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基于多普勒效应的双峰荧光光谱激光稳频技术研究

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摘 要:提出了一种使用斜光诱导荧光光谱进行激光稳频的方法,该荧光光谱是通过铯原子束与斜入射激光相互作用产生,具有结构简单、可靠性高的优点。斜光诱导荧光光谱的中心峰的半高宽是可调谐的,在不同入射角度下,分别得到了半高宽为 16.3 MHz, 18.7 MHz, 21.7 MHz, 24.4 MHz 及 26.9 MHz 的谱线,这些比单峰荧光光谱(实际测量值为 42.5 MHz)的半高宽更加窄。当使用中心峰为 24.4 MHz 的双峰荧光光谱进行激光稳频时,激光频率波动的峰峰值被限制在 70 kHz 以内,频率稳定度为 2.7×10^{-11} @ 1s, 1.2×10^{-11} @ 10 s, 4.0×10^{-12} @ 100 s, 1.4×10^{-12} @ 1 000 s, 优于使用单峰荧光光谱进行激光稳频时的频率稳定度, 3.9×10^{-11} @ 1 s, 1.4×10^{-11} @ 10 s, 4.7×10^{-12} @ 100 s, 3.4×10^{-12} @ 1 000 s。该方法有利于提高小型光抽运铯束频标的性能及可靠性。

关键词:激光稳频;双峰荧光光谱;多普勒效应;频率稳定度;光抽运铯束频标

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0 Introduction

Laser frequency stabilization played a crucial role in optically pumped Cs beam frequency standard^[1-2], which is also important for some other applications, such as optical frequency standard, high-resolution molecular and atomic spectroscopy, laser cooling and trapping of atoms, optical communications^[3-6]. Improving the stability of laser frequency by reducing frequency fluctuation can enhance the performance of the optically pumped Cs beam frequency standard^[7]. In the past several decades, it has developed various methods to stabilize the laser frequency. The most popular approach is using the Saturated Absorption Spectroscopy (SAS)^[8-10], which has realized the 5×10^{-13} at ~ 1 s, but immediately reached the flicker floor^[11]. There are also some other methods, such as Polarization Spectroscopy (PS)^[12-13], dichroic atomic-vapor laser lock (DAVLL)^[14-15], Frequency Modulation (FM) spectroscopy^[16], Doppler-free bichromatic spectroscopy^[17]. These methods above are mostly based on the thermal vapor cell in a system which is kind of complicated, and facing the problems of the temperature of vapor, the light power and the magnetic field, which caused the unstable frequency of laser. The technique of Modulation Transfer Spectroscopy (MTS) with its short-term frequency stability has recently reached 2.6×10^{-13} at averaging time of 5 s^[18], can provide a signal which is independent of fluctuations in polarization, temperature and beam intensity^[19], but at the cost of a more complex system.

For compact optically pumped Cs beam frequency standard, reliability is an important thing that need to be considered, which is probably easier to realize through a simple structure. And the method of laser frequency stabilization by laser-induced (one-peak) fluorescence spectroscopy is developed^[20-24]. This method has

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enhanced the system reliability, but introduce a problem of linewidth broadening, which caused by divergent atomic beam. For Doppler-free spectroscopic technique in Ref. [25], its dispersive signal has a high signal-to-noise ratio and no Doppler-background, but the drawback of this work is that the locking point may be not located at the center of the atomic transition if two light beams are not well managed.

In this paper, we present a method which has a very simple and reliable structure to stabilize the laser frequency by Two-Peak Fluorescence Spectroscopy (TPFS). The results indicate that the performance of this method is better than the method of laser frequency stabilization by One-Peak Fluorescence Spectroscopy (OPFS). With this method, we get the narrower half-width of the fluorescence signal, and the better laser frequency stability, which is suitable for a compact optically pumped Cs beam frequency standard. In the next, we will show the details of this method

1 Experimental setup

The experimental setup for stabilizing the laser frequency by TPFS is illustrated in Fig. 1. A light beam with 852 nm from an External-Cavity Diode Laser (ECDL) is operated at $6S_{1/2}$, $F=4 \leftrightarrow 6P_{3/2}$, $F'=5$ cycling transition of Cs atoms. Its free-running linewidth is less than 200 kHz. And its frequency can be accurately tuned by adjusting the injection current with a rate of 2.5 MHz/ μ A. The laser is divided into two beams by a Polarization Beam Splitter (PBS) after passing through an isolator, which is used to prevent the optical feedback. One beam is used for generating a calibration reference. Another is split into two parts by PBS for frequency stabilization, with a Half-Wave Plate (HWP) added to adjust the laser power of two paths. After reflecting by mirrors, the two beams propagate contrarily and coincide exactly with each other, they will interact with Cs atomic beam and emit fluorescence when the laser frequency is scanned to the cycling transition. After fluorescence collection and photoelectric conversion, the fluorescence spectroscopy is generated. A sine modulation signal with a frequency of about 2 kHz and the first (or third) derivative signal are sent to the laser controller. When the electronic servo circuit is operating, the laser is locked with a 2 MHz wide scan at a frequency of about 2 kHz.

The Cs beam tube is arranged in the experiment with the atomic beam is un-perpendicular to the laser beam, as shown in Fig. 1. For Cs atoms with the speed of v , the frequency shift due to the first-order Doppler effect is given by $\Delta\omega = v\omega_0 \cos(\pi/2 + \theta)/c$, where $\Delta\omega$ is frequency shift in radian, ω_0 is the atomic resonance frequency, c is the speed of light, $(\pi/2 + \theta)$ is the angle between the direction of atoms and light beam. The frequency shift of the central frequency of fluorescence spectroscopies induced by these two light beams are $\pm v\omega_0 |\sin\theta|/c$ separately. When these two fluorescence spectroscopies add up, the two-peak fluorescence spectroscopy is formed. The tube was packaged by magnetic shields and the magnetic field intensity inside is smaller than 1 mG. The Cs atomic beam is emitted by a coaxial hole array of collimator, its temperature is controlled at 100 °C with fluctuation less than $\pm 0.1^\circ\text{C}$, which is more stable owing to the vacuum environment

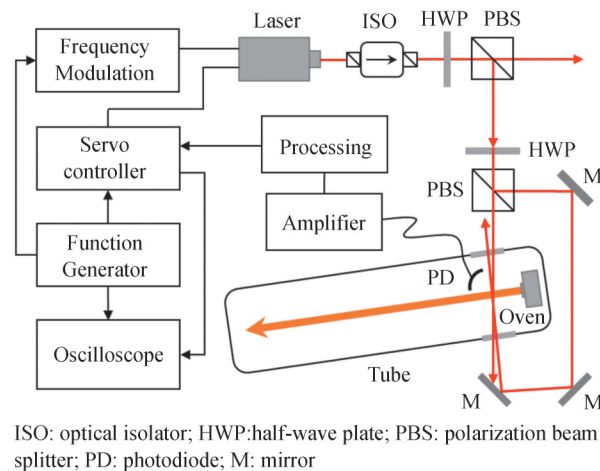


Fig. 1 Schematic of experimental arrangement

than the temperature of vapor cell in the atmosphere. The distance between the interaction region and the collimator is about 30 mm, where the atomic beam have a bigger divergent angle.

Relevant hyperfine energy levels of Cs atoms associated with the experiment are shown in Fig. 2. The $6S_{1/2}$, $F=4 \leftrightarrow 6P_{3/2}$, $F'=5$ is a cycling transition, and the natural linewidth of the $6P_{3/2}$ state is about 5.2 MHz.

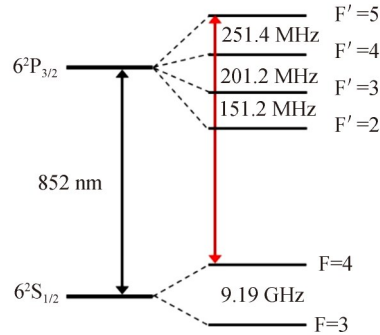


Fig. 2 Relevant energy levels of Cs atoms

2 Experimental results and discussion

The TPFS of cycling transition is obtained by oblique-incidence laser at different angles, as shown in Fig.3. The magnification of the fluorescence signal is different from each other, for ensuring that the amplitude of the fluorescence peak is about same. Although, the laser beams are not perpendicular to the Cs atomic beam, but there is no crossover line in the fluorescence spectroscopy. Because the number of atoms corresponding to the appropriate velocity is small, owing to the velocity distribution in the atomic beam and the slight oblique angle. Besides, the TPFS of 4~5 line strength is much stronger than others for the reasons of electric dipole selection rules and optical pumping principle^[26].

The half-width (as marked by a pair of arrows in Fig.3) of the measured TPFS is changed with oblique angle θ . And we get 16.3 MHz, 18.7 MHz, 21.7 MHz, 24.4 MHz and 26.9 MHz at different angles separately, which are larger than the natural linewidth (5.2 MHz) but smaller than the linewidth of measured OPFS (about 42.5 MHz, in Fig. 6). In addition, the amplitude of the central peak will decrease and tend to zero when the θ decreases and tend to zero, which result in the deterioration of the signal-to-noise ratio. The inset figure in Fig.3 shows the first derivative A and inverted third derivation B frequency discriminating curve, which the TPFS generates with a linewidth of about 24.4 MHz. And we can see that the first derivative and inverted third derivative of TPFS are similar. The under line is the calibration reference.

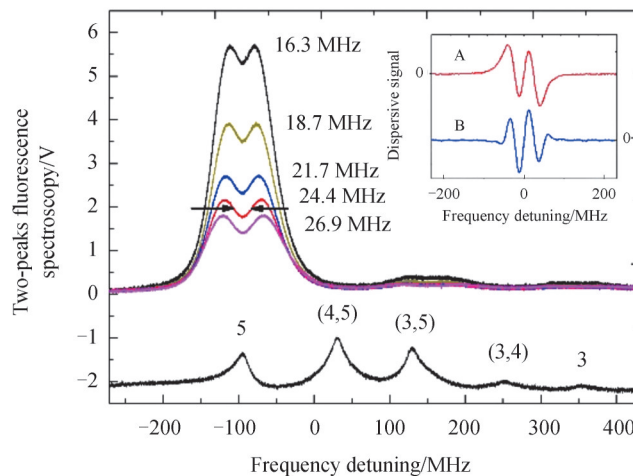


Fig. 3 Measured TPFSs of Cs-D₂ line induced by oblique-incidence laser at different angles

In this experimental scheme, the locking point will be located at the center of the atomic transition when the laser power of two beams is the same and the two beams coincide, which is easy to realize. The laser frequency is locked in the $6S_{1/2}, F=4 \leftrightarrow 6P_{3/2}, F'=5$ transition of the Cs-D₂ line. Frequency fluctuation can be estimated by the voltage of the error signal. Fig. 4 (a) shows the results of the laser is free-running with a recording time of about 300 s. The estimated frequency drift is about 2.3 MHz, which is due to the temperature, current, and mechanical fluctuations [27]. When the electronic servo circuit operating, the laser fluctuation is suppressed to about 70 kHz peak-peak, as shown in Fig. 4 (b).

The laser frequency stability is obtained by the self-evaluation method, and the result of Allan deviation is shown in Fig. 5. The square with the red line shows the stability of the laser stabilized by TPFS and the circles with the blue line represent the stability of the laser stabilized by OPFS. The recording time of both of them is about one day. The laser frequency stability of the red line is about 2.7×10^{-11} at 1s which is better than 1×10^{-10} , and less than 2.0×10^{-12} at 500 s which is better than 5×10^{-12} . These meet the stability requirements of laser light for the optically pumped Cs atomic clock [25]. The Allan deviation $\sigma_y(\tau)$ is proportional to $\tau^{-1/2}$ for $\tau < 1000$ s, then reach to the flicker floor. And the minimum $\sigma_y(\tau)$ is about 1.4×10^{-12} at 1000 s.

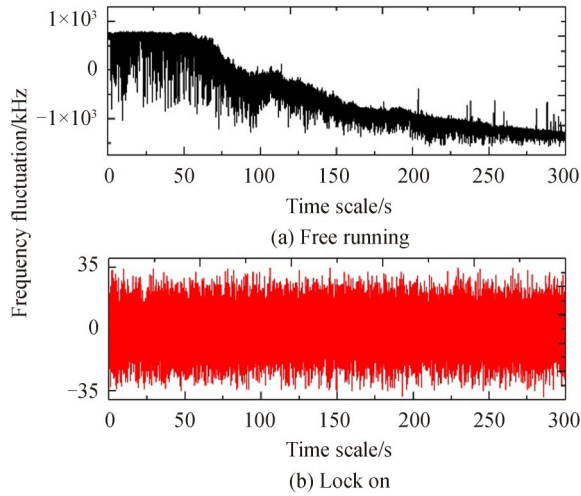


Fig. 4 Frequency fluctuation of error signal without and with the locking circuit operating

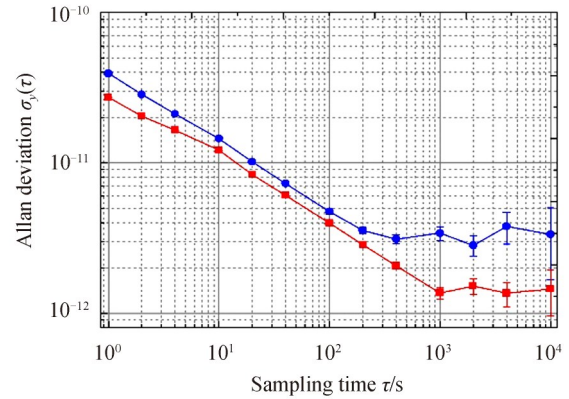


Fig. 5 The Allan deviation of the stabilized 852 nm laser by TPFS and OPFS separately

In experiment, the temperature fluctuation of the oven is controlled within ± 0.1 °C, so the relative fluctuation of beam flux is about 0.6%. The oven and Cs atomic beam are shelled by three magnetic shielding material layers, with a residual magnetic field of less than 1 mG. The measured vacuum degree is better than 10^{-5} Pa, which will not cause a collision between atoms. Besides, the center of TPFS is not affected by the intensity of laser or beam flux. The influence of the above factors on frequency stability has been reduced to negligible level. So, the flicker floor is probably caused by the light beam angle shift, which will limit the medium- and long-term frequency stability [22].

For OPFS, the measured linewidth is about 42.5 MHz, as shown in Fig. 6, due to the 30 mm distance between the interaction region and the collimator where the divergence angle of the atomic beam is bigger.

Replacing TPFS by OPFS, keeping the amplitude of dispersive signals roughly same and the other parameters invariable, we obtain the error signal after the locking circuit operating with the

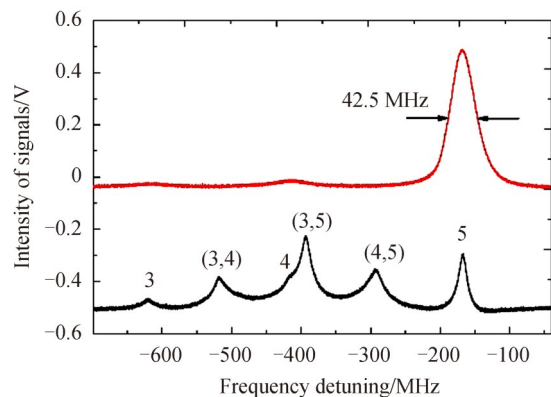
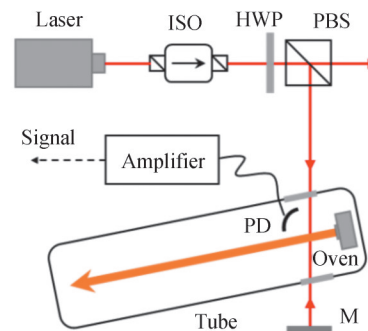


Fig. 6 The OPFS of Cs-D₂ line. The under line is the calibration reference

frequency fluctuation of about 100 kHz peak-peak. In Fig. 5, the circles with blue line represent the stability of laser stabilized by OPFS. The Allan deviation $\sigma_y(\tau)$ is about 3.9×10^{-11} at 1 s and about 3.2×10^{-12} at 500 s, which also fulfill the stability requirements of laser light for the optically pumped Cs atomic clock [25]. And the $\sigma_y(\tau)$ is proportional to $\tau^{-1/2}$ for $\tau < 400$ s, then reach to the flicker floor, and the $\sigma_y(\tau)$ is about 3.4×10^{-12} at 1 000 s.

By comparing these two methods, its results have been shown in Fig. 5. We can see that the frequency stability of laser locking by TPFS is better than the frequency stability of laser locking by OPFS. We have used the same low-pass filter to reduce the noise of frequency-discrimination signals for both these two methods. The differences between these two methods in our experiment are that they have different half-width (about 24.4 MHz and 42.5 MHz separately) and different signal-to-noise ratio (the amplitude ratio between them is about 1/3). The method of laser frequency locking by TPFS has lower signal-to-noise ratio but has a narrower half-width.

For the generation of TPFS, further work can be done to simplify the arrangement of the experiment. As shown in Fig. 7, after being split by PBS, one of the laser beams spread straightly through the optical windows and returns along its original path after reflection by a mirror. A TPFS can also be obtained but exist left-right asymmetry, which will cause the locking point to be shifted from the center of $6S_{1/2}, F=4 \leftrightarrow 6P_{3/2}, F'=5$ transition. But this character is suitable for laser cooling, because of the frequency locking point can be easily tuned by changing the propagation direction of the Cs atomic beam.



ISO: optical isolator; HWP: half-wave plate; PBS: polarization beam splitter; PD: photodiode; M: mirror

Fig. 7 The simplified experimental arrangement for generating the TPFS

3 Conclusion

In conclusion, we have presented a very simple scheme to stabilize the laser frequency by TPFS, and the performance of this method is better than the performance of laser stabilization by OPFS. The linewidth of TPFS is tunable, and we get 16.3 MHz, 18.7 MHz, 21.7 MHz, 24.4 MHz and 26.9 MHz at the different oblique incidence angles separately. The frequency fluctuation of stabilized laser is about 70 kHz peak-peak by TPFS with a linewidth of about 24.4 MHz, and the frequency instability is 2.7×10^{-11} at 1 s, and less than 2.0×10^{-12} at 500 s, which are fulfill the requirements of the compact optically pumped Cs beam frequency standard. Future works will focus on replacing the ECDL by Distributed Feedback (DFB) diode laser and reducing the noise of the laser.

References

- [1] VANIER J, AUDOIN C. The classical caesium beam frequency standard: fifty years later[J]. Metrologia, 2005, 42: S31-S42.
- [2] AUDOIN C. Caesium beam frequency standard: classical and optically pumped[J]. Metrologia, 1992, 29: 113-134.
- [3] GILL P. Optical frequency standards[J]. Metrologia, 2005, 42: S125-S137.
- [4] LIN H, WANG T, MOSSBERG T W. Experimental demonstration of swept-carrier time-domain optical memory[J]. Optics Letters, 1995, 20(1): 91-93.
- [5] VANIER J. Atomic clocks based on coherent population trapping: a review[J]. Applied Physics B, 2005, 81: 421-442.
- [6] PHILLIPS W D. Laser cooling and trapping of neutral atoms[J]. Reviews of Modern Physics, 1998, 70(3): 721-741.
- [7] DIMARCQ N, GIORDANO V, CERREZ P, et al. Analysis of the noise sources in an optically pumped cesium beam resonator[J]. IEEE Transactions on Instrumentation and Measurement, 1993, 42(2): 115-120.
- [8] HALL J L, BORDE C J, UEHARA K. Direct optical resolution of the recoil effect using saturated absorption spectroscopy[J]. Physical Review Letters, 1976, 37(20): 1339-1342.
- [9] VANSTEENKISTE N, GERZ C, KAISER R, et al. A frequency-stabilized LNA laser at 1.083 um: application to the manipulation of helium 4 atoms[J]. Journal de Physique II, 1991, 1(12): 1407-1428.
- [10] IM K B, JUNG H Y, OH C H, et al. Saturated absorption signals for the Cs D₂ line[J]. Physical Review A, 2001, 63

- (3): 034501.
- [11] ROVERAE G D, SANTARELL G, CLAIRON A. A laser diode system stabilized on the Caesium D₂ line[J]. Review of scientific instruments, 1994, 65(5): 1502-1505.
- [12] WIEMAN C, HANSCH T W. Doppler-free laser polarization spectroscopy[J]. Physical Review Letters, 1976, 36(20) : 1170-1173.
- [13] HARRIS M L, ADAMS C S, CORNISH S L, et al. Polarization spectroscopy in rubidium and cesium [J]. Physical Review A, 2006, 73(6): 062509.
- [14] CORWIN K L, LU Z T, HAND, et al. Frequency-stabilized diode laser with the Zeeman shift in an atomic vapor[J]. Applied Optics, 1998, 37(15): 3295-3298.
- [15] SU Dianqiang, LIU Ruijuan, ZHANG Chuanbiao, et al. Laser frequency stabilization in sub-nanowatt level using nanofibers[J]. Journal of Physics D: Applied Physics, 2018, 51: 465001.
- [16] GAHEY C B. Frequency-modulation spectroscopy: a new method for measuring weak absorptions and dispersions [J]. Optics Letters, 1980, 5(1): 15-17.
- [17] VANOOIJEN E D, KATGERT G, VANDERSTRATEN P. Laser frequency stabilization using Doppler-free bichromatic spectroscopy[J]. Applied Physics B, 2004, 79 :57-59.
- [18] SHANG Haosen, ZHANG Tongyun, MIAO Jianxiang, et al. Laser with 10⁻¹³ short-term instability for compact optically pumped cesium beam atomic clock[J]. Optics Express, 2020, 28(5): 6868-6880.
- [19] MCCARRON D J, KING S A, CORNISH S L. Modulation transfer spectroscopy in atomic rubidium[J]. Measurement Science and Technology, 2008, 19: 105601.
- [20] WANG Depel, XIE Linzhen, WANG Yiqiu. GaAlAs laser diode frequency locked at the D₂ line of Cs atoms in an atomic beam[J]. Optics Letters, 1988, 13(10): 820-822.
- [21] DIMARCO N, GIORDANO V, THEOBALD G, et al. Comparison of pumping a cesium beam tube with D₁ and D₂ lines [J]. Journal of Applied Physics, 1991, 69(3): 1158-1162.
- [22] WANG Qing, DUAN Jun, QI Xianghui, et al. Improvement of laser frequency stabilization for the optical pumping cesium beam standard[J]. Chinese Physics Letters, 2015, 32(5): 054206.
- [23] SHI Hao, MA Jie, LI Xiaofeng, et al. Design of an atom source collimator for a compact frequency-stabilized laser[J]. Applied Optics, 2018, 57(22): 6620-6625.
- [24] ZHAO Xingwen, CHEN Haijun, YANG Lin, et al. Stabilizing the optical frequency by laser-induced fluorescence in optically pumped Cs beam frequency standard [J]. Journal of Astronautic Metrology and Measurement, 2020, 40(1) : 17-22.
- [25] WANG Qing, QI Xianghui, Liu Shuyong, et al. Laser frequency stabilization using a dispersive line shape induced by Doppler Effect[J]. Optics. Express, 2015, 23(3): 2982-2990.
- [26] AVILA G, GIORDANO V, CANDELIER V, et al. State selection in a cesium beam by laser-diode optical pumping[J]. Physical Review A, 1987, 36(8): 3719-3728.
- [27] ZHAO Y T, ZHAO J M, HUANG T, et al. Frequency stabilization of an external-cavity diode laser with a thin Cs vapour cell[J]. Journal of Physics D: Applied Physics, 2004, 37: 1316-1318.

Laser Frequency Stabilization by Two-peak Fluorescence Spectroscopy Induced by Doppler Effect with Laser at Oblique Incidence

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Abstract: Improving the stability of laser frequency by reducing frequency fluctuation can enhance the performance of the optically pumped Cs beam frequency standard. It has developed various methods to stabilize the laser frequency, such as the Saturated Absorption Spectroscopy (SAS), Polarization Spectroscopy (PS), Dichroic Atomic-Vapor Laser Lock (DAVLL), Frequency Modulation (FM) spectroscopy, Doppler-free bichromatic spectroscopy. These methods above are mostly based on the thermal vapor cell in a system which is kind of complicated and facing the problems of the temperature of vapor, the light power and the magnetic field, which caused the unstable frequency of laser. For compact optically pumped Cs beam frequency standard, reliability is an important thing that need to be considered,

which is probably easier to realize through a simple structure. Therefore, the method of laser frequency stabilization by laser-induced (one-peak) fluorescence spectroscopy is developed. This method has enhanced the system reliability, but introduce a problem of linewidth broadening, which caused by divergent atomic beam. In this paper, we propose a method to stabilize the 852 nm laser frequency by fluorescence spectroscopy with two peaks, which have a simple and reliable structure, and can reduce the Doppler effect caused by divergent atomic beam. The fluorescence spectroscopy with two peaks is induced by two laser beams and atomic beam at oblique incidence. The two laser beams propagate contrarily and coincide exactly with each other. After fluorescence collection and photoelectric conversion, the two-peak fluorescence spectroscopy is formed. By changing the oblique angle θ , the half-width of the central peak of the two-peak fluorescence spectroscopy can be tuned, and we get 16.3 MHz, 18.7 MHz, 21.7 MHz, 24.4 MHz and 26.9 MHz at the different oblique incidence angles separately, which is narrower than the linewidth of one-peak fluorescence spectroscopy whose measured linewidth is about 42.5 MHz. By using the two-peak fluorescence spectroscopy to stabilize the laser, the locking point will be located at the center of the atomic transition when the power of two laser beams is the same and the two laser beams coincide with each other, which is easy to realize. But this method has a drawback of deterioration of the signal-to-noise ratio when the oblique angle θ decreases and tend to zero. When the laser frequency is stabilized by the central peak of a two-peak fluorescence signal with a half-width of about 24.4 MHz, the frequency fluctuation is suppressed to 70 kHz peak-peak and the estimated frequency stability is about 2.7×10^{-11} at 1 s, 1.2×10^{-11} at 10 s, 4.0×10^{-12} at 100 s, 1.4×10^{-12} at 1 000 s. In order to compare the performance between the two-peak and one-peak fluorescence spectroscopies, we keep the amplitude of dispersive signals roughly same and the other parameters invariable. And we obtain the error signal by one-peak fluorescence signal with the Allan deviation $\sigma_y(\tau)$ is about 3.9×10^{-11} at 1 s, 1.4×10^{-11} at 10 s, 4.7×10^{-12} at 100 s, 3.4×10^{-12} at 1 000 s. The results show that the method of laser frequency stabilization by two-peak fluorescence spectroscopy is better. This method would help improving the performance and reliability of the compact optically pumped Cs beam frequency standard. For the generation of two-peak fluorescence spectroscopy, further work can be done to simplify the arrangement of the experiment. The laser beam spreads straightly through the optical windows and returns along its original path after reflecting by a mirror. The two-peak fluorescence spectroscopy can also be obtained but exist left-right asymmetry, which will cause the locking point to be shifted from the center of transition. But this character is suitable for laser cooling, because of the frequency locking point can be easily tuned by changing the propagation direction of the Cs atomic beam.

Key words: Laser frequency stabilization; Two-peak fluorescence spectroscopy; Doppler effect; Frequency stability; Optically pumped Cs beam frequency standard

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