Femtosecond Lasers for Optical Frequency Measurement

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Abstract

Equidistance modes in the frequency domain can represent femtosecond laser pulses in the time domain. By stabilizing two parameters of these equidistance modes, namely, the repetition rate and the offset frequency, these modes can be called an Optical Frequency Comb (OFC). Optical frequency measurement was a complicated task before introducing the first OFC. However, the certain procedures have to be followed to assure accurate measurement, such as the determination of the mode number and the sources of uncertainty. This review article will present an overview of femtosecond lasers in the time and frequency domains. In addition, the principle of optical frequency measurement using OFCs is explained. For example, an optical frequency measurement using an OFC on the hyperfine components of the two-photon transition $5S_{1/2}$ - $5D_{3/2}$ at 778 nm of a rubidium atom is demonstrated measurement and the uncertainty is calculated. Several other accurate transitions can be measured using OFC.

Keywords: ***Optical frequency standards, two-photon transition in rubidium, Femtosecond frequency comb.***

1. Introduction

Since the invention of lasers in 1960s, ultrashort pulsed lasers have been hot topic of research in optics and photonics. However, it was not until the 1990s since the introduction of the first reliable self-mode-locked Ti:Sapphire femtosecond laser [1], which is considered a transition point in femtosecond pulse generation. Femtosecond pulses have superior characteristics in time and frequency domains, making them appealing for several applications. These superior characteristics include [2, 3]:

1- The ultrashort pulse duration.
2- The high peak power of the short pulses.
3- The wide spectral bandwidth and therefore, called supercontinuum.
4- The ability to know precisely the frequency of each mode of the equidistant modes in the frequency domain.
5- The collimated nature of the laser beam.

During the last decades, several enhancements to the characteristics of the femtosecond lasers occurred to make them a fascinating tool for millions of applications. To mention some of them, I have to classify those applications depending on their usage, whether in time or in frequency domain. In time domain, the ultrashort pulse of the femtosecond laser has been exploited in the following applications:

1- Time Resolved Spectroscopy (TRS): TRS means that femtosecond pulses can measure ultrafast phenomena using techniques such as the pump-probe spectroscopy or cavity ring down spectroscopy [4]. This is an important tool to monitor the recovery time of saturable absorbers, the melting time of a sample after being exposed to intense pulse, and diffusion speed of photoexcited carriers.
2- In metrology, absolute distance can be measured with high resolution by exploiting the ultrashort pulse width of the femtosecond laser [5, 6]. According to the definition of the meter, if the time-of-flight of the femtosecond pulse to a target is measured accurately, the distance to that target can be directly calculated from the measured time for a constant speed of light. However, to measure such ultrashort pulse accurately, an autocorrelation technique is required.
3- In surgery: Ultrashort pulse delivers laser energy to the target tissues only while preserving the surrounding tissues from damage. Femtosecond pulses employ the plasma-mediated ablation which uses the first few pulses femtosecond to ionize the molecule. Hence creating a submicrometric bubble of plasma that can remove the target tissue with negligible heat transfer and damage to the neighbouring tissues. This process is used in eye surgery of what is well-known as LASIK eye surgery [7]. Since this topic is out of the scope of this article, the discussion of this topic is limited to this part only.

In the frequency domain, the train of ultrashort pulses in the time-domain is equivalent to a spectrum of equidistant modes, which is known as OFC. The bandwidth of the spectrum becomes wider as the pulse becomes narrower. I will mention here some of the applications that exploit the superior characteristics of the femtosecond laser in the frequency domain.

1- Optical frequency measurement: If the spacing between the equidistant modes of the comb spectrum and their offset frequency from the zero is stabilized to the Cs clock, the frequency of each mode of the spectrum will be well-known. This means that if a beat is made between an external laser and that well-known comb mode, the frequency of the external laser could be measured precisely [8].
2- Dimensional measurement: If a single-mode from the OFC is filtered [9], it can be used in interferometry to measure the displacement of a mirror. The mirror could be scanned over the object, which dimension needs to be measured. Therefore, the measurement will be traceable directly to the SI unit of second.
3- In optical communication: each mode of the evenly spaced OFC modes is considered as a high purity laser source. Since the OFC contains a million of modes. A million of laser sources could be transmitted into a single optical fiber to increase the transmission bandwidth. This could be implemented mainly in coherent communication, an emerging optical communication field [10].

In this review article, optical frequency measurement with OFC based on femtosecond laser will be demonstrated as an application of the femtosecond lasers. In addition, a sample optical frequency measurement is performed on the hyperfine transition of the two-photon absorption in Rb.

2. Principle of femtosecond lasers

T Femtosecond lasers are based mainly on passive mode-locking in broadband laser cavities. To explain the idea behind the passive mode-locking, the oscillating modes of laser oscillations supported by the broadband spectrum of the gain medium should be considered. Several modes within the laser gain curve can oscillate within a cavity. However, all modes oscillate with random phase relationships between each other. If a technique is used to lock the phase of all modes in the laser cavity, the laser will produce short pulses. If the number of the oscillating modes within the laser cavity increases, the pulses will become shorter, with a large number of phase-locked modes a femtosecond laser pulse will produce, see fig. 1.

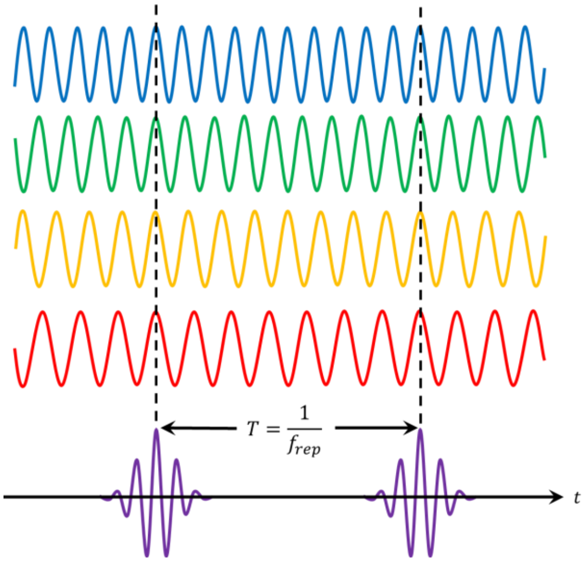

Fig. 1: Phase-locked laser cavity modes corresponding to pulses in the time domain. The pulses will become shorter when oscillatory modes are phase-locked together.

Mode-locking can be either passive or active depending on the loss modulator within the laser resonator. To demonstrate the mode-locking operation, a laser cavity with two reflectors surrounding gain and loss media are depicted in fig. 2. The partial reflector reflects a tiny fraction of the circulating pulse in the laser resonator each round-trip of the pulse within the resonator. The round-trip time of the pulse within the resonator is related to the length of the resonator (L) from the relation ($T = 2\,L/v_g$), where v_g is the group velocity of light [11].

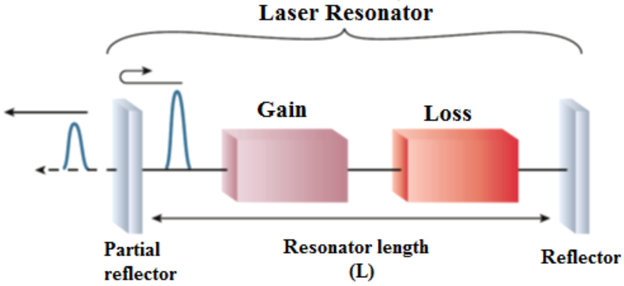

Fig. 2: Mode-Locking in laser cavities

For active mode-locking, the loss modulator can be an acousto-optic or an electro-optic modulator controlled using an external signal. To achieve mode-locking, the saturation in the loss modulator should take place with a period equal to the round-trip time. The saturable absorber is used as a loss modulator for passive mode-locking to obtain self-amplitude modulation for the light inside the cavity. The saturable absorber introduces loss in the cavity so that it greatly attenuates light propagating in the cavity with low intensities and insignificantly attenuates light with high intensities. Therefore, a pulse is formed since the high intensity is in the peak of the pulse and the low intensity at its wings, see fig. 3 [11].

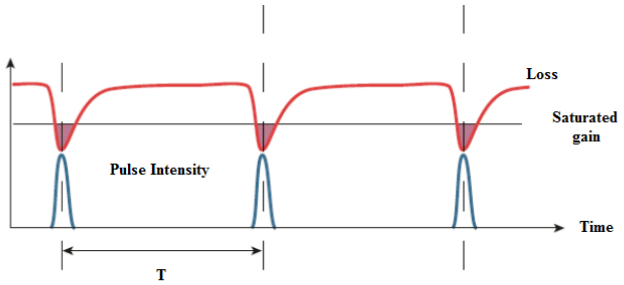

Fig. 3: The function of loss modulator: tiny loss for high intensities and large loss for small intensities

Passive mode-locking produces shorter pulses than active mode-locking since the recovery time of the saturable absorber is very fast, which causes fast loss modulation. Saturable absorbers are materials that decrease their absorption as light intensity increases. Saturable absorbers can be classified according to their wavelength range, recovery time, and saturation intensity. In the past, saturable absorbers were mainly dyes. Although they have a wide wavelength range, they are toxic and have short lifetimes. Nowadays, there are several kinds of saturable absorbers depending on the type of femtosecond lasers; three of them only will be mentioned here because they are used mainly to generate OFCs that are based either on optical fibers or Ti:Sapphire lasers:

1- <u>*Kerr-Lens Mode-Locking (KLM)*</u>

Kerr lens effect can be explained by the dependence of the refractive index of light on its intensity in a nonlinear medium according to the following relation: $\Delta n = n_2 I(r,t)$, where n_2 is the nonlinear refractive index, $I(r,t)$ is the radial-temporal intensity [11]. Therefore, when short pulses propagating in a nonlinear medium, the phase delay is largest for high-intensity light at the center of the beam and smallest for the low-intensity light at the beam's edges. This causes the short pulses to focus as there is a lens. Therefore, the cavity favours the high-intensity pulses to oscillate within the cavity, which is the aim of the saturable absorber.

2- <u>*Nonlinear Polarization Rotation (NPR)*</u>

Nonlinear Polarization Rotation (NPR) is considered an artificial saturable absorber used mainly in fiber-based mode-locked lasers. It can be explained as follows: The Kerr nonlinearity in fiber causes rotation of the polarization state in the light propagating in the fiber as a function of its intensity [2]. The output polarizer can then be aligned to allow high-intensity light to pass and block low-intensity light. Therefore, the NPR acts as a fast saturable absorber.

3- Nonlinear Loop Mirror (NLM)

NLM is an artificial saturable absorber which is used mainly in fiber-based mode-locked lasers. The function of the NLM is explained as following [12]. If two beams of light are propagating in opposite directions in a Sagnac-like interferometer, one of the directions is subject to a phase shift of more than the other. Suppose the phase shift occurs for the high-intensity light more than the low-intensity light is adjusted so that it interferes instructively with the beam from the other direction while low-intensity light interferes destructively with the light from the opposite direction, mode-locking will occur.

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3. Optical Frequency Combs

Scientists noticed that lasers that emit a continuous train of ultrashort light pulses have millions of different colors. This can be explained by the inverse relationship between time and frequency, which made the train of closely spaced optical pulses in the time domain are equivalent to a comb of closely spaced oscillations in the frequency domain. The train of femtosecond laser pulses in the time domain can be represented by a series of oscillating modes [13], see fig. 4:

$$E(t) = \sum_n A_n e^{i((\omega + n\omega_{rep})t + \varphi_n)} \qquad (1)$$

where, ω is the laser center oscillation frequency, A_n is the amplitude of each mode, and ϕ_n is the phases of each mode, and ω_{rep} is the repetition frequency of the pulses ($\omega_{rep} = \pi c \, / \, L$). If the modes of the laser are free oscillating, A_n and φ_n can take any random values leading to CW operation; however, for mode-locked lasers, A_n and φ_n are constant.

In the frequency domain, the train of femtosecond pulses can be represented by a comb of modes in the frequency domain such that the spacing between the modes corresponding to the repetition frequency (ω_{rep}= 2π f_{rep}), see fig. 4 (b). The optical frequency ($\omega = 2\pi\upsilon$) of the mode number (n) can be obtained from equation 1 as follows:

$$\upsilon = nf_{rep} + f_o \qquad (2)$$

The pulse train is not perfectly periodic since the oscillations of the electric field of the carrier wave is constantly shifted concerning the pulse envelope, see fig. 4 (a). The dispersion and nonlinearities cause this carrier-envelope offset (CEO) from pulse to pulse within the cavity. The CEO frequency is calculated from $f_o = \Delta\varphi/2\pi T$, where $\Delta\varphi$ is the phase shift between the carrier wave and the envelope of the pulses, T is the pulse repetition period.

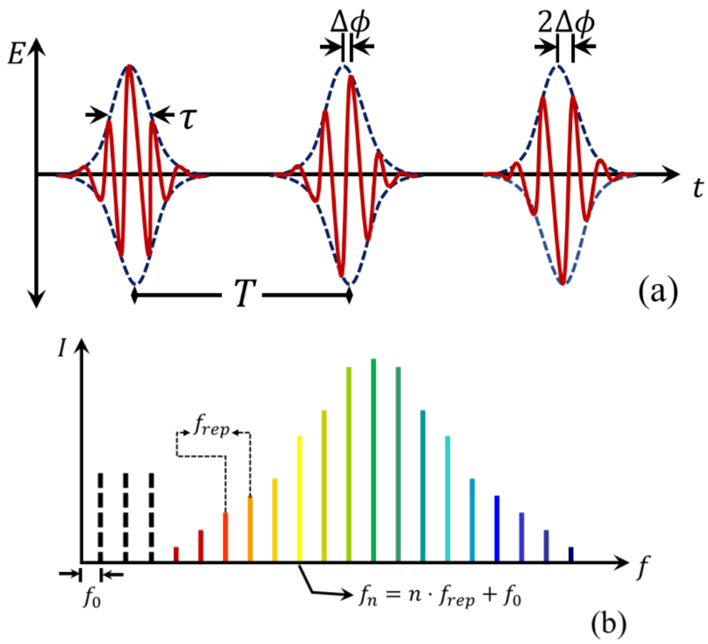

Fig. 4: OFC (a) time-domain representation of femtosecond pulses which shows cycle slips in the CEO phase (b) frequency-domain representation of the OFC which shows the frequency of each mode which is calculated from the offset frequency of the CEO and the repetition rate. τ is the pulse width.

OFC has two degrees of freedom, namely, the repetition rate (f_{rep}) and the carrier-envelope offset (CEO) frequency (f_o). Depending on the application, one or both degrees of freedom must be stabilized. For example, when measuring optical frequencies, OFC act as a frequency ruler, therefore, both degrees of freedom must be stabilized. However, it is enough to have control over the repetition rate when measuring distance. The process of frequency stabilization can be divided into two steps, namely, detection and control.

For the repetition rate, detection (f_r) is made using high-speed photodetector, while control is performed by controlling the length of the laser cavity. For free-space femtosecond lasers or fiber lasers with mirror in their cavities, control moves the mirror by attaching it to a piezoelectric element. For all-fiber cavities control is made through a fiber stretcher attached to the laser cavity. electro-optic modulators (EOM) can be used [14]. Some researchers reported stabilizing the repetition rate using an electronic polarization controller (EPC) [15].
For CEO frequency (f_o) stabilization, both detection, and control are challenging. The detection of the CEO frequency is made using the *f-2f* self-referencing interferometer scheme, see Fig. 5. This scheme is applied by broadening the OFC spectrum until it covers an optical octave (the higher-end of the spectrum is exactly double of the lower end of the spectrum). The broadening of the spectrum can be performed by the self-phase modulation in any high-nonlinear fiber such as the photonic crystal fiber. Then a beat is made between the frequency-doubled lower-end of the spectrum with the higher-end of the spectrum.

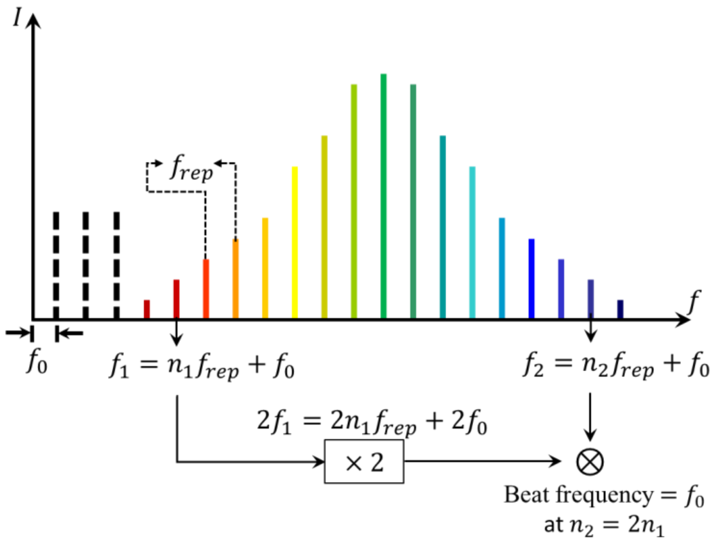

Fig. 5: CEO detection by using the f-2f scheme.

The beat will be between:

1- The frequency-doubled component of the lower frequency which is:

$$2f_1 = 2n_1 f_{rep} + 2f_o \qquad (3)$$

2- The higher frequency which match the frequency-doubled component ($f_2 = 2f_1$) which is:

$$f_2 = n_2 f_{rep} + f_o \qquad (4)$$

Then the beat frequency is the difference between equation (3) and (4) is the CEO frequency (f_o). A typical method for the compensation of CEO phase fluctuations is to feedback the fluctuations as an error signal to the current of the pump laser to control its optical power [15].

4. Optical frequency measurement with OFCs

The absolute frequency of each comb mode is well known if the offset frequency (f_o) and the repetition rates (f_{rep}) are locked. Therefore, to measure the absolute frequency of any laser pulse, a beat is made between that laser pulse and the nearest mode from the comb. If the number (m) of the comb mode is known, the absolute frequency of the laser can be calculated from the equation:

$$\upsilon_{cw} = m\, f_r \pm \Delta f_{cw} \pm f_o \qquad (5)$$

Where, Δf_{LC} is the frequency of the beat between the comb mode and the laser.

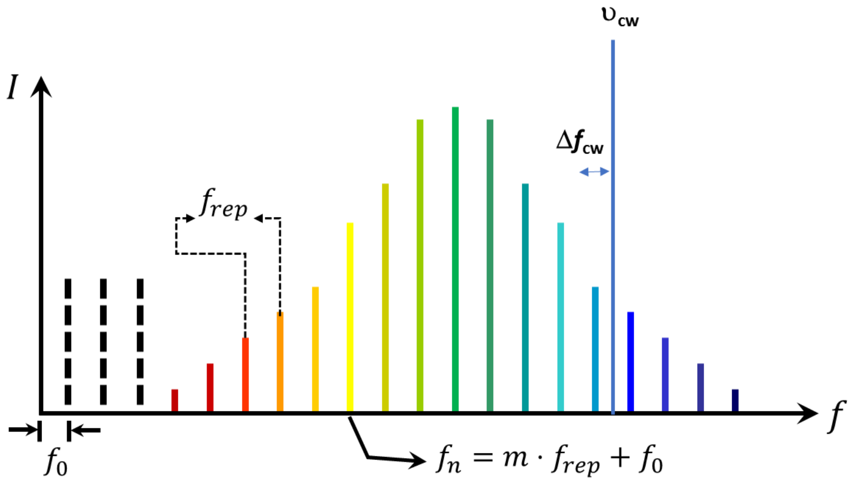

Fig. 6. The spectrum of an OFC with equidistant modes with spacing of f_{rep}. All Modes are shifted by the offset frequency from the zero.

In order to determine the absolute frequency of the laser, two main parameters must be determined, namely the signs of f_o and f_{CL} beats and the OFC mode number with which the laser beats. The sign of the f_{cw} beat could be determined by changing the repetition frequency of the comb. The sign should be as follows:

Increase Δf_{rep} (↑), if beat increased(Δf_{cw} ↑)→(f_{cw}→-ve).

The sign of the f_o is determined by changing the offset beat frequency as follows:

Increase (Δf_o) (↑): if (Δf_{cw} ↑) → (f_o) has the opposite sign of (Δf_{cw}).
: if (Δf_{cw} ↓) → (f_o) has the same sign of (Δf_{cw}).

The mode number can be determined by measuring the frequency of the CW laser with an accurate wavemeter with accuracy better than $f_{rep}/2$. It can be determined also by sweeping the repetition rate until the laser beat with comb return to its position again several times (around 7 times) and using the following equation to determine the mode number:

$$m_1 = \frac{\Delta m f_{rep2} + \Delta f_{cw}}{\Delta f_{rep}} \pm \frac{\Delta_{beat}}{\Delta f_{rep}} \qquad (6)$$

Where $\quad \Delta f_{rep} = f_{rep1} - f_{rep2} \qquad \Delta f_{beat} = f_{beat2} - f_{beat1}$

$\Delta_{beat} = \sqrt{\Delta_1^2 + \Delta_2^2}$ (Uncertainty in beats measurements)

However, the second term in equation 6 (uncertainty of the mode) should be less than 0.5 to avoid the ambiguity between two consecutive modes. The laser must be efficiently stabilized to avoid drift during the sweeping of repetition rate by not more than 0.1 MHz.

5. Optical frequency measurement of the Two-photon absorption in Rb

Recently, remarkable progress is achieved by having several ultra-narrow linewidth lasers at a broad wavelength range. Consequently, the observation of narrow transitions in trapped ions and cold atoms becomes possible. This impacts the studies of the fundamental constants, the redefinition of the second, metrology, and optical communications [16]. An optical frequency standard based on the two-photon absorption in Rb atom is presented, and its hyperfine transitions are measured using the OFC.

Doppler broadening, which results from the thermal motion of atoms, is so broad (order of GHz) for laser frequency stabilization. Hyperfine transitions are naturally broadened lines that has a linewidth in the kHz range; therefore, can be used for laser stabilization. Several techniques have been proposed to stabilize the laser on the hyperfine transitions directly, while excluding the effect of Doppler broadening. Two-photon absorption (TPA) is one of those techniques. It occurs when two photons are absorbed from counter-propagating directions. Since the photons from the other direction have different sign, the Doppler shifts should be cancelled. In other words, the two counter-propagating beams appear to atoms as they have the frequencies $\omega_o(1+kV/c)$ and $\omega_o(1-kV/c)$, where ω_o is the center frequency. Consequently, the Doppler shift is cancelled since all atoms at different velocities can absorb two photons.

Therefore, the signal to noise ratio (SNR) is higher for the two-photon transitions than for other transitions like the saturation absorption since all atoms contribute to the transition. A fluorescence light at 420 nm is emitted when the two-photon absorption occurs. Fig. 7 illustrates the two-photon transition together for the hyperfine levels of the two-photon transitions $5S_{1/2}$–$5D_{3/2}$ in rubidium.

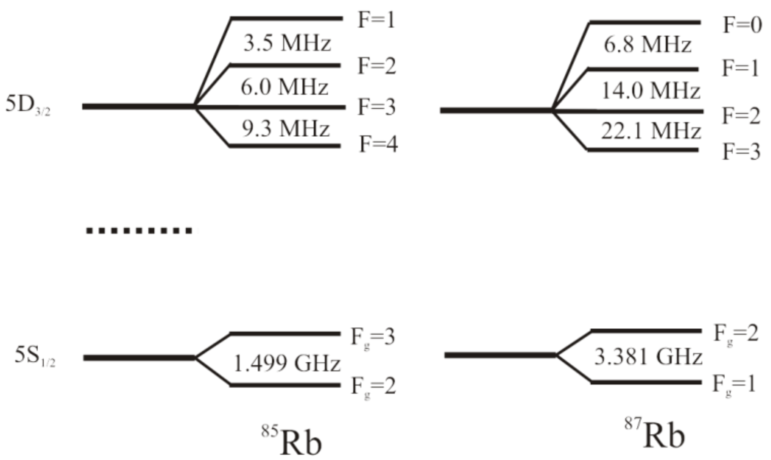

Fig. 7 Hyperfine levels for the $5S_{1/2}$–$5D_{3/2}$ two-photon transition in rubidium

The Hyperfine components spectrum of the two-photon transition ($5S_{1/2}$-$5D_{3/2}$) in rubidium is obtained by scanning the fiber laser frequency over them. The linewidth of the fiber laser of 3.4 kHz resolves sufficiently the narrow hyperfine components, as depicted in Fig.8.

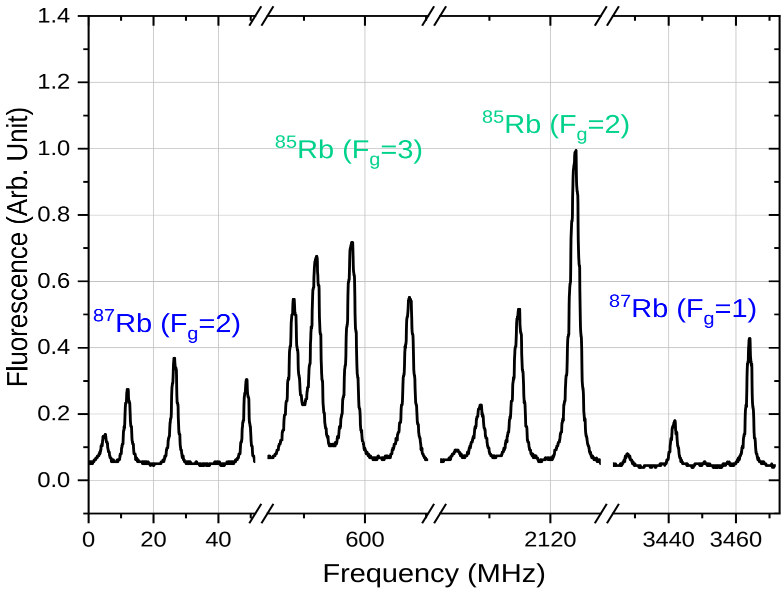

Fig. 8. Hyperfine components of the two photon transition ($5S_{1/2}$-$5D_{3/2}$) in rubidium.

The frequency stabilization system of a laser to the two-photon transition in Rb consists of a fiber laser with a linewidth of 3.4 kHz. The laser power is amplified using an Erbium-doped fiber amplifier (EDFA) to around 150 mW, which is necessary for frequency doubling through the periodically poled Lithium Niobate (WG-PPLN) crystal. Since the frequency doubling process is polarization sensitive, a polarization controller is introduced before the WG-PPLN crystal. The light is collimated out of the fiber of the WG-PPLN using a collimator, then filtered using 780 nm bandpass filter to remove the remaining 1556 nm from the frequency doubling process. A 260 mm lens is used to focus the beam to a diameter of 100 µm at the center of the rubidium gas cell. Another focusing lens and a reflector are used to reflect the beam with the same diameter at the center of the cell again. To heat the Rb cell to a temperature of 95 ± 1 °C, the cell is placed in a thermally conducting box with two thermoelectric elements installed below it. The fluorescence signal at 420 nm is detected using a photomultiplier tube (PMT). The fluorescence beam is focused at the PMT cell using a 50 mm lens and filtered using a filter at 420 nm to eliminate any stray light from the room. To eliminate the effect of the Earth's magnetic field on the transition frequencies, the Rb cell is placed into a magnetic shield that is made from the µ-metal. The PMT output is directed to the frequency locking electronics, consisting of a waveform generator, a lock-in amplifier, a high-speed PID controller, and an Oscilloscope. The system is depicted in fig. 9.

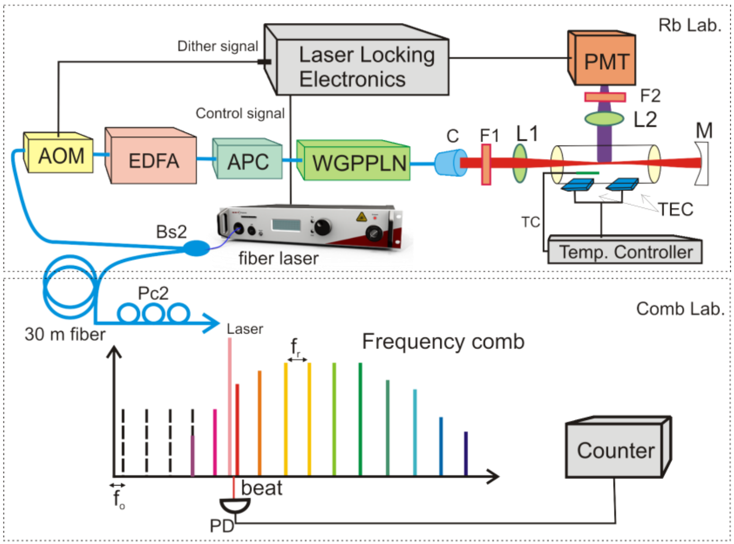

Fig. 9. Absolute frequency measurement setup of the $5S_{1/2}$ – $5D_{3/2}$ TPT in rubidium. WGPPLN: waveguide periodically-poled Lithium-Niobate crystal, APC: Active polarization controller, EDFA: Erbium-doped fiber amplifier, C: triplet collimator, L: Convex Lens, M: flat sliver mirror, F: filter, PC: polarization controller, BS: beam splitter, f_r: repetition rate, f_o: offset frequency, PD: photodetector, TEC: Thermo-electric element, AOM: Acousto-Optic Modulator.

A modulation signal is applied to the AOM to dither the laser frequency over the hyperfine component. The is a derivative like signal output of lock-in amplifier is used to lock the laser to the hyperfine component.

To measure the absolute frequency of the hyperfine components with the OFC, the laser fundamental wavelength at is transmitted through 30 m fiber to the OFC laboratory, as depicted in Fig. 9. The OFC has a spectral bandwidth of 34 nm with frequency spacing of 250 MHz centered at 1567 nm. The repetition rate and the offset frequency of the OFC are stabilized to a GPS-disciplined quartz oscillator with short-term stability of 5×10^{-12} at 1 s and long-term stability of 5×10^{-13} at 400 s. After the stabilization, all OFC modes should have the same stability of the RF reference. The stabilized laser beats with the nearest mode from the OFC and is counted using a zero dead-time counter. The changing repetition frequency method determines the mode number of the OFC mode which beats with the laser.
The absolute values of all the hyperfine components in the $5S_{1/2}$–$5D_{3/2}$ two-photon transition in rubidium are given in table 1.

Table 1. The absolute frequencies of the hyperfine components of the TPT ($5S_{1/2}$-$5D_{3/2}$) in rubidium (after correction of systematic shifts) together with the expected values.

Isotope	**Component** $F_g \rightarrow F_e$	**Measured frequency (kHz)**	**STD (kHz)**
^{85}**Rb**	3 → 1	385 240 679 711.8	3.5
	3 → 2	385 240 683 216.6	2.9
	3 → 3	385 240 689 193.2	2.3
	3 → 4	385 240 698 496.0	2.8
	2 → 1	385 242 197 573.3	3.7

	2 → 2	385 242 201 081.2	1.4
	2 → 3	385 242 207 056.9	3.3
	2 → 4	385 242 216 363.7	2.8
^{87}**Rb**	2 → 0	385 240 094 975.3	2.6
	2 → 1	385 240 101 726.6	1.7
	2 → 2	385 240 115 693.9	1.2
	2 → 3	385 240 137 803.5	1.4
	1 → 3	385 243 519 073.4	2.2
	1 → 2	385 243 533 032.4	2.6
	1 → 1	385 243 555 142.3	3.4

The effect of the various systematic shifts on the measured frequency of the hyperfine components are estimated as follows, see table 2:

1- The "AC Stark shift" is determined at each measurement by changing the laser power between 5 and 10 mW and found to be -6.8 ± 0.2 kHz/mW.

2- The "Pressure shift" Since the cell is operating at 95 ^{o}C (17 mPa), the pressure shift is calculated to be -1224 ± 34 Hz.

4- The Earth magnetic field is measured after shielding to be < 10 mG which is not believed to cause a considerable shift.

4- Black-body radiation shifts can introduce a shift of -209 ± 13 Hz.

5- The second-order Doppler shift is estimated to be -229 ± 1 Hz.

Table 2: Uncertainty budget and corrections for the $5S_{1/2}$-$5D_{3/2}$ transition in natural rubidium.

Systematic effect	Correction	Uncertainty
AC Stark shift	-34 kHz	1 kHz
Pressure shift	-1224 Hz	34 Hz
Electronics Error	0	723 Hz
Zeeman shift	0	0
Blackbody radiation shift	-209 Hz	13 Hz
Second-order Doppler shift	-229 Hz	1 Hz
Total	-35.662 kHz	±1235 Hz*

*the uncertainties are summed in quadrature.

Two-photon stabilized laser can be used to characterize the OFC which is used as a transfer oscillator in ultraprecise comparison between optical frequency standards [17]. This comparison uses different outputs branches from the OFC. Therefore, the relative stability between branches should be better than the stability of the optical frequency standards under comparison. Several noisy processes can destroy the relative stability if not common between branches. Such processes may include amplification using Erbium-doped fiber amplifier (EDFA), spectral broadening using self-phase modulation (SPM), and propagation through optical fibers. The fiber refractive index is sensitive to environmental perturbations, introducing phase noise to the optical frequencies propagating through it [18, 19]. Optical fibers are used as patch cables but also included in EDFA.

6. Conclusions

In this review article, the principle of the operation of mode-locked femtosecond lasers is explained. It has been demonstrated that the femtosecond pulses in the time domain correspond to a spectrum of equidistant modes in the frequency domain, which is called the OFC. Each mode can have a fixed, well-known frequency if the spacing between modes and their offset frequency is locked to high stability frequency standards, such as the Caesium clock. If this lock is performed, the OFC can be used for frequency measurement. The principle of optical frequency measurement with OFC is demonstrated. The OFC is used afterward to measure the hyperfine component of the two-photon transition in Rb. The absolute frequency of each component is represented, and the measurement uncertainty budget is calculated. The measurement uncertainty is less than 4 kHz.

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