High-sensitive and precise measurement of Doppler-free two-photon absorption spectra of Rb using dual-comb spectroscopy

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Introduction

Dual-comb techniques are applicable to high-resolution and precise spectroscopies [1,2]. In this work, we demonstrated Doppler-free two-photon absorption dual-comb spectroscopy of Rb. We obtained high signal-to-noise ratio spectra of the $5S_{1/2} - 5D_{5/2}$ transition and precisely determined absolute frequencies of the hyperfine spectra.

Experiment

Figure 1(a) depicts the principle of the two-photon absorption spectroscopy using the dual-comb technique [3]. In spite of the weak comb mode power, two-photon transitions which are nonlinear processes are observable due to the contribution of many comb mode pairs with the same sum frequency. We observe the fluorescence from the excited states, and obtain the spectra by taking the fast Fourier-transform of the fluorescence signals. The sensitivity of the dual-comb measurement is significantly improved by the fluorescence detection scheme.

The experimental setup is illustrated in Fig. 1(b). We used second-harmonic generations (SHG) of mode-locked Er-doped fiber lasers. The center wavelength of the SHG spectra were adjusted near 778 nm by temperature control of PPLN crystals

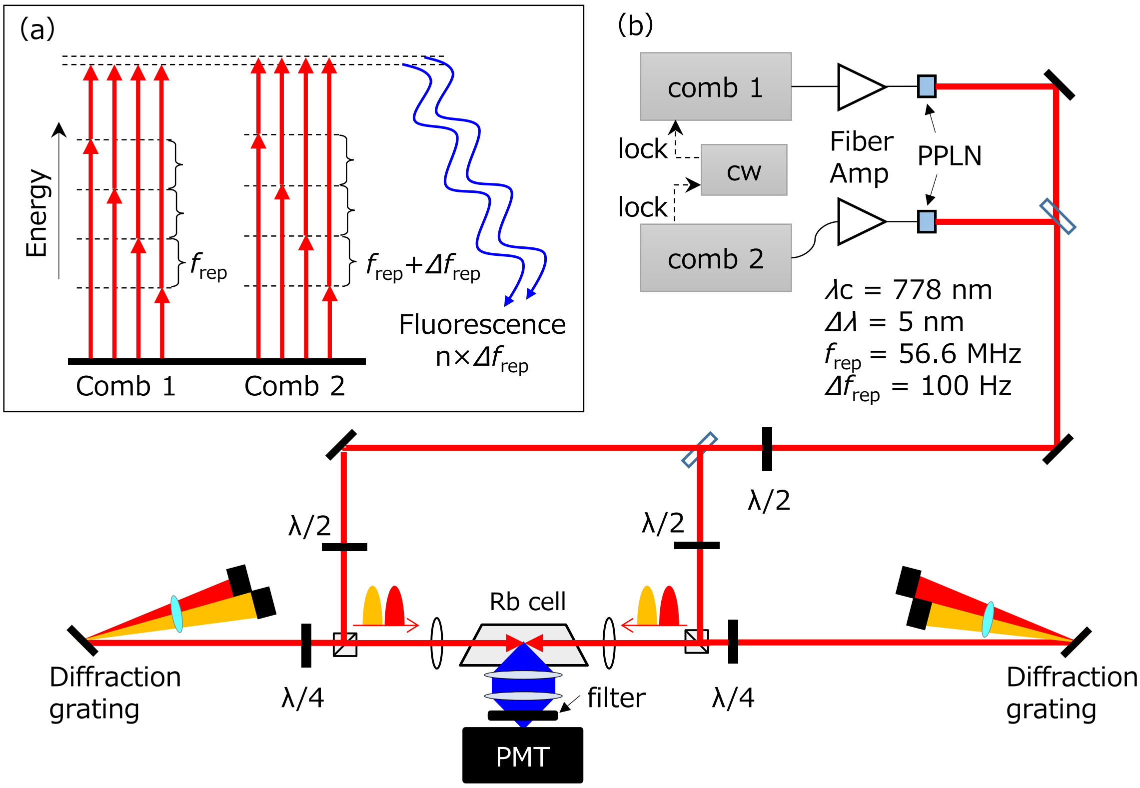

Fig. 1 (a) Principle of the two-photon dual-comb spectroscopy. (b) Experimental setup. PPLN: periodically-poled lithium niobate, PMT: photomultiplier tube.

to observe the $5S_{1/2}$ – $5D_{5/2}$ transition at 778.1 nm. The two outputs were overlapped and split into two arms. In each arm, two color parts (>778 nm, <778 nm) of the spectra were separated using a diffraction grating and two mirrors with an adjustable delay. The delay of the two color pulse was used to eliminate Doppler-broadening background, then we can observe background-free spectra which were caused by only counter-propagating pulses. The counter-propagating pulses were overlapped in the center of a Rb vapor cell, and the fluorescence interferogram was detected by a photomultiplier tube (PMT).

Results

Figures 2 (a) shows Doppler-broadened spectrum of the $5S_{1/2}$ – $5D_{5/2}$ transition observed with only unidirectional incident beam without the delay. The spectral width of transitions from a hyperfine level of the ground state of ^{87}Rb was about 1.3 GHz. Figure 2 (b) shows the Doppler-free and background-free spectrum observed with counter-propagating pulses with the delay of 1 cm, and magnified view of the $5S_{1/2}$ (F''=1) – $5D_{5/2}$ (F'=1, 2, 3) transitions of ^{87}Rb is shown in Fig. 2(c). The observed spectra had a flat baseline and showed fully-resolved hyperfine structure of the excited state. Center frequencies of the background-free spectra were precisely determined with the uncertainty of less than 100 kHz.

The spectral widths of the hyperfine spectra were approximately 5 MHz which was broader than the natural linewidth of the transition. As shown in Fig. 2(c), the observed sub-Doppler spectra were fitted to Gaussian profiles, because the main contribution to the broadening was residual Doppler shifts. In the dual-comb two-photon absorption scheme, many pairs of modes introduce a transition, and frequency differences of the mode pairs cause the residual Doppler shift. The broadband comb spectrum increases the variety of mode pairs, then the observed sub-Doppler spectral widths are broadened.

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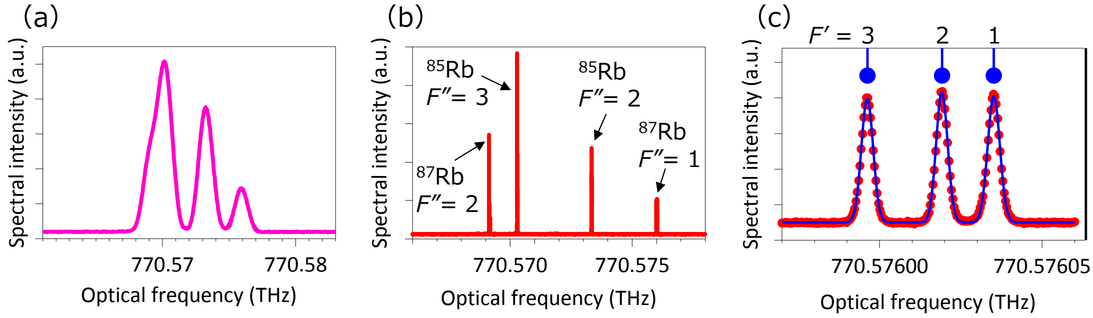

Fig.2. (a) Doppler-limited spectrum of $5S_{1/2}$ – $5D_{5/2}$ transition of Rb. (b) Doppler-free spectrum observed with counter-propagating beams. (c) Magnified view of the Doppler-free spectra of $5S_{1/2}$ (F'' = 1) – $5D_{5/2}$ (F' = 3, 2, 1) of ^{87}Rb.