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Investigation on the Monochromatic Two-Photon Transition Spectroscopy of Rubidium by Using Intensity Modulation Method

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We experimentally study the high-resolution two-photon spectroscopy of rubidium using a single laser. The $5S_{1/2} - 5P_{3/2} - 5D_{5/2}$ transition is realized via 778.1 nm laser beams acting on the vapor with a counter-propagating configuration. The intensity modulation method is employed to improve the signal-to-noise ratio of the spectrum by two orders of magnitude compared with that for direct measurement. The effects of the vapor temperature, the frequency-scanning speed of the laser, the polarization combination of the laser beams, and the laser power on the spectra are investigated to obtain the optimal experimental parameters. This research is expected to be beneficial for potential applications since it can be used to realize a compact and high-precision frequency standard for optical communication.

1. Introduction

Precision spectroscopy has great prospects in determining fundamental physical constants,[1,2] laser frequencies,[3] atomic structures,[4] and so forth.[5] However, the existence of atomic natural linewidth, motion, and collisions broadens the spectrum linewidth,[6] which greatly limits the development of high-resolution spectroscopy. Among all the factors that broaden the spectrum linewidth, the reduction or elimination of the Doppler effect is the most effective and accessible way to increase the spectroscopy resolution via various methods, such as polarization spectroscopy,[7] saturated absorption spectroscopy,[8] and two-photon spectroscopy.[9]

The $5S_{1/2} - 5P_{3/2} - 5D_{5/2}$ two-photon transition of the rubidium atom has been extensively studied owing to its large transition probability, its superior Doppler free background[10] caused by the small difference between the real energy level and the virtual energy level, and its potential application as a frequency standard[11] due to its relatively narrow natural linewidth and low sensitivity to the external environment. The traditional method of two-photon transition spectroscopy is achieved by two laser beams coupling with the real energy levels, which was well studied by Moon and co-workers. They illustrated the on-resonant and off-resonant two-photon absorption in a $5S_{1/2} - 5P_{3/2} - 5D_{5/2}$ ladder-type atomic system in ^{87}Rb atoms[12] and the relationship between two- and three-photon coherence in terms of the transition routes.[13] With the development of the optical frequency comb, the accuracy and stability of the frequency have provided two-photon spectroscopy with a new research platform. Martin et al. studied a compact optical atomic clock based on a $5S_{1/2} - 5P_{3/2} - 5D_{5/2}$ two-photon transition in rubidium with an optical frequency comb.[14] Lopez et al. studied coherent blue emission generated by $5S_{1/2} - 5P_{3/2} - 5D_{5/2}$ two-photon excitation by using diode and femtosecond lasers.[15] However, the above-mentioned system is complicated and expensive, making it very difficult to integrate. As a result, a simple and effective method of two-photon transition spectroscopy is required for research applications of optical frequency standards,[16] and optical communication.[17]

The two-photon transition spectroscopy of the $5S_{1/2} - 5P_{3/2} - 5D_{5/2}$ transition by a single laser of 778.1 nm is expected to benefit from convenient system integration and better elimination of the Doppler effect due to the absolutely same laser frequency. However, the small transition probability compared with that when using real energy levels will result in a low signal-to-noise ratio (SNR) of the spectrum, which is clearly an obstacle to research.

In this paper, we report high-resolution $5S_{1/2} - 5P_{3/2} - 5D_{5/2}$ two-photon transition spectroscopy in the rubidium atom excited by a single 778.1 nm laser. Both experimental and theoretical studies are performed to investigate the influence of experiment parameters, such as the vapor temperature, the laser scanning speed, the polarization combination of laser beams, and the laser power. Furthermore, the improved SNR of the spectrum is obtained by an intensity modulation method. This research provides a solid experimental basis for potential applications based on two-photon spectroscopy.

2. Experiment Setup

The related energy levels are shown in Fig. 1(a). The transitions from the ground state ($5S_{1/2}$) to the intermediate excited state ($5P_{3/2}$) and from the intermediate excited state to the high excited state ($5D_{5/2}$) correspond to 780 and 776 nm, respectively. The $5S_{1/2}$ and $5D_{5/2}$ levels can also be coupled with two 778.1 nm laser beams via a virtual energy level, which is labeled with a dashed line in Fig. 1(a). The unstable excited atoms will spontaneously radiate to the $6P_{3/2}$ level and then to the $5S_{1/2}$ ground level with 420 nm fluorescence emission. The fast spontaneous radiation of $5D_{5/2} - 6P_{3/2}$ can be ignored; thus, the 420 nm fluorescence can represent the population of the $5D_{5/2}$ state.[18]

The experimental setup is shown in Fig. 1(b). The laser is a Littrow external cavity diode laser (Toptica DL pro) with a tunable range of 15 nm and a linewidth of 2 MHz. Two counter-propagating laser beams acting on rubidium vapor are achieved by a high-reflection mirror (M). The laser intensity is modulated by a chopper wheel (Stanford Research Systems SR540). A quartz tube of 100 mm length and 25 mm diameter is filled with rubidium vapor, which is shielded with a μ-metal to reduce the effect of any stray magnetic field. The temperature of the vapor can be accurately controlled by a self-feedback system. The waists

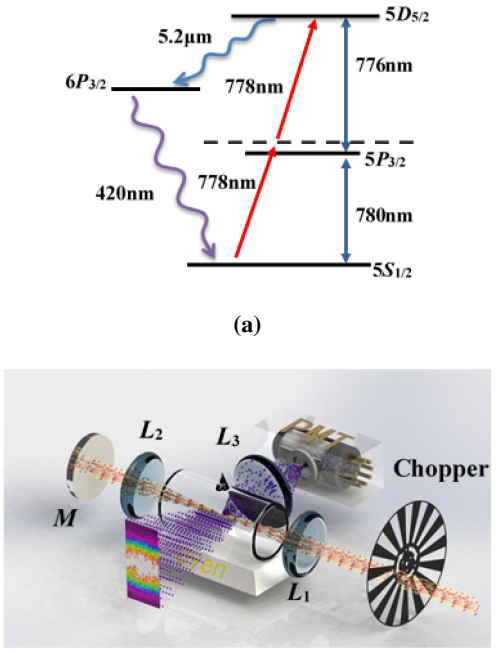

Fig. 1. (Color online) (a) Energy levels of the experiment system. The solid lines are the real energy levels and the dashed line represents the virtual energy level. (b) Experimental setup. L, lens; M, high-reflection mirror; PMT, photomultiplier tube.

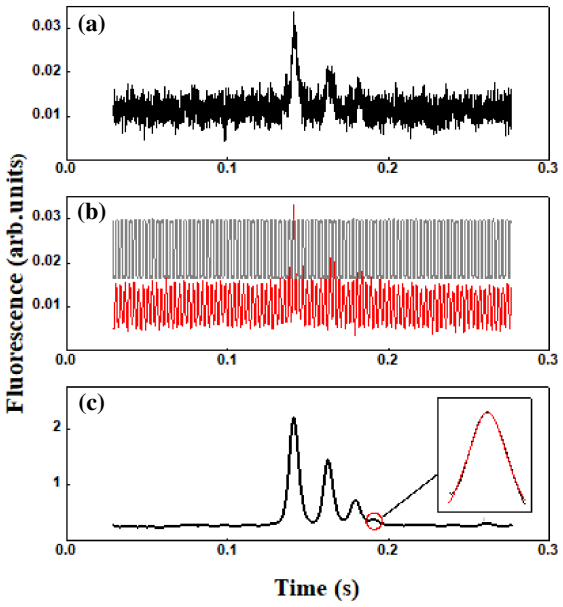

Fig. 2. (Color online) (a) Two-photon transition spectrum of ^{87}Rb $5S_{1/2}(F=2)-5D_{5/2}(F''=4,3,2,1)$ directly detected by the PMT. (b) Modulation signal of the laser (above) and the modulated spectrum (below). (c) Demodulated two-photon transition spectrum [inset: $5D_{5/2}(F''=1)$ peak, the red line is the result of fitting by a Gaussian function].

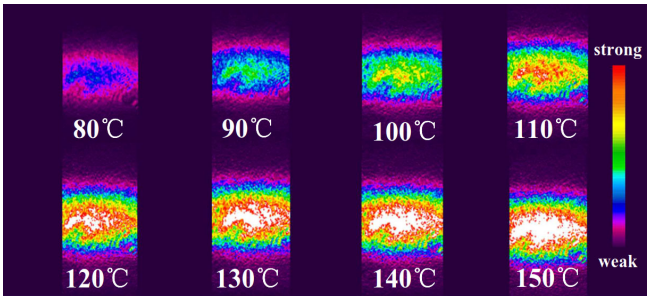

Fig. 3. (Color online) Intensity of 420 nm fluorescence for different rubidium vapor temperatures.

of the focused laser beams in the middle of the cell are about 100 μm. The 420 nm fluorescence from the cascade decay of the upper $5D_{5/2}$ state is filtered with an interference plate (center wavelength 420 nm, 10 nm pass band) to isolate the fluorescence from the background light and then detected by a side-window photomultiplier tube (Hamamatsu PMT CR131). The output signal of the PMT is sent to a lock-in amplifier (Stanford Research Systems SR830) referenced to the chopper wheel. The demodulated two-photon transition spectrum with a high SNR is monitored on an oscilloscope. A charge-coupled device (CCD) is placed near the vapor to detect the profile of the 420 nm fluorescence.

3. Results and Discussion

The two-photon transition spectrum of ^{87}Rb $5S_{1/2}(F=2)-5D_{5/2}(F''=4,3,2,1)$ directly detected by the PMT is shown in Fig. 2(a). The SNR is relatively low and the $5D_{5/2}(F''=1)$ peak cannot be distinguished. A chopper is used to modulate the laser intensity with a modulation frequency of 250 Hz. The modulation signal of the laser and the modulated spectrum are shown in Fig. 2(b) by the gray line above and red line below. The spectrum demodulated by a lock-in amplifier is shown in Fig. 2(c). The SNR of the spectrum has been significantly improved by about two orders of magnitude compared with Fig. 2(a). Also, the $5S_{1/2}(F=2)-5D_{5/2}(F''=1)$ two-photon transition peak can be observed clearly as shown in the inset of Fig. 2(c).

A two-photon transition spectrum with a high SNR can be obtained by optimization of the experimental parameters. The fluorescence will be strongly affected by the density of atoms, which is directly determined by the vapor temperature. The fluorescence intensity at temperatures from 80 to 150 °C is studied while the power of the laser is kept at 25 mW, as shown in Fig. 3. The fluorescence intensity gradually increases with the vapor temperature. However, when the temperature is higher than 150 °C, the fluorescence intensity tends to saturate for the self-absorption of the $6P_{3/2}-5S_{1/2}$ transition. This result agrees with the observations reported by Ryan et al.[19] and Cao et al.[20]

The two-photon transition spectra of ^{87}Rb $5S_{1/2}(F=2)-5D_{5/2}(F''=4,3,2,1)$ with a variety of scanning speeds of the laser are plotted in Fig. 4, in which the black lines are the experimental results and the red lines are corresponding multipeak curves fitted with a Gaussian function. With increasing scanning speed, the spontaneous emission of fluorescence from $6P_{3/2}-5S_{1/2}$ is limited by the atomic transition, resulting a low resolution. The hyperfine structure cannot be clearly distinguished and is obscured by the background, as shown in Figs. 4(a) and 4(b). As the scanning speed decreases, the resolution of the spectrum is improved. When the scanning speed reaches 1.40 MHz/ms, the resolution is high enough to identify each hyperfine structure of ^{87}Rb $5S_{1/2}(F=2)-5D_{5/2}(F''=4,3,2,1)$.

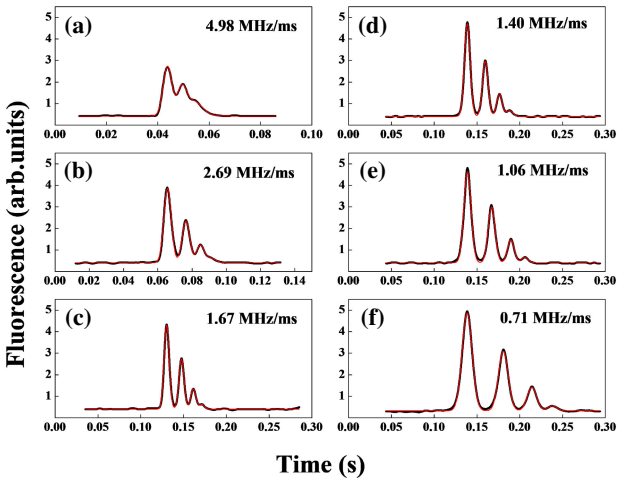

Fig. 4. (Color online) Hyperfine spectra of ^{87}Rb $5S_{1/2}(F=2)-5D_{5/2}(F''=4,3,2,1)$ transition with different scanning speeds of the laser (black lines). The scanning speeds are 4.98 MHz/ms (a), 2.69 MHz/ms (b), 1.67 MHz/ms (c), 1.40 MHz/ms (d), 1.06 MHz/ms (e), and 0.71 MHz/ms (f). Red lines are corresponding multipeak fittings of the data with a Gaussian function.

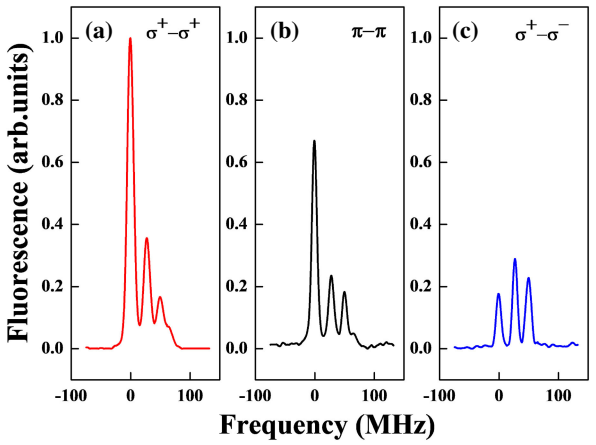

Fig. 5. (Color online) Two-photon transition spectra of ^{87}Rb $5S_{1/2}(F=2)-5D_{5/2}(F''=4,3,2,1)$ with different polarization combinations of the laser beams. π is the linearly polarized laser, σ^+ is the right-handed circularly polarized laser, and σ^- is the left-handed circularly polarized laser.

The two-photon transition can be interpreted as a process in which an atom simultaneously absorbs two photons from the initial state to the final state through an intermediate state. The transition probability from the $5S_{1/2}$ state to the final state $|5D_{5/2}F''\rangle$ is given by second-order perturbation theory as follows:[21]

$$P(5S_{1/2}F,5D_{5/2}F'') = \left(\frac{I_1 I_2}{4\varepsilon_0^2 c^2 \hbar^4}\right)\left(\frac{1}{2F+1}\right)\frac{\gamma_{5D_{5/2}}}{[\omega_{5S_{1/2}F:5D_{5/2}F''}-(\omega_1+k_1\cdot v)-(\omega_2+k_2\cdot v)]^2+\left(\frac{\gamma_{5D_{5/2}}}{2}\right)^2}$$
$$\times \Sigma_{M_F,M_{F''}}\left|\Sigma_{F',M_{F'}}\frac{\langle 5D_{5/2}F''M_F''|\hat{e}_2\cdot d|5P_{3/2}F'M_F'\rangle\langle 5P_{3/2}F'M_F'|\hat{e}_1\cdot d|5S_{1/2}FM_F\rangle}{\omega_{5S_{1/2}F:5P_{3/2}F'}-(\omega_1+k_1\cdot v)-i\frac{\gamma_{5P_{3/2}}}{2}}\right|^2 \quad (1)$$

The first term in Eq. (1) arises from the conservation of energy;[22] ω_1 and ω_2 are the frequencies of the two photons; I_1 and I_2 are the intensities of the two laser beams; M_F, M_F', and M_F'' are the magnetic quantum numbers for hyperfine momenta F, F', and F'', respectively; v is the velocity of atoms; k_1 and k_2 are the wave vectors for the photons; $\hat{e}_1$ and $\hat{e}_2$ are the unit vectors along the direction of the axis of quantization for the two laser beams; d is the electric dipole operator; γ_{nL} is the homogeneous line width of the state $|nL_J\rangle$; and $\omega_{nL_JF:n'L_J'F'}$ is the resonant angular frequency of the transition $|nL_JF\rangle \rightarrow |n'L_J'F'\rangle$.

The second term in Eq. (1) describes the transition probability for the two-photon transition, which can also be described as:[23,24]

$$\langle n'L_J'F'M_F'|\hat{e}\cdot d|nL_JFM_F\rangle = \langle J\|d\|J'\rangle(-1)^{2F'+M_F+I+J}\sqrt{(2F+1)(2F'+1)(2J+1)} \times \begin{pmatrix} F' & 1 & F \\ M_F' & M_F-M_F' & -M_F \end{pmatrix}\begin{Bmatrix} J & J' & 1 \\ F' & F & I \end{Bmatrix}. \quad (2)$$

Here $\langle J\|d\|J'\rangle$ is the reduced matrix element, which remains constant for a hyperfine transition, I is the nuclear spin, and the terms in brackets and curly brackets are the 3-J and 6-J symbols, respectively. Thus, the transition probabilities of different polarization combinations are calculated. The peak intensity ratios of the hyperfine transition for ^{87}Rb $5S_{1/2}(F=2)-5D_{5/2}(F''=4,3,2,1)$ with the combinations σ^+–σ^+, π–π, and σ^+–σ^- are $1:0.34:0.13:0.05$, $0.67:0.23:0.21:0.03$, and $0.17:0.26:0.22:0.03$,[25] respectively, after normalization by the intensity of the $5S_{1/2}(F=2)-5D_{5/2}(F''=4)$ transition with the σ^+–σ^+ combination.

The influence of the different polarization combinations of laser beams on the two-photon transition of ^{87}Rb $5S_{1/2}(F=2)-5D_{5/2}(F''=4,3,2,1)$ in this experiment is shown in Fig. 5, in which the laser power is 25 mW and the vapor temperature is kept at 140 °C. Figure 5(a) plots the result with two right-handed circularly polarized beams (σ^+–σ^+), Fig. 5(b) shows the result with two linearly polarized beams (π–π), and Fig. 5(c) illustrates the result of two beams with different-handedly circularly polarized (σ^+–σ^-). These experimental results agree well with the above theoretical calculations. The polarization combination of σ^+–σ^+ causes the highest transition probability of the $5D_{5/2}(F''=4)$ peak. The polarization combination of σ^+–σ^- has slightly different transition probabilities with different hyperfine transition peaks, and lower probabilities are obtained. Thus, the π–π polarization combination is chosen for our experimental study because suitable transition probabilities allow all transition peaks to be clearly distinguished.

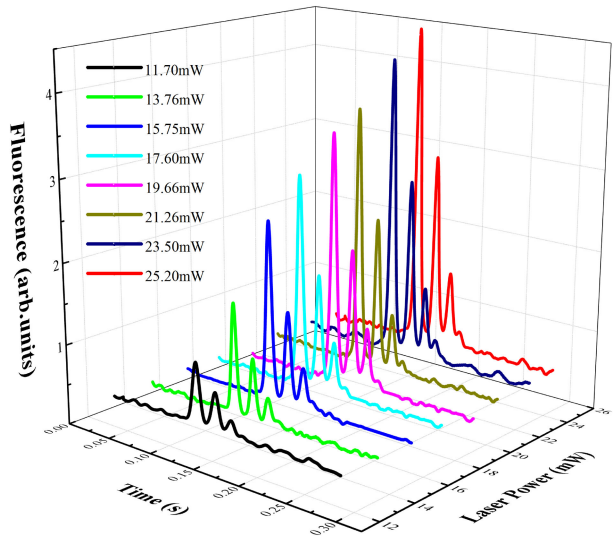

Fig. 6. (Color online) $5S_{1/2}(F=2)-5D_{5/2}(F''=4,3,2,1)$ two-photon transition spectra in ^{87}Rb as a function of laser power.

The spectra with laser powers from 11.70 to 25.20 mW are shown in Fig. 6. From Eq. (1), we find that the two-photon transition probability is quadratic to the laser intensity, and the result of the experimental study is consistent with the theoretical simulation, which is similar to the result obtained at real energy levels.[26] Moreover, as the laser power increases, the linewidth becomes larger due to the power-broadening effect.[27]

4. Conclusion

In conclusion we have experimentally studied the high-resolution $5S_{1/2}-5P_{3/2}-5D_{5/2}$ two-photon spectroscopy of ^{87}Rb using a single diode laser. The SNR of the spectrum is effectively improved by two orders of magnitude with the intensity modulation method. The fluorescence intensity strongly depends on the vapor temperature, which is directly related to the atomic density. A distinguishable hyperfine structure spectrum is obtained at a laser scanning speed of 1.40 MHz/ms. The experiment results for different polarization combinations and the power dependence of the laser beams agree well with theoretical simulation. This technique provides a simple and effective method for obtaining a high-resolution spectrum using a single diode laser, which is helpful for the study of integrated equipment and optical quantum devices.

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