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# Refractive-index saturation-mediated multiple line emission in polymer thin-film distributed-feedback lasers

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We report experimental and theoretical investigations of multiple laser-line emission in a distributed-feedback dye laser pumped by two coherent optical beams. We have used a Lloyd interferometer configuration with second- and third-order Bragg reflections to study the interaction between the two incident pumps in an organic thin film. We demonstrated theoretically that the number of laser emission lines can be interpreted with reference to the saturation effect in the refractive index.

Since the first papers by Kogelnik and Shank<sup>1,2</sup> about distributed-feedback lasers, many groups have successfully attempted, both experimentally<sup>3,4</sup> and theoretically,<sup>5,6</sup> the study of various configurations to obtain a laser emission. Optimized architectures have been obtained with high conversion efficiency and low divergence.<sup>7</sup> The Bragg reflector needed for laser emission is obtained in different configurations, some of them using a permanent grating<sup>8–11</sup> or a dynamic grating<sup>12,13</sup> obtained by means of an interferometer. If the pumps are perfectly superposed, many interactions can occur in the media. For example, Khan and Hall produced five and nine lines by using two and three pairs of pump beams, respectively.<sup>14,15</sup> Their experiments were performed in dye solutions, and their model,<sup>16</sup> based on the geometry of the system, gives the wavelengths of the laser lines as a function of the incidence angles. In this Letter we present the study of the emission spectrum of a distributed-feedback laser of a Rhodamine 6G (Rh6G) doped polymer excited by two coherent pumps beams in a spin-coated thin film. By using the flexibility of the Lloyd mirror device, we have obtained multiple lines corresponding to second- and third-order Bragg reflections written in the thin film. In addition, we show that the content of the emission spectra can be explained by the saturation effect that occurs in the refractive index during the pumping step. A similar effect has been evidenced in a fiber Bragg grating.<sup>17</sup>

The thin films were composed of a polymer (polymethylmethacrylate) and an organic dye (Rh6G) and were spin coated on glass substrates. The concentration of the polymer was 150 g/l and that of Rh6G solution in 1,2,3-trichloroethan solvent was  $4 \times 10^{-3}$  M/l. The  $2 \mu\text{m}$  thick film acts as a single-mode waveguide and avoids the spectral effects due to multimode propagation.<sup>18</sup> Figure 1 presents the experimental setup. The sample was excited by a pulsed

Nd:YAG laser ( $\lambda = 532 \text{ nm}$ ,  $\Delta t = 35 \text{ ps}$ , 5 Hz). The temporary superposition of the two pumps was monitored by a delay line. The pair of pump beams forms a complex pattern in the thin film, and this intensity modulation creates a phase grating that induces more than two laser lines simultaneously. For each beam, the output wavelength  $\lambda_i$  ( $i = 1, 2$ ) is linked to the grating period by the Bragg condition as  $\lambda_i = n_{\text{eff}} \lambda_p / (m \sin \theta_i)$ , where  $n_{\text{eff}}$  is the effective refractive index,  $\lambda_p$  is the pump wavelength, and  $m$  is the Bragg reflection order. This formula cannot describe the interaction between the coherent pumps, and further theoretical insight is needed to explain the spectral signature of this interaction. We consider two incident pumps, with wavelengths  $\lambda_p$  at angles  $\theta_1$  and  $\theta_2$ , and the reflected beams (see Fig. 2). The electric field amplitude is

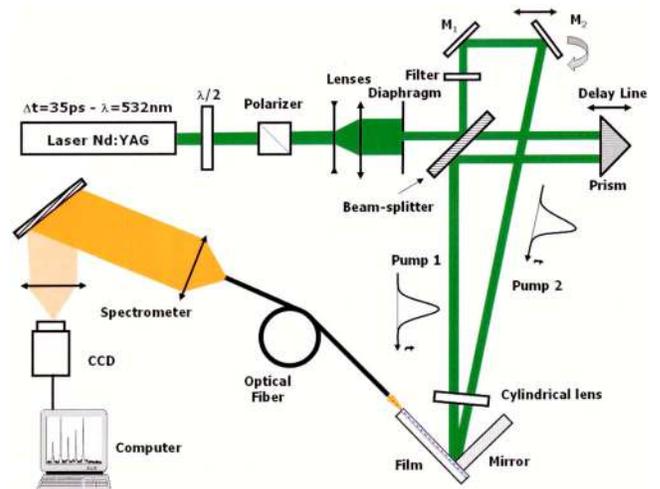


Fig. 1. (Color online) Experimental setup. The emitted beam is collected by an optical fiber (length 1 m) and dispersed by a 1200 lines/mm grating spectrometer.

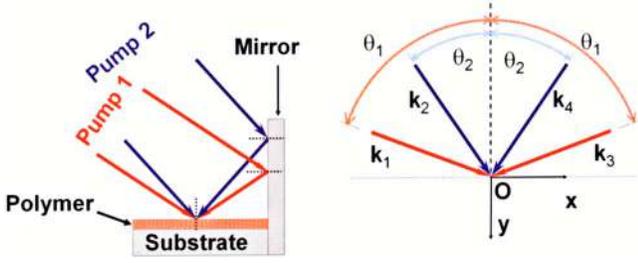


Fig. 2. (Color online) Configuration of incident wave vectors on the sample.

$$E = \sum_{j=1}^4 E_j e^{i\vec{k}_j \cdot \vec{r}}, \quad (1)$$

where  $\vec{k}_j = 2\pi/\lambda_p(\sin \theta_j, \cos \theta_j, 0)$  for  $j=1, 2, 3, 4$ , with  $\theta_3 = -\theta_1$  and  $\theta_4 = -\theta_2$ . The interference pattern is given by the intensity

$$I = |E|^2 = \sum_{j,l=1}^4 E_j E_l^* e^{i(\vec{k}_j - \vec{k}_l) \cdot \vec{r}}. \quad (2)$$

Because the emission is confined inside the polymeric film, only  $\vec{r} = (x, 0, 0)$  and the  $x$  components  $k_j$  of the wave vectors are involved. The nonlinear refractive index  $\Delta n$ , including its imaginary part that corresponds to nonlinear gain or absorption, is, in a first approximation, proportional to  $I$ . However, a saturation occurs above some value  $I_s$  of the intensity. It can be accounted for by the expression  $\Delta n = n_2 I / (1 + I/I_s)$ . Because of saturation, the Fourier spectrum of  $\Delta n$  involves all harmonics of the frequencies present in the spectrum of  $I$ . Since  $k_{3,4} = -k_{1,2}$ , the latter can be written as the sums  $(k_j + k_l)$ , with  $j, l = 1, 2, 3, 4$ . The nonlinear index can be expanded as

$$\Delta n = \sum_{j,l=1}^4 a_{j,l} e^{i(k_j+k_l)x} + \sum_{j,l,m,p=1}^4 b_{j,l,m,p} e^{i(k_j+k_l+k_m+k_p)x} + \sum_{j,l,m,p,q,s=1}^4 c_{j,l,m,p,q,s} e^{i(k_j+k_l+k_m+k_p+k_q+k_s)x} + \dots, \quad (3)$$

where  $a_{j,l}$ ,  $b_{j,l,m,p}$ , and  $c_{j,l,m,p,q,s}$  are the amplitudes of the fundamental frequencies, second harmonics, and third harmonics, respectively.

We now proceed to coupled-wave theory, as introduced by Kogelnik and Shank.<sup>2</sup> They consider a sinusoidal nonlinear index  $\Delta n = n_1 \cos(k_0 x)$ , where  $n_1$  is the amplitude of the modulation and  $k_0$  characterizes its spatial frequency. This factor couples a right-propagating wave  $S(x) \exp(ikx)$  to a left-propagating one  $R(x) \exp(-ikx)$ . The parametric interaction between the two waves involves a spatial dependency  $\exp(2ikx)$  and must be matched with  $\exp(\pm ik_0 x)$  according to the expression of  $\Delta n$ . Therefore we must have  $k = k_0/2$ . In principle, a periodical perturbation of the medium generates an infinity of diffraction orders, but only two orders are phase matched around the Bragg frequency. When the second order of the Bragg reflection is considered, the coupling is due to the second harmonic terms in the Fourier expansion

[Eq. (3)] of the nonlinear index  $\Delta n$ . The intensities of the higher harmonics are assumed to be negligible. Under these conditions,  $k_0 = k_j + k_l + k_m + k_p$ ; hence the wave vectors of the emitted light have the form  $k_0 = (k_j + k_l + k_m + k_p)/2$ , where  $j, l, m, p = 1, 2, 3, 4$ . In order for  $k$  to belong to a spectral domain where gain is present, all  $k_j$  must have the same sign. We get the corresponding wavelengths as

$$\lambda = \frac{n_{\text{eff}} 2\pi}{k} = \frac{2n_{\text{eff}} \lambda_p}{(\sin \theta_j + \sin \theta_l + \sin \theta_m + \sin \theta_p)}, \quad (4)$$

where  $\theta_j, \theta_l, \theta_m$ , and  $\theta_p$ , are either  $\theta_1$  or  $\theta_2$ . When the third order is considered, the coupling is due to the third harmonic, and the possible wave vectors are  $k_0 = (k_j + k_l + k_m + k_p + k_q + k_s)/2$ , while the corresponding wavelengths are

$$\lambda = \frac{2n_{\text{eff}} \lambda_p}{(\sin \theta_j + \sin \theta_l + \sin \theta_m + \sin \theta_p + \sin \theta_q + \sin \theta_s)}. \quad (5)$$

The Bragg reflection order is not explicitly involved in the formula. As mentioned above, we can observe laser lines only in the spectrum interval, which corresponds to the amplified spontaneous emission (ASE) of the dye. In our case, the Rh6G shows a maximum ASE near 560 nm, and the effective refractive index of the film is close to 1.5. Thus the laser lines in this interval are obtained with incident angles close to  $45^\circ$ , corresponding to the second Bragg order ( $m=2$ ) or close to  $20^\circ$  for the third order ( $m=3$ ). The obtained wavelengths originate in the coherent superposition for all Bragg reflection orders. We performed our experiments with  $m=2$  and  $m=3$  to compare the experimental results with the theory. When the incident angle is  $45^\circ$ , the overlap between the reflected and the direct beam in the sample is perfect. Thus the size of the grating is optimal, and the efficiency of the spectral selectivity is maximal. The angle between the pumps is chosen smaller than  $2.8^\circ$  to fall in the region of highest gain of the ASE spectrum of Rh6G. Thus the pump beam is divided into two parts; a set of two mirrors allows the tuning of the incidence angle of the second pump, whereas the incidence angle of the first pump is fixed. We use  $s$  polarization for the pumps and an intensity just above the threshold to avoid multimode propagation.<sup>19</sup> The energy of each pump beam was approximately  $100 \mu\text{J}/\text{pulse}$ , and the conversion efficiency is typically 1%. The linewidth of the laser peaks is 0.5 nm, which is the limit of resolution of our spectrometer.

For  $m=2$  ( $m=3$ ), we used  $\theta_1 = 44.92^\circ$ ,  $\theta_2 = 44.47^\circ$  ( $\theta_1 = 28.16^\circ$ ,  $\theta_2 = 27.90^\circ$ ),  $n_{\text{eff}} = 1.485$ , and  $\lambda_p = 532$  nm. The spectrum of the output is shown as the dashed curve in Fig. 3(a) [Fig. 3(b)]. It appears that the theoretical values calculated with Eq. (4), 559.41, 560.52, 561.63, 562.74, and 563.87 [Eq. (5), 558.00, 558.79, 559.58, 560.38, 561.17, 561.98, and 562.78] almost perfectly match the experimental results: 559.40, 560.45, 561.54, 562.66, and 563.88 (557.96, 558.82, 559.56, 560.37, 561.08, 561.90, and 562.72).

Laser lines 1 and 7 for  $m=3$  (1 and 5 for  $m=2$ ) correspond to the emission of the gratings separately, while the lines 2–6 for  $m=3$  (2–4 for  $m=2$ ) were induced by the coherent interaction between the pump beams. Notice that Eqs. (4) and (5) give the positions of the laser lines, but not their amplitudes.

The basis of the theoretical approach is expansion (3), which requires a numerical confirmation. Indeed, in the case of a periodical pattern, the formation of harmonics due to the saturation of the nonlinear index follows from the properties of the Fourier series. Here the situation is more complicated because the pattern is quasi-periodic with two fundamental frequencies corresponding to  $k_1$  and  $k_2$ . It is therefore interesting to investigate numerically which harmonics will or will not be produced. The electric field  $E$ , the intensity  $I$ , and the nonlinear index  $\Delta n$  are first computed; the spectrum of  $\Delta n$  is then obtained by using the fast Fourier transform algorithm. The coupled wavelength  $\lambda$  is related to the wave vector  $k$  as in the first equality in Eq. (4). We can then plot the spectrum against  $\lambda$  as shown in Fig. 3(a) (solid curve) for  $m=2$  and Fig. 3(b) (solid curve) for  $m=3$ . Notice that the plots are expected to reproduce the locations

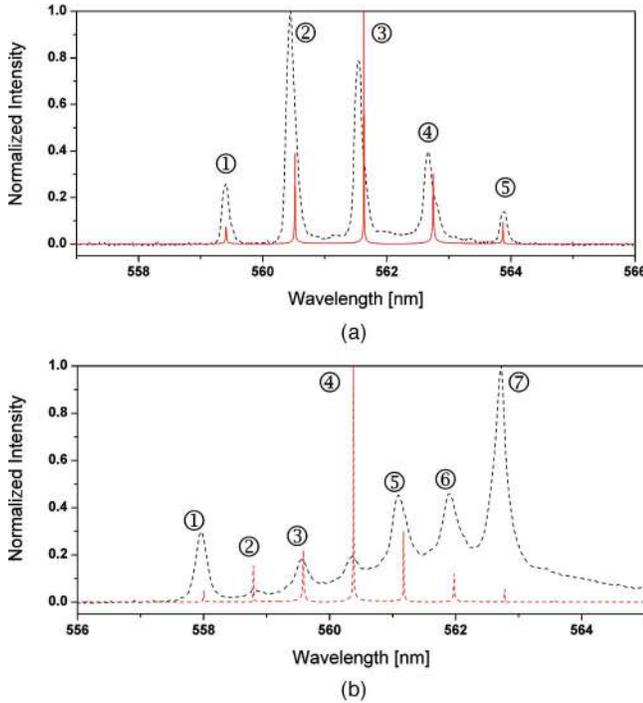


Fig. 3. (Color online) Comparison between theory and experimental results. (a) Experimental spectrum (dashed curve) obtained with  $\theta_1=44.92^\circ$  and  $\theta_2=44.47^\circ$  and plot of a part of the spectrum of the nonlinear index  $\Delta n$  (solid curve) against the corresponding coupled wavelengths with  $m=2$ . The peaks correspond to the wavelengths emitted by the laser. The circles assign an arbitrary number to the laser lines. (b) Experimental spectrum (dashed curve) and calculated spectrum (solid curve) with  $m=3$  obtained with  $\theta_1=28.16^\circ$  and  $\theta_2=27.90^\circ$ . The lines are superposed onto a slope pattern because of residual ASE. The imperfect spatial matching of direct and reflected beams in the sample is probably at the origin of this effect.

of the emitted wavelengths but by no means the corresponding amplitudes. In the numerical simulation, other wavelengths appear corresponding to higher-order harmonics. They are not located between the peaks corresponding to the emission of the independent gratings 1–5 (or 1–7) but outside this range. Their amplitude is smaller and depends strongly on the value of the saturation intensity  $I_s$ . It can also be reduced by taking into account some partial incoherences and amplitude dissymmetry.

In conclusion, we have shown that the multiple laser peaks observed in the distributed-feedback laser pumped by two beams are neither due to a nonlinear interaction between the fundamental lines nor to some higher-order wave coupling; they are due to the presence of higher-order spatial harmonics in the structure of the grating. Taking the higher-order harmonics into consideration allows one to predict the observed laser frequencies with great precision. The formation of the harmonics was interpreted as the consequence of the saturation of the nonlinear refractive index, as shown by the results of our numerical computations.

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