A direct frequency comb for two-photon transition spectroscopy in a cesium vapor*

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(Received 9 January 2012; revised manuscript received 4 June 2012)

A phase-stabilized femtosecond frequency comb is used to measure high-resolution spectra of two-photon transition $6^2S_{1/2}$-$6^2P_{1/2,3/2}$-$8^2S_{1/2}$ in a cesium vapor. The broadband laser output from a femtosecond frequency comb is split into counter-propagating parts, shaped in an original way, and focused into a room-temperature cesium vapor. We obtain high-resolution two-photon spectroscopy by scanning the repetition rate of femtosecond frequency comb, and through absolute frequency measurements.

Keywords: broadband laser, two-photon spectroscopy, femtosecond frequency comb

PACS: 37.10.De, 34.35.+a, 34.10.+x **DOI:** 10.1088/1674-1056/21/11/113701

1. Introduction

A variety of techniques have been implemented to eliminate Doppler effects in optical frequency measurements of atomic transitions. These include saturated absorption spectroscopy in vapor cells,[1,2] magneto–optic traps,[3] thermal atomic beams,[4,5] and two-photon spectroscopy in vapor cells.[6,7] The two-photon spectroscopy experiments in vapor cells are not only easy to perform but also precise. The resonantly enhanced Doppler-free two-photon high-resolution precision spectroscopy in an atomic vapor cell was first discussed and experimentally demonstrated in Na by Bjorkholm and Liao.[6,7] In previous measurements of two-photon transitions in Cs, the final uncertainty was limited by such systematic effects and optical frequency measurement techniques. In our experiments, we use a femtosecond frequency comb to eliminate systematic effects and improve frequency measurement. The development of the femtosecond frequency comb has revolutionized precision spectroscopy and fundamental physics owing to its ability to provide a precise and direct link between microwave and optical frequencies.[8,9] Serving as a frequency counter or clockwork, an optical frequency comb can be used directly for spectroscopy, which is thus named direct frequency comb spectroscopy (DFCS).[10] In particular, the spectrum versatility provided by a modern frequency combs allows simultaneous study of multiple atomic transitions with a single experimental setup.

Precision measurement techniques have provided us with new ways to test fundamental theories in atomic physics. Measurements of nuclear structure and tests of the fine structure constant as well as the weak interaction are possible using accumulated information about transition amplitudes and frequencies obtained through precision measurements.[11−14] Cesium, being one of the most thoroughly studied heavy atoms, is very suitable for these investigations, because the accuracy of atomic theory in this system is on the order of 1%.[15] In this respect, measurements of transition frequencies play a very important role because the experimental accuracy can be many orders higher than that obtained through atomic structure calculations.

In this work, we applied the DFCS to determine absolute atomic transition frequencies within the comb bandwidth for two-photon processes. We used an octave-spanning femtosecond comb for precision

*Project supported by the National Basic Research Program of China (Grant No. 2012CB921603), the International Science & Technology Cooperation Program of China (Grant No. 2011DFA12490), the International Science & Technology Cooperation Program of Shanxi Province, China (Grant No. 2011081030), the National Natural Science Foundation of China (Grant Nos. 61008012, 10934004, 60978001, 11174187, and 11074154), the NSFC Project for Excellent Research Team of China (Grant No. 61121064), and the Specialized Research Fund for the Doctoral Program of Higher Education, China (Grant No. 20101401120004).

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spectroscopy in cesium atomic vapor, and used a self-referenced optical frequency comb to excite Doppler-free two-photon transitions from the ground state to $8^2S_{1/2}$ state in cesium. The center-of-gravity frequency is measured to be 729009798.923(32) MHz, which is consistent with previous measurements on the $8^2S_{1/2}$ state.[16,17] The technique of velocity-selective two-photon excitation to measure the center-of-gravity frequency for the $8^2S_{1/2}$ state proved to be a robust method to effectively improve the measurement accuracy of multiple transitions. The versatility of the DFCS is demonstrated.

2. Experimental setup

Figure 1 shows an energy level diagram of the hyperfine levels involved in the $6^2S_{1/2}$–$6^2P_{1/2,3/2}$–$8^2S_{1/2}$ transitions in ^{133}Cs atoms. We detect the two-photon excitation as blue fluorescence emission as the atom relaxes from the $7^2P_{1/2,3/2}$ state to the ground state.

The experimental setup is schematically shown in Fig. 2. The sample of cesium atoms was prepared in a cesium vapor cell at room temperature. We excite the cascaded two-photon transition using a self-referenced femtosecond frequency comb (Menlo systems: FC1500 optical frequency synthesizer). The wavelength range we used of the output spectrum extends from 300 to 1000 nm.

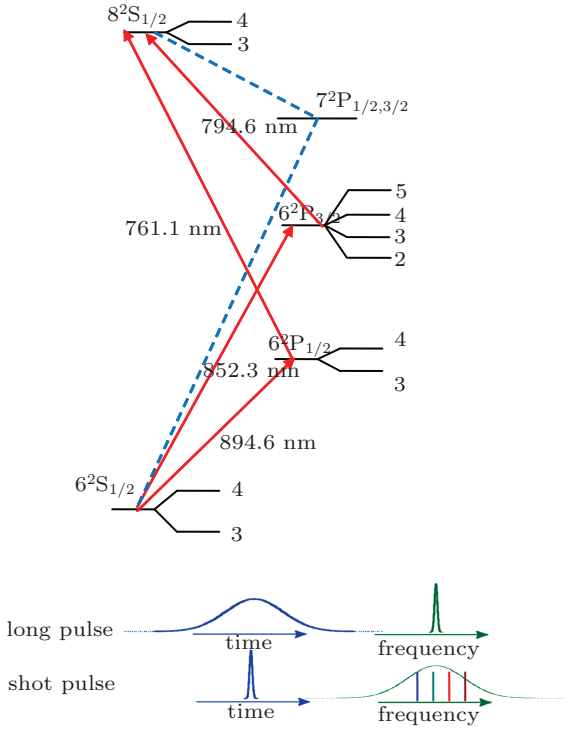


Fig. 1. (colour online) (a) Energy levels of ^{133}Cs (I=7/2). Solid lines: excitation pathways; dashed lines: decay channels used in the detection. (b) Diagram of frequency comb.

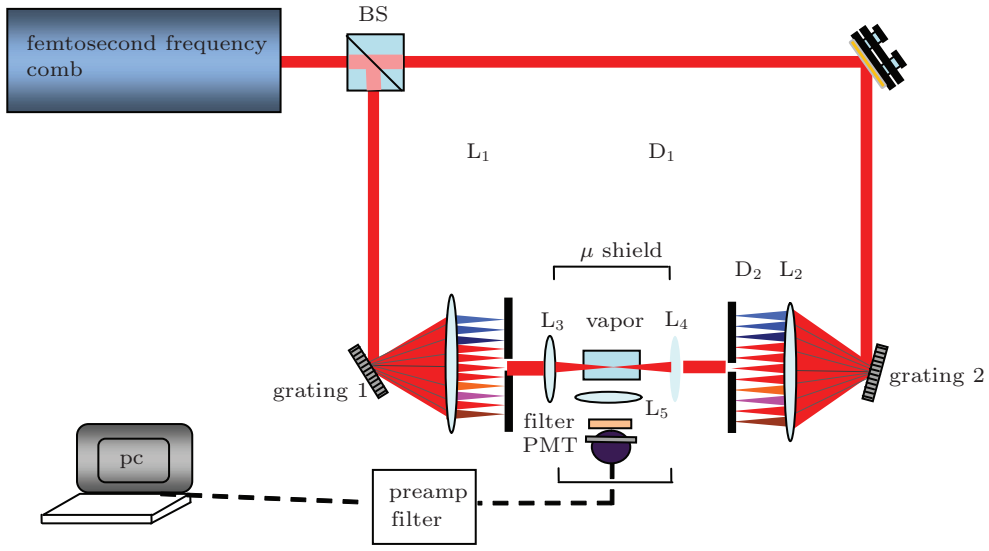


Fig. 2. (colour online) Experimental setup for two-photon transition in cesium atoms at room temperature in a Cs vapor cell. BS denotes the non-polarizing beam splitter. L_1–L_5 are convex Lens. D_1 and D_2 are diaphragms.

The optical frequency comb provides a precise and direct link between microwave and optical frequencies.[1,2] The time-domain Fourier transform of the frequency domain (see Fig. 1(b)) is expressed as

$$E(t) = \sum_n E_{\rm env}(t - n\tau)e^{i(\omega_c t - n\omega_c \tau + n\Delta\phi + \phi_0)} = \sum_n E_{\rm env}(t - n\tau)e^{i(\omega_c t + n(\Delta\phi - \omega_c \tau) + \phi_0)}, \quad (1)$$

where $E_{\rm env}(t)$ is the envelope, ω_c is the carrier fre-

quency, ϕ_0 is the overall phase offset, and τ is the time between two adjacent pluses.

The inverse transform of Eq. (1) is given by

$$\begin{aligned} E(\omega) &= \int \sum_n E_{\rm env}(t-n\tau)\mathrm{e}^{\mathrm{i}[\omega_{\rm c}t+n(\Delta\phi-\omega_{\rm c}\tau)+\phi_0]} \\ &\quad \times \mathrm{e}^{-\mathrm{i}\omega t}\mathrm{d}t \\ &= \sum_n \mathrm{e}^{\mathrm{i}[n(\Delta\phi-\omega_{\rm c}\tau)+\phi_0]} \\ &\quad \times \int E_{\rm env}(t-n\tau)\mathrm{e}^{-\mathrm{i}[(\omega-\omega_{\rm c})t]}\mathrm{d}t, \qquad (2) \\ \widetilde{E}(\omega) &= \int E_{\rm env}(t)\mathrm{e}^{-\mathrm{i}\omega t}\mathrm{d}t, \\ \int f(x-a)\mathrm{e}^{-\mathrm{i}\alpha x}\mathrm{d}x &= \mathrm{e}^{-\mathrm{i}\alpha a}\int f(x)\mathrm{e}^{-\mathrm{i}\alpha x}\mathrm{d}x, \\ E(\omega) &= \sum_n \mathrm{e}^{\mathrm{i}[n(\Delta\phi-\omega_{\rm c}\tau)+\phi_0]}\mathrm{e}^{-\mathrm{i}n(\omega-\omega_{\rm c})\tau}\widetilde{E}(\omega-\omega_{\rm c}) \\ &= \mathrm{e}^{\mathrm{i}\phi_0}\sum_n \mathrm{e}^{\mathrm{i}(n\Delta\phi-n\omega\tau)}\widetilde{E}(\omega-\omega_{\rm c}). \qquad (3) \end{aligned}$$

In a femtosecond resonant cavity, the phase change is an integer multiple of 2π

$$\begin{aligned} &\Delta\phi-\omega_n\tau = 2n\pi, \\ &f_0 = \Delta\phi/2\pi\tau, \quad f_{\rm rep} = 1/\tau, \\ &f_n = f_0 + nf_{\rm rep}, \qquad (4) \end{aligned}$$

where ω_n is the carrier frequency, and $\Delta\phi$ is the pulse-to-pulse carrier envelope phase shift. Thus, the frequency of each optical mode $f_{\rm n}$ can be related to two microwave frequencies: the carrier-envelope offset frequency f_0, and the laser repetition rate $f_{\rm rep}$. The parameter n is an integer mode number smaller than 5×10^6. The carrier-envelope-offset frequency is stabilized by an f-to-$2f$ interferometer, thus self referencing all modes of the comb. The offset frequency f_0 was stabilized to 20 MHz. The repetition rate of the femtosecond frequency comb is almost 250 MHz.

The broad band laser beam from the femtosecond frequency comb was split by a 50%–50% nonpolarizing beam-splitting (BS) cube. The two output laser beams were linearly polarized along the same axis, overlapped with each other, and propagated inversely. The broadband laser beam was expanded by two gratings (1400 lines), and the special wavelength was chosen by a diaphragm and a spectrometer. The counter-propagating beams were focused by lenses of 10-cm focal length to a size of about 200 μm inside the vapor cell. The Earth's magnetic field was cancelled by wrapping three layers of a μ-metal sheet around the Rb cell.[18,19]

Two-photon resonance signals were monitored by measuring $7^2\mathrm{P}_{1/2,3/2}$–$6^2\mathrm{S}_{1/2}$ fluorescence at 459 nm and 455 nm. The fluorescence collected by a convex lens at a right angle to the counter-propagating beams was detected by using a photomultiplier tube (PMT CR131). The detector avoided background light and chose the light from the excitation laser beams by a 450-nm filter with a 10-nm pass band. The detector output was amplified and filtered (mini-circuits amplifier@500 MHz), and then sampled and recorded by a computer. The femtosecond frequency comb controlled the scan of the repetition rate by stepping the frequency of the signal generator to which the repetition rate was locked. Scan data were collected by changing the frequency of signal generator from 249.990645 to 249.991045 MHz with a step of 1 Hz. The repetition rate of 400 Hz results in a variation of the two-photon frequency of 1–2 Hz.

3. Spectral theory

We can use the second-order time-dependent perturbation theory to analyze the two-photon transition spectra. We start by considering the transition from the ground state $6^2\mathrm{S}_{1/2}$ to an excited state with a total angular momentum F in cesium atom with two continuous-wave (cw) lasers with angular frequencies ω_1 and ω_2, intensities I_1 and I_2, and wave vectors $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$. Here n refers to the principal quantum number of the excited state. We further assume that ω_1 is close to the resonant frequency for the $6\mathrm{S}_{1/2}$–$6\mathrm{P}_{J'}$ transition, where J' (= 1/2, 3/2) is the angular momentum quantum number. The transition probability of the atom is given by[20]

$$\begin{aligned} &P(6\mathrm{S}_{1/2}F, n''L_{J''}F'') \\ &\propto \frac{1}{2F+1}\frac{I_1I_2}{[\omega 6\mathrm{S}_{1/2}F : n''-(\omega_1+\boldsymbol{K}_1\cdot\boldsymbol{V})-(\omega_1+\boldsymbol{K}_1\cdot\boldsymbol{V})]^2+\left(\frac{\gamma_{n''L_{J''}}}{2}\right)^2} \\ &\times \sum_{M_F,M_{F''}}\left|\sum_{F',M_{F''}}\frac{\langle n''L''_{J''}F''M''_F\,|\boldsymbol{e}_1\cdot\boldsymbol{d}|\,6\mathrm{P}_{J'}F'M'_{F'}\rangle\,\langle 6\mathrm{P}_{J'}F'M'_F\,|\boldsymbol{e}_1\cdot\boldsymbol{d}|\,6\mathrm{S}_{1/2}FM_F\rangle}{\omega 6\mathrm{S}_{1/2}F : 6\mathrm{P}_{J'}F'-(\omega_1+\boldsymbol{K}_1\cdot\boldsymbol{V})-\mathrm{i}\dfrac{\gamma 6\mathrm{P}_{J'}}{2}}\right|^2, \qquad (5) \end{aligned}$$

where V is the velocity of the atom, M_F, M'_F, and M''_F are the projections of the total angular momenta F, F', and F'' along the axis of quantization, $\boldsymbol{d}$ is the electric dipole operator, $\boldsymbol{e}_1$ ($\boldsymbol{e}_2$) is the unit vector along the polarization direction for the first (second) laser beam, $\gamma_{n''}L_{J''}/2$ is the homogeneous linewidth of the state $|nL_J\rangle$, and $\omega_{nL_JF:n'L'_{J'}F'}$ is the resonant angular frequency of the transition $|nL_JF\rangle \rightarrow |n'L'_{J'}F'\rangle$. We calculated the excitation probabilities by numerically integrating Eq. (5) over a Gaussian velocity distribution for optical modes in the near-resonant condition. The calculated spectra agree quite well with the experimental spectra.

The resonant frequencies for a transition from the ground state with total angular momentum F to an excited state with total angular momentum F'' are related to the center-of-gravity frequency and the hyperfine coupling constant[21]

$$V_{6S_{1/2}F:n''L''J''F''} = V_{6S_{1/2}F:n''L''J''F''} - \frac{1}{2}A_{6S_{1/2}}k(F,J) + \frac{1}{2}A_{n''L''J''}k(F'',J'') + B_{n''L''_{J''}}\frac{\frac{3}{4}k(F'',J'')^2+\frac{3}{4}k(F'',J'')-I(I+1)J''(J''+1)}{2I(2I-1)J''(2J''-1)}, \quad (6)$$

where $k(F,J) = F(F+1) - I(I+1) - J(J+1)$. For $6S_{1/2,3/2}$ state of ^{133}Cs $I = 7/2$. The repetition rate at which the calculated resonances occur depends on the center-of-gravity frequencies in the two stages of the excitation and the hyperfine constants A and B of the ground, intermediate, and final states. Spectra were calculated with different values of the two-photon center-of-gravity frequency and hyperfine coupling constants of the final excited state.

4. Experimental observations and discussion

Figure 3 shows the spectrum collected as the repetition rate of the frequency comb was scanned without use of gratings 1 and 2, convex lens $L_{1,2}$, and diaphragm $D_{1,2}$ when all broadband lasers (300–1000 nm) are working in two photon excitation. Hence, the spectrum not only comes from the excitation to 8S state but also from the excitations to 9S, 7D, 10S, 8D, and 6D. The spectrum consists of many narrow Doppler-free peaks and a Doppler-broadened back ground. The narrow peaks are due to the excitations to many states of a cesium atom via two counter-propagating photons, while the Doppler-broadened background is due to the excitations by two co-propagating photons. The observed spectrum is quite complicated because of numerous discrete frequencies present in the comb, combined with the multiple atomic energy levels and accessible transition pathways.

The spectrum is greatly simplified and the Doppler background is eliminated by introducing gratings 1 and 2, convex lens $L_{1,2}$, and diaphragm $D_{1,2}$, as shown in Fig. 2.

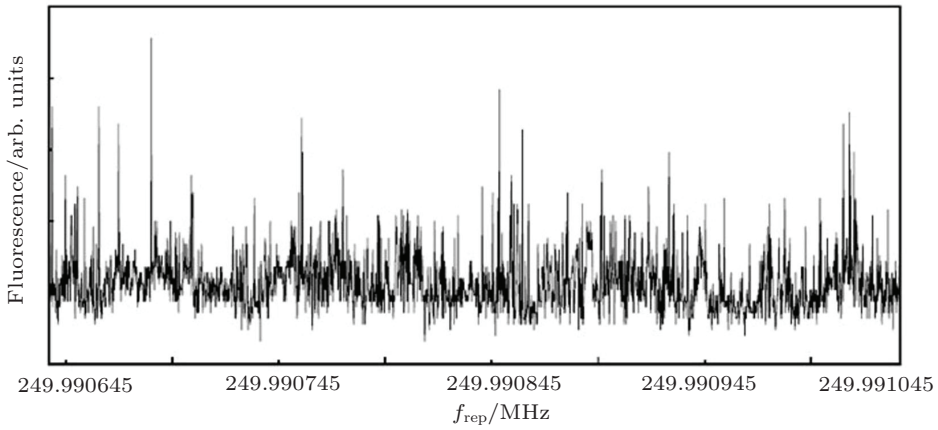


Fig. 3. Fluorescence spectrum collected by scanning the repetition rate of the frequency comb without using gratings 1 and 2, convex lens $L_{1,2}$, and diaphragm $D_{1,2}$.

The use of the expanded equipment of a broadband laser beam restricts the transitions and enables the selection of individual excitation pathways. Figure 4 shows that the fluorescence of one of the two counterpropagating laser beams is restricted between 850 and 855 nm and the other between 790 and 800 nm. The first beam can only excite the $6^2S_{1/2}$–$6^2P_{3/2}$ transition at 852 nm (the D_2 transition of cesium) and the second beam can only excite the $6^2P_{1/2}$–$8^2S_{1/2}$ transition at 795 nm . This technique uses a grating and a diaphragm to eliminate the excitations to 9S, 7D, 10S, 8D, and 6D states as well as the Doppler-broadened background. Each spectral peak is the result of excitation from a given ground-state hyperfine level to a given excited-state hyperfine level, and individual peaks in the group correspond to specific transition pathways through every allowable hyperfine level of the intermediate state. We cannot observe the transition from $F = 3$ states to $F = 4$ states because the power of each mode of the broadband laser is lower. Based on Eq. (4), the center-of-gravity frequency is found to be 729009798.923(32) MHz, which is consistent with previous measurements on the $8S_{1/2}$ state (729009799.020(26) MHz).[16,17]

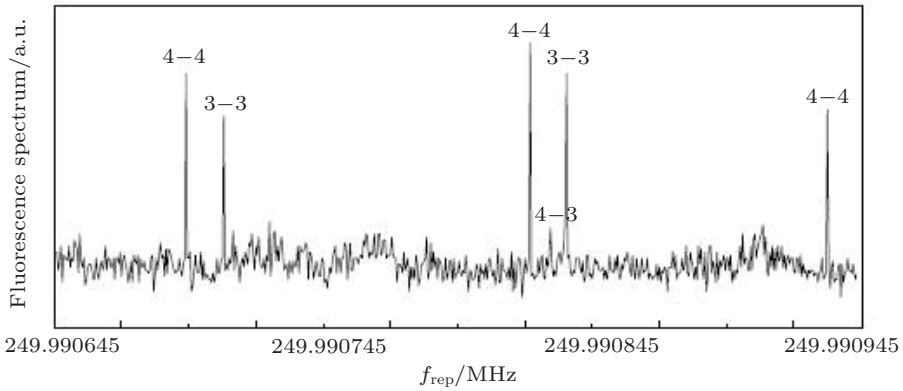


Fig. 4. Fluorescence spectrum for $7^2P_{1/2,3/2}$–$6^2S_{1/2}$ when Cs atoms are excited from $6^2S_{1/2}$ state to $8^2S_{1/2}$ state. The group of peaks are labeled by F–F'' to denote the hyperfine ground states and hyperfine excited states involved in the transition. The peaks of each ground state correspond to different hyperfine components of the intermediate $6^2P_{3/2}$ state.

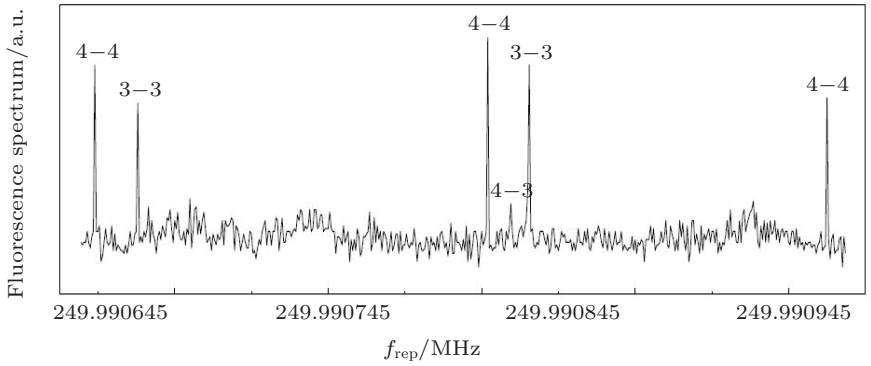


Fig. 5. Fluorescence spectrum for $7^2P_{1/2,3/2}$–$6^2S_{1/2}$ when Cs atoms are excited from $6^2S_{1/2}$ state to $8^2S_{1/2}$ state. The group of peaks are labeled by F–F" to denote the hyperfine ground state and hyperfine excited states involved in the transition. The peaks of each ground state correspond to different hyperfine components of the intermediate $6^2P_{1/2}$ state.

Figure 5 shows that the fluorescence of one of the counter-propagating laser beams is restricted between 890 and 900 nm, and the other between 750 and 770 nm. The first beam can only excite the $6^2S_{1/2}$–$6^2P_{1/2}$ transition at 895 nm (the D_1 transition of cesium), and the second beam can only excite the $6^2P_{1/2}$–$8^2S_{1/2}$ transition at 761 nm. This technique uses a grating and a diaphragm to eliminate the excitations to 9S, 7D, 10S, 8D , and 6D states, as well as the Doppler-broadened background. Each spectral

peak is the result of excitation from a given ground-state hyperfine level to a given excited-state hyperfine level, and individual peaks in the group correspond to specific transition pathways through every allowable hyperfine level of the intermediate state. We cannot find the transition from $F = 3$ states to $F = 4$ states because the power of each mode of broadband laser is lower. By means of Eq. (4), the center-of-gravity frequency is found to be 729009798.909(42) MHz, which is consistent with previous measurements[16,17] for the $8S_{1/2}$ state (729009799.020(26) MHz).

5. Conclusion

We have obtained two-photon spectroscopy of cesium atoms in a cesium vapor cell at room temperature by detecting fluorescence signal of 7P–6S from $6^2S_{1/2}$–$6^2P_{1/2,3/2}$–$8^2S_{1/2}$ excitation. Our measured center-of-gravity transition frequency (729009798.923(32) MHz) is in good agreement with previous measurements, even with higher accuracy for the $8^2S_{1/2}$ state. In the measurement, the uncertainty are limited mainly by signal-to-noise (SNR) ratio,[22] and ac stark shift factors.

The two-photon spectroscopy in the cesium vapor cell at room temperature based on a femtosecond frequency comb proves to be a robust method to effectively improve the measurement sensitivity of atomic energies and hyperfine constant A. We can carry out further research using this technique, such as measurements of 9S, 7D, 10S, 8D, and 6D of the cesium atom, as well as energy levels of alkali atoms such as Rb, Li, and Ca. In particular, the DFCS spectral versatility provided by the femtosecond frequency comb allows the simultaneous study of multiple atomic transitions in only one experimental process.

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