

# Dual optical–microwave molecular clock

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**Abstract.** Molecular transitions provide a versatile platform for exploring fundamental physics and advancing timekeeping technologies. We report a dual-output molecular clock system based on the carbon monoxide transition R(9) at 1567 nm. The system achieves a frequency stability of  $6 \times 10^{-14}$  and bridges optical and microwave domains using a frequency comb. Comparative tests with rubidium and hydrogen clocks suggest promising performance for precision measurements and portable applications. The dual-output capability of this molecular clock holds significant potential for applications in metrology, navigation, and fundamental physics research.

**Keywords:** frequency reference; laser frequency stabilization; molecular spectrum.

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## 1 Introduction

High-precision timekeeping is essential for a wide range of scientific applications, from testing the foundations of physics<sup>1–5</sup> to enabling modern technologies such as global positioning systems.<sup>6–9</sup> Atomic clocks, particularly those based on cesium and strontium, have been at the forefront of this field, achieving remarkable accuracy and stability with uncertainties on the order of  $10^{-19}$ .<sup>10</sup> These clocks have enabled fundamental tests of general relativity<sup>11</sup> and are critical for precise time synchronization in telecommunication and satellite-based systems. However, their reliance on complex environments—such as ultra-high vacuum chambers and precise magnetic shielding—limits their scalability and portability. Consequently, developing more robust, portable, and scalable timekeeping systems has become a key challenge for both applied and fundamental science.

In contrast, molecular rovibrational transitions offer unique advantages due to their sensitivity to physical constants and environmental factors. This sensitivity has led to growing interest in their application for determining fundamental constants,<sup>12–15</sup> testing fundamental symmetries,<sup>16</sup> and exploring new physics beyond the Standard Model.<sup>1,17</sup> In addition, the long natural lifetimes of molecular transitions make them attractive candidates for frequency metrology.<sup>8,9</sup> However, the complexity of

molecular structures has limited successful applications of laser cooling techniques to only a few molecules.<sup>18,19</sup> Consequently, many molecular clocks are operated under buffer-gas-cooled<sup>8</sup> or room-temperature conditions, which makes them susceptible to Doppler shifts. Despite these challenges, the relatively simple auxiliary system requirements make molecular clocks promising candidates for portable and versatile applications,<sup>20,21</sup> albeit with typically lower-frequency precision than laser-cooled atomic clocks.

Another significant advantage of molecular transitions as frequency references is their rich energy-level structure, which provides access to multiple transition options within a single molecular system. This capability enables molecular clocks to offer frequency references across a broad range of optical bands, achieving excellent optical frequency stability. Furthermore, molecules with zero nuclear spin are less susceptible to magnetic field disturbances, providing increased robustness against stray magnetic interference compared with atomic clocks. Decades of research on molecular clocks have seen widespread use of iodine molecules for laser stabilization,<sup>22–25</sup> with iodine transitions near 532 nm, for example, stabilizing lasers to a level of  $2.3 \times 10^{-14}$  at 1 s.<sup>26</sup> However, iodine transitions are sensitive to external magnetic fields due to hyperfine splittings, limiting their application in certain environments. Other molecular transitions, such as those of  $^{13}\text{C}_2\text{H}_2$  at 1.5  $\mu\text{m}$  and at 1064 nm,<sup>27,28</sup>  $\text{CH}_4$  at 3.39  $\mu\text{m}$ ,<sup>29</sup> and  $\text{OsO}_4$  near 10.6  $\mu\text{m}$ ,<sup>30</sup> have also been explored as optical frequency standards.

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In this work, we present a molecular clock system with dual optical and microwave outputs, utilizing the CO transition at 1567 nm. This transition, located within the 1.55- $\mu\text{m}$  telecommunications band, is particularly advantageous due to minimal optical fiber loss at this wavelength. The CO transition has been measured with sub-kHz accuracy<sup>31,32</sup> and exhibits negligible pressure shifts at low pressures, enabling stable, long-term operation. The well-isolated transitions of  $^{12}\text{C}^{16}\text{O}$  and the absence of hyperfine structure further reduce susceptibility to electric and magnetic field perturbations, simplifying the shielding requirements for the system. To meet the growing demand for stable microwave references, such as 10 MHz or 1 GHz, we employ a femtosecond optical frequency comb referenced to a simple quartz crystal oscillator to convert the optical reference to the microwave. By combining optical and microwave outputs, this system paves the way for a compact, self-sustained molecular clock, offering broad applications in metrology, navigation, and other fields.

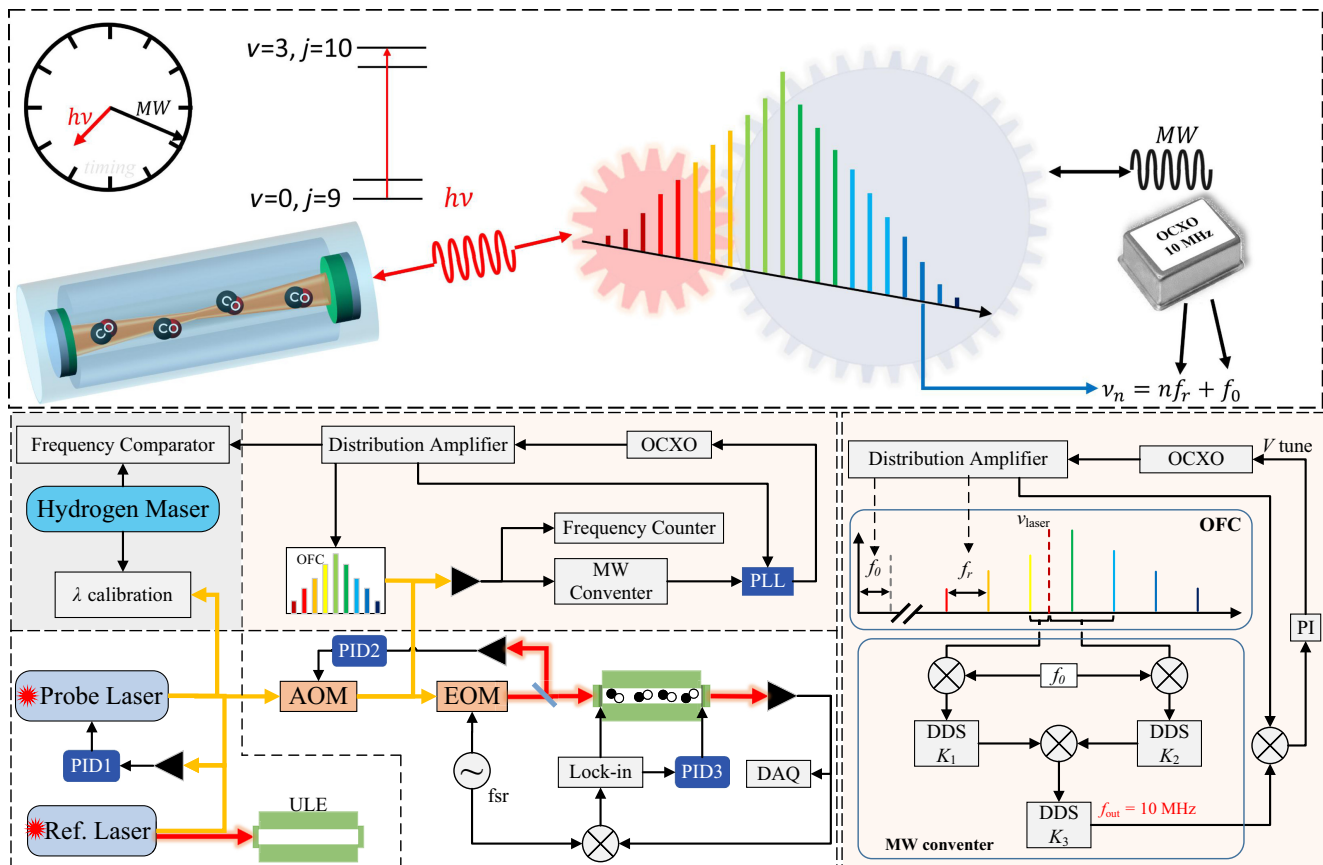
## 2 Method

The optical setup, illustrated in Fig. 1, is composed of three key components: 1) Narrow-linewidth laser system: This system functions as a high-quality optical oscillator for the molecular

clock. It combines a broad tuning range with a narrow linewidth, delivering exceptional frequency stability and precision essential for accurate clock operation. 2) Molecular frequency discriminator: The frequency discriminator stabilizes the optical oscillator by locking it to a specific molecular absorption line. A cavity-enhanced saturated absorption spectroscopy setup is employed to enhance signal strength and improve the accuracy and stability of the frequency lock. 3) Clock output and frequency conversion: To enable broader applicability, the optical clock output is converted to microwave frequencies. This conversion retains the high precision of the optical clock while making it compatible with microwave-based systems and technologies. Each of these components plays a critical role in ensuring the molecular clock's high performance and reliability.

### 2.1 Narrow-Linewidth Laser System

An external cavity diode laser (ECDL) was used to probe the spectrum. The probe laser, based on a gain chip (Thorlabs SAF1550P2, Newton, New Jersey, United States) in a Littrow configuration, has a linewidth of  $\sim 100$  kHz over a 1-s timescale and provides up to 35-mW output with a tunable range of 1530 to 1570 nm. We adopted another narrow-linewidth fiber laser (NKT Koheras AdjustiK E15) as the reference source,



**Fig. 1** Experimental setup of the molecular clock system. A  $\lambda$ -calibration system consisting of an optical frequency comb referenced to a hydrogen maser was used to assess the performance of the optical output of the molecular clock. Another optical frequency comb (OFC) was phase-locked and referenced to the 10-MHz signal from the oven-controlled crystal oscillator (OCXO) and also used to convert the optical reference to the 10-MHz microwave reference. The right panel shows the diagram of the optical-to-microwave conversion process.

which was frequency-stabilized to a ULE-glass Fabry–Pérot cavity via the Pound–Drever–Hall (PDH) locking technique. The 10-cm ULE cavity was temperature-stabilized at its zero-expansion point, effectively minimizing thermal drift. The frequency drift of the reference laser was measured to be  $\sim 0.7$  kHz/h. The ECDL was phase-locked to the reference laser by stabilizing the beat frequency between the two lasers to a designated microwave signal. A proportional–integral–derivative (PID) feedback circuit (PID1 in Fig. 1) controlled the locking servo. This configuration ensured that the ECDL inherited the high stability and precision of the reference laser.

## 2.2 Molecular Frequency Discriminator

The R(9) line in the (3 to 0) band of  $^{12}\text{C}^{16}\text{O}$  was selected as the optical frequency reference, as illustrated in Fig. 1. According to the HITRAN database,<sup>33</sup> this transition line is located at  $6383.08961\text{ cm}^{-1}$ , with an intensity of  $2.074 \times 10^{-23}\text{ cm/molecule}$  at room temperature and an Einstein coefficient of  $0.007652\text{ s}^{-1}$ . In this work, we employ the noise-immune cavity-enhanced optical heterodyne molecular spectroscopy (NICE-OHMS) method to lock the probe laser frequency to the R(9) line center of CO. This technique was selected for its exceptional detection sensitivity, enabling the acquisition of a saturated absorption signal with a high signal-to-noise ratio (SNR).

Another Fabry–Pérot cavity was used for spectral detection. The cavity was composed of a glass–ceramic spacer with a thermal expansion coefficient of  $\sim 10^{-7}/^\circ\text{C}$ , effectively minimizing cavity length variations caused by temperature drift. Two piezoelectric transducers (PZTs) were bonded at both ends of the spacer: a 13.5-mm PZT for cavity length locking servo and a 2-mm PZT for cavity length modulation. Two high-reflectivity mirrors were used, one flat and one concave with a curvature of 2 m. These mirrors, separated by 32.7 cm, had a reflectivity of 99.9974% at 1567 nm, as measured using the ring-down method. The cavity exhibited a finesse of 120,000, a free spectral range (FSR) of 458 MHz, and a cavity mode width of 3.8 kHz.

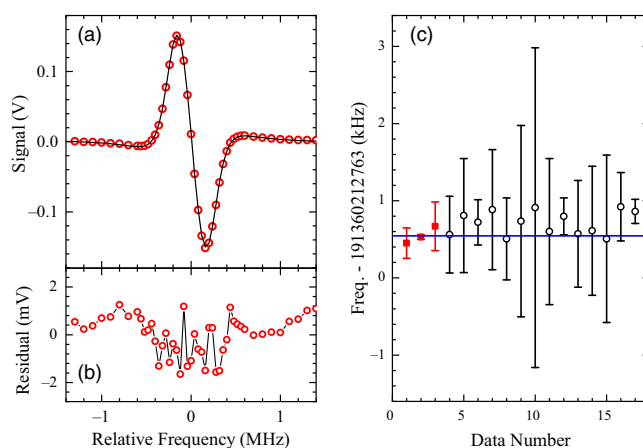
The probe beam from the ECDL passed through a fiber-coupled acousto-optic modulator (AOM) and a fiber-coupled electro-optic modulator (EOM) prior to entering the spectroscopy cavity. The AOM induced a frequency shift of  $\sim 78$  MHz. A feedback servo (not shown in Fig. 1) stabilized the intensity of the first-order diffracted light by regulating the modulation amplitude applied to the AOM. This servo reduced the laser power drift to less than 0.01% before spectroscopy measurements. The EOM was modulated at two distinct frequencies: 21 and 458 MHz. The 21 MHz modulation generated a PDH error signal, which was used in a feedback loop (PID2 in Fig. 1) to lock the diffracted beam frequency to a longitudinal mode of the spectroscopy cavity by adjusting the AOM driving frequency. We used the DeVoe–Brewer method to lock the 458-MHz modulation frequency to the cavity’s free spectral range. The transmitted light from the cavity was detected by a high-speed photodetector, and the signal was demodulated at 458 MHz to produce the NICE-OHMS signal. One challenge in this setup was the birefringence effect in the electro-optic crystal of the EOM, which introduced residual amplitude modulation (RAM) and degraded the sensitivity of laser frequency locking and spectral detection. To mitigate RAM, precise temperature control was implemented for the EOM, maintaining temperature fluctuations within  $0.01^\circ\text{C}$ .

The AOM was driven at a frequency set close to 78 MHz, corresponding to the difference between the ECDL frequency and the cavity resonance frequency. This drive frequency was locked to a reference radio frequency of 78 MHz using feedback control to the 13.5 mm PZT attached to the cavity spacer. By fine-tuning the reference radio frequency, the AOM driving frequency could be adjusted, enabling simultaneous tuning of the cavity resonance and the probe laser frequency. NICE-OHMS signals for various reference radio frequencies were recorded via a data acquisition card, enabling precise measurements of the CO R(9) saturation absorption signal.

To improve the SNR of the NICE-OHMS signal, which was initially weak, an additional modulation was applied to the 2-mm PZT at one end of the cavity spacer. This modulation had an amplitude of 160 mV and a frequency of 330 Hz. The second harmonic of the NICE-OHMS signal was demodulated using a lock-in amplifier (SRS SR860), producing the  $2f$  wavelength-modulated NICE-OHMS signal ( $2f$  wm-NICE-OHMS). Figure 2(a) presents a sample  $2f$  wm-NICE-OHMS Lamb-dip spectrum of the R(9) line.

The absolute laser frequency was calibrated using a  $\lambda$ -calibration system, which consists of an optical frequency comb (OFC) referenced to the hydrogen maser (Model VCH-1003M). The  $\lambda$ -calibration system provided a fractional frequency uncertainty of  $10^{-16}$ .

The spectrum exhibits an antisymmetric profile, which was also used as the error signal for a feedback servo (PID3 in Fig. 1). The feedback signal was applied to the 13.5-mm PZT to stabilize the cavity length. Together with the feedback servo of PID2, which locked the laser to the cavity mode, this configuration ultimately stabilized the probe laser frequency (after the AOM) to the CO R(9) transition. Accordingly, the frequency-stabilized laser beam acts as the optical clock output referenced to the CO molecular transition. Meanwhile, monitoring the AOM driving frequency enables real-time tracking of the intrinsic frequency drift of the free-running probe laser.



**Fig. 2** (a) Example of the  $2f$  wm-NICE-OHMS saturation absorption spectrum of the R(9) transition in the (3 to 0) band of  $^{12}\text{C}^{16}\text{O}$ . Scattered circles are experimental data, and the solid line represents the simulated spectrum. (b) Fitting residuals. (c) Experimental centers of the R(9) line. Black circles were derived by fitting the NICE-OHMS spectra with Lorentzian line profiles. Red squares were obtained by locking the laser to the R(9) transition.

To verify the long-term stability of the optical clock, we conducted measurements over the course of one year by intentionally turning the system on and off and recording the clock output using the  $\lambda$ -calibration system. The measurement results also provided the center frequency of the CO R(9) transition. Figure 2(c) shows the results from multiple runs conducted throughout the year. The red solid squares represent the measurements obtained by locking the laser directly to the CO line using the PID3 servo. For comparison, we also turned off the PID3 servo, recorded the NICE-OHMS spectra, and determined the line centers by fitting the spectral profiles. These results are depicted as black open circles. The uncertainties associated with each data point primarily reflect statistical variations, influenced by the number of measurements performed in each individual run of the experiment. A weighted average of all the data yields the center frequency of the R(9) line of CO as 191,360,212,763.545 kHz, with a statistical uncertainty of 0.07 kHz.

### 2.3 Clock Output and Frequency Conversion

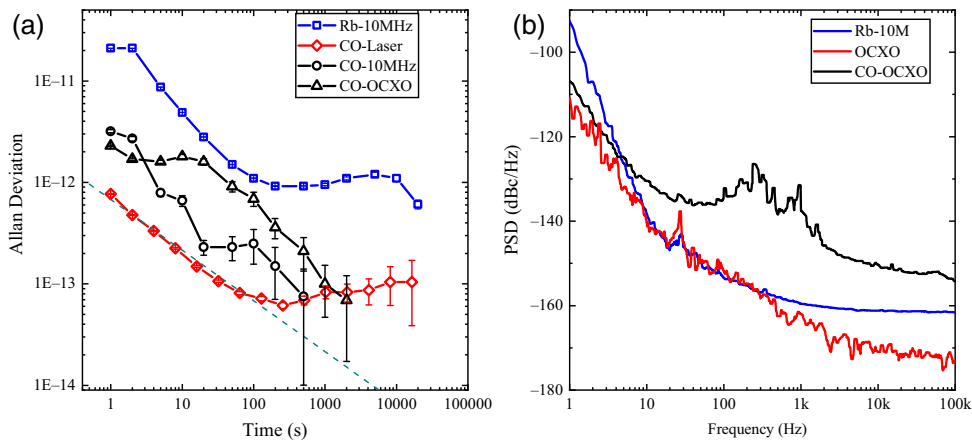
We obtained an optical output at 1567 nm from the clock system by locking the laser to the  $2f$  wm-NICE-OHMS signal of the R(9) line. Its frequency was measured by the  $\lambda$ -calibration system, which was referenced to the hydrogen maser. As shown in Fig. 3, the Allan deviation of the optical frequency was  $7.7 \times 10^{-13}$  at 1 s and reached  $7 \times 10^{-14}$  at 100 s. This stability was primarily limited by the signal-to-noise ratio of the  $2f$  wm-NICE-OHMS saturation signal. The observed stability is consistent with the SNR of 3000 shown in Fig. 2 and the 300-kHz linewidth of the  $2f$  wm-NICE-OHMS spectrum.

The frequency-stabilized laser can serve as a direct optical frequency reference. However, due to its monochromatic nature and the limited bandwidth of available photodetectors, it can only be used as a reference for other lasers operating within the same spectral band. This constraint limits its practical applicability as a versatile optical frequency reference. To address

this limitation, we extended the functionality of the optical clock by binding it to an optical frequency comb. The comb enabled the optical clock frequency to be transferred to other wavelengths and converted into a microwave frequency. For this purpose, we employed another OFC with a repetition frequency ( $f_r$ ) of  $\sim 205$  MHz and an offset frequency ( $f_0$ ) of 250 MHz. The OFC was referenced to an oven-controlled crystal oscillator (OCXO, model EP-01060HU), which provided a low-noise 10-MHz reference signal through a distribution amplifier (model SRS FS-735). The OCXO exhibited a frequency stability of  $2 \times 10^{-12}$  at 1 s. The system structure used for this frequency conversion is illustrated in Fig. 1.

Beat signals between the frequency-stabilized laser and two adjacent teeth of OFC were recorded simultaneously. As shown in Fig. 1, both radio frequency (RF) signals were filtered and amplified separately. The two beat frequencies, denoted as  $f_{b+}$  and  $f_{b-}$ , corresponded to comb teeth with indices  $N$  and  $N + 1$ , respectively, as described in Eqs. (1) and (2). These beat frequencies incorporate contributions from both the repetition frequency  $f_r$  and the carrier-envelope offset frequency  $f_0$  of the comb.

To suppress noise associated with the carrier-envelope offset frequency  $f_0$ , the two beat signals were mixed using double-balanced mixers (DBMs). The resulting signals were processed in direct digital synthesis (DDS) systems, where they were divided by factors of  $M/N$  and  $M/(N + 1)$ , as defined in Eqs. (3) and (4). After further processing in the DDS systems, the output RF frequencies were mixed again using another DBM to eliminate noise associated with the comb repetition frequency  $f_r$ . The final output frequency,  $f_{\text{out}}$ , was derived, depending only on the stabilized laser frequency  $f_\nu$  and predefined system parameters. As a result, the output frequency inherited the stability and accuracy of the frequency-stabilized laser. Since  $f_{\text{out}}$  could deviate slightly from the target frequency of exactly 10 MHz, a third DDS system was employed to adjust the frequency by a factor of  $K$  [as shown in Eq. (6)]. This adjustment produced a highly stable microwave RF signal at 10 MHz (or 5 MHz if desired), making it suitable for conventional practical applications.



**Fig. 3** (a) Frequency stability of the 1567-nm optical reference (red diamonds) and the 10-MHz microwave reference (black triangles and circles) stabilized via CO molecular transitions, in comparison with a commercial rubidium clock (blue squares). The dashed line marks the noise floor of the optical reference at 130 Hz/√Hz. (b) Power spectral density of the 10-MHz signals: blue line for the rubidium clock, red line for the free-running OCXO, and black line for the output of the OCXO-based CO clock.

$$f_{b+} = f_{\nu} - f_0 - Nf_r, \quad (1)$$

$$f_{b-} = f_0 + (N + 1)f_r - f_{\nu}, \quad (2)$$

$$f_+ = \frac{M}{N}(f_{b+} + f_0) = \frac{M}{N}f_{\nu} - Mf_r, \quad (3)$$

$$f_- = \frac{M}{N+1}(f_{b-} - f_0) = Mf_r - \frac{M}{N+1}f_{\nu}, \quad (4)$$

$$f_{\text{out}} = f_+ + f_- = \left(\frac{M}{N} - \frac{M}{N+1}\right)f_{\nu}, \quad (5)$$

$$f_{\text{RF}} = Kf_{\text{out}}. \quad (6)$$

Using this approach, we utilize a frequency-stabilized laser locked to a CO molecular absorption line in conjunction with an OFC to generate a highly stable and accurate 10-MHz microwave reference. This system enables the precise conversion of the stabilized optical signal into a robust microwave reference, making it suitable for a wide range of practical applications, such as advanced measurement systems and timing synchronization. By harnessing the inherent stability of the molecular clock, this method ensures that the generated microwave reference preserves the exceptional precision of the optical frequency standard, providing a reliable and versatile solution for applications requiring high-frequency accuracy.

### 3 Results and Discussion

We evaluated the frequency stability of the 10-MHz microwave reference output from the molecular clock using a high-precision frequency comparator (model VCH-314) integrated within the hydrogen maser. The active hydrogen maser was employed as the reference standard, exhibiting short-term frequency stability on the  $10^{-14}$  level and long-term stability approaching  $10^{-16}$ . As illustrated in Fig. 3(a), the 10-MHz microwave reference (denoted as CO-10 MHz) was characterized via this comparator. Its fractional frequency stability achieves  $2.5 \times 10^{-13}$  at an integration time of 100 s and  $7 \times 10^{-14}$  at 500 s. In comparison, the original optical frequency of the molecular clock achieves a relative stability of  $7 \times 10^{-14}$  at 100 s. Such results reveal degraded frequency stability after down-converting the molecular optical frequency to the 10-MHz microwave frequency. One plausible reason lies in the excess noise introduced by electronic circuitry during the optical-to-RF conversion. Although RF bandpass filters and low-noise amplifiers are adopted for RF signal conditioning and amplification, these discrete electronic components unavoidably add extra noise to the 10-MHz output signal.

The OFC in optical-RF-conversion was referenced to an OCXO, which features ultra-low phase noise and outstanding short-term frequency stability. Therefore, we phase-lock this OCXO to the 10-MHz output of the CO molecular clock, generating an optimized microwave reference signal denoted as CO-OCXO. As shown in Fig. 3(b), the OCXO exhibits superior intrinsic phase noise performance compared with conventional rubidium clocks. A self-developed phase-locked loop (PLL) is adopted to realize precise locking between the OCXO and the 10-MHz molecular clock reference. Taking full advantage of the OCXO's ultra-low inherent phase noise, the proposed scheme effectively suppresses spurious noise originating from discrete RF circuits.

As illustrated in Fig. 3(b), the phase noise results are characterized using a phase noise analyzer (R&S FSWP-26) and show that the OCXO delivers excellent low-frequency phase noise within the frequency range of 1 to 10 Hz, contributing to improved short-term frequency stability of the system. Nevertheless, its phase noise performance still deteriorates significantly in the frequency range from 100 Hz to 2 kHz, with a prominent noise peak observed at  $\sim 300$  Hz. This artifact is attributed to the 330-Hz wavelength modulation applied to the laser of the optical molecular clock. Since the bandwidth of the PID feedback circuit extends to the kilohertz regime, the 330-Hz modulation component cannot be sufficiently filtered, thereby introducing considerable additional phase noise. Eventually, the 10-MHz reference achieves a fractional frequency stability of  $7 \times 10^{-14}$  at an integration time of 2000 s, almost equivalent to the stability level of the original optical clock output.

These results confirm that the molecular clock exhibits excellent long-term frequency stability in both its optical and microwave outputs. Since the OFC is locked to the OCXO-derived microwave output, the entire frequency conversion chain is ultimately traceable to the molecular transition. Nevertheless, cumulative phase noise generated during optical-to-RF conversion significantly degrades the short-term stability of the microwave reference. Such performance deterioration can be effectively mitigated by utilizing low-phase-noise RF components and high-stability OCXOs with narrow-bandwidth PLLs, enabling the microwave output to closely approach the intrinsic stability limit of the optical clock signal.

### 4 Conclusion

In this study, we demonstrated a dual-output molecular clock system based on the carbon monoxide (CO) R(9) transition at 1567 nm, achieving both optical and microwave frequency references with remarkable stability. Using noise-immune cavity-enhanced optical heterodyne molecular spectroscopy and optical frequency combs, we stabilized the optical frequency and converted it to a microwave reference. Notably, this system, including the OFC, can operate autonomously without requiring an external precision reference, relying solely on the molecular transition. The clock achieved optical and microwave stability of  $7 \times 10^{-14}$ , underscoring its potential for high-precision applications. To optimize clock performance, employing molecular transitions with stronger intensity and narrower spectral linewidth will substantially improve frequency stability. Furthermore, in the frequency conversion architecture, the combination of low-phase-noise RF components and crystal oscillators could improve the frequency stability of the microwave reference to be comparable to that of the optical reference.

This molecular clock offers significant advantages over traditional atomic clocks, including portability, robustness, and reduced susceptibility to environmental factors such as magnetic fields and pressure variations. Its compact design and dual-output capability make it a promising candidate for applications in metrology, satellite navigation, and global time synchronization. The ability to generate both optical and microwave frequency references within a single, self-contained system could revolutionize timekeeping technology, delivering remarkable versatility and stability.

Beyond its practical utility, the dual-frequency output of the molecular clock holds profound implications for fundamental science. This system enables precision measurements requiring both optical and microwave references and could play a pivotal

role in advancing our understanding of fundamental physics. Potential applications include measuring fundamental constants, testing relativistic effects, and exploring physics beyond the standard model. Its high stability and simplified operation, free from complex auxiliary systems, position it as a transformative tool for both research and real-world applications.

Looking ahead, advancements in molecular clock technology—such as enhanced frequency stability, further miniaturization, and integration into field-deployable systems—could unlock new frontiers in timekeeping and precision measurement. These developments would enable experiments in space-time physics and contribute to the evolution of measurement techniques across diverse scientific disciplines.

## Disclosures

The authors declare no conflicts of interest.

## Acknowledgments

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