Quantum Electronics **30** (3) 370 – 372 (2000)

PACS numbers: 42.62.Fi; 42.55.Mv
DOI: 10.1070/QE2000v030n04ABEH001708

Application of the three-mode operation of a broad-band laser in intracavity dispersion frequency-domain spectroscopy

I P Konovalov

Abstract. The beats of a three-mode dye laser, containing a component with a frequency close to the difference between neighbouring mode spacings, the technical fluctuations of which cancel out, are considered. The aim is to develop a laser spectroscopic method with a detection limit of the order of 10^{-10} – 10^{-11} cm^{-1} Hz$^{-1/2}$, determined by the natural frequency fluctuations and by the spectral resolution limited by the homogeneous width of the resonance line. Use was made of the unequal changes in two mode spacings of an equidistant three-mode spectrum, caused by the dispersion of the absorption line of an intracavity absorber, and of the detection of the resulting difference frequency.

The optical beat spectrum of a broad-band laser with a homogeneous gain line and slightly nonequidistant frequencies of three stably generated modes ($\omega_1 - \omega_2 \equiv \omega_{12} \neq \omega_{23} \equiv \omega_2 - \omega_3$) contains a difference frequency component $\Omega \equiv \omega_{12} - \omega_{23}$ [1]. The correlated technical fluctuations of the mode frequency spacings, associated in particular with the limited rigidity of the construction of the cavity, also give rise to the difference frequency $\delta\Omega = (\omega_{12} - \omega_{23})(\delta L/L)$, which vanishes completely in the equidistant ($\omega_{12} = \omega_{23}$) three-mode regime. Apart from the noise in the photoelectric lasing channel, uncorrelated natural frequency fluctuations remain. According to the estimates, their spectral density does not exceed $\sim 0.1 - 1$ Hz Hz$^{-1/2}$. The order of magnitude of the experimental noise in a three-mode dye laser is the same as the upper limit of the above estimate [1].

The physical characteristics of the competition and stability of the modes in the dye laser are of a kind general for lasers with an external cavity based on a whole series of active media with a homogeneous gain line which is ultrabroad compared with the reasonable spacing $\omega_{12} \sim 1$ GHz: cavities based on F centres, on transition metal ion-activated crystals, on semiconductors, etc. [2]. In a beat spectrum with a slight departure from the equidistant distribution of the three modes, a low-frequency low-noise component should appear. Since its appearance may be induced by various causes, there is a prospect of developing a frequency-detection laser metrology with detection limits physically bounded only by the natural fluctuations of the optical lasing frequencies. In the present study, this prospect is considered in relation to the development of an intracavity dispersion frequency-domain spectroscopy.

I P Konovalov P N Lebedev Physics Institute, Leninskii prospekt 53, 117924 Moscow, Russia

Received 15 June 1999
Kvantovaya Elektronika **30** (3) 370 – 372 (2000)
Translated by A K Grzybowski

The investigation was based on the linear repulsion of the modes of an optical cavity from the centre of the line of the resonant absorber introduced into it, caused by unsaturated dispersion and recorded in the radiofrequency intermode beats [3]. If the mode frequency exceeds the central frequency ω_0 of the absorption line, then the mode frequency increases after the introduction of an absorber. Otherwise it decreases. The change in the beat frequency of these two modes $\Delta\omega_{12}$ represents the analytical signal proportional to the resonant-particle concentration. Its independence of the power of the lasing field is due to the linearity of the effect employed. In the two-mode lasing regime, the maximum frequency photoresponse is attained for the symmetrical tuning of the modes relative to ω_0 and in the mode spacing equal to the Doppler width $2\Delta\nu_D$ of the line dispersion of the absorbing transition. This photoresponse has the sensitivity

$$\left(\frac{\Delta\omega_{12}}{\alpha}\right)_{\max} \approx \frac{cd}{L} = 3 \times 10^{10}\,\frac{d}{L}\ \text{Hz cm}\,, \qquad (1)$$

where α is the absorption coefficient; c is the velocity of light; L and d are the lengths of the cavity and of the internal absorbing cell. This estimate has been conformed experimentally in a jet rhodamine 6G laser with Na vapour within the cavity [3].

The weakest detectable absorption $\alpha_{\min}$ is determined by the minimum detuning $\Delta\omega_{12}$ which can be recorded experimentally. The recording of the beat frequency $\omega_{12} \approx 1$ GHz of a two-mode laser, used in the experiments of Ref. [3], with an uncertainty of ~ 1 kHz has been reported [4]. At the same time, a study was made [4] of the lasing regime for three equidistant modes in the same mode spacing $\omega_{12} = \omega_{23}$. Within the framework of the Lamb formalism, it was shown that in a dye laser this regime should be treated as the locking of three modes when the side-frequency beats are in phase with the central frequency (ω_1, ω_2 and ω_2, ω_3). The experimental instability of the frequency proved to be an order of magnitude smaller (~ 100 Hz) than in the two-mode regime. Evidently, for the symmetrical disposition of three equidistant modes on the absorption line and for $\omega_{12} = \omega_{23} = \Delta\nu_D$, the shift $\Delta\omega_{12} = \Delta\omega_{23}$ of the beats is smaller by a factor of two than in the case of two-mode beats. However, a greater stability ensures a lower detection limit $\alpha_{\min}$, i.e. the three-mode lasing regime is metrologically preferable to the two-mode regime. Nevertheless, in this case, the detection limits (~ 100 Hz) are several orders of magnitude greater than the normal fluctuations of the intermode beats, which bound physically the absolute detection limit ($\sim 10^{-10}$ – 10^{-11} cm^{-1} Hz$^{-1/2}$). Despite the fact that the technical fluctuations of the eigenfrequencies of the cavity are correlated (each mode is a virtually ideal optical heterodyne for the other), the fluctuations of the mode spacings remain: $\delta\omega_{nm}/\omega_{nm} = \delta L/L$.

If we resort to the frequency component Ω, it is likely that the situation may be radically improved. The frequency component should arise when the moduli of the lasing frequencies undergo different changes if the initially equidistant three-mode spectrum is tuned asymmetrically relative to ω_0. (To make the exposition specific, we may note that the frequency 2Ω also arises [2]. However, according to the nature of the problem, Ω is small and in a real experiment the apparatus averages out these components, so that for the sake of simplicity we take into account only the first harmonic Ω.) In the general case, the deviation of each of the generated frequencies is the sum of the linear and nonlinear dispersion effects [3]: $\Delta\omega_n = \Delta\omega_n^{(1)} + \Delta\omega_n^{(3)}$. In the limit corresponding to the inhomogeneous absorption-line broadening $\Delta\nu_{\mathrm{D}} \gg \gamma$, where γ is the homogeneous line half-width, the following expressions are valid:

$$\Delta\omega_n^{(1)} = A\,\frac{2(\omega_n-\omega_0)}{\Delta\nu_{\mathrm{D}}(\ln 2)^{1/2}}\exp\left[-\left(\frac{\omega_n-\omega_0}{\Delta\nu_{\mathrm{D}}(\ln 2)^{1/2}}\right)^2\right],$$
$$\Delta\omega_n^{(3)} = -A\sum_{m=1}^{3}\frac{2\gamma(\omega_n+\omega_m-2\omega_0)}{4\gamma^2+(\omega_n+\omega_m-2\omega_0)^2}\,I_m I_0^{-1}. \qquad (2)$$

Here, $A = \alpha c(d/L)$; I_0 is the absorption-line saturation intensity; I_m is the intensity of the mth mode. For $m = n$, the term $\Delta\omega_n^{(3)}$ in the sum describes the saturation of the mode dispersion at the frequency ω_n by its own field, whereas for $m \neq n$ it describes its cross-saturation by the fields of the other two modes. It is easily seen that, for the symmetrical tuning of the three equidistant frequencies relative to ω_0 ($\omega_2 = \omega_0$, $\omega_{12} = \omega_{23}$), the linear repulsion of the side modes, which is the same in magnitude but of opposite sign, does not lead to a departure from the equidistant distribution and to the appearance of the difference-frequency component $\Omega^{(1)} = [\Delta\omega_1^{(1)} - \Delta\omega_2^{(1)}] - [\Delta\omega_2^{(1)} - \Delta\omega_2^{(1)}]$ whatever the ratio $\omega_{12}/\Delta\nu_{\mathrm{D}}$. If $|\omega_2 - \omega_0| \sim \gamma$ under these conditions, then the nonlinear corrections to all the modes in terms of the approximation involving the equality of their intensities are the same and the contribution in the difference component is $\Omega^{(3)} = 0$.

In the other characteristic case involving tuning to the centre of the absorption line of one of the side modes, for example ω_1, its frequency does not undergo a linear dispersion shift: $\Delta\omega_1^{(1)} = 0$. The nonzero shifts of the other two modes are different. They do in fact lead to the appearance of a difference linear component $\Omega^{(1)} \neq 0$. If $\omega_{12}/\Delta\nu_{\mathrm{D}}$, then depending on the ratio $\omega_{12} = \omega_{23} \sim \Delta\nu_{\mathrm{D}}$, the maximum three-mode photoresponse $\Omega^{(1)}$ may be both somewhat smaller and somewhat greater than the two-mode photoresponse. Ignoring these spectrometrically insignificant differences, we assume that the sensitivity in terms of absorption is the same in both lasing regimes and replace $\Delta\omega_{12}$ in formula (1) by $\Omega^{(1)}$. For the reasonable ratio $d/L = 1/3$, the width of the low-frequency 'noise path' [1] then corresponds to a detection limit of $10^{-9}-10^{-10}$ cm Hz$^{-1/2}$ and its decrease to its quantum-fluctuation estimate makes it possible to expect a reduction of the detection limit at least by another order of magnitude. The spectral resolution (selectivity of the analysis) is then limited, as happens generally in linear spectroscopy, by the Doppler broadening of the absorption line. In the course of tuning over the range $|\omega_1 - \omega_0| \gtrsim \gamma$, we obtain from formula (2)

$$\Delta\omega_1^{(3)} = -A\,\frac{\gamma(\omega_1-\omega_0)}{\gamma^2+(\omega_1-\omega_0)^2}\,I_1 I_0^{-1}\,,$$
$$\Delta\omega_2^{(3)} = \Delta\omega_3^{(3)} = 0\,, \qquad (3)$$

i.e. the nonlinear resonance $\Omega^{(3)} \equiv \Delta\omega_1^{(3)}$ arises against the background of the linear photoresponse $\Omega^{(1)} \approx \mathrm{const}$. In terms of its parameters, the resonance is physically identical to the saturated-dispersion resonance in the beats of a two-mode laser [5] and ensures to the same extent the spectral resolution characteristic of nonlinear laser spectroscopy. It is limited by the known homogeneous (ultimately radiative) width of the absorbing transition. The dependence of the parameters of this resonance on intensity does not affect the sensitivity. The latter is determined by the linear pedestal $\Omega^{(1)}$, which is independent of the lasing characteristics.

Thus the prospect arises of the development of a new laser-spectroscopic method for a dispersion-induced departure from the equidistant distribution of the three generated modes and the detection of the low-noise, low beat frequency arising under these conditions. The principal condition for this is the experimental detection in Ref. [1] of the presence in the beat spectrum of three slightly nonequidistant laser modes with a broad homogeneous gain line of the component with a frequency equal to the difference between the mode frequency spacings. The technical fluctuations of the optical cavity length are compensated in this difference. There is no reason to doubt that there is a departure from an equidistant distribution of the resonance-dispersion modes of the intracavity absorber and there is little doubt that the sensitivity given by expression (1) can be reached. It is based on the calculation of the linear dispersion [formula (2)], for which the estimate (1) has been confirmed quantitatively in a two-mode experiment [3]. The observation of the nonlinear resonance [formula (3)], which is associated with the self-saturation of the dispersion of one of the modes and is well known not only in theory but has also been observed experimentally [5], is entirely realistic. However, overall we are dealing with the implementation of a physical mechanism of the conversion of the interaction of the optical field with matter into a signal with a sonic (infrasonic) frequency. An attractive feature is that such a radical conversion does not require any technical manipulations with laser radiation and is achieved automatically by virtue of the very physical nature of the laser. The possibility of employing the usual photodetection technique with a slow response, which cuts off the high-frequency components of the main beat spectrum, is also quite important.

The experimental implementation reduces mainly to a rational engineering solution of the problem of achieving the necessary asymmetric tuning and to the monitoring of the positions of the lasing frequencies against the absorption line profile. One of the realistic versions involves the preliminary tuning of the three-mode spectrum to frequency resonance of the saturated dispersion [formula (3)], which has been used successfully as a comparison reference in precision laser metrology [5]. We need not be troubled by a possible increase in the complexity of the apparatus when a negative feedback loop is introduced in it. For example, a scheme characterised by at least four stabilised interrelated parameters has been proposed [6] for the optical matching of an external measuring interferometer of ultrahigh quality (a finesse of 10^4) to the laser cavity and for the modulation of the radiation of the latter at a frequency of ~ 1 GHz. Its deviation had to be recorded with an error up to 0.01 Hz. According to the nature of the problem considered here, we are dealing only with an imprecise location of ω_1 within the $c/2L$ frequency spacing, which 'embraces' ω_0. This spacing is an order of magnitude smaller than the characteristic distance between the lasing frequencies $\omega_{12} = \omega_{23} \sim \Delta\nu_{\mathrm{D}}$ [1 – 4]. For the linear photoresponse $\Omega^{(1)}$, the instability of ω_1 within it is not critical.

As regards the possibilities for the precise optical stabilisation of the equidistant three-mode spectrum, it appears preferable to employ the absorption line with $\gamma \ll \Delta\nu_{\mathrm{D}} \ll \omega_{12} = \omega_{23}$ when the frequency ω_2 of the central mode is tuned to ω_0. Within the limits $|\omega_2 - \omega_0| \sim \gamma$, there is no linear $\Omega^{(1)}$ pedestal. The nonlinear resonance $\Omega^{(3)} \equiv 2|\Delta\omega_2^{(3)}|$, where $\Delta\omega_2^{(3)}$ is given by expression (3) when the subscripts in it are interchanged $(1 \leftrightarrow 2)$, should be generated directly against the low-frequency background. It will have an even symmetry in respect of $\omega_2 - \omega_0$, in contrast to the odd-symmetric resonance profile [formula (3)], whereas the low-frequency oscillations in the different wings of this 'butterfly' should be antiphase.

References

1. Konovalov I P Kvantovaya Elektron. (Moscow) **24** 833 (1997) [Quantum Electron. **27** 813 (1997)]
2. Konovalov I P Kvantovaya Elektron. (Moscow) **24** 325 (1997) [Quantum Electron. **27** 315 (1997)]
3. Konovalov I P Spectrochim. Acta B **48** 575 (1993)
4. Konovalov I P, Loginov F V Kvantovaya Elektron. (Moscow) **23** 238 (1996) [Quantum Electron. **26** 231 (1996)]
5. Tyurikov D A, Gubin M A, Shelkovnikov A S, Koval'chuk E V IEEE Trans. Instrum. Meas. **44** 166 (1995)
6. Mugino A, Yamamoto T, Omatsu T, Gubin M A, Morinaga A, Takeuchi N Opt. Rev. **3** 243 (1996)