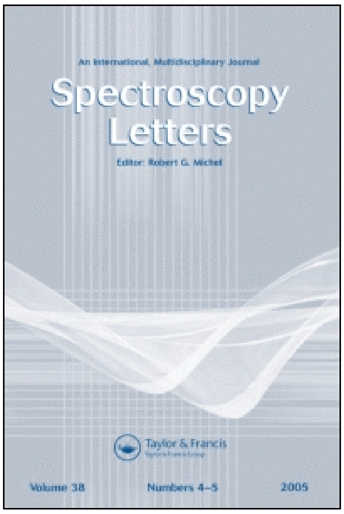

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METHOD OF SELECTIVE INTRACAVITY ABSORPTION USING DYE LASERS

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INTRODUCTION

The method of the selective intracavity absorption is based on the high sensitivity of the laser intensity on the absorption of the sample placed into the resonator. The method for Nd-glass laser with inhomogeneous broadening of the gain spectrum was grounded and experimentally checked by the authors of the papers[1,2]. Later demands for the spectral region expanding of the method have led to application of dye lasers for the intracavity measurements. The capability to broad-band emission in the different spectral regions makes of dye lasers the unique instruments for intracavity spectroscopy. A number of the successful experiments on the dye lasers applications to the sensitive detection of Na, Ba, Ho, Pr, I_2, Br_2 small concentrations was published until now[3-6]. At the same time one cannot consider the theoretical analysis of the intracavity absorption characteristics of the dye lasers, i.e. the lasers with homogeneous spectral broadening of active media as sufficiently complete and reliable. The theoretical estimation of the sensitivity of the method for CW laser operation was given in papers[7,8]. But analytical formulae for sensitivity in reference[7] are valid only for near-threshold le - vels of pumping power which does not often correspond to experimental conditions and leaves open the question about potentialities of the method at the higher pumping power (two or more times exceeding the threshold). The authors of the reference[8] consider the problem at any pumping levels, but their approach is based on the simplified model which seems to be doubtful and requires the special foundation. In particular they suppose the existence in the active media of the number of N_j particles, which interact only with one mode j and do not interact with other laser modes, and also of the number of N_0 molecules (so called reservoir) equally interacting with every mode. In the case of the homogeneous spectral broadening such dividing the molecules into types does not seem to be rightful. In addition in the both papers[7,8] the gain and loss of all modes (except one) are assumed to be of the same values so the total number of modes is given arbitrary and not connected with resonator, active media and pumping parameters. Because of this reason one cannot calculate correctly the value of the method sensitivity or analyze it's dependence on the parameters.

In this paper the attempt is made to describe analytically the characteristics of the intracavity absorption in dye lasers under arbitrary pumping power level for steady-state laser operation taking into account the spectral dependence of the gain.

INITIAL EQUATIONS AND ANALYTICAL FORMULAE

The approach in use is based on the rate equations method. In the case of length dependent gain and loss the condition of steady-state operation of i-mode could be evidently given as

$$\int_0^\ell K_g^i(x)U_i(x)dx = \int_0^\ell K_i(x)U_i(x)dx\ , \tag{1}$$

where $K_g^i(x)$ and K_i (x) are the gain and loss coefficients per cm of active media length and $U_i(x)$ the volume density of i - mode radiation at the point x. The value of $K_i(x)$ denotes the laser radiation losses due to the inactive absorption (including the absorption by the excited molecules) and scattering, edge losses and losses connected with output of radiation throughout the mirrors of the resonator. Assuming all these losses independent on x, we may transform (1) to

$$K_i = \frac{\int_0^\ell K_g^i(x)U_i(x)dx}{\int_0^\ell U_i(x)dx} \tag{2}$$

For the sake of the simplicity we shall consider a small overlapping of luminescence and absorption bands when for $K^i(x)$ the expression below is valid

$$K_g^i(x) = \varkappa_i \frac{n^*(x)}{n}\ , \tag{3}$$

where $\varkappa_i = \frac{h\nu}{v} n B_i$ is the gain coefficient per full number of molecules n , B_i the Einstein coefficient, and $n^*(x)/n$ the relative population of the excited level at the point x .Using (3) and representing the density of laser radiation as a production

$$U_i(x) = U_i^0 \cdot f_i(x) \tag{4}$$

where u_i^0 is the amplitude of $u_i(x)$, one may rewrite (2) in the form

$$\frac{k_i}{\varkappa_i} = \frac{n^*}{n} . \qquad (5)$$

We denoted by n_i^* the value

$$n_i^* = \frac{\int_0^\ell n^*(x) f_i(x)\,dx}{\int_0^\ell f_i(x)\,dx} \qquad (6)$$

which is an average density of the excited molecules interacting with mode i. To find n_i^* we shall use the balance equation for the steady-state regime

$$nBu_p = n^*(x)\left(Bu_p + \frac{1}{\tau}\right) + n^*(x)\,B_i u_i(x) + n^*(x)\sum_{j\neq i} B_j u_j(x) . \qquad (7)$$

Here $B_i u_i(x)$ and $B_j u_j(x)$ are the probabilities of induced emission at i and j frequencies, and τ is the lifetime of the excited state. The rate of pumping Bu_p and the total concentration $n = n_1(x) + n^*(x)$ are assumed to be independent on x .

Multiplying the equation (7) by $f_i(x)$, then integrating the left-hand and right-hand sides by x from 0 to ℓ and dividing the result into $\int_0^\ell f_i(x)\,dx$ one can obtain the equation for the unknown value of n_i^* :

$$\frac{n}{n_i^*}\tau Bu_p = 1 + \tau Bu_p + \beta_i \tau B_i u_i^0 + \sum_{j\neq i} \alpha_{ij} \tau B_j u_j^0 . \qquad (8)$$

Here

$$\beta_i = \frac{\int_0^\ell n^*(x) f_i^2(x)\,dx}{\int_0^\ell n^*(x) f_i(x)\,dx} , \qquad (9)$$

$$\alpha_{ij} = \frac{\int_0^\ell n^*(x) f_i(x) f_j(x)\, dx}{\int_0^\ell n^*(x) f_i(x)\, dx} . \tag{10}$$

If modes under consideration are plane standing waves the distribution function $f_i(x)$ may be represented as

$$f_i(x) = \frac{1}{2}\left[1 - \cos\frac{2\pi(k+i)x}{\ell}\right] \tag{11}$$

where k and i are the whole numbers. In terms of negligibly small oscillations of the value $n^*(x)$ over standing wave period[9] we may simplify (9) and (10) to

$$\beta_i = \frac{\int_0^\ell f_i^2(x)\, dx}{\int_0^\ell f_i(x)\, dx} , \tag{12}$$

$$\alpha_{ij} = \frac{\int_0^\ell f_i(x) f_j(x)\, dx}{\int_0^\ell f_i(x)\, dx} . \tag{13}$$

Substituting (11) into (12) and (13) one finds $\beta = 3/4, \alpha = 1/2$. Independence of the parameters on the mode numbers i and j means that both parameters are of the same values for any modes. Keeping this in mind one obtains from (5) and (8)

$$(\beta-\alpha)\tau B_i U_i^0 + \alpha\sum_{j=1}^{m}\tau B_j U_j^0 = \tau B U_p\left(\frac{\varkappa_i}{k_i} - 1\right) - 1 . \tag{14}$$

The sum $\sum_{j=1}^{m}\tau B_j U_j^0$ in (14) can be found by summarizing the left-hand and the right-hand sides of equation (14) by i index from 1 to m (m is the total number of oscillating modes). Having substituted the result back to (14) one can find after some simple algebra the following expression for the radiation density of i mode

$$u_i^{o} = \frac{h\nu n}{v\tau\beta\varkappa_i}\left\{ \frac{\tau B u \rho}{1-\gamma}\left[\frac{\varkappa_i}{k_i} - \frac{\gamma\left(\sum_i \frac{\varkappa_i}{k_i} - 1\right)+1}{\gamma(m-1)+1}\right] - \frac{1}{\gamma(m-1)+1}\right\} \qquad (15)$$

Here $\gamma = \alpha/\beta$ (for the plane standing waves $\gamma = 2/3$).

Now we shall take into account the spectral dependence of the gain and the loss coefficients. Let the gain band be described by a dispersion curve. If the beginning of the mode number count is put at the maximum of the gain band the spectral function $\varkappa_i$ assumes the form

$$\varkappa_i = \frac{\varkappa_0}{1 + i^2\left(\frac{\delta\nu}{a}\right)^2} \qquad (16)$$

where $2a$ is the gain halfwidth and $\delta\nu$ is the spectral space between neighbour modes. Taking into account that spectral selector (prism, diffraction grating and others) can be inserted into a resonator and assuming the additional losses Δk_j and Δk_t are present at the frequencies j and t, respectively, we shall write down the loss coefficient in the form

$$k_i = k_0\left[1 + i^2\left(\frac{\delta\nu}{b}\right)^2\right] + \delta_{ij}\,\Delta k_j + \delta_{it}\,\Delta k_t \qquad (17)$$

where $2b$ is the halfwidth of the selector transmission and δ_{ij} , δ_{it} are the delta functions.

From formulae (15) - (17) using the connection between total number of oscillating modes m and the maximum number of the last oscillating mode i_0 ($m = 2i_0 + 1$) and taking into account the total spectral width of laser emission $i_0\,\delta\nu$ being always less than the halfwidth of the gain band (a) (or the halfwidth of the selector transmission band b, if any) one can come to expression

$$U_i^0 = \frac{h\nu n\left[1+i^2\left(\frac{\delta\nu}{a}\right)^2\right]}{v\tau\beta\varkappa_0}\cdot\left\{\frac{\tau B U_p}{1-\gamma}\left[\frac{\varkappa_0/K_0}{1+i^2\left(\frac{\delta\nu}{c}\right)^2+\left[1+i^2\left(\frac{\delta\nu}{a}\right)^2\right]\left[\delta_{ij}\frac{\Delta K_j}{K_0}+\delta_{it}\frac{\Delta K_t}{K_0}\right]}\right.\right. - \tag{18}$$

$$\left.\left.\frac{\gamma\left\{\frac{\varkappa_0}{K_0}(2i_0+1)\left[1-\frac{1}{3}i_0^2\left(\frac{\delta\nu}{c}\right)^2\right]-1\right\}+1}{2\gamma i_0+1}\right]-\frac{1}{2\gamma i_0+1}\right\}$$

Deduction of (18) is made under conditions $K_0/\varkappa_0 \ll 1$, $i_0 \gg 1$, which are practically always valid. The value of

$$c = \frac{ab}{\sqrt{a^2+b^2}}$$

is equivalent to a if there is no a selector in the resonator ($b \to \infty$) or to b when the selector is used with $b \ll a$. Solving the equation (18) at $i = i_0$, which makes $U_0(i = i_0) = 0$ one can find the maximum number of oscillating mode

$$i_0 = \left(\frac{c}{\delta\nu}\right)^{2/3}\sqrt[3]{\frac{3}{4}\left(\frac{1}{\gamma}-1\right)\left(1-\frac{1}{X_0}\right)}\ . \tag{19}$$

Here $X_0 = \frac{\tau B U_p}{K_0/\varkappa_0}$ is the ratio of the pumping power to the threshold power for mode $i = 0$ (it is easy to see that threshold for $i = 0$ is $\tau B U^0_{thr} = K_0/\varkappa_0$) . For plane standing waves ($\gamma = 2/3$) the total number of oscillating modes is

$$m \simeq 2i_0 = 1.44\left(\frac{c}{\delta\nu}\right)^{2/3}\sqrt[3]{1-\frac{1}{X_0}} \tag{20}$$

which is in exact coincidence with the corresponding expression in the paper[9] obtained by other means.

From (18) it is easy to find the gap depth in the laser emission spectrum at the mode j to be

$$\Delta U_j^0 = U_j^0 - U_j^0(\Delta K_j = 0) = \frac{h\nu n}{v\tau\beta K_0}\cdot\frac{\tau B U_p}{1-\gamma}\left[1-j^2\left(\frac{\delta\nu}{b}\right)^2\right]^2\frac{\Delta K_j/K_0}{1+\frac{\Delta K_j}{K_0}\left[1-j^2\left(\frac{\delta\nu}{b}\right)^2\right]} \tag{21}$$

when a selector is used or

$$\Delta U_j^0 = \frac{h\nu n}{v\tau\beta} \cdot \frac{\tau B U_p}{1-\gamma} \cdot \frac{1}{K_0} \quad \frac{\Delta K_j/K_0}{1+\Delta K_j/K_0} \tag{22}$$

without a selector. If $\Delta k_j = 0$ expression (18) may be simplified to

$$U_j(\Delta k_j=0)=U_j^* \simeq \frac{h\nu n[1+j^2(\frac{\delta\nu}{a})^2]}{2 v\tau\beta\varkappa_0\gamma i_0} \cdot \left\{ X_0 \left[1 - \frac{2i_0}{1/\gamma - 1}(j^2 - \frac{1}{3}i_0^2)(\frac{\delta\nu}{c})^2\right] - 1 \right\} \tag{23}$$

The ratio of (21) or (22) to (23) gives the relative value of the gap depth $\Delta U_j^0/U_j^*$. For the central frequency $j = 0$ the value is

$$\frac{\Delta U}{U} = \frac{X_0}{1/\gamma - 1} \cdot \frac{\Delta K/K_0}{1+(\Delta K/K_0)} \cdot \frac{2i_0}{X_0\left[1+\frac{2i_0^3}{3(\frac{1}{\gamma}-1)}\cdot(\frac{\delta\nu}{c})^2\right]-1} \tag{24}$$

or after substituting (19) for i_0

$$\frac{\Delta U}{U} = A \cdot \frac{\Delta K/K_0}{1+(\Delta K/K_0)} \tag{25}$$

where

$$A = \left[\frac{4\bar{n}LC_0}{3(\frac{1}{\gamma}-1)(1-\frac{1}{X_0})} \right]^{2/3}, \tag{26}$$

$\bar{n}$ is the mean refraction index of the media in the resonator, L the resonator base and $C_0 = 2C$ the halfwidth of the active media gain band (or selector transmission band). If the relative error of experiment is $(\Delta U/U)_{min} = \theta$ the absolute sensitivity of the intracavity absorption measurements at frequency $j = 0$ is

$$(\Delta K_0 \ell_0)_{min}^{in\,cav} = \frac{\theta K_0 \ell}{A} \tag{27}$$

where Δk_0 is the absorption coefficient at this frequency and ℓ_0 the thickness of the sample.

It is easy to see the absolute sensitivity of the optical density measurements with the conventional method to be equal to the relative error

$$(\Delta k_0 \ell_0)_{min}^{conven} = \theta .$$

If the parameter θ is of the same value in both cases the enhancement of the sensitivity for intracavity method is

$$\xi = \frac{(\Delta k_0 \ell_0)_{min}^{conven}}{(\Delta k_0 \ell_0)_{min}^{in\ cav}} = \frac{A}{k_0 \ell} . \qquad (28)$$

For comparison we shall give the corresponding formulae for the single frequency laser operation. In this case instead of the spectral gap one registers the difference Δu of intensities with and without the absorbing sample in the resonator. From (5) and (7) under $\sum_{j \neq i} u_j = 0$ one obtains

$$\left(\frac{\Delta u}{u}\right)_{single} \simeq \frac{\Delta k/k_0}{1 + \Delta k/k_0} \cdot \frac{1}{1 - \frac{1}{x_0}} , \qquad (29)$$

$$(\Delta k_0 \ell_0)_{min}^{single} = \theta k_0 \ell \left(1 - \frac{1}{x_0}\right) , \qquad (30)$$

$$\xi_{single} = \frac{\Delta k_0 \ell_0{}_{min}^{conven}}{(\Delta k_0 \ell_0)_{min}^{single}} = \frac{1}{k_0 \ell \left(1 - \frac{1}{x_0}\right)} . \qquad (31)$$

It is quite evident the formulae (29) - (31) are also valid for any frequency of multi-mode laser emission if the gain spectrum is completely inhomogeneous and each mode interacts only with its own type of molecules.

To analyze the above formulae one needs to know the loss coefficient k_0. According to reference[10] it may be presented in the form of

$$k_0 = \rho_0 + \frac{1}{2l} \ln \frac{1}{z_1 z_2} + \frac{L\varphi}{ld} , \qquad (32)$$

where ρ_0 is scattering and inactive absorption coefficient, z_1 and z_2 are resonator mirror reflectivities, φ is angle divergency, and d is an aperture.

DISCUSSION

If there is no a selector in the resonator the absolute value of the gap depth at the frequency of the sample absorption, according to (22), does not depend on the mode number j (i.e. on the position of the absorption line of the sample relative to the center of the laser emission spectrum) and is totally determined by the value of the sample absorption loss Δk_j. In the presence of a selector (see (21)) the gap depth decreases as frequency moves off the maximum of the selector transmission band. In both cases the absolute gap depth is proportional to the pumping power. As is seen from (21) and (23) in contrast to the absolute value (Δu_j^o) the relative one ($\Delta u_j^o / u_j^*$) increases as frequency moves off the gain band center.

Taking into account the ratio $\Delta u_j^o / u_j^*$ cannot exceed 1 ($\Delta u_j^o / u_j^* = 1$ corresponds to zero intensity of the laser emission at the frequency j) one can conclude from (21), (23) and (19) that for any mode number

$$j \geqslant \sqrt{i_0^2 - \frac{\Delta k_j / k_0}{1 + \Delta k_j / k_0} \cdot \left(\frac{a}{\delta \nu}\right)^2} \tag{33}$$

the presence of the additional losses Δk_j will reduce the intensity of the mode to zero (in (33) it is assumed for the sake of the simplicity $c = a$). The maximum value of Δk_j over which the full gap of the intensity at frequency j = 0 will be observed is

$$\Delta k_{max} = \frac{k_0}{A} \tag{34}$$

Considering the absorption loss of the sample $\Delta k \leqslant k_{max}$ and assuming $A \gg 1$ we may transform (25) into the simple linear function

$$\Delta u / u \simeq \Delta k / k_{max} . \tag{35}$$

According to (34), (35) and (26) the linear dependence of $\Delta u / u$ on Δk is true at any level of pumping. This property is of the great practical value and may serve as the basis for the simple quantitative method of the intracavity selective absorption measurements. As it follows from (25) and (26) the linear function (35) is valid only under the condition $A \gg 1$. The A estimation for the typical parameters ($L \simeq 50$ cm, $c_0 \simeq 2000\ cm^{-1}$, $x_0 \gg 1$, $\gamma = 2/3$, $\bar{n} \sim 1$) gives $A = 4.10^3$, which means the inequality $A \gg 1$ for dye lasers is fully guaranteed. In contrast to the multimode regime the single frequency operation (or multimode operation with completely inhomogeneous gain spectrum of the active media) is characterized by nonlinear dependence of $\Delta u / u$ on Δk , making it inconvenient for the practical usage.

From (21) and (22) one can draw a conclusion of the method spectral resolution which is of the most impoŕtant characteristics. It is seen from (21) the gap depth in the laser spectrum at j mode is determined by the additional losses at frequency j and independent on the value of the additional losses at frequency t. As this is valid for any j and t uncluding $t = j \pm 1$, the conclusion can be made that the spectral resolution corresponds to the intermode distance $\delta\nu$, i.e. rather high (for example $\delta\nu = 10^{-2}$ cm^{-1} if $L = 50$ cm).

As it follows from (22) the ratio of the gap values at frequencies j and t (under $\Delta k_j / k_0 \ll 1$) is proportional to that of the loss coefficients at the frequencies

$$\Delta u_j^0 / \Delta u_t^0 = \Delta k_j / \Delta k_t \ . \tag{36}$$

It is easy to show the relationship is true as well if the additional losses are present not only at two frequencies (j and t) but at any number m^* of frequencies as far as m^* is sufficiently less than the total number of modes $m = 2i_0 + 1$. It means the simple measurements of the undistorted absorption line shape can be made by the method if only the line width under investigation is sufficiently narrower than the full width of the laser emission spectrum. One should keep in mind the shape of the absolute (Δu_j^0) but not relative (Δu_j^0 / u_j^*) intensity gap gives the right shape of the absorption line.

Now let us consider the sensitivity of the intracavity absorption measurements. As is seen from (28) and (26) the sensitivity enhancement factor depends on the laser radiation losses per a resonator pass ($K_0 \ell$), the resonator base (L), the gain band halfwidth (C_0), the pumping power excess over the threshold (x_0) and parameter γ which describes the spatial overlapping of modes in the active media. One can see the dependence on the pumping power is essential only at low pumping levels. The sensitivity reaches its maximum if a laser operates at the very threshold. But usually such a regime is insufficiently stable and inconvenient for practical use. If the laser pumping exceeds the threshold by a factor of 1,5 or more the relative sensitivity becomes practically independent on it. But the absolute gap depth increases permanently along with the pumping power (see formula (22)).

The broader the gain band of the laser active media (C_0) the higher sensitivity of the intracavity method. Insertion of any spectral selectors into the resonator resulting in the narrowing of the laser emission spectrum (C_0 decreases) reduces the sensitivity . On the contrary the partial compensation of the gain spectral dependence leading to the broadening of the laser emission spectrum could result in higher sensitivity of measurements.

The analysis of enhancement factor dependence on parameter L shows the existence of the optimum resonator base

$$L^{opt} = \frac{2d}{\varphi}\left(\rho_0 \ell + \frac{1}{2} \ln \frac{1}{z_1 z_2}\right) \qquad (37)$$

corresponding to the maximum sensitivity of measurements. For typical values $\varphi \simeq 5.10^{-3}$ rad, $2d = 0.5$ cm, $\rho_0 \ell + \frac{1}{2} \ln \frac{1}{z_1 z_2} = 0.5$ formula (37) gives $L^{opt} \simeq 50$ cm.

Parameter γ amounts 2/3 while using the plane mirror resonator. But it can be increased the resonator being exchanged for a spherical one. This change is accompanied by mode-competition strengthening and as is seen from (26) and (28) by selective absorption sensitivity rise.

The value of the laser radiation loss per a resonator pass ($k_0 \ell$) is of a great importance for the method. Sensitivity enhancement factor ξ is inversely dependent on the parameter $k_0 \ell$. As it follows from (32) this parameter can be reduced by increasing of the mirror reflectivities, decreasing of the angle divergency and increasing of an aperture. Unfortunately, the value of $k_0 \ell$ for pulsed dye lasers is comparatively large ($k_0 \ell \sim 1$). In such a case enhancement factor ξ equals parameter A , which typically has a value of 4.10^3. If relative experimental error is $\theta \sim 5\%$the optical density values as low as $\Delta k_0 \ell_0 \sim 10^{-5}$ can be registered. Parameter $k_0 \ell$ for CW dye lasers apparently could be reduced by a factor of 10^2 or 10^3 which should permit the increasing of the absolute sensitivity up to the optical densities $\Delta k_0 \ell_0 \sim 10^{-7} - 10^{-8}$.

In the case of the single frequency operation the sensitivity of the intracavity method is higher than conventional one only at $k_0 \ell < 1$ (we consider $x_0 \gg 1$). If $k_0 \ell > 1$ the conventional measurements are preferred(the same is true for multifrequency laser operation if the gain spectrum of active media is completely inhomogeneous). The typical sensitivity of the multimode intracavity method exceeds one of the single frequency operation by a factor of $A \sim 4.10^3$.

The results presented above differ greatly from the conclusion of the paper[8]. It is assertained in the paper that at the high pumping levels the sensitivities of the intracavity absorption measurements with homogeneous and completely inhomogeneous spectrum broadening of active media are adequate. In both cases according to the reference[8] enhancement parameter ξ is equal to $1/\kappa_0 \ell$ which is three orders less than the value ξ given by formula (28) above.

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