Hyperfine-structure-constant determination and absolute-frequency measurement of the Rb $4D_{3/2}$ state

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The frequency of a laser diode is stabilized onto the line centers of double-resonance optical pumping spectra, and the absolute frequencies of the hyperfine-structure components of the $5P_{3/2}$-$4D_{3/2}$ excited-state transition for two isotopes of ^{87}Rb and ^{85}Rb atoms are measured using a fiber femtosecond frequency comb system. The $4D_{3/2}$ state hyperfine-structure constants of the two Rb isotopes are determined by utilizing the measured absolute optical frequency and the ratio of the hyperfine-structure constants for the two Rb isotopes from the accurately determined values of other states of rubidium. The magnetic-dipole constants A are determined to be 24.75(12) MHz for ^{87}Rb and 7.329(35) MHz for ^{85}Rb. The uncertainties of both results are reduced by one order compared to previous results. The electric-quadrupole constants B are determined to be 2.19(11) and 4.52(23) MHz for ^{87}Rb and ^{85}Rb, respectively

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Since the initial determination of hyperfine-structure (HFS) constants in atomic beam experiments, atomic hyperfine splitting has been investigated with various experimental techniques [1,2]. Accurate measurement of HFS constants has been of interest in a variety of fields, including tests of fundamental physics, electron-cloud–nucleus interaction, atomic parity nonconservation, laser cooling of atoms, high-resolution spectroscopy, and frequency standards [3–10]. To experimentally investigate the HFS of alkali metal atoms, various experimental methods have long been successfully employed, such as optical spectroscopy, optical double resonance, atomic beam magnetic resonance, Doppler-free laser spectroscopy, and other methods [1,2]. Although each technique has its own advantages and disadvantages, optical methods with lasers have been much stronger tools for HFS observations.

In excited states without a direct electric-dipole transition from the ground state, HFS was investigated by radiofrequency cascade spectroscopy, optical double resonance, and two-photon spectroscopy [1,2]. However, in some cases of excited states, the magnetic-dipole constant A could be determined but the electric-quadrupole constant B could not, as the B value was too small to be measured or the signal-to-noise ratio (SNR) was too low [1]. For the $4D_{5/2}$ state of the ^{85}Rb atom, Sinclair *et al.* successfully determined the B value in laser-cooled atoms using the optical double-resonance method [9]. More recently, by applying a unique experimental technique known as the double-resonance optical pumping (DROP), Lee *et al.* determined the B value of the $4D_{5/2}$ state of the ^{87}Rb atom in a vapor cell [10–12].

However, for the $4D_{3/2}$ state of the Rb atom, only the A values are known to be 25.1(9) MHz for the ^{87}Rb atom and 7.3(5) MHz for the ^{85}Rb atom, as the B value is too small to be measured with the previous method of cascaded radio-frequency spectroscopy [1,13]. One can expect that optical methods can solve this problem. Conventionally, the optical-optical double-resonance (OODR) method has widely been used for transitions between excited states, with a number of successful results [9,14]. However, with the OODR method in the $5P_{3/2}$-$4D_{3/2}$ transition, the SNR of the spectrum is too poor to be used reliably for laser frequency stabilization [11,12]. With the DROP technique [11,12], the clean spectra of this excited-state transition can be obtained by utilizing the advantage of detecting the population of the ground state rather than that of the excited state.

In this study, the HFS components of the $4D_{3/2}$ state of the two isotopes ^{85}Rb and ^{87}Rb are investigated using the DROP technique. The results of the absolute frequencies of the $5P_{3/2}$-$4D_{3/2}$ transitions of Rb atoms measured using a fiber femtosecond frequency comb are presented. Moreover, the HFS constants, A and B, of the $4D_{3/2}$ state of the two isotopes ^{85}Rb and ^{87}Rb are determined through the use of the measured absolute frequencies of the HFS components. It was possible to obtain only two of the HFS components in the $5P_{3/2}$-$4D_{3/2}$ transition due to the selection rule; thus, the previously known ratios of the HFS constants of the two Rb isotopes were used to determine the HFS constants. The magnetic-dipole coupling constant with reduced uncertainty was found to be in good agreement with previous reports [13]. Additionally, this study contains the determination of the electric-quadrupole constant.

Figure 1 shows an energy-level diagram of the $5S_{1/2}$-$5P_{3/2}$-$4D_{3/2}$ transitions of the two isotopes ^{85}Rb and ^{87}Rb. When the frequency of the first laser diode (LD1) is resonant with the cycling transition of the $5S_{1/2}$-$5P_{3/2}$ transition of the Rb D_2 line, only two HFS components of the $4D_{3/2}$ ($F''=2$ and 3) of ^{87}Rb and the $4D_{3/2}$ ($F''=3$ and 4) of ^{85}Rb can be obtained due to the selection rule. In Fig. 1, the HFS components interacting with two lasers are represented by black lines while the other components are represented by gray lines. The lifetime of the $4D_{3/2}$ state is known to be approximately 78.7 ns (with a linewidth of 2.2 MHz) [15].

The DROP experiment with two counterpropagating beams offers a spectrum with a high SNR as well as a narrow

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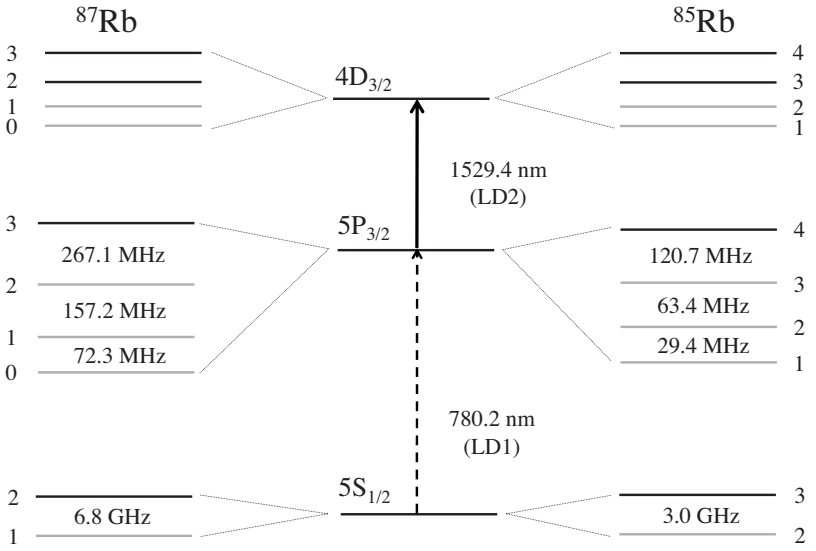

FIG. 1. Energy-level diagram of the $5S_{1/2}$-$5P_{3/2}$-$4D_{3/2}$ transitions of two Rb isotopes.

linewidth in the $5P_{3/2}$-$4D_{3/2}$ excited-state transition of Rb [12]. The frequency of the laser diode is stabilized onto the line center of the DROP spectrum of the HFS components of the $5P_{3/2}$-$4D_{3/2}$ excited-state transition of the two isotopes ^{85}Rb and ^{87}Rb. The absolute frequency of the HFS components is measured using a fiber femtosecond frequency comb system. The experimental setup used in this study is similar to those described in previous works [10].

Figure 2 shows the experimental schematic of the DROP experiment and the absolute-frequency measurement. For the DROP experiment, two grating-feedback external cavity diode lasers (ECDLs) operated in a single mode were used. The frequency of LD1 is fixed on the cycling transition of the $5S_{1/2}$-$5P_{3/2}$ transition of the Rb D_2 line using a conventional modulation/first-harmonic lock-in detection system of saturated absorption spectroscopy (SAS) in a 5-cm-long Rb vapor cell. A modulation depth of approximately 5 MHz at a modulation frequency of 5 kHz was provided by the piezoelectric transducer (PZT) in the external cavity of LD1. To obtain the DROP spectrum in the $5P_{3/2}$-$4D_{3/2}$ transition, the frequency of the second laser diode (LD2) was scanned over the range of the upper states in the $5P_{3/2}$-$4D_{3/2}$ transition and LD1 was probed. The optical powers of LD1 and LD2 entering the Rb cell were measured to be 14 and 26 μW, respectively. The counterpropagating LD1 and LD2 beams were overlapped within 1 mrad using two apertures (AP) with a diameter of 1.5 mm. The polarizations of both lasers were linear and parallel to each other in the 10-cm-long Rb vapor cell at room temperature. With three layers of a μ-metal sheet around the Rb cell, the residual magnetic field

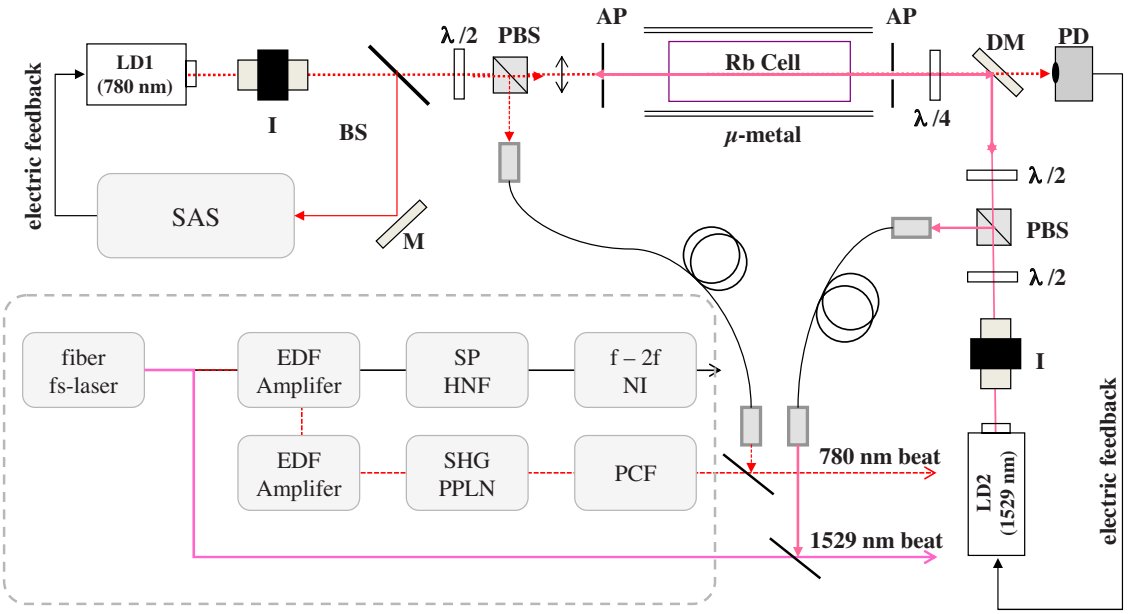

FIG. 2. (Color online) Experimental setup of the DROP spectroscopy and the absolute-frequency measurement; PBS: polarizing beam splitter; SAS: saturated absorption spectroscopy setup; DM: dichroic mirror (reflection at 1529 nm, transmission at 780 nm); AP: aperture; PD: Si photodiode; I: optical isolator; λ/2: half wave plate; SHG PPLN: periodically polled lithium niobate for the second-harmonic generation; HNF: high-nonlinear fiber; SP: spectral broadening; NI: nonlinear interferometer; PCF: photonic crystal fiber.

in the Rb cell was reduced to less than 1.5 μT. The transmission of LD1 through the Rb cell and a dichroic mirror was measured with a Si photodiode to obtain the DROP spectrum. To achieve the frequency stabilization of LD2 onto the HFS components of the $5P_{3/2}$-$4D_{3/2}$ transition, the first derivative signal of the DROP spectrum was obtained using a phase-sensitive detector attached to a lock-in amplifier, and the first derivative signal of the DROP was electrically fed back into LD2.

The optical frequencies of transitions between excited states have seldom been measured using the optical frequency comb technique, as it is not easy to obtain the spectrum of the transitions between the excited states with a high SNR. To measure the absolute frequencies of LD1 and LD2, an optical frequency comb capable of covering two octaves was used in the experiment. The frequency spacing between the neighboring modes of the optical frequency comb was approximately 250 MHz. This was stabilized using a hydrogen maser as a frequency reference. The carrier-envelope-offset frequency of the comb was also stabilized by the same hydrogen maser. The fiber optical frequency comb is similar to that used in an earlier study [10]. The outputs of the frequency-stabilized LD1 and LD2 were delivered to the optical frequency comb using the coupling lenses and two 30-m-long single-mode fibers for the respective wavelengths. The absolute frequency of LD1 was measured using the 780 nm part of the comb simultaneously with that of LD2, via the 1529 nm part of the comb, while counting the heterodyne beat frequency between the comb and each laser. Each laser beam was arranged using a polarization beam splitter so that it overlapped the comb output. The beat frequency between LD2 and the 1529 nm comb was measured using an avalanche photodetector and a frequency counter. The laser output of the LD2 was amplified by an erbium-doped fiber amplifier (EDFA) to obtain a beat signal with a sufficiently high SNR (greater than 30 dB in a resolution bandwidth of 400 kHz) to ensure correct frequency counting. The absolute frequency of LD1 was measured using a similar process.

While the frequency of the LD2 laser was scanned over the range of the HFS components of the $4D_{3/2}$, the DROP spectra of the $5P_{3/2}$-$4D_{3/2}$ transition were obtained by measuring the transmission of the LD1 laser, as shown in Fig. 3. Figure 3 shows the DROP spectra and the first derivative signals of the two Rb isotopes. Figures 3(a) and 3(b) show the two HFS components ($F''=2$ and 3) of the ^{87}Rb $4D_{3/2}$ state and the two HFS components ($F''=3$ and 4) of the ^{85}Rb $4D_{3/2}$ state, respectively. The spectral widths were measured to be approximately 6 MHz, which agrees with the estimated results under the two-photon resonance condition [12]. As is well known, the SNR of the DROP spectra is more than ten times higher compared to that of the OODR [12]. A clean spectrum could be obtained with a SNR as high as 170:1 for the DROP spectrum and 480:1 for the first derivative signal when utilizing the DROP method. The DROP spectra were demodulated by the 5 kHz modulation frequency of LD1 for stabilization using SAS, and the first derivative signals were obtained without direct frequency modulation of LD2. The first derivative signal in Fig. 3 was fed back to LD2 to lock the frequency of LD2 onto one of the HFS components of the $5P_{3/2}$-$4D_{3/2}$ transitions of Rb.

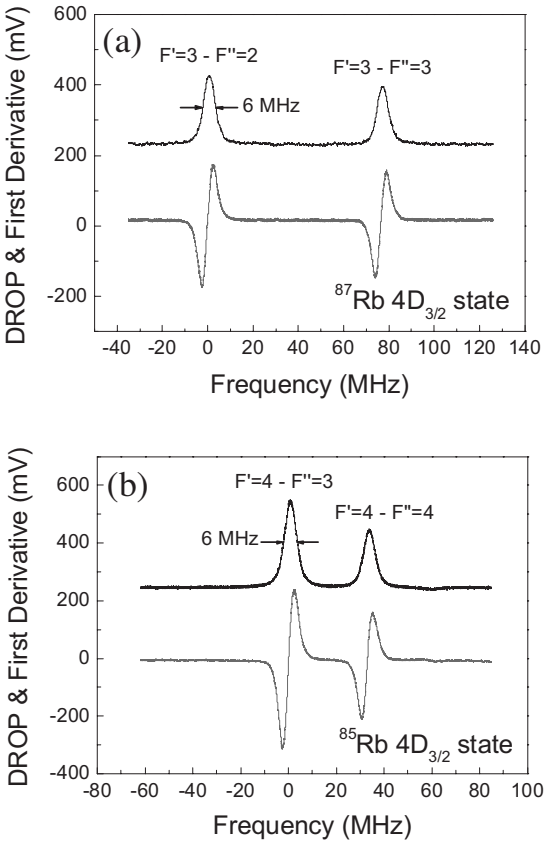

FIG. 3. DROP spectrum (upper trace) and its first derivative signal (lower trace) of (a) the $5P_{3/2}(F'=3)$-$4D_{3/2}(F''=2,3)$ transition of the ^{87}Rb and (b) the $5P_{3/2}(F'=4)$-$4D_{3/2}(F''=3,4)$ transition of ^{85}Rb.

Measuring the beat frequency between the stabilized laser and the frequency comb, the absolute frequencies of the $5S_{1/2}$-$5P_{3/2}$ transition and the $5P_{3/2}$-$4D_{3/2}$ transition of Rb were determined simultaneously. The absolute frequency of

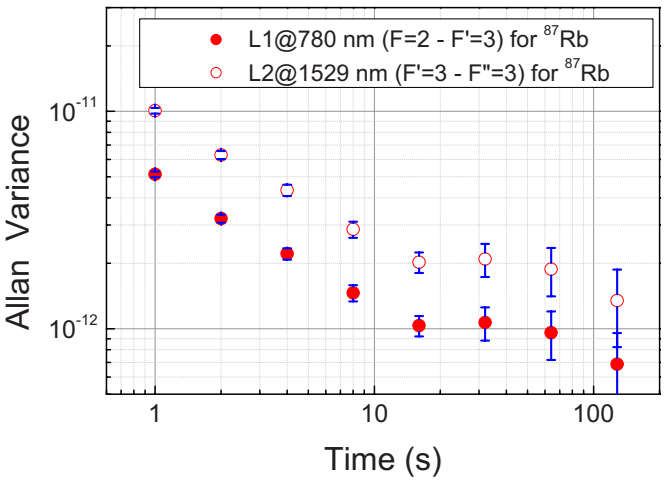

FIG. 4. (Color online) Allan variances of the LD1 laser stabilized to the $5S_{1/2}(F=2)$-$5P_{3/2}(F'=3)$ cycling transition of the ^{87}Rb atom using SAS (closed circles) and of the LD2 laser stabilized to the $5P_{3/2}(F'=3)$-$4D_{3/2}(F''=3)$ transition of the ^{87}Rb atom using DROP (open circles).

TABLE I. The results of the absolute frequencies of the ^{87}Rb, $5P_{3/2}(F'=3)$-$4D_{3/2}(F''=2,3)$ transition and the $5S_{1/2}(F=2)$-$4D_{3/2}(F''=2,3)$ transition.

HFS component of ^{87}Rb $4D_{3/2}$	Absolute frequency of the transition from ^{87}Rb $5P_{3/2}(F'=3)$ (MHz)	Absolute frequency of the transition from ^{87}Rb $5S_{1/2}(F=2)$ (MHz)
$F''=2$	196037137.33(13)	580265252.53(13)
$F''=3$	196037213.77(12)	580265328.98(12)

the $5S_{1/2}(F=2)$-$5P_{3/2}(F'=3)$ transition of ^{87}Rb was reported to be 384 228 115 208 kHz with an uncertainty of 17 kHz in a previous study by the authors [10]. Additionally, in this work the absolute frequency of the $5S_{1/2}(F=3)$-$5P_{3/2}(F'=4)$ transition of ^{85}Rb was found to be 384 229 241 999 kHz with an uncertainty of 17 kHz. This result is in good agreement with the findings of a previous study [16]. Figure 4 shows the frequency stability of the laser in terms of the Allan variance calculated from the beat frequency between the frequency-stabilized laser and the frequency comb. The frequency stability of LD1 [stabilized to the $5S_{1/2}(F=2)$-$5P_{3/2}(F'=3)$ transition of ^{87}Rb] was 5.0×10^{-12} at 1 s of averaging time; this was reduced to 7×10^{-13} at 128 s, as shown in Fig. 4.

The absolute frequencies of LD2, which was locked onto one of the HFS components of the $5P_{3/2}$-$4D_{3/2}$ transitions of Rb, were measured simultaneously with those of LD1 using the fiber comb IR output. The frequency stability of LD2 was 1.0×10^{-11} at 1 s, decreasing to 1.3×10^{-12} at 128 s, as shown in Fig. 4. Tables I and II show the absolute frequency of the $4D_{3/2}$ state of ^{87}Rb and ^{85}Rb measured by a femtosecond frequency comb, respectively. Sasada measured the absolute frequencies of the $5P_{3/2}$-$4D_{3/2}$ transition of Rb with an uncertainty of 40 MHz [14]. The present results provide a far more precise value of this frequency compared to earlier investigations with the uncertainty reduced by nearly 3 orders of magnitude. The absolute energy levels of the HFS components of the $4D_{3/2}$ state of the Rb atom from the $5S_{1/2}$ ground state, that is, the absolute frequency of the $5S_{1/2}$-$4D_{3/2}$ transition, were also measured by adding the simultaneously measured absolute frequency of the $5S_{1/2}$-$5P_{3/2}$ transition, as shown in Tables I and II. The frequency uncertainties include the polarization, the lock dc offset, the repeatability, the laser powers of both of LD1 and LD2, and the modulation depth of LD1. The estimated frequency uncertainties are 130 and 120 kHz for the $5P_{3/2}(F'=3)$-$4D_{3/2}(F''=2,3)$ transitions of ^{87}Rb and 84 and 86 kHz for the $5P_{3/2}(F'=4)$-$4D_{3/2}(F''=3,4)$ transitions of ^{85}Rb, respectively. The major part of the uncertainty is caused by the polarization angle, which is attributed to the residual Zeeman effect due to the uncancelled magnetic field. The effects from the relativistic shift, the collision shift, the black body radiation, and the neighboring transition are ignored because these are relatively small.

The HFS constants of the $4D_{3/2}$ state of two Rb isotopes were then examined using the measured absolute energy levels. The HFS splitting is related to the HFS constants. From Tables I and II, two equations were obtained, as follows:

$$f(3'-3'',87)-f(3'-2'',87)=3A_{87}+B_{87}=76.44(18)\ \text{MHz}, \quad (1)$$

$$f(4'-4'',85)-f(4'-3'',85)=4A_{85}+\frac{4}{5}B_{85}=32.93(12)\ \text{MHz}, \quad (2)$$

where $f(F'-F'',N)$ is the absolute frequency of the $5P_{3/2}(F')$-$4D_{3/2}(F'')$ transition of a rubidium isotope with a atomic mass number of N. Additionally, A_N and B_N are the magnetic-dipole constant and the electric-quadrupole constant, respectively.

As only two equations exist for four variables, the previously known ratios of the HFS constants of the two Rb isotopes were also used. These ratios are often used for consistency checks of experimental measurement data [17]. Although the HFS constants differ according to the isotope and the atomic state, the ratio of the HFS constants between two isotopes in a state is nearly identical regardless of the state, as the ratio of A is given approximately by the nuclear magnetic-dipole moments and the nuclear spins of ^{87}Rb and ^{85}Rb, while the ratio of B is given by the nuclear electric-quadrupole moments. As highly accurate HFS constants of the $5S_{1/2}$ state are known well [1], the A value of 3417.341 306 42(15) MHz for ^{87}Rb and the value of 1011.910 813(2) MHz for ^{85}Rb in the $5S_{1/2}$ state were used to obtain the relationship of

TABLE II. The results of the absolute frequencies of the ^{85}Rb, $5P_{3/2}(F'=4)$-$4D_{3/2}(F''=3,4)$ transition and the $5S_{1/2}(F=3)$-$4D_{3/2}(F''=3,4)$ transition.

HFS component of ^{85}Rb $4D_{3/2}$	Absolute frequency of the transition from ^{85}Rb $5P_{3/2}(F'=4)$ (MHz)	Absolute frequency of the transition from ^{87}Rb $5S_{1/2}(F=3)$ (MHz)
$F''=3$	196037161.680(84)	580266403.679(86)
$F''=4$	196037194.613(86)	580266436.612(88)

$$\frac{A_{87}}{A_{85}} = 3.377\ 117\ 096\ 2(67). \quad (3)$$

Using the B value of the $5P_{3/2}$ state (12.52(9) MHz for ^{87}Rb and 25.88(3) MHz for ^{85}Rb [1]) gives the equation of

$$\frac{B_{87}}{B_{85}} = 0.4838(35). \quad (4)$$

With these two additional equations, the four HFS constants of the $4D_{3/2}$ state of a rubidium atom can be determined. The magnetic-dipole coupling constants were determined to be $A_{87}=24.75(12)$ MHz and $A_{85}=7.329(35)$ MHz, which shows good agreement with previous reports [11] with the uncertainty reduced by sevenfold and 14-fold, respectively. The electric-quadrupole constants are determined to be $B_{87}=2.19(11)$ MHz and $B_{85}=4.52(23)$ MHz.

In conclusion, the absolute frequency of the HFS components of the $5P_{3/2}$-$4D_{3/2}$ excited-state transition of the two isotopes of ^{85}Rb and ^{87}Rb atoms were measured using a fiber femtosecond frequency comb system. The frequency of LD2 was successfully stabilized onto the line centers of the DROP spectra of the HFS components of the $5P_{3/2}$-$4D_{3/2}$ excited-state transition of the Rb atoms without the direct frequency modulation of LD2. The absolute energy level of the HFS components of the $4D_{3/2}$ state from the $5S_{1/2}$ ground state was determined by simultaneously measuring the absolute frequency of the $5S_{1/2}$-$5P_{3/2}$ transition. Using the HFS constant ratios between two isotopes as well as the measured hyperfine splitting between two HFS components, the magnetic-dipole coupling constants of the $4D_{3/2}$ state of ^{87}Rb and ^{85}Rb were determined with nearly tenfold reduced uncertainty. In particular, the electric-quadrupole constant of the $4D_{3/2}$ state of Rb was determined.

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