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## A HIGHLY SENSITIVE TECHNIQUE OF DETECTING SUPERNARROW SPECTRAL LINES BASED ON THE FREQUENCY RESONANCES OF A DOUBLE-MODE LASER

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**Abstract.** - The present work investigates a highly sensitive technique of obtaining narrow resonances of saturated dispersion detected by the mode beat signal of a double-mode laser with an internal absorption cell which allows to register weak absorption lines with the  $\sim 10^{-11} \text{ cm}^{-1} \text{ Hz}^{-1/2}$  sensitivity determined by the natural frequency fluctuations of the laser. The resonances of 2.2 kHz HWHM have been separated in the 80 cm cell with the light beam of the 30 mm in diameter in experiments on the He-Ne/CH<sub>4</sub> double-mode laser ( $\lambda = 3.39 \text{ m}\mu\text{m}$ ).

**Introduction.** - To detect narrow spectral reference transitions various techniques of sub-Doppler saturation spectroscopy have been widely used [1-3]. The narrow resonance in the registered laser intensity (inverted Lamb-dip) results from the saturation of the inhomogeneously broadened absorption line with the oppositely directed radiation waves of a one-mode laser (OML). To this end various experimental geometries from external or internal cells with an absorption gas to beam sources have been applied.

So far this technique has permitted to obtain a very high spectral resolution in the optical range and to separate Lamb-dips with the Q-factor of  $10^{10} - 10^{11}$  for the transitions of CH<sub>4</sub>, Ca, OsO<sub>4</sub> [4-7]. The detection of that high Q-spectral lines demands the use of atomic beams or molecular gases of low densities. The value of inverted Lamb-dip ( $a_{-}$ ) is known to be proportional to  $p_{-}^3 V_{-}$  (where  $p_{-}$  is the pressure of the absorption gas and  $V_{-}$  is the caustic volume in the absorption medium) and to decrease quickly with the drop of the  $p_{-}$ . As a result the value of  $a_{-}$  has a low contrast as compared to the 'background intensity' in the detector. This in turn enhances the difficulties of detecting a signal among the noise due to the background intensity (the detector shot noise, fluctuations in the laser intensity, etc.). A few modified techniques of registration were suggested to eliminate the influence of the background intensity on the sensitivity of detecting Lamb-dips. In particular registering may be performed with the help of a change in the polarization state of the probe wave in the cell ("polarization spectroscopy" [8]) or with a fluorescence signal from the upper state of the reference transition [1]. The latter is especially efficient for beam experiments on atoms when multiple fluorescence on the adjacent transition can be sometimes ensured. For molecular lines multiple fluorescence is hardly realizable because of the complex structure of molecular energy states.

The enlisted resonance detection techniques may be called 'amplitude' by analogy with radio-technique as they all imply registration of the intensity (or amplitude) of the light wave. However, it is known that the saturation of an inhomogeneously broadened line can be detected by the change of the phase-frequency characteristics of the

laser radiation as the frequency as well as the phase of the laser wave depends on the change of the refractive index in the absorption medium. With the inhomogeneously broadened dispersion line saturated by the oppositely directed waves refractive index in the centre of the absorption line undergoes resonance changes which in the external cell can be detected by a phase shift of the wave with a great accuracy by means of a frequency modulated method suggested in [9,10]. The saturation of the dispersion line in a laser with an internal cell is detected as a nonlinear dependence of the laser frequency on the tuning frequency of the 'empty' cavity (the effect of a nonlinear frequency pulling to the line centre [11, 12]).

In terms of Lamb's theory the frequency of a OML with an internal absorption cell at  $|\omega_1 - \omega_-| \sim \gamma_-$  can be written as

$$\omega_1 = \Omega_1 + \sigma_+ + \beta_+ E_1^2 - \sigma_- - \frac{\alpha_- c l_-}{l_0} \cdot \frac{d_{12}^2 E_1^2}{\hbar^2 \gamma_-^2} \frac{\gamma_- (\omega_1 - \omega_-)}{\gamma_-^2 + (\omega_1 - \omega_-)^2} \quad (1)$$

where  $\Omega_1$  is the empty cavity resonance frequency,  $E_1^2$  is the mode intensity,  $\omega_1$  is the emission frequency,  $\omega_-$  is the centre of the absorption line,  $\sigma_{\pm}$  are the addends describing the linear (non-saturated) dispersion lines of the active and absorbing media and the term  $\beta_+ E_1^2$  describes the saturation of the dispersion line of the active medium. The last term in Eq.(1) is a narrow resonance of a dispersion kind pointing to the nonlinear pulling effect of the laser frequency to the centre of the absorption line:  $\alpha_-$  is the nonsaturated absorption factor per unit length,  $c$  is the light velocity,  $l_-$  is the cell length,  $l_0$  is the full cavity length,  $\hbar$  is Plank's constant,  $\gamma_-$  is the homogeneous width of the absorption line and  $d_{12}$  is the matrix element of the dipole moment of the transition. The factor  $d_{12}^2 E_1^2 / \hbar^2 \gamma_-^2 \equiv I_1$  is the parameter of the transition saturation.

For convenience we shall shorten the resonance dependence in Eq.(1) to the 'frequency resonance' (FR) and the inverted Lamb dip in the radiation intensity to the 'amplitude resonance' (AR).

The FR in a OML with an internal absorption cell has been thoroughly investigated [13-15]. In [14, 15] it was used to stabilize frequency of a He-Ne/CH<sub>4</sub> OML. From Eq.(1) it follows that the value of the FR ( $\Delta\omega = \alpha_- c l_-^4 I_1 / l_0$ ) in contrast to the AR is linearly depends on  $I_1$  and in addition as it was shown in [14] the frequency stabilization of the FR reduces shifts of the stabilized frequency due to the stray amplitude modulation.

A frequency adjusted laser-heterodyne ( $\omega_{het}$ ) detecting FR by the beat signal at the frequency  $\omega_{het} = |\omega_1 - \omega_{het}|$  is needed to observe FR in a OML. In this case the noise level is determined by a relative stability of a beat frequency  $\omega_{het}$ . As the technical frequency fluctuations of the  $\omega_{het}$  are not correlated by those of the laser under investigation even at the absolutely perfect heterodyne stabilization the noise level be high as the achieved laser passive laser frequency stability cannot be more than  $10^3 \div 10^4 \text{ Hz} \cdot \text{Hz}^{-1/2}$ . This forbids an efficient application of FR in the OML.

2. This work investigates a more efficient technique of detecting FR when they are separated by a beat signal of a double-mode laser (DML) with an internal absorption cell [16, 17]. The physics of the mechanism of the FR in a DML is similar to that of a OML where the second mode is used as a 'heterodyne' to detect a beat signal which reduces technical frequency noise to several orders. This

method may be defined as 'double frequency intracavity spectroscopy'. From [11, 16] it can be seen that the frequency difference of the two modes in the laser with a nonlinear absorption at  $|\omega_1 - \omega_2| \sim \gamma$  is represented as

$$\omega_{12} \equiv \omega_1 - \omega_2 \approx (\Omega_1 - \Omega_2) + \Delta\sigma_+ + \Delta\beta_+ (E_1^2, E_2^2) + \frac{\alpha - c\ell_-}{\ell_0} \cdot I_1 \cdot \frac{\gamma - (\omega_1 - \omega_2)}{\gamma^2 + (\omega_1 - \omega_2)^2}, \quad (2)$$

where the second and third terms show the influence of the linear and nonlinear parts in the dispersion of the active medium and the factor of "2" is related to the second mode parameters. Eq.(2) does not take into account small non-resonance additions of the absorbing medium. If the frequency difference of the empty cavity satisfies the condition

$$\gamma \ll \Omega_1 - \Omega_2 \ll \gamma_+, kL_+ \quad (3)$$

(where  $\gamma_+$ ,  $kL_+$  - homogeneous and Doppler widths of the <sup>active</sup> medium) it is not difficult to prove that the influence of technical fluctuations due to the addends of the active medium may be neglected. In this case the frequency noises of mode beats are related only to the technical fluctuations of frequency difference in the empty cavity. As the technical fluctuations of  $\Omega_1$  and  $\Omega_2$  occur in a correlated way then their contribution to the noise is a few orders of magnitude lower than in a OML with an external heterodyne. As is shown in [17] the contribution of other noise sources (in particular additive noise of the photodetector) is small and natural frequency fluctuations in the mode frequency become a dominant noise factor. Estimates made for the He-Ne/CH<sub>4</sub> system (the value of the FR  $\Delta\omega \sim 10^5$  Hz at methane pressure  $p \sim 1$  Torr,  $I_1 \sim 1$ ) show that at the spectral density of natural laser frequency fluctuations  $(10^{-1} - 10^{-2})$  Hr.Hz<sup>-1/2</sup> the proposed technique allows to register the absorption coefficients for the weak lines with the sensitivity of  $\sim 10^{-11}$  cm<sup>-1</sup>.Hz<sup>-1/2</sup>.

**3. Experiment.** - An experimental investigation of narrow FR was made in a He-Ne laser with an internal methane cell. The experimental arrangement shown in Fig.1 consisted of three lasers: a DML with a telescopic light beam expander (the coustic diameter  $d_c$  of 30 mm, the cell length  $\ell_- = 0.8$  m, the cavity length  $\ell_0 = 1.8$  m), a reference DML ( $d_c = 3$  mm,  $\ell_- = 0.7$  m,  $\ell_0 = 1.6$  m) with a narrow radiation spectrum ( $\sim 10$  Hz) and a laser-heterodyne one-mode operated and 0.6 MHz shifted from the reference DML. To reduce the line-width of the lasers the frequency of the reference DML was stabilized along the non-resolved F<sub>2</sub>(2) methane component ( $p_- = 3$  m Torr,  $\gamma \sim 200$  kHz) by a broadband two loop AFC system. The AR of the reference DML worked as the discriminator of the 'slow' loop, while the 'fast' loop used the FR [17]. The frequency bandwidth of the combined system was about 20 kHz. The narrow radiation spectrum of the reference DML by means of the laser frequency coupling systems was transmitted via the laser-heterodyne to the DML with telescopic expander.

The stable two-mode operation in both He-Ne/CH<sub>4</sub> lasers was ensured by a Fabry-Perot cavity with two  $\lambda/4$  phase plates. The properties of such a cavity were investigated in [19,20]. [20,21] showed that when the angle  $\theta$  between the optical axes of the plates approximates zero or  $\pi/2$ , then at  $\lambda = 3.39$  mkm Ne transition a stable generation of two modes of the same transverse structure with the mutually orthogonal linear polarizations occurs. The dis-

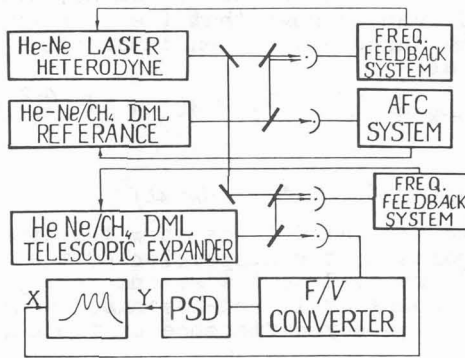


Fig.1. - The schematic view of the full experimental set-up.

tance between modes ( $\omega_{12}$ ) is adjusted by the angle  $\theta$  in the range 0.2 MHz and greater. This readily permits to fulfil the necessary requirement (3) for the investigated laser, as  $kL + \delta \gg 100$  MHz. The difference of this scheme from the one in [17] is that the active medium of the DML with telescopic beam expander was in the weak transverse magnetic field and the methane cell design incorporated liquid nitrogen cooling. The registration scheme of the FR and its derivatives in a DML is shown in Fig.2.

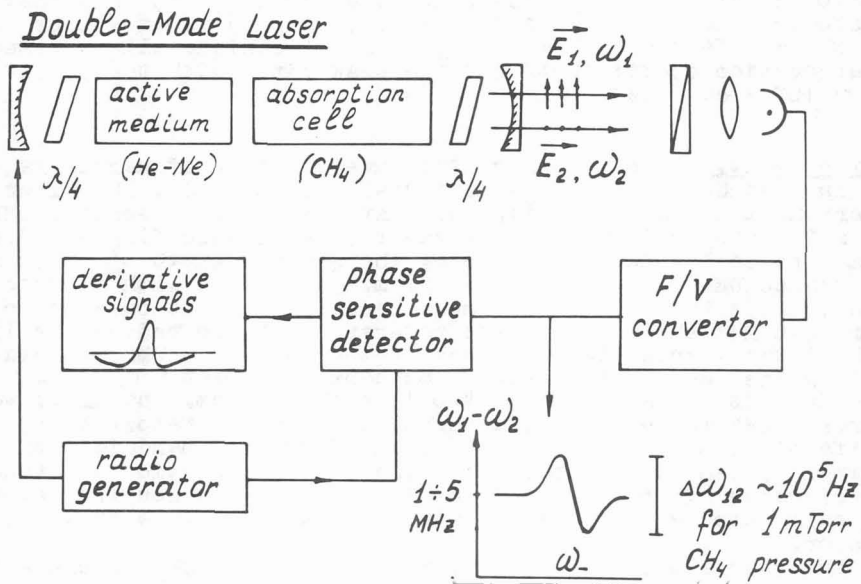


Fig.2. - The registration scheme of the frequency resonances and its derivatives in a double-mode laser. The double-mode operation of the laser may be possibly determined by the two phase plates inside the cavity (see the text).

Fig.3 gives the spectrograms of the first and second derivatives of the FR for the  $F_2^{(2)}$  methane line obtained at the gas pressure  $p = 60 \mu\text{Torr}$ ,  $T = 77^\circ\text{K}$ , the saturation parameter of the separated component of MHPS  $I_1 \sim 0.4$  and radiation frequency deviation 1500 Hz peak-to-peak (modulation frequency 400 Hz). With these parameters the HWHM of the detected resonances was minimum and about 2.2 kHz (the spectrograms was processed with the account of the doublet split of each component of the MHPS due to the recoil effect [3,4]). The spectrograms in Fig.4 point to the influence of the saturation parameter on the spectral component resolution.

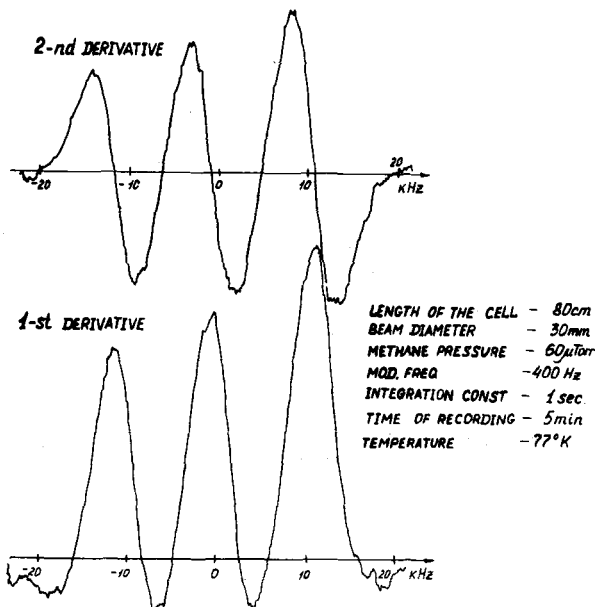


Fig.3. - The signals of the first and second derivatives of the frequency resonances in the double-mode He-Ne/ $\text{CH}_4$  laser. The MHPS of the  $F_2^{(2)}$  methane line is resolved.

It should be stressed that cells of considerably greater length and transverse dimensions are used to obtain AR signals in the OML with the similar resolution and signal-to-noise ratio. To ensure the low level of the saturated power the DML was operated with a considerable difference between intensities of the first and second modes. The intensity difference in the modes was provided by a transverse magnetic field  $H$  along the polarization of the second (heterodyne) mode. The calculation and experiments proved that owing to the Zeeman line split of the gain curve at  $\delta$  and  $\pi$  components the first mode saturating the centre of the absorption line is reduced while the second mode increases. The theoretical interpretation of the behaviour of the mode intensities in the magnetic field is given in Fig.5.

$$E_2^2 \gg E_1^2$$

The experiment qualitatively verified this dependence. Hence the relation for a stable laser double-mode generation can be obtained. At the same time the natural noise of the frequency and intensity increase negligibly (approximately two times for  $E_2^2/E_1^2 \sim 100$ ) which

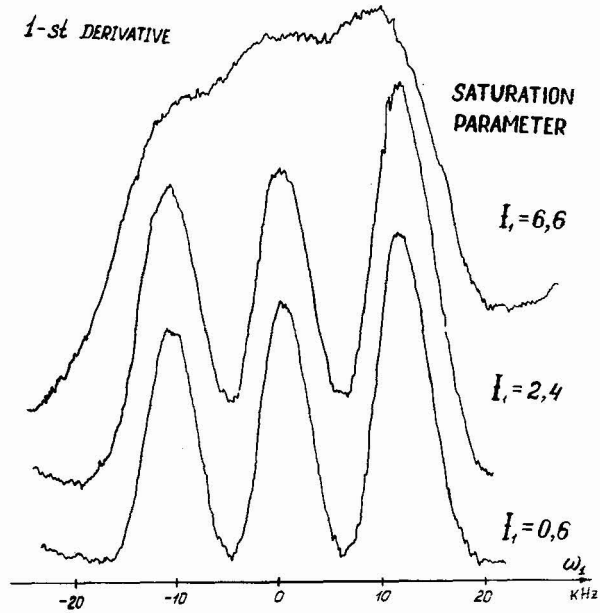


Fig.4. - The same signals as in Fig.3 under different saturation parameters of the first mode ( $I_1$ ) coincident with the absorption line centre.

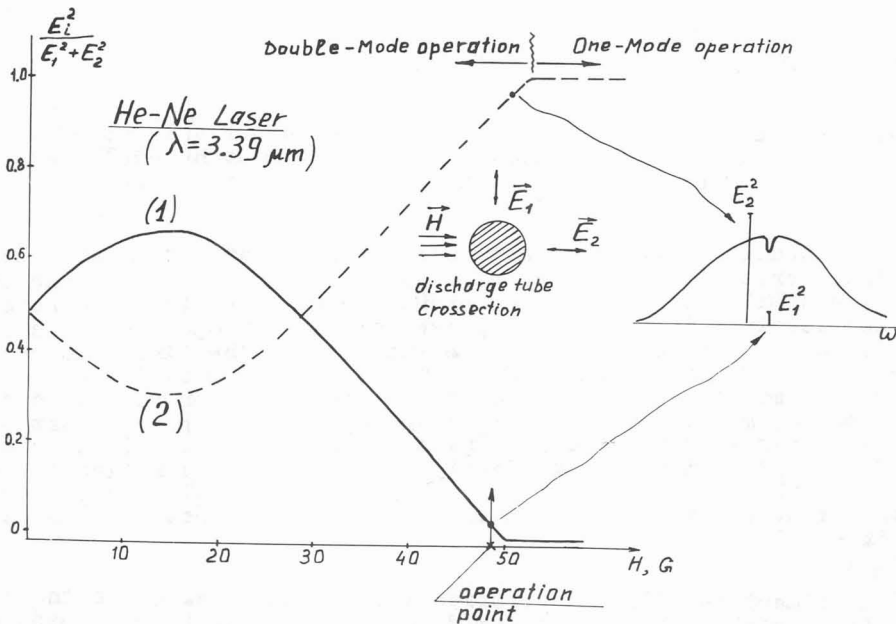


Fig.5. - The theoretical dependence of the mode intensities in the double-mode He-Ne laser ( $\lambda = 3,39 \mu\text{m}$ ) via transverse magnetic field. The direction of the magnetic field coincides with the polarization direction of the first (weak) mode saturating the methane line centre.

may be accounted by the deep saturation of the gain curve by a strong generation at the frequency close to that of the heterodyne mode ( $\omega_{12}$  was chosen in a range 1-5 MHz). In the case of a OML the decrease in the saturation parameter requires as a rule an application of large telescopic systems.

Conclusion. - The developed technique of the double frequency intracavity spectroscopy based on the frequency detection of the signal is shown to be highly sensitive and promising in laser spectroscopy and in particular for the detection of narrow reference lines. The application of frequency resonances of the double-mode laser as an optical frequency discriminator ensures small technical shifts, high short-term and long-term stabilities and an efficient reduction of the field broadening. The application of the above technique in the He-Ne/CH<sub>4</sub> laser provided optical reference lines of  $\sim 2.2$  kHz width when using the cell of 0.8 m length, the beam diameter of 30 mm and the time of 1 sec for the signal integration.

This technique is multi-purposed and might be extended to other types of lasers for providing supersensitive linear as well as non-linear laser spectroscopy.

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