Multichromatic Operations in cw Dye Lasers

Hong Fu and H. Haken
Institut für Theoretische Physik und Synergetik, Universität Stuttgart, D-7000 Stuttgart 80, Federal Republic of Germany
(Received 7 October 1987)

A series of stable multichromatic solutions is presented explicitly based on a band model for cw dye lasers. The results explain recent experimental observations of bichromatic operation in cw dye lasers and predict still higher-order multichromatic steady states.

PACS numbers: 42.55. Mv, 42.55.Bi, 42.60.Gd

Recently, a series of experiments demonstrated a single-frequency laser bifurcating into a bichromatic laser with 20–340-Å spectral splittings in cw rhodamine-6G ring dye lasers.[1-4] The main signatures of the experiments have proved resistant to a qualitative understanding on the basis of the usual laser theory for two-level atoms.[5]

To explain these experiments, we proposed a band model for dye lasers[6] and showed that there may exist multichromatic solutions.[7] It is assumed that the relevant energy-level diagram of cw dye lasers consists of a multilevel ground state and an excited single level as illustrated in Fig. 1. The upper level can interact with each of the lower sublevels, but there is no interaction among the sublevels themselves.

In this paper, we shall present analytically a series of stable multichromatic solutions which will operate in sequence with increasing pump power in cw dye lasers. Our results not only fit the well-known experimental observations, but predict even higher-order stable states; e.g., we predict that the bichromatic operation will become unstable at a threshold, above which a stable trichromatic solution will operate. This prediction provides a criterion for experimentalists to check the three different theories proposed so far.[1,4]

In comparison with our previous work,[7] where the refractive indices were in fact assumed to be unity, we shall solve for them in a self-consistent way in this paper. This is crucial to ensure the stability of the solutions. We point out in advance that the bifurcation point to be given here differs essentially from the instability discussed in Ref. 6.

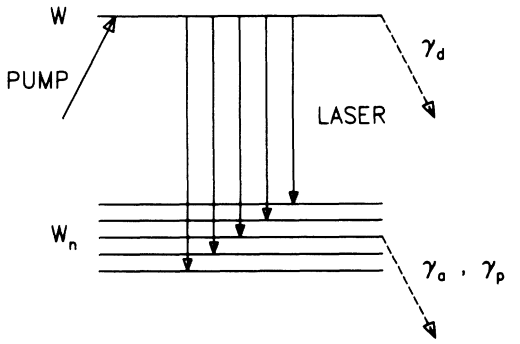


FIG. 1. The relevant energy-level diagram of a dye molecule in a cw dye laser.

The Maxwell-Bloch equations of the band model read as follows[6,7]:

$$\dot{E} = -\kappa E - c\,\partial_z E + \kappa \textstyle\sum_n P_n, \tag{1}$$

$$\dot{P}_n = (-\gamma_p + i\Delta_n)P_n + \gamma_p E f_n (D - A_n), \tag{2}$$

$$\dot{A}_n = -\gamma_a A_n + (\gamma_d/4)(P_n E^* + P_n^* E), \tag{3}$$

$$\dot{D} = \gamma_d \left[D_0 - D - \tfrac{1}{4} \textstyle\sum_n (P_n^* E + P_n E^*) \right]. \tag{4}$$

Here E is the electric field strength, P_n and A_n the polarization and population with respect to the nth sublevel, respectively, and D the population of the upper level. D_0 is the unsaturated population of the upper level due to pump and relaxation processes. All the E, P_n, A_n, and D are normalized quantities.[7] κ, γ_p, γ_a, and γ_d are the relaxation rates of E, P_n, A_n, and D, respectively. Δ_n is the detuning of the nth sublevel from the band center. f_n is proportional to the absolute square of the dipole moment between the nth sublevel and the excited level and satisfies $\sum_n f_n/(1+\Delta_n^2) = 1$.

The lower lasing band is characterized by the detunings $\{\Delta_n\}$ and the dipole moments $\{f_n\}$. For brevity, we assume the sublevels to have an equal spacing Δ, and the distribution of the dipole moments to be a Lorentzian of width Γ within the band and to vanish beyond it, i.e., $f_n \propto 1/[1+(\Delta_n/\Gamma)^2]$ for $|n| \le L$ and $f_n = 0$ for $|n| > L$, where $N_s \equiv 2L+1$ is the number of the sublevels. In this case, the band is characterized by N_s, the bandwidth $B_w \equiv N_s\Delta$, and Γ. The band shape is sketched in Fig. 2.

In what follows, we discuss (1)–(4) under the approximation[7]

$$\gamma_a/\gamma_p, \gamma_d/\gamma_p \to 0, \tag{5}$$

and under the periodic boundary condition and the mean-field limit.

We assume that $E(z,t)$ and $P_n(z,t)$ are composed of

several modes:

$$E(z,t)=\sum_j E(j)\exp[-i\Omega_j(t-z/c)+i\kappa K_j z/c],$$
$$P_n(z,t)=\sum_j P_n(j)\exp[-i\Omega_j(t-z/c)+i\kappa K_j z/c], \quad (6)$$

where the summations include all the lasing modes, e.g., one mode $j=0$ in the single-frequency case and two modes $j=\pm 1$ in the bichromatic case; $E(j)$ and $P_n(j)$ are functions of z and t; Ω_j and $1+\kappa K_j/\omega$ are the splitting from the central frequency ω and the refractive index of the jth mode, respectively. All $E(j)$, $P_n(j)$, Ω_j, and K_j are to be solved. The hypothesis (6) implies a self-consistency requirement; i.e., the resulting mode configuration must be stable. This requirement not only puts constraints on a multichromatic solution, but also determines the actual occurrence of specific multichromatic operations. If we take (6) in (1)-(4), it can be shown under the limit (5) that, unlike the Rabi oscillation, the populations A_n and D do not pulsate at the intermode frequency.[7] This fact decouples the different modes in (1)-(4) and leads to

$$\dot{E}(j)=-\kappa(1+iK_j)E(j)-c\,\partial_z E(j)+\kappa\sum_n P_n(j), \quad (7)$$
$$\dot{P}_n(j)=[-1+i(\Delta_n+\Omega_j)]P_n(j)+f_n E(j)(D-A_n), \quad (8)$$
$$\dot{A}_n=-\gamma_a A_n+(\gamma_d/4)\sum_j[P_n^*(j)E(j)+P_n(j)E^*(j)], \quad (9)$$
$$\dot{D}=\gamma_d\left\{D_0-D-\tfrac{1}{4}\sum_j\sum_n[P_n^*(j)E(j)+P_n(j)E^*(j)\right\}. \quad (10)$$

In terms of the notations $R\equiv\gamma_d/\gamma_a$, $I(j)\equiv(R/2)\times|E(j)|^2$, and $D_n\equiv(D-A_n)$, a stationary solution of (7)-(10) is seen to satisfy the relations

$$D_0=D+(1/R)\sum_j I(j), \quad (11)$$
$$\sum_n \frac{f_n D_n(\Delta_n+\Omega_j)}{1+(\Delta_n+\Omega_j)^2}=K_j \text{ for all } j, \quad (12)$$
$$D_n=D\left[1+\sum_j \frac{f_n I(j)}{1+(\Delta_n+\Omega_j)^2}\right]^{-1}, \quad (13)$$
$$\sum_n \frac{f_n D_n}{1+(\Delta_n+\Omega_j)^2}=1 \text{ for all } j. \quad (14)$$

However, (11)-(14) do not uniquely determine the solution. The additional condition is provided by the requirement of the linear stability of the solution.

To this end, we put the deviations $e(j)$, $p_n(j)$, a_n, and d, which correspond to $E(j)$, $P_n(j)$, A_n, and D, respectively, and have the form $\exp[\lambda t+i\beta z/c]$, in (7)-(10) and find that $p_n(j)$, a_n, and d affect the eigenvalue through the factors $\gamma_p/(i\beta-\gamma_p)$, $\gamma_d/(i\beta-\gamma_a)$, and $\gamma_d/(i\beta-\gamma_d)$, respectively. Therefore, if $\beta\simeq\gamma_d,\gamma_a$, then, because of (5), we can adiabatically eliminate P_n and show that there is no instability; if $\beta\simeq\gamma_p$, then a_n and d would have negligibly small influence on λ, so that λ is determined by

$$\lambda e(j)=-\kappa(1+iK_j)e(j)-i\beta e(j)+\kappa\sum_n p_n(j), \quad (15)$$
$$\lambda p_n(j)=[-1+i(\Delta_n+\Omega_j)]p_n(j)+f_n e(j)D_n. \quad (16)$$

The sign of the eigenvalue is identical to $G(\beta,j)\equiv\mathrm{Re}\lambda_1$, where λ_1 is defined by $\lambda=-i\beta+\lambda_1\kappa+\cdots$.[6] From (15) and (16) we find

$$G(\beta,j)=-1+\sum_n \frac{f_n D_n}{1+(\Delta_n+\Omega_j+\beta)^2}. \quad (17)$$

In consideration of (14), a stable solution must satisfy $\partial_\beta G(\beta,j)|_{\beta=0}=0$, i.e.,

$$\sum_n \frac{f_n D_n(\Delta_n+\Omega_j)}{[1+(\Delta_n+\Omega_j^2]^2}=0 \text{ for all } j. \quad (18)$$

A multichromatic solution is completely determined by (11)-(14) and (18) for a given pump.

The stable multichromatic solutions up to the tetrachromatic operation are shown in Fig. 3. From Fig. 3(a) we see that (i) the spectral splittings are on the order of γ_p, which, for rhodamine-6G, corresponds to some 100 Å splitting in wavelength; (ii) each instability threshold of a multichromatic operation is precisely the operation threshold of the next higher-order multichromatic solution; i.e., there is no bistability. More detailed study shows that there is no jump in the laser intensity either. This result agrees with some of the experiments, but disagrees with others. We mention in passing that the

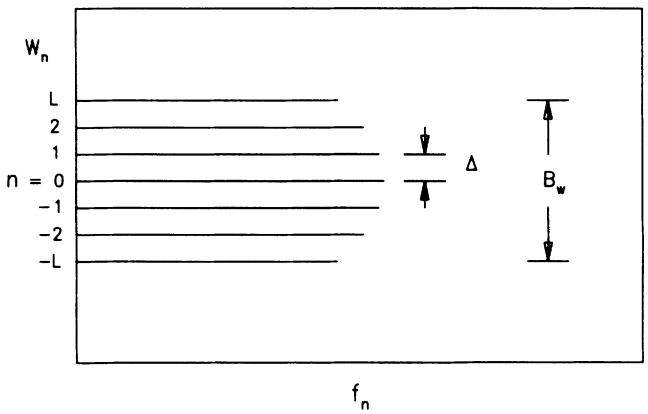


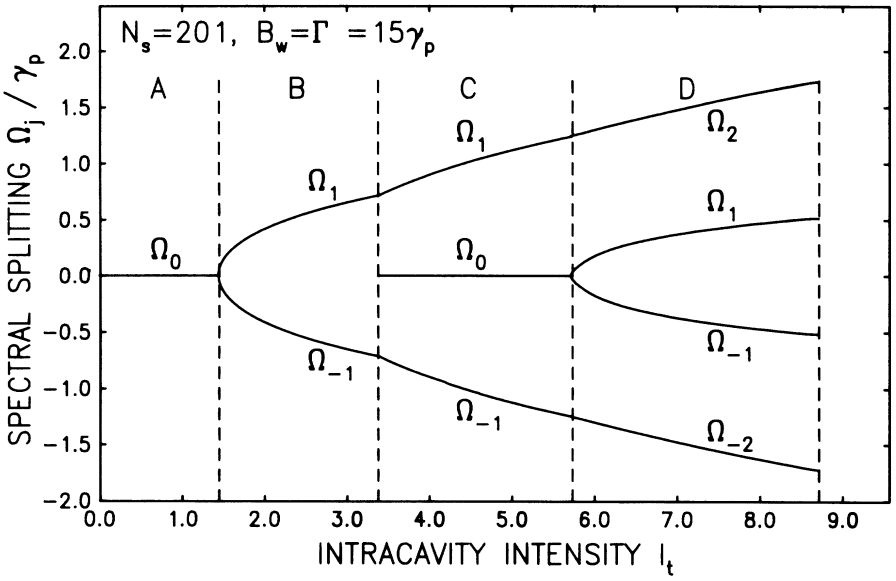
FIG. 2. The distribution of dipole moments on the sublevels.

discontinuity shown in the experiments[1] may be caused by the dispersive prism in the laser cavity and the excited-state absorption of the dye molecules, which have not been taken into account in the present model.

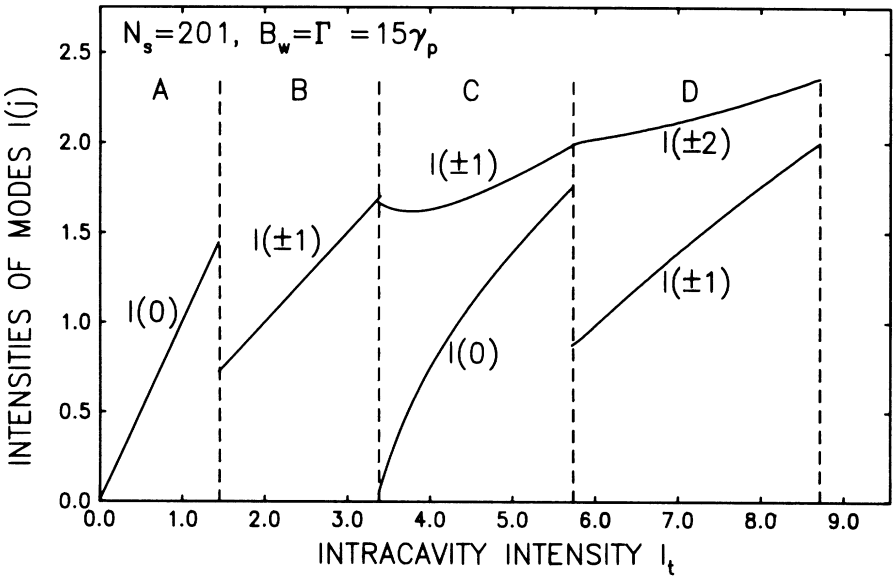
Figure 3(b) shows the energy partition among the modes of a multichromatic operation. Clearly, the instability of a multichromatic solution is always related to the emergence or bifurcation of the central mode.

Physically, the multichromatic solutions can be viewed as the consequence of the balance of two distributions on the sublevels, namely, the dipole moment $\{f_n\}$ and the population $\{A_n\}$, which determine the activity of the sublevels. The former tends to prevent a splitting because the central sublevels have stronger dipole moments while the latter, being determined by the stimulated emission, is conducive to the splitting

For example, in the weak-field case the distribution of the dipole moments dominates and hence the single-frequency operation is expected. This single mode then induces a peak in the distribution of population in the center of the band. When this peak is so high that the sublevels on the two sides of the center become more ac-



(a)



(b)

FIG. 3. Multichromatic solutions as functions of the dimensionless intracavity intensity I_t. *A*, *B*, *C*, and *D* indicate the stable regions for the monochromatic, bichromatic, trichromatic, and tetrachromatic operations, respectively.

tive, the bifurcation will inevitably occur. In the case of the bichromatic operation, two such peaks are induced by the modes of the field. These two peaks tend to move away from each other so that they do not pile up to deactivate the sublevels. This effect becomes more prominent with an increasing lasing field. Therefore, the splitting between the two modes increases with the increasing pump. When these two peaks are separated far away from each other, the center of the band will lose the population and retain the activity. Consequently, a new central mode will grow, leading to the trichromatic operation.

To make a one-to-one comparison with the experiments, data for rhodamine-6G in ethylene glycol are necessary. However, the energy relaxation rates and the density and the dipole moments of the sublevels are not well known as yet; e.g., only the lifetimes of higher vibrational levels have been measured, which may be several orders of magnitude shorter than those of the lower sublevels.[8] If this is indeed the case, then the sensitivity of the bifurcation to the detuning[3] may be explained by our saying that there are some long-living sublevels which are responsible for the bifurcation.

Even assuming the band structure to be of the simplest form as treated in this paper, and using the data of the higher-energy sublevels, i.e., $\gamma_a^{-1} \approx 1$ ps,[9] and taking $\gamma_d^{-1} \approx 1$ ns, which yields $R \approx 0.001$, we are still able to explain the low bifurcation threshold, which was 2 or 3 times the lasing threshold in the experiments; e.g., a band with $N_s = 201$, $B_w = 69\gamma_p$, $\Gamma \rightarrow \infty$, and $R = 0.001$ leads to a bifurcation threshold $D_0 \simeq 2.4$ times the lasing threshold.

We are grateful to C. R. Stroud's group at Rochester University and N. M. Lawandy's group at Brown University for sending us preprints and for valuable discussions. This research was supported in part by the Volkswagenwerk Foundation, Hannover.

[1]L. W. Hillman, J. Krasinski, R. W. Boyd, and C. R. Stroud, Jr., Phys. Rev. Lett. **52**, 1605 (1984).

[2]C. R. Stroud, Jr., K. Koch, and S. Chakmakjian, in *Optical Instabilities,* edited by R. W. Boyd, M. G. Raymer, and L. M. Narducci (Cambridge Univ. Press, Cambridge, 1986), p. 274.

[3]C. R. Stroud, Jr., K. Koch, S. Chakmakjian, and L. W. Hillman, in *Optical Chaos,* edited by J. Chrostowski and N. B. Abraham, The International Society for Optical Engineering Proceedings No. 667 (SPIE, Bellingham, WA, 1986), p. 48.

[4]N. M. Lawandy, R. S. Afzal, and W. S. Rabinovich, Phys. Rev. A **36**, 1759 (1987).

[5]L. A. Lugiato, L. M. Narducci, E. V. Eschenazi, D. K. Bandy, and N. B. Abraham, Phys. Rev. A **32**, 1563 (1985).

[6]Hong Fu and H. Haken, Phys. Rev. A **36**, 4802 (1987).

[7]Hong Fu and H. Haken, to be published.

[8]H. Graener, R. Dohlus, and A. Laubereau, in *Ultrafast Phenomena V,* edited by F. P. Schaefer, Springer Series in Chemical Physics Vol. 46 (Springer-Verlag, Berlin, 1986), p. 458.

[9]D. Ricard and J. Ducuing, J. Chem. Phys. **62**, 3616 (1975).