High-performance molecular iodine-stabilized laser

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***Abstract*—This study focuses on the development and optimization of the frequency-stabilized laser system based on molecular iodine, aiming to meet the demands of high-precision measurements. We have developed two high-performance molecular iodine optical frequency references based on modulation transfer spectroscopy, both referencing the R(56)32−0: a₁ transition of molecular iodine. The 30 cm long saturated absorption cell is used by the first system and operated at a pressure of 3.3 Pa to achieve high short-term stability, with a fractional frequency instability of 3.3 × 10^{-15} at 2 s. The unsaturated absorption cell with a length of 25 cm is employed by the second system and maintained at a pressure of 0.88 Pa to achieve high long-term stability, with a fractional frequency instability of 1.7 × 10^{-15} at 10000 s.**

Keywords*— *Frequency-stabilized laser, molecular iodine, frequency instability

I. Introduction

Optical frequency standards have extensive potential applications in spectroscopy, laser ranging, optical clocks, and testing fundamental physic. There is an urgent need to create portable, robust, and simple optical clocks for both industrial and scientific applications. Especially for space applications, which has strict requirements for power consumption, volume and mass, the most promising method is still to use Doppler-free spectroscopy to lock the laser on the transition lines of atoms or molecules. Among them molecular iodine is one of the optimal candidates because its spectral lines cover a wide wavelength range and have narrow linewidth, strong absorption, which means high signal-to-noise ratio (SNR). Its fractional frequency instability can meet the 10^{-15} level in the averaging time of 1-10000 s [1-9].

In this study, we demonstrate the development of the Nd: YAG solid-state laser that is locked to the R (56)32-0: a_1 transition line of the molecular iodine near 532 nm based on modulation transfer spectroscopy (MTS). We have developed two systems, primarily differentiated by their use of iodine absorption cell. A saturated absorption iodine cell is utilized by the first system (called MI1), which operates at a pressure of 3.3 Pa and exhibits a higher signal-to-noise ratio (SNR). This cell demonstrates a short-term frequency instability of 3.3 $\times 10^{-15}$ at 2 s averaging time [6]. In contrast, an unsaturated absorption iodine cell is used by the second system (called MI2), which is maintained at a pressure of 0.88 Pa without the implementation of active temperature control. This cell achieves superior long-term stability, exhibiting a frequency instability of 1.7$\times 10^{-15}$ at 10,000 s averaging time [9].

II. Experimental Setup

In the experiment, the Nd: YAG solid-state laser is locked to the R (56)32-0: a_1 transition line of the molecular iodine near 532 nm based on the MTS. The schematic diagram of the iodine-stabilized laser system is shown in Fig. 1a. The 532 nm laser beam is divided into pump and probe beams, each modulated by acousto-optic modulators (AOMs) operating at 100 MHz and 80 MHz frequencies, respectively. This modulation not only ensures power stability but also mitigates interference artifacts stemming from differential frequency effects. Subsequently, the pump beam passes through the wedged electro-optic modulator (EOM) and is directed into the MTS module via an optical fiber. The optical path diagram of the spectroscopy module is shown in Fig. 1b. The physical devices of the two systems are completely identical except for the iodine absorption cell. In the saturated absorption cell system, the optical power for the pump and probe beams is allocated at 9.8 mW and 2.6 mW, respectively. Conversely, in the unsaturated absorption cell system, the optical power for these beams is distributed at 5.6 mW and 1.4 mW, respectively.

(a)

(b)

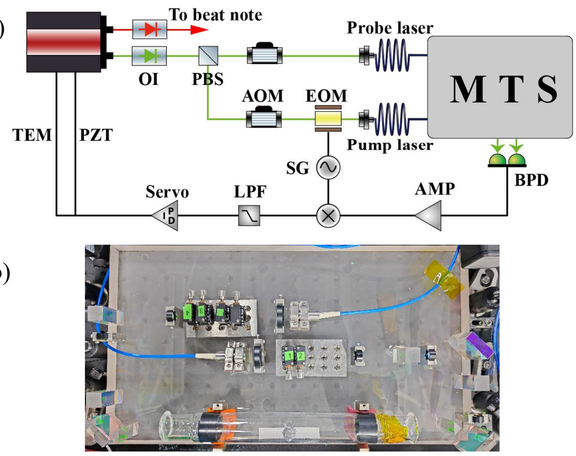

Fig. 1. (a) Schematic diagram of the iodine-stabilized laser system. AOM: Acousto-optic modulator. MTS: Modulation Transfer Spectroscopy. PBS: polarized beam splitter; BPD: balanced photodetector, OI: optical isolator, SG: signal generator; EOM: electro-optic modulator, AMP: amplifier. (b) Photograph of the integrated optical path module of the iodine spectroscopy setup.

III. Results

Figure 2 illustrates the observed hyperfine transition lines, demonstrating that the a_1 hyperfine transition exhibits a superior SNR in comparison to the BIPM-recommended a_{10} hyperfine transition. Consequently, we have implemented laser locking on the a_1 hyperfine transition. The SNR of the a_1 hyperfine transition, achieved with a saturated cell setup, is measured at 420 with a 10 kHz resolution bandwidth, yielding a broadened linewidth of approximately 680 kHz. In contrast, utilizing an unsaturated cell, the SNR of the a_1 hyperfine transition is recorded at 70, within the same resolution bandwidth, with the corresponding broadened linewidth being approximately 340 kHz.

Figure 3 shows the Allan standard deviation of the iodine stabilized Nd: YAG solid-state laser. The frequency instability of the saturated absorption cell is evaluated by beating with an ultra-stable cryogenic sapphire cavity laser, and the results are shown in Fig. 3 with the black line. It shows that the instability reaches a minimum value 3.3×10^{-15} at 2 s and 4 s averaging time and the frequency instability result of the laser system is better than 6×10^{-15} at averaging times from 1 s to 10000 s.

The short-term performance (1 s- 100 s averaging time) of the unsaturated absorption cell is evaluated with the saturated absorption cell. The long-term performance (100 s-10000 s averaging time) of the system is evaluated with the frequency comb, which has a very good performance at longer time since it is locked to a hydrogen maser. And the results are shown in Fig. 3 with the red line. The calculated Allan deviation shows the fractional frequency instability of the system is 1.4×10^{-14} at an averaging time of 1 s, and decreases to a level of 1.7×10^{-15} at an averaging time of 10,000 s without any detrending.

MI2 exhibits relatively lower short-term stability in comparison to MI1. This is because the temperature of the cold finger in MI1 is -2 °C, which means it has a larger vapor pressure. The SNR in MI1 is seven times larger while the linewidth is only two times larger, so the short-term stability of MI1 is better. Conversely, MI2 demonstrates superior long-term stability. The result indicates that the unsaturated cell has superior long-term frequency stability and are less sensitive to the environment.

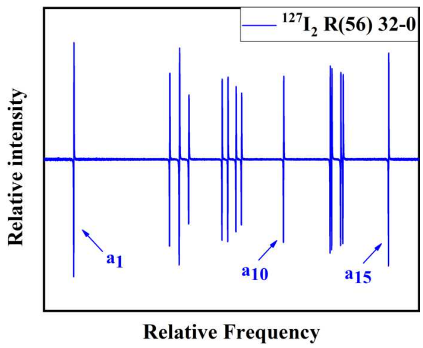

Fig. 2. The R (56)32-0 transition of $^{127}I_2$ at 532 nm in a bandwidth of 10 kHz.

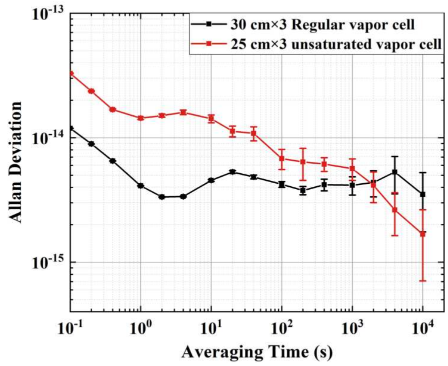

Fig. 3. Frequency instability of the iodine stabilized laser system, given as Allan deviation for a given time span τ. The measured short-term stability of the saturated absorption cell (black line) reaches 3.3×10^{-15} at 2 s. The long-term performance of the unsaturated absorption cell (red line) reaches 1.7×10^{-15} at 10000 s.

IV. Conclusions

We have successfully achieved the high-performance molecular iodine optical frequency reference based on MTS. The frequency instability of both systems reached the level of 10^{-15}. The saturated absorption iodine cell works at a higher pressure of 3.3 Pa, has a higher SNR, and achieves a short-term stability of 3.3×10^{-15} at 2 s. The long-term stability of the unsaturated absorption iodine cell without active temperature control reached 1.7×10^{-15} in the averaging time of 10000 s.

Distinct noise peak characteristics are observed in each of the two systems at the 2 seconds. Future work will involve a detailed analysis of noise sources to enhance system stability and miniaturize the system for portable operation, with the goal of housing the entire system within a 3U chassis.

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