1,2}$ between the two-step $5S$-$5P$-$5D$ transitions, i.e., $f_{\rm rep} = \Delta f_{1,2}/N$ (N is an integer), there are always some velocity groups of atoms to be excited by different mode pairs of the comb and contribute to the S-TPTs. Figure 2(b) shows the contributions of the atoms to the TPT intensity when $f_{\rm rep}$ is 144.563 MHz ($\Delta f_{1,2}/14612$). It is shown that the interval between the comb-like lines for S-TPTs (part 1), which corresponds to a Doppler frequency shift of $f_{\rm rep}$, is twice of that for D-TPTs (part 2). When $f_{\rm rep}$ is being scanned within a small region of several tens of hertz, the contribution of part 2 is also constant and results in constant background.

However, when the $f_{\rm rep}$ is far detuned from $\Delta f_{1,2}/N$, the two-resonance excitation condition is hardly satisfied for any group of atoms, which would decrease the S-TPT intensity [15]. Taking account of all the fourteen possible $5S_{1/2}$-$5P_{3/2}$-$5D_{5/2}$ ^{87}Rb S-TPT routes, in Fig. 3 we demonstrate the experimental results of the total Doppler broadening background variation by scanning $f_{\rm rep}$ for a range of 20 kHz. The result indicates that the Doppler broadening background is very strong when $f_{\rm rep}$ is close to the resonances of the fourteen routes. However, when $f_{\rm rep}$ is far from the resonance values,

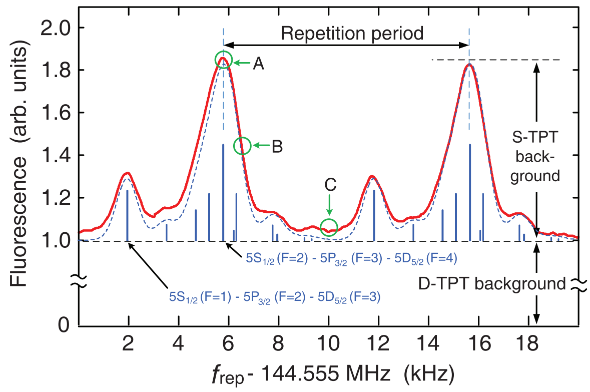

FIG. 3. (Color online) Variation of the Doppler broadening background with the f_{rep} scanned around 144.555 MHz. The contribution of D-TPTs performs a constant background, while the contribution of the S-TPTs varies greatly with the f_{rep}. The red lines show the measured background intensity. The blue bars demonstrate the calculated strengths and positions of the resonant S-TPT lines, and the dashed blue line shows the corresponding simulated contribution of these bars to the S-TPT background intensity.

the S-TPT background is suppressed to a relatively low level. This property can be used to suppress the S-TPT background.

D-TPTs and S-TPTs excited by the two counterpropagating beams generate the Doppler-free TPT signals. So the excitations of the Doppler-free TPTs require that the comb's "symmetric line" should be equal to half the $5S$-$5D$ transition frequency, whatever the velocity of the atoms. This is a common excitation condition for both Doppler-free D-TPTs and S-TPTs. It should be mentioned that f_{rep} is not required to be $\Delta f_{1,2}/N$ here for Doppler-free S-TPTs. The reason is that, from the point of a moving atom, the sum frequency of the counterpropagating photons remains almost constant, while the difference frequency between them changes with the atom's velocity. This property is opposite to that of the S-TPTs excited by one unidirectional beam. So once the "symmetric line" condition is satisfied, whatever the f_{rep}, there are always some comb-like distribution velocity groups of atoms being excited by corresponding mode pairs and contributing to the Doppler-free S-TPT signals. It is worth mentioning that the Doppler-free D-TPTs occur in the limited region where counterpropagating pulses overlap, while the Doppler-free S-TPTs occur along the whole laser path within the cell with the "stepwise" property.

According to above analysis, we use different methods to eliminate the D-TPT and S-TPT Doppler broadening background. To eliminate the D-TPT background, we utilize the pulse spectrum splitting method [11] as demonstrated in Fig. 4, where each pulse is split into two subpulses with a time delay τ between them. The corresponding electric field in this case can be described as

$$E_2(t) = \left[\sum_{n=1}^{\infty} \epsilon_1(t - nT_r)e^{in\Phi_{R1}}\right] e^{i\omega_1 t} + \left[\sum_{n=1}^{\infty} \epsilon_2(t - \tau - nT_r)e^{in\Phi_{R2}}\right] e^{i\omega_2(t-\tau)}, \quad (3)$$

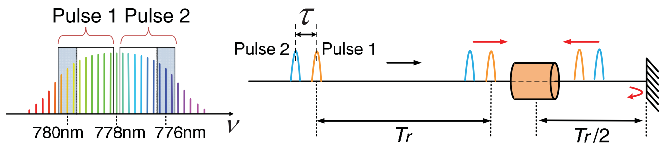

FIG. 4. (Color online) TPT excitation scheme using spectrum split pulses. Each of the pulses is split into two subpulses with >778 nm and <778 nm spectral components, respectively, and a time delay τ is introduced between the first and second subpulses.

where $\epsilon_1(t - nT_r)$ and $\epsilon_2(t - \tau - nT_r)$ are the electric field envelopes of the first and second subpulses of the nth pulse, respectively, and $\omega_1(\omega_2)$ corresponds to the laser's central frequency 779 nm (777 nm) of the first (second) subpulse. In this way, the unidirectional beam-excited D-TPTs could not take place anymore because the two photons no longer reach the atoms simultaneously. However, this method does not work effectively for the S-TPT background for the "stepwise" property. For the S-TPT background, it can be eliminated by tuning the f_{rep} far from the resonance values, as we discussed and demonstrated in Fig. 3.

To illustrate the effects of the two schemes for background elimination, Fig. 5 shows the simulated and experimental background intensity versus the time delay τ for different f_{rep}. When τ is smaller than the pulse duration, the photons from the <778 nm and >778 subpulses could reach the Rb atoms simultaneously and hence excite the D-TPTs. Especially when $\tau = 0$, the two subpulses' electrical field have the strongest constructive interference, which would result in a rather high TPT intensity [16]. For these two reasons, each of the curves reaches its maximum at $\tau = 0$. When τ is detuned far from zero, i.e., $|\tau| > 3$ ps, the two subpulses do not overlap anymore, and as a result, the D-TPTs cannot take place correspondingly. In this case, the background mainly comes from the S-TPTs. Comparison between the three curves (A, B, and C), which correspond to the cases marked in Fig. 3, indicates that the farther we tune f_{rep} from the resonance values for S-TPTs, the more background will be eliminated. Besides, the background performs a relatively lower intensity when

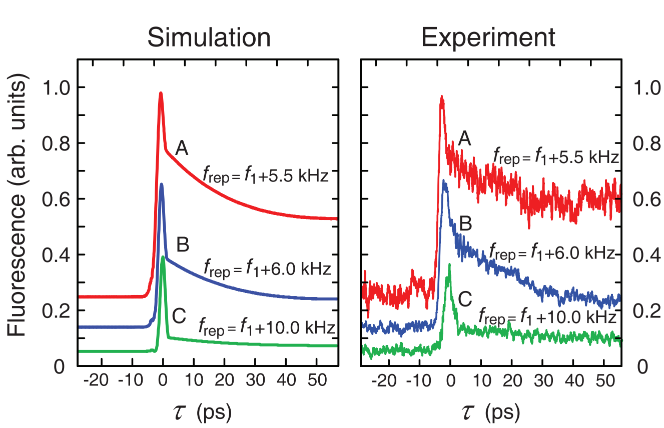

FIG. 5. (Color online) (a) Simulated and (b) experimental Doppler broadening background intensity versus the time delay τ between the two subpulses at different f_{rep} ($f_1 = 144.555$ MHz). A, B, and C correspond to the three f_{rep} cases as marked in Fig. 3.

τ is negative than when it is positive. This is because when $\tau < 0$, the $5P$ population has been decaying for a time of $T_r - \tau$ ($\sim$7 ns) rather than τ (several ps) before the $5P$-$5D$ transition, which results in a lower S-TPT intensity. Therefore, the D-TPTs and S-TPTs background can be respectively eliminated by introducing a negative time delay τ between the subpulses and choosing a suitable f_{rep}, and a combination of the two methods enables a more than 95% suppression of the total Doppler broadening background.

It is worth mentioning that these two background suppression methods have no qualitative influence on the intensity of Doppler-free D-TPTs and S-TPTs signals, except that the peak intensity of the pulses is reduced after being split. In this scheme, the Doppler-free D-TPTs take place at the two overlapping spots of counterpropagating subpulses with different spectra, while the Doppler-free S-TPTs take place along the whole laser path and their intensity would not decrease when choosing a f_{rep} far from the resonance values, because the two-resonance excitation condition becomes a "symmetric line" condition in hot vapor systems, as we discussed before.

In order to obtain a high quality TPT spectrum of ^{87}Rb with Doppler broadening background eliminated, we choose τ to be -20 ps, and f_{rep} to be around 144.565 MHz (case C in Fig. 5). By scanning f_{rep} and detecting the 420 nm $6P$-$5S$ transition fluorescence, we obtained the TPT spectrum as illustrated in Fig. 6. Compared with the $5S_{1/2}$-$5D_{3/2}$ TPT lines, the $5S_{1/2}$-$5D_{5/2}$ ones have much higher strengths and all of them can be clearly distinguished in the spectrum. The Doppler broadening background is greatly suppressed with over 95% efficiency and remains in a quite low level with respect to the Doppler-free TPT signals. For instance, the strength of the $5S_{1/2}$ ($F = 2$) to $5D_{5/2}$ ($F = 4$) line is 50 times stronger than the background. Each of the transition lines performs a linewidth of $\sim$2 MHz, which is larger than the natural linewidth ($\sim$300 kHz). This linewidth broadening is due to the residual Doppler broadening, transit time broadening, and laser power broadening.

Also we can see in the figure that the observed TPT spectrum repeats itself every time the f_{rep} is scanned $\sim$27 Hz, which corresponds to a repetition of $f_{\mathrm{rep}}/2$ at the 778 nm optical frequency band. For the Doppler free D-TPTs, the "symmetric line" resonance condition is satisfied every time the comb's optical frequency sweeps for $f_{\mathrm{rep}}/2$. So its contribution to the TPT spectrum has a repetition period of $f_{\mathrm{rep}}/2$, and a similar result can be found in Refs. [10,12]. With respect to the Doppler-free S-TPTs, as the excitation condition reduces to the "symmetric line" resonance condition in the hot vapor system, it also illustrates a repetition period of $f_{\mathrm{rep}}/2$. In contrast, in a cold atomic system, the excitation of S-TPTs requires that one pair of comb modes should precisely equal the two stepwise excitation frequencies respectively, which results in a repetition period of f_{rep} [1].

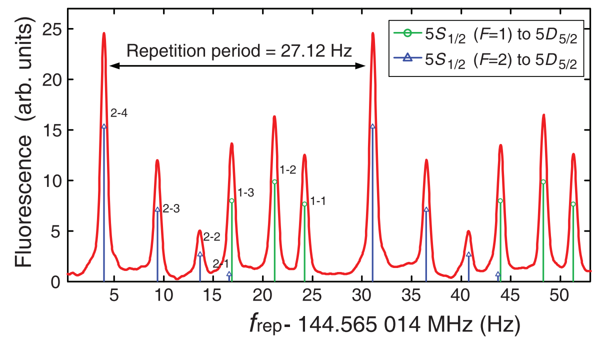

FIG. 6. (Color online) The observed spectrum of ^{87}Rb TPTs (red line) with f_{rep} scanned near 144.565 MHz. The blue and green bars show the calculated strengths and center positions of the TPT transition lines. The label "i-j" denotes the $5S_{1/2}$ ($F = i$) to $5D_{5/2}$ ($F = j$) TPT transition.

In conclusion, we have demonstrated the observation of the Rb $5S$-$5D$ TPT spectrum excited directly by a frequency-doubled Er:fiber laser comb in a hot vapor system. Both of the D-TPTs and S-TPTs are incorporated and contribute to the total transitions, which could take full advantage of the frequency-doubled comb power and enhance the total TPT signals. Various influences of the types of TPTs and atomic velocity distribution on the TPT spectrum are theoretically analyzed and simulated. By combining two techniques of comb spectral control, i.e., introducing a time delay between two spectral parts of the pulses and detuning the comb f_{rep} far from resonance values, we obtain high-resolution DFCS with Doppler broadening background effectively eliminated. This paves the way for further establishing a high-stability OFC at optical communication bands. These techniques would benefit the high-resolution spectroscopy measurement and quantum coherent control in hot atomic vapor systems.

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