Dynamics of DFB dye lasing by polarization modulation: simulations and experiment

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Abstract
Distributed feedback (DFB) dye lasing by polarization modulation is studied theoretically on the basis of modified rate equations. The numerical solution of these equations allows one to obtain the generated power dynamics and the dependence of laser energy on pump energy. The results of the calculations are in good qualitative agreement with the experimental data.

Keywords: DFB dye laser, polarization grating, rate equations

(Some figures may appear in colour only in the online journal)

1. Introduction

Distributed feedback (DFB) lasers are compact and easy-to-use sources of narrowband coherent radiation. In DFB lasers, the feedback is provided via backward Bragg light scattering from the spatially periodic structure formed inside the active medium itself and, hence, is distributed throughout its length. The idea of these lasers was put forward and implemented by the authors of [1, 2]. As applied to dye solutions as active media, the spatially periodic structures can be either of a stationary [1] or dynamic (rapidly reversible) [2] character. It should be noted that dynamic DFB dye lasers exhibit the highest performance characteristics. A number of optical schemes of these lasers have been developed. Their advantages have been demonstrated, such as a small spectral linewidth, wide tuning range, high efficiency, the possibility of a high repetition rate operation without dye solution flow as well as simultaneous generation at two or more wavelengths with independent spectral tuning etc [3–11]. At the same time, the most interesting and important feature of dynamic DFB dye lasers is their ability to produce picosecond pulses both at picosecond and nanosecond excitation [12–18]. In the latter case, the use of these lasers represents the simplest way of obtaining single pulses of a few tens of picosecond duration with the possibility of smooth wavelength tuning.

Due to their important merits, DFB dye lasers are still the subject of steady attention and intensive study by scientists from different research centres (see some recent publications [19–26]). In dynamic DFB dye lasers, the excitation of the active medium is generally provided by two converging pump beams with a vertical orientation of the electric field vector (s-polarization). In this case, the spatially periodic modulation of the resultant pump field intensity is provided along the excited dye region. A fundamentally different type of modulation occurs when the pump beams are orthogonally polarized (for example, the first one is s-polarized, while the second one is p-polarized). Under such conditions, the pump field intensity is uniform over the excited zone of the dye, while a periodic change of the resultant excitation field polarization takes place. Due to the anisotropy of the light absorption and emission by dye molecules, the above pump field polarization modulation results in the formation of a transient gain dichroism (i.e. polarization) grating inside the dye solution.

Dynamic DFB dye laser action by polarization modulation was experimentally investigated in [27–30], where the spectral, threshold and polarization characteristics of a DFB laser pumped by the second harmonic from a nanosecond Nd:YAG laser were reported. Unlike the works mentioned above, in our studies we used the second harmonic generation from a subnanosecond diode-pumped solid-state (DPSS) Nd:LSB microlaser to initiate the DFB lasing by polarization modulation in the dye solution [31, 32]. Under such conditions, the DFB laser exhibited a spectral linewidth of $\Delta \lambda_{0.5} < 0.008$ nm (full width at half-maximum, FWHM) and an energy conversion efficiency as high as $\eta_{\text{max}} \sim 48\%$. It should be noted for comparison that under identical pumping conditions the DFB
lasing by intensity modulation showed a similar value of $\Delta \lambda = 0.5$ and $n_{\text{max}} \sim 50\%$. Thus, in terms of the spectral linewidth and peak lasing efficiency, the DFB laser based on the dynamic polarization grating is comparable with that employing traditional (i.e. intensity modulation) pumping geometry. To the best of our knowledge, no similar results regarding the spectral line narrowness and efficiency of DFB dye lasing by polarization modulation have been reported previously. At the same time, our results seem to be of undoubted interest and indicate the potential of these devices. It is obvious that, for its practical realization, a more in-depth insight into the operation of these DFB dye lasers is required. The latter is hardly possible without an appropriate theoretical model capable of adequately predicting their performance. Since, as far as we know, all existing studies still lack any theoretical treatment of the properties of these laser devices, here we attempt to fill this gap.

In this paper, we propose a simple theoretical model of DFB lasing by polarization modulation and report on the numerical simulations of its dynamics for the case of subnanosecond excitation. The results obtained reveal that, at moderate pumping rates, a multi spike emission is provided by a DFB laser, while single picosecond pulse generation takes place at the pump intensities not far from the threshold. As proof of the consistency and usefulness of the proposed model, the output characteristics of the Rhodamine 6G DFB laser excited by two orthogonally polarized second harmonic beams from a subnanosecond DPSS Nd:LSB microlaser are presented.

2. Theoretical model

The geometry of the model is shown in figure 1. The field of the pump beam which forms the DFB grating is a sum of the two plane waves crossing inside the medium at an angle $2\beta$. We denote $\beta$ the angle between the pump wave vector and the unit vector along the $z$ axis. Then, the spatial distribution of the total pump electric field can be written (after time averaging) as follows,

$$
\vec{E} = \vec{E}_1 e^{ik_1(y \cos \beta + z \sin \beta)} + (\vec{E}_2 \sin \beta + \vec{E}_3 \cos \beta) A_2 e^{ik_2(y \cos \beta - z \sin \beta)},
$$

where $A_1$ and $A_2$ are the amplitudes of the pump waves, $\vec{E}_{1,2,3}$ are the unit vectors along the corresponding axes and $k_1 = 2\pi / \lambda_1$ is the wavenumber of the pump waves with the wavelength $\lambda_1$. The field (1) gives the periodic change of the polarization state along $z$ axis, while its intensity $|\vec{E}|^2$ does not depend on the position.

Interaction of a dye molecule with the field is governed by the value of $|\vec{d} \cdot \vec{E}|^2$, the direction of the molecular dipole moment vector being defined as

$$
\vec{d} = |\vec{d}|(\vec{e}_x \sin \theta \cos \varphi + \vec{e}_y \cos \theta \sin \varphi + \vec{e}_z \cos \theta),
$$

where the angles of the spherical coordinate system $\theta$ (zenith angle) and $\varphi$ (azimuth angle) are counted from $z$ and $x$ axes, correspondingly. Using equations (1) and (2), the excitation efficiency for the molecule with the dipole moment oriented along angles $\theta$ and $\varphi$ can be calculated by

$$
f_{\text{exc}}(\theta, \varphi) = |\vec{d} \cdot \vec{E}|^2 = A(\theta, \varphi) + B(\theta, \varphi) \cos(2\pi \Lambda / \lambda_1).
$$

Here $\Lambda = \pi / \lambda \sin \beta = \lambda / 2 \sin \beta$ is the grating period, and the auxiliary functions are

$$
A(\theta, \varphi) = I_1 \sin^2 \theta \cos^2 \varphi + I_2 \sin^2 \beta \sin^2 \varphi + \cos^2 \beta \cos^2 \theta + \frac{1}{2} \sin 2\beta \sin 2\theta \sin \varphi,
$$

$$
B(\theta, \varphi) = \sqrt{I_3} \sin \beta \sin^2 \theta \sin 2\varphi + \cos \beta \sin 2\theta \cos \varphi,
$$

where $I_{1,2} = A_1^2 / 2(\Lambda_1^2 + \Lambda_2^2)$ are the coefficients which define the part of the entire pump power contained in both incident waves.

Thus, for every molecule orientation, we have the excitation grating which is the result of polarization modulation and is described by equation (3). In order to calculate the lasing dynamics, we use the idea of [33]. Let us consider molecules of given orientation separately in the maxima and minima of the excitation grating, i.e. for two values of efficiency

$$
f_{\text{exc}}^\pm(\theta, \varphi) = A(\theta, \varphi) \pm B(\theta, \varphi).
$$

These two values should be substituted in the rate equations which describe the temporal dynamics of the excited molecules' concentration and the density of the generated photons. To obtain these equations, we use the rate equations given in [15, 34] for DFB laser action by intensity modulation and generalize them in the case of polarization modulation. As a result, we have the following equations for the densities $n_{\pm}(\theta, \varphi)$ of the excited molecules in the maxima and minima of the excitation grating,

$$
\frac{dn_{\pm}(\theta, \varphi)}{dt} = 3 I_p(t) n_{\pm}(\theta, \varphi)[N/4\pi - n_{\pm}(\theta, \varphi)]
$$

$$
- \frac{n_{\pm}(\theta, \varphi)}{\tau_2} - 3 \frac{c}{\eta} n_{\pm}(\theta, \varphi) (q_s \cos^2 \varphi + q_i \sin^2 \varphi) \sin^2 \theta,
$$

where $q_s$ and $q_i$ are the absorption and emission cross-sections, $I_p(t)$ is the intensity profile of the pump pulse, $q_s$ and $q_i$ are the average densities of the photons polarized along the $x$ and $y$ axes, respectively, $N$ is the density of the dye molecules, $\tau_2$ is the life-time of the excited state, $\eta$ is the refractive index of the dye solution, $c$ is the vacuum speed of light and the coefficient $3$ is the result of the usage of the cross-sections averaged over molecular orientations. The first term on the right-hand side of equation (4) describes the process of molecular excitation under pump influence, while the second and third terms are responsible for the spontaneous and stimulated decay of the excited state, respectively.

In order to obtain the equations for $q_s$ and $q_i$, we need the expressions for the average gain coefficients for the light
polarized along the \( x \) and \( y \) axes (s- and p-polarizations). These values are defined as

\[
k_{x,y} = \frac{1}{2}(k_{x,y}^+ + k_{x,y}^-),
\]

where the gain coefficients in the (maxima and minima of the excitation grating can be calculated through

\[
k_{x,y}^\pm = 3\alpha_0 \int_0^\pi \int_0^{2\pi} n_{s,p}(\theta, \varphi) \sin^2 \varphi \sin^2 \theta \, d\varphi \, d\theta,
\]

Then, the equations for the average densities of the s- and p-polarized photons are as follows,

\[
\frac{d \rho_{s,p}}{dt} = \frac{c}{\eta} k_{s,p} q_{s,p} - \frac{q_{s,p}}{\tau_{s,p}} + \frac{\Omega}{\tau} \bar{n}.
\]

Here, the photon life-time in the cavity \( \tau_{s,p} = \eta \lambda L \alpha_{s,p}^2 / 8 c \pi^2 \) is proportional to the squared amplitude of the gain grating [15], which in our case is equal to \( \alpha_{s,p} = (k_{x,y}^+ - k_{x,y}^-)/2 \); \( L \) is the length of the DFB structure. The last term in equation (6) describes the spontaneous emission [34] with a coefficient \( \Omega = b / \pi \alpha_0 L^2 S \), where \( b \) is the height of the excited volume, \( S \) is the spectral factor which determines the fraction of the spontaneous emission falling into the laser bandwidth. The average density of the excited molecules is calculated by \( \bar{n} = \langle n_+ \rangle + \langle n_- \rangle / 2 \) with \( n_{\pm} = \int_0^{2\pi} n_s(\theta, \varphi) \sin \theta \, d\varphi \, d\theta \).

Finally, the lasing power can be determined by the expression [15, 34]

\[
P_{\text{out}} = P_s(t) + P_p(t) = \frac{h c}{2 \lambda} L \text{Im} \left[ \frac{q_s(t)}{\tau_s(t)} + \frac{q_p(t)}{\tau_p(t)} \right],
\]

where \( h \) is the Planck constant, and the penetration depth \( a \) of the pump beam into the dye solution can be estimated from

\[
\frac{1}{a} = 3\alpha_0 \frac{N}{4\pi} \int_0^{\pi} \int_0^{2\pi} A(\theta, \varphi) \sin \theta \, d\varphi \, d\theta = \alpha_0 N.
\]

Equations \(4\) and \(6\) are the main relations of our model. Solving them numerically and substituting this solution into equation \(7\), we can describe the dynamics of power and the polarization properties of the DFB lasing by polarization modulation.

3. Numerical results

Let us consider some results of the numerical solution of the rate equations presented in the previous section. Calculations were performed for the set of parameters which can be considered typical for the dye-laser systems: pump wavelength \( \lambda_p = 532 \text{ nm} \), laser wavelength \( \lambda_L = 565 \text{ nm} \), absorption and emission cross-sections (Rhodamine 6G) \( \alpha_0 = 3.8 \cdot 10^{-16} \text{ cm}^2 \) and \( \sigma = 2.15 \cdot 10^{-16} \text{ cm}^2 \), concentration of the dye solution \( C = 0.243 \text{ mM} \), refractive index of the solution at the laser wavelength \( \eta = 1.36 \), excited state lifetime \( \tau = 4 \text{ ns} \), the length and the height of the DFB structure \( L = 1 \text{ cm} \) and \( b = 0.01 \text{ cm} \), respectively, and spectral factor \( S = 10^4 \). The pulse of the pump intensity has a Gaussian envelope with FWHM 0.5 ns. The ratio of the intensities of the pump beams with s- and p-polarizations is 2 : 1, i.e. the intensity parameters are \( I_1 = 0.67 \) and \( I_2 = 0.33 \). The angle of incidence for the pump beams is \( \beta = \text{arcsin} \lambda_p / \lambda_L = 70.32^\circ \).

We start our analysis with the laser power dynamics calculated at different pumping levels. We are interested in the qualitative relations rather than in the absolute values of the laser characteristics. Therefore, we measure pump energy \( W_p \) with normalized parameter \( \gamma = W_p / W_{\text{th}} \), where \( W_{\text{th}} \) is the threshold pump energy. This threshold is determined as the point of abrupt rise of the output power. In absolute units, our calculations give \( W_{\text{th}} = 2.5 \mu J \) for the parameters listed above.

According to our calculations, only s-polarized light is generated at low excesses over the pump threshold. In this case, laser radiation forms a single pulse which, at the threshold, appears at the trailing edge of the pump pulse and arises earlier and earlier as the pump energy grows. The example of this s-polarized pulse is shown in figure 2(a) at \( \gamma = 1.28 \). This single-pulse lasing corresponds to the sharp decrease in the average density \( \bar{n} \) of the excited molecules presented in the
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It can be seen that at first the value $n$ grows smoothly as a result of the pump and then, when it becomes larger than a certain level (which in general depends on the pump energy), the density of the excited molecules drops, releasing the stored energy in the form of laser radiation. As for the generation of p-polarized light under these conditions, it has a low intensity and cannot be described by the characteristic bell-shaped envelope (see the lower panel in figure 2(a)).

When $\gamma \geq 1.5$, the generation of the second pulse starts. This second pulse is p-polarized, which is illustrated by the profiles calculated at $\gamma = 2$ (figure 2(b)). In this case, the dynamics of the average density of the excited molecules has two well-pronounced peaks which correspond to both laser pulses. The third pulse appears for $\gamma \geq 2.25$ and is s-polarized like the first one. The example of the calculated power profiles and behaviour of $\hat{n}$ with three maxima are shown in figure 2(c) for $\gamma = 2.76$. Finally, when $\gamma \geq 3$, the fourth pulse

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**Figure 2.** Results of calculation of the generated pulse profiles (solid lines) and dynamics of the average density of excited molecules (dotted lines) at different pump levels: (a) $\gamma = 1.28$, (b) 2, (c) 2.76, (d) 4. The density of excited molecules is normalized by the full density of the dye molecules $N$. The dashed line shows the profile of the pump pulse.
Figure 3. Laser output energy versus pump energy calculated with our model. The arrows indicate pump energies where a new pulse appears at the laser output.

As can be seen in figure 2(d), at $\gamma = 4$, this fourth pulse contains comparable quantities of both s- and p-polarized radiation.

After these examples, we should consider the energetic characteristics of laser action described by our model. Figure 3 demonstrates the full output energy and the energy of s- and p-polarized laser light taken separately as the functions of pump energy. One can see that the appearance of subsequent pulses is characterized by inflection points in the curves depicted in figure 3. Note that before this inflection point is reached, a plateau occurs, where the generated energy changes very slightly with the pump energy. This property of the laser system under consideration seems to be conducive to obtaining linearly polarized single pulses with high stability of the output energy.

4. Experimental results

The output characteristics of a DFB dye laser based on dynamic polarization grating were investigated under subnanosecond excitation. The optical scheme of a DFB laser and registering apparatus were practically the same as in [35, 36] (figure 4). A 0.26 mM ethanol solution of Rhodamine 6G was used as the active medium. The vertically polarized second harmonic generation ($\lambda = 532$ nm; $\Delta \lambda_{0.5} < 0.003$ nm; beam quality factor $M^2 \leq 1.2$) from a DPSS STA01SH-500 Nd:LSB microlaser (Standa Ltd, Lithuania) delivering $\sim$0.5 ns (FWHM) pulses with an energy of $E_P \leq 80 \mu J$ and a repetition rate as high as $f = 500$ Hz was employed as a pump source. The DFB laser oscillator represented a right-angle isosceles K-8 glass prism, the hypotenuse side of which is in optical contact with a dye in a cell with nonparallel windows. In this scheme, the 532 nm pump beams are symmetrically coupled into the active medium through the side faces of the prism, thereby resulting in the first-order DFB laser operation. Laser oscillations occur at a wavelength $\lambda_L$ given by

$$\lambda_L = \frac{n_s \lambda_P}{2 n_p \sin \alpha},$$

where $n_s$ is the refractive index of the dye solution at the laser wavelength, $n_p$ is the refractive index of the prism material at the pump wavelength $\lambda_P$ and $\alpha$ is the incident angle of the pump beam at the prism-dye solution interface (it differs from the angle $\beta$ inside the solution). To obtain a pair of pump beams with s- and p-polarizations, respectively, the electric field vector of one of the two microlaser beams was turned by 90° with the help of a half-wave plate. The intensity of s-polarized beam was approximately two times higher than that of the p-polarized one. The maximum total pump beams energy falling onto the surface of the dye solution in a cell did not exceed $E_P = 35 \mu J$.

To provide the optimal pumping geometry, the output beam from a microlaser with an initial diameter of about 0.02 cm and a full-angle divergence of $\sim 10$ mrad was collimated in the vertical plane by a cylindrical lens (focus length 18 cm), while the distance between the microlaser head and the DFB laser input window was set to $\sim 99$ cm. In these conditions, the excited zone of the dye in a cell represented a narrow horizontal stripe with a $\sim 0.9$ to 1.2 cm length and $\sim 0.01$ cm height, depending on the intersection angle 2$\alpha$ of the pump beams.

During the experiments, the pump input and the DFB laser output energies were measured simultaneously with calibrated FD-24K photodiodes and an ADC-2OM/10-2 two-channel analogue-to-digital converter (ADC). To adjust the pump power in a continuous manner, a rotatable half-wave plate in combination with a dielectric polarizer was employed. A fibre-coupled S3804 automated diffraction grating spectrograph (spectral resolution up to $\sim 0.08$ nm) and an IT51-30 Fabry–Perot interferometer were used for the coarse and fine spectral measurements, respectively. The transient behaviour of the pump and DFB laser pulses was studied using an Agat-SF3 ultrafast streak camera (time resolution up to $\sim 2$ ps). Both the interferograms and streak camera images were captured by means of a SDU-R205 charge-coupled device (CCD) USB-TV camera with subsequent computer acquisition and processing of the obtained data.

Under the above-mentioned pumping conditions, a DFB laser exhibited smooth wavelength tunability within 549–592 nm on a spectral linewidth of $\Delta \lambda_{0.5} < 0.008$ nm and an optical-to-optical energy conversion efficiency up to $\eta_{max} \sim 60\%$. The dependence of the output characteristics of a DFB laser on the experimental pumping conditions was investigated in detail at $\lambda_L = 565$ nm, which falls into the dye amplification band maximum. In this case, the length of the pumped region of active medium was $L_{DFB} \approx 1$ cm.

First, the influence of the pump pulse intensity on the temporal course of the DFB laser output was studied in detail. It was found that when pumped well above the threshold, the DFB laser generally produces a multi-spike emission. Both the overall length and the number of spikes in the pulse train depend on the pumping level $\gamma$ and tend to decrease with decreasing the $\gamma$ value. At pumping levels not very far from the threshold, single ultrashort pulses of a few tens of picoseconds duration are generated.

As an example, figure 5 reveals the streak camera traces of the DFB laser pulses registered at different pumping levels corresponding to the next pulse appearing in the laser output. Note that, unlike figure 2, the zero position of the time scale is
of an arbitrary character and does not reflect the real build-up dynamic of the DFB lasing. It should also be noted that, due to the relatively low streak camera scan rate ($v_{sc} \sim 1$ ns cm$^{-1}$) used during these measurements, both the individual pulses in the pulse trains and their durations are not fully resolved.

It can be seen from figure 5(a) that at a pumping level of $\gamma \approx 3.5$ a DFB laser produces a train of four pulses with an overall width of $\sim 900$ ps. When the pump power is lowered to $\gamma \approx 3.0$ (figure 5(b)) and $2.2$ (figure 5(c)), the number of emitted pulses reduces to three and two, respectively, while the overall pulse train lengths shorten to $\sim 700$–$800$ ps. At pumping levels $1 < \gamma < 1.7$ the stable generation of single picosecond pulses was observed (figure 5(d)). The single pulse duration was rather sensitive to the pumping level and progressively broadened on decreasing the $\gamma$ value. The shortest pulses were obtained near the threshold of the second pulse ($\gamma \approx 1.7$). In this case, their duration was measured to be about $\tau_{0.5} \approx 37$ ps (FWHM). Note that before performing the above measurements the streak camera was set to operate with a scan rate of $v_{sc} \sim 0.25$ ns cm$^{-1}$, thus providing an approximately four-fold increase in the time resolution. Under such conditions, the spectral linewidth of the DFB laser emission amounted to $\Delta \lambda_{0.5} \approx 0.0075$ nm, while its energy reached $E_0 \sim 0.13$ $\mu$J. Based on these data, the time-bandwidth product $\nu_0 \tau_{0.5}$ and peak power $P$ of a single pulse are estimated to be 0.3 and 3.5 kW, respectively. The obtained value of $\nu_0 \tau_{0.5}$ is evidence of the transform-limited character of the single picosecond pulses generated by a DFB dye laser.

Figure 6 shows the dependence of the DFB laser output energy $E_L$ on the pump energy $E_P$ measured for the range of pumping levels $1 < \gamma \leq 4$ (i.e. close to that in figure 5).

It can be seen that $E_L$ gradually grows on increasing the pump intensity. Along with this, at least three alterations in the slope may be noticed corresponding to the inflection points in figure 3. The first one is observed around a pump energy of $\sim 1.85$ $\mu$J representing the lasing threshold, while the second and third ones correspond to the $E_p \approx 3.15$ and 4.1 $\mu$J, respectively. Based on the two latter values of pump energies, we find that the appropriate pumping levels $\gamma$ are accordingly $\sim 1.7$ and $\sim 2.2$. From the streak camera measurements presented in figures 5(d) and (c) it follows that the specified pumping levels of $\gamma \approx 1.7$ and $2.2$ are indicative of the second and third picosecond pulses appearing in the DFB laser output.

The above features regarding the course of the input–output characteristic of a DFB laser operating not far from the threshold seem to be of undoubted practical interest. Indeed, based solely on the energy measurement, it becomes possible without using a high-cost streak camera to precisely determine the range of pump intensities within which single picosecond pulse generation is provided by a DFB dye laser.

5. Conclusion

In conclusion, we have proposed the theoretical model of DFB dye laser action by polarization modulation. This fundamentally simple model based on the consideration of values averaged over excitation grating allows us to describe the lasing dynamics and the energetic and polarization characteristics of these lasers. The mathematical basis of the model is the modified rate equations for the density of excited molecules and the concentration of laser photons.
A comparison of our calculation results with the experimental data shows good qualitative agreement between theory and experiment. In particular, the model allows us to describe the multi-spike lasing and approximately predict the threshold values for successive pulse generation. The polarization characteristics of these DFB lasers will be studied in detail in a separate work.

As far as we know, we have performed the first realistic description of the lasing dynamics of a DFB dye laser based on polarization modulation. However, to obtain not only qualitative, but also quantitative agreement between calculations and measurements, one needs an advanced theory. In future, we plan to develop a semiclassical treatment of these lasers, which is expected to give a closer fit to the experimental data.

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