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Tunable two-mode Cr^{2+}:ZnSe laser with a frequency-noise spectral density of 0.03 Hz $Hz^{-1/2}$

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Abstract. **An optically pumped cw laser on a Cr^{2+}:ZnSe crystal with a tunable (in the range of 2.3 – 2.6 μm) wavelength, operating with generation of two axial modes, has been developed. It is shown that the minimum laser frequency-noise spectral density does not exceed 0.03 Hz $Hz^{-1/2}$. Application of this laser in problems of Doppler and Doppler-free spectroscopy makes it possible to detect spectral absorption lines of gases with sensitivities of 5×10^{-12} and 2×10^{-10} cm^{-1}, respectively (averaging time $\tau = 1$ s). Having stabilised this laser with respect to the Doppler-free resonances of saturated dispersion of methane molecule, one can obtain a short-term frequency stability of 10^{-15} – 10^{-16} ($\tau = 1$ s).**

Keywords: *Cr^{2+}:ZnSe laser, IR lasers, tunable lasers, solid-state lasers, two-mode lasers, optical frequency standards, frequency noise, laser spectroscopy.*

1. Introduction

Many methods for selecting weak spectral lines with Doppler and sub-Doppler resolution, based on the use of cw single-frequency lasers, have been developed to implement highly sensitive optical spectroscopy and optical frequency standards [1, 2]. In the Doppler-free saturation spectroscopy narrow resonances with a homogeneous width γ are recorded in the laser radiation intensity, frequency, or phase spectra upon interaction of counterpropagating laser waves with absorption lines of low-pressure gases. Among the methods of saturation spectroscopy, NICE-OHMS (noise-immune cavity-enhanced optical heterodyne molecular spectroscopy) has the highest sensitivity to detection of weak absorption lines [3]. The sensitivity of this method is determined by the quantum limit (Poisson noise of the number of photons), which is obtained using the frequency-modulation method [4], with a cell containing absorbing gas placed in a high-Q Fabry – Perot interferometer.

The frequency-modulation method effectively eliminates the influence of technical fluctuations of the laser radiation frequency and intensity, and the cavity increases the effective radiation – molecule interaction length, thus increasing the amplitude of narrow spectral resonances by a factor of $2\mathcal{F}/\pi$, where $\mathcal{F}$ is the Fabry – Perot interferometer finesse. When observing Doppler-free resonances at ultraweak transitions, where the intracavity field does not cause supersaturation of lines, one can use ultra-high-Q Fabry – Perot interferometers ($\mathcal{F} > 10^5$). In particular, using this instrument, Ma et al. [3] detected absorption on the order of 10^{-12} – 10^{-13}, with an averaging time $\tau = 1$ s, on the overtones of C_2HD molecule [3].

The two-mode laser spectroscopy [5], which is used in this study, has a sensitivity to weak absorption lines that is comparable with the NICE-OHMS sensitivity; it is applied to problems in which ultra-high-Q Fabry – Perot interferometers cannot be used and the optimal finesse is 10^2 – 10^3. This method is successfully applied to select narrow Doppler-free resonances when implementing optical frequency standards based on cw gas lasers with an internal absorption cell, in particular, in design of femtosecond optical clock [6 – 9].

The two-mode method has two modifications: detection of saturated-absorption (SA) resonances from the change in the mode intensities ['amplitude resonances' (ARs)] and detection of saturated-dispersion (SD) resonances from the change in the intermode frequency ['frequency resonances' (FRs)]. The FR method is more promising [10]. Its advantage is in the high detection sensitivity, which is determined by the natural frequency noise of radiation (Schawlow – Townes limit), the linearity of the response to the absorption introduced into the cavity (weak influence of mode competition), and less strict requirements to the output laser power (the possibility of designing compact devices without loss in sensitivity). In fact, when applied to some problems, the two-mode method allows one to reach the same sensitivity as the NICE-OHMS method but with a much simpler spectroscope scheme.

In the 1990s the two-mode method was extended to cw tunable dye lasers (for Doppler spectroscopy problems) [11] and lasers on colour centres in RbCl:Li crystals (for Doppler-free spectroscopy problems) [12]. In particular, a two-mode spectroscope for the range of 2.7 – 3.3 μm was designed and SD resonances were obtained on a number of lines of the ν_3 methane band in [13]. However, no systems convenient for practical applications have been developed because of the difficulties related to the low lifetime of RbCl:Li crystals and their need for cryogenic cooling, as well as the absence of appropriate pump lasers. To date, the situation with solid-tate lasers in the spectral range we are interested in (2 – 3.5 μm) has radically changed due to the progress in the growth technology of high-quality A_2B_6 laser crystals, doped

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with divalent transition metal ions [14, 15]. For example, Cr^{2+}:ZnSe and Cr^{2+}:CdSe crystals provide lasing in the ranges of 1.88–3.1 μm [16] and 2.26–3.61 μm [17], respectively; they can be used at room temperature and have a wide absorption band near λ = 1.9 μm, which allows one to pump them by commercial thulium-doped fibre lasers or directly by laser diodes.

These lasers were shown to be promising for high-resolution spectroscopy. For example, a cw tunable Cr^{2+}:ZnSe laser operated in the single-frequency lasing mode with an output power of 150 mW and a linewidth of 120 MHz [18]. In the cw Cr^{2+}:CdSe laser smooth tuning of the lasing wavelength was implemented in the range of 2.35–3.45 μm; in this case, the output laser power in the maximum of the tuning curve exceeded 200 mW. It was shown that this laser effectively operates at a wavelength of 3.28 μm, which is interesting for developing an optical frequency standard on the methane molecule [19]. Single-frequency lasing with a linewidth not larger than 60 MHz was obtained in this spectral range [20].

In this study we developed a cw laser on a Cr^{2+}:ZnSe crystal to find out the potential possibilities of applying two-mode spectroscopy in lasers of this type. The wavelength of the laser developed can be gradually tuned in the vicinity of 2.4 μm, where methane lines promising for selecting Doppler-free resonances in small absorbing cells are located. Stable cw lasing in two neighbouring axial modes was implemented and the limiting sensitivity to detection of absorption lines was estimated by measuring the spectral density of intermode frequency fluctuations for different output powers in the frequency range under consideration: 2–12 kHz.

2. Experimental technique

The optical scheme of the experimental setup is shown in Fig. 1. The laser active element (AE) was made of a Cr^{2+}:ZnSe single crystal grown on a single-crystal seed by the physical vapour transport method with simultaneous doping during growth, using the technology developed for growing A_2B_6 single crystals [14, 15]. We used a crystal with a Cr^{2+} concentration of 1.1×10^{19} cm^{-3}. The AE had a thickness of 2.2 mm and transverse sizes of 1.5 × 8 mm. To provide effective heatsink, the AE was clamped (with indium interlayers) between two copper plates, which were cooled by continuous-flow water with a temperature of 17 °C. The AE (oriented at the Brewster angle) was placed in the middle of a four-mirror cavity, assembled with astigmatism compensation [21]. The plane mirror M1 and intermediate spherical mirrors M2 and M3 had reflectances close to 100% in the spectral range of 2.35–2.65 μm. The transmittance of the output mirror M4 was 2%. The total cavity length was 1 m. To tune the lasing spectrum and perform its preliminary narrowing, we applied an interference-polarisation (Lyot) filter, made of a crystalline quartz plate 1.1 mm thick, with an optical axis oriented parallel to the plate plane. The Fabry–Perot intracavity etalon was a plane-parallel CaF_2 plate 8 mm thick; it provided stable two-mode operation of the Cr^{2+}:ZnSe laser. The general loss in the cavity did not exceed 10%.

Optical pumping of the Cr^{2+}:ZnSe laser was performed by unpolarised radiation of a cw commercial thulium-doped fibre laser with a wavelength of 1.94 μm and a maximum output power of 2.8 W. The pump beam passed by the cavity spherical mirror M2 and was focused in the AE by the spherical mirror into a spot ~0.1 mm in diameter. The pump beam

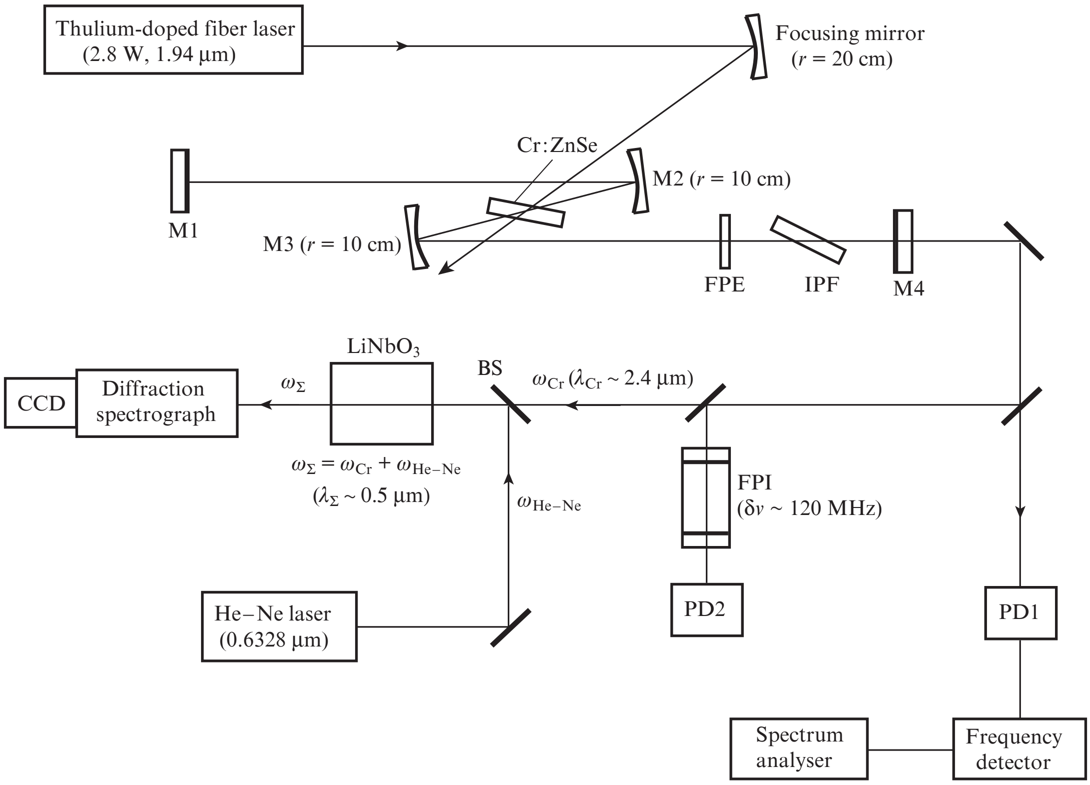

Figure 1. Schematic of the experimental setup: (FPE) a Fabry–Perot etalon, (IPF) an interference-polarisation filter, (FPI) a Fabry–Perot interferometer, and (BS) a beam splitter.

propagation direction made a small angle (~3.5°) with the optical axis of the Cr^{2+}:ZnSe laser cavity. In this way we excluded the influence of the pump radiation reflected from the elements of the Cr^{2+}:ZnSe laser cavity on the operation of the thulium-doped fibre laser. In this geometry, due to the small angle between the pump and laser beams (~1.5°), the pump loss in the crystal, caused by the mismatch between the regions of pump and laser modes, were insignificant. The main pump loss was related to the reflection of vertically polarised radiation from the AE surface and the incomplete absorption of the pump radiation by the crystal. Under our experimental conditions the maximum power absorbed in the AE did not exceed 1.6 W at a maximum power of the incident radiation of 2.8 W.

The Cr^{2+}:ZnSe laser radiation spectrum was preliminarily analysed, and the lasing wavelength was measured using a diffraction spectrograph with a resolution of 0.04 cm^{-1}. To this end, the IR radiation of Cr^{2+}:ZnSe laser (with a frequency ω_{Cr}) was transformed into visible spectral range ($\lambda_\Sigma \sim$ 0.5 μm) during generation of the sum frequency ($\omega_\Sigma = \omega_{Cr} + \omega_{He-Ne}$) upon its mixing in a nonlinear $LiNbO_3$ crystal (90° synchronism with temperature tuning) with monochromatic He–Ne laser radiation having a wavelength of 0.6328 μm (frequency ω_{He-Ne}) and a power of 5 mW. The radiation transformed into visible light was recorded by the spectrograph using a multichannel optical analyser based on a CCD line array. This detection technique allowed us to perform online monitoring of the IR laser radiation spectrum. The fine structure of the laser spectrum was analysed using a scanning Fabry–Perot interferometer with a free spectral range of ~2.3 GHz and a resolution $\delta\nu \sim$ 120 MHz.

The Cr^{2+}:ZnSe laser radiation was partially directed to a photodiode PD1 to detect the beat signal of two neighbouring axial modes. Then this signal was demodulated by a frequency detector (FD), and its noise characteristics were studied by an FFT spectrum analyser.

3. Experimental results

Using a Lyot filter, one can tune the Cr^{2+}:ZnSe laser at any wavelength in the range of 2.3–2.6 μm (the tuning range was limited mainly by the mirrors used). In the maximum of the tuning curve (λ = 2.45 μm) the output power P_{out} was 74 mW at an absorbed pump power of 1 W (the threshold pump power was 0.195 W). At a wavelength of 2.36 μm (near the methane absorption line that shows good prospects), we had P_{out} = 61 mW at an absorbed power of 1 W and a threshold power of 0.212 W.

Since the standing waves of neighbouring axial laser modes are shifted by $\lambda/4$ with respect to each other at the cavity centre, one can place the AE in this region to reduce to minimum the interaction between the neighbouring-mode fields. An analysis of the beat spectrum with the photodiode PD1 (the photodiode bandwidth is ~500 MHz) confirmed stable lasing in two neighbouring axial modes separated by $\omega_{12} = c/(2L) \sim$ 150 MHz (L is the cavity length).

In contrast to the single-frequency lasing, the two-mode regime makes it possible to estimate the spectral density of laser frequency fluctuations from the intermode frequency fluctuations $\delta\omega_{12}(f)$ (f is the Fourier frequency) without an external optical frequency discriminator. The $\delta\omega_{12}(f)$ values were measured in the low-frequency spectral range (f < 12 kHz), which is most important for frequency standards.

Along with the intermode frequency fluctuations $\delta\omega_{12}(f)$, the FD also contributes to the observed noise. Particular attention was paid to reduction of its noise in the frequency range under consideration (f < 12 kHz). We performed experiments with several analog FDs having an ultimate intrinsic noise of 0.03–0.05 Hz $Hz^{-1/2}$ and a slope of 0.2–0.1 mV Hz^{-1}. The best results were obtained with a FD designed according to the scheme of phase frequency self-tuning with a tunable generator based on an *LC* circuit. The phase noise of this generator determined the sensitivity to intermode frequency fluctuations. The FD noise was calibrated by a signal from a reference generator. It is planned to reduce the FD noise even more using digital demodulation schemes with a quartz generator as a reference.

The measurement results are shown in Figs 2 and 3. The curves in Fig. 2 were obtained at similar laser pump powers but using FDs with different characteristics. The minimum noise, obtained at P_{out} = 65 mW, was 0.03 Hz $Hz^{-1/2}$ (FD2, f = 10–12 kHz). The dashed line is the calculated lower limit of radiation frequency noise, which is determined by the natural noise of the generator, $\delta\omega_{nat}$ (or Schawlow–Townes noise). The estimation was based on the formula [22]

$$\delta\omega_{nat} = \Delta\omega_{res}\sqrt{\frac{2h\omega}{P_{out}}},$$

where $\Delta\omega_{res}$ is the cavity linewidth and ω is the lasing frequency. For the laser under study, $\Delta\omega_{res} \sim$ 5 MHz (the cavity loss is $T \sim 10\%$), ω = 125 THz, and $P_{out} \sim$ 65 mW; thus, $\delta\omega_{nat} \sim$ 0.008 Hz $Hz^{-1/2}$.

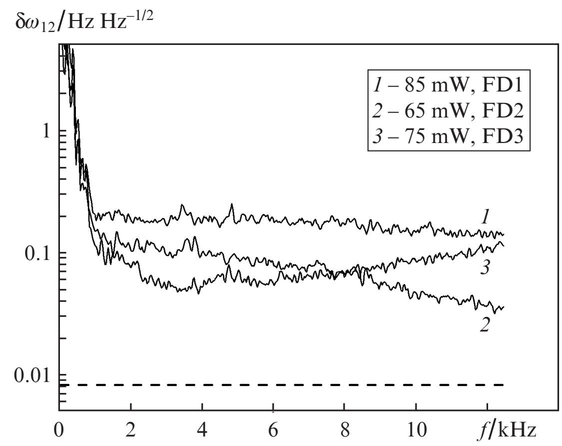

Figure 2. Spectral densities of intermode frequency fluctuations $\delta\omega_{12}$, obtained at similar laser powers using different FDs. The dashed line shows the calculated level of Schawlow–Townes noise.

Figure 3 shows the laser noise spectrum, recorded using FD3, and the intrinsic noise spectrum of FD3. A comparison of curves (*1*) and (*2*) shows that the intrinsic laser noise in the frequency range of 2–12 kHz [the range of low-frequency technical noise (f < 2 kHz) is excluded from consideration] barely exceeds the noise level of the FD used. Note that the spectral density of natural frequency noise of radiation of the monolithic-cavity Nd:YAG laser (which is most developed for precise measurements), determined at the analysed frequencies f > 100 kHz, was ~0.2 Hz $Hz^{-1/2}$ [23] at P_{out} = 40 mW. The measurements in [23] were performed using a

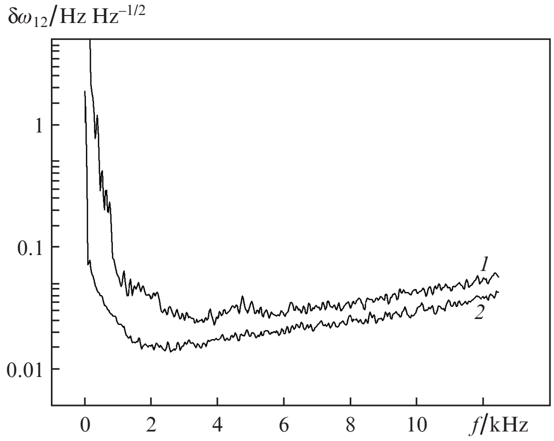

Figure 3. (*1*) Spectral density of intermode frequency fluctuations $\delta\omega_{12}$, obtained with FD3, and (*2*) the spectral density of FD3 intrinsic noise.

single-frequency laser, stabilised with respect to the Fabry–Perot interferometer.

4. Discussion

The frequency-noise spectral density obtained by us makes it possible to estimate the limiting sensitivity of the detector of weak absorption lines based on the laser developed.

For linear absorption on Doppler-broadened lines the change in the beat frequency $\Delta\omega_{12}$ is related to the introduced differential absorption coefficient $\varkappa$ by the expression [11]

$$\Delta\omega_{12} = \frac{c\varkappa l}{2\pi L}|F_1 - F_2|, \qquad (1)$$

where l is the absorbing-cell length and F_i is the dispersion shape of Doppler line, dependent on the mode frequency ω_i. When the condition $\omega_{12} \sim \Delta\omega_{\rm D}$ is satisfied ($\Delta\omega_{\rm D}$ is the Doppler line width), one can assume that the form factor $|F_1 - F_2| \sim 1$.

Relation (1) can be used to estimate the detection limit of the differential absorption coefficient $\varkappa_{\rm D}^{\rm min}$ for Doppler-broadened absorption lines in the case under consideration. On the assumption that $\Delta\omega_{12} = \delta\omega_{12}\sqrt{\Delta f}$ (Δf is the bandwidth of recorded frequencies), a case corresponding to the signal-to-noise ratio equal to unity, we obtain

$$\varkappa_{\rm D}^{\rm min} = \frac{2\pi L}{cl}\delta\omega_{12}\sqrt{\Delta f} = \frac{2\pi L}{cl}\frac{\delta\omega_{12}}{\sqrt{2\pi\tau}},$$

where $\tau = 1/(2\pi\Delta f)$ is the averaging time. The $\varkappa_{\rm D}^{\rm min}$ value for the experimentally obtained noise was estimated to be $\sim 5 \times 10^{-12}$ cm^{-1} at an averaging time $\tau = 1$ s and the length ratio $l/L = 1/2$.

To estimate the detection limit for the absorption coefficient in the case of SD Doppler-free resonances, where the frequency difference $\omega_{12} \gg \gamma$ (γ is the absorption-line homogeneous width), (1) is replaced by the relation [24]

$$\Delta\omega_{12} = \frac{c\varkappa l}{2\pi L}R(I_{\rm sat})D_1.$$

Here, D_1 is the dispersion function describing an SD resonance with a homogeneous width γ in dependence of the mode frequency ω_1 ($|D_1| = 0.5$) and $R(I_{\rm sat})$ is the function describing the dependence of the SD resonance magnitude on the parameter of absorbing-medium saturation ($I_{\rm sat}$) by the cavity field. The saturation parameter optimal for obtaining a maximum resonance signal is $I_{\rm sat} = I_0 = 4.83$, a value at which the derivative of the SD resonance with respect to frequency at the line centre, $S_0 = {\rm d}\omega_{12}/{\rm d}\omega_1 \approx \Delta\omega_{12}^{\rm max}/\gamma$ (the 'self-stabilisation coefficient', according to [1]), is maximum. At this $I_{\rm sat}$ value the peak-to-peak value of the SD resonance is

$$\Delta\omega_{12}^{\rm max} = \frac{c\varkappa l}{4\pi L}\frac{\sqrt{1+I_0}-1}{I_0\sqrt{1+I_0}} \approx 0.03\varkappa l\frac{c}{2L}.$$

When passing to Doppler-free spectroscopy, the detection limit for the differential absorption coefficient can be written as

$$\varkappa_{\rm SD}^{\rm min} = \frac{10^2 L}{cl}\frac{\delta\omega_{12}}{\sqrt{2\pi\tau}}.$$

For the laser under consideration, at $l/L = 1/2$ and averaging time $\tau = 1$ s, we have $\varkappa_{\rm SD}^{\rm min} \sim 2 \times 10^{-10}$ cm^{-1}.

This laser is planned to be used to design frequency standards and select SD resonances on lines of combination vibrational–rotational bands of methane in the wavelength range of 2.33 – 2.5 μm. Note that the methane pressure in the cell should be sufficiently low (no more than 1 mTorr) to retain the introduced linear absorption at a level of no more than 1%–2% and exclude reduction of the laser cavity Q factor.

The geometric parameters of the laser cavity and, primarily, the caustic transverse size in the cell (which determines the transit width) allow one to obtain resonances with a homogeneous width $\gamma \sim 100$ kHz at the same (or larger) resonance peak-to-peak amplitude $\Delta\omega_{12}^{\rm res}$ as on the methane line F^2 with $\lambda = 3.39$ μm (which is used in combination with the He–Ne laser radiation), where it amounts to $\sim$50 kHz. Estimations have shown that the derivative $S_0 \approx \Delta\omega_{12}^{\rm res}/\gamma$ at the centre of SD resonance at some methane lines with $\lambda = 2.3–2.5$ μm can be $\sim$1. Hence, the ultimate short-term stability σ_y (relative Allan deviation [25]), attainable in the Cr^{2+}:ZnSe/CH_4 standard under development ($\omega \approx 125$ THz) at an averaging time $\tau = 1$, can be estimated as

$$\sigma_y = \frac{\delta\omega_{12}^{\rm nat}}{S_0\omega\sqrt{2\tau}} \sim 2 \times 10^{-16}.$$

This value corresponds to the best results obtained in this field.

To increase the sensitivity to weak lines and attain the limiting short-term frequency stability using the laser developed, one needs an FD with reduced intrinsic noise. As far as we know, such devices are not produced commercially. As for the development of this frequency demodulator, we should note that a digital FD is characterised by a lower noise than the analog version we used. The main factors determining its sensitivity are the additive photodetector noise and the digital discretisation noise [26]. The total effect of these factors was estimated from above (by the effective value of fluctuations of the input beat frequency) to be $\sim 10^{-3}$ Hz Hz$^{-1/2}$.

5. Conclusions

We developed a cw low-noise optically pumped two-mode laser on a Cr^{2+}:ZnSe crystal, with wavelength tuned in a range of 2.3–2.6 μm. The spectral density of intermode frequency fluctuations was measured to be 0.03 – 0.05 Hz $Hz^{-1/2}$ in the frequency band 2 – 12 kHz; it was determined by the sensitivity of the FD used. The use of this laser for Doppler and Doppler-free spectroscopies provides limiting sensitivities to detection of differential absorption coefficient of 5×10^{-12} and 2×10^{-10} cm^{-1}, respectively, at an averaging time τ = 1 s. The estimated limiting short-term frequency stability, which can be reached by stabilising the Cr^{2+} : ZnSe laser with respect to Doppler-free SD resonances of the methane molecule, gives grounds to expect a short-term frequency stability $\sigma_y = 10^{-15} - 10^{-16}$ (τ = 1 s) in the Cr^{2+} : ZnSe/CH_4 optical standard.

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